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Generation of coherent hard-x-ray radiation in crystalline solids by high-intensity femtosecond laser pulses

Péter Kálmán
Department of Experimental Physics, Technical University of Budapest, Budafoki út. 8 FII10, H-1521 Budapest, Hungary

Thomas Brabec
Abteilung für Quantenelektronik und Lasertechnik, Technische Universität Wien, Gußhausstrasse 27, A-1040 Wien, Austria

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A process for coherent hard-x-ray generation is suggested, which is based on the idea that high-intensity, ultrashort laser pulses can be coupled into a crystal before the lattice structure is destroyed. In the presence of the lattice periodicity, additional momentum can be transferred to the electrons that are moving in the strong light field. This additional momentum supplied by the crystal results in a substantial decrease of the threshold pump laser intensity required for hard-x-ray generation as compared to free electrons. The lattice is also utilized to select the frequency of the emitted x rays via Bragg coupling. A numerical estimation for the yield of the proposed mechanism gives an output power of some 10 W in the range of 1–10 keV of photon energies by using a state-of-the-art optical pulse with a peak intensity of about 10^{17} W/cm^2 and a pulse duration of 30 fs.

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Continuous effort is being made to develop and improve coherent x-ray sources [1]. However, the generation of coherent x-ray radiation with photon energies higher than 1 keV is an unsolved problem to date. The purpose of this paper is to propose a process for the generation of coherent hard-x-ray radiation in crystalline solids. It is the following. A high intensity laser field produces free electrons in a crystal. The free electrons are dressed by the laser field and emit hard-x-ray photons due to scattering by the crystal lattice.

Free electrons in a strong laser field can generate low-order harmonics [2], where the order of the highest harmonic is proportional to the product of two physical quantities: the change in electronic momentum and the magnitude of the electric field strength of the laser radiation. The momentum change of free electrons in a light field is determined by energy-momentum conservation; e.g., see the intense-field Compton effect. Therefore, the only possibility to create x-ray radiation by using free electrons is to increase the laser intensity. As the required intensities are difficult to realize experimentally, it is important to address alternative routes to coherent x-ray generation. The analysis performed here demonstrates that in the presence of a periodic crystal lattice the laser intensities required for the generation of x-ray radiation can be significantly reduced.

The presence of the periodic lattice structure is essential for the following reasons: (i) a large change in electron momentum can be supplied by electron-lattice scattering; (ii) the generated x rays are coupled via Bragg reflection so that the radiation forms a standing-wave pattern perpendicular to its direction of propagation. For a proper choice of polarization vector the transmission losses for the standing-wave x-ray modes can be drastically reduced (Borrmann effect) [3].

The mechanism proposed here is based on the potential of a new class of high power femtosecond solid state lasers [4] supporting pulses with durations of some 10 fs. This pulsewidth is much shorter than the electron-phonon relaxation time, which is a few hundreds of femtoseconds [5]. Therefore, in spite of the high intensities, the lattice can be supposed to participate in the laser-induced x-ray generation process [6].

The theoretical study of this paper reveals the following main results. For a given pump laser intensity a spectrum of x-ray frequencies is generated due to scattering by the crystal lattice. The lower cutoff of the frequency band is determined by the Bragg condition, whereas the upper cutoff frequency is determined by the pump-laser intensity due to the threshold condition. Our results show that the momentum exchange between the electron and the crystal requires a three-dimensional lattice and the mechanism does not work for one-dimensional periodic structures [7]. Formulas for the cutoff frequencies, the threshold pump laser intensity, and the x-ray power generated per pump pulse are obtained.

Our analysis starts with the assumption that free electrons are created in the crystal due to the interaction with the laser field [8]. The coupling between the free electrons and the laser field is so strong that it must be treated nonperturbatively. Therefore, the calculation is performed in two steps. First, a Volkov solution is dealt with, which is used to describe the free electronic states dressed by the laser field. Note that the wavelength of the x-ray radiation is comparable to the distance between the lattice sites; hence, the dipole approximation cannot be employed and the space dependence of the radiation field has to be taken into account. In a second step, the interaction of the electron with the lattice and with the x-ray field is taken into account by perturbation theory, where the dressed free electron solutions are used as initial and final states.

Free electronic states Ψ dressed by an intense laser field are described by the Schrödinger equation

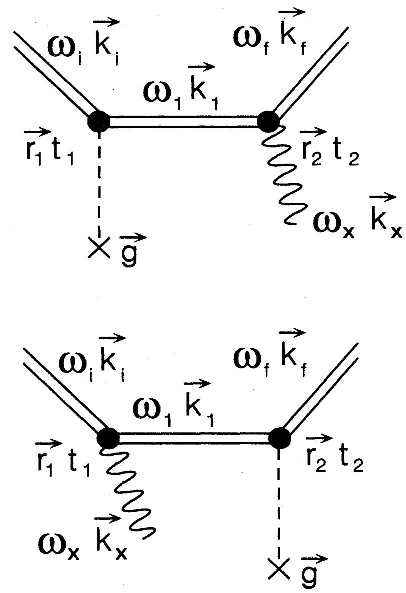

FIG. 1. Depiction of two graphs of x-ray generation in a crystal lattice. The double line with indices i, 1, f indicates the initial, intermediate, and final Volkov states of the electron. The dashed and the curved lines denote lattice potential and the x-ray radiation, respectively.

$$i\hbar\frac{\partial\Psi}{\partial t}=\frac{1}{2m}\left(-i\hbar\vec{\nabla}-\frac{e}{c}\vec{A}_p\right)^2\Psi, \tag{1}$$

where $\vec{A}_p,e,m,c$ denote the vector potential of the laser (pump) field, the electron charge, the electron mass, and the light velocity, respectively. The solution of Eq. (1) is the nonrelativistic Volkov state [9]

$$\Psi=\frac{1}{\sqrt{V}}e^{i(\omega t-\vec{k}\cdot\vec{r})}e^{-i[\vec{\alpha}(t)\cdot\vec{k}\sin\phi]}. \tag{2}$$

Here, V, $\vec{r}$, t, ω, and $\vec{k}$ denote the normalization volume, space-coordinate, time-coordinate, electron angular frequency, and electron wave vector, respectively. The parameter $\phi=(\omega_p t-\vec{k}_p\cdot\vec{r})$ is the phase of the laser field, where ω_p and $\vec{k}_p$ are the laser angular frequency and the laser wave vector. The refractive index of the material at the laser frequency is n, so that $|\vec{k}_p|=\omega_p n/c$. The coupling of the electron to the laser radiation is determined by $\vec{\alpha}(t)=\vec{e}_p\alpha_0\exp(-t^2/\tau^2)$. Here, τ is the width of the laser pulse, which is assumed to be Gaussian. The coupling strength at the pulse center is $\alpha_0=eE_p/(m\omega_p^2)$ and $\vec{e}_p$ denotes the laser polarization, which is assumed to be linear. Finally, $E_p=\omega_p A_p/c$ is the amplitude of the electric field. Note that Eq. (2) is an approximation of the relativistic Volkov solution [9] and contains the space dependence of the phase of the laser light. The pulse shape dependence in Eq. (2) has been included by using a slowly varying envelope approximation [10]. The space dependence of the pulse envelope is neglected.

The electron-lattice interaction potential can be written as

$$V(\vec{r})=\sum_{\vec{g}}V(\vec{g})e^{i\vec{g}\cdot\vec{r}}. \tag{3}$$

We assume a model lattice that consists of Coulomb potentials in an fcc lattice with lattice distance d, unit cell volume $V_c=d^3$. For this model the Fourier coefficients are $V(\vec{g})=4\pi e^2/(g^2V_c)$. The reciprocal lattice vector is denoted by $\vec{g}=g_0\vec{G}$, where $\vec{G}$ denotes a vector with integer components, and $g_0=2\pi/d$ is the magnitude of the smallest reciprocal lattice vector.

The interaction between the dressed electron and the x-ray radiation is described by the operator $Q=(ie\hbar/mc)\vec{\nabla}\cdot\vec{A}_x+e^2/(mc)\vec{A}_p\cdot\vec{A}_x$, where the vector potential of the x-ray field is denoted by $\vec{A}_x$. The first term of Q does not depend on the laser intensity and hence can be neglected in high intensity light-matter interactions. The remaining term is

$$Q=-\sum_x Q_x(e^{i\phi}+e^{-i\phi})e^{i(\omega_x t-\vec{k}_x\cdot\vec{r})}a_x^\dagger, \tag{4}$$

$$Q_x=\sqrt{\frac{2\pi\hbar c^2}{V\omega_x}}\frac{e^2E_p(t)}{2mc\omega_p^2}\vec{e}_p\cdot\vec{e}_x. \tag{5}$$

Here, the vector potential $\vec{A}_x$ is given in a quantized form; the operator $a_x^\dagger$ accounts for the creation of an x-ray photon with angular frequency ω_x, wave vector $\vec{k}_x$, and state of polarization $\vec{e}_x$. The sum over x denotes summation with respect to $\vec{k}_x$ and $\vec{e}_x$. As the refractive index at the x-ray frequency is approximately unity, the magnitude of the wave vector is $|\vec{k}_x|=\omega_x/c$.

The two lowest-order graphs describing our x-ray generation process are depicted in Fig. 1, from which the scattering matrix element S_{fi} can be determined by using the following definitions. The double line in Fig. 1 denotes the Volkov solution, Eq. (2), and $i,1,f$ refer to initial, intermediate, and final electron states with angular frequency $\omega_{i,1,f}$ and wave vector $\vec{k}_{i,1,f}$, respectively. The dashed and curved lines indicate the lattice potential and the x-ray radiation. The number eigenstates of the x-ray photon are denoted by $|0\rangle$ and $|1\rangle$. Then, from Fig. 1 and Eqs. (2)–(5) we have

$$S_{fi}=\frac{-1}{2\hbar^2}\frac{V}{(2\pi)^3}\int_{-\infty}^{\infty}dt_2\int_{-\infty}^{t_2}dt_1\int_{-\infty}^{\infty}d^3r_1d^3r_2d^3k_1\Psi_{1i}(1)\Psi_{f1}(2)[V(1)\langle 1|Q(2)|0\rangle+V(2)\langle 1|Q(1)|0\rangle]. \tag{6}$$

For the sake of brevity, we have introduced the following notation: $\Psi_{1i}=\Psi_1^*\Psi_i$, $\Psi_{f1}=\Psi_f^*\Psi_1$, $(1)=(\vec{r}_1,t_1)$, and $(2)=(\vec{r}_2,t_2)$. For the evaluation of Eq. (6) the Jacobi-Anger formula [11] is used to reexpress the Volkov solution as a sum over high harmonics of the laser field. The integrations in Eq. (6) are performed in a closed form using the following assumptions: $\omega_x \gg \omega_p$, $\omega_i \approx 0$, and the slowly varying pulse approximation $\tau\omega_p \gg 1$, which allows the evaluation of the t_1 integral [12]. Note that $\omega_i \approx 0$ is equivalent to the assumption that a free electron originates with zero velocity. This assumption has proven to be very successful in describing high harmonic generation in gases [13].

For the rest of the analysis we choose an fcc lattice with basis vectors aligned along the x, y, and z directions, $\vec{k}_x \perp \vec{z}$, $\vec{k}_p \| \vec{y}$, and $\vec{e}_p \| \vec{e}_x \| \vec{z}$; see Fig. 2. The last assumption fulfills the Borrmann condition for minimum propagation losses; i.e., the state of polarization of the x-ray radiation is parallel to the atomic plane responsible for Bragg coupling [3].

Because of the coherence of the process the total scattering matrix element is $\bar{S}_{fi}=S_{fi}n_e$, where n_e is the number of electrons in the interaction volume. Integrating $|\bar{S}_{fi}|^2$ over the phase space of the electron in the final state and multiplying by the phase-space element of the outgoing x-ray photon determines the number of x-ray photons $dN_x/(d\Omega_x d\omega_x)$ generated per solid space angle $d\Omega_x$ and frequency interval $d\omega_x$. Introducing $P_x=N_x\hbar\omega_x/\tau$, which is the average x-ray power generated per pump pulse in the interaction volume, and performing the integration, we obtain

$$\frac{dP_x}{d\omega_x d\Omega_x}=\frac{\tau I_p \lambda_p^2 n_e^2 r_e^4}{2\pi^3 d^4}\sum_{\vec{G},M}\frac{|\vec{G}\cdot\vec{u}_x|^2}{|\vec{G}|^4} I_M^2 . \tag{7}$$

Here the parameter $r_e=e^2/mc^2$ is the classical electron radius, M denotes the order of the high harmonic, I_p is the laser peak intensity, and $\vec{u}_x$ is the unit vector in the direction of $\vec{k}_x$. The spectral characteristic of the x-ray signal is determined by the Fourier integral

$$I_M=\int_{-\infty}^{\infty} dt J_M(g_0\alpha_0\vec{G}\cdot\vec{e}_p e^{-t^2})e^{i\epsilon t}, \tag{8}$$

where $\epsilon=\tau(\omega_{fi}+\omega_x-M\omega_p)$ is the spectral parameter and J_M is the Bessel function of order M. The parameter $\omega_{fi}\approx(\hbar\vec{G}g_0)^2/(2m)$ denotes the change in electron frequency between initial and final states, and is determined by the relation for momentum conservation [14]. From ω_{fi} we find that ϵ is a function of M and $\vec{G}$. Therefore, the summation in Eq. (7) is confined to values M and $\vec{G}$ for which ϵ lies within the bandwidth of I_M.

Significant generation of x-ray radiation can only be expected when the argument of the Bessel function for $t=0$ in Eq. (8) is comparable to its order, i.e., $f(\vec{G})=g_0\alpha_0 G_z/M \geq 1$, where $\vec{G}=(G_x,G_y,G_z)$. This relation leads to a threshold condition for the generation of x-ray radiation, as follows. As the bandwidth of I_M is small compared with ω_x we can use $\epsilon=0$ to obtain a relation between M and $\vec{G}$ that gives

$$f(\vec{G})=\frac{2mg_0\omega_p\alpha_0 G_z}{2m\omega_x+\hbar g_0^2(G_x^2+G_y^2+G_z^2)} . \tag{9}$$

Then, the vector $\vec{G}_0$ for which f has a maximum is determined by solving $\vec{\nabla}_{\vec{G}}f=\vec{0}$. This yields $G_{x0}=G_{y0}=0$ and $G_{z0}=\sqrt{2m\omega_x/(\hbar g_0^2)}$. The threshold condition $f(\vec{G}_0)=1$ from which the threshold intensity is obtained as

$$I_{th}=\frac{\omega_x}{\omega_p}\,1.24\times10^{13}\,\frac{\mathrm{W}}{\mathrm{cm}^2} . \tag{10}$$

At a given pump intensity I above the threshold a spectrum of x-ray frequencies is generated. Equation (10) can be used to obtain the upper cutoff frequency $\omega_{x,max}$ of the spectrum. The lower cutoff frequency $\omega_{x,min}$ is determined by the Bragg condition. As stated above, a considerable x-ray signal can only be obtained when the transmission losses are reduced by Bragg coupling. The Bragg condition for a reciprocal lattice vector $\vec{g}$ is given by

$$\sin\theta=\frac{|\vec{g}|c}{2\omega_x} , \tag{11}$$

where θ is the Bragg angle; see Fig. 2. The Bragg condition at $\theta=\pi/2$ with the smallest possible vector $|\vec{g}|=2g_0$ defines $\omega_{x,min}=cg_0$.

For the numerical evaluation we choose crystalline LiF with $d=0.403$ nm, laser wavelength $\lambda_p=$ 800 nm, and pulsewidth $\tau=30$ fs. For these parameters the lower cutoff photon energy is 3.08 keV and the threshold intensity $I_{th}=2.45\times10^{16}$ W/cm^2. The power dP_x of the generated x-ray radiation is evaluated at the lower cutoff frequency (θ = $\pi/2$) for a peak pump intensity $I=7\times10^{16}$ W/cm^2. The rest of the parameters required for the evaluation of Eq. (7) are $\tau d\omega_x$ = 30, $d\Omega_x=10^{-6}$, and $n_e=4.6\times10^{12}$, which corresponds to one free electron per unit cell and an interaction volume $10\times10\times3$ μm^3. Then, $dP_x=1.08\times10^7\Sigma$, where Σ denotes the sum in Eq. (7); numerical computation gives $\Sigma=7\times10^{-6}$ and finally we get $dP_x=$ 80 W.

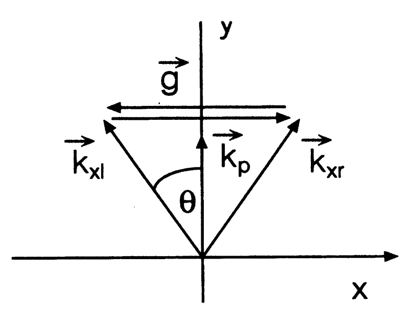

FIG. 2. A vector diagram for Bragg coupling of the generated x-ray fields with wave vectors $\vec{k}_{xl}$ and $\vec{k}_{xr}$; the Bragg vector is denoted by $\vec{g}$. x and y are crystallographic directions of an fcc lattice. The Bragg condition determines the angle of propagation θ that depends on the x-ray frequency ω_x. The states of polarization of the laser ($\vec{e}_p$) and the x-ray radiation ($\vec{e}_x$) are both perpendicular to the plane of the figure. $\vec{k}_p$ denotes the direction of propagation of the laser light.

It is informative to compare our numerical result with the characteristics of synchrotron radiation facilities. Our relative bandwidth and intensity are 2×10^{-4} and 270 MW/cm^2, respectively. The photon number in one pulse is 4870, which corresponds to a brilliance in a pulse of 4.8×10^{22} photon/(s $mrad^2$ mm^2), but the brilliance is usually defined by taking the time average of the pulse train. If we assume a laser repetition rate of 10^3 and a relative bandwidth of 10^{-3} generally used for synchrotron radiation sources we obtain a brilliance of 7.2×10^{12} photon/(s $mrad^2$ mm^2). This is comparable to the brilliance of synchrotrons, which is in the range of $10^{13}-10^{15}$ photon/(s $mrad^2$ mm^2) [15].

Concluding, based on the experimental progress in ultrafast laser technology, coherent x-ray radiation is expected to be generated by ultrashort intense laser beams in crystalline solids. The central concept of the suggested mechanism is that laser radiation of high intensity can be coupled to a crystal before the periodic lattice structure is destroyed. In the presence of a periodic lattice, additional momentum is transferred to the electrons dressed by the laser field. This reduces substantially the threshold intensity required for hard x-ray generation as compared to free electrons. The lattice is also utilized to select the frequency of the emitted x rays via Bragg coupling. Our analysis indicates that at pump intensities $\approx 10^{16}-10^{17}$ W/cm^2 coherent x-ray radiation with a photon energy in the keV range and with a power of some 10 W can be expected.

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