Density matrix description of resonant coherent excitation of swift highly charged ions in oriented crystals

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Abstract. We report selected results of our latest resonant coherent excitation studies obtained within density matrix approach and present suggestions for new RCE experiments.

1. Introduction
Swift ion in matter is an open quantum system involved into both coherent and incoherent interactions with its surroundings. Starting from this understanding, we (contrary to theoretical approaches of other groups [1–5]) take advantages of density matrix formalism to describe resonant coherent excitation (RCE) of multiply charged ions in crystals and apply it to analysis of a number of experiments. Beginning our studies from the RCE of light ions at rather low energy [6, 7], the density matrix approach allowed us to achieve a unified description of both characteristics of the RCE process known from the pioneering papers of Okorokov [8] and early RCE experiments [9,10]. These are the charge state distribution at the exit of the target (survival fraction) and yield and angular distribution of the characteristic X-ray radiation produced by excited ions. Current experiments with relativistic highly charged ions reveal some new aspects of RCE process. We extend our approach in accordance with these new trends.

2. Density matrix analysis of recent RCE experimental data
2.1. Alignment of channeled RCE ion and angular anisotropy of its X-ray radiation
Polarization of the electric field of the crystal lattice leads to alignment of the angular momentum of resonant coherently excited ions. General procedure of our calculations [6, 7] begins with solving the generalized Master equation to obtain time evolution of the density matrix for individual ion trajectories. Then, averaged over the whole ensemble of ions taking part in the RCE process, it is projected on the subspace of ionic states emitting photons whose yield, angular distribution and polarization are then calculated by using standard density matrix (statistical tensors) technique [11]. A strong evidence of the alignment effect was obtained in 2006 by the Tokyo RCE collaboration [12] by the measurements of angular asymmetry of photon yield from 423 MeV/u Fe²⁴⁺ ions in (220) planar channel of 21 µm-thick Si crystal. Our calculation showed close agreement with these data [13]. To go further to the origin of this effect, we undertook
detailed investigation [14] of how certain geometrical properties of the in-crystal electric field manifest themselves in these X-ray observables.

2.2. RCE resolved by ion trajectories
Resolution of survival fraction RCE data by ion trajectory was done in [18] by using a thin (∼1 μm) silicon crystal of thickness corresponding to one-quarter of the period of Ar$^{17+}$ ion transverse oscillations in the channel. Being motivated by this work, we performed calculations [14] of trajectory resolved anisotropy of X-rays from 423 MeV/u Fe$^{24+}$ ions in (220) channel of 0.9 μm Si crystal, coherently excited at the same (k, l) = (2, −1) resonance as in work [12] on the angular anisotropy of X-rays from Fe$^{24+}$ ions in thick target. Calculation results show the origin of the horizontal (in the plane) to vertical (perpendicular to the plane) photon yield asymmetry ratio $H/V \approx 2$ observed in the experiment.

Experimental verification of these results demands coincident detection of the characteristic X-ray photons with emerging ions resolved by the deflection angle [10]. Inspiring results of recent photon-ion coincidence experiments [15] in near-lying domains of physics of highly charged ions support the idea of similar coincidence measurements in RCE studies as well.

2.3. RCE without channeling (3D RCE)
The recent experiment [16] proved the possibility of observation of RCE without channeling. When ions are sent through the crystal in non-channeling directions ($\theta_t$, $\phi_t$), tuning to any specific (k, l, m) harmonic may be achieved according to a general Okorokov resonance condition

$$\Delta E = \hbar \gamma (\mathbf{G}_{klm} \cdot \mathbf{v}) = \frac{2\pi \hbar}{a} \gamma v \left( \sqrt{2}(k \cos \phi_t + m \sin \phi_t) \cos \theta_t + l \sin \theta_t \right).$$

We performed calculations for 391 MeV/u Ar$^{17+}$ ions passing through 1 μm Si crystal. In Fig.1 we compare our results with the data [16] for the survival fraction. Considerable angular anisotropy is expected for corresponding in-target and out-of-target characteristic X-ray radiation.

3. Theoretical arguments for new RCE measurements
3.1. Auger electrons from double RCE
The experiment [19] on “ladder type” double RCE of autoionizing states of helium-like Ar$^{16+}$ ions passing through the (220) planar channel of a 27 μm-thick Si crystal connects physics of autoionization phenomena and channeling. Our calculation [14] performed for 387.90 MeV/u Ar$^{16+}$ ions, planar channelled in Si crystal and resonantly excited under simultaneous action of

![Figure 1](image-url)

**Figure 1.** Calculated survival fraction (normalized) and differential X-ray yield under 3D RCE (k, l) = (1, 1) conditions for 391 MeV/u Ar$^{17+}$ in 1 μm thick Si crystal. Differential X-ray yield (summed contributions from excited ions decay inside and outside the target) is presented in two directions: in (220) plane (solid line) and perpendicular to this plane (dashed line). To fit the experimental data [16], ionization cross-sections [17] for n=2 states were multiplied by the factor $\sim 0.45$. 
the \((k, l) = (1, -2)\) and \((k, l) = (1, 1)\) harmonics, has revealed considerable Stark mixing between closely lying \(2p^2 \cdot {^1}D_2\) and \(2s2p \cdot {^1}P_1\) autoionizing states due to the Lindhard potential. As estimated in paper [14], the total yield of Auger electrons emitted in this process is about 40\% of the total probability of the electron loss by the ion at the resonance maximum. Calculations also show that doubly excited states produced in the RCE process are aligned and, as a result, angular distribution of the Auger electrons is strongly anisotropic.

3.2. Metastable ion production
Resonant monopole E0 transitions, such as \(1s \rightarrow 2s\), are strongly suppressed in photoexcitation of free ions but turn out to be open in crystals due to their Stark mixing with corresponding optically allowed RCE transitions. Under channeling, the Lindhard continuous potential is responsible for this mixing; contribution of the metastable state to the survival fraction of the initial charge state is expected to be of the order of 10\% (Fig. 2).

Without channeling such mixing was observed by the Tokyo collaboration [21] using double resonance technique in 3D resonant coherent excitation of Ar\(^{17+}\). Ionic excited states were coupled by resonant strong oscillating electric field resulting in Autler-Townes effect. The density matrix calculation for the yield of the metastable \(1s2s \cdot {^1}S_0\) fraction in this experiment at the finest tuning on the resonance \(1s2s \cdot {^1}S_0 \rightarrow 1s2p \cdot {^1}P_1\) is presented in Fig. 3.

![Figure 2](image1.png)

**Figure 2.** Calculated survival fraction (thin solid line) and excited states fractions (dashed lines — \(2p\), thick solid line — metastable \(2s\)) for 390 MeV/u Ar\(^{17+}\) passing through 0.74 \(\mu\)m thick silicon crystal under planar channeling conditions. The experimental data (squares) are taken from [20].

![Figure 3](image2.png)

**Figure 3.** Calculated fraction of metastable \(1s2s \cdot {^1}S_0\) state at the exit of the 1\(\mu\)m silicon crystal for 416 MeV/u Ar\(^{16+}\) under double resonance in non-channeling conditions corresponding to the setup of the recent experiment [21].

3.3. Stokes parameters of X-ray radiation from RCE devices
We consider observation of strong alignment of the angular momentum of relativistic RCE ions [12] as a signal to special experimental and theoretical investigations of this process as a candidate for a tunable source of polarized X-ray radiation. In Fig. 4 we demonstrate our calculation for the laboratory frame Stokes parameters of radiation from relativistic Fe\(^{24+}\) ions under planar channeling in two cases of thick 21 \(\mu\)m and thin 0.9 \(\mu\)m Si crystal. The former illustrates high level of possible linear polarization of photons from \(1s2p \cdot {^1}P_1 \rightarrow 1s^2 \cdot {^1}S_0\) deexcitation transition and possibility to vary its linear polarization degree in the interval \(0 \leq P_L \leq 1\) on the \(\theta \approx 40^\circ\) cone around the direction of the incoming ion beam.

In Fig. 4b we suggest a special organization of RCE X-ray experiment to produce vector polarized RCE ions and, hence, to be able to observe its circular polarized radiation. Contrary
to usual RCE X-ray measurements, photon detection should be done in coincidence with the exit ions leaving the channel asymmetrically relative to the channel plane. The example shown corresponds to RCE of 423 MeV/u Fe$^{24+}$ ions in thin 0.9 µm-thick Si crystal.

![Diagram](image)

**Figure 4.** Angular distribution (dashed lines) and polarization of photons in the laboratory frame from 423 MeV/u Fe$^{24+}$ ions: a) linear polarization $P_L$ (solid line) of photons on the 41° cone, b) circular polarization $P_3$ (solid line) in the channel plane in coincidence with ions leaving the channel on one side from the channel plane.

### 4. Conclusion

We have reported selected results from our latest RCE studies and presented suggestions for new RCE experiments. Other aspects of performed calculations will be reported elsewhere.

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