PROCEEDINGS OF THE YEREVAN STATE UNIVERSITY

Physical and Mathematical Sciences 2017, **51**(1), p. 85–88

Physics

X-RAY THIRD-ORDER NONLINEAR RENNINGER EFFECT AND ROCKING CURVES

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In the work third-order nonlinear Takagi's equations for X-ray monochromatic waves are investigated for the forbidden reflection case. The forbidden dynamical diffraction in the nonlinear case is related to the presence in the nonlinear equations of the terms proportional to the zero order and the second order nonzero Fourier coefficients of the third-order nonlinear susceptibility. As a consequence, in the third-order nonlinear Bragg diffraction case, a nonlinear analogue of the well known Renninger effect takes place, which is considered theoretically and numerically. The numerical calculations show that in the Bragg geometry the nonlinear reflection curve's behavior is the same as for not forbidden reflection, but for forbidden reflection the rocking curves are by several orders more sensitive to the input intensity value.

***Keywords*:** third-order nonlinearity, Bragg diffraction, nonlinear Takagi's equations, nonlinear Renninger effect, rocking curves.

Introduction. Successes of high intensity X-ray synchrotron sources and XFELs bring to theoretical and experimental investigations of nonlinear X-ray diffraction and other nonlinear effects of the X-ray interaction with matter as well. X-ray dynamical diffraction is described by Takagi's equations [1]. In the wave equation, replacing the linear susceptibility by the third-order nonlinear one, the stationary and time dependent nonlinear Takagi's equations (NTE) are established [2–6] and third-order nonlinear stationary and time-dependent effects are investigated. In [7], using cold plasma model, the linear dynamical diffraction of the formed X-ray second-order harmonic in a perfect crystal under two wave diffraction conditions has been studied. The backward influence of two Bragg-diffracted waves on the amplitude of the incidence wave was not considered. In [8, 9] without using the cold plasma model, the kinematical diffraction of an intense X-ray plane wave under the second order nonlinearity conditions with parametric-down conversion of an X-ray photon into an X-ray low frequency photon and an UV photon was investigated. Using the third-order nonlinear cold plasma model, in [10] the direct propagation of an intense X-ray beam was investigated. For forbidden reflections the third-order nonlinear two-wave dynamical diffraction takes place due to nonlinear nonzero susceptibilities Fourier-coefficients of second-order diffraction vector. Thus a third-order nonlinear Renninnger's effect [11] analogue takes place. In this paper, basing on the stationary NTE the third-order nonlinear Renninger effect both theoretically and numerically is investigated. It is shown that the third-order nonlinear rocking curves in the forbidden reflection case are very sensitive to the input energy and its variation.

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Basic Formulas. In nonlinear non-magnetic medium, the wave equation for a monochromatic wave $\mathbf{E}(\mathbf{r},t)=\mathbf{E}(\mathbf{r},\omega)\exp(-i\omega t)+c.c.$ has the form

$$rot\,rot\mathbf{E}+\frac{1}{c^2}\cdot\frac{\partial^2\mathbf{E}}{\partial t^2}=-\frac{1}{\varepsilon_0 c^2}\cdot\frac{\partial^2\mathbf{P}}{\partial t^2}, \tag{1}$$

where the polarization $\mathbf{P}(\mathbf{r},t)=\mathbf{P}(\mathbf{r},\omega)\exp(-i\omega t)+c.c.$ is the sum of the linear and third-order nonlinear components:

$$\mathbf{P}(\mathbf{r},\omega)=\mathbf{P}^{(1)}(\mathbf{r},\omega)+\mathbf{P}^{(3)}(\mathbf{r},\omega). \tag{2}$$

The linear and nonlinear polarizations are presented via linear and nonlinear susceptibilities

$$\begin{gathered}\mathbf{P}^{(1)}(\mathbf{r},\omega)=\varepsilon_0\chi^{(1)}(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega),\\ P_i^{(3)}(\mathbf{r},\omega)=3\varepsilon_0\chi_{ijkl}(\omega;\,\omega,\omega,-\omega,\mathbf{r})E_j(\mathbf{r},\omega)E_k(\mathbf{r},\omega)E_l^*(\mathbf{r},\omega),\end{gathered} \tag{3}$$

where the summation is performed over the dummy indices, and ε_0 is the permittivity of the free space. From Eq. (1) we obtain the wave equation for the monochromatic wave amplitude:

$$rot\,rot\mathbf{E}(\mathbf{r},\omega)-\frac{\omega^2}{c^2}\Big(1+\chi^{(1)}(\mathbf{r},\omega)\Big)\mathbf{E}(\mathbf{r},\omega)=\frac{\omega^2}{\varepsilon_0 c^2}\mathbf{P}^{(3)}(\mathbf{r},\omega). \tag{4}$$

It is assumed that the crystal, as in the linear theory, may be considered as isotropic media. In this case $\chi^{(1)}(\mathbf{r},\omega)$ is a scalar and the third-order nonlinear susceptibility is expressed as [12]

$$\chi_{ijkl}^{(3)}(\omega;\omega,\omega,-\omega,\mathbf{r})=\frac{1}{3}\chi^{(3)}(\omega,\mathbf{r})(\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_j k), \tag{5}$$

where [2]

$$\chi^{(3)}(\omega,\mathbf{r})\approx\frac{n(\mathbf{r})e^4a_0^4}{\varepsilon_0\hbar^3\omega^3}\approx(10^{-31}-10^{-33})\;m^2/V^2, \tag{6}$$

here $n(\mathbf{r})\approx(10^{-28}-10^{-30})\;m^{-3}$ is the electron concentration and $a_0=5.3\cdot10^{-11}\;m$ is the Bohr radius. Both $\chi^{(1)}(\mathbf{r},\omega)$ and $\chi^{(3)}(\omega,\mathbf{r})$ are $3D$ periodic functions of coordinates and can be expanded into Fourier-series in terms of reciprocal lattice vectors. Inserting these quantities into Eq. (4), presenting the electrical field strength as the sum of quasi-Bloch waves and repeating the derivation of the Takagi's equations, the NTE for two-wave diffraction case, has been obtained [2]. For a forbidden reflection $\chi^{(1)}=0$ and $\eta^{(3)}=0$ these equations for σ-polarization can be written in the form

$$\begin{gathered}\frac{2i}{k}\cdot\frac{\partial E_0}{\partial s_0}+\chi_0^{(1)}E_0+\eta_0^{(3)}\left(|E_0|^2+|E_h|^2\right)E_0+(\eta_0^{(3)}E_0E_h^*+\eta_{2\bar{h}}^{(3)}E_0^*E_h)E_h=0,\\ \frac{2i}{k}\cdot\frac{\partial E_h}{\partial s_h}+\chi_0^{(1)}E_h+\eta_0^{(3)}\left(|E_0|^2+|E_h|^2\right)E_h+(\eta_0^{(3)}E_0^*E_h+\eta_{2h}^{(3)}E_0E_h^*)E_0=0,\end{gathered} \tag{7}$$

where $\eta^{(3)}=3\chi^{(3)}$, s_0 and s_h are the coordinates along the propagation directions of the transmitted and diffracted waves, $\chi_{0,h,2h}^{(1)}$ and $\eta_{0,h,2h}^{(3)}$ are the Fourier coefficients of linear and nonlinear susceptibilities respectively. From Eqs. (7) it is seen that even if the reflection is forbidden, due to the presence in the equations the terms proportional to $\eta_{0,2h}^{(3)}$, the nonlinear reflection is not forbidden. This is the third-order nonlinear analogue of the well known Renninger effect [11].

Nonlinear Rocking Curves for Forbidden Reflection. The solution of Eqs. (7) for diffracted wave amplitude both for Laue and Bragg geometries is zero, since such solution satisfies zero boundaries conditions for diffracted wave and Eqs. (7). However, for real crystals, the susceptibilities of forbidden reflections are not exactly zero due to some concentrations of dislocations and other localized defects. It must be assumed that in real crystals the susceptibilities are by several orders smaller than for non-forbidden reflections, but are not exactly zero.

An analytical solution for the third-order nonlinear Bragg case dynamical diffraction in non-absorbing crystals and for $2h$ forbidden reflection has been found in [3], which shows the dependence of the reflection coefficient on the deviation parameter from the Bragg exact condition and on the intensity of the incidence wave.

Let us consider the case of forbidden h reflection, when the $2h$ reflection is not forbidden and for definiteness assume that the Fourier coefficients of forbidden reflection are by two orders less than for not forbidden $2h$ reflection. The Eqs. (7) can be solved numerically using the modified half-step algorithm [2]. Let us consider the case of forbidden $\mathrm{Si}(200)$ MoK_α reflection and symmetrical Bragg geometry (Fig. 1). The following details of dynamical diffraction must be considered: $1)$ the dependence of the rocking curve center on the intensity of the incident wave (y is the standard deviation parameter from the Bragg exact condition [11]); $2)$ the dependence of the reflection coefficient on y for various values of the incident wave intensity. The intensities will be shown in the units of $I_{cr}/3$, where the critical intensity $I_{cr} = E_{cr}^2 = |\chi^{(1)}|/\chi^{(3)}$ and $E_{cr} = \left(\hbar^3\omega/(me^2a_0^4)\right)^{1/2} \approx 1.2 \cdot 10^{13}\ V/m$ [2].

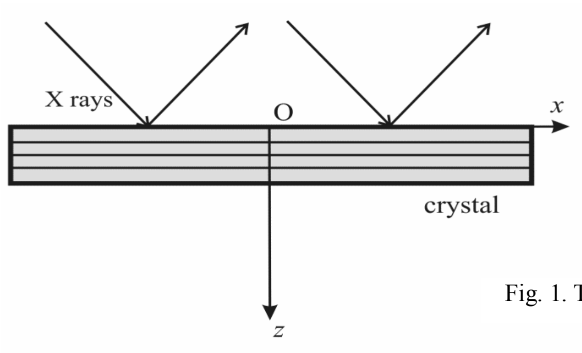

Fig. 1. The Bragg geometry of the diffraction.

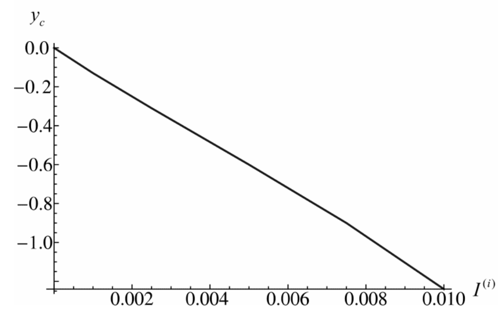

Fig. 2. The dependence of the angular position of the rocking curve on the intensity of incident wave.

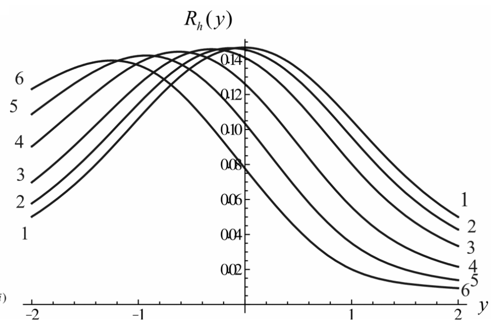

Fig. 3. The rocking curves for various values of input intensity: 1 – 0.0001; 2 – 0.001; 3 – 0.0025; 4 – 0.005; 5 – 0.0075; 6 – 0.01.

The results of numerical calculations are shown in the Figs. 2 and 3. Fig. 2 shows the dependence of the angular position of the rocking curve, and Fig. 3 shows the rocking curves for various values of the incident wave intensity. The dependence of the angular position rocking curve on the input energy value is a line, i.e. is the same as for non-forbidden reflection [3]. With the increase of the intensity the position of the rocking curve shifts towards the low angles. According to Fig. 3, the dependence of reflection coefficient on the input energy is also the same as for non-forbidden reflection, i.e. with the increase of the intensity the rocking curve shifts towards the low angles. The main difference between nonlinear rocking curves for forbidden and non-forbidden reflections is associated with much higher sensitivity of the nonlinear rocking curves to the input energy value (the difference in the sensitivity is two orders as a result of assumptions made related to susceptibility $\chi_h^{(1)}$). For forbidden reflection, the variation of the input energy by 0.001 results in a considerable shift of the reflection curve, meanwhile for non-forbidden reflection the reflection curve shifts by the same value for the intensity 0.1.

Conclusion. The third-order nonlinear two-wave dynamical diffraction is analyzed for the forbidden reflection theoretically and numerically. The numerical solutions in the case of the forbidden Bragg reflection show that main features of rocking curves are the same as for non-forbidden reflection. However, rocking curves of forbidden nonlinear reflection are more sensitive (by several orders) to the incident wave intensity than those in the case of non-forbidden reflection. This sensitivity provides a new opportunity to investigate the nonlinear dynamical diffraction effects.

Received 22.12.2016

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