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

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Recently [Phys. Rev. A **52**, R21 (1995)] a process for coherent hard-x-ray generation in crystalline solids was suggested by us. In the current work the theoretical treatment of this process is formulated in the language of nonlinear optics. An approximate solution of the coupled Schrödinger and Maxwell equations is presented that governs the creation and the evolution of the x-ray field. The theory generalizes our previous study by providing a description of the pulse evolution and of the temporal shape of the x-ray pulse.

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The realization of efficient hard-x-ray sources is an unsolved problem. In a recent paper [1] we have proposed a process for the generation of coherent hard-x-ray radiation in crystalline solids which is based on the following mechanism. A high-intensity laser field creates free electrons in a crystal. The electrons are dressed by the electromagnetic field and can emit hard-x-ray photons due to additional momentum transfer supplied by electron-lattice scattering.

The proposed mechanism is based on the potential of a class of high power femtosecond solid state lasers [2] supporting pulses with peak intensities of about 10^{17} W/cm^2 and with durations of some 10 fs. This pulse width is much shorter than the electron-phonon relaxation time, which is a few hundred femtoseconds [3]. Therefore, in spite of the high intensities, the lattice can be supposed to participate in the laser induced x-ray generation process.

In Ref. [1] an estimation for the efficiency of x-ray generation in crystalline solids has been given, however, questions related to the pulse shape have not been addressed. The main goal of this Brief Report is to develop a theoretical approach that provides a description of the evolution of the x-ray pulse. This is performed by solving the coupled Schrödinger and Maxwell equations approximately. The result is found to be in agreement with our former analysis [1] which is based on the *S*-matrix formalism.

The derivation of the x-ray pulse evolution proceeds as follows. First, the Schrödinger equation is solved where the lattice periodicity is assumed as a perturbation and the unperturbed wave functions are Volkov solutions. From the solutions of the Schrödinger equation the transition current is determined [4]. The transition current is used as a source term for the generation of x-ray radiation in the Maxwell equations. The resulting wave equation is solved by supposing that the electromagnetic field which is created during the crystal-laser interaction takes the form of a Borrmann mode [5]. These are the modes which experience minimum losses during propagation in the crystal.

The electron wave function Ψ in a strong laser field and a periodic lattice potential is described by the Schrödinger equation

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

Here, e, m, c, $\vec{r}=(x,y,z)$, t denote the electron charge, the electron mass, the light velocity, the space coordinate, and the time coordinate, respectively. The vector potential of the laser (pump) field is defined by $\vec{A}_p=A_p\vec{e}_p f(T/\tau)\cos\Phi$ with $A_p=cE_p/\omega_p$. The function f defines the laser pulse envelope and the parameter $T=t-y/v_g$ is a retarded time frame moving with the group velocity of the pulse, v_g. The phase of the pump field is defined by $\Phi=\omega_p t-\vec{k}_p\cdot\vec{r}$, where ω_p and $\vec{k}_p$ are the laser angular frequency and the laser wave vector. The parameters $\vec{e}_p$ and E_p denote the state of polarization and the peak value of the electric field strength of the laser radiation. Finally, for the pump field the following definitions are assumed: $\vec{e}_p\|\vec{z}$ and $\vec{k}_p\|\vec{y}$. In the Fourier representation the lattice potential has the form $V(\vec{r})=\Sigma_{\vec{g}}V(\vec{g})e^{i\vec{g}\cdot\vec{r}}$, where $\vec{g}$ denotes a reciprocal lattice vector. We assume a model lattice that consists of Coulomb potentials in a fcc lattice with lattice distance d and 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 $\vec{G}=(G_x,G_y,G_z)$ is a vector with integer components and $g_0=2\pi/d$ denotes the magnitude of the smallest reciprocal lattice vector. Here, we have assumed that the basis vectors of the reciprocal lattice are aligned along the x, y, and z directions.

In the presence of the laser field an electron current is generated. Due to the high intensity of the laser pulse only terms containing the vector potential of the laser radiation are taken into account. Therefore the relevant part of the current density for the emission of x-ray radiation is

$$\vec{J}=-n_e\frac{e^2}{mc}\vec{A}_p\mathrm{Re}\sum_f\Psi_f^*\Psi_i, \tag{2}$$

where n_e is the number of free electrons. The indices i and f denote the initial state of the electron after the ionization and

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the final state of the electron after the emission of an x-ray photon, respectively. In Eq. (2) we have assumed that all electrons originate with zero velocity. This assumption has proven to be very successful in describing high harmonic generation in gases [6].

The propagation of the x-ray signal is described by the Maxwell equations, which contain the current density as a source term for the generation of x-ray radiation. Eliminating the magnetic field components in the Maxwell equations, the propagation of the electric field component of the x-ray radiation, $\vec{E}$, is governed by the wave equation

$$\nabla^2\vec{E}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{E}=\frac{4\pi}{c^2}\frac{\partial}{\partial t}\vec{J}. \tag{3}$$

From x-ray diffraction theory it is known that x-ray radiation in a periodic lattice experiences high absorption losses, with the exception of waves coupled via Bragg reflection. Bragg coupling results in the formation of a standing wave pattern perpendicular to the direction of propagation. The transmission losses are minimized for Bragg waves that fulfill the Borrmann condition [5]; i.e., the state of polarization of the x-ray radiation is parallel to the atomic plane responsible for Bragg coupling and the nodal points of the standing wave pattern coincide with the atomic lattice sites. As x-ray generation is only efficient for low transition losses, we choose for the analysis of Eq. (3) an ansatz function for the electric field of the x-ray radiation which contains the characteristics of a Borrmann mode. The wave vector, the circular frequency, and the state of polarization of the x-ray field are denoted by $|\vec{k}_x|=\omega_x/c$, ω_x, and $\vec{e}_x$, respectively. For the Borrmann ansatz the following assumptions are used. The vector responsible for Bragg coupling $\vec{g}=(g_x,0,0)$ is assumed to point in the x direction. Then, the wave vector of the x-ray field has the form $\vec{k}_x=(g_x/2,k_y,0)$, where k_y is the y component of the x-ray wave vector. For the direction of polarization of the x-ray field we assume $\vec{e}_p\|\vec{e}_x\|\vec{z}$. With the above definitions the ansatz for the electric field component of the x-ray field can be written as $\vec{E}=E(y,t)\vec{e}_x\sin(g_x x/2)\sin(\omega_x t-k_y y)$ [7]. The pulse envelope of the x-ray field is denoted by $E(y,t)$. Inserting the ansatz function in Eq. (3) and applying the slowly varying wave approximation, an equation for the pulse envelope is obtained that is

$$-\left(2k_y\frac{\partial}{\partial y}-\frac{2\omega_x}{c^2}\frac{\partial}{\partial t}\right)\vec{e}_xE(y,t)\sin(\omega_x t-k_y y)\sin(g_x x/2)=\frac{4\pi}{c^2}\frac{\partial}{\partial t}\vec{J}. \tag{4}$$

The equation describes the generation and propagation of a spectrum of x-ray frequencies which differ in the magnitude of k_y. In Ref. [1] it has been found that the threshold intensity for x-ray generation is lowest for $k_y=0$. Therefore the rest of the paper is devoted to the analysis of the wave equation (4) for $k_y=0$.

The source for the current densities $\vec{J}$ are free electrons that have been created in ionization by the intense laser field. The electrons can be regarded as quasifree as the influence of the laser field is much stronger as compared to the effect of the lattice atoms. Neglecting the influence of the lattice the wave function for the free electron is a Volkov solution

$$\Psi = \frac{1}{\sqrt{V}}e^{-i(\omega t-\vec{k}\cdot\vec{r})-i\vec{k}\cdot\vec{\alpha}(t)\sin\Phi_p}. \tag{5}$$

The function $\vec{\alpha}(t)=\alpha_0\vec{e}_pf(T/\tau)$ characterizes the coupling of the free electron to the laser field, with $\alpha_0=eE_p/(m\omega_p^2)$. The quantities V, ω, and k denote some normalization volume, electron angular frequency, and electron wave vector, respectively. The effect of the lattice periodicity on the Volkov wave function is taken into account by using perturbation theory and yields

$$\begin{aligned}\delta\Psi=&\frac{1}{(2\pi)^3}\frac{1}{\sqrt{V}}\int_{-\infty}^{\infty}d^3k_1e^{-i(\omega_1t-\vec{k}_1\cdot\vec{r})-i\vec{\alpha}(t)\cdot\vec{k}_1\sin(\omega_pt-\vec{k}_p\cdot\vec{r})}\\&\times\int_{-\infty}^{t}dt_1\int_{-\infty}^{\infty}d^3r_1e^{i(\omega_1t_1-\vec{k}_1\cdot\vec{r}_1)+i\vec{\alpha}(t_1)\cdot\vec{k}_1\sin(\omega_pt_1-\vec{k}_p\cdot\vec{r}_1)}\\&\times\sum_{\vec{g}}\frac{V(\vec{g})}{i\hbar}e^{i\vec{g}\cdot\vec{r}_1}e^{-i(\omega t_1-\vec{k}\cdot\vec{r}_1)-i\vec{\alpha}(t_1)\cdot\vec{k}\sin(\omega_pt_1-\vec{k}_p\cdot\vec{r}_1)}.\end{aligned} \tag{6}$$

The integrals in Eq. (6) are evaluated utilizing the same assumptions as in Ref. [1]. After performing the integrations over dt_1, d^3r_1, and d^3k_1 the perturbed wave function is inserted in Eq. (2). To obtain the final expression for the transition current density the sum over all final states in Eq. (2) has to be evaluated. The sum is reexpressed by using the energy and momentum conservation relations and the electronic differential phase space element $V/(2\pi)^3d^3k$ which yields the correspondence $\Sigma_f\rightarrow V/(2\pi)^3g_0^3\vec{G}^2d\omega_xd\Omega_x/(\pi\omega_x)$. Here, the differential phase space element of the electron in the final state has been expressed by the differential solid space angle $d\Omega_x$ and the relative bandwidth $d\omega_x/\omega_x$ of the x-ray signal. In the final expression only terms containing the functional dependence $\sin(g_xx/2)\sin(\omega_xt-k_yy)$ for Borrmann propagation are retained. Combination of Eqs. (2) and (3) yields the electric field component of the x-ray field, $E=-2\pi/\omega_x\vec{J}\vec{e}_p$. Finally, inserting the transition current in this expression we obtain

$$\frac{E}{E_p}=n_e\kappa S(T)\frac{d\omega_x}{\omega_x}\frac{d\Omega_x}{\pi}, \tag{7}$$

where $\kappa=r_e\alpha_f\lambda_p/(\pi^3d^2)$, and the parameters α_f, r_e, and λ_p are the fine structure constant, the classical electron radius, and the wavelength of the pump laser, respectively. The function $S(T)$ is defined by the sum

$$S=f\left(\frac{T}{\tau}\right)\sum_{M,\vec{G}}J_M\left(\vec{G}\cdot\vec{e}_p\alpha_0g_0f\left(\frac{T}{\tau}\right)\right). \tag{8}$$

The summation index M denotes the Mth order of the fundamental pump frequency ω_p and is related to $\vec{G}$ by the condition for energy conservation, i.e., $M=\hbar g_0^2\vec{G}^2/(2m\omega_p)+\omega_x/\omega_p$.

Near the threshold the sum in Eq. (8) can be further modified to obtain a simple expression for the temporal shape of the x-ray pulse. Using the approximation $J_M(\lambda z) \approx \lambda^M J_M(z)$ [8] in Eq. (8) gives

$$S=\sum_M [f(T/\tau)]^{M+1}\sum_{\vec{G}} J_M(\vec{G}\cdot\vec{e}_p\alpha_0 g_0). \tag{9}$$

From Eq. (9) an estimation for the x-ray pulse length τ_x can be obtained. The lowest possible order of M in the x-ray generation process is determined by the threshold number of the absorbed pump (laser) photons $M_{th}=\omega_x/\omega_p$. Inserting M_{th} in Eq. (9) we obtain $\tau_x \leqslant M_{th}^{-1/2}\tau$.

For the numerical evaluation of the x-ray radiation yield we have chosen the same parameters as in Ref. [1]; i.e., crystalline LiF with $d=0.403$ nm, laser wavelength $\lambda_p=800$ nm, pulse width $\tau=30$ fs, and pump peak intensity $I_p=7\times10^{16}$ W/cm^2. The rest of the parameters required for the evaluation of Eq. (8) are $d\omega_x/\omega_x=2\times10^{-4}$, $d\Omega_x=10^{-6}$, and $n_e=1.0\times10^{11}$, which corresponds to one free electron per unit cell in the interaction volume [9]. To get an estimation for the peak intensity of the generated x-ray radiation I_x the quantity $S_{max}=S(T=0)$ is evaluated numerically, which gives $S_{max}=15$. Inserting these parameters in Eq. (7) we finally get $I_x/I_p=(E/E_p)^2=1\times10^{-7}$ or $I_x=7\times10^9$W/cm^2.

Converting the peak intensity of the x-ray field to an average intensity $I_{av}=I_x\tau_x/\tau$ we get $I_{av}=160$ MW/cm^2. This is in reasonable agreement with the average intensity per pump pulse obtained in Ref. [1], $I_{av}=270$ MW/cm^2.

In conclusion, we have investigated the generation and evolution of x-ray radiation in crystalline solids by high power femtosecond pulses. The periodic potential of the crystal lattice is important for two reasons. First, the momentum transfer due to the lattice periodicity enhances the efficiency of the x-ray generation. Second, in the presence of the lattice potential the x-ray modes with minimum absorption losses are selected via Bragg coupling. In this paper a theoretical formalism has been developed which contains both the generation and the propagation of the x-ray field. The electron in the laser field is described by quantum mechanics. Approximate solution of the Schrödinger equation is substituted in the transition current which is used as a source term for x-ray generation in the Maxwell equations. Solution of the coupled equations gives a formula determining the generated x-ray pulse intensity. Assuming a laser pulse intensity of 7×10^{16} W/cm^2 and a laser pulse length of 30 fs we obtain an average x-ray intensity per pulse of 160 MW/cm^2 and a peak intensity 7×10^9 W/cm^2.

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