which are then excited lead to an exchange between trapped and free electrons until $|n_e^{tr} - n_{eo}^{tr}|$ is decreased to a value satisfying condition (5.25). The characteristic time for this process is $\lesssim \omega_{pi}^{-1}$. Finally, we note that if we use model Boltzmann distribution functions for the trapped particles with an arbitrary temperature and neglect the contribution from the resonance ions, Eqs. (5.24), (5.19) lead to expressions which are equivalent to those obtained by Schamel.[5]

[1)]In the Ott-Sudan Eq. (4.13) the electrons, on the other hand, give the main contribution, at least when $T_e/T_i \gtrsim 4$.
[2)]If we used (5.30) to evaluate $A$ and $B$, which is equivalent to taking only terms $\propto \nu^2$ into account in (5.13) and (5.14), we would find $A = B$, i.e., $\Psi_- = 0$.
[3)]It makes no sense to take higher-order terms into account as they were dropped when we derived the perturbed KdV Eq. (3.10).


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# Contribution to the theory of nonlinear photoelasticity of solids


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Nonlinear modulation of the dielectric constant of a crystal by a sound wave is considered theoretically. An expression quadratic in the sound-wave amplitude is obtained for the amplitude of the change of the dielectric constant of the crystal; this expression is analyzed for various optical frequency bands. It is shown that the effect of nonlinear modulation of the dielectric constant of a crystal by a sound wave is particularly pronounced in the resonant case $\hbar\omega \approx E_g$ for dielectrics, or else at $\hbar\omega \approx E_g + F$ for degenerate semiconductors, when relatively low sound power is needed for the onset of nonlinear effects (here $\omega$ is the frequency of the light wave, $E_g$ is the width of the forbidden band of the crystal, $F = m_c\epsilon_F/\mu$, where $m_c$, $\mu$, and $\epsilon_F$ are respectively the effective mass of the conduction electrons, the reduced effective mass of the conduction and valence electrons, and the Fermi energy of the conduction electrons).



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The diffraction of light by sound (the acousto-optical (AO) interaction) has by now been the subject of a large number of studies, owing to the extensive practical use of this phenomenon in physical research and in modern technology (see the reviews[1,2]). In all the theoretical studies of the AO interaction it is assumed that one of its basic mechanisms is the change of the dielectric constant of the crystal upon propagation of the sound wave. It is customarily proposed that in view of the smallness of the relative deformation of the crystal by the passage of the sound, its dielectric constant $\varepsilon_{ik}$ in the presence of the sound wave can be expressed in the following form (see, e.g., Refs. 1–3)

$$\varepsilon_{ik} = \varepsilon_{ik}^0 + \Delta^1\varepsilon_{ik} \equiv \varepsilon_{ik}^0 - \varepsilon_{il}^0\varepsilon_{km}^0 p_{lmnj} u_{nj}, \quad (1)$$

where $\varepsilon_{ik}^0$ is the dielectric constant of the crystal in the absence of sound, $u_{nj}$ is the crystal strain tensor in the sound wave, and $p_{lmnj}$ is the crystal photoelasticity tensor (summation over repeated indices is understood from now on). Terms of higher order in $u_{ik}$ are as a rule neglected in (1).

It should be noted, however, that in some cases a sound wave of even relatively small power is capable producing in the dielectric constant of the crystal a modulation that is essentially nonlinear in the parameter $u_{ik}$. In fact, in the simplest case at $\hbar\omega < E_g$ there is no absorption of light in the crystal (neglecting the absorption "tail"). On the other hand, in the presence of a sound wave modulation of the quantity $E_g$ sets in, and a situation becomes possible wherein the relation $\hbar\omega > E_g - \Delta E_g$ is satisfied in a region of a decreased

gap width ($\Delta E_g$ is the amplitude of the modulation of the gap width), i.e., in such a region the light will be absorbed. It is clear that under these conditions the sound essentially modulates the absorption coefficient of the light nonlinearly (the same situation holds also for the real part of $\varepsilon_{ik}$). Assume now that we cannot confine ourselves in the expansion of $\varepsilon_{ik}$ to terms linear in $u_{ik}$, and it is necessary generally speaking to use the entire series.[1)] Nonlinear modulation of $\varepsilon_{ik}$ by sound will lead, for example, to a situation wherein a noticeable AO interaction over the length of the crystal becomes possible in the case of a monochromatic sound wave in the Bragg diffraction regime at different light incidence angles, and even in the case of low efficiency the diffraction will depend essentially nonlinearly on the sound power.

In the present paper, under the assumption that the main contribution to the polarizability of the crystal is made by the electron polarizability, we consider the terms quadratic in $u_{ik}$ of the expansion of $\varepsilon_{ik}$ in the parameter $u_{ik}$. It will be shown that in some cases (in particular, in resonant diffraction of light by sound) these terms (as well as terms of higher order in $u_{ik}$) can become substantial also at relatively low sound power.

The presence of terms quadratic in $u_{ik}$ in $\varepsilon_{ik}$ (we shall designate the sum of the terms by $\Delta^2\varepsilon_{ik}$) leads to the appearance, in the term linear in $\Delta^2\varepsilon_{ik}$ approximation, of induced currents at frequencies $\omega\pm2\Omega$ ($\Omega$ is the frequency of the sound. This causes in turn the appearance of corresponding diffraction orders, whose intensity can become noticeable at sufficiently large length of the AO interaction if the spatial synchronism condition is satisfied between the zeroth order of diffraction and one of the ±1-st diffraction orders (with frequencies $\omega\pm2\Omega$) (see Ref. 2 on this subject). We shall henceforth pay principal attention to just this part of $\Delta^2\varepsilon_{ik}$ [which we designate $\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)$], since it causes the appearance of these diffraction orders.[2)]

To calculate the corresponding quantities $\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)$, we used the following connection between the density of the stimulated induced current $\mathbf{j}^{\omega\pm2\Omega}$ at the frequencies $\omega\pm2\Omega$ and the electric field $\mathbf{E}$ of the electromagnetic wave at the frequency $\omega$:

$$j_i^{\omega\pm2\Omega}(\mathbf{r},t)=-\frac{i(\omega\pm2\Omega)}{4\pi}\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)E_k(\omega); \tag{2}$$

here

$$\mathbf{j}^{\omega\pm2\Omega}(\mathbf{r},t)=\mathrm{Tr}\left(\hat\rho_1(\omega\pm2\Omega)\hat{\mathbf{j}}_1+\hat\rho_2(\pm2\Omega)\hat{\mathbf{j}}_2\right),$$

$$\hat{\mathbf{j}}_1=\frac{e}{2m}\left[\delta(\mathbf{r}-\mathbf{r}_e)\hat{\mathbf{p}}_e+\hat{\mathbf{p}}_e\delta(\mathbf{r}-\mathbf{r}_e)\right],$$

$$\hat{\mathbf{j}}_2=-\frac{e^2}{mc}\mathbf{A}(\mathbf{r}_e,t)\delta(\mathbf{r}-\mathbf{r}_e),$$

$\mathbf{A}(r_e,t)$ is the vector potential of the electromagnetic wave at the frequency $\omega$, and is assumed to be proportional to the exponential $\exp(-i\omega t)$; $\hat\rho_{1,2}$ are the terms of the single-particle electron density matrix and are proportional respectively to $\exp[-i(\omega+2\Omega)t]$ and $\exp[\pm2i\Omega t]$; $\hat{\mathbf{p}}_e=(\hbar/i)\partial/\partial\mathbf{r}_e$; $\delta(\mathbf{r}-\mathbf{r}_e)$ is a delta function; $\mathbf{r}_e$ is the spatial coordinate which enters in the wave function of the electron; $e$ and $m$ are the charge and mass of the free electron, and $c$ is the speed of light in vacuum.

To find the quantities $\hat\rho_{1,2}$ it is necessary to solve the equation for the density matrix $\hat\rho$, in the form

$$i\hbar\partial\hat\rho/\partial t=[\hat H_0+\hat V,\hat\rho]+\hat I_{\mathrm{col}}. \tag{3}$$

Here $\hat I_{\mathrm{col}}$ is the integral term of (3) and describes the collisions of the electrons with the scatterers; $\hat H_0$ is the Hamiltonian of the unperturbed system of electrons (in the absence of the electromagnetic wave and the sound wave), and $\hat V$ is the Hamiltonian of the perturbation, which can be written in our case in the form

$$\hat V=-\frac{e}{2mc}(\hat{\mathbf{p}}\hat{\mathbf{A}}+\hat{\mathbf{A}}\hat{\mathbf{p}})+\hat f e^{i\Omega t}+\hat f_1 e^{2i\Omega t}+\hat f^+e^{-i\Omega t}+\hat f_1^+e^{-2i\Omega t}. \tag{4}$$

In this approximation, the last four terms describe the electron-phonon interaction in the approximation linear in the amplitude of the sound wave, i.e., $\hat f=\hat f_{nj}\bar u_{nj}$, $\hat f_1=\hat f_{nj}\bar u^1_{nj}$, where $\bar u_{nj}$ and $\bar u^1_{nj}$ are the amplitudes of the strain tensor of the fundamental harmonic (with frequency $\Omega$) and of the harmonic and frequency $2\Omega$, which is produced as a result of the nonlinear propagation of the sound wave through the crystal. To simplify the calculation we confine ourselves henceforth to the simplest approximation of the $\hat I_{\mathrm{col}}$, assuming that it is possible to introduce a constant electron relaxation time $\tau$. In this case the expression for $\hat I_{\mathrm{col}}$ can be written in the form

$$\hat I_{\mathrm{col}}=-i\hbar(\hat\rho-\hat\rho_s)/\tau, \tag{5}$$

where $\hat\rho_s$ is the equilibrium density matrix of the electrons and corresponds to their local concentration (see e.g., Ref. 8).

Next, when solving (3), we use the method of successive approximations. To calculate the quantity $\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)$ it is necessary to find the terms $\hat\rho_2$ and $\hat\rho_1$ of respectively zeroth and first order in the electron-proton interaction operator and of second order in the electron-phonon interaction operator. Assuming that the electromagnetic wave is purely transverse (the corresponding generalization entails no difficulty), we obtain the following equation for the density matrix $\hat\rho_s$:

$$[\hat H_0+\hat\Phi+\hat\Phi^+,\hat\rho_s]=0, \tag{6}$$

where $\hat\Phi$ is a certain stationary operator, linear in $u_{ik}$ (without its time dependence) under the assumptions made here. We assume that the intraband matrix elements of the operator $\hat\Phi$ are constant in the electron momentum-space region of importance to the integration. We assume further that the collisions of the electrons with scatterers produce practically no change in the local concentration of the electrons in each of the energy bands. We then obtain the following solutions of Eq. (6) in first and second orders of perturbation theory in the operator $\hat\Phi$:

$$\langle\mathbf{p}|\hat\rho_s^1|\mathbf{p}\pm\mathbf{q}\rangle=\frac{f_0(\mathbf{p})-f_0(\mathbf{p}\pm\mathbf{q})}{E_{\mathbf{p}}-E_{\mathbf{p}\pm\mathbf{q}}}\langle\Phi(\pm\mathbf{q})\rangle, \tag{7}$$

$$\langle\mathbf{p}|\hat\rho_s^2|\mathbf{p}\pm2\mathbf{q}\rangle=\left(\frac{f_0(\mathbf{p})-f_0(\mathbf{p}\pm\mathbf{q})}{E_{\mathbf{p}}-E_{\mathbf{p}\pm\mathbf{q}}}-\frac{f_0(\mathbf{p}\pm\mathbf{q})-f_0(\mathbf{p}\pm2\mathbf{q})}{E_{\mathbf{p}\pm\mathbf{q}}-E_{\mathbf{p}\pm2\mathbf{q}}}\right)\frac{\langle\Phi(\pm\mathbf{q})\rangle^2}{E_{\mathbf{p}}-E_{\mathbf{p}\pm2\mathbf{q}}}. \tag{8}$$

Here

$$\langle\hat{\Phi}(+\mathbf{q})\rangle=\langle\hat{\Phi}(-\mathbf{q})\rangle^{*}=\langle\hat{f}\rangle L/(L_0+R_0);$$

$$L_0=\frac{1}{V}\sum_{\mathbf{p}}\frac{f_0(\mathbf{p}+\mathbf{q})-f_0(\mathbf{p})}{E_{\mathbf{p}}-E_{\mathbf{p}+\mathbf{q}}},\qquad L=\frac{1}{V}\sum_{\mathbf{p}}\frac{f_0(\mathbf{p}+\mathbf{q})-f_0(\mathbf{p})}{E_{\mathbf{p}}-E_{\mathbf{p}+\mathbf{q}}+\hbar\Omega-is};$$

$$R_0=\frac{is}{V}\sum_{\mathbf{p}}\frac{f_0(\mathbf{p}+\mathbf{q})-f_0(\mathbf{p})}{(E_{\mathbf{p}}-E_{\mathbf{p}+\mathbf{q}})(E_{\mathbf{p}}-E_{\mathbf{p}+\mathbf{q}}+\hbar\Omega-is)};$$

$\mathbf{p}$ is the wave vector of the electrons in the energy band, $E_p$ and $f_0(\mathbf{p})$ are the energy of the electrons with wave vector $\mathbf{p}$ and their distribution function in the absence of sound; $\mathbf{q}$ is the wave vector of the sound wave; $\langle\hat{f}\rangle$ is the matrix element of the operator $\hat{f}$ in the considered band; $s=\hbar/\tau$; $V$ is the volume of the crystal (the asterisk denotes the complex-conjugation operation). In the derivation of expressions (7) and (8) we neglected the intervand matrix elements of the operator $\hat{f}$.

It is seen from (7) and (8) that in the considered approximations in dielectrics, as well as in conducting crystals in the collisionless regime $ql\gg1$ (where $l$ is the characteristic mean free path of the electrons that make the substantial contribution to the considered effect) we have $\hat{\rho}_s\approx\hat{\rho}_0$, where $\hat{\rho}_0$ is the equilibrium density matrix in the absence of perturbation.

Using expressions (7) and (8) for the density matrix $\hat{\rho}_s$, we solve Eq. (3) by successive approximations and obtain an expression for $\hat{\rho}_{1,2}$, and then the induced current $j^{\omega\pm2\Omega}$, and finally, using relation (2), we derive an expression for the quantity $\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)$. We assume in the calculation that $\omega\tau\gg1$. The general expression for $\Delta^2\varepsilon_{ik}(\omega+2\Omega)$ is quite cumbersome, and we therefore write down here its simplest form, which is obtained when $\hat{\rho}_s\approx\hat{\rho}_0$:

$$\Delta^2\varepsilon_{ik}(\omega-2\Omega)=-\frac{4\pi e^2e^{i(2\Omega t-2\mathbf{qr})}}{m\omega(\omega-2\Omega)V}\sum_{1-4}\left\{\frac{[f_0(3)-f_0(4)]c_{ik}}{\Delta_1^{4}(-\Omega)\Delta_1^{3}(-2\Omega)\Delta_2^{3}(\omega'')}\right.$$
$$+\frac{[f_0(1)-f_0(4)]c_{ik}}{\Delta_1^{4}(-\Omega)\Delta_1^{3}(-2\Omega)\Delta_2^{3}(\omega'')}+\frac{[f_0(4)-f_0(3)]c_{ki}}{\Delta_1^{3}(-\Omega)\Delta_1^{3}(-2\Omega)\Delta_1^{4}(\omega'')}$$
$$+\frac{[f_0(4)-f_0(1)]c_{ki}}{\Delta_1^{4}(-\Omega)\Delta_1^{3}(-2\Omega)\Delta_1^{2}(\omega'')}+\frac{[f_0(2)-f_0(3)]c_{ki}}{\Delta_1^{2}(\omega')\Delta_1^{2}(\omega'')\Delta_2^{2}(\omega)}$$
$$+\frac{[f_0(1)-f_0(2)]c_{ik}}{\Delta_2^{1}(\omega)\Delta_2^{4}(\omega')\Delta_2^{3}(\omega'')}+\frac{[f_0(2)-f_0(1)]b_{ik}}{\Delta_2^{1}(\omega)\Delta_2^{4}(\omega')\Delta_3^{4}(\omega'')}$$
$$+\frac{[f_0(2)-f_0(1)]b_{ik}}{\Delta_3^{1}(\omega')\Delta_2^{4}(\omega)\Delta_3^{4}(\omega'')}+\frac{[f_0(1)-f_0(4)]c_{ik}}{\Delta_2^{4}(\omega')\Delta_1^{4}(-\Omega)\Delta_2^{3}(\omega'')}$$
$$+\frac{[f_0(4)-f_0(3)]c_{ki}}{\Delta_1^{3}(-\Omega)\Delta_1^{2}(\omega')\Delta_1^{2}(\omega'')}+\frac{[f_0(4)-f_0(1)]b_{ik}}{\Delta_2^{4}(\omega')\Delta_1^{4}(-\Omega)\Delta_3^{4}(\omega'')}$$
$$\left.+\frac{[f_0(2)-f_0(3)]b_{ik}}{\Delta_3^{2}(-\Omega)\Delta_3^{1}(\omega')\Delta_3^{4}(\omega'')}\right\}. \tag{9}$$

The numbers 1–4 denote here the electronic states of the unperturbed system, which are characterized in the crystal by the quasimomentum $\hbar\mathbf{p}$ and by the band index $n$;

$$\Delta_k^{i}(\omega)=E_i-E_k+\hbar\omega+is,\quad \omega'=\omega-\Omega,\quad \omega''=\omega-2\Omega,$$
$$c_{ik}=\langle1|\hat{f}|4\rangle\langle4|\hat{f}|3\rangle\langle3|e^{-i\mathbf{k}_1\mathbf{r}}\hat{p}_i|2\rangle\langle2|e^{i\mathbf{kr}}\hat{p}_k|1\rangle,$$
$$c_{ki}=\langle1|\hat{f}|4\rangle\langle4|\hat{f}|3\rangle\langle3|e^{i\mathbf{kr}}\hat{p}_k|2\rangle\langle2|e^{-i\mathbf{k}_1\mathbf{r}}\hat{p}_i|1\rangle,$$
$$b_{ik}=\langle1|\hat{f}|4\rangle\langle4|e^{-i\mathbf{k}_1\mathbf{r}}\hat{p}_i|3\rangle\langle3|\hat{f}|2\rangle\langle2|e^{i\mathbf{kr}}\hat{p}_k|1\rangle,$$

$\mathbf{k}$ is the wave vector of the electromagnetic wave in the medium, and $\mathbf{k}_1=\mathbf{k}-2\mathbf{q}$.

For the quantity $\Delta^2\varepsilon_{ik}(\omega+2\Omega)$ we obtain an analogous expression with the substitutions $\Omega,\ q\to-\Omega,\ -q$ and $\hat{f}\to\hat{f}^{*}$.

Neglecting the exponentials in the matrix elements (9) and changing from $\langle1|\hat{\mathbf{p}}|2\rangle$ to the metrix elements of the coordinate $\mathbf{r}$, we rewrite expression (9), after some algebraic transformations, and $s\langle|\Delta_k^{i}(-\Omega)|,\ |\Delta_k^{i}(\omega)$ in the following from (see also Ref. 9 on this subject):

$$\Delta^2\varepsilon_{ik}(\omega-2\Omega)=-\frac{4\pi e^2}{V}e^{i(2\Omega t-2\mathbf{qr})}$$
$$\times\sum_{1-4}\left\{\frac{[f_0(1)-f_0(4)]c_{ik}}{\Delta_1^{4}(-\Omega)\Delta_1^{3}(-2\Omega)\Delta_2^{1}(\omega)}+\frac{[f_0(3)-f_0(4)]c_{ik}}{\Delta_1^{3}(-\Omega)\Delta_1^{3}(-2\Omega)\Delta_2^{3}(\omega'')}\right.$$
$$+\frac{[f_0(4)-f_0(2)]c_{ik}}{\Delta_2^{1}(\omega)\Delta_2^{4}(\omega')\Delta_2^{3}(\omega'')}-\frac{f_0(1)b_{ik}}{\Delta_2^{1}(\omega)\Delta_3^{1}(\omega')\Delta_1^{4}(-\Omega)}$$
$$\left.+\frac{f_0(2)b_{ik}}{\Delta_2^{1}(\omega)\Delta_2^{4}(\omega')\Delta_3^{2}(-\Omega)}+X\right\}. \tag{10}$$

Here $X$ denotes the preceding expression with the following changes:

$$p_k\rightleftarrows p_i,\quad \omega+is\rightleftarrows-(\omega''+is),\quad \omega'+is\rightleftarrows-(\omega'+is)$$

and the quantities $c_{ik}$ and $b_{ik}$ contain in place of the operator $\hat{\mathbf{p}}$ the operator $\hat{\mathbf{r}}$, while the corresponding exponentials are absent. Expression (10) is particularly convenient in estimates of the quantity $\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)$ for the case of dielectrics at $\hbar\omega\ll E_g$.

The quantity of $\Delta^2\varepsilon_{ik}(\omega\mp2\Omega)$, given by expression (9), is due to an induced nonlinear action of the sound wave of the fundamental harmonic on the electron system, as a result of which an electromagnetic wave of frequency $\omega$, when propagating through the crystal, induces even in the approximation linear $\Delta^2\varepsilon_{ik}(\omega\mp2\Omega)$ a stimulated current $j^{\omega\mp2\Omega}$ at the frequency $\omega\mp2\Omega$, which in final analysis (when the appropriate conditions are satisfied) can lead to the appearance of noticeable diffraction orders at the frequencies $\omega\mp2\Omega$. In addition to the indicated terms of expression (9), a contribution to $j^{\omega\mp2\Omega}$ is made also by terms due to the presence of the second harmonic of the sound wave at the frequency $2\Omega$ [we shall denote their sum by $\Delta^1\varepsilon_{ik}(u_{nj}^1)$]. We shall not present here the expression for $\Delta^1\varepsilon_{ik}(u_{nj}^1)$, since it is in fact the ordinary expression, linear in the parameter $u_{nj}^1$, for the amplitude of the change of the quantity $\varepsilon_{ik}$ under the influence of the second harmonic of the sound wave. We use in the estimates instead the already available results on $\Delta^1\varepsilon_{ik}$ (see Refs. 2, 4–7, 9, 10). We note here that in the case when the higher harmonics in the sound wave are small, the quantity $|\Delta^1\varepsilon_{ik}(u_{nj}^1)|$ is much less than $|\Delta^1\varepsilon_{ik}(u_{nj})|$, which is obtained in the approximation linear in $u_{nj}$ [see (1)].[3)]

We consider next some particular cases of expression (9) for different frequency bands of the electromagnetic wave.

1. The case of low frequencies $\omega$, when $\hbar\omega\ll E_g$ ($E_g$ is the width of the forbidden band of the crystal). In this case the expression for

$$\Delta\varepsilon_{ik}=\Delta^2\varepsilon_{ik}(\omega-2\Omega)+\Delta^1\varepsilon_{ik}(u_{nj}^1)$$

can be written in the following form (accurate to terms of order $\Omega/\omega$):

$$\Delta\varepsilon_{ik}=-\left(\frac{4\pi e^2}{\omega^2}n^{2\Omega}(m^{*})_{ik}^{-1}+Q_{iknj}\bar{u}_{nj}^{1}+Q_{iknjlm}\bar{u}_{nj}\bar{u}_{lm}\right)e^{i(2\Omega t-2\mathbf{qr})}, \tag{11}$$

where $(m^{*})_{ik}^{-1}$ is the tensor of the effective mass of the

free carriers, $n^{2\Omega}$ is the amplitude of the electron-concentration wave at the frequency $2\Omega$, due to the nonlinear response of the electron system to the perturbation produced by the fundamental harmonic of the sound wave and to the linear response to the perturbation produced by the second harmonic of the sound wave; the quantities $Q_{iknjlm}$ and $Q_{iknj}$ are determined in the main by the electrons of the filled bands of the dielectric (with account taken of the filling of a certain number of levels in the conduction band and the possible presence of holes in the valence band of the semiconductor).

The main contribution of the first term of (11) to $\Delta\varepsilon_{ik}$ is due to the nonlinear modulation of the concentration of the free carriers by the sound in the crystal for the low-frequency collisionless case (i.e., at $\hbar\omega \ll E_g$ and $\omega\tau \gg 1$). For semiconductors, in the case when the sound power reaches a value such that $|\varphi| \sim \langle\varepsilon\rangle$ and concentration nonlinearity sets in (here $|\varphi$ is the energy of the electron in the field of the sound wave and $\langle\varepsilon\rangle$ is the average energy of the electron in the crystal), the value of $n^{2\Omega}$ becomes of the order of the equilibrium electron concentration $n_0$ (see, e.g., Refs. 12 and 13), and the contribution of the first term of (11) to $\Delta\varepsilon_{ik}$ can be substantial precisely at low frequencies $\omega$, in analogy to the situation for the quantity $\Delta^1\varepsilon_{ik}(u_{nj})$, where at low frequencies $\omega$ the contribution of the electron concentration wave $n^{\Omega}$ to $\Delta^1\varepsilon_{ik}(u_{nj})$ can become predominant (see, e.g., Ref. 2).[4)]

As for the second and third terms of (11), starting from Eq. (10) and from the expression for $Q_{iknj}$ (see Ref. 9) we can easily obtain at $\hbar\omega \ll E_g$ the following estimate for a dielectric:

$$|Q_{iknjlm}\bar{u}_{lm}| \sim |Q_{iknj}||\langle f_{lm}\rangle \bar{u}_{lm}/E_g|. \tag{12}$$

Thus, in order to obtain the value $\Delta_{ik} \sim \Delta^1\varepsilon_{ik}(u_{nj})$ in a dielectric at $\hbar\omega \ll E_g$ we must use a powerful sound wave that disturbs noticeably the electron system in the crystal when $\langle f_{lm}\rangle u_{lm} \sim E_g$, or else generates noticeable higher harmonics as a result, for example, of lattice nonlinearity in the course of sound propagation. At weaker sound waves, a substantial value of $\Delta\varepsilon_{ik}$ can be obtained in the present case for semiconductors on account of the first term of (11) under conditions of nonlinear modulation of the concentration of the free carriers by the sound wave, when the higher harmonics in the propagation of the sound wave can still be small.[13,14]

2. The resonant case, when $\hbar\omega \sim E_g$. It will be assumed here that the power of the sound wave is insufficient to generate higher harmonics, and then we certainly have

$$|\Delta^1\varepsilon_{ik}(u_{nj}')| \ll |\Delta^1\varepsilon_{ik}(u_{nj})|$$

and we shall consider henceforth the resonant properties of $\Delta\varepsilon_{ik}$, due to the terms $\Delta^2\varepsilon_{ik}(\omega \mp 2\Omega)$. One of the characteristic resonant terms $I_{ik}$, contained in (9), is of the form

$$I_{ik}^{r} = -\frac{4\pi e^2}{m^2\omega(\omega-2\Omega)}\frac{2}{(2\pi)^3}\int d^3\mathbf{p}\,\frac{f_0(E^c_{\mathbf{p}+\mathbf{q}}) - f_0(E^v_{\mathbf{p}+2\mathbf{q}-\mathbf{k}})}{E^c_{\mathbf{p}} - E^v_{\mathbf{p}-\mathbf{k}_1} - \hbar\omega'' - is} \times \frac{\langle c,\mathbf{p}|\hat{f}|c,\mathbf{p}+\mathbf{q}\rangle\langle c,\mathbf{p}+\mathbf{q}|\hat{f}|c,\mathbf{p}+2\mathbf{q}\rangle}{E^c_{\mathbf{p}+\mathbf{q}} - E^v_{\mathbf{p}-\mathbf{k}_1} - \hbar\omega' - is} \times \frac{\langle c,\mathbf{p}+2\mathbf{q}|e^{i\mathbf{k}r}\hat{p}_k|v,\mathbf{p}-\mathbf{k}_1\rangle\langle v,\mathbf{p}-\mathbf{k}_1|e^{-i\mathbf{k}_1 r}\hat{p}_i|c,\mathbf{p}\rangle}{E^c_{\mathbf{p}+2\mathbf{q}} - E^v_{\mathbf{p}-\mathbf{k}_1} - \hbar\omega - is}, \tag{13}$$

where $|c,\mathbf{p}\rangle$ and $|v,\mathbf{p}\rangle$ are the states of the electron with quasimomentum $\hbar\mathbf{p}$ in the conduction and valence bands, respectively.

In the case of a dielectric, the term under consideration is the principal resonant term at $\hbar\omega \approx \varepsilon_g$ (there are also analogous terms with matrix elements $\langle\hat{f}\rangle$ over the valence band), while for a degenerate semiconductor there are added resonant terms due, as will be shown later, to modulation of the Fermi energy of the free electrons by the sound wave. We shall not write out here all the resonant terms in explicit form [this can be easily done by using, for example, expression (9)], and present immediately the result of integration of these terms for the case of an $n$-type degenerate semiconductor, neglecting the presence of holes in the valence band, under the assumption that the matrix elements are constant in the electron momentum-space region that is essential for the integration, and also assuming the electron spectra in the bands to be parabolic and neglecting the wave vector of the light and the sound.

Under the foregoing assumptions, the expression for $\Delta^2\varepsilon_{ik}^{r}$, when only the resonant terms are retained in (9), takes the form

$$\Delta^2\varepsilon_{ik}^{r} = A_{ik}\left(\frac{\Lambda_c^2(m_c/\mu)^2(\Delta+F)+4(m_c/\mu)F\Lambda_c(\Lambda_v-\Lambda_c)}{2F^{1/2}(\Delta-F)^2} + (\Lambda_c-\Lambda_v)^2 f(\omega)\right), \tag{14}$$

where

$$A_{ik} = \left(\frac{2\mu}{\hbar^2}\right)^{1/2}\frac{e^2}{\pi m^2\omega^2}\langle c|\hat{p}_k|v\rangle\langle v|\hat{p}_i|c\rangle,$$
$$\Lambda_c = \langle c|\hat{f}|c\rangle, \quad \Lambda_v = \langle v|\hat{f}|v\rangle,$$
$$\Delta = \hbar\omega - E_g + is, \quad F = \hbar^2 p_F^2/2\mu$$

($p_F$ is the Fermi wave vector of the electrons);

$$f(\omega) = \frac{1}{(\hbar\Omega)^2}(\chi(\omega)+\chi(\omega'')-2\chi(\omega')),$$
$$\chi(\omega) = \Delta^{1/2}\left(\pi i + \ln\left|\frac{F^{1/2}+\Delta^{1/2}}{F^{1/2}-\Delta^{1/2}}\right| - i\,\mathrm{arctg}\,\frac{2F^{1/2}\,\mathrm{Im}\,\Delta^{1/2}}{|\Delta|-F}\right)$$

[in the expression for $\chi(\omega)$, the function $f = \tan^{-1}x$ is defined in the upper half-plane]. In the derivation of (14) it was assumed that

$$|\hbar\omega - E_g|, \hbar\Omega, F \ll \hbar^2 k_m^2/2\mu,$$

where $k_m$ is the wave vector of the band boundary, and it was also assumed that $ql \gg 1$ or[5)] $ql \ll 1$ ($l = \hbar p_F\tau/m_c$),

$$v_s = \Omega/q \ll \hbar p_F/m_c, \quad p_F \gg q, k.$$

In the case of dielectrics, only the last term, in which $\chi(\omega) = \pi i\Delta^{1/2}$, is left in expression (14).

It should be noted that, under the assumptions made, expression (14) is valid also in the immediate vicinity of the resonances for the case of dielectrics (i.e., as $|\Delta| \to 0$), provided the following condition is satisfied

$$\hbar^2 q_0^2/2\mu \ll \max(s, \hbar\Omega)$$

[here $q_0 = \max(q,k)$], and in the case of degenerate semiconductors (i.e., as $|\Delta| \to F$) it is valid if the following conditions are satisfied

$$\hbar^2 p_F q_0/\mu,\ m_c T/\mu \ll s$$

($T$ is the temperature of the electrons in energy units),

when we can neglect both the quantities $q$ and $k$ and the thermal spread of the electron distribution function in the calculation of $\Delta^2\varepsilon_{ik}^r$. If these conditions are not satisfied, then expression (14) is valid only far from the resonances, when

$$|\Delta|>\hbar^2q_0^2/2\mu \quad \text{as} \quad |\Delta|\to 0,$$
$$|\Delta-F|>\max\left(\frac{\hbar^2p_Fq_0}{\mu}, \frac{m_cT}{\mu}\right) \quad \text{as} \quad |\Delta|\to F;$$

while in the region of the resonance a more rigorous calculation of $\Delta^2\varepsilon_{ik}^r$ must be made on the basis of a general expression [e.g., (9)] with account taken of the fact that $q$ and $k$ are finite quantities, and also of the thermal spread of the distribution function ( a more rigorous calculation of $\Delta^2\varepsilon_{ik}^r$ is necessary also at $ql\sim 1$). The estimates show that in this case that the maximum values of $\Delta^2\varepsilon_{ik}^r$ in the region of the resonances are reached as a rule within the limits of the validity of (14). But in those cases when the finite character of $q$, $k$, and $T$ must be taken into account, the corresponding maximum values of $\Delta^2\varepsilon_{ik}^r$ usually decrease. We shall therefore use as our basis expression (14) for $\Delta^2\varepsilon_{ik}^r$, which describes the characteristic resonant singularities of this quantity and as a rule gives the highest estimates of the considered effect.

We consider now on the basis of (14) the characteristic resonant singularities of the quantity $\Delta^2\varepsilon_{ik}^r$.

a) $|\Delta|\to 0$. In this case the principal resonant term of (14) at $|\hbar\omega-E_g|>\max(s,\hbar\Omega)$ is of the form

$$\Delta^2\varepsilon_{ik}^r\approx\alpha(\omega)\frac{\pi A_{ik}(\Lambda_c-\Lambda_v)^2}{4|E_g-\hbar\omega|^{3/2}}, \tag{15}$$

where

$$\alpha(\omega)=\begin{cases}1 & \text{if } \hbar\omega<E_g,\ |\Delta|>F;\\ -i & \text{if } \hbar\omega>E_g+F;\\ 0 & \text{if } E_g<\hbar\omega<E_g+F.\end{cases}$$

(b) $|\Delta|\to F$. The analogous expression for $\Delta^2\varepsilon_{ik}^r$ is here the following:

$$\Delta^2\varepsilon_{ik}^r\approx\frac{A_{ik}F^{1/2}(\Lambda_c(m_c/\mu-1)+\Lambda_v)^2}{(\Delta-F)^2}. \tag{16}$$

If (14) is valid in the immediate vicinity of the resonance, then $\max(\Delta^2\varepsilon_{ik}^r)$ in the region of the resonances can be obtained from (15) and (16) in the following manner: as $|\Delta|\to 0$ we obtain $\max(\Delta^2\varepsilon_{ik}^r)$ from expression (15) by replacing $|\hbar\omega-E_g|$ by $\max(s,\hbar\Omega)$, and as $|\Delta|\to F$ by the corresponding replacement of $|\Delta-F|$ by $\max(s,m_cT/\mu)$. Thus, at $|\Delta|\approx F$ the thermal spread of the distribution function decreases the value of $\max(\Delta^2\varepsilon_{ik}^r)$ if $m_cT/\mu>s$.

Using (15), (16), and the corresponding expressions obtained in the analogous approximations for $\Delta^1\varepsilon_{ik}^r$ (see Ref. 7), we get for the relation

$$\xi=|\Delta^2\varepsilon_{ik}^r/\Delta^1\varepsilon_{ik}^r(u_{nj})|$$

the following estimates:

a) $|\Delta|\to 0$ for a dielectric

$$\xi\approx\left|\frac{\Lambda_c-\Lambda_v}{\max(|\Delta|,\hbar\Omega,s)}\right|; \tag{17}$$

(b) $|\Delta|\to F$ for a degenerate semiconductor

$$\xi\approx\left|\frac{\Lambda_c(m_c/\mu-1)+\Lambda_v}{\max(|\Delta-F|,s,m_cT/\mu)}\right|. \tag{18}$$

Thus, to obtain a maximum value of $|\Delta^2\varepsilon_{ik}^r|$ of the order of $|\Delta^1\varepsilon_{ik}(u_{nj})|$, the sound in the resonant case must have a power such that, for example, in the case of dielectrics,

$$|\Lambda_c-\Delta_v|\sim\max(\hbar\Omega,s).$$

Comparing the required value of $\bar{u}_{ik}$ in the resonant case ($u_{ik}^r$) with the required value of $\bar{u}_{ik}$ for $\hbar\omega\ll E_g$ ($\bar{u}_{ik}^n$), when $|\Delta^2\varepsilon_{ik}|\sim|\Delta^1\varepsilon_{ik}(u_{nj})|$, we see that for dielectrics

$$\left|\frac{\bar{u}_{ik}^r}{\bar{u}_{ik}^n}\right|\sim\left|\frac{\max(s,\hbar\Omega)}{E_g}\right|\ll 1.$$

The similar estimate for degenerate semiconductors under conditions of essentially nonlinear modulation of the concentration of the free carriers, and at a noticeable contribution of the free carriers to the dielectric constant of the crystal, takes the form

$$\left|\frac{\bar{u}_{ik}^r}{\bar{u}_{ik}^n}\right|\sim\left|\frac{\max(s,m_cT/\mu)}{\varepsilon_F}\right|\ll 1 \quad \text{as} \quad \frac{m_c}{\mu}T\ll\varepsilon_F$$

(here $\varepsilon_F$ is the Fermi energy of the conduction electrons).

Thus, the nonlinear modulation of the dielectric constant of the crystal by a sound wave manifests itself most strongly in the resonant case, when much lower sound power is necessary to attain an essentially nonlinear modulation of $\varepsilon_{ik}$ by the sound wave than in the nonresonant case.

To obtain an estimate, we consider the interaction via the strain potential,[6)] when $\langle\hat{f}\rangle\sim\Lambda_{ik}u_{ik}$. At a sound power $W\sim 5$–$10$ W/cm$^2$, a sound velocity $v_s\sim 10^5$ cm/sec, and $\Lambda_{ik}\sim 10$ eV we have $\langle\hat{f}\rangle\sim 10^{-3}$ eV, i.e., at $s\lesssim 10^{-3}$ eV (which corresponds, for example, to a momentum relaxation time $\tau\gtrsim 10^{-12}$ sec), sound having this power produces in the resonant case an essentially nonlinear modulation of the dielectric constant $\varepsilon_{ik}$ of the crystal.

The concrete case of small $|\mathbf{q}|$ considered above [see (14)–(16)] admits of a simple interpretation if $\Omega$ can also be neglected. In this approximation, the action of the sound on the electron system is in fact analogous to a perturbation that is homogeneous in space and in time, and in the resonant region the quantities $\Delta^1\varepsilon_{ik}^r(u_{nj})$ and $\Delta^2\varepsilon_{ik}^r$ are expansions of the dielectric constant $\varepsilon_{ik}$ of the crystal in series in the parameters

$$\Delta E_g=\Lambda_c-\Lambda_v, \quad \Delta F=\frac{m_c}{\mu}\Delta\varepsilon_F=-\frac{m_c}{\mu}\Lambda_c,$$

where $\Delta E_g$ and $\Delta F$ are the changes of $E_g$ and $F$ due to the modulation of the width of the forbidden band and of the concentration of the free carriers by the sound wave propagating through the crystal.

Consequently, in this approximation the expressions for $\Delta^1\varepsilon_{ik}^r(u_{nj})$ and $\Delta^2\varepsilon_{ik}^r$ take the form

$$\Delta^1\varepsilon_{ik}^r(u_{nj})=\frac{\partial\varepsilon_{ik}^0}{\partial E_g}\Delta E_g+\frac{\partial\varepsilon_{ik}^0}{\partial F}\Delta F, \tag{19}$$

$$\Delta^2\varepsilon_{ik}^r=\frac{1}{2}\frac{\partial^2\varepsilon_{ik}^0}{\partial E_g^2}(\Delta E_g)^2+\frac{1}{2}\frac{\partial^2\varepsilon_{ik}^0}{\partial F^2}(\Delta F)^2+\frac{\partial^2\varepsilon_{ik}^0}{\partial E_g\partial F}\Delta F\Delta E_g. \tag{20}$$

It is clear from (19) and (20) that in this approximation the resonant singularities of $\Delta^2\varepsilon_{ik}^r$ are connected with the singularities of the derivatives $\partial\varepsilon_{ik}^0/\partial E_g$ and $\partial\varepsilon_{ik}^0/\partial F$, and the nonlinear modulation of the quantity $\varepsilon_{ik}$ by the sound wave is produced at sound powers such that when the parameters $E_g$ and $F$ are altered by the sound wave a noticeable change takes place also in these derivatives. It should be noted that these simple considerations do not hold when it is necessary to take into account the finite character of $q$ and $\hbar\Omega$; in this case it is necessary to use a more general expression for $\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)$, for example expression (9).

It should also be noted that at $\xi\sim1$ the calculation of the quantities $\Delta^1\varepsilon_{ik}$, $\Delta^2\varepsilon_{ik}$, etc. by perturbation theory becomes substantially more complicated, for in this case we cannot neglect the contribution of the higher orders of perturbation theory. Generally speaking in this case the quantities $\Delta^1\varepsilon_{ik}$, $\Delta^2\varepsilon_{ik}$, $\Delta^2\varepsilon_{ik}$, ... are of the same order, but all are usually small at $|\langle\hat{f}\rangle|\ll\varepsilon_F$ compared with $\varepsilon_{ik}^0(\omega)$, i.e., the sound produces in this case a small but an essentially nonlinear modulation of $\varepsilon_{ik}$.

We see thus from the foregoing that at $\xi\sim1$, in the resonant case, the modulation of the dielectric constant of the crystal by the sound is essentially nonlinear. When light is diffracted by the sound that produces the nonlinear modulation of $\varepsilon_{ik}$, the intensity of the diffraction orders will have a nonlinear dependence on the sound power also at low diffraction effectiveness, while the effective AO interaction over the length of the crystal in the Bragg diffraction regime in the case of monochromatic sound is possible at different incidence angles $\theta_{inc}$ of the light on the sample, for example, at $\sin\theta_{inc}=nqc/2\omega$, where $n=\pm1, \pm2, \ldots$, for the case of isotropic diffraction.[7)]

The possibility of nonlinear modulation of $\varepsilon_{ik}$ by a sound wave in the resonant case is indeed ensured by the fact that $\Omega\ll\omega$ (actually $\Omega\sim10^{-5}\omega$) i.e., at of resonance with the frequency $\omega$, resonance occurs in fact also with the frequencies $\omega\pm\Omega$, $\omega\pm2\Omega$, .... This constitutes the essential difference between the effect considered here and the effect, e.g., of the onset of nonlinear susceptibilities in the field of the electromagnetic wave of frequency $\omega$ itself (see Ref. 16 and 17), where resonance at this frequency, for example $\hbar\omega\sim E_g$, is not accompanied generally speaking by resonance with the light frequencies $2\omega$, $3\omega$, ... As a result even under resonance conditions at real optical powers the nonlinear susceptibilities decrease rapidly in magnitude for the frequencies $2\omega$, $3\omega$, ... However, an effect similar to that considered in the present paper is apparently possible under definite condition also if the dielectric constant is electro-optically modulated by an electromagnetic wave of frequency $\omega_0\ll\omega$ in the case when resonant conditions similar to those considered above obtain for the frequency $\omega$.

[1)]Much attention has been paid recently to resonant diffraction of light by sound, which takes place when $\hbar\omega\approx\Delta E$ (here $\Delta E$ is the characteristic energy difference between the electron levels of the crystal). This is caused by the interesting singularities that the AO interaction has in this frequency region (see Refs. 2 and 4–7).

[2)]The quantity $\Delta^2\varepsilon_{ik}$ has also a component that is constant in time and in space and is quadratic in $u_{ik}$, i.e., in this approximation the sound wave produces also a constant increment to $\varepsilon_{ik}^0$. This effect, however, will not be considered in detail in the present paper.

[3)]In semiconductors the higher harmonics are generated as a rule because of electronic nonlinearity, whereas the generation of the higher harmonics in dielectrics calls for higher sound powers.[11]

[4)]Light diffraction by electron "walls" that are bunched by sufficiently strong sound were considered in Ref. 14.

[5)]Calculation has shown that under these assumptions expression (14) is valid only at $ql\ll1$ (in the calculation of $\Delta^2\varepsilon_{ik}^r$ it is necessary here to use the general expression for $\Delta^2\varepsilon_{ik}(\omega\pm2\Omega)$ obtained with relations (7) and (8) taken into account.

[6)]In the presence of a sufficiently high concentration of free, carriers as in dielectrics, the principal electron-phonon interaction mechanism contributing to $\Delta^2\varepsilon_{ik}^r$ is interaction via the strain potential, which generally speaking cannot be simultaneously screened out in the valence and in the conduction bands[7] (a similar effect takes place in multivalley semiconductors[15]).

[7)]If the monochromatic sound modulates $\varepsilon_{ik}$ linearly, then effective AO interaction over the length $d$ of the interaction of the sound with the light is really possible for isotropic Bragg diffraction at light-incidence angles $\sin\theta_{inc}\approx\pm qc/2\omega$, but in the case when $\sin\theta_{inc}\approx nqc/2\omega$, $n=\pm2, \pm3\ldots$ it is easy to show that for effective AO interaction it is necessary to have interaction lengths with the sound larger by a factor $Q^{|n|-1}$ [here $Q=q^2d/k\gg1$ is the parameter of the Bragg diffraction (see Refs. 1 and 2)].


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