Charge-State Distribution of MeV He Ions Scattered from the Topmost Atomic Layer of the SnTe(001) Surface

Kenji Kimura, Hisashi Ohtsuka, and Michi-hiko Mannami
Department of Engineering Science, Kyoto University, Kyoto 606, Japan
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The charge-state distribution of 0.5-MeV He ions scattered from a SnTe(001) surface has been investigated using a new technique of high-resolution high-energy ion scattering spectroscopy. The ions scattered from successive atomic layers can be resolved in the energy spectra of the scattered ions. The observed charge-state distribution of the ions scattered from the topmost atomic layer coincides with that of ions scattered from the subsurface region and does not depend on the incident charge state but depends on the exit angle. The observed exit-angle dependence is explained by a model which includes the charge-exchange process with the valence electrons in the tail of the electron distribution at the surface.

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Charge-exchange phenomena of energetic ions passing through solids have been extensively studied for many years [1]. It was shown that charge-state equilibrium is attained on passage of MeV He ions through even the thinnest of self-supporting foils [2]. Using a backscattering technique, Lurio and Ziegler tried to determine the path length over which the equilibrium is attained [3]. They concluded that the charge-state equilibrium of MeV He ions is attained in the 0.5–1 nm between the backscattering collision and the surface. A more sophisticated experiment was performed by Haight *et al.* with well-characterized surfaces under UHV conditions [4]. They measured the neutralization of 75–180-keV He^+ ions backscattered from clean and Cs-covered Si(100)-(2×1) surfaces using a surface-sensitive channeling technique (a depth resolution of 0.7 nm was attained). It was found that the ion fraction did not depend on the depth from which the ion was scattered. This suggests that charge-state equilibrium is attained by only one backscattering collision. However, the depth resolution of 0.7 nm corresponds to five atomic layers and is long enough to attain charge-state equilibrium. Thus it is still an open question whether charge-state equilibrium is attained by one hard collision with a target atom or not. In this Letter, we report on the first measurement of the charge-state distributions of He ions scattered from the topmost surface atomic layer of SnTe(001) using monolayer-resolvable high-resolution high-energy ion scattering spectroscopy (HRHEISS).

A single crystal of SnTe(001), which has an NaCl-type crystal structure, was prepared by epitaxial growth *in situ* by vacuum evaporation of pure SnTe on a cleavage (001) surface of KCl in a UHV chamber and was used as a target. The surface structure of SnTe(001) was determined to be a bulk exposed surface using reflection high-energy electron diffraction and Rutherford-backscattering-channeling techniques [5]. A beam of 0.5-MeV He^+ ions from a 4-MV Van de Graaff accelerator at Kyoto University was collimated to 2.5 mm×2.5 mm by a series of apertures. The beam current was monitored by a vibrating beam chopper. The typical beam current was about 5 nA. Ions scattered at a scattering angle of 35° were energy analyzed by a 90° sector magnetic spectrometer (radius 300 mm) with inclined boundaries (26.6°) for two-directional focusing. The dispersion of the analyzer was 1200 mm. An aperture of diameter 0.5 mm was installed at the entrance focus point which was placed 200 mm away from the target. The analyzed ions were detected by a position-sensitive detector (resolution 0.13 mm) consisting of microchannel plates located in the focal plane. The energy spectra of scattered ions were measured with an energy resolution of ~0.1%.

Figure 1 shows an example of the observed energy spectra of the scattered He^+ ions at an exit angle $\theta_e = 2.1°$ measured from the (001) surface plane (the incident angle θ_i measured from the surface was 32.9°). There are peaks at ~493, ~489, and ~485 keV. These peaks are equally spaced and their yields are almost the

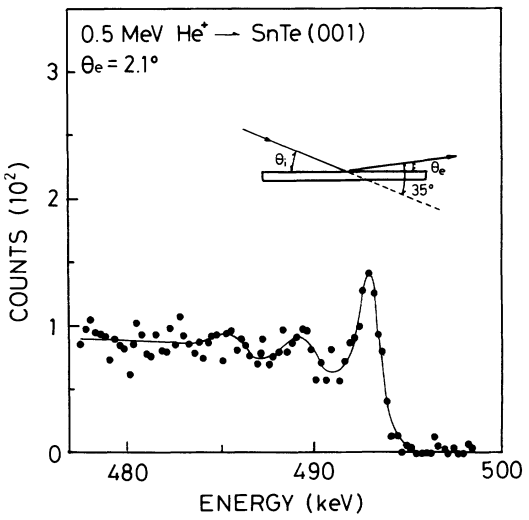

FIG. 1. The energy spectrum of He^+ ions scattered at 35° when 0.5-MeV He^+ ions are incident on the SnTe(001) surface ($\theta_e = 2.1°$). The contributions from successive atomic layers are resolved as separated peaks. The solid curve is a guide to the eye.

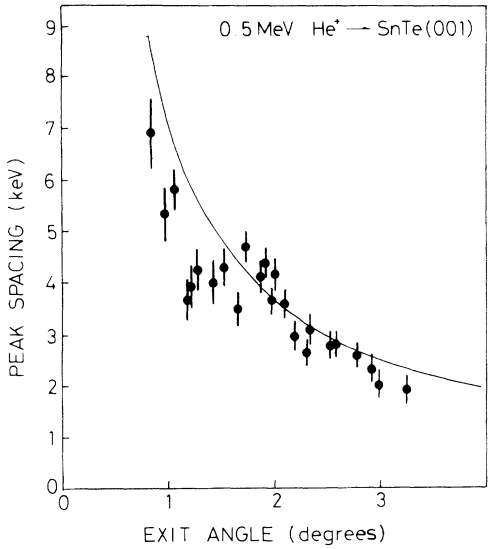

FIG. 2. The exit-angle dependence of the energy spacing between the first and second peaks. The solid line shows the calculated energy difference between the ion scattered from the topmost atomic layer and that from the second atomic layer.

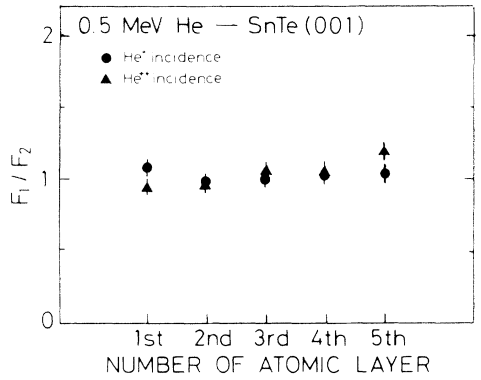

FIG. 3. The ratio of the fraction of He^+ ions to that of He^{2+} ions for the ions scattered from successive atomic layers. The circles show the ratio at $\theta_e = 2°$ for 0.5-MeV He^+ ion incidence and the triangles show that for 0.5-MeV He^{2+} ion incidence.

same. This suggests that these peaks correspond to ions scattered from the first, second, and third atomic layers, respectively. The difference in kinematic factors for He-Sn and He-Te scattering is so small (0.08%) that the ions scattered by Sn and Te atoms cannot be distinguished in the energy spectrum. Similar energy spectra consisting of several peaks were measured only at exit angles smaller than $\sim 3.5°$. The observed exit-angle dependence of the peak spacing is shown in Fig. 2. The peak spacing decreases with increasing exit angle. The energy difference between the scattered He^+ ions from adjacent atomic layers is given by $\Delta E = Sd_p(1/\sin\theta_i + 1/\sin\theta_e)$, where S is the stopping power and d_p is the interplanar distance (the scattering angle $\theta_s = \theta_i + \theta_e$ was kept to 35° throughout the experiment). The calculated ΔE using tabulated values for random stopping power [6] is shown by the solid line in Fig. 2. The agreement between the observed peak spacing and the calculated ΔE is very good. This indicates that the observed peaks correspond to ions scattered from successive atomic layers. Recently, Vrijmoeth *et al.* succeeded in resolving the monolayer contributions in backscattered ion energy spectra using a blocking technique [7]. The contribution from the second atomic layer was seen as a distinct shoulder in the energy spectrum at a blocking condition. Here, we demonstrate that the contributions from the successive atomic layers can be resolved as well-separated peaks in the energy spectrum even in random directions.

Energy spectra of scattered He^{2+} ions were also measured in order to determine the charge-state distribution of the scattered ions. We could not measure the neutral atoms but the fraction of He^0 was smaller than 5% at 0.5 MeV in the previous foil-transmission experiment [8]. So we neglect the scattered He^0 atoms throughout this work. The ratio of the fraction of He^+, F_1, to the fraction of He^{2+}, F_2, was obtained for the ions scattered from successive atomic layers. Figure 3 shows the results at $\theta_e = 2°$ for 0.5-MeV He^+ incidence and that for 0.5-MeV He^{2+} incidence. The observed ratio does not depend on either the incident charge state or the atomic layers from which the ions are scattered. This suggests that the charge-state distribution attains equilibrium after only one hard collision with a target atom on the topmost atomic layer. However, the exit angle $\theta_e = 2°$ is so small that the surface may affect the charge-state distribution during the long outgoing trajectory near the surface and the ion may forget its charge state prior to the interaction with the surface.

Figure 4 shows the observed exit-angle dependence of the charge-state distribution. The ratios F_1/F_2 for the ions scattered from the topmost atomic layer are shown by solid circles (He^+ incidence) and solid triangles (He^{2+} incidence). We could measure the charge-state distribution only at $\theta_e < 3.5°$, because the ions scattered from the topmost atomic layer could not be resolved at $\theta_e > 3.5°$. The ratios for ions scattered from the subsurface region (ions whose inelastic energy loss in solid is smaller than 10 keV) are also shown by open circles (He^+ incidence) and open triangles (He^{2+} incidence). The observed ratios cluster into a universal curve which is a function of the exit angle. The ratio F_1/F_2 for 0.5-MeV He ions passing through a self-supporting SnTe foil was measured to be 1.5 in a previous work [8]. This previous result coincides with the present result at large exit angles. The observed exit-angle dependence at small exit angles shows clearly that the surface plays an important role in the charge-exchange process.

We have measured the charge-state distribution of MeV He ions specularly reflected from SnTe(001) sur-

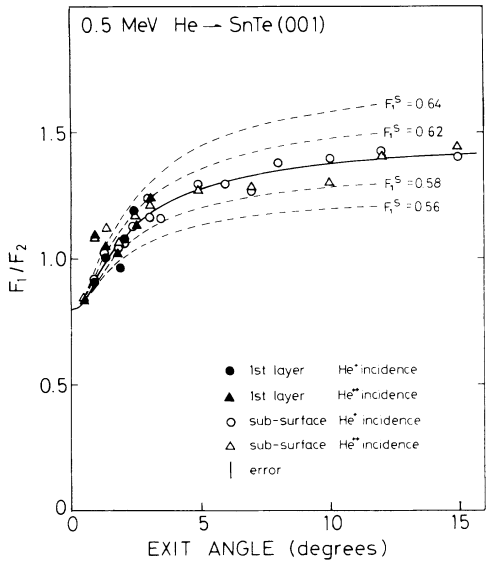

FIG. 4. The exit-angle dependence of the ratio F_1/F_2 for the ions scattered from the topmost atomic layer for incidence of 0.5-MeV He^+ ions (●) and 0.5-MeV He^{2+} ions (▲) and that for ions scattered from the subsurface region for incidence of 0.5-MeV He^+ ions (○) and 0.5-MeV He^{2+} ions (△). The calculated result for the ions scattered from the subsurface region is shown by the solid curve. The results for the ions scattered from the topmost atomic layer calculated with several values of F_1^{single} are shown by dashed curves. The values of F_1^{single} are indicated.

faces [9]. The observed distribution of the ions, which were reflected from the surface without penetration through the surface atomic layer, did not depend on either the incident angle (3–7 mrad) or the incident charge state. This indicates that charge-state equilibrium is attained during interaction with the surface. However, the observed distribution was very different from the equilibrium distribution of ions passing through a self-supporting SnTe foil (the ratio F_1/F_2 was 0.8 at 0.5 MeV for the specularly reflected ions, whereas it was 1.5 for the foil-transmitted ions). This difference is attributed to the fact that the specularly reflected ion interacts only with valence electrons during its long trajectory near the surface. The observed charge-state distribution was considered to be the equilibrium distribution of He ions in the valence electron cloud. Because the velocity of the valence electron is much slower than MeV He ions, the electron capture probability in the valence electron cloud is smaller than that inside the solid. Consequently, the He^+ fraction for the specularly reflected ions is smaller than that for foil-transmitted ions [9].

In the present case, the ions scattered from the subsurface region attain charge-state equilibrium when the ions leave the atomic surface. These ions interact with the valence electrons in the tail of the electron distribution after leaving the atomic surface and change their charge state. Neglecting He^0 atoms, the He^+ fraction after leaving the valence electron cloud can be given by

$$F_1 = F_1^{bulk} e^{-P} + F_1^{surf}(1 - e^{-P}), \quad (1)$$

where $P = \int n(r)(\sigma_c + \sigma_l)dx$, F_1^{bulk} is the equilibrium He^+ fraction in the solid, $F_1^{surf} = \sigma_c/(\sigma_c + \sigma_l)$ is that in the valence electron cloud, $n(r)$ is the valence electron density, σ_c is the electron capture cross section for the He^{2+} ion in collision with a valence electron near the surface, σ_l is the electron loss cross section for the He^+ ion for collision with a valence electron, and the integral is performed along the outward trajectory of the ion after leaving the atomic surface. Using a straight-line approximation for the ion trajectory, P is written as

$$P = (\sigma_c + \sigma_l)\langle n \rangle d_p / 2\sin\theta_e, \quad (2)$$

where $\langle n \rangle$ is the mean density of the valence electrons. Using $\sigma_c + \sigma_l$ as a fitting parameter and substituting $F_1^{bulk} = 0.6$ (obtained from the foil-transmission experiment) and $F_1^{surf} = 0.44$ (obtained from the specular-reflection experiment) into Eq. (1) the fraction of He^+ ions was calculated. The best fit shown by the solid curve in Fig. 4 was obtained with $\sigma_c + \sigma_l = 1.0 \times 10^{-17}$ cm^2. The agreement with the experimental result is very good. Using this result and the relation $F_1^{surf} = \sigma_c/(\sigma_c + \sigma_l)$, the cross sections are calculated as $\sigma_c = 4.4 \times 10^{-18}$ cm^2 and $\sigma_l = 5.6 \times 10^{-18}$ cm^2. This electron loss cross section is 3 times as large as the experimental ionization cross section of He^+ ions from the impact of electrons with the same velocity to the 0.5-MeV He ion [10]. As the electron loss cross section is inversely proportional to the binding energy of the electron, this difference can be ascribed to the existence of the excited He^+ ions in the present experiment. Although the analysis presented here is very crude, it explains the experimental result reasonably well. We need a precise theory describing the charge-exchange process of fast ions at surfaces in order to understand the obtained cross sections. Recently, Flores has developed a theory of charge exchange and energy loss of swift ions in solids [11]. The theory was successfully applied to the surface scattering of keV He ions [12]. The theory might be applied to explain the present result.

The charge-state distribution of the ions scattered from the topmost atomic layer is also affected by the collision with valence electrons and the effect can be calculated with Eq. (1), where F_1^{bulk} is replaced by the He^+ fraction of ions scattered by an isolated target atom, F_1^{single}. The results calculated with several values of F_1^{single} are shown by dashed curves in Fig. 4. Comparing the calculated results with the experimental ones (solid circles and solid triangles in Fig. 4) F_1^{single} can be determined to be 0.61 ± 0.03 for both He^+ and He^{2+} incidences. This means that the charge-state distribution after one hard collision with a target atom is almost independent of the incident charge state and the distribution is almost the same as the equilibrium distribution inside the solid. In the ion-atom collisions, it was found that the charge-state

distributions of ~MeV/u heavy ions after close single collisions with rare gases depend very weakly on the incoming charge state, suggesting that charge-state equilibrium is attained in the single collision [13]. The present result shows that the same thing happens in the ion-solid collision.

The exit-angle dependence of the charge-state distribution was studied both experimentally and theoretically. Chateau-Thierry and Gladieux observed a decrease of the neutral fraction of 1.4-MeV proton beam emerging from foils of Al and Au at oblique angles [14]. However, Cross pointed out that their result can be explained assuming the presence of a thin layer of light atoms on the surface [15]. The present result of the exit-angle dependence cannot be explained by the presence of a thin contamination layer because the ratio F_1/F_2 for the contaminated surface is not 0.8 but 1.3.

We have shown that surface steps affect the charge-state distribution of specularly reflected MeV He ions [16], where θ_i and θ_e were of the order of 0.1° and the step density was about 10^{-2} nm^{-1}. In the present experiment, however, the effect of surface steps was much smaller and so we neglected it because θ_i and θ_e were almost 10 times as large as those of the specular reflection.

We could not determine the charge-state equilibration length in the present experiment. Although the equilibration length is usually estimated using charge-exchange cross sections measured with gaseous targets [15], it is a question whether the estimation gives a correct value or not. Experimental work which can determine the equilibration length using HRHEISS is now in progress in order to resolve the question.

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