Feature of Kinematic Diffraction of Nonmonochromatic Divergent X-Ray Beams in a Crystal with a Periodically Strained Lattice

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Abstract—Coherent scattering of a nonmonochromatic divergent beam of X-ray quanta in a crystal excited by an acoustic wave is described using a kinematic approach. The possibility of manifestation of a peculiar effect of amplification of the diffracted X-ray beam by the acoustic wave is predicted using this approach.

INTRODUCTION

Increasing interest in the study of high-energy electromagnetic processes in crystals under the action of external fields (e.g., acoustic waves) is due to the possibility of controlling the yield of such processes and detecting new effects important for applications; of special interest is the enhancement of X-ray beams diffracted in a crystal that is excited by an acoustic wave. The observation of the effect discussed here was reported for the first time in the 1930s [1–3]. In subsequent experiments, a considerable increase in the intensity of diffracted beams upon an increase in the amplitude of the acoustic wave was observed [4–7]. Theoretical analysis of the effect revealed that no increase in the integrated intensity of radiation scattered into a Bragg reflection takes place in the framework of kinematic diffraction; i.e., the effect is purely dynamic (see, for example, [8]). The development of theoretical approaches to studying dynamic diffraction of X-rays in strained crystals [9–12] has made it possible to describe the effect in detail [13–15].

Here, we consider peculiarities of kinematic diffraction of X-rays in a crystal with a lattice periodically strained by an acoustic wave, which are associated with a finite spectral width and angular divergence of the primary beam, as well as specific features of the experimental setup that is conventionally used for studying the amplification effect, in which the axis of the primary photon beam incident on the sample, as well as the position of the radiation detector, is fixed. We consider high-frequency modulation [16] indicating the smallness of the acoustic wave length as compared to the cross section of the primary beam of X-ray quanta. It will be shown that under certain conditions, an analog of amplification of scattered radiation due to excitation of an acoustic wave in the crystal can be observed, which may result in a considerable increase in the integrated yield.

We are using the relativistic system of units, in which $\hbar = c = 1$.

INTEGRATED YIELD OF RADIATION SCATTERED INTO BRAGG REFLECTION

Let us consider the diffraction of X rays in a crystal based on the wave equation for the Fourier transform of the electric field,

$$(k^2 - \omega^2)\mathbf{E}_{\omega\mathbf{k}} - \mathbf{k}(\mathbf{k}\mathbf{E}_{\omega\mathbf{k}}) + \int d^3k' G(\mathbf{k}' - \mathbf{k})\mathbf{E}_{\omega\mathbf{k}'} = 0,$$
$$G(\mathbf{k}' - \mathbf{k}) = \frac{Ze^2}{2\pi^2 m}\frac{1}{1 + (\mathbf{k}' - \mathbf{k})^2 R^2}\sum_l \exp(i(\mathbf{k}' - \mathbf{k})\mathbf{r}_l, \quad (1)$$

where the last term in the equation is proportional to the induced current of electrons from the target, calculated in the high-frequency approximation $\omega \gg I$, in which atomic electrons can be treated as free in the course of radiation scattering; I is the average ionization potential of the atom, Z is the number of electrons in the atom, and R is the electron screening radius for the atomic nucleus; summation in the last formula of (1) is performed over all atoms of the target.

Decomposing function G into the averaged ($\bar{G}(\mathbf{k}' - \mathbf{k}) = \omega_0^2\,\delta(\mathbf{k}' - \mathbf{k})$, ω_0 is the plasma frequency of the target) component responsible for refraction of photons and the fluctuation component $\tilde{G}(\mathbf{k}' - \mathbf{k})$ responsible for diffraction of photons, we will solve system (1) by iterations in powers of $\tilde{G}$. In the zeroth approximation, we obtain from (1) the following expression for the primary field in the target:

$$\mathbf{E}^{(i)}_{\omega\mathbf{k}} = \int d^2\theta_i f(\theta_i)\sum_{j=1}^{2}\mathbf{e}_{ij}(\theta_i)E_{\omega j}\delta(\mathbf{k} - \omega\sqrt{\varepsilon}\mathbf{n}_i(\theta_i)), \quad (2)$$

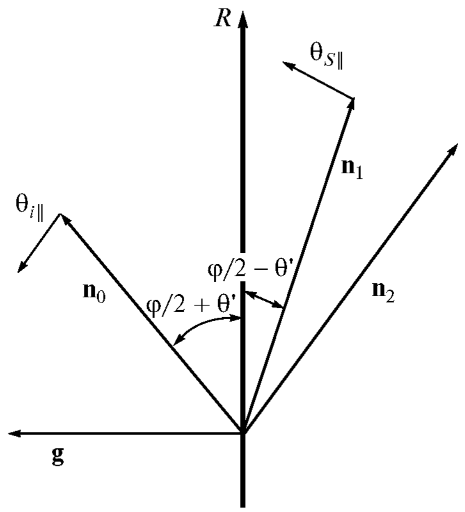

Fig. 1. Geometry of scattering of an X-ray beam: $\mathbf{n}_0$ and $\mathbf{n}_1$ are the axes of the incident and scattered beams, respectively; φ is θ' are the angles of orientation of the sample, which are varied with the help of a goniometer, $\theta_{i\|}$ and $\theta_{S\|}$ are the components of 2D angles lying in the diffraction plane, which describe the angular structure of the incident and scattered beams, respectively; R is the reflecting crystallographic plane, and $\mathbf{g}$ is the reciprocal lattice vector specifying this plane.

where function $f(\theta_i)$ describes the angular spread in the beam of primary quanta, the spectrum is determined by function $E_{\omega j}$, vectors $\mathbf{e}_{ij}$ and $\mathbf{n}_i$ denote the polarization and direction of propagation of photons, $\mathbf{e}_{ij}\mathbf{n}_i = 0$, $\mathbf{n}_i = \mathbf{n}_0(1 - \theta_i^2/2) + \boldsymbol{\theta}_i$, $\mathbf{n}_0\boldsymbol{\theta}_i = 0$, $\mathbf{n}_0$ being the axis of the primary photon beam, and $\varepsilon = 1 - \omega_0^2/\omega^2$ is the conventional permittivity of the medium in the X-ray frequency range. Taking into account the statistical independence of photons, we can write the relations

$$\langle f(\boldsymbol{\theta}_1) f^*(\boldsymbol{\theta}_2)\rangle = f_0(\boldsymbol{\theta}_1)\delta(\boldsymbol{\theta}_1 - \boldsymbol{\theta}_2),$$

where f_0 is the angular distribution functions for primary photons, and

$$\langle E_{\omega j} E^*_{\omega l}\rangle = \frac{1}{2}|\mathbf{E}_\omega|^2\delta_{ij}.$$

In the next approximation, we obtain the diffracted field

$$\mathbf{E}^{(S)}_{\omega\mathbf{k}} = -\frac{1}{k^2 - \omega^2\varepsilon}\int d^2\theta_i f(\theta_i)\tilde{G}(\omega\sqrt{\varepsilon}\mathbf{n}_i - \mathbf{k}) \times \sum_{j=1}^{2}\left(\mathbf{e}_{ij} - \mathbf{k}\frac{\mathbf{k}\mathbf{e}_{ij}}{\omega^2\varepsilon}\right)E_{\omega j}. \tag{3}$$

The spectral–angular distribution of scattered quanta following from this expression has the form

$$\omega\frac{dN}{d\omega d\Omega} = 2\pi^2|\mathbf{E}_\omega|^2\int d^2\theta_i f_0(\theta_i)(1 + (\mathbf{n}_S\mathbf{n}_i)^2) \times \langle|\tilde{G}\omega\sqrt{\varepsilon}(\mathbf{n}_i - \mathbf{n}_S)|^2\rangle, \tag{4}$$

where $\mathbf{n}_S = \mathbf{n}_1(1 - \theta_S^2/2) + \boldsymbol{\theta}_S$ is the unit vector in the direction of propagation of the scattered quantum, $\mathbf{n}_1$ is the axis of the scattered photon beam, and two-dimensional angle $\boldsymbol{\theta}_S$ describes the angular distribution of scattered quanta; angle brackets denote averaging over the positions of target atoms. In this averaging, we take into account the fact that the positions of atoms in a crystal with the lattice modulated by an acoustic wave are given by the formula $\mathbf{r}_l = \mathbf{R}_l + \mathbf{u}_l + \mathbf{a} = \sin(\boldsymbol{\xi}\mathbf{R}_l)$, in which quantities $\mathbf{R}_l$ indicate periodically arranged equilibrium positions of atoms; $\mathbf{u}_l$ are thermal displacements, and $\mathbf{a}$ and $\boldsymbol{\xi}$ are the amplitude and wavevector of the acoustic wave. The result of averaging is the sum of incoherent (small) and coherent terms, the latter term being the sum of reflections each of which corresponds to the contribution to scattering from the crystallographic plane defined by the fixed vector $\mathbf{g}$ of the reciprocal lattice.

In the case of long-wave vibrations of the lattice considered here ($\xi \ll \mathbf{g}$), the distribution of photons coherently scattered by the fixed plane has the form

$$\omega\frac{dN_g}{d\omega d^2\theta_S} \approx \frac{\pi}{4}V\omega_g^4(1 + \cos^2\varphi)|\mathbf{E}_\omega|^2\sum_p J_p^2(\mathbf{g}\cdot\mathbf{a}) \times \int d^2\theta_i f_0(\theta_i)\delta(\omega\sqrt{\varepsilon}(\mathbf{n}_i - \mathbf{n}_S) + p\boldsymbol{\xi} - \mathbf{g}), \tag{5}$$

where V is the volume of the target, $\omega_g^2 = \omega_0^2\exp(-g^2u^2/2)(1 + g^2R^2)^{-2}$, and φ is the fixed scattering angle as shown in Fig. 1 illustrating the scattering geometry.

The effect of modulation of the beam of scattered quanta considered here is associated with bending vibrations of the reflecting plane (formula (5) contains only the amplitude component a perpendicular to the plane).

Let us consider the orientation dependence of the integrated yield of quanta into a fixed reflection. Assuming that the angular and frequency distributions of the beam of primary quanta are Gaussian,

$$f_0(\theta_i) = \frac{1}{\pi\theta_0^2}\exp\left(-\frac{\theta_i^2}{\theta_0^2}\right),$$

$$|\mathbf{E}_\omega|^2 = \frac{|\mathbf{E}_0|^2}{\sqrt{\pi}\Delta\omega}\exp\left[-\left(\frac{\omega - \bar{\omega}}{\Delta\omega}\right)^2\right] \equiv |E_{\max}|^2\exp\left[-\left(\frac{\omega - \bar{\omega}}{\Delta\omega}\right)^2\right],$$

we obtain from relation (5) the final formula

$$N_g(\theta') = N_0 \frac{\theta_*}{\sqrt{\theta_0^2+\theta_*^2}} \sum_p J_p^2(\mathbf{g}\cdot\mathbf{a}) \exp\left(-\frac{\eta_p^2}{\theta_0^2+\theta_*^2}\right)$$

$$\times \operatorname{erf}\left(\frac{\Delta\theta_\perp}{2\theta_0}\right)\left\{\operatorname{erf}\left[\frac{\sqrt{\theta_0^2+\theta_*^2}}{\theta_0\theta_*}\left(\frac{\Delta\theta_\parallel}{2}+\mu_p\right)\right]\right. \tag{6}$$

$$\left.+\operatorname{erf}\left[\frac{\sqrt{\theta_0^2+\theta_*^2}}{\theta_0\theta_*}\left(\frac{\Delta\theta_\parallel}{2}-\mu_p\right)\right]\right\},$$

$$N_0 = \frac{\pi}{4}\frac{V\omega_g^4}{g^2\omega_B}|\mathbf{E}_{\max}|^2(1+\cos^2\varphi),$$

$$\eta_p = \theta' - z - \frac{p}{g\cos(\varphi/2)}(\mathbf{n}_0\xi\cos\varphi + \mathbf{n}_2\boldsymbol{\theta}\sin\varphi),$$

$$\mu_p = \frac{\theta_0^2+2\theta_*^2}{\theta_0^2+\theta_*^2}\theta' + \frac{\theta_0^2}{\theta_0^2+\theta_*^2}z$$

$$-\frac{p}{g\cos(\varphi/2)}\left(\mathbf{n}_0\xi\frac{\theta_0^2+2\theta_*^2\cos^2(\varphi/2)}{\theta_0^2+\theta_*^2} + \mathbf{n}_2\xi\frac{\theta_*^2\sin\varphi}{\theta_0^2+\theta_*^2}\right),$$

where $\omega_B = g/2\,\sin(\varphi/2)$ is the Bragg frequency with the reflection spectrum concentrated in its neighborhood, $\Delta\theta_\perp$ and $\Delta\theta_\parallel$ are the angular dimensions of the collimator of radiation, $\theta_* = \frac{\Delta\omega}{\omega_B}\tan(\varphi/2)$, and $z = \frac{\omega_B - \bar{\omega}}{\omega_B}\tan(\varphi/2)$. The latter coefficients characterize the effect of the finiteness of the spectral width for primary photons and detuning of the Bragg resonance for the average frequency of the beam on the orientation dependence of scattered radiation yield considered here.

GENERAL ANALYSIS OF SCATTERED RADIATION

Let us analyze the result. Above all, it should be noted that the effect of the wave can be significant only for a large wave amplitude, for which the condition $\mathbf{g}\cdot\mathbf{a} > 1$ is satisfied, which indicates that the amplitude is comparable with the interplanar spacing in the crystal. Indeed, for $\mathbf{g}\cdot\mathbf{a} \ll 1$, only the terms proportional to $J_0^2(\mathbf{g}\cdot\mathbf{a}) \approx 1$ in sum (6) remains significant. We can prove that the conditions optimal for controlling the orientation curve are observed when the wave propagates over the reflecting crystallographic plane and there is no detuning ($z = 0$). In this case,

$$\eta_p = \theta' - p\frac{\xi}{g}, \quad \mu_p = \frac{\theta_0^2+2\theta_*^2}{\theta_0^2+\theta_*^2}\eta_p$$

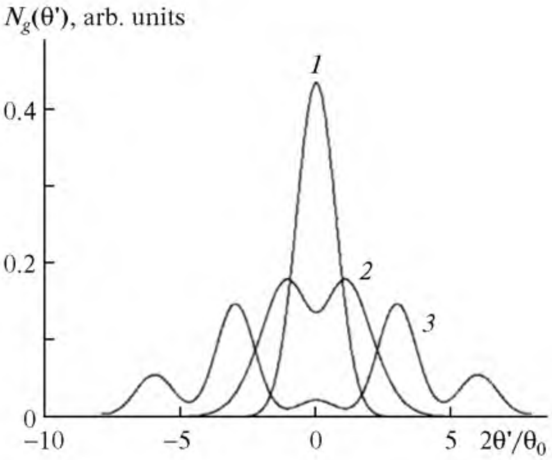

Fig. 2. Orientation dependence of the scattered radiation yield. The curves are calculated by formula (7) for fixed values of parameters $2\Delta\theta/\theta_0 = 0.2$, $\mathbf{g}\cdot\mathbf{a} = 2$, and for various values of parameter $2\xi/g\theta_0$: 0.1 (*1*), 1 (*2*), and 3 (*3*).

and the orientation curve is the sum of identical peaks with different amplitudes, which are displaced relative to one another by a distance multiple of ξ/g.

Obviously, the acoustic wave leads to broadening of the orientation curve. The extent of broadening can be estimated easily. Since a considerable contribution to the sum over p in Eq. (6) comes from the terms with $|p| \le \mathbf{g}\cdot\mathbf{a}$ (properties of Bessel's functions), the sought broadening can be estimated as $\Delta\theta' \approx 2\mathbf{g}\cdot\mathbf{a}\xi/g \approx 2\xi a$; i.e., the broadening increases with the amplitude of the acoustic wave and upon a decrease in the wavelength. The $N_g(\theta')$ dependence has the simplest form in the case of a broadband ($\theta_0^2 \ll \theta_*^2$) or narrowband ($\theta_0^2 \gg \theta_*^2$) beam of primary photons. In accordance with Eq. (6), under the given conditions, the exponential in Eq. (6) becomes a slow function as compared to the combination of error functions in the square brackets in (6). Moreover, in the vicinity of the minimum of this combination, the exponential differs from unity insignificantly and can be omitted. For example, in the first limiting case, general formula (6) can be reduced to the simple result

$$N_g(\theta') = N_0 \operatorname{erf}\left(\frac{\Delta\theta_\perp}{2\theta_0}\right)\sum_p J_p^2(\mathbf{g}\cdot\mathbf{a})\left[\operatorname{erf}\left(\frac{\Delta\theta_\parallel}{2\theta_0}+\frac{2\theta'}{\theta_0}-\frac{2p\xi}{g\theta_0}\right)\right.$$
$$\left.\times\operatorname{erf}\left(\frac{\Delta\theta_\parallel}{2\theta_0}+\frac{2\theta'}{\theta_0}-\frac{2p\xi}{g\theta_0}\right)\right], \tag{7}$$

which shows that the characteristic scale of elementary peaks constituting the $N_g(\theta')$ dependence is the angular spread of the beam of primary photons under the given conditions. It should be noted that for $2\xi/g\theta_0 < 1$, individual satellites in expression (7) overlap. In this case, the $N_g(\theta')$ dependence is smooth. On

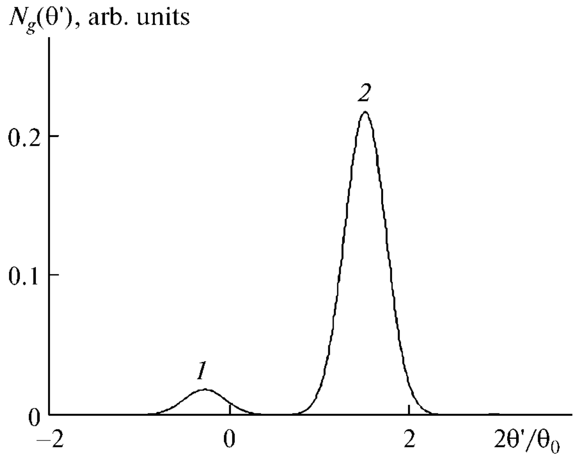

Fig. 3. Effect of an acoustic wave on the scattered radiation yield. The curves are calculated by general formula (6) for fixed values of parameters $\varphi = \pi/2$, $2\Delta\theta_{\|}/\sqrt{2}\,\theta_0 = 0.3$, $\theta_0 = \theta_*$, $\mathbf{g}\cdot\mathbf{a} = 2$, $\xi/2g\theta_0 = 1.5$, and $\mathbf{n}_0\cdot\xi = \xi$. Resonance condition (8) was satisfied for the satellite with number $p = 1$. Curve *1* corresponds to scattering from an unperturbed crystal; curve *2* describes the orientation dependence of the scattered radiation yield in the presence of an acoustic wave.

the other hand, this dependence becomes oscillating in the opposite case.

Function $N_g(\theta')$ is represented in Fig. 2 by the curves calculated by formula (7) for different values of the parameters of the problem. The curves confirm the experimentally observed effect of a considerable change in the orientation dependence of the yield of diffracted photons upon excitation by an acoustic wave in the crystal [17, 18]. It should also be noted that the characteristic shape of the orientation curves determined by the behavior of Bessel's functions in the vicinity of the point at which the order and argument are identical is the same as that observed in experiment [18].

AMPLIFICATION OF A DIFFRACTED X-RAY BEAM BY AN ACOUSTIC WAVE

Let us now analyze the possibility of manifestation of an increase in the yield of X-rays scattered by a crystalline target by an acoustic wave. It should be noted above all that the total yield of radiation scattered by a periodically strained system of atomic planes in the crystal does not exceed the analogous quantity for the unperturbed system (this statement follows from Eq. (6) in the limit $\Delta\theta_\perp, \Delta\theta_{\|} \longrightarrow \infty$ taking into account the relation $\sum_p J_p^2(\mathbf{g}\cdot\mathbf{a}) = 1$). Thus, the amplification effect is not observed in pure form in the framework of kinematic diffraction.

Nevertheless, an analog of enhancement can be manifested owing to specific features of the conventional experimental setup, in which the axes of the incident and scattered beams are fixed (angle φ is determined by the photon channels for the primary and scattered beams in experiments in a vacuum chamber or determined by the position of the detector of radiation with a finite size). In such experiments, Bragg's resonance condition $\bar{\omega} = \omega_B$ or $z = 0$ must be satisfied exactly. However, this is not always possible (i.e., in the case of a primary beam with an asymmetric frequency distribution). Tuning of the crystal in orientation angle θ' can compensate this inaccuracy; however, the scattered beam in this case may leave the boundaries of the fixed collimator. This peculiarity is reflected in formula (6) containing the product of two orientation-dependent bell-shaped factors (exponential and combination of error functions in the square brackets). The positions of the maxima of these factors, which are determined by the conditions $\eta_p = 0$ and $\mu_p = 0$, generally do not coincide. In the case of an unperturbed crystal, the angular separation $\delta\theta' = 2z$ between the peaks may prove to be sufficient for complete or partial suppression of the yield of radiation scatted by the given crystallographic plane. On the other hand, $\delta\theta'$ can be compensated at the relevant peaks of the sum in (6). It can easily be shown that the positions of the peaks of the exponential and the pth term in the sum in (6) coincide when the following condition is satisfied:

$$z = \frac{p}{g}\tan\left(\frac{\varphi}{2}\right)\left[\mathbf{n}_0\xi\sin\left(\frac{\varphi}{2}\right) - \mathbf{n}_2\xi\cos\left(\frac{\varphi}{2}\right)\right]. \qquad (8)$$

The scattered radiation yield can sharply increase in this case. Figure 3 illustrates an example of observation of the effect of a considerable increase in the yield of the scattered beam under the effect of an acoustic wave; the curves show the $N_g(\theta')$ dependence in the case of an unperturbed crystals and a crystal with a wave-modulated lattice.

It should be borne in mind that the effect considered here can be observed only in the conditions when the effects of angular divergence and finiteness of the spectral width for the primary beam are approximately the same ($\theta_0^2 \approx \theta_*^2$) because if one of these parameters prevails, the role of the exponential factor in Eq. (6) becomes insignificant. In this case, expression (6) is transformed into formula (7), which predicts only a shift in the peak in the orientation curve at $z \neq 0$.

CONCLUSIONS

Thus, we have analyzed possible results of experiments on diffraction of nonmonochromatic divergent X-ray beams in crystals with a lattice periodically strained by an acoustic wave using a simple kinematic approach in diffraction theory. The main result is formula (6) which makes it possible to take into account the yield of scattered quanta for all parameters of the incident beam and experimental conditions and to determine optimal conditions for controlling the characteristics of the scattered beam in the short-wavelength case [16], when the cross section of the incident X-ray beam considerably exceeds the wavelength of acoustic perturbation.

Our analysis proved a substantial dependence of the characteristics of scattered X-ray beams on the relation between the angular and frequency spreads in the primary beam as well as on the detuning of the average frequency of the primary beam from the Bragg frequency for a fixed reflecting plane and for the averaged scattering angle determined by the geometry of the setup.

The approach developed here makes it possible to indicate specific experimental conditions, in which the general conclusion of the kinematic theory concerning the absence of enhancement of the integrated yield of scattered radiation due to modulation of the crystal lattice by an acoustic wave is violated. It is shown that the observed effect of enhancement can be quite significant.

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