Energy straggling of light-ion beams

T. Kaneko and Y. Yamamura

Department of Applied Physics, Okayama University of Science, Ridai-cho 1-1, Okayama 700, Japan

(Received 9 September 1985)

The dielectric function method is applied to investigate the energy straggling of protons and helium ions using the atomic and the solid local-electron-density models. A partially stripped ion is treated as well as point charges. At low energies, the conventional stragglings of a proton ($\Omega^2_{\mathrm{H}^+}$) and of helium ions ($\Omega^2_{\mathrm{He}^+},\Omega^2_{\mathrm{He}^{2+}}$) due to the fluctuation in electronic excitations are proportional to the kinetic energies of the ions even when the local-electron-density models are adopted, and at high energies they approach the values predicted by Bohr. Here we do not consider the bunching term since we assume the probability of exciting an electron is small. The straggling ratio $\Omega^2_{\mathrm{He}^+}/\Omega^2_{\mathrm{H}^+}$ shows a remarkable feature that it is nearly constant up to $\sim$50 keV/amu, and increases gradually beyond this energy for solid targets. The estimation of the collisional straggling of helium-ion beams is performed using the charge-state fractions, resulting in displaying Z_2 (target atomic number) oscillations similar to those of $\Omega^2_{\mathrm{He}^+}$ and $\Omega^2_{\mathrm{He}^{2+}}$. The straggling caused by charge-state fluctuations, which enhance the Z_2 oscillations of the total straggling of helium-ion beams more sharply at the energies considered, is also estimated.

I. INTRODUCTION

The energy loss of impinging charged particles has received wide attention recently in ion-beam—material interactions, since it is a fundamental problem to investigate the composition, the depth distribution, and the location of lattice sites of implanted atoms in the host material. The energy spread around the average energy loss, i.e., the energy straggling, limits ultimately the spatial resolution of such implanted atoms. The energy straggling is caused by statistical fluctuations in the collision processes that particles are subjected to during the passage. For collisional (hereafter this term is used instead of "conventional") energy straggling, several theories are available. Bohr[1] derived the straggling formula for a particle with atomic number Z_1 and velocity v penetrating a target material with Z_2,

$$\Omega_B^2 = 4\pi Z_1^2 Z_2 e^4 N x \ , \tag{1.1}$$

in the high-velocity region, where N and x denote the number density of target atoms and the path length of the particle, respectively. The typical feature of (1.1) is that Ω_B^2 is independent of v and proportional to x. In order to apply the above formula for lower velocities, Lindhard and Scharff[2] (LS) have extended Bohr's formula by multiplying by the velocity-dependent factor

$$\Omega^2/\Omega_B^2 = \begin{cases} (\frac{1}{2})L(y) & \text{for } y \lesssim 3 \\ 1 & \text{for } y > 3 \ , \end{cases} \tag{1.2}$$

where the variable y is defined by $y=(1/Z_2)(v/v_0)^2$ (v_0 is the Bohr velocity) and $L(y)$ denotes the stopping number, which has an approximate form

$$L(y) = 1.36 y^{1/2} - 0.016 y^{3/2} \ , \tag{1.3}$$

if the Thomas-Fermi model is used. Refinements of the LS theory have been performed by Bonderup and Hvelplund,[3] who assumed the Lenz-Jensen atomic model. By adopting a more realistic electron distribution derived from the Hartree-Fock wave functions, Chu[4] obtained the crossover feature for different target atoms at the same proton and helium-ion velocities, which results in the well-known Z_2 oscillation. Apart from collisional straggling, the extra contributions coming from, e.g., target thickness variation, the "bunching" effect in atomic and molecular targets, and charge-state fluctuations have also been discussed recently,[5] since experiments supply us with straggling values greater than the ones predicted from the conventional part.

To obtain the stopping powers and the straggling, the local-electron-density models (LEDM's) are successfully used for describing the spatial target electron distributions. For an atomic target, the local electron density is easily obtained from Hartree-Fock wave functions, while for solid targets, it should be modified to include free electrons through a constant electron density in the outer region of the Wigner-Seitz cell, where the collective excitation mode is characterized by the bulk plasma frequency.[6]

The aim of this paper is to evaluate the straggling of protons and helium ions penetrating solid media, where the charge-changing contribution Ω^2_{CC} is treated as well as the conventional contribution Ω^2_{coll}. Estimation of Ω^2_{CC} has been emphasized because none of these calculations have been done systematically for various targets. The present calculation of the straggling of a single particle is based on the Lindhard-Winther (LW) theory[7] together with the LEDM's. One difficulty in estimating Ω^2_{CC} theoretically lies in the stopping cross section for a partially stripped ion (PSI). To attack the problem, the dielectric function method extended to a PSI is used with the LEDM. Moreover, a reasonable knowledge of charge-changing cross sections and charge-state fractions is necessary. In our case, for these quantities, a consistent treatment is prescribed using experimental data and theories. In Sec. II, our procedure is described, and Sec. III is assigned to present numerical results and discussions. Atomic units are used unless otherwise is stated.

II. PROCEDURE

As energetic particles pass through matter, they are subjected to successive electron capture and loss and they also excite the target in collisions with target atoms. After traversing a sufficiently long distance, the charge states of the particles reach the equilibrium state. Considering the particle transport, we write down the basic equation discussed previously as follows:[8]

$$dG(i;x,n)/dx=\sum_{j(\neq i)}[B_{ij}G(j;x,n)-B_{ji}G(i;x,n)]+\sum_{k=1}^{n}\begin{bmatrix}n\\k\end{bmatrix}G(i;x,n-k)\int_0^\infty d\omega\,P_i(\omega)\omega^k\ , \tag{2.1}$$

where $G(i;x,n)$ is defined as the contribution to the nth moment of energy loss E at path length x from the particles in charge state i at x. B_{ij} is the charge-changing probability per unit length from charge state j to i, and $P_i(\omega)d\omega$ is the probability per unit length of transfering energy ω of the particle in charge state i to the target electrons. Here we neglect the deflection of penetrating particles, namely, the contribution of elastic collisions with particles and target atoms. And we assume the probability of exciting an electron during the passage of a projectile is very small. Therefore, the "bunching" term is not introduced at this moment. In (2.1), the terms for energy transfer accompanied by charge exchange in a collision are also neglected because they are regarded as the first-order correction to the leading term. In the case where the emergent particles are detected in all charge states allowed in matter, the average energy transfer $\langle E\rangle$ and the average of the square of the energy transfer $\langle E^2\rangle$ to the matter from the particles are given by

$$\langle E\rangle=\sum_i G(i;x,1)$$

and (2.2)

$$\langle E^2\rangle=\sum_i G(i;x,2)\ ,$$

as a function of x. Then, the energy straggling of ion beams is defined as

$$\Omega^2=\langle E^2\rangle-\langle E\rangle^2\ . \tag{2.3}$$

To get the explicit form of Ω^2, as an example, for the ion beams with three dominant charge components labeled by 0, 1, and 2, the differential equation is constructed

$$d(\Omega^2)/dx=\sum_i dG(i;x,2)/dx-2\sum_i G(i;x,1)\times\sum_j dG(j;x,1)/dx\ . \tag{2.4}$$

Substituting into (2.4) the right-hand side of Eq. (2.1) with $n=2$, and using the solution of $G(i;x,1)$ $(i=0,1,2)$, we get, under the charge equilibrium condition, the following after integrating over x:

$$\begin{aligned}&\Omega^2=\Omega^2_{\rm coll}+\Omega^2_{\rm CC}\ ,\\&\Omega^2_{\rm coll}=\Omega_0^2\phi_0+\Omega_1^2\phi_1+\Omega_2^2\phi_2\ ,\\&\Omega^2_{\rm CC}=Nx(2/D)\{\alpha_t[(S_2-S_0)^2\phi_0\phi_2+(S_2-S_1)^2\phi_1\phi_2+(S_1-S_0)^2\phi_0\phi_1]\\&\qquad-(S_2-S_1)^2\phi_2A_{12}-(S_1-S_0)^2\phi_1A_{01}\}\ ,\end{aligned} \tag{2.5}$$

with

$$\begin{aligned}&\alpha_t=A_{21}+A_{12}+A_{10}+A_{01}\ ,\\&D=A_{01}A_{12}+A_{10}A_{12}+A_{10}A_{21}\ ,\\&\phi_0=A_{01}A_{12}/D,\ \ \phi_1=A_{10}A_{12}/D,\ \ \phi_2=A_{10}A_{21}/D\ ,\end{aligned} \tag{2.6}$$

where N is the number density of target atoms, and S_i and ϕ_i $(i=0,1,2)$ denote the stopping cross section and the equilibrium charge fraction, replacing $G(i;x,0)$ at a sufficiently large x, respectively. Here only the cross sections A_{ij} $(=B_{ij}/N)$ for changing the charge by ±1 are included. In (2.5), $\Omega^2_{\rm coll}$ is the so-called collisional energy straggling, resulting from the square-energy transfer to target electrons from the particles in each charge state, and this quantity has been discussed for point-charge intruders. In this paper, besides the charge state distribution (CSD) the straggling of a partially stripped ion is also treated and, therefore, the size effect of a PSI is included. It is noted here that the contribution of the elastic collisions between incident particles and target atoms is neglected. The collisional part $\Omega^2_{\rm coll}$ is straightforwardly obtained for ion beams with an arbitrary number of charge states allowed in matter, resulting in

$$\Omega^2_{\rm coll}=\sum_i\Omega_i^2\phi_i\ . \tag{2.7}$$

The additional contribution $\Omega^2_{\rm CC}$ is due to charge-state fluctuations of particles during the passage, in which the differences of the stopping cross sections play a significant role. If we set $A_{21}=0$ and $A_{12}\neq0$ in (2.5), then $\Omega^2_{\rm CC}$ reduces to the formula that has already been obtained for the case of two dominant charge components[5]

$$\begin{aligned}&\Omega^2_{\rm CC}=2(S_1-S_0)^2\phi_0\phi_1Nx/(A_{01}+A_{10})\ ,\\&\phi_0=A_{01}/(A_{01}+A_{10}),\ \ \phi_1=A_{10}/(A_{01}+A_{10})\ .\end{aligned} \tag{2.8}$$

This formula for $\Omega^2_{\rm CC}$ indicates that the charge-fluctuation effect on the straggling is reflected only via the squares of the difference in the stopping cross sections, apart from the charge-changing cross sections.

The collisional straggling of a partially stripped ion in charge state i with bound electrons is expressed in the dielectric function theory as

$$\Omega_i^2 = Nx \int_0^\infty d\omega\, \omega^2 \int_{\omega/v}^\infty dk (2/\pi k v^2) \,|\, Z_1 - \rho_i(k) \,|^2 \,\mathrm{Im}[1/\epsilon(k,\omega)] \;, \tag{2.9}$$

where $\epsilon(k,\omega)$ is the dielectric function of a solid and $\rho_i(k)$ is defined by $\rho_i(k) = \int \rho_i(r) \exp(i\mathbf{k}\cdot\mathbf{r}) d^3r$. The spatial distribution of the bound electrons, $\rho_i(r)$, is assumed to be spherically symmetric so that its Fourier transform $\rho_i(k)$ has the same symmetry. In contrast with the stopping cross sections, the collective excitation branch contributes negligibly in comparison with the pair excitation branch.[3] Therefore, we consider only the electron-hole pair excitation in the spectrum of the target excitation. In order to estimate Ω_i^2, local-electron-density models are used. The local electron density of neutral atoms, which is assumed spherically symmetric, is given by statistical distributions, e.g., the Thomas-Fermi or the Lenz-Jensen model, and in a more realistic way, by Hartree-Fock wave functions.[9] On the other hand, in solid targets, the local electron density $\rho(r)$ should be modified in such a way that $\rho(r)$ includes the free electrons as well as the core electrons. For simple metals (or semiconductors), this modification is rather easily performed by introducing a constant electron density n_0 in the outer region of the Wigner-Seitz cell, where n_0 is characterized by the frequency of the collective motion, i.e., the bulk plasma frequency ω_p through $\omega_p = (4\pi n_0)^{1/2}$ in atomic units. More detail descriptions of and comments on the present LEDM are given in Refs. 6 and 10.

One important point in the paper is to include the charge-state distribution (CSD) in the formulation. Our concern here is not to determine the CSD from the first principles, but to use the CSD that is inferred consistently from the stopping cross sections for ion beams. In order to arrange the experimental data of charge-state fractions, ϕ's, the following expression for the ratio, ϕ_i/ϕ_j, is successfully used:

$$\phi_i/\phi_j = AE^{-B} \;, \tag{2.10}$$

where E is the energy of the projectile, and A and B are constants independent of E. In spite of its simple form, the predicted CSD from Eq. (2.10) using both the data of the stopping cross sections for helium-ion beams and the calculation of those for helium ions in each charge state provides a good agreement with the CSD data and the resulting average charge.[8] From the discussion of the linear screening effect on external charges in solids,[11] a hydrogen atom, H^0, and a helium one, He^0, cannot generally exist inside solids except in alkali metals. Therefore, for helium-ion beams passing through solids, formula (2.8) for two-charge-component cases is sufficient to estimate the charge-changing contribution Ω_{CC}^2. And Ω_{CC}^2 can be neglected for a proton beam in such solids. On the other hand, in gaseous targets, neutral particles H^0 and He^0 are able to exist during the passage so that the formula Ω_{CC}^2 in Eq. (2.5) for three charge components will be used in principle for helium-ion beams. In addition, this formula will

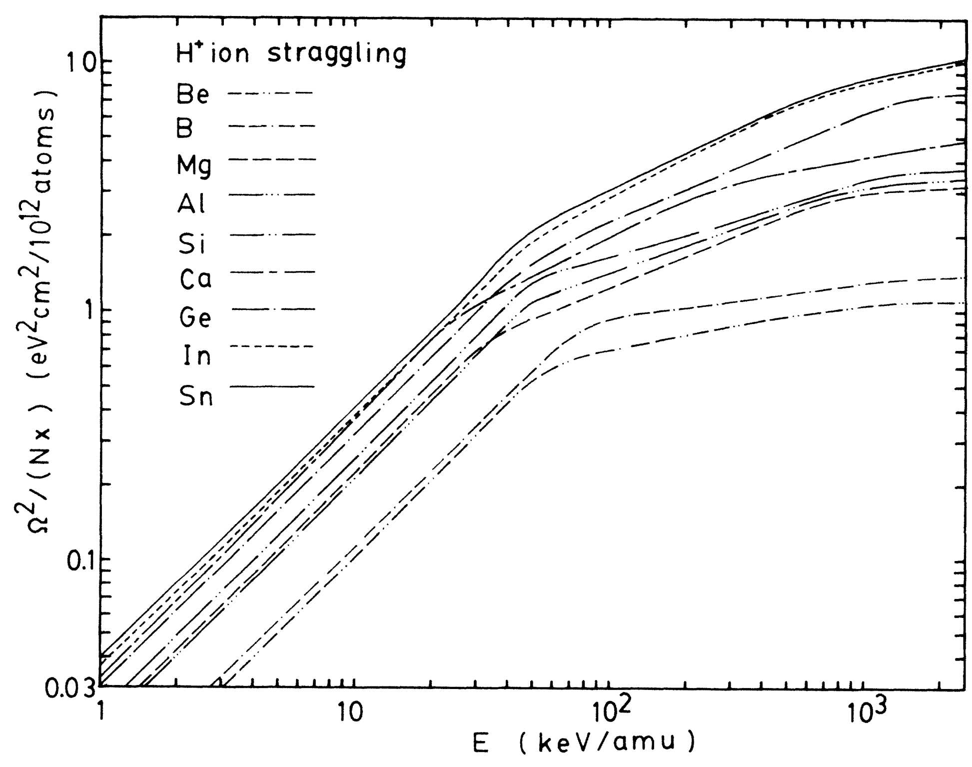

FIG. 1. The collisional straggling of a H^+ ion incident on solids calculated from Eq. (2.9) with the solid LEDM.

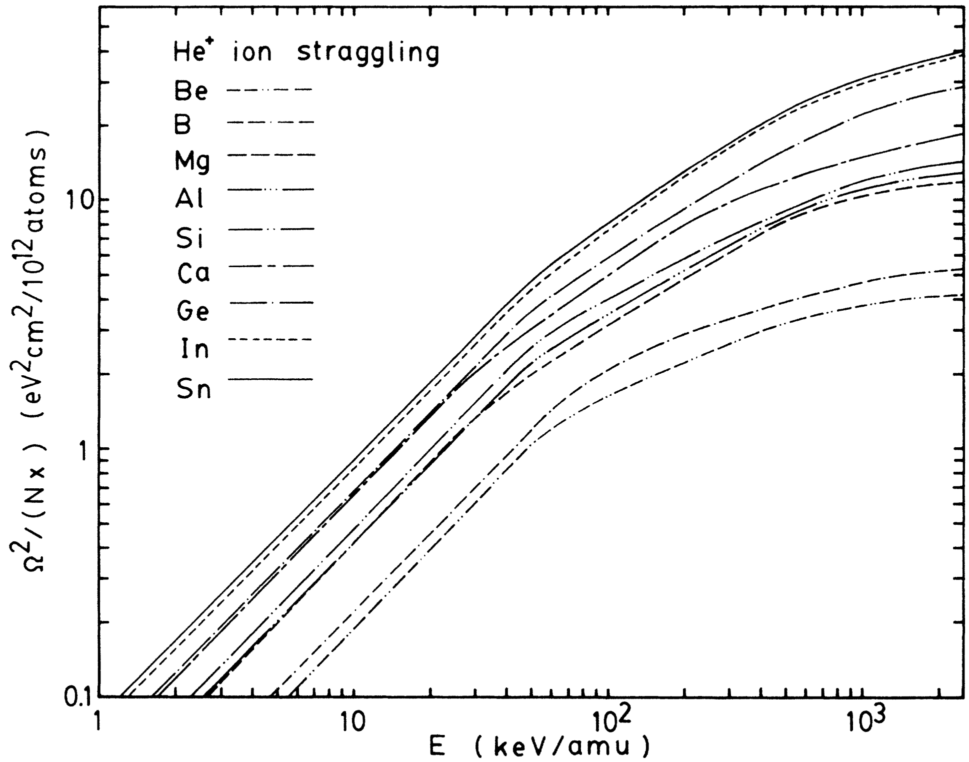

FIG. 2. The collisional straggling of a He^+ ion incident on solids calculated from Eq. (2.9) with the solid LEDM.

be applicable to Ω_{CC}^2 of heavy ion beams, of course, when the charge components are limited to two or three.

III. NUMERICAL RESULTS AND DISCUSSIONS

Based on the local electron density models, the calculated collisional stragglings of a proton and a singly charged helium ion are shown in Figs. 1 and 2, respectively, where the number of free electrons are assumed to be two for Be, Mg, Ca, three for B, Al, In, and four for Si, Sn. From the LW theory, Ω_{coll}^2 is proportional to the kinetic energy E in the low-E region. This relation is also valid even when the LEDM is adopted here, and, in addition, whether it is a point charge (H^+) or a partially stripped ion (He^+). At

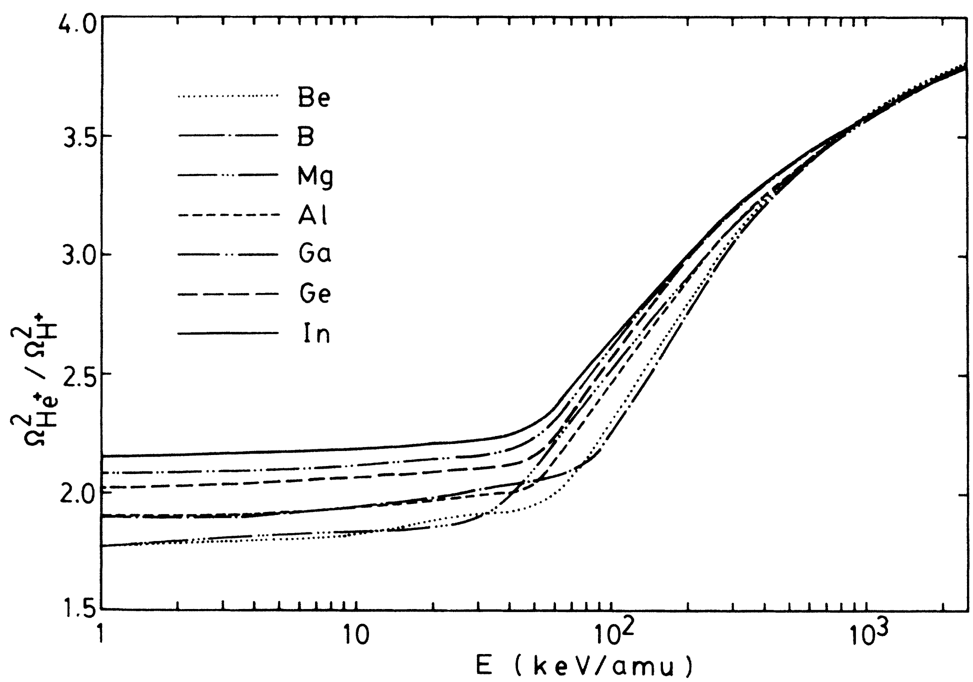

FIG. 3. The ratio of the collisional stragglings, $\Omega_{He^+}^2/\Omega_{H^+}^2$, calculated for solids.

low energies, the free electrons contribute a major part of Ω^2_{coll} especially for light elements, where they produce more than 80% of the total Ω^2_{coll} of a H^+ ion up to ~ 100 keV. With increasing energy, the core electrons contribute dominantly and, in the extreme case, the binding effect can be neglected so that only the number of electrons is important to estimate Ω^2_{coll}. Then the Bohr formula becomes valid. In Fig. 3, the ratio of the stragglings of a He^+ ($\Omega^2_{He^+}$) and of a H^+ ($\Omega^2_{H^+}$) ion are illustrated with respect to E. We note here the remarkable feature that the ratio $\Omega^2_{He^+}/\Omega^2_{H^+}$ is nearly constant up to ~ 50 keV/amu and increases gradually beyond this energy until it reaches 4.0 asymptotically. This is explained in the same manner as the stopping ratio S_{He^+}/S_{H^+} by the fact that at low energies, the momentum transfer k ranging from 0 to $\sim 2k_F$ (k_F is the Fermi wave number) contributes dominantly to the straggling. And the external charge of a He^+ ion in Fourier space is not very much different from 1.0 in the region $0 \lesssim k \lesssim 2k_F$. On the other hand, at high energies the contributing momentum-transfer region in the k-ω plane depends on the particle energy, where the electrons in the matter tends to be scattered by the nucleus rather than by the bound electron in a He^+ ion. Then, the external charge in the Fourier space, i.e., $2-\rho_i(k)$, approaches 2.0 as a result.

In Figs. 4 and 5, the Z_2 dependence of the stragglings are illustrated for helium ions with 200 and 400 keV/amu energies, where the charge-exchange contributions are estimated as well as the collisional ones of a He^+ and a He^{2+} ($\Omega^2_{He^{2+}}$) ion using both the atomic and the solid LEDM's. In the solid LEDM, the observed plasma frequencies[12] are used to deduce the number of free electrons

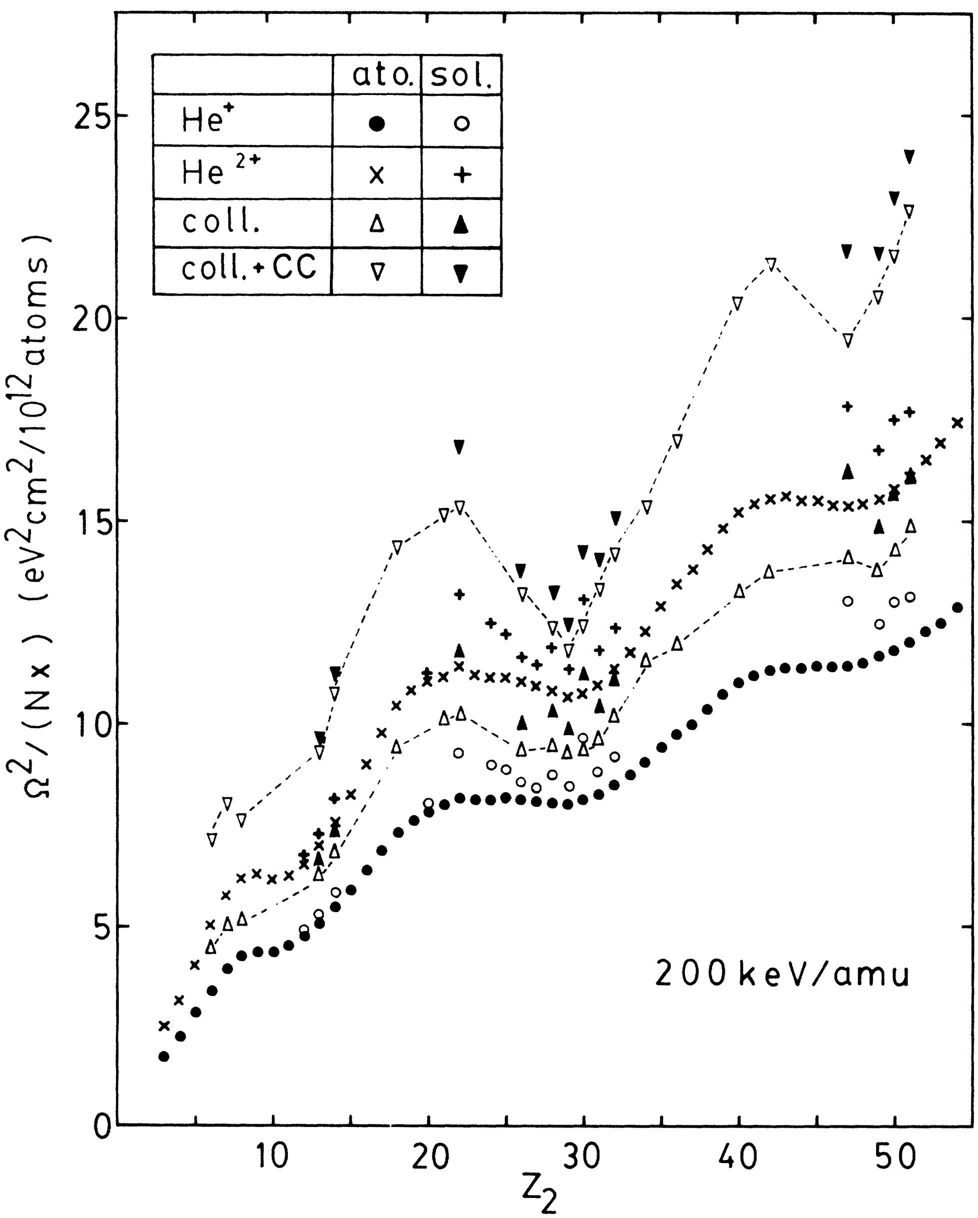

FIG. 4. The collisional stragglings of a He^+ and a He^{2+}, and the collisional and the charge-changing stragglings of a helium-ion beam at 200 keV/amu with respect to Z_2, calculated in both the atomic and the solid LEDM's.

per Wigner-Seitz cell for nonsimple metals. To be able to get Ω_{CC}^2 reasonably, accurate information of the magnitude of charge-changing and stopping cross sections with respect to Z_2 and v, is necessary, and moreover so is a consistent treatment of the cross sections and the charge-state fractions (CSF's). Our procedure is the following: the first step is to obtain the CSF from experiments,[13–15] or to predict the CSF from the stopping cross sections for helium ions.[8] For the latter case, the constants A and B in Eq. (2.10) are tabulated in Table I for some solids with the observed ω_p. As far as the stopping cross sections for a He^+ and a He^{2+} ion, i.e., S_{He^+} and $S_{He^{2+}}$, are concerned, we need partly the aid of the semiempirical data.[16] As shown previously, the LEDM is practically useful in spite of its simplicity. In order to avoid any discrepancy, we adopt the semiempirical values for the proton stopping cross section S_{H^+} so that $S_{He^{2+}}$ is obtained as $4S_{H^+}$. Furthermore, as the stopping ratio γ $(=S_{He^+}/S_{H^+})$ is computed by the theory,[8,10] the modified S_{He^+} is straightforwardly obtained by multiplying the semiempirical S_{H^+} value by γ. It is not easy to estimate successfully the electron capture cross sections for various targets within a

TABLE I. Parameters A and B for some solids in Eq. (2.10) predicted from the stopping cross sections for helium ions and for helium-ion beams. The energy E is measured in MeV.

Element	$_{22}$Ti	$_{26}$Fe	$_{28}$Ni	$_{29}$Cu	$_{30}$Zn	$_{47}$Ag
A	0.313	0.467	0.554	0.553	0.626	0.291
B	1.88	1.18	1.77	1.62	2.47	2.25

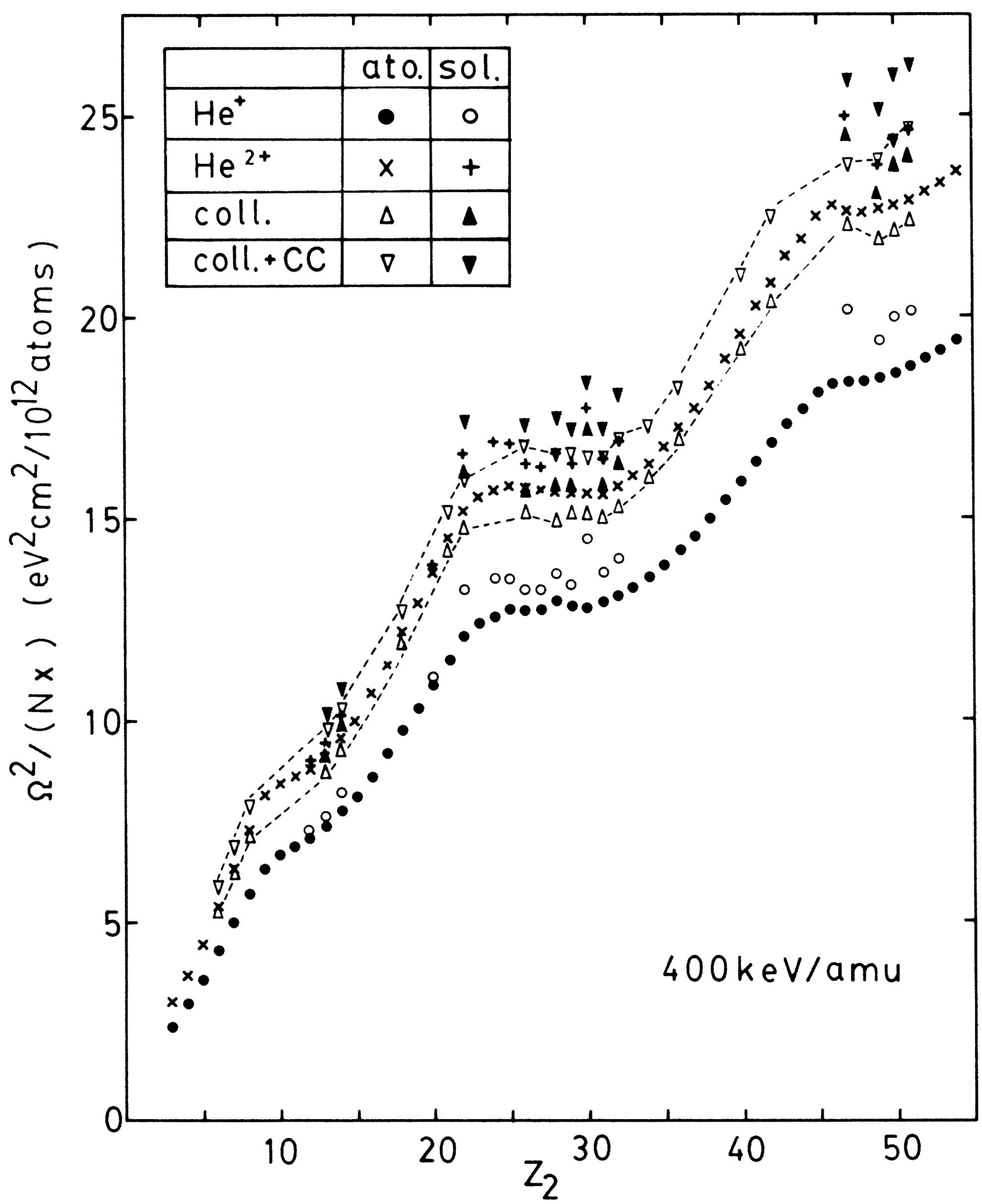

FIG. 5. Same as in Fig. 4 except for at 400 keV/amu.

reasonable order of magnitude, partly because we have to treat three-body problems at least, and partly because they sometimes display nonmonotonical behavior with respect to Z_2. Instead we adopt the idea of obtaining capture cross sections from the charge-state ratios and the loss cross sections. Recently, the unitarized impact-parameter method has been presented to obtain the electron loss cross sections,[17] and yields a good agreement with the data both in the monotonical Z_2 dependence and in the energy dependence. This idea enables us to determine the capture cross sections much more easily. The procedure stated up to here brings us a consistent evaluation of $\Omega^2_{\rm CC}$, which have been estimated explicitly only for a few target elements. In the figures we should remark the following points: the first is that the straggling of a He^+ ion shows almost the same structure as that of a He^{2+} ion with respect to Z_2, the second is that the total straggling Ω^2 $(=\Omega^2_{\rm coll}+\Omega^2_{\rm CC})$ of the helium-ion beams is greater than the straggling of a He^{2+} ion at the energies considered, and the third is that the charge-changing contributions enhance the Z_2 oscillation compared with the collisional ones. It is noted, in addition, that the solid LEDM tends to yield larger collisional, and consequently larger total stragglings than the atomic LEDM.

The charge changing part $\Omega^2_{\rm CC}$ is closely related to the width σ of the charge-state distribution, defined by $\sigma^2=\sum_q (q-\bar{q})^2\phi_q$ where the ϕ_q's and $\bar{q}$ denote the charge-state fraction and the average charge, respectively. In the case where two charge components are dominant, we obtain

$$\sigma^2=\phi_0\phi_1\ , \tag{3.1}$$

which is actually contained in Eq. (2.8). Therefore, $\Omega^2_{\rm CC}$ is expected to contribute greatly at energies where the charge distribution is broad as well as where the stopping cross sections are large.

Figure 6 shows the comparison of the calculated results with the experimental data[18–22] for an Ag target where $\omega_p=23$ eV. Below 100 keV/amu, the total straggling of the helium beam is less than that of a He^{2+} ion, since a partially stripped ion He^+ can exist in the He-ion beam inside Ag, and also since $\Omega^2_{\rm CC}$ is small. In the specific energy range of $100<E<500$ keV/amu, the charge-fluctuation part is the main contribution, enhancing the total straggling and decreasing the discrepancy. Our total straggling curve also improves the LS curve at low energies, and explains the experimental data at least qualitatively. When we see the large differences, e.g., as much as a factor of 2, between measurements performed by different research groups with the same target material, further measurements especially by the transmission technique are necessary to get accurate and universal behavior. In this paper we concentrate only on the charge-state fluctuation effect on the straggling among the extra contributions in order to examine it systematically.

In conclusion, based on our procedure, the straggling of helium ions and of helium-ion beams are estimated using LEDM's, where a partially stripped ion as well as point charges were treated. Here we rely on the possibility of defining charge states in a solid. This subject may be controversial. For example, it is claimed by the authors[5] that charge states are only well defined in gases. However, our opinion is different, based on the following points: One is the observation of the Z_2 oscillation in the average charge of MeV helium-ion beams. This effect is well predicted by electron capture and loss cross sections for a single ion in the charge states considered,[13,23,24] the reason for which is that the core electrons rather than the free electrons in a solid play a dominant role in charge-changing processes

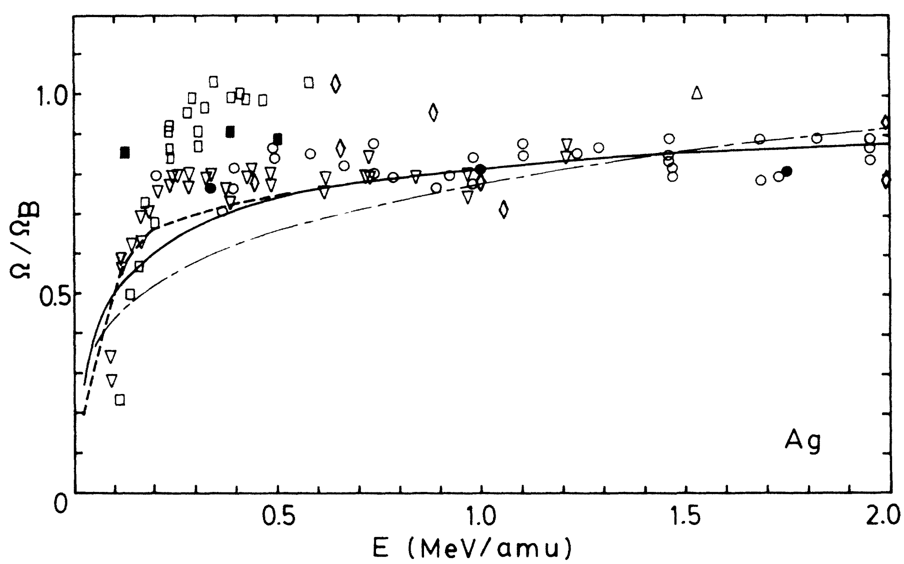

FIG. 6. Comparison of the calculated stragglings with the experimental ones in Ag target: the collisional straggling of a point charge in the solid LEDM (——), the total straggling of helium-ion beams (— — —), and the data [○-protons, ▽-deuterons, □-alpha particles (Ref. 18), ●-protons (Ref. 19), ◇-protons (Ref. 20), △-averaged alpha particles (Ref. 21), and ■-alpha particle (Ref. 22)]. The Lindhard-Scharff (LS) result is denoted by –·–·–.

except in the low-energy region. Moreover, the above situation is also supported for a proton.[25] The other point is the analysis of the stopping cross sections performed in Ref. 8 based on (2.1). The experimental stopping data for helium around the stopping maximum are located between the theoretical curve for a He^+ ion and that for a He^{2+} ion. This implies that it is important to take into account the charge states in estimating the stopping cross sections there. These situations mean that charge states are also well defined in a solid.

The ratio of the collisional straggling, $\Omega^2_{He^+}/\Omega^2_{H^+}$, shows remarkable energy dependence for various solids, providing curves similar to those of the stopping ratio, S_{He^+}/S_{H^+}. This reflects the size effect of a He^+ ion. As to the charge-changing contribution to the straggling, the formula for ion beams with three charge components is derived. The charge-changing contribution is evaluated through the careful consideration of the stopping data and the charge-changing cross sections, resulting in the enhanced Z_2 oscillations. Compared with the straggling data, the collisional contributions of helium-ion beams are smaller, even if we take into account the relatively large experimental errors. Based on the linear screening, the formula (2.8) of Ω^2_{CC} for two-charge-component cases is valid for solid targets. The charge-changing part, therefore, decreases the discrepancy a little bit. It is pointed out that the bunching term is important at low energies for atomic and molecular targets, where the probability for a projectile to excite an electron is not so small.[5] In our case, however, this term is neglected because we concentrate on the analysis based on the local electron density models which assume a small probability of exciting an electron. To get a more satisfactory agreement, the other extra contributions will be examined quantitatively in the future.

ACKNOWLEDGMENTS

We are indebted to Professor Y. H. Ohtsuki at Waseda University for useful discussions. This work is financially supported by Special Project Research on Ion Beam Interaction with Solids from the Ministry of Education, Science and Culture, Japan, and in part by Institute of Plasma Physics at Nagoya University, Japan.

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