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Coherent control of X-rays by laser-produced optical phonons and generation of extremely short X-ray pulses

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Abstract. By solving analytically and numerically dynamical diffraction equations, we study the modulation of X-rays in ideal crystals by a co-propagating wave of impulsively excited optical phonons. Our results for a diamond crystal suggest that the output diffracted X-ray field can have a time structure much shorter than the period of the excited vibrational mode (T_v= 25fs). We explain this dynamics in terms of high-order Raman scattering of X-rays by optical phonons. The potential of this ultrafast modulation technique for controlling X-rays from synchrotron and FEL sources is discussed.

1. Introduction

In recent years a great variety of new fascinating experiments utilizing time-resolved X-ray diffraction have been reported from fields as diverse as solid-state physics [1,2], chemistry [3], and biology [4]. This breakthrough has been made due to the development of novel ultrafast hard X-ray sources, e.g. high-brilliance synchrotrons delivering few-ps X-ray pulses [5] and, especially, laser-plasma-based sources [6,7] able to generate bursts of X-rays of a sub-ps duration.The shortest presently available hard X-ray pulses (≈100fs) are, however, too long to resolve many ultrafast processes in liquids and solids, and much less to visualize the dynamics of inner shell electrons developing on the time scale of a few femtoseconds. Here we note the recent advances in the soft X-ray region, where attosecond X-ray sources have become available and made possible unique spectroscopic studies [11].

Recent attempts to generate extremely short hard X-ray pulses have included different techniques [7-10]. One of the most promising approaches is the idea that very short bursts of X-rays can be produced by controlling X-ray diffraction with a fs-laser pulse. In [7] a fs-laser was used to melt the crystal surface to interrupt X-ray Bragg reflection. Modulation of X-ray diffraction by acoustic [9] and optical [10] phonons generated by a fs-pulse photoabsorption was also demonstrated. However, the efficiency of these processes is still low due to the fact that the interaction occurs within the optical absorption depth which is much smaller than the X-ray diffraction length. On the other hand, increase of the driving field intensity is limited by the onset of the unwanted effects (irreversible structural changes, nonthermal melting etc.[1]).

In this paper we discuss a new mechanism of generation of ultrashort X-ray pulses which uses a scattering of X-rays by a co-propagating wave of coherent optical phonons excited by a fs-laser pulse [19]. Unlike the available X-ray modulation techniques [6-10], the method explores the transparency region of the crystal for both X-ray and laser radiation, thus allowing one to take advantage of a minimum loss and a maximum interaction length. Propagation effects play a central role in this

method, leading to a new physics: We show that the modulation rate achievable with this technique is not limited by the period of the excited phonon mode and may lie in the sub-vibrational-period region, as a consequence of a higher-order Raman scattering. We start from the fact that significant vibrational amplitudes can be generated without excitation of electronic transitions using nonresonant impulsive Raman scattering (ISRS)[12,14]. As a result, large volumes can be excited uniformly. Another important moment is the principle feasibility to synchronize a diffracted X-ray field with a wave of laser produced phonons to enhance the interaction. This can be done either by adjusting the Bragg angle (by choosing an appropriate reflection) or by tuning the X-ray energy [13]. The use of the Laue (transmission) geometry is especially attractive, since unlike the Bragg (reflection) scheme it enables accumulating the interaction over many diffraction lengths, similar to the travelling wave Raman scattering scheme in nonlinear optics [14]. In addition, in Bragg modulators [12], it might be difficult to separate the weak laser-induced satellite reflection from the background of the main Bragg reflection. We show that the Laue scheme offers a principally new possibility: The generated ultrafast bursts of X-rays can be accurately sliced from the long input bunch using the "pendulumlike" behavior of X-rays in a Laue crystal.

2. Theoretical model

Let us consider an interaction of X-rays with a wave of optical vibration in terms of the scheme shown in Figure 1, where the X-ray field propagates in conditions of symmetric Laue scattering, and the excitation laser pulse moves parallel to the crystal planes along the Z-axis. We assume that both the laser and the X-ray field are infinite and uniform in the transverse(X-Y) direction. The interaction is then independent of the transverse coordinates, and the evolution of the X-ray field in the crystal is described by one-dimensional nonstationary equations of dynamical theory for the amplitudes of the forward diffracted A_0 and reflected A_g wave [15]:

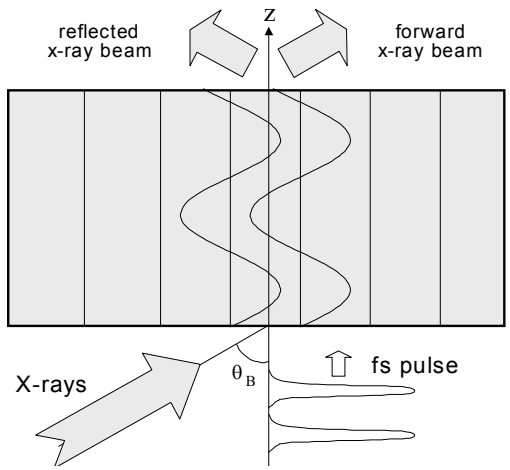

Figure 1.

(Ia) $$\cos(\theta_B)\frac{\partial A_0}{\partial z}+\frac{1}{c}\frac{\partial A_0}{\partial t}=\frac{\pi}{\lambda_x}\{i\chi_0 A_0+i\chi_{\bar{g}}A_g\}$$

(Ib) $$\cos(\theta_B)\frac{\partial A_g}{\partial z}+\frac{1}{c}\frac{\partial A_g}{\partial t}=\frac{\pi}{\lambda_x}\{i\chi_0 A_g+i\chi_g A_0+i2\sin(2\theta_B)\Delta\theta A_g\}$$

In equations (1) the X-ray fields are represented in the form: $E_0(\vec{r},t)=\frac{1}{2}A_0\exp(i\vec{k}\vec{r}-i\omega t)+c.c.$, $E_g(\vec{r},t)=\frac{1}{2}A_g\exp(i(\vec{k}+\vec{g})\vec{r}-i\omega t)+c.c.$, where $\vec{g}$ is the corresponding reciprocal lattice vector; $\chi_m=V^{-1}\int\chi(\vec{r})e^{i(\vec{g}_m\vec{r})}d^3\vec{r}$, $(m=0,\pm g)$ is the Fourier component of the crystal susceptibility, λ_X is the X-ray wavelength, and $\Delta\theta$ is the detuning from the Bragg angle θ_B . The Fourier components are related through $\chi_m=-(e^2/mc^2)(\lambda_X^2/\pi V)F_{g_m}$ to the structure factor of a unit cell $F_{g_m}=\sum_{j=1}^{N}f_j(\vec{g}_m)e^{i(\vec{g}_m,\vec{r}_j)}$, which depends on the positions of atoms in the cell and the atomic scattering factor $f_j(\vec{g}_m)$. We focus on the mechanism of modulation of the structure factor F_{g_m} due to the excitation of lattice vibrations.

The vibrations change positions of the atoms in the cell: $\vec{r}_j(t)=\vec{r}_j(0)+\vec{u}_j(t)$ ($\vec{u}_j(t)$ is the deviation of the j-th atom from the equilibrium $\vec{r}_j(0)$), but do not change the cell's size and, thus, the gross periodicity is unchanged. The structure factor F_{g_m} is assumed to vary adiabatically slow (e.g. the characteristic temporal (T_v) and spatial (Λ_v) scale of the variation are $T_v >> 2\pi/\omega_X$ and $\Lambda_v >> \lambda_X$), leading to a slow change of the amplitudes $A_0(z,t)$ and $A_g(z,t)$ in the crystal. The deviations $\vec{u}_j(t)$ can be represented as a superposition of normal vibrational modes $Q_\alpha(t)$ of the crystal $\vec{u}_j(t)=\sum Q_\alpha(t)\,\vec{\psi}_\alpha^j$ where $\vec{\psi}_\alpha^j$ is the polarization vector of the α-th vibration, and we have:

$$F_{g_m}(t) \approx \sum_{j=1}^{N} f_j \; e^{i(\vec{g}_m,\,\vec{r}_j(0))} + i\sum_j \sum_\alpha Q_\alpha(t)\,(\vec{g}_m,\vec{\psi}_\alpha^j)\; f_j\, e^{i(\vec{g}_m,\,\vec{r}_j(0))} \qquad (2)$$

The laser pulse propagating in the crystal excites optical vibrations via stimulated Raman process, and dynamics of a normal mode $Q_\alpha(t)$ obeys the equation of a Raman oscillator:

$$\frac{\partial^2 Q_\alpha}{\partial t^2} + \frac{2}{T_\alpha}\frac{\partial Q_\alpha}{\partial t} + \Omega_\alpha^2\, Q_\alpha = \sum_{ij}\left(\frac{\partial \hat{\chi}_{ij}}{\partial Q_\alpha}\right) E_i E_j \qquad (3)$$

where Ω_α is the characteristic frequency and T_α is the relaxation constant of the oscillator, $E_i(z,t)$ (i=x,y,z) is a component of the laser field, and $(\partial\hat{\chi}_{ij}/\partial Q_\alpha)$ is the Raman scattering tensor.

3. Ultrafast modulation of X-rays by impulsively excited phonons

When a single fs-pulse with a duration $\tau_p << (2\pi/\Omega_\alpha)$ interacts with the oscillator, a vibrational motion produced in the "impulsive" regime has the form of damped oscillations, $Q_\alpha(t)=q_\alpha \sin[\Omega_\alpha t]\exp(-t/T_\alpha)$, with the amplitude $q_\alpha = \Omega_\alpha^{-1}(\partial\chi/\partial Q)\int_{-\infty}^{\infty}|E(t)|^2\,dt$ proportional to the integrated laser intensity [14]. The first Fourier component of the susceptibility, which ís responsible for the modulation of X-ray diffraction, is found by summing over all the impulsively excited vibrational modes:

$$\chi_g(t) = \overline{\chi}_g + \sum_\alpha \delta\chi_g^\alpha \sin[\Omega_\alpha t]\exp(-t/T_\alpha) \qquad (4)$$

where $\delta\chi_g^\alpha = -\frac{e^2}{mc^2}\frac{\lambda_X^2}{\pi V}\sum_j i\, q_\alpha(\vec{g},\vec{\psi}_\alpha^j)\, f_i\, e^{i(\vec{g},\,\vec{r}_j(0))}$ is the amplitude of the deviation of $\chi_g(t)$ from the unperturbed value $\overline{\chi}_g$ due to α-th optical vibration. We note that the sum in $\delta\chi_g^\alpha$ over the indexes j of the atoms within a unit cell differs from that appearing in the case of acoustic vibrations, where all the atoms of a cell experience equal displacement, which leads to a phase shift in F_g and χ_g [18]. When optical phonons are excited, the amplitude of χ_g is generally changed [15]. To get an insight into the physics of the modulation process, we consider that as a result of ISRS-excitation the susceptibility acquires only amplitude modulation. Assuming further that only the strongest mode is excited, the susceptibility wave generated by the pulse is described by $\chi_g(z,t)=\overline{\chi}_g+\delta\chi_g \sin[\Omega(t-z/v_L)]$. By passing in equations (1) to new variables $(z,\tau=t-z/v_\parallel)$, where $v_\parallel = c\cos(\vartheta_B)$ and considering $\Delta\theta \approx 0$, equations (1) can be solved exactly:

$$A_0(z,\tau) = A_0(0)e^{i\beta\chi_0 z}\cos\{\pi(z/L_{ext}) + \xi(z)\sin[\Omega(\tau-\Delta z/2)]\} \qquad (5a)$$

$$A_g(z,\tau) = i\,A_0(0)e^{i\beta\chi_0 z + i\varphi_g}\sin\{\pi(z/L_{ext}) + \xi(z)\sin[\Omega(\tau-\Delta z/2)]\} \qquad (5b)$$

where $A_0(0)$ is the amplitude of the incident X-ray bunch at z=0, $\beta = \pi/(\lambda_X \cos(\theta_B))$, and φ_g is the phase of the susceptibility: $\bar{\chi}_g = \exp(i\varphi_g)\left|\bar{\chi}_g\right|$. As follows from (5), in the absence of the modulation term (e.g. of the time-dependent term in brackets), the formulas describe the well-known pendulumlike solution [18], in which the energy of the X-ray beam flows over from the forward diffracted to the reflected wave periodically in the z direction on the spatial scale of the extinction length $L_{ext} = \lambda_X \cos(\theta_B)/\left|\bar{\chi}_g\right|$. The relative role of the modulation term in the X-ray diffraction depends on the magnitude of the modulation amplitude in equations (5) given by:

$$\xi(z) = \frac{\delta\chi_g}{\bar{\chi}_g}\,\frac{\pi}{L_{ext}}\,\frac{\sin\left[\Omega\Delta z/2\right]}{\Omega\Delta/2} \qquad (6)$$

where the parameter $\Delta = (1/v_L - 1/v_{||})$ characterizes the group velocity (GV) walk-off between the excitation pulse and the diffracted X-ray fields. In the absence of synchronism, an increase of the modulation amplitude $\xi(z)$ with the distance is limited by the length $z_{sat} \approx \pi/\Omega\Delta$ at which the phase of the lattice vibrations modifying diffraction of the co-propagating X-ray field changes its sign. However, when the fields are synchronized (Δ=0), the amplitude grows linearly with the length: $\xi(z) = \pi(\delta\chi_g/\bar{\chi}_g)(z/L_{ext})$. This reflects the fact that the effect of the modulation is accumulated over the propagation length and, as we show below, dramatically modifies the X-ray diffraction. Indeed, by representing the X-ray fields (5) as a series:

$$A_0(z,\tau) = A_0(0)\, e^{i\beta\chi_0 z}\sum_{n=-\infty}^{\infty} J_n(\xi)\cos\left\{\pi(z/L_{ext}) + n\Omega\tau\right\} \qquad (7a)$$

$$A_g(z,\tau) = i\,A_0(0)\,e^{i\beta\chi_0 z + i\varphi_g}\sum_{n=-\infty}^{\infty} J_n(\xi)\sin\left\{\pi(z/L_{ext}) + n\Omega\tau\right\} \qquad (7b)$$

we can see that the perturbation leads to a generation of an infinite number of harmonics of the vibrational frequency, which are Stokes and anti-Stokes Raman components of the X-ray field: $\omega_n = \omega \pm n\Omega$. The number of components making a major contribution to the X-ray field (7) is determined by the argument $\xi(z)$ of the Bessel functions $J_n(\xi)$. A direct consequence of this fact is that for $z > (L_{ext}/\pi)\left|\bar{\chi}_g/\delta\chi_g\right|$ the X-ray spectrum is enriched with higher (n>1) order harmonics, that is, the time scale of the field variation becomes faster than the period of lattice vibrations. Note that the ultrafast modulation can also be thought of as originating from the time modulation of the effective extinction length, $L_{eff}(\tau) = L_{ext}[1+(\delta\chi_g/\bar{\chi}_g)\sin(\Omega\tau)]^{-1}$, defining a spatial scale on which the X-ray field is reflected by the atomic planes.

Since the X-ray fields have the form of ultrafast transients developing against a background of the long input X-ray bunch, the important question is how to cut the transients from the background. This appears to be possible since the net phase of the pendulum-like solutions (5) contains the time independent phase shift $\Phi_0(z) = (\pi z / L_{ext})$. By choosing the crystal thickness L so that in the absence of the perturbations the output deflected (or forward scattered) X-ray field is zero due to the pendulum-like behavior (this is the case when $(L/L_{ext}) = k$ or $(2k+1)/2$, respectively, (k=1,2,...)), the background can be completely removed. In fact, the fs-burst of X-rays emerges from the crystal only after the crystal was exposed to the laser field.

4. Discussion and conclusions

In order to make quantitative predictions, we studied equations (1)-(4) numerically for different crystals. A diamond crystal is especially interesting because of its very high optical phonon frequency: $\Omega = 1332\,cm^{-1}$ (T_{vibr}= 25fs). In addition, synchronism can be achieved in the spectral regions where X-

rays and the pump laser pulse exhibit a low absorption. For example, for a pump pulse at λ=0.8μm (Ti:Sapphire laser), GV-synchronism takes place for $(\bar{2}02)$ reflection with X-ray energy E=5.3KeV (λ_X = 2.3 Å , θ_B=63°). According to [12,14], ISRS can produce atomic displacements ~10^{-1} Å per 1J/cm^2, and we estimated that for the pump pulse with τ_p =20fs and energy 0.1mJ focused to a diameter 300μm the change of the susceptibility will be $|\delta\chi_g / \bar{\chi}_g| \approx 0.1$. Because the pendulum oscillation length depends on the Bragg detuning, $L_{ext}(\Delta\theta)=L_{ext}(0)[1+y^2]^{-1/2}$ where $y=(\Delta\theta)\sin(2\theta_B)/|\bar{\chi}_g|$ [18] , the optimum crystal thickness L and the generated pulse shape are functions of Δθ . Figures 2a,b show the temporal structure of the output reflected X-ray field for different Δθ .
In Figure 2a the optimum crystal thickness L(Δθ) is small (for instance, for Δθ=0 the latter is L(0)= 17.1 μm), and the modulation regime is close to linear (ξ<<1). The output X-ray field is seen to replicate the sinusoidal behavior of the perturbed susceptibility. In a thicker crystal (Figure2b, L(0)=85.5 μm), high-order scattering comes to play (ξ>1) and leads to the specific temporal splitting of the generated structures. The most efficient modulation is observed within a 10-arcsec angle near the Bragg angle, where the output X-ray field is comparable in intensity to the long input X-ray bunch.

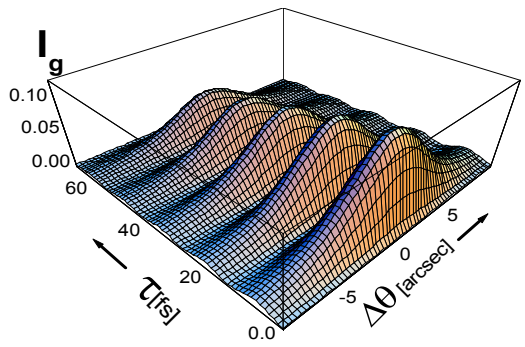

Figure 2a.

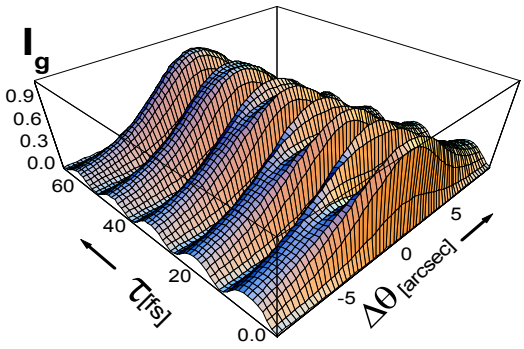

Figure 2b.

Intensity of the output diffracted X-rays modulated by a wave of optical phonons in diamond $(\bar{2}02)$ versus the Bragg detuning (the pump pulse: λ_p= 0.8μm, τ_p=20fs, I_p=2.5TW/cm^2). The crystal length is chosen such that the background is removed due to the pendulumlike behavior: **Figure 2a** - L=3L_{ext}, **Figure 2b** - L=15L_{ext}, where $L_{ext}(\Delta\theta)$= 5.7(μm)(1+0.04 $(\Delta\theta)^2)^{-1/2}$ with Δθ in arcsec.The intensity is normalized by the input intensity.

Whereas duration of the transients in Figure 2a,b are determined by the relaxation time of the excited vibrations, much faster structures can be produced by applying two identical fs-laser pulses. If the second pulse is delayed by the half vibrational period (12.5fs) it will turn off the coherent lattice vibration, and a half-oscillation vibration will be produced [16]. Figures 3a,b display the X-ray transients generated by the half-oscillation vibration excited by two 7fs-laser pulses (I_p=5TW/cm^2) in diamond for different crystal lengths: (a)- L(0)=171 μm, and (b)- L(0)=256,5 μm. Owing to the low dispersion of diamond at λ=0.8 μm ($k_{\omega\omega} \approx 10^3$fs^2/cm), reshaping of the pump pulses in the crystal is small. As can be seen from Figures 3a,b, the output X-ray field exhibits temporal modulations on the time scale essentially shorter than T_{vibr}=25fs. Fourier transform of the time structure in Figure 3b suggests that Raman sidebands up to 10-order contribute to the output X-ray field. Note that the X-ray intensity in Figure 3b is somewhat smaller than that in Figure3a due to linear attenuation (L_a=300 μm).
To observe the sub-10fs structures, the diffraction properties of the X-ray beam must be well-pronounced over interaction lengths of tens of L_{ext}. This implies a rather good quality of the X-ray beam.We estimated that a beam divergence below 10 arcsec is required, and therefore the technique could be used to control X-rays from modern synchrotron sources [9]. In view of the ongoing progress

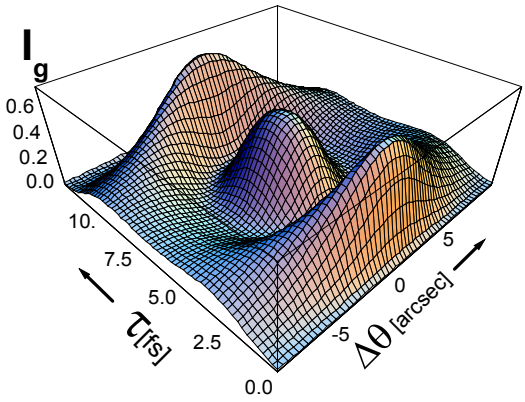

Figure 3a

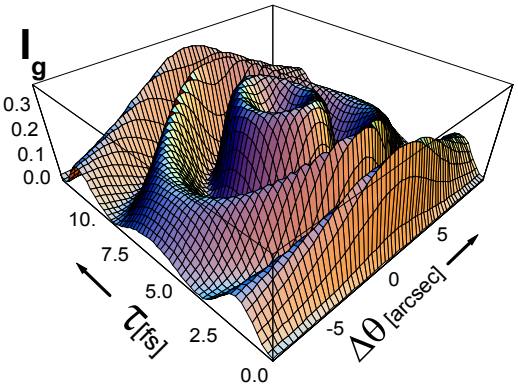

Figure 3b

The intensity of the diffracted X-ray field modulated by a half-wavelength lattice vibration in diamond, which is produced by a sequence of two 7fs-laser pulses (λ_p= 0.8μm, τ_p=20fs, I_p=2.5TW/cm^2). The crystal length is chosen such that the background is removed: **Figure 3a** - L=25L_{ext}, **Figure 3b** - L=45L_{ext}, where $L_{ext}(\Delta\theta)$= 5.7(μm)(1+0.04 $(\Delta\theta)^2)^{-1/2}$ with Δθ in arcsec. The intensity is normalized by the input intensity.

in intensity and brilliance of X-rays sources, it could also be interesting for shaping of X-rays generated by future sources, such as hard X-ray free electron lasers [17].

In conclusion, we studied the modulation of X-rays by a wave of coherently excited optical phonons. We found that the modulation rate achievable with this process may lie in the sub-vibrational-period region, as a result of higher order Raman scattering of X-rays. The results of modeling of X-ray diffraction in diamond under the conditions of impulsive excitation of the phonon mode at Ω=1332 cm^{-1} suggests that ultrafast X-ray transients with duration as short as only few femtosecond can be generated. Fundamentally, our results show that travelling wave interaction schemes (analogous to those long known in nonlinear optics) can give access to a significant increase of the efficiency of laser-assisted processes in the angström region.

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