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Trajectory dependent resonant coherent excitation of planar-channeled ions in a thin Si crystal

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Abstract

We observed resonant coherent excitation (RCE) of 1s electron to the $n = 2$ states in 390 MeV/u Ar^{17+} ions passing through thin Si crystals of about 1 μm in the {220} planar-channeling condition by measuring both the exit charge state and the exit angle of the emerged ions simultaneously in a list mode. The yield of the de-excitation X-rays from the Ar^{17+} ions was also measured. The thin crystal is suitable to study trajectory dependent RCE dynamics, because the exit angle of the high energy planar-channeling ion is uniquely related to the ion trajectory. From the de-excitation X-ray yield on resonance, we learned that RCE occurs with a fairly large probability within the traveling length of about 1 μm, and a majority of the excited ions survive from ionization by the collisions with target electrons or nuclei, and de-excite into the ground state by emitting the X-rays. On the other hand, the observed resonance profile of the exit charge state is shallow and broadened by the Stark effect. We found that the ionization of the excited ions takes place when the ions travel close to the atomic plane from the observed relation between the charge state and the exit angle of the emerged ions.

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Keywords: Heavy ion; Channeling; Electronic excitation; Stark shift

1. Introduction

Energetic ions passing through a single crystal feel temporally oscillating fields originating from the spatially periodic structure. When one of the frequencies corresponds to the transition energy of the ion, the oscillating field has a chance to resonantly excite the internal state of the ion. This process called resonant coherent excitation (RCE) was first pointed out by Okorokov [1] in 1965. Since then, many experiments dealing with the electronic excitation of the channeled ions through RCE have been performed using energetic ions in the axial- or planar-channeling condition, or in the surface-scattering condition [2–4]. For a decade we have been devoted to the experimental studies of RCE of several 100 MeV/u heavy ions ($Z \geqslant 18$) in the planar-channeling condition, where RCE is induced by a periodic array of atomic strings on an atomic plane [5–8]. The once excited ions through RCE experience two competing channels of subsequent ionization by the collisions with the target electrons or nuclei, and the spontaneous de-excitation to the ground state by emitting X-rays in addition to the resonant coherent de-excitation (the reversed process of RCE). The observation of RCE has been performed by the measurements of the exit charge state distribution of the ions and the yield of the de-excitation X-rays from them. The observed features are

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asymmetric and structured resonance profiles due to the Stark shift induced by the static planar potential in the channels. The information on the trajectories of the channeled ions has also been obtained through the deposited energy of the ions by adopting Si solid-state detectors as the target crystals, and the ion trajectory dependence of the transition energies reflecting the Stark shift of the excited ion states was clearly observed.

In this paper, we report the observation of trajectory dependent RCE of high energy (390 MeV/u) hydrogen-like Ar^{17+} ions passing through thin (about 1 μm) Si crystals in the {220} planar-channeling condition. We measured the charge state of the emerged ions after passing through the crystal and their exit angle simultaneously in a list mode (event by event).

Generally, a planar-channeled ion oscillates in a planar channel many times. Even though both incident and exit angles are experimentally identified, several trajectories with the different numbers of bounces satisfy the corresponding condition, and it is impossible to specify the trajectory uniquely. The difference in the number of bounces naturally results from the differences in the oscillation period and the oscillation amplitude due to the unharmonicity of the planar potential. Since these differences are reflected in the energy loss of the ions, we can distinguish the trajectories with the different number of bounces by measuring the energy loss [9,10]. These experiments have been performed with thin crystals, but the energy of the channeled ions were around 1 MeV/u, which results in the bouncing of about ten times.

In the present experiments, we prepared the crystals of the thickness (about 1 μm) much smaller than the oscillation period of the high energy ions (about 4 μm). Under this situation the traveling ions bounce at the atomic plane merely once at maximum, i.e., they experience the half collision with the atomic plane, which enables us to specify the trajectory of the channeled ion experimentally by measuring the exit angle of the ions. A typical example of the ion trajectory simulation corresponding to the present experimental conditions is shown in Fig. 1, where the exit angle of the ion is uniquely related to the ion trajectory and the position x (the distance from the channel center in the direction perpendicular to the plane) at the entrance. The ion trajectory approaching closer to the atomic plane emerges from the crystal with the larger exit angle.

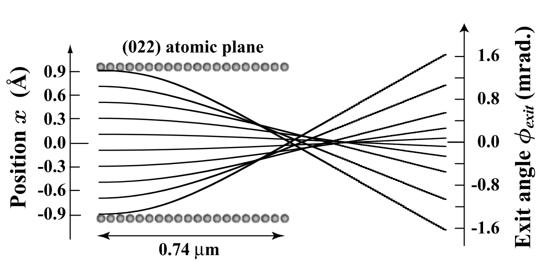

Fig. 1. Simulated ion trajectories of 390 MeV/u Ar^{17+} ions injected in the parallel direction to the (022) plane of the 0.74 μm-thick Si crystal. The planar potential is represented by a sum of atomic potentials, for which we adopted the Moliére potential with thermal vibration folded. The position x is defined from the channel center in the direction perpendicular to the (022) plane.

We observed RCE of them in the {220} planar-channeling condition by changing the tilt angle, θ, from the direction of the $\langle 110 \rangle$ axis in the {220} plane keeping the ion velocity v constant [2,5]. The resonance condition for the transition energy ΔE is given by $\Delta E = h\nu$, where ν is the frequency of the oscillating field induced by an array of the atomic strings on the planes in the case of RCE in the planar-channeling condition. The transition energy ΔE under the resonance condition is associated with the tilt angle θ by the equation of

$$\Delta E = h\gamma v(k\cos\theta/(a/\sqrt{2}) + l\sin\theta/a), \quad (1)$$

where a is the lattice constant of a Si crystal, h Planck's constant, and γ the Lorentz factor. A two-dimensional Miller index, (k, l), specifies the atomic string. We studied the transitions from the 1s to the $n = 2$ states of the ions under the $(k, l) = (2, 1)$ resonance through the measurements of the exit charge state and the exit angle of the ions simultaneously in a list mode, and the measurements of the de-excitation X-rays under the $(k, l) = (1, 1)$ resonance. The difference between $(k, l) = (2, 1)$ and $(1, 1)$ does not basically affect our results and discussion below.

2. Experimental

The experiments were performed at heavy ion medical accelerator in Chiba (HIMAC), and the experimental setup was already described elsewhere except the collimator conditions [5,6]. The tightly-collimated ions passed through the thin Si crystal mounted on a goniometer. The Si crystal was placed so that the {220} plane was horizontal. A 50 mm-thick collimator with a hole of 0.6 mm in diameter was located 650 cm upstream of the target crystal. Another 50 mm-thick horizontal slit of 0.2 mm in width were added to reduce the divergence in the vertical direction at the position 50 cm upstream. The exit charge state and the exit angle of the individual emerged ions were measured in a list mode with a combination of a charge separation magnet and a two-dimensional position-sensitive Si detector (2D-PSD) located 120 cm and 550 cm downstream of the target, respectively. Note that the charge state is measured in the horizontal direction and the exit angle in the vertical direction by the 2D-PSD. Thus, the beam divergence was reduced down to 0.04 mrad and the beam spot size on the 2D-PSD was 0.3 mm both in the horizontal and vertical directions.

Emitted X-rays were detected by a Si(Li) X-ray detector placed on the horizontal plane at an angle of 41° with respect to the beam direction [7,8]. The angle corresponds to about 90° in the projectile frame through the Lorentz transformation. That is, we measured X-rays emitted from the Ar ions into the directions nearly parallel to the {220} plane in the projectile frame. The intensity of the incident ions for the X-ray measurements was monitored by Kα X-rays from a thin Cu-foil placed at the downstream end.

In the manufacturing process of the thin Si crystal of about 10 mm in a diameter, its surface normal direction is selected to be the [1 0 0] direction. Nonuniformity in thickness was measured to be about ±0.15 μm through the energy loss of alpha particles emitted from ^{241}Am. We utilized the center area of the Si crystal, where the thickness is 0.74 μm. We adopted two different effective thicknesses of the same target crystal in the {2 2 0} planar-channeling conditions. One is 0.74 μm for the incident beam direction close to the [1 0 0] direction in the (0 2 2) planar-channeling condition. The other is 1.05 μm for the incident beam direction close to the [$1\bar{1}0$] direction in the (2 2 0) planar-channeling condition, for which we tilted the crystal by about 45°. The tilt angle θ is defined as the incident beam direction with respect to the [$01\bar{1}$] and [$1\bar{1}0$] axes in the former and latter cases, respectively.

3. Results and discussion

We first investigated the survived Ar^{17+} fraction of the emerged ions from the 0.74 μm-thick crystal as a function of the tilt angle ϕ defined to be orthogonal to θ across the (0 2 2) plane under the non-RCE condition. As shown in Fig. 2, the survived Ar^{17+} fraction has a peak of 0.86 in the (0 2 2) planar-channeling condition. As ϕ moves away from the channeling condition, the Ar^{17+} fraction decreased down to 0.77 at $\phi = 0.015°$, and again recovers to a constant value of 0.79 at $\phi > 0.020°$, where the ions were not channeling. The value in the non-channeling condition is consistent with the mean free path (mfp) of the collisional ionization of 4.3 μm for the non-excited ions which was theoretically calculated [11]. The small dips appeared at both sides of the channeling peak originate from the so-called quasi-channeling: the ion travels along the atomic plane, where the densities of electrons and nuclei are higher than in the non-channeling condition and lead to the enhancement of the collisional ionization.

Fig. 3(a) shows the θ dependence of the survived Ar^{17+} fraction of the emerged ions from the 0.74 μm-thick crystal under the $(k, l) = (2, 1)$ resonance in the (0 2 2) planar-channeling condition. We observed a broad and shallow resonance profile. The Ar^{17+} fraction decreased only by 2%

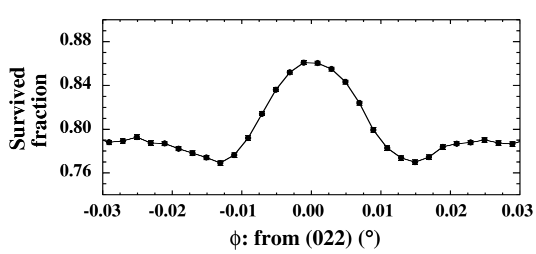

Fig. 2. Survived Ar^{17+} fraction of the emerged ions from the 0.74 μm-thick crystal under the non-RCE condition as a function of ϕ defined to be orthogonal to θ across the (0 2 2) plane. The condition of $\phi = 0$ corresponds to the planar-channeling.

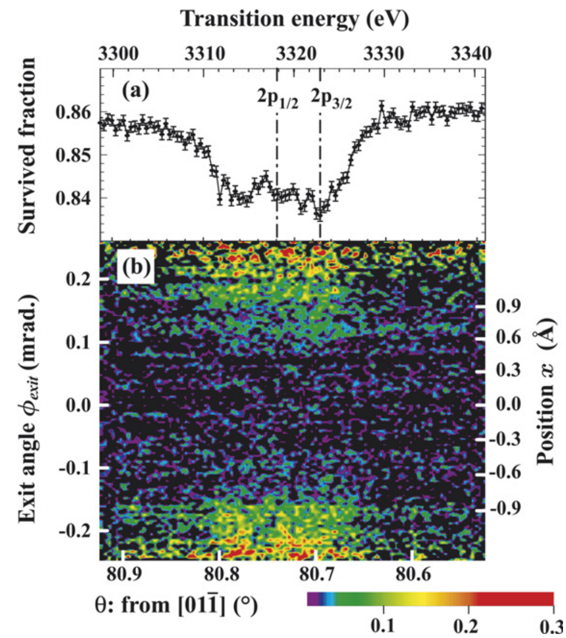

Fig. 3. (a) θ dependence of the survived Ar^{17+} fraction of the emerged ions from the 0.74 μm-thick crystal under the $(k, l) = (2, 1)$ resonance in the (0 2 2) planar-channeling condition. (b) A contour map of the fraction of RCE with the subsequent ionization, $F(\theta, \phi_{exit})$ at the incident angle θ and the exit angle of the emerged ions ϕ_{exit}.

at most from 0.86, and the fine structure of the $2p_{1/2}$ and $2p_{3/2}$ states, previously observed for the thicker crystals (21 and 94.7 μm) in the same conditions, is not resolved clearly [5–8]. We here give a qualitative explanation of this difference as follows. Since the crystal thickness is reduced by twenty times or more, the probabilities of RCE and the subsequent ionization of the channeled ions are, as a whole, depressed to a large extent, which results in the shallow resonance profile. The observed resonance profile of the charge state emphasizes the ion trajectories close to the atomic plane in the crystal, where the subsequent ionization probability of the excited ions by the collisions with the target electrons or nuclei is large. These ions feel the strong static crystal field inducing the large Stark shift, which results in the broadening of the resonance profile. On the other hand, the excited ions traveling not close to the atomic plane escape from the ionization and decay into the ground state by emitting X-rays as explained later.

Then, the relation between the exit charge state, namely the Ar^{17+} fraction and the exit angle of the emerged ions ϕ_{exit} at the incident angle θ was examined by the list mode measurements. The results are shown in Fig. 3(b) as a contour map of the fraction of RCE with the subsequent ionization $F(\theta, \phi_{exit})$, which is defined as $F(\theta, \phi_{exit}) = 1 - f(\theta, \phi_{exit})/f(\theta_{off}, \phi_{exit})$. Here $f(\theta, \phi_{exit})$ is the survived fraction of the emerged ions with the exit angle ϕ_{exit} observed at the incident angle θ and $f(\theta_{off}, \phi_{exit})$ is that observed under the non-RCE condition. Namely, $F(\theta, \phi_{exit})$ shows the probability for ions with exit angle ϕ_{exit} to be ionized subsequent to RCE at a specific θ normalized to the survived

Ar^{17+} fraction under the non-RCE condition. We found that RCE with the subsequent ionization obviously takes place at the large exit angles corresponding to the ion trajectories close to the atomic plane where the severe Stark shift occurs, which is consistent to the explanation above. We point out that the ambiguity in the exit angle measurements comes mainly from the finite beam divergence of 0.04 mrad and the beam spot size of 0.3 mm, and they are equivalent to 0.2 Å of the position x, which is unfortunately non-negligible compared with the width of the {220} planar channel of 1.92 Å.

Furthermore, we also measured the de-excitation X-ray yield of the emerged ions from the 1.05 μm-thick crystal as a function of θ under the $(k, l) = (1, 1)$ resonance in the (220) planar-channeling as well as the exit charge state measurements. We multiplied the X-ray yields by $8\pi/3$ assuming the X-ray emission through the dipole transition to estimate the lower limit of the total X-ray yield. As seen in Fig. 4, the difference of the resonance profiles of the charge state between the thicknesses of 0.74 and 1.05 μm is small except a little enhancement of the resonance dip for the latter case. However, the resonance profile of the X-ray yield per incident ion shows significant differences from the resonance profile of the exit charge state. First, the resonance profile is characterized by two peaks reflecting the fine structure of $2p_{1/2}$ and $2p_{3/2}$ states, and the doublet structure is folded to the $2p_{1/2}$ peak. Second, the X-ray yields are large, and 0.15 photons are emitted per incident ion for the $2p_{3/2}$ peak after subtracting the non-resonant component.

The traveling path length of the ions for the transition from the ground state to the excited state by RCE is theoretically estimated to be of a few μm depending on the ion trajectories. The spontaneous decay rate of the 2p excited state is $6.67 \times 10^{13}\ s^{-1}$ corresponding to the mfp of 4.6 μm. It is noted that attenuation length of the de-excitation X-rays with energy of 5.0 keV, which is heavily-Doppler shifted from the intrinsic energy of 3.3 keV, is 16 μm, and all of the X-rays practically penetrate the 1 μm-thick crystal. The mfp of collisional ionization of the Ar^{17+} ions is estimated to be 1.0 μm for the excited (2p) ions in the random incidence from the calculation [11] and it is expected to be longer in the planar-channeling condition. Taking all of them in account, we conclude that RCE occurs with a fairly large probability within the about 1 μm-thick crystal. A small fraction of the once excited ions by RCE are ionized by the collisions with the target electrons or nuclei. A majority of the excited ions by RCE de-excite into the ground state by emitting X-rays not only in the crystal but also in the vacuum. This channel is favored for the trajectory in the region of the channel center, where the Stark effect does not heavily distort the spectra. However, it is noted that the obviously observed doublet of the $2p_{1/2}$ peak in the resonance yield by the de-excitation X-rays is resulted from the Stark effect, and the smallness of excitation just in the midst of the channel center.

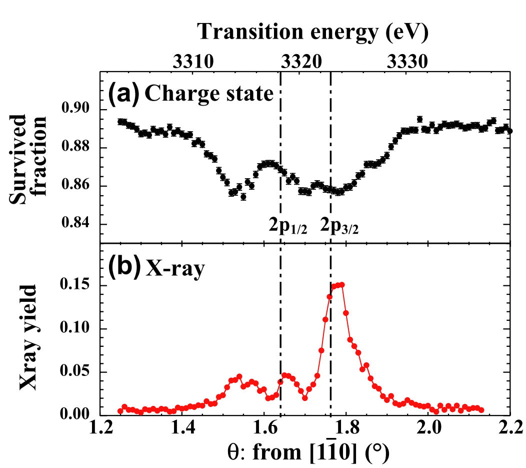

Fig. 4. θ dependences of (a) the survived Ar^{17+} fraction and (b) the yield of de-excitation X-rays of the emerged ions from the 1.05 μm-thick crystal under the $(k, l) = (1, 1)$ resonance in the (220) planar-channeling condition. Note a large difference of the scale of the y-axis in (a) and (b).

4. Summary

We observed RCE of 1s electron to the $n = 2$ states in 390 MeV/u Ar^{17+} ions passing through the thin Si crystal of about 1 μm in the {220} planar-channeling condition by measuring both the exit charge state and exit angle of the emerged ions simultaneously in a list mode. The yield of the de-excitation X-rays was also measured. We found the following features.

(1) RCE takes place with a probability of more than 15% at the maximum within the about 1 μm-thick crystal.
(2) A small fraction of the excited ions by RCE are ionized by the collisions with the target electrons or nuclei. The collision probability is large for the case that the ion travels close to the atomic plane where the densities of target electrons or nuclei are high. Accordingly, the excited states suffer the severe Stark shift resulting in the broad resonance profile of the charge state. The ion trajectory dependence of this feature is clearly observed in the relation between the exit charge state and the exit angle of the emerged ions.
(3) A majority of the excited ions by RCE de-excite into the ground state by emitting X-rays. This channel is favored for the trajectory not close to the atomic plane but in the region of the channel center, where the Stark effect does not heavily distort the spectra. However, the observed doublet of the $2p_{1/2}$ peak in the resonance yield by the de-excitation X-rays is resulted from the Stark shift, and the smallness of excitation just in the midst of the channel center.

Thus, we obtained the information on the trajectory dependent dynamics of the ion states relevant to the RCE process as well as ionization and de-excitation, and we indeed conclude that RCE occurs within the about 1 μm path length.

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References

[1] V.V. Okorokov, Yad. Fiz. 2 (1965) 1009 [Sov. J. Nucl. Phys. 2 (1966) 719].

[2] H.F. Krause, S. Datz, Adv. Atom. Mol. Opt. Phys. 37 (1996) 139.

[3] F.J. García de Abajo, V.H. Ponce, Adv. Quantum Chem. 46 (2004) 65.

[4] C. Cohen, D. Dauvergne, Nucl. Instr. and Meth. B 225 (2004) 40.

[5] K. Komaki, T. Azuma, T. Ito, Y. Takabayashi, Y. Yamazaki, M. Sano, M. Torikoshi, A. Kitagawa, E. Takada, T. Murakami, Nucl. Instr. and Meth. B 146 (1998) 19.

[6] T. Azuma, T. Ito, K. Komaki, Y. Yamazaki, M. Sano, M. Torikoshi, A. Kitagawa, E. Takada, T. Murakami, Phys. Rev. Lett. 83 (1999) 528;
T. Azuma, T. Ito, Y. Yamazaki, K. Komaki, M. Sano, M. Torikoshi, A. Kitagawa, E. Takada, T. Murakami, Nucl. Instr. and Meth. B 135 (1998) 61;
T. Azuma, Phys. Scripta T80 (1999) 79;
Y. Nakai, T. Ikeda, Y. Kanai, T. Kambara, N. Fukunishi, K. Komaki, C. Kondo, T. Azuma, Y. Yamazaki, Nucl. Instr. and Meth. B 230 (2005) 90.

[7] T. Ito, Y. Takabayashi, K. Komaki, T. Azuma, Y. Yamazaki, S. Datz, E. Takada, T. Murakami, Nucl. Instr. and Meth. B 164–165 (2000) 68.

[8] T. Azuma, T. Muranaka, Y. Takabayashi, T. Ito, C. Kondo, K. Komaki, Y. Yamazaki, S. Datz, E. Takada, T. Murakami, Nucl. Instr. and Meth. B 205 (2003) 779.

[9] S. Datz, C.D. Moak, T.S. Noggle, B.R. Appleton, H.O. Lutz, Phys. Rev. 179 (1969) 315.

[10] T. Azuma, K. Komaki, Y. Yamazaki, N. Kakutani, S. Ninomiya, K. Maki, T. Takahira, M. Sekiguchi, T. Hattori, T. Hasegawa, Nucl. Instr. and Meth. B 115 (1996) 306.

[11] J. Rozet, C. Stephan, D. Vernhet, Nucl. Instr. and Meth. B 107 (1996) 67.