X-ray production from resonant coherent excitation of relativistic HCIs in crystals as a model for polarization XFEL studies in the keV range

V.V.Balashov, A.A.Sokolik, A.V.Stysin

D.V.Skobeltsyn Institute of Nuclear Physics, M.V.Lomonosov Moscow State University, Moscow 119991, Russia

EMMI, GSI; June 7-9, 2010
Fourier series of the potential of the in-target electric field and its Lorentz transformation to the ion rest frame;

Scalar potential $\varphi(\vec{r})$ of the electric field of the crystal is composed in the laboratory frame into its Fourier (klm) harmonics

$$\varphi(\vec{r}) = \sum_{kln} \Phi_{kln} e^{i\vec{G}_{kln} \cdot \vec{r}},$$

with wave vectors $\vec{G}_{kln}$ being reciprocal lattice vectors. After a set of successive coordinate frame transformations this potential is transformed into the the scalar and vector electromagnetic potentials in the ion rest frame:

$$\varphi'(\vec{r}', t') = \gamma \sum_{kln} e^{i\nu t'}(\vec{G}_{kln})_x \Phi_{kln} e^{\pi i (z_{ion}/d+1/2)} e^{s \vec{G}_{kln} \cdot \vec{r}'},$$

$$\vec{A}'(\vec{r}', t') = -\vec{e}_x \gamma \frac{\nu}{c} \sum_{kln} e^{i\nu t'}(\vec{G}_{kln})_x \Phi_{kln} e^{\pi i (z_{ion}/d+1/2)} e^{s \vec{G}_{kln} \cdot \vec{r}'}.$$

with vectors $\vec{G}_{kln}' = \vec{G}_{kln} + (\gamma - 1)(\vec{G}_{kln})_x \vec{e}_x$ standing for the reciprocal lattice vectors in the ion rest frame, which $x$-component is $\gamma$ times larger, due to relativistic lattice contraction, than $x$-component of $\vec{G}_{kln}$. Hamiltonian $H_0$ of the free ion and the operator

$$V = -e\varphi' + \frac{e}{2mc}(\vec{p}\vec{A}' + \vec{A}'\vec{p})$$

of its interaction with the potentials $\varphi'(\vec{r}', t')$ and $\vec{A}'(\vec{r}', t')$ form the total Hamiltonian of the channeled ion $H = H_0 + V$ entering the generalized Master equation

$$i \frac{\partial \rho}{\partial t} = [H, \rho] + R\rho,$$

which describes time evolution of the density matrix $\rho$ of the ion.
\[ \Delta E = \begin{cases} 
\frac{2\pi h}{\alpha} \gamma v \left( \sqrt{2} k \cos \theta_t + \ell \sin \theta_t \right), & \text{channeling (2D)}; \\
\frac{2\pi h}{\alpha} \gamma v \left( \sqrt{2} (k \cos \phi_t + m \sin \phi_t) \cos \theta_t + \ell \sin \theta_t \right), & \text{non-channeling (3D)}. 
\end{cases} \]
**423 MeV/u Fe$^{24+}$, 1s2p:$^1P_1 \rightarrow 1s^2:$^1S_0**

Energy range of produced photons in the lab frame: 
**2.67 – 16.82 keV**

**Anisotropic X-ray emission:**
**experiment and calculations**

423 MeV/u Fe$^{24+}$ → (220) Si, thickness 21 μm

**Wavelengths**
- SASE 1: 0.1 nm
- SASE 2: 0.1-0.4 nm
- SASE 3: 0.4-1.6 nm

**Laserlike/coherent**
- SASE 1: yes
- SASE 2: yes
- SASE 3: yes

**Polarization**
- SASE 1: linear
- SASE 2: linear
- SASE 3: circular, linear

* V.V. Balashov, A.A. Sokolik, A.V. Stysin, JETP 107 (2008) 133

Compare:
The European XFEL
Technical Design Report
Energy range of produced photons in the lab frame: 2.67 – 16.82 keV

Anisotropic X-ray emission: experiment and calculations

423 MeV/u Fe\(^{24+}\) → (2\^20) Si, thickness 21 μm

σ\(_0\) = 0.03° (k,l) = (2,-1)

calculations
\( z_{max} = 0.9(d/2), \chi = 0.6 \)

calculations

V.V.Balashov, A.A.Sokolik, A.V.Stysin, JETP 107 (2008) 133

Compare:
The European XFEL
Technical Design Report

Wavelengths
SASE 1 0.1 nm
SASE 2 0.1-0.4 nm
SASE 3 0.4-1.6 nm

Laserlike/coherent
yes yes yes

Polarization
linear linear circular, linear

RCE

X-ray

Fe\(^{24+}\)
Stokes parameters of the characteristic X-ray radiation from RCE ions

The calculations show that since the photon yield and their polarization parameters in conditions typical or close to current RCE experiments are expected to be large, the resonance coherent excitation method can be considered as a candidate for a tunable source of polarized X-ray radiation in the keV region.

V.V.Balashov, V.K.Dolinov, A.A.Sokoli -- JETP Letters 89 (2009) 399
1965 – 2006: RCE exclusively with channeled ions –
V.V.Okorokov, Yad. Fiz. 2 (1965) 1009; Pis'ma Zh. Eksp. Teor.Fiz 2 (1965) 175
K.Komaki, T.Azuma et al., NIM B 146 (1998) 19,
and many other.

Since 2006: RCE measurements with both channeled ions and without channeling at all

Present-day portrait of the RCE process (the Okorokov effect) from Tokyo RCE measurements of 2006-2009:

Anisotropic X-ray emission from helium-like Fe24+ ions aligned by RCE with a periodic crystal potential –
T.Azuma et al., PRL 97 (2006) 145502;

Three-dimensional RCE of nonchanneling ions in a crystal –
C.Kondo et al., PRL 97 (2006) 135503;

Trajectory dependent RCE of planar-channeled ions in a thin Si crystal –
C.Kondo et al. NIM B 256 (2007) 157;

Doubly-resonant coherent excitation of HCI planar channeling in a Si crystal –

Dressed atoms in flight through a periodic crystal field: X-UVU double resonances –
Y.Nakai et al., PRL 101 (2008) 113201;

RCE of lithium-like Li15+ ions in a thin Si crystal –

Polarization control in three-dimensional RCE –
Y.Nakano et al., PRL 102 (2009) 085502;
A unified concept for theoretical analysis of RCE data and suggestions for new measurements (Moscow State University: 1998-2009)

Characteristic X-ray production in the RCE process

Metastable ion production in the RCE process
V.V. Balashov, I.V. Bodrenko -- NIM B 245 (2006) 52

Resonant coherent excitation of Ar$^{17+}$ ions in planar channel of a silicon crystal
V.V. Balashov, A.A. Sokolik -- Optics and Spectroscopy 103 (2007) 761

Angular anisotropy of characteristic X-radiation and Auger electrons during the resonance coherent excitation of relativistic ions under planar channeling
V.V.Balashov, A.A.Sokolik, A.V.Stysin -- JETP 107 (2008) 133.

Characteristic X-ray radiation and Auger electrons from resonant coherently excited highly charged ions under channeling

Kinetics of double resonant coherent excitation of relativistic multicharged ions in crystals beyond the channeling conditions

Angular anisotropy of the RCE X-rays under planar channeling as manifestation of geometrical properties of the in-crystal electric field

Polarization of photons emitted in the process of resonant coherent excitation of relativistic ions under planar channeling

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

Polarization and correlation aspects of resonant coherent excitation of fast highly charged ions in crystals
Fast highly charged ion in matter – an open quantum system

Density matrix approach; the generalized Master equation

\[ i \frac{\partial \hat{\rho}}{\partial t} = [\hat{H}, \hat{\rho}] + \hat{R} \hat{\rho}; \]

\[ \hat{H} = \hat{H}_0 + \hat{V}(t); \quad \hat{V}(t) = \hat{V}^{(\text{lattice})}(t) + \hat{V}^{(\text{wake})}(t). \]

A system of coupled equations to solve:

\[
\frac{\partial \rho_{pp}(t)}{\partial t} = 2 \cdot \sum_{q=1}^{N} \text{Im} \left[ V_{pq}(t) \cdot \rho_{qp}(t) \right] - \lambda_p(t) \rho_{pp}(t) + \sum_{q \neq p} \lambda_{qp}(t) \rho_{qp}(t); \\
\frac{\partial \rho_{pq}(t)}{\partial t} = -i \cdot \sum_{r=1}^{N} \left[ V_{pr}(t) \rho_{rq}(t) - \rho_{pr}(t) V_{rq}(t) \right] - \frac{1}{2} \left[ \lambda_p(i) + \lambda_q(i) + \lambda_{pq}^{(\text{elastic})}(i) \right] \rho_{pq}(t); \\
\rho_{pq}(t = 0) = \delta_{p1} \delta_{q1}. \]

for relativistic ions
wake potential is very small
From the generalized Master equation to RCE observables

- from density matrices for individual trajectories
to the averaged density matrix for the whole
ensemble of ions participating in the RCE process
and to the survival fraction $S$

$$\langle \hat{\rho}(t) \rangle = \frac{1}{2z_{\text{max}}} \int_{-z_{\text{max}}}^{z_{\text{max}}} \hat{\rho}_{z_{\text{in}}}(t) dz_{\text{in}}$$

$$S = \text{Tr} \{ \langle \hat{\rho}(t = t_{\text{out}}) \rangle \}$$

from density matrix to statistical tensors of X-ray emitting states

$$\langle \hat{\rho}(t) \rangle \Rightarrow \langle \hat{\rho}(t)^{\text{rad}} \rangle \Rightarrow \langle JM | \hat{\rho}(t)^{\text{rad}} | J'M' \rangle \Rightarrow \rho_{pq}(t)$$

$$W_{\gamma}(\theta, \varphi) = \frac{1}{4\pi} \left[ 1 + \alpha^2 \sqrt{\frac{4\pi}{5}} \sum_{q=-2}^{2} \frac{\rho_{2q}(t)}{\rho_{00}(t)} Y_{2q}(\theta, \varphi) \right]$$

The density matrix approach has shown its universality in treating the RCE process in different conditions of its observation and when performing theoretical analysis of various RCE measurements by covering, on the same theoretical ground, a wide scope of RCE observables.
This feature manifests itself especially clearly when applied to treat “two-color” excitations when two harmonics of the electric field of the crystal act on the electron cloud of the moving ion like two lasers of corresponding frequencies.
Okorokov effect in two-colour (double resonant) excitation of ionic autoionizing states
\[ \text{Ar}^{16+} (387.9 \text{ MeV/u}) \rightarrow \text{Si} (220) \]
Auger electrons from double RCE

$\text{Ar}^{16+} (387.90 \text{ MeV/u}) \rightarrow \text{Si} (220)$

**Fig. 3.** Survival fraction of $\text{Ar}^{16+}$ ions upon excitation of a single (the dashed curve is our calculation, and the circles are the experimental data from [12]) and double (the solid curve is our calculation, and the squares are the data from [12]) resonances.
RCE without channeling

\[
(G_{klm})_x = \frac{2\pi}{a} \left( \sqrt{2k} \cos \theta \cos \phi + 
+ l \sin \theta + \sqrt{2m} \cos \theta \sin \phi \right),
\]

\[
(G_{klm})_y = \frac{2\pi}{a} \left( -\sqrt{2k} \sin \theta \cos \phi + 
+ l \cos \theta - \sqrt{2m} \sin \theta \sin \phi \right),
\]

\[
(G_{klm})_z = \frac{2\pi}{a} \left( -\sqrt{2k} \sin \phi + \sqrt{2m} \cos \phi \right).
\]

\[
E'_{klm}(t') = -i \Phi_{klm} \left[ (G_{klm})_x e_x + \gamma (G_{klm})_y e_y + 
+ \gamma (G_{klm})_z e_z \right] \exp \left( i \left( G'_{klm} \right)_x vt' \right),
\]

\[
\omega_{k,l,m} = \frac{2\pi \hbar}{a} \gamma v \left( \sqrt{2(k \cos \phi + m \sin \phi)} \cos \theta + l \sin \theta \right)
\]
Autoionization in double RCE without channeling

Exp.: Tokyo
Theory: Moscow,
also strongly anisotropic angular distribution of the Auger electrons predicted
Autler-Townes effect in resonant coherent excitation of relativistic highly charged ions in crystals

PHYSICAL REVIEW
VOLUME 100, NUMBER 2
OCTOBER 15, 1955

Stark Effect in Rapidly Varying Fields*

S. H. Autler† and C. H. Townes
Columbia University, New York, New York
(Received May 31, 1955)

A method is developed for calculating the effects of a strong oscillating field on two states of a quantum-mechanical system which are connected by a matrix element of the field. Explicit approximate solutions are obtained for a variety of special cases, and the results of numerical computations are given for others. The effect of an rf field on the $J=2\rightarrow1$ $l$-type doublet microwave absorption lines of OCS has been studied in particular both experimentally and theoretically. Each line was observed to split into two components when the frequency of the rf field was near 12.78 Mc or 38.28 Mc, which are the frequencies separating the $J=1$ and $J=2$ pairs of levels, respectively. By measuring the rf frequency, $\nu_0$, at which the microwave lines are split into two equally intense components, one may determine the separation between the energy levels. The measured value of $\nu_0$ depends upon the intensity of the rf field and the form of this dependence has been calculated and found to be in good agreement with the experimental results.
Observation of the A-T doublet in current P&P measurements with lasers (an example)

PHYSICAL REVIEW A 78, 013802 (2008)

Observation of narrow Autler-Townes components in the resonant response of a dense atomic gas

Vladimir A. Sautenkov,1,2 Yuri V. Rostovtsev,1 and Eric R. Eliel3
1Department of Physics and Institute of Quantum Studies, Texas A&M University, College Station, Texas 77843-4242, USA
2P. N. Lebedev Institute of Physics, 119991 Moscow, Russia
3Huygens Laboratory, Leiden University, P.O. Box 9504, 2300 RA Leiden, The Netherlands
(Received 21 April 2008; published 1 July 2008)

We have experimentally studied the reflection of a weak probe beam from a dense atomic potassium vapor in the presence of a strong laser field tuned to the atomic resonance transition. We have observed an Autler-Townes doublet under hitherto unexplored conditions, namely, that the Rabi frequency induced by the strong laser field is much smaller than the self-broadened width of the resonance transition of the unexcited vapor. We attribute our observation to a reduction of the atomic decoherence by the strong drive field. We present a theoretical model of nonlinear processes in a dense atomic gas to explain the observed results.

- Probe 770 nm laser power ~ 10^-4 W
- Pump 766 nm laser power ~ 0.5 W
- Pump detuning = 3 GHz = 0
- Autler-Townes splitting ~ 8 GHz

![Diagram showing laser configurations and observed spectra](image-url)
Autler-Townes doublet at resonant coherent excitation (RCE) of relativistic ions in crystals without channeling

\[ \omega_{k,l,m} = \frac{2\pi \hbar}{a} \gamma \nu \left( \sqrt{2} (k \cos \phi + m \sin \phi) \cos \theta + l \sin \theta \right) \]

**Diagram:**

- **1s2p: \(^1P_1\)**
- **1s2s: \(^1S_0\)**
- **1s: \(^1S_0\)**

**Probing (1,-1,-2)**

- **9.3 \times 10^{-15} \text{ sec}**
- **\(\Delta E_{1s-2s} = 3124.52 \text{ eV}\)**
- **\(\Delta E_{1s-2p} = 3139.55 \text{ eV}\)**

**Coupling (0,0,-2)**

- **2.3 \times 10^{-9} \text{ sec}**
- **\(\Delta E_{2s-2p} = 15.03 \text{ eV}\)**

**Detuning \(\delta_c\)**

**Ar \(^{16+}\), 416 MeV/u**

**Target Thickness (Si) \(t\) \(\sim 1 \mu m\)**

Tuning in **coupling** и **probing** resonances – by target rotation
Both coupling and probing electric fields in the ion rest frame are strong:

e.g.: $E_{(0,0,-2)} (1s2s^1S \rightarrow 1s2p^1P) \sim 3.5 \cdot 10^9 \text{ V/cm}$

That corresponds to radiation energy flux of 15 eV - VUV laser of about $5 \cdot 10^{15} - 1.5 \cdot 10^{16} \text{ W/cm}^2$. We calculated that a little lower energy flux of about $1.5 \cdot 10^{15} \text{ W/cm}^2$ corresponds to 3140 eV – X-ray harmonic $(0,1,2)$ of the probing field.

According to Technical Design Report of the European XFEL (pp. 264-274), the latter rate is near the lowest level of intensity of the flux for such suggested experiments on linear and non-linear processes as:

6.4.1. Small Quantum Systems

1. Inner shall ionization in atomic ions,
2. X-ray photons scattered at trapped ion crystals
3. Molecular dynamics following X-ray photoionization
4. Cluster experiments
5. Multiphoton studies in the X-ray spectral region
6. Dynamics of aligned small molecules and molecular wave packet dynamics
7. Time-resolved photo-fragmentation of small molecules
Autler-Townes doublet at resonant coherent excitation (RCE) of relativistic ions in crystals without channeling

\[ \omega_{k,l,m} = \frac{2\pi\hbar}{a} \gamma V \left( \sqrt{2} (k \cos \phi + m \sin \phi) \cos \theta + l \sin \theta \right) \]

Ar\(^{16+}\), 416 MeV/u

t (Si) \sim 1 \mu m

Tuning in coupling и probing resonances – by target rotation
Survival fraction measurements


“Kinetics of double resonant coherent excitation of relativistic multicharged ions in crystals beyond channeling conditions” – V.V. Balashov, A.A. Sokolik, A.V. Stysin - *JETP* **108** (2009) 1010

Red lines – calculation with 5x5 density matrix in basis
1s\(^2\):1S\(_0\); 1s2s:1S\(_0\); 1s2p:1P\(_1\)(M=0;±1).

No fitting parameters.
\[ \Delta E_d^{(\pm)} = \Delta E_{2P-2S} + \frac{1}{2}[\hbar \delta_c \pm \sqrt{\hbar \Omega_0^2 + (\hbar \delta_c)^2}] \]

\( \Omega_0 \) as a fitting parameter in the Autler-Townes formula = 3.72 eV;

\( \Omega_0 \) as calculated matrix element

\(< 1s2p:1P_1|V_{0,0,2}|1s2s:1S_0 >\)

of the field

= 3.55 eV
Drastic change in both experiment and calculation [JETP 108 (2009) 1010] in profile of the Autler-Townes doublet for X-ray photons detected in the (2,-2,0) plane direction [horizontal] and perpendicular to this plane [vertical] - clear demonstration of the potential of the Autler-Townes scheme in X-ray measurements to control polarization characteristics of double excitation of highly charged ions.

Here - indication to “fine structure” magnetic quantum number splitting of the Autler-Townes dublet $1s2s^21S_0 - 1s2p^2^1P_{M=0;\pm 1}$. 
Conclusion

The insightful prediction on resonant coherent excitation of fast ions in crystals made 45 years ago by Okorokov opened a way to wide experimental and theoretical investigations of this nice phenomenon. Basic parameters of currently studied RCE processes with relativistic highly charged ions (produced photon energy, radiation energy flux in the ion rest frame) look close to those usually related to XFELs. Also, one cannot ignore a unique feature of RCE as a tunable source of polarized X-ray radiation. True, RCE will never compete with lasers in whole. But wide experience gained in experimental and theoretical RCE studies, especially concerning polarization aspects of dynamics of various multi-photon processes in keV energy region, will be a good support among others for future XFEL experiments.

Deep thanks

to my RCE co-authors and co-workers
    I.Bodrenko,
    A.Sokolik,
    A.Stysin,
    V.Dolinov;

to Prof. T.Azuma, Dr. Y.Nakano and their colleagues (Tokyo Metropolitan University) for the long-term pleasure of interesting and stimulating contacts on RCE;

to Profs. P.Mokler, T.Stoehlker (GSI),
    D.Dauvergne (IPN, Lyon),
    J.Ullrich (MPI Heidelberg),
    J.Rost (MPIKS, Dresden) –
for many useful discussions and general support;

to Russian Foundation for Basic Research for regular supporting grants.
Thank you very much!
Angular Correlation of the Cascade Photons in the Course of Dielectronic Recombination of Channeled Ions

V. V. Balashov and A. V. Styshin

Institute of Nuclear Physics, Moscow State University, Leninskoe gory, Moscow, 119992 Russia
e-mail: balvse@anna19.npi.msu.ru
Received December 25, 2006

Fig. 1. Splitting (Δ) of the 1s2p \(^1\!P_1\) level in an ion from argon Ar\(^{16+}\) to iron Fe\(^{24+}\) in the plane (220) channel of the silicon crystal. (1) at the center of the channel and on departure from the center by (2) 0.3 and (3) 0.6 channel half-width; (4) radiation width Γ of this level.