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Angular dependence of the neutral fraction of medium-energy hydrogen emerging from $NiSi_2(111)$

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A strong angular dependence has been found first in the neutral fraction of medium-energy hydrogen emerging from $NiSi_2(111)$. The angular dependence can be explained well in terms of the atomic arrangement and composition of the $NiSi_2(111)$ surface and impact-parameter- (trajectory) dependent charge-exchange processes at a few atomic layers beneath the exit surface.

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Charge exchange of energetic ions is one of the most fundamental processes of ion-beam interaction with a solid and a solid surface. In the present paper, we report an experimental result on the crystallographic azimuthal-angle dependence of charge fractions of medium-energy ions emerging from a single-crystalline compound surface. In medium (10–500 keV/u) and high ($\gtrsim$ 500 keV/u) energy ranges, a distinct angular dependence was observed for the charge fraction in a channeling penetration condition [1,2]. For random directions, however, the angular dependence has not been studied in detail, and so far no crystallographic effect has been reported. In contrast, in low-energy (in the $\sim$ keV range) ion scattering (LEIS), the neutralization probability of ions depends on the crystal azimuth and the scattering trajectory [3]. This influences the quantitative analysis of the scattering yield because only scattered ions are detected with an electrostatic analyzer. For the same reason, the understanding of the neutralization of the medium-energy ions in the solid and on its surface is also important for medium-energy ion scattering (MEIS) [4].

Recently, we found that the neutral fraction of medium-energy hydrogen emerging from $NiSi_2(111)$ depends strongly on the exit angle (the angle between the outgoing direction and the surface plane) [5]. In the medium-energy range, the neutral hydrogen in the emerging beam is formed by the charge-exchange processes within the outermost surface layers of the exit surface [6] because of very large charge-exchange cross sections ($\sim 10^{-16}$ cm^2) [7]. In addition, the fraction depends strongly on target atomic species [8]. Thus, the exit-angle dependence can be attributed to the high sensitivity of the neutral fraction to the atomic composition of the outermost surface layers of $NiSi_2(111)$ [5]. In the previous experiment [5], however, the crystallographic direction of the outgoing hydrogen was not specified, and the effect of the crystalline structure on the fraction was not understood at all.

In the present study, we measured the crystallographic azimuthal-angle dependence of the neutral fraction for medium-energy hydrogen emerging from $NiSi_2(111)$. The data will be discussed in terms of the surface structure of $NiSi_2(111)$ and the impact-parameter (trajectory) dependence of electron capture and loss probabilities of hydrogen.

A 100-nm-thick $NiSi_2$ crystal of a type-B orientation was grown epitaxially on a Si(111) wafer by Tung's template method [9]. The crystallographic orientation of the fabricated silicide film was checked by taking channeling spectra along the $\langle 110 \rangle$ and $\langle 114 \rangle$ axes [10]. The neutral-fraction measurements were done in an ultrahigh-vacuum chamber (base pressure 5×10^{-9} Pa) coupled to the Kyoto University 1.7-MV tandem Cockcroft-Walton accelerator. The experimental setup is shown schematically in Fig. 1. The measurements were carried out in the following way, as described in detail previously [5,8]. The surface of the target was cleaned by a sequential procedure of 1-keV Ne^+ sputtering and $\sim$500 °C annealing. A 504-keV H^+ beam was then incident on the target. The backscattered neutrals and ions were detected with a surface barrier detector (SSD) which was positioned at a fixed scattering angle of 30° and had an acceptance angle of about 0.5°. The neutral fraction was determined from the two spectra for the neutrals alone and for both the neutrals and ions, which were taken with and without a magnetic deflecting field applied vertically to the path of charged particles coming to the detector [5,8]. Both spectra spanned the entire energy range from zero to 502 keV (the energy of hydrogen scattered from a Ni surface atom), and the neutral fractions for any exit energies (scattering depths) were determined within a passage through only a few atomic layers beneath the exit surface. The data were taken for azimuthal angles ϕ of the target rotation from $-15°$ to 75° relative to the $\langle \bar{2}11 \rangle$ outgoing direction at three different exit angles θ = 23°, 19.4°, 15°. Here, we take the exit angle θ as the angle between the detector and the surface plane. We reannealed the target every 2000 s of the H^+ bombardment to keep the surface clean and to restore the beam-induced surface damage. By this procedure, the characteristic angular patterns were obtained reproducibly.

Figure 1 shows the obtained azimuthal-angle ϕ dependences of the neutral fractions of 250-keV hydrogen emerging from $NiSi_2(111)$ at the exit angles θ of (a) 23°, (b) 19.4°, and (c) 15°. The neutral fraction depends strongly on both the azimuthal and the exit angles. Main features of the data are as follows: broad minima are found at ϕ = 0° and 60° for all exit angles, and narrow peaks with angular widths of $<1°$ are superimposed on the broad minima. At the lowest exit angle of θ = 15°,

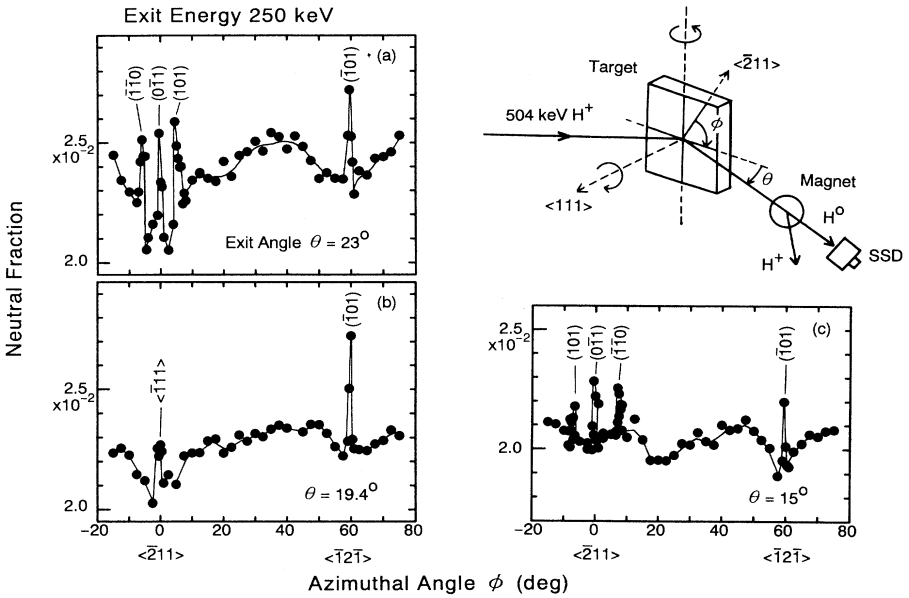

FIG. 1. Measured azimuthal-angle ϕ dependences of neutral fractions for 250-keV hydrogen emerging from $NiSi_2(111)$ at exit angles θ of (a) 23°, (b) 19.4°, and (c) 15°. The solid curves are drawn to guide the eye.

another minimum appears around $\phi = 20°$. The data have similar angular patterns in an exit-energy range of 100–500 keV. (The data shown in Fig. 1 are typical examples.) However, no significant angular dependence was observed at energies $\lesssim 100$ keV.

The narrow peaks can be attributed to the channeling of the backscattered beam on the basis of the following facts: the peaks were observed when the outgoing direction was parallel to a low index crystallographic plane or axis of the $NiSi_2$ crystal (so-called "blocking" direction), and the angular widths of the peaks were consistent with the estimated critical angles for channeling of the emerging hydrogen. It is known that the charge fraction changes drastically around the channeling direction since the channeled particles have larger impact parameters than the randomly directed ones [1,2]. In our case, particles detected along the blocking direction moved under the channeling condition from the backscattering points to the exit surface; as a result, the neutral fraction changed drastically around the blocking directions. Our result differs from higher-energy channeling data by Gaillard *et al.* [2], who observed a remarkable reduction of the neutral fraction at the channeling direction for 0.4–2-MeV hydrogen transmitted through thin crystals. Whether the neutral fraction is enhanced or reduced at the channeling direction depends critically on both the impact-parameter dependence of the charge-exchange probabilities and the flux distributions of the channeled particles in the crystal. In the present study, however, we cannot quantitatively discuss the channeling effect from our limited data since it is difficult to know the flux distributions for the particles which were captured in the crystal channel after the large-angle scattering.

Our interest in the present study is concentrated on the broad minima. The azimuthal data for all exit angles are drawn together in Fig. 2(a), where the channeling peaks are eliminated. The pattern in the azimuthal dependence can be attributed to a surface structural effect by the following three reasons. First, the neutral fraction of hydrogen scattered in any depths is determined within the outermost surface layers because of the very large charge-exchange cross sections. Second, in the present energy range, the charge exchange in the solid is a binary collision between the projectile and the target atom, which can occur at impact parameters as small as a few a.u. (atomic units). Third, the neutral fraction for a pure Ni target is much higher than that for a pure Si target [8]; the fact reflects the larger electron capture cross section for a Ni atom than for a Si atom. In the case of 250-keV hydrogen, the neutral fractions for a pure Si and Ni

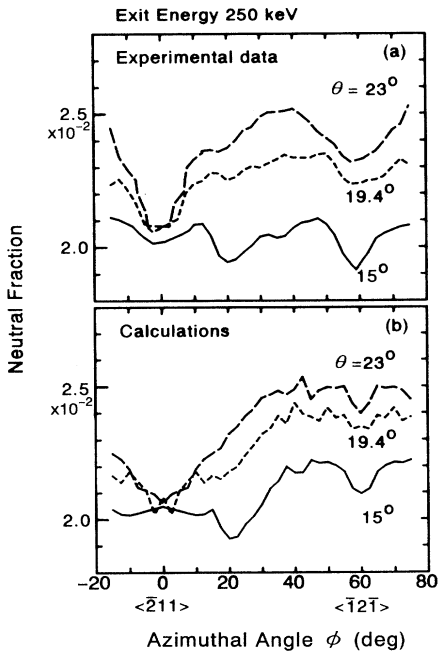

FIG. 2. Comparison of the measured (a) and the calculated (b) azimuthal-angle ϕ dependences. The channeling peaks are eliminated from the experimental data.

are 1.2×10^{-2} and 4.3×10^{-2}, respectively. Thus, for hydrogen emerging from $NiSi_2(111)$, it is expected that the neutral fraction is reduced for the outgoing trajectories that pass close to the Si atom at the exit surface, whereas it is enhanced for the trajectories that pass close to the Ni atom and far from the Si atom. Therefore, the azimuthal dependence of the neutral fraction can reflect the atomic arrangement of Si and Ni at the exit surface. It should be noted that, since the cross section for large-angle ($\gtrsim 1°$) deflection is much smaller than that for charge exchange in the present energy range, the projectiles suffer only a small deflection during the charge-exchange collision, and thus most of them pass through the exit-surface layers along straight-line trajectories.

So far, two structural models have been proposed for the $NiSi_2(111)$ surface. One is a bulk-terminated model in which the surface was bulk-terminated with Si as the topmost layer and Ni as the second one [Figs. 3(a) and 3(b)] [11], and the other is a bilayer model in which the surface consists of a Si terminated layer with an additional Si bilayer on the top [Fig. 3(c)] [12]. Our experimental results in Fig. 2(a) can reasonably be explained by using the bulk-terminated model. That is, at $\phi = 0°$ and 60°, most of the particles that pass close to the second layer Ni atom finally pass close to the adjacent top-layer Si atoms at emergence. This will cause the reduction of the neutral fraction. In contrast, the neutral fraction is enhanced around $\phi = 40°$ because the particles that pass close to the second-layer Ni atom can escape from the surface without any charge-exchange collisions with Si atoms on the top layer. At the lowest exit angle of $\theta = 15°$, the neutral fraction is reduced also around $\phi = 20°$. The minimum can be due to the charge exchange between the projectile and the top-layer Si atom located at the $\langle\bar{5}41\rangle$ direction (corresponding to $\phi = 19.1°$) with respect to the second-layer Ni atom [see Fig. 3(a)]. On the contrary, the present data cannot be explained by the bilayer model. Evidently, if the additional Si bilayer lies on the bulk-terminated surface as shown in Fig. 3(c), the minimum at $\phi = 20°$ must be observed also for $\theta > 15°$.

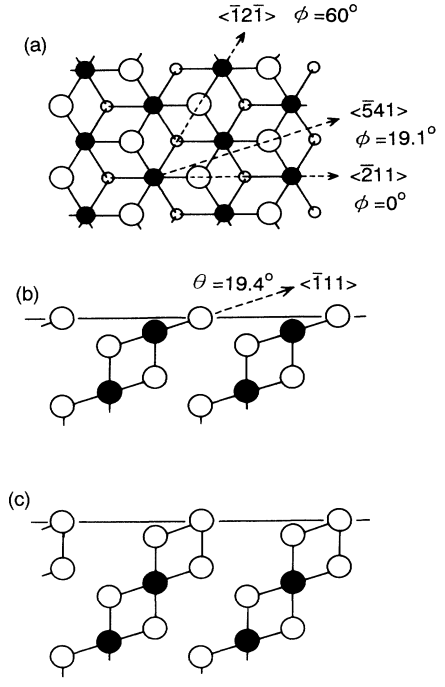

FIG. 3. Surface structural models of $NiSi_2(111)$. (a) Top view of the bulk-terminated model. Open and solid circles are Si and Ni atoms, respectively. (b) Side view of the bulk-terminated model. (c) Side view of the bilayer model. The surface contains an additional Si bilayer on top of the bulk-terminated surface.

It should be noted that, in the case of a clean Si(100) target, the neutral fraction of hydrogen hardly depends on the crystal azimuth except for several narrow peaks at the blocking directions [16]. A similar result was obtained also by Haight *et al.* for 75–180-keV helium [13]. Therefore, the strong azimuthal dependence observed for $NiSi_2(111)$ should come from the fact that the surface consists of a regular array of two different atomic species between which the charge-exchange cross sections of hydrogen are very different.

In order to discuss the detailed azimuthal patterns in Fig. 2(a), a crude numerical calculation was done for the bulk-terminated surface, by taking account of charge-exchange collisions within the last ten layers. First, starting points of 400 projectiles were uniformly distributed over a unit cell of the lattice in the tenth layer below the surface. The outgoing trajectories in the crystal were approximated by straight lines. The neutral fraction was calculated at each point of the closest approach to the target atom, using impact-parameter-dependent electron-capture and loss probabilities. The electron loss probabilities were calculated by the classical-trajectory Monte Carlo (CTMC) method [14] by using the Thomas-Fermi-Molière potential [15] as the target potential field. It is difficult to calculate the impact-parameter-dependent electron-capture probabilities for multielectron atoms (such as Si or Ni). Thus, in the present calculation, we assumed the capture probabilities to be $C\exp(-b/w)$, where b is the impact parameter, C and w are parameters that were fitted to the azimuthal patterns. Finally, the fractions were averaged over all the trajectories. Figure 2(b) shows the calculated azimuthal dependences. The calculation can well reproduce the main features of the azimuthal patterns in Fig. 2(a), such as the broad minima at $\phi = 0°$ and 60° for all exit angles and the minimum at $\phi = 20°$ for $\theta = 15°$. (On the contrary, the bilayer model cannot reproduce the experimental results with any choice of the fitting parameters [17].) The calculated neutral fraction depends strongly on the trajectory of the emerging hydrogen, and reflects the atomic arrangement of Si and Ni of the outermost surface layers. The calculation indicates also that the detailed angular dependence is sensitive to the width of the impact-parameter dependence of the charge-exchange probability. The reproduction is obtained here on the condition that the electron loss occurs dominantly at impact parameters $\lesssim 3$ a.u., whereas the capture occurs at values $\lesssim 1$ a.u. We consider that more detailed measurements and calculations can reveal the detailed distributions of the impact-parameter-dependent charge-

exchange probabilities of ions in solid and on its surface.

In summary, we found first the strong angular dependence of the neutral fraction of medium-energy hydrogen emerging from $NiSi_2(111)$. The angular dependence is the reflection of the geometrical arrangement of Si and Ni atoms in the $NiSi_2(111)$ surface. The result indicates also that the detailed angular dependence is sensitive to the impact-parameter dependence of the electron capture and loss probabilities. The simple calculation including the impact-parameter-dependent charge-exchange probabilities can well reproduce the main features of the obtained angular dependences.

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[17] Especially, the neutral fraction calculated by the bilayer model is much smaller than the experimental data, and rather close to the fraction for pure Si [5].