

Chapter 2

Ion Channeling

2.1 Basic Facts

A positively charged particle impinging on a single crystal in a direction close to a major crystallographic axis will very probably experience a correlated series of many small angle collisions with the crystal's atoms [51]. As a result, an oscillatory particle motion in the space between the atomic strings of the crystal, i.e., along the axial crystal channels, is established. The process is called axial particle channeling. Under such conditions, the yields from close encounter processes involving the particle and crystal's atoms are very low. If the particle stays in one channel during the propagation through the crystal, the process is referred to as hyperchanneling. Similar behavior is expected if the incident particle direction is in a plane close to a major crystallographic plane, and the process is called planar channeling.

The possibility of channeling of positively charged particles in crystals was mentioned for the first time by Stark and Wendt in 1912 [52, 53]. However, the process was experimentally observed first about 50 years after that by Rol et al. [54], and then by Almén and Bruce [55]. Those authors showed that the yield of ion sputtering of monocrystalline target surfaces depended significantly on their orientation. In between the appearances of those two studies, Davies et al. [56, 57] reported on the anomalously long ranges of ions moving through polycrystalline targets. In a reaction to the results presented in [55], Robinson [58] emphasized the importance of the direction of ion penetration through crystals. Soon after that, Robinson and Oen [59, 60] reported on the computer simulations of slowing down of Cu atoms of kinetic energies between 1 and 10 keV in various monocrystalline targets, in which very large penetration depths of the ions with the incident directions close to the major crystallographic axes were observed. Those results were crucial for explaining the results of the above-mentioned three experimental studies – by the process of ion channeling. It became clear how an ion of a kinetic energy of the order of a keV or even MeV can move long in a potential well of a

depth of the order of an eV. Lehman and Leibfried [61, 62], Nelson and Thompson [63], Lindhard [64], and Erginsoy [65] continued the theoretical exploration of the process. They introduced the concept of continuum interaction potential of a channeled ion and an atomic string and, using the approach of statistical mechanics, founded the analytical theory of ion channeling. The theory was described in detail by Lindhard [66].

The analytical theory of axial ion channeling is concentrated on a single atomic string and is based on the assumptions that: (i) the ion-string interaction is well described applying classical mechanics, (ii) the string can be treated as continuous, rather than discrete, and (iii) the transverse ion energy is conserved [66]. These assumptions enable an analytical description of the ion motion along the atomic string. In order to calculate the flux of channeled ions, one has to introduce an additional assumption – that the ion has an equal probability of finding itself at any point in the region of the transverse plane accessible to it, which is determined by its transverse energy [66]. However, this assumption, referred to as the assumption of statistical equilibrium in the transverse plane, makes the theory applicable only for thick crystals [67]. This theory has been improved by abandoning the assumption of conservation of the transverse ion energy, i.e., taking into account the energy losses of the ions in their collisions with the crystal's nuclei and electrons, and treating their motion along the atomic string as the diffusion in the transverse plane [68, 69].

2.2 Interaction Potentials

It is usually assumed that the interaction of an ion and an isolated atom can be sufficiently accurately described as a two-body interaction, i.e., the interaction of the nuclei of the collision partners screened by their electron clouds [51]. One also supposes that the ion-atom interaction potential does not depend on their relative velocity and that it is not influenced by the inelastic processes during the encounter. The interaction of an ion and a crystal is most often treated starting from such an interaction potential. This approach is applied in studying the process of charged particle channeling as well.

It has been demonstrated that the (statistical) Thomas-Fermi atomic model can lead to sufficiently accurate atom-atom interaction potentials [51]. With this approximation, the interaction potential reads

$$U(r_a) = \frac{Z_1 Z_2 e^2}{r_a} \varphi_s \left(\frac{r_a}{a_s} \right), \quad (2.1)$$

where Z_1 and Z_2 are the projectile and target atomic numbers, respectively, e is the elementary charge, r_a is the atom-atom distance, $\varphi_s(r_a/a_s)$ is the atom-atom screening function, and a_s is the atom-atom screening length. The function φ_s describes the screening of the nuclei of the collision partners with their electron clouds, but without taking care of the shell structures of the clouds. It is given as the numerical

solution of a differential equation [70]. Molière [71] found a good analytical approximation of the screening function,

$$\varphi_M\left(\frac{r_a}{a_s}\right) = \sum_{\ell=1}^3 \alpha_{\ell} \exp\left(-\frac{\beta_{\ell} r_a}{a_s}\right), \quad (2.2)$$

where $(\alpha_{\ell})=(0.10, 0.55, 0.35)$ and $(\beta_{\ell})=(6.0, 1.2, 0.3)$ are the fitting parameters. This expression has been commonly used in treating atomic collisions in solids.

Lindhard [66] proposed another approximation of the Thomas-Fermi atom-atom screening function, which reads

$$\varphi_L\left(\frac{r_a}{a_s}\right) = 1 - \frac{1}{\left[1 + C_L(r_a/a_s)^{-2}\right]^{1/2}}, \quad (2.3)$$

where C_L is a fitting parameter whose standard value is 3. In this expression, the two terms represent the nuclear and electronic contributions to the atom-atom interaction potential, respectively. This screening function has been widely used in studying the process of ion channeling.

In the Thomas-Fermi atomic model, the atomic screening length is

$$a_{TF} = \left(\frac{9\pi^2}{128}\right)^{1/3} Z_2^{-1/3} a_0, \quad (2.4)$$

where $a_0 = \hbar^2/(m_e e^2) = 0.0529$ nm is the Bohr radius, m_e is the electron mass, and $\hbar$ is the reduced Planck constant [72]. Firsov [73] showed that, when an atom-atom collision is considered, one should use the screening length

$$a_F = \left(\frac{9\pi^2}{128}\right)^{1/3} \left(Z_1^{1/2} + Z_2^{1/2}\right)^{-2/3} a_0. \quad (2.5)$$

This expression reduces to Eq. (2.4) when $Z_1 \ll Z_2$. An alternative expression for the atom-atom screening length,

$$a_L = \left(\frac{9\pi^2}{128}\right)^{1/3} \left(Z_1^{2/3} + Z_2^{2/3}\right)^{-1/2} a_0, \quad (2.6)$$

was proposed by Lindhard [72]. This means that for a_s , appearing in Eqs. (2.2) and (2.3), one should choose a_{TF} , a_F , or a_L . It has been established [51] that, when the projectile is a bare nucleus, the most appropriate choice is a_{TF} . If the projectile is a screened nucleus, one should choose a_F or a_L .

Ziegler, Biersack, and Littmark (ZBL) [74–76] introduced an approximation of the screening function based on the Hartree-Fock method and the assumption that the shell structures of the electron clouds of the partners did not change during the collision process. They applied it to 261 atomic pairs. This approximation, which is

designated as the universal atom-atom screening function, is often used in treating atomic collisions. It reads

$$\varphi_{\text{ZBL}}\left(\frac{r_a}{a_{\text{ZBL}}}\right) = \sum_{\ell=1}^4 \gamma_{\ell} \exp\left(-\frac{\delta_{\ell} r_a}{a_{\text{ZBL}}}\right), \quad (2.7)$$

with

$$a_{\text{ZBL}} = \left(\frac{9\pi^2}{128}\right)^{1/3} (Z_1^p + Z_2^p)^{-1} a_0 \quad (2.8)$$

as the atom-atom screening length, where $(\gamma_{\ell})=(0.1818, 0.5099, 0.2802, 0.02817)$, $(\delta_{\ell})=(3.2, 0.9423, 0.4028, 0.2016)$, and $p=0.23$ are the fitting parameters.

2.3 Computer Simulations

2.3.1 Applicability of Classical Mechanics

Let us imagine a collision of two bare nuclei, the projectile and target, with atomic numbers Z_1 and Z_2 , whose masses are m_1 and m_2 , respectively, with the condition $m_2 \gg m_1$. The projectile velocity and impact parameter are v and b , respectively. Initially, the target is at rest and, being much heavier than the projectile, can be taken to stay that way. The relativistic effects are ignored. We treat the projectile as a wave packet. According to Bohr [77], there are two contributions to the uncertainty of the projectile scattering angle. The first contribution is due to the uncertainty of its initial position while the source of the second contribution is its diffraction. The resulting two conditions reduce to Bohr's condition for application of classical mechanics, which reads

$$\frac{2Z_1 Z_2 e^2}{\hbar v} \gg 1, \quad (2.9)$$

where e is the elementary charge and $\hbar$ is the reduced Planck constant. According to this expression, there is a projectile kinetic energy above which classical mechanics cannot be used.

Lindhard [66] showed that, when two screened nuclei collide and the corresponding screening function is given by Eq. (2.3), the condition for application of classical mechanics is

$$\frac{2Z_1 Z_2 e^2}{\hbar v} \gg 1 + \frac{3b^2}{C_L a_s^2}. \quad (2.10)$$

This means that for a sufficiently high ion kinetic energy or a sufficiently large ion impact parameter, i.e., a sufficiently small ion scattering angle, quantum mechanics must be used. For example, if a proton of kinetic energy of 2 MeV is incident on a Si atom, $C_L=3$, and $b=a_s$, the left-hand side of the inequality (2.10) is ~ 3 and its right-hand side is 2, i.e., the inequality is not valid. Thus, since in ion channeling b is above $\sim a_s$, making, in this case, the right-hand side of the inequality even larger than 2, one might conclude that classical mechanics cannot be used for describing the process of 2 MeV proton channeling in an axial channel of a Si crystal. However, Lindhard [66] demonstrated that, even though an individual small angle collision may not be amenable to classical treatment, the situation is opposite for a correlated series of such collisions. He did that in cases of a real (discrete) atomic string of a crystal, consisting of a number of equally displaced atoms, and of a continuous string, obtained after a uniform longitudinal smearing of its atoms. In case of the real atomic string, the uncertainty of the ion scattering angle does not grow during the series of collisions. Also, the necessity to use quantum mechanics decreases as the ion kinetic energy increases. In case of the continuous atomic string, Lindhard obtained the following condition for application of classical mechanics:

$$2 \left(\frac{Z_1 Z_2 m_1 a_s^2}{m_e d a_0} \right)^{1/2} \gg 1, \quad (2.11)$$

where m_e is the electron mass, d is the distance between the atoms of the atomic string, and a_0 is the Bohr radius. This condition is independent of the projectile velocity and is certainly satisfied for $m_1 \gg m_e$.

As it has been said in the Prologue, we shall discuss here about ion channeling in various crystals and carbon nanotubes, and about positron channeling in carbon nanotubes. In accordance with the inequality (2.11), the cases of ion channeling will be described using classical mechanics, and the cases of positron channeling using quantum mechanics [66].

2.3.2 Continuum String Model

From a classical point of view, a positively charged particle is considered to be channeled with respect to an atomic string of a crystal if it is incident on the string at an angle, φ , small enough that it is scattered away from the string by a correlated series of many glancing collisions with the atoms of the string [51]. As φ becomes greater than

$$\psi_c = \left(\frac{2Z_1 Z_2 e^2}{dE} \right)^{1/2}, \quad (2.12)$$

where E is the particle kinetic energy, referred to as the critical angle for axial channeling, for which the distance of closest approach of the particle to the atomic string is $\sim a_s$ [66], the particle begins to approach the string so closely that it no longer remains channeled. Instead, it begins to interact dominantly with the individual atoms and is scattered away from the atomic string upon one or a few larger angle collisions, i.e., it becomes dechanneled. For $\varphi \gg \psi_c$, one can take that the particle moves randomly with respect to the atomic string. Thus, in the process of axial particle channeling, it is central that the particle interaction with each atomic string includes many consecutive small angle collisions and no collisions with the individual atoms. Under such conditions, one can use the continuum string model, i.e., assume that the particle interacts with a continuous atomic string, which is obtained after a uniform longitudinal smearing of its atoms [61–66]. In this case, it is also said that the continuum approximation is employed.

Let us take that the z axis of the reference frame is the longitudinal axis, and the x and y axes are the transverse axes, being the vertical and horizontal axes, respectively. The atomic strings are parallel to the z axis, with the median plane of the crystal coinciding with the xy plane. The thermal vibrations of the atoms of the strings are ignored. The continuum interaction potential of the particle and the i th atomic string is defined as

$$V_i(\rho_i) = \frac{1}{d} \int_{-\infty}^{+\infty} U[(\rho_i^2 + z^2)^{1/2}] dz, \quad (2.13)$$

where U is given by Eq. (2.1), $\rho_i^2 = (x - x_i)^2 + (y - y_i)^2$ is the particle-string distance squared, x and y are the transverse components of the particle position vector, x_i and y_i are the transverse coordinates of the string, and z is the longitudinal component of the particle position vector [51]. Hence, one obtains

$$V_i(\rho_i) = \frac{2Z_1Z_2e^2}{d} f_s\left(\frac{\rho_i}{a_s}\right), \quad (2.14)$$

with

$$f_s(\xi_i) = \int_0^\infty (\xi_i^2 + \zeta^2)^{-1/2} \varphi_s[(\xi_i^2 + \zeta^2)^{1/2}] d\zeta \quad (2.15)$$

and $\xi_i = \rho_i/a_s$. For Molière's atom-atom screening function, given by Eq. (2.2),

$$f_s(\xi_i) = \sum_{\ell=1}^3 \alpha_\ell K_0(\beta_\ell \xi_i), \quad (2.16)$$

where K_0 denotes the zero-order modified Bessel function of the second kind [51]. For Lindhard's atom-atom screening function, defined by Eq. (2.3),

$$f_s(\xi_i) = \ln \left(1 + \frac{C_L}{\xi_i^2} \right)^{1/2} \quad (2.17)$$

[66]. Finally, for the ZBL atom-atom screening function, determined by Eqs. (2.7) and (2.8),

$$f_s(\xi_i) = \sum_{\ell=1}^4 \gamma_{\ell} K_0(\delta_{\ell} \xi_i) \quad (2.18)$$

[51]. The particle-string continuum interaction potential can be regarded as the zero-order term of the Fourier expansion of the real (periodic) interaction potential [51]. The coefficients of the remaining terms of the real interaction potential decrease rapidly with the particle distance from the atomic string [61, 62]. The particle-crystal continuum interaction potential is the sum of the particle-string interaction potentials.

Usually, the particle velocity is such that the time of its interaction with an atom of a string is much shorter than the period of thermal vibrations of the atom [51]. This means that, in the case in question, the atomic string can be treated as stationary. Thus, the particle in fact experiences the continuum potential of the atomic string averaged over the thermal displacements of its atoms from their equilibrium positions. The resulting particle-string continuum interaction potential is obtained as it was shown by Appleton et al. [78]. In such a derivation, it is assumed that the atoms of the string vibrate independently, and that, for each translational degree of freedom, the probability of an atomic displacement is described by a Gaussian distribution function. The standard deviation of this distribution, i.e., the one-dimensional atomic thermal vibration amplitude, σ_{th} , is determined using the Debye theory of thermal vibrations [79]. The obtained thermally averaged interaction potential begins to deviate considerably from the corresponding static interaction potential when the particle distance from the atomic string becomes comparable to σ_{th} [51].

If the particle energy losses in its collisions with the crystal's nuclei and electrons, i.e., its nuclear and electronic energy losses, can be neglected, the longitudinal component of the particle velocity vector can be treated as constant. Thus, if the continuum string model is used, only the transverse particle motion needs to be treated.

Let us assume that the process of axial ion channeling we are interested in can be accurately described using classical mechanics and the continuum string model. A computer simulation of the process requires that we select: (i) an ion-atom interaction potential, among those given in Sect. 2.2, (ii) a one-dimensional atomic thermal vibration amplitude, (iii) an adequate expression for the ion nuclear energy loss rate, (iv) adequate expressions for the ion electronic energy loss rate and the rate of the corresponding uncertainty of the ion scattering angle, and (v) an

uncertainty of the ion incident angle, corresponding to the incident ion beam divergence [80].

Then, the transverse components of the initial ion position vector, in the entrance plane of the crystal, are chosen from a (two-dimensional) uniform distribution over the region of a crystal channel. The choosing is done randomly, by a random number generator, or uniformly. In doing this, we avoid the initial position vectors lying within the small circular regions around the atomic strings defining the channel since the resulting channeling process would obviously include larger angle collisions. These regions are limited by the ion-atom screening length, a_s . Upon that, the transverse components of the initial ion velocity vectors are chosen from a two-dimensional Gaussian distribution defined by the selected uncertainty of the ion incident angle. This choosing is also performed randomly or uniformly.

With the initial conditions for the ion propagation along the channel defined, its equations of motion in the transverse plane are solved, and its trajectory is obtained. At the end of each step of the calculation, the longitudinal component of the ion velocity vector is changed on the basis of the calculated ion nuclear and electronic energy losses during the step, whereas the transverse components of the ion velocity vector are randomly changed on the basis of the calculated uncertainties of the components of the ion scattering angle during the step. In most cases, the nuclear energy loss can be disregarded [66].

When the ion equations of motion in the transverse plane are solved, one can calculate the transverse components of the final ion position vector, in the exit plane of the crystal, and the components of the final ion channeling angle, i.e., the ion transmission angle. The spatial and angular distributions of transmitted ions are generated with a sufficiently large number of incident ions. If the transverse components of the initial ion position and velocity vectors are chosen randomly, one says that the distributions are obtained using the Monte Carlo method.

The spatial and angular distributions of transmitted ions generated following such a computer simulation approach will be shown and analyzed in Chaps. 3, 4 and 5. In all these cases, the nuclear energy loss will be neglected [66].

2.3.3 *Binary Collision Model*

In the binary collision model of ion channeling in a crystal, the crystal is treated as real, i.e., a three-dimensional symmetric arrangement of atoms. It is assumed that the ion has only one important interaction at a time as it moves through the crystal. In each collision, the ion scattering angle is computed using the momentum approximation [61, 62], or this is done by numerical integration. Two computer simulation procedures for using this model have been developed. In both of them, it is assumed that the condition for application of classical mechanics is satisfied. In the former procedure [59, 60], the unit cell of the crystal and its translational properties are defined, and the computer code is left to search and find the crystal's atom with which the next ion interaction will happen. In the latter procedure

[81, 82], the crystal is viewed as composed of the symmetrically arranged atomic strings, consisting of the equally displaced atoms. In this case, the next important ion collision occurs with the next atom of the atomic string currently involved in the channeling process, referred to as the current central string. The critical moment in the calculation is the one at which the attention is transferred from the current central atomic string to the next one. The latter procedure has proven to be more efficient than the former one [82]. Often, it is necessary to include the contributions of the surrounding atomic strings to the ion scattering angle. This is done by treating these atomic strings as continuous, i.e., employing the continuum approximation.

In a computer simulation going on according to the above-described latter procedure, the ion trajectory is a straight line between the equidistant transverse planes with the spacing equal to the distance between the atoms of the current central atomic string. In each of these planes, the components of the ion position and velocity vectors are updated on the basis of the outcomes of the four events [83]: (i) the ion collision with the nearest atom of the string, being a screened nucleus, taking into account its thermal displacement from the equilibrium position, (ii) the ion scattering from the surrounding atomic strings governed by their thermally averaged continuum interaction potentials, (iii) the ion energy loss due to its collision with the nearest screened nucleus, and (iv) the ion energy loss and the changes of its scattering angle due to its collisions with the crystal's electrons. As it has been said in the previous subsection, in most cases, the nuclear energy loss can be neglected [66]. If the updated ion velocity vector makes a large angle with the atomic string, it is assumed that the ion motion has become random. For the ion-atom interaction potential, one of those mentioned in Sect. 2.2 is selected.

The initial conditions for the ion motion along the channel are chosen in the same way as in the computer simulation employing the continuum string model, which has been described in the previous subsection. One may also include in the calculations the mosaic spread of the crystal, the disorder of the crystal surface, the correlations of the thermal vibrations of the adjacent atoms of the current central string, and the changes of the ion charge during the propagation.

The output quantities in such a computer simulation code may include: (i) the probability that an ion experiences a sufficiently close encounter with a crystal's atom to induce an event such as a large angle scattering, an energetic atom recoiling, or a nuclear reaction, (ii) the relative number of recoiled atoms having the kinetic energy above a certain value, and (iii) the spatial and angular distributions of transmitted ions. Two such codes that have been used most so far are the LAROSE code, made by Barrett [81, 82], and the FLUX code, developed by Smulders and Boerma [83]. In Subsect. 3.1.2, we shall present and analyze the angular distributions generated with the former code.

Rainbows in Channeling of Charged Particles in Crystals
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