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DIFFRACTION AND SCATTERING
OF IONIZING RADIATIONS

X-Ray Third-Order Nonlinear Dynamical Diffraction in a Crystal

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Abstract—The dynamic diffraction of an X-ray wave in a crystal with a third-order nonlinear response to external field strength has been theoretically investigated. General equations for the wave propagation in crystal and nonlinear Takagi equations for both ideal and deformed crystals are derived. Integrals of motion are determined for the nonlinear problem of dynamic diffraction. The results of the numerical calculations of reflectivity in the symmetric Laue geometry for an incident plane wave and the intensity distributions on the output crystal surface for a point source are reported as an example.

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INTRODUCTION

The X-ray dynamic diffraction in crystals is described by the Takagi equations [1], which include the linear part of crystal susceptibility. In view of the development of high-intensity synchrotron X-ray sources and X-ray free-electron lasers, the theoretical study of the dynamic diffraction with allowance for the nonlinearity of crystal susceptibility becomes urgent. Supplementing the linear polarization of crystal with the third-order nonlinear part, one can derive the propagation equations of dynamic diffraction by analogy with the derivation of the Takagi equations. One can consider crystal an isotropic medium with a rather high accuracy, as in the linear theory. In this case, the expression for the third-order nonlinear polarization is simplified. Note that the second-order nonlinear polarization is completely absent in centrosymmetric crystals. In noncentrosymmetric crystals, the influence of the second-order nonlinearity, i.e., the second-harmonic generation (SHG), is significant when certain phase-matching conditions are fulfilled. If not (as will be assumed below), the SHG effect is insignificant and can be neglected. Thus, there is only third-order nonlinearity of susceptibility in the case under consideration. These statements hold true for the model accepted in the theory of visible light optics [2]. Two models are considered in the literature on the nonlinear interaction of X rays with crystals; one of them was noted above, and the other is the model of cold plasma formed in a crystal by an ultrashort X-ray pulse of ultrahigh intensity passing through it. Apparently, the cold-plasma model is valid for media composed of light elements, where scattering occurs mainly from electrons weakly bound with atoms. For media containing atoms with an intermediate serial number or atoms of heavy elements, scattering is mainly from electrons of inner atomic shells; thus, the theory of scattering from inner atomic shells is more appropriate in this case [3]. In addition, it is reasonable to consider the cold-plasma model at very high intensities of incident radiation, when the contribution from the nonlinear part of susceptibility to scattering becomes equal to the contribution made by the linear part. The linear two-wave second-harmonic diffraction was considered in [4] within the cold-plasma model. The reverse dynamic influence of the two newly formed Bragg waves on the primary wave was disregarded. The kinematic Bragg diffraction of an intense plane X-ray wave under conditions of second-order nonlinearity with parametric conversion of an incident X-ray photon into an X-ray photon of lower frequency and a UV photon was considered in [5, 6] without application of the cold-plasma model. To implement this process, phase-matching conditions also must be fulfilled. The direct transmission of an intense X-ray beam through a crystal under the conditions of third-order crystal response to the radiation field strength was analyzed in [7] in terms of the cold-plasma model.

At low intensities, it is only the linear part of susceptibility that contributes to scattering. With a subsequent gradual increase in the incident radiation intensity, the nonlinear susceptibility, as well as the linear part of susceptibility, is formed due to the scattering from inner-shell electrons. This scattering for intensities below critical (at which the contribution of the nonlinear susceptibility to scattering becomes equal to the contribution of the linear susceptibility) can physically be described using the model known in optics. In this study we use the conventional model known in the visible light optics [2] to theoretically analyze the two-wave dynamic diffraction of X-ray waves in a crystal under the conditions of third-order response of the crystal to the radiation field strength.

PROPAGATION EQUATIONS

Let us consider the dynamic diffraction of an X-ray wave in a nonmagnetic perfect crystal without free charges under conditions of a third-order response of the crystal to the electric field strength. The crystal is assumed to be isotropic, as in most cases of linear theory. The second-order nonlinear susceptibility is absent in centrosymmetric crystals, and, if the phase-matching conditions are not fulfilled, the influence of the second-order nonlinearity can be neglected. We denote the electric field strength as $\tilde{\tilde{\mathbf{E}}}(\mathbf{r},t)$, where $\mathbf{r}$ is the radius vector of the observation point and t is time. The electric field satisfies the wave equation [2]:

$$\operatorname{curl}\operatorname{curl}\tilde{\tilde{\mathbf{E}}} + \frac{1}{c^2}\frac{\partial^2\tilde{\tilde{\mathbf{E}}}}{\partial t^2} = -\frac{1}{\varepsilon_0 c^2}\frac{\partial^2\tilde{\tilde{\mathbf{P}}}}{\partial t^2}, \qquad (1)$$

where c is the speed of light, $\varepsilon_0 = 8.85\times10^{-12}$ F/m is the permittivity of free space, and $\tilde{\tilde{\mathbf{P}}}(\mathbf{r},t)$ is the polarization of crystal. Equation (1) must be supplemented with the equation for induction $\tilde{\tilde{\mathbf{D}}} = \varepsilon_0\tilde{\tilde{\mathbf{E}}} + \tilde{\tilde{\mathbf{P}}}$:

$$\operatorname{div}\tilde{\tilde{\mathbf{D}}} = 0. \qquad (2)$$

In the case of monochromatic radiation with a frequency ω, the electric field, polarization, and induction can be presented in the form:

$$\begin{aligned}\tilde{\tilde{\mathbf{E}}}(\mathbf{r},t) &= \tilde{\mathbf{E}}(\mathbf{r})\exp(-i\omega t) + \text{c.c.},\\ \tilde{\tilde{\mathbf{P}}}(\mathbf{r},t) &= \tilde{\mathbf{P}}(\mathbf{r})\exp(-i\omega t) + \text{c.c.},\\ \tilde{\tilde{\mathbf{D}}}(\mathbf{r},t) &= \tilde{\mathbf{D}}(\mathbf{r})\exp(-i\omega t) + \text{c.c.},\end{aligned} \qquad (3)$$

where c.c. is a complex conjugate value. Substituting (3) into (1) and using (2), we obtain

$$\operatorname{curl}\operatorname{curl}\tilde{\mathbf{E}} - k^2(\tilde{\mathbf{E}} + \tilde{\mathbf{P}}/\varepsilon_0) = 0, \qquad (4)$$

where $k = \omega/c$. Equation (4) is a nonlinear analog of the corresponding equation of the standard linear theory of dynamic diffraction [8, 9].

It is convenient to represent polarization as a sum of linear and nonlinear parts:

$$\tilde{\mathbf{P}}(\mathbf{r}) = \tilde{\mathbf{P}}^{(1)}(\mathbf{r}) + \tilde{\mathbf{P}}^{(3)}(\mathbf{r}). \qquad (5)$$

The polarization and electric field are related as follows [2]:

$$\begin{aligned}\tilde{\mathbf{P}}^{(1)} &= \varepsilon_0\chi^{(1)}(\omega,\mathbf{r})\mathbf{E}(\mathbf{r}),\\ \tilde{P}_i^{(3)}(\mathbf{r}) &= 3\varepsilon_0\chi^{(3)}_{ijkl}(\omega,\mathbf{r})\tilde{E}_j(\mathbf{r})\tilde{E}_k(\mathbf{r})\tilde{E}_l^*(\mathbf{r}),\end{aligned} \qquad (6)$$

where $\chi^{(1)}(\omega,\mathbf{r})$ is the linear part of susceptibility (it is a scalar in an isotropic medium); $\chi^{(3)}_{ijkl}(\omega,\mathbf{r})$ is the tensor of third-order nonlinear susceptibility and indices i, j, k, l acquire values of 1, 2, 3, which correspond to the coordinate axes x, y, z, respectively (repeating indices imply summation over all values); and * indicates a complex conjugate value. In the general case, the tensor of the nonlinear part of susceptibility has 81 components; however, only 21 nonzero component remain for an isotropic medium because of symmetry; two of these components are independent, and the others can be presented as [2]

$$\chi^{(3)}_{ijkl} = \chi^{(3)}_{1122}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) + \chi^{(3)}_{1221}\delta_{il}\delta_{jk}, \qquad (7)$$

where δ is the Kronecker delta.

Substituting (7) into the second equation of (6), we find

$$\tilde{\mathbf{P}}^{(3)}(\mathbf{r}) = \varepsilon_0 A(\mathbf{r})(\tilde{\mathbf{E}}\tilde{\mathbf{E}}^*)\tilde{\mathbf{E}} + \varepsilon_0 B(\mathbf{r})(\tilde{\mathbf{E}}\tilde{\mathbf{E}})\tilde{\mathbf{E}}^*, \qquad (8)$$

where $A(\mathbf{r}) = 6\chi^{(3)}_{1122}(\omega,\mathbf{r})$, $B(\mathbf{r}) = 3\chi^{(3)}_{1221}(\omega,\mathbf{r})$; ω is omitted in the arguments for brevity.

Within the classical susceptibility theory, the electrons of crystal atoms behave like nonlinear oscillators: they oscillate under an electric field to induce an additional current and the corresponding polarization [2]. Let us denote the maximum resonance frequency of the oscillators as $\omega_{\max}$. In most cases the following inequality is fulfilled: $\omega \gg \omega_{\max}$; i.e., the external-field frequency greatly exceeds the resonance frequencies of the electrons of the medium [10]. In this case one can neglect the oscillator resonance frequency in the denominators of the final expressions for susceptibilities (as if scattering occurred from free electrons), as well as the oscillator damping coefficient. Within this approximation, according to [2],

$$\begin{aligned}\chi^{(1)}(\omega,\mathbf{r}) &= -\frac{n(\mathbf{r})e^2}{\varepsilon_0 m\omega^2},\\ \chi^{(3)}_{ijkl}(\omega,\mathbf{r}) &= \chi^{(3)}(\omega,\mathbf{r})\frac{(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})}{3},\end{aligned} \qquad (9)$$

where $\chi^{(3)}(\omega,\mathbf{r}) = n(\mathbf{r})e^4 b/(\varepsilon_0 m^3\omega^8)$; $n(\mathbf{r})$ is the electron concentration; e is the elementary charge; m is the electron mass; and b is a phenomenological constant, to which the third-order nonlinear part of the restoring force on electron is proportional. Let us estimate the third-order susceptibility based on quantum mechanics. The electron perturbation Hamiltonian in the radiation field has the form

$$H' = H_1' + H_2' = \frac{i\hbar e}{m}\tilde{\mathbf{A}}\nabla + \frac{e^2}{2m}\tilde{\mathbf{A}}^2, \qquad (10)$$

where $\tilde{\mathbf{A}}$ is the vector potential of the radiation field. Then, to estimate the third-order susceptibility, we will consider the first term in (10). The current density operator $\mathbf{j}$ in the radiation field is known to have the form [11, 12]:

$$\hat{\mathbf{j}}(\mathbf{r},\mathbf{r}_e) = \frac{e(\hat{\mathbf{p}}_e\delta(\mathbf{r}-\mathbf{r}_e)+\delta(\mathbf{r}-\mathbf{r}_e)\hat{\mathbf{p}}_e)}{2m} - \frac{e^2}{m}\tilde{\mathbf{A}}(\mathbf{r}_e,t)\delta(\mathbf{r}-\mathbf{r}_e), \quad (11)$$

where $\hat{\mathbf{p}}_e = -i\hbar\nabla_e$ is the operator of electron canonical momentum, $\hbar$ is Planck's constant, $\mathbf{r}_e$ is the electron coordinate operator, and δ is the Dirac delta. If the electron wave function u is presented in the form of an asymptotic series $u = u^{(0)}+u^{(1)}+\ldots$ in small perturbation powers, the expression for the mean quantum-mechanical current density will contain terms that are linear, quadratic, third-order, etc., with respect to perturbation. Let us consider the third-order terms with respect to the perturbation. The third-order current perturbation can be written as

$$\left\langle \mathbf{j}^{(3)}(\mathbf{r})\right\rangle = \left\langle u^{(0)} \mid \hat{\mathbf{j}} \mid u^{(3)}\right\rangle + \left\langle u^{(3)} \mid \hat{\mathbf{j}} \mid u^{(0)}\right\rangle + \left\langle u^{(1)} \mid \hat{\mathbf{j}} \mid u^{(2)}\right\rangle + \left\langle u^{(2)} \mid \hat{\mathbf{j}} \mid u^{(1)}\right\rangle + \ldots. \quad (12)$$

Here, the ellipsis indicates other terms that are of the third order with respect to perturbation. Generally, matrix elements (12) are considered within the dipole approximation, both for optical radiation and for X-ray waves [13]. In this approximation, the perturbation Hamiltonian can be written as $H' = -\hat{\boldsymbol{\mu}}\tilde{\tilde{\mathbf{E}}}$, where $\hat{\boldsymbol{\mu}} = e\mathbf{r}_e$ is the electron dipole moment operator [2]. Then, the mean quantum-mechanical value of the third-order correction to the dipole moment of one electron has the form

$$\left\langle \mathbf{p}^{(3)}\right\rangle = \left\langle u^{(0)} \mid \hat{\boldsymbol{\mu}} \mid u^{(3)}\right\rangle + \left\langle u^{(3)} \mid \hat{\boldsymbol{\mu}} \mid u^{(0)}\right\rangle + \left\langle u^{(1)} \mid \hat{\boldsymbol{\mu}} \mid u^{(2)}\right\rangle + \left\langle u^{(2)} \mid \hat{\boldsymbol{\mu}} \mid u^{(1)}\right\rangle. \quad (13)$$

The total dipole moment of unit volume is obtained as a result of multiplying (13) by the electron concentration. A detailed calculation for optical waves was performed in [2]. Based on the general formula derived in [2] (for example, formulas (4.3.12)–(4.3.14) in [2]), the third-order susceptibility of an isotropic medium in the case of low frequencies with respect to the resonance frequencies of the electrons of the medium was estimated to be

$$\chi^{(3)} = \frac{8n(\mathbf{r})e^4a^4}{\varepsilon_0\hbar^3\omega_0^3}, \quad (14)$$

where ω_0 is the characteristic resonance frequency of the electrons of the medium and a is the characteristic atomic radius, which can be assumed to be equal to the Bohr radius: $a = 5.3\times10^{-11}$ m. For X-ray waves, the external field frequency generally exceeds the resonance frequencies of the electrons of the medium. Using the same formulas [2] for the case where the frequency of incident X-ray waves is much larger than the resonance frequencies, we obtain the estimate

$$\chi^{(3)} \approx \frac{n(\mathbf{r})e^4a^4}{\varepsilon_0\hbar^3\omega^3}, \quad (15)$$

which contains, in contrast to (14), the frequency of incident X rays instead of the resonance frequency of the medium. Having compared (15) with (9), one can assign a certain value to the phenomenological constant.

Using the values $n(\mathbf{r}) \approx 10^{28}$–$10^{30}$ m^{-3} and $\omega \approx 10^{19}$ s^{-1}, we obtain the following estimate from (15): $\chi^{(3)}(\omega,\mathbf{r}) \approx 10^{-31}$–$10^{-33}$ m^2/V^2. The quantum-mechanical calculation of the third-order susceptibility [3] yielded an estimate for light elements (with a serial number $Z < 10$) far from the resonance frequencies: $\chi^{(3)}(\omega,\mathbf{r}) \approx 2\times10^{-40}$ m^2/V^2. It was also concluded in [3] that, in the range of X-ray frequencies far from resonance, the third-order susceptibility from bound electrons can be neglected for media with light atoms.

Let us denote $\left|\chi^{(1)}\right|/\chi^{(3)} = E_{cr}^2 = I_{cr}$, where E_{cr} is the critical electric field strength of the incident radiation at which the contribution of the third-order susceptibility becomes of the same order of magnitude as the contribution of linear susceptibility and I_{cr} is the corresponding critical intensity. According to (9) and (15), we have

$$E_{cr} = \left(\frac{\hbar^3\omega}{me^2a^4}\right)^{1/2} \approx 1.2\times10^{13}\ \text{V/m}. \quad (16)$$

Let us now estimate the incident-wave peak power at which the contribution of the third-order nonlinear susceptibility to propagation equation (4) becomes equal to the contribution of the linear susceptibility. The incident-wave peak power can be written as $P^{(i)} = \left|\mathbf{S}^{(i)}\right|\sigma_0$, where $\left|\mathbf{S}^{(i)}\right|$ is the modulus of the time-averaged Poynting vector (energy density flux), σ_0 is the incident-wave cross-sectional area, and the superscript (i) indicates the incident wave. Proceeding from formula (15) and the estimate $\chi^{(3)}(\omega,\mathbf{r}) \approx 10^{-31}$ m^2/V^2 (based on this formula), we obtain $\left|\mathbf{S}^{(i)}\right| \approx 0.5\varepsilon_0cE_{cr}^2 \approx 2\times10^{23}$ W/m^2. If an incident wave has sizes of 100 μm in the diffraction plane and 10 nm in the direction perpendicular to this plane, $\sigma_0 = 10^{-13}$ m^2 and the estimated power is $P^{(i)} \approx 100$ GW, a value that can be attained in X-ray free-electron lasers, according to [3]. Note that the above estimate is the upper limit of application of the perturbation theory, whereas nonlinear effects can be observed at much lower (one or even two orders of magnitude) incident radiation intensities, both in the case of plane waves and in experiments with a point source. Therefore, there is no need to start consideration with the plasma model; it is reasonable to apply it at intensities close to critical or higher. With a gradual increase in the incident radiation intensity, the nonlinear susceptibility, as well as the linear part of susceptibility, arises due to the scattering from inner-shell electrons. At intensities below critical, this scattering can physically be described using the model known in optics; this description will be performed below.

The electron concentration in a homogeneous medium is independent of the observation-point coordinates, whereas the electron concentration in a crystal is a three-dimensional periodic function of coordinates. Therefore, both linear and nonlinear parts of susceptibility in a crystal are three-dimensional periodic functions of coordinates. Note that, according to (10), the classical theory of susceptibility yields the following result: $\chi^{(3)}_{1122}(\omega,\mathbf{r}) = \chi^{(3)}_{1221}(\omega,\mathbf{r})$; i.e., 21 nonzero components of the third-order susceptibility tensor in an isotropic medium are expressed in terms of one component, the role of which can be played by $\chi^{(3)}_{1122}(\omega,\mathbf{r})$. In this case, as follows from formula (10) and definition of A and B, we have $A = 2\chi^{(3)}$, $B = \chi^{(3)}$, and $B = A/2$. Generally, the quantum-mechanical theory may lead to different values of susceptibilities: $\chi^{(3)}_{1122}(\omega,\mathbf{r}) \neq \chi^{(3)}_{1221}(\omega,\mathbf{r})$ [2].

Below we introduce the designation $\eta^{(3)} = A + B$; within the classical theory of susceptibility, $\eta^{(3)} = 3\chi^{(3)}$.

NONLINEAR TAKAGI EQUATIONS

According to (9), $A(\mathbf{r})$, $B(\mathbf{r})$ are three-dimensional periodic functions; they can be expanded in Fourier series in vectors $\mathbf{g}$ of crystal reciprocal lattice [8, 9]. At the same time, as in the linear theory, the electric field strength can be presented as a series of quasi-planar waves [1]. Thus, we arrive at

$$\mathbf{A} = \sum_{\mathbf{g}} \mathbf{A}_{\mathbf{g}} e^{i\mathbf{g}\mathbf{r}},$$
$$\mathbf{B} = \sum_{\mathbf{g}} \mathbf{B}_{\mathbf{g}} e^{i\mathbf{g}\mathbf{r}}, \qquad (17)$$
$$\tilde{\mathbf{E}} = \sum_{\mathbf{g}} \tilde{\mathbf{E}}_{\mathbf{g}} e^{i\mathbf{K}_{\mathbf{g}}\mathbf{r}},$$

where $\mathbf{g}$ is the vector of the crystal reciprocal lattice and $\mathbf{K}_{\mathbf{g}} = \mathbf{K}_0 + \mathbf{g}$, where $\mathbf{K}_0$ is the carrier wave vector of the transmitted wave (this vector is chosen arbitrarily [1]); $|\mathbf{K}_0| = k$. Amplitudes $\tilde{\mathbf{E}}_{\mathbf{g}}$ are slowly varying (at atomic distances) functions of coordinates, whereas the exponentials are microscopic values and change rapidly at distances on the order of atomic. Let us rewrite (4) in the form

$$\Delta\tilde{\mathbf{E}} - \operatorname{grad}\operatorname{div}\tilde{\mathbf{E}} + k^2(\tilde{\mathbf{E}} + \tilde{\mathbf{P}}/\varepsilon_0) = 0. \qquad (18)$$

Substituting (17) into (18) and carrying out a derivation similar to the derivation of the Takagi equations [1] in the linear theory, we arrive at nonlinear Takagi equations. Let us recall the key points of this derivation. Only the first derivatives of amplitudes are retained in the first term of (18). In the second term of (18), it is taken into account that, in view of the smallness of susceptibility in the range of X-ray frequencies (it is on the order of 10^{-5}–10^{-6}), $\operatorname{div}\tilde{\mathbf{E}}$ is also small (the electric field is almost transverse, see (2)). Based on this circumstance, all derivatives of amplitudes are rejected in the second term and only rapidly oscillating exponentials are differentiated. Thus, we obtain an infinite system of equations:

$$2i(\mathbf{K}_{\mathbf{g}}\nabla)\tilde{\mathbf{E}}_{\mathbf{g}} + k^2\tilde{\mathbf{E}}_{\mathbf{g}} - \mathbf{K}^2_{\mathbf{g}}\tilde{\mathbf{E}}_{\mathbf{g}[\mathbf{g}]} + \mathbf{K}^2_{\mathbf{g}}\left(\sum_{\mathbf{g}'}\chi^{(1)}_{\mathbf{g}-\mathbf{g}'}\tilde{\mathbf{E}}_{\mathbf{g}'} + \sum_{\mathbf{g}'}A_{\mathbf{g}-\mathbf{g}'}\mathbf{Q}_{1\mathbf{g}'} + \sum_{\mathbf{g}'}B_{\mathbf{g}-\mathbf{g}'}\mathbf{Q}_{2\mathbf{g}'}\right) = 0. \qquad (19)$$

Here,

$$\mathbf{Q}_{1\mathbf{g}'} = \sum_{\mathbf{g}_1,\mathbf{g}_2}(\tilde{\mathbf{E}}_{\mathbf{g}'+\mathbf{g}_1-\mathbf{g}_2}\tilde{\mathbf{E}}^*_{\mathbf{g}_1})\tilde{\mathbf{E}}_{\mathbf{g}_2},$$
$$\mathbf{Q}_{2\mathbf{g}'} = \sum_{\mathbf{g}_1,\mathbf{g}_2}(\tilde{\mathbf{E}}_{\mathbf{g}'-\mathbf{g}_1+\mathbf{g}_2}\tilde{\mathbf{E}}_{\mathbf{g}_1})\tilde{\mathbf{E}}^*_{\mathbf{g}_2}, \qquad (20)$$

and $\tilde{\mathbf{E}}_{\mathbf{g}[\mathbf{g}]}$ is the component of vector $\tilde{\mathbf{E}}_{\mathbf{g}}$ that is perpendicular to $\mathbf{K}_{\mathbf{g}}$ and equal to $[\mathbf{K}_{\mathbf{g}}[\mathbf{E}_{\mathbf{g}}\mathbf{K}_{\mathbf{g}}]]/\mathbf{K}^2_{\mathbf{g}}$. For an absorbing crystal, susceptibilities are assumed to be complex values, the imaginary parts of which determine the absorption in the crystal. For a deformed crystal, the following transition is implemented in Eq. (19): $\chi^{(1)}_{\mathbf{g}}$, $A_{\mathbf{g}}$, $B_{\mathbf{g}} \Rightarrow \chi^{(1)}_{\mathbf{g}}\exp(-i\mathbf{g}\mathbf{u})$, $A_{\mathbf{g}}\exp(-i\mathbf{g}\mathbf{u})$, $B_{\mathbf{g}}\exp(-i\mathbf{g}\mathbf{u})$, where $\mathbf{u}(\mathbf{r})$ is the displacement vector of atoms from their equilibrium positions in an ideal crystal [1]. System of equations (19) is the system of nonlinear Takagi equations.

Let us consider the two-wave approximation, in which two waves corresponding to sites 0 and $\mathbf{h}$ of the reciprocal lattice propagate in the crystal. The Oy axis is assumed to be perpendicular to the diffraction plane, where s_0 and s_h are coordinates in the propagation directions of the transmitted and diffracted waves, respectively. It is expedient to choose carrier wave vector $\mathbf{K}_0$ in system (19) to be oriented exactly at the Bragg angle with respect to the reflecting planes; in this case, $|\mathbf{K}_0| = |\mathbf{K}_h| = k$. The two-wave-diffraction equations can be simplified by introducing amplitudes $\mathbf{E}_{0,\mathbf{h}} = \tilde{\mathbf{E}}_{0,\mathbf{h}}\exp[-ik\chi^{(1)}_0 z/(2\cos\theta)]$, where θ is the Bragg angle and z is the coordinate along the reflecting planes; the Ox axis lies in the diffraction plane and is antiparallel to diffraction vector $\mathbf{h}$. Then, the terms with factors $\mathbf{K}_{0,\mathbf{h}}^2\chi_0^{(1)}$ can be rejected. System (19) takes the form

$$\begin{gathered}\frac{2i}{k}\frac{\partial \mathbf{E}_0}{\partial s_0} + (A_0(|\mathbf{E}_0|^2 + |\mathbf{E}_\mathbf{h}|^2) + A_{\bar{\mathbf{h}}}(\mathbf{E}_\mathbf{h}\mathbf{E}_0^*) \\ + A_\mathbf{h}(\mathbf{E}_0\mathbf{E}_\mathbf{h}^*))\exp[-\mu z/\cos(\theta)]\mathbf{E}_0 + (B_0(\mathbf{E}_0\mathbf{E}_0) \\ +2B_{\bar{\mathbf{h}}}(\mathbf{E}_\mathbf{h}\mathbf{E}_0) + B_{2\bar{\mathbf{h}}}(\mathbf{E}_\mathbf{h}\mathbf{E}_\mathbf{h}))\exp[-\mu z/\cos(\theta)]\mathbf{E}_0^* \\ +(\chi_{\bar{\mathbf{h}}}^{(1)}\exp[\mu z/\cos(\theta)] + A_0(\mathbf{E}_0\mathbf{E}_\mathbf{h}^*) \\ + A_{\bar{\mathbf{h}}}(|\mathbf{E}_0|^2 + |\mathbf{E}_\mathbf{h}|^2) + A_{2\bar{\mathbf{h}}}(\mathbf{E}_\mathbf{h}\mathbf{E}_0^*))\exp[-\mu z/\cos(\theta)]\mathbf{E}_\mathbf{h} \\ + (2B_0(\mathbf{E}_\mathbf{h}\mathbf{E}_0) + B_\mathbf{h}(\mathbf{E}_0\mathbf{E}_0) \\ + B_{\bar{\mathbf{h}}}(\mathbf{E}_\mathbf{h}\mathbf{E}_\mathbf{h}))\exp[-\mu z/\cos(\theta)]\mathbf{E}_\mathbf{h}^* = 0, \qquad (21) \\ \frac{2i}{k}\frac{\partial \mathbf{E}_\mathbf{h}}{\partial s_\mathbf{h}} + (A_0(|\mathbf{E}_0|^2 + |\mathbf{E}_\mathbf{h}|^2) + A_\mathbf{h}(\mathbf{E}_0\mathbf{E}_\mathbf{h}^*) \\ + A_{\bar{\mathbf{h}}}(\mathbf{E}_\mathbf{h}\mathbf{E}_0^*))\exp[-\mu z/\cos(\theta)]\mathbf{E}_\mathbf{h} + (B_0(\mathbf{E}_\mathbf{h}\mathbf{E}_\mathbf{h}) \\ +2B_\mathbf{h}(\mathbf{E}_\mathbf{h}\mathbf{E}_0) + B_{2\mathbf{h}}(\mathbf{E}_0\mathbf{E}_0))\exp[-\mu z/\cos(\theta)]\mathbf{E}_\mathbf{h}^* \\ +(\chi_\mathbf{h}^{(1)}\exp[\mu z/\cos(\theta)] + A_0(\mathbf{E}_\mathbf{h}\mathbf{E}_0^*) + A_\mathbf{h}(|\mathbf{E}_0|^2 + |\mathbf{E}_\mathbf{h}|^2) \\ + A_{2\mathbf{h}}(\mathbf{E}_0\mathbf{E}_\mathbf{h}^*))\exp[-\mu z/\cos(\theta)]\mathbf{E}_0 + (2B_0(\mathbf{E}_\mathbf{h}\mathbf{E}_0) \\ + B_\mathbf{h}(\mathbf{E}_0\mathbf{E}_0) + B_{\bar{\mathbf{h}}}(\mathbf{E}_\mathbf{h}\mathbf{E}_\mathbf{h}))\exp[-\mu z/\cos(\theta)]\mathbf{E}_0^* = 0.\end{gathered}$$

Here, μ is the linear absorption coefficient of the crystal. In the case of nonabsorbing crystal, $\chi_\mathbf{g}^* = \chi_{\bar{\mathbf{g}}}$, $A_\mathbf{g}^* = A_{\bar{\mathbf{g}}}$, $B_\mathbf{g}^* = B_{\bar{\mathbf{g}}}$ for arbitrary $\mathbf{g}$. With allowance for this fact, multiplying the first and second equations of system (21) by $\mathbf{E}_0^*$, $\mathbf{E}_\mathbf{h}^*$, respectively, and the equations complex-conjugate to them by $\mathbf{E}_0$, $\mathbf{E}_\mathbf{h}$ and summing the four thus obtained equations, we arrive at

$$\frac{\partial|\mathbf{E}_0|^2}{\partial s_0} + \frac{\partial|\mathbf{E}_\mathbf{h}|^2}{\partial s_\mathbf{h}} = 0. \qquad (22)$$

Equation (22) is considered in the linear theory [7]; it describes the energy flux conservation law for a nonabsorbing crystal. Let us introduce, as in [7, 8], unit polarization vectors: $\mathbf{e}_\sigma$ (oriented parallel to the Oy axis), $\mathbf{e}_{0\pi} = [\mathbf{s}_0\mathbf{e}_\sigma]$, and $\mathbf{e}_{\mathbf{h}\pi} = [\mathbf{s}_\mathbf{h}\mathbf{e}_\sigma]$, where $\mathbf{s}_0$ and $\mathbf{s}_\mathbf{h}$ are the unit vectors in the propagation directions of the transmitted and diffracted waves, respectively.

If the incident wave has both σ-, and π-polarized components, as follows from (21), due to the presence of nonlinear terms, both polarizations are interrelated by the same system of equations (i.e., their interactions propagate) and their propagation equations are not separated. If the incident wave has only σ- or only π-polarized component, system of equations (21) yields that π- and σ-polarized components have zero amplitudes in the first and second cases, respectively. The reason is as follows. In the first case, according to the boundary conditions, the transmitted-wave amplitude is zero on the crystal surface, and, under this boundary condition, system (21) allows for zero solution for the amplitudes of π-polarized component. Just the same, the amplitudes of σ-polarized components are zero in the second case. Thus, system of equations (21) is reduced to the following system for an incident σ-polarized wave:

$$\begin{gathered}\frac{2i}{k}\frac{\partial E_0}{\partial s_0} + (\eta_0^{(3)}(|E_0|^2 + |E_h|^2) \\ + \eta_h^{(3)}E_0E_h^* + \eta_{\bar{h}}^{(3)}E_0^*E_h)\exp[-\mu z/\cos(\theta)]E_0 \\ +(\chi_{\bar{h}}^{(1)}\exp[\mu z/\cos(\theta)] + \eta_0^{(3)}E_0E_h^* + \eta_{\bar{h}}^{(3)}(|E_0|^2 + |E_h|^2) \\ + \eta_{2\bar{h}}^{(3)}E_0^*E_h)\exp[-\mu z/\cos(\theta)]E_h = 0, \qquad (23) \\ \frac{2i}{k}\frac{\partial E_h}{\partial s_h} + (\eta_0^{(3)}(|E_0|^2 + |E_h|^2) + \eta_h^{(3)}E_0E_h^* \\ + \eta_{\bar{h}}^{(3)}E_0^*E_h)\exp[-\mu z/\cos(\theta)]E_h \\ +(\chi_h^{(1)}\exp[\mu z/\cos(\theta)] + \eta_0^{(3)}E_0^*E_h + \eta_h^{(3)}(|E_0|^2 + |E_h|^2) \\ +\eta_{2h}^{(3)}E_0E_h^*)\exp[-\mu z/\cos(\theta)]E_0 = 0,\end{gathered}$$

($\eta^{(3)} = A + B$). For simplicity, subscripts $\mathbf{h}, \bar{\mathbf{h}}$ in (23) and below are taken in the form $h, \bar{h}$.

In the case of incident π-polarized wave, we multiply the first and second equations of (21) by $\mathbf{e}_{0\pi}$ and $\mathbf{e}_{\mathbf{h}\pi}$, respectively. As a result, we arrive at the following system of equations:

$$\begin{gathered}\frac{2i}{k}\frac{\partial E_0}{\partial s_0} + (\eta_0^{(3)}(|E_0|^2 + |E_h|^2) - B_0\sin^2 2\theta|E_h|^2 \\ + \eta_h^{(3)}\cos 2\theta(E_0E_h^*) \\ +\eta_{\bar{h}}^{(3)}\cos 2\theta(E_hE_0^*))\exp[-\mu z/\cos(\theta)]E_0 \\ + (\chi_{\bar{h}}^{(1)}\exp[\mu z/\cos(\theta)] + \eta_0^{(3)}\cos 2\theta(E_0E_h^*) \\ +\eta_{\bar{h}}^{(3)}(|E_0|^2 + |E_h|^2) + (\eta_{2\bar{h}}^{(3)}\cos 2\theta + B_{2\bar{h}}\tan 2\theta\sin 2\theta) \\ \times(E_hE_0^*))\exp[-\mu z/\cos(\theta)]E_h\cos 2\theta = 0, \\ \frac{2i}{k}\frac{\partial E_h}{\partial s_h} + (\eta_0^{(3)}(|E_0|^2 + |E_h|^2) - B_0\sin^2 2\theta|E_0|^2 \qquad (24) \\ + \eta_h^{(3)}\cos 2\theta(E_0E_h^*) \\ +\eta_{\bar{h}}^{(3)}\cos 2\theta(E_hE_0^*))\exp[-\mu z/\cos(\theta)]E_h \\ + (\chi_h^{(1)}\exp[\mu z/\cos(\theta)] + \eta_0^{(3)}\cos 2\theta(E_hE_0^*) \\ +\eta_h^{(3)}(|E_0|^2 + |E_h|^2) + (\eta_{2h}^{(3)}\cos 2\theta + B_{2h}\tan 2\theta\sin 2\theta) \\ \times(E_0E_h^*))\exp[-\mu z/\cos(\theta)]E_0\cos 2\theta = 0.\end{gathered}$$

Within the classical theory of susceptibility, $B_{0,2h,2\bar{h}} = \eta_{0,2h,2\bar{h}}^{(3)}/3$.

Systems of equations (23) and (24) show that, instead of constant values of susceptibilities responsible for scattering in the linear approximation, the susceptibility coefficients in the nonlinear theory are modulated by diffracted-wave amplitudes in a crystal. These systems of equations can be used to determine the amplitudes by numerical methods for an incident wave of an arbitrary form in both ideal and deformed crystals.

INCIDENT PLANE WAVE: SYMMETRIC LAUE CASE

Let us consider the diffraction of an incident plane σ-polarized wave in an ideal crystal in the symmetric Laue case (the reflecting planes are perpendicular to the input crystal surface). We introduce an $Oxyz$ coordinate system, where the Oz axis is directed into the crystal bulk perpendicular to the input surface, the Ox axis lies in the diffraction plane and is antiparallel to the diffraction vector, and the Oy axis is perpendicular to the diffraction plane. The incident-wave electric field on the input crystal surface ($z = 0$) can be written as

$$E^{(i)}(x,0) = E_0^{(i)} \exp(ik\sin\theta^{(i)}x), \tag{25}$$

where $E_0^{(i)}$ is a constant amplitude and $\theta^{(i)}$ is the angle between the incident-wave propagation direction and the reflecting planes. Let us denote the deviation from the exact Bragg direction as $\Delta\theta = \theta^{(i)} - \theta$. In this case, the solution to (23) can be presented as

$$E_{0,h} = F_{0,h}(z)\exp(ipx), \tag{26}$$

where parameter p must be determined from the boundary conditions. The boundary conditions on the input surface ($z = 0$) have the same form as in the linear theory, i.e.,

$$\begin{gathered} E_0(x,0) = E_0^{(i)} \exp(ik\cos\theta\Delta\theta x), \\ E_h(x,0) = 0. \end{gathered} \tag{27}$$

It follows from (25)–(27) that

$$\begin{gathered} F_0(0) = E_0^{(i)}, \\ F_h(0) = 0, \\ p = k\cos\theta\Delta\theta. \end{gathered} \tag{28}$$

Substituting (26) into (23), one can write the propagation equations as

$$\begin{gathered} 2ik\cos\theta\frac{dF_0}{dz} - 2kp\sin\theta F_0 \\ + k^2\Big[\eta_0^{(3)}(|F_0|^2 + |F_h|^2) \\ + \eta_h^{(3)}F_0F_h^* + \eta_{\bar{h}}^{(3)}F_0^*F_h\Big]\exp[-\mu z/\cos(\theta)]F_0 \\ + k^2\Big[\chi_{\bar{h}}^{(1)}\exp[\mu z/\cos(\theta)] + \eta_0^{(3)}F_0F_h^* + \eta_{\bar{h}}^{(3)}(|F_0|^2 + |F_h|^2) \\ + \eta_{2\bar{h}}^{(3)}F_0^*F_h\Big]\exp[-\mu z/\cos(\theta)]F_h = 0, \\ 2ik\cos\theta\frac{dF_h}{dz} + 2kp\sin\theta F_h \\ + k^2\Big[\eta_0^{(3)}(|F_0|^2 + |F_h|^2) \\ + \eta_h^{(3)}F_0F_h^* + \eta_{\bar{h}}^{(3)}F_0^*F_h\Big]\exp[-\mu z/\cos(\theta)]F_h \\ + k^2\Big[\chi_h^{(1)}\exp[\mu z/\cos(\theta)] + \eta_0^{(3)}F_0^*F_h + \eta_h^{(3)}(|F_0|^2 + |F_h|^2) \\ + \eta_{2h}^{(3)}F_0F_h^*\Big]\exp[-\mu z/\cos(\theta)]F_0 = 0. \end{gathered} \tag{29}$$

In this case, the energy-flux conservation law for a nonabsorbing crystal (22) takes the form

$$|F_0(z)|^2 + |F_h(z)|^2 = \text{const} = |E_0^{(i)}|^2 = I, \tag{30}$$

where I is the incident-wave intensity. The value I of the first integral was obtained using boundary conditions (28). Multiplying the first and second equations of (29) by dF_0^*/dz and dF_h^*/dz, respectively, and the first and second equations of the complex-conjugate system by dF_0/dz and dF_h/dz, respectively, and summing the thus derived four equations, we obtain the second integral of motion in a nonabsorbing crystal:

$$\begin{gathered} p\sin\theta\frac{|F_h|^2 - |F_0|^2}{k} + \mathrm{Re}[\chi_h^{(1)}F_0F_h^*] \\ + \frac{\eta_0^{(3)}}{2}|F_0|^2|F_h|^2 + \mathrm{Re}[\eta_h^{(3)}IF_0F_h^*] \\ + \frac{1}{2}\mathrm{Re}[\eta_{2h}^{(3)}F_0^2F_h^{*2}] = \text{const} = -\frac{p\sin\theta I}{k}. \end{gathered} \tag{31}$$

System of equations (29) can efficiently be used to find numerically both exact plane-wave solutions and amplitudes for an ideal crystal.

EXAMPLE

As an example, we will consider the following conditions: Si(220) reflection, incident radiation wavelength $\lambda = 0.71$Å (17.46 keV), symmetric Laue case, σ-polarized wave, and extinction length corresponding to linear susceptibility: $\Lambda^L = \lambda\cos\theta/|\chi_{hr}^{(1)}| = 36.6$ μm. The values of the real and imaginary parts of linear susceptibility are taken from [8]. Based on formulas (8), (9), (15), and (16), we assume that $\chi_{0r,hr,2hr}^{(3)} = -\chi_{0r,hr,2hr}^{(1)}/E_{cr}^2$ for the real parts of the third-order susceptibility and that $\chi_{0i,hi,2hi}^{(3)} = 0.01\chi_{0r,hr,2hr}^{(3)}$ for the imaginary parts (this assumption is in correspondence with the ratio of these values for the linear part of susceptibilities) and substitute these values into the system of equations (23) and (29). Then the wave amplitudes in the crystal are normalized by ampli-

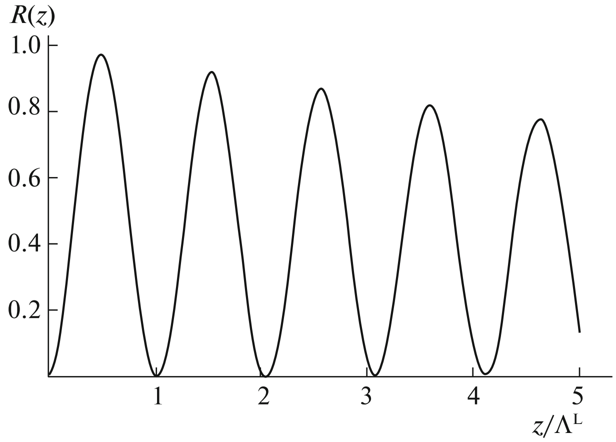

Fig. 1. Pendulum oscillations of the reflectivity of third-order nonlinear plane-wave dynamic diffraction (numerical calculation).

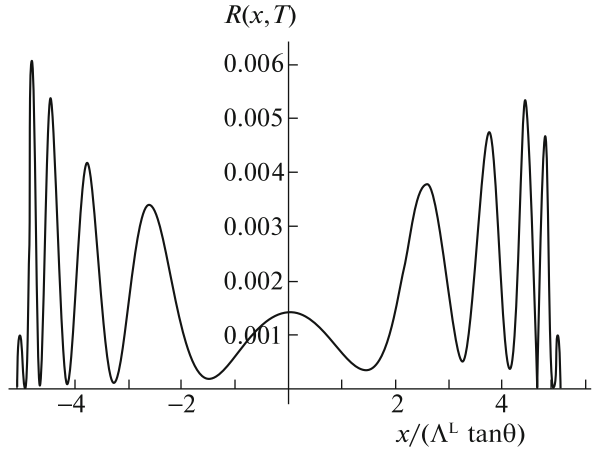

Fig. 2. Intensity distribution on the output crystal surface for third-order nonlinear dynamic diffraction in the case of a point source (numerical calculation).

tude $E_{cr}/\sqrt{3}$ and the intensities are normalized by intensity $I_{cr}/3$. The same values will be used to normalize, respectively, amplitude $E_0^{(i)}$ and intensity I of the incident wave. Let us solve numerically system (29) for a plane wave incident on a crystal at the exact Bragg angle and having intensity $I = 0.03$ (in $I_{cr}/3$ units). This can be done using an algorithm of any standard (providing necessary accuracy) numerical method for solving systems of ordinary differential equations. The computational burden is rather small. The numerically obtained dependence of reflectivity

$$R(z) = \frac{\exp\left(-\frac{k\chi_{0i}^{(1)}}{\cos\theta}z\right)I_h(z)}{I} = \frac{\exp\left(-\frac{k\chi_{0i}^{(1)}}{\cos\theta}z\right)\left|E_h(z)\right|^2}{\left|E_0^{(i)}\right|^2} \quad (32)$$

on z for a crystal with thickness $T = 5\Lambda^L$ is shown in Fig. 1. It can be seen in this figure that the field in the crystal, as in the case of linear polarization, is oscillatory and has a corresponding nonlinear extinction length. In the case of linear polarization, the reflectivity on the output crystal surface would be zero (minimum). However, since the nonlinear extinction length differs from linear, the reflectivity has no minimum. It can also be seen in Fig. 1 that the nonlinear extinction length exceeds linear, which is explained by the renormalization (reduction) of the effective susceptibility (responsible for scattering in crystal). This follows from system of equations (28), where $\chi_{hr}^{(1)}$ and $\eta_{hr}^{(3)}$ have opposite signs. Calculations show that the increase in the extinction length is significant even for the incident radiation intensity equal to 0.01; in addition, this increment is accumulated with an increase in the crystal thickness. Thus, to observe the plane-wave nonlinear pendulum effect, one can use intensities lower than critical by two orders of magnitude.

In the case of inhomogeneous σ-polarized wave incident on a crystal, system (23) must be numerically solved. We will use the half-step algorithm of numerical integration of Takagi equations, which is well known in the linear theory [9, 14], with only one difference: according to (23), in each step of calculating amplitudes, one has effective susceptibilities modulated by the amplitudes of transmitted and diffracted waves at the output of a given layer instead of constant values of susceptibilities; the values calculated at the input of this layer are used for these amplitudes. The calculated intensity distribution

$$R(x,T) = \frac{\exp\left(-\frac{k\chi_{0i}^{(1)}}{\cos\theta}T\right)I_h(x,T)}{I} = \frac{\exp\left(-\frac{k\chi_{0i}^{(1)}}{\cos\theta}T\right)\left|E_h(x,T)\right|^2}{\left|E_0^{(i)}\right|^2} \quad (33)$$

of the diffracted wave on the output surface ($z = T$), as a function of x, for the same conditions as in the previous example but for a point source located on the crystal surface (Kato's case [8, 9]) and for $I = 0.1$ is shown in Fig. 2. In linear theory, this dependence is symmetric with respect to x. As can be seen in Fig. 2, this dependence becomes asymmetric in the nonlinear theory, which is explained by the difference in the reflectivities of rays with different deviations from the exact Bragg direction. Similar calculations show that this asymmetry is pronounced for incident-wave intensities of 0.03 or higher. Therefore, nonlinear dynamic effects for a point source can be observed for intensities two orders of magnitude lower than critical.

CONCLUSIONS

We theoretically considered the two-wave dynamic diffraction of an X-ray wave in an ideal crystal with a nonlinear third-order response to electric field strength and presented a nonlinear analog of Takagi equations: a system of propagation equations for transmitted and diffracted waves in both ideal and deformed crystals. Integrals of motion in a nonabsorbing crystal were obtained in the symmetric Laue case for a plane wave incident on a crystal. These propagation equations can be used to efficiently determine amplitudes numerically. As an example, we presented the results of a numerical calculation of the reflectivity (pendulum effect) of nonlinear dynamic diffraction for a plane wave incident on a crystal at the exact Bragg angle and for a point source located on the crystal surface. The calculations showed that dynamic effects can be observed at intensities 1–2 orders of magnitude lower than critical.

Furthermore, it will be interesting to solve exactly the presented nonlinear equations, numerically investigate other dynamic effects in the nonlinear mode (such as the Borrmann effect, rocking curves, and influence of asymmetry on the nonlinear diffraction), analyze the nonlinear diffraction in the Bragg geometry, etc.

Corresponding experiments can be performed on X-ray synchrotron radiation sources, in particular, using X-ray free-electron lasers.

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