Anisotropic X-Ray Emission from Heliumlike Fe^{24+} Ions Aligned by Resonant Coherent Excitation with a Periodic Crystal Potential

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We have measured deexcitation x rays emitted from the resonant coherently excited 2^1P_1 state of heliumlike Fe^{24+} ions of 423 MeV/amu, planar channeling through a Si crystal. Large anisotropy in the angular distribution of deexcitation x-ray emission is observed: the x-ray emission in the direction parallel to the channeling plane is favored by a factor of 2 compared to the perpendicular direction. This anisotropy originates from the direction of the periodic crystal field, which populates specific m states in resonant coherent excitation and aligns the excited states.

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A crystal is a regular arrangement of atoms located on lattice positions. Energetic ions traveling through a crystal along an axis or plane can be trapped in the static continuum potential well formed by atomic strings or planes; i.e., the ions are "channeled" in the crystal. A channeled ion experiences less scattering and energy loss than an ion traveling through the crystal in a "random" direction [1]. In addition, when the ion travels along a periodic array of atoms or across a periodic array of atomic strings on an atomic plane, it experiences a crystal field oscillating periodically in time in the projectile frame. When one of the frequencies of the oscillation corresponds to an atomic transition energy of the ion, ΔE, it has a chance to be resonant coherently excited (RCE), and this phenomenon is often called the Okorokov effect [2–6]. The oscillating crystal field has a character analogous to that of a photon, which is a field oscillating transversely, i.e., perpendicular to the direction of the propagation in vacuum. However, the oscillating crystal field in the projectile frame has both transverse and longitudinal components with respect to the propagation direction. The amplitude and the direction of polarization are determined by Fourier components of the periodic crystal potential. We have a chance to excite the ions into specific m states utilizing polarization of the oscillating crystal field and hence align the excited states. Since the corresponding energy of the oscillating crystal field practically covers the x-ray region for the high-energy projectiles, it is possible to excite $1s$ electrons of heavy ions by this technique.

We have been observing RCE of high-energy heavy ions in the transmission channeling condition for years. A sharp resonance is obtained for projectiles of moderately relativistic energy (several hundreds MeV/amu), since the collision process with target electrons that would otherwise destroy the coherence is suppressed to a large extent for that velocity regime. The RCE of a $1s$ electron to the $n = 2$ excited states in hydrogenlike or heliumlike ions is experimentally observed through two alternative processes. The ions in the excited states are more easily ionized by the target-electron impact in the crystal than in the ground state. Accordingly the RCE is observed as the increases both in the ionized fraction of the transmitted ions [7,8], and in the convoy electron yield emitted into the same direction as the projectile ion [9].

When the excited states do not suffer from ionization, they nonradiatively deexcite through the inverse process of RCE or radiatively deexcite to the ground state. The latter process is generally more favored for heavy ions due to the short lifetime of the excited state through spontaneous emission, where the transition rate is roughly proportional to Z^4 (Z: nuclear charge of ion) and its ionization cross section is proportional to Z^{-2}. Since the first observation of deexcitation x rays by Fujimoto *et al.* [10], several experiments by Datz *et al.* have been reported [11]. They challenged anisotropy measurements in the angular distribution of the x-ray emission, but a decisive conclusion had not been obtained. In those experiments, the analysis is complicated because the Stark effect, not only from the static crystal field, but also from the wakefield induced by the dynamic response of the target electrons, has to be taken into account in the chosen projectile energy region of MeV/amu. We have recently reported a precise measurement for the resonance profile of the deexcitation x rays from high-energy hydrogenlike Ar^{17+} ions under the RCE condition [12]. In addition, the x-ray yields in the two directions were also measured, although significant anisotropy was not observed [13].

In this Letter, we report observation of a large anisotropy in the angular distribution of the deexcitation x rays emitted from resonant coherently excited heliumlike Fe^{24+} ions. This observation is the first direct evidence of the aligned excited states produced by the inherently "polarized" crystal field. We observed RCE of 423 MeV/amu (i.e., 23.7 GeV, $\beta = 0.73$, $\gamma = 1.45$) heliumlike Fe^{24+} ions $(2\bar{2}0)$ planar channeled through a 21 μm-thick Si crystal by changing the incident angle of the ions θ with respect to the direction of the [110] axis in the $(2\bar{2}0)$ plane through rotating the crystal [3,8]. The transition energy, ΔE, under the resonance condition, is uniquely associated with the tilt angle by the equation of

$$\Delta E = h\gamma\beta c[k\cos\theta/(a/\sqrt{2}) + l\sin\theta/a], \qquad (1)$$

where a is the lattice constant of a Si crystal and c is the light velocity. The two-dimensional Miller indices, (k, l), specify the atomic string [8]. Since the transition energy of $1s$ electrons in heavy ions is large, an ion velocity of several hundreds MeV/amu is necessary to excite them with large probability. We observed the transition from the $1s^2$ to $1s2p$ of heliumlike Fe^{24+} ions under the $(k, l) = (2, -1)$ resonance through measurements of the deexcitation x rays from the Fe ions. The incident ion energy was carefully chosen so that the RCE condition does not overlap with those of the levels of Fe^{25+} ions produced by the target-electron impact on the incident Fe^{24+} ions in the crystal. The Fe^{24+} ions were provided at the Heavy Ion Medical Accelerator at Chiba (HIMAC).

As shown in Fig. 1, we set the $(2\bar{2}0)$ plane horizontal. Hereafter, we define the z axis as the direction of the ion velocity, the x axis perpendicular to the $(2\bar{2}0)$ plane, and the y axis normal to both of them. The channeled Fe ions travel in the z direction between two $(2\bar{2}0)$ atomic planes which are parallel to the $(y$-$z)$ plane guided by the static crystal electric field, i.e., the derivative of the planar continuum potential. Emitted x rays were detected by the two Si(Li) x-ray detectors placed on the vertical $(x$-$z)$ and horizontal $(y$-$z)$ planes at an angle of 41° with respect to the beam direction (z direction) as shown in Fig. 1 [12,13]. The angle corresponds to $\sim$90° in the projectile frame for both detectors (i.e., nearly on the x and y axes) through the Lorentz transformation. That is, we measured x rays emitted from the Fe ions into both directions nearly perpendicular (x direction) and parallel (y direction) to the $(2\bar{2}0)$ plane in the projectile frame. The intensity of the incident ions was monitored by $K\alpha$ x rays from a thin Cu foil placed at the downstream end. It was kept to be less than 10^6 cps to avoid the pileup effect in the preamplifiers for the x-ray detectors.

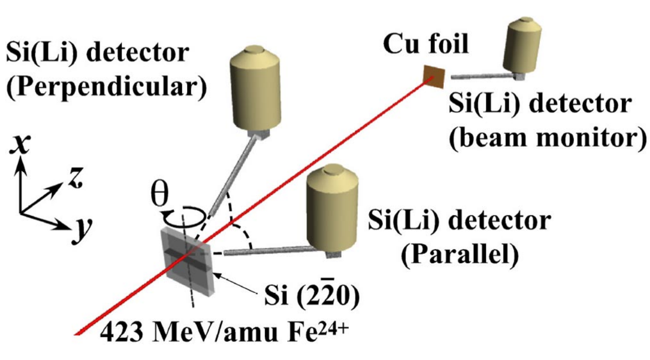

FIG. 1 (color online). Schematic layout of the experimental setup. The $(2\bar{2}0)$ plane of the Si crystal is set horizontal. The crystal is rotated as a function of the tilt angle, θ, from the [110] axis, the surface normal, in the horizontal $(2\bar{2}0)$ plane.

The crucial issue on the RCE of the planar channeled ions is that the amplitude and the direction of the oscillating crystal field are functions of the ion position, X, i.e., the distance from the channel center in the x direction. Furthermore, we have to take account of the effect of the static crystal field on the electrons bound to the ion, i.e., the Stark effect. In Fig. 2(a), we present squared transition matrix elements, which are proportional to transition probabilities, for excitation of one of the two ground state electrons $1s^2$ of Fe^{24+} ions to $2s$, $2p_x$, $2p_y$, and $2p_z$ as a function of the ion position X for the $(k, l) = (2, -1)$ resonance. They are calculated by adopting the periodic crystal potential directly as a perturbation without dipole approximation. The transition probability to $2s$ is negligibly small. We naïvely regarded that electron excitation of $1s^2$, $1^1S_0 \rightarrow 1s2p$, 2^1P_1 is properly dealt with in a manner similar to hydrogenlike ions by assuming that the nuclear charge is completely screened by the other bound electron, and electron-electron correlation is neglected [8]. In general, the oscillating crystal field felt by the ions has a tendency to increase in the region near the atomic plane, i.e., larger X. In the $(k, l) = (2, -1)$ case, both the x and z components of the oscillating crystal field are always stronger than the y component, which is directly reflected in the relative magnitude of the transition matrix elements. In the case of heliumlike ions, the Stark mixing between $1s2s$ and $1s2p$ states is small due to the large difference in their unperturbed level energies but removes degeneracy among $1s2p_x$, $1s2p_y$, and $1s2p_z$ states. The static crystal field intermixes $1s2s$ and $1s2p_x$ to a small extent depending on the ion position X. It is noted that the dynamic wakefield is quite weak in the present energy region but it further intermixes $1s2p_z$ slightly with these two components. Thus we have excited singlet states consisting of one isolated and three nearly degenerate levels, the former being mostly composed of $1s2s$ 2^1S_0. Two of the latter

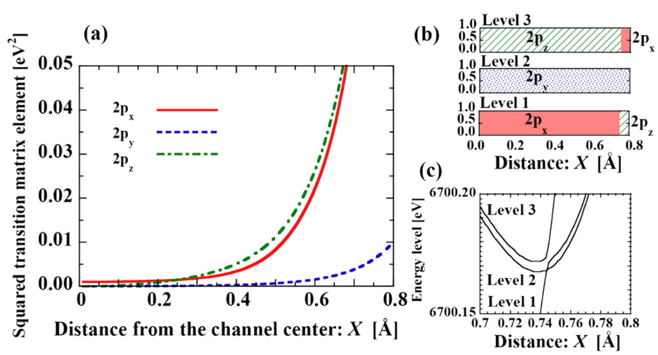

FIG. 2 (color online). (a) Squared transition matrix elements for $2p_x$, $2p_y$, and $2p_z$ components of the $n = 2$ states for the $(k, l) = (2, -1)$ resonance, and (b) their relative fraction in Level 1–3 of 2^1P_1 as a function of the distance from the channel center, X, in the planar channel. (c) Energy levels of the Level 1–3 in the region close to an avoided crossing as a function of X. The distance from the channel center to the $(2\bar{2}0)$ atomic plane is 0.96 Å.

are mostly composed of $1s2p_x$ and $1s2p_z$, respectively, while the other of the latter is composed purely of $1s2p_y$. Hereafter we call these three sublevels of the excited state, 2^1P_1, as Level 1–3 in the increasing order of energy eigenvalue at the channel center. Figure 2(b) shows the fraction of $2s$, $2p_x$, $2p_y$, and $2p_z$ components in eigenstate Level 1–3 of 2^1P_1 as a function of the distance X from the channel center. It is noted that the $2p_x$-rich and $2p_z$-rich natures are exchanged between Level 1 and 3 through an avoided crossing at the ion position $X = \sim 0.74$ Å when the ion travels in the x direction. Nevertheless, the nature of the wave function is shown to remain unchanged via the diabatic transition between two sublevels owing to the small splitting and the large ion velocity in the x direction according to the Landau-Zener criteria. Thus, the 2^1P_1 state of the heliumlike ions practically consists of three degenerate levels of $2p_x$, $2p_y$, and $2p_z$ in the crystal. In addition, we note that mixing of singlet and triplet states for heliumlike Fe^{24+} is $\sim$10% due to the jj coupling nature. However, through a relativistic configuration interaction (CI) type calculation, we confirmed that general tendency of fraction of $2s$, $2p_x$, $2p_y$, and $2p_z$ components as a function of the field strength is explained by the present crude approximation [14].

When we detect the deexcitation x rays in the parallel direction, we observe x rays emitted mainly from $2p_x$ and $2p_z$ levels, because x rays due to the dipole transition are emitted preferentially in the perpendicular direction with respect to the dipole. On the other hand, we observe x rays emitted mainly from $2p_y$ and $2p_z$ levels, when we detect them in the perpendicular direction. As a whole, we expect larger x-ray yield in the parallel direction than in the perpendicular direction.

The x rays emitted from the Fe ions were observed at 10.2 keV with the Si (Li) detectors, an energy which is $\sim$50% Doppler shifted from the intrinsic transition energy due to the ion motion. Typical examples of the raw spectra detected by the Si(Li) x-ray detector have been shown in [12]. Since the attenuation length of a 10.2 keV x ray in silicon, 145 μm [15], is sufficiently greater than the target crystal thickness, absorption is negligibly small. Figure 3 shows the yield of the deexcitation x rays of 10.2 keV as a function of the tilt angle, which relates to the transition energy by Eq. (1). An increase in the x-ray yield under the resonance condition corresponding to $1s^2$, $1^1S_0 \rightarrow 1s2p$, 2^1P_1 was clearly observed together with the small peak corresponding to 2^1S_0 or 2^3P_1. The origin of the latter peak is complicated: the 2^3P state is mixed with 2^1P as already described, and the direct 2^1S excitation through the nondipole transition associated with the Stark mixing may contribute to this peak, the detail of which will be discussed elsewhere. The two arrows in Fig. 3 show the theoretical transition energies in vacuum [16]. The most striking feature we found is that the x-ray yield in the direction parallel to the $(2\bar{2}0)$ channeling plane is larger than in the direction perpendicular by a factor of 2 in the

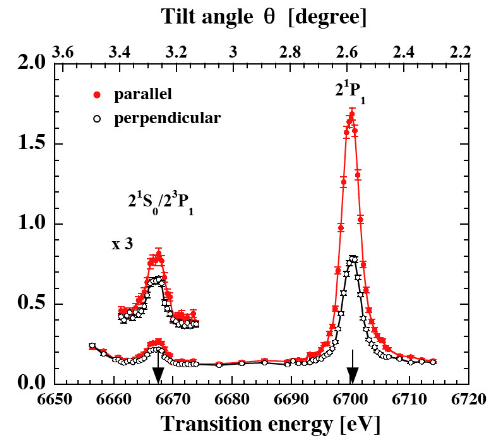

FIG. 3 (color online). The RCE profiles of $(k, l) = (2, -1)$ resonance in $(2\bar{2}0)$ planar channeling through a 21 μm-thick Si crystal as a function of the tilt angle, θ, from the [110] axis in the horizontal $(2\bar{2}0)$ plane for deexcitation x rays from Fe ions measured with the Si(Li) detectors at the parallel position to the plane (●), and at the perpendicular position (○).

resonance condition for 2^1P_1. The present large anisotropy originates from the fact that the oscillating crystal field in the x direction is much stronger than in the y direction as already described, which leads to a selective transition to the $2p_x$ level with large probability through RCE and alignment of the excited states.

We point out that intrashell mixing due to target-electron impact does not play a crucial role, since this effect would otherwise obscure the initial alignment of the excited states. The direct dipole transition between three degenerate states of heliumlike ions is not allowed. Accordingly intrashell mixing occurs only through two-step transition like $2p_x \rightarrow 2s \rightarrow 2p_y$, which is considered not to occur frequently prior to x-ray emission.

The angular distribution of the x-ray emission through the dipole transition from $2p_x$ and $2p_z$ components obeys the $\sin^2\theta$ law, and the transition to $2p_y$ is small as shown in Fig. 2(a). Thereby, we multiplied the x-ray yield in the parallel direction by $8\pi/3$ to estimate the absolute total x-ray yield roughly. This procedure results in 1.7 photon emission per incident ion for the 2^1P_1 peak after subtracting the nonresonant component. It implies the Fe ions experience excitation and deexcitation once on the average during the passage through the 21 μm-thick crystal due to the short lifetime of the 2^1P_1 excited state [4.57×10^{14} s^{-1} [16] corresponding to the mean free path (mfp) of 0.69 μm] and large RCE probability. The mfp of RCE is estimated to be in the order of a few μm. Another decay process, i.e., ionization, depends strongly on the local density of target electrons along the trajectory. However, the ionized fraction of 0.13 per incident ion at the peak of the resonance profile measured through the charge state distribution of the transmitted ions suggests that deexcitation is much preferred to ionization. We performed a crude Monte Carlo simulation of RCE and x-ray emission taking these effects into account and obtained reasonable agreement with the observed tendency. Alignment effect on ionization probability by target-electron impact is so far not considered because of the minor contribution of ionization as well as the small size of the bound electron cloud of the Fe^{24+} ion compared with the width of the planar channel of 1.92 Å, but a future work elucidating this effect is desired.

Here we briefly mention the absence of anisotropy in x-ray emission of hydrogenlike Ar^{17+} ions previously reported [13]. The $n = 2$ states are split into two states by the $l \cdot s$ interaction and further into four states by the Stark effect. Each of them is composed of $2s$, $2p_x$, $2p_y$, and $2p_z$. The relative fraction of these components varies depending on the ion position in a complicated manner. The deexcitation x rays are emitted according to the relative fraction of these components at the moment of emission, which does not necessarily reflect the polarization of the oscillating crystal field. Compared with heliumlike Fe^{24+} ions, the longer lifetime of the excited state and collisional intrashell-mixing effect also contribute to the absence.

In conclusion, we studied RCE of 423 MeV/amu heliumlike Fe^{24+} ions $(2\bar{2}0)$ planar channeled through a Si crystal. The deexcitation x rays emitted from resonant coherently excited Fe^{24+} ions into the direction parallel to the channeling plane are twice as intense as those emitted into the plane perpendicular. The observed anisotropy in the angular distribution originates from the direction of the oscillating crystal field: the component in the perpendicular direction to the channeling plane is larger than the parallel component regardless of the ion position X. Therefore, the larger RCE probability in the perpendicular direction to the channeling plane results in alignment of the excited states. The $n = 2$ states of heliumlike ions are free from the severe Stark mixing usually associated with the static crystal field. In addition, heavy ions have a short lifetime of the excited state and small cross sections for ionization and intrashell mixing. These features contribute to the large anisotropy. Recently we have started extensive anisotropy measurements of x rays emitted from resonant coherently excited hydrogenlike and heliumlike Ar, Fe, and Kr ions, and we obtained general trends supporting our conclusion.

Observation of such phenomena related with photon polarization in the x-ray energy region has been limited. A few possibilities like polarized light from synchrotron orbital radiation using undulator devices or x-ray lasers are listed as candidates for polarized x-ray sources. The present RCE technique readily prepares aligned ions, and this opens a new field of interesting research in future: applying the unique capabilities of RCE for investigation of alignment of highly charged heavy ions.

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