Doubly-resonant coherent excitation of HCl planar channeled in a Si crystal

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Abstract. We investigated resonant coherent excitation of H-like Ar¹⁷⁺ and He-like Ar¹⁶⁺ ions planar channeled in a Si crystal under the V-type and ladder-type double resonance conditions. In both cases, we observed distinct enhancement in the ionized fraction of the transmitted ions when the double resonance conditions were satisfied. In the ladder-type configuration, the enhancement indicates that the doubly-excited 2p² state of He-like Ar¹⁶⁺ was produced through doubly-resonant coherent excitation.

1. Introduction

When energetic ions interact with a crystal, one can see characteristic phenomena attributed to the anisotropic or periodic structure of the crystal. In the case that the incident ion beam is parallel to an atomic plane, some of the ions travel in the inter-planar spacing without traversing the atomic planes. This effect is well known as planar channeling, reflecting the anisotropic structure. In addition, the periodicity of the crystal induces resonant coherent excitation (RCE), in which the traveling ions are excited by feeling the periodic oscillating field in the crystal. For decades, RCE has been studied with few-electron ions/atoms (Z = 1–26) under axial, planar and surface channeling conditions [1, 2]. However, observation of RCE is so far limited to two level atomic systems driven by a monochromatic oscillating field, although the field in reality consists of numerous frequency components. In this report, we demonstrate RCE in three-level systems using two frequency components of the field simultaneously, to which we hereafter refer as doubly-resonant coherent excitation (dRCE).

In the planar channeling conditions, the ions feel the oscillating field by traversing the arrays of atomic strings. Each array is specified by a two-dimensional reciprocal lattice vector gₖ,ₙ, where (k, l) is the Miller index of the 2D crystal (atomic plane). Then the corresponding frequency component is expressed as ωₖ,ₙ = γgₖ,ₙ⋅v, where v is the ion velocity and γ is the Lorentz factor. In the case of the (220) planar channeling through a diamond structured crystal, the resonance
condition for a transition of energy $\Delta E$ is written as

$$\Delta E = h\nu_{k,l}(v; \theta) = \frac{h\gamma v}{a} \left( \sqrt{2k \cos \theta + l \sin \theta} \right),$$

(1)

where $h$ is Planck’s constant, $a$ is the lattice constant and $\theta$ is the incident angle with respect to the [110] axis [3]. Even in a fixed condition of $v$ and $\theta$, we can expect a variety of $h\nu_{k,l}$ depending on a pair of $(k, l)$. When two of these $h\nu_{k,l}$ match with two different transition energies between three levels of the system, dRCE occurs. In order to satisfy the dRCE conditions, the incident energy of the ion has to be tuned with the accuracy of 0.01% considering the resonance width. Since we can measure the incident energy by the RCE spectrum of a well-known transition, the beam energy was precisely adjusted by tuning the radio frequency of the synchrotron accelerator in five or more digits.

2. Experiment

Ar$^{17+}$ and Ar$^{16+}$ ion beams of $\sim 400$ MeV/u were supplied from HIMAC (the heavy ion medical accelerator in Chiba), which was well collimated to be less than 0.05 mrad (rms) in divergence both in horizontal and vertical directions. The ions travel through a 27 $\mu$m-thick Si crystal for about $1 \times 10^{-14}$ s and 30% of them emerge with the initial charge state under the (220) planar channeling condition. When RCE takes place, the survived fraction of the transmitted ions decreases because the excited ions lose their electrons more easily than the ground-state ions by the collisions with target electrons in the crystal. Note that both cross sections of mechanical electron capture (MEC) and radiative electron capture (REC) are negligibly small in that high-velocity regime, although the latter process is more favored than the former.

We performed experiments in V-type and ladder-type three-level systems as illustrated in Fig. 1. In the V-type double resonance, we employed the frequency components specified by $(k, l) = (1, -1)$ and $(1, 3)$ for the transitions $1s - 2p_{3/2}$ and $1s - 3p_{3/2}$, respectively. In the ladder-type double resonance, we tried to produce the doubly-excited $2p^2$ state by the successive excitation of $1s^2 - 1s2p$ and $1s2p - 2p^2$ using the components of $(k, l) = (1, -2)$ and $(1, 1)$, respectively. It is noted that the second excitation must occur within the finite lifetime of the intermediate state in the ladder type configuration, while both transitions are from the ground state in the V-type configuration.

**Figure 1.** Energy diagrams of (a) V-type and (b) ladder-type configurations of H-like Ar$^{17+}$ and He-like Ar$^{16+}$. The solid arrows represent the resonant processes (resonant coherent excitation/de-excitation) with the employed Miller index $(k, l)$. The dotted arrows represent spontaneous decay with their lifetimes.
3. Results and Discussion

3.1. V-type

The double resonance condition was calculated to be \( E = 438.69 \text{ MeV/u} \) and \( \theta = 3.57^\circ \), where \( E \) is the incident energy of the ions corresponding to the incident velocity \( v \). The incident energy for the single resonance measurement was fixed to be 440.60 MeV/u, slightly detuned from the double resonance condition. We show the survived fraction of \( \text{Ar}^{17+} \) as a function of \( \theta \) under the single resonance measurement in Fig. 2. The transition energies corresponding to the oscillating field for \((k,l) = (1,-1)\) and \((1,3)\) are indicated in the figure. RCE from \( 1s \) to \( n = 2 \) and \( n = 3 \) were individually observed as decreases in the survived fraction because the ionization probability becomes higher when RCE occurs. The resonance profiles show a complicated structure due to the Stark effect induced by the static planar potential of the \((220)\) planes [3].

When we precisely tune the incident energy of the ions to be 438.69 MeV/u, the resonance conditions for \( 1s - 2p_{3/2} \) and \( 1s - 3p_{3/2} \) are simultaneously satisfied at \( \theta = 3.57^\circ \) if the Stark shift is not taken into account. The two individual spectra of \( n = 2 \) and \( n = 3 \) overlap with each other and the ionized fraction is enhanced as shown in Fig. 3. This enhancement indicates that the population of the excited states are increased due to dRCE. We point out that RCE of \( 2p_{3/2} - 3d_{3/2} \) also occurs by the \((k,l) = (0,4)\) frequency component under the present double resonance condition due to the finite resonance width, which would somewhat contribute to the enhancement of the ionized fraction.

3.2. Ladder type

In the ladder type double resonance, the incident energy for dRCE was determined to be 387.90 MeV/u. We again slightly detuned the incident energy for single resonance measurement to 388.54 MeV/u as in the case of the V-type double resonance. In Fig. 4, we show the \( \text{Ar}^{16+} \) survived fractions for the single resonance and double resonance measurements as a function of \( \theta \) with different \( x \)-axes. The \( x \)-axes are adjusted so that the resonance dips of \( 1s^2 - 1s2p \) locate at the same position. The Stark effect is not prominent in this case because the energies of the unperturbed states of He-like ions are sufficiently far from each other. In the double resonance measurement, the resonance conditions for \( 1s^2 - 1s2p \) and \( 1s2p - 2p^2 \) are simultaneously satisfied at \( \theta = 1.23^\circ \), that is, the dRCE condition. Note that no resonance dip appears at the resonance condition solely for \( 1s2p - 2p^2 \) because the population of the initial \( 1s2p \) state is negligibly small in the case of the single resonance condition. The ladder-type double resonance results in an
enhancement of the resonance by a factor of 1.4. This decrease of the survived fraction at the resonance confirms the production of the $2p^2$ state in the crystal because the cross section of collisional ionization for $2p^2$ state is larger than that for $1s2p$ states. In addition, the doubly-excited state has a channel of auto-ionization through the Auger process within a lifetime of $3.1 \times 10^{-15}$ s [4].

In summary, we succeeded in observing doubly-resonant coherent excitation (dRCE) in both V-type and ladder-type three-level systems. Our result demonstrates that multiple resonances in the x-ray region are possible by the multi-colored crystal field. This method can be utilized for the production of hollow atoms or metastable ions such as $^1S$ states. Finally we remark that we can vary the frequencies of the oscillating field by two-axis control of the crystal using RCE under non-channeling conditions (3D-RCE) [5]. This allows us to satisfy the double resonance condition readily without tuning the incident energy.

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References