Harmonic Sputtering Theory

Zhu Lin Zhang
Wayne State University

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HARMONIC SPUTTERING THEORY

by

ZHU LIN ZHANG

DISSERTATION

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of Wayne State University,

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Approved by:

_________________________________________________________________

Advisor Date
DEDICATION

To my parents, my wife and my son, whose support and guidance has been invaluable in helping me accomplish my academic and professional goals.
ACKNOWLEDGMENTS

First of all, I am grateful to Dr. K. R. Padmanabhan, my research advisor, Associate Professor in Department of Physics and Astronomy of Wayne State University in U.S.A., without whose initial supervision (1983-1986) this work could not have been done. Special thanks are due to Dr. P. K. Kuo, Professor in the same Department, for recommending me as a graduate student in the Department in 1980. I am grateful to Peter Sigmund, Professor in Physics Department, University of Southern Denmark for his encouragement and constructive criticisms face to face in 1999. My first heart felt thanks go to my family—my mother S.S. Wang (passed away), my wife B.C. Zhang and my son L. Zhang—for their year after year long time unwavering support.
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CHAPTER 1

INTRODUCTION

1.1 SPUTTERING PHYSICS

When incident particles irradiate a target, they will set off collision cascades in it due to momentum exchange between the ions and atoms in the material. If some of atoms in such cascades recoil and reach the target surface with energy above the surface binding energy, the atoms can be ejected, or sputtered. Sputtering was discovered experimentally more than a century ago. Both experimental and theoretical interest in this phenomenon has increased constantly for many years, due to remarkable applications in different fields.

Sputtering is a typical non-equilibrium, even far-from-equilibrium phenomenon. For example, the Maxwell distribution in Equilibrium Statistics Mechanics couldn’t describe the speed distribution of sputtered particle in general. Though non-equilibrium phenomena abound in nature, but they are still only very poorly understood at a fundamental level. Professor T.D. Lee, Nobel Laureate in Physics, pointed out: “How does a mechanical system approaches equilibrium from non-equilibrium still is unsolved problem up to now….. At near equilibrium, still could get results by using expansion around an equilibrium state. At far-from-equilibrium, one will meet serious difficulty”[134]. From some point of view, sputtering research may open a new window to study non-equilibrium physics.

In 1969, by the aid of the Lindhard power cross-section, Sigmund developed the well-known modern sputtering theory [7]. Why can I make the determination and persist in doing hard work on sputtering research for along time? The answer is a long story.

1.2 PHYSICS FOUND ME AND I FOUND PHYSICS

Thirty-one years ago, on June 9, 1978, I was selected as a graduate student in Institute of Theoretical Physics (ITP), the Chinese Academy of Sciences, Beijing, PRC. My major was Gravitation theory and Relativistic Astrophysics.
30 years before in Beijing, Dr. George Leung, Professor in University of Massachusetts Dartmouth, U.S.A., presented me to Dr. P. K. Kuo, Professor in Department of Physics and Astronomy, Wayne State University, U.S.A., he decided to recommend me as a graduate student in the Department after a short time conversation. In spring 1983, I made a final decision to work on sputtering for PhD under the supervision of Prof. K. R. Padmanabhan in the same department. At the first day, Prof. K. R. Padmanabhan gave me some research reference papers published by P. Sigmund [33] and Y. Yamamura [69]. At the beginning, as a layman, I didn’t know anything about sputtering and related phenomena. In the following days and weeks, I was trying my best to comprehend more and more related reference papers [12][16][70] and build up my background. In the meantime, I was working hard in the Lab to get used to the new and strange research area (for me at that time) as soon as possible. Beyond my original imagination, I felt in love with sputtering due to the logical rigor and internal beauty of Sigmund sputtering theory which is similar to my previous major—the theory of relativity. The true essence of beauty is harmonic. I love harmonic!

According to the Plan of Work made by Prof. K R Padmanabhan, the title of my PhD Dissertation was “SPUTTERING YIELD AND ITS ANGULAR DISTRIBUTION FROM COMPOUND TARGET”. The research work for the proposed dissertation concerns:

i) The measurement of the angular distribution of material sputtered from stoichiometric Group IVa metal nitride compound targets.

ii) The investigation of surface segregation of the constituent atoms of the target.

iii) The explanation of the experimental results based on Sigmund Sputtering theory [33][16].

This work would concentrate on the sputtering yield and surface segregation measurement of the Group IVa metal nitrides. These nitrides had been chosen because of their importance as hard coating materials. The sputtering yield and yield angular distribution under various conditions of ion energy, ion mass and target composition would be investigated. The surface segregation of the constituent atoms from these targets due to ion bombardment would also be studied. This work could
provide fundamental information in understanding the problems associated with composition and phases formed in R. F. sputter deposition process of such hard coatings. Since nitrogen is a volatile element, it certainly will play an unusual roll in sputtering these metal nitride compound targets.

1.3 WHAT IS THE REAL MEANING OF “SURFACE HARNESS ENHANCED BY ION IMPLANTATION”?

Unfortunately, I couldn’t complete my research and had to return to China in 1986. Up until summer 1986, in the experiment respect, only one experimental work had been down, i.e. the sputtering yield angular distribution of Ti and N had been determined by RBS, for TiN target bombarded by 100 keV Xe\(^+\) ion (Chapter-9). In the theory respect, I found a mistake in Yamarmura’s paper [7][69][189]. Later, starting from this finding, I established “Zhang” and “M-Zhang expressions” (Chapter-8). Besides, I have never forgotten Prof. P.K. Rol, Dean of College of Engineering, Wayne State University at that time, because he sent me some his research notes just before I left US for China. Among these notes, I found an article published by J. B. Sanders et.al. [71]. The article helped me to understand “frozen state (Chapter-2). In September 1986, I came back China with “these seeds”. I believed that “these seeds” would help me to stand up. In other words, these seeds enhanced my willpower. This is the real meaning of “surface hardness enhanced by ion implantation”.

1.4 MIND HAS NEVER LEFT SPUTTERING

1.4.1 PRELIMINARY RESULTS

Since came back China, I have concentrated on the analytical theoretical research of Sputtering, because, it’s not possible for me to do any experimental work or computer simulation due to the lack of least conditions. During the last couple of decades, most of published papers are experimental results and computer simulation studies, whereas the analytical work on basic research seems to be relatively rare. In spring, 1987, I obtained the first interest result, i.e. found an intrinsic relation between two different phenomena: one is the definite systematic deviation of the energy
dependence of the low energy heavy ion induce sputtering yield from the original Sigmund’s formula [1][7], another one is the “under cosine” like sputtering yield angular distribution due to the same ion incident. According to the intrinsic relation, by using only one fitting energy parameter in the anisotropic correction term [7][69][140], both phenomena can be calculated and fit the experimental results very well [84][116][141] (Chapter-8).

Following Sigmund theory [16], I derived the nonstoichiometry factors of the anisotropic correction term for a multi-component target. Specially, I calculated the relative abundance of isotopes sputtered in the normal direction compared to oblique direction. The calculated results agree with the experimental measurement [86] reasonably well (chapter 8). Until now, to my knowledge, no correct analytical calculation of the isotopic effect in the low energy heavy ion sputtering has been published. Sigmund had pointed out, "large isotope enrichments observed at very low ion energy are asserted to be caused mainly by recoil implantation."[17]. Later, Sigmund has published some new simulation of energy dependent isotope sputtering [101]. These results gave my analytical formula a very strong support (Chapter 8). All of these facts indicate that, the sign of the anisotropic correction term in the modified Sigmund’s formula must be negative. Then, what is the physical origin of this term? The physical origin must be due to the momentum deposited on the target surface. Furthermore, the negative sign of the anisotropic correction term indicates that the momentum must direct inward of target surface for low energy heavy ion incident. Therefore, the momentum depth distribution turns to be important.

1.4.2 BASIC ASSUMPTION OF THE MOMENTUM DEPOSITED ON THE TARGET SURFACE

Sigmund et.al, go so far as to disregard the facts, they are still insist on that, the momentum must direct outward, because the back sputtering yield must carry the momentum away (out ward) [40], even if the integrated the momentum density deposited on the negative half space outside of target determine a ‘mean velocity of sputtered particle [20]. Therefore, they believe that, the
anisotropic term contributed by momentum deposition, if exist, only cause a positive correction to isotropic flux by narrowing the angular distribution (over cosine like) and broadening the energy spectrum of sputtered particle [7]. Sigmund criticized J. B. Sanders and Y. Yamamura et.al.: “give the wrong sign of anisotropy, because of ignoring the variation with depth of the deposited momentum”[51]. If was positive, the anisotropic term would provide an enhancement in the sputtering yield and would increase in importance with decreasing energy and increasing surface binding energy [51]. Of course, they knew that all of the features directly contradict the experimental measurements and computer simulation results. Therefore, they further pointed out, “this implies that the type of momentum asymmetry discussed here can not be the dominating factor determining the threshold behavior of the sputter yield”[51].

It’s very clear that the key point is the direction of the momentum deposited on the target surface for the low energy heavy ion incident. In 1982, taking the threshold energy $E_1$ into account, J. B. Sanders et.al. computed the spatial distribution of deposited energy and momentum for self-atom bombardment [12]. They came to the conclusion that, the deposited energy density on target surface is approximately independent of $E_1/E$ for $0.1 \geq E_1/E > 0$, but, the momentum density is very sensitive to $E_1/E$, their calculation showed that the momentum is preferably directed inwards as long as $E_1/E > 0.045$. But, P. Sigmund criticized J. B. Sanders et. al. “the use of an arbitrary, undefined, and irrelevant energy parameter $E_1$ in the analysis.”[142]. Later, he emphasized once again [40], “in the outer regions of a cascade, the drift velocity implies that the local drift velocity must have a component opposite to the initial velocity in certain regions of space.” Due to the same reason, L. G. Glazov also pointed out, “The curve (in [5]) for the case $E_1/E = 0.1$ indicates positive values of the distribution for $x \leq 0$. It is a very strange result from a physical point of view.” Therefore, it’s necessary analyze the physical meaning of the momentum deposited on the target surface in detail (Chapter-5). At first, we should note that, the momentum, deposited in the
target surface layer $\bar{P}(x \sim 0)dx$, consists of two different parts. One part is $\bar{P}_b(x \sim 0)dx$, may be carried by back-sputtered particles. Of course, the drift velocity of them must be directed outward. This part was taken account by P. Sigmund and L. G. Glazov et.al. Another part is $\bar{P}_t(x \sim 0)dx$, may be carried by transmission sputtered particles (relative to the surface layer)[7]. The drift velocity of which certainly is directed inward, this part was neglected by P. Sigmund et.al. i.e.

$$\bar{P}(0) = \bar{P}_b(0) + \bar{P}_t(0) \neq \bar{P}_b(0).$$

Obviously, the correct expression is

$$\bar{P}_b(0) = \bar{P} - \int_{0}^{\infty} \bar{P}(x)dx = \int_{-\infty}^{0} \bar{P}(x)dx \neq \bar{P}(0).[20]$$

Secondly, it's necessary to introduce a threshold energy $E_t$ in the analysis [12], because, each target must have a certain value of surface and bulk binding energy ($U_b$). In this dissertation, a plenty of momentum deposition depth profiles had been calculated and plotted for many ion-target systems, perpendicular incidence and $m = 0.2$ in chapter 5. Our calculation confirmed the conclusion given by J. B. Sanders et.al. [12]. For each ion-target pair and $m$, must exist a certain threshold, as long as incident goes energy beyond the threshold. Just as Yamamura pointed out early [69][70], in a collision cascade induced by a low energy ion incidence, after few scattering, both recoil and scattered atoms still prefer to keep the incident direction, the drift velocity of them certainly is directed inward. The basic assumption in this work is that the momentum deposited in the target surface directed parallelly to the ion incidence for the low energy and heavy ion bombardment.

### 1.5 BLOSSOM AND BEAR FRUIT

In the year 1997, I submitted my two virgin papers to Nuclear Instruments and Methods in Physics Research Section B (NIM-B) for a publication [18]. Both papers were successfully published in 1999. After that, Peter Sigmund, Professor in Physics Department, University of Southern Denmark, invited me to present ICACS - 18 (International Conference on Atomic Collisions in Solids, August 3-8, 1999, Odense, Denmark). As a chairman of the Conference, Prof.
Sigmund was very busy, but drove a car to Odense railway station to greet me and arrange my accommodations by himself. I was really moved and couldn't believe it really happen. As very serious physics, Prof. Sigmund gave me tremendous encouragements and at the same time, I received a lot of constructive criticisms. Ever since, I have published more than 20 articles in professional journals and was the thesis adviser to one master degree student in China. The Project “BASIC THEORETICAL STUDIES OF COLLISIONAL SPUTTERING” received the Third-Class Prize in Natural Science of Anhui Province of China in 2008.

In the year 2005, Dr. Dietmar Fink, Editor-in-Chief of Radiation Effects and Defects in Solids, wrote me: “As we know from your recent contributions to our journal that you are an outstanding scientist in the field of theoretical description of radiation effects, we would like to offer you to become the Regional Editor for our journal for China, specifically for manuscripts treating theoretical problems”. Up to now, the most important research results are following:

i) Instead of regular power scattering cross section, the exact scattering cross section was introduced to Sigmund theory for a power potential interaction. All traditional transport equations can be solved asymptotically [19]. One of the applications of the above exact scattering cross section is reducing the large discrepancy between Sigmund theory and experimental results of the depth of origin of sputtered atoms. Sigmund sputtering formulae remain unchanged, the hard sphere collision sputtering theory partly survived [40].

ii) Taking the momentum deposition into account, a new modified Sigmund Sputtering Theory has been developed to describe anisotropic sputtering phenomena induced by low energy and heavy ion bombardment, including sputtering yield energy and angular distributions as well as isotopic effect. The momentum deposition usually ignored, but could play an important role in the atom collisions in solid, such as the anisotropic transport in the ion mixing [73]. For BN target bombarded by low energies \( Xe^+ \) ion, our new theory predicted that N is preferentially sputtered and the surface enrichment of B and light isotope of boron \( ^{10}B \) preferentially sputtered.
CHAPTER 2
LINEAR TRANSPORT EQUATIONS
IN SPUTTERING THEORY

2.1 BASIC TRANSPORT EQUATIONS

2.1.1 BACKWARD BOLTZMANN EQUATIONS

Consider a random, infinite n-components medium with \( \alpha_j N \) atoms of type \( j \) (atomic number \( Z_j \), atomic mass \( M_j \)) per unit volume. \( \alpha_j (0 \leq \alpha_j \leq 1; \sum_j \alpha_j = 1) \) is the concentration of \( j \)-atoms, and \( N \) the atom number density \([\text{atoms/cm}^3]\) [1]. Let’s assume an i-atom starting its motion at \( t = 0 \) and \( r = 0 \) with velocity \( \vec{v}_i \). The basic quantity of interest is \( G_{ij}(\vec{r}, \vec{v}_i, \vec{v}_0, t) d^3\vec{v}_0 d^3\vec{r} dt \), which is the statistical average number of \( j \)-atoms moving in a time interval \((t, dt)\) and in a volume element \((\vec{r}, d^3\vec{r})\) with velocity \((\vec{v}_0, d^3\vec{v}_0)\). After a time \( \delta t \), it may or may not have made a collision. Because of the translation invariance in space and time, see Fig. 2.1.1, we have

\[
N_v \delta t \sum_k \alpha_k \left[ d\sigma_{ik}(\vec{v}_i; \vec{v}_k, \vec{v}_s^*) \left[ G_{ij}(\vec{r}, \vec{v}_i, \vec{v}_0, t) + G_{kj}(\vec{r}, \vec{v}_s^*, \vec{v}_0, t) \right] \right] + \left[ 1 - N_v \delta t \sigma_i(\vec{v}_i) \right] G_{ij}(\vec{r} - \vec{v}_i \delta t, \vec{v}_i, \vec{v}_0, t - \delta t) = G_{ij}(\vec{r}, \vec{v}_i, \vec{v}_0, t)
\]

(2.1.1)

Here \( d\sigma_{ik}(\vec{v}_i; \vec{v}_k, \vec{v}_s^*) = \sigma_{iik}(\vec{v}_i; \vec{v}_k, \vec{v}_s^*) \) is the differential cross section for scattering between an i-atom moving with velocity \( \vec{v}_i \) and a k-atom at rest. \( \vec{v}_s^* \) and \( \vec{v}_s \) represent the velocities of scattered and recoiled atoms respectively, see Fig. 2.1.2, \( \sigma_{i}(\vec{v}_i) \equiv \int d\sigma_{iik}(\vec{v}_i; \vec{v}_k, \vec{v}_s) \) is the corresponding total cross-section, and \( \sigma_i(\vec{v}_i) \equiv \sum_k \alpha_k \sigma_{iik}(\vec{v}_i) \).

The initial condition is given by

\[
G_{ij}(\vec{r}, \vec{v}_i, \vec{v}_0, t = 0) = \delta_j \delta(\vec{r}) \delta(\vec{v}_i - \vec{v}_0)
\]

(2.1.2)

Since \( N_v \delta t \sigma_i(\vec{v}_i) \) represents the probability for making a collision, so must be less than unity. Thus,
Eqs. (2.1.1) is tenable only for finite $\sigma_i(\bar{\nu})$ [2]. The solution may be extended to $\sigma_i(\bar{\nu}) = \infty$ finally, if it exists. But, even so, $\delta t$ approaching to zero should be executed first, then $\sigma_i(\bar{\nu}) = \infty$ to guarantee against the occurrence of $N_0 \delta t \sigma_i(\bar{\nu}) \to \infty$.  

After expansion of this Eq. (2.1.1) in powers of $\delta t$, from the first order terms, one obtained the Backward Boltzmann:

$$
-\left( \frac{1}{v} \frac{\partial}{\partial \tau} + \bar{\nu} \cdot \nabla_r \right) G_y(r, \bar{\nu}, \bar{\nu}_0, t) = N \sum_{k=1}^{n} \alpha_k \int \mathsf{d} \sigma_{ik}(\bar{\nu}; \bar{\nu}', \bar{\nu}^\sigma) \\
\times \left[ G_y(\bar{r}, \bar{\nu}, \bar{\nu}_0, t) - G_y(\bar{r}, \bar{\nu}', \bar{\nu}_0, t) - G_y(\bar{r}, \bar{\nu}^\sigma, \bar{\nu}_0, t) \right]
$$

(2.1.3)

Great successes of which made many authors take it for granted that Eqs. (2.1.3) must be equivalent to Eqs. (2.1.1). If the total cross-section is infinite, this equivalence may not always be true. For example, a Russian theoretician, Dr. Lev G. Glazov, created a paradox [3][4][5] in the calculation of energy and momentum depth distribution by using the famous power cross-section [1][2]. The paradox here refers to a self-contradiction: an equation derived based on the space translation invariance has such a solution that violates the invariance.

Fig.2.1.1 Geometry for derivation of transport equation.
2.1.2 EQUIVALENCE BETWEEN FORWARD AND BACKWARD BOLTZMANN EQUATION

As everyone knows, the Boltzmann Equation in the Statistical Mechanics \[6\] reads

\[
\left( \frac{\partial}{\partial t} + \mathbf{u} \cdot \nabla \right) f_j(\mathbf{r}, \mathbf{u}, t) = \sum_{k=1}^{N} \int \sigma_{jk}(\mathbf{u}, \mathbf{u}_1; \mathbf{u}', \mathbf{u}'') d^3 \mathbf{u}_1 d^3 \mathbf{u}' d^3 \mathbf{u}''
\]

\[
\times \left[ f_j(\mathbf{u}'') f_k(\mathbf{u}') - f_j(\mathbf{u}) f_k(\mathbf{u}_1) \right]
\]

\[ (2.1.4) \]

For a dilute cascade, this equation can be linearized to Forward Boltzmann equation \[7\].

\[
-\left( \frac{1}{\mathbf{v}} \frac{\partial}{\partial t} + \frac{\mathbf{v}}{\mathbf{u}} \cdot \nabla \right) \Psi_j(\mathbf{r}, \mathbf{u}, t)
\]

\[
= N \sum_k \int d^3 \mathbf{u}' \int d^3 \mathbf{u}'' \left[ \alpha_k \Psi_j(\mathbf{r}, \mathbf{u}', t) \sigma_{jk}(\mathbf{u}_1; \mathbf{u}', \mathbf{u}'') - \alpha_j \Psi_k(\mathbf{r}, \mathbf{u}'', t) \sigma_{kj}(\mathbf{u}_1; \mathbf{u}', \mathbf{u}'') \right]
\]

\[ (2.1.5) \]

Here \( \Psi_j(\mathbf{r}, \mathbf{u}, t) \equiv \mathbf{u} G_{ij}(\mathbf{r}, \mathbf{u}_1, \mathbf{u}, t) \) is the flux of j-atom.

Introducing propagator functions \( G_{ij}(\mathbf{r}_0, \mathbf{u}_0, t_0; \mathbf{r}, \mathbf{u}, t) \) by way of the definition

\[
g_j(\mathbf{r}, \mathbf{u}, t) = \sum_m \int d^3 \mathbf{r}_0 \int d^3 \mathbf{u}_0 g_m(\mathbf{r}_0, \mathbf{u}_0, t_0) G_{mj}(\mathbf{r}_0, \mathbf{u}_0, t_0; \mathbf{r}, \mathbf{u}, t)
\]

it’s straightforward to prove equivalence between Backward Boltzmann equations (2.1.3) and Forward Boltzmann equations (2.1.5) in a multi-component medium \[8\], see Appendix IIA.

In most of the applications, we are only interested in the functions \[1\]
\[ G_i(\vec{r}, \vec{u}, \vec{v}_0) \equiv \int_0^t G_{ij}(\vec{r}, \vec{u}, \vec{v}_0, t) dt \]

Taking the initial conditions (2.1.2) into account, directly integrating Eqs. (2.1.3, 5), we obtained

\[
N \sum_{k=1}^{n} \alpha_k \int d\sigma_{ik} (\vec{u}; \vec{u}', \vec{u}'') \left[ G_{ij}(\vec{r}, \vec{u}, \vec{v}_0) - G_{ij}(\vec{r}, \vec{u}', \vec{v}_0) - G_{kij}(\vec{r}, \vec{u}'', \vec{v}_0) \right] = \frac{1}{v_0} \delta_{ij} \delta(\vec{u} - \vec{v}_0) - \vec{n} \cdot \nabla G_{ij}(\vec{r}, \vec{u}, \vec{v}_0),
\]

(2.1.6)

\[
N \sum_{k} \int d^3\vec{u}' \int d^3\vec{u}'' \left[ \alpha_k \Psi_j(\vec{r}, \vec{u}) \sigma_{jk}(\vec{u}; \vec{u}', \vec{u}'') - \alpha_k \Psi_j(\vec{r}, \vec{u}') \sigma_{jk}(\vec{u}; \vec{u}', \vec{u}'') - \alpha_j \Psi_k(\vec{r}, \vec{u}'') \sigma_{kj}(\vec{u}'; \vec{u}'', \vec{u}') \right] = \delta_{ij} \delta(\vec{u} - \vec{v}_0) - \vec{n} \cdot \nabla \Psi_j(\vec{r}, \vec{u})
\]

(2.1.7)

Here \( \vec{n} \equiv \vec{u} / v \) and \( \Psi_j(\vec{r}, \vec{u}) = v G_{ij}(\vec{r}, \vec{u}_1, \vec{u}) \).

\[ 2.2 \] FREEZING STATE OF COLLISION CASCADE

\[ 2.2.1 \] THREE DENSITIES: \( F_{ij}^r, F_{ij}^s \) AND \( F_{ij}^c \)

Freezing recoil density \( F_{ij}^r \), scattering density \( F_{ij}^s \) and collision density \( F_{ij}^c \) were introduced and derived asymptotically by Sanders, et al. for a monatomic medium \([9][10][11][12]\). Following their track \([13][14][15]\), a particle with energy above a threshold energy \( U \) is able to create a cascade and slows down till it’s energy \( E_0 \equiv \frac{1}{2} M \nu^2 \leq U \), then the particle will keep on moving with it’s final energy \( E_0 \). In other words, if a incident particle with energy \( E \equiv \frac{1}{2} M \nu^2 > U \), a collision cascade is induced until a steady state established. Finally, there aren’t any particles with energy \( E_0 > U \), i.e.

\[
F_{ij}^c(\vec{r}, \vec{u}, \vec{v}_0, U) = F_{ij}^r(\vec{r}, \vec{u}, \vec{v}_0, U) = F_{ij}^s(\vec{r}, \vec{u}, \vec{v}_0, U) = 0, \quad \text{if} \quad E_0 > U.
\]

(2.2.1)

An incident particle with energy \( E \leq U \) has no capability to create any collision cascade, thus

\[
F_{ij}^c(\vec{r}, \vec{u}, \vec{v}_0, U) = F_{ij}^s(\vec{r}, \vec{u}, \vec{v}_0, U) = \delta_{ij} \delta(\vec{u} - \vec{v}_0), \quad \text{if} \quad E \leq U.
\]

(2.2.2)

For \( E > U \geq E_0 \), \( F_{ij}^r \), \( F_{ij}^s \) and \( F_{ij}^c \) are related to particle fluxes in the following way

\[
F_{ij}(\vec{r}, \vec{u}, \vec{v}_0, U) = N \alpha_j \sum_k \int d^3\vec{u}_p \Psi_k(\vec{r}, \vec{u}_p) \int_{\vec{u}_p \leq U} d\sigma_{kj}(\vec{u}_p; \vec{u}', \vec{u}'') \delta(\vec{u}' - \vec{v}_0)
\]

(2.2.3)
\[ F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) = N \sum_k \alpha_k \int d^3 \bar{\nu}_p \Psi_j(\bar{r}, \bar{\nu}_p) \int_{\bar{\nu} \leq U} d\sigma_{jk}(\bar{\nu}_p; \bar{\nu}', \bar{\nu}'') \delta(\bar{\nu}' - \bar{\nu}_0) \]  
\[ (2.2.4) \]

\[ F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) = F^s_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) + F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) \]  
\[ (2.2.5) \]

Freezing free can be considered as a special case of Freezing: \( U = E_0 \), i.e.

\[ H^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0) \equiv H^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U = E_0) \]  
\[ (2.2.6) \]

Here, \( H_{ij} \) stands for one of \( F^s_{ij} \), \( F^c_{ij} \) and \( F^c_{ij} \). In Freezing free case, the collision cascade can go on forever theoretically \([16][17][18]\), no steady state could be reached.

Substituting (2.2.3~5) into Eqs. (2.1.6), we have the following equations for \( F^c_{ij} \):

\[
\begin{aligned}
& N \sum_k \alpha_k \int d\sigma_{ik}(\bar{\nu}; \bar{\nu}', \bar{\nu}'') \left[ F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) - F^c_{ij}(\bar{r}, \bar{\nu}', \bar{\nu}_0, U) \right] \\
& \quad - F^c_{ij}(\bar{r}, \bar{\nu}', \bar{\nu}_0, U) = -\bar{n} \cdot \nabla F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) \\
& F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) = \delta_{ij} \delta(\bar{\nu} - \bar{\nu}_0) \quad \text{if} \quad E \leq U.
\end{aligned}
\]  
\[ (2.2.7) \]

(2.2.2) appeared in (2.2.7) stands for the incident particle, it’s usually ignored by others authors \([11][13][15]\), but, it certainly plays a critical role in the entire linear transport theory of sputtering \([11]\). No incident particle causes no collision cascade.

### 2.2.2 ENERGY AND MOMENTUM DEPOSITIONS: \( F_{ij}^E \), \( \bar{P}_{ij} \), RECOIL AND SCATTERING NUMBERS: \( N_{ij} \), \( \bar{N}_{ij} \) IN A FREEZING STATE

According to the “frozen in” picture, it’s natural to define space distribution functions as following \([11][13][19][20]\),

\[ F_{ij}(\bar{r}, \bar{\nu}, U) = \int_0^U d^3 \bar{\nu}_0 \cdot \left( \frac{1}{2} M_j \bar{\nu}_0^2 \right) \cdot F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) \]  
\[ (2.2.8) \]

\[ \bar{P}_{ij}(\bar{r}, \bar{\nu}, U) = \int_0^U d^3 \bar{\nu}_0 \cdot \left( M_j \bar{\nu}_0 \right) \cdot F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) \]  
\[ (2.2.9) \]

\[ N_{ij}(\bar{r}, \bar{\nu}, U) = \int_0^U d^3 \bar{\nu}_0 \cdot F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) \]  
\[ (2.2.10) \]

\[ \bar{N}_{ij}(\bar{r}, \bar{\nu}, U) = \int_0^U d^3 \bar{\nu}_0 \cdot F^c_{ij}(\bar{r}, \bar{\nu}, \bar{\nu}_0, U) \]  
\[ (2.2.11) \]

(2.2.8,9) are equivalent to the definitions of energy and momentum distribution functions in \([11]\).
The energy and momentum deposition space distribution functions of i-particle are defined as,

\[
F_{(i)}(\vec{r}, \vec{\nu}, U) = \sum_j F_{(ij)}(\vec{r}, \vec{\nu}, U) \quad (2.2.12)
\]

\[
\vec{P}_{(i)}(\vec{r}, \vec{\nu}, U) = \sum_j \vec{P}_{(ij)}(\vec{r}, \vec{\nu}, U) \quad (2.2.13)
\]

By using (2.2.8,9) and (2.2.12,13), directly taking integration and summation of Eqs. (2.2.7), we have the following equations for \( F_{(i)}(\vec{r}, E, U) \) and \( \vec{P}_{(i)}(\vec{r}, \vec{\nu}, U) \) respectively:

\[
\begin{align*}
\int N \sum_k \alpha_k \int d\sigma_k (\vec{\nu}; \vec{\nu}', \vec{\nu}^*) [H_{(i)}(\vec{r}, \vec{\nu}, U) &- H_{(j)}(\vec{r}, \vec{\nu}', U)] \\
&- \vec{n} \cdot \nabla_i H_{(i)}(\vec{r}, \vec{\nu}, U) \end{align*} \quad (2.2.14)
\]

Here, \( H_{(i)} \) stands for one of \( F_{(i)} \) and \( \vec{P}_{(i)} \). Directly integrating Eqs. (2.2.14) over entire space yielded

\[
\int d^3\vec{r} \cdot F_{(i)}(\vec{r}, \vec{\nu}, U) = E \quad (2.2.15)
\]

\[
\int d^3\vec{r} \cdot \vec{P}_{(i)}(\vec{r}, \vec{\nu}, U) = M_i \vec{\nu} \quad (2.2.16)
\]

(2.2.15,16) represent the energy and momentum conservations respectively. Even though (2.2.15,16) are important for space moments computation, but they still can’t take the place of incident conditions in Eqs. (2.2.14).

### 2.3 PARTICLE ENERGY SPECTRUMS AND DISTRIBUTIN FUNCTIONS

#### 2.3.1 PARTICLE ENERGY SPECTRUMS

Total particle energy spectrums are defined by entire space integration and then expanded in terms of Legendre Polynomials [16~19]:

\[
H_{ij}(\vec{\nu}, \vec{\nu}_0) = \int d^3\vec{r} \cdot H_{ij}(\vec{r}, \vec{\nu}, \vec{\nu}_0) = (4\pi)^{-3} \sum_{L=0}^{\infty} (2L+1)H_{ij}^L(E, E_0)P_L(\eta_0) \quad (2.3.1)
\]

Here, \( H_{ij} \) stands for one of \( G_{ij}, F_{ij}^s, F_{ij}^c, \vec{n}_0 = \vec{\nu}_0 / \nu_0, \eta_0 = \vec{n} \cdot \vec{n}_0 \) and \( P_L(\eta_0) \) are Legendre Polynomials. By using (2.2.3,4), directly integrating Eqs.(2.1.6) over entire space and expanding them in terms of Legendre Polynomials respectively, finally we obtained,
\[ \sigma^L (E_0) G^L_0 (E, E_0) = (N_{\nu_0})^{-1} \delta_0 \delta (E - E_0) \]  
(2.3.2)

\[ \sigma^L (U) F^L_0 (E, E_0, U) = \alpha_j P_L [\sqrt{E_0 / E_m}] \sigma^L_0 (E, E_0), \]
(2.3.3)

\[ \sigma^L (U) F^L_0 (E, E_0, U) \]

\[
= \delta_0 \sum_k \alpha_k P_L [U_{jk} \sqrt{E_0 / E} + V_{jk} \sqrt{E / E_0}] \sigma^L_{jk} (E, E - E_0)
\]
(2.3.4)

\[ F^L_0 (E, E_0, U) = F^L_0 (E, E_0, U) + F^L_0 (E, E_0, U) \]

\[ N \sum_k \int_0^{E_k} dT \left[ \alpha_k \sigma^L_{jk} (E, T) \psi_j^L (E - \alpha_k \sigma^L_{jk} (E + T)) P_L \left( \cos \psi^*_{jk} \right) \psi_j^L (E + T) \right. \]

\[ - \left. \alpha_j \sigma^L_{jk} (E + T, E) P_L \left( \cos \psi^*_{jk} \right) \psi_j^L (E + T) \right] = \delta_0 \delta (E - E_0) \]
(2.3.5)

Here \( d \sigma^L_{ik} (E, T) = \sigma^L_{ik} (E, T) dT = d \sigma^L_{ik} (\vec{u}; \vec{u}', \vec{u}''); \gamma_{ik} \equiv 4M_i M_k (M_i + M_k)^{-2}; \)
\( T_m = \gamma_{ik} E; U_{ik} \equiv (M_i + M_k)/(2M_i); V_{ik} \equiv (M_i - M_k)/(2M_i) \) and
\[ \begin{cases} 
\cos \psi^*_{jk} = U_{ik} \sqrt{(E - T)/E} + V_{ik} \sqrt{E/(E - T)} \\
\cos \psi^*_{jk} = \sqrt{T/T_m}
\end{cases} \]
(2.3.6)

\( \sigma^L (U) \), acting on \((i, E)\), is such a linear operator that, for any set of functions \( H^L_i (E) \), we have
\[ \sigma^L (U) H^L_i (E) = \sum_k \alpha_k \int_0^{E_k} d \sigma^L_{ik} (E, T) H^L_i (E) - \\
- \left[ I - \theta (U - E + T) \right] P_L \left( \cos \psi_{ik} \right) H^L_i (E - T) - \left[ I - \theta (U - T) \right] P_L \left( \cos \psi_{ik} \right) H^L_i (T). \]
(2.3.2~4) were derived by P. Sigmund [16][17] for freezing free and \( L = 0 \). From now on, we make an agreement: if \( L = 0 \), “0” can be neglected, such as \( H_{ij} (E, E_0, U) = H^0_{ij} (E, E_0, U) \).

### 2.3.2 NUMBER OF FREANKEL

Based on a simple physics consideration, for single element medium, we can define
\[ N^i (E) = \int_{0.5U}^{U} F^c (E, E_0, U) dE_0 \]
as the average number of Frenkel-pairs (vacancy- interstitial) [21][22]created by a primary knock-on atom with energy \( E \). Directly integrating (2.3.3,4) yields
\[ \sigma (U) N^i (E) = \int_{0.5U}^{U} \left[ d \sigma (E, T) + d \sigma (E, E - T) \right] \]
or
(2.3.7)
\[
\int_0^E d\sigma(E, T)[N_f(E) - N_f(E - T) - N_f(T)] = 0
\]
\[
N_f(E) = 0(U - E) - 0(0.5U - E), \quad \text{if } E \leq U
\]

Here \(0.5U\) is effective displacement threshold energy. The later equation appeared in [22].

### 2.3.3 ENERGY & MOMENTUM PARTITIONING AND PARTIAL PARTICLE NUMBERS

Energy & momentum partitioning, partial recoil & scattering number [11][13][19] are defined

\[
H_{ij}(E, U) = \int_{-\infty}^\infty d^3r \cdot H_{ij}(\vec{r}, \vec{v}, U)
\]

(2.3.8)

Here \(H_{ij}\) stands for one of \(F_{ij}, \bar{P}_{ij}, N_{ij}\) and \(N_{ij}^a\). By using (2.2.8~11), directly integrating Eqs.(2.2.7) over entire space and taking the incident condition into account, we derived

\[
\sigma(U)F_{ij}(E, U) = \alpha_j \int_0^U d\sigma_{ij}(E, T)T + \delta_{ij} \sum_k \alpha_k \int_{E-U}^E d\sigma_{ik}(E, T)(E - T)
\]

(2.3.9)

\[
\sigma(U)P_{ij}(E, U) = \alpha_j \int_0^U d\sigma_{ij}(E, T)(\cos \phi_{ij}^a)^{\sqrt{2M_jT}}
\]

+ \delta_{ij} \sum_k \alpha_k \int_{E-U}^E d\sigma_{ik}(E, T)(\cos \phi_{ik}^a)^{\sqrt{2M_j(E - T)}}

(2.3.10)

\[
\sigma(U)N_{ij}(E, U) = \alpha_j \int_0^U d\sigma_{ij}(E, T)
\]

(2.3.11)

\[
\sigma(U)N_{ij}^a(E, U) = \delta_{ij} \sum_k \alpha_k \int_{E-U}^E d\sigma_{ik}(E, T)
\]

(2.3.12)

Here \(\bar{P}_{ij}(E, U) \equiv \vec{n} \cdot P_{ij}(E, U)\). Substituting (2.2.3~5) into (2.2.8~11) respectively and using Eqs. (2.3.5), we have: [11][13~19]

\[
F_{ij}(E, U) = [1 - \theta(U - E)] \cdot \omega_j(U) + \delta_{ij} \theta(U - E)E
\]

(2.3.13)

\[
P_{ij}(E, U) = [1 - \theta(U - E)] \cdot \xi_j(U) + \delta_{ij} \theta(U - E)\sqrt{2M_jE}
\]

(2.3.14)

\[
N_{ij}(E, U) = [1 - \theta(U - E)] \cdot \zeta_j(U)
\]

(2.3.15)

\[
N_{ij}^a(E, U) = [1 - \theta(U - E)] \cdot \chi_j(U) + \delta_{ij} \theta(U - E)
\]

(2.3.16)

Here \(\omega_j(U) \equiv N \sum_k \int_0^E dE \left[ \psi_k(E')\alpha_j \int_0^U d\sigma_{kj}(E', T) + \psi_j(E')\alpha_k \int_{E-U}^E d\sigma_{jk}(E', T)(E' - T) \right] \)

\[
\xi_j(U) \equiv N \sum_k \int_0^E dE' \left[ \psi_k(E')\alpha_j \int_0^U d\sigma_{kj}(E', T)dT(\cos \phi_{kj}^a)^{\sqrt{2M_jT}}
\]

+ \psi_j(E')\alpha_k \int_{E-U}^E d\sigma_{jk}(E', T)dT(\cos \phi_{jk}^a)^{\sqrt{2M_j(E' - T)}} \}

\[
\zeta_j(U) \equiv \int_0^E dE'N\alpha_j \sum_k \psi_k(E') \int_0^U d\sigma_{kj}(E', T)
\]

(2.3.17)
Using Eqs. (2.3.5), taking the derivative of (2.3.13-16) with respect to $U$, \cite{11}\cite{13}\cite{19}, then

\[
\begin{align*}
\mathcal{F}_{(ij)}(E, U)/\partial U + N \sum_k \left[ \psi_j(U) \alpha_k S_{jk}(U) - \psi_k(U) \alpha_j S_{ij}(U) \right] &= 0 \\
F_{(ij)}(E, U) &= \delta_{ij} E, \quad \text{if} \quad E \leq U. \tag{2.3.17}
\end{align*}
\]

\[
\begin{align*}
\mathcal{P}_{(ij)}(E, U)/\partial U + N \sum_k \left[ \psi_j(U) \alpha_k \epsilon_{jk}(U) - \psi_k(U) \alpha_j \epsilon_{ij}(U) \right] &= 0 \\
P_{(ij)}(E, U) &= \delta_{ij} \sqrt{2M_j E}, \quad \text{if} \quad E \leq U. \tag{2.3.18}
\end{align*}
\]

\[
\begin{align*}
\mathcal{N}_{(ij)}(E, U)/\partial U &= F^c_j(U) - F_j(U) \\
N_{(ij)}(E, U) &= 0, \quad \text{if} \quad E \leq U. \tag{2.3.19}
\end{align*}
\]

\[
\begin{align*}
\mathcal{N}_{(ij)}^*(E, U)/\partial U &= F_j(U) \\
N_{(ij)}^*(E, U) &= \delta_{ij}, \quad \text{if} \quad E \leq U. \tag{2.3.20}
\end{align*}
\]

Here $F_j(U) = \int_U^E dE' \sum_k \psi_k(E') \sigma_{kj}(E', U);$$F^c_j(U) = N \alpha_j \sum_k \psi_k(U) \sigma_{kj}(U);$ $S_{jk}(U) = \int_0^U d\sigma_{ij}(U, T) T$ is the nuclear stopping cross section and $T_{jk}(U) = \int_0^U d\sigma_{jk}(E, T) (\cos \phi^*_{jk}) \sqrt{2M_j T} = (M_j + M_k) S_{jk}(U)/\sqrt{2M_j U}$ is the momentum transfer cross section \cite{11}.

By using Eqs. (2.3.17,18) and (2.3.20), it’s straightforward to prove that $F_{(ij)}(E, U)$, $P_{(ij)}(E, U)$ and $N_{(ij)}^*(E, U)$ are continuous functions of $U$ at $U = E$, at least for $\sigma_i(E) = \infty$.

If further assume

\[
F_{(ij)}(E, U) = P_{(ij)}(E, U) = N_{(ij)}^*(E, U) = 0, \quad \text{if} \quad E \leq U \tag{2.3.21}
\]

Eqs. (2.3.17,18,20) will be mathematically equivalence to the corresponding equations appeared in earlier references \cite{11}\cite{13}. It appears that the authors of these references ignored the incident condition (2.2.2), because, (2.3.21) contradicts the second equations (2.3.17,18) and (2.3.20), which’s derived from (2.2.2).
2.4 MOMENTS EQUATIONS FOR DEPOSITED ENERGY AND MOMENTUM DEATH DISTRIBUTION FUNCTIONS

For simplicity, let’s consider the planar Geometry [11][18][20]. For a semi-infinite target with a plane surface at \( z = 0 \), the \( z \)-axis is chosen along the inward surface normal with a unit vector \( \hat{z} \).

Thus, as long as we make the following replacements:

\[
\text{d}^3 \vec{r} \rightarrow \text{d}z, \quad \vec{n} \cdot \nabla_r \hat{H}(\vec{r}) \rightarrow \eta \frac{\partial \hat{H}(z)}{\partial z}, \quad \vec{r} \rightarrow z, \quad \eta \equiv n \cdot \hat{z}, \quad \vec{r} - \vec{v} \delta t \rightarrow z - \eta v dt,
\]

all of the expressions mentioned previously turns out to be suitable for the planar Geometry.

In this work, we are only interested in \( z \) component of momentum deposition \( P_{Dz}(z, \vec{v}, U) \).

The moments are defined and expanded in terms of Legendre Polynomials as following:

\[
H^a_{(i)}(\vec{v}, U) = \int_{-\infty}^{\infty} \text{d}z \cdot z^n \hat{H}^a_{(i)}(z, \vec{v}, U) = \sum_{L=0}^{\infty} (2L + 1) H^a_{(i)L}(E, U) P_L(\eta) \quad (2.4.1)
\]

Here, \( H \) stands for one of \( F \) and \( P \). Substituting (2.4.1) into (2.2.14–16), we derived

\[
\begin{align*}
\Delta = \sigma_{(i)L} & = \hat{H}^a_{(i)L}(E, U) = \sum_{L=0}^{\infty} (2L + 1) H^a_{(i)L}(E, U) P_L(\eta) \\
N \sigma_{(i)L} & = \Delta H^a_{(i)L}(E, U) = H^a_{(i)L}(E, U)
\end{align*}
\]

Here \( \Delta \), acting on \( (n, L) \), is such an operator that, for any set of numbers \( H^a_{L} \), we have

\[
\Delta H^a_{L} = \frac{n}{2L + 1} \left[ L H^a_{L-1} + (L + 1) H^a_{L+1} \right].
\]

APPENDIX II A:

Statement: The equivalence between Forward and Backward Boltzmann Equation in multi-component medium

In the transport theory of sputtering, many authors [1][18][19] use backward Boltzmann Equations as basic equations, many other authors [13][23] like to use forward Boltzmann Equations as a starting point. Of course, these two equations are obviously identical in physics. Sigmund introduced a propagator function and gave a simple proof of equivalence between forward and
backward Boltzmann equations in a monatomic medium by using the propagator [7]. In view of development of sputtering theory, author of this work thinks that it is necessary to generalize the theorem to a multi-component medium.

As is well known, Boltzmann Equation determines each component distribution function $f_j(\mathbf{r}, \mathbf{v}, t)$, $(i = 1, 2, \ldots, n)$, where $f_j(\mathbf{r}, \mathbf{v}, t) d^3r d^3v$ is a statistical average over the number of j-atom in a volume element $(\mathbf{r}, d^3r)$ moving with a velocity $(\mathbf{v}, d^3v)$ at time $t$, for n-components medium. In general, Boltzmann Equation reads [89]

$$\frac{\partial}{\partial t} f_j(\mathbf{r}, \mathbf{v}, t) + \mathbf{v} \cdot \nabla f_j(\mathbf{r}, \mathbf{v}, t) = \sum_{k=1}^{n} \sigma_{jk}(\mathbf{v}, \mathbf{v}_1; \mathbf{v}', \mathbf{v}'') d^3v_1 d^3v' d^3v'' \cdot [\mathbf{v} - \mathbf{v}_1] \left[ f_j(\mathbf{v}') f_k(\mathbf{v}'') - f_j(\mathbf{v}) f_k(\mathbf{v}_1) \right]$$

where $\sigma_{jk}(\mathbf{v}, \mathbf{v}_1; \mathbf{v}', \mathbf{v}'') d^3v_1 d^3v' d^3v''$ is the differential cross section for a j-projectile with initial velocity $\mathbf{v}$ and a k-target atom moving with initial velocity $\mathbf{v}_1$ to scatter each other into velocities $(\mathbf{v}', d^3v')$ and $(\mathbf{v}'', d^3v'')$ respectively. In addition, $f_j \equiv f_j(\mathbf{v}) \equiv f_j(\mathbf{r}, \mathbf{v}, t)$ and $\mathbf{F}_j \equiv \mathbf{F}(\mathbf{r}, \mathbf{v}, t)$ is an external force acting on the moving j-atom. According to a similar discussion in [6], we have

$$\sigma_{jk}(\mathbf{v}, \mathbf{v}_1; \mathbf{v}', \mathbf{v}'') = \sigma_{kj}(\mathbf{v}', \mathbf{v}''; \mathbf{v}, \mathbf{v}_1)$$

$$\sigma_{kj}(\mathbf{v}_1, \mathbf{v}; \mathbf{v}'', \mathbf{v}') = \sigma_{kj}(\mathbf{v}'', \mathbf{v}'; \mathbf{v}_1, \mathbf{v})$$

(IIA.2)

For our purpose, external forces can usually be ignored ($\mathbf{F}_j = 0$) and the target atom can assumed at rest initially, for sufficiently fast projectiles, i. e.

$$f_j(\mathbf{r}, \mathbf{v}_1, 0) = N \alpha_j \delta(\mathbf{v}_1)$$

(IIA.3)

where $\delta$ is the Dirac delta function, $N$ is the target atomic number density \([ \text{atoms/cm}^3 \])$, and $\alpha_j$ \((1 \geq \alpha_j \geq 0, \sum_j \alpha_j = 1\)$ is the concentration of j-atoms.

Two important limiting cases can be specified. If only the fate of the projectile is of interest, the motion of target atom need not be followed, and by using (IIA.2), (IIA.3) can be reduced to
Conversely, when the motion of recoil atoms is important, (IIA.1) can be linearized for a dilute cascade, i.e. where only a small fraction of atoms participate the collision cascade.

\[
f_j(\vec{r},\vec{u},t) = N\alpha_j \delta(\vec{u}) + F_j(\vec{r},\vec{u},t)
\]

(IIA.5)

where \( F_j(\vec{r},\vec{u},t) \) is “small”, by using (IIA.2), Eq.(IIA.1) can be simplified to

\[
-\left( \frac{\partial}{\partial t} + \vec{u} \cdot \vec{\nabla}_r \right) F_j(\vec{r},\vec{u},t) = N\sum_k \alpha_k \int d^3\vec{u}' \int d^3\vec{u}'' \left[ \nu f_j(\vec{u}) \sigma_{jk}(\vec{u};\vec{u}'') - \nu' f_j(\vec{u}') \sigma_{jk}(\vec{u}';\vec{u}'') \right]
\]

(IIA.6)

Here \( F_j \equiv F_j(\vec{r},\vec{u},t), F'_j \equiv F_j(\vec{r},\vec{u}',t) \) and \( F''_j \equiv F_j(\vec{r},\vec{u}'',t) \).

\[\sigma_{jk}(\vec{u};\vec{u}',\vec{u}'') \equiv \sigma_{jk}(\vec{u},0;\vec{u}',\vec{u}'') \text{ and } \sigma_{jk}(\vec{u};\vec{u}') \equiv \int d^3\vec{u}'' \sigma_{jk}(\vec{u};\vec{u}',\vec{u}'').\]

Eq.(IIA.4) can be used as a basis for ion penetration theory, while Eq.(IIA.6) can serve as a basis for the theory of linear collision cascades and usually be called Forward Boltzmann equation.

Introduce propagator functions \( G_{ij} \) by way of the definition

\[
F_j(\vec{r},\vec{u},t) = \sum_m \int d^3\vec{r}_0 \int d^3\vec{u}_0 F_m(\vec{r}_0,\vec{u}_0;\vec{t}_0) G_{mj}(\vec{r},\vec{u},\vec{t};\vec{r}_0,\vec{u}_0,\vec{t}_0)
\]

(IIA.7)

Here \( t \geq t_0 \), such that

\[
G_{ij}(\vec{r},\vec{u},t_0;\vec{r}_0,\vec{u}_0,t_0) = \delta_{ij} \delta(\vec{r}-\vec{r}_0) \delta(\vec{u}-\vec{u}_0)
\]

(IIA.8)

Now, take the derivative of (IIA.7) with respect to \( t_0 \), and insert (IIA.6) for

\[
\frac{\partial}{\partial t_0} F_m(\vec{r}_0,\vec{u}_0,t_0) \text{ then yield}
\]

\[
0 = I_1 - I_2 - I_3 + I_4 + I_5
\]

(IIA.9)

Here \( I_1 = \sum_m \int d^3\vec{r}_0 \int d^3\vec{u}_0 F_m(\vec{r}_0,\vec{u}_0;\vec{t}_0) \frac{\partial}{\partial t_0} G_{mj}(\vec{r},\vec{u},\vec{t};\vec{r}_0,\vec{u}_0,\vec{t}_0) \)

\[
I_2 = \sum_m \int d^3\vec{r}_0 \int d^3\vec{u}_0 G_{mj}(\vec{r},\vec{u},\vec{t};\vec{r}_0,\vec{u}_0,\vec{t}_0) \vec{u}_0 \cdot \vec{\nabla}_{u_0} F_m(\vec{r}_0,\vec{u}_0;\vec{t}_0)
\]
Partial integration of $I_2$ with respect to $r_0$ yields

$$I_2 = \sum_m \int d^3 \tilde{r}_0 \int d^3 \tilde{u}_0 \mathcal{F}_m(\tilde{r}_0, \tilde{u}_0, t_0) \tilde{u}_0 \cdot \nabla \tilde{u}_0 \sum_m \alpha_k \int d^3 \tilde{u}' \int d^3 \tilde{u}''$$

Replacing integration variable in $I_{3,4}$ gives

$$I_3 = \sum_m \int d^3 \tilde{r}_0 \int d^3 \tilde{u}_0 \mathcal{F}_m(\tilde{r}_0, \tilde{u}_0, t_0) \sum_k \alpha_k \int d^3 \tilde{u}' \int d^3 \tilde{u}'' \sigma_{mk}(\tilde{u}_0; \tilde{u}', \tilde{u}'')$$

$$I_4 = \sum_m \int d^3 \tilde{r}_0 \int d^3 \tilde{u}_0 \mathcal{F}_m(\tilde{r}_0, \tilde{u}_0, t_0) \sum_k \alpha_k \int d^3 \tilde{u}' \int d^3 \tilde{u}'' \sigma_{mk}(\tilde{u}_0; \tilde{u}', \tilde{u}'')$$

Interchanging $\tilde{u}_0$ and $\tilde{u}''$, $k$ and $m$ yield

$$I_5 = \sum_m \int d^3 \tilde{r}_0 \int d^3 \tilde{u}_0 \mathcal{F}_m(\tilde{r}_0, \tilde{u}_0, t_0) \sum_k \alpha_k \int d^3 \tilde{u}' \int d^3 \tilde{u}'' \sigma_{mk}(\tilde{u}_0; \tilde{u}', \tilde{u}'')$$

Substituting these relations into (IIA.9), we obtained

$$\left( \frac{\partial}{\partial t_0} + \tilde{u}_0 \cdot \nabla_0 \right) G_{mj}(\tilde{r}, \tilde{u}, t; \tilde{r}_0, \tilde{u}_0, t_0) = \sum_k \alpha_k \int d^3 \tilde{u}' \int d^3 \tilde{u}'' \sigma_{mk}(\tilde{u}_0; \tilde{u}', \tilde{u}'') \left[ G_{mj}(\tilde{r}, \tilde{u}, t; \tilde{r}_0, \tilde{u}_0, t_0) - G_{mj}(\tilde{r}, \tilde{u}, t; \tilde{r}_0, \tilde{u}_0, t_0) - G_{kj}(\tilde{r}, \tilde{u}, t; \tilde{r}_0, \tilde{u}_0, t_0) \right]$$

(IIA.10)

Since $\mathcal{F}_m(\tilde{r}_0, \tilde{u}_0, t_0)$ are arbitrary distribution functions. Because of translational invariance in space and time, we have

$$G_{mj}(\tilde{r}, \tilde{u}, t; \tilde{r}_0, \tilde{u}_0, t_0) \equiv G_{mj}(\tilde{r} - \tilde{r}_0, \tilde{u}, \tilde{u}_0, t - t_0)$$
so that

\[
\left( \frac{\partial}{\partial t_0} + \bar{v}_0 \cdot \nabla r_0 \right) G_{mj} = - \left( \frac{\partial}{\partial t} + \bar{v}_0 \cdot \nabla r \right) G_{mj} \tag{IIA.11}
\]

Inserting (IIA.11) into (IIA.10), setting \( t_0 = 0, t_0 = 0 \) and interchanging \( \bar{v} \) and \( \bar{v}_0 \), we found

\[
- \left( \frac{\partial}{\partial t} + \bar{v} \cdot \nabla r \right) G_{mj}(\bar{r}, \bar{v}, \bar{v}_0, t) = N \nu \sum_k \alpha_k \int d^3 \bar{v}' \int d^3 \bar{v}'' \sigma_{mk}(\bar{v}_0; \bar{v}', \bar{v}'').
\]

\[
\left[ G_{mj}(\bar{r}, \bar{v}, \bar{v}_0, t) - G_{mj}(\bar{r}, \bar{v}', \bar{v}_0, t) - G_{kj}(\bar{r}, \bar{v}', \bar{v}_0, t) \right]
\]

\[
(\text{IIA.12})
\]

Here \( d\sigma_{mk}(\bar{v}; \bar{v}', \bar{v}'') = \sigma_{mk}(\bar{v}; \bar{v}', \bar{v}'')d^3 \bar{v}'d^3 \bar{v}'' \)

and (IIA.8) reduced to

\[
G_{ij}(\bar{r}, \bar{v}, \bar{v}_0, t = 0) = \delta_{ij} \delta(\bar{r}) \delta(\bar{v} - \bar{v}_0)
\]

\[
(\text{IIA.13})
\]

Therefore, the propagators \( G_{ij} \) satisfy the Backward Boltzmann equation (IIA.12).

For a collision cascade initiated by one \( i \) incident particle, we use \( F_{im}(\bar{r}_0, \bar{v}_0, t_0) \) to represent the distribution function of atom-m type, then, we have

\[
F_{im}(\bar{r}_0, \bar{v}_0, t_0 = 0) = \delta_{im} \delta(\bar{r}_0) \delta(\bar{v}_0 - \bar{v}_i)
\]

\[
(\text{IIA.14})
\]

Substituting (IIA.14) into (IIA.7), we obtained

\[
F_{ij}(\bar{r}, \bar{v}, t) = G_{ij}(\bar{r}, \bar{v}, t; 0, \bar{v}_1; 0) = G_{ij}(\bar{r}, \bar{v}_1, \bar{v}, t)
\]

and (IIA.6) turned out to be

\[
- \left( \frac{\partial}{\partial t} + \bar{v} \cdot \nabla r \right) F_{mj}(\bar{r}, \bar{v}, t) = N \sum_k \int d^3 \bar{v}' \int d^3 \bar{v}'' [\alpha_k \nu F_{mj} \sigma_{jk}(\bar{v}; \bar{v}', \bar{v}'')].
\]

\[
- \alpha_k \nu' F_{mj} \sigma_{jk}(\bar{v}; \bar{v}'', \bar{v}') - \alpha_k \nu'' F_{mk} \sigma_{kj}(\bar{v}'', \bar{v}', \bar{v})
\]

\[
(\text{IIA.15})
\]

Since the propagator \( G_{ij} \) are identical with the distribution function \( F_{ij} \), of course, they must also satisfy the Forward Boltzmann Equation (IIA.15).
CHAPTER 3
SOLUTIONS FOR SEPARABLE SCATTERING CROSS-SECTIONS

3.1 LAPLACE TRANSFORMS OF SOLUTIONS

For the following separable scattering cross sections

\[ d\sigma_{ij}(E, T) = Q_i(E) A_{ij}(X) dX \] (3.1.1)

where, \( 1 \geq X \equiv T/T_m \geq 0, T_m = \gamma E, \) and \( \gamma \equiv \gamma_{ij} \), following Sigmund method [16~20], let’s introduce the new variables \( E = U^u, \ T = U^v \) and take the Laplace Transform on both sides of Eqs. (2.3.2) with respect to \( u \), we obtained (see Appendix IIIA) a set of algebraic equations,

\[ \sum k \beta_{ik}^l(s)[x_{ik}^l(s)\tilde{G}_{ij}^l(s) - \tilde{G}_{kij}^l(s)] = \delta_{ij}/[N_0 E_0 Q_i(E_0)] \] (3.1.2)

then solving it exactly yields

\[ \tilde{G}_{ij}^l(s) = \frac{\Pi_{ji}^l(s)}{N_0 E_0 Q_j(E_0)} \] (3.1.3)

By using the same method, it’s easy to derive the Laplace Transforms of solutions of Eqs. (2.3.3,4), (2.3.7) and (2.3.9~12),

\[ \tilde{F}_{ij}^l(s) = E_0^{-1} \cdot \sum_k [D_{ij}^l(s)]_{kl} \Pi_{kl}^l(s) R_{kj}^l(s) \] (3.1.4)

\[ \tilde{F}_{ij}^l(s) = E_0^{-1} \cdot \Pi_{ji}^l(s) \sum_k S_{kj}^l(s) \] (3.1.5)

\[ \tilde{N}_i(s) = \frac{1}{sD(s)} \left\{ (2^s - 1) \int_0^{0.5} \frac{dx}{\sqrt{x}} \left[ A(x) + A(1 - x) \right] x^s \right. \\
\left. + \int_0^{0.5} \frac{dx}{\sqrt{x}} \left[ A(x) + A(1 - x) \right] (1 - x^s) \right\} \] (3.1.6)

\[ \tilde{F}_{(ij)}(s) = U \cdot \{ - \sum_k \Pi_{ki}^l(s) B_{kj}^l(s) + \Pi_{ji}^l(s) \sum_k [E_{jk}^l(s) + B_{jk}^l(s)] \} \] (3.1.7)

\[ \tilde{P}_{(ij)}(s) = \sqrt{2M_i U} \cdot \{ - \sum_k \Pi_{ki}^l(s) B_{kj}^l(s) \}
\left. + \Pi_{ji}^l(s) \sum_k [E_{jk}^l(s) + B_{jk}^l(s) + 0.5] \right\} \] (3.1.8)

\[ \tilde{N}_{ij}(s) = \sum_k \Pi_{ki}^l(s) [\beta_{kj}(0) - \beta_{kj}(s)] \] (3.1.9)
\[
\tilde{N}_y(s) = \prod_{\beta}(s) \sum_k \beta_{jk}(s) e_{jk}(s) \quad (3.1.10)
\]

Where \( \tilde{H}(s) = \int_0^\infty du \cdot \exp(-su)H(U_e^{a}, E_0) \) and \( H \) stands for one of \( G_{ij}^L \), \( F_{ij}^L \), \( F_{ij}^{al} \), \( N_{ij} \), \( F_{ij} \), \( P_{ij} \), \( N_{ij}^s \), \( D_{ij}^L(s) \) is the element \( (i, j) \) of the determinant \( D^L(s) \) and \([D^L(s)]_{ij}\)

is the algebraic cofactor for element \( (i, j) \).

\[
\begin{cases}
D_{ij}^L(s) = -\beta_{ij}^L(s), & \text{for } i \neq j \\
D_{ii}^L(s) = \sum_k \beta_{ik}^L(s)\left[e_{ik}^L(s) - \delta_{ik}\right]
\end{cases}
\]

\[
\Pi_{ij}^L(s) = [D^L(s)]_{ij}/[D^L(s)]
\]

\[
\beta_{ij}^L(s) = \alpha_j \gamma^{s} \int_0^\infty dX A_{ij}(X)X^s P_L(\sqrt{X})
\]

(3.1.11)

\[
e_{ij}^L(s) = \alpha_j \left[e_{ij}^L(s)\right]^{-1} \\
\int_0^1 dX A_{ij}(X)\left[1 - (1 - \gamma X)^s P_L\left[U_{ij}(1 - \gamma X)^{0.5} + V_{ij}(1 - \gamma X)^{-0.5}\right]\right]
\]

(3.1.12)

\[
R_{ij}^L(s) = \alpha_j \left(U/E_0\right)^\gamma \int_{\frac{E_n}{U}}^{\infty} dX A_{ij}(X)X^s P_L(\sqrt{X})
\]

(3.1.13)

\[
S_{ij}^L(s) = \alpha_j \left(U/E_0\right)^\gamma \\
\int_{\frac{E_n}{U}}^{\gamma-1} dX A_{ij}(X)(1 - \gamma X)^s P_L\left[U_{ij}(1 - \gamma X)^{0.5} + V_{ij}(1 - \gamma X)^{-0.5}\right]
\]

(3.1.14)

\[
B_{ij}^L(s) = [\beta_{ij}^L(s) - \beta_{ij}^L(1 - 0.5L)]/(s + 0.5L - 1), \quad \text{for } L = 0, 1
\]

\[
E_{ij}^L(s) = \beta_{ij}^L(1 - 0.5L)[e_{ij}^L(s) - (M_{ij}/M_i)^{0.5L}]/(s + 0.5L - 1), \quad \text{for } L = 0, 1
\]

In the freezing free case, \( U = E_o \), (3.1.13,14) reduced to

\[
R_{ij}^L(s) = \beta_{ij}^L(s) \quad \text{and} \quad S_{ij}^L(s) = \beta_{ij}(0) - \beta_{ij}^L(s)e_{ij}^L(s)
\]

(3.1.15)

Specially, we have

\[
e_{ij}(s) = s^{1-s}\left[\langle s | 1 \rangle_{ij} + \frac{1}{2!}(1-s)\gamma<s|X>_{ij} + \cdots
\]

\[
+ \frac{1}{n!}(1-s)(2-s)\cdots(n-1-s)\gamma^{n-1}\langle s | X^{n-1} >_{ij} + \cdots
\]
Here, \( \langle s | f(x) \rangle_{ij} = \left[ \int_0^s dx A_{ij}(x)x^s \right]_{ij}^{-1} \int_0^1 dx A_{ij}(x)f(x) \)

A simple calculation shows

\[
\varepsilon_{ij}(0) = 0 \\
\varepsilon_{ij}(1) = \langle 1 | 1 \rangle_{ij} = 1, \\
\varepsilon_{ij}^1(0.5) = (M_j / M_i)^{0.5} \\
R_{ij}(s + 0.5) = (\gamma U/E_0)^{0.5} \cdot R_{ij}(s + 0.5) \\
S_{ii}(s + 0.5) = (U/E_0)^{0.5} \cdot S_{ii}(s) \\
B_{ij}(s + 0.5) = \gamma^{0.5} \cdot B_{ij}(s) \\
E_{ii}(s + 0.5) = E_{ii}^1(s) \\
[D(I)]_{ji} = [D(1)]_{ji} \\
[D^1(0.5)]_{ji} = [D^1(0.5)]_{ji} \cdot (M_i / M_j)^{0.5} \\
D(s) = (s - 1) \sum_{jk} [D(s)]_{kj} E_{kj}(s) \quad (3.1.16) \\
D^1(s) = (s - 0.5) \sum_{jk} [D^1(s)]_{kj} E_{kj}^1(s)(M_k / M_j)^{0.5} \quad (3.1.17)
\]

The next step is to evaluate the inverse Laplace transform of \( \tilde{H}(s) \). The main problem is to find the poles and corresponding residues of \( \tilde{H}(s) \) [7].

### 3.2 EXPLAINING MONTE CARLO SIMULATIONS BY SIGMUND THEORY

In 1992, H. M. Urbassek and U. Conrad presented a wider variety of solutions of Eqs. (2.3.5)
in binary media ($\alpha_1 + \alpha_2 = 1$) for general power law cross sections ("UC" theory),

$$d\sigma_{ij}(E, T) = C_0 E^{-m_0} \cdot T^{-1-m_1} dT \quad \gamma_{ij} = 1$$

(3.2.1)

In addition, these equations were also solved numerically by means of a Monte Carlo simulation with $\gamma_{ij} = 1$. The authors of [23] demonstrated that "UC" theory agreed with the simulation results in the asymptotic regime of small recoil energies. Seeing that the general power law cross sections (3.2.1) couldn’t fit our separable scattering cross sections (3.1.1), thus, Sigmund method [17] couldn’t be used. In 1999, the present author uses conventional power cross sections

$$d\sigma_{ij}(E, T) = C_0 E^{-m_0} \cdot T^{-1-m_1} dT \quad \gamma_{ij} = 1$$

(3.2.2)

to approximate (3.2.1) by choosing a proper $m_1$, then, Sigmund method can be used. The energy $E_0$ is measured relative to the bombarding energy $E$, i.e. $x = E_0 / E$, and distribution functions are normalized as

$$\psi_{ij}(x) = N C_{11} E^{(1-m_{ij})} \cdot \psi_{ij}(E, E_0),$$

(3.2.3)

$$f_{ij}(x) = E F_{ij}(E, E_0)$$

(3.2.4)

to make them dimensionless. The values of the coefficients in cross sections (3.2.1,2) are given in a normalized way as

$$\tilde{C}_{ij} = \frac{\alpha_j C_{ij}}{\alpha_i C_{11}} \cdot E^{2(m_{ij} - m_0)}$$

With the help of the above considerations, the inverse Laplace transforms of (3.1.3,4) are

$$\begin{align*}
x\psi_{11}(x) &= x^{2m_1} L^{-1}\left[(s-m_1)[((\alpha_1 \tilde{C}_{21} + \alpha_2 \tilde{C}_{22}) v_{22}(s) - \alpha_2 \tilde{C}_{22}]/d(s)]\right] \\
x\psi_{12}(x) &= x^{2m_1} \alpha_2 \tilde{C}_{12} L^{-1}\left[(s-m_2)/d(s)\right] \\
x\psi_{21}(x) &= x^{2m_1} \alpha_1 \tilde{C}_{21} L^{-1}\left[(s-m_1)/d(s)\right] \\
x\psi_{22}(x) &= x^{2m_1} L^{-1}\left[(s-m_2)[((\alpha_1 \tilde{C}_{11} + \alpha_2 \tilde{C}_{12}) v_{11}(s) - \alpha_1 \tilde{C}_{11}]/d(s)]\right]
\end{align*}$$

(3.2.5)
\[
\begin{align*}
\int x^2 f_1(x) &= xL^{-1}\left\{a_1\tilde{C}_{11}(\alpha_1\tilde{C}_{21} + \alpha_2\tilde{C}_{22})\varepsilon_{22}(s) - d(0)\right\}/d(s) \\
\int x^2 f_2(x) &= xL^{-1}\left\{a_2\tilde{C}_{12}(\alpha_1\tilde{C}_{21} + \alpha_2\tilde{C}_{22})\varepsilon_{22}(s)\right\}/d(s) \\
\int x^2 f_3(x) &= xL^{-1}\left\{a_2\tilde{C}_{21}(\alpha_1\tilde{C}_{11} + \alpha_2\tilde{C}_{12})\varepsilon_{11}(s)\right\}/d(s) \\
\int x^2 f_4(x) &= xL^{-1}\left\{a_2\tilde{C}_{22}(\alpha_1\tilde{C}_{11} + \alpha_2\tilde{C}_{12})\varepsilon_{11}(s) - d(0)\right\}/d(s)
\end{align*}
\]

(3.2.6)

Here \(d(s) = [(\alpha_1\tilde{C}_{11} + \alpha_2\tilde{C}_{12})\varepsilon_{11}(s) - \alpha_2\tilde{C}_{11}] \cdot [(\alpha_1\tilde{C}_{21} + \alpha_2\tilde{C}_{22})\varepsilon_{22}(s) - \alpha_2\tilde{C}_{22}] \]

\[-\alpha_2\alpha_2\tilde{C}_{12} - \tilde{C}_{21} \]

(3.2.7)

\[
\varepsilon_{ii}(s) = 1 + \frac{s}{m_i} \frac{\Gamma(1 - m_i)}{\Gamma(s - m_i)} \frac{\Gamma(s)}{\Gamma(1 - m_i)}
\]

\[
= s(s - m_i) \left[ \frac{1}{1!(1 - m_i)} + \frac{(1 - s)}{2!(2 - m_i)} + \ldots + \frac{(1 - s)(2 - s)(n - 1 - s)}{n!(n - m_i)} + \ldots \right]
\]

Specially, we have

\[
d(0) = \alpha_1\alpha_2(\tilde{C}_{11}\tilde{C}_{22} - \tilde{C}_{12}\tilde{C}_{21})
\]

\[
d(1) = 0
\]

(3.2.8)

\[
d'(l) = \alpha_1\tilde{C}_{21}(\alpha_1\tilde{C}_{11} + \alpha_2\tilde{C}_{12})[\varepsilon_{11}(1)]' + \alpha_2\tilde{C}_{12}(\alpha_1\tilde{C}_{21} + \alpha_2\tilde{C}_{22})[\varepsilon_{22}(1)]'
\]

\[
d'(0) = -\alpha_2\tilde{C}_{22}(\alpha_1\tilde{C}_{11} + \alpha_2\tilde{C}_{12})[\varepsilon_{11}(0)]' - \alpha_1\tilde{C}_{11}(\alpha_1\tilde{C}_{21} + \alpha_2\tilde{C}_{22})[\varepsilon_{22}(0)]'
\]

\[
[\varepsilon_{ii}(1)]' = [\Gamma_m]^{-1}, \quad \Gamma_m = m/[(\psi(l) - \psi(1 - m)].
\]

\[
[\varepsilon_{ii}(0)]' = -m \left[ \frac{1}{1!(1 - m_i)} + \frac{1}{2!(2 - m_i)} + \ldots + \frac{1}{n!(n - m_i)} + \ldots \right]
\]

Some typical values of \(\Gamma_m\) have been calculated and collected in table (3.2.1).

Obviously, (3.2.8) provided the highest pole \(s = 1\), and gave the leading terms of (3.2.5,6),

\[
\begin{align*}
\int x\psi_{11}(x) &= x\psi_{21}(x) = 2\Lambda(1 - m_i)\alpha_1\tilde{C}_{21} \cdot x^{2m_i - 2} \equiv x\psi_1(x) \\
\int x\psi_{12}(x) &= x\psi_{22}(x) = 2\Lambda(1 - m_2)\alpha_2\tilde{C}_{12} \cdot x^{2m_2 - 2} \equiv x\psi_2(x)
\end{align*}
\]

(3.2.9)

**Table 3.2.1** Some typical values of \(\Gamma_m\)

<table>
<thead>
<tr>
<th>(m)</th>
<th>0.2</th>
<th>0.24</th>
<th>0.30</th>
<th>0.40</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\Gamma_m)</td>
<td>0.51574</td>
<td>0.49693</td>
<td>0.46670</td>
<td>0.41519</td>
</tr>
</tbody>
</table>
\[
\begin{align*}
\left\{\begin{array}{l}
x^2 f_{11}(x) &= x^2 f_{21}(x) = \Lambda \alpha \tilde{C}_{21} \cdot (\alpha \tilde{C}_{11} + \alpha \tilde{C}_{12}) \\
x^2 f_{12}(x) &= x^2 f_{22}(x) = \Lambda \alpha \tilde{C}_{22} \cdot (\alpha \tilde{C}_{21} + \alpha \tilde{C}_{22})
\end{array}\right.
\end{align*}
\] (3.2.10)

where \( \Lambda = \frac{\Gamma_{m_1} \Gamma_{m_2}}{\alpha \tilde{C}_{21} (\alpha \tilde{C}_{11} + \alpha \tilde{C}_{12}) \Gamma_{m_2} + \alpha \tilde{C}_{12} (\alpha \tilde{C}_{21} + \alpha \tilde{C}_{22}) \Gamma_{m_1}} \)

All simulations in [23] have been performed for \( \alpha_1 = \alpha_2 = 0.5 \), thus, (3.2.5~7) further reduced to

\[
\begin{align*}
\left\{\begin{array}{l}
x \psi_{11}(x) &= 0.5x^{2m_1} L^{-1} \left[ (s - m_1) \left( (\tilde{C}_{21} + \tilde{C}_{22}) e_{22} (s) - \tilde{C}_{22} \right) / d(s) \right] \\
x \psi_{12}(x) &= 0.5x^{2m_2} L^{-1} \left[ (s - m_2) / d(s) \right] \\
x \psi_{21}(x) &= 0.5x^{2m_1} L^{-1} \left[ (s - m_1) / d(s) \right] \\
x \psi_{22}(x) &= 0.5x^{2m_2} L^{-1} \left[ (s - m_2) \left( (\tilde{C}_{11} + \tilde{C}_{12}) e_{11} (s) - \tilde{C}_{11} \right) / d(s) \right]
\end{array}\right.
\end{align*}
\] (3.2.11)

\[
\begin{align*}
\left\{\begin{array}{l}
x^2 f_{11}(x) &= 0.25x L^{-1} \left[ (\tilde{C}_{11} + \tilde{C}_{22}) e_{22} (s) - \tilde{C}_{11} \tilde{C}_{22} + \tilde{C}_{12} \tilde{C}_{21} \right] / d(s) \\
x^2 f_{12}(x) &= 0.25x L^{-1} \left[ (\tilde{C}_{11} + \tilde{C}_{22}) e_{22} (s) / d(s) \right] \\
x^2 f_{21}(x) &= 0.25x L^{-1} \left[ (\tilde{C}_{11} + \tilde{C}_{12}) e_{11} (s) / d(s) \right] \\
x^2 f_{22}(x) &= 0.25x L^{-1} \left[ (\tilde{C}_{22} (\tilde{C}_{11} + \tilde{C}_{12}) e_{11} (s) - \tilde{C}_{11} \tilde{C}_{22} + \tilde{C}_{12} \tilde{C}_{21} \right] / d(s) \right]
\end{array}\right.
\end{align*}
\] (3.2.12)

\[
4d(s) = [(\tilde{C}_{11} + \tilde{C}_{12}) e_{11} (s) - \tilde{C}_{11}] \cdot [(\tilde{C}_{21} + \tilde{C}_{22}) e_{22} (s) - \tilde{C}_{22}] - \tilde{C}_{12} \cdot \tilde{C}_{21}
\] (3.2.13)

The subsequent terms stem from other poles excepts \( s = 1 \). It may be sufficient to analyze the possibility of poles occurring between zero and one for several special cases simulated by Urbassek and Conrad [23].

### 3.2.1 “DETAILED BALANCE”

Parameters in (3.2.1) for Monte Carlo simulations in [23] are

\[
\begin{align*}
m_{11} &= 0.30 \quad m_{12} = 0.40 \quad m_{21} = 0.24 \quad m_{22} = 0.20 \\
\tilde{C}_{11} &= 1 \quad \tilde{C}_{12} = 100 \quad \tilde{C}_{21} = 100 \quad \tilde{C}_{22} = 1
\end{align*}
\] (3.2.14)

Since \( \tilde{C}_{12} = \tilde{C}_{21} >> \tilde{C}_{11} = \tilde{C}_{22} \), the most frequent collision in the cascade takes place between atom-1 and atom-2. The energy distributions of the two species are closely coupled together and a situation of “detailed balance” is established [23]. Therefore, it is natural to choose the following parameters
\[ m_1 = 0.40 \quad \text{and} \quad m_2 = 0.24 \]  

(3.2.15)

for (3.2.2). Then (3.2.13) reads

\[ 4d(s) = 10l[\varepsilon_{11}(s)\varepsilon_{22}(s) - \varepsilon_{11}(s) - \varepsilon_{22}(s) - 99] \]  

(3.2.16)

and has been evaluated and plotted in part (A) of Fig. (3.2.1) as a function of \( s \), for \( l \geq s \geq 0 \), see curve “i”. Except \( s = 1 \), obviously, no extra zero of \( d(s) \) can be found. Thus, (3.2.9,10) themselves turn out to be the best solutions, i.e.

\[
\begin{align*}
\psi_1(x) &= \psi_{11}(x) = \psi_{21}(x) = 2.6862 \cdot 10^{-3} \cdot x^{-0.20} \\
\psi_2(x) &= \psi_{12}(x) = \psi_{22}(x) = 3.4025 \cdot 10^{-3} \cdot x^{-0.52} \\
x^2f_1(x) &= x^2f_{11}(x) = x^2f_{21}(x) = 0.2261 \\
x^2f_2(x) &= x^2f_{12}(x) = x^2f_{22}(x) = 0.2261
\end{align*}
\]  

(3.2.17)

(3.2.18)

They are plotted in part (A) of Fig. (3.2.2) and Fig. (3.2.3) respectively. One can see that (3.2.17,18) reproduce the Monte Carlo simulation results very precisely for \( E_0 < 0.02E \).

\[ \begin{array}{c}
\text{Fig. 3.2.1} \text{ Calculated results of (3.2.16), as a function of } s. \text{ (A) Curve “i” stands for “Detailed balance”. Curve “iv” stands for “Ignorance & Detailed balance”. No any extra zeros can be found. (B) “Dominance”, two extra zeros can be found. (C) “Ignorance”, one extra zero can be found.}
\end{array} \]
Fig. 3.2.2 Normalized energy distributions. Thick: Monte Carlo Simulation results are copied from [23]; Thin: Sigmund analytical theory (3.2.9). (A) “Detailed balance”, calculated by (3.2.17); (B) “Dominance”, calculated by (3.2.21); (C) “Ignorance”, calculated by (3.2.25); (D) “Ignorance & Detailed balance”, calculated by (3.2.29).

Fig. 3.2.3 Normalized recoil densities. Thick: Monte Carlo Simulation results are copied from [23]; Thin: Sigmund analytical theory (3.2.10). (A) “Detailed balance”, calculated by (3.2.18); (B) “Dominance”, calculated by (3.2.22); (C) “Ignorance”, calculated by (3.2.26).
3.2.2 “DOMINANCE”

Parameters in (3.2.1) for Monte Carlo simulations in [23] are

\[
\begin{align*}
m_{11} &= 0.30, & m_{12} &= 0.40, & m_{21} &= 0.21, & m_{22} &= 0.24, \\
\tilde{C}_{11} &= 1, & \tilde{C}_{12} &= 100, & \tilde{C}_{21} &= 1, & \tilde{C}_{22} &= 100.
\end{align*}
\]  

(3.2.19)

Since \( \tilde{C}_{12} = \tilde{C}_{22} >> \tilde{C}_{11} = \tilde{C}_{21} \), the most frequent collision is that atom-1 strikes atom-2 and atom-2 strikes atom-2. The species 2 behaves as in a monatomic medium, while the energy distribution of species 1 is determined by the one of species 2 in a complicated way. This case is called the “dominance” of species 2 over species 1 [23]. Therefore, it is natural to choose parameters (3.2.15) for (3.2.2). Then (3.2.13) reads

\[
4d(s) = 10\left[\varepsilon_{s1} - 100\varepsilon_{s1} - \varepsilon_{s2} - \varepsilon_{s2}(s)\right]
\]  

(3.2.20)

and has been evaluated and plotted in part (B) of Fig. (3.2.1) as a function of \( s \), for \( 1 \geq s \geq 0 \). Except \( s = 1 \), two extra zeros of \( d(s) \) can be found, \( s_0 = 0.3986 \) and \( s_1 = 0 \). Taking account of contributions of \( s = 1, s_0 \) and \( s_1 \), (3.2.11,12) turn out to be

\[
\begin{align*}
x\psi_{11}(x) &= x\psi_1(x) \cdot \left(1 - 0.6068 \cdot x^\alpha + 140.371 \cdot x\right) \\
x\psi_{12}(x) &= x\psi_2(x) \cdot \left(1 - 0.5889 \cdot x^\alpha - 0.6649 \cdot x\right) \\
x\psi_{21}(x) &= x\psi_{11}(x) \cdot \left(1 + 6.7748 \cdot 10^{-3} \cdot x^\alpha - 1.4037 \cdot x\right) \\
x\psi_{22}(x) &= x\psi_{22}(x) \cdot \left(1 + 6.5548 \cdot 10^{-3} \cdot x^\alpha + 6.6472 \cdot 10^{-3} \cdot x\right)
\end{align*}
\]  

(3.2.21)

\[
\begin{align*}
x^2f_{11}(x) &= x^2f_1(x) \cdot \left(1 - 0.2906 \cdot x^\alpha\right) \\
x^2f_{12}(x) &= 100x^2f_{11}(x) \\
x^2f_{21}(x) &= f_2(x) \cdot \left(1 + 3.2552 \cdot 10^{-3} \cdot x^\alpha\right) \\
x^2f_{22}(x) &= 100x^2f_{21}(x)
\end{align*}
\]  

(3.2.22)

Here \( \alpha \equiv 1 - s_0 = 0.6014; \)

\[
\begin{align*}
x\psi_1(x) &= 5.8281 \cdot 10^{-5} \cdot x^{0.20} \\
x\psi_2(x) &= 7.3822 \cdot 10^{-3} \cdot x^{0.52}
\end{align*}
\]  

(3.2.22 a)

(3.2.21, 22) are calculated and plotted in part (B) of Fig. (3.2.2) and Fig. (3.2.3) respectively. One
can see that $\psi_{11}(x)$, $\psi_{12}(x)$, $\psi_{22}(x)$, $f_{12}(x)$ and $f_{22}(x)$ reproduce the Monte Carlo simulation results very precisely. $\psi_{21}(x)$, $f_{11}(x)$ and $f_{11}(x)$ agree with the simulation results reasonably well. Particularly, for $x\psi_{11}(x)$, the large deviation from the corresponding asymptotic solution

$$x\psi_1(x) = 5.8281 \cdot 10^{-5} \cdot x^{-0.2}$$

is contributed by the third highest pole $s_1 = 0$, i.e. the third term in the bracket of the first relation of (3.2.21). However, the pole makes no contribution to $xf_{ij}(x)$ at all.

### 3.2.3 “IGNORANCE”

Parameters in (3.2.1) for Monte Carlo simulations in [23] are

$$
\begin{align*}
\tilde{C}_{11} &= 1 \\
\tilde{C}_{12} &= 0.01 \\
\tilde{C}_{21} &= 0.01 \\
\tilde{C}_{22} &= 1
\end{align*}
$$

(3.2.23)

Since $\tilde{C}_{11} \gg \tilde{C}_{22} \gg \tilde{C}_{21}$, the most frequent collision is that atom-1 strikes atom-1 and atom-2 strikes atom-2, each species shows the same energy dependence as in a monatomic medium. Therefore, this case is called “ignorance”. Therefore, it is natural to choose parameters (3.2.15) for (3.2.2). Then (3.2.13) reads

$$4d(s) = 1.0 \left[ 1.01c_{11}(s)c_{22}(s) - 100c_{11}(s) - c_{22}(s) + 0.99 \right]$$

(3.2.24)

and has been evaluated and plotted in part (C) of Fig. (3.2.1) as a function of $s$, for $1 \geq s \geq 0$. Except $s = 1$, one extra zero of $d(s)$ can be found, $s_0 = 0.9909515$. The curve showed in large circle is enlargement part of $d(s)$ near $s = 1$. Taking account of contributions of $s = 1$ and $s_0$, (3.1.16,17) turn out to be

$$
\begin{align*}
x\psi_{11}(x) &= x\psi_1(x) \cdot (1 + 0.8306 \cdot x^\alpha) \\
x\psi_{12}(x) &= x\psi_2(x) \cdot (1 - 0.99554 \cdot x^\alpha) \\
x\psi_{21}(x) &= x\psi_1(x) \cdot (1 - 0.99234 \cdot x^\alpha) \\
x\psi_{22}(x) &= x\psi_2(x) \cdot (1 + 1.1893 \cdot x^\alpha)
\end{align*}
$$

(3.2.25)
Here $\alpha \equiv 1 - s_0 = 0.0090585$

\[
\begin{align*}
\alpha &= 1 - s_0 = 0.0090585 \\
\psi_1(x) &= 0.2686 \cdot x^{-0.2} \\
\psi_2(x) &= 0.3402 \cdot x^{-0.52} \\
x^2f_1(x) &= x^2f_2(2) = 0.2261.
\end{align*}
\]

(3.2.25,26) are calculated and plotted in part (C) of Fig. (3.2.2) and Fig. (3.2.3) respectively. One can see that $\psi_{11}(x), \psi_{22}(x), \psi_{21}(x)$, $f_{11}(x)$ and $f_{22}(x)$ reproduce the Monte Carlo simulation results accurately, and $\psi_{12}(x), f_{12}(x)$ and $f_{21}(x)$ agree with the simulation results reasonably well. Because, second highest pole $s_0 = 0.9909581 \approx 1$, very close to the main pole $s = 1$, it’s very easy to be ignored. In the energy window concerned, the first and second highest poles provide almost identical contributions in magnitude, thus neither of them can be neglected. This was the reason why Urbassek et.al couldn’t give the analytical solutions, but still could determine the energy distributions in this regime [23].

3.2.4 “IGNORANCE & DETAILED BALANCE ”

Parameters in (3.2.1) for Monte Carlo simulations in [23] are

\[
\begin{align*}
m_{11} &= 0.40 & m_{12} &= 0.30 & m_{21} &= 0.20 & m_{22} &= 0.24 \\
C_{11} &= 1 & C_{12} &= 100 & C_{21} &= 100 & C_{22} &= 1
\end{align*}
\]

Due to the same reason as in “Detailed balance”, it is natural to choose

\[
m_1 = 0.30 \quad \text{and} \quad m_2 = 0.24
\]

for (3.2.2). Then (3.2.13) reduced to (3.2.16) again and has been evaluated and plotted in part (A) of Fig. (3.2.1) as a function of $s$, for $1 \geq s \geq 0$, see curve “iv”. Except $s = 1$, obviously, no extra zero of
\( d(s) \) can be found. Thus, \((3.2.9,10)\) themselves turn out to be the best solutions, i.e.

\[
\begin{align*}
\psi_1(x) &= x\psi_{11}(x) = x\psi_{21}(x) = 3.396 \cdot 10^{-3} \cdot x^{-0.4} \\
\psi_2(x) &= x\psi_{12}(x) = x\psi_{22}(x) = 3.881 \cdot 10^{-3} \cdot x^{-0.6}
\end{align*}
\]

(3.2.29)

which has been plotted in part (D) of Fig. (3.2.2). One can see that (3.2.29) reproduce the Monte Carlo simulation results very precisely for \( E_0 < 0.02E \).

In conclusion, on the one hand, the Monto Carlo simulations, which had been performed by H. M. Urbassek et [23], provided a very strong support for Sigmund theory. On the other hand, Sigmund theory gave the all of these simulation results [23] a best explanation.

### 3.3 Statistical Distribution Functions

#### 3.3.1 Asymptotic Solutions

The excellent agreement between Sigmund theory and the Monte Carlo simulation results in [23] is full proof that each inverse Laplace transform of \((3.1.3-10)\) must be the exact solution of corresponding integral equation. Unfortunately, it has not been possible, so far, to evaluate the inverse Laplace transform in closed form for general case. But, in the most of applications, we are looking for solutions in the energy region \( E >> U \), i.e. large values of \( u \). The procedure of determining asymptotic solutions is a generalization of the one described in detail in [16]. The main problem is to find the highest single pole of \((3.1.3-10)\). The poles may occur at the zeros of the determinant \( D^1(s) \). From \((3.1.16,17)\), Sigmund gave a beautiful proof [16] which showed that \( D(s) \) has a highest zero at \( s = 1 \). In Appendix III B, we provided a simular proof which showed that \( D^1(s) \) has a highest zero at \( s = 0.5 \). Therefore, from \((3.1.16,17)\), these two highest poles generate the following asymptotic solutions of Eqs. \((2.3.3,4), (2.3.7)\) and \((2.3.9-12)\) respectively [11][19][24][25]:

For \( L = 0 \)
\[ \begin{align*}
\text{For } L = 1: \\
G_i(E, E_0) &= \frac{\Pi_j}{N_0 q E_0 Q_j(E_0)} \cdot \frac{E}{E_0} = \frac{\Gamma_j}{v_0 [dE_0 / dx]_{ij}} \cdot \frac{G_j}{E_0} \\
F_i(E, E_0, U) &= E (UE_0)^{-1} \cdot \sum_k \Pi_k R_{kj}(1) = F_j(E, E_0, U) \\
F_i^*(E, E_0, U) &= E (UE_0)^{-1} \cdot \sum_k \Pi_j S_{jk}(1) = F_j^*(E, E_0, U) \\
N_j(E) &= \xi_j E / U, \\
F_{ij}(E, U) &= E \left\{ - \sum_k \Pi_k B_{kj}(1) + \Pi_j \sum_k \left[ E_{jk}(1) + B_{jk}(1) \right] \right\} = F_{ij}(U) \\
N_j^*(E, U) &= EU^{-1} \cdot \sum_k \Pi_j \left[ \beta_{kj}(0) - \beta_{kj}(1) \right] = N_j(U) \\
N_j^*(E, U) &= EU^{-1} \cdot \sum_k \beta_{jk}(1) = N_j^*(1)(U) \\
\end{align*} \]

Here \([D(1)]' = [d[D(s)]/ds]_{s=1} = \sum_k [D(1)]_{kk} \sum_j E_{kj}(1)\]

\[ [D(1)]_{jj} = [D(1)]_{jj} \]

\[ \Pi_j = [D(1)]_{jj} / [D(1)]' \]

\[ [\varepsilon_j(1)]' = [d\varepsilon_j(s)/ds]_{s=1} = 1 - \ln \gamma - \left\{ \ln x \right\}_{ij} - \sum_{n=1}^{\infty} \left( \frac{n}{n+1} \right) \left\{ \ln x_n \right\}_{ij} \]

\[ B_j(1) = [d\beta_j(s)/ds]_{s=1} = [\beta_j(1)]' \]

\[ E_j(1) = \beta_j(1)[\varepsilon_j(1)]' \]

\[ [D^1(0.5)]' = \{d[D^1(s)]/ds\}_{s=0.5} = \sum_k [D^1(0.5)]_{kk} \sum_j E_{kj}(0.5) \]
\[ [D^l(0.5)]_{ij} = [D^l(0.5)]_{ij} \cdot \sqrt{M_i/M_j} \]

\[ \Pi^l_j = [D^l(0.5)]_{ij}/[D^l(0.5)]' \]

\[ [\varepsilon^l_{ij}(0.5)]' = [d\varepsilon^l_{ij}(s)/ds]_{s=0.5} \]

\[ = \sqrt{\gamma} \left\{ 1 - U_{ij} \left[ \ln \gamma + \langle 1 | \ln x \rangle_{ij} \right] + \sum_{n=1}^{\infty} \gamma^n \left[ V_{ij} - \frac{1}{n+1} \langle 1 | x^n \rangle_{ij} \right] \right\} \]

\[ = \sqrt{\gamma} - \frac{M_j}{M_i} \left\{ \ln \gamma + \langle 1 | \ln x \rangle_{ij} + \sum_{n=1}^{\infty} \frac{\gamma^n}{(n+1)} \left[ V_{ij} - \frac{1}{n+1} \frac{M_i - M_j}{M_i + M_j} \langle 1 | x^n \rangle_{ij} \right] \right\} \]

\[ B^l_{ij}(0.5) = [d\beta^l_{ij}(s)/ds]_{s=0.5} \equiv [\beta^l_{ij}(0.5)]' \]

\[ E^l_{ij}(0.5) = \beta^l_{ij}(0.5) [\varepsilon^l_{ij}(0.5)]' \]

\[ \varepsilon_{ii} = \varepsilon^l_{ii} = 1 - \langle 1 | \ln x \rangle_{ii} - \sum_{n=1}^{\infty} \frac{1}{n(n+1)} \langle 1 | x^n \rangle_{ii} \equiv 1/\Gamma_i \]

\[ \tilde{P}_i = \tilde{n} \cdot \sqrt{2M_iE} ; \quad \tilde{P}_{ij} = \tilde{n}_0 \cdot \sqrt{2M_jE_0} ; \]

\[ \xi_F = 2\Gamma \left[ \int_0^1 dX A(X) \right]^{-1} \int_0^{0.5} dX [A(X) + A(1 - X)] \]

\[ G_j/(G) = \beta_{ij}(1)\Pi^l_j/(\alpha_j \Gamma_j) \quad \text{and} \quad H_j/(H_j) = \beta_{ij}(1)\Pi^l_j/(\alpha_j \Gamma_j) \quad \text{are nonstoichiometric factors appeared in literatures [7].} \]

### 3.3.2 ANISOTROPIC DIFFERENTIAL PARTICLE FLUX

Inserting (3.3.1) and (3.3.8) into (2.3.1), only the \( L=0,1 \) terms are considered. We obtain

\[ G_j(0,0) = K_j \cdot \left[ \frac{E}{E_0} + 3A_j \frac{\tilde{n}_0 \cdot \tilde{P}_i}{P_{ij}} \right] \quad \text{(3.3.12)} \]

where

\[ K_j = \frac{\Pi^l_j}{N\tilde{n}_0 E_0 Q_j(E_0)} , \quad A_j = \frac{\Pi^l_j}{\Pi_j} \]

Particularly, for a binary medium (3.3.12) reads,
\[
G_{i1}(\tilde{\nu}, \tilde{\nu}_0) = K_1 \cdot \left[ \frac{E}{E_0} + 3 \cdot \frac{\bar{n}_0 \cdot \bar{P}_i}{\sqrt{2M_1E_0}} \cdot A \right], \quad i = 1, 2
\] (3.3.13)

Here \( D = \beta_{21}(1)[E_{11}(1) + E_{12}(1)] + \beta_{12}(1)[E_{21}(1) + E_{22}(1)] \)

\[
D' = U_{21}\beta_{21}(1)[E_{11}(1) + E_{12}(0.5)] + U_{12}\beta_{12}(1)[E_{21}(0.5) + E_{21}(1)]
\]

\[
A = \frac{(M_1 + M_2)}{2\sqrt{M_1M_2}} \cdot \frac{D}{D'}
\]

\[
K_1 = \frac{1}{N\nu_0E_0Q_1(E_0)} \cdot \frac{\beta_{21}(1)}{D}
\]

and

\[
K_2 = \frac{1}{N\nu_0E_0Q_2(E_0)} \cdot \frac{\beta_{12}(1)}{D}
\]

(3.3.13) was first derived by Sigmund et al. for a binary medium with nearly equal masses \((M_1 \approx M_2)\)[17]. Furthermore, replacing \(E\) and \(\bar{P}_i\) by deposited energy and momentum spatial distribution functions respectively (3.3.13) turns out to be the form which was given by Sigmund et al. in 1996 [11].

### 3.3.3 SOME IMPORTANT IDENTITIES

From (3.3.5,11), one can see that the asymptotic energy and momentum are independent of \(U\), then, substituting (3.3.5,11) into (2.3.17,18) respectively, we obtained

\[
\sum_k [\psi_j(U)\alpha_kS_{jk}(U) - \psi_k(U)\alpha_jS_{kj}(U)] = 0, \quad \text{for} \ E \gg U
\] (3.3.14)

\[
\sum_k [\psi_j^{1}(U)\alpha_kT_{jk}(U) - \psi_k^{1}(U)\alpha_jT_{kj}(U)] = 0, \quad \text{for} \ E \gg U
\] (3.3.15)

Directing summation of (3.3.14) or (3.3.15) over \(j\) only yields \(0 = 0\). Therefore, the identities (3.3.14) or (3.3.15) couldn’t be considered as a set of equations for the particle fluxes \(\psi_j(U)\) or \(\psi_j^{1}(U)\), because, these equations are not independent.
In addition, direct summation of (3.3.5,11) yielded

\[ F_{ij}(E, U) = \sum_j F_{ij}(E, U) = E \quad (3.3.16) \]
\[ \bar{P}_{ij}(E, U) = \sum_j \bar{P}_{ij}(E, U) = \bar{P}_i \quad (3.3.17) \]

It’s similar to (2.2.15,16) that even though (3.3.5,11) are asymptotic energy & momentum partitions \((E >> U)\), must also satisfy energy & momentum conservation laws respectively.

In freezing free case [16][17], substituting (3.1.15) into (3.3.2, 3), we have

\[ \sum \beta \Pi \cdot = -k_{ij}^{00} j^1 \]
\[ \sum \beta \Pi \cdot = -k_{ij}^{00} j \]

Substituting (3.3.1) into (3.3.18,19), we obtain

\[ \sum \sum \psi = \beta \Pi \cdot = -k_{ij}^{00} j^1 \]
\[ \sum \sum \psi = \beta \Pi \cdot = -k_{ij}^{00} j \]

Here \( L_j^t (E_o) = [NQ_j (E_o) \sum_k \beta_{jk} (0)]^{-1} \) is the mean free path of j-atom with energy \( E_o \) and \( L_j (E_o) = [NQ_j (E_o) \sum_k \beta_{jk} (1)]^{-1} \) is the mean slowing down distance with energy \( E_o \).

### 3.3.4 ATOM EJECTION PROCESS

For a single element target, (3.3.1) and (3.3.18-21) reduced to

\[ \psi (E_o) = \Gamma L(E_o) E E_o^{-2} \quad ; \quad (3.3.22) \]
\[ F(E_o) = \Gamma E E_o^{-2} \quad ; \quad (3.3.23) \]
\[ \psi (E_o) = L(E_o) F(E_o) = L^t(E_o) F^t(E_o) \quad ; \quad (3.3.24) \]

From (3.3.20,21), one can see that (3.3.24) is tenable only for a single element medium. In order to explain (3.3.24) in relation to the average depth of origin of sputtered atom [28~31], lets derive the
atom flux produced by a homogeneous and isotropic atom source \( f(E_0) \) in an infinite uniform medium. First of all, we knew that the atom flux must be homogeneous and isotropic, due to a simple symmetry consideration. Thus, it can be treated as a one dimensional problem (x-axis). To determine the energy spectrum \( \psi(E_0) \) of the atom flux, Falcone and Sigmund have proposed two different theories.

i) Falcone-Sigmund theory

The basic assumption of Falcone-Sigmund theory is that each flux atom must come from some atom sources, and the source atom slows down continuously along a straight line to contribute the atom flux\[18][32][33]. Thus, all particles in the atom flux \( \psi(E_0) \) at the origin O of the x-axis must slow down from a uniformly distributed atom source \( f(E) \) on the positive (or negative) side of the x-axis, i.e. \( x \in (0, \infty) \). The energy loss of each atom is

\[
\frac{dE}{dx} = -NS_n(E) \quad \text{or} \quad x = \int_{E_0}^{E} \frac{dE}{[NS_n(E)]} \tag{3.3.25}
\]

which defines a function \( E = E(E_0, x) \). A direct differentiation of (3.3.25) yields

\[\frac{dx}{(dE)_{E_0}} = \frac{[NS_n(E)]}{dE_{E_0}} \quad \text{and} \quad \frac{(dE)_x}{S_n(E)} = \frac{dE_0}{S_n(E_0)} = NL(E_0)E_0^{-1}dE_0.\]

Then we have,

\[\frac{(dE)_x}{S_n(E)} \cdot dx = L(E_0)E_0^{-1}dE_0(dE)_{E_0} \tag{3.3.26}\]

where \( (dH)_w \) represents the differential of H, with W remaining constant [6]. The energy spectrum of the atom flux at the origin O is given by

\[\psi(E_0)dE_0 = \int_{E_0}^{\infty} f(E)(dE)_x \ dx \tag{3.3.27}\]

Inserting (3.3.26) into (3.3.27) yields

\[\psi(E_0) = L(E_0)F(E_0) \tag{3.3.28}\]

Here \( F(E_0) = \left( L/E_0 \right) \int_{E_0}^{\infty} f(E)dE \). If \( F(E_0) = f(E_0) \), we have \( f(E_0) \propto E_0^{-2} \), which is satisfied by (3.3.23). Therefore, considering the recoil density \( F(E_0) \) as an atom source, Falcone-Sigmund theory
can give a correct atom flux (3.3.22). In other words, any loss of particles due to scattering from a
given direction of motion is compensated by an equivalent gain [32][33]. However, Falcone-Sigmund theory is tenable only for single element medium.

ii) Falcone theory

Different from Falcone-Sigmund theory, G. Falcone believes that each flux atom must come from some atom sources without undergoing collision on its way [34][35]. In this case, we have to choose collision density $F^c(E_0)$ as an atom source to generate the correct atom flux (3.3.20). However, in order to estimate the average depth of origin of the sputtered atoms, Falcone chose the recoil density $F(E_0)$ as an atom source in his analysis, i.e. $\psi(E_0) = L^c(E_0)F(E_0)$. As we see, this relation contradicts our basic transport theory (3.3.20), thus, can not be a tenable theory. In general, $F^c(E_0) \neq F(E_0)$.

APPENDIX IIIA:

Statement: If

$$W_{ik}^L(E) = \int_0^{\infty} A_{ik}(T/T_m) \left[ H_i^L(E) - P_L(\cos \phi_{ik})H_i^L(E-T) \right. \right.$$

$$\left. - P_L(\cos \phi_{ik}^*)H_i^L(T) \right] dT/T_m$$

(III.A.1)

where $H_i^L(E < U) = 0$. (III.A.2)

and

$$\begin{cases} A_{ik}(X) \geq 0, & \text{for } 1 \geq X \geq 0 \\ A_{ik}(X) = 0, & \text{for } 1 > 1 \text{ or } X < h \end{cases}$$

(III.A.3)

Introducing the new variables

$$E = Ue^u, \quad T = Ue^v$$

(III.A.4)

and taking the Laplace Transform with respect to $u$, we have

$$\alpha_k \tilde{W}_{ik}^L(s) = \beta_{ik}^L(s)\tilde{H}_i^L(s) - \tilde{H}_k^L(s)$$

(III.A.5)

Here $1 > h \geq 0$, $T_m \equiv \gamma E$, $\gamma \equiv \gamma_{ik}$ and

$$\tilde{W}_{ik}^L(s) = \int_0^\infty du \cdot \exp(-su)W_{ik}^L(Ue^u).$$
Proof:

\[ W_1(E) = \int_0^{T_m} A_{ik} \left( \frac{T}{T_m} \right) H_i^L(E) dT/T_m = H_i^L(E) \int_0^1 A_{ik}(X) dX \]

\[ \therefore \quad \tilde{W}_1(s) = \tilde{H}_i^L(s) \int_0^1 A_{ik}(X) dX \quad (\text{III.A.6}) \]

Considering (III.A.2) and (III.A.3), we have

\[ W_2(E) = \int_0^{T_m} A_{ik} \left( \frac{T}{T_m} \right) \mathcal{P}_L(\cos \phi'_{ik}) H_i^L(E - T) dT/T_m \]

\[ = \int_U A_{ik} \left( \frac{E - T}{\gamma E} \right) dT \mathcal{P}_L \left( \frac{\sqrt{E - T}}{\sqrt{E}} \right) H_i^L(T) \quad (\text{III.A.7}) \]

Substituting (III.A.4) into (III.A.7), we have

\[ W_2(U^{e^u}) \]

\[ = \int_0^T A_{ik} \left[ 1 - e^{-(u-v)/\gamma} \right] \mathcal{P}_L \left[ U_{ik} e^{-0.5(u-v)} + V_{ik} e^{0.5(u-v)} \right] H_i^L(U^{e^u}) \quad (\text{III.A.8}) \]

\[ = A_{ik} \left[ (1 - e^{-u})/\gamma \mathcal{P}_L \right] \left( U_{ik} e^{-0.5u} + V_{ik} e^{0.5u} \right) \mathcal{H}_i^L \left[ U^{e^u} \right] \]

Where, $\otimes$ stands for the convolution. Therefore, taking the Laplace Transform of (III.A.8) with respect to $u$, we have

\[ \tilde{W}_2(s) = \tilde{H}_i^L(s) \int_0^1 dX A_{ik} \left[ (1 - X)/\gamma \right] \mathcal{P}_L \left[ U_{ik} X^{0.5} + V_{ik} X^{-0.5} \right] X' dX/\gamma \]

\[ = \tilde{H}_i^L(s) \int_0^1 dX A_{ik}(X) \left[ (1 - \gamma X)/\gamma \right] \mathcal{P}_L \left[ U_{ik} (1 - \gamma X)^{0.5} + V_{ik} (1 - \gamma X)^{-0.5} \right] \quad (\text{III.A.9}) \]

Considering (III.A.2) and (III.A.3), we have

\[ W_3(E) = \int_0^{T_m} A_{ik} \left( \frac{T}{T_m} \right) \mathcal{P}_L(\cos \phi''_{ik}) H_i^L(T) dT/T_m \]

\[ = \int_E^T A_{ik} \left( \frac{T}{T_m} \right) \mathcal{P}_L(\cos \phi''_{ik}) H_i^L(T) dT/T_m \quad (\text{III.A.10}) \]

Substituting (III.A.4) into (III.A.10), we have

\[ W_3(U^{e^u}) = \int_0^E A_{ik} \left[ e^{-(u-v)/\gamma} \right] \mathcal{P}_L \left[ e^{-0.5(u-v)/\gamma} \right] \mathcal{H}_i^L(U^{e^u}) \]

\[ = A_{ik} \left[ e^{-u}/\gamma \mathcal{P}_L \right] \left( e^{-0.5u}/\gamma \right) \mathcal{H}_i^L \left[ U^{e^u} \right] \quad (\text{III.A.11}) \]

Therefore, taking the Laplace Transform of (III.A.11) with respect to $u$, we have

\[ \tilde{W}_3(s) = \int_0^1 dX A_{ik}(X) X' \mathcal{P}_L \left( \sqrt{X} \right) \tilde{H}_i^L(s) \]

\[ = \tilde{H}_i^L(s) \gamma' \int_0^1 dX A_{ik}(X) X' \mathcal{P}_L \left( \sqrt{X} \right) \quad (\text{III.A.12}) \]

Thus, from (III.A.6)- (III.A.9)- (III.A.12), finally we have
\[
\alpha_k \tilde{W}_k^I (s) = \alpha_k \left[ \tilde{W}_1 (s) - \tilde{W}_2 (s) - \tilde{W}_3 (s) \right] \\
= \tilde{H}_1^I (s) \beta^I_{ik} (s) \varepsilon^I_{ik} (s) - \beta^I_{ik} (s) \tilde{H}_1^I (s) \\
= \beta^I_{ik} (s) [\varepsilon^I_{ik} (s) \tilde{H}_1^I (s) - \tilde{H}_1^I (s)] \\
\]

which is (III A.5). End of proof.

**APPENDIX III-B:**

**Statement:** The determinant \( D^I (s) \) has the highest zero \( s = 0.5 \).

**Proof:** Substituting \( \beta^I_{ik} (0.5) = \beta^I_{ik} \) and \( \varepsilon^I_{ik} (0.5) = (M_1 / M_2)^{0.5} \) into \( D^I (s) \), we have

\[
D^I (0.5) = \begin{vmatrix}
\beta^I_{12} \sqrt{M_3 / M_1} + \beta^I_{13} \sqrt{M_3 / M_1} + \cdots + \beta^I_{1n} \sqrt{M_n / M_1}, & -\beta^I_{12}, & -\beta^I_{13}, & \cdots, & -\beta^I_{1n} \\
-\beta^I_{21}, & \beta^I_{21} \sqrt{M_1 / M_2} + \beta^I_{23} \sqrt{M_3 / M_2} + \cdots + \beta^I_{2n} \sqrt{M_n / M_2}, & -\beta^I_{23}, & \cdots, & -\beta^I_{2n} \\
\vdots & \ddots & \ddots & \ddots & \ddots \\
-\beta^I_{n1}, & \cdots, & -\beta^I_{n(n-1)}, & \beta^I_{n1} \sqrt{M_2 / M_n} + \beta^I_{n2} \sqrt{M_3 / M_n} + \cdots + \beta^I_{n(n-1)} \sqrt{M_n / M_n} \\
\end{vmatrix}
\]

\[
\sum_{k=1}^{n} \frac{M_k}{M_1} \cdot (k\text{th.column}) = \begin{pmatrix} 0 \\ \vdots \\ 0 \end{pmatrix},
\]

\[
D^I (0.5) = 0, \ D^I (s) \text{ has a zero } s = 0.5.
\]

Next step, we want to show that the determinant \( D^I (s) \) has no any zeros for \( s > 0.5 \). From (3.1.11), one can see

\[
\beta^I_{ik} (s) = \alpha_k \gamma^s \int_0^s dX A_{ik} (X) X^{s+0.5} > 0 \text{ and is a strictly decreasing function of } s, \text{ thus}
\]

\[
\beta^I_{ik} (0.5) > \beta^I_{ik} (s) > 0, \quad \text{for } s > 0.5 \quad (\text{III} \text{B.1})
\]

From (3.1.12), one can see

\[
\beta^I_{ij} (s) \varepsilon^I_{ij} (s) = \alpha_j \gamma^s \int_0^s dX A_{ij} (X) \left[ U_{ij} (1 - \gamma X)^{s+0.5} + V_{ij} (1 - \gamma X)^{s-0.5} \right]
\]

is a strictly increasing function of \( s \), for \( s > 0.5 \), thus
\[
\beta_j^i(s)e^i_j(s) > \alpha_j \cdot \int_0^1 dX A_{ij}(X) \left[ I - V_{ij} - U_{ij}(1-\gamma X) \right]
\]
\[
= \beta_j^i(0.5)(M_i/M_j)^{0.5}, \quad \text{for } s > 0.5.
\]

Considering (III B.1), we have
\[
\varepsilon_{ik}^1(s) > (M_i/M_j)^{0.5}, \quad \text{for } s > 0.5. \quad \text{(III B.2)}
\]

Now, let \( \beta = (\beta_{ik}) \) be an arbitrary \( n \times n \) matrix with positive elements, \( \beta_{ik}^1 > 0 \) and let \( \varepsilon_{ik}^1 \) (\( i, k = 1, 2, \ldots, n \)) be a set of \( n^2 \) arbitrary elements, we define another \( n \times n \) matrix \( \Delta = (\Delta_{ik}) \) by
\[
\Delta_{ik} = \delta_{ik} \cdot \sum_j \beta_{ij}^1 \varepsilon_{ij}^1
\]
and will now prove the following theorem for the determinant
\[
\det(\Delta - \beta^1) = \begin{vmatrix}
\beta_{11}^1(\varepsilon_{11}^1 - 1) + \beta_{12}^1 \varepsilon_{12}^1 + \cdots + \beta_{1n}^1 \varepsilon_{1n}^1, & -\beta_{12}^1, & -\beta_{13}^1, & \cdots, & -\beta_{1n}^1 \\
-\beta_{21}^1 & \beta_{22}^1 \varepsilon_{22}^1 + \cdots + \beta_{2n}^1 \varepsilon_{2n}^1, & -\beta_{23}^1, & \cdots, & -\beta_{2n}^1 \\
\vdots & \vdots & \ddots & \ddots & \vdots \\
-\beta_{n1}^1 & -\beta_{n2}^1 & \cdots & -\beta_{n(n-1)}^1 & \beta_{nn}^1 \varepsilon_{nn}^1 + \sum_{i=1}^{n-1} \beta_{ni}^1 \varepsilon_{ni}^1 + \cdots + \beta_{nn}^1 (\varepsilon_{nn}^1 - 1)
\end{vmatrix}
\]
If all elements \( \varepsilon_{ik}^1 > (M_k/M_i)^{0.5} \), then \( \det(\Delta - \beta) > 0. \)

The theorem is proved by induction, increasing the dimension of the matrix from \( (n-1) \) to \( n. \)

\begin{itemize}
  \item[i)] The case \( n = 1 \) is trivial: \( \det(\Delta - \beta) = \beta_{11}^1(\varepsilon_{11}^1 - 1) > 0. \)
  
  \item[ii)] The general step \( n-1 \rightarrow n \) : we first note that if all \( \varepsilon_{ik}^1 = (M_k/M_i)^{0.5} \), then \( \det(\Delta - \beta) = 0. \) It is, therefore, sufficient to show that for \( \varepsilon_{ik}^1 > (M_k/M_i)^{0.5} \), \( \det(\Delta - \beta) \) is a strictly increasing function of all the all \( \varepsilon_{ik}^1, \) or
\[
\left\{ \frac{\partial \det(\Delta - \beta)}{\partial \varepsilon_{ik}^1} > 0 \right\} \quad \text{for all } \varepsilon_{ik}^1 > (M_k/M_i)^{0.5}.
\]

For seasons of symmetry, it is sufficient to consider the case \( i = 1, \) we get by differentiation:
The last determinant is a \((n-1) \times (n-1)\) determinant, but in a form not suited for induction.

However, the matrix can be brought into a suitable form by defining new quantities \(\bar{e}_{ii}\) and \(\bar{\beta}_{ii}\) so that for \(i = 2, \cdots, n\)

1) \(\beta_{ii}^{1}(\bar{e}_{ii}^{1} - 1) + \beta_{ii}^{1}e_{ii}^{1} = \bar{\beta}_{ii}^{1}(\bar{e}_{ii}^{1} - 1)\)

2) \(\bar{\beta}_{ii}^{1} > 0\)

3) \(\bar{e}_{ii}^{1} > 1\).

which is evidently possible. For \(i \neq k\), we define \(e_{ii}^{1} = \bar{e}_{ii}^{1}\) and \(\beta_{ii}^{1} = \bar{\beta}_{ii}^{1}\). Thus,

\[
\frac{\partial}{\partial \bar{e}_{ik}^{1}} \det(\Delta - \beta^{1}) = \begin{vmatrix}
\beta_{11}^{1}(e_{11}^{1} - 1) + \beta_{12}^{1}e_{12}^{1} + \cdots + \beta_{1n}^{1}e_{1n}^{1} & -\beta_{12}^{1} & -\beta_{13}^{1} & \cdots & -\beta_{1n}^{1} \\
-\beta_{21}^{1}, \beta_{22}^{1} + \beta_{23}^{1}(e_{22}^{1} - 1) + \cdots + \beta_{2n}^{1}e_{2n}^{1} & -\beta_{22}^{1} & -\beta_{23}^{1} & \cdots & -\beta_{2n}^{1} \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
-\beta_{n1}^{1}, -\beta_{n2}^{1}, \cdots, -\beta_{n(n-1)}^{1}, [\beta_{n1}^{1}e_{n1}^{1} + \beta_{n2}^{1}e_{n2}^{1} + \cdots + \beta_{nn}^{1}(e_{nn}^{1} - 1)] & -\beta_{n2}^{1} & -\beta_{n3}^{1} & \cdots & -\beta_{nn}^{1}
\end{vmatrix}
\]

Here \(\beta_{ik}^{1} > 0\) and the determinant is positive too, since it is a \((n-1) \times (n-1)\) determinant of the type considered. This proves the theorem.
CHAPTER 4
SOLUTIONS FOR ARBITRARY SCATTERING CROSS-SECTIONS

4.1 APPROXIMATE ASYMPTOPIC SOLUTIONS FOR SEMI-SEPARABLE SCATTERING CROSS-SECTIONS

4.1.1 “AS” THEORY

For the semi-separable scattering cross sections [23] [25]

\[ d\sigma_{ij}(E, T) = E^{-2m_{ij}} A_{ij}(X) dX \]  

(4.1.1)

the pool of asymptotic solutions (3.3.1-11) derived in previous chapter can not be used, because cross sections (4.1.1) couldn’t be separable to fit (3.1.1), due to \( E^{-2m_{ij}} \neq Q_{ij}(E) \). In order to solve the problem, we proposed AS theory [25]. Multiplying both sides of Eqs. (2.3.2) by \( E^{-2m_{ij}} \), introducing the new variables \( E = E_0 e^u \), \( T = E_0 e^v \) and taking the Laplace Transform on both sides with respect to \( u \), we obtained the following sets of equations,

\[ \sum_k E_0^{-M_{ij}} \beta_{ik} (s + M_{ik}) [\Gamma_{ij}(s + M_{ik}) - \Gamma_{ij}(s + M_{ik})] = E_0^{-2m_{ij}} \delta_{ij} / (N\sigma_0 E_0) \]  

(4.1.2)

rather than Eqs. (3.1.2) in general. \( m_i \) are free parameters remained to choose for final solutions. If \( m_{ik} \) is independent of \( k \), it is natural to choose \( m_i = m_{ik} \), then \( M_{ik} = 0 \), (4.1.2) reduced to (3.1.2), then (3.3.1) gives the exact asymptotic solution. Unfortunately, if \( m_{ik} \) is dependent on \( k \), it’s almost not possible to solve Eqs. (4.1.2) analytically at least for multi-component media. But, in most of applications, we are looking for solution in energy region \( E >> E_0 \). In the linear sputtering, based on both physics and mathematics considerations, we should have

\[ G_{ij}(E, E_0) = G_{ij} E/E_0 \], \quad \text{for } E >> E_0 \]  

(4.1.3)

If \( m_{ik} \) is related to \( k \), we have to choose such \( m_i \) so that (4.1.3) satisfies the basic symmetric
property of asymptotic solutions, i.e.

\[ G_{ij} = G_{2j} = \cdots = G_{nj} \equiv G_j, \quad j = 1, 2, \cdots, n \]  \hspace{1cm} (4.1.4)

(4.1.3) means that we have the following exact relations,

\[ G_{ij} = \lim_{s \to 0} [sG_{ij}(1 + s)] \]  \hspace{1cm} (4.1.5)

For a non-zero but very small \( s \), we have the following approximate relations,

\[ G_{ij} \approx sG_{ij}(1 + s) \]  \hspace{1cm} (4.1.6)

Substituting \( s = 1 \) into Eqs. (4.1.2) and using (4.1.5, 6), we obtained

\[ \sum_k D_{ik}(E_0)G_{kj} = E_0^{2m_2}\delta_{ij}/(\nu_0E_0) \]  \hspace{1cm} (4.1.7)

where

\[
\begin{align*}
D_{ij}(E_0) &= -E_0^{-M_{ij}}\beta_{ij}(1 + M_{ij})/M_{ij}, \quad \text{for } i \neq j \\
D_{ii}(E_0) &= \sum_k E_0^{M_{ik}}\beta_{ik}(1 + M_{ik})[\nu_{ik}(1 + M_{ik}) - \delta_{ik}]/M_{ik}
\end{align*}
\]

Solving Eqs. (4.1.7) yields \( G_{ij} \), then choosing \( m_i \) to satisfy (4.1.4), finally (4.1.3) generates approximate asymptotic solutions, named AS theory in this work. In principle, this method is suitable for all of other equations (2.3.3,4), (2.3.7) and (2.3.9~12). For simplicity, we only consider a binary media \((\alpha_1 + \alpha_2 = 1)\). The approximate asymptotic solutions are

\[
\begin{align*}
G_{11}(E,E_0) &= EE_0^{2m_2}\cdot D_{22}/(\nu_0[D]) \\
G_{21}(E,E_0) &= EE_0^{2m_2}\cdot (-D_{21})/(\nu_0[D]) \\
G_{12}(E,E_0) &= EE_0^{2m_2}\cdot (-D_{12})/(\nu_0[D]) \\
G_{22}(E,E_0) &= EE_0^{2m_2}\cdot D_{11}/(\nu_0[D])
\end{align*}
\]  \hspace{1cm} (4.1.8)

where

\[
\begin{align*}
D_{11} &= E_0^{-M_{11}}\cdot E_{11}(1 + M_{11}) - D_{12}\cdot \varepsilon_{12}(s + M_{12}) \\
D_{12} &= -E_0^{-M_{12}}\cdot \beta_{12}(s + M_{12})/M_{12} \\
D_{21} &= -E_0^{-M_{21}}\cdot \beta_{21}(s + M_{21})/M_{21} \\
D_{22} &= -D_{21}\cdot \varepsilon_{21}(s + M_{21}) + E_0^{-M_{22}}\cdot E_{22}(1 + M_{22})
\end{align*}
\]
For the recoil density, by using the same method, it’s easy to derive the approximate asymptotic solutions,

\[
\begin{align*}
[F_1(E_0, E_0)] &= E E_0^{-2} \cdot [E_0^{-M_{11}} \cdot D_{22} \beta_1 (1 + M_{11}) - E_0^{-M_{21}} \cdot D_{12} \beta_2 (1 + M_{21})] / [D] \\
[F_2(E_0, E_0)] &= E E_0^{-2} \cdot [E_0^{M_{12}} \cdot D_{12} \beta_1 (1 + M_{12}) - E_0^{-M_{12}} \cdot D_{12} \beta_1 (1 + M_{11})] / [D] \\
[F_1(E_0, E_0)] &= E E_0^{-2} \cdot [E_0^{M_{22}} \cdot D_{22} \beta_2 (1 + M_{22}) - E_0^{-M_{22}} \cdot D_{22} \beta_2 (1 + M_{22})] / [D] \\
[F_2(E_0, E_0)] &= E E_0^{-2} \cdot [E_0^{M_{22}} \cdot D_{12} \beta_2 (1 + M_{22}) - E_0^{-M_{22}} \cdot D_{22} \beta_1 (1 + M_{12})] / [D]
\end{align*}
\]

The condition of asymptotic solutions (4.1.4) provided only one choice for \( m_1 \), i.e.

\[
m_1 = m_{12} \quad \text{and} \quad m_2 = m_{21},
\]

Thus

\[
\begin{align*}
M_{11} &= 2m_{11} - 2m_{12} \\
M_{12} &= M_{21} = 0 \\
M_{22} &= 2m_{22} - 2m_{21}
\end{align*}
\]

Substituting (4.1.10,11) into (4.1.8,9), we obtained sputtered particle fluxes and recoil density

\[
\begin{align*}
\psi_1(E_0) &= EE_0^{2m_{12} - 2} \beta_2 (1) / (ND) \\
\psi_2(E_0) &= EE_0^{2m_{21} - 2} \beta_1 (1) / (ND)
\end{align*}
\]

\[
\begin{align*}
F_1(E_0) &= EE_0^{-2} \cdot [E_0^{2m_{12} - 2m_{11}} \beta_1 (1 - 2m_{12} + 2m_{11}) + \beta_{12}(1)] \cdot \beta_2 (1) / D \\
F_2(E_0) &= EE_0^{-2} \cdot [\beta_2 (1) + E_0^{2m_{12} - 2m_{22}} \cdot \beta_1 (1 - 2m_{21} + 2m_{22})] \cdot \beta_{12} (1) / D
\end{align*}
\]

where

\[
D = E_0^{2m_{12} - 2m_{11}} \beta_2 (1) E_{11}(1 - 2m_{12} + 2m_{11}) + \beta_{12}(1) E_{21}(1) + \beta_2 (1) \beta_{12}(1)
\]

\[
+ E_0^{2m_{12} - 2m_{22}} \beta_1 (1) E_{21}(1 - 2m_{21} + 2m_{22})
\]

(4.1.12,13) belongs to “AS” theory.

### 4.1.2 COMPARING “AS” THEORY WITH “US” THEORY

In 1992, H. M. Urbashek and U. Conrad presented a wider variety of solutions of Eqs. (2.3.2,3)
for general cross sections (3.2.1)[23]. P. Sigmund set a high value on these asymptotic solutions [51], because (3.2.1) does not fit the separable scattering cross sections (3.1.1). However, in their case [23], the semi-separable scattering cross sections (4.1.1) reduced to (3.2.1) as long as $A_{ij}(X) = X^{-1-m_{ij}}$. Therefore, UC theory is a special case of “AS” theory (4.1.12,13).

Specific case I: “Detailed balance”

If $m_{12} > m_{11}$ and $m_{21} > m_{22}$, AS theory (4.1.12,13) reduced to

$$
\begin{align*}
\psi_1(E_o) &= \frac{1}{E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{N^2 E_{0}^{m_{11}}} \\
\psi_2(E_o) &= \frac{\beta_{12}(1)}{\beta_{21}(1)E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{N^2 E_{0}^{m_{11} + 2m_{21} - 2m_{21}}} \\
F_1(E_o) &= \frac{\beta_{11}(1)}{E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{E_{0}^{2-m_{11}}} \\
F_2(E_o) &= \frac{\beta_{12}(1)}{E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{E_{0}^{2-m_{11} + 2m_{21}}}
\end{align*}
$$

(4.1.16,17) are approximate asymptotic solutions, because (4.1.6) is also used for the derivation.

Because all of (4.1.6) related terms have been canceled in the derivation process, (4.1.14,15) derived in this work are exact asymptotic solutions. (4.1.14,15) have already been derived by H. M. Urbassek and U. Conrad [23], they named (4.1.14,15) “Detailed balance”.

Specific case II: “Dominance”

If $m_{12} < m_{11}$ and $m_{21} > m_{22}$, AS theory (4.1.12,13) reduced to

$$
\begin{align*}
\psi_1(E_o) &= \frac{E}{E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{N^2 E_{0}^{m_{11}}} \\
\psi_2(E_o) &= \frac{\beta_{12}(1)}{\beta_{21}(1)E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{N^2 E_{0}^{m_{11} + 2m_{21} - 2m_{21}}} \\
F_1(E_o) &= \frac{\beta_{11}(1)}{E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{E_{0}^{2-m_{11}}} \\
F_2(E_o) &= \frac{\beta_{12}(1)}{E_{11}(1 - 2m_{12} + 2m_{11})} \cdot \frac{E}{E_{0}^{2-m_{11} + 2m_{21}}}
\end{align*}
$$

Two similar formula have been published by H. M. Urbassek and U. Conrad [23],
This case was named “Dominance”[23]. Comparing (4.1.18,19) with (4.1.16,17) one function by one function in detail, one can see bit difference in each formula. One may naturally link the origin of the bit differences with the choice of $m_i$. We know that (4.1.10) is necessary and sufficient condition to guarantee $G_{1j} = G_{2j}$ and $F_{1j} = F_{2j}$. In order to search the origin of the differences between (4.1.18,19) and (4.1.16,17), lets try to choose different values for $m_i$, i.e.

\[
\begin{align*}
m_1 &= m_{11} \text{ and } m_2 = m_{11} - m_{12} + m_{21}, \quad \text{then} \\
M_{11} &= 0 \\
M_{12} &= M_{21} = 2m_{11} - 2m_{12} < 0 \\
M_{22} &= 2m_{22} - 2m_{21} + 2m_{12} - 2m_{22} < 0
\end{align*}
\]

Substituting (4.1.20) into (4.1.8,9) and considering $m_{12} < m_{11}$ and $m_{21} > m_{22}$, we obtained,

\[
\begin{align*}
\Psi'_{11}(E_0) &= \frac{1}{E_{11}(I)} \cdot \frac{E}{NE_0^{2-2m_{11}}} \\
\Psi'_{12}(E_0) &= \frac{\beta_{12}(1 - 2m_{11} + 2m_{12})}{\beta_{21}(1 - 2m_{11} + 2m_{12})E_{11}(I)e_{21}(I - M_{21})} \cdot \frac{E}{NE_0^{2-2m_{11}+2m_{12}}} \\
F_{11}(E_0) &= \frac{\beta_{11}(I)}{E_{11}(I)} \cdot \frac{E}{E_0^2} \\
F_{12}(E_0) &= \frac{\beta_{12}(1 - 2m_{11} + 2m_{12})}{E_{11}(I)} \cdot \frac{E}{E_0^{2-2m_{11}+2m_{12}}} \quad (4.1.21) \\
\Psi'_{21}(E_0) &= \frac{1}{e_{21}(I - M_{21})E_{11}(I)} \cdot \frac{E}{NE_0^{2-2m_{11}}} \\
\Psi'_{22}(E_0) &= \frac{(2m_{12} - 2m_{11})}{\beta_{21}(1 + M_{12})e_{21}(I + M_{12})} \cdot \frac{E}{NE_0^{2-2m_{11}}} < 0 \quad (4.1.22)
\end{align*}
\]
\[
\begin{align*}
&\left\{ \begin{array}{l}
F_{21}(E_0) = \frac{\beta_{11}(l) + (2m_{12} - 2m_{11})E_{11}(l)}{E_{11}(l)} \cdot \frac{E}{E_0^2} \\
F_{22}(E_0) = \frac{(2m_{12} - 2m_{11})\beta_{22}(1 + M_{22})}{\beta_{21}(1 + M_{12})\varepsilon_{22}(1 + M_{12})} \cdot \frac{E}{E_0^{2-2m_{11}+2m_{12}}} < 0
\end{array} \right. \\
(4.1.24)
\end{align*}
\]

Obviously, (4.1.21~24) are incorrect formulae, because \( \Psi_{ij} \neq \Psi_{2j} \) and \( F_{ij} \neq F_{2j} \). Besides, formulae (4.1.23, 24) predict the ridiculous “negative particle flux \( \Psi_{2j} \) and recoil density” \( F_{2j} \).

Comparing UC formulae (4.1.18,19) with (4.1.21,22), we can see that

\[
\begin{align*}
&\left\{ \begin{array}{l}
\Psi_1(E_0) = \Psi_{11}(E_0) \\
\Psi_2(E_0) = \Psi_{12}(E_0) \cdot \varepsilon_{21}(1 - 2m_{11} + 2m_{12}) \\
F_1(E_0) = F_{11}(E_0) \\
F_2(E_0) = F_{12}(E_0)
\end{array} \right. \\
(4.1.25)
\end{align*}
\]

Therefore, UC formula (4.1.19) is equivalent to the illegal formulae (4.1.22) exactly, UC formula (4.1.18) is almost equivalent to the illegal formulae (4.1.25) except a extra factor \( \varepsilon_{21}(1 - 2m_{11} + 2m_{12}) \) which closes to unity.

However, in view of \( |m_{ii} - m_{i2}| \) is small in our derivation, the difference between AS theory (4.1.16,17) [25] and UC theory (4.1.18,19) [23] should not be very important qualitatively.

### 4.2 “SC” THEORY

#### 4.2.1 INTRODUCTION TO “SC” THEORY

For arbitrary interaction cross sections, M. Vicanek, H. M. Urbassek and U. Conrad (VUC) [13][23] introduced the stopping cross sections \( S_{ij} \) and the (partial) energy slowing down cross sections \( \sigma_{ij}^s \), \( \sigma_{ij}^r \) and \( \tilde{\sigma}_{ij} \) as following,

\[
S_{ij}(E) \equiv \int_0^T T d\sigma_{ij}(E, T) \\
(4.2.1)
\]

\[
\sigma_{ij}^s(E) \equiv -\int_0^T [1 - (T/E)] \ln[1 - (T/E)] d\sigma_{ij}(E, T) > 0 \\
(4.2.2)
\]

\[
\sigma_{ij}^r(E) \equiv -\int_0^T (T/E) \ln(T/E) d\sigma_{ij}(E, T) > 0 \\
(4.2.3)
\]
In particular, for the separable scattering cross sections (3.1.1), the functions (4.2.1-5) reduced to

\begin{align*}
\tilde{\sigma}_j(E) &\equiv \sigma_j(E) + \sigma_j(E) \\
\tilde{\sigma}_i(E) &\equiv \sum_j \alpha_j \tilde{\sigma}_j(E).
\end{align*}

(4.2.4)

(4.2.5)

For simplicity, we only consider a binary media \((\alpha_1 + \alpha_2 = 1)\). The asymptotic solutions (3.3.1) and (3.3.2) can be written,

\begin{align*}
\left[ \psi_1(E_\alpha) = EE_\alpha^{-2} \cdot \alpha_1 S_{21}(E_\alpha) / [NDE_\alpha] \right] \\
\left[ \psi_2(E_\alpha) = EE_\alpha^{-2} \cdot \alpha_2 S_{12}(E_\alpha) / [NDE_\alpha] \right]
\end{align*}

(4.2.6)

\begin{align*}
\left[ F_1(E_\alpha) = EE_\alpha^{-3} \cdot [\alpha_1 S_{11}(E_\alpha) + \alpha_2 S_{12}(E_\alpha)] \cdot \alpha_1 S_{21}(E_\alpha) / D(E_\alpha) \right] \\
\left[ F_2(E_\alpha) = EE_\alpha^{-3} \cdot [\alpha_1 S_{21}(E_\alpha) + \alpha_2 S_{22}(E_\alpha)] \cdot \alpha_2 S_{12}(E_\alpha) / D(E_\alpha) \right]
\end{align*}

(4.2.7)

where \(D(E_\alpha) = \alpha_1 S_{21}(E_\alpha) \tilde{\sigma}_1(E_\alpha) + \alpha_2 S_{12}(E_\alpha) \tilde{\sigma}_2(E_\alpha)\).

We noticed that \(\sigma_j(E, T)\) appears entirely in (4.2.1-7) without separation, i.e.

\[ Q_1(E) \int_0^\infty dX A_j(X) H(X) = \int_0^E d\sigma_j(E, T) H(T/\gamma E), \]

where \(H(X)\) is an arbitrary function. The asymptotic solutions (4.2.6,7) can serve as approximately asymptotically correct ones for arbitrary cross sections [19]. We named (4.2.6,7) “SC” theory in this work. (4.2.6) has already been derived by VUC based on the condition of detailed balance (3.3.14) for a binary media, see Eq. (45) on page 622 in Ref. [13]. On the same pager, “VCU” pointed out: “For general power cross sections, however, Eq.(45) does not represent the correct asymptotic behavior.” In order to check the vanity of the statement, lets substitute the semi-separable scattering cross sections (4.1.1) into (4.2.6,7), then we obtained
\[
\begin{align*}
\Psi_1(E_0) &= EE_0 2m_{12} - 2 \beta_{21}(l)/(ND) \\
\Psi_2(E_0) &= EE_0 2m_{12} - 2 \beta_{12}(l)/(ND)
\end{align*}
\] (4.2.8)

\[
\begin{align*}
F_1(E_0) &= EE_0 2 \left[ E_0 2m_{12} - 2m_{11}, \beta_{11}(l) + \beta_{12}(l) \right] \cdot \beta_{21}(l)/D \\
F_2(E_0) &= EE_0 2 \left[ \beta_{21}(l) + E_0 2m_{12} - 2m_{12}, \beta_{11}(l) \right] \cdot \beta_{12}(l)/D
\end{align*}
\] (4.2.9)

Here \( D = E_0 2m_{12} - 2m_{11}, \beta_{21}(l)E_{11}(l) + \beta_{12}(l)E_{21}(l) + E_{12}(l)\beta_{21}(l) + E_0 2m_{12} - 2m_{12}, \beta_{12}(l)E_{22}(l) \)

In view of \( |m_{11} - m_{12}| \) is small in the derivation of AS theory, therefore, “SC” theory (4.2.8, 9) is almost equivalent AS theory (4.1.12,13).

### 4.2.2 COMPARING “SC” THEORY WITH “AS” THEORY

Both (4.2.8,9) reduced to (3.3.1,18) respectively if \( m_{11} = m_{12} \), otherwise, (4.2.8,9) will approach one of the following formulae of AS theory asymptotically when \( E_0/E \to 0 \):

i) “Detailed balance”

If \( m_{12} > m_{11} \) and \( m_{21} > m_{22} \), (4.2.8,9) exactly reduced to (4.1.14,15) respectively.

ii) “Dominance”

If \( m_{12} > m_{11} \) and \( m_{22} > m_{21} \), (4.2.8,9) reduced to

\[
\begin{align*}
\Psi_1(E_0) &= \frac{1}{E_{11}(l)} \cdot \frac{E}{N_{E_0} E_0 2m_{11}} \\
\Psi_2(E_0) &= \frac{\beta_{12}(l)}{\beta_{21}(l)E_{11}(l)} \cdot \frac{E}{N_{E_0} E_0 2m_{12} + 2m_{12} - 2m_{21}}
\end{align*}
\] (4.2.10)

\[
\begin{align*}
F_1(E_0) &= \frac{\beta_{11}(l)}{E_{11}(l)} \cdot \frac{E}{E_0} \\
F_2(E_0) &= \frac{\beta_{12}(l)}{E_{11}(l)} \cdot \frac{E}{E_0} 2m_{12} + 2m_{12}
\end{align*}
\] (4.2.11)

respectively. In view of \( |m_{11} - m_{12}| \leq 0.1 \) is small, (4.2.10,11) are very close to (4.1.16,17) or (4.1.18,19) respectively.

iii) “Ignorance”

If \( m_{11} > m_{12} > m_{22} > m_{21} \) and \( m_{11} - m_{12} > m_{22} - m_{21} \), (4.2.8) reduced to (4.2.10)
for particle flux, but, instead (4.2.11), (4.2.9) reduced to

\[
\begin{align*}
F_1(E_0) &= \frac{\beta_{11}(l)}{E_{11}(l)} \cdot \frac{E}{E^2} \\
F_2(E_0) &= \frac{\beta_{12}(l)}{\beta_{21}(l)[E_{11}(l)]^2} \cdot \frac{E}{E_0}^{\frac{n}{2}} \cdot 2^{2m_{11}} \cdot 2^{2m_{12}} \cdot 2^{2m_{22}}
\end{align*}
\]

(4.2.12)

for recoil density.

iv) “New ignorance”

If \( m_{11} > m_{12} \), \( m_{22} > m_{21} \) and \( m_{11} - m_{12} = m_{22} - m_{21} \), (4.2.8, 9) reduced to

\[
\begin{align*}
\psi_1(E_0) &= EE_0^{4m_{12} - 2m_{11} - 2} \cdot \beta_{21}(l)/(ND) \\
\psi_2(E_0) &= EE_0^{4m_{12} - 2m_{22} - 2} \cdot \beta_{12}(l)/(ND) \\
F_1(E_0) &= EE_0^{2} \cdot \beta_{12}(l)/D \\
F_2(E_0) &= EE_0^{2} \cdot \beta_{21}(l)/D
\end{align*}
\]

(4.2.13, 4.2.14)

Here \( D = \beta_{21}(l)E_{11}(l) + \beta_{12}(l)E_{22}(l) \).

Three formulae (4.2.12, 13, 14) have never been published anywhere.

### 4.2.3 COMPARING “SC” THEORY WITH MONTE CARLO SIMULATIONS

A wider variety of solutions derived by H. M. Urbassek and U. Conrad in [23] have been showed to fit corresponding Monte Carlo Simulations [23]. Following same procedure, “SC” theory (4.2.8, 9) can be further written as

\[
\begin{align*}
x \psi_1(x) &= x^{2m_{12} - 2m_{11}} \beta_{21}/D(x) \\
x \psi_2(x) &= x^{2m_{21} - 2m_{12}} \beta_{12}/D(x)
\end{align*}
\]

(4.2.15)

\[
\begin{align*}
x^2 f_1(x) &= x^{(2m_{12} - 2m_{11})} \beta_{11} + \beta_{12} \beta_{21}/D(x) \\
x^2 f_2(x) &= \beta_{21} + x^{(2m_{21} - 2m_{22})} \beta_{22} \beta_{12}/D(x)
\end{align*}
\]

(4.2.16)

Here \( D(x) = x^{2m_{12} - 2m_{11}} \beta_{11} \beta_{21} \Gamma_{m_{11} - 1} + \beta_{12} \beta_{21} \left( \Gamma_{m_{12} - 1} + \Gamma_{m_{21} - 1} \right) + x^{2m_{21} - 2m_{22}} \beta_{21} \beta_{22} \Gamma_{m_{22} - 1}, \)

\[
\beta_{ij} = 0.5 \tilde{C}_{ij}/(1 - m_{ij})
\]

and some typical values of \( \Gamma_m \) have been calculated and collected in table 3.2.1.

Substituting all of parameters (3.2.14), (3.2.19), (3.2.23) and (3.2.27) into “SC” theory
(4.2.15,16) respectively, we obtained the following numerical results.

i) “Detailed balance”
\[
\begin{align*}
\psi_1(x) & = 263.2 \cdot x^{-0.20} / D(x) = 2.71 \cdot 10^{-3} \cdot x^{-0.20} \\
\psi_2(x) & = 333.2 \cdot x^{-0.52} / D(x) = 3.44 \cdot 10^{-3} \cdot x^{-0.52} \\
\end{align*}
\]
(4.2.17)
\[
\begin{align*}
x^2 f_1(x) & = (188 \cdot x^{0.2} + 21930) / D(x) = 0.226 \\
x^2 f_2(x) & = (21930 + 208 \cdot x^{0.04}) / D(x) = 0.226 \\
\end{align*}
\]
(4.2.18)

Here \( D(x) = 403 \cdot x^{0.2} + 96997 + 404 \cdot x^{0.08} = 96997. \)

Comparing (4.2.17,18) with (3.2.17,18) respectively, we can see that they are very close each other. Both of them reproduce the Monte Carlo simulation results well. Both (4.1.12,13) are tenable in this case.

ii) “Dominance”
\[
\begin{align*}
\psi_1(x) & = 2.5 \cdot x^{-0.20} / D(x) = 5.66 \cdot 10^{-5} \cdot x^{-0.12} \\
\psi_2(x) & = 333.3 \cdot x^{-0.6} / D(x) = 7.54 \cdot 10^{-3} \cdot x^{-0.52} \\
\end{align*}
\]
(4.2.19)
\[
\begin{align*}
x^2 f_1(x) & = (1.786 \cdot x^{0.2} + 208.3) / D(x) = 4.72 \cdot 10^{-3} \cdot x^{0.08} \\
x^2 f_2(x) & = (208.3 + 2193 \cdot x^{-0.08}) / D(x) = 0.496 \\
\end{align*}
\]
(4.2.20)

Here \( D(x) = 3.826 \cdot x^{0.2} + 905.7 + 44178 \cdot x^{-0.08} = 44178 \cdot x^{-0.08}. \)

Comparing (4.2.19,20) with (3.2.22a, 22b) respectively, we can see that \( \psi_2(x) \) and \( x^2 f_2(x) \) in both cases are close each other. \( \psi_1(x) \) and \( x^2 f_1(x) \) given here are even more accurate than (3.2.22a, 22b), because, (4.2.19,20) predicted slight different \( x \) dependences. Both (4.2.10,11) are tenable in this case.

iii) “Ignorance”
\[
\begin{align*}
\psi_1(x) & = 0.025 \cdot x^{-0.40} / D(x) \rightarrow 0.4982 \cdot x^{-0.20} \\
\psi_2(x) & = 0.02857 \cdot x^{-0.60} / D(x) \rightarrow 0.5694 \cdot x^{-0.40} \\
\end{align*}
\]
(4.2.21)
\[
\begin{align*}
x^2 f_1(x) & = (0.02083 \cdot x^{-0.2} + 1.7857 \cdot 10^{-4}) / D(x) \rightarrow 0.4151 \\
x^2 f_2(x) & = (1.7857 \cdot 10^{-4} + 0.01880 \cdot x^{-0.08}) / D(x) \rightarrow 0.3747 \cdot x^{0.18} \\
\end{align*}
\]
(4.2.22)
Here  \( D(x) = 0.05018 \cdot x^{-0.2} + 7.2887 \cdot 10^{-4} + 0.03787 \cdot x^{-0.08}. \)

Unfortunately, formulas (4.2.21,22) couldn’t reproduce the Monte Carlo simulation results in the energy window \((10^0 \geq x \geq 10^{-4}).\) The reason of the failure is that the simulated particle energy spectrums do not satisfy the basic symmetric property of asymptotic solutions (4.1.4), or the Monte Carlo simulation has not reached the asymptotic state yet. In a much lower energy window, for example \((10^{-10} \geq x \geq 10^{-14}),\) formulas (4.2.21,22) should predict the asymptotic behavior of particle spectra well. But, the authors of [23] had never published any simulation results in the energy window yet. Both (4.2.10, 12) should be tenable in this case.

iv) “Ignorance & Detailed balance”

\[
\begin{align*}
7 \cdot 3 \cdot 10^9 
&= \frac{250 \cdot x^{-0.4}}{D(x)} = 3.43 \cdot 10^{-3} \cdot x^{-0.4} \\
7 \cdot 3 \cdot 10^8 
&= \frac{286 \cdot x^{-0.6}}{D(x)} = 3.92 \cdot 10^{-3} \cdot x^{-0.6} \\
D(x) &= 501.8 \cdot x^{-0.2} + 72887 + 378.7 \cdot x^{-0.08}.
\end{align*}
\]

Comparing (4.2.23) with (3.2.29), we can see that they are very close each other. Both of them reproduce the Monte Carlo simulation results well.

In the energy window \((10^0 \geq x \geq 10^{-4}),\) we can see

\[
D(x) = 501.8 \cdot x^{-0.2} + 72887 + 378.7 \cdot x^{-0.08} = 72887, \quad \text{thus,}
\]

(4.1.12) is tenable, therefore, this case belongs to “Detailed balance”. But, in a much lower energy window \((10^{-20} \geq x \geq 10^{-24}),\) we can see

\[
D(x) \rightarrow 501.8 \cdot x^{-0.2}, \quad \text{thus,}
\]

\[
\begin{align*}
\frac{x \psi_1(x)}{D(x)} &= 250 \cdot x^{-0.4} \rightarrow 0.4982 \cdot x^{-0.20} \\
\frac{x \psi_2(x)}{D(x)} &= 286 \cdot x^{-0.6} \rightarrow 0.5699 \cdot x^{-0.40}
\end{align*}
\]

which is almost equivalent to (4.2.21) or (4.2.10). Therefore, this case belongs to “Dominance”. However, the authors of [23] have never published any simulation results in the energy window.

In conclusion, our calculations here demonstrated that (4.2.6,7) can generate all asymptotic formulae including some solutions never been simulated yet.
4.2.4 CONSTRUCTING “SC” THEORY WITH EXPRESSIONS

In view of the success of “SC” theory for particle and recoil density, it's possible to build up all approximate asymptotic solutions for arbitrary cross sections based on the asymptotic solutions (3.3.1-11). We named all of these “build up” solutions “SC” theory in this work. “SC” theory consists of two steps: The first step — "S", is Separating approximation. We use \( Q_i(E)A_{ij}(X)dX \) to approximate arbitrary cross sections \( d\sigma_y(E, T) \), i.e. (3.1.1) in all equations Eqs. (2.3.2-4), (2.3.7) and (2.3.9-12), even if \( d\sigma_y(E, T) \) may not be separated, and then solve these equations for asymptotic solutions (3.3.1-11). The second step — "C", is Combining approximation. Using \( d\sigma_y(E, T) \) to replace \( Q_i(E)A_{ij}(X)dX \) in asymptotic solutions (3.3.1-11), we obtained all approximately asymptotic solutions for arbitrary cross sections for \( E >> E_0 \) or \( U \).

\[
\psi_j(E_0) = E E_0^{-2} \Pi_j(E_0) / N \\
F_j(E_0) = E E_0^{-3} \sum_k \Pi_k(E_0) \alpha_j S_{kj}(E_0),
\]

\[
F_{(ij)}(U) = E \left[ \alpha_j \sum_k \Pi_k(U) \sigma_{kj}^t(U) + \Pi_j(U) \sum_k \alpha_k \sigma_{jk}^a(U) \right]
\]

\[
\psi_j^i(E_0) = P_i \left( N E_0 P_{ij} \right)^{-1} \Pi_j(E_0)
\]

\[
P_{(ij)}(U) = P_i \left[ \alpha_j \sum_k \Pi_k(U) U_{kj} \sigma_{kj}^t(U) + \Pi_j(U) \sum_k \alpha_k \sigma_{jk}^a(U) \right] \ldots,
\]

where \( D_{ij}^L(E_0) \) is the element \( [i, j] \) of the determinant \( [D^L(E_0)] \) and \( [D^L(E_0)]_{ij} \) is the algebraic cofactor for element \( [i, j] \) for \( L = 0, 1, \)

\[
\begin{cases}
E_0 \bar{D}_{ij}^L(E_0) = -y^{-0.5L} \alpha_j S_{ij}^t(E_0), \text{ for } i \neq j \\
E_0 \bar{D}_{ij}^L(E_0) = \sum_k U_{ik}^L \alpha_k S_{kj}(E_0)
\end{cases}
\]

\[
\Pi_j^L(E) = [D^L(E)]_{kk} / [D^L(E)]
\]

\[
\sigma_{ij}^a(U) = \sigma_{ij}^t(U) U_{ij} \sqrt{M_i / M_j};
\]
\[ \sigma_{ij}^{(3)}(E) = - \int_{y}^{T_{y}} [1 - (T/E)U_{ij}] \ln[1 - (T/E)]d\sigma_{ij}(E, T) > 0 \]

\[ \tilde{\sigma}_{ij}^{(1)}(E) = \sigma_{ij}^{(3)}(E) + \sigma_{ij}^{(4)}(E) \]

\[ \tilde{\sigma}_{ij}^{(4)}(E) = \sum_{j} \alpha_{j} \tilde{\sigma}_{ij}(E). \]

\[ \sum_{k} \prod_{k}^{l}(E)\sigma_{ij}^{(4)}(E) = 1 \quad (4.2.30) \]

(4.2.6,7) are the special cases of (4.2.25,26) respectively. Direct summation of (4.2.27,29) and using (4.2.30) yielded

\[ \sum_{j} F_{ij}(U) = E \quad (4.2.31) \]
\[ \sum_{j} P_{ij}(U) = \sqrt{2M_{i}E} \quad (4.2.32) \]

Substituting (3.3.1) into (3.3.5,11) respectively, we have

\[ F_{ij}(U) = N \sum_{k} \left[ \alpha_{k} \sigma_{jk}^{(3)}(U)U^{2}\psi_{j}(U) + \alpha_{j} \sigma_{ij}^{(4)}(U)U^{2}\psi_{k}(U) \right] \quad (4.2.33) \]
\[ P_{ij}(U) = N \sum_{k} \left[ \sqrt{2M_{j}} \cdot \alpha_{k} \sigma_{jk}^{(3)}(U)U^{3/2}\psi_{j}^{(1)}(U) + \sqrt{2M_{k}} \cdot \alpha_{j} \sigma_{ij}^{(4)}(U)U^{3/2}\psi_{k}^{(1)}(U) \right] \quad (4.2.34) \]

Substituting (4.2.33,34) into (4.2.31,32) respectively, we have

\[ NU^{2} \sum_{k} [\tilde{\sigma}_{k}(U)\psi_{k}(U)] = E \quad (4.2.35) \]
\[ NU^{1.5} \sum_{k} [\tilde{\sigma}_{k}^{(1)}(U)\sqrt{2M_{k}}\psi_{k}^{(1)}(U)] = \sqrt{2M_{i}E} \quad (4.2.36) \]

In addition, (3.3.14,15) are approximately asymptotic correct for arbitrary cross sections and can be used as the conditions of detailed balance [13].

We should point out that all of above statistical distribution functions and relations including (3.3.14,15) are approximately asymptotic \((E >> U)\) correct for arbitrary cross sections. However, if the scattering cross section is separable i.e. (3.1.1), each of them reduced to the corresponding one derived in previous chapter and became asymptotic \((E >> U)\) correct. (4.2.6). (4.2.31) and (4.2.33) have been derived by “VUC” by using a much more complicate methods (so-called age theory [26]) and published in [13].
4.3 “VCU” THEORY

4.3.1 INTRODUCTION TO “VCU” THEORY

In 1993, M. Vicanek, U. Conrad and H. M. Urbassek (called “VCU” in this work) proposed a general method to study energy distributions of recoil atoms in collision cascades in composite media [13]. According to “VCU”, Eqs. (2.3.5) \((L = 0)\) can be reduced to computationally much simpler systems of differential equations for arbitrary interaction cross sections,

\[
\begin{aligned}
\left[ N \sum_k \left[ - \alpha_k S_{jk}(U) \psi_j(U) + \alpha_j S_{kj}(U) \psi_k(U) \right] \right. \\
+ \left. \frac{d}{dU} \left[ \alpha_k \sigma_{jk}^0(U) U^2 \psi_j(U) + \alpha_j \sigma_{kj}^0(U) U^2 \psi_k(U) \right] \right] \\
+ \delta_{ij} U \delta(U - E) = 0
\end{aligned}
\]  

(4.3.1)

with the “obvious” initial condition,

\[
\psi_j(U > E) = 0.
\]  

(4.3.2)

Adding “\(j\)” up in (4.3.1), one obtained

\[
\left. \frac{d}{dU} \left( N \sum_j [\tilde{\sigma}_j(U) U^2 \psi_j(U)] \right) \right] + U \delta(U - E) = 0
\]

Integrating it and using the initial condition (4.3.2), one got

\[
\sum_j [\tilde{\sigma}_j(U) \psi_j(U)] = E/(NU^2)
\]  

(4.3.3)

which is equivalent to (4.2.35). Replacing one of the equations of Eqs. (4.3.1), \((i = j)\), one obtained a set of equations which is equivalent to Eqs. (4.3.1),

\[
\begin{aligned}
\left[ N \sum_k \left[ - \alpha_k S_{jk}(U) \psi_j(U) + \alpha_j S_{kj}(U) \psi_k(U) \right] \right. \\
+ \left. \frac{d}{dU} \left[ \alpha_k \sigma_{jk}^0(U) U^2 \psi_j(U) + \alpha_j \sigma_{kj}^0(U) U^2 \psi_k(U) \right] \right] = 0, \text{ for } j \neq i.
\end{aligned}
\]  

(4.3.4)

Considering the “obvious” initial condition (4.3.2) and integrating \(U\) on the both sides of Eqs. (4.3.1) over \((E-, E+)\), we obtained the initial condition (see Postulation of “VCU” theory),
Eqs. (4.3.4,5) is equivalent to “VCU” theory Eqs. (4.3.1,2) [13] and can be solved for arbitrary cross sections. We noticed that the strange initial condition (4.3.5) derived here means that $\psi_j(E)$ couldn’t be all positive. In general, Eqs. (4.3.4,5) may determine a unique solution. However, we have to keep our mind that $\psi_j(U)$ must approximately and asymptotically satisfy (3.3.14), if it really expresses approximate asymptotic fluxes.

### 4.3.2 “VCU” THEORY FOR A BINARY MEDIA

Specifically for a binary media, Eqs. (4.3.4,5) read

\[
\begin{align*}
\left\{ \frac{d}{dx} &\left[\left(1 - r_2\right)\phi_2(x) + r_1\phi_1(x)\right] - \frac{1}{x}s_2\phi_2(x) + \frac{1}{x}s_1\phi_1(x) = 0 \\
\phi_1(x) + \phi_2(x) = 1
\end{align*}
\] (4.3.6)

\[
\begin{align*}
\left[\left(1 - r_1(1)\right)\phi_1(1) + r_2(1)\phi_2(1) = 1 \\
r_1(1)\phi_1(1) + \left[1 - r_2(1)\right]\phi_2(1) = 0
\end{align*}
\] (4.3.7)

respectively. The notations here are the same in ref. [13]:

\[
x = U/E
\]

\[
\phi_j(x) = (N/E)\bar{\sigma}_j(U)U^2\psi_j(U), \quad (j = 1, 2)
\] (4.3.8)

\[
s_1(x) = \alpha_2 S_{12}(U)/[U\bar{\sigma}_1(U)], \quad r_1(x) = \alpha_2 \sigma_{12}(U)/\bar{\sigma}_1(U),
\]

\[
s_2(x) = \alpha_1 S_{21}(U)/[U\bar{\sigma}_2(U)], \quad r_2(x) = \alpha_1 \sigma_{21}(U)/\bar{\sigma}_2(U),
\]

Solving Eqs. (4.3.7), one obtained the real initial conditions for solving Eqs. (4.3.6),

\[
\begin{align*}
\left\{ \phi_1(1) &\equiv \left[1 - r_2(1)\right]/\left[1 - r_1(1) - r_2(1)\right] \\
\phi_2(1) &\equiv -r_1(1)/\left[1 - r_1(1) - r_2(1)\right]
\end{align*}
\] (4.3.9)

Changing variables from $(x, \phi_j)$ to $(\eta, \Gamma)$,

\[
\Gamma \equiv (1 - r_2) - (1 - r_1 - r_2)\phi_1 \quad \text{and} \quad \eta \equiv -\ln x.
\]

Eqs. (4.3.7,9) reduced to
\[\frac{df}{d\eta} = -[\alpha(f - 1 + r_2) + s_2]\]

\[f(0) = 0\]  \hspace{1cm} (4.3.10)

Here \(\alpha = \left( s_1 + s_2 \right)/(1 - r_1 - r_2) \neq 0\)  \hspace{1cm} (4.3.11)

Eqs. (4.3.12) can be solved exactly,

\[f(-\ln x) = 1 - h(x) \left[ 1 + \int_x^1 \frac{(\alpha r_2 + s_2)}{h(x')} dx' \right] \]  \hspace{1cm} (4.3.12)

Here \(h(x) = \exp\left[ -\int_x^1 \alpha dx'/x' \right]\). Thus,

\[
\begin{align*}
(1 - r_1 - r_2)\phi_1(x) &= 1 - r_2(x) - f(-\ln x) \\
(1 - r_1 - r_2)\phi_2(x) &= -r_1(x) + f(-\ln x)
\end{align*}
\]  \hspace{1cm} (4.3.13)

\[
\begin{align*}
E^{-1}F_{(ij)}(E, U) &= 1 - f(-\ln x); \\
E^{-1}F_{(ij)}(E, U) &= f(-\ln x).
\end{align*}
\]  \hspace{1cm} (4.3.14)

\[E^{-1}F_{(ij)}(E, U \rightarrow E) = \delta_{ij}.\]  \hspace{1cm} (4.3.15)

H. M. Urbassek et. al. published a different formula for the particle flux [13]:

\[
\begin{align*}
(1 - r_1 - r_2)\phi_1(x) &= [1 - r_2(1)]h(x) + h(x) \int_x^1 \left[ \frac{s_2}{x'} + \frac{dr_2}{dx'} \right] h(x') dx' \\
(1 - r_1 - r_2)\phi_2(x) &= -r_1(1)h(x) + h(x) \int_x^1 \left[ \frac{s_1}{x'} + \frac{dr_1}{dx'} \right] h(x') dx'
\end{align*}
\]  \hspace{1cm} (4.3.16)

which couldn’t satisfy \(\phi_1(x) + \phi_2(x) = 1\), therefore, (4.3.16) is incorrect.

If \(\alpha > 0\) or \((1 - r_1 - r_2) > 0\), (4.3.12) turns out to be

\[f = 1 - r_2 - s_2/\alpha = (s_1 + r_1s_2 - r_2s_1)/(s_1 + s_2), \quad \text{for } x \to 0,\]  \hspace{1cm} (4.3.17)

Thus, (4.3.13,14) reduced to

\[
\begin{align*}
\phi_1(x \to 0) &= s_2/(s_1 + s_2) \\
\phi_2(x \to 0) &= s_1/(s_1 + s_2)
\end{align*}
\]  \hspace{1cm} (4.3.18)

\[
\begin{align*}
E^{-1}F_{(ij)}(E, U \to 0) &= (s_1 + r_1s_2 - r_2s_1)/(s_1 + s_2); \\
E^{-1}F_{(ij)}(E, U \to 0) &= (s_2 + r_2s_1 - r_1s_2)/(s_1 + s_2).
\end{align*}
\]  \hspace{1cm} (4.3.19)

Substituting (4.3.18) into (4.3.6), we obtained “SC” solution (4.2.6). The condition of detailed balance (4.2.37) holds. Therefore, it’s not surprised to see that “VCU” solved Eqs. (4.3.1,2) and
found approximate asymptotic solutions for some scattering cross-sections\[26\].

If \( \alpha < 0 \) or \( (1 - r_1 - r_2) < 0 \), (4.3.12) only can generate \( f(\infty) \rightarrow \infty \), thus, for \( x \rightarrow 0 \), (4.3.13,14) turns out to be,

\[
\begin{align*}
\phi_1 (x \rightarrow 0) \cdot \phi_2 (x \rightarrow 0) &< 0 \\
\left\{ E^{-1}F_{(1)} (E, U \rightarrow 0) \rightarrow -\infty; \right. \\
\left. E^{-1}F_{(2)} (E, U \rightarrow 0) \rightarrow +\infty. \right. 
\end{align*}
\]

Therefore, both “VCU” equations (4.3.1,2) and the condition of detailed balance (4.2.37) fail.

### 4.3.3 COMPARING “VCU” THEORY WITH MONTE CARLO SIMULATION

For Kr-C potential interaction cross-section, directly solving Eqs. (4.3.10) numerically by the Runge–Kutta methods, or using the formula (4.3.12), we obtained \( f(-\ln x) \) for HfC system. According to (4.3.14), \( f(-\ln x) \) is the energy partition function. By using Kr-C potential interaction cross section (see Chapter 6), we have calculated \( f(-\ln x) \) for HfC bombarded by incident Hf or C atoms with energy 6 keV or 100 keV respectively. The calculated results agree with the correspondent curves in Fig. 1 of ref.[13] very well. Substituting \( f(-\ln x) \) into (4.3.13) then (4.3.8), we obtained particle fluxes. Again, the calculated results agree with the correspondent curves in Fig. 3 of ref.[13] very well too. These agreements showed that the “VCU” must use similar methods (4.3.10-15) to do their calculations. The all energy spectra of particle fluxes have been showed to agree with correspondent simulations very well. The SU Solutions (4.2.6) predicted the approximately asymptotic behavior of all energy spectra for HfC

For general power law cross sections (3.2.1), we derived and obtained,

\[
\begin{align*}
\sigma_j &= x^{-2m_{j1}} \cdot \alpha_1 \bar{C}_{j1} \big/ \left[ (l - M_{j1}) r_{m_{j1}} \right] + x^{-2m_{j2}} \cdot \alpha_2 \bar{C}_{j2} \big/ \left[ (l - M_{j2}) r_{m_{j2}} \right] \\
\left[ s_1 (x) = x^{-2m_{j1}} \cdot \alpha_2 \bar{C}_{j2} \big/ \left[ (l - M_{j2}) r_1 \right] \right] \\
\left[ s_2 (x) = x^{-2m_{j2}} \cdot \alpha_1 \bar{C}_{j1} \big/ \left[ (l - M_{j1}) r_2 \right] \right]
\end{align*}
\]
\[
\begin{align*}
    r_1(x) &= s_1(x)/(1 - m_{12}) \\
    r_2(x) &= s_2(x)/(1 - m_{21}) \\
    \psi_j(x) &= \phi_j(x)/\sigma_j,
\end{align*}
\] (4.3.24)

\[
\psi_j(x) = \phi_j(x)/\sigma_j,
\] (4.3.25)

In order to compare “VCU” theory with Monte Carlo Simulations [23], let’s follow the same procedure for each simulation: (1) Substituting all of parameters (3.2.14), (3.2.19), (3.2.23) and (3.2.27) into (4.3.23,24) for \( s_1(x), s_2(x), r_1(x) \) and \( r_2(x) \); (2) Solving Eqs. (4.3.10) and substituting the solution \( f(\ln x) \) into (4.3.13) for \( \phi_j(x) \); (3) substituting \( \phi_j(x) \) and \( \sigma_j(x) \) into (4.3.25) for \( \psi_j(x) \); (4) Plotting \( x\psi_j(x) \) as a function of \( x \) in Fig. 4.3.1 and directly comparing with the corresponding simulation.

i) “Detailed balance”

\[
\begin{align*}
    s_1(x) &= x^{-0.80} \cdot 83.333/\sigma_1 \\
    s_2(x) &= x^{-0.48} \cdot 65.789/\sigma_2 \\
    r_1(x) &= x^{-0.80} \cdot 138.89/\sigma_1 \\
    r_2(x) &= x^{-0.48} \cdot 86.565/\sigma_2 \\
    \sigma_1 &= 1.5305 \cdot x^{-0.66} + 200.71 \cdot x^{-0.80} \\
    \sigma_2 &= 132.54 \cdot x^{-0.48} + 1.2119 \cdot x^{-0.40}
\end{align*}
\]

Calculated curves (4.3.25) have been plotted in part (A) of Fig. 4.3.1. “VCU” theory couldn’t reproduce their Monte Carlo Simulations at all. One can see that both \( \psi_{12} \) and \( \psi_{21} \) “fly into the sky” and \( \psi_{11} \) and \( \psi_{22} \) “sink to the hell”, because “VCU” theory predicted \( \psi_{11} < 0 \) and \( \psi_{22} < 0 \) in the entire energy region \( 0 < x \leq 1 \).

ii) “Dominance”

\[
\begin{align*}
    s_1(x) &= x^{-0.80} \cdot 83.333/\sigma_1 \\
    s_2(x) &= x^{-0.40} \cdot 0.625/\sigma_2 \\
    r_1(x) &= x^{-0.80} \cdot 138.89/\sigma_1 \\
    r_2(x) &= x^{-0.40} \cdot 0.781/\sigma_2
\end{align*}
\]
\[
\begin{align*}
\sigma_1 &= 1.5305 \cdot x^{-0.60} + 200.71 \cdot x^{-0.80} \\
\sigma_2 &= 1.2119 \cdot x^{-0.40} + 132.54 \cdot x^{-0.48}
\end{align*}
\]

Calculated curves (4.3.25) have been plotted in part (B) of Fig. (4.3.1). It's easy to see that "VCU" theory can reproduce the Monte Carlo simulation results approximately, but, the deviation is serious in higher energy window. Particularly, "VCU" theory predicted $\psi_{21} < 0$ for $0.4973 < x \leq 1$ and $\psi_{12} < 0$ for $0.4258 < x \leq 1$.

iii) "Ignorance"

\[
\begin{align*}
    s_1(x) &= x^{-0.60} \cdot 83.333 \cdot 10^{-4} / \sigma_1 \\
    s_2(x) &= x^{-0.40} \cdot 0.00625 / \sigma_2 \\
    r_1(x) &= x^{-0.60} \cdot 138.89 \cdot 10^{-4} / \sigma_1 \\
    r_2(x) &= x^{-0.40} \cdot 0.00781 / \sigma_2 \\
    \sigma_1 &= 2.0071 \cdot x^{-0.80} + 0.017865 \cdot x^{-0.60} \\
    \sigma_2 &= 0.01212 \cdot x^{-0.40} + 1.3254 \cdot x^{-0.48}
\end{align*}
\]

Calculated curves (4.3.25) have been plotted in part (C) of Fig. (4.3.1). "VCU" theory can reproduce the Monte Carlo simulation results for $\psi_{11}$ and $\psi_{22}$, approximately reproduce the simulation results for $\psi_{12}$ and $\psi_{21}$. But, It's easy to see $\psi_{12} < 0$ for $0.2889 < x \leq 1$ and $\psi_{21} < 0$ for $0.3122 < x \leq 1$.

iv) "Ignorance & Detailed balance"

\[
\begin{align*}
    s_1(x) &= x^{-0.60} \cdot 71.428 / \sigma_1 \\
    s_2(x) &= x^{-0.40} \cdot 62.5 / \sigma_2 \\
    r_1(x) &= x^{-0.60} \cdot 102.04 / \sigma_1 \\
    r_2(x) &= x^{-0.40} \cdot 78.125 / \sigma_2 \\
    \sigma_1 &= 2.0071 \cdot x^{-0.80} + 153.05 \cdot x^{-0.60} \\
    \sigma_2 &= 121.19 \cdot x^{-0.40} + 1.3245 \cdot x^{-0.48}
\end{align*}
\]

Calculated curves (4.3.25) have been plotted in part (D) of Fig. (4.3.1). The situation is same as in the case "Detailed balance". "VCU" theory couldn't reproduce their Monte Carlo Simulations at all.
One can see that both $\psi_{12}$ and $\psi_{21}$ “fly into the sky” and $\psi_{11}$ and $\psi_{22}$ “sink to the hell”, because VCU theory predicted $\psi_{11} < 0$ and $\psi_{22} < 0$ in the entire energy region $0 < \chi \leq 1$.

![Normalized energy distributions](image)

**Fig. 4.3.1** Normalized energy distributions calculated by using “VCU” theory and simulation respectively. The general power cross-sections (3.2.1) with same parameters were used for both “VCU” theory and Monte Carlo Simulation. Thick line: Monte Carlo Simulation results [23].Thin line: Calculated by “VCU” theory (4.3.1,2), or (4.3.22–25). (A) and (D), “VCU” theory fails. (B) and (C) CU theory succeeds.

### 4.3.4 “VCU” ANALYTICAL THEORY

As “VCU” theory (4.3.25) is not analytical function of $\chi$, the conclusion may not very clear. In order to obtain the “VCU” analytical theory, we have to use the separable scattering cross sections (3.1.1), thus,

$$s_1 = \beta_{12}/(\beta_{11}\epsilon_{11} + \beta_{12}\epsilon_{12}) > 0; \quad s_2 = \beta_{21}/(\beta_{21}\epsilon_{21} + \beta_{22}\epsilon_{22}) > 0;$$

$$r_1 = -B_{12}/(\beta_{11}\epsilon_{11} + \beta_{12}\epsilon_{12}) > 0; \quad r_2 = -B_{21}/(\beta_{21}\epsilon_{21} + \beta_{22}\epsilon_{22}) > 0.$$

Considering all of $s_1, s_2, r_1, r_2$ and $\alpha$ are independent of $\chi$, solving Eqs. (4.3.10), we obtained

$$f(-\ln \chi) = (s_1 + s_2)^{-1}[s_1 - r_2s_1 + s_2r_1][1 - \chi^\alpha] \quad (4.3.26)$$
In the previous chapter, we have used conventional power cross sections (3.2.2) to approximate general power law cross sections (3.2.1) and derived some analytical expressions to explain Monte Carlo Simulations [23] successfully. Seeing that cross sections (3.2.2) belongs to (3.1.1), all of \( s_1, s_2, r_1, r_2 \) and \( \alpha \) are independent of \( x \),

\[
\begin{align*}
\phi_1(x) &= s_2/(s_1 + s_2) + cx^\alpha \\
\phi_2(x) &= s_1/(s_1 + s_2) - cx^\alpha
\end{align*}
\]

(4.3.27)

Here \( c = (1 - r_2)/(1 - r_1 - r_2) - s_2/(s_1 + s_2) \).

Substituting (4.3.28,29) into (4.3.26,27), then (4.3.14, 25), we obtained the energy partition \( \sigma_1 = x^{-2m_i} \cdot (\alpha_1 \tilde{C}_{i1} + \alpha_2 \tilde{C}_{i2}) / (\Gamma_{m_i} (1 - m_i)) \)

(4.3.30)

Substituting (4.3.28,29) into (4.3.26,27), then (4.3.14, 25), we obtained the energy partition \( f(-\ln x) \) and particle flux \( x\psi_j(x) \) respectively. Substituting \( \alpha_j = 0.5 \) and \( \tilde{C}_{ij} \) in (3.2.14), (3.2.19), (3.2.23) and (3.2.27) into \( x\psi_j(x) \) respectively for the individual simulation. All of results have been plotted Fig. 4.3.1.

i) “Detailed balance” — represented by point “A” in Fig.4.3.2

\[
\begin{align*}
(m_1 = 0.40; \quad m_2 = 0.24)
\end{align*}
\]

\[
\begin{align*}
x\psi_{11}(x) &= x\psi_1(x) \cdot (1 - 2.9555 \cdot x^\alpha) < 0 \\
x\psi_{12}(x) &= x\psi_2(x) \cdot (1 + 3.5326 \cdot x^\alpha) \\
x\psi_{21}(x) &= x\psi_1(x) \cdot (1 + 2.5790 \cdot x^\alpha) \\
x\psi_{22}(x) &= x\psi_2(x) \cdot (1 - 3.0827 \cdot x^\alpha) < 0
\end{align*}
\]

(4.3.31)

where \( \alpha = -2.7204 < 0; \ x\psi_2(x) \) and \( x\psi_2(x) \) are given by (3.2.17). (4.3.31) confirmed that both \( \psi_{12} \) and \( \psi_{21} \) “fly into the sky” and \( \psi_{11} \) and \( \psi_{22} \) “sink to the hell”.

ii) “Dominance” — represented by point “B” in Fig.4.3.2

\[
\begin{align*}
(m_1 = 0.40; \quad m_2 = 0.24)
\end{align*}
\]
\[
\begin{align*}
\psi_{11}(x) &= x\psi_1(x) \cdot (1 + 271.71 \cdot x^\alpha) \\
\psi_{12}(x) &= x\psi_2(x) \cdot (1 - 3.2482 \cdot x^\alpha) \\
\psi_{21}(x) &= x\psi_1(x) \cdot (1 - 2.7752 \cdot x^\alpha) \\
\psi_{22}(x) &= x\psi_2(x) \cdot (1 + 0.03318 \cdot x^\alpha)
\end{align*}
\] (4.3.32)

Here \( \alpha = 1.3489 > 0 \); \( \psi_2(x) \) and \( \psi_2(x) \) are given by (3.2.21). \( \psi_{12} < 0 \) for \( 0.4175 < x \leq 1 \); \( \psi_{21} < 0 \) for \( 0.4692 < x \leq 1 \).

iii) “Ignorance” — represented by point “C” in Fig.4.3.2

\[
\begin{align*}
\psi_{11}(x) &= x\psi_1(x) \cdot (1 + 0.8492 \cdot x^\alpha) \\
\psi_{12}(x) &= x\psi_2(x) \cdot (1 - 1.0120 \cdot x^\alpha) \\
\psi_{21}(x) &= x\psi_1(x) \cdot (1 - 1.0120 \cdot x^\alpha) \\
\psi_{22}(x) &= x\psi_2(x) \cdot (1 + 1.2100 \cdot x^\alpha)
\end{align*}
\] (4.3.33)

Here \( \alpha = 0.009147 > 0 \); \( \psi_2(x) \) and \( \psi_2(x) \) are given by (3.2.25). \( \psi_{12} < 0 \) for \( 0.2714 < x \leq 1 \); \( \psi_{21} < 0 \) for \( 0.2714 < x \leq 1 \).

iv) “Ignorance & Detailed balance”

\[
\begin{align*}
\psi_{11}(x) &= x\psi_1(x) \cdot (1 - 3.3111 \cdot x^\alpha) < 0 \\
\psi_{12}(x) &= x\psi_2(x) \cdot (1 + 3.6615 \cdot x^\alpha) \\
\psi_{21}(x) &= x\psi_1(x) \cdot (1 + 3.0782 \cdot x^\alpha) \\
\psi_{22}(x) &= x\psi_2(x) \cdot (1 - 3.4039 \cdot x^\alpha) < 0
\end{align*}
\] (4.3.34)

Here \( \alpha = -3.2625 < 0 \); \( \psi_2(x) \) and \( \psi_2(x) \) are given by (3.2.29). (4.3.34) confirmed that both \( \psi_{12} \) and \( \psi_{21} \) “fly into the sky” and \( \psi_{11} \) and \( \psi_{22} \) “sink to the hell”. Calculated curves (4.3.31-34) have been plotted in part (A), (B), (C) and (D) of Fig. 4.3.1 respectively to comparing with the corresponding “VCU” numerical theory, we can see that both curves agree very well. Therefore, “VCU” analytical theory confirmed the ridiculous “negative particle fluxes” or “sink to the hell”.
### 4.3.5 COMPARING “VCU” THEORY WITH SIGMUND THEORY

Introducing the new variables

\[ X \equiv \alpha_2 \tilde{C}_{12} / (\alpha_1 \tilde{C}_{11}), \quad Y \equiv \alpha_1 \tilde{C}_{21} / (\alpha_2 \tilde{C}_{22}), \quad \text{and} \quad W_i \equiv \Gamma_{m_i} / (1 - m_i), \]

zeros of \( d(s) \) defined by (3.2.7) have given by the following equation

\[
[(X + 1)\varepsilon_{11}(s) - 1] \cdot [(Y + 1)\varepsilon_{22}(s) - 1] = XY
\]  

(4.3.35)

Obviously, we know that \( s = 1 \) is the highest solution of Eqs. (4.3.35), then, the next highest solution \( s = 0 \) defines a function \( s(X, Y) = 0 \). At any point on the curve

\[
XY(W_1 + W_2 - 1) + (X - 1)W_1 + (Y - 1)W_2 = 1
\]  

(4.3.36)

we have \( r_1 + r_2 = 1 \) and \( \alpha = \pm \infty \). For \( m_1 = 0.40 \) and \( m_2 = 0.24 \), we have plotted \( s(X, Y) = 0 \) and (4.3.36) in Fig. (4.3.2). Two contour lines divided the entire region into three parts, \( I \cup II \cup III \). In region \( II \cup III \), (3.2.7) has no any positive zeros, except the highest zero \( s = 1 \). Therefore, Sigmund theory (3.2.9) is accurate enough to describe the energy spectra of particle flux. The case “Detailed balance” represented by point “A” is an example. Unfortunately, in region \( III \), due to \( r_1 + r_2 > 1 \) or \( \alpha < 0 \), “VCU” theory (4.3.10) fails. In region \( I \cup II \), “VCU” theory (4.3.10) can give an asymptotic description of the energy spectra of particle flux. In the region \( I \), we have consider the contribution from other poles in Sigmund theory for more accurate energy spectra, both cases “Dominance” and “Ignorance” are examples represented by points “B” and “C” respectively.

### 4.3.6 POSTULATION OF “VCU” THEORY

In view of both particle flux \( \psi_j(U) \) and energy partition \( F_{ij}(E, U) \) in (2.3.17) are exact without any approximation, we have
Fig. 4.3.2 Contour lines were calculated by (4.3.35) and (4.3.36). The power cross-sections (3.2.2) with $m_1 = 0.40$ and $m_2 = 0.24$ have been used for computations. In the region III, “VCU” theory (4.3.10) fails.

\[ F_{(ij)}(E, U = E-) = \delta_{ij}E \]  
\[ F_{(ij)}(E, U \ll E) = F_{(ij)}(U) \]
\[ = N \sum_k \left[ \alpha_k \sigma_{jk}^s(U)U^2 \psi_j(U) + \alpha_j \sigma_{kj}^i(U)U^2 \psi_k(U) \right] \]

For separable cross-section (3.1.1), (4.3.38) reduced to (3.3.5) which is exact asymptotic behavior and independent of $U$ and the type of incident particle. For arbitrary cross sections, (4.3.38) is approximate asymptotic and is relatively independent of $U$ and the type of incident particle. The postulation of “VCU” theory [13] is the following:

Formula (4.3.38) expresses the energy partition function in the entire energy region $E \geq U > 0$.

\[ F_{(ij)}(E, U) = N \sum_k \left[ \alpha_k \sigma_{jk}^s(U)U^2 \psi_j(U) + \alpha_j \sigma_{kj}^i(U)U^2 \psi_k(U) \right] \]

It’s obvious that $\psi_j(U)$ in (4.3.39) are related to the type of incident particle, i.e. no longer satisfy the basic symmetric property of asymptotic solutions (4.1.4). Therefore, “VCU” sacrificed accuracy of $\psi_j(U)$ to match the energy partition function in the energy region near the incident energy $E$. 

\[ \delta_{ij} = \begin{cases} 
1 & \text{if } i = j \\
0 & \text{otherwise} 
\end{cases} \]

\[ \psi_j(U) = \begin{cases} 
\text{asymptotic behavior} & \text{if } U \ll E \\
\text{exact behavior} & \text{if } E \geq U > 0 
\end{cases} \]
Considering the continuity of energy partition functions and substituting (4.3.37) into (4.3.39), “VCU” obtained the initial condition (4.3.5).

Substituting (4.3.39) into (2.3.17), “VCU” obtained a system of differential equations (4.3.1,2) or equivalent one (4.3.4,5). This is the origin of “VCU” theory (4.3.1,5). “VCU” derived it by using a very clumsy way [13].

Since \( \sigma^{s}_{jk}(E) > 0 \) and \( \sigma^{l}_{kj}(E) > 0 \), from the initial condition (4.3.5), we can see that at least one of \( \psi_{j}(E) \) must be negative in any case. If the negative particle flux \( \psi_{j}(E) \) is spread by “VCU” equation (4.3.1) over entire energy region, “VCU” theory “breaks down”.

In addition, following the previous procedure, we can make another postulation: the momentum partition function (4.2.34) holds in the entire energy region \( (E \geq U > 0) \),

\[
P_{(ij)}(E, U) = P_{(ij)}(U) = N \sum_{k} \left[ \sqrt{2M_j} \cdot \alpha_k \sigma^{sl}_{jk}(U) U^{3/2} \psi_{j}'(U) + \sqrt{2M_k} \cdot \alpha_j \sigma^{rl}_{kj}(U) U^{3/2} \psi_{k}'(U) \right].
\] (4.3.40)

Considering the continuity of momentum partition functions, we obtained the initial condition,

\[
P_{(ij)}(E, U = E-) = N \sum_{k} \left[ \sqrt{2M_j} \cdot \alpha_k \sigma^{sl}_{jk}(E) E^{3/2} \psi_{j}'(E) + \sqrt{2M_k} \cdot \alpha_j \sigma^{rl}_{kj}(E) E^{3/2} \psi_{k}'(E) \right] = \delta_{ij} \sqrt{2M_i E}
\] (4.3.41)

Substituting (4.3.40) into (2.3.18), we obtained a system of differential equations

\[
\begin{bmatrix}
N \sum_{k} \left[ -\alpha_k T_{jk}(U) \psi_{j}'(U) + \alpha_j T_{kj}(U) \psi_{k}'(U) \right] \\
+ \frac{d}{dE} N \sum_{k} \left[ \alpha_k \sigma^{sl}_{jk}(U) U^{3/2} \sqrt{2M_j} \psi_{j}'(U) + \alpha_j \sigma^{rl}_{kj}(U) U^{3/2} \sqrt{2M_k} \psi_{k}'(U) \right] = 0, \quad i \neq j \\
N U^{3/2} \sum_{k} \left[ \sigma^{l}_{kj}(U) \sqrt{2M_k} \psi_{k}'(U) \right] = \sqrt{2M_i E}, \quad i = j
\end{bmatrix}
\] (4.3.42)

(4.3.42) with the initial condition (4.3.41) may be called as expanded “VCU” theory which Could be solved for \( \psi_{j}'(E) \).
4.3.7 COMPARING “VCU” THEORY WITH “SU” THEORY

If the solutions $\psi_j^U(U)$ of Eqs. (4.3.4,5) and Eqs. (4.3.41,42) really expresses approximate asymptotic fluxes we are looking for, then, the solutions $\psi_j^U(U)$ must approximately and asymptotically satisfy (3.3.14,15) respectively. Thus, instead of Eqs. (4.3.4,42), we have

$$\sum_k [\psi_j(U)\alpha_k S_{jk}(U) - \psi_k(U)\alpha_j S_{kj}(U)] = 0, \quad j \neq i. \quad (4.3.43)$$

$$\sum_j [\tilde{\sigma}_j(U)\psi_j(U)] = E/(NU^2)$$

$$\sum_k [\psi_j^U(U)\alpha_k T_{jk}(U) - \psi_k^U(U)\alpha_j T_{kj}(U)] = 0, \quad i \neq j \quad (4.3.44)$$

$$NU^{3/2} \sum_k [\tilde{\sigma}_k(U)\sqrt{2M_k}\psi_k(U)] = \sqrt{2M_i E}, \quad i = j$$

Only “SC” solutions (4.2.25, 28) satisfy them. We have a verity of “SC” solutions for arbitrary cross sections. “SC” solutions are all analytic and much easy to use. Why we have make a large loop to go much more tortuous way to pick up the same apple?

4.4 CONCLUSION

We have solved the transport equations describing a collision cascade for general power-low cross section and obtained an approximate asymptotic solution. The analytical results given by H. M Urbassek et. in [23] are included as special cases.

Based on above solutions, we have proposed "SC" theory. This theory may yield the approximate asymptotic solutions for arbitrary scattering cross-section.

“VCU” theory and its applications have been discussed in some detail. The postulation of “VCU” theory has been revealed. Based on the postulation, “VCU” differential equations have been derived. Both success and failure originated from the postulation.

If “VCU” theory success, “SC” solution can approach “VCU” solution approximately and asymptotically. “SC” solution can give approximate and asymptotic description for all statistical distribution functions and relations, even if in the case “VCU” theory fails.
A pool of Monte Carlo simulations published by “UC” [23] to solve Eqs (2.3.2) for a general form of power cross sections (3.2.1) can be served to check the vanity of any analytical method solving transport equations. The final results for Sigmund analytical method, “UC” theory, “VCU” theory and “SC” theory (4.3.18) have been list in the Table 4.4.1. Immediately, one can see that Sigmund analytical method is the best one.

Therefore, Sigmund analytical method is a very powerful theory tool to solve the transport equations.

**Table 4.4.1** Comparison of four major analytical methods with simulations

<table>
<thead>
<tr>
<th>Analytical results</th>
<th>Sigmund theory</th>
<th>“UC” theory</th>
<th>“VCU” theory</th>
<th>“SC” theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>i) “Detailed balance”</td>
<td>Good</td>
<td>Good</td>
<td>Fail</td>
<td>Fail</td>
</tr>
<tr>
<td>ii) “Dominance”</td>
<td>Good</td>
<td>Good</td>
<td>Good</td>
<td>Good</td>
</tr>
<tr>
<td>iii) “Ignorance”</td>
<td>Good</td>
<td>Fail</td>
<td>Good</td>
<td>Good</td>
</tr>
<tr>
<td>iv) “Ignorance &amp; Detailed balance”</td>
<td>Good</td>
<td>Fail</td>
<td>Fail</td>
<td>Fail</td>
</tr>
</tbody>
</table>
CHAPTER 5
DEPOSITED ENERGY AND MOMENTUM
DEPTH DISTRIBUTIONS

5.1 LAPLACE TRANSFORM OF RECURRENT RELATION

Let’s specify \( Q_i(E) = E^{-2m} \), the scattering cross-sections (3.1.1) reduced to

\[
\text{d}\sigma_{ij}(E, T) = E^{-2m} A_{ij}(X) \text{d}X
\]  

(5.1.1)

and the moments equations (2.4.2) can be solved by using Sigmund method (see Chapter 3).

Introducing the new variables \( E = Ue^\omega \), \( T = Ue^\upsilon \) and taking the Laplace Transform on both sides of Eqs. (2.4.2) with respect to \( u \), we obtained a system of algebraic equations. Solving them rigorously yields the following recurrent relation exactly.

\[
\begin{align*}
\mathcal{F}_{ij}^0(s) &= \frac{U\delta_{i0}}{s - 1}, \\
\mathcal{F}_{ij}^0(s) &= \frac{\delta_{i1}\sqrt{2M_iU}}{3(s - 0.5)} \\
\mathcal{H}_{ij}^n(s) &= \frac{U^{2m}}{ND^n(s)} \sum_k [D^L(s)]_{ki} \cdot \Delta\mathcal{H}_{ij1}^n(s - 2m)
\end{align*}
\]  

(5.1.2)

The recurrent relations (5.1.2) determine Laplace Transform of moments \( \mathcal{H}_{ij1}^n(s) \). Except some obvious poles, two zeros \( D(s = 1) = 0 \) and \( D^L(s = 0.5) = 0 \) should be considered for the evaluation of inverse Laplace transform. In addition, a particular attention has to be paid on some higher order poles in order to remove the “spurious divergences” [36].

5.2 ASYMPTOTIC MOMENTS OF DEPOSITED ENERGY DEPTH DISTRIBUTIONS

5.2.1 IN THE CASE \( m \neq 0.25 \)

Considering the obvious scaling property of Eqs. (5.1.2) [37~39], it’s suitable to define

\[
\begin{align*}
F_{(i)}(z, \bar{u}, U) &= F_{(i)}^{[0]}(z, \bar{u}, U) + F_{(i)}^{[1]}(z, \bar{u}, U) + F_{(i)}^{[2]}(z, \bar{u}, U) \\
F_{(i)}^{n}(E, U) &= E \cdot [E^{2m}/N]^n \cdot A_{(i)}^{n}(s)
\end{align*}
\]  

(5.2.1)
Here \( F_{(i)L}^{[0]n} (E, U) = E \cdot [E^2/N]^n \cdot A_{(i)L}^{[0]n} \) \( (5.2.2) \)

\[ F_{(i)L}^{[1]n} (E, U) = E \cdot [E^2/N]^n \cdot [-(U/E)^{0.5+2m}] \cdot A_{(i)L}^{[1]n} \] \( (5.2.3) \)

\[ F_{(i)L}^{[2]n} (E, U) = E \cdot [E^2/N]^n \cdot [(U/E)^{4m}] \cdot A_{(i)L}^{[2]n} \] \( (5.2.4) \)

Thus \( A_{(i)L}^{[n]} = A_{(i)L}^{[0]n} - [A_{(i)L}^{[1]n} - (U/E)^{2m-0.5}] \cdot A_{(i)L}^{[2]n}, (U/E)^{0.5+2m} \) \( (5.2.5) \)

For \( n = 1, (5.1.2) \) reads

\[ \tilde{F}_{(i)}^{[1]} (s) = \frac{U^{1+2m}}{3N[s-(1+2m)]D^1(s)} \cdot \sum_k[D^1(s)]_{ki} \]

The first two leading terms arise from two simple poles \( s = 1 + 2m \) and \( s = 0.5 \), we obtained,

\[ A_{(i)}^{[1]} = \frac{1}{2} \cdot \sum_k \Pi_{ki} (1 + 2m) \] \( (5.2.6) \)

\[ A_{(i)}^{[1]} = \frac{1}{2} \cdot (0.5 + 2m)^{-1} \cdot \sum_k \sqrt{M_i/M_k} \] \( (5.2.7) \)

For \( n = 2, L = 0, (5.1.2) \) reads

\[ \tilde{F}_{(i)}^{[2]} (s) = \frac{2U^{1+4m}}{N^2 D^2(s)} \cdot \sum_k[D^2(s)]_{ki} \cdot \left[ \frac{A_{(i)}^{[0][k]l} - A_{(i)}^{[1][k]l}}{s-(1+4m)} - \frac{A_{(i)}^{[1][k]l}}{s-(0.5+2m)} \right] \] \( (5.2.8) \)

The first three leading terms of \( (5.2.8) \) arise from three simple poles \( s = (1+4m), (0.5+2m) \) and \( s = 1 \), we obtained,

\[ A_{(i)}^{[1]} = 2 \cdot \sum_k \Pi_{ki} (1 + 4m)A_{(i)}^{[0][k]l} \] \( (5.2.9) \)

\[ A_{(i)}^{[2]} = 2 \cdot \sum_k \Pi_{ki} (0.5 + 2m)A_{(i)}^{[0][k]l} \] \( (5.2.10) \)

\[ A_{(i)}^{[1]} = 2 \cdot \sum_k \Pi_{k} \cdot [A_{(i)}^{[0][k]l} /(-4m) + A_{(i)}^{[1][k]l} / (2m-0.5)] \] \( (5.2.11) \)

For \( n = L = 2, (5.1.2) \) reads

\[ \tilde{F}_{(i)}^{[2]} (s) = \frac{4}{5} \frac{U^{1+4m}}{N^2 D^2(s)} \cdot \sum_k[D^2(s)]_{ki} \cdot \left[ \frac{A_{(i)}^{[0][k]l} - A_{(i)}^{[1][k]l}}{s-(1+4m)} - \frac{A_{(i)}^{[1][k]l}}{s-(0.5+2m)} \right] \]

The first two leading terms arise from two simple poles \( s = (1 + 4m) \) and \( (0.5 + 2m) \), we obtained

\[ A_{(i)}^{[0][k]l} = 0.8 \cdot \sum_k \Pi^2_{ki} (1 + 4m)A_{(i)}^{[0][k]l} \] \( (5.2.12) \)
\[ A_{(i)2}^{[1]} = 0.8 \cdot \sum_k \Pi_{ki}^2 (0.5 + 2m) A_{(k)1}^{[0]} \] (5.2.13)

In general, for \( n \geq 3 \), only considering the first three terms and substituting (5.2.2-4) into (5.2.1), immediately, we have

\[ F_{(i)2}^n (E, U) = N^{-n} \left[ E^{1+2mn} \cdot A_{(i)2}^{[0]} - U^{0.5+2m} \cdot E^{0.5+2m(n-1)} \cdot A_{(i)1}^{[n]} + U^{4m} \cdot E^{1+2m(n-2)} \cdot A_{(i)2}^{[2n]} \right] \] (5.2.14)

Obviously, the Laplace Transform of (5.2.14) is given by

\[ \tilde{F}_{(i)2}^n (s) = \frac{U^{1+2mn}}{N^n} \cdot \frac{s - (1 + 2mn)}{s - [0.5 + 2m(n - 1)]} \]
\[ - \frac{s - [1 + 2m(n - 2)]}{s - [0.5 + 2m(n - 1)]} \] (5.2.15)

Substituting (5.2.15) into (5.1.2) yielded,

\[ \tilde{F}_{(i)2}^n (s) = \frac{U^{1+2mn}}{N^n D^L (s)} \cdot \sum_k [D^L (s)]_{ki} \cdot \frac{\Delta A_{(k)2}^{[0]n}}{s - (1 + 2mn)} \]
\[ - \frac{\Delta A_{(k)1}^{[2]n}}{s - [0.5 + 2m(n - 1)]} + \frac{\Delta A_{(k)2}^{[2]n}}{s - [1 + 2m(n - 2)]} \] (5.2.16)

The three leading terms arise from the simple poles \( s = (1 + 2mn), [0.5 + 2m(n - 1)] \) and \([1 + 2m(n - 2)]\), we obtained,

\[ A_{(i)2}^{[0]n} = \sum_k \Pi_{ki}^L (1 + 2mn) \Delta A_{(k)2}^{[0]n} \] (5.2.17)

\[ A_{(i)1}^{[2]n} = \sum_k \Pi_{ki}^L [0.5 + 2m(n - 1)] \Delta A_{(k)1}^{[2]n} \] (5.2.18)

\[ A_{(i)2}^{[2]n} = \sum_k \Pi_{ki}^L [1 + 2m(n - 2)] \Delta A_{(k)2}^{[2]n} \] (5.2.19)

Putting (5.2.6,7), (5.2.9-13) and (5.2.17-19) together, we obtained three recurrent relations:

\[
\begin{align*}
A_{(i)2}^{[0]} &= \delta_{L0} \cdot A_0 \\
A_{(i)1}^{[2]} &= \sum_k \Pi_{ki}^L (1 + 2mn) \cdot \Delta A_{(k)1}^{[0]} \quad \text{for} \ n > 0 \\
A_{(i)2}^{[2]} &= \delta_{L1} \cdot A_1 \\
A_{(i)1}^{[2]} &= \sum_k \Pi_{ki}^L [0.5 + 2m(n - 1)] \cdot \Delta A_{(k)1}^{[2]} \quad \text{for} \ n > 1
\end{align*}
\] (5.2.20)
\[
\begin{aligned}
A_{(i)_{L}}^{[2]_{2}} & = \delta_{L0} \cdot A_{2} \\
A_{(i)_{L}}^{[2]_{n}} & = \sum_{k} \Pi_{k}^{1} [1 + 2m(n - 2)] \cdot \Delta A_{(k)_{L}}^{[2]_{n}}, \quad \text{for } n > 2 \\
\end{aligned}
\]

Where
\[
\begin{aligned}
A_{0} & = 1 \\
A_{1} & = \frac{1}{3} (0.5 + 2m)^{-1} \cdot \sum_{k} \Pi_{k}^{1} \sqrt{M_{1}/M_{k}} \\
A_{2} & = 2 \sum_{k} \Pi_{k}^{1} \cdot [A_{(k)_{L}}^{[0]}/(-4m) + A_{(k)_{L}}^{[1]}/(2m - 0.5)] \\
\end{aligned}
\]

Obviously, the non-vanishing moments \( A_{(i)_{L}}^{[\mu]_{n}} \) are
\[
\begin{aligned}
\mu = 0 : & \quad A_{0}^{0}, A_{1}^{1}, A_{2}^{2}, A_{3}^{3}, A_{4}^{4}, A_{5}^{5}, \ldots; \\
\mu = 1 : & \quad A_{0}^{1}, A_{0}^{1}, A_{2}^{2}, A_{3}^{3}, A_{4}^{4}, A_{5}^{5}, \ldots; \\
\mu = 2 : & \quad A_{0}^{2}, A_{1}^{2}, A_{0}^{1}, A_{2}^{3}, A_{3}^{3}, A_{4}^{4}, A_{5}^{5}, \ldots; \\
\end{aligned}
\]

In addition, two simple poles in (5.2.8) \( s = 0.5 + 2m \) and \( s = 1 \) turn out to be one double pole in the case \( m = 0.25 \). If treat the double pole as two simple poles, we will meet singularity. For example, the inverse Laplace transform of
\[
\tilde{f}(s) = [(s - x)(s - y)]^{-1}
\]
is given by
\[
f(E, U) = (x - y)^{-1}[(U/E)^{x} - (U/E)^{y}] \quad \text{for } x \neq y
\]
due to two simple poles \( s = x \) and \( s = y \). We can see that \( x = y \) is a singularity. On the other hand, if \( x = y \), these two simple poles turn out to be one double pole \( s = x \), we know that the inverse Laplace transform of
\[
\tilde{f}(s) = (s - x)^{-2}
\]
is
\[
f(E, U) = (U/E)^{x} \ln(U/E)
\]
which also can be derived by limit process,
\[
\lim_{y \to x} f(E, U) = (U/E)^{x} \lim_{y \to x} \left( (x - y)^{-1} \left[ 1 - (U/E)^{y-x} \right] \right)
\]

Thus, the \( x = y \) is only a removable singularity [36]. However, \( \ln(U/E) \) term will be spread out
to other moments to cancel all of singularities, such as “renormalization” in Quantum Electrodynamics [90]. Therefore, this term will certainly make some changes in the structure of recurrent relations (5.2.20-22). Naturally, \( \ln(U/E) \) term also appears in the expressions of moments of deposited momentum depth distribution in the case \( m = 0.25 \).

### 5.2.2 IN THE CASE \( m = 0.25 \)

In order to avoid the obvious singularities in (5.2.10,11), let’s redefine \( F_{(i)L}^n(E) \) as following

\[
F_{(i)L}^n(E, U) = E \cdot \left[ \frac{E^{2m}}{N} \right]^n \cdot B_{(i)L}^n
\]

Here \( F_{(i)L}^{[0]}(E, U) = E \cdot \left[ \frac{E^{2m}}{N} \right]^n \cdot B_{(i)L}^{[0]} \)

\[
F_{(i)L}^{[1]}(E, U) = E \cdot \left[ \frac{E^{2m}}{N} \right]^n \cdot \left[ -\left( \frac{U}{E} \right)B_{(i)L}^{[1]} \right]
\]

\[
F_{(i)L}^{[2]}(E, U) = E \cdot \left[ \frac{E^{2m}}{N} \right]^n \cdot \left[ -\left( \frac{U}{E} \right)\ln(U/E)B_{(i)L}^{[2]} \right]
\]

Thus \( B_{(i)L}^n = B_{(i)L}^{[0]} - \left[ B_{(i)L}^{[1]} + \ln(U/E) \cdot B_{(i)L}^{[2]} \right] \cdot \left( \frac{U}{E} \right) \)

For \( n = 1 = 1 \), (5.2.6,7) reduced to

\[
B_{(i)L}^{[1]} = \frac{1}{3} \sum_k \Pi_k (1.5)
\]

\[
B_{(i)L}^{[2]} = \frac{1}{3} \sum_k \Pi_k^1 \cdot \sqrt{M_i/M_k}
\]

For \( n = 2 \), \( L = 0 \), (5.2.8) reads

\[
\tilde{F}_{(i)0}^2(s) = \frac{2U^{1+4m}}{N^2D(s)} \sum_k [D(s)]_{kl} \left[ \frac{A_{(k)L}^{[0]}}{(s-2)} - \frac{A_{(k)L}^{[1]}}{(s-1)} \right]
\]

The three leading terms arise from two simple poles \( s = 2 \) and one double poles \( s = 1 \), we obtained,

\[
B_{(i)0}^{[0]} = 2\sum_k \Pi_k (2)B_{(k)i}^{[0]}
\]

\[
B_{(i)0}^{[1]} = 2\sum_k \left[ \Pi_k B_{(k)i}^{[0]} + \Sigma_k B_{(k)i}^{[1]} \right]
\]

\[
B_{(i)0}^{[2]} = 2\sum_k \Pi_k \cdot B_{(k)i}^{[2]}
\]

Here, \( \Sigma_k \equiv \lim_{s \to 1} [d((s-1)\Pi_k(s))/ds] \).
For $n = L = 2$, (5.2.12,13) read

$$B_{(i)L}^{(1)2} = 0.8 \cdot \sum_{k} \Pi_{ki}^2 (2) B_{(k)L}^{(0)j}$$  \hspace{1cm} (5.2.34)

$$B_{(i)L}^{(1)2} = 0.8 \cdot \sum_{k} \Pi_{ki}^2 (1) B_{(k)L}^{(1)j}$$  \hspace{1cm} (5.2.35)

In general, for $n \geq 3$, only considering the first three terms and substituting (5.2.25-27) into (5.2.24), immediately, we have

$$F_{(i)L}^n (E, U) = N^{-n}[E^{1+0.5n} \cdot B_{(i)L}^{(0)n} - U \cdot E^{0.5n} \cdot B_{(i)L}^{(1)n} - U \cdot E^{0.5n} \cdot \ln(E/U)B_{(i)L}^{(2)n}]$$

Obviously, its Laplace Transform is given by

$$\bar{F}_{(i)L}^n(s) = \frac{U^{1+0.5n}}{N^n} \cdot \left[ \frac{B_{(i)L}^{(0)n}}{s - (1 + 0.5n)} - \frac{B_{(i)L}^{(1)n}}{s - 0.5n} - \frac{B_{(i)L}^{(2)n}}{(s - 0.5n)^2} \right]$$  \hspace{1cm} (5.2.36)

Substituting (5.2.36) into (5.1.2) yielded,

$$\bar{F}_{(i)L}^n(s) = \frac{U^{1+0.5n}}{N^n D_L(s)} \cdot \sum_{k} D_L(s)_{ki} \cdot \left[ \frac{\Delta B_{(k)L}^{(0)n}}{s - (1 + 0.5n)} - \frac{\Delta B_{(k)L}^{(1)n}}{s - 0.5n} - \frac{\Delta B_{(k)L}^{(2)n}}{(s - 0.5n)^2} \right]$$

The three leading terms arise from two simple poles $s = (1 + 0.5n)$, $s = 0.5n$ and one double pole $s = 0.5n$, we obtained,

$$B_{(i)L}^{(0)n} = \sum_{k} \Pi_{ki}^L (1 + 0.5n) \cdot \Delta B_{(k)L}^{(0)n}$$  \hspace{1cm} (5.2.37)

$$B_{(i)L}^{(1)n} = \sum_{k} [\Pi_{ki}^L (0.5n) \cdot \Delta B_{(k)L}^{(1)n} + \Sigma_{ki}^L (0.5n) \cdot \Delta B_{(k)L}^{(2)n}]$$  \hspace{1cm} (5.2.38)

$$B_{(i)L}^{(2)n} = \sum_{k} \Pi_{ki}^L (0.5n) \cdot \Delta B_{(k)L}^{(2)n}$$  \hspace{1cm} (5.2.39)

Here, $\Sigma_{ki}^L (s) \equiv d\Pi_j^L (s)/ds$.

Putting (5.2.29-35) and (5.2.37-39) together, we obtained three recurrent relations:

$$B_{(i)L}^{(0)n} = [A_{(i)L}^{(0)n}]_{n=0.25}$$  \hspace{1cm} (5.2.40)

$$B_{(i)L}^{(1)n} = \delta_{L1} \cdot \frac{1}{3} \sum_{k} \Pi_{ki}^L \sqrt{M_i/M_k}$$

$$B_{(i)L}^{(12)} = \delta_{L2} \cdot 2 \sum_{k} [\Pi_{ki} B_{(k)L}^{(2)n} + \Sigma_{ki} B_{(k)L}^{(1)n}] + \delta_{L2} \cdot 0.8 \sum_{k} [\Pi_{ki}^2 (1)B_{(k)L}^{(1)n}]$$  \hspace{1cm} (5.2.41)

$$B_{(i)L}^{(1n)} = \sum_{k} [\Pi_{ki}^L (0.5n) \cdot \Delta B_{(k)L}^{(1)n} + \Sigma_{ki}^L (0.5n) \cdot \Delta B_{(k)L}^{(2)n}]$$, for $n > 2$
\[ \begin{align*}
B_{(i)L}^{[2]n} &= \delta_{L0} \cdot 2 \sum_k \Pi^k \cdot B_{(k)L}^{[1]n} \\
B_{(i)L}^{[2]n} &= \sum_k \Pi^k \cdot (0.5n) \cdot \Delta B_{(k)L}^{[2]n}, \quad \text{for } n > 2
\end{align*} \] (5.2.42)

The non-vanishing moments \( B_{(i)L}^{[n]n} \) are same as \( A_{(i)L}^{[n]n} \) given by (5.2.23). No singularities can be seen in (5.2.40-42).

### 5.2.3 CONTINUITY AT \( m = 0.25 \)

**Statement:** \( \lim_{m \to 0.25} A_{(i)L}^{n} = B_{(i)L}^{n} \) (5.2.43)

**Proof:** First of all, we noticed the following relations,

\[ \lim_{m \to 0.25} A_{(i)L}^{[0]n} = B_{(i)L}^{[0]n} \] (5.2.44)

\[ \lim_{m \to 0.25} A_{(i)L}^{[1]k} = B_{(i)L}^{[1]k}, \quad k = 1, 2 \] (5.2.45)

\[ \lim_{m \to 0.25} (2m - 0.5)A_{(i)L}^{[2]1} = B_{(i)L}^{[2]1} \] (5.2.46)

\[ \lim_{m \to 0.25} [A_{(i)L}^{[1]2} - A_{(i)L}^{[2]2}] = 2 \sum_k [\Pi^k B_{(k)L}^{[0]} + \Sigma_k B_{(k)L}^{[1]}] = B_{(i)L}^{[2]} \] (5.2.47)

By using (5.2.46), comparing (5.2.42) with (5.2.22), we can see

\[ \lim_{m \to 0.25} (2m - 0.5)A_{(i)L}^{[2]n} = B_{(i)L}^{[2]n} \] (5.2.48)

Then, let’s define

\[ R_{(i)L}^{n} \equiv \lim_{m \to 0.25} [A_{(i)L}^{[1]n} - (U/E)^{(0.5-2m)} \cdot A_{(i)L}^{[2]n}] \] (5.2.49)

\[ \overline{R}_{(i)L}^{n} \equiv B_{(i)L}^{[1]n} + \ln(U/E) \cdot B_{(i)L}^{[2]n} \] (5.2.50)

and proof

\[ R_{(i)L}^{n} = \overline{R}_{(i)L}^{n} \] (5.2.51)

by induction

i) For \( n = 1 \), (5.2.45) means

\[ R_{(i)k}^{k} = B_{(i)k}^{[1]k} = \overline{R}_{(i)k}^{k}, \quad k = 1, 2 \] (5.2.52)

For \( n = 2 \), using (5.2.46,57), we obtained,
\[ \mathbf{R}^2 (i)_{0} = \lim_{m \to 0,25} \left\{ \mathbf{A}^{(1)_{i0}} - \mathbf{A}^{(2)_{i0}} + [1 - (\mathbf{U}/\mathbf{E})^{0.5 - 2m}] \cdot \mathbf{A}^{(2)_{i0}} \right\} = \mathbf{B}^{(1)_{i0}} + \ln(\mathbf{U}/\mathbf{E}) \cdot \mathbf{B}^{(2)_{i0}} = \bar{\mathbf{R}}^2 (i)_{0} \] \quad (5.2.53)

ii) For \( n-1 \rightarrow n \), let’s assume, 

\[ \mathbf{R}^{n-1}_{(i)L} = \bar{\mathbf{R}}^{n-1}_{(i)L} \] \quad (5.2.54)

Substituting (5.2.21,22) into (5.2.49), we obtained, 

\[ \mathbf{R}^{n}_{(i)L} = \sum_{k} \lim_{m \to 0.25} \left\{ \Pi^{(1)_{i}}_{kL} [0.5 + 2m(n - 1)] \cdot \Delta \mathbf{A}^{(1)_{i}}_{(kL)} - (\mathbf{U}/\mathbf{E})^{0.5 - 2m} \cdot \Pi^{(2)_{i}}_{kL} [1 + 2m(n - 2)] \cdot \Delta \mathbf{A}^{(2)_{i}}_{(kL)} \right\} \] \quad (5.2.55)

Substituting the Taylor expansion

\[ \Pi^{(1)_{i}}_{kL} [1 + 2m(n - 2)] = \Pi^{(1)_{i}}_{kL} [0.5 + 2m(n - 1) - (2m - 0.5)] = \Pi^{(1)_{i}}_{kL} [0.5 + 2m(n - 1)] - (2m - 0.5) \cdot \Sigma^{(1)_{i}}_{kL} [0.5 + 2m(n - 1)] + \text{higher order terms of } (2m - 0.5) \]

into (5.2.55) and using (5.2.49), we obtained

\[ \mathbf{R}^{n}_{(i)L} = \sum_{k} \left[ \Pi^{(1)_{i}}_{kL} (0.5n) \cdot \Delta \mathbf{R}^{(1)_{i}}_{(kL)} + \Sigma^{(1)_{i}}_{kL} (0.5n) \cdot \Delta \mathbf{B}^{(2)_{i}}_{(kL)} \right] \] \quad (5.2.55)

According to our basic assumption (5.2.54), we have

\[ \Delta \mathbf{R}^{n}_{(k)L} = \frac{n}{(2L + 1)} \left[ LR^{n-1}_{(k)L - 1} + (L + 1) \bar{R}^{n-1}_{(k)L - 1} \right] = \frac{n}{(2L + 1)} \left[ LR^{n-1}_{(k)L - 1} + (L + 1) \bar{R}^{n-1}_{(k)L - 1} \right] = \Delta \bar{\mathbf{R}}^{n}_{(k)L} \] \quad (5.2.56)

Substituting (5.2.56) into (5.2.55), and using (5.2.41,42) and (5.2.50), we obtained

\[ \mathbf{R}^{n}_{(i)L} = \sum_{k} \left[ \Pi^{(1)_{i}}_{kL} (0.5n) \cdot \Delta \mathbf{B}^{(1)_{i}}_{(kL)} + \Sigma^{(1)_{i}}_{kL} (0.5n) \cdot \Delta \mathbf{B}^{(2)_{i}}_{(kL)} \right] + \ln(\mathbf{U}/\mathbf{E}) \cdot \sum_{k} \left[ \Pi^{(1)_{i}}_{kL} (0.5n) \cdot \Delta \mathbf{B}^{(2)_{i}}_{(kL)} \right] \] \quad (5.2.57)

This proves (5.2.51).

Finally, using (5.2.5), (5.2.44) and (5.2.51), we obtained,

\[ \lim_{m \to 0.25} \mathbf{A}^{n}_{(i)L} = \lim_{m \to 0.25} \mathbf{A}^{(0)_{i0}}_{(i)L} - \mathbf{R}^{n}_{(i)L} - (\mathbf{U}/\mathbf{E}) = \mathbf{B}^{(0)_{i1}}_{(i)L} - \bar{\mathbf{R}}^{n}_{(i)L} - (\mathbf{U}/\mathbf{E}) = \mathbf{B}^{n}_{(i)L} \]
This proves Statement (5.2.43), which means that $m = 0.25$ is nothing but a removable singularity of $A_{(i)L}^n$ [36].

In general, following the same procedure, it’s easy to show that all of singularities appearing in $A_{(i)L}^n$ are removable. Therefore, moments $F_{(i)L}^n (E, U)$ determined by Eqs. (2.4.2) must be continuous functions of $m$ for $l > m > 0$.

5.2.4 ION RANGE

Following the previous procedure to derive (5.2.20), we obtained the following recurrent relations for asymptotic moments of ion range [37][38],

$$R_{(i)L}^n (E) = \left[ E^{2m} / N \right]^n \cdot r_{(i)L}^n, \quad$$

$$\begin{cases}
    r_{(i)L}^0 = \delta_{L0} \\
    r_{(i)L}^n = \Delta r_{(i)L}^n / \sum_k \beta_k^n \cdot (2mn) \cdot e_k^n (2mn), \quad \text{for } n > 0 
\end{cases} \quad (5.2.58)$$

The non-vanishing moments $r_{(i)L}^n$ are same as $A_{(i)L}^{0n}$ given by (5.2.23) with $\mu = 0$.

5.3 ASYMPTOTIC MOMENTS OF DEPOSITED MOMENTUM DEPTH DISTRIBUTIONS

5.3.1 IN THE CASE $m \neq 0.25$

Considering the obvious scaling property of Eqs. (5.1.2) [39–41], it’s suitable to define

$$P_{(i)}(z, \bar{u}, U) = P_{(i)}^{[0]}(z, \bar{u}, U) + P_{(i)}^{[1]}(z, \bar{u}, U) + P_{(i)}^{[2]}(z, \bar{u}, U) + \ldots$$

$$P_{(i)L}^n (E, U) = \sqrt{E} \cdot \left[ E^{2m} / N \right]^n \cdot a_{(i)L}^n \quad (5.3.1)$$

Here

$$P_{(i)L}^{[0n]} (E, U) = \sqrt{E} \cdot \left[ E^{2m} / N \right]^n \cdot \bar{a}_{(i)L}^{[0n]} \quad (5.3.2)$$

$$P_{(i)L}^{[1n]} (E, U) = \sqrt{E} \cdot \left[ E^{2m} / N \right]^n \cdot \left[ -(U/E)^{2m-0.5} \cdot \bar{a}_{(i)L}^{[1n]} \right] \quad (5.3.3)$$

$$P_{(i)L}^{[2n]} (E, U) = \sqrt{E} \cdot \left[ E^{2m} / N \right]^n \cdot \left[ -(U/E)^{4m} \cdot \bar{a}_{(i)L}^{[2n]} \right] \quad (5.3.4)$$
\[ P_{(i)_{L}}^{[3]_{n}} (E, U) = \sqrt{E} \cdot \left[ E^{2m}/N \right]^{n} \cdot \left[ + (U/E)^{6m-0.5} \cdot a_{(i)_{L}}^{[3]_{n}} \right] \]  

(5.3.5)

Thus we have

\[ a_{(i)_{L}}^{n} = [a_{(i)_{L}}^{[0]_{n}} - (U/E)^{2m-0.5} \cdot a_{(i)_{L}}^{[1]_{n}} ] \\
- [a_{(i)_{L}}^{[2]_{n}} - (U/E)^{2m-0.5} \cdot a_{(i)_{L}}^{[3]_{n}} ] \cdot (U/E)^{4m} \]  

(5.3.6)

Following the previous procedure, we obtained the following recurrent relations,

\[
\begin{cases}
    a_{(i)_{L}}^{[0]_{n}} = \delta_{L1} \cdot a_{0} \\
    a_{(i)_{L}}^{[1]_{n}} = \sum_{k} \Pi_{k\mid i}^{L} (0.5 + 2mn) \cdot \Delta a_{(i)_{L}}^{[0]_{n}}, \quad \text{for } n > 0 \\
    a_{(i)_{L}}^{[1]_{n}} = \delta_{L0} \cdot a_{1} \\
    a_{(i)_{L}}^{[2]_{n}} = \sum_{k} \Pi_{k\mid i}^{L} [1 + 2m(n - 1)] \cdot \Delta a_{(k)_{L}}^{[1]_{n}}, \quad \text{for } n > 1 \\
    a_{(i)_{L}}^{[2]_{n}} = \delta_{L1} \cdot a_{2} \\
    a_{(i)_{L}}^{[3]_{n}} = \sum_{k} \Pi_{k\mid i}^{L} [0.5 + 2m(n - 2)] \cdot \Delta a_{(k)_{L}}^{[2]_{n}}, \quad \text{for } n > 2 \\
    a_{(i)_{L}}^{[3]_{n}} = \delta_{L0} \cdot a_{3} \\
    a_{(i)_{L}}^{[4]_{n}} = \sum_{k} \Pi_{k\mid i}^{L} [1 + 2m(n - 3)] \cdot \Delta a_{(k)_{L}}^{[3]_{n}}, \quad \text{for } n > 3
\end{cases}
\]  

(5.3.7 - 5.3.10)

Where

\[ a_{0} = \frac{1}{2} \cdot \sqrt{2M_{i}} \]
\[ a_{1} = \frac{1}{2} \cdot (2m - 0.5)^{-1} \cdot \sum_{k} \Pi_{k} \sqrt{2M_{k}} \]
\[ a_{2} = \frac{2}{3} \cdot \frac{\sum_{k} \Pi_{k}^{L} \sqrt{M_{i} / M_{k}} \cdot \left\{ a_{(i)_{L}}^{[0]_{n} \mid (k)_{0}} + 2a_{(i)_{L}}^{[0]_{n} \mid (k)_{2}} \right\} / (4m) - a_{(i)_{L}}^{[1]_{n} \mid (k)_{0}} / (0.5 + 2m)}{a_{3} = 3 \sum_{k} \Pi_{k} \left[ -a_{(i)_{L}}^{[0]_{n} \mid (k)_{0}} / (6m - 0.5) + a_{(i)_{L}}^{[1]_{n} \mid (k)_{2}} / (4m) + a_{(i)_{L}}^{[2]_{n} \mid (k)_{L}} / (2m - 0.5) \right]}
\]

Obviously, the non-vanishing moments \( a_{(i)_{L}}^{[\mu]_{n}} \) are

\[
\begin{align*}
\mu = 0 : & a_{0}^{0}, a_{0}^{1}, a_{2}^{0}, a_{2}^{1}, a_{2}^{2}, a_{3}^{0}, a_{3}^{1}, a_{3}^{2}, a_{3}^{3}, a_{4}^{0}, a_{4}^{1}, a_{4}^{2}, a_{4}^{3}, a_{5}^{0}, a_{5}^{1}, a_{5}^{2}, a_{5}^{3}, \ldots;
\mu = 1 : & a_{0}^{1}, a_{1}^{0}, a_{1}^{1}, a_{2}^{0}, a_{2}^{1}, a_{3}^{0}, a_{3}^{1}, a_{3}^{2}, a_{4}^{0}, a_{4}^{1}, a_{4}^{2}, a_{5}^{0}, a_{5}^{1}, a_{5}^{2}, a_{6}^{0}, a_{6}^{1}, \ldots;
\mu = 2 : & a_{2}^{2}, a_{3}^{0}, a_{3}^{1}, a_{3}^{2}, a_{4}^{0}, a_{4}^{1}, a_{5}^{0}, a_{5}^{1}, a_{5}^{2}, a_{6}^{0}, a_{6}^{1}, a_{6}^{2}, a_{6}^{3}, \ldots;
\mu = 3 : & a_{3}^{3}, a_{4}^{0}, a_{4}^{1}, a_{5}^{0}, a_{5}^{1}, a_{5}^{2}, a_{6}^{0}, a_{6}^{1}, a_{6}^{2}, a_{6}^{3}, \ldots.
\end{align*}
\]  

(5.3.11)
5.3.2 IN THE CASE  \( m = 0.25 \)

Considering the obvious scaling property of Eqs. (5.1.2), it’s suitable to define

\[
P_{(i)}(z, \bar{v}, U) = P_{(i)}^{[0]}(z, \bar{v}, U) + P_{(i)}^{[1]}(z, \bar{v}, U) + P_{(i)}^{[2]}(z, \bar{v}, U) + P_{(i)}^{[3]}(z, \bar{v}, U)
\]

\[
P_{(i)}^n(E, U) = \sqrt{E} \cdot [E^{2m}/N]^n \cdot b_{(i)L}^n
\]

(5.3.12)

Here \( P_{(i)}^{[0]}(E, U) = \sqrt{E} \cdot [E^{2m}/N]^n \cdot b_{(i)L}^{[0]} \)

(5.3.13)

\[
P_{(i)}^{[1]}(E, U) = \sqrt{E} \cdot [E^{2m}/N]^n \cdot [\ln(U/E) \cdot b_{(i)L}^{[1]}]
\]

(5.3.14)

\[
P_{(i)}^{[2]}(E, U) = \sqrt{E} \cdot [E^{2m}/N]^n \cdot [-(U/E) \cdot b_{(i)L}^{[2]}]
\]

(5.3.15)

\[
P_{(i)}^{[3]}(E, U) = \sqrt{E} \cdot [E^{2m}/N]^n \cdot [-(U/E) \ln(U/E) \cdot b_{(i)L}^{[3]}]
\]

(5.3.16)

Thus \( b_{(i)L}^n = [b_{(i)L}^{[0]} + \ln(U/E) \cdot b_{(i)L}^{[1]}] - [b_{(i)L}^{[2]} + \ln(U/E) \cdot b_{(i)L}^{[3]}] \cdot (U/E) \)

(5.3.17)

Following the previous procedure, we obtained the following recurrent relations,

\[
\begin{align*}
b_{(i)L}^{[0]} &= \delta_{L,0} \cdot \frac{1}{3} \sum_k \Pi_{ki} \sqrt{2M_k} + \delta_{L,2} \cdot \frac{2}{3} \sum_k \Pi_{ki}^2 \cdot (1) \sqrt{2M_k} \\
b_{(i)L}^{[0]} &= \sum_k \left[ \Pi_{ki} \cdot [0.5(n+1)] \cdot \Delta b_{(k,L)}^{[0]} + \sum_k \Pi_{ki} \cdot [0.5(n+1)] \cdot \Delta b_{(k,L)}^{[1]} \right] \text{ for } n > 0
\end{align*}
\]

(5.3.18)

\[
\begin{align*}
b_{(i)L}^{[1]} &= \delta_{L,0} \cdot \frac{1}{3} \sum_k \Pi_{ki} \sqrt{2M_k} \\
b_{(i)L}^{[1]} &= \sum_k \Pi_{ki} \cdot [0.5(n+1)] \cdot \Delta b_{(k,L)}^{[1]} \text{, for } n > 1
\end{align*}
\]

(5.3.19)

\[
\begin{align*}
b_{(i)L}^{[2]} &= \delta_{L,0} \cdot \frac{2}{3} \sum_k \Pi_{ki} \cdot \sqrt{M_i / M_k} \cdot [b_{(k,0)}^{[0]} + 2b_{(k,2)}^{[0]} - b_{(k,0)}^{[1]}] \\
b_{(i)L}^{[2]} &= \sum_k \left[ \Pi_{ki} \cdot [0.5(n+1)] \cdot \Delta b_{(k,L)}^{[2]} + \sum_k \Pi_{ki} \cdot [0.5(n+1)] \cdot \Delta b_{(k,L)}^{[3]} \right] \text{ for } n > 2
\end{align*}
\]

(5.3.20)

\[
\begin{align*}
b_{(i)L}^{[3]} &= \delta_{L,0} \cdot \frac{3}{3} \sum_k \Pi_{ki} \cdot b_{(k,0)}^{[2]} \\
b_{(i)L}^{[3]} &= \sum_k \Pi_{ki} \cdot [0.5(n+1)] \cdot \Delta b_{(k,L)}^{[3]} \text{, for } n > 3
\end{align*}
\]

(5.3.21)

No singularities [36] can be seen in (5.3.11) and

\[
\lim_{m \to 0.25} a_{(i)L}^n = b_{(i)L}^n
\]

which means that \( m = 0.25 \) is nothing but a removable singularity of \( a_{(i)L}^n \). Therefore, moments

\[
P_{(i)L}^n(E, U) \text{ determined by Eqs. (2.4.2) must be continuous functions of } m \text{ for } 1 > m > 0.
\]
5.4 BASIC PROPERTIES OF DEPTH DISTRIBUTION FUNCTIONS
OF DEPOSITED ENERGY AND MOMENTUM

5.4.1 ENERGY AND MOMENTUM DIFFUSIONS

The recurrent relations (5.2.20-22) and (5.3.7-10) can be written respectively as following,

\[
\begin{align*}
A^{[\mu]}_{(i)L} &= \delta_{La(\mu-1)} \cdot A_\mu, \\
A^{[\mu]}_{(i)L} &= \sum_k \Pi^L_{kiL}(s) \cdot \Delta A^{[\mu]}_{(k)L}, \quad \text{for } n > \mu \\
& \quad s \equiv 1 - 0.5a(\mu - 1) + 2m(n - \mu) \\
& \quad \mu = 0, 1, 2.
\end{align*}
\]

(5.4.1)

Here \( a(\mu) \equiv 0.5[1 + (-1)^\mu] \). For \( \mu = 1, 2, 3 \), the second equation of (5.4.2) turns out to be

\[
a^{[\mu]}_{(i)L} = \sum_k \Pi^L_{kiL}(s) \cdot \frac{n}{(2L + 1)} \left[ L \cdot a^{[\mu]}_{(k)L-1} + (L + 1) \cdot a^{[\mu]}_{(k)L+1} \right] \quad \text{for } n > \mu \quad \text{or}
\]

\[
\sum_k \Pi^L_{kiL}(s) \cdot \frac{(n-1)}{(2L + 1)} \left[ L \cdot a^{[\mu]}_{(k)L-1} + (L + 1) \cdot a^{[\mu]}_{(k)L+1} \right], \quad \text{for } n > \mu
\]

(5.4.3)

Substituting

\[
a^{[\mu]}_{(i)L} = [a_\mu / (\mu A_{\mu-1})] \cdot nA^{[\mu-1]}_{(i)L}^{-1}
\]

(5.4.4)

into (5.4.3), we obtained (5.4.1), thus, (5.4.4) is a correct relation associating (5.4.2) with (5.4.1).

Following a similar procedure, we can derived

\[
A^{[\mu]}_{(i)L} = [A_\mu / (\mu A_{\mu-1})] \cdot nA^{[\mu-1]}_{(i)L}^{-1}
\]

(5.4.5)

By using (5.2.2-4) and (5.3.3-5), we can see that (5.4.4,5) are equivalent to

\[
\begin{align*}
P^{[\mu]}_{(i)}(z, E, U) &= -D_\mu(U) \cdot \partial P^{[\mu]}_{(i)}(z, E, U) / \partial z, \quad \mu = 1, 2, 3 \\
\Gamma^{[\mu]}_{(i)}(z, E, U) &= -D_\mu(U) \cdot \partial \Gamma^{[\mu]}_{(i)}(z, E, U) / \partial z, \quad \mu = 1, 2.
\end{align*}
\]

(5.4.6, 5.4.7)

Here \( D_\mu(U) \equiv N^{-1} \cdot U^{2m-0.5 \cdot [a_\mu / (\mu A_{\mu-1})]} \) and \( D_{\mu}^{\parallel}(U) \equiv N^{-1} \cdot U^{2m+0.5 \cdot [A_\mu / (\mu A_{\mu-1})]} \)
are independent of the incident particle.

Thus, if only the first two terms \( (\mu = 0, 1) \) are considered, we have \( [11][40][41] \):

\[
P_{(i)}(z, \bar{\nu}, U) = P_{(i)}^{(0)}(z, \bar{\nu}, U) - D_{(i)}(U) \cdot \partial F_{(i)}(z, \bar{\nu})/\partial z \tag{5.4.8}
\]

\[
F_{(i)}(z, \bar{\nu}, U) = F_{(i)}^{(0)}(z, \bar{\nu}) - D'_{(i)}(U) \cdot \partial P_{(i)}(z, \bar{\nu})/\partial z \tag{5.4.9}
\]

From (5.2.2), (5.2.25) (5.3.2) and (5.2.13), one can see that

\[
P_{(i)}(z, \bar{\nu}) \equiv P_{(i)}^{(0)}(z, \bar{\nu}, U) \quad \text{and} \quad F_{(i)}(z, \bar{\nu}) \equiv F_{(i)}^{(0)}(z, \bar{\nu}, U)
\]

are the depth distribution functions with neglecting threshold energy \( U \) [3][4]. or

\[
P_{(i)}(z, \bar{\nu})= \lim_{(U/E) \to 0} [P_{(i)}(z, \bar{\nu}, U)], \quad \text{if it exists;}
\]

\[
F_{(i)}(z, \bar{\nu}) = \lim_{(U/E) \to 0} [F_{(i)}(z, \bar{\nu}, U)], \quad \text{if it exists.}
\]

The diffusion coefficients \( D(U) \) and \( D'(U) \) are given by

\[
D_{(i)}(U) = \frac{\Gamma_m}{3(0.5 - 2m)} \sum_k \frac{G_k}{(G)_k} \frac{\sqrt{2M_k U^3}}{N_k(U)} \tag{5.4.10}
\]

\[
D'_{(i)}(U) = \frac{\Gamma_m}{(0.5 + 2m)} \sum_k \frac{H_k}{(H)_k} \frac{U^2}{N_k(U) \sqrt{2M_k U}} \tag{5.4.11}
\]

Here \( \Gamma_m = \beta_n (I)[\int_0^1 dX A_n (X)[-X \ln(X) - (1 - X) \ln(1 - X)]. \)

\[
(5.4.12)
\]

Repeat substituting (5.4.6,7) gives

\[
P_{(i)}^{(2)}(z, E, U) = [D_{(i)}(U)D_{(i)}'(U)] \cdot \partial^2 P_{(i)}(z, E)/\partial z^2;
\]

\[
F_{(i)}^{(2)}(z, E, U) = [D_{(i)}(U)D_{(i)}'(U)] \cdot \partial^2 F_{(i)}(z, E)/\partial z^2;
\]

\[
P_{(i)}^{(3)}(z, E, U) = [D_{(i)}(U)D_{(i)}'(U)] \cdot \partial^3 P_{(i)}(z, E)/\partial z^3.
\]

Thus, (5.4.8,9) can be generalized to

\[
P_{(i)}(z, \bar{\nu}, U) = [1 + D_{(i)}(U)D'_{(i)}(U) \cdot \partial^2 /\partial z^2 + \cdots] \cdot P_{(i)}(z, \bar{\nu}) - D_{(i)}(U)[1 + D_{(i)}(U)D'_{(i)}(U) \cdot \partial^2 /\partial z^2 + \cdots] \cdot \partial F_{(i)}(z, \bar{\nu})/\partial z \tag{5.4.13}
\]

\[
F_{(i)}(z, \bar{\nu}, U) = [1 + D_{(i)}(U)D'_{(i)}(U) \cdot \partial^2 /\partial z^2 + \cdots] \cdot F_{(i)}(z, \bar{\nu}) - D'_{(i)}(U)[1 + D_{(i)}(U)D'_{(i)}(U) \cdot \partial^2 /\partial z^2 + \cdots] \cdot \partial P_{(i)}(z, \bar{\nu})/\partial z \tag{5.4.14}
\]
Therefore, the continuity and differentiability of $P_{(i)}(z, \bar{u})$ and $F_{(i)}(z, \bar{u})$ play a very important role in researching depth distribution functions of deposited energy and momentum.

### 5.4.2 CONTINUITY AND DIFFERENTIABILITY OF DEPTH DISTRIBUTION

Similar to (2.1.1), to first order in $dz$, and using the space translational invariance of the medium [37][39–42], $H_{(i)}(z, \bar{u}, U)$ must satisfy

$$H_{(i)}(z, \bar{u}, U) = \left[1 - N\delta z\sigma_{(i)}(\bar{u})\right]H_{(i)}(z - \eta\delta z, \bar{u}, U) + N\delta z \sum_{k=1}^{\infty} \alpha_k \int d\sigma_{ik} (\bar{u}; \bar{v}', \bar{v}^*) [H_{(i)}(z, \bar{u}', U) + H_{(k)}(z, \bar{u}^*, U)]$$

or

$$H_{(i)}(z - \eta\delta z, \bar{u}, U) - H_{(i)}(z, \bar{u}, U) = N\delta z \sum_{k=1}^{\infty} \alpha_k \int d\sigma_{ik} \left[ H_{(i)}(z - \eta\delta z, \bar{u}, U) - H_{(i)}(z, \bar{u}', U) - H_{(k)}(z, \bar{u}^*, U) \right] \quad (5.4.15)$$

Here, $H$ stands for one of $F$ and $P$, same as (2.4.1). (5.4.15) holds for the ion range if canceling the third term in the bracket of the right hand. $H_{(i)}(z, \bar{u}) \equiv H_{(i)}(z, \bar{u}, U \to 0)$ for the case neglecting threshold energy $U$, if it exists. The following is a new theory about the continuity and differentiability of $H_{(i)}(z, \bar{u}, U)$ and it’s proof.

**Theorem:** $H_{(i)}(z, \bar{u}, U)$ determined by Eqs. (5.4.15) must be continuous and infinitely differentiable functions of $z$, if they exist.

**Proof:** The critical point is that Eqs. (5.4.15) was derived based on finite $\sigma_{(i)}(\bar{u})$ [2]. The solution of Eqs. (5.4.15) may be extended to $\sigma_{(i)}(\bar{u}) = \infty$ finally, if they exist. But, even so, $\delta z$ approaching to zero ($\delta z \to 0$) in Eqs. (5.4.15) must be executed first, then $\sigma_{(i)}(\bar{u}) = \infty$ to guarantee against the occurrence of $N\delta z\sigma_{(i)}(\bar{u}) \to \infty$.

For finite $\sigma_{(i)}(\bar{u})$, $H_{(i)}(z, \bar{u}, U) = \text{finite}$,

$$\int d\sigma_{ik} \left[ H_{(i)}(z - \eta\delta z, \bar{u}, U) - H_{(i)}(z, \bar{u}', U) - H_{(k)}(z, \bar{u}^*, U) \right] = (\text{finite quantity})$$

Thus, the discontinuity of $H_{(i)}(z, \bar{u}, U)$ is determined by (5.4.15)
\[
\lim_{\sigma \to \infty} \lim_{\nu \to 0} [H_{(i)}(z - \eta \delta z, \nu) - H_{(i)}(z, \nu, U)] = N \lim_{\sigma \to \infty} \lim_{\nu \to 0} [\delta z \cdot (\text{finite quantity})] = 0.
\]

Therefore, for both \( \sigma, (\nu) = \text{finite and } \infty \), \( H_{(i)}(z, \nu, U) \) must be continuous functions of \( z \) and

\[
H_{(i)}^{(r)}(z, \nu, U) = \text{finite}
\]

Here \( H_{(i)}^{(r)}(z, \nu, U) \equiv \frac{\partial^n H_{(i)}(z, \nu, U)}{\partial z^n}, \quad n = 1, 2, 3, \ldots. \)

Assuming that \( H_{(i)}^{(r-1)}(z, \nu, U) \) is finite for \( \sigma = \text{finite} \), to first order in \( dz \), and using the space translational invariance of the medium, \( H_{(i)}^{(r-1)}(z, \nu, U) \) must satisfy

\[
H_{(i)}^{(r-1)}(z - \eta \delta z, \nu, U) - H_{(i)}^{(r-1)}(z, \nu, U) = N\delta z \sum_{k=1}^{6} \alpha_k \int d\sigma_{\nu} \left[H_{(i)}^{(r-1)}(z - \eta \delta z, \nu, U) - H_{(i)}^{(r-1)}(z, \nu', U) - H_{(k)}^{(r-1)}(z, \nu'', U)\right]
\]

The same argument resulted in that for both \( \sigma, (\nu) = \text{finite and } \infty \), \( H_{(i)}^{(r-1)}(z, \nu, U) \) must be continuous function of \( z \) and

\[
H_{(i)}^{(r)}(z, \nu, U) = \text{finite}.
\]

Therefore, \( H_{(i)}(z, \nu, U) \) must be infinitely differentiable respect to \( z \), as long as it exists, even if for \( \sigma, (\nu) = \infty \). This completes the proof.

In literatures [1~11], authors usually expand the first term on the right-hand of Eqs. (5.4.15) to the first order in \( \delta z \) and obtain the linear Boltzmann equations determining \( H_{(i)}(z, \nu, U) \):

\[
- \eta \frac{\partial H_{(i)}(z, \nu, U)}{\partial z} = N\delta z \sum_{k=1}^{6} \alpha_k \int d\sigma_{\nu} \left[H_{(i)}(z, \nu, U) - H_{(i)}(z, \nu', U) - H_{(k)}(z, \nu'', U)\right] (5.4.16)
\]

which is obviously equivalent to Eqs. (5.4.15) if \( \sigma, (\nu) = \text{finite} \). The conclusion is correct for the ion range.

Great successes of Eqs. (5.4.16) made many authors take it for granted that Eqs. (5.4.16) is equivalent to (5.4.15) in any cases, even if \( \sigma, (\nu) = \infty \). In Ref.[3][4][5], L.G. Glazov had seen dear signs of non-equivalence of Eqs. (5.4.16) and (5.4.15).
5.5 ON GLAZOV’S PARADOX

5.5.1 BASIC EQUATIONS AND TWO DIFFERENT POWER LAW CROSS-SECTIONS

For mono-element amorphous or polycrystalline infinite medium, the depth distributions of deposited energy \( F(z, \bar{u}, U) \) and momentum \( P(z, \bar{u}, U) \) are defined so that \( F(z, \bar{u}, U) \) and \( P(z, \bar{u}, U)dz \) are the average amount of energy and parallel component of momentum respectively located in a layer \((z, dz)\). Eqs. (5.4.15,16) reduced to

\[
\begin{align*}
\int \left( \nu' - \nu - \nu \right) \delta - \sigma \delta &= \nu - \nu \eta \delta - \int \delta H_{ij}(z, \bar{u}, U) - H_{ij}(z, \bar{u}', U) - H_{ij}(z, \bar{u}^*, U) \right) \] \\
&= N \int \delta H_{ij}(z, \bar{u}, U) / \partial z \\
&= N \int \delta H_{ij}(z, \bar{u}, U) - H_{ij}(z, \bar{u}', U) - H_{ij}(z, \bar{u}^*, U) \\
\tag{5.5.1}
\end{align*}
\]

Here \( j = 0, 1 \). \( H_{i0}(z, \bar{u}, U) \equiv F(z, \bar{u}, U) \) and \( H_{ij}(z, \bar{u}, U) \equiv P(z, \bar{u}, U) \).

The most critical question is whether Eq. (5.5.2) is equivalent to (5.5.1)?

To answer this question needs scattering cross-section. Let’s introduced the following modified power law cross-section

\[
\begin{align*}
\sigma(E, T, \xi) &= \begin{cases} 
CE^{-m T^{-1}} dT, & E \geq T \geq \xi E \\
0, & T < \xi E
\end{cases} \\
\tag{5.5.3}
\end{align*}
\]

where \( 1 > \xi \geq 0 \). The total cross-section \( \sigma = (C/m)(\xi^{-m} - 1)E^{-2m} \) is finite for \( \xi \neq 0 \).

If \( \xi = 0 \), (5.5.3) reduced to the regular power cross-section [7]

\[
\sigma(E, T) = CE^{-m T^{-1}} dT, \quad E \geq T \geq 0, \quad 1 > m > 0. \tag{5.5.4}
\]

which we usually see in literatures [3][4][5], however, the total cross-section of (5.5.4) is infinite.

The power law cross-section (5.5.3) is a special case of (5.1.1),

\[
A(X) = \begin{cases} 
CX^{-1-m}, & 1 \geq X \geq \xi \\
0, & X < \xi
\end{cases} \tag{5.5.5}
\]

\( \xi = 0 \) corresponds to the regular power cross-section (5.5.4).
5.5.2 NEGLECTING THRESHOLD ENERGY $U$

Besides scattering cross-section, solving Eq. (5.5.2) needs conditions (2.2.15, 16), thus,

$$\int_{-\infty}^{\infty} H_{ij}(z, \nu) \, dz = E^{1/2} \left( \eta \sqrt{2M} \right)^{j}$$

(5.5.6)

Seeing (5.5.5), (5.5.3, 4) are special cases of (5.1.1), according to the previous results (5.2.20) and (5.3.7), $H_{ij}(z, \nu)$ satisfy a simple scaling law, and can be represented in the form [3][4]:

$$H_{ij}(z, \nu) = 3^{-j} (2M)^{j/2} NCE^{1-j/2-2m} F_{ij}(x, \eta)$$

(5.5.7)

Here $x \equiv z \frac{NC}{E^{2m}}$. The dimensionless functions $F_{ij}(x, \eta)$ can be expanded in terms of Legendre polynomials:

$$F_{ij}(x, \eta) = \sum_{L=0}^{\infty} (2L + 1) P_{L}(\eta) F_{ijk}(x)$$

(5.5.8)

Substituting (5.5.7, 8) into Eq. (5.5.6) yielded the following equation [3][4]:

$$\left\{ \begin{array}{l}
- \left[ LF_{ij}^{(j)}(x) + (L + 1) F_{ij}^{(j+1)}(x) \right] = (2L + 1) \int_{\xi}^{1} t^{-1-m} dt F_{ijkl}(x) \\
- P_{L}(\sqrt{1-t})(1-t)^{1-j/2-2m} F_{ijkl}(x/(1-t)^{2m}) - P_{L}(\sqrt{1-t}) t^{1-j/2-2m} F_{ijkl}(x/t^{2m}) \\
\int_{-\infty}^{\infty} F_{ijkl}(x) \, dx = \delta_{ij}
\end{array} \right.$$  

(5.5.9)

which also holds for the ion range if canceling the third term in the bracket of the right hand. In general, the behavior of solution of Eq. (5.5.9) is dependent on $\xi$. Eq. (5.5.9) is obviously equivalent to Eq.(5.5.1) if $\xi \neq 0$, otherwise, we have

**Theorem:** The equivalence between Eq. (5.5.9) and Eq. (5.5.1) can not be extended to $\xi = 0$ for $1 > m > 0$, because $\lim_{\xi \to 0} [F_{ijkl}(x)]$ does not exists, or

$$\lim_{\xi \to 0} \lim_{x \to \infty} x^a F_{ijkl}(x) = \lim_{x \to \infty} \lim_{\xi \to 0} x^a F_{ijkl}(x)$$

$a \in (-\infty, \infty)$

(5.5.10)

**Proof:** Introducing the spatial moments

$$F_{ijkl}^{n} = \int_{-\infty}^{\infty} x^n F_{ijkl}(x) \, dx$$
multiplying both sides of the first equation of Eq. (5.5.9) by \( x^n \) and integrating \( x \) over the range \((\infty, \infty)\), one can easily obtained the recursion relations:

\[
\begin{align*}
F_{nl}^0 &= \delta_{nj}, \\
F_{nl}^n &= \frac{n L F_{nl-1}^{n-1} + (L + 1) F_{nl+1}^{n-1}}{(2L + 1) I_L(\xi, 2mn + 1 - j/2)}, \quad n = 1, 2, 3, 4, \ldots
\end{align*}
\]

(5.5.11)

Here, \( I_L(\xi, s) = \int_0^\xi t^{i-m} dt \left[ - P_L(\sqrt{1-t}) \cdot (1-t)^{i} - P_L(\sqrt{t}) \cdot t^i \right] \)

\[
\lim_{\xi \to 0} I_L(\xi, s) = I_L(0, s) \equiv I_L(s)
\]

\[
= C^L_0 I_0[s + 0.5L] + C^L_{L-2} I_0[s + 0.5(L - 2)] + C^L_{L-4} I_0[s + 0.5(L - 4)] + \ldots
\]

\[+ \begin{cases} C^L_0 I_0(s + 0.5), & \text{if } L \text{ is odd} \\ C^L_0 I_0(s), & \text{if } L \text{ is even} \end{cases} \]

\[
I_0(s) = \int_0^s t^{i-m} dt \left[ - (1-t)^{i} - t^i \right] = (s/m) \left[ B(s, 1-m) - (s-m)^{-1} \right]
\]

\( B(x, y) = \Gamma(x) \Gamma(y)/\Gamma(x+y) \) is B-function.

\( C^L_0 \) are the coefficients of Legendre polynomials, i.e.

\[
P_L(x) = C^L_0 x^L + C^L_{L-2} x^{L-2} + C^L_{L-4} x^{L-4} + \ldots + \begin{cases} C^L_1 x, & \text{if } L \text{ is odd} \\ C^L_0 a_0, & \text{if } L \text{ is even} \end{cases}
\]

From the recursion relation, one can easily obtained non-vanishing and finite moments:

\[
\begin{align*}
L = 0 : & \quad F^0_{[00]}, \quad F^2_{[00]}, \quad F^4_{[00]}, \quad F^6_{[00]}, \quad F^8_{[00]}, \quad \ldots \\
L = 1 : & \quad F^1_{[01]}, \quad F^3_{[01]}, \quad F^5_{[01]}, \quad F^7_{[01]}, \quad \ldots \\
L = 2 : & \quad F^2_{[02]}, \quad F^4_{[02]}, \quad F^6_{[02]}, \quad F^8_{[02]}, \quad \ldots \\
& \quad \ldots \ldots
\end{align*}
\]

(5.5.12)

for deposited Energy, i.e. \((j = 0)\) and

\[
\begin{align*}
L = 0 : & \quad F^0_{[10]}, \quad F^2_{[10]}, \quad F^4_{[10]}, \quad F^6_{[10]}, \quad \ldots \\
L = 1 : & \quad F^0_{[11]}, \quad F^2_{[11]}, \quad F^4_{[11]}, \quad F^6_{[11]}, \quad \ldots \\
L = 2 : & \quad F^1_{[12]}, \quad F^3_{[12]}, \quad F^5_{[12]}, \quad F^7_{[12]}, \quad \ldots \\
& \quad \ldots \ldots
\end{align*}
\]

(5.5.13)

for deposited Momentum i.e. \((j = 1)\). (5.5.12) and (5.5.13) agree with (5.2.23) and (5.3.11) respectively for \( \mu = 0 \).
(5.5.12,13) can be expressed by \( F_{j L}^{n} = 0 \) for \((j + L + n) = 1, 3, 5, 7, \ldots\). Therefore, \( F_{j L}(x) \) is even or odd finite functions if \((j + L)\) is even or odd respectively. Thus, if \( \xi \neq 0 \), \( F_{j L}(x) \) must be a continuous and infinitely differentiable function of \( x \), and

\[
\lim_{x \to 0} [F_{j L}(x)] = 0, \quad \text{for } (j + L + n) = 1, 3, 5, 7, \ldots. \tag{5.5.14}
\]

Here \( F_{j L}^{(n)}(x) \equiv \frac{d^n F_{j L}(x)}{dx^n} \).

Taking \( n \)-order derivative of both sides of the first equation of Eq. (5.5.9) respect \( x \), then:

\[
- \left[ L F_{j L}^{(n+1)}(x) + (L + 1) F_{j L+1}^{(n+1)}(x) \right] = (2L + 1) \int_{\xi}^{1} t^{1-m - 1} dt \left[ F_{j L}^{(n)}(x) \right] - P_{L} (\sqrt{1 - t})(1 - t)^p F_{j L}^{(n)}(x/(1 - t)^2m) - P_{L} (\sqrt{t}) t^p F_{j L}^{(n)}(x/t^2m)
\]

where \( n = 0, 1, 2, 3, \ldots \) and \( p \equiv 1 - j/2 - 2m(n + 1) \). Eq. (5.5.15) is also equivalent to Eq. (5.5.1) if \( \xi \neq 0 \). On the one hand, if \((j + L + n)\) is odd, substituting (5.5.14) into Eq. (5.5.15), immediately we obtained

\[
\lim_{\xi \to 0} \lim_{x \to 0} [L F_{j L}^{(n+1)}(x) + (L + 1) F_{j L+1}^{(n+1)}(x)] = 0 \tag{5.5.16}
\]

which agrees with (5.5.14). On the other hand, seeing that

\[
\int_{0}^{1} t^{1-m - 1} dt \left[ F_{j L}^{(n)}(x) \right] - P_{L} (\sqrt{1 - t})(1 - t)^p F_{j L}^{(n)}(x/(1 - t)^2m) = \text{finite}
\]

as long as the integer number \( n > (2 + 2q - j)/(4m) - 1.5 \), one always can have

\[
\lim_{x \to 0} \lim_{\xi \to 0} [L F_{j L}^{(n+1)}(x) + (L + 1) F_{j L+1}^{(n+1)}(x)]
\]

\[
\sim (2L + 1) \lim_{x \to 0} \int_{0}^{1} t^{p-1-m} dt P_{L} (\sqrt{t}) F_{j L}^{(n)}(x/t^2m)
\]

\[
\sim (2L + 1) \lim_{x \to 0} x^{(p+q)/(2m)-0.5} \int_{0}^{\infty} y^{-(p+q)/(2m) - 0.5} F_{j L}^{(n)}(y) dy \]

\( = \infty \)

where \( q \equiv 0 \) if \( L \) is even and \( q \equiv 0.5 \) if \( L \) is odd. As one can see that (5.5.17) contradicts (5.5.16) directly, i.e. (5.5.10). Thus, the validity of Eq. (5.5.9) can not extend to \( \xi = 0 \) for \( 1 > m > 0 \). This completes the proof.

A typical example was given by Glazov in Ref.[4] for momentum deposition \( (j = 1) \):
substituting \( m = 0.5 \) and \( n = L = 0 \), \((5.5.16,17)\) read

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(1)}_{\xi \xi}(x) \right] = 0
\]

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(1)}_{\xi \xi}(x) \right] \sim \lim_{x \to 0} \int_{x}^{\infty} t^{-2.5} dt F^{(0)}_{\xi \xi}(x/t) \\
\sim \lim_{x \to 0} \left[ x^{-1.5} \int_{x}^{\infty} y^{0.5} F^{(0)}_{\xi \xi}(y) dy \right] = \infty
\]

respectively. In Ref.\([4][5]\), Glazov used the regular power cross-section \((5.5.4)\) for the derivation, in the other word, \( \xi \to 0 \) executed from the very beginning. Another example is given here for energy deposition \((j = 0)\): substituting \( m = 0.5 \), \( n = 1 \) and \( L = 0 \), \((5.5.16,17)\) read

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(2)}_{\xi \xi}(x) \right] = 0
\]

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(2)}_{\xi \xi}(x) \right] \sim \lim_{x \to 0} \int_{x}^{\infty} t^{-2.5} dt F^{(0)}_{\xi \xi}(x/t) \\
\sim \lim_{x \to 0} \left[ x^{-1.5} \int_{x}^{\infty} y^{0.5} F^{(0)}_{\xi \xi}(y) dy \right] = \infty
\]

respectively. Furthermore, even \( m \) is as small as \( m = 10^{-4} \), the contradiction \((5.5.10)\) still holds.

For momentum deposition \((j = 1)\): substituting \( m = 10^{-4} \), \( L = 0 \) and \( n = 2500 \), \((5.5.16,17)\) read

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(2501)}_{\xi \xi}(x) \right] = 0
\]

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(2501)}_{\xi \xi}(x) \right] \sim \lim_{x \to 0} \int_{x}^{\infty} t^{-1.0003} dt F^{(2500)}_{\xi \xi}(x/t^{0.0002}) \\
\sim \lim_{x \to 0} \left[ x^{-1.5} \int_{x}^{\infty} y^{0.5} F^{(2500)}_{\xi \xi}(y) dy \right] = \infty
\]

respectively. If the regular power cross-section \((5.5.4)\) is used for solving Eq. \((5.5.2)\), \( F_{\xi \xi}(x) \) and it’s all order derivatives will be continuous until 2499-order, but it’s 2500-order derivative diverges at \( x = 0 \). A similar phenomenon happened for energy deposition \((j = 0)\): substituting \( m = 0.0001 \), \( L = 0 \) and \( n = 4999 \), \((5.5.16,17)\) read

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(5000)}_{\xi \xi}(x) \right] = 0
\]

\[
\lim_{x \to 0} \lim_{\xi \to 0} \left[ F^{(5000)}_{\xi \xi}(x) \right] \sim \lim_{x \to 0} \int_{x}^{\infty} t^{-1.0001} dt F^{(4999)}_{\xi \xi}(x/t^{0.0002}) \\
\sim \lim_{x \to 0} \left[ x^{-0.5} \int_{x}^{\infty} y^{-0.5} F^{(4999)}_{\xi \xi}(y) dy \right] = \infty
\]
respectively. If the power cross-section (5.5.4) is used for solving Eq. (5.5.2), its derivatives will be continuous until 4998-order, but it's 5000-order derivative diverges at \( x = 0 \).

5.5.3 CONSIDERING THRESHOLD ENERGY EFFECTS U

In the frozen picture, solving Eq. (5.5.2) needs conditions

\[ H_{ij}(z, \bar{u}) = g(z, \sigma)E^{1-j/2}(\eta\sqrt{2M})^j, \quad \text{for} \quad E < U. \quad (5.5.18) \]

Here \( g(z, \sigma) \equiv \theta(z)(N\sigma/\eta)\exp(-zN\sigma/\eta) \).

If \( \sigma = \infty \), \( g(z, \sigma) = \delta(z) \) and (5.5.18) reduced to the second equations of Eqs. (2.2.14).

For \( E > U > 0 \), a collision cascade will be initiated. Eq. (5.5.2) can be written as

\[
\begin{align*}
\eta\frac{\partial H_{ij}(z, \bar{u})}{\partial z} &= -Q_{ij}(z, \bar{u}) \\
&+ Ng(z, \sigma)(2M)^{j/2}\left[\int_{-U}^{E} d\sigma (\eta^*)^j(E - T)^{1-j/2} + \int_{0}^{U} d\sigma (\eta^*)^j(T)^{1-j/2}\right]
\end{align*}
\]  

(5.5.19)

Here \( Q_{ij}(z, \bar{u}) = \int_{0}^{E} d\sigma H_{ij}(z, \bar{u}) - \int_{-U}^{E} d\sigma H_{ij}(z, \bar{u}') - \int_{U}^{E} d\sigma H_{ij}(z, \bar{u}^*). \)

On the one hand, considering that each term on the left-hand of Eq. (5.5.19) is finite in the case \( \sigma = \text{finite} \), integrating the both sides of Eq. (5.5.19) over a small region \( a > z > -a \) yielded

\[ \lim_{\sigma \to \infty} \lim_{a \to 0} [\Delta H_{ij}(\bar{u})] = H_{ij}(z = +0, \bar{u}) - H_{ij}(z = -0, \bar{u}) = 0 \quad (5.5.20) \]

Thus, \( H_{ij}(z, \bar{u}) \) makes absolutely no jump at \( z = 0 \). On the other hand, for perpendicular incidence, substituting \( \eta = 1, \eta' = \sqrt{1 - T/E} \) and \( \eta^* = \sqrt{T/E} \), (5.5.19) reduced to

\[
\begin{align*}
\frac{\partial H_{ij}(z, \bar{u})}{\partial z} &= Ng(z, \sigma)(2M/E)^{j/2}\left[\int_{E-W}^{E} d\sigma \cdot (E - T) + \int_{0}^{W} d\sigma \cdot T\right] - Q_{ij}(z, \bar{u})
\end{align*}
\]  

(5.5.21)

In the case \( \sigma = \infty \), integrating the both sides of Eq. (5.5.21) over a small region including \( z = 0 \) and ignoring \( \int_{0}^{b+} dz Q_{ij}(z, E, \eta) \), Glazov obtained [5] [41]

\[ \lim_{\sigma \to \infty} \lim_{a \to 0} [\Delta H_{ij}(\bar{u})] = N(2M/E)^{j/2}\left[\int_{E-W}^{E} d\sigma \cdot (E - T) + \int_{0}^{W} d\sigma \cdot T\right] \neq 0 \quad (5.5.22) \]

which is different from (5.5.20) due to
In particular, substituting (5.5.4) into (5.5.22), we obtained
\[
\lim_{a \to 0} \lim_{ \sigma \to \infty} \int_{-a}^{a} d \theta(z, \sigma) = \lim_{a \to 0} \lim_{ \sigma \to \infty} \int_{-a}^{a} d \delta(z) = 1 \neq 0 = \lim_{a \to 0} \lim_{ \sigma \to \infty} \int_{-a}^{a} d \theta(z, \sigma)
\]

The last formula was derived by Glazov for momentum deposition depth distributions [5] and proved by simulation results [11]. As one can see that (5.5.23) contradicts (5.5.20) directly, similar to (5.5.10). The conclusion is also correct for the ion range.

### 5.5.4 GLAZOV’S PARADOX IS IMPORTANT, BUT NOT SERIOUS

Glazov’s paradox [3][4][5] revealed the following fact at first time: the regular power cross-section (5.5.4) couldn’t be used to calculate depth distributions of deposed energy, momentum and the ion range in the strict sense because of infinite total cross-section.

On the other hand, from the physics point of view, any total cross-section couldn’t be infinite due to the Quantum Mechanics consideration. As we known, (5.5.4) can only describe the realistic atomic collision approximately. In the case neglecting threshold energy, at least for \(0 < m < 0.5\), all of \(F_{ij}(x)\) are continuous functions. For \(m = 0.2\), \(F_{0ij}(x)\) is first-order differentiable function.

In the case considering threshold energy effects, for the deposited momentum depth distribution functions, the Monte Carlo simulation results [11] has been showed to fit the corresponding curves calculated by Padé approximant [12] reasonable good for \(m = 0.2\) and \(1/3\). For \(m = 0.2\) and \(U/E = 0.1\), the simulation results [11] showed a small “discontinuity” at the target surface in deed.

But, in both case \(m = 0.2\) and \(1/3\), one can see the momentum deposited at the target surface is positive as \(U/E = 0.1\). Therefore, \(F_{ij}(x, \eta)\) still can describe these depth distributions approximately at least in the sputtering theory concerned.
5.6 RECONSTRUCTION OF DEPTH DISTRIBUTIONS

5.6.1 THE SERIES FOR FOURIER TRANSFORM

L. G. Glazov introduced Fourier transforms $f(k, \eta)$ and $f_L(k)$ for $F(x, \eta)$ and $F_L(x)$ respectively

$$
\begin{align*}
\left\{ f(k, \eta) \right\} &= \int_{-\infty}^{\infty} e^{-ikx} \left\{ F(x, \eta) \right\} dx \\
\left\{ f_L(k) \right\} &= \int_{-\infty}^{\infty} e^{-ikx} \left\{ F_L(x) \right\} dk 
\end{align*}
$$

and the corresponding inverse Fourier transforms

$$
\begin{align*}
\left\{ F(x, \eta) \right\} &= \int_{-\infty}^{\infty} e^{ikx} \left\{ f(k, \eta) \right\} dk \\
\left\{ F_L(x) \right\} &= \int_{-\infty}^{\infty} e^{ikx} \left\{ f_L(k) \right\} dk
\end{align*}
$$

Substituting Taylor Series Expansions of Exponential Function

$$
\exp(z) = 1 + \frac{z}{1!} + \frac{z^2}{2!} + \frac{z^3}{3!} + \cdots 
$$

and $z = -ikx$ into (5.6.1), we obtained

$$
\begin{align*}
\left\{ f(k, \eta) \right\} &= \sum_{n=0}^{\infty} \left( \frac{-ik}{n!} \right)^n \left\{ F^n(\eta) \right\} \\
\left\{ f_L(k) \right\} &= \sum_{n=0}^{\infty} \left( \frac{-ik}{n!} \right)^n \left\{ F_L^n(\eta) \right\}
\end{align*}
$$

We repeated Glazov’s computations for the momentum deposition. The moments of the distribution were calculated according to the recurrence formulae (5.5.11) with high accuracy ($n = 300$ with 28-decimal-digits precision). Then the function $f(k, \eta)$ was calculated by using the formula (5.6.4) and plotted in Fig. 5.6.1. The same group of parameters taken by Glazov’s work: $m = 1/2, \eta = 1 (1), \eta = 0.6 (2)$ and $\eta = 0.4 (3)$ were used for the calculation. The present calculated curves agree with Glazov’s results very well up to $k = 30$, but diverge after $k = 37$. We rose the accuracy up to $n = 980$ with 500-decimal-digits precision, the formula (5.6.4) diverges after $k = 67$. This is not strange, Taylor Series Expansions of Exponential Function (5.6.3) converges in the calculus, but diverges in numerical calculation. This divergence is called Runge phenomenon in numerical calculation. It’s well known that Taylor Series Expansions of Exponential
Function can be approximated by a converged rational expression, called Padé approximant Method [91]. In order to avoid diverges, we have to turn to Padé approximant Method [2][11][20].

![Fourier transform of momentum deposition depth distribution. The curves obtained by calculating the formula (5.6.4) directly.](image)

**5.6.2 PADÉ APPROXIMANT METHOD**

Given a function \( f(z) = \sum_{i=0}^{\infty} c_i z^i \) and two integers \( m \geq 0 \) and \( n \geq 0 \), the Padé approximant of order \((m, n)\) is the rational function

\[
R_{nm}(z) = \frac{a_0 + a_1 z + \cdots + a_n z^n}{1 + b_1 z + \cdots + b_m z^m}
\]

which agrees with \( f(z) \) to the highest possible order, which amounts to

\[
R_{nm}^{(j)}(0) = f^{(j)}(0), \quad j = 0, 1, 2, \cdots, n + m
\]

Equivalently, if \( R_{nm}(z) \) is expanded in a Taylor Series at 0, its first \( n + m + 1 \) terms of \( R_{nm}(z) \) would cancel the first \( n + m + 1 \) terms of \( f(z) \), and as such

\[
f(z) - R_{nm}(z) = c_{n+m+1} z^{n+m+1} + c_{n+m+2} z^{n+m+2} + \cdots.
\]

The Padé approximant is unique for given \( m \) and \( n \), that is, the coefficients \( a_0, a_1, \cdots, a_n \) and
$b_1, b_2, \cdots, b_m$ can be uniquely determined by the following Padé equations,

$$
\begin{pmatrix}
c_m & c_{m-1} & \cdots & c_{m+n+1} \\
c_{m+1} & c_m & \cdots & c_{m+n+2} \\
\vdots & \vdots & \ddots & \vdots \\
c_{m+n-1} & c_{m+n-2} & \cdots & c_m \\
\end{pmatrix}
\begin{pmatrix}
b_1 \\
b_2 \\
\vdots \\
b_n \\
\end{pmatrix}
= -
\begin{pmatrix}
c_{m+1} \\
c_{m+2} \\
\vdots \\
c_{m+v} \\
\end{pmatrix},
$$

$$
\begin{pmatrix}
a_0 \\
a_1 \\
a_2 \\
\vdots \\
a_m \\
\end{pmatrix}
= 
\begin{pmatrix}
c_0 & 0 & 0 & \cdots & 0 \\
c_1 & c_0 & 0 & \cdots & 0 \\
c_2 & c_1 & c_0 & \cdots & 0 \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
c_m & c_{m-1} & c_{m-2} & \cdots & c_0 \\
\end{pmatrix}
\begin{pmatrix}
b_0 \\
b_1 \\
b_2 \\
\vdots \\
b_n \\
\end{pmatrix}.
$$

The Padé approximant defined above is also denoted as $\left[ \frac{n}{m} \right](z) \equiv R_{nm}(z)$ since $R_{nm}(z)$ is the rational function of $z$, it’s easy to determine the inverse Fourier transform, i.e., the depth distribution functions, as long as, $c_i = \frac{1}{i!} \left\{ F^i(\eta) \right\}$.

We have computed (5.6.2) by Padé approximant Method $\left[ \frac{490}{490} \right](z)$ with ultrahigh accuracy 500-decimal-digits precision for the Fourier transform of the momentum deposition depth distribution functions. The group of parameters: $m = 1/2$, $\eta = 1$ (1), $\eta = 0.8$ (2), $\eta = 0.6$ (3), $\eta = 0.4$ (4) and $\eta = 0$ (6) and were used for the calculation. The calculated curves $f(k, \eta)$ and $f_L(k)$ were plotted in Fig. 5.6.2 and Fig. 5.6.3 respectively. The calculated results showed that both $f(k, \eta)$ and $f_L(k)$ converge very well until $k = 630$ at least, therefore, Padé approximant Method is a powerful tool to research these depth distribution functions. The calculated results also demonstrated that

$$
\left| (2L+1)f_L(k) \right| < 0.001, \quad \text{for } L > 20.
$$

Neglecting the terms $f_{L>20}(k)$, (5.5.8) reduced to

$$
f(k, \eta) = \sum_{L=0}^{20} (2L+1)P_L(\eta)f_L(k) \quad (5.6.5)
$$

The calculated curves $f(k, \eta)$ by (5.6.4) and (5.6.5) agree each other very well.
Fig. 5.6.2 Fourier transform of momentum deposition depth distribution. The curves obtained by calculating the formula (5.6.4) with Padé approximant Method. The present calculated curves converge very well.

Fig. 5.6.3 Same as Fig. 5.6.2, but for $f_L(k)$, $L = 0, 1, 2, 3, 4, 5, 6, 7$. 
CHAPTER 6
TRANSPORT THEORY OF SPUTTERING

6.1 CLASSICAL SCATTERING CROSS-SECTION

6.1.1 BASIC FORMULAE

Consider an particle with mass $M_i$ and energy $E$, colliding with another particle at rest, with mass $M_k$, see Fig.2.1.2. For elastic collisions ($M_i \rightarrow M_k$), using the conservation law of energy and momentum yields formula (2.3.6) [18][44-49] and

$$X \equiv \frac{T}{T_m} = \cos^2 \varphi^* = \sin^2 \left( \frac{\theta_M}{2} \right) = \cos^2 \psi. \quad (6.1.1)$$

$T$ is the energy transfer (recoil energy) in the laboratory system (L.S.), (see Fig.2.1.2), $\theta_M$ is the scattering angle in the central mass system (C.M.S.), see Fig.6.1.1.

![Fig. 6.1.1 Scattering of a moving i-atom collides with a k-atom at rest in the center-of-mass system.](image)

We are normally concerned with a conservative and central force (described by a interaction potential function $V_{ik}(r)$), implying the conservation of angular momentum. Thus, the classical equation of motion can be derived [44],

$$\frac{d^2 u}{d\theta^2} + u + \frac{1}{2E_c b^2} \cdot \frac{dV}{du} = 0 \quad (6.1.2)$$

where, $(r, \theta)$ stands for the position of projectile in the polar coordinate system and
\[ r = r(\theta) \] represents the orbit. \( u \equiv 1/r, \ V = V_{ik}(1/u) \) and \( E_c = EM_k/(M_i + M_k) \) is the incident energy of projectile in (C.M.S.). The impact parameter \( b \) gives the initial condition:

If \( \theta = \pi \), then \( u = 0 \) and \( du/d\theta = 1/b \).

Equations (6.1.2,3) can be solved either analytically, or by a numerical calculation, such as Runge-Kutta.

Because the orbit must be symmetric about a line from the center to the nearest point, \( r = r_m \) on the orbit, see Fig.6.1.1. As \( r \) decreases from infinity to \( r_m \), i.e. \( u \) increases from zero to \( u = 1/r_m \), \( \theta \) decreases from \( \pi \) to \( \psi \), then, we obtained the relation between \( \psi \) and \( b \). Integrating Eq.(6.1.2) with the initial condition (6.1.3), we obtained

\[
\begin{align*}
\psi & = b \int_{r_m}^{\infty} \frac{dr}{\sqrt{r^2 (1-V(r)/E_c)} - b^2} \\
& = \int \left( 1 - \frac{V(r_m)}{E_c} - \left( \frac{b}{r_m} \right)^2 \right) = 0
\end{align*}
\]

(6.1.4)

For repulsive scattering, by changing the variable of integration to

\[
\rho^2 \equiv \left( r - r_m \right)/r = 1 - r_m/r \quad \text{or} \quad r = r_m/(1 - \rho^2).
\]

The second equation of (6.1.4) remains unchanging but the first one reduced to

\[
\psi = 2b \int_{0}^{1} \frac{d\rho}{\sqrt{(r_m/\rho)^2 - [V(r_m) - V(r)]/E_c + b^2 (2 - \rho^2)}}
\]

(6.1.5)

Since \( \lim_{r \to r_m} \left( V(r_m) - V(r) \right)/(r_m - r) = -F(r_m) \) is finite, the integration (6.1.5) allows naturally to use Gauss-Legendre quadrature schemes.

Solving (6.1.4) or (6.1.5) for \( b \) and using (6.1.1), we obtained \( b = b(E,X) \) and the energy transfer cross-section in L.S.

\[
d\sigma_{ik}(E,T) = -\pi db^2 = -(2\pi b d\theta dX) dX
\]

(6.1.6)

For a very large impact \( b, \theta_m << 1 \), the orbit is an almost straight line, the momentum approximation [49] can be used. Thus, it’s easy to derive the following expression [95] for \( \theta_m \),

\[
\theta_m = -\frac{b}{E_c} \int_{b}^{\infty} \frac{dr}{r \sqrt{1 - (b/r)^2}}
\]

(6.1.7)
6.1.2 SCALING POTENTIAL INTERACTION

For \( V_{ik}(r) = \chi_{ik} \Phi(x) \) and \( x = r/a_{ik} \), (6.1.4,5) read,

\[
\psi = 2b \int_0^1 \left\{ \frac{d\rho}{\sqrt{(x_M/\rho)^2 \cdot [\Phi(x_M) - \Phi(x)]/\epsilon + b^2 (2 - \rho^2)}} \right\}
\]

\[
1 - \Phi(x_M)/\epsilon - (b/x_M)^2 = 0
\]  

(6.1.8)

where, \( \rho^2 = 1 - x_M/x \); A natural energy scale is LSS-unit

\[
\epsilon_0 = 10^{-3} (M_i + M_k) \chi_{ik} / M_k \text{ (KeV)}
\]  

(6.1.9)

and the reduced energy is given by \( \epsilon = E(\text{keV})/\epsilon_0 \). Solving (6.1.8) for \( b \) and using (6.1.1), we obtained \( b = b(\epsilon, X) \) and the energy transfer cross-section in L.S.

\[
d\sigma_{ik}(E, T) = \pi a_{ik}^2 \cdot F(\epsilon, X) dX
\]  

(6.1.10)

Here \( F(\epsilon, X) = -2b \) \( d\rho/dX \) is the universal cross-section. J. Linhard introduced the reduced variable \[92\],

\[
t \equiv \epsilon \cdot X \quad \text{or} \quad X \equiv \epsilon \cdot t \epsilon^{-2}
\]  

(6.1.11)

and rewrote (6.1.10) in the following form

\[
d\sigma = 0.5 \pi a_{ik}^2 \cdot t^{-1.5} f(t^{0.5}) dt
\]  

(6.1.12)

thus, we obtained

\[
f(t^{0.5}) = (\pi a_{ik}^2)^{-1} \cdot t^{1.5} \frac{d\sigma}{dt} = 2t^{1.5} \epsilon^{-2} F(\epsilon, X) = 2t^{0.5} X F(\epsilon, X)
\]  

(6.1.13)

Let’s define the moments

\[
F^n(\epsilon) \equiv \int_0^1 X^n F(\epsilon, X) dX, \quad n = 0, 1, 2, 3, \ldots
\]  

(6.1.14)

then, the nuclear stopping power \( S_n(E) \) and the (partial) energy slow down cross-sections \( \sigma_{\gamma i}(E) \)

and \( \sigma_{\gamma j}(E) [13] \) in chapter 4 turned out to be

\[
\begin{align*}
[S_n(E)] &= K \cdot s_n(\epsilon) \\
K &= 10^{-3} \gamma_{ik} \pi a_{ik}^2 \epsilon_0 = 4\pi \chi_{ik} a_{ik}^2 M_i/(M_i + M_k)
\end{align*}
\]  

(6.1.15)
Here \( s_\gamma(\varepsilon) = eF^I(\varepsilon) \) is the reduced nuclear stopping power;

\[
\sigma'(\varepsilon) = \int_0^1 X \ln X \cdot F(\varepsilon, X) dX = \sigma'(\varepsilon, \gamma = 1)
\]

(6.1.19)

\[
\sigma^s(\varepsilon, \gamma \rightarrow 0) = F^I(\varepsilon)
\]

(6.1.20)

Formulae (6.1.15-17) and (6.1.19,20) have been published by Urbaskek et. al in ref.[13]. Substituting (6.1.15-20) into (4.2.25-29), we obtained all SC solutions for Thomas-Fermi scaling potential interaction.

Three special potential functions are particularly important:

i) Thomas-Fermi interaction potential [92]

In the case, \( V_{ik}(r) = \chi_{ik} x^{-1} \Phi_0(x), \quad x = r/a_{ik} \)

Here \( \chi_{ik} = Z_i Z_k Z / 32.523(\text{keV}) \), \( Z \equiv (Z_i^{2/3} + Z_k^{2/3})^{1/2}, \quad a_{ik} = 0.8853a_o / Z \),

the Bohr Radius \( a_o = 0.0523189379 \text{nm} \) and

\[
[\Phi_0(x)]^{-1} = 1 + 0.02747 x^{1/2} + 1.423 x - 0.1468x^{3/2} + 0.2302 x^2 + 0.007298 x^{5/2} + 0.006944 x^3
\]

(6.1.21)

which was given in Ref. [92] approximately. Thus, (6.1.9) and (6.1.16) turn out to be

\[
\begin{align*}
\varepsilon_0 &= Z_i Z_k Z (M_i + M_k) / (32.523 M_k) (\text{keV}) \\
K &= 84.445 Z_i Z_k M_i / [Z(M_i + M_k)] (\text{eV} \cdot 0.01 \text{nm}^2)
\end{align*}
\]

(6.1.22)
Inserting (6.1.21) into (6.1.8), the universal cross section $F(\varepsilon, X)$ and its moments (6.1.14) can be computed. But, about the stopping cross section, P. Sigmund had pointed out long time ago in Ref.[7], Thomas-Fermi reduced nuclear stopping power $s_n(\varepsilon)$, which is the one used, in comparison of experimental sputtering yield with theory, is known to be too high. For practical calculations, the reduced nuclear stopping power $s_n(\varepsilon)$ in (6.1.15) were simplified respectively to

$$s_n(\varepsilon) = \frac{0.5 \ln(1 + 1.1383\varepsilon)}{\varepsilon + 0.01321\varepsilon^{0.21226} + 0.19593\varepsilon^{0.5}}$$ \hspace{1cm} [53], \hspace{1cm} (6.1.23)

$$s_n(\varepsilon) = \frac{0.5 \ln(1 + 1.2288\varepsilon)}{\varepsilon + 0.0081\varepsilon^{0.1504} + 0.1728\varepsilon^{0.5}}$$ \hspace{1cm} [102], \hspace{1cm} (6.1.24)

$$s_n^{K-C}(\varepsilon) = \frac{0.5 \ln(1 + 1.2288\varepsilon)}{\varepsilon + 0.1728\sqrt{\varepsilon} + 0.0081\varepsilon^{0.1504}}$$ \hspace{1cm} [94], \hspace{1cm} (6.1.25)

$$s_n(\varepsilon) = \frac{3.441\sqrt{\varepsilon} \ln(\varepsilon + 2.718)}{1 + 6.355\sqrt{\varepsilon} + \varepsilon(1.708 + 6.882\sqrt{\varepsilon})}$$ \hspace{1cm} [82], \hspace{1cm} (6.1.26)

We have plotted the reduced nuclear stopping power $s_n(\varepsilon)$ expressed by (6.1.23-26) respectively in Fig. 6.1.2 as a function of the reduced energy and some corresponding experiment results have been also plotted for comparison. The electronic stopping curve has been included in Fig. 6.1.2. One can see that both (6.1.23,24) fit the experiment points very well [102] for $\varepsilon \leq 0.1$. This was the reason we prefer to use (6.1.23) for sputtering calculation [73]. (6.1.25), representing the system Kr-C, predicts the higher value for $s_n(\varepsilon)$ in the same reduced energy region. (6.1.26), Yamamura et. al liked to use for sputtering theory research [82], even worse than (6.1.25). The electronic stopping ($s_e(\varepsilon) = 0.1\varepsilon^{0.5}$) can be neglected in the low energy region.

ii) Born-Mayer interaction potential [57]

In the case, $V(r) = A \exp(-r/a)$, \hspace{1cm} (6.1.27)

Here $A = 52(Z_iZ_k)^{3/4}eV$, $a = 0.0219nm$, then, (6.1.9) and (6.1.15) turned out to be

$$\begin{aligned}
\varepsilon_0 &= 52(Z_iZ_k)^{3/4}(M_i + M_k)/(1000M_k)(keV) \\
K &= 208\pi a^2(Z_iZ_k)^{3/4}M_i/(M_i + M_k)\cdot(eV \cdot 0.01nm^2)
\end{aligned} \hspace{1cm} (6.1.28)$$
Fig. 6.1.2 Nuclear stopping power as function of particle energy in LSS-units. The small circles denote experimental data; the letters at the theoretical curves designate: A, B, C and D were calculated by (6.1.23), (6.1.24), (6.1.25) and (6.1.26) respectively.

Inserting \( \exp(-r/a) \) into (6.1.8), the universal cross section \( F(\varepsilon, X) \) and its moments (6.1.14) can be computed and plotted in the coordinate system \([t^{1/2}, f(t^{1/2})]\). For convenience and reliability, we just made a copy from [57] for further comparison, (see part (b) of Fig. 6.2.1).

iii) Kr-C interaction potential [13]

If only the first three moments are considered in (6.1.17),

\[
\sigma^* (\varepsilon, \gamma) = F^1(\varepsilon) - (\gamma/2)F^2(\varepsilon) - (\gamma^2/6)F^3(\varepsilon)
\]  

(6.1.29)

Specially, we have

\[
\begin{aligned}
\sigma^*(\varepsilon, 1) &= F^1(\varepsilon) - (1/2)F^2(\varepsilon) - (1/6)F^3(\varepsilon) \\
\sigma^*(\varepsilon, 1/2) &= F^1(\varepsilon) - (\gamma/4)F^2(\varepsilon) - (\gamma^2/24)F^3(\varepsilon)
\end{aligned}
\]  

(6.1.30)

Solving Eqs. (6.1.30) yields,

\[
\begin{aligned}
F^2(\varepsilon) &= 6F^1(\varepsilon) - 8\sigma^*(\varepsilon, 1/2) + 2\sigma^*(\varepsilon, 1) \\
F^3(\varepsilon) &= -12F^1(\varepsilon) + 24\sigma^*(\varepsilon, 1/2) - 12\sigma^*(\varepsilon, 1)
\end{aligned}
\]  

(6.1.31)

Substituting (6.1.31) into (6.1.29), we obtained

\[
\sigma^*(\varepsilon, \gamma) = (1 - 3\gamma + 2\gamma^2) \cdot s_\varepsilon(\varepsilon)/\varepsilon \\
+ 4\gamma(1 - \gamma)\sigma^*(\varepsilon, 1/2) + \gamma(2\gamma - 1)\sigma^*(\varepsilon, 1)
\]  

(6.1.32)
Fig. 6.1.3 Calculated curves of $\Gamma(\varepsilon, \gamma)$ for Kr-C interaction potential [13] as a function of reduced energy. (6.1.33) was used for the calculation. Numerical results of scaled cross-sections were taken from ref.[13]. Two short straight lines were calculated by (6.3.19) and (6.3.20) respectively.

predicts the higher value for $s_n(\varepsilon)$ in the same reduced energy region. (6.1.26), Yamamura et. al liked to use for sputtering theory research [82], even worse than (6.1.25). The electronic stopping $(s_e(\varepsilon) = 0.1e^{0.5})$ can be neglected in the low energy region.

Scaled cross-sections $s_n(\varepsilon)/\varepsilon$, $\sigma^t(\varepsilon)$, $\sigma^s(\varepsilon, 1/2)$ and $\sigma^s(\varepsilon, 1)$ for the Kr-C interaction potential have been plotted by Urbassek et. al (see Fig.6 in ref.[13]). Inserting $s_n(\varepsilon)/\varepsilon$, $\sigma^t(\varepsilon)$ and (6.1.32) into (6.1.15-17), we obtained the nuclear stopping power $S_n(E)$ and the (partial) energy slow down cross-sections $\sigma^s_y(E)$ and $\sigma^s_y(E)$. Substituting theses cross sections into VCU Eqs. (4.3.1), we have calculated the energy distribution of recoils in a HfC compound, bombarded by incident Hf or C atoms with energy 6 keV respectively. The calculated results agree with the correspondent curves in Fig. 3 of ref.[13] very well. These agreements demonstrated that (6.1.32) is a good approximate formula. According to Urbassek’s date [13], we have calculated

$$\Gamma(\varepsilon, \gamma) = \frac{\bar{s}(\varepsilon)/\varepsilon}{\bar{\sigma}^s(\varepsilon, \gamma) + \bar{\sigma}^t(\varepsilon, \gamma)},$$

(6.1.33)

and plotted it in Fig. 6.1.3 as a function of the reduced energy $\varepsilon$ for further comparison.
6.2 TWO CROSS-SECTIONS OF POWER POTENTIAL INTERACTION

6.2.1 EXACT CLASSICAL SCATTERING CROSS-SECTION

For a power potential interaction \( i \rightarrow j \), substituting

\[
V_{ik}(r) = \chi_{ik} r^{-\gamma}, \quad \text{for} \quad l \geq m = l/s > 0
\]  

(6.2.1)

and new variables

\[
\left( \frac{y}{t} \right) \equiv \left( \frac{E_s}{\chi_{ij}} \right)^m \left( \frac{r_M}{b} \right)
\]

into (6.1.4), for \( l \geq m > 0 \), we obtained [18][87][95]

\[
\begin{align*}
\psi &= ty^{0.5s-1} \int_0^t dx \frac{1}{\sqrt{y^s - t^2 y^{s-2} x^2 + x^s}} \\
y^s &= t^2 y^{s-2} + 1
\end{align*}
\]  

(6.2.2)

Where \( m \) depends on incident particle in general. The “up sign” and the “low sign” in (6.2.2) stand for repulsive and attractive respectively. Obviously, (6.2.2) determine \( \psi = \psi(t) \) or \( t = t(X) \)

because of (6.1.1), thus, a universal scattering cross-section can be defined,

\[
F_m(X) = -\pi dt^2 / dX = -2\pi t dt / dX
\]  

(6.2.3)

According to (6.1.6), the energy transfer cross-section turns out to be
(6.2.4)
\[
d\sigma_{ik}(E, T) = \left(\chi_{ik}/E_c\right)^{2m} F_m(X) dX = C_{ik} \gamma_{ik}^{-m} Z_m(X) X^{1-m} dX
\]

Here \( C_{ik} \equiv \pi m (s_{ik} s_{ik}) \cdot (M_i/M_j)^m \). (6.2.5)

\[
Z_m(X) \equiv (\pi m)^{-1} (2m/s)_{ik}^{2m} F_m(X) X^{1-m}
\]

and \( \gamma_{ik} \equiv 0.5B[0.5, 0.5(s+1)] \), (see Table 6.2.1 and Table 6.2.2.)

One extremely important case is \( m = 0 \) for the repulsive scattering, (6.2.2) fails. We noticed that the following limit exists: \( d_{ik} \equiv \lim_{m \to 0} [(\chi_{ik})^m] \), then

\[
\lim_{m \to 0} [V_{ik}(r)] = \lim_{m \to 0} [\chi_{ik} r^{-3}] = \lim_{s \to \infty} \lim_{r \to 0} (d_{ik}/r)^s = \begin{cases} 0, & \text{if } r > d_{ik} \\ \infty, & \text{if } r < d_{ik} \end{cases}
\]

which stands for the hard sphere interaction [97] with the radius \( d_{ik} \). Therefore, only considering the geometry, we have:

\[
b^2 = (d_{ik})^2 \sin^2 \psi = (\sigma_{ik}/\pi)(1 - X), \quad \sigma_{ik} \equiv \pi (d_{ik})^2
\]

According to (6.1.6), the energy transfer cross-section turns out to be

\[
d\sigma_{ik}(E, T) = -\pi d^2 = \sigma_{ik} dX
\]

Obviously, the total cross-section is finite.

<table>
<thead>
<tr>
<th>( S )</th>
<th>( \gamma_S )</th>
<th>( m )</th>
<th>( p )</th>
<th>( q )</th>
<th>( [\Gamma_m]_Z )</th>
<th>( \Gamma_m )</th>
<th>( [\Gamma_m]_P )</th>
<th>( Z_m(I) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \infty )</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>1</td>
<td>0.6079</td>
<td>( \infty )</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>0.122</td>
<td>0.01</td>
<td>0.03978</td>
<td>0.8525</td>
<td>0.969</td>
<td>0.968</td>
<td>0.604</td>
<td>93.84</td>
</tr>
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<td>50</td>
<td>0.173</td>
<td>0.02</td>
<td>0.08447</td>
<td>0.8388</td>
<td>0.939</td>
<td>0.939</td>
<td>0.599</td>
<td>44.67</td>
</tr>
<tr>
<td>20</td>
<td>0.277</td>
<td>0.05</td>
<td>0.2116</td>
<td>0.8649</td>
<td>0.862</td>
<td>0.862</td>
<td>0.586</td>
<td>15.85</td>
</tr>
<tr>
<td>10</td>
<td>0.387</td>
<td>0.1</td>
<td>0.3667</td>
<td>0.9522</td>
<td>0.764</td>
<td>0.763</td>
<td>0.563</td>
<td>6.837</td>
</tr>
<tr>
<td>5</td>
<td>0.533</td>
<td>0.2</td>
<td>0.6059</td>
<td>1.1093</td>
<td>0.617</td>
<td>0.620</td>
<td>0.516</td>
<td>2.835</td>
</tr>
<tr>
<td>2</td>
<td>0.785</td>
<td>0.5</td>
<td>0.9768</td>
<td>1.4984</td>
<td>0.363</td>
<td>0.363</td>
<td>0.361</td>
<td>1.032</td>
</tr>
</tbody>
</table>
\[
\int_0^{T_m} d\sigma_{ik}(E,T) = \sigma_{ik} \int_0^1 dX = \sigma_{ik}
\]

(6.2.8)

Thus, the power potential interaction (6.2.1) has been extended to \( m = 0 \). Therefore, (6.2.4) and (6.2.7) turned out to be the exact differential scattering cross-section \([18]\),

\[
d\sigma_{ik}(E,T) = \begin{cases} 
C_{ik} E^{-m} Z_m(X) T^{-m} dT & \text{for } \ 1 \geq m > 0 \\
\sigma_{ik} T^{-1} dT, & \text{for } \ m = 0
\end{cases}
\]

(6.2.9)

Except the hard sphere interaction, there are still two exact differential scattering cross-sections can be written analytically or in closed form:

i) For Rutherford Scattering \( (s = m = 1) \),

Solving the second equation of (6.2.2) yielded

\[
y = 0.5\left(\sqrt{1 + 4t^2} \pm 1\right)
\]

(6.2.10)

Substituting (6.2.10) into the first equation of (6.2.2) and completing the integration, we obtained

\[
y = 0.5\left(\sqrt{1 + 4t^2} \pm 1\right)
\]

(6.2.10)

\[
\psi = 0.5\pi \pm \arcsin(1 + 4t^2)^{-0.5}, \quad \text{or} \quad \cos \psi = \pm (1 + 4t^2)^{-0.5}
\]

thus, (6.1.1) turns out to be

Table 6.2.2 Calculated values of \( \gamma_s, p, q, \Gamma_m \) and \( Z_m(1) \) by using (6.2.23)

<table>
<thead>
<tr>
<th>( S )</th>
<th>( \gamma_s )</th>
<th>( m )</th>
<th>( p )</th>
<th>( q )</th>
<th>( [\Gamma_m]_Z )</th>
<th>( \Gamma_m )</th>
<th>( [\Gamma_m]_p )</th>
<th>( Z_m(1) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>0.277</td>
<td>0.05</td>
<td>13.65</td>
<td>0.7503</td>
<td>0.840</td>
<td>0.862</td>
<td>0.586</td>
<td>15.85</td>
</tr>
<tr>
<td>10</td>
<td>0.387</td>
<td>0.1</td>
<td>5.327</td>
<td>0.6214</td>
<td>0.741</td>
<td>0.763</td>
<td>0.563</td>
<td>6.837</td>
</tr>
<tr>
<td>7</td>
<td>0.457</td>
<td>1/7</td>
<td>3.184</td>
<td>0.6067</td>
<td>0.687</td>
<td>0.695</td>
<td>0.543</td>
<td>4.353</td>
</tr>
<tr>
<td>5</td>
<td>0.533</td>
<td>0.2</td>
<td>1.796</td>
<td>0.5820</td>
<td>0.620</td>
<td>0.621</td>
<td>0.516</td>
<td>2.835</td>
</tr>
<tr>
<td>4</td>
<td>0.589</td>
<td>0.25</td>
<td>1.125</td>
<td>0.5483</td>
<td>0.565</td>
<td>0.565</td>
<td>0.492</td>
<td>2.144</td>
</tr>
<tr>
<td>3</td>
<td>0.667</td>
<td>1/3</td>
<td>0.5226</td>
<td>0.5145</td>
<td>0.488</td>
<td>0.489</td>
<td>0.450</td>
<td>1.523</td>
</tr>
<tr>
<td>2</td>
<td>0.785</td>
<td>0.5</td>
<td>0.0314</td>
<td>0.7764</td>
<td>0.363</td>
<td>0.363</td>
<td>0.361</td>
<td>1.032</td>
</tr>
</tbody>
</table>
\[ X = \cos^2 \psi = (1 + 4t^2)^{-1}, \quad \text{or} \quad t^2 = 0.25(X^{-1} - 1) \]

then, (6.2.3) reduced to
\[ F_m(X) = 0.25 \pi X^{-2} \quad \text{(6.2.11)} \]

finally, substituting (6.2.11) into (6.2.6) yielded
\[ Z_1(X) = 1 \quad \text{(6.2.12)} \]

ii) For Bohr formula [96] \( (s = 2, \ m = 0.5) \)

(6.2.2) reduced to
\[
\begin{cases}
\psi = t \int_0^1 \frac{dx}{\sqrt{y^2 - (t^2 \pm 1)x^2}} \\
y^2 = t^2 \pm 1
\end{cases}
\]

Solving it yielded
\[
\psi = \pm \frac{t}{\sqrt{t^2 \pm 1}} \cdot \frac{1}{\sqrt{1 - x^2}} = \frac{\pi}{2} \cdot \frac{t}{\sqrt{t^2 \pm 1}}, \quad \text{or}
\]
\[
t^2 = \pm \frac{\psi^2}{(\pi^2/4 - \psi^2)}
\]

thus, (6.2.3) reduced to
\[
F_{0.5}(X) = \frac{\pi^3}{8} \cdot \frac{\psi}{\sqrt{X(1 - X)}} \cdot \frac{d\psi}{dX}
\]

Considering (6.1.1), \( X = \cos^2 \psi \), or
\[
\psi = 0.5\pi \mp \sin^{-1} \sqrt{X}, \quad \left( \sin^{-1} z \equiv \arcsin z \right), \quad \text{then}
\]
\[
\frac{d\psi}{dX} = \mp \frac{1}{2\sqrt{X(1 - X)}}
\]
\[
F_{0.5}(X) = \frac{\pi^3}{8\sqrt{X(1 - X)}} \cdot \frac{\psi}{\left(\pi^2/4 - \psi^2\right)} \quad \text{(6.2.13)}
\]

Substituting (6.2.13) into (6.2.6) yielded
\[
Z_{0.5}(X) = 2\left(2\pi f_2\right)^{-1} F_{0.5}(X) X^{1.5}
\]
\[
=\frac{\pi X \left(\pi \mp 2 \sin^{-1} \sqrt{X}\right)}{\sqrt{1 - X} \cdot \left(\sin^{-1} \sqrt{X}\right)^2} \left(\pi \mp \sin^{-1} \sqrt{X}\right)^2 \quad \text{(6.2.14)}
\]
Two limits of \( Z_{0.5}(X) \) may be interest:

\[
Z_{0.5}(X \to 0) = 1
\]  \hspace{1cm} (6.2.15)

\[
Z_{0.5}(X \to 1) = \begin{cases} 
32\pi^{-3} = 1.032049102\cdots, & \text{for repulsive scattering} \\
(32/9)\cdot \pi^{-2}/(1 - X) \to \infty, & \text{for attractive scattering}
\end{cases}
\]  \hspace{1cm} (6.2.16)

### 6.2.2 IMPACTS BOTH LARGE AND SMALL

Although, it’s not possible to obtain closed form for all exact scattering cross-sections, we can still derive two asymptotic behaviors for impacts both large and small:

i) Small angle scattering \([45][89][92]\)

If the impact parameter \( b \) is very large, the angle scattering \( \theta_m \sim 0 \). Substituting \( b = t \), \( E_c = 1 \) and \( V(r) = \pm r^{-s} \) into (6.1.7) yielded \( \theta_m = \pm (s \gamma_s)^{1-s} \), then, using (6.1.1), we have

\[
X = \sin^2 \left( \frac{\theta_m}{2} \right) = \left( \frac{\theta_m}{2} \right)^2 = (0.5s \gamma_s)^2 t^{-2s} \to 0 \quad \text{and}
\]

\[
t^2 = (0.5s \gamma_s)^{2m} X^{-m}
\]

Inserting it into (6.2.3) yielded

\[
F_m(X \to 0) = -\pi \frac{dt^2}{dX} = \pi m(0.5s \gamma_s)^{2m} X^{-m-1}
\]

then, (6.2.6) reduced to

\[
Z_m(0) = 1
\]  \hspace{1cm} (6.2.17)

for both attractive and repulsive scattering. (6.2.15) is the special case of (6.2.17).

ii) Large angle repulsive scattering

If the impact parameter \( b \) is very small, i.e. both \( \psi \) and \( t \sim 0 \), the solution of Eq. (6.2.2)

\[
\psi = t \int_0^t dx \left( 1 - x^s \right)^{-0.5} = tmB(m, 0.5).
\]

Thus (6.1.1) reads

\[
X = \cos^2 \psi = 1 - \sin^2 \psi = 1 - \psi^2 = 1 - t^2[mB(m, 0.5)]^2 \to 1, \quad \text{or}
\]

\[
t^2 = (1 - X)[mB(m, 0.5)]^{-2}.
\]

Inserting it into (6.2.3) yielded
\[ F_m (X \rightarrow 1) = -\pi \frac{dt^2}{dX} = \pi \left[ mB(m, 0.5) \right]^2 = [\Gamma(0.5 + m)/\Gamma(1 + m)]^2 \]

then, (6.2.6) reduced to

\[ Z_m (X \rightarrow 1) = (\pi m)^{-1} (2m/\gamma_x)^{2m} [\Gamma(0.5 + m)/\Gamma(1 + m)]^2 X^{1+m} \text{ and} \]

\[ Z_m (1) = (\pi m)^{-1} (2m/\gamma_x)^{2m} [\Gamma(0.5 + m)/\Gamma(1 + m)]^2. \quad (6.2.18) \]

Substituting \( m = 0.5 \) into (6.2.18) yields \( Z_{0.5}(1) = 32\pi^{-3} \) which agrees with (6.2.16). The boundary values (6.2.18) are entirely consistent with the errors in the power approximation which were given by Lindhard et al. in ref.[46]. Some of numerical results of \( Z_m (1) \) are collected in Table 6.2.1, 2.

### 6.2.3 DOUBLE AND SIMPLE POWER CROSS-SECTIONS

In general, we have to use the numerical computation method. It’s similar to (6.1.5), by changing the variable of integration and using (6.1.1), (6.2.2) reduced to

\[
\begin{align*}
X &= \cos^2 \left[ 2t \int_0^1 \frac{dp}{\sqrt{t^2 (2 - \rho^2) \pm d(\rho^2)y^{-s}}} \right] \\
y^s &= t^2 y^{-s-2} \pm 1, \quad d(x) = x^{-1}[1-(1-x)^s] 
\end{align*}
\]

Since \( \lim_{x \to 0} d(x) = 0 \), the integration can be completed by using Gauss-Legendre quadrature schemes and defines \( t = t(X) \).

On the other hand, we noticed that (6.2.3) can be considered as a scattering cross-section of one particle with energy \( E_c = 1 \) in a central field \( V(r) = \pm r^{-s} \). (6.1.2) and (6.1.3) turn out to be

\[
\begin{align*}
\frac{du}{d\theta} &= v \\
\frac{dv}{d\theta} &= -u + 0.5st^{-2}u^{-1} 
\end{align*}
\]

with the initial condition: if \( \theta = \pi \),

\[
\begin{align*}
u &= 0 \\
v &= 1/t 
\end{align*}
\]

(6.2.20) and (6.2.21) can be solved by any numerical methods, such as Runge-Kutta, and define \( u = u(\theta) \). For each \( t \), there must be a \( \text{Max}(u) = u(\theta_M) \), thus, \( t = t(X) \) is determined.
Finally, substituting \( t = t(X) \) into (6.2.3) and (6.2.6) generated \( Z_m(X) \). The program had been written in BASIC. \( Z_m(X) \) had been computed for different \( m \) \((1 > m > 0)\) and repulsive scattering. The calculated results for \( Z_m(X) \) form a Data Table for further use, and some numerical results are plotted in Fig. 6.2.2. Even if \( Z_m(X) \) may be calculated to any accuracy, it is still inconvenient to use for some applications. However, we found that \( Z_m(X) \) can be approximated by

\[
Z_m(X) \approx X^{1+m} Z_m(1)[(1-p) + pX^{-q}] \quad \text{for } 0.2 > m \geq 0 \tag{6.2.22}
\]

\[
Z_m(X) \approx 1 + pX^q \quad \text{for } 1 \geq m > 0.1. \tag{6.2.23}
\]

where \( p \) and \( q \) are such parameters that make (6.2.22,23) best fit the numerical curves of \( Z_m(X) \). Some calculated data for the \( p, q \) parameters in (6.2.22,23) are collected in Table 6.2.1,2 respectively. The corresponding analytical results (6.2.22,23) are plotted in parts (a) and (b) of Fig. 6.2.2 respectively for comparison. One can see that (6.2.22,23) fit the numerical curves well. Substituting (6.2.22,23) into (6.2.9) respectively, we obtained the double power cross sections. In contrast with (6.2.9), the well know Lindhard simple power cross-section, was given a long time ago [1][7][46][49].

**Fig. 6.2.2** Universal correction function \( Z_m(X) \) defined in the differential cross section (6.2.9).

Thick line: Numerical results calculated by (6.2.2,3) and (6.2.6). Thin line: Approximate analytical results calculated by (6.2.22) in (a) and by (6.2.23) in (b).
for the same power potential interaction (6.2.1). A large discrepancy can be seen between $Z_m(X)$ and unity for $0.5 > m \geq 0$ in Fig. 6.2.1 [46]. In other words, (6.2.24) couldn’t properly describe the scattering of a power potential interaction (6.2.1) for $m \sim 0$ [46]. In addition, by using the momentum approximation [49], Lindhard derived (6.2.24) for $1 \geq m > 0$, just as (6.2.17). But, this approximation can’t be suitable for the hard sphere interaction [97], therefore, simply extending (6.2.24) to $m = 0$ may be still questionable.

Obviously, scattering cross-sections (6.2.9) and (6.2.24) satisfy (5.1.1), as long as defined

$$[A_{ij}(X)]_Z = \begin{cases} C_{ij}^{-m} Z_m(X) X^{-1-m}, & \text{for } 1 \geq m > 0 \\ \sigma_{ij}, & \text{for } m = 0 \end{cases}$$  \hspace{1cm} (6.2.25)$$

$$[A_{ij}(X)]_P = C_{ij}^{-m} X^{-1-m}$$  \hspace{1cm} (6.2.26)$$

Hereafter, $[H]_Z$ and $[H]_P$ stand for the results of $H$ calculated by (6.2.25) and (6.2.26) respectively. Naked $H$ stands for the results calculated by both. Thus, all of the solutions appeared in previous chapter can be evaluated.

### 6.2.4 BOTH SCATTERING CROSS-SECTIONS FOR REAL INTERACTION

For a real potential interaction collision, the approximate classical scattering cross section is derived in two steps: The first step is to approximate the real interaction potential by a power form (6.2.1). This is feasible over limited ranges of $r$ [49]. The second step is to derive the classical scattering cross section from the power potential (6.2.1). It is clear that the error in the presently used cross sections only originates from the first step, while, the error in the simple power cross section comes from both the first and second step. Even so, the simple power cross section could still provide the better approximation due to some possible error cancellations. In 1969, regarding (6.2.24) as exact, Robinson derived the corresponding interatomic potential function by using an inversion procedure [98]. Robinson found that the exact potential approaches the power potential limit (6.2.1) at high energy and large interatomic separation. However, the author also realized that
the exact potentials are softer than the asymptotic forms and significantly energy dependent. Both features become more pronounced, as \( s \) increases. Robinson’s findings are intrinsically consistent with those of the present work. Therefore, for a low energy incidence, \( m \sim 0 \), (6.2.24) couldn’t describe an energy independent collision.

As examples, let’s consider the Born-Mayer interaction [57]. In this case, the interatomic potential (6.1.23) is given by

\[
V(r) = A \exp(-r/a) \sim (A k_s a^s) r^{-s} \quad \text{or} \quad V(r)/A = \exp(-r/a) \sim k_s (r/a)^{-s}
\]

for \( 0 < m < 4/3 \). Then, (6.2.5) reduced to

\[
C_{ik} = 0.5 \pi \alpha_m a^2 \left( M_i/M_k \right)^m \cdot (2A)^2 m
\]

\[
\lambda_m = 2m \left( 0.5 s \gamma_s k_s \right)^2 m
\]

Using (6.2.9), (6.2.24) and (6.2.28, 29), it is easy to derive the reduced nuclear-stopping [1] as a function of reduced energy \( \varepsilon \)

\[
\left[ S_n(\varepsilon) \right]_Z = 0.5 \left[ \lambda_m \right]_Z \cdot \varepsilon^{1-2m} \cdot \int Z_m(X) X^{-m} dX
\]

\[
\left[ S_n(\varepsilon) \right]_p = 0.5 \left[ \lambda_m \right]_p \cdot \varepsilon^{1-2m} \cdot 1/(1-m)
\]

and the Lindhard scaling function in scattering cross section [1]

\[
\left[ f(t^{1/2}) \right]_Z = \left[ \lambda_m \right]_Z \cdot Z_m \left( t \varepsilon^{-2} \right) (t^{1/2})^{1-2m}, \quad t \equiv X \varepsilon^{-2}
\]

\[
\left[ f(t^{1/2}) \right]_p = \left[ \lambda_m \right]_p \cdot (t^{1/2})^{1-2m}
\]

Matching both \([S_n(\varepsilon)]_Z\) and \([S_n(\varepsilon)]_p\) with the numerical curve of \( S_n(\varepsilon) \) in part (a) of Fig. 6.2.1, we obtained,

\[
[S_n(\varepsilon)]_Z = [S_n(\varepsilon)]_p = 6.59 \cdot \varepsilon^{0.8} \quad \text{for} \quad 10^{-2} > \varepsilon > 10^{-5}
\]

Thus, (6.2.28-31), (6.2.33) and TABLE 6.2.1 give all the necessary parameters.
$m = 0.1, \quad p = 0.3667, \quad q = 0.9522$

$$[\lambda_m]_x = 3.05 \quad \text{and} \quad [\lambda_m]_p = 12.5 \quad (6.2.34)$$

Inserting these parameters (6.2.34) back into (6.2.32) yielded

$$\left[ f(t^{1/2}) \right]_x = 13.2 \cdot \varepsilon^{0.8} (t^{1/2} \cdot \varepsilon^{-1})^3 \cdot [1 + 0.579 \cdot (t^{1/2} \cdot \varepsilon^{-1})^{-1.904}]$$

$$\left[ f(t^{1/2}) \right]_p = 12.5 \cdot (t^{1/2})^{0.8}$$

which is plotted $S_n(\varepsilon)$ in part (b) of Fig. 6.2.1 for comparing with the numerical curves of $f(t^{1/2})$. It is clear that $[f(t^{1/2})]_x$ fits them very well, for $10^{-2} \geq \varepsilon \geq 10^{-5}$, while, $[f(t^{1/2})]_p$ does not.

In both parts (a) and (b) of Fig. 6.2.1, the numerical curves $S_n(\varepsilon)$ and $f(t^{1/2})$ are copied from a and b of Figure 6 in ref. [57].

On the other hand, inserting parameters (6.2.34) back into (6.2.29) yielded,

$$[k_s]_x = 4.062 \cdot 10^5$$

$$[k_s]_p = 4.938 \cdot 10^8 \quad (6.2.35)$$

Substituting (6.2.35) into (6.2.27) we obtained

$$\left[ V(r)/A \right]_x = 4.062 \cdot 10^5 \cdot (a/r)^{10} \quad (6.2.36)$$

$$\left[ V(r)/A \right]_p = 4.938 \cdot 10^8 \cdot (a/r)^{10} \quad (6.2.37)$$

Plotting (6.2.36,37) in part (c) of Fig. 6.2.1, it's clearly to see that, $\left[ V(r)/A \right]_x$ curve (6.2.36) fits (6.2.27) very well, within the region $20 > r/a > 5$. But, $\left[ V(r)/A \right]_p$ curve (6.2.37) is too high, about 1000 times than (6.2.27)! On the other hand, let's look at this point from the opposite direction, instead of (6.2.37), taking (6.2.36) as a power function to approximate the Born-Mayer potential (6.2.27), i.e. inserting $\lambda_m = 3.05$ into the second equation of (6.2.31), we will get $S_n(\varepsilon) = 1.678 \cdot \varepsilon^{0.8}$, which is too low to match the numerical curve in part (a) of Fig.6.2.1.

Therefore, just using the cross-section (6.2.24), it's not possible to choose such a value of $k_s$ (or $\lambda_m$) that both numerical curves in both parts (a) and (c) of Fig. 6.2.1 can be fit simultaneously.
6.3 TWO CROSS-SECTIONS IN TRANSPORT THEORY OF SPUTTERING

6.3.1 INTRINSIC DIFFERENCES BETWEEN

Substituting (6.2.25) and (6.2.26), we derived $\Gamma_m \equiv \Gamma_i$,

$$\left[ \Gamma_m \right]_Z = \left[ 1 - \langle |\ln x| \rangle_m - \sum_{n=1}^{\infty} \frac{1}{n(n+1)} \langle |x^n| \rangle_m \right]^{-1}, \quad \text{if } 1 \geq m > 0 \quad (6.3.1).$$

$$\left[ \Gamma_m \right]_P = \left[ 1 + \frac{1}{(1-m)} - (1-m) \sum_{n=1}^{\infty} \frac{1}{n(n+1)(n+1-m)} \right]^{-1}$$

$$= m/\Psi(1) - \Psi(1-m)], \quad \Psi(x) \equiv d\log \Gamma(x)/dx. \quad (6.3.2).$$

$$\left[ \Gamma_0 \right]_Z = 1 \quad \text{ (6.3.3).}$$

$$\left[ \Gamma_0 \right]_P = 6\pi^2 \approx 0.6079. \quad \text{ (6.3.4)}$$

$$\left[ \Gamma_1 \right]_Z = \left[ \Gamma_1 \right]_P = 0 \quad \text{ (6.3.5)}.$$

Here $\langle |f(X)| \rangle_m = \left[ \int_0^1 dX Z_m(X)X^{-m} \right]^{-1} \int_0^1 dX Z_m(X)X^{-m} f(X)$.

Both $[\Gamma_m]_Z$ and $[\Gamma_m]_P$ have been calculated and plotted in Fig.6.3.1 as continuous functions of $m$ for $1 \geq m \geq 0$. Some of numerical results of $\Gamma_m$ are collected in Table 6.2.1, where $[\Gamma_m]_Z$ and $[\Gamma_m]_P$ are calculated by (6.3.1-5) respectively. It is clear that $[\Gamma_m]_Z \approx [\Gamma_m]_P$ for $1 \geq m > 0.5$. However, a great discrepancy can be seen for $0.25 \geq m \geq 0$. Certainly, this discrepancy will certainly cause some intrinsic differences in the transport theory. For simplicity, only a monatomic medium is concerned for the following discussions.

![Fig. 6.3.1 Calculated curve of $\Gamma_m$ as a continuous function of $m$, defined by (6.3.1–3). Thick line: calculated by Exact theory (6.3.1,3). Thin line: calculated by Sigmund theory (6.3.2).](image-url)
6.3.2 NUMBER OF RECOILS

The number of recoils \( N^+(E_0) \) was introduced by Urbassek et.al. in ref.[13] as the average number of particles slowing down via collision from an energy \( E > E_0 \) to energy below \( E_0 \):

\[
N^+(E_0) = \int_{E_0}^{E} \int_{E_0}^{E'} dE' \Psi(E') \int_{E-E_0}^{E'} d\sigma(E',T)
\]

which has been given by (2.2.11), (2.3.8), (3.3.7) and (3.3.23). Therefore, substituting (6.2.25,26) into (3.3.23), the asymptotic formula for \( N^+(E_0) \) can be derived

\[
N^+(E_0) = \Gamma_m E^{m-1} \quad \text{for} \quad E > E_0
\]

(6.3.6)

which makes an opportunity of measuring \( \Gamma_m \). In ref.[13], Urbassek et. al. assigned a local power exponent \( m \) to the Kr-C stopping cross section: \( m = 0.12 \) for Hf-Hf collisions and \( m = 0.23 \) for C-C collisions at 10 eV interaction energy. Thus, the number of recoils \( N^s \) generated above 10 eV in a collision cascade initiated by 100 keV Hf ion is predicted by Eq.(6.3.6),

\[
[N^s]_Z = 7290, \quad [N^s]_p = 5500 \quad \text{for} \quad \text{Hf} \rightarrow \text{Hf}
\]

\[
[N^s]_Z = 5890, \quad [N^s]_p = 5000 \quad \text{for} \quad \text{Hf} \rightarrow \text{C}
\]

where \( N^s = N^+(10\text{eV}) \). Seeing Fig.2(c) on page 623 in ref.[13], we realize that \([N^s]_Z\) reproduce the corresponding data precisely. However, \([N^s]_p\) is too low. Because \( N^s \) data were generated by using the Kr-C potential interaction in ref.[13][57], they naturally provide evidence supporting the point that replacing the actual interatomic potential by a power potential (6.2.1) is a less severe simplification than replacing the actual cross section by a simple power cross section (6.2.24).

6.3.3 MEAN SLOWING DOWN DISTANCE

Substituting (6.2.25) and (6.2.26) into (3.3.22), the atom flux turns out to be,

\[
\psi(E_0) = [L_m(E_0)\Gamma_m] \cdot E_0^{-2}
\]

(6.3.7)
Here \[ L_m(E_0)_Z = \frac{(E_0)^{2m}}{N[C_m]_Z} \int_0^1 dXZ_m(X)X^{-m}, \] if \( 1 \geq m > 0 \).

\[ [L_m(E_0)_p] = \frac{(1-m)(E_0)^{2m}}{N[C_m]_p} \]

At low energies \((m \sim 0)\), the collisions become less penetrating than predicted by Sigmund’s theory [46]. Particularly for \( m = 0 \), both cross-sections (6.2.9) and (6.2.24) reduce to

\[ [d\sigma]_Z = \sigma E^{-1}dT \quad \text{and} \quad [d\sigma]_p = C_0 T^{-1}dT \]

(6.3.8)

respectively. Thus, for \( m = 0 \), we have

\[ [L_0]_Z \equiv [L_0(E_0)]_Z = 2(N\sigma)^{-1} \quad \text{and} \quad [L_0]_p \equiv [L_0(E_0)]_p = (NC_0)^{-1} \quad [57]. \] (6.3.9)

Adjusting \( \chi \) of (6.2.5) makes \([\psi(E_0)]_Z = [\psi(E_0)]_p\), and then

\[ [L_m(E_0)\Gamma_m]_Z = [L_m(E_0)\Gamma_m]_p \]

(6.3.10)

As a result, we can see

\[ [L_m(E_0)_Z \approx [L_m(E_0)]_p \quad \text{for} \quad 1 \geq m \geq 0.5 \]

\[ [L_m(E_0)_Z \approx [L_m(E_0)]_p \quad \text{for} \quad 0.5 > m \geq 0 \]

Inserting (6.3.3,4) and (6.3.9) into (6.3.10), we have

\[ \sigma = \pi^2 C_0 / 3 \] (6.3.11)

Both cross-sections yield identical atom fluxes, but two different mean slowing down distances

\[ [L_0]_Z \approx 0.6079[L_0]_p \] (6.3.12)

This is exactly the point resulted in the ‘depth of origin’ puzzle.

**6.3.4 MUBER OF RENKEL-PAIRS**

Substituting (6.2.25,26) into (3.3.4), we derived \( \xi_f (m) \equiv \xi_f \),

\[ [\xi_f (m)]_Z = 2[\Gamma_m]_Z \left[ \int_0^1 dXZ_m(X) \right]^{-1} \int_0^{0.5} dX[Z_m(X) + Z_m(1-X)]X. \] (6.3.13)

\[ [\xi_f (m)]_p = 2[\Gamma_m]_p (2^m - 1)/m, \] (6.3.14)
\[ [\xi_F(0)]_Z = 1 \quad \text{and} \quad [\xi_F(0)]_p = 12\pi^2 \ln 2 \approx 0.84. \] (6.3.15)

Both \([\xi_F(m)]_p\) and \([\xi_F(m)]_Z\) have been calculated and plotted in Fig. 6.3.2 as continuous functions of \(m\) for \(1 \geq m \geq 0\). The calculated results showed that \([\xi_F(m)]_p < [\xi_F(m)]_Z\) for \(0.25 \geq m > 0\). Therefore, \([\xi_F(m)]_Z\) has confirmed Lindhard’s statement [46] and removed the limitation of hard-sphere scattering model pointed out by Sigmund [22].

**Fig. 6.3.2** Calculated curve of \(\xi_F(m)\) as a continuous function of \(m\), defined by (6.3.13–15). Thick line: calculated by Exact theory (6.3.13,15). Dash line: calculated by Sigmund theory (6.3.14).

### 6.3.5 RECOIL DENSITY

The Recoil density \(F(E_0)\) was introduced by Urbassek et al. in ref. [13],

\[ F(E_0) = K(E_0) \cdot E E_0^{-2} \] (6.3.16)

\[ K(E_0) = \frac{S(E_0)}{E_0 \tilde{\sigma}(E_0)} = \frac{S(E_0)}{E_0 [\sigma'(E_0) + \sigma'(E_0) ]} \] (6.3.17)

(6.3.16,17) have been given by (4.2.7), a long as \(\alpha_z = 0\). For Kr-C interaction potential [13], (6.3.17) reduced to

\[ K(E_0) = \Gamma(\varepsilon, l), \quad \varepsilon = E_0 / \varepsilon_0 \] (6.3.18)
which is a special case of (6.1.33) for $\gamma = 1$.

If use a power potential to approximate the Kr-C interaction potential, i.e. substituting (6.2.9) and (6.2.24) into (6.1.15-17) and (6.1.33) respectively, we derived

$$
\Gamma_m(\gamma) \equiv \Gamma(\varepsilon, \gamma),
$$

and

$$
\left[ \Gamma_m(\gamma) \right]_Z = 1 - \ln \gamma - \left\{ 1 \left| \ln \gamma \right\}_m - \sum_{n=1}^{\infty} \frac{\gamma^n}{n(n+1)} \left\{ 1 \left| x^n \right\}_m , \quad \text{if } 1 \geq m > 0. \tag{6.3.19}
$$

$$
\left[ \Gamma_m(\gamma) \right]_p = 1 - \ln \gamma + \frac{1}{1-m} - (1-m) \sum_{n=1}^{\infty} \frac{\gamma^n}{n(n+1)(n+1-m)} \tag{6.3.20}
$$

Comparing (6.3.19,20) with (6.3.1,2), we can see $\Gamma_m = \Gamma_m(l) = \Gamma(\varepsilon, l)$.

Let’s use Urbassek’s formula [13]

$$
1 - 2m = \frac{d \ln S_{\gamma}(E)}{d \ln E} \tag{6.3.21}
$$

to estimate $m$. According to Urbassek’s data [13], we have calculated and got $m = 0.12$ within the energy window $5 \cdot 10^{-4} > \varepsilon > 10^{-5}$. Then, substituting $m = 0.12$ into (6.3.19) and (6.3.20) respectively, we obtained

$$
\left[ \Gamma_m \right]_Z = 0.730, \quad \left[ \Gamma_m(0.5) \right]_Z = 0.430 \quad \text{and} \tag{6.3.22}
$$

$$
\left[ \Gamma_m \right]_p = 0.554, \quad \left[ \Gamma_m(0.5) \right]_p = 0.371 \tag{6.3.23}
$$

and plotted these data in Fig.6.1.3 to compare with the numerical curves of (6.1.33). We have found that (6.3.22) agree with the left side of the curves very well. But, (6.3.23) are too low.

### 6.4 SPUTTERING PARTICLE FLUX

If the power potential (6.2.1) satisfies the following conditions,

$$
\begin{align*}
\text{(m)}_1 = \text{(m)}_2 = \cdots = \text{(m)}_n \equiv m \\
\chi_{ij} = \chi_{ji}
\end{align*}
\tag{6.4.1}
$$

we have the following theories for the sputtering atomic flux of multi-component target. [15][18]

#### 6.4.1 ANDERSEN-SGMUND RELATION

Substituting (6.4.1) and (6.2.25,26) into (3.1.11) respectively, we obtained
\[
\frac{\beta_{ij}^L}{\beta_{ij}^L} = \frac{\alpha_i}{\alpha_j} \left( \frac{M_j}{M_i} \right)^{2m}, \quad \text{for } L = 0, 1
\]  

(6.4.2)

where \( \beta_{ij}^L = \beta_{ij}^L (1 - 0.5L) \). Furthermore, using (6.4.2), it’s easy to derive

\[
\frac{\beta_{ij}^L}{\beta_{ij}^L} \frac{\beta_{k\ell}^L}{\beta_{k\ell}^L} \cdots \frac{\beta_{n_k\ell_k}^L}{\beta_{n_k\ell_k}^L} = \left( \frac{\beta_{ij}^L}{\beta_{ij}^L} \right) \cdots \left( \frac{\beta_{n_k\ell_k}^L}{\beta_{n_k\ell_k}^L} \right)
\]

\[
= \frac{\alpha_i}{\alpha_j} \left( \frac{M_j}{M_i} \right)^{2m} \cdot \frac{\alpha_{r_1}}{\alpha_{k_1}} \left( \frac{M_{r_1}}{M_{k_1}} \right)^{2m} \cdots \frac{\alpha_{k_k}}{\alpha_{r_1}} \left( \frac{M_{k_k}}{M_{r_1}} \right)^{2m} \cdot \frac{\alpha_i}{\alpha_j} \left( \frac{M_{r_1}}{M_{r_1}} \right)^{2m}
\]

(6.4.3)

One the other hand, a direct expansion gives

\[
\left( \frac{D_{ij}^L}{D_{ij}^L} \right) = \sum A^L (r_1, r_2, \cdots r_{k-1}, r_k) \left( \frac{\beta_{ij}^L}{\beta_{ij}^L} \right) \cdots \left( \frac{\beta_{n_k\ell_k}^L}{\beta_{n_k\ell_k}^L} \right)
\]

(6.4.4)

Inserting (6.4.3) into (6.4.4), we have

\[
\frac{D_{ij}^L}{D_{ij}^L} = \frac{\alpha_j}{\alpha_i} \left( \frac{M_i}{M_j} \right)^{2m}
\]

(6.4.5)

Substituting (6.4.5) into (3.3.1) and (3.3.8), the ratio between the fluxes of moving atoms of the two species can be derived [16][18]

\[
\frac{\psi_i^L(E_0)}{\psi_i^L(E_0)} = \frac{G_{kji}(E, E_0)}{G_{kij}(E, E_0)} = \frac{D_{ji}^L}{D_{ij}^L} \cdot \left( \frac{M_j}{M_i} \right)^{0.5L} = \frac{\alpha_j}{\alpha_i} \left( \frac{M_j}{M_i} \right)^{2m-0.5L}
\]

(6.4.6)

Obviously, this relation is tenable for both scattering cross sections and only depends on the pair of elements related to in a multi-component medium, independent of others. In ref.[16], P. Sigmund et. had derived (6.4.6) only for a binary medium Later, H. M. Urbassek et. had demonstrated (6.4.6) can be applied to a ternary system with an arbitray third component[99][100]. Besides, P. Sigmund had shown that (6.4.6) is only approximately correct for a special case of a poly-isotopic target.[17][100].

### 6.4.2 Anisotropic Differential Particle Flux

By using (6.4.5), it’s easy to proof \( A_j / A_i = M_j / M_i \). Formula (3.3.12) turned out to be
Here \( K_j = E_0^{2m-1} \cdot \Pi_j /(N\nu_0) \). Formula (6.4.7) is tenable for any multi-component medium. Even if Formula (3.3.13) was derived by Sigmund for a binary medium with nearly equal masses \( M_1 \approx M_2 \) \([17][51] \), P. Sigmund himself, still worried about his analytical results of (3.3.13), he has written down in ref. [101]: "In transport theory, small effects may be analyzed by perturbation theory but systematic errors caused by the neglect of potentially significant features may limit the predictive power."

### 6.5 HARD SPHERE SCATTERING CROSS-SECTION

#### 6.5.1 RANGE OF VALIDITY OF ASYMPTOTIC SOLUTIONS

The validity of the asymptotic solutions for \( L = 0 \) in Chapter 3 is directly related to the correction terms in the asymptotic expansions of solutions. These correction terms are determined by the residues at subsequent poles of \( H_j(s) \), i.e. zeros of \( D(s) \), in most cases. For a binary medium \( (\alpha_1 + \alpha_2 = 1) \),

\[
D(s) = \beta_{11}(s)[\varepsilon_{11}(s) - 1] \beta_{21}(s)\varepsilon_{21}(s) + \beta_{22}(s)[\varepsilon_{22}(s) - 1] \beta_{12}(s)\varepsilon_{12}(s) + \beta_{11}(s)\varepsilon_{11}(s) - 1] \beta_{22}(s)\varepsilon_{22}(s) - 1] + \beta_{12}(s)\beta_{21}(s)[\varepsilon_{12}(s)\varepsilon_{21}(s) - 1] 
\]

(6.5.1)

For simplicity, only the case of \( m = 0 \) is treated. Thus, substituting (6.3.8) into (3.1.11) and (3.1.12) respectively, we obtained,

\[
[\beta_{ij}(s)]_2 = \alpha_j \sigma \gamma^s (s + 1)^{-1} 
\]

(6.5.2)

\[
[\beta_{ij}(s)]_1 = \alpha_j C_0 \gamma^s s^{-1} 
\]

(6.5.4)

\[
[\varepsilon_{ij}(s)]_2 = \gamma^{-s-1}[(s + 1)\gamma - 1 + (1 - \gamma)^{s+1}] 
\]

(6.5.3)
\[
[\varepsilon_y(s)]_p = s^2 \gamma^{-s} \left[ (1 \cdot 1!)^{-1} + (1-s)(2 \cdot 2!)^{-1} \gamma + (1-s)(2-s)(3 \cdot 3!)^{-1} \gamma^2 + \cdots + (1-s)(2-s) \cdots (n-1-s)(n \cdot n!)^{-1} \gamma^{n-1} + \cdots \right]
\]

(6.5.5)

\[
[\varepsilon_y]_Z = \gamma^{-1} + (\gamma^{-1} - 1)^2 \cdot \ln(1-\gamma) - \ln \gamma
\]

\[
[\varepsilon_y]_p = 2 - \ln \gamma - \sum_{n=1}^{\infty} \gamma^n \left[ \ln(n+1)^2 \right].
\]

Here, \( \gamma \equiv \gamma_y \). In particular, if \( \gamma = 1 \), we have

\[
[\varepsilon_y(s)]_Z = s,
\]

\[
[\varepsilon_y(s)]_p = 1 + \psi(s), \quad \psi(x) \equiv \frac{d \ln \Gamma(x)}{dx}.
\]

\[
[\Gamma_0]_Z = [\varepsilon_y]_Z = 1 \quad \text{and} \quad [\Gamma_0]_p = [\varepsilon_y]_p = 6 \pi^{-2} \approx 0.6079.
\]

Substituting (6.5.2-5) into (6.5.1), except \( s = 1 \), the extra zero \( s_0 \) of \( D(s) \) is calculated and plotted in Fig. 6.5.1 as a function of the concentration \( \alpha_1 \) (or \( \alpha_2 \)) for three typical binary systems HfN, ZrN and TiN in both Sigmund and Hard sphere theories. One can see \( [s_0]_p -0.5 \) and \( [s_0]_Z < 0 \) for TiN and ZrN. This fact shows that Sigmund's asymptotic solutions may not be suitable for HfN and ZrN in Sigmund theory, but are acceptable in Hard sphere theory. Therefore, the Hard sphere theory has a much larger range of validity.

For further simplicity, let's assume \( \alpha_1 = \alpha_2 = 0.5 \), thus Eq.(6.5.1) can be factorized, i.e.

\[
D(s) = \beta_{11}(s) \cdot D_+(s)D_-(s)
\]

Here \( D_\pm(s) \equiv \varepsilon_{11}(s) - 1 + \gamma^\pm [\varepsilon_{12}(s) \pm 1] \) and \( \gamma \equiv \gamma_{12} \).

\[
[\tilde{G}_\pm(s)] = \frac{1}{N\nu_0 E_0} \cdot \frac{1}{2\beta_{11}(s)} \left[ \frac{1}{D_-(s)} \pm \frac{1}{D_+(s)} \right]
\]

(6.5.6)

Here \( G_+ \equiv G_{11} = G_{22} \), \( G_- \equiv G_{12} = G_{21} \), \( D_\pm(s) \equiv \varepsilon_{11}(s) - 1 + \gamma^\pm [\varepsilon_{12}(s) \pm 1] \).

A simple calculation shows that \( D_-(s) \) has one positive zero \( s = 1 \). Therefore, the inverse Laplace transforms of the first term of (6.5.6) yields asymptotic solution, i.e.
Fig. 6.5.1 Additional positive zero of $D(s)$, as a function of the chemical concentration for three typical systems TiN, ZrN and HfN. In Hard sphere theory, the highest additional zero is 0.174 for HfN. No such zero exists for ZrN and TiN. In Sigmund theory, the highest additional zero is about 0.5 for ZrN and HfN [16].

Fig. 6.5.2 Additional positive zero of $D(s)$, as a function of reduced atomic mass for a binary system with chemical concentration 0.5. Thick line: calculated by Hard sphere theory. Dash line: calculated by Sigmund theory.

\[
G_\pm (E_0)_{AS} = \frac{E}{N^0_0 E_0^2} \cdot \frac{1}{2\beta_{11} (1)(\epsilon_{11} + \gamma \epsilon_{12})}
\]  

(6.5.7)

which is just the special case of (3.3.1). In addition, $D_+ (s)$ has a $\gamma$ dependent zero $s_0$, which is calculated and plotted in Fig. 6.5.2 as a function of $\gamma$ in both Sigmund and Hard sphere theories.
In Sigmund theory, see Fig. 6.5.2, if and only if \( \gamma \sim 1 \) or \( M_1 \sim M_2, s_0 \sim 0 \), thus the asymptotic solutions (6.5.7) are acceptable [51]. Taking account of the contribution of \( s_0 \), the inverse Laplace transforms of (6.5.6) are given by

\[
\left[ G_{\pm}(E_0) \right]_p = \frac{\Gamma_0 E}{N_0 C_0 E_0^2} \cdot \frac{G}{(G)} \cdot \left(1 \pm Ax^{1-s_0}\right)
\]  

(6.5.8)

Here \( G/(G) = \left(\pi^2/6\right) \left(\varepsilon_{11} + \gamma \varepsilon_{12}\right)^{-1} \),

\[ A = s_0 \left( s_{11} + \gamma s_{12} \right) \left[ D'_+(s_0) \right]^{-1} \]  

and

\[ D'_+(\alpha) = \left[ dD_{\pm}(s)/ds \right]_{s=\alpha}. \]

For HfN and HfC, we have calculated \( s_0, G/(G) \) and \( A \) by using Sigmund theory and given the relative discrepancy between (6.5.7) and (6.5.8)

\[ [\delta]_p = A \cdot x^{1-s_0} \]  

(6.5.9)

which is calculated and plotted in Fig. 6.5.3 as a function of \( x \equiv E_0/E \) for the HfN and HfC systems respectively.

In Hard sphere theory, \( \gamma \left[ D_+(s) \right]_Z = 2\gamma s - 1 + (1 - \gamma)^{s+1} \pm \gamma^{s+1} \). Excepts the \( \gamma \) dependent zero \( s_0 \), \( \left[ D_+(s) \right]_Z \) has an extra constant zero \( s = 0 \).

In particular, if \( \gamma_1 \) satisfies

\[ 2\gamma_1 + (1 - \gamma_1) \ln(1 - \gamma_1) + \gamma_1 \ln \gamma_1 = 0, \]  

i.e.

\[ \gamma_1 = 0.3092493810\cdots, \]  

i.e. \( (M_1/M_2)^{x_1} = 10.84231365\cdots \),

then, \( s = 0 \) is the second order zero, i.e. \( \left[ D_+(s) \right]_Z \sim s^2 \), for \( s \sim 0 \). Thus

\[
G_{\pm}(E_0) = \frac{2E}{N_0 \sigma E_0^2} \cdot \frac{G}{(G)} \cdot \left(1 \mp Ax \ln x\right)
\]  

(6.5.10)

Here \( G/(G) = 0.5580026975\cdots \) and \( A = 1.064765133\cdots \).

In general, if \( \gamma \neq \gamma_1 \), a simple calculation shows that \( D_+(s) \) has a non-vanishing
zero, \( s_0 < 0 \) for \( 1 \geq \gamma > \gamma_0 \) and \( s_0 > 0 \) for \( \gamma_0 \geq \gamma > 0 \). Taking account of the contributions of these three poles \( s = 0, 1 \) and \( s_0 \), the inverse Laplace transforms of (6.5.6) are given by

\[
G_\pm(E_0) = \frac{2E}{\nu_0 \sigma E_0^2} G \left(1 \pm A x^{1-s_0} \mp B x\right),
\]

(6.5.11)

\[
\frac{G}{(G)} = \frac{\gamma}{D_+'(1)}, \quad A = \frac{(s_0 + 1)}{2} \frac{D_+'(1)}{D_+(s_0)} \quad \text{and} \quad B = -\frac{1}{2} \frac{D_+'(1)}{D_+(0)}.
\]

For HfN and HfC, we have calculated \( s_0 \), \( G/(G) \) and \( A \) by using using Hard sphere theory, written (6.5.10-11) together and given the relative discrepancy

\[
[\delta]_x = \begin{cases} 
Ax \ln x, & \text{for } \gamma = 0.309 \ldots \\
Ax^{1-s_0} - Bx, & \text{others}
\end{cases}
\]

(6.5.12)

which is calculated and plotted in Fig.(6.5.3) as a function of \( x \) for the HfN and HfC systems. Fig.(6.5.3) shows that Hard sphere theory generates a more accurate asymptotic solution (6.5.7) than the one given by Sigmund theory for the same system. For instance, if \( E_0/E = 0.01 \), then \( [\delta]_x \sim 10\% \) and \( [\delta]_p \sim 30\% \). Therefore, in Sigmund theory, the correction term in the asymptotic expansions of solution for \( L = 0 \) not only could be as large as the leading term for \( L = 1 \), but could also disturb the isotropic leading term for \( L = 0 \), thus complicate the sputtering yield energy distribution in the case where \( \gamma \) is very small [56].

### 6.5.2 EXACT SOLUTIONS

Another remarkable feature of hard sphere scattering cross section is that all of the transport equations can be solved analytically, even exactly. For a monatomic target bombarded by a self-atom, Eq. (3.1.3, 4) is reduced to

\[
\tilde{G}_\pm(s) = L_0 v_0^{-1} [(2E_0)^{-1} + \tilde{F}_\pm(s)],
\]

\[
\tilde{F}_\pm(s) = \tilde{F}_\pm(s) = E_0^{-1} \left[1 - 2 \int_0^\infty dXX^s P_L \left(\sqrt{X}\right)\right]^{-1} \int_0^\infty dXX^s P_L \left(\sqrt{X}\right)
\]

(6.5.13)

Taking account of the contributions of all poles, including complex variable poles of Eq. (6.5.13), the inverse Laplace transforms of Eq. (6.5.13) are given exactly as follows,
\begin{align*}
f(x) &= 1 \\
f^1(x) &= x^{0.5} \\
f^2(x) &= x^{1.5} \cos\left(0.5\sqrt{3} \ln x\right) \\
f^3(x) &= \sqrt{15/11} \cdot x^{2} \sin\left(0.5\sqrt{11} \cdot \ln x + \Phi_0\right), \quad \Phi_0 = \tan^{-1}\left(0.5\sqrt{11}\right)
\end{align*} \tag{6.5.14}

Here \( F^1(E, E_0) = EE_0^{-2} f^1(x) \). \( f^1(x) \) has been plotted as a function of \( x \equiv EE_0^{-1} \) respectively for \( L = 0, 1, 2, 3 \cdots 10 \) in Fig.(6.5.4). In addition, substituting (6.5.14) into Eq.(2.3.1), the numerical calculation has shown that Eq.(2.3.1) converges very well as \( L \) increases infinitely.

Fig. 6.5.3 The relative errors calculated by using Sigmund theory and Hard sphere theory respectively. Obviously, Hard sphere theory can generate a more accurate asymptotic solution.

Fig. 6.5.4 Exact solutions for recoil densities for a monatomic target bombarded by a self-atom in Hard sphere theory, for \( L = 0, 1, 2, \cdots, 10 \).
6.6 DEPTH DISTRIBUTIONS OF DEPOSITED ENERGY AND MOMENTUM AND ION RANGE

6.6.1 UNIFIED DIMENSIONLESS DEPTH DISTRIBUTION FUNCTIONS

Considering the obvious scaling property, the depth distributions of deposited energy, momentum and ion range can be written,

\[ F_{(i)}(z, \bar{u}, U) = E \cdot L^{-1}_i(E) \cdot f_{(i)}[zL^{-1}_i(E), UE^{-1}, \eta] \quad (6.6.1) \]

\[ P_{(i)}(z, \bar{u}, U) = \sqrt{2M_i E \cdot L^{-1}_i(E) \cdot p_{(i)}[zL^{-1}_i(E), UE^{-1}, \eta]} \quad (6.6.2) \]

\[ R_{(i)}(z, \bar{u}) = L^{-1}_i(E) \cdot r_{(i)}[zL^{-1}_i(E), \eta] \quad (6.6.3) \]

Here \( L_i(E) = [E^{-2m}N\beta_i]^{-1} \) is the mean slowing down distance defined by (3.3.20) and \( \beta_i = \sum_k \beta_{ik}(1) \). Therefore, \( f_{(i)}(x, u, \eta) \), \( p_{(i)}(x, u, \eta) \) and \( r_{(i)}(x, \eta) \) are unified dimensionless depth distribution functions. \( x \equiv zL^{-1}_i(E) = zE^{-2m}N\beta_i \) is a dimensionless depth and \( u \equiv U/E \) is a dimensionless threshold energy. Substituting

\[ E \cdot L^{-1}_i(E) = E^{1-2m}N\beta_i = NS_n(E) = N \sum_k \alpha_k[S_n(E)]_{ik} \quad (6.6.4) \]

into (6.6.1-3), we obtained

\[ F_{(i)}(z, \bar{u}, U) = NS_n(E) \cdot f_{(i)}(x, u, \eta) \quad (6.6.5) \]

\[ P_{(i)}(z, \bar{u}, U) = \sqrt{2M_i E \cdot NS_n(E) \cdot p_{(i)}(x, u, \eta)} \quad (6.6.6) \]

\[ R_{(i)}(z, \bar{u}) = E^{-1} \cdot NS_n(E) \cdot r_{(i)}(x, \eta) \quad (6.6.7) \]

Here \([S_n(E)]_{ik} = E^{1-2m}\beta_{ik}(1) = \int_0^{\tau_n} T d\sigma_{ik}(E, T)\).

By using SC theory [19] described in Chapter 4, \( d\sigma_{ik}(E, T) \) can be considered as an arbitrary scattering cross section. Therefore, \([S_n(E)]_{ik}\) appeared in (6.6.4) can be replaced by (6.1.15) and one of (6.1.23-26). The next key problem is to determine the unified dimensionless depth distribution functions.

Substituting (6.2.25) and (6.2.26) into all recursion relations (5.2.20-22), (5.2.40-42) for
energy deposition, (5.3.7-10), (5.3.18-21) for momentum deposition and (5.2.58) for ion range respectively, one can derived all moments:
\[
f^{(1)}_{iL} = \left[ \beta_i \right]^n \cdot \begin{cases} A^n_{iL}, & \text{for } m \neq 0.25 \\ B^n_{iL}, & \text{for } m = 0.25. \end{cases} \quad (6.6.8)
\]
\[
p^n_{(1)L} = (2M_i)^{0.5} \left[ \beta_i \right]^n \cdot \begin{cases} a^n_{iL}, & \text{for } m \neq 0.25 \\ b^n_{iL}, & \text{for } m = 0.25. \end{cases} \quad (6.6.9)
\]
\[
t^n_{(1)L} = \left[ \beta_i \right]^n \cdot q^n_{iL} \quad (6.6.10)
\]

By using the approximation \(Z_i/Z_j = M_i/M_j \equiv \rho_{ij}\) giving in Ref. [37], we obtained
\[
\chi_{ij}/\chi_{\mu\nu} = (Z_i/Z_j)/(Z_\mu Z_\nu) = \rho_{ij} \rho_{\mu\nu}, \text{ and}
\]
\[
C_{ij}/C_{\mu\nu} = \rho_{ij}^{3m} \cdot \rho_{\mu\nu}^{(5m-2)/3} \cdot \left[ (1 + \rho_{ij}^{2/3})/(1 + \rho_{\mu\nu}^{2/3}) \right]^{m-1} \quad \text{for the Thomas-Fermi potential,}
\]
\[
C_{ij}/C_{\mu\nu} = \rho_{ij}^{5m/2} \cdot \rho_{\mu\nu}^{m/2} \quad \text{for the Born-Mayer potential. Thus,}
\]
we have
\[
\alpha^n(\eta) = \sum_L (2L+1)\alpha^n_L \eta^L \quad (6.6.11)
\]

Here \(\alpha\) stands for one of \(f, p\) and \(r\). The subscript \(i\) has been dropped for simplicity. The next step is the reconstruction of unified dimensionless depth distribution function \(\alpha(x, \eta)\) by Padé approximant Method\([\mu/\nu](x)\) from the moments \(\alpha^n(\eta)\) in (6.6.11).

A small program has been written in BASIC for the calculation about 15 years ago. The program has been run for some momentum depositions depth distribution functions simulated by P. Sigmund et. al.[11]. The single power cross section (6.2.24) was used for the calculation. The calculated curves agree the computer simulations in [11] very well. The program was successful to be executed for various Ion-Target systems including Y-Ba-Cu-O compounds [103] under different conditions. This program now can be executed up to \(n = 980\) moments with 500-decimal-digits precision on any one of Laptops, see chapter 5.
6.6.2 NUMERICAL RESULTS

About 15 years before, the calculations were down on the SHARP PC-1500 pocket calculator for a few typical Ion-Target systems at a perpendicular incidence for \( m = 0.2 \) by using both single and double power scattering cross sections. In the case, (6.2.23) reduced to

\[
Z_{0.2}(X) = 1 + 1.796X^{0.5820}.
\]  

(6.6.12)

For energy depth distribution [18][37], our calculated curves for three Ion-Target systems have been plotted in Fig.6.6.1. According to (6.6.5), the energy deposited in the target surface is

\[
F_D(0, E, \eta) = N\alpha S_n(E).
\]  

(6.6.13)

Here \( \alpha \equiv f(0, u, \eta) \) is a dimensionless function of all atomic mass ratios and the incident angle [1]. The remarkable finding points are:

i) Even though (6.6.12) shows a great discrepancy of \( Z_m(X) \) from unity for \( m = 0.2 \), both double and single power cross section (6.2.23,24) generate almost the same value of \( \alpha \), the relative error being less than 10% [18], i.e.

\[
[\alpha]_Z \approx [\alpha]_P
\]  

(6.6.14)

ii) All calculated curves of \( f(x, u, \eta) \), corresponding to different \( u \) (here \( 0.1 \geq u \geq 0 \)), almost across at the same point near the target surface. This means that

\[
\alpha \approx f(0, \eta)
\]

A similar conclusion has been obtained for \( m = 1/3 \) in literature [12], i.e. \( \alpha \) can be considered as a dimensionless function of atomic mass ratios and incident angle. Therefore, the energy deposited in the target surface (6.6.13), given by P. Sigmund [1], can be used for the sputtering of single element target bombarded by low energy heavy ion at least.
Fig. 6.6.1 Depth distribution of deposited energy for a target bombarded by a projectile at perpendicular incidence. The moments are evaluated by using (5.2.20-22) with of $M = 0.2$. The reconstruction of the depth distribution functions is done by Padé method. (a) Calculated by using Single power scattering cross section (6.2.24). (b) Calculated by using Double power scattering cross section (6.2.9) and (6.2.23).

Fig. 6.6.2 Depth distribution of deposited momentum for Molybdenum target bombarded by Argon ion at perpendicular incidence. The moments are evaluated by using (5.3.7-10) with of $M = 0.2$. The reconstruction of the depth distribution functions is done by Padé method.

Fig. 6.6.3 Unified ion range distribution function in a target bombarded by an incident ion perpendicularly. The moments are evaluated by using (5.2.58) with of $M = 0.2$. Thick line: Calculated by using Single power cross section (6.2.24). Dash line: Calculated by using Double power cross section (6.2.9,23).
A similar conclusion has been obtained for $m = 1/3$ in literature [12], i.e. $\alpha$ can be considered as a dimensionless function of atomic mass ratios and incident angle. Therefore, the energy deposited in the target surface (6.6.13), given by P. Sigmund [1], can be used for the sputtering of single element target bombarded by low energy heavy ion at least.

For momentum depth distribution [39][40], our calculated curves for Molybdenum target bombarded by Argon ion have been plotted in Fig. 6.6.2. Figures 6.6.1 and 6.6.2 are similar. Both analytical calculation and computer simulation showed that the parallel component of momentum deposited in the target surface is very sensitive to factor $\eta = U/E$ and could be positive as long as $U/E > 0.05$ for $m = 1/3$ [12] and $U/E > 0.1$ for $m = 0.2$ [11]. In our calculation in Fig. 6.6.2, one can come to a similar conclusion. For the low energy and heavy ion bombardment, according (6.6.6), we obtained the momentum deposited in target surface:

$$P_{\alpha}(0, \eta, U) = \sqrt{2M_i/E \cdot NpS_n(E)}$$

(6.6.15)

Here $p = p(0, \eta, \eta)$ is a dimensionless function, but related to $\eta$, all atomic mass ratios and the incident angle [1]. According to our basic assumption in Chapter 1, $p$ could be a positive number for the low energy and heavy ion bombardment.

For Ion range [38], our calculated curves for three Ion-Target systems have been plotted in Fig. 6.6.3. We has also found that for the low energy ion (few hundreds eV) incidence, the depth of ion implantation must be smaller, the ion range distribution must be narrower than the results predicted theoretically by using the single power cross section [37]. This conclusion agrees with our work on the depth of sputtered atom [18] and Frenkel-Pair Production [21]. Nevertheless, as long as the characteristic length (3.3.20) is used as the depth unit, the unified ion range $r(x)$ calculated by using both double and single cross section are about the same.
CHAPTER 7

ISOTROPIC SPUTTERING

7.1 ISOTROPIC SPUTTERING THEORY

7.1.1 ISOTROPIC SPUTTERED PARTICLE SPECTRUM

P. Sigmund divided a sputtering event into two steps: Primary stage—the creation of a primary recoil atom by an incident ion, which mainly determines the deposited energy and momentum depth distribution functions, and low energy stage—the subsequent development of a cascade of recoil, which are ejected through the target surface as sputtered atom flux [7]. According to this statement, completed solutions of all transport equations appeared in chapter-2 can be constructed as long as replacing the energy E and momentum P of incident particle by corresponding the space distribution functions of deposited energy and momentum respectively. In particular, replacing the energy E of incident particle in (3.3.1) by energy deposited in the target surface [1][7], Sigmund derived the outward particle current

\[ J_j \left( E_o, \Omega_o \right) = \frac{G_j}{\left( G \right)_j} \cdot \frac{\Gamma_m}{\left( \frac{dE_o}{dx} \right)_j} \cos \theta_i \cdot \frac{F_{i j}(0, E, \theta)}{E_o} \frac{d^2 \Omega_j dE_j}{4\pi} \]  

(7.1.1)

Where \( F_{i j}(z, \vec{u}) = F_{i j}(z, E, \eta) \) is the depth distribution function of deposited energy (6.6.5). In principal, (7.1.1) can be considered as a basis for isotropic theory of sputtering of any target [1][16][53]. Taking account of the refraction by the planar surface binding energy \( U_{oj} \),

\[ E_1 \cos^2 \theta_1 = E_o \cos^2 \theta_0 - U_{oj} \]
\[ E_1 \sin^2 \theta_1 = E_o \sin^2 \theta_0 \]

(7.1.2)

(7.1.1) turns out to be the isotropic flux of sputtered atom-j moving with energy \( E_1 \) and along the emission direction \((\theta_1, \phi_1)\) through the target plane surface.

i) Energy distribution of sputtered atoms-Thompson distribution

\[ \frac{d^3 Y_j}{dE_j d^2 \Omega_j} = Y_{mj} \frac{2 |\cos \theta_i|}{\pi U_{oj}} \frac{\epsilon}{(\epsilon + 1)^{3/2}} \], \quad \epsilon \equiv \frac{E_1}{U_{oj}} \]  

(7.1.3)
ii) Cosine like angular distribution of sputtered atoms

\[
\frac{d^2Y_j}{d^2\Omega_j} = Y_{mj} \left| \cos \theta_j \right| \frac{1}{\pi};
\]  

(7.1.4)

iii) Total partial sputtering yield: \( Y_j = Y_{mj} = B \Lambda_{mj}(0) \)  

(7.1.5)

iv) Andersen Sigmund relation is the partial sputtering yield ratio between two components i and j [16][19], which is given by

\[
\frac{Y_i}{Y_j} = \frac{\alpha_i}{\alpha_j} \left( \frac{M_j}{M_i} \right)^{2m} \left( \frac{U_{ij}}{U_{oi}} \right)^{1-2m}
\]  

(7.1.6)

Here \( B^{-1} = (1 - m)(1 - 2m) \), \( \Lambda_{mj} = \Lambda_{mj}(0) \) and

\[
\Lambda_{mj}(z) \equiv \frac{G_j}{(G_j)^{1/8}} \frac{\Gamma_m}{NB_{ji}} \frac{\alpha_j}{\alpha_i} \frac{F_{(1)}(z,E,\eta)}{U_{ij}^{1-2m}}
\]

Andersen Sigmund relation (7.1.6) predicts the isotropic preferential sputtering.

### 7.1.2 ENERGY AND ANGULAR DISTRIBUTIONS OF SPUTTERED ATOMS

The emission energy and angular distributions of atoms sputtered from elemental targets has been measured many times. Typical examples of energy distributions were given for elemental targets Al, Ca and Ag bombarded by 8,4 and 5 keV respectively [55]. The experimental data can be well approximated by Thompson distribution (7.1.3) with \( m = 0 \) and the surface binding energies \( U_0 = 1.4 \text{eV}, \ 1.5 \text{eV} \) and \( 2.2 \text{eV} \) should be chosen respectively [55], see Fig.7.1.1.

![Fig.7.1.1 Energy distribution of neutral atoms sputtered from elemental surfaces under bombardment with indicated projectiles. Solid curves were calculated by Thompson distribution (7.1.3) with \( m = 0 \) and the surface binding energies 1.4 eV, 1.5 eV and 2.2 eV should be chosen respectively](image-url)
Fig. 7.1.2 Emission energy distributions of Hf and C atoms sputtered from HfC target under bombardment with incident energy 6-keV. Solid curves were calculated by Thompson distribution (7.1.3) with m = 0 and the surface binding energies: 4.8 eV for carbide atom and 6.7 eV for hafnium atom should be chosen.

For multi-component targets, the phenomenon can be seen. Typical example published by M. Szymoński is sputtering binary compound target HfC bombarded by Xe$^+$ ion with incident energy $E = 6\text{keV}$ [56]. Again, Thompson distribution (7.1.3) with $m = 0$ can reproduce the experimental results well, as long as the surface binding energies were chosen: $U_0 = 4.8\text{eV}$ for carbide atom and $U_0 = 6.7\text{eV}$ for hafnium atom see Fig. 7.1.2. Seeing the atomic mass of hafnium is almost 15 times heaver than carbon, the parameter $m$ should be different between them. H. M Hrbasseck et. al. worked out the same system and assumed [13] $m_{\text{HfF}} = 0.12$, $m_{\text{CHf}} = 0.17$ and $m_{\text{HFC}} = 0.13$. But their calculated energy distribution mismatch the corresponding experiment curves badly, see the Fig. 5 (smooth curves) on page 625 in reference [13].

In addition, even if the transport theory of sputtering only studied a structureless medium, we still believe that “the caging of a small light species by a big and heavy species shields the light species some what from collision”[13], if the simple power scattering cross section $(d\sigma = C dT/T)$ is used. Thompson distribution (7.1.3) derived based on the highest pole of (6.5.1), i.e. $s = 1$, transport theory. The range of validity of sputtering theory [16] is determined by the next highest pole $s_0$. Simple calculation
showed that the next highest pole \([s_0]_p > 0.6\) for HfC target, the contribution from this pole could disturb the sputtering yield energy distribution to make the theory be failed [19]. But using hard sphere scattering cross section \((d\sigma = \sigma dE/E)\), the next highest pole \([s_0]_Z < 0.15\) for HfC target, the pole only make minor effect [19]. Therefore, Szymoński’s experimental results [56] confirmed our hard sphere theory [19], even if the atomic mass ratio of the binary target is larger than ten. This fact indicates that the “caging” could be broken through by our hard sphere.

Typical angular distribution was given for elemental target Ag bombarded by 5 keV Ar⁺ ion [54]. The experimental data satisfies cosine-like angular distribution (7.1.4).

![Fig.7.1.3 Polar plot of the angular distribution of Ag sputtered from a polycrystaling silver sample by 5keV Argon ion. Solid curve is cosine curve.](image)

### 7.1.3 DEPTH DISTRIBUTION OF ORIGIN OF SPUTTERED ATOMS

For a single element target, the recoil density, \(F(z,E',\Omega')\), i.e.

\[
d^4F(z,E',\Omega') = F(z,E',\Omega')dE'd^2\Omega'dz
\]

(7.1.7)

is defined as the number of atom per incident ion (energy E) recoiling from depth layer \((z, dz)\) at
energy \((E', dE')\) into solid angle \((\Omega', d^2\Omega')\). Substituting (3.3.23) into (7.1.7) and replacing \(E\) by the depth distribution function of deposited energy, we obtained

\[
d^4F(z, E', \Omega') = \frac{2}{\pi} (\varepsilon')^{-2} \varepsilon' d^2\Omega' d\Lambda_m(z)
\]

(7.1.8)

Here \(\varepsilon' = E'/U_o\); \(z = XL_m\); \(L_m = L_m(U) = U^{2m}/(NC_m)\).

On the one hand, according to Falcone-Sigmund theory \([32][33]\), each recoil atom slows down continuously along a straight line, thus

\[
\cos \theta_o \cdot (dE/dZ) = -NS_n(E) = -(NC_m)E^{1-2m}
\]

(7.1.9)

Solving Eq. (7.1.9) with initial energy \(E'\) and final energy \(E_o\) yields

\[
\varepsilon' = \varepsilon_o \left(1 + \frac{2mX}{\varepsilon_o^{2m} \cos \theta_o}\right)^{1/2m} \quad \varepsilon_o \equiv E_o/U
\]

(7.1.10)

which determines the energy \(E_o\) of a recoil atom with initial energy \(E'\) after having traveled from \(z\) to the surface. Substituting (7.1.10) into (7.1.8) yields the atom flux just under the surface,

\[
d^4F(z, E_o, \Omega_o) = \frac{2}{\pi} (\varepsilon_o)^{-2} \left(1 + \frac{2mX}{\varepsilon_o^{2m} \cos \theta_o}\right)^{1-1/2m} \varepsilon_o d^2\Omega_o d\Lambda_m(z)
\]

(7.1.11)

Taking account of the refraction (7.1.2) and substituting

\[
\varepsilon_o = \varepsilon_1 + 1,
\]

\[
\cos \theta_o = \sqrt{(\varepsilon_1 \cos^2 \theta_1 + 1)/(\varepsilon_1 + 1)} \quad \text{and}
\]

\[
d^2\Omega_o = d^2\Omega_1 \cdot \frac{\varepsilon_1 \cos \theta_1}{[(\varepsilon_1 + 1)(\varepsilon_1 \cos^2 \theta_1 + 1)]^{0.5}}
\]

into (7.1.11), we obtained the double differential sputtering yield originated from depth \((z, dz)\),

\[
d^4F(z, E_1, \Omega_1) = \frac{2 \varepsilon_1 \cos \theta_1}{\pi (\varepsilon_1 + 1)^{(3-2m)}} d\varepsilon \varepsilon d^2\Omega d\Lambda_m(z) g(\varepsilon_1, \theta_1, X)
\]

(7.1.12)

Here \(g(\varepsilon, \theta, X) = (\varepsilon + 1)^{(0.5-2m)} [(\varepsilon \cos^2 \theta + 1)^{(0.5-2m)} (\varepsilon \cos^2 \theta + 1)^{0.5}]^{-1-1/2m}\)

(7.1.13)

Integrating (7.1.12) over \(X\) gives the double differential sputtering yield originated from all depth.
\[
\frac{d^3F(E_1, \Omega_1)}{\pi} = \frac{e_1 \cos \theta_1}{(e_1 + 1)^{(3-2m)}} \cdot d\varepsilon d^2\Omega \int_0^\infty dX\Lambda_m(z)g(e_1, \theta_1, X) \] 

(7.1.14)

Due to \( \int_0^\infty dXg(\varepsilon, 0, X) = 1 \), the escape depth of sputtered atom with energy and orientation \((E_1, \theta_1)\) is given by-

\[
\bar{X}(e_1, \theta_1) = \int_0^\infty dXg(e_1, \theta_1, X)X 
= \frac{1}{(1-2m)} \left( \frac{(e_1 + 1)^{(0.5-2m)}}{(e_1 + 1)^{(0.5-2m)}} \right) 
\] 

(7.1.15)

Integrating (7.1.12) over \((X, E_1)\) gives the angular differential sputtering yield originated from all depth,

\[
d^2F(\Omega) = \frac{B}{\pi} \cos \theta d^2\Omega \int_0^\infty dX\Lambda_m(z)h(\theta_1, X) 
\] 

(7.1.16)

Here \( h(\theta, X) \equiv \frac{4}{B\sin^2 \theta} \int_0^1 dy(l - y^2) \left[ 1 + \frac{2mX(y^2 - \cos^2 \theta)}{y} \right]^{2m} \] 

(7.1.17)

Due to \( \int_0^\infty dXh(0, X) = 1 \), the escape depth of sputtered atom with orientation \( (\theta_1) \) is given by

\[
\bar{X}(\theta) = \int_0^\infty dXh(0, X)X 
= \frac{4(1-m)}{(\sin^2 \theta)^{(1-2m)} \int_0^1 dy(l - y^2) \frac{y^2}{(y^2 - \cos^2 \theta)^{2m}}} 
\] 

(7.1.18)

Integrating (7.1.16) over solid angle, we finally obtained the total sputtering yield,

\[
Y = B \int_0^\infty dX\Lambda_m(XL_m)H(X) 
\] 

(7.1.19)

Here \( H(X) = \frac{4}{B} \int_0^1 dy \int_0^{y^2} dt \left( 1 + \frac{2mX}{y} \right)^{-1-2m} \] 

(7.1.20)

Since \( \int_0^\infty dXH(X) = 1 \), the escape depth of sputtered atoms is given by

\[
\bar{X} = \int_0^\infty dXH(X)X. 
= \frac{4(1-m)}{(5-8m)(1-4m)} \ \text{for} \ 0.25 > m \geq 0 
\] 

(7.1.21)

On the other hand, by using (6.6.13), (7.1.5) or (7.1.19) reduced to

\[
Y = \frac{B}{8} \frac{\Gamma_m}{N C_m} \frac{F_i(0, E, \eta)}{U_o^{1-2m}} = \frac{B[L_m(U_0) \Gamma_m]}{8U_o} N\alpha S_n(E) 
\] 

(7.1.22)
Considering \( \alpha_p \approx \alpha \) given by (6.6.14) and adjusting \( C_m \) makes \( Y_Z = Y_p \), thus, we came back to (6.3.10) and got
\[
\left[ L_m(U_0) \right]_Z / \left[ L_m(U_0) \right]_p = \left[ \Gamma_m \right]_p / \left[ \Gamma_m \right]_Z \quad \text{for} \quad 0.25 > m \geq 0
\] (7.1.23)

In a particular case, \( m = 0 \), (7.1.13,15), (7.1.17,18) and (7.1.20,21) reduced to
\[
\begin{align*}
g(\epsilon, 0, X) &= \left[ 1 / \tilde{g}(\epsilon, 0) \right] \exp[-X / \tilde{g}(\epsilon, 0)] \\
\tilde{g}(\epsilon, 0) &= \left[ (\epsilon \eta^2 + 1) / (\epsilon + 1) \right]^{0.5}
\end{align*}
\] (7.1.24)
\[
\begin{align*}
h(\theta, X) &= 4 \sin^{-4} \theta \cdot \int_0^H dy (1 - y^2) \exp(-X/y) \\
\tilde{h}(\theta) &= \frac{4}{\pi^2} [3\eta + (\eta + 1)^{-1} + (\eta + 1)^{-2}]
\end{align*}
\] (7.1.25)
\[
\begin{align*}
H(X) &= 4E_4(X) \sim \frac{4}{3} \exp(-\frac{4}{3}X) \\
\bar{H} &= 0.8
\end{align*}
\] (7.1.26)

Here \( \eta \equiv \cos \theta \) and \( E_n(Z) = \int_0^Z dy \cdot y^n \exp(-Z y) \) is the fourth order expint function. Formula (7.1.25,26) were derived by P. Sigmund et. al.[32]. Thus, (7.1.22) and (7.1.23) turns out to be
\[
Y_p = \frac{6}{\pi^2} \cdot \frac{\alpha S_n(E)}{8C_0 U_o}
\] (7.1.27)
\[
\begin{align*}
[\bar{Z}]_Z / [\bar{Z}]_p &= 6\pi^2 \approx 0.6097 \sim 0.5, \\
[\bar{Z}]_p &= 0.8/(NC_0)
\end{align*}
\] (7.1.28)

(7.1.27) is the famous Sigmund formula [1]. Sigmund estimated \( \bar{Z}_p = 0.75/(NC_0) \) [1]. One can clearly see that any adjustment of the constant \( C_0 \) influences not only the depth of sputtered atoms but also the total sputtering yield in Sigmund sputtering theory [57]. Our new theory very naturally gives a credible answer to the 'depth of origin' puzzle of ref. [28–31] [57–60], and means that hard sphere collisions dominant in the low energy cascade. Our new theory is obvious different from the traditional hard sphere model of sputtering [110][111], because the soft sphere \( (m > 0) \) collisions dominant in the primary or high energy stage, which is same as Sigmund theory. Therefore, except some new explanations, entire Sigmund theory of sputtering remains unchanged.
7.2 SURFACE COMPOSITION CHANGE DUE TO SPUTTERING OF ALLOY

7.2.1 STEADY STATE

In sputter process at steady-state, as a consequence of preferential sputtering, the surface concentration of a virgin target is changed during ion bombardment until a surface concentration $\alpha_k^s$ \((0 \leq \alpha_k^s \leq 1; \Sigma \alpha_k^s = 1)\) reached the steady-state, from which the atoms are sputtered with partial yield proportional to the bulk concentration $C_k$ \((1 \geq C_k \geq 0; \Sigma C_k = 1)\), i.e.,

$$Y_1/C_1 = Y_2/C_2 = \cdots = Y_n/C_n \quad \text{(7.2.1)}$$

Here the partial sputtering yield is given by $Y_j = Y_j^c \alpha_j^c$. Andersen Sigmund relation (7.1.6)

gives the sputtering yield ratio between component i and j,

$$\frac{Y_i^c}{Y_j^c} = \left(\frac{M_j}{M_i}\right)^{2m} \cdot \left(\frac{U_{0j}}{U_{0i}}\right)^{1-2m} \quad \text{(7.2.2)}$$

Substituting (7.2.2) into (7.2.1) yields

$$\left(\frac{\alpha_i^c}{\alpha_j^c}\right) = \eta_{ij} \left(C_i/C_j\right) \quad \text{(7.2.3)}$$

Here $\eta_{ij} = Y_i^c/Y_j^c = \left(\frac{M_j}{M_i}\right)^{2m} \cdot \left(\frac{U_{0j}}{U_{0i}}\right)^{1-2m}$, $\eta_{ij} = 1/\eta_{ij}$, $\eta_{ij} = \eta_{ik} \eta_{kj}$ and

$$\eta_{i2} \eta_{23} \eta_{31} \cdots \eta_{ni} = 1 \quad \text{for } n > 2 \quad \text{(7.2.4)}$$

If the surface binding energy ratio $\left(U_{0j}/U_{0i}\right)$ between two elements i and j is independent of alloy composition, $\eta_{ij}$ are constants and then, (7.2.3) can be solved easily

$$\alpha_i^c = C_i \left[\sum_{j=1}^{n} \eta_{ji} C_j\right]^{-1} \quad \text{(7.2.5)}$$

This relation has been proved approximately by Betz’s experimental measurements for a numerous binary alloy [104], such as Ag-Cu, Ag-Au, Cu-Pd, etc. For our purpose in this work, only three alloys Ag-Pd, Ag-Au and Cu-Pd need to be analyzed, because these alloys are directly related to a ternary alloy system Ag-Pd-Au, which usually is used for the comparison between different theoretical models [105][62].
Fig.7.2.1 Ratio of measured Auger peak heights versus ratio of bulk concentrations. Horizontal coordinate represents the ratio of bulk concentrations; Vertical coordinate represents the ratio of measured Auger heights. Dot line is the ratio of Auger peak heights before sputtering. Thin line is the ratio of Auger peak heights in steady state. Thick line represents the calculated results of (7.2.3).
(a) Ag-Pd alloy; (b) Pd-Au alloy (c). Au-Ag alloy.

i) Binary alloy systems

Several alloys of different bulk composition of the Ag-Pd system were sputtered by 2-keV Ar ions by Betz et al. [104]. They observed the surface enrichment of Pd. Experimental data \( \left( \frac{\alpha_{Ag}^s}{\alpha_{Pd}^s} \right) \) have been plotted in Fig.7.2.1(a) as a function of \( \left( \frac{\alpha_{Ag}}{\alpha_{Pd}} \right) \). If choosing \( \eta_{Ag-Pd} = 1/2.54 \), one can see that (7.2.3) fits experimental results reasonably well. For Pd-Au system, results have been plotted in Fig.7.2.1(b). The surface enrichment of Pd was observed. If choosing \( \eta_{Pd-Au} = 1.49 \), (7.2.3) fits experimental results qualitatively except in the region near \( c_{Pd}/c_{Au} \sim 3 \). For Ag-Au system, results have been plotted in Fig.7.2.1(c). The surface enrichment of Pd was observed. If choosing \( \eta_{Au-Ag} = 1.70 \), (7.2.3) fits experimental results very well.

ii) Ternary alloy system

As to a ternary alloy system Ag-Pd-Au (1 = 1-Ag; 2-Pd; 3-Au) under 2-keV Ar\(^+\) ion bombardment, the experimental results [104] are summarized in Table 7.2.1. Since

\[
\eta_{Ag-Pd} \cdot \eta_{Pd-Au} \cdot \eta_{Au-Ag} = \left( \frac{1}{2.54} \right) \cdot \left( 1.49 \right) \cdot \left( 1.70 \right) = 0.997 \approx 1,
\]

thus, (7.2.4) approximately holds, substituting \( \eta_{12} = \eta_{Ag-Pd} \), \( \eta_{23} = \eta_{Pd-Au} \) and \( \eta_{31} = \eta_{Au-Ag} \).
The calculated data are collected in Table 7.2.1 (New) and expressed by arrowheads in Fig. 7.2.2 respectively. The experimental data are expressed by small squares in Fig. 7.2.2 for the comparison. The agreement between calculated values and experimental data is much better than the results given in [105] at the point numbers: 3 → 3′, 5 → 5′, 6 → 6′, 7 → 7′, 8 → 8′ and 9 → 9′. This is not surprising conclusion, the previous authors neglected the nonstoichiometry factor [7][105] in sputtering of multi-component target, because the authors only used sputtering yield data of single element targets in their theory [105]. However the phenomena of bombardment-induced Gibssian Segregation during Alloy Sputtering [28][125] is ignored in the analysis.

Table 7.2.1 The surface composition change of ternary system Ag-Au-Pd under 2 keV argon ion bombardment

<table>
<thead>
<tr>
<th></th>
<th>$\alpha_{\text{Au}}$ *100</th>
<th>$\alpha_{\text{Ag}}$ *100</th>
<th>$\alpha_{\text{Pd}}$ *100</th>
</tr>
</thead>
<tbody>
<tr>
<td>1′</td>
<td>56.4 54.3 58.0  53.2</td>
<td>37.4 5.3 7.2  6.4</td>
<td>37.4 40.4 34.8  40.4</td>
</tr>
<tr>
<td>2′</td>
<td>36.6 33.2 37.2  31.9</td>
<td>57.2 4.9 7.0  7.0</td>
<td>57.2 61.9 55.8  61.1</td>
</tr>
<tr>
<td>3′</td>
<td>78.8 79.4 80.4  79.2</td>
<td>15.0 5.8 7.5  5.9</td>
<td>15.0 14.8 12.1  14.9</td>
</tr>
<tr>
<td>4′</td>
<td>28.0 25.8 28.6  24.8</td>
<td>60.5 10.1 14.3 14.3</td>
<td>60.5 64.1 57.1  60.9</td>
</tr>
<tr>
<td>5′</td>
<td>40.0 39.1 40.6  38.5</td>
<td>42.8 17.2 22.8 20.7</td>
<td>42.8 43.7 36.6  40.8</td>
</tr>
<tr>
<td>6′</td>
<td>63.8 64.8 63.5  65.7</td>
<td>16.3 19.1 23.8 19.0</td>
<td>16.3 16.1 12.7  15.3</td>
</tr>
<tr>
<td>7′</td>
<td>31.6 30.5 31.3  30.3</td>
<td>44.4 24.0 31.3 28.7</td>
<td>44.4 45.5 37.5  41.0</td>
</tr>
<tr>
<td>8′</td>
<td>37.7 37.4 34.5  40.2</td>
<td>20.0 44.0 51.7 43.7</td>
<td>20.0 18.6 13.8  16.1</td>
</tr>
<tr>
<td>9′</td>
<td>56.5 56.5 54.4  58.0</td>
<td>17.3 26.6 32.6 26.4</td>
<td>17.3 16.9 13.0  15.6</td>
</tr>
</tbody>
</table>

Note: “Exp.” stands for the experimental results given in [104]. “New” stands for theoretical data calculated by (7.2.5) in this work. A and B stand for theoretical data calculated by using the methods provided in [105] and [62] respectively.
Fig. 7.2.2 Concentration change during sputtering and the steady state established for the ternary system Ag- Au - Pd. The dark points stand for the bulk concentration. The target surface concentrations of the steady state are calculated by (7.2.5) and expressed by arrowheads. The experimental data are expressed by small squares for comparisons.

It’s contrary to Andersen Sigmund relation (7.1.6), if the partial sputtering yield ratio of two elements is related to another third element in the target, this phenomenon is called “matrix effect” proposed by Galdikas et. el [62]. Galdikas et. el introduced a matrix $\{Y_{ij}\}$ which satisfied

$$Y_i^c = \sum_j Y_{ij} \alpha_j^s$$  \hspace{1cm} (7.2.6)

Besides, the mass conservation (7.2.1) gives

$$\frac{Y_i^c}{Y_j^c} = \frac{C_i \cdot \alpha_j^s}{C_j \cdot \alpha_i^s}$$  \hspace{1cm} (7.2.7)

Thus, the matrix elements yield $Y_{ij}$ for a given energy of bombarding ion can be computed and are independent to the composition of target. Specifically for Ag-Pd-Au alloys under 2-keV $Ar^+$ ion bombardment, by using the matrix $Y_{ij}$ given by Galdikas et. el. in [62], solving Eqs. (7.2.6) and (7.2.7) simultaneously yields the surface composition $\alpha_i^s$ in the steady state. The calculated data are collected in Table 7.2.1, one can see that Andersen Sigmund relation (7.1.6) provided calculated values much better than the results given here at the point numbers: $2 \rightarrow 2'$, $4 \rightarrow 4'$, $6 \rightarrow 6'$, $7 \rightarrow 7'$, $8 \rightarrow 8'$ and $9 \rightarrow 9'$. 
Besides, seeing the experimental results showed that \( \eta_{ij} \) are approximately independent of the bulk concentration ratio \( C_i/C_j \) in many alloy sputtering [104], \( \eta_{ij} \) defined in (7.2.3) can be considered as constants. Thus, solving Eqs. (7.2.6) and (7.2.7) yields matrix elements
\[
Y_{ij} = Y_j \eta_{ji}, \quad Y_{ji} > 0 .
\] (7.2.8)
and (7.2.6) reduced to
\[
Y_{ii} = Y_{ii} \eta_{ii}, \quad Y_{jj} > 0 .
\] (7.2.8)

The target surface concentration \( \alpha^+_j \) in the steady state turned out to be (7.2.5). For Ag-Pd-Au alloys under 2-keV Ar\(^+\) ion bombardment, the matrix elements can be written as
\[
(Y_{ij}) = \begin{pmatrix}
Y_{11} & 2.54Y_{22} & 1.70Y_{22} \\
Y_{11}/2.54 & Y_{22} & Y_{11}/1.49 \\
Y_{33}/1.70 & 1.49Y_{22} & Y_{33}
\end{pmatrix}, \quad Y_{ji} > 0
\]
which reproduce the calculated data collected in Table 7.2.1 (New). Therefore, the “Matrix effect” (7.2.5) is nothing but an illusive effect.

In addition, does the “Matrix effect” theory can really describe “non-linear effect” appears in Pd-Au alloy sputtering [104]? The answer is negative. The “non-linear effect” means that \( \eta_{ij} \) is related to the bulk concentration ratio non-linearly. For simplicity, only a binary alloy is concerned, based on (7.2.6,7), it is easy to derive
\[
\frac{Y_{11}x + Y_{12}}{Y_{21}x + Y_{22}} = \frac{x}{y} \quad (7.2.9)
\]
Here \( x \equiv (\alpha_1/\alpha_2) \); \( y \equiv (\alpha_1^+/\alpha_2^+) \). For Pd-Au (Pd-1; Au-2) alloy, experimental results [104] yielded
\[
\begin{align*}
\text{Point 1} & : x = y = 2.33 \\
\text{Point 2} & : x = 0.2; y = 0.3
\end{align*} \quad (7.2.10)
\]

Substituting (7.2.10), \( Y_{11} = 3.90 \) and \( Y_{22} = 4.90 \) [62] into (7.2.9), one obtained the matrix element of sputtering yields
\[
(Y_{ij}) = \begin{pmatrix}
3.90 & 2.68 \\
2.95 & 4.90
\end{pmatrix} \quad (7.2.11)
\]
Solving Eqs. (7.2.6,7) yielded,
For \( x = 10 \), (7.2.12) gives \( y = 8.37 \) which means the surface enrichment of Au. But, the experiment measurements [104] showed the surface enrichment of Pd. Therefore, the “matrix effect” theory cannot predict the “non-linear effect” appears in Pd-Au alloy sputtering [104].

7.2.2 TRANSIENT STATE

A model for the evolution of the surface composition proposed by J. M. Mayer et. al. for a binary target [65]. Mayer’s model predicted the surface chemical composition change of PtSi target surface bombarded by energetic ion as a function of the ion flux until the steady state established.

We are now developing the model to a multi-component target and find exact solution. By using the mass conservation

\[
N \int_0^\infty dz \left[ \frac{d\alpha_i(\phi, z)}{d\phi} \right] = Y_i - Y_i(\phi), \quad i = 1, 2, \cdots, n
\]

(7.2.13)

Here \( \phi \) is the incident ion dose per unit area (cm\(^2\)). \( Y_i(\phi) \) is the partial sputtering yield of \( i \)-atoms as a function of \( \phi \), and \( Y_i(\infty) = Y_i \). \( N\alpha_i(\phi, z) \) stands for the density of \( i \)-atoms per unit volume as a function of the depth \( z \), \( \sum_j \alpha_j(\phi, z) = 1 \). Specially, we have \( \alpha_i(0, z) = C_i \) and \( \alpha_i(\infty, z) = \alpha_i^i \). By using the steady-state condition (7.2.1), we have derived

\[
N \int_0^\infty dz \left[ C_i^{-1} \alpha_i(\phi, z) - C_j^{-1} \alpha_j(\phi, z) \right] = C_j^{-1} Y_j(\phi) - C_i^{-1} Y_i(\phi) \quad \text{or} \quad \text{or}
\]

(7.2.14)

\[
N \int_0^\infty dz \left[ C_i^{-1} \alpha_i(\phi, z) - C_j^{-1} \alpha_j(\phi, z) \right] = C_j^{-1} \int_0^\infty Y_j(\phi')d\phi' - C_i^{-1} \int_0^\infty Y_i(\phi')d\phi'
\]

(7.2.15)

Mayer’s equation (Eq.(1a) on page 5302 in Ref. [65]) is obviously a special case of Eqs. (7.2.15). Mayer et. al. further approximated the profiles \( \alpha_i(\phi, z) \) by simple linear functions with a depth parameter \( W \). The depth parameter \( W \) plays a role of the thickness of the altered layer [131], \( \alpha_i(\phi, z = 0) = \alpha_i(\phi) \) and \( \alpha_i(\phi, z \geq W) = C_i \). By using the formula of area of triangle, we got

\[
\int_0^\infty dz \left[ C_i^{-1} \alpha_i(\phi, z) - 1 \right] = 0.5W[C_i^{-1} \alpha_i(\phi) - 1],
\]

(7.2.16)
Substituting (7.2.16) into the left-hand side of Eqs. (7.2.14), we obtained

\[
\beta^{-1} \frac{d}{d\phi} \left[ C_i^{-1} \alpha_i(\phi) - C_j^{-1} \alpha_j(\phi) \right] = C_j^{-1} Y_j(\phi) - C_i^{-1} Y_i(\phi)
\]  
(7.2.17)

Here \( \beta^{-1} \equiv 0.5WN \). Further substituting \( Y_j(\phi) = Y_j^\alpha \alpha_j(\phi) \) into Eqs. (7.2.17), we finally derived a set of equations,

\[
\beta^{-1} \frac{d}{d\phi} \left[ C_i^{-1} \alpha_i(\phi) - C_j^{-1} \alpha_j(\phi) \right] = C_j^{-1} Y_j^\alpha \alpha_j(\phi) - C_i^{-1} Y_i^\alpha \alpha_i(\phi)
\]  
(7.2.16)

which can be solved exactly with initial condition \( \alpha_i(0) = C_i \) or \( \alpha_i(\infty) = \alpha_i^\alpha \). Here, \( \alpha_i^\alpha \) also stats for the surface concentration of steady-state and is given by (7.2.5).

i) Binary alloy systems

The exact solution of Eqs. (7.2.16) turns out to be

\[
\begin{align*}
\left[ (C_1 Y_1^c + C_2 Y_1^c) \right] \alpha_1(\phi) &= C_1 \cdot \left[ Y_2^c + C_2 \left( Y_1^c - Y_2^c \right) \exp(-\beta AY\phi) \right] \\
\left[ (C_1 Y_2^c + C_2 Y_1^c) \right] \alpha_2(\phi) &= C_2 \cdot \left[ Y_1^c - C_1 \left( Y_1^c - Y_2^c \right) \exp(-\beta AY\phi) \right]
\end{align*}
\]  
(7.2.17)

Here \( A \equiv (C_1 Y_1^c + C_2 Y_1^c)/(C_1 Y_1^c + C_2 Y_2^c) \) and \( Y \equiv C_1 Y_1^c + C_2 Y_2^c \). J. M. Mayer et. al. presumed the total sputtering yield

\[
Y(\phi) \equiv \alpha_1(\phi) Y_1^c + \alpha_2(\phi) Y_2^c = (Y_1^c - Y_2^c) \alpha_1(\phi) + Y_2^c
\]

as a constant, so couldn’t give analytical expressions for their final results [65]. Although a similar solution was given by Harrid et. al. long time ago [127], however, (7.2.17) is much easier to use in practice. In particular, the surface concentration ratio, as a function of \( \phi \), can be easily derived,

\[
\frac{\alpha_1(\phi)}{\alpha_2(\phi)} = \frac{C_1 \cdot r - C_1(r-1) \exp(-\beta AY\phi)}{C_2 \cdot 1 + C_1(r-1) \exp(-\beta AY\phi)}
\]  
(7.2.18)

Here \( r \equiv Y_2^c/Y_1^c \) and \( A \equiv (C_1 r + C_2)/(C_1 + C_2 r) \).
Fig.7.2.3 Increase of the surface Pt/Si concentration ratio with argon ion dose in 80-keV argon ion sputtering of PtSi. The measured values were taken from literature [65]. The solid curve was calculated by using our formula (7.2.19). The theoretical curve of Mayer’s model [65] is also plotted for the comparison.

Comparing with experiment I: The sputtering of PtSi by Ar$^+$ ions of 80-keV was carried out by J. M. Mayer et. al. [65] and increase of the surface Pt/Si concentration ratio 
\[
\left( \frac{N_{Pt}}{N_{Si}} = \frac{\alpha_{Pt}}{\alpha_{Si}} \right)
\]
was plotted in Fig.7.2.3 as a function of \( \phi \). The theoretical curve of Mayer’s model [65] was also plotted in Fig.7.2.3 for the comparison. In our theory, substituting 
\[
C_{Pt} = C_{Si} = 0.5, \quad NW = 4.5 \times 10^{17} \text{ cm}^{-2} \quad (W \sim 75 \text{nm}) \quad [65], \quad A = 1, \quad Y = 4 \quad [65]
\]
and 
\[
r = Y_{Si}^c / Y_{Pt}^c = 1.94
\]
into (7.2.18), we got 
\[
\frac{\alpha_{Pt}(\phi)}{\alpha_{Si}(\phi)} = 1.94 \times \frac{1 - 0.2423 \exp[-(16/9)F]}{1 + 0.4700 \exp[-(16/9)F]}, \quad \phi \equiv F \times 10^{17} \text{ cm}^{-2} \quad (7.2.19)
\]
which was plotted in Fig.7.2.3 which demonstrates both theoretical curve agree with experimental measurement very well.

Comparing with experiment II: The sputtering of AgAu alloy by Ar$^+$ ions of 100-keV was carried out by H. H. Andersen et. al. [130] and decrease of the surface Ag/Au concentration ratio 
\[
\left( \frac{N_{Ag}}{N_{Au}} = \frac{\alpha_{Ag}}{\alpha_{Au}} \right)
\]
was plotted in Fig.7.2.4 as a function of \( \phi \). In our theory, substituting 
\[
C_{Ag} = 0.427, \quad C_{Au} = 0.573, \quad NW = 224 \text{ layers} \quad (W \sim 85 \text{nm}), \quad A = 0.912 \quad \text{and}
\]

\[ r = \frac{Y_{Ag}^c}{Y_{Au}^c} = 1.92 \] into (7.2.18), we got

\[
\frac{\alpha_{Ag}(\phi)}{\alpha_{Au}(\phi)} = 0.388 \times \left\{ \frac{1 + 0.393 \exp[-(1/15.4)F]}{1 - 0.275 \exp[-(1/15.4)F]} \right\}; \quad \phi \equiv F \times 8 \text{ layers.} \tag{7.2.20}
\]

which was plotted in Fig.7.2.4 which demonstrates our theoretical curve agrees with experimental measurement very well.

**Fig.7.2.4** Decrease of the surface Ag/Au concentration ratio with argon ion dose in 100-keV argon ion sputtering of AgAu alloy. The measured values were taken from literature [130]. The solid curve was calculated by using our formula (7.2.20).

**ii) Ternary alloy system**

Eqs. (7.2.16) turns out to be

\[
\begin{align*}
\beta^{-1} d\alpha_1(\phi)/d\phi &= C_1 Y_3^c - \left[ Y_1^c + C_1 (Y_1^c - Y_4^c) \right] \alpha_1(\phi) + C_2 (Y_2^c - Y_3^c) \alpha_2(\phi) \\
\beta^{-1} d\alpha_2(\phi)/d\phi &= C_2 Y_3^c + C_2 (Y_1^c - Y_3^c) \alpha_1(\phi) - \left[ Y_2^c + C_2 (Y_3^c - Y_2^c) \right] \alpha_2(\phi)
\end{align*}
\tag{7.2.21}
\]

Let \( X \equiv \beta Y\phi \), \( \overline{\alpha}_i(X) \equiv \alpha_i(\phi) - \alpha_i^* \), \( Y \equiv C_1 Y_1^c + C_2 Y_2^c + C_3 Y_3^c \),

Eqs. (7.2.21) reduced to

\[
\frac{d}{dX} \left( \overline{\alpha}_1(X) \right) = \begin{pmatrix}
 r_1 - C_1 (r_1 - r_3) & -C_1 (r_2 - r_3) \\
 -C_2 (r_1 - r_3) & r_2 - C_2 (r_2 - r_3)
\end{pmatrix} \left( \overline{\alpha}_i(X) \right)
\tag{7.2.22}
\]

Here \( r_i \equiv Y_i^c/Y \) or \( r_1 = (C_1 + C_2 \eta_{31} + C_3 \eta_{13})^{-1} \), \( r_i = (C_1 \eta_{12} + C_2 + C_3 \eta_{32})^{-1} \) and
\[ r_i = (C_{1} \eta_{31} + C_{2} \eta_{32} + C_{3}^{-1})^{-1}. \] Eqs. (7.2.22) can be solved exactly with initial condition
\[ \overline{\alpha}_i(0) = C_i - \alpha_i, \text{ and } \overline{\alpha}_i(\infty) = 0. \] Substituting
\[
\begin{pmatrix}
\overline{\alpha}_1(X) \\
\overline{\alpha}_2(X)
\end{pmatrix} =
\begin{pmatrix}
u \\
v
\end{pmatrix}
\exp(-\lambda X)
\] (7.2.23)
into Eqs. (7.2.22), we obtained
\[
\begin{pmatrix}
\lambda - r_i + C_1 (r_1 - r_3) & C_1 (r_2 - r_3) \\
C_2 (r_1 - r_3) & \lambda - r_2 + C_2 (r_2 - r_3)
\end{pmatrix}
\begin{pmatrix}
u \\
v
\end{pmatrix} = 0
\] (7.2.24)
Solving the characteristic equation of Eq.(7.2.24)
\[
\begin{vmatrix}
\lambda - r_i + C_1 (r_1 - r_3) & C_1 (r_2 - r_3) \\
C_2 (r_1 - r_3) & \lambda - r_2 + C_2 (r_2 - r_3)
\end{vmatrix} = 0
\] (7.2.25)
we got both eigenvalues \( \lambda_1 \) and \( \lambda_2 \). Solving Eq. (7.2.24) for each eigenvalue, \( \lambda_i \) yields a normalized eigenvector \( (u_i, v_i)^T \). The linear combination of (7.2.23) gives the general solution of Eqs. (7.2.22), finally we have
\[
\begin{pmatrix}
\alpha_1(\phi) \\
\alpha_2(\phi)
\end{pmatrix} =
\begin{pmatrix}
\alpha_1^s \\
\alpha_2^s
\end{pmatrix} + x\begin{pmatrix}
u_1 \\
v_1
\end{pmatrix}
\exp(-\lambda_1 \beta \phi) + y\begin{pmatrix}
u_2 \\
v_2
\end{pmatrix}
\exp(-\lambda_2 \beta \phi)
\] (7.2.26)
The initial condition
\[
\begin{cases}
u_1 x + \nu_2 y = c_1 - \alpha_1^s \\
u_1 x + \nu_2 y = c_1 - \alpha_1^s
\end{cases}
\] (7.2.27)
determines coefficients \( x \) and \( y \) in (7.2.26).

The entire procedure can be easily generalized to any multi-component medium \( n > 3 \).

Comparing with experiment III: The sputtering a ternary alloy system Ni-Cu-Pt \( (i=1-Ni; 2-Cu; 3-Pt) \) by \( \text{Ar}^+ \) ions of 2-keV was carried out by G. Betz [104] and explained by Q. Zhe theoretically [105]. In the case, \( C_1 = 34.8\%, C_2 = 33.7\% \) and \( C_3 = 32.0\%. \) In our theory, using the date of corresponding binary alloy [104], we obtained \( \eta_{23} = 1.5, \eta_{31} = 1.39 \) and \( \eta_{21} = 2.09 \). Immediately, we have \( \alpha_1^s = 42.0\%, \alpha_2^s = 26.8\% \) and \( \alpha_3^s = 31.3\%. \) In the
literature, we got that sputtering 40 layers target material off needs 3.574 minutes time [104]. Substituting $\eta_i$, $C_i$ and $NW = 40$ layers into Eqs. (7.2.22-25), solving them, we got $\lambda_1 = 1.117$ and $\lambda_2 = 0.897$. Finally, we obtained the following exact solution [132],

$$ \begin{aligned} 
\alpha_{Ni}(t) &= (34.3\%) \left[ 1.2263 - 0.01602 \exp(-t/1.25) - 0.2103 \exp(-t/1.55) \right] \\
\alpha_{Cu}(t) &= (33.7\%) \left[ 0.7958 + 0.1592 \exp(-t/1.25) + 0.0449 \exp(-t/1.55) \right] \\
\alpha_{Pt}(t) &= (32.0\%) \left[ 0.9789 - 0.1173 \exp(-t/1.25) + 0.1386 \exp(-t/1.55) \right] 
\end{aligned} $$  

(7.2.26)

Here, the time $t$ is in minute. The solution (7.2.26) was plotted in Fig. 7.2.5. The figure demonstrates our theoretical curve agrees with experimental measurement very well. The remarkable point is that two exponential terms appeared in the expression of $\alpha_{Pt}$ seem to be cancel each other, therefore, $\alpha_{Pt}$ couldn’t change much in the entire sputtering process.

![Fig. 7.2.5](image)

**Fig. 7.2.5** The surface composition change process of the triplet target Ni-Cu-Pt sputtered by 2-keV argon bombardment. The measured values were taken from literature [104]. Three solid curves were calculated by using our formula (7.2.26).
CHAPTER 8
ANISOTROPIC SPUTTERING

8.1 ANISOTROPIC SPUTTERING THEORY

8.1.1 ANISOTROPIC SPUTTERED PARTICLE SPECTRUM

According to Sigmund “two steps” theory \([7][19][24]\), replacing the energy \(E\) and momentum \(P\) of incident particle in (6.4.7) by energy and momentum deposited in the target surface, we derived the outward particle current \(J_j dE \, d^2\Omega\) of target atoms of type \(j\) \([1][7]\),

\[
J_j(E_0, \Omega_0) = \frac{\Gamma_m \cos \theta_j}{4\pi} \left[ \frac{F_{(i)}(0,E,\eta)}{E_0} + 3A_i \frac{M_j}{M_i} \frac{\bar{n}_j \cdot \bar{P}_{(i)}(0,E,\eta)}{\sqrt{2M_jE_0}} \right]. \tag{8.1.1}
\]

The energy deposited in the target surface was given by (6.6.13), i.e.

\[
F_{(i)}(0,E,\eta) = N\alpha S_a(E). \tag{8.1.1a}
\]

\(\alpha\) is a dimensionless functions of atomic mass ratios and incident angle. For low energy and heavy ion bombardment, according to (6.6.15) and the basic assumption of the momentum deposited in target surface in Chapter 2, we have

\[
\bar{P}_{(i)}(0,\bar{v},U) = \sqrt{2M_j/E} \cdot N\alpha S_a(E) \cdot \bar{v}/\nu \quad \text{and} \quad \alpha > 0 \tag{8.1.1b}
\]

\(\alpha\) is not only a dimensionless functions of atomic mass ratios and incident angle, but also relates to \(U/E\). Obviously, we have \(\alpha = p(0,u,1)\) for the perpendicular incidence. In principal, (8.1.1) can be considered as a basis for anisotropic theory of sputtering of multi-component target \([7][19][24]\).

Substituting (8.1.1a) and (8.1.1b) into (8.1.1), we have

\[
J_j(E_0, \Omega_0) = \frac{G_j}{(G) j} \frac{\Gamma_m \alpha S_a(E)}{\bar{P}_{(i)}E_0^2} \frac{\cos \theta_j}{4\pi} \left[ 1 - 3\cos \hat{\theta} \cdot \left( A_i \frac{\alpha}{\alpha} \right) \frac{M_jE_0}{M_iE} \right], \tag{8.1.2}
\]

Here \(\cos \hat{\theta} = \cos \theta \cos \theta_0 - \sin \theta \sin \theta_0 \cos \phi_0\).

(1) Primary sputter process

For a perpendicular incidence, taking account of the refraction by the planar surface (7.1.2),
(8.1.2) turned out to be the anisotropic flux of sputtered atom-j moving with energy \( E_j \) and along the emission direction \((\theta_j, \phi_j)\) through the target plane surface.

i) Energy distribution of sputtered atoms

\[
\frac{d^1 Y_j}{dE_j d\Omega_j} = Y_{mj} \frac{\cos \theta_j}{\pi U_{oj}} \frac{\varepsilon}{(\varepsilon + 1)^{3/2m}} \left[ 2 - H_j(E) \sqrt{\varepsilon \cos^2 \theta_j + 1} \right]
\]

(8.1.3)

Here \( E_T = 36U_0 (A_j \alpha/\alpha)^2 \) is just a parameter and does not play the role of sputtering threshold energy \( E_{th} \) in literatures \([4-6]\) and \([12]\),

\[
H_j(E) = \sqrt{M_j U_{oj} E_T / (M_i U_{oj} E)}
\]

(8.1.4)

ii) Under-cosine like angular distribution of sputtered atoms \([54]\) Integrating (8.1.3) over energy \( E_j \) and considering a natural boundary condition

\[
J_j(E_j < \gamma E_i \Omega) = 0
\]

(8.1.5)

we obtained the angular spectrum of sputtered atoms,

\[
\frac{d^2 Y_j}{d^2 \Omega_j} = Y_{mj} \frac{\cos \theta_j}{4\pi} A_j \left[ 4 - \gamma_m(\theta_j) H_j(E) \right]
\]

(8.1.6)

Here \( \frac{1}{2} \pi \geq \theta_j \geq 0 \), \( \xi_j \equiv \gamma_j E / U_{oj} \),

\[
A_j \equiv \frac{2}{(1-2m)} (1 - \xi_j^{2m-1}) - \frac{1}{(1-m)} (1 - \xi_j^{2m-2})
\]

\[
\gamma_m(\theta_j) \equiv \frac{4}{A_j} \int_0^{\xi_j^{-1}} \frac{\varepsilon dx}{(\varepsilon + 1)^{3/2m}} \sqrt{\varepsilon \cos^2 \theta_j + 1}
\]

iii) Total partial sputtering yield

Further integrating (8.1.5) over \( \Omega_j \) gave the total sputtering yield

\[
Y_j = Y_{mj} A_j [1 - B_j H_j(E)]
\]

(8.1.7)

Here \( B_j \equiv \frac{1}{3A_j} \left[ \frac{2}{(0.5-2m)} (1 - \xi_j^{2m-0.5}) - \frac{1}{(1-m)} (1 - \xi_j^{2m-2}) \right] \)

(2) Sputter process at steady state

If total partial sputtering yields (8.1.7) are not proportion to the corresponding concentrations
on the target surface, preferential sputtering happen. As a consequence, the surface concentration of a virgin target is changed during ion bombardment until a surface concentration \( \alpha_k \) reached the steady-state, from which the atoms are sputtered with partial yields proportional to the relative atomic concentration \( C_k \) in the bulk, i.e. (7.2.1). By using (7.1.6), solving (7.2.1) in conjunction with (8.1.7) yielded,

\[
\alpha_j = C_j \cdot \sum_k \alpha_k \frac{A_k [1 - B_k H_k(E)]}{A_j [1 - B_j H_j(E)]} \left( \frac{M_j}{M_k} \right)^{2m} \left( \frac{U_{oi}}{U_{ok}} \right)^{1-2m}, \tag{8.1.8}
\]

which can be solved by using an iteration method with initial condition: \( \alpha_k = C_k \). Then, substituting \( \alpha_j \) into (8.1.3) and (8.1.6,7), we can obtain the energy and angular distributions of sputtered atoms, as well as the partial sputtering yield at the steady-state.

**8.1.2 COMPARING WITH COMPUTER SIMULATIONS: ISOTOPE PUZZLE OF SPUTTERING**

If the target is isotopic mixture medium, the surface binding energy for different components is the same, i.e. \( U_{0j} = U_{0i} \). Therefore, the isotopic medium provides the simplest model systems for the study of the sputtering of multi-component materials. In this case, only kinematics effects due to mass differences between the isotopes contribute to their preferential emission during sputtering. Besides, up to now, Sigmund conventional (isotropic or \( L = 0 \)) sputtering theory \[17\] seems couldn’t describe the unusual isotopic medium sputtering effect. A critical problem is the very large isotope enrichments (the isotopic puzzle) observed by Wehner et al.\[85\][86] and Gnaser et al. \[106\] at very low ion energy \( \leq 300\text{ev} \). In order to solve this problem, a variety of numerical simulations have been down by Shulga et al.\[101\][107] and Eckstein et al \[108\] to study the unusual behaviors of isotope sputtering.

1. Primary sputter process

Taking the ratio of the sputtering yields of the two species, by the use of the general relation (7.1.6), we obtained
The sputtering yield ratio and enrichment exponent $m_{\text{eff}}$ for a 50/50% isotope mixture of polycrystalline $^{100}\text{Mo}$-$^{92}\text{Mo}$, have been simulated by Shulga et al. [101][107], as functions of $\text{Ar}^+$-ion energy. In our analytic theory, substituting the parameters $m = \frac{\gamma}{6}$ [15], $M_1 = 40$ (Ar), $M_2 = 100$, $U_0 = 6.82$ eV (8.1.11) and $\alpha_1 = \alpha_2 = 0.5$, $M_2 = 98$, $E_T = 32$ eV into (8.1.10), $Y_1/Y_2$ and $m_{\text{eff}}$ were calculated and plotted the results in Fig.8.1.1(a), one can see that our analytic results agree with the simulations very well. In addition, $Y_1/Y_2$, $m_{\text{eff}}$ and dependence of sputter yield ratio on ejection angle for different ion energies, also have been simulated for a 50/50% isotope mixture of polycrystalline $^{100}\text{Mo}$-$^{50}\text{Mo}$ [101][107]. In this case, substituting (8.1.11) and $\alpha_1 = \alpha_2 = 0.5$, $M_2 = 50$, $E_T = 36$eV into (8.1.9,10), we calculated them and plotted the results in Fig. 8.1.1 (a) and (b). Again, one can see that our analytic results agree with the correspondent simulations very well. A remarkable feature of our analytic theory is that, using only one common parameter $E_T = 36$ eV, (8.1.9,10) can reproduce both simulation curves in Fig.8.1.1 (a) and (b), particularly in low energy region ($E \sim 100$eV). In addition, for the fixed reference mass, $M_1 = 100$, $E_T$ seems insensitive to $M_2$ within the region $100 \geq M_2 \geq 50$. The nonstoichiometric emission of sputtered atoms different from the target composition can be described by the fractionation $\delta$ [108].

By using (8.1.9), we obtained

$$
\delta_Y = \left( \frac{M_2}{M_1} \right)^{2m} \frac{A_1[1-B_1H_1(E)]}{A_2[1-B_2H_2(E)]} - 1
$$

(8.1.12)

Owing to the fixed reference mass, $M_1 = 100$, the fractionation $\delta_Y$ has been simulated by
Eckstein et al. [108] versus the isotope mass $M_2$ ($100 \geq M_2 \geq 92$) for the bombardment of Mo with $Ar^+$ at different incident energies and normal incidence. In our analytic theory, substituting the parameters (8.1.11) and $E_T = 29.6\text{eV}$ into (8.1.12), $\delta_Y$ was calculated and plotted in Fig. 8.1.2 (a), one can see that our analytic results agree with the simulations well in low energy region. Eckstein et al. [108] also simulated the fractionation versus the incident energy for four different isotope mass $M_2$ ($M_2 = 92, 94, 96,$ and $98$). In our analytic theory, substituting the same group of parameters, the formulae (8.1.10) once again reproduces the simulations, as shown in Fig. 8.1.2 (b). The reason for choosing two different energy parameters $E_T$ for a same ion-target combination in Ref. [101], [107] and [108] obviously is that two simulations may use two different inter-atomic potentials. By using (8.1.3), we can derive the fractionation $\delta_E$ of the energy distribution of sputtered atoms,

$$\delta_E = \left(\frac{M_2}{M_1}\right)^{2m} \left[\frac{2 - H_1(E)}{2 - H_2(E)} \sqrt{\varepsilon \cos^2 \theta_i + 1}\right] - 1 \quad (8.1.13)$$

$\delta_E$ has been simulated by Eckstein et al. [108] for $E = 0.05, 0.1, 0.5,$ and $5\text{keV} Ar^+$ normally incident on Mo consisting of $50\%$ mass $100$ and mass $92$, the results were plotted in Fig. 8.1.2 (c). On the other hand, inserting the parameters (8.1.11), $M_2 = 92, E_T = 29.6\text{eV}$ and $\theta_i = 0$ into (8.1.13), the simulation results have been reproduced reasonably well in Fig. 8.1.2 (c).

**Fig. 8.1.1** Comparison of anisotropic ($L = 0,1$) sputtering theory (AST) with simulations given by Shulga and Sigmund [101][107]. (a): The sputtering yield ratio and enrichment effective exponent $m$ for a $50/50\%$ isotope mixture of a polycrystalline target as a function of argon ion energy. (b): Dependence of the sputter yield ratio on ejection angle for different ion energies.
(a),(b),(c): Fractionation of sputtering yield versus the isotope for the bombardment with argon ion at different incident energies. (d): Differential sputtering yield and fraction versus the cosine of the polar emission angle at the steady-state.

(2) Surface concentration at the steady state

For the isotope sputtering, (8.1.8) reduced to

\[
\alpha_j = C_j \cdot \sum_k \alpha_k \frac{A_k [1 - B_k H_k (E)]}{A_j [1 - B_j H_j (E)]} \left( \frac{M_j}{M_k} \right)^{2m} \]  \hspace{1cm} (8.1.14)

The influence dependence of the relative surface concentration has been simulated by Eckstein et al [108] for \( E = 100eV \) Ar\(^+\) incident on Mo target consisting of 50% mass 100 and mass 92. In our analytic theory, substituting (8.1.11) and \( C_1 = C_2 = 0.5, M_2 = 92 \) and \( E_T = 29.6eV, E = 100eV \) into (8.1.14), immediately we obtained \( \alpha_1 = 0.526 \) and \( \alpha_2 = 0.474 \). The results agree with the simulations very well, as shown in Fig.18 of [108]. Another example is the fractionation of the relative surface concentration versus the incidence for a natural Mo target [108] bombarded with 100eV Ar\(^+\) at normal incidence. The simulation results have been plotted in
Fig. 19 of [108]. In our analytic theory, substituting $M_k, C_k$, the parameters describing the target given in Table 8.1.1, and $m = \gamma [101][107]$, $M_i = 40$ (Ar), $U_0 = 6.82$ eV, $E_T = 29.6$ eV, and $E = 100$ eV into (8.1.14), by using the iteration method, we obtained the surface concentrations $\alpha_k$ at the steady state. Then, inserting $\alpha_k$ into

$$\delta_k = \frac{(\alpha_k C_7)}{(\alpha_7 C_k)} - 1, \ k = 1, 2, \ldots, 7,$$

one obtained the fractionation of the relative surface concentration $\delta_k$. The calculated data for $\alpha_k$, and $\delta_k$ are collected in Table 8.1.1, the simulation results for $\delta_k$ given in [108] are also listed in Table 8.1.1 for comparison, one can see that our analytic results agree with the simulations reasonably well.

In addition, the sputtering yield angular distributions at the steady state have been simulated for the same ion-target system in [108]. Inserting (8.1.11) and $\alpha_1 = 0.526, \ \alpha_2 = 0.474, \ M_\gamma = 92$ into (8.1.6) and (8.1.9), plotting the results in Fig. 8.1.2 (d), one can see that our analytic results agree with the simulations very well and the present theory predicts “under-cosine” angular distribution. Anisotropic ($L = 0, 1$) sputtering formula (8.1.1) predicted that the ratio of the anisotropic term and the isotropic term must be negative, and proportional to $\cos \theta_j \sqrt{M_j E_0 / E}$ in magnitude for a normal incidence. This is the intrinsic reason why isotopic mixtures are characterized by higher erosion rates for lower isotopic masses and vice versa [101][107] for a low energy ion bombardment. In particular, the anisotropic term could be comparable with the isotropic term in magnitude for the near threshold energy ion incidence. The former may cancel the most part of the latter. Thus, the isotopic effect could be magnified and become even more pronounced in some ratios. Therefore, the anisotropic ($L = 0, 1$) sputtering formula (8.1.1) successfully solved the isotope puzzle in low energy sputtering.
Table 8.1.1 Calculated values of $\alpha_k$, $\delta_k$ and Simulation data of $\delta_k$

<table>
<thead>
<tr>
<th>k</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
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<tr>
<td>$M_k$</td>
<td>92</td>
<td>94</td>
<td>95</td>
<td>96</td>
<td>97</td>
<td>98</td>
<td>100</td>
</tr>
<tr>
<td>$\alpha_k$ (%)</td>
<td>14.06</td>
<td>9.00</td>
<td>15.70</td>
<td>16.67</td>
<td>9.68</td>
<td>24.8</td>
<td>10.14</td>
</tr>
<tr>
<td>$\delta_k$ (%)</td>
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<td>-7.546</td>
<td>-6.323</td>
<td>-5.087</td>
<td>-3.836</td>
<td>-2.572</td>
<td>0</td>
</tr>
<tr>
<td>$\delta_k$ (%)</td>
<td>-8.2</td>
<td>-5.5</td>
<td>-4.6</td>
<td>-3.5</td>
<td>-2.8</td>
<td>-1.7</td>
<td>0</td>
</tr>
</tbody>
</table>

8.2 ANISOTROPIC SPUTTERING THEORY AND EXPERIMENTS OF MONO-ATOMC TARGET

8.2.1 MODIFIED SIGMUND’S FORMULAE

Since the anisotropic sputtering formula (8.1.1) provided a unique successful solution for the isotope puzzle in simulation results of low energy ion sputtering, it hints broadly that the theory can be used to explain the experimental results of anisotropic sputtering induced by low energy heavy ion bombardment. The most remarkable feature of this theory is that, only one free energy parameter, $E_z$, can determine both differential and total sputtering yields.

i) Energy distribution of sputtered atoms [67][68]

For elemental targets, only the primary sputter process needs to be considered. As we mentioned previously, in order to explain experimental data [55] of sputtering yield energy distribution, we have to choose $m = 0$. Thus, (8.1.2) reduced to

$$J(E_0, \Omega_0) = Y_p \frac{U_0}{E_0^2} \frac{2|\cos \theta_0|}{\pi} \left[ 1 - \frac{1}{2} \cos \theta \sqrt{\frac{E_0 E_{th}}{U_0 E}} \right]$$

(8.2.1)

Here $E_{th} = E_T \cdot \left( \frac{M_1}{M_2} \right) = 36U_0 \left( \frac{A_1}{A} \right)^2 \left( \frac{M_1}{M_2} \right)$. Taking account of the refraction by
the planar surface (7.1.2), formula (8.2.1) turns out to be the differential recoil flux of sputtered atoms moving with energy $E$ and along the emission direction $\left(\theta_1, \phi_1\right)$ through the target plane surface,

$$
\frac{d^3Y}{dE \ d^2\Omega} = Y_p \frac{2|\cos \theta_1|}{\pi U_0} \cdot \frac{\varepsilon}{(\varepsilon + 1)^3} \cdot 
\left\{1 - \frac{1}{2} \sqrt{\frac{E_{\text{th}}}{E}} \cos \theta \sqrt{\varepsilon \cos^2 \theta_1 + 1 + \sqrt{\varepsilon} \sin \theta \sin \theta_1 \cos \phi_1}\right\}, \quad \varepsilon = \frac{E}{U_0}.
$$

(8.2.2)

ii) Angular distribution of sputtered atoms [69–73]

Neglecting (8.1.5), integrating (8.2.2) over energy $E$ from zero to infinite, we obtained,

$$
\frac{d^2Y}{d^2\Omega} = Y_p \frac{|\cos \theta_1|}{\pi} \left\{1 - \frac{1}{4} \sqrt{\frac{E_{\text{th}}}{E}} \left[\cos \theta \gamma(\theta_1) + \frac{3}{2} \pi \sin \theta \sin \theta_1 \cos \phi_1\right]\right\}.
$$

(8.2.3)

Here $\gamma(\theta_1) = \frac{3 \sin^2 \theta_1 - 1}{\sin^2 \theta_1} + \cos^2 \theta_1 \left(\frac{3 \sin^2 \theta_1 + 1}{2 \sin^2 \theta_1}\right) \chi \left[\frac{1 + \sin \theta_1}{1 - \sin \theta_1}\right]$, or

$$
\gamma(\theta_1) = \frac{16}{3} \left[\frac{2 \sin^2 \theta_1}{15} + \frac{3 \sin^4 \theta_1}{7 \cdot 15} + \cdots + \frac{(k + 1) \sin^{2k} \theta_1}{(2k - 1)(2k + 11)(2k + 3) + \cdots}\right]
$$

Specially, we have $\gamma(\theta_1 = 0) = 16/3$ and $\gamma(\theta_1 = \pi/2) = 2$.

iii) Total sputtering yield [69,70]:

Further integrating (8.2.3) over $\Omega_1$ gave the total sputtering yield

$$
Y = Y_p \left(1 - \cos \theta \sqrt{\frac{E_{\text{th}}}{E}}\right)
$$

(8.2.4)

Here $Y_p = 0.042\alpha \left(M_2/M_1, \theta\right) S_n(\varepsilon) \pi U_0$ is Sigmund sputtering formula [1]. $E_{\text{th}}$ appeared in (8.2.2-4) is not only a free energy parameter but also plays the role of sputtering threshold energy [69–73]. Even if formulae (8.3.2) is called as Zhang’s expression by A. P. Yalin in literatures [74][75], but the formulae was derived by Y. Yamamura [69][70] and H. E. Roosendaal et. al.[71][72] first. For a perpendicular incidence ($\theta = 0$), (8.2.4) reduced to Matsunami’s expression, [69],

$$
Y = Y_p \left(1 - \sqrt{\frac{E_{\text{th}}}{E}}\right)
$$

(8.2.5)

Different from other works [71][72], the contribution of this work is finding the intrinsic relation
among (8.2.1-3), i.e. a parameter \( E_m \) couple (8.2.1-3) together. This intrinsic relation has been used to calculate the sputtering yield angular and energy distribution. Good agreement between theory and the most of experimental results has been reached [73].

### 8.2.2 EFFECT OF ADSORBED IMPURITY LAYER

If the target surface covered by a very thin layer with the thickness \( h \), such as adsorbed impurity layer in a poor vacuum [109], (8.2.1) reduced to,

\[
J(E_0, \Omega_0) = Y_p \left( \frac{U_0}{E_0^2} \right) \left( \frac{2|\cos \theta_0|}{\pi} \right) \left[ 1 - \frac{1}{2} \cos \theta_0 \cdot \sqrt{\frac{E_0 E_{th}}{U_0 E}} \right] \exp \left( - \frac{A h \cos \theta}{\cos \theta_0} \right)
\]  

(8.2.6)

for a perpendicular incidence. \( A \) is the adsorption coefficient and \( A^{-1} \) can considered as “mean free path” in Falcone’s ejection theory [34].

i) Angular distribution of sputtered atoms

Taking account of the refraction by the planar surface (7.1.2), (8.2.6) turns out to be the differential recoil flux of sputtered target atoms moving with energy \( E_1 \) and along the emission direction \((\theta_1, \phi_1)\). Integrating it over energy \( E_1 \) from zero to infinite, we obtained,

\[
\frac{d^2 Y}{d^2 \Omega_1} = Y_p \left( \frac{\cos \theta_1}{\pi} \right) \left[ \phi(\theta_1) - \frac{1}{4} \phi_1(\theta_1) \right] \left( \frac{E_{th}}{E} \right)
\]

(8.2.7)

Here

\[
\phi(\theta_1) = 4 \frac{\cos^2 \theta_1}{\sin^4 \theta_1} \int_0^{\theta_1} (\cos^2 \theta - \cos^2 \theta_1) \sin \theta d\theta \cdot \exp \left( - A h \cdot \frac{\cos \theta}{\cos \theta_1} \right)
\]

\[
= \frac{4}{\sin^4 \theta_1} \left[ E_3(Ah) - \cos^2 \theta_1 \cdot E_3 \left( \frac{Ah}{\cos \theta_1} \right) - E_5(Ah) + \cos^4 \theta_1 \cdot E_5 \left( \frac{Ah}{\cos \theta_1} \right) \right].
\]

\[
\phi_1(\theta_1) = 4 \frac{\cos^2 \theta_1}{\sin^2 \theta_1} \int_{\cos \theta_1}^{\theta_1} dt \cdot t^2 (1 - t^2) (t^2 - \cos^2 \theta_1)^{-0.5} \exp(-Ah/t).
\]

\[
= 8 \frac{\cos^2 \theta_1}{\sin^2 \theta_1} \int_0^{\theta_1} \frac{(\cos^2 \theta - \cos^2 \theta_1)}{\cos^2 \theta} \cdot \exp \left( - \frac{Ah \cdot \cos \theta}{\cos \theta_1} \right).
\]

ii) Total sputtering yield

Further integrating (8.2.7) over \( \Omega_1 \) gave the sputtering yield

\[
Y = Y_p \left( 1 - \sqrt{E_{th}/E} \right) \cdot \left[ 4E_5(Ah) \right]
\]

(8.2.8)
(See Appendix VIII A). If \( h = 0 \), we have \( \phi(\theta) = 4E_s(Ah) = 1 \) and \( \phi(\theta) = \gamma(\theta) \).

Then, (8.2.7) and (8.2.8) are reduced to (8.2.3) and (8.2.5) respectively.

The most remarkable results is (8.2.8), even if the target surface covered by a very thin adsorbed impurity layer, Matsunami’s expression (8.2.5), or the threshold energy \( E_{th} \) remains unchanged, as long as the value of \( \alpha \) is modified to

\[
\alpha_{eff} = \alpha \cdot [4E_s(Ah)] \leq \alpha.
\]  

(8.2.9)

On the other hand, if we use

\[
\frac{d^2 Y}{d^2 \Omega_i} \propto |\cos \theta| \left[ 1 - \frac{1}{4} \gamma(\theta) \sqrt{\frac{E_{th}}{E}} \right]
\]

to match (8.2.7), the threshold energy will reduce to \( E_{th}^{eff} \leq E_{th} \), thus, the intrinsic relation between the total sputtering yield and the sputtering yield angular distribution will become more complicated.

### 8.2.3 MODIFIED ZHANG’S EXPRESSIONS

If the natural boundary condition (8.1.5) is taken into account, (8.2.2-4) will be modified to

**i) Energy distribution of sputtered atoms** [76][77]

\[
\frac{d^3 Y}{dE_i d^2 \Omega_i} = Y_p \frac{2 |\cos \theta_i|}{\pi U_0} \frac{\varepsilon}{(\varepsilon + 1)^3} \left[ 1 - \frac{1}{2} \sqrt{\frac{E_z}{E}} \left( \cos \theta \sqrt{\varepsilon \cos^2 \theta_i + 1 + \sqrt{\varepsilon} \sin \theta \sin \theta_i \cos \phi_i} \right) \right] \]  

(8.2.10)

**ii) Angular distribution of sputtered atoms** [78]

\[
\frac{d^2 Y}{d^2 \Omega_i} = Y_p \frac{|\cos \theta_i|}{\pi} \cdot B \left[ 1 - \frac{1}{4} \sqrt{\frac{E_z}{E}} \left( \cos \theta \Gamma(\theta) + \beta \sin \theta \sin \theta_i \cos \phi_i \right) \right].
\]

(8.2.11)

**iii) Total sputtering yields:** \( Y = Y_p B \left[ 1 - \cos \theta \cdot A \sqrt{E_z/E} \right] \)  

(8.2.12)

Here \( B = (1 - D)^2 \), \( D = U_0 / E \), and

\[
A = \left( 3 - 4 \sqrt{D} + D^2 \right) / (3B),
\]

\[
\Gamma(\theta) = \int_{\theta_1}^{\theta_2} \frac{X \cos^2 \theta_i}{(X + 1)^3} \sqrt{X \cos^2 \theta_i + 1} \quad \text{and}
\]

\[
\int_0^{D-1} XdX \frac{X \cos^2 \theta_i}{(X + 1)^3} \sqrt{X \cos^2 \theta_i + 1}
\]
\[ \beta = \frac{8}{B} \int_0^{1/2} \frac{X^{3/2} dX}{(X + 1)^3}. \]

\( E_Z \) appeared in (8.2.10–12) is no longer the sputtering threshold energy. (8.2.11) is called as Modified Zhang expressions by A. P. Yalin [74][75]. For perpendicular incidence: \( \theta = 0 \), (8.2.8) reduced

\[ Y(E) = Y_p(E)B[1 - A_N \sqrt{E_Z/E}] \quad (8.2.13) \]

**8.2.4 COMPARING ZHANG AND M-ZHANG’S EXPRESSIONS WITH OTHER THEORIES**

i) Sigmund theory

If the incident energy \( E >> E_{th} \) or \( E_{Z} \), the Anisotropic term can be neglected, Zhang and MZ expressions turn out to be Sigmund’s sputtering expressions. In particular, the energy distribution of sputtered atoms reduced to the Thompson formula (7.1.3) with \( m = 0 \).

\[ \frac{d^3Y}{dE_i d^2\Omega_i} \propto \frac{E_1}{(E_i + U_0)^3} \quad (8.2.14) \]

ii) Huang’s expressions [109]Neglecting the Anisotropic term, (8.2.7,8) reduced to Huang’s expressions [109],

\[ \frac{d^2Y}{d^2\Omega_i} = Y_p \left( \frac{\cos \theta_i}{\pi} \right) \phi(\theta_i) \quad (8.2.15) \]

\[ Y = Y_p \cdot \left[ 4E \xi (Ah) \right] \quad (8.2.16) \]

which does not effect the relative sputtering yields for 22 different ions at 45 keV for Si, Cu, Ag and Au targets [81]. W. Huang used (8.2.15) to explain the over-cosine like sputtering yield angular distribution [109], unfortunately, (8.2.16) was completely ignored. If both sputtering yield angular distribution \( d^2Y/d^2\Omega_i \) and total sputtering yield incident energy dependence \( Y(E) \) had been measured under the same vacuum condition, the experimental measurement would have come to a definite conclusion and therefore would have been more meaningful in physics.
iii) Falcone’s formula [35][79][80] The sputtering yield energy distribution formulated by Falcone is of the form,

\[
\frac{dY}{dE_i d\Omega_i} = \frac{C}{(E_i + U_0)^{5/2}} \cdot \ln \left( \frac{\gamma(1 - \gamma)E_i}{E_{1AU}} \right), \quad E_{1AU} < \gamma(1 - \gamma)E_i.
\]

However, if the energy \( E \) of incident ion is very large \( (E \gg E_i) \), Falcone’s formula will not approach to the Thompson distribution (7.1.14).

**8.2.5 COMPARING ZHANG AND M-ZHANG’S EXPRESSIONS WITH EXPERIMENTS**

i) Incident ion energy dependence of total sputtering yields [81–83]

Both Matsunami’s expression (8.2.5) and Eq. (8.2.13) were used to compute the ion incident energy dependence of total sputtering yield [81–83] for 15 ion-target systems. These ion-target systems were chosen for the further research on the differential sputtering yield. The values of \( \alpha \) and \( U_0 \) were cited from the literature [7] and collected in the Table 8.2.1. The sputtering yield calculation needs the nuclear stopping cross section, formula (6.1.26) was used for three ion-target systems: \( \text{Ar}^+ \rightarrow \text{Cu} \), \( \text{Ar}^+ \rightarrow \text{W} \) and \( \text{Ar}^+ \rightarrow \text{Al} \); for other ion-target systems, formula (6.1.23) was used with replacing \( Z = \left( Z_i^{2/3} + Z_k^{2/3} \right)^{1/2} \) by \( Z = Z_i^{0.23} + Z_k^{0.23} \) in (6.1.22). For \( \text{Ar}^+ \rightarrow \text{Ti} \), \( \text{Hg}^+ \rightarrow \text{Fe} \), \( \text{Hg}^+ \rightarrow \text{U} \) and \( \text{Hg}^+ \rightarrow \text{Ge} \), we have to consider the effect of adsorbed impurity layer on the target surface to match the corresponding experiment results [82], the specific values of \( (\text{Ah}) \) for each system are collected in the Table 8.2.1. Specifically for \( \text{Hg}^+ \rightarrow \text{Ge} \), if neglected Yamamura’s new experiment results [82], the values of \( (\text{Ah}) \) will be vanish, but, \( E_{th} = 130\text{eV} \) and \( E_T = 190\text{eV} \) are too high to be used for the calculation of sputtering yield angular distributions. On the other hand, if considered Yamamura’s new experiment results [82], \( \text{Ah} = 0.23 \) can not be vanish, but, \( E_{th} = 90\text{eV} \) and \( E_T = 140\text{eV} \) are excellent for the calculation of sputtering yield angular distributions.
Table 8.2.1 Parameters for computing total sputtering yields

<table>
<thead>
<tr>
<th>Ion</th>
<th>Target</th>
<th>(S_{0}(\varepsilon))</th>
<th>(U_{0}) (eV)</th>
<th>(\alpha)</th>
<th>(\alpha_{\text{eff}})</th>
<th>(E_{\text{th}}) (eV)</th>
<th>(E_{Z}) (eV)</th>
<th>Fig. (8.2.1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>Mo</td>
<td>(6.1.23)</td>
<td>6.82</td>
<td>0.21</td>
<td>0.21</td>
<td>0</td>
<td>114</td>
<td>188</td>
</tr>
<tr>
<td>Hg</td>
<td>Ni</td>
<td>(6.1.23)</td>
<td>4.45</td>
<td>0.19</td>
<td>0.19</td>
<td>0</td>
<td>86</td>
<td>138</td>
</tr>
<tr>
<td>Hg</td>
<td>Pt</td>
<td>(6.1.23)</td>
<td>5.77</td>
<td>0.27</td>
<td>0.27</td>
<td>0</td>
<td>75</td>
<td>130</td>
</tr>
<tr>
<td>Hg</td>
<td>Ge</td>
<td>(6.1.23)</td>
<td>3.85</td>
<td>0.20</td>
<td>0.20</td>
<td>0</td>
<td>130</td>
<td>190</td>
</tr>
<tr>
<td>Hg</td>
<td>Cu</td>
<td>(6.1.23)</td>
<td>3.51</td>
<td>0.19</td>
<td>0.19</td>
<td>0</td>
<td>60</td>
<td>105</td>
</tr>
<tr>
<td>Hg</td>
<td>W</td>
<td>(6.1.23)</td>
<td>8.82</td>
<td>0.26</td>
<td>0.26</td>
<td>0</td>
<td>100</td>
<td>180</td>
</tr>
<tr>
<td>Xe</td>
<td>Mo</td>
<td>(6.1.23)</td>
<td>6.82</td>
<td>0.23</td>
<td>0.23</td>
<td>0</td>
<td>96</td>
<td>162</td>
</tr>
<tr>
<td>Xe</td>
<td>Al</td>
<td>(6.1.23)</td>
<td>3.38</td>
<td>0.18</td>
<td>0.18</td>
<td>0</td>
<td>80</td>
<td>135</td>
</tr>
<tr>
<td>Ar</td>
<td>Cu</td>
<td>(6.1.26)</td>
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<td>82</td>
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<td>Ar</td>
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<td>0.51</td>
<td>0</td>
<td>80</td>
<td>149</td>
</tr>
<tr>
<td>Ar</td>
<td>Al</td>
<td>(6.1.26)</td>
<td>3.38</td>
<td>0.23</td>
<td>0.23</td>
<td>0</td>
<td>80</td>
<td>124</td>
</tr>
<tr>
<td>Ar(^{+})</td>
<td>Ti</td>
<td>(6.1.23)</td>
<td>4.90</td>
<td>0.30</td>
<td>0.21</td>
<td>0.23</td>
<td>60</td>
<td>95</td>
</tr>
<tr>
<td>Hg</td>
<td>Fe</td>
<td>(6.1.23)</td>
<td>4.31</td>
<td>0.194</td>
<td>0.146</td>
<td>0.18</td>
<td>100</td>
<td>160</td>
</tr>
<tr>
<td>Hg</td>
<td>U</td>
<td>(6.1.23)</td>
<td>5.55</td>
<td>0.306</td>
<td>0.214</td>
<td>0.23</td>
<td>70</td>
<td>120</td>
</tr>
<tr>
<td>Hg</td>
<td>Ge</td>
<td>(6.1.23)</td>
<td>3.85</td>
<td>0.20</td>
<td>0.14</td>
<td>0.23</td>
<td>90</td>
<td>140</td>
</tr>
</tbody>
</table>

![Graph a](image1.png)  ![Graph b](image2.png)
Fig. 8.2.1 Ion-energy dependence of total sputtering yields from single element target by normal incident heavy ion bombardment. The calculated results of Sigmund formula (7.1.27) were given for comparison. The solid lines were calculated by formulae (8.2.5) and (8.2.12), these calculated curves show good agreement with the experimental measurements [82]. (a), (b), (c), (d), (e) and (f) demonstrated Mo, Ni, Pt, Ge, Cu and W targets bombarded by mercury ion respectively; (g) and (h) demonstrated Mo and Al targets bombarded by Xenon ion respectively; (i), (j), (k) and (l) demonstrated Cu, W, Al and Ti targets bombarded by argon ion respectively; (m), (n) and (o) demonstrated Fe, U and Ge targets bombarded by mercury ion respectively. $Ah > 0$.

Recently, many authors are interesting in the sputtering data for molybdenum target bombarded by low energy xenon ions for their own purpose [114][115][117][118][121]. We found that formulae (8.2.5) and (8.2.13) fit the experiment data published by Yalin et. al.[117] and Weijsenfeld et. al.[113] very well, but the experiment data published by Wehner et.al.[82] seams little too higher. On the contrary, instead of (6.1.23), if the nuclear stopping cross-section (6.1.26) is used for the same calculation, both formulae (8.2.5) and (8.2.13) will fit Wehne’s experiment data [82] very well as long as $E_{th} = 80eV$ and $E_{Z} = 140eV$. In this case, the experiment data published by Yalin et. al.[117] and Weijsenfeld et. al.[113] seams little too low. This example demonstrated that the nuclear stopping cross-section must play a crucial role in the sputtering calculation. In other word, both theoretical and experimental research on the low energy sputtering could provide important information of the nuclear stopping cross-section in low energy region [55].

The best fitting values of $E_{th}$ and $E_{Z}$ were determined by matching experimental results of
ion energy dependence of total sputtering yield to Eq. (8.2.5) and Eq. (8.2.13) respectively. The parameters for the computing are collected in Table 8.2.1. The calculated curves were plotted in Fig. 8.2.1. The calculated curves of both (8.2.5) and (8.2.13) agree with the corresponding experiment Data well and our formula (8.2.13) fits experiment Data better than Matsunami’s expression (8.2.5).

ii) Sputtering yield angular distributions

Substituting the parameters $E_{th}$ and $E_Z$ in Table 8.2.1 into Zhang and M-Zhang expressions (8.2.3) and (8.2.11) respectively, we obtained the corresponding angular distributions of sputtered atoms[73][78]. The related parameters were collected in Table 8.2.2 and the calculated curves were plotted in Fig.8.2.2 (a)-(q) respectively. It’s surprise that both calculated curves of Eq. (8.2.3) and (8.2.11) agree each other very well for all of the ion-target systems. For the angular distributions of sputtered material from Mo, Hi, Pt, Fe and Ge targets by normally and obliquely incident $E = 250, 300 \text{eV}$ mercury ion bombardments, the calculated curves show the excellent agreement with the correspondent Wehner’s experimental results $\left(\theta_i < 50^0\right)$ given in Ref.[84], see Fig. 8.2.2 (a)-(f) and Fig. 8.2.2 (g) and (h). For Pt and Fe targets bombarded by normally incident $E = 250, 500, 750, 1000 \text{eV}$ mercury ion, the calculated curves show a good agreement with the correspondent Wehner’s experimental results $\left(\theta_i < 50^0\right)$ given in Ref.[84], see Fig. 8.2.2 (l) and (m). The same calculation for Ge target, only reasonably good, see Fig. 8.2.2 (q). For Mo target, if $E_{th} = 114 \text{eV}$ and $E_Z = 188 \text{eV}$ are used, the calculated curves fit Wehner’s experimental results $\left(\theta_i < 50^0\right)$ very well only for $E = 250 \text{eV}$ as mentioned previously, but for $E = 500, 750, 1000 \text{eV}$, we have to use $E_{th} = 210 \text{eV}$ and $E_Z = 300 \text{eV}$, see Fig. 8.2.2 (o). A similar phenomenon can be seen for Xe$^+ \rightarrow$ Mo system. Substituting $E_{th} = 60 \text{eV}$ and $E_Z = 105 \text{eV}$, Zhang and M-Zhang expressions (8.2.3) and (8.2.11) can describe the sputtering yield angular distribution for incident energy $E = 200 \text{eV}$, see Fig. 8.2.2 (i). For higher incident
energies $E = 350, 500\text{eV}$, we have to chose much high $E_h$ and $E_Z$, see Table 8.2.2 and Fig. 8.2.2 (j), (k). Unfortunately, Zhang and M-Zhang expressions (8.2.3) and (8.2.11) couldn't give any reasonable descriptions for the angular distributions of sputtered material from Ni target by normally incident $E = 500, 750, 1000\text{ eV}$ Hg$^+$ ion.

<table>
<thead>
<tr>
<th>Ion</th>
<th>Target</th>
<th>Incident Energy (eV)</th>
<th>Incident Angle (Deg)</th>
<th>Experimental Data Source</th>
<th>Fig. (8.2.1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>Mo</td>
<td>250</td>
<td>0</td>
<td>Wehner [84]</td>
<td>(a)</td>
</tr>
<tr>
<td>Hg</td>
<td>Ni</td>
<td>250</td>
<td>0</td>
<td>Wehner [84]</td>
<td>(b)</td>
</tr>
<tr>
<td>Hg</td>
<td>Ni</td>
<td>300</td>
<td>0</td>
<td>Wehner [84]</td>
<td>(c)</td>
</tr>
<tr>
<td>Hg</td>
<td>Pt</td>
<td>250</td>
<td>0</td>
<td>Wehner [84]</td>
<td>(d)</td>
</tr>
<tr>
<td>Hg</td>
<td>Fe</td>
<td>250</td>
<td>0</td>
<td>Wehner [84]</td>
<td>(e)</td>
</tr>
<tr>
<td>Hg</td>
<td>Ge</td>
<td>250</td>
<td>0</td>
<td>Wehner [84]</td>
<td>(f)</td>
</tr>
<tr>
<td>Xe</td>
<td>Mo</td>
<td>200</td>
<td>0</td>
<td>Shutthanandan [114][121]</td>
<td>(g)</td>
</tr>
<tr>
<td>Xe</td>
<td>Mo</td>
<td>350</td>
<td>0</td>
<td>Yalin [118]</td>
<td>(h)</td>
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<tr>
<td>Xe</td>
<td>Mo</td>
<td>500</td>
<td>0</td>
<td>Shutthanandan [114][121]</td>
<td>(i)</td>
</tr>
<tr>
<td>Hg</td>
<td>Mo</td>
<td>250</td>
<td>25</td>
<td>Wehner [84]</td>
<td>(j)</td>
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<td>250</td>
<td>21</td>
<td>Wehner [84]</td>
<td>(k)</td>
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<tr>
<td>Hg</td>
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<td>Wehner [84]</td>
<td>(l)</td>
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<td>250-1000</td>
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<td>Wehner [84]</td>
<td>(m)</td>
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<td>Mo</td>
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<td>(p)</td>
</tr>
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<td>Hg</td>
<td>Ge</td>
<td>250-1000</td>
<td>0</td>
<td>Wehner [84]</td>
<td>(q)</td>
</tr>
</tbody>
</table>
Fig. 8.2.2 Polar diagram of material sputtered from mono-atomic target. Both Zhang and M-Zhang expressions (8.2.3,11) were used for the calculations. The calculated curves show the excellent agreement with the correspondent Wehner’s experimental results given in Ref.[84]. Mo, Ni, Pt, Fe and Ge targets bombarded by normally incident 250eV mercury ion in (a), (b), (d), (e) and (f). Ni target bombarded by normally incident 300eV mercury ion in (c). Mo and Ni targets bombarded by obliquely incident 250eV mercury ion in (g) and (h). The ion incident angles are 21 degree and degree respectively in (g) and (h). Mo target bombarded by normally incident 200eV, 350eV and 500eV Xenon ion respectively in (i), (j) and (k). Pt, Fe, Mo, Ni, and Ge targets bombarded by normally incident 250eV, 500eV, 750eV and 1000eV mercury ion respectively in (l), (m), (n), (o), (p) and (q)
iii) Sputtering yield energy distributions [67][76][77]

Seeing the most of the experimental results cited here published by Goehlich et. al [79][80],

it’s necessary to make clear that the relation between the orientation angles \( \theta, \phi, \theta_1, \) and \( \phi_1 \) in this work and the ones \( \Psi, \Phi \) in [79][80] is

\[
\theta = \Psi, \quad \theta_1 = |\Phi| \quad \text{and} \quad \cos \phi_1 = -1, \quad \text{if} \quad \Phi \geq 0
\]

or \( \cos \phi_1 = +1, \quad \text{if} \quad \Phi < 0. \)

For the normal incidence and perpendicular emission \( \Psi = \Phi = 0 \), Eq.(8.2.10) gave the

normalized energy distribution of sputtered atoms is

\[
\bar{S}(E; \theta, \theta_1, \phi_1, \phi) = \frac{C}{(E_1 + U_0)^3} \left[ 1 - \frac{1}{2} \sqrt{-\frac{E}{E_z}} \sqrt{E + U_1 + U_0} \right] \tag{8.2.18}
\]

If \( E_T \) here is replaced by \( E_{th} \), Eq. (8.2.18) will become the old formula (8.2.1). As we mentioned

previously, substituting \( E_T \) and \( E_{th} \) into (8.2.3) & (8.2.11) and (8.2.5)&(8.2.12) respectively, both

theories predict almost the same results for the total sputtering yield and the yield angular
distribution. But, the situation could be different in sputtering yield energy distribution. For copper

target bombarded by normal incident argon ion, substituting \( U_0 = 3.51 \text{eV}, \ E_z = 82 \text{eV} \) (new

theory) or 50eV (old theory) and \( E = 200 \text{eV} \) or \( E = 400 \text{eV} \) into Eqs.(8.2.18), we obtained

the sputtering yield energy distribution and plotted in Fig.8.2.3(A). The calculated curve shows

good agreement with the corresponding experimental results [67][116]. No cut off has been found

Falcone in Ref. [68] in sputtering yield energy distribution. There is no distinct difference between

new theory and old theory has been seen, but the new theory fits experimental results slightly better

[67][76][77].Therefore, only new theory Eq.(8.2.10) will be used for the further calculations.

For tungsten and aluminum targets bombarded by normal incident argon ion, substituting \( U_0 = 8.82 \text{eV}, \ E_z = 149 \text{eV}, \ E = 225, 300, 500, 5000 \text{eV} \) respectively for tungsten

target and \( U_0 = 3.38 \text{eV}, \ E_z = 135 \text{eV}, \ E = 200, 500 \text{eV} \) respectively for aluminum target, into

Eqs.(8.2.18), we obtained the sputtering yield energy distribution and plotted in Fig.8.2.3 (B) and
in Fig.8.2.3 (C) respectively. The calculated curves show good agreement with the corresponding experimental results [79][80].

The sputtering yield energy distribution formulated by Falcone (8.2.17) was shown to yield a good approximation to Goehlich’s experimental results for tungsten target bombarded by normally incident argon ion (in Figures.6 (b~d)) [79][80]. However, Falcone theory (8.2.17) can not predict correct experimental results (in Figures.6 (a)) [79][80]. It is not surprised, if the energy E of incident ion is very large (E₁ << E), Eq. (8.2.17) will approach to

\[ S(E,0;E₁ << E,0,φ₁) = \frac{CE₁}{(E₁ + U₀)^{1/2}} \]

which is not Thompson distribution (7.1.3). In particular, for copper target bombarded by normally incident argon ion, Eq. (8.2.17) couldn’t predict correct experimental results in (8.2.3)(A). Besides, the relation between Falcone theory (8.2.17) and Sigmund formula (7.1.27) is unclear.

For the normal incidence, i.e. \( Ψ = 0 \), Eq.(8.2.10) gave the normalized energy distribution of sputtered atoms is

\[ S(E,0;E₁,0,φ₁) = \frac{CE₁}{(E₁ + U₀)^{1/2}} \left[ 1 - \frac{1}{2} \sqrt{\frac{E_Z}{EU₀}} \sqrt{E₁ \cos² θ₁ + U₀} \right] \] (8.2.19)\]

For Titanium target bombarded by normal incident \( \text{Ar}^+ \), substituting two groups of parameters:

\[ U₀ = 4.90\text{eV}, \ E_Z = 160\text{eV}, \ E = 300\text{eV} \text{ and } \Phi = 0⁰, 40⁰, 60⁰, 80⁰ ; \]

\[ U₀ = 4.90\text{eV}, \ E_Z = 160\text{eV}, \ E = 225\text{eV}, 300\text{eV}, 5000\text{eV} \text{ and } \Phi = 0⁰, 60⁰ \]

into Eqs. (8.2.19), the theoretical energy distribution are plotted in Fig.8.2.3 (D) and (E) respectively. The calculated curves show good agreement with the corresponding experimental results [79][80]. The problem is that the parameter \( E_Z = 95\text{eV} \) in the Table 8.2.1 seems too low.

For the oblique incidence, i.e. \( Ψ \neq 0 \), the normalized energy distribution of sputtered atoms
\[ S(E, \theta; E_i, \theta_i, \phi_i) = \frac{C E_i}{(E_i + U_0)^3} \left( 1 - \frac{1}{2} \sqrt{\frac{E_Z}{E U_0}} \left[ \cos \theta \sqrt{E_i \cos^2 \theta_i + U_s} + \sqrt{E_i \sin \theta \sin \theta_i \cos \phi_i} \right] \right) \]  

(8.2.20)

For oblique incidence of argon ion, \( U_0 \) and \( E_z \) in Eqs.(8.2.20) have been given by Table 8.2.1: \( U_0 = 8.82 \text{eV} \) and \( E_z = 149 \text{eV} \) for W target; \( U_0 = 3.38 \text{eV} \) and \( E_z = 124 \text{eV} \) for Al target, \( U_0 = 4.90 \text{eV} \), \( E_z = 160 \text{eV} \), \( E_z = 95 \text{eV} \) in table 8.2.1 for Titanium target. The other parameters in Eqs.(8.2.20) are

\[ \Psi = 50^\circ, \Phi = -50^\circ, -10^\circ, 10^\circ, 30^\circ, 50^\circ, 70^\circ \text{ and } E = 200 \text{eV} \text{ for W target}, \]
\[ \Psi = 50^\circ, \Phi = -30^\circ, -10^\circ, 10^\circ, 30^\circ, 50^\circ, 70^\circ \text{ and } E = 200 \text{eV} \text{ for Al target}, \]
\[ \Psi = 50^\circ, \Phi = -30^\circ, 10^\circ, 30^\circ, 70^\circ \text{ and } E = 300 \text{eV} \text{ for W target}, \]
\[ \Psi = 70^\circ, \Phi = -50^\circ, 10^\circ, 30^\circ, 50^\circ, 70^\circ \text{ and } E = 500 \text{eV} \text{ for W target}, \]
\[ \Psi = 50^\circ, \Phi = -50^\circ, -30^\circ, -10^\circ, 10^\circ, 30^\circ, 50^\circ \text{ and } E = 500 \text{eV} \text{ for Ti target}. \]

The calculated energy distributions of sputtered atoms are plotted in Fig. 8.2.3(F), Fig. 8.2.3(G), Fig. 8.2.3(H), Fig. 8.2.3(I) and Fig. 8.2.3(J) respectively. For the oblique incidence of Xenon ion, \( U_0 \) and \( E_z \) in Eqs.(8.2.20) have been given by Table 8.2.1: \( U_0 = 3.38 \text{eV} \) and \( E_z = 135 \text{eV} \) for Al target. The other parameters in Eqs.(8.2.20) are \( \Psi = 50^\circ, \Phi = -70^\circ, -30^\circ, -10^\circ, 30^\circ, 50^\circ, 70^\circ \text{ and } E = 250 \text{eV} \text{. The calculated energy distribution of sputtered atoms are plotted in Fig. 8.2.3(K). The calculated curves show a good agreement with the corresponding experimental results [79][80]: (e) in Fig. 8.2.3 (F); (d) and (e) in Fig.8.2.3(G); (c) and (d) in Fig. 8.2.3(H); (c) and (d) in Fig. 8.2.3(I); (b), (c), (d), (e) and (f) in Fig. 8.2.3(J); \( \Phi = -70^\circ, -30^\circ \text{ in Fig. 8.2.3(K). The calculated curves show a reasonable agreement with experimental results [79][80]: (d) in Fig. 8.2.3(F); (a), (c) and (f) in Fig. 8.2.3(G); (b) in Fig. 8.2.3(H); (e) in Fig. 8.2.3(I); (a) in Fig. 8.2.3(J); \( \Phi = -10^\circ \text{ in Fig. 8.2.3(K). However, a great discrepancy exists between the} \)
predictions of new theory and experimental results [79][80]: (a), (b) and (c) in Fig. 8.2.3(F); (b) in Fig. 8.2.3(G); (a) in Fig. 8.2.3(H); (a) and (b) in Fig. 8.2.3(I); \( \Phi = 30^0, 50^0, 70^0 \) in Fig. 8.2.3(K). The great discrepancy probably may be due to the influence of direct recoil atoms on the energy spectrum.

Finally, we feel it necessary to make mention of Prof. A. Goehlich’ contributions in sputtering research [79][80]. In order to understand the anisotropic sputtering behavior, Goehlich et. al. used a similar form of (8.2.19),

\[
\overline{S}(E, \theta; E_i, \theta_i, \phi_i) \propto \frac{E_i}{(E_i + U_\theta)^3} \left[ 1 + \lambda \cdot \sqrt{E_i \cos^2 \theta_i + U_\theta} \right]
\] (8.2.21)

to describe the angular variation of the energy distributions. \( \lambda \) in (8.2.21) is treated by them an empirical fitting parameter, considered as meaningful only parameter. But, for low energy bombardment, they realized that the fitting parameter \( \lambda \) turns out to become negative to approximate their experimental distributions. The negative value of \( \lambda \) indicates a negative correction term to the isotropic flux. But, they still deny any relation between the correction term and the deposited momentum density.

8.2.6 CONCLUSION

At least for the flux of sputtered atoms induced by low energy and heavy particle bombardment, the momentum term in (8.1.1) could cause a negative anisotropy correction to Sigmund’s isotropic differential sputtering yield by broadening the angular distribution and narrowing the energy spectrum of sputtered particle increases in importance with decreasing energy \( E \) and increasing surface binding energy.
Table 8.2.3 Parameters for computing sputtering yield energy distributions

<table>
<thead>
<tr>
<th>Ion</th>
<th>Target</th>
<th>Ion incident Energy (eV)</th>
<th>Ion incident Angle (Deg)</th>
<th>Sputtered atoms Out Angle (Deg)</th>
<th>Experimental Data Source</th>
<th>Fig. 8.2.3</th>
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<tr>
<td>Ar</td>
<td>Cu</td>
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<td>0</td>
<td>0</td>
<td>[67][116]</td>
<td>(A)</td>
</tr>
<tr>
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<td>225,300, 500,5000</td>
<td>0</td>
<td>0</td>
<td>Goehlich [80]</td>
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<td>200, 500</td>
<td>0</td>
<td>0</td>
<td>Goehlich [80]</td>
<td>(C)</td>
</tr>
<tr>
<td>Ar</td>
<td>Ti</td>
<td>300</td>
<td>0</td>
<td>0, 40, 60, 80</td>
<td>Goehlich [80]</td>
<td>(D)</td>
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<tr>
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<td>225,300, 5000</td>
<td>0</td>
<td>0, 60</td>
<td>Goehlich [80]</td>
<td>(E)</td>
</tr>
<tr>
<td>Ar</td>
<td>Al</td>
<td>200</td>
<td>50</td>
<td>-10, 10, 30, 50,70</td>
<td>Goehlich [80]</td>
<td>(F)</td>
</tr>
<tr>
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<td>W</td>
<td>200</td>
<td>50</td>
<td>-50, -10, 10, 30, 50,70</td>
<td>Goehlich [80]</td>
<td>(G)</td>
</tr>
<tr>
<td>Ar</td>
<td>W</td>
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<td>50</td>
<td>-30, 10, 30, 70</td>
<td>Goehlich [80]</td>
<td>(H)</td>
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<tr>
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<td>W</td>
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<td>70</td>
<td>-50, 10, 30, 50,70</td>
<td>Goehlich [80]</td>
<td>(I)</td>
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<td>50</td>
<td>-50, -30, -10, 10, 30,50,</td>
<td>Goehlich [80]</td>
<td>(J)</td>
</tr>
<tr>
<td>Xe</td>
<td>Al</td>
<td>250</td>
<td>50</td>
<td>-70, -30, -10, 30, 50, 70</td>
<td>Goehlich [80]</td>
<td>(K)</td>
</tr>
</tbody>
</table>
Fig. 8.2.3 Normalized energy distribution of sputtered neutral atoms. Small squares and triangles stand for experimental measurement results [67][116]. The solid thin lines are theoretical curves calculated by using parameters listed in Table (8.2.1). New: calculated by (8.2.10). Old: calculated by (8.2.2) [67]. Fal: calculated by Falcone's formula (8.2.17) [79]. Cu, W, Al and Ti targets bombarded by normally incident Argon ion in (A), (B), (C), (D) and (E). W, Al and Ti targets bombarded by obliquely incident Argon ion in (F), (G), (H), (I) and (J). Al target bombarded by obliquely incident Xenon ion in (K).
8.3 ANISOTROPIC SPUTTERING THEORY AND EXPERIMENTS OF
MULTI-COMPONENT TARGET

8.3.1 ISOTOPIC ATOMIC MASS EFFECT

Isotopic mixtures sputtering preferentially under ion bombardment has attracted considerable
interest for many years [61]. First, isotopic medium provides the simplest model systems for the
study of the sputtering of multi-component materials. In this case, only kinematic effects due to mass
differences between the isotopes contribute to their preferential emission during sputtering. Second,
isotopic medium provides more dimensions to look into the elemental target sputtering. Actually,
most of natural elemental materials consist of several isotopes. Thus, this system can offer a chance
to reveal the possible errors concealed in the elemental target sputtering theory [1]. The last is the
most important one. Up to now, Sigmund conventional (isotropy) sputtering theory [1][7] seems
couldn’t describe the unusual isotopic medium sputtering effect.

In the primary sputter process, considering a perpendicular incidence $\theta = 0^\circ$, and using
$m = 0$, $U_{0j} = U_0$ and $M_j \approx M = \sum_k C_k M_k$, taking account of the refraction by the planar
surface binding energy, (8.1.2) turned out to be the sputtered atom flux. Then, taking the boundary
conditions $J_j(E_0 < \gamma E, \Omega_0) = 0$ into account, integrating over energy and solid angle, we obtained
the partial sputtering yield of component $j$,

$$Y_j = \alpha_j Y_b B[1 - \alpha H_j(E)], \quad (8.3.1)$$

Here $H_j(E) \equiv \sqrt{\left(M_j/M\right)(E_z/E)} \quad (8.3.2)$

In sputter process at steady-state, as a consequence of preferential sputtering, the surface
concentration of a virgin target is changed during ion bombardment until a surface concentration
$\alpha_k (0 \leq \alpha_k \leq 1; \sum \alpha_k = 1)$ reached the steady-state, from which the atoms are sputtered with
partial yields proportional to the bulk concentration. Solving (7.2.1) with (8.3.1) yielded,
\[ \alpha_j = C_j \cdot \sum_k \frac{B_j[1 - AH_k(E)]}{B_j[1 - AH_j(E)]} \]  
(8.3.3)

which can be solved by an iteration method with initial condition: \( \alpha_k = C_k \). Thus, we obtained the angular distribution of the relative abundance of component \( j \),

\[ [\text{Abundance}]_j(\theta_i) = \frac{\alpha_j B_j[4 - \Gamma(\theta_i)H_j(E)]}{\sum_k \alpha_k B_k[4 - \Gamma(\theta_i)H_k(E)]} \]  
(8.3.4)

\[ \left[ \frac{\text{Abundance normal}}{\text{abundance oblique}} \right]_j = \frac{[\text{Abundance}]_j(\theta_i = 0^\circ)}{[\text{Abundance}]_j(\theta_i = 90^\circ)} \]  
(8.3.5)

(8.3.5) has been measured experimentally by G. K. Wehner et. al [85][86] for Cu, Mo, W and U targets bombarded by mercury ion with incident energy \( E = 60, 100, 200, 300 \) eV respectively. The experimental results have been collected in Table 8.3.1. In present theory, substituting \( M_k, C_k \), the parameters describing the target given Table 8.3.1. By using the iteration method, we obtained the surface concentrations \( \alpha_k \) at the steady state. Then, inserting \( \alpha_k \) into (8.3.4,5) yields the theoretical results are also collected in Table 8.3.1 for comparing with experimental data. Again, one can see that the present theory predicts experimental data as long as a suitable energy parameter \( E \) is chosen for each ion-target combination. In addition, the present theory naturally predicts "under-cosine" angular distribution of material ejected from target bombarded at low ion energies [73][78]. (8.1.1) and (8.1.2) predict a negative asymmetric term for a normal incidence. This is the intrinsic reason why isotopic mixtures are characterized by higher erosion rates for lower isotopic masses and vice versa [61] for a low energy ion bombardment. In particular, the asymmetric term could be comparable with the isotropic term in magnitude for the near threshold energy ion incidence. The former may cancel the most part of the latter. Thus, this effect could be magnified and become even more pronounced. Therefore, the present theory successfully solved the isotope puzzle in low energy sputtering. However, non of parameters \( E \) chosen here is equivalent to the corresponding one given in the Table 8.3.1. In the case, we may have to consider the contribution of
pronounced composition gradients [136].

More recently, a few papers [122-124] (SZR works) published by a research group from the Tuskegee University describe the mass spectrometer measurements of the secondary ions or neutral atoms ejected from the surface under the energetic xenon ion bombardment. One of the interesting results is that at low energies (below 350 eV for BN), heavy isotope of boron ($^{11}\text{B}$) is preferentially sputtered, while at higher energies light isotope ($^{10}\text{B}$) is preferentially sputtered. Similar results were observed for copper and silver. The conclusion in SZR works seems contradict to our theory. The key point to solve the problem is the geometry of the experimental setup in SZR works. The ion incident angle is 50 degree and the emission angle of sputtered atoms is 40 degree. The emission line of sputtered atoms is perpendicular to the ion incident line exactly in SZR works. Therefore, it’s not very strange to see that heavy isotope of boron ($^{11}\text{B}$) is preferentially sputtered, the detail calculated curves are expressed Part (b) of Fig.8.3.1. We are interesting in the preferentially sputtered isotope in the case the ion incidence line is almost parallel to emission line of sputtered atoms. Fortunately, by using QCM system, Yalin et. al. have measured differential sputter yields of HBC grade BN target bombarded by $\text{Xe}^+$ ion with different energies and incident angles [88]. Considering that the QCM only measures the angular distribution of sputtered boron atoms, following the same procedure (8.3.1-5) and using the parameters ($E_{\text{th}} \equiv E^*$) published by Yalin et. al. [88], we have calculated the fractionation of relative surface concentration and the angular distribution of the relative concentration relative of isotopes $^{10}\text{B}$ and $^{11}\text{B}$ sputtered from BN target bombarded by $\text{Xe}^+$ ion, the calculated results collected in Table 8.3.2. The angular distributions of sputtering yield

$$Y(\theta) = \frac{2 \cos \theta_i}{(1 - \cos \theta \cdot \sqrt{E_{\text{th}}/E})} \left[ 1 - \frac{1}{4} \frac{E_{\text{th}}}{E} \left[ \cos \theta_i (\theta_1) + \frac{3}{2} \pi \sin \theta \sin \theta_i \cos \phi_i \right] \right]$$ (8.3.6)

and the relative concentration relative of isotopes

$$\delta(\theta) = \frac{Y_1(\theta)C_2}{[Y_2(\theta)C_1]} - 1,$$ (8.3.7)
have been plotted in Part (a) and Part (b) of Fig. 8.3.1 respectively, here, $j = 1$ stands for $^{10}\text{B}$ and $j = 2$ stands for $^{11}\text{B}$. $Y^{-1}Y(\theta)$ determined by (8.3.6) for sputtered Boron have been measured by Yalin et al. by using QCM [88], therefore measuring $\delta(\theta)$ determined by (8.3.7) turns out to be very urgent. We hope we could measure $\delta(\theta)$ by using NRA or NAA in near future.

### Table 8.3.1 Theoretical and experimental results of sputtered isotopes.

<table>
<thead>
<tr>
<th>Target</th>
<th>E (eV) for Hg$^+$</th>
<th>Isotope abundance On target surface</th>
<th>Abundance $\frac{\text{normal}}{\text{oblique}}$</th>
<th>$E_T$</th>
<th>$E_{th}$</th>
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<tbody>
<tr>
<td></td>
<td></td>
<td>Isotopes</td>
<td>Natural (%)</td>
<td>Theory (%)</td>
<td>Experiment</td>
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<tr>
<td>Cu</td>
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<td>63</td>
<td>69.1</td>
<td>68.77</td>
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<td></td>
<td>65</td>
<td>30.9</td>
<td>31.23</td>
<td>0.976 ± 0.009</td>
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<tr>
<td></td>
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<td>1.008 ± 0.003</td>
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<td>31.05</td>
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<td>0.982 ± 0.007</td>
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<td>99.3</td>
<td>99.302</td>
<td>0.9999 ± 0.0001</td>
<td>0.9998</td>
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Table 8.3.2 Theoretical results of relative surface concentration and its angular distribution of isotopes $^{10}$B and $^{11}$B sputtered from BN target bombarded by Xe$^+$ ion.

<table>
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<tr>
<th>Ion Energy (eV)</th>
<th>Incident Angle (Deg)</th>
<th>$Y_{\text{Cond.}}$ (mm$^3$/C)</th>
<th>$Y_{\text{Marg.\ Max.}}$ (mm$^3$/C)</th>
<th>$E_{\text{th}}$ (eV)</th>
<th>Theoretical results of $\delta$ on the target surface</th>
<th>Figs. (8.3.1)</th>
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<td>0.974</td>
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</table>
8.3.2 SURFACE BINDING EFFECT

For convenience, let’s consider a binary target \((j = 1, 2)\) and \(M_1 \approx M_2\), thus, the nonstoichiometric factors \(G_j / \langle G \rangle_j = \alpha_j\). Neglecting condition (8.1.5), formulae (8.1.7) and

\[
Y_j = \alpha_j \cdot \frac{Y_p U_0}{U_{0j}} \left[ 1 - \sqrt{\frac{E_{\text{thj}}}{E}} \right] \tag{8.3.8}
\]

\[
Y_j(\theta_i) = 2\pi \frac{d^2 Y_j}{d \Omega_1} = \alpha_j \cdot \frac{Y_p U_0}{8 U_{0j}} \cos \theta_i \left[ 4 - \gamma(\theta_i) \right] \sqrt{\frac{E_{\text{thj}}}{E}} \tag{8.3.9}
\]

Here \(Y_p = 0.042\alpha S_n(E)/U_0\) and \(E_{\text{thj}} = E[H_j(E)]^2\). Using (8.1.4) gave

\[
\frac{E_{\text{th2}}}{E_{\text{th1}}} = \frac{M_2 U_{02}}{M_1 U_{01}} \tag{8.3.10}
\]

At the steady-state, the condition (8.1.8) reduced to

\[
\frac{C_1}{C_2} = \frac{Y_1}{Y_2} = \frac{\alpha_1 U_{02}}{\alpha_2 U_{01}} \left[ 1 - \sqrt{\frac{E_{\text{th1}}}{E}} \right] \left[ 1 - \sqrt{\frac{E_{\text{th2}}}{E}} \right] \tag{8.3.11}
\]

\[
\frac{\alpha_1 U_{02}}{\alpha_2 U_{01}} = \frac{C_1}{C_2} \left[ 1 - \sqrt{\frac{E_{\text{th2}}}{E}} \right] \left[ 1 - \sqrt{\frac{E_{\text{th1}}}{E}} \right] = A \quad \text{and} \tag{8.3.12}
\]

\[
\alpha_1 / \alpha_2 = A \cdot \left( \frac{U_{01}}{U_{02}} \right) \tag{8.3.13}
\]
By using (8.3.8), the total sputtering yield is given

\[ Y = Y_1 + Y_2 = (A + 1) \frac{\alpha_2 Y_p U_0}{U_{02}} \left( 1 - \sqrt{\frac{E_{th}}{E}} \right) \]  

(8.3.14)

Here \( \sqrt{E_{th}} = \frac{A}{(A + 1)} \sqrt{E_{th1}} + \frac{1}{(A + 1)} \sqrt{E_{th2}} \)  

(8.3.15)

By using (8.3.9), the angular distributions of the total sputtering yield and the ratio of partial sputtering yield,

\[ Y(\theta_1) = Y_1(\theta_1) + Y_2(\theta_1) = (A + 1) \frac{\alpha_2 Y_p U_0}{8U_{02}} \cos \theta_1 \left[ 4 - \gamma(\theta_1) \sqrt{\frac{E_{th}}{E}} \right] \]  

(8.3.16)

\[ \frac{Y_1(\theta_1)}{Y_2(\theta_1)} = A \cdot \frac{[4 - \gamma(\theta_1) \sqrt{E_{th1}/E}]}{[4 - \gamma(\theta_1) \sqrt{E_{th2}/E}]} \]  

(8.3.17)

i) Fe-Ni alloy

Let's consider Fe-Ni target (18-82%) bombarded by 1 keV Hg\(^+\) ion perpendicularly [121],

\[ M_1 = 55.847, \quad M_2 = 58.7 \quad \text{and} \quad C_1/C_2 = 0.2195 \]  

(8.3.18)

Here \( 1 = \text{Fe} \) and \( 2 = \text{Ni} \).

Let's assume \( U_{01}/U_{02} = 2.1381 \) and \( E_{th1} = 323.11\text{eV} \)

then (8.3.10) gives

\[ E_{th2} = \left( \frac{M_2 U_{02}}{M_1 U_{01}} \right) \cdot E_{th1} = 158.84\text{eV} \]  

(8.3.19)

Substituting \( E = 1000\text{eV} \) and (8.3.18,19) into (8.3.12,13) and (8.3.15), one obtained

\[ A = 0.3064, \quad E_{th} = 192.18\text{eV} \quad \text{and} \quad \alpha_1/\alpha_2 = 0.6540 \]  

(8.3.20)

which predicts that Ni is preferentially sputtered and the surface enrichment of Fe for the steady-state. Then (8.3.16) and (8.3.17) yield the total sputtering yield angular distribution

\[ Y(\theta_1) \propto \cos \theta_1 \left[ 1 - 0.1097 \gamma(\theta_1) \right] \]  

(8.3.21)

and the angular distribution of the ratio of partial sputtering yield,
\[
\frac{C_2 Y_1(\theta_i)}{C_1 Y_2(\theta_i)} = 1.3959 \cdot \frac{[4 - 0.5689\gamma(\theta_i)]}{[4 - 0.3985\gamma(\theta_i)]},
\]

(8.3.22)

which’s normalized with respect to the bulk composition ratio. The analytical results \( Y_j(\theta_i)/C_j \), \( Y(\theta_i) = Y_1(\theta_i) + Y_2(\theta_i) \) and \( C_2 Y_1(\theta_i)/[C_1 Y_2(\theta_i)] \) have been plotted in Fig. 8.3.2 (a), (b) and (c) respectively. The calculated curves of (8.3.21,22) agree Wehner’s experimental results well. Once again, we ignored the bombardment-induced Gibssian Segregation [125] in the analysis. Such Gibssian Segregation would cause a great change of the angular distribution of partial sputtering yield ratio, if it exists.

**Fig. 8.3.2** Angular distribution of sputtered Fe and Ni from FeNi target by normally incident mercury ion with energy 1000eV. Experimental data are cited from R.R. Olson and G.K. Wehner [121]. The calculated curves show good agreement with experimental data. (a) Calculated angular distributions of sputtered Fe and Ni respectively by using (8.3.19); (b) Deposited film thickness (Fe+Ni) Solid line stands for theoretical results (8.3.21). (c) Angular distribution of the ratio of sputtered Fe and Ni. The ratio here refers to the Auger-peaks normalized to the identical ratio in the target value. Solid line stands for theoretical results (8.3.22).
ii) Boron nitride (BN)

Some of more complicate systems, such as BNSiO₂ and BNAIn [137], may not fit our theoretical model described in Chapter 2. In these systems, the binding energies between B and N, Si and O, Al and N may be significant higher than ones between BN and SiO₂, BN and AlN, therefore the majority of sputtered particles are clusters and molecules. In another word, assuming if BN, SiO₂ and AlN are “element” particles respectively, systems BNSiO₂ and BNAIn will be considered binaries and fit our theoretical model. In the cases BNSiO₂ target irradiated, BN molecule are sputtered preferentially due to lighter molecule mass and lower surface binding energy. The sputtering of these targets may provide an excellent opportunity to study emission of clusters and molecules. However, Boron nitride target couldn’t be considered as single “element” (BN block) material due to it’s structure. Yalin et. al. have measured the total sputtering yields and sputtering yield angular distribution by using weight loss and QCM [117][118], see Table 8.3.2 here, or the Table 1 on page 7 of [88]. One can see $Y_{\text{Marg.et.Max.}} \approx 2Y_{\text{Cond.}}$ rather than $Y_{\text{Marg.et.Max.}} \approx Y_{\text{Cond.}}$. Yalin’s experiments showed that most of B atoms and N (may be N₂ molecules) sputtered individually, rather than BN block at least in the low energy ion bombardment [127]. The most of BN must be separated into B and N atoms and amorphous layer should be established in the target surface during the sputtering process [138]. Therefore, Boron nitride (BN) target can be treated as a binary in our theoretical model.

Boron nitride is of high interest due to its remarkable characters and tremendous applications [88][119]. As far as the sputtering mechanism research is concerned, boron nitride is a typical kind of nitrides, because the atomic mass difference between the two components is small, Yalin et. al. have measured the ion-energy dependence of total sputtering yields of BN target bombarded at normal incidence with Xe⁺ ion by using weight loss and QCM [117][118] and obtained two groups of data. They found that the weight loss measurements are lower, by a factor of
approximately 2 to 3, but they are very confident of the QCM measurements. Yalin’s both two
groups of Data (QCM and weight loss) have been plotted in Fig. 8.3.3 with computer simulation
results. Seeing the computer simulation results agree with QCM measurements (E = 100 eV to 350
eV), we used Formula (8.3.14), i.e.

\[
Y_B(E) = \sigma_C B [S_n(E)]_B + C_N [S_n(E)]_N \left( 1 - \frac{E_{th}}{\sqrt{E}} \right)
\]

(8.3.23)

to match the data of QCM measurements and simulations, see Fig. 8.3.3, one can see that

\[ E_{th} \approx 65 \text{eV} \]

(8.3.24)

On the other hand, by using QCM system, Yalin et. al. have measured differential sputter yields of
HBC grade BN target bombarded by normal incidence \( \text{Xe}^+ \) ion with energy of 100, 250, 350, and
500 eV [88]. As mentioned previously, Yalin et. al. only measured the condensable fraction of
sputter products, Boron. Thus, using Zhang expression (8.2.3), i.e.

\[
Y_B(\theta) = \sigma_C (\cos \theta) \left( 1 - \frac{\gamma(\theta)}{4} \frac{E_{thB}}{\sqrt{E}} \right)
\]

(8.3.25)

to match their measured differential sputter yields of boron [88]. Yalin et. al. obtained

\[
\begin{cases}
E_{th} = 104 \text{ eV} & \text{for } E = 250 \text{eV} \\
E_{th} = 88.1 \text{ eV} & \text{for } E = 350 \text{eV} \\
E_{th} = 123 \text{ eV} & \text{for } E = 500 \text{eV}
\end{cases}
\]

(8.3.26)

for boron, see the Table 1 on page 7 of [88]. In principle, a set of equations (8.3.10), (8.3.11), (8.3.12)
and (8.3.15) determines \( E_{th2}, U_{02}/U_{01}, \alpha_i/\alpha_2 \) or \( \alpha \), as long as \( E, E_{th} \) and \( E_{th1} \) are given.

Substituting (8.3.23, 25), the set of equations can be easily solved by using the iteration method.

Thus, we obtained

\[
\begin{cases}
A = 2.23 & \text{and } E_{thN} = 10.91 \text{eV}, \text{ for } E = 250 \text{eV} \\
A = 1.33 & \text{and } E_{thN} = 39.71 \text{eV}, \text{ for } E = 350 \text{eV} \\
A = 1.73 & \text{and } E_{thN} = 78.91 \text{eV}, \text{ for } E = 500 \text{eV}
\end{cases}
\]

(8.3.27)

Polar plot of the analytical results of (8.3.6) have been plotted in Fig. 8.3.4 for \( Y_B(\theta) \) and
Y_N(\theta_i) respectively, if the nitrogen yield is atomic. Since Y_B(\theta_i) has already been measured by Yalin et. al.[88], therefore, measuring the angular distribution of sputtered nitrogen atoms is also very urgent. It’s different from other’s conclusion [146] that our theory predicted N is preferentially sputtered and the surface enrichment of B for the steady-state in the energy region. It is interesting to notice that Y_N(\theta_i) is closed to a cosine function even if Y_B(\theta_i) still is “under-cosine” like.

Thus, although the energy distribution of sputtered Boron predicted by (8.2.10) or (8.2.19) should be significantly narrower than Thompson-Sigmund distribution (8.2.14). If our theory is tenable in the future, I will never forget the encouragement from Prof. A.P. Yalin due to his intensive discussion with me.

APPENDIX VIII: DERIVATION OF (8.2.9)

Integrating (8.2.7) over \( \varepsilon \) and \( \Omega_1 \), we obtained the total sputtering yield,

\[
Y = \int_0^\infty dE_1 d^2\Omega_1 J(E_1, \Omega_1) = 4Y_p \left[ (\text{term})_0 - \sqrt{\frac{E_{th}}{E}} \cdot (\text{term})_1 \right] \quad (\text{VIII.A.1})
\]

Here \((\text{term})_0 = \frac{1}{2} \int_0^\infty \frac{e l e d \varepsilon}{(\varepsilon + 1)^3} \int_0^1 d\mu \exp \left[ - \left( \frac{Ah}{\varepsilon + 1} \right)^{0.5} \right] \)

\((\text{term})_1 = \frac{1}{4} \int_0^\infty \frac{e l e d \varepsilon}{(\varepsilon + 1)^{3.5}} \int_0^1 d\mu \left( \frac{\varepsilon + 1}{\varepsilon + 1} \right)^{0.5} \exp \left[ - \left( \frac{Ah}{\varepsilon + 1} \right)^{0.5} \right] \)

(\text{VIII.A.2}) and (\text{VIII.A.3}) reduced to

\[
(\text{term})_0 = 2\int_0^1 x dx \int_x^1 t dt \exp \left( - \frac{Ah}{t} \right) = 2\int_0^1 t dt \exp \left( - \frac{Ah}{t} \right) \int_0^1 x dx
\]

(\text{VIII.A.4)}
(term)$_i$ = \int_0^1 dx \int_0^t t^2 dt \exp\left(- \frac{Ah}{t}\right) = \int_0^t t^2 dt \exp\left(- \frac{Ah}{t}\right)\int_0^1 dx \\
= \int_0^1 t^2 dt \exp\left(- \frac{Ah}{t}\right) = E_5(Ah).

(VIII.5)

Substituting (VIII.4) and (VIII.5) into (VIII.1), one obtained

\[ Y = Y_p \left(1 - \sqrt{\frac{E_{th}}{E}}\right) \left[4E_5(Ah)\right] \]

(VIII.6)

which is (8.2.9). The end of proof.

**DEDUCTION**

Since directly integrating (8.2.8) over \( \Omega \) gave (8.2.9), we have the following two mathematical identities

\[ \int_0^{\pi/2} \phi(\theta) \sin \theta \cos \theta \, d\theta = 2E_5(Ah) \]

(VIII.7)

\[ \int_0^{\pi/2} \phi_i(\theta) \sin \theta \cos \theta \, d\theta = 8E_5(Ah) \]

(VIII.8)

If \( h = 0 \), (VIII.7) and (VIII.8) reduced to

\[ \int_0^{\pi/2} \sin \theta \cos \theta \, d\theta = 1/2 \]

(VIII.9)

\[ \int_0^{\pi/2} \gamma(\theta) \sin \theta \cos \theta \, d\theta = 2 \]

(VIII.10)

(VIII.9) and (VIII.10) played the critical role in our work [73].

---

**Fig.8.3.3** Ion-energy dependence of total sputtering yields of BN target bombarded at normal incidence with Xenon ion. The solid curves were calculated by using Formulae (8.3.23,24).
Fig. 8.3.4 Polar plot angular distribution of sputtered B and N from BN target by normally incident Xenon ion with energy 250eV, 350eV and 500eV respectively. The solid curves were calculated by using Formulae (8.3.6, 26 and 27). It is remarkable that the angular distribution of atomic nitrogen seems “cosine-like” even if the one of boron still is “under-cosine” like.
CHAPTER 9

SPUTTERING YIELD ANGULAR DISTRIBUTION
MEASURED BY RBS AND NRA

9.1 SPUTTERING TITANIUM NITRIDE

Angular distribution of sputtered atoms from a stoichiometric TiN target had been measured by using Rutherford Backscattering Spectrometry (RBS) for Ti and \(^{14}\text{N} (d, \alpha)^{12}\text{C}\) Nuclear Reaction Analysis (NRA) for nitrogen. A tantalium foil was used to collect nitrogen yield. Unfortunately, NRA technique couldn’t provide meaningful nitrogen signal, because sputtered nitrogen atoms are not condensable.[117][118]. Carbon discs were used for collecting sputtered titanium atoms. After sputtering, RBS technique was used to determine atom number density on the collector surfaces. It was successful to use RBS to measure the angular distribution of condensable, sputtered Ti atoms. Therefore, only sputtering yield of Ti component will be discussed more detail in this section.

As well known [114][120], a necessary condition for obtaining reproducible sputtering yield data is the irradiation is performed with a well-defined ion beam both with regard to ion species and energy. Such kind of Xenon ion beam is obtained from ion implantation (100 KV Varian Extrion) facilities (100 keV ion-implanter in Department of physics and astronomy of WSU), and focused onto the target with a well-defined direction of incidence for sputtering experiment. At least as important as a precise characterization of the incoming beam are parameters describing the target conditions. This condition should be satisfied by using a high quality commercial target. However, the target properties may change during irradiation due to contamination by background gas in the vacuum. For example, N atom could be replaced by O atom on the target surface of TiN absorption of back-ground gas due to poor vacuum may cause a great change of sputtering yield angular distribution [6]. In our experiment, the vacuum in Sputtering Chamber was \(3 \times 10^{-6}\) \(\tau\).
Fig. 9.1 Schematic diagram of the target-collector assembly. Semicircular cylinder is machined from aluminium, and Carbon discs are mounted behind the cylinder uniformly to allow sputtered particles to impact on the polished surface of them.

The experiment setup shown in Fig.9.1 is mounted in a vacuum chamber connected to ion implantation (100 kV Varian Extrion) facilities (100 keV ion implanter in the Department of physics and astronomy, WSU). Xenon ions created from the 100 keV accelerator pass through a 2mm diameter defining aperture and a central hole in a semi-cylindrical collector to impinge on the target. In view of the beam dosimeter is of primary importance in Sputtering yield measurement, the target and yield collector system are mounted in a secondary–electron– suppressor i.e. Faraday cup, see Fig. 9.1. The beam influence is measured by integrating the target current. The electrical insulation of the target from ground is better than 100MΩ. Therefore, the target current measurement should be accurate enough.

The experimental procedures:

1. Cut a carbon bar as 1 mm thick, 5mm diameter, Carbon disc, and polish each carbon disc with sand paper.

2. Mount those polished carbon discs behind the collector, Fig. 9.1, and mount the collector into the Faraday Cup, Fig. 9.1.

3. Close the Implanter beam line valve, close the mean valve of the target room and vent it.
(4) Put the Faraday cup into the target chamber, close the shutter and connect wires of shutter, target, secondary-electro-suppressor to each correspond electrical feed-through.

(5) Close the target chamber and pump down.

(6) Open the ion beam line valve, and adjust the ion Implanter (ion source pressure, focus, etc.) to get the highest shutter current.

(7) Apply a secondary-electron-suppressor voltage $-2.5\text{keV}$.

(8) Set up target current integrator, each count corresponds $10^{-6}$ coulombs, i.e. $6.24 \times 10^{12}$ ions.

(9) Open the shutter, then Sputtering begins to run. The Sputtering conditions are

a) $100\text{ keV} \ Xe^+ \text{ ion beam impinges on} \ TiN \text{ target perpendicularly}$

b) Target current: $0.6\mu\text{A} \sim 3.8 \times 10^{12} \text{ atoms/second}$

c) Vacuum in the Implanter: $4.6 \times 10^{-7} \tau$.

Beam line: $4.6 \times 10^{-7} \tau$.

Target chamber: $3 \times 10^{-6} \tau$.

(10) Running about 1h, the counter number of target current integrator reaches 2517, then close the shutter, close the beam line and vent the target chamber.

9.2 RUTHERFORD BACKSCATTERING SPECTROMETRY (RBS)

A 4.75 MV Van de Graaff accelerator (in the Department of physics and astronomy, W.S.U.) had been used for the surface analysis to determine the number density of sputtered atom deposited on carbon discs [128]. Charged particles are generated in an ion source. Their energy is then raised to several mega electron volts by the accelerator. The high-energy beam and filter it for a selected type of particle and energy. The beam then enters the scattering chamber and impinges on the sample to be analyzed. Some of the backscattered particles impinge on the detector, where they generate an
electrical signal. This signal is amplified and processed with fast analog and digital electronics. The final stage of the data usually has the form of a (digitized) spectrum.

**9.2.1 BASIC PHYSICAL CONCEPTS**

When a projectile of atomic mass $M_1$, atomic number $Z_1$, and energy $E_1$, collides elastically with a stationary target atom of $M_2$ and $Z_2$, the energy will be transferred from $M_1$ to $M_2$. This simple elastic collision problem had been solved by applying the principles of conservation of energy and momentum. Finally, one obtained the Kinematic factor [128],

$$K \equiv \frac{E_1}{E} = \left[ \left( 1 - \mu \sin^2 \theta \right)^{0.5} + \mu \cos \theta \right]^2, \quad \mu \equiv \frac{M_1}{M_2} \quad \text{(9.2.1)}$$

and the differential cross section in the laboratory frame of reference,

$$\frac{d\sigma}{d^2\Omega} = \left( \frac{Z_1 Z_2 e^2}{4E} \right) \frac{4}{\sin^4 \theta} \cdot \frac{\left( (1 - \mu \sin^2 \theta)^{0.5} + \cos \theta \right)^2}{\left[ 1 - \mu \sin^2 \theta \right]^{0.5}} \quad \text{(9.2.2)}$$

**9.2.2 SYSTEM CALIBRATION**

RBS can be used to detect surface impurities on a light-element substrate [128]. For example, a carbon substrate is often used as a control sample to check the quality of a vacuum-deposited layer. Any surface impurity with an atomic mass greater than that of carbon will be visible in a RBS spectrum. These samples can also be used to determine the channel width $\varepsilon$ of the multi-channel analyzer. In our experiment, a RBS spectrum for 2-MeV $^4\text{He}^+$ ions backscattering was at an angle $\theta$ of 160° from a carbon target on which, aluminum, cobalt and gold are present as surface depositions. Three peaks for the depositions at channel number 276, 396.5 and 489. The right-hand scale gives Kinematic values for scattering, with $k = 0.5600$ for Al, 0.7683 for Co, and 0.9242 for Au. (Kinematic values are given by (9.2.1)). By using the least-squares fit, one can find the best fit straight line of Data in TABLE (9.2.1) as following,

$$E_i = (3.4219 \times \text{Ch.No.} + 177.17)\text{keV}. \quad \text{(9.2.3)}$$
Table 9.2.1 Calibration data for RBS.

<table>
<thead>
<tr>
<th>Element</th>
<th>Atomic Mass (amu)</th>
<th>$K_i$</th>
<th>$K_iE_i$ (keV)</th>
<th>Ch. No.</th>
</tr>
</thead>
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<td>Al</td>
<td>27</td>
<td>0.5600</td>
<td>1120</td>
<td>276</td>
</tr>
<tr>
<td>Co</td>
<td>58.9</td>
<td>0.7683</td>
<td>1537</td>
<td>396.5</td>
</tr>
<tr>
<td>Au</td>
<td>197</td>
<td>0.9242</td>
<td>1848</td>
<td>489</td>
</tr>
</tbody>
</table>

Table 9.2.2 Channel number of Carbon, Nitrogen and Titanium

<table>
<thead>
<tr>
<th>Element</th>
<th>Atomic Mass (amu)</th>
<th>$K_i$</th>
<th>$K_iE_i$ (keV)</th>
<th>Ch. No.</th>
</tr>
</thead>
<tbody>
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<td>C</td>
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<td>0.2600</td>
<td>520</td>
<td>100</td>
</tr>
<tr>
<td>N</td>
<td>14</td>
<td>0.3192</td>
<td>638.4</td>
<td>135</td>
</tr>
<tr>
<td>Ti</td>
<td>47.9</td>
<td>0.7231</td>
<td>1446.2</td>
<td>370</td>
</tr>
</tbody>
</table>

9.2.3 NUMBER DENSITY OF SPUTTERED ATOMS DEPOSITED ON CARBON DISC

The calibration formula (9.2.3) can be used to determine the Ch. No. for any element. Specially, the Ch. NO of Carbon, Nitrogen and Titanium had been calculated and collected in TABLE (9.2.2).

In our experimental measurement, a uniform 2-MeV $^4$He$^+$ beam impinges at normal incidence on a uniform target that is larger than the area of beam cross section (Diameter. 2mm). The RBS spectrum is obtained from a thick carbon disc target. The sputtered material deposited on the carbon disc is about a mono-atomic layer $\left(\sim 10^{15} \text{atoms} \cdot \text{cm}^{-2}\right)$. It then follows from Eq.(3.70) in literature [128] that the spectrum high for scattering from the top surface layer can be written in the surface energy approximation as
\[ H_0 = \sigma(E_0)\Omega Q \varepsilon/[\varepsilon_0] \]  

(9.2.4)

Where \( Q \) is the total number of particles hit the target, \( \varepsilon \) is energy width of one channel, \([\varepsilon_0]\) is the stopping cross section and \( \Omega \) is very small detector solid angle.

On the other hand, the number density \( S_i \) of impurities can be calculated directly from the area of the signals \( A_i \) given by the total number of counts integrated over the region of interest. For a given impurity denoted by the subscript \( i \), \( A_i \) can be expressed by Eq.(5.1) in [128],

\[ A_i = \sigma_i(E_0)\Omega Q S_i \]  

(9.2.5)

for normal incidence. One way to determine \( S_i \) is to use the thick-target signal of the substrate as a reference. The yield \( H_{c,o} \) for scattering from the carbon surface is given by (9.2.4). By combining (9.2.4) and (9.2.5), one obtained.

\[ S_i = \frac{A_i}{H_{c,o}} \cdot \frac{\sigma_i(E_0)}{\sigma_i(E_0)} \cdot \frac{\varepsilon}{[\varepsilon_0]^c} \]  

(9.2.6)

where the subscripts \( c,o \) refer to the carbon substrate and surface energy approximation. Substituting (9.2.2) into (9.2.6), one got

\[ S_i = \frac{A_i}{H_{c,o}} \cdot \left( \frac{6}{Z_i} \right)^2 \cdot \frac{\varepsilon}{[\varepsilon_0]^c} \]  

(9.2.7)

Further substituting \([\varepsilon_0]^c = 2.597 \times 10^{-14} \text{ ev} \cdot \text{cm}^2 \) (see p.363 in [128]) and \( \varepsilon = 3.4219 \text{ keV} \) (see (9.2.3)) into (9.2.7), obtained,

\[ S_i = \frac{A_i}{H_{c,o}} \cdot \left( \frac{6}{Z_i} \right)^2 \times 1.3176 \times 10^{17} \text{ atom} \cdot \text{[cm]}^{-1} \]  

(9.2.8)

For Nitrogen and Titanium, substituting \( Z_N = 7 \) and \( Z_{Ti} = 22 \) into (9.2.8) respectively, we got,

\[ S_N = \frac{A_N}{H_{c,o}} \times 0.9680 \times 10^{17} \text{ atom} \cdot \text{[cm]}^{-1} \text{ and} \]

\[ S_{Ti} = \frac{A_{Ti}}{H_{c,o}} \times 0.9800 \times 10^{16} \text{ atom} \cdot \text{[cm]}^{-1} \]
Our experimental conditions for RBS are:

1. $2$-MeV $^4\text{He}^+$ beam incidents on carbon disc sample perpendicularly.

2. Beam Current: 220 nA

3. F. S. Ampers: $6 \times 10^{-7}$ C

4. Pulse Number: 1000. Thus,

$$H_{e,o} = 7.75 \times 10^4.$$  \hfill (9.2.9)

Substituting (9.2.9) and measurement results in TABLE 9.2.3 into (9.2.8), we obtained sputtered material densities on each carbon disc sample. The calculated results are collected in TABLE 9.2.4. One can see that, the number density of sputtered atom is about $\sim 10^{15}$ atoms $\cdot$ cm$^{-2}$, i.e. a mono-atomic layer.

<table>
<thead>
<tr>
<th>Angle from normal (Deg)</th>
<th>$A_{Ti}$</th>
<th>$A_{N}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Right</td>
<td>Left</td>
</tr>
<tr>
<td>5</td>
<td>14285.7</td>
<td>12890.8</td>
</tr>
<tr>
<td>15</td>
<td>13317.8</td>
<td>9818.3</td>
</tr>
<tr>
<td>25</td>
<td>10141.2</td>
<td>9836.5</td>
</tr>
<tr>
<td>35</td>
<td>9366.3</td>
<td></td>
</tr>
<tr>
<td>45</td>
<td>8021.5</td>
<td>6916.8</td>
</tr>
<tr>
<td>55</td>
<td>6933.8</td>
<td>5786.8</td>
</tr>
<tr>
<td>65</td>
<td>5490.5</td>
<td>5170.0</td>
</tr>
<tr>
<td>75</td>
<td>3767.5</td>
<td>3166.0</td>
</tr>
<tr>
<td>85</td>
<td></td>
<td>1224.3</td>
</tr>
</tbody>
</table>
Table 9.2.4 Number density of sputtered atom on carbon discs

<table>
<thead>
<tr>
<th>Angle from normal (Deg)</th>
<th>$S_{Ti}$ [10^{14} \text{atoms} \cdot \text{cm}^{-2}]$</th>
<th>$S_{N}$ [10^{14} \text{atoms} \cdot \text{cm}^{-2}]$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Right</td>
<td>Left</td>
</tr>
<tr>
<td>5</td>
<td>18.06</td>
<td>16.3</td>
</tr>
<tr>
<td>15</td>
<td>16.84</td>
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<td>12.82</td>
<td>12.44</td>
</tr>
<tr>
<td>35</td>
<td>11.84</td>
<td></td>
</tr>
<tr>
<td>45</td>
<td>10.14</td>
<td>8.75</td>
</tr>
<tr>
<td>55</td>
<td>8.77</td>
<td>7.32</td>
</tr>
<tr>
<td>65</td>
<td>6.94</td>
<td>6.54</td>
</tr>
<tr>
<td>75</td>
<td>4.76</td>
<td>4.00</td>
</tr>
<tr>
<td>85</td>
<td></td>
<td>1.55</td>
</tr>
</tbody>
</table>

9.3 EXPERIMENTAL RESULTS OF DIFFERENTIAL SPUTTERING YIELD

The differential sputtered yield relates to the sputtered material density on carbon disc through the following relation,

$$\frac{d^3Y_i}{d^2\Omega_i} = r^2 \cdot \frac{S_i}{N_0}$$  \hspace{1cm} (9.3.1)

where, $r$ is the radius of the sputtering yield collector, $N_0$ is the number of incident atom. For our experiments,

$$r = 3 \text{cm}; \quad N_0 = 10^{-6} \times 2517C/(1.602 \times 10^{-19} \text{C}) = 1.57 \times 10^{16}.$$  

Substituting $r$ and $N_0$ into (9.3.1), one obtained the total sputtering yield of titanium

$$Y_{Ti} = 2.67$$  \hspace{1cm} (9.3.2)

and the corresponding differential sputtering yield in Table 9.3.1. Due to the perpendicular incidence of Xenon ion beam, in consideration of the obvious symmetric property, let's define the average differential sputtering yield as
\[
\frac{d^2Y}{d^2\Omega_i} = \frac{1}{2} \left[ \frac{d^2Y}{d^2\Omega_i} \right]_{\text{Right}} + \frac{d^2Y}{d^2\Omega_i} \right]_{\text{Left}} \quad (9.3.3)
\]

and plotted the results in Fig. 9.2. One can see that (9.3.3) satisfies cosine-like angular distribution:

\[
d^2Y/d^2\Omega_i = 0.85 \cos \theta_i. \quad (9.3.4)
\]

Besides, the sputtering yield of TiN bombarded by 0.5 keV Cd\textsuperscript{+} had been given \[133\].

The experimental result is 0.7. Considering the steady state, it's natural to take,

\[
Y_{\text{Ti}} = 0.35 \quad \text{for} \quad 0.5 \text{keV} - \text{Cd}^+ \rightarrow \text{TiN}. \quad (9.3.5)
\]

let's define the ratio,

\[
R \equiv \frac{Y_{\text{Ti}}(0.5 \text{keV} - \text{Cd}^+ \rightarrow \text{TiN})}{Y_{\text{Ti}}(100 \text{keV} - \text{Xe}^+ \rightarrow \text{TiN})} \quad (9.3.6)
\]

Thus, the experimental results (9.3.2) and (9.3.5) give

\[
R_{\text{exp}} = \frac{0.35}{2.67} = 0.131.
\]

On the other hand, by using Sigmund's theory, \( R \) in (9.3.6) can be calculated,

\[
R_{\text{sig}} = \frac{[\alpha]_{\text{Ti}}^{\text{Cd}^+} \cdot [S_n(0.5 \text{keV})]_{\text{Ti}}^{\text{Cd}^+} + [\alpha]_{\text{Ti}}^{\text{Cd}^+} \cdot [S_n(0.5 \text{keV})]_{\text{Ti}}^{\text{Cd}^+}}{[\alpha]_{\text{Ti}}^{\text{Xe}^+} \cdot [S_n(100 \text{keV})]_{\text{Ti}}^{\text{Xe}^+} + [\alpha]_{\text{Ti}}^{\text{Xe}^+} \cdot [S_n(100 \text{keV})]_{\text{Ti}}^{\text{Xe}^+}}
\]

\[
= \frac{0.166 \times 393.3 + 0.205 \times 764.1}{0.166 \times 2457 + 0.205 \times 5493} = 0.149.
\]

Therefore, \( R_{\text{exp}} \) agrees with \( R_{\text{sig}} \) reasonably well. But \( R_{\text{exp}} \) is smaller than \( R_{\text{sig}} \) about 20% is probable due to ignoring the an-isotropic correction discussed intensively in the previous Chapter.
Table 9.3.1 Differential Sputtering Yields (Atoms/ion x sterad)

<table>
<thead>
<tr>
<th>Angle from normal (Deg)</th>
<th>$\frac{d^2Y_{Ti}}{d^2\Omega_1}$</th>
<th>$\frac{d^2Y_{N}}{d^2\Omega_1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Right</td>
<td>Left</td>
</tr>
<tr>
<td>5</td>
<td>1.04</td>
<td>0.93</td>
</tr>
<tr>
<td>15</td>
<td>0.97</td>
<td>0.71</td>
</tr>
<tr>
<td>25</td>
<td>0.73</td>
<td>0.71</td>
</tr>
<tr>
<td>35</td>
<td>0.68</td>
<td></td>
</tr>
<tr>
<td>45</td>
<td>0.58</td>
<td>0.50</td>
</tr>
<tr>
<td>55</td>
<td>0.50</td>
<td>0.42</td>
</tr>
<tr>
<td>65</td>
<td>0.40</td>
<td>0.37</td>
</tr>
<tr>
<td>75</td>
<td>0.27</td>
<td>0.23</td>
</tr>
<tr>
<td>85</td>
<td></td>
<td>0.089</td>
</tr>
</tbody>
</table>

Fig. 9.2 Polar diagram of Titanium sputtered from TiN target by normally incident Xenon with energies 100keV. The solid stands for $0.85 \cos \theta$. 
CHAPTER 10
CONCLUSIONS AND FUTURE RESEARCH

10.1 CONCLUSIONS

10.1.1 SIGMUND THEORY IS THE LIGHTHOUSE

Sputtering was discovered experimentally more than 150 years ago. Both experimental and theoretical interest in this phenomenon has increased constantly for many years, due to remarkable applications in different fields. In 1969, by the aid of the Lindhard power cross-section (6.2.24), Sigmund developed the well-known modern sputtering theory. The theory has been found to agree reasonably well with corresponding experimental data. Because Sigmund directly writes his basic equations in a backward form, therefore he can give analytical results. Since then, a lot of analytical works on sputtering and related phenomena have been published. These works mainly focused on two aspects. The first aspect is dealing with a monatomic target. In this case, a number of atomic distribution functions, such as the slowing-down, recoil, scattering and collision densities, etc., are introduced and derived to describe particle flux in a linear atomic collision cascade. Furthermore, taking the anisotropic (or momentum) term \( L = 1 \) into account, these distribution functions have been derived asymptotically in order to explain the energy and angular distributions of sputtered atoms. The second aspect is dealt with multi-component targets. In the latter case, almost only isotropic term \( (L = 0) \) has been taken cared of, and the anisotropic correct term \( (L = 1) \) has never been discussed in detail. Thus, it is tured out to be mandatory to develop a new anisotropic sputtering theory in multi-component case. Since we understood that any new sputtering theory must automatically reduce to Sigmund theory, if the necessary conditions are satisfied, therefore, Sigmund theory can be considered as a Guiding line, or lighthouse.

10.1.2 MY CONTRIBUTIONS IN SPUTTERING THEORY

The first part of research comprising my dissertation has focused on the transport theory of
sputtering. In Chapter 2, we have derived all Linear Transport Equations appeared in Sputtering theory based on the standard Botzmann equation in classical Statistics Mechanics. The relations among these Linear Transport Equations have been discussed in detail. In Chapter 3, substituting separable scattering cross-sections, we have solved these equations by using Laplace Transformation for both isotropic term \((L = 0)\) and anisotropic term \((L = 1)\). The accuracy of these analytical solutions is demonstrated by comparison with Monte Carlo simulations published by Urbassek et al.[23]. In Chapter 4, we proposed a new method (“SC” theory) to build up approximate asymptotic solutions for arbitrary cross sections. We have showed that “VCU” theory [13] even could generate non-physics negative sputtered particle flux in some cases simulated by the author themselves and found the reasons of the mistake. In Chapter 5, by using Laplace Transformation, we have derived a pool of recursion relations for asymptotic moments of deposited energy, momentum depth distribution and ion range. We also have given the Glazov’s paradox an exact explanation.

The second part of research comprising my dissertation has focused on the two major problems appeared in sputtering theory: one is the depth of origin of sputtered atoms \((L = 0)\) and another one is the anisotropic sputtering theory \((L = 0, 1)\).

Based on the transport theory, we confirmed the atom ejection theory proposed by Falcone and Sigmund [32], denied the theory proposed by Falcone [34] and derived an explicit expression for the distribution of the depth of origin of sputtered atoms. Thus, the mean sputter depth was estimated about twice longer than the experiment measurement. In Chapter 6 and 7, we introduced (6.2.9) into Sigmund sputtering theory. Compared with (6.2.24), (6.2.9) can give a much better approximation to the Born-Mayer scattering in the low energy region \((m \sim 0.1)\). By using both power cross-sections, we have solved the linear transport equations describing the sputtering problem asymptotically. In the Chapter 3, it has further been proved that Falcone’s theory of the atom ejection process contradicts the transport theory. In the Chapter 6, the Andersen-Sigmund relation for partial
sputtering yield ratios between two elements in an arbitrary multi-component target has been derived by both cross-sections. The energy deposited in the target surface layers has been computed for a few typical ion-target combinations. The numerical curves show that both cross-sections generate almost the same results (error<10%) for \( m = 0.2 \). In the Chapter 7, it is also shown that, if the sputtering yield equals the corresponding one in Sigmund’s theory, the depth of origin of sputtered atoms may be only about one half of the latter as long as \( m = 0 \), because the hard sphere collision is dominant in this low energy cascade. Therefore, the entire Sigmund sputtering theory has been rebuilt and the problem on the depth origin of sputtered atoms has been solved. In Chapter 8, taking the momentum deposition into account, a new modified Sigmund Sputtering Theory has been developed to describe anisotropic sputtering phenomena induced by low energy and heavy ion bombardment, including sputtering yield energy and angular distributions as well as isotopic effect.

The momentum deposition usually ignored [1], but could play an important role in the atom collisions in solid, such as the anisotropic transport in the ion mixing [151–152]. We have clearly demonstrated the intrinsic relation between the ion energy dependence of total sputtering yields and the angular distribution of sputtered atoms induced by low-energy heavy ion bombardment. The sputtering yield angular distributions have calculated based on the intrinsic relation for many ion-target combinations. The calculated curves show excellent agreement with the corresponding Wehner’s experimental results of sputtering yield angular distribution. The fact clearly confirmed the intrinsic relation. This intrinsic relation had been ignored in Yamamura’s papers (1981, 1982) due to some obvious mistakes. Our new anisotropy sputtering theory has been used to calculate the sputtering yield energy distributions and showed to fit the corresponding experimental results except the cases where the larger ion incident angle and larger sputtering emission angles were considered. The larger discrepancy between the new theory and experiment result in the latter cases probably is due to the influence of direct recoil atoms on the energy spectrum. The fact clearly demonstrated the great importance of momentum deposited on the target surface in the physical sputtering
Our new anisotropic sputtering theory predicted that the ratio of the asymmetric term \((L = 1)\) and the isotropic term \((L = 0)\) must be negative, and proportional to \(\cos \theta \sqrt{M_1 E_0 / E}\) in magnitude for a normal incidence. This is the intrinsic reason why isotopic mixtures are characterized by higher erosion rates for lower isotopic masses and vice versa for a low energy ion bombardment. In particular, the asymmetric term \((L = 1)\) could be comparable with the isotropic term \((L = 0)\) in magnitude for the near threshold energy ion incidence. The former may cancel the most part of the latter. Thus, this effect could be magnified and become even more pronounced in some ratios. Therefore, our theory successfully solved the isotope puzzle in low energy sputtering.

For BN target bombarded by low energies \(Xe^+\) ion, the experiment measurements [122-124] showed that heavy isotope of boron \(^{11}\text{B}\) is preferentially sputtered. Similar results were observed for copper and silver. The key point to solve the problem is the geometry of the experimental setup. The ion incident angle is 50 degree and the emission angle of sputtered atoms is 40 degree. The emission line of sputtered atoms is perpendicular to the ion incident line exactly. Due to the same reason, they found boron, rather Nitrogen, sputtered preferentially [148].

It’s different from other’s conclusion [122-124][146][148], our new theory predicted N is preferentially sputtered and the surface enrichment of B; light isotope of boron \(^{10}\text{B}\), rather than heavy isotope of boron \(^{11}\text{B}\), preferentially sputtered for the steady state in the energy region.

The third part of research comprising my dissertation is one experiment. It has been down for the measurement of sputtering yield and yield angular distribution. In Chapter 9, by using RBS for determining titanium (Ti) and NRA for nitrogen (N), we have measured the angular differential sputtering yield of Ti and N sputtered from stoichiometric TiN target bombarded by 100keV xenon ion perpendicularly. We have found that the angular differential sputtering yield of Ti can be approximately expressed by

\[
d^2 Y / d^2 \Omega = 0.85 \cos \theta.
\]
10.2 FUTURE WORK

10.2.1 EXPERIMENTAL RESEARCH

Sputtering Boron nitride target by low heavy ion bombardment provided excellent opportunity to study the basic mechanism of collisional sputtering. The angular distributions of sputtered $^{10}\text{B}$ and $^{11}\text{B}$ atoms can be measured by using NRA [143] [144]. It may be a rather challenging work to measure the sputtering yield angular distribution of atom N or molecular N$_2$. The reference [145] showed that “the sticking probability is almost unity up to coverage equivalent to 3 to 5 mono-layers for O$_2$ and to about one monolayer for N$_2$ ....”. This result can be used to determine the angular distribution of sputtered oxygen or nitrogen collected on tantalum films. The key point for collecting nitrogen on tantalum film is the temperature, i.e. 77 degree kelvin (liquid nitrogen). In addition, an ultra high vacuum (UHV) system for measuring sputtered N or O atoms is also necessary.

10.2.2 THEORRETICAL RESEARCH

Future theoretical work should involve computer simulation.

The real situation of sputtering is not as clear if multi-component materials are bombarded. In the case, the surface composition is often changed due to preferential sputtering, bombardment induced mixing, surface segregation, etc. Moreover, the phase structures of the bombarded material will exert a large influence on the development of ion induced surface topography [149]. As a consequence, the actual system will in general be much different from what is assumed in theoretical approaches. Experimentally, much less data exist on sputtering yields of this kind of materials, and no tool has been published which allows an accurate estimate of unknown yield values. However, we could use computer simulation to make some predictions.

Computer simulation of ion channeling is another interest and important project in Ion Beam Science, because it allows a straightforward control of the physics involved in the motion of ions in the crystal [150].
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and Meth. B 27 (1987) 1-20


ABSTRACT

HARMONIC SPUTTERING THEORY

by

ZHU LIN ZHANG

December 2009

Advisor: Dr. Karur R. Padmanabhan

Major: Physics

Degree: Doctor of Philosophy

Based on the standard Botzmann equation in Classical Statistics Mechanics, we have derived a variety of Linear Transport Equations appeared in Sputtering Theory for a random, infinite multi-components medium. The pertinent relations among these Linear Transport Equations have been studied in detail. We have introduced exact classical scattering cross-sections for power potential interaction collision into these Transport Equations and solved them asymptotically by using Laplace Transformation for both isotropic term and anisotropic term. A pool of analytical asymptotic solutions has been given for both Lindhard power cross-section and our exact cross-section, such as slowing-down, recoil, scattering and collision densities, etc. The accuracy of these analytical solutions is demonstrated by comparison with Monte Carlo simulations. For the sputtered atom ejection process, based on the intrinsic relation between recoil density and sputtering calculations, we have proved that the Falcone “mean free path” theory directly contradicts the transport theory, and confirmed Falcone-Sigmund “slow down straight” theory in mono-atomic medium case. After the exact classical scattering cross-section, instead of Lindhard power cross-section, was introduced into the linear transport theory, only one significant different between them has been found: the depth of origin of sputtered atoms must be shorter about one half due to the hard sphere collision is dominant in the low energy cascade. Therefore, the entire
Sigmund sputtering theory can be rebuilt and the problem on the depth origin of sputtered atoms has been solved in a natural way. In addition, we have found that a theory proposed by Urbassek et al. (1993) could generate non-physics negative sputtered particle flux in some cases simulated by the author themselves. We also have given the Glazov’s paradox (1994, 1995) an exact explanation.

We have derived an expression for the anisotropic sputtered particle flux at first time for multi-components target. Taking the momentum deposition into account, a new modified Sigmund Sputtering Theory has been developed to describe anisotropic sputtering phenomena induced by low energy and heavy ion bombardment, including sputtering yield energy and angular distributions as well as isotopic effect. The momentum deposition has been showed to be important in the atom collisions in solid, even if usually ignored in literetures. We have clearly demonstrated the intrinsic relation between the ion energy dependence of total sputtering yields and the angular distribution of sputtered atoms induced by low-energy heavy ion bombardment. The sputtering yields energy and angular distributions have calculated based on the intrinsic relation for many ion-target combinations. Our new theory has been shown to fit the corresponding experimental results of sputtering yield energy distributions well except the cases where the larger ion incident angle and larger sputtering emission angles were considered.

Our new anisotropic sputtering theory predicted that the ratio of the asymmetric term and the isotropic term must be negative, and proportional to $\cos \theta_0 \sqrt{M_1 E_0 / E}$ in magnitude for a normal incidence. This is the intrinsic reason why isotopic mixtures are characterized by higher erosion rates for lower isotopic masses and vice versa for a low energy and heavy ion bombardment. In particular, the asymmetric term could be comparable with the isotropic term in magnitude for the near threshold energy ion incidence. The former may cancel the most part of the latter. Thus, this effect could be magnified and
become even more pronounced in some ratios. Therefore, the present theory successfully solved the isotope puzzle in low energy sputtering.

It's different from other authors, our new theory predicted N is preferentially sputtered and the surface enrichment of B; light isotope of boron $^{10}\text{B}$, rather than heavy isotope of boron $^{11}\text{B}$, preferentially sputtered and the surface enrichment of $^{11}\text{B}$ in the steady-state for BN target sputtered by low energy xenon ion bombardment. Nuclear reaction analysis (NRA) has been proposed to do the experimental measurements to prove the statements.

By using RBS for determining titanium (Ti) and NRA for nitrogen (N), we have measured the angular differential sputtering yield of Ti and N sputtered from stoichiometric TiN target bombarded by 100keV xenon ion perpendicularly. We have found that the angular differential sputtering yield of Ti can be approximately expressed by

$$\frac{d^2Y}{d^2\Omega_i} = 0.85 \cos \theta_i.$$
AUTOBIOGRAPHICAL STATEMENT

Education:
1965~1970, University of Science and Technology of China (USTC), The People’s Republic of China: Bachelor of Condensed Matter Physics

Publications:

Note: “Nucl. Instr. and Meth. B” stands for “Nuclear Instruments and Methods in Physics Research Section B”.