Resonant Coherent Excitation of Surface Channeled Ions

K. Kimura, H. Ida, M. Fritz, and M. Mannami

Department of Engineering Science, Kyoto University, Kyoto 606-01, Japan

(Received 18 July 1995; revised manuscript received 4 December 1995)

We observe the resonant coherent excitation (Okorokov effect) of [100] surface channeled B^{4+} ions from $n = 1$ to $n = 2$ on a SnTe(001) surface through the reduction of the B^{4+} fraction which is caused by the ionization of the excited B^{4+} ions. On an atomically flat surface, the reduction of the B^{4+} fraction is almost erased by subsequent charge exchange processes near the surface. We show that these subsequent charge exchange processes can be suppressed by utilizing surface steps. [S0031-9007(96)00139-1]

PACS numbers: 79.20.Rf, 34.50.Fa, 61.85.+p

When an ion passes through an axial channel in a crystal, it feels a periodic perturbation of frequencies $\nu = kv/d$, where $k = 1, 2, 3, \ldots$, v is the ion velocity, and d is the atomic spacing along the axis. If one of the frequencies coincides with the excitation energy of the ion, i.e., if the following condition is fulfilled,

$$2\pi\hbar \frac{kv}{d} = \Delta E \,, \tag{1}$$

a resonant coherent excitation (RCE) can occur, where ΔE is the excitation energy. This effect, predicted by Okorokov [1] 30 years ago, was first observed by Datz *et al.* [2] through the change in the charge state distribution of channeled ions and, recently, through the enhancements of projectile x-ray emission [3,4] and of convoy electron emission [5].

The possibility of RCE of surface channeled ions (surface Okorokov effect) was first pointed out by Kupfer, Gabriel, and Burgdörfer [6]. They showed that resonant coherent excitation leads to a strongly polarized excited state. Elci suggested that surface channeling has a considerable advantage over the transmission geometry for obtaining stimulated emission of soft x rays from ions [7]. Recently, Garcia de Abajo, Ponce, and Echenique studied the resonant coherently induced electron loss to the continuum for the case of surface channeling [8]. They suggested a new spectroscopy for analyzing the dynamical interaction of electronic bound states in the vicinity of surfaces. In spite of many theoretical studies on the RCE of surface channeled ions, there has been no experimental study on this subject so far. In this Letter, we report the first observation of the RCE of surface channeled ions.

A single crystal of SnTe(001) was prepared by epitaxial growth *in situ* by vacuum evaporation on a cleaved surface of KCl at 250 °C in a UHV chamber. The crystal was mounted on a five-axis precision goniometer. As SnTe has a NaCl-type crystal structure, Sn atoms and Te atoms are alternately arranged along the [100] direction with a spacing $d = 6.0$ a.u. Strictly speaking, there are additional resonance frequencies of half integers ($k = 0.5, 1.5, 2.5$, etc.). However, the small difference between the atomic numbers of Sn and Te makes the amplitudes of these additional frequencies very small. Here, we study the RCE of $n = 1$ to $n = 2$ transition of the surface channeled B^{4+} ion traveling along the [100] axis on the SnTe(001) surface through the change in the charge state distribution of the ions. Using Eq. (1), the resonance energy is calculated to be 5.46 MeV for $k = 2$.

Beams of 4.6–6.4 MeV B^{3+} ions from the 1.7 MV Tandetron accelerator of Kyoto University were incident on the crystal at glancing angles of 2–6 mrad measured from the surface plane. The beams were collimated by a series of apertures to less than 0.1×0.1 mm^2 and to a divergence angle less than 0.3 mrad. The azimuthal angle of the crystal was aligned to the [100] direction. The ions scattered at the specular angle were energy analyzed by a 90° sector magnetic spectrometer, which could be rotated around the target. The spectrometer had an aperture ($\phi = 0.2$ mm) at the entrance focal point 40 cm downstream from the target. Microchannel plates located in the focal plane of the spectrometer served as a position-sensitive detector (length 55 mm, resolution 0.13 mm) for the ions. Including the energy spread of the incident beam, the energy resolution of the spectrometer was better than 0.1%.

The charge state distribution of the reflected ions was measured as follows: The magnetic field of the spectrometer was changed periodically so that the ions of each charge state could reach the detector alternately. The energy spectrum of q^+ ions was registered in the qth memory group of a multichannel analyzer. This was done to reduce the error caused by the fluctuation of the incident beam. In the present energy region, the charge state distribution of the scattered ions depends weakly on the incident charge state [9], and the B^{4+} fraction is dominant (larger than 50%), irrespective of the incident charge state showing a large charge exchange probability near the surface. Although we studied the RCE of B^{4+} ions, the B^{3+} beam was used as an incident beam because the accelerator could not produce a B^{4+} beam of the required intensity. Approaching the surface, however, the incident B^{3+} ions lost their electrons, and the produced B^{4+} ions were excited by the RCE at the resonance energy.

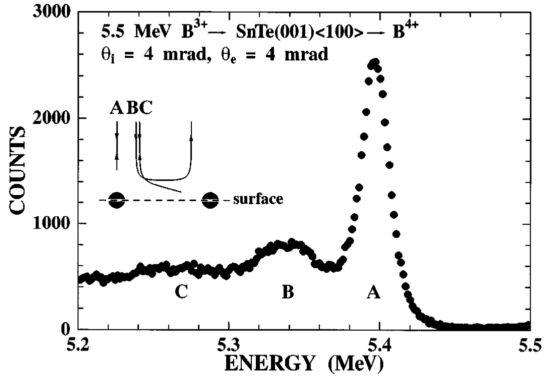

FIG. 1. Energy spectrum of the [100] surface channeled B^{4+} ions at the incidence of 5.5 MeV B^{3+} ions on SnTe(001). The ions were observed at the specular angle, i.e., the angle of incidence was $\theta_i = 4$ mrad, and the exit angle was $\theta_e = 4$ mrad. There are several well-defined peaks which correspond to the specific trajectories shown by the inset. The closed circles in the inset show the atomic strings. The highest energy peak corresponds to the trajectory *A*, the second and third peaks correspond to *B* and *C*.

Figure 1 shows an example of the observed energy spectrum of the scattered B^{4+} ions when 5.5 MeV B^{3+} ions were incident along the [100] axis at a glancing angle $\theta_i = 4$ mrad. There are a few peaks at ~5.4, ~5.34, and ~5.26 MeV. In our previous study on the energy loss of surface channeled ions, we have shown that these peaks correspond to the particular ion trajectories [10]. The corresponding ion trajectories projected on the plane perpendicular to the atomic string are shown in the inset. The peak of highest energy corresponds to the on-string trajectory labeled *A*, which has the shortest path near the surface. Here, we concentrate on this trajectory. For the ion that follows this trajectory, the probability for the occurrence of the RCE is expected to be large because of the shortest distance of the closest approach to the atomic string. As the electron loss probability for the excited ion is much larger than that for the ion in the ground state, a reduction of the B^{4+} fraction should occur at the resonance energy, as had been observed in the transmission experiment [2].

Figure 2 shows the ratio of the observed B^{4+} fraction to the B^{5+} fraction. The statistical error is smaller than the symbols. It is known that the ratios of the charge state fractions of foil-transmitted 0.2–6.5 MeV He ions can be fitted with a simple expression of the form $F(q)/F(q') = AE^n$, where A and n are constants [11]. This expression can also be applied to 0.5–2.0 MeV He ions [12] and 0.5–2.0 MeV C ions [13] which are specularly reflected from crystal surfaces. The present results are therefore shown by a log-log plot to emphasize the effect of the RCE. The result at $\theta_i = 2$ mrad shows a small reduction of the B^{4+} fraction around 5.5 MeV which agrees with the calculated resonance energy (5.46 MeV), suggesting that the observed B^{4+} reduction is caused by the RCE. At $\theta_i = 4$ mrad, the resonance becomes broader and shifts towards higher energies.

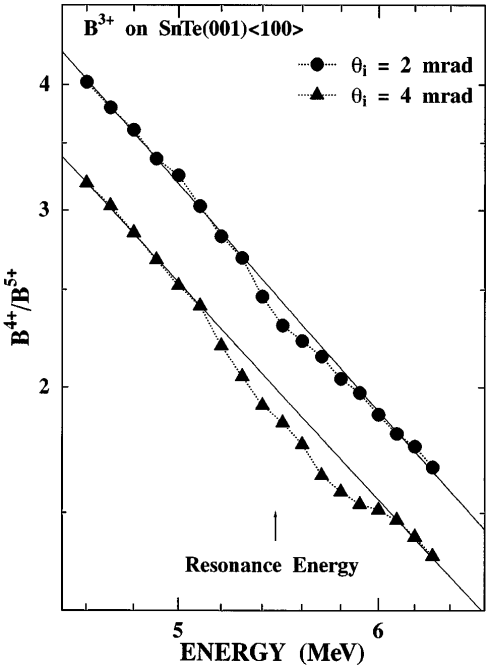

FIG. 2. Ratio of the B^{4+} fraction to the B^{5+} fraction observed at the specular angle when B^{3+} ions are incident at $\theta_i = 2$ and 4 mrad on a SnTe(001) surface along the [100] axis. The lines are drawn to guide the eye. The statistical error is smaller than the symbols.

The observed θ_i dependence of the resonance shape can be explained in terms of the Stark effect. The energy levels of the surface channeled ions are shifted by the Stark effect in the same way as those of ions channeling through bulk crystals [14]. The energy levels of the surface channeled B^{4+} ions traveling along the trajectory *A* were calculated by means of first order perturbation

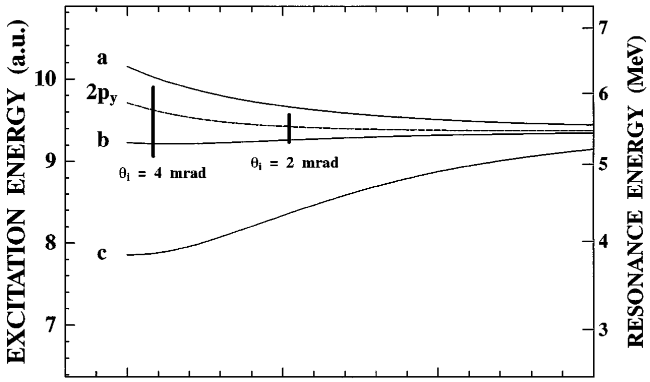

FIG. 3. Calculated excitation energies from 1*s* to $n = 2$ states for 5.5 MeV B^{4+} ions near the SnTe[100] string. The 2*s* mixes with $2p_x$ and $2p_z$ to make substates *a*, *b*, and *c*. The excitation energies derived from the observed B^{4+} reduction are shown by bars.

theory. The surface potential, which consists of the surface wake potential [15] and the string potential, was treated as a perturbation. The difference from the theory of Crawford and Ritchie was the usage of the surface wake potential instead of the wake potential. Figure 3 shows the calculated excitation energies from ls to $n = 2$ states for 5.5 MeV B^{4+} ions at the surface. The $2s$ mixes with $2p_x$ and $2p_z$ (the x axis is parallel to the string direction and the z axis is perpendicular to the surface) to make substates labeled a, b, and c. The $2p_y$ does not mix with other states because it is antisymmetric with respect to the y axis, while other states ($2s$, $2p_x$, and $2p_z$) and the surface potential are symmetric. The Fourier components of the electric field along the trajectory A has only x and z components. As a result, the $2p_y$ state cannot be excited by the RCE in the dipole approximation.

The excitation energies estimated from the observed B^{4+} reduction (5.3–5.7 MeV at $\theta_i = 2$ mrad and 5.1–6.1 MeV at $\theta_i = 4$ mrad) are shown by bars at the distances of the closest approach ($z = 1.52$ a.u. at $\theta_i = 2$ mrad and $z = 1.08$ a.u. at $\theta_i = 4$ mrad). Since the amplitude of the periodic field increases very rapidly with decreasing distance from the string, the RCE occurs mainly at the closest approach. In the present energy region, however, the charge state was not frozen as mentioned above. The observed B^{4+} reduction is a result of the competition between the resonance process and the charge exchange process. Moreover, the width of the resonance is determined by the number of effective interactions with ion cores. The resonance width for surface channeling is much wider since the surface channeled ions interact with a relatively small number of ion cores around the turning point. Although, due to those aspects, the comparison between the calculated excitation energies and the observed resonance is not straightforward, the calculated result qualitatively explains the observed result quite well, as can be seen in Fig. 3.

Thus the RCE could be a reasonable explanation for the observed reduction of the B^{4+} fraction, though the effect is very small probably due to the large charge exchange probability at the surface. If it were possible to measure the charge state distribution at closest approach, i.e., to get rid of subsequent charge exchange processes, a more pronounced reduction of the B^{4+} fraction might be observed. This can be done by utilizing surface steps [16]. The surface we used was not an ideal surface but had defects such as surface steps. If an ion hits a down step, this ion hardly interacts with the surface after passing over the step, which is due to the fact that the step height [6.0 a.u. for a monatomic height step on SnTe(001)] is very large. The ion does not change its state after passing over the step, except for a small possibility of autoionization processes. The exit angle of the ion scattered by the step deviates from the specular angle, as shown in the inset of Fig. 4. This suggests that it should be possible to observe the charge state distribution during the closest approach by observing ions at exit angles smaller than the specular angle.

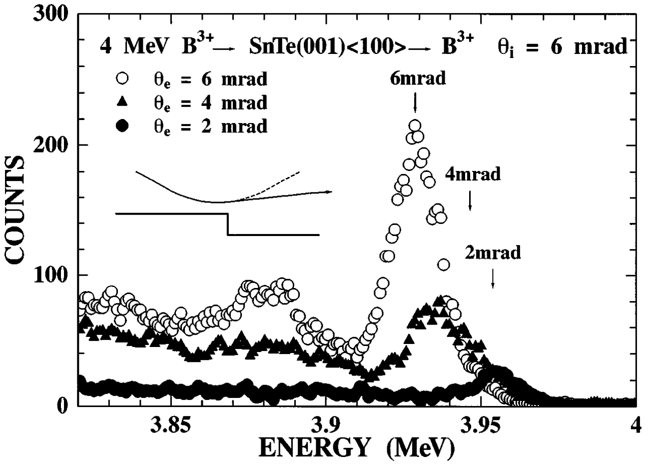

FIG. 4. Displayed are energy spectra of [100] surface channeled B^{3+} ions observed at various exit angles at the incidence of 4 MeV B^{3+} ions on the SnTe(001). The inset shows the trajectory of the ion whose exit angle is, due to hitting a down step, smaller than the specular angle. The arrows indicate the calculated energies of ions scattered by down steps.

Figure 4 shows the observed energy spectra at various exit angles. With decreasing exit angle from the specular angle ($\theta_e = 6$ mrad), the peak energy increases. Although only the spectra of scattered B^{3+} ions are shown in the figure, the same features were seen for other charge states [17]. In a previous study [10], we have shown that the energy loss of surface channeled MeV He ions can be explained by the theoretical position-dependent stopping power based on an inhomogeneous electron gas model [18] and a binary encounter model [10]. The energy loss of the surface channeled boron ion was calculated with the theoretical stopping power. In the calculation, we chose the effective charge $Z_{\text{eff}} = 4.1$ to reproduce the observed energy loss at the specular angle. The arrows in Fig. 4 show the calculated results. The calculated energy loss at $\theta_e = 2$ mrad agrees very well with the observed result. This indicates that the ions observed at $\theta_e = 2$ mrad correspond to ions scattered by down steps as shown in the inset. The energy loss of an ion hitting an up step is also smaller than that of specularly reflected ions. However, this ion appears at an exit angle larger than the specular angle. Thus we can exclude ions scattered at up steps by choosing the exit angle smaller than the specular angle.

There are other mechanisms which cause angular deviation such as scattering by thermally vibrated surface atoms and scattering by surface electrons. The energy loss of ions deflected by these processes is almost independent of the deviation angle. The calculated energy loss at $\theta_e = 4$ mrad is slightly smaller than the experimental one showing that some ions observed at $\theta_e = 4$ mrad are deflected by these additional processes.

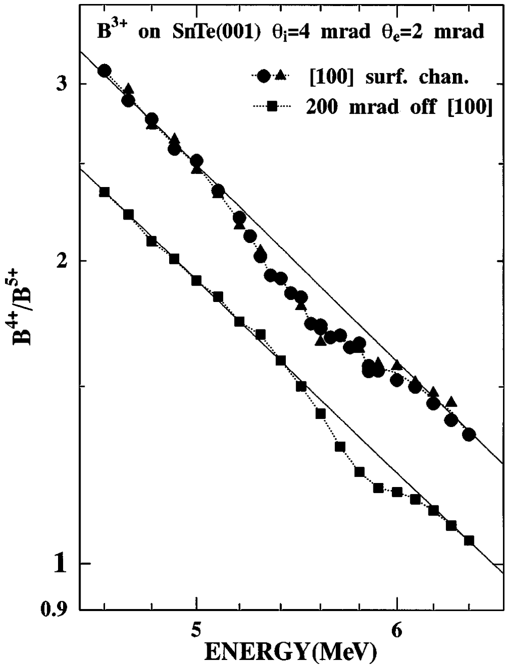

FIG. 5. Ratio of the B^{4+} fraction to the B^{5+} fraction observed at the off-specular angle ($\theta_e = 2$ mrad) when B^{3+} ions are incident on the SnTe(001) surface along the [100] axis with $\theta_i = 4$ mrad. The solid circles and solid triangles show the result of different energy scans. The result observed at the azimuthal angle of 200 mrad off the [100] axis with $\theta_i = 4$ mrad and $\theta_e = 2$ mrad is also shown by solid squares. The lines are drawn to guide the eye. The statistical error is smaller than the symbols.

Figure 5 shows the ratio of the B^{4+} fraction to the B^{5+} fraction observed at $\theta_e = 2$ mrad when B^{3+} ions were incident with $\theta_i = 4$ mrad. The results of two energy scans are shown by circles and triangles. The statistical error is smaller than the symbols. Another experimental error, estimated from the difference between the two scans, is comparable to the size of the symbols. The reduction of the B^{4+} fraction about 5.6 MeV is more pronounced than that shown in Fig. 2, indicating the efficacy of surface steps.

In order to exclude other possible mechanisms, which might be responsible for the observed B^{4+} reduction, the charge state distribution was measured at an azimuthal angle $\phi = 200$ mrad from the [100] axis. Even at the off-axis conditions, the RCE can occur. However, the resonance condition is different from Eq. (1) and is given by [19]

$$2\pi\hbar \frac{k\cos\phi + l\sin\phi}{d}\nu = \Delta E\,, \tag{2}$$

where k and l are integers. Substituting $\phi = 0$, this equation is reduced to Eq. (1). The observed ratios of the B^{4+} fraction to the B^{5+} fraction at $\phi = 200$ mrad are shown by squares in Fig. 5. Other experimental conditions were the same as for the [100] surface channeling experiment. There is a dip at ~5.85 MeV which is about 1.04 times larger than the resonance energy (~5.6 MeV) observed at the [100] surface channeling. The observed energy shift agrees with the calculated shift at $\phi = 200$ mrad for $k = 2$ and $l = 0$ ($1/\cos^2\phi = 1.041$). This clearly indicates that the observed B^{4+} reduction was caused by the RCE.

In the present work, we observed the RCE for [100] surface channeled B^{4+} ions by measuring the change in the charge state distribution. The reduction of the B^{4+} fraction due to the RCE is partly erased by the subsequent charge exchange processes at the surface. It is shown that these subsequent charge exchange processes can be suppressed by utilizing surface steps. Since the charge exchange probability decreases with increasing ion energy, the RCE may be easily observed with high energy ions, although the smaller critical angle for surface channeling might cause an additional experimental difficulty.

We would like to thank the members of the Department of Nuclear Engineering of Kyoto University for the use of the Tandetron accelerator. We are grateful to Professor J. Burgdörfer for fruitful discussions. This work was supported in part by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture.

[1] V. V. Okorokov, Pis'ma Zh. Eksp. Teor. Fiz. **2**, 175 (1965) [JETP Lett. **2**, 111 (1965)].
[2] S. Datz *et al.*, Phys. Rev. Lett. **40**, 843 (1978).
[3] F. Fujimoto *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **33**, 354 (1988).
[4] S. Datz *et al.*, Radiat. Eff. Def. Solids **117**, 73 (1991).
[5] K. Kimura *et al.*, Phys. Rev. Lett. **66**, 25 (1991).
[6] E. Kupfer, H. Gabriel, and J. Burgdörfer, Z. Phys. A **300**, 35 (1981).
[7] A. Elci, Phys. Rev. Lett. **53**, 1696 (1984).
[8] F. J. Garcia de Abajo, V. H. Ponce, and P. M. Echenique, Phys. Rev. Lett. **69**, 2364 (1992).
[9] K. Narumi *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **100**, 1 (1995).
[10] Y. Fujii *et al.*, Phys. Rev. A **47**, 2047 (1993).
[11] J. C. Armstrong *et al.*, Proc. Phys. Soc. **86**, 1283 (1965).
[12] K. Kimura *et al.*, Phys. Rev. B **38**, 1052 (1988).
[13] K. Matsuda, Master thesis, Kyoto University, 1983 (unpublished).
[14] C. H. Crawford and R. H. Ritchie, Phys. Rev. A **20**, 1848 (1979).
[15] Y. H. Ohtsuki, *Charge Beam Interaction with Solids* (Taylar & Francis, London, 1983), p. 230.
[16] Y. Fugii *et al.*, Phys. Rev. A **49**, 1897 (1994).
[17] M. Fritz *et al.* (to be published).
[18] M. Kitagawa, Nucl. Instrum. Methods Phys. Res., Sect. B **33**, 409 (1988).
[19] S. Datz *et al.*, Nucl. Instrum. Methods **170**, 15 (1980).