The phase effect of electronic stopping power*

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Abstract A corrective factor ($\psi(E,\rho) \leq 1$) dependent on ion energy and mass density of material for energy loss has been introduced into Bethe-Bloch formula, so that the energy deposition process of fast ion penetrating through the allotropic solid films are well discussed with the two-component assumption. An analysis expression of electronic stopping power for different phase structures has been derived from the contribution of "valence" and "core" electrons. The two thirds of inelastic scattering arisen from valence electron was revealed by comparing the theoretical calculation and experimental results on both random and oriented lattice site. The corrective factor representative to the role of inner electrons increases with the projectile energy but decreases with mass density of solids.

Keywords ELPU (Energy loss per unit length), Electronic stopping power, Valence electron gas, Energetic ion

1 Introduction

When the charged particle with energy of $E_0(keV \leq E_0 \leq$MeV/u) passes through the projected range, ds, in solids, it will lose its energy by inelastic collision with target atom and cause the host atom to be excited, ionized and displaced. The amount $(\mathrm{d}s/\cos\theta)(\mathrm{d}E/\mathrm{d}x)$ of ion energy is deposited on the path, which is not only correlative to the energy and effective charge of projectiles and the angle of ion incidence but also associated with the mean ionization energy, electronic density, elemental composition and solid structure of target.[1] In the present, basing on the previous experimental results[2,3], the physical mechanism of energy deposition for proton passing through the three kinds of silicon films has been tentatively discussed.

2 Theoretical method

As the energetic ion goes into the solid medium, the elastic and inelastic collisions between the projectile and the target atom will occur. Under Coulomb interaction the electrons at different states are excitd or ionized by the exchange of momentum and energy. Therefore, the incident ion transfers part of its energy to the target electrons. The collective excitation in plasma form and the individual ionization in independent-particle form of these electrons are closely relative to the probability of obtaining the energy from ion beam. Although the binding and ionization energy of 'core' or 'valence' electrons is independent on the atomic arrangement of solids, the energy loss of ion is closely associated with the arrangement of atoms in solids due to the correlation of the ionization of valence electron with atomic match. The contribution of solid electron to the energy loss was usually divided into two parts: one is from valence electrons; the other is from inner shell electrons.[4,5]

For the amorphous solid, the electronic stopping power of ion in Bethe-Bloch energy region can be expressed by

$$-(\mathrm{d}E/\mathrm{d}x)_{random} = -(\mathrm{d}E/\mathrm{d}x)_{val} + [4\pi Z_1^2 e^4/(m_e v^2)] \cdot N \sum_i Z_i f_i \cdot \ln(m_e v^2/E_i) \qquad (1)$$

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where Z_1 is the charge state of projectile, N the atomic density of target; e the electron charge; m_e the static mass of electron; f_i the strength of harmonic oscillator; E_i the ionization energy of electron; v the velocity of incident projectile; and Z_i the electron number corresponding to E_i and f_i. The first term in Eq.(1) arises from the valence electrons. The second one is the summation of individual inner shell electron's contributions.

When 1.0 MeV proton moving in Si, the energy loss per unit length (ELPU) from inner shell electron of silicon atom can be estimated as: at K shell, Z_K=2, E_K=1845 eV, the value of ELPU is about 1.43 eV/nm; at L shell, Z_L=8, the mean ionization energy E_L=115.7 eV (i.e. E_{L_1}= 148.7 eV, E_{L_2,L_3}=99.2 eV), the value of ELPU is about 12.6eV/nm. While the electronic stopping power of 1.0 MeV proton in random silicon is 42 eV/nm[6], it can be estimated that the 3.4% and 30% of ELPU are from K shell and L shell electrons, respectively. Thus the 66% of ELPU comes from the valence electrons, in other words, the two thirds of ELPU is caused by the 'outer shell' electrons in amorphous silicon.

In the same way, as the charged particle moves in long-range order solids (single crystal), the stopping action caused by solid electrons to the projectile into axial or plane channels can also be expressed in the sum of the excitation and ionization effects from 'outer' and 'inner' shell electrons[7]as following

$$-(\mathrm{d}E/\mathrm{d}x)_{ch} = [4\pi Z_1^2 e^4/(m_e v^2)] \cdot N\left\{Z_{val} \cdot \ln(v/v_f) + Z_{loc} \cdot \ln[2m_e v v_f/(h\nu_p)]\right\} + [2Z_1^2 e^4/(m_e v^2)] \cdot [n_0 f/(b^2 d)] \quad (2)$$

In Eq.(2), the collective excitation of valence electrons and the individual excitation of local electrons are given in first term, and the action of inner shell electrons is in second part. Z_{val} and Z_{loc} are the number of valence and local electrons acting on the projectiles in the channels, respectively. While NZ_{val} and NZ_{loc} represent the effective electron density for producing the plasma- and individual-excitation, respectively. The other parameters are defined as: $v_f = (h/2\pi m_e)(3\pi^2 n_e)^{1/3}$ is Fermi velocity; $\omega_p = 2\pi\nu_p = 2\pi(4\pi n_e e^2/m_e)^{1/2}$ is plasma frequency; h is Planck constant; n_e is the target electron density; n_0 is the closer axial number forming the ion channel; f is the number of inner shell electron contributing to the energy loss; b is the axial channel radius; and d is the atomic separation along the channel orientation. Simply along the orientation of Si(110), b=0.145 nm, the energy loss contributed from the inner shell electrons of silicon atom is about 1~0.5 eV/nm for the proton energy of 0.5~1.0 MeV. However, according to the equipartition principle and the experimental as well as theoretical results[8,9], the energy loss of proton into the channel of Si is approximately half of that in the amorphous Si, i.e. 32~21 eV/nm. In this channeling direction in Si, therefore, the energy loss of inner shell electron to proton is below 3.0% and mainly arisen from L subshell.

Generally, the Fermi velocity of valence electron gas in Eq.(2) can be simplified to be $v_f = (\mathrm{h}/2\pi m_e)(3\pi^2 N Z_{val})^{1/3} \approx 3.6(NZ_{val})^{1/3} \times 10^8$cm/s, and the plasmon energy is about $\mathrm{h}\nu_p \approx 37(NZ_{val})^{1/2}$ eV. When the effective electron density value $[\mathrm{n}_e^*(r)]$ for ion-atom system under IPM potential[10] was used and the contribution of inner shell electron to the energy loss was neglected in evaluating the local electron distribution, then the number of local electron was calculated to be $Z_{loc} \approx 4.0$ by replacing the NZ_{val} with $n_e^*(r = a_0, a_0$ the Bohr radius), and the Z_{val}=4.0. Therefore, the four valence electrons of Si atom are crosswise distributed in a channel of crystal and act as the plasma excitation and individual scattering. The most energy of the projectile has been deposited on these conductive electrons.

For those solid-phase structure materials with different mass density but being composed of the same elemental composition, there is not exactly crystallographic feature definition to classify them as crystalline, polycrystalline

or amorphous states. In this case, the allotropic effect on electronic stopping power for light ions (such as H,He) in silicon and carbon had been verified by previous works[11~13], and the similar phenomenon was also observed in this investigation. To our knowledge, there is no proper theory to describe the effect, and we could not simply apply the general formula as Eqs.(1,2) to the allotropic solids, because the projectiles in the solid, actually, move neither in random nor in oriented directions into the long-range or short-range order lattice sites. For the purpose of elucidating the solid phase effect of electronic stopping power, a corrective factor, $\psi(E,\rho)$, which is correlative with mass density, was introduced in the summation formula of energy loss resulted from "valence" and "core" electrons.

$$-(\mathrm{d}E/\mathrm{d}x)_{all} = (\mathrm{d}E/\mathrm{d}x)_{val} - \psi(E,\rho)(\mathrm{d}E/\mathrm{d}x)_{core} = [4\pi Z_1^2 e^4/(m_e v^2)] \cdot (NZ_{val}) \cdot \ln[2m_e v^2/(h\nu_p)]$$

$$+\psi(E,\rho) \cdot [4\pi Z_1^2 e^4/(m_e v^2)] \cdot N \sum_i Z_i f_i \cdot \ln(m_e v^2/E_i) \quad (3)$$

The Eq.(3) can be changed to the specific energy loss deposited on per unit area-mass density as following

$$-(1/\rho)(\mathrm{d}E/\mathrm{d}x)_{all} = [4\pi Z_1^2 e^4/(m_e v^2)] \cdot (N_0/A) \cdot \Big\{ Z_{val} \cdot \ln[2m_e v^2/(h\nu_p)]$$

$$+\psi(E,\rho) \cdot \sum_i Z_i f_i \cdot \ln(m_e v^2/E_i) \Big\} \quad (4)$$

where ρ is volume density (g/cm^3) of target, A is atomic mass, N_0 is Avogadro's number, Z_{val} is the number of valence electron corresponding to the plasma, and $\psi\ (E,\rho)$ is unitless factor relative to projectile energy and volume density of target. The strength of harmonic oscillator of inner shell electron in Bloch equation is related to the number of subshell electron (n_i), approximately defined as $f_i \sim n_i/Z_i$, and the plasma energy of collective excitation is about $h\nu_p$=31.05 eV.

3 Results and discussions

In order to well-understand the stopping mechanism of target electrons to energetic ion going into allotropic solids, a corrective factor has been introduced in formula (3) or (4), which is only linked to the "core" electrons and dependent on the ion energy and atomic density of target. The estimation of this factor for proton in silicon can be derived from the experimental results[3] and Eq.(4). The typical dependence of the factor vs area-mass density and ion energy is shown in Fig.1. With the increase in area-mass density the factor decreases, which means that the interaction of inner shell electron with projectile is decreased when atomic network of Si films is in short-range order. However, the factor increases with the increase in incident ion energy in Bethe-Bloch energy region. A plausible interpretation for the appearance of this trend is that the inelastic collision probability between the energetic ion and closed atomic core has been increased as the projectile has the ability to polarize the atom. Further study, we have found that the factor which can reflect the inner shell electron behavior has a form in $\psi(E,\rho) = E^{\alpha}\rho^{\beta}$ ($\alpha > 1, \beta < 1$).

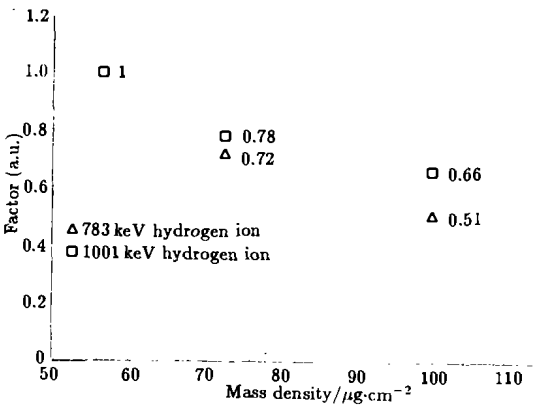

Fig.1 The dependence of the corrective factor on area-mass density of Si films and proton energy

As just mentioned above for the two-component assumption of electronic stopping power for light ions in solids, the valence electron gas in solids plays a dominant role for stopping the projectiles. On the other side, the change of the bonded atoms and the electronic states in different mass density solids will affect the charge exchange of moving projectile with target electrons, resulting in the variation of charge screening of the projectile and the polarization and ionization of inner shell electrons. These possible processes should lead to the difference of single-inelastic Coulomb scattering with atomic structures. So that the probability of approaching collisions in dense solids are statistically decreased.

In summary, under the two-component assumption of electronic stopping power, the essential role of core electrons in allotropic solids has been discussed by introducing a corrective factor dependent on the ion energy and mass density. The allotropic effect appeared in the energy absorption experiments has been explained. The fast charged particle, through the excitation and ionization interaction with target atom, transfers the most of its energy to the valence electrons (or plasma), only a small amount of its energy is shifted to inner shell electrons by Coulomb scattering. The more the atomic density of allotropic solids, the more the energy obtained by inner shell electrons, corresponding to the more energy deposition from ions.

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Over 50 papers have been published in Chinese and foreign journals, which dealed with the interaction between ion and solids, the synthesis, characterization and modification of metal, semiconductor, ceramics and insulator materials. Specific research interests presently include diamond film growth, doping of diamond by ion implantation, laser annealing of radiation damage in damond, the interaction of ion beam with carbon based materials, synthesis of the ultra-hard carbon-nitride thin films, wide band gap nitrides, carbon nanostructures and the application of advanced solid analysis techniques to advanced materials systems.