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Charge-state distribution of MeV He ions specularly reflected from a SnTe(001) surface

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The charge exchange process of fast He ions at solid surfaces has been investigated by the specular reflection of MeV He ions from a clean (001) surface of a SnTe single crystal. A large difference is observed between the charge-state distribution of specularly reflected ions and that of ions transmitted through a self-supporting foil. The observed results indicate that the charge-state distribution of the specularly reflected ions is determined by a charge exchange process with the valence electrons in the tail of the electron distribution at the solid surface, whereas that of the foil-transmitted ions is mainly determined inside the foil. The electron-capture cross section of He^{2+} ions in collisions with valence electrons near the surface of SnTe(001) is estimated from the observed charge-state distributions. It is shown that the surface plays an important role in the charge exchange process of MeV He ions for takeoff angles of the order of 10 mrad.

I. INTRODUCTION

Charge exchange phenomena of energetic ions passing through solid targets have received considerable attention in studies of ion-solid interactions. Since the pioneering work of Phillips,[1] which showed that the exit surface region plays an important role in the neutralization of protons passing through solid foils, much of the interest has been concentrated on charge exchange between ions and the solid surface.[2-8] In the low-energy region, it is widely accepted that ions capture and/or lose electrons at the surface by the mechanisms of Auger neutralization, resonance neutralization, and resonance ionization.[9] However, the role of the surface is not so clear in the high-energy region. The capture probabilities of electrons from the tail of the conduction-electron distribution outside of the surface have been calculated by several authors,[2,3] and agree well with the observed neutralization probabilities of protons passing through solids. However, Cross showed that the charge-state distributions can be understood in terms of electron-capture and -loss processes within the solid.[10] He concluded that the surface plays no special role.

Many experimental studies have been performed in order to reveal the role of the surface,[4-8] although only a few studies have been performed with a well-characterized surface. An excellent study on the neutralization of medium-energy He ions backscattered from clean and cesium-covered Si(100)-(2×1) surfaces was performed by Haight *et al.*[7] The ions scattered from surface atoms and those from bulk atoms were separated using surface-sensitive channeling techniques. A depth resolution of 0.7 nm, which means 5 atomic layers, was attained. They found that the ion fraction did not depend on either the ion-takeoff angle or the depth from which the ion was scattered, but that it depends on the work function. They concluded from these experimental results that the ion fractions are determined at the solid surface, during the ion's outward path; the ion retains no memory of the path within the solid. In their experimental situation, the backscattered ion experienced a hard collision with a target atom. The hard collision is also expected to cause charge exchange because the ion penetrates inside the atom where the electron density is large. So the charge-state distributions of the scattered ions are determined by both the charge exchange at the surface and during the hard collision, even if the ions are scattered by the surface atoms. In order to more clearly reveal the role of the surface in charge exchange, an experiment is desired for the situation where the ion interacts only with the surface.

In the present paper we report on a study of the charge exchange of MeV He ions specularly reflected from a clean (001) surface of SnTe single crystals at glancing-angle incidence. The specularly reflected ions do not penetrate through the surface atomic layer, and so they interact only with the surface.[11-14]

II. EXPERIMENTAL PROCEDURE

The details of the experimental apparatus were described elsewhere.[11] So only a brief description is given here. The single crystal of SnTe(001) was prepared by epitaxial growth *in situ* by vacuum evaporation on a cleaved KCl(001) surface in a scattering chamber with a base pressure of 10^{-10} Torr. A well-defined (1×1) pattern from the surface was observed by reflection high-energy electron diffraction. A beam of He ions from the 4-MV Van de Graaff accelerator of Kyoto University was collimated by apertures to 0.1×0.1 mm^2 and to a divergence angle less than 0.5 mrad. The ions scattered at an angle θ_s in the plane containing the incident beam and the normal to the surface were chosen by a movable aperture. The acceptance angle of this aperture was 0.9 mrad for the scattered ions. The ions passing through the aperture were resolved into their charge states by a magnetic analyzer, and the energy spectrum of the ions of each charge state was measured by a solid-state detector. The neutral He ions were not measured, but the neutral fraction was determined to be less than 1% for the present experimental conditions. The ratio of the fraction of

He^+ ions, F_1, to that of He^{2+} ions, F_2, was measured as follows: The magnetic field of the analyzer was changed periodically so that the He^+ ions and He^{2+} ions reach the detector alternately. The energy spectra of He^+ ions and He^{2+} ions were registered in two memory groups of a multichannel analyzer separately. This was done to eliminate the error in the ratio F_1/F_2 caused by the fluctuation of the incident beam. The typical period of change in the magnetic field was 3 s, and the measurement was performed for 100 periods.

III. EXPERIMENTAL RESULTS

When fast ions are incident on the clean SnTe(001) surface with a small glancing angle, the energy spectra of the scattered ions have an oscillatory structure.[14] An example of the energy spectra is shown in the inset of Fig. 1. The scattered ions of the first peak (the peak of highest energy) are reflected from the surface atomic plane, and others penetrated through the surface and traveled for a few wavelengths of oscillatory motion in the (001) channel before appearing from the surface.[14] Thus we can identify the ions reflected from the surface atomic plane. The fractions $F_1(\mathrm{I})$ and $F_2(\mathrm{I})$ of He^+ and He^{2+} ions, which were reflected from the surface atomic plane, were obtained from the corresponding peak areas of the first peaks of the energy spectra of He^+ and He^{2+}, respectively. Even if the incident He ions were directed towards a low-index crystallographic axis parallel to the surface (the condition of surface channeling), the energy spectra had a peak composed of the ions scattered from the surface atomic rows, and they can be distinguished from the ions which had penetrated through the surface. It must be noted that reflected ions interact with many surface atoms. So the results represent some kind of an average for Sn and Te atoms.

The ratio of the fraction of He^+ ions, $F_1(\mathrm{I})$, to that of He^{2+} ions, $F_2(\mathrm{I})$, in the beam reflected from the surface atomic layer was measured for various conditions. Figure 1 shows the ratio $[F_1(\mathrm{I})]/[F_2(\mathrm{I})]$ as a function of the

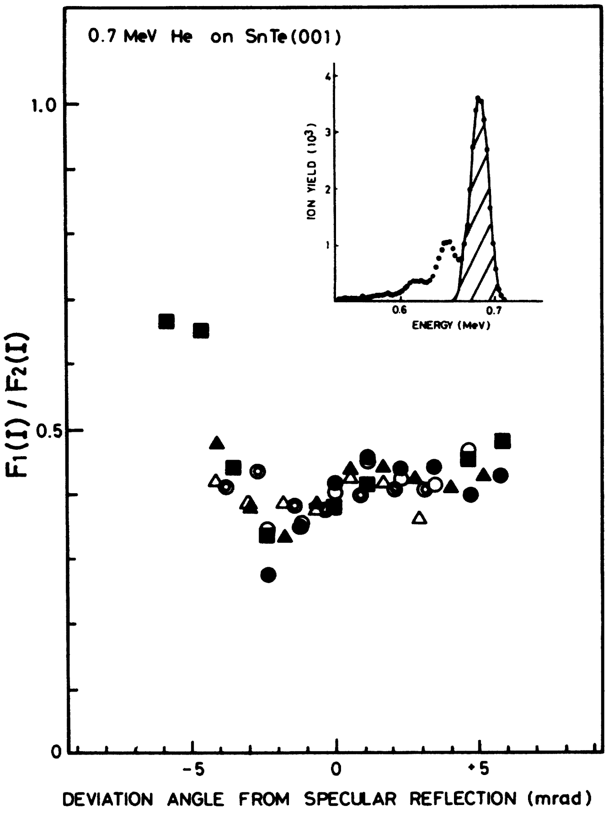

FIG. 1. The ratio of the fraction of He^+ ions, $F_1(\mathrm{I})$, to that of He^{2+} ions, $F_2(\mathrm{I})$, for the ions of first peak as a function of the deviation angle from the specular reflection. The ions of the first peak are reflected from the surface atomic plane. The ratios for 0.7-MeV He^+ incidence with glancing angles 2.9 mrad (●), 4.9 mrad (▲), 6.9 mrad (■), and 0.7-MeV He^{2+} incidence with a glancing angle 5.2 mrad (◎) are shown. The ratios for 0.7 MeV He^+ incidence with glancing angles 2.9 mrad (○) and 4.9 mrad (△) in the condition of [210] surface channeling are also shown.

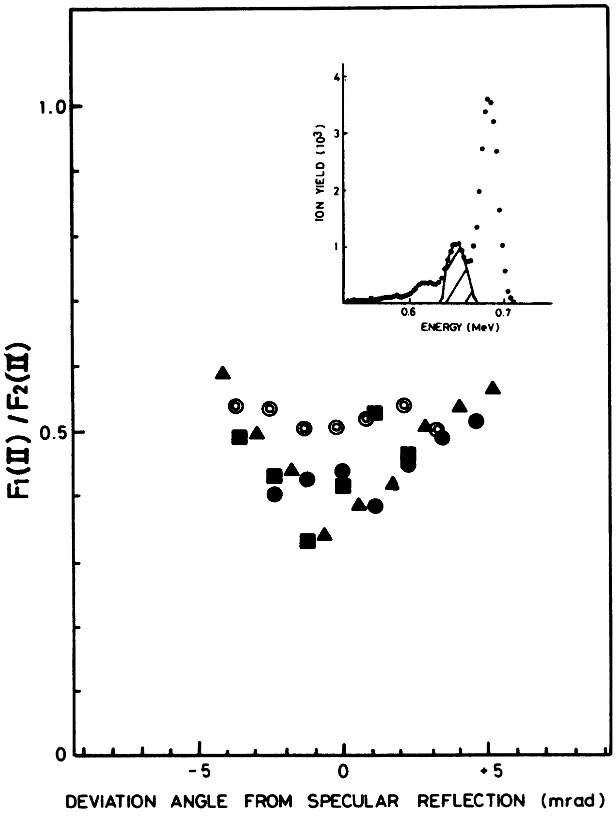

FIG. 2. The ratio of the fraction of He^+ ions, $F_1(\mathrm{II})$, to that of He^{2+} ions, $F_2(\mathrm{II})$, for the ions of second peak as a function of the deviation angle from the specular reflection. The ions of second peak travel for one period of oscillatory motion in the (001) planar channel. The ratios for 0.7-MeV He^+ incidence with glancing angles 2.9 mrad (●), 4.9 mrad (▲), 6.9 mrad (■), and for 0.7-MeV He^{2+} incidence with a glancing angle 5.2 mrad (◎) are shown.

deviation angle from the specular reflection, i.e., as a function of the scattering angle minus twice the glancing angle, for scattering of 0.7-MeV He^+ and He^{2+} ions. The results for the [210] surface-channeled ions is also shown. The observed ratios cluster into a near-universal curve which is a function of the deviation angle from the specular reflection. This curve depends neither on the glancing angle, the incident charge state, nor the scattering mode (whether there is surface channeling or not). The fractions $F_1(\mathrm{II})$ and $F_2(\mathrm{II})$ were also obtained from peak areas for the second peaks of the energy spectra, which show the fractions of He^+ and He^{2+} ions which experienced one period of oscillatory motion in the (001) planar channel. The ratio $[F_1(\mathrm{II})]/[F_2(\mathrm{II})]$ is shown in Fig. 2. Although the scatter is rather large, the ratio $[F_1(\mathrm{II})]/[F_2(\mathrm{II})]$ shows almost the same deviation-angle dependence as $[F_1(\mathrm{I})]/[F_2(\mathrm{I})]$. This suggests that the ion loses the memory of the passage in the (001) planar channel after appearing from the surface. These results indicate that the surface plays an important role in the charge exchange process at the glancing-angle scattering.

For comparison, the charge-state distribution of He ions transmitted through a SnTe self-supporting foil with a clean exit surface was also measured. The foil thickness was about 500 nm and was thick enough for the attainment of the equilibrium charge-state distribution. The observed ratio of the fraction of He^+ ions, $F_1(T)$, to that of He^{2+} ions, $F_2(T)$, in the transmitted beam, and the ratio $[F_1(\mathrm{I})]/[F_2(\mathrm{I})]$ for the specular reflection measured with a glancing angle of 2.9 mrad, are shown as a function of the exit energy in Fig. 3. The ratio $[F_1(\mathrm{I})]/[F_2(\mathrm{I})]$ for the specular reflection is about a half of the ratio $[F_1(\mathrm{T})]/[F_2(\mathrm{T})]$ for the transmission experiment.

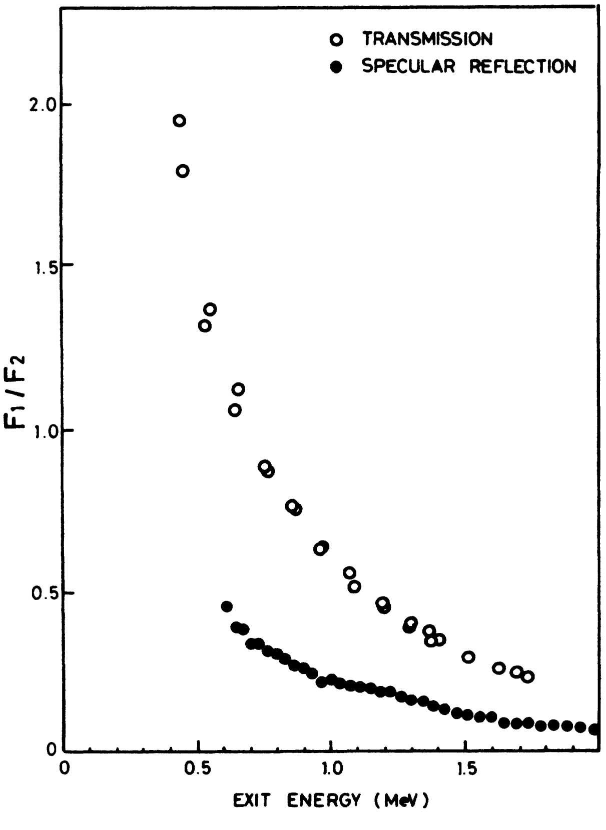

FIG. 3. The energy dependence of the ratio $[F_1(\mathrm{I})]/[F_2(\mathrm{I})]$ for the specular reflection with a glancing angle 2.9 mrad (●) and that of the ratio $[F_1(T)]/[F_2(T)]$ for the ions passing through a self-supporting SnTe foil (○) are shown.

IV. DISCUSSION

For 0.7-MeV He ions scattered from the surface of clean SnTe(001) surfaces, the observed charge-state distribution does not depend on the charge state of the incident ions, the glancing angle, or the incident azimuthal angle with respect to the crystallographic axis, as can be seen in Fig. 1. This indicates that the charge-state distribution achieves equilibrium during the interaction with the surface at the glancing angles studied. However, the observed distribution is very different from the equilibrium charge-state distribution obtained by He ions transmitted through a self-supporting SnTe foil as shown in Fig. 3. This difference can be explained by the following: The velocity of 0.7-MeV He ions is nearly equal to the velocity of the N-shell electrons of Sn and Te atoms. So the He ions passing through the SnTe foil capture electrons mainly from the N shells. However, this is not true for the specularly reflected He ions. Figure 4 shows the density distributions $n_i(x)$ of electrons for individual shells near the surface of SnTe(001) calculated using the Hartree-Fock approximation for isolated Sn and Te atoms,[15] where the surface was assumed to be a bulk exposed surface, $x=0$ was chosen at the center of surface atoms, and the continuum approximation was employed, i.e., the electron distributions were averaged in the plane parallel to the surface. The upper abscissa indicates the glancing angle for 1-MeV He ions whose closest approach is equal to the distance x from the surface shown by the lower abscissa. It can be seen that the specularly reflected He ions should interact mainly with the tail of the valence-electron distribution. These ions have small chance of capturing electrons as compared with the He ions passing through a foil. So the ratio $[F_1(\mathrm{I})]/[F_2(\mathrm{I})]$ for specularly reflected ions is smaller than the ratio $[F_1(T)]/[F_2(T)]$ for foil-transmitted ions.

The charge exchange cross sections for He ions in collisions with valence electrons near the surface can be estimated. Let σ_{cap} be the electron-capture cross section for the He^{2+} ion in collisions with valence electrons near the surface of SnTe(001), and let σ_{loss} be the electron-loss cross section of the He^+ ion for collisions with valence electrons. As the neutral fraction is very small in the present experiment, the He^+ fraction of specularly reflected ions, F_1, can be written as

$$F_1 = F_1^0 e^{-S} + F_1^{\mathrm{eq}}(1-e^{-S}) , \tag{1}$$

where F_1^0 is the incident He^+ fraction,

$$F_1^{eq} = \sigma_{cap}/(\sigma_{cap}+\sigma_{loss}) ,$$

$$S = (\sigma_{cap}+\sigma_{loss}) \int n_v(x) dl ,$$

$n_v(x)$ is the density distribution of the valence electrons, and the integral is performed along the ion trajectory. The present experimental results show that the charge-state distribution of specularly reflected He ions reaches equilibrium, i.e., $F_1 = F_1^{eq}$. The ionization cross section of He^+ ions from impact of electrons with velocity v can be substituted for the σ_{loss} of the He^+ ions with the same velocity v, if the He^+-ion velocity is large enough that motion of the valence electron can be ignored in the collision. This condition holds for MeV He ions. Thus the electron-capture cross section σ_{cap} can be estimated from the experimental charge-state distribution and the ionization cross section of He^+ by the impact of electrons reported in the literature.[16] Figure 5 shows the capture cross sections obtained as a function of He-ion energy. The capture cross section decreases with increasing energy, and it is nearly proportional to E^{-2} in the high-energy region.

The electron-capture cross section of He^{2+} ions in an electron gas can be roughly estimated with the use of the Bohr-Lindhard model.[17] According to this model, the electron-capture process is divided into two steps: a release process of electrons from target atoms and a capture process of the released electrons. In the case of valence-electron capture, however, valence electrons are not bound to particular atoms, and they behave like free electrons. So we need not take account of the release process. A He^{2+} ion passing near an electron with an impact parameter ρ captures the electron if $2e^2/\rho > mv^2/2$, where m is the electron mass and v is the ion velocity. Thus the electron-capture cross section can be written as

$$\sigma = 16\pi a_0^2 (v_0/v)^4 , \tag{2}$$

where a_0 is the Bohr radius and v_0 is the Bohr velocity. The energy dependence of the capture cross sections obtained from the present experiments is in agreement with this theoretical prediction, Eq. (2), although the absolute value is about one-tenth of the theoretical cross section. A more precise theory is needed before a detailed comparison with experimental results is justified.

The contribution of the surface to the charge-state distribution of ions transmitted through a foil can be estimated with Eq. (1), where F_1^0 is regarded as the He^+ fraction in the solid and the integral in S is performed along the ion trajectory after the surface. The measure of

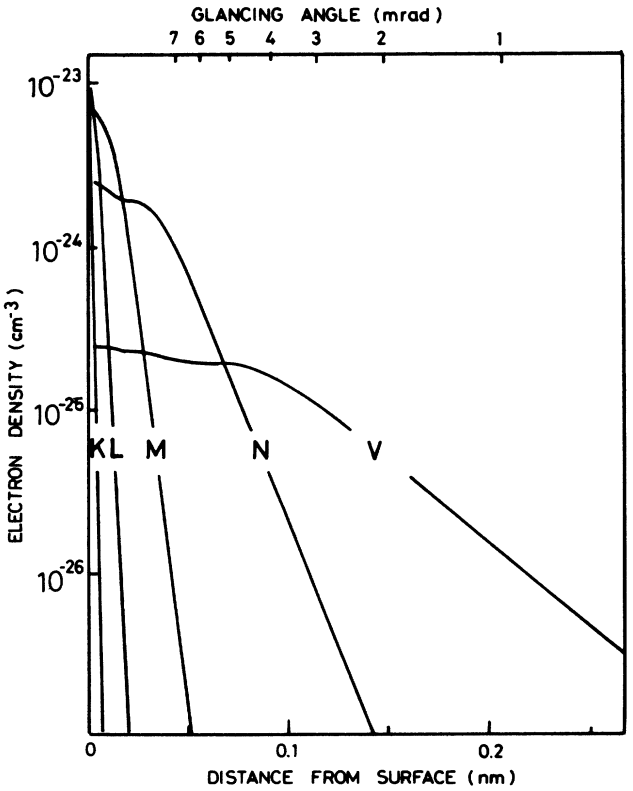

FIG. 4. The electron-density distributions near the surface of SnTe(001) for individual electron shells. The upper abscissa indicates the glancing angle for the 1-MeV He ion whose closest approach is equal to the distance from the surface shown by the lower abscissa.

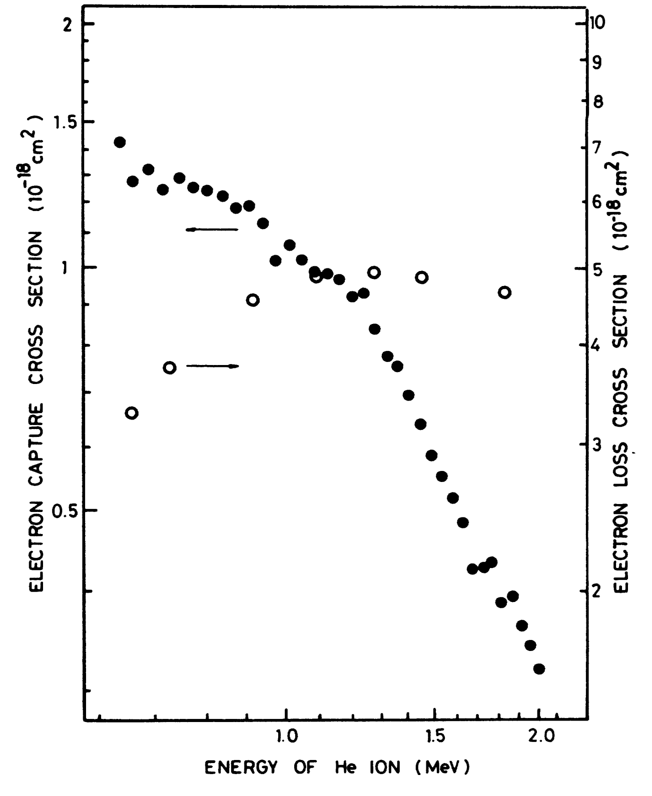

FIG. 5. The electron-capture cross section of He^{2+} ions at collisions with valence electrons of SnTe. The electron-loss cross section of He^+ ions at collisions with the valence electrons is also shown by an open circle, which is estimated from the ionization cross section of He^+ ions by the impact of electrons (Ref. 16).

the surface contribution is given by $1-e^{-S}$. If the ion trajectory is approximated as a straight line, S can be written as

$$S=(\sigma_{\text{cap}}+\sigma_{\text{loss}})nd_p/2\sin\phi \ ,$$

where n is the mean density of the valence electrons in solid, d_p is the interplanar distance, and ϕ is the takeoff angle. The contribution is about 1% for 1-MeV He ions exiting normally from a clean SnTe surface. The contribution becomes larger with decreasing emergence angle. It is about 50% at an emergence angle of 20 mrad. Thus it can be concluded that the influences of the surface on the charge-state distribution in the measurements reported here for MeV He ions is small for the transmission geometry with large emergence angle and is dominant for the very small takeoff angles of the order of 10 mrad used in the specular reflection measurements. This conclusion is in agreement with a previous investigation of the neutralization of energetic light ions at solid surfaces.[5]

Very striking agreement between the charge-state distribution of (25–120)-keV H particles backscattered from surface atoms and that of foil-transmitted particles was reported.[8] The agreement was also reported between the charge-state distribution of (75–180)-keV He ions backscattered from surface atoms and that from bulk atoms;[7] the latter is expected to be the same distribution for ions transmitted through a foil. The present large difference in the charge-state distribution between the specular reflection and the foil transmission seems to be inconsistent with these reports. However, there are differences in the experimental conditions between the present experiment and the reports, i.e., ion energies, target materials, etc. The main difference is whether the surface scattering contains a hard collision (large-angle scattering) or not. During the hard collision the ion penetrates inside the target atom and interacts with many inner-shell electrons. Therefore the ion has a large chance to capture and/or lose electrons. The measure of the number of the collisions with electrons during the hard collision is given by

$$\tilde{n}=2\int_b^\infty n_{\text{atom}}(r)dr \ ,$$

where $n_{\text{atom}}(r)$ is the electron density of the atom and b is the closest approach distance. When MeV He ions are scattered by Sn or Te atoms, $\tilde{n}$ is about 6×10^{17} cm^{-2} for N-shell electrons. The mean density of the N-shell electrons in the SnTe crystal is 5.7×10^{23} cm^{-3}. Therefore, one hard collision corresponds to the passage of 10 nm in the solid with respect to the collision with N-shell electrons. The passage of 10 nm in the solid is long enough for the attainment of the equilibrium charge-state distribution. So it can be expected that the charge-state distribution of backscattered MeV He ion from the surface atoms at large angles agrees approximately with that of the foil-transmitted ions.

The present experimental results show that the charge-state distribution depends slightly on the deviation angle from the specular reflection, as can be seen in Fig. 1. In order to explain this deviation-angle dependence, a knowledge of the details of the trajectory of the ion is needed. A qualitative explanation of this deviation-angle dependence of the charge-state distribution is given here. In specular reflection from a static surface, ions experience a correlated series of small-angle scattering with surface atoms, and they are scattered at the scattering angle equal to twice the incidence angle relative to the surface. The deviation of the scattering angle from the specular reflection angle comes about from scatterings by thermally vibrating surface atoms near the apex of the trajectory where ions experience their closest approach.[13] The impact parameters of these scatterings are comparable with the radius of the N shell, e.g., the impact parameter is about 0.025 nm for 2-mrad scattering of a 1-MeV He ion by a Sn atom, and so an additional charge exchange takes place during these scatterings. It can be calculated from Eq. (1) that about one-tenth of the reflected ions do not change their charge state after leaving the apex of the trajectory. An ion that experiences scattering from thermally vibrating surface atoms retains some memory of the additional charge exchange that occurred during the scattering. Thus the charge-state distribution depends slightly on the deviation angle from the specular reflection angle.

In conclusion, it is shown that for very-small-angle reflection from surfaces that (1) the observed charge-state distribution of scattered MeV ions does not depend on the charge state of the incident ions, the glancing angle, or the incident azimuthal angle with respect to the crystallographic axis, (2) the charge-state distribution is determined in the tail of the valence-electron distribution outside the surface, (3) the cross section of the He^{2+} ion for capture of a valence electron can be estimated from the experimental results, and (4) the surface does not play an important role in the charge exchange process of MeV He ions transmitted through a foil, except for very small emergent angles.

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