Stimulated Emission from Surface-Scattered Ions

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We discuss coherent excitation of ions incident on crystal surfaces at grazing angles and show that substantial gains for soft x rays can be achieved for reasonable beam currents.

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Coherent excitation of ions by the periodic potential of a crystal was proposed by Okorokov some time ago[1] and has been observed by different groups who passed highly ionized atoms through thin foils.[2,3] The effect arises from the fact that an ion moving in a crystal with speed v experiences a time-dependent potential with harmonics $\Omega_n = (2\times\pi v n)/d$, where n is a positive integer and d is the lattice spacing in the direction of ionic motion. If v and d are such that Ω_n matches an ionic transition frequency, then ions are resonantly excited by the nth harmonic. The probability that ions are in an excited state becomes a periodic function of the thickness of the foil,[4] which can be viewed as a consequence of the Rabi oscillations[5] of ions in their rest frame. The Okorokov effect depends critically on the finiteness of the probability that ions keep their charge states constant while inside the crystal.

One can expect the same physical picture to hold for ions scattering from crystal surfaces at grazing incidences. This may be called the surface-Okorokov effect and offers interesting possibilities for obtaining stimulated emission of soft x rays from ions. For such a purpose, the surface scattering geometry has considerable advantage over the beam-foil geometry. The damage to the surface resulting from ion bombardment is minimized at grazing incidences and the same surface can be used repeatedly.[6] Stopping powers of surfaces can be lower than stopping powers of bulk materials, and therefore, ions can resonantly interact with surfaces for longer distances than with foils.

In this paper I consider the scattering of hydrogenic ions, such as Li^{+2}, Be^{+3}, etc., from crystal surfaces at grazing incidences, and show that ions execute extremely rapid Rabi oscillations in their rest frame. It is assumed that the ionic beam simultaneously interacts with a coherent electromagnetic field whose frequency matches one of the ionic transition frequencies corresponding to an effective population inversion, and it is shown that the inhomogeneously broadened gain of the signal can be substantial.

The geometry of the problem is shown in Fig. 1. An ion is incident from the left, moving in the z' direction. In the laboratory frame, the periodic crystal potential can be described by

$$V_c'(\vec{x}') = \theta(x_0 - x')\sum_{\vec{G}} \mathscr{V}_{\vec{G}} e^{i\vec{G}\cdot\vec{x}'}. \quad (1)$$

Here, x_0 designates the distance between the center of mass of the ion and the surface (x_0 is negative if the ion is above the surface, positive if the ion penetrates the surface). $\vec{G}$'s are the reciprocal lattice vectors. The crystal is assumed to terminate sharply on a plane at x_0. This plane need not coincide with the plane of the first layer of surface atoms; rather, it is an effective plane where the influence of the surface on the ion begins to be appreciable.

In the ionic rest frame, $V_c'(\vec{x}')$ is transformed into a vector $[\vec{A}_c(\vec{x},t)]$ and a scalar potential $[V_c(\vec{x},t)]$. $\vec{A}_c$ is purely longitudinal, and can be eliminated by a gauge transformation on the wave function of the ionic electron, $\psi(x,t) = \exp[i\Lambda(\vec{x},t)]\phi(\vec{x},t)$, such that $\nabla\Lambda = (e/\hbar c)\vec{A}_c$. ϕ satisfies

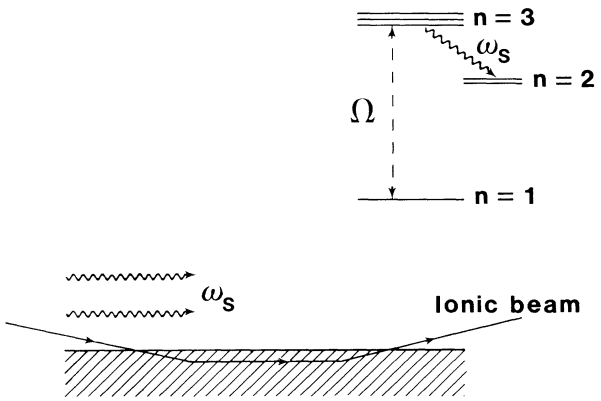

FIG. 1. The schematic of the grazing-incidence surface scattering.

the Schrödinger equation

$$i\hbar\partial_t\phi=[(2m)^{-1}(\vec{p}-e\vec{A}_s c^{-1})^2-Ze^2|\vec{x}|^{-1}+V_\perp+V_\parallel]\phi, \tag{2a}$$

where

$$V_\perp(\vec{x}_\perp)=e\gamma\theta(x_0-x)\sum_{s_\perp}\mathscr{V}_{\vec{G}}e^{i\vec{G}\cdot\vec{x}},\quad s_\perp=\{\vec{G}|\hat{z}\cdot\vec{G}=0\}, \tag{2b}$$

$$V_\parallel(\vec{x},t)=e\gamma^{-1}\theta(x_0-x)\sum_{s_\parallel}\mathscr{V}_{\vec{G}}\exp[i\vec{G}\cdot(\vec{x}_\perp+\gamma\vec{z}+\hat{z}\gamma vt)],\quad s_\parallel=\{\vec{G}|\hat{z}\cdot\vec{G}\neq 0\}, \tag{2c}$$

$\vec{A}_s=\hat{y}A_s\cos(k_sz-\omega_st)$ is the signal field, and $\gamma=(1-v^2/c^2)^{-1/2}$. $V_\perp$ is a static potential and depends on the transverse coordinates of the ionic electron. Note that going from the laboratory frame to the ionic rest frame is equivalent to taking a time average of the crystal potential along the ionic trajectory; $V_\perp$ is this time-averaged potential.[7,8] The level shifts induced by $V_\perp$ on ionic spectra in foils are estimated in Ref. 8. Here, however, we will ignore $V_\perp$ and its level shifts. For us the important term is the time-dependent potential $V_\parallel$. If $\vec{G}=l\vec{G}_1+m\vec{G}_2+n\vec{G}_3$, where l,m,n are integers, then the harmonics of $V_\parallel$ are given by $\gamma v\hat{z}\cdot(l\vec{G}_1+m\vec{G}_2+n\vec{G}_3)$. In the following, we will assume that one of the fundamental frequencies obtained from this expression equals or is near an ionic transition frequency. Specifically, we will assume that only $n=1$ and $n=3$ levels of the ion are involved in the transitions induced by the crystal, and that an effective population inversion occurs between $n=3$ and $n=2$ levels as shown in Fig. 1. One can then suppress all $\vec{G}$'s in the set $s_\parallel$ except for those which give a constant frequency Ω which is either resonant or nearly so with the ionic transition frequency ω_{31}. Thus we define the set of vectors $\vec{g}$ such that $\vec{g}=\hat{x}G_x+\hat{y}G_y+\hat{z}\gamma G_z$ and $|\vec{g}\cdot\hat{z}|=\text{const}=\Omega/v$. Ignoring the signal field for the moment, the amplitude equations in the rotating-wave approximation are

$$i\dot{c}_{100}=\exp[i(\Omega-\omega_{31})t]M_{100;3lm}c_{3lm}, \tag{3a}$$

$$i\dot{c}_{3lm}=\exp[-i(\Omega-\omega_{31})t]M^*_{100;3lm}c_{100}, \tag{3b}$$

where

$$M_{nlm;n'l'm'}=e(\hbar\gamma)^{-1}\sum_{\vec{g}}\mathscr{V}_{\vec{g}}\int d^3x\,u^*_{nlm}(\vec{x})\theta(x_0-x)e^{i\vec{g}\cdot\vec{x}}u_{n'l'm'}(\vec{x}), \tag{3c}$$

and u_{nlm} are the hydrogenic wave functions. Assuming $c_{100}(0)=1$ and $c_{3lm}(0)=0$, the solution of (3a) and (3b) is

$$c_{100}(t)=(\Omega^+e^{-i\Omega^-t}-\Omega^-e^{-i\Omega^+t})/(\Omega^+-\Omega^-), \tag{4a}$$

$$c_{3lm}(t)=M^*_{100;3lm}(e^{i\Omega^+t}-e^{i\Omega^-t})/(\Omega^+-\Omega^-), \tag{4b}$$

where

$$\Omega^\pm(lm)=(\omega_{31}-\Omega)/2\pm[(\omega_{31}-\Omega)^2/4+\Omega^2_{Rlm}]^{1/2}$$

and $\Omega_{Rlm}=|M_{100;3lm}|$ is the Rabi frequency.

If the ion penetrates the surface and $|x_0|\sim 1$ Å $>(a_B/Z)$, one can essentially ignore the step function in (3c) since u_{nlm}'s are localized to a radius of approximately $(na_B)/Z$. One then finds

$$M^{(p)}_{100;300}=3(3\pi)^{1/2}e(4\hbar\gamma)^{-1}\sum_{\vec{g}}\mathscr{V}_{\vec{g}}\beta_g^4(\beta_g^2+3)(\beta_g^2+1)^{-4}, \tag{5a}$$

$$M^{(p)}_{100;3lm}=i(\pi)^{1/2}e(2\hbar\gamma\sqrt{3})^{-1}\sum_{\vec{g}}\mathscr{V}_{\vec{g}}\beta_g^3(\beta_g^2+2)(\beta_g^2+1)^{-3}Y_{lm}(\hat{g}), \tag{5b}$$

$$M^{(p)}_{100;32m}=\frac{9e^2}{2\hbar\gamma}\left(\frac{3}{\pi}\right)^{1/2}\sum_{\vec{g}}\mathscr{V}_{\vec{g}}Y_{2m}(\hat{g})\beta_g^3 \times\left\{\frac{\beta_g(3\beta_g^3+4)}{6(\beta_g^2+1)^4}-\left(\frac{2}{\pi}\right)^{1/2}\sum_{s=0}^{\infty}\frac{\Gamma(s+\frac{5}{2})\Gamma(s-\frac{3}{2})}{2^s\Gamma(s+1)\Gamma(s+\frac{7}{2})}\left[1-\frac{\beta_g}{(\beta_g^2+1)^{1/2}}\right]^{s+(5/2)}\right\}, \tag{5c}$$

where $\beta_g = 4Z/(3ga_B)$ and the superscript p is meant to imply a penetrating ion. Note that only $M^{(p)}_{100;310}$ describes an electric dipole transition; the others arise from higher-order multipole transitions. This is in contrast to the formulation of the Okorokov effect in Refs. 4 and 9, where the dipole approximation is assumed to hold. In justification of (5a)–(5c), note that whether one can make the dipole approximation depends on the ratio of the crystal period to the radius of the ion's electronic orbital, that is on whether $|\vec{g}|(a_B n/Z) \ll 1$. For typical crystals one can easily obtain $|\vec{g}|(a_B n/Z) \sim 1$ even for low-lying orbitals. The dipole approximation breaks down. Incidentally, this fact can be exploited to populate those states which are not dipole connected to the ground state and therefore, relatively long lived.

If one takes the pseudopotential values of $\mathscr{V}_{\vec{g}}$ that are used in structure calculations,[10] and uses just the smallest and the next smallest $\vec{g}$'s in the sums of (5), one finds for a crystal like Si that $|M^{(p)}_{100;300}|$, $|M^{(p)}_{100;32m}| \sim 10^{16}$ sec^{-1} and $|M^{(p)}_{100;310}| \sim 10^{17}$ sec^{-1}. The resonance condition $v\hat{z}\cdot(\vec{G}_1+\vec{G}_2+\vec{G}_3) \sim \omega_{31} \sim 193$ eV can be satisfied for a Be^{+3} ion moving with speed $v \sim 10^9$ cm/sec. It follows that the ion can be completely excited after traveling a "Rabi distance" $l_{Rlm} = \pi v/\Omega_{Rlm} \sim 3-30$ Å.

In (5b) and (5c) the contributions of different terms are proportional to $Y_{lm}(\hat{g})$. There are, therefore, orientation-dependent interferences among $\mathscr{V}_{\vec{g}}$ components. By varying the direction of the ionic beam, it is possible to pick different sets of $\vec{g}$ vectors, and thus obtain information about different $\mathscr{V}_{\vec{g}}$ components from a comparison of Ω_{Rlm}.

We now treat the coupling of the ion to the coherent signal field as a perturbation imposed upon the evolution described by (4a) and (4b). In the calculation of the gain, the steps are: (a) determine $c_{2lm}(t)$ to first order in A_s with use of (4b); (b) calculate the transverse current density resulting from $(3lm) \rightarrow (2l'm')$ transitions; (c) substitute it into the Maxwell equation for A_s to obtain a dispersion relation between a real ω_s and a complex k_s. Imk_s gives the small signal gain for ions moving at a fixed speed v. Finally, (d) multiply the gain at v with the probability distribution of ion velocities, $\rho_{ion}(v)$, and replace Ω by $v|\hat{z}\cdot\vec{g}|$. It is assumed that $\rho_{ion}(v)$ is sufficiently narrow as not to effect the overall choice of the set of $\vec{g}$'s, and integration is over $-\infty < v < \infty$. In the limit that the lifetimes of the states in the $n=2$ shell go to ∞, the inhomogeneously broadened gain is given by

$$\mathscr{G}_s = \frac{2\pi^2\hbar e^2 P_{cs} N_{ion}}{\omega_s m^2 c} \sum_{l=0}^{2} \sum_{m=-l}^{+l} \frac{D_{lm}\rho_{ion}(v_{lm})}{[(\omega_s-\omega_{32})^2+\Omega^2_{Rlm}]}, \tag{6a}$$

where N_{ion} is the density of ions, P_{cs} is the probability that the charge state of ions remains unchanged, and v_{lm} is a tuning velocity given by

$$v_{lm} = (1/|\hat{z}\cdot\vec{g}|)\{\omega_s - \omega_{32} - [\Omega^2_{Rlm}/(\omega_s-\omega_{32})] + \omega_{31}\}. \tag{6b}$$

D_{lm} is a coefficient related to the momentum matrix elements,

$$D_{lm} = \sum_{l'm'}\sum_{l''m''} \mathscr{P}_{2l'm';3l''m''}\mathscr{P}^*_{2l'm';3lm} M_{100;3lm} M^*_{100;3l''m''}, \tag{6c}$$

where

$$\mathscr{P}_{2lm;3l'm'} = \int d^3x\, u^*_{2lm}(-i\nabla)u_{3l'm'}$$
$$= -(Z/a_B)\delta_{m,0}(\delta_{m',1}+\delta_{m',-1})[\delta_{l,0}\delta_{l',1}(\eta_1)^{1/2}+\delta_{l,1}\delta_{l',2}(\eta_2)^{1/2}]. \tag{6d}$$

Here $\eta_1 = 2^{13}\cdot 3^2\cdot 5^{-10} \simeq 7.6\times 10^{-3}$ and $\eta_2 = 2^{18}\cdot 3^3\cdot 5^{-10} \simeq 0.72$ are pure numbers.[11] A representative gain curve versus ω_s is depicted in Fig. 2 just for one particular (lm) in the sum (6a), and for a Gaussian probability distribution of average velocity v_0 and width Δv: $\rho_{ion}(v) = (\Delta v\pi)^{-1}\exp[-(v-v_0)^2/\Delta v^2]$. It has double peaks surrounding the exact resonance at $\omega_s = \omega_{32}$, where it vanishes. $\mathscr{G}_s$ is the sum of several such curves which have differing heights and widths, but which vanish on exact resonance.

According to (6a) $\mathscr{G}_s$ has an effective population inversion factor $N_{ion}\Omega^2_{Rlm}/[(\omega_s-\omega_{32})^2+\Omega^2_{Rlm}]$ instead of the usual $(N_{upper}-N_{lower})$. Thus all ions participate in the emission. The degree of their cooperation is determined by Ω_{Rlm}.

Going back to the Be^{+3} example and assuming $P_{cs} \sim 0.5$, one finds $\mathscr{G}_s \sim (N_{ion}/10^{12}\ \text{cm}^{-3})\times(v/\Delta v)\times 10^{-4}\ \text{cm}^{-1}$ for a signal of $\omega_s \sim 30$ eV. Thus, for $\Delta v/v \sim 10^{-4}$ and an ionic current density $j_{ion} \sim 4.8$ A/mm^2, one has $\mathscr{G}_s \sim 1$ cm^{-1}.

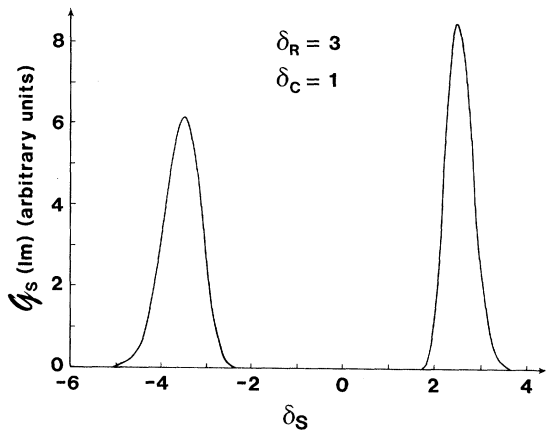

FIG. 2. The gain for a particular (lm) vs $\delta_S = (\omega_s - \omega_{32})(|\hat{z}\cdot\vec{g}|\Delta v)^{-1}$. For illustrative purposes, $\delta_R = \Omega_R(|\hat{z}\cdot\vec{g}|\Delta v)^{-1}$ and $\delta_C = (\omega_{31} - v_0|\hat{z}\cdot\vec{g}|)(|\hat{z}\cdot\vec{g}| \times \Delta v)^{-1}$ are taken to be 3 and 1, respectively.

As an ion moves inside the crystal or on its surface, it loses energy to the material electrons, and its center of mass slows down. This can cause the ion to detune from the resonance with the crystal field. Scaling from the stopping powers of protons in bulk materials,[12] one finds $S \sim 10^9$ eV/cm for Be^{+3} with $v \sim 10^9$ cm/sec for a Si target. For grazing-angle scatterings one can expect S to be lower since the number of material electrons may be depleted near the effective plane at x_0 as a result of the formation of inversion layers further down in the interior.[13] It is also possible to choose the beam direction in such a way that ions can move between the crystal planes, avoiding heavy concentration of material electrons. Such channeling of ions can decrease S by orders of magnitude.[14] For an ion moving the Rabi distance l_R, the minimum energy loss is $E_{\text{loss}} \sim S_{lR}$. Taking $S \sim 10^8$ eV/cm for channeled ions and $l_R \sim 3-30$ Å, one finds $\Delta E_{\text{loss}}/E_{\text{ion}} \sim 10^{-6} - 10^{-5}$ for Be^{+3}, which is quite small.

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