Hard Photon and Slow Neutron Scattering in Semiconductor Crystals under Conditions of Sound Instability

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It is shown that under sound-instability conditions, i.e., when phonon generation is produced by an external electric field such that the drift velocity of the carriers exceeds the phase velocity of the sound wave, the scattering of x-rays, γ-rays and slow neutrons is sharply altered: the amplitude of coherent scattering in this case falls sharply, while the incoherent (i.e., diffuse) peak increases sharply. An explicit expression is obtained for the Debye-Waller factor in a crystal under the conditions of sound instability and it is shown that due to the production of phonons it becomes sharply anisotropic. Attention is drawn to the possibility of modulating the intensity of the scattered x- and γ-radiation with the aid of a time dependent process for phonon production in an external electric field.

It is well known that the thermal motion of atoms in crystals leads to the fact that the intensity of the diffraction maxima is decreased by the Debye-Waller factor and that, moreover, diffuse scattering occurs the maxima of which, generally speaking, do not coincide with the maxima of structural scattering.[1] On the other hand, in semiconductors and semimetals when the drift velocity of the carriers exceeds the phase velocity of the sound wave spontaneous generation of thermal phonons occurs,[2] and therefore, naturally, the picture of the diffraction of x-rays in this case will be sharply altered. From physical considerations it is immediately clear that the intensity of the diffraction maxima for the coherent scattering in this case can only be diminished, while the intensity of diffuse scattering will be relatively increased.

Experiment shows that the intensity of production of phonons in semiconductors can exceed the thermal background by four-five orders of magnitude, and therefore one should expect that the changes in the picture of scattering will be quite considerable. The characteristic time for the establishment of the local value of the phonon density or, in other words, the characteristic time for the growth of the phonon flux is of the order of magnitude of the transit time for a phonon across the sample and for a crystal of dimensions of 10^{-1} cm will amount to 10^{-6}–10^{-7} sec. The latter statement means that with such a frequency one can produce a modulation in the intensity of the x- and γ-radiation. Moreover, by controlling the characteristic frequency of phonon generation (for example, by means of a change in the concentration of the carriers in a zone), as will be shown below, one can achieve a deflection of the x- and γ-rays by a certain angle which can attain a value of several minutes. Here we have in mind the spatial deflection of the diffuse scattering peak, the intensity of which under thermodynamically equilibrium conditions is usually very small. But, in the nonequilibrium case, when the number of phonons grows sharply the diffuse scattering is increased and, in principle, can turn out to be of the same order of magnitude as the structural scattering in an equilibrium crystal. Moreover, due to the spatial dependence of the number of phonons in a crystal the intensity of scattering at the maximum of the diffuse peak will be proportional no longer to the first power of the scattering volume, as in the equilibrium case, but to a higher power of this volume. This means that the relative halfwidth of the diffuse peak is diminished in the nonequilibrium case. Experimental observation of the scattering anomalies indicated above can yield very valuable information concerning a number of nonequilibrium characteristics of semiconductors under the conditions of phonon generation, and in particular to determine the spatial, angular and spectral distributions of the phonons being produced. At the same time, in contrast to the optical methods based on the scattering of light from a laser,[3] the x-ray methods have a greater resolving power and are also applicable to optically nontransparent crystals.

From the preceding it follows that an investigation of the problem of the scattering of x- and γ-radiation in a nonequilibrium crystal, when acoustical (or optical) lattice oscillations are excited by means of some mechanism, is of definite interest.

1. STRUCTURAL SCATTERING. THE DEBYE-WALLER FACTOR UNDER CONDITIONS OF SOUND INSTABILITY

The intensity of the diffraction maxima in the scattering of the types of radiation indicated above diminishes with increasing temperature, and this is described by the Debye-Waller factor e^{-2W}, where $w \propto \overline{u^2}$ which is the mean squared displacement of the atom of the lattice in a direction perpendicular to the mirror plane. We now obtain the explicit form of the Debye-Waller factor in a crystal under conditions of sound instability. We shall carry out our discussion on the example of the scattering of x-rays, but the same results are also obtained for the scattering of γ-rays and of slow neutrons.

We consider the case of a monochromatic plane wave. Then the effective scattering cross section σ, defined as the ratio of the intensity of radiation diffracted into the solid angle dΩ to the density of energy flux in the incident unpolarized wave, will be equal, as is well known, to[1]

$$\frac{d\sigma}{d\Omega}=\frac{1}{2}\left(\frac{e^2}{mc^2}\right)^2(1+\cos^2\vartheta)\left|\int dV\,n(\mathbf{r})e^{i(\mathbf{k}-\mathbf{k}')\mathbf{r}}\right|^2. \quad (1)$$

Here ϑ is the angle between the directions of incidence and scattering, i.e., between the propagation vectors $\mathbf{k}$ and $\mathbf{k}'$, $n(\mathbf{r})$ is the microscopic value of the electron density, V is the volume. The electron density, just as any other function in a crystal, can be represented in the form

$$n(\mathbf{r})=\sum_{\mathbf{b}} n_{\mathbf{b}} e^{2\pi i \mathbf{b}\mathbf{r}}, \tag{2}$$

where the summation is taken over all possible values of the inverse lattice vector $\mathbf{b}$. The variation in the microscopic electron density at each point of the crystal can be regarded as the result of a simple shift of the lattice by an amount equal to the local value of the displacement vector $\mathbf{u}(\mathbf{r}, t)$ which arises due to phonon generation. Thus, in the presence of sound waves we have

$$n(\mathbf{r})=n(\mathbf{r}+\mathbf{u}(\mathbf{r}, t)). \tag{3}$$

The scattering cross section in the form (1) describes both the coherent diffraction scattering and the diffuse scattering. In order to separate from the beginning both these types of scattering we shall represent the integral over the volume appearing in (1) in the form

$$I \equiv I_1+I_2 \equiv \langle I\rangle+(I-\langle I\rangle), \tag{4}$$

where the angular brackets $\langle\ \rangle$ denote averaging over an ensemble of systems. Now substituting (2) into (1), taking (3) into account and averaging, we see that the term proportional to $I_1 = \langle I\rangle$, will describe the coherent part of the scattering and the term proportional to $I_2 = I - \langle I\rangle$ will describe the diffuse part of the scattering.[1)] Further, it is not difficult to obtain expressions describing the effective cross sections for diffuse and coherent scattering:

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{diff}}=2\pi^2\left(\frac{e^2}{mc^2}\right)^2(1+\cos^2\vartheta)\sum_{\mathbf{b},\alpha}(\mathbf{b}\mathbf{e}^\alpha)\,|n_{\mathbf{b}}|^2\cdot$$
$$\cdot\int dV\,|u^\alpha(\mathbf{q}=\mathbf{k}-\mathbf{k}'+2\pi\mathbf{b}, \mathbf{r})\,|^2, \tag{5}$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}}=\frac{1}{2}\left(\frac{e^2}{mc^2}\right)^2(1+\cos^2\vartheta)\,\Big|\int dV\sum_{\mathbf{b}} n_{\mathbf{b}}\cdot$$
$$\times\exp\{2\pi i\mathbf{b}\mathbf{r}+i\mathbf{k}\mathbf{r}-i\mathbf{k}'\mathbf{r}-w(\mathbf{r})\}\Big|^2; \tag{6}$$

where

$$w(\mathbf{r})=2\pi^2\sum_\alpha(\mathbf{b}\mathbf{e}^\alpha)^2\int d^3\mathbf{q}\,|u^\alpha(\mathbf{q}, \mathbf{r})\,|^2 \tag{7}$$

is the term describing the decrease in the scattering cross section due to the oscillations of the atoms; $\mathbf{e}^\alpha$ is the unit polarization vector for the sound waves, α is the polarization index; $u^\alpha(\mathbf{q}, \mathbf{r})$ is the Fourier amplitude for elastic displacement, and its dependence on the coordinate $\mathbf{r}$ emphasizes the fact that under conditions of sound instability the intensity of phonon generation increases along the direction of the supersonic electron flux. In the course of calculating the diffuse scattering cross section the integration over the volume has already been carried out and this led to the appearance of the condition $\mathbf{q} = \mathbf{k} - \mathbf{k}' + 2\pi\mathbf{b}$ which determines the spatial direction of diffuse scattering. Under conditions of thermodynamic equilibrium, i.e., in the absence of electron drift, w does not depend on the coordinates and then e^{-2W} (after averaging over the Planck distribution) reduces to the well known Debye-Waller factor.

We first consider the change in the structure scattering peak near some maximum when phonon generation occurs. Let the Laue condition be satisfied for the given inverse lattice vector $\mathbf{b}$, i.e.,

$$\mathbf{k}-\mathbf{k}'-2\pi\mathbf{b}=0. \tag{8}$$

Then the scattering cross section at the maximum will evidently be given by

$$\left(\frac{d\sigma}{d\Omega}\right)^{max}_{\text{coh}}=\frac{1}{2}\left(\frac{e^2}{mc^2}\right)^2(1+\cos^2\vartheta)\,|n_{\mathbf{b}}|^2\left\{\int dV\,e^{-w(\mathbf{r})}\right\}^2. \tag{9}$$

If w does not depend on $\mathbf{r}$, then the scattering cross section at the maximum turns out to be proportional to V^2.

We now find the scattering cross section at the maximum under conditions of phonon generation. For this it is necessary to calculate explicitly the average value of the square of the amplitude of elastic displacement. We shall consider the case of a piezosemiconducting crystal, where the interaction of electrons with phonons occurs through the piezoelectric field. (For crystals with a deformation interaction analogous phenomena are also possible.[2]) For the determination of $\langle|u^\alpha(\mathbf{q}, \mathbf{r})|^2\rangle$ one must consider the kinetic equation for phonons in a piezosemiconductor (cf., [2]):

$$\left[\mathbf{v}_g^\alpha\frac{\partial}{\partial\mathbf{r}}+2\gamma^\alpha(q)\right]\langle|u^\alpha(\mathbf{q}, \mathbf{r})\,|^2\rangle$$
$$=\frac{2}{(2\pi)^3}\frac{1}{\rho q^2 v_\alpha^2}\left(T_e\frac{\Gamma^\alpha(\mathbf{q})}{1-\mathbf{q}\mathbf{v}_d/qv_\alpha}+T_l\frac{\mu_\alpha q^2}{2\rho}\right), \tag{10}$$

where $\mathbf{v}_g^\alpha$ is the vector for the group velocity of acoustic waves of polarization α, v_α is the phase velocity (below we shall not take into account the difference between the phase and the group velocity of the waves), ρ is the crystal density, T_e and T_l are respectively the temperatures of the electron gas and of the lattice, $\mathbf{v}_d$ is the electron drift velocity in an external electric field, $\mathbf{v}_d = \mu\mathbf{E}_d$, where μ is the electron mobility, $\mathbf{E}_d$ is the external electric field, $\gamma^\alpha(\mathbf{q})$ is the increment in the generation of acoustic waves of polarization α, $\Gamma^\alpha(\mathbf{q})$ is its electron part, μ_α is the viscosity responsible for the nonelectronic mechanism for the absorption of phonons in the crystal, where

$$\gamma^\alpha(\mathbf{q})=\frac{\mu_\alpha q^2}{2\rho}+\Gamma^\alpha(\mathbf{q}),$$
$$\Gamma^\alpha(\mathbf{q})=\frac{2\pi(\beta_{\mathbf{q},\mathbf{q}p}e_p{}^\alpha)^2}{\rho v_\alpha{}^2\varepsilon_0}\,q\,\mathrm{Im}\left(\frac{\varepsilon_0}{\varepsilon_\parallel(\omega, \mathbf{q})}\right)_{\omega=qv_\alpha}. \tag{11}$$

Here $\beta_{i,kl}$ is the tensor for the piezomoduli of the crystal "with respect to the deformation," ϵ_0 is the dielectric permittivity of the lattice, $\epsilon_\parallel(\omega, \mathbf{q})$ is the longitudinal permittivity of the medium:

$$\varepsilon_\parallel(\omega, \mathbf{q})=\varepsilon_0+\frac{4\pi\sigma_0}{i\omega}\left(1-\frac{\mathbf{q}\mathbf{v}_d}{\omega}+\frac{iq^2\mu T_e}{e\omega}\right)^{-1} \tag{12}$$

where $\sigma_0 = en_0\mu$ is the longitudinal d.c. conductivity of the crystal. From formula (12) it follows that in the case when the electron drift velocity is greater than the phase velocity of the sound waves, the imaginary part of the dielectric permittivity of the medium, i.e., the conductivity, becomes negative, and in the crystal generation of sound occurs, instead of electronic absorption of sound if, of course, $\gamma^\alpha(\mathbf{q}) < 0$. When $\gamma^\alpha(\mathbf{q}) < 0$, then, as follows immediately from the kinetic equation for the phonons (10), the average value of the square of the am-

[1]It is necessary to note that here in the course of averaging only pair correlations are taken into account, while correlators of the type <uuu> and of still higher order are neglected.

plitude for elastic displacement will grow in spece, so that the solution of equation (10) under zero boundary conditions has the form

$$\langle |u^\alpha(\mathbf{q}, \mathbf{r})|^2\rangle = \frac{1}{(2\pi)^3 \rho v_\alpha^2 q^2}\left[T_e \frac{\Gamma^\alpha(\mathbf{q})}{1-\beta^\alpha(\mathbf{q})} + T_p \frac{\mu_\alpha q^2}{2\rho}\right] \frac{1-\exp[-Q^\alpha(\mathbf{q},\theta)x]}{\gamma^\alpha(q)}, \quad Q^\alpha(\mathbf{q},\theta) = 2\gamma^\alpha(\mathbf{q})/v_\alpha \cos\theta \tag{13}$$

(ϑ is the angle between the electric field vector $\mathbf{E}_d$ and the propagation vector $\mathbf{q}$).

In deriving (13) it was assumed that the electron drift occurs along the x direction; phonon generation also occurs in the same direction and $\beta^\alpha(\mathbf{q}) = \mathbf{q}\mathbf{v}_d/qv_\alpha$.[2)] If the electric field is absent, then from (13) follows the well-known classical value

$$\langle |u^\alpha(\mathbf{q})|^2\rangle = T/(2\pi)^3 \rho v_\alpha^2 q^2. \tag{14}$$

If now one substitutes (14) into (7) then one obtains immediately the well-known expression for the Debye-Waller factor in which, however, integration over the modulus of the propagation vector q should be carried out from 0 to q_{max}, where q_{max} (just as in the case of the Debye theory of specific heat) is determined from the condition of equality of the total number of oscillations to the number of degrees of freedom in the crystal. Generation of acoustic phonons occurs only within the Cerenkov cone, and therefore integration over the angle θ (in spherical coordinates) in formula (7) can be conveniently separated into two intervals: $0 \le \theta \le \theta_0$ and $\theta_0 \le \theta \le \pi$, where θ_0 is the angle for which $\gamma^\alpha(\mathbf{q}) = 0$. Then, it is evident, that in the angular interval $\theta_0 \le \theta \le \pi$ no phonon generation occurs, and if we do not take into account the insignificant change in the phonon distribution factor due to the effect of the phonon drag by the subsonic electron flux (cf., [4]), then we can assume that for these angles the Debye-Waller factor is determined by the thermodynamic equilibrium value (14). Taking this circumstance into account formula (7) can be written in the form

$$w(\mathbf{r}) = \tfrac{1}{2} w_T (1+\cos\theta_0) + w_{ne}(\mathbf{r}), \tag{15}$$

where $w_{ne}(\mathbf{r})$ is the nonequilibrium increment due to phonon generation which has the form

$$w_{ne}(\mathbf{r}) = \frac{1}{2}\sum_\alpha (2\pi \mathbf{b}\mathbf{e}^\alpha)^2 \int_0^{q_{max}} dq \int_0^{2\pi} d\varphi \int_0^{\theta_0} d\theta \sin\theta \times \frac{1}{(2\pi)^3}\frac{1}{\rho v_\alpha^2}\left\{T_e \frac{\Gamma^\alpha(\mathbf{q})}{1-\beta^\alpha(\mathbf{q})} + T_p \frac{\mu_\alpha q^2}{2\rho}\right\} \frac{1-\exp[-Q^\alpha(\mathbf{q},\theta)x]}{\gamma^\alpha(\mathbf{q})}. \tag{16}$$

Under the conditions of generation it is possible that $|Q^\alpha(\mathbf{q}, \theta)| > 1$ and then scattering by nonequilibrium phonons will be the decisive factor in the diminution of the intensity at the peak.

We make an estimate of $w_{ne}(\mathbf{r})$. If we do not take into account the difference in the angular dependence for equilibrium and nonequilibrium phonons (it is evident that this difference cannot essentially alter the ratio of these quantities), then for purposes of making an estimate one can assume that

$$w_{ne} \sim w_T \exp\{-Q^\alpha(\tilde{\mathbf{q}}, \bar{\theta})x\}, \tag{17}$$

where $\tilde{\mathbf{q}}$ is the characteristic propagation vector for the phonons being generated, while $\bar{\theta}$ is a certain angle. In piezosemiconducting crystals of the type of CdS, CdSe and ZnO the spatial increments in the phonon generation can attain values of the order of 10^2 cm^{-1}, i.e., one can assume that by a suitable choice of the parameters for these crystals it is not difficult to obtain conditions under which $Q^\alpha(\tilde{\mathbf{q}}, \bar{\theta}) \approx 10^2$ cm^{-1}. (Such values have already been repeatedly attained experimentally, cf. the review article[2]). From here follows the estimate that $w_{ne} \sim w_T e^{100x}$, where x [cm] is the path length traversed by a phonon in the crystal in the direction of the electric field. If the dimensions of the crystal are of the order of 1 cm, then it is clear that the intensity of the radiation scattered at the peak must practically completely disappear when phonon generation begins.

We now estimate the characteristic time during which the nonequilibrium number of photons grows in a crystal of dimensions L. It can be shown (cf. the review article [2]) that the phonon density at a given point x attains a stationary state during a time equal to x/v_α. Therefore, if we consider a sufficiently thin crystal, say, of dimensions $L \approx 0.1$ cm, then this time will be 5×10^{-7} sec for transverse waves and 2×10^{-7} sec for longitudinal waves. (Here we have in mind a crystal of the type of CdS, for which $v_\parallel = 4.8 \times 10^5$ cm/sec, $v_\perp = 2 \times 10^5$ cm/sec.) Thus, with the same characteristic times one can bring about modulation of the intensity of x- and γ-radiation in an individual peak of structure scattering. The intensity of scattering of thermal neutrons also diminishes when lattice oscillations are present, and this decrease is also described by the Debye-Waller factor, where only in place of k and k′ the neutron propagation vectors will occur. Therefore, as in the case of x-ray radiation, modulation of the intensity of scattered slow neutrons is possible.

2. DIFFUSE SCATTERING

We now consider the behavior of the diffuse peak in the scattering under conditions of phonon generation. Substituting into formula (5) the value of $\langle |u^\alpha(\mathbf{q}, \mathbf{r})|^2\rangle$ from (13) and, just as above, considering some one fixed value of the inverse lattice vector, we obtain for the maximum

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{diff}} = \frac{S}{4\pi}\left(\frac{e^2}{mc^2}\right)^2 (1+\cos^2\vartheta) \sum_\alpha |n_b|^2 (\mathbf{b}\mathbf{e}^\alpha)^2 \times \frac{1}{\rho q^2 v_\alpha^2}\left[T_e \frac{\Gamma^\alpha(\mathbf{q})}{1-\beta^\alpha(\mathbf{q})} + T_p \frac{\mu^\alpha q^2}{2\rho}\right] \times \frac{L_x + [\exp\{-Q^\alpha(\mathbf{q},\theta)x\} - 1]/Q^\alpha(\mathbf{q},\theta)}{\gamma^\alpha(\mathbf{q})}, \tag{18}$$

where $S = \int dy\,dz$ is the area, while L_x is the dimension of the sample in the direction of the x axis. In formula (18) the propagation vector $\mathbf{q}$ satisfies the condition

$$\mathbf{q} = \mathbf{k} - \mathbf{k}' + 2\pi\mathbf{b}. \tag{19}$$

From expression (18) it can be seen that the intensity of the diffuse peak under conditions of phonon generation has increased by a factor of approximately $|\gamma^\alpha(\mathbf{q})|^{-1} \times \exp\{|2\gamma^\alpha(\mathbf{q})| L_x/v_\alpha\}$, i.e., very considerably. The

[2)]Here zero-point oscillations are not taken into account, and it is assumed that the temperature of the electrons and of the lattice is not very low, so that $\hbar\omega < T$, where ω is the characteristic frequency of the generated phonons.

spectrum of the generated acoustic phonons is quite narrow, therefore in addition to an increase in the amplitude of the peak also a narrowing of the diffuse scattering line must take place due to the fact that phonon generation occurs only inside the Cerenkov cone. But the distribution of the intensity of phonon generation inside the Cerenkov cone for a piezosemiconducting crystal is a very complicated function of the mutual orientation of the crystallographic directions and the direction of the external electric field, and therefore the behavior of the halfwidth of the diffuse peak as a result of variation of some parameters of the system which affect phonon generation can be very complicated.

The spectrum of the generated acoustic phonons and, in particular, their propagation vector, depend on a number of electronic characteristics of the crystal: the density of the carriers, the electric field, the temperature etc. (cf., [2]). Varying in the course of an experiment any one of these parameters one can realize a spatial displacement of the diffuse scattering peak. The maximum "angle of refraction" that can be obtained in the case of scattering by acoustic phonons will evidently be given by

$$\Delta\varphi \sim |q/k| \ll 1. \tag{20}$$

If the frequency of generation of acoustic phonons is $\omega \approx 10^{11}\ \mathrm{sec}^{-1}$ (which can be quite readily obtained in crystals of the type of CdS), then for an x-ray quantum of energy $E \approx 14$ keV ($k \approx 7.4 \times 10^8\ \mathrm{cm}^{-1}$) this angle can attain a value of $2'-3'$. Thus, by changing the spectrum of the generated acoustic oscillations, one can, in principle, realize refraction of x-rays.

We note that here we have restricted ourselves to an investigation of the picture of scattering due to the generation of acoustic phonons. On the other hand at the present time a number of mechanisms is known with the aid of which one can realize a sufficiently strong generation of optical phonons. If generation of optical phonons is achieved with the aid of an electron beam, then the necessary conditions for the generation is $v_e > \omega_0/q$, where v_e is the velocity of the electrons in the beam. Since for optical phonons the frequency ω_0 is practically constant, then by changing the velocity v_e one can achieve generation of optical phonons with different propagation vectors $q \sim \omega_0/v_e$. The latter condition means that the angle of deflection of the diffuse peak will vary as a function of v_e. The propagation vector for an optical phonon is $q \approx 10^8\ \mathrm{cm}^{-1}$, and as can be seen from (20), the "angle of refraction" for diffuse scattering by optical phonons can attain a value of several degrees. This problem shall be discussed separately.

In a crystal of limited size phonon generation occurs over a discrete set of frequencies corresponding to the eigenoscillations of an acoustic resonator. Therefore in the inelastic scattering of radiation a series of new lines —satellites will appear which correspond to processes of absorption and emission of phonons.

The changes in the picture of scattering of x-rays in piezosemiconducting crystals indicated above under the conditions of sound instability have been observed experimentally by Lemke, Müller and Schnürer.[5] The experiment was carried out using a film of cadmium sulphide. In the absence of phonon generation a narrow line of coherent scattering was observed at the Bragg angle, but under the conditions of phonon generations the intensity of coherent scattering sharply decreased and two diffuse peaks appeared at the sides of this line. The angular scatter between the coherent line and the diffuse peak experimentally amounted to approximately 20′, which for a crystal of the CdS type corresponds to generation of acoustic phonons at a frequency of ~500 MHz.[3)]

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[3)]We note that the theoretical discussion of the problem of the scattering of x-rays in a crystal under conditions of sound instability undertaken in connection with this experiment in [5] does not take into account the change in the diffraction picture as a result of phonon generation, assuming at the same time that the interaction between an x-quantum and a phonon occurs in the same manner as the interaction between a light photon and a phonon in a continuous medium. Such lack of taking into account the diffraction properties of the medium leads to the fact that in the theory the Debye-Waller factor does not arise in general, while the shape of the line of the scattered radiation turns out to be quite sensitive to the spectral composition of the emitted phonons.

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90