Cyclotron–phonon resonance in the case of a nonlinear electron–phonon polarization interaction with allowance for nonequidistant Landau bands

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A theory of cyclotron–phonon resonance is developed for the case when the difference between the Landau levels is equal to the energy of two-phonon combinations ($2LO$, $LO+TO$, $2TO$). An allowance is made for nonequidistant Landau bands. A comparison of the value of the splitting energy under pinning conditions with the experimental values is used to estimate the constants of the nonlinear electron–phonon polarization interaction.

Many experimental investigations of the magnetoabsorption in semiconductors have revealed pinning (with a characteristic discontinuity of the curve representing the dependence of the maximum absorption coefficient on the magnetic field) in the case of a cyclotron–phonon resonance (CPR) which can be explained by means of one-phonon[1,2] and two-phonon[3,4] interaction Hamiltonians allowing for various branches of the crystal lattice vibrations. The experimental investigations reported in Refs. 3–8 have shown that pinning can be due to the interaction of electrons with the following combinations of optical phonons: $2LO(m)$, $2TO(m)$, $LO(m)+TO(m)$, where $m=X$, L, and Γ are the high-symmetry points in the Brillouin zone.[9] The interaction of the electron subsystem with these phonon combinations should give rise to the observed effects not only in the CPR case, but, for example, in the case of a magnetophonon resonance, under Raman scattering conditions, etc.

In this connection it would be useful to estimate the constants governing the electron–phonon interaction in a two-phonon Hamiltonian. We shall therefore develop a theory of the CPR lineshape in the case of a nonlinear electron–phonon polarization interaction, allowing for a nonequidistant distribution of the Landau bands.

The two-phonon processes are manifested most clearly under pinning conditions when the frequencies of the longitudinal $\omega_{LO(m)}$ and transverse $\omega_{TO(m)}$ phonons and the separation Ω between the nth and ground Landau levels satisfy

$$\Omega_n = p\omega_{LO(m)} + q\omega_{TO(m)}, \qquad (1)$$

where n, p, and q are arbitrary integers; $\omega_{LO(m)}$ is the frequency of a longitudinal phonon; $\omega_{TO(m)}$ is the frequency of a transverse phonon; $\Omega_n=(\varepsilon_n-\varepsilon_0)/\hbar$ [ε_n is given by Eq. (2)]. Under these conditions the upper electron level, to which an electron is transferred as a result of the CPR absorption, is very unstable because the probability of its decay is high.[10,11] If p and q assume the values 0, 1, and 2, so that we have $p+q=2$ (which corresponds to the experimental situation discussed in Ref. 4), then the nth level has the same energy as the $n=0$ level plus the energy of two phonons if the condition (1) is satisfied. An allowance for the interaction between these levels lifts the degeneracy and splits the CPR absorption line into its components.[10,11]

We consider an n-type semiconductor having cubic symmetry with a nonparabolic dispersion law subjected to a quantizing magnetic field at temperatures such that electrons fill only the $n=0$ Landau band and optical phonons are not excited. The transition of an electron to higher Landau levels on absorption of an electromagnetic wave is due to a linear optical polarization interaction.[12]

The electron energy levels in a magnetic field, considered without requiring the Landau bands to be equidistant or the conduction band to be parabolic, can be written in the form (see Ref. 13)

$$\varepsilon_{n\pm} = -\frac{1}{2}\varepsilon_g + \frac{\varepsilon_g}{2}\left\{1+\frac{4f_1(\varepsilon)}{\varepsilon_g}\left[\hbar\Omega_{(n+1/2)} \mp \frac{1}{2}g\mu H f_4(\varepsilon)\right]\right\}^{1/2}\left(1+A_n\frac{\hbar^2k_z^2}{2m^*}\right),$$

$$f_1(\varepsilon)=\frac{(\varepsilon_g+\Delta)(\varepsilon+\varepsilon_g+2\Delta/3)}{(\varepsilon_g+2\Delta/3)(\varepsilon+\varepsilon_g+\Delta)},$$

$$f_4(\varepsilon)=\frac{\varepsilon_g+2\Delta/3}{\varepsilon+\varepsilon_g+2\Delta/3}, \qquad (2)$$

$$g=2+\left(1-\frac{m}{m^*}\right)\frac{2\Delta}{3\varepsilon_g+2\Delta},$$

$$A_n=\frac{2f_1(\varepsilon)}{\varepsilon_g(1+4f_1(\varepsilon)\hbar\Omega_{(n+1/2)}/\varepsilon_g)}.$$

Here ε_g is the width of the band gap, m^* is the effective mass of an electron, m is the mass of a free electron, Δ is the spin–orbit splitting; μ is the Bohr magneton, Ω is the cylotron frequency, and H is the magnetic field intensity. The systems of equations (2) is derived on the assumption that the states with low values of k_z play the dominant role in the CPR process when the condition (1) is satisfied.

A nonlinear polarization interaction Hamiltonian can be defined as in Ref. 14:

$$H=\sum_{\alpha\alpha'\mathbf{q}\mathbf{q}'jj'}\{C_{jj'}(\mathbf{q},\mathbf{q}')J_{\alpha\alpha'}(\mathbf{q}+\mathbf{q}')b_{\mathbf{q}j}b_{\mathbf{q}'j'}+\text{H.c.}\}a_\alpha^+a_{\alpha'}, \qquad (3)$$

where a combination of $2LO(m)$ phonons corresponds to

$$C_{jj'}(\mathbf{q},\mathbf{q}')=\frac{iA\hbar\omega_{LO(m)}}{|\mathbf{q}+\mathbf{q}'|V}\Gamma_{jj'}(\mathbf{q},\mathbf{q}')\delta_{j1}\delta_{j'1},$$

whereas for a combination of $LO(m)+TO(m)$ phonons we have

$$C_{jj'}(\mathbf{q},\mathbf{q}')=iD\hbar[\omega_{LO(m)}\omega_{TO(m)}]^{1/2}\Gamma_{jj'}(\mathbf{q},\mathbf{q}')\times\{\delta_{j1}(\delta_{j'2}+\delta_{j'3})+\delta_{j'1}(\delta_{j2}+\delta_{j3})\},$$

and in the case of a combination of $2TO(m)$ phonons, we find that

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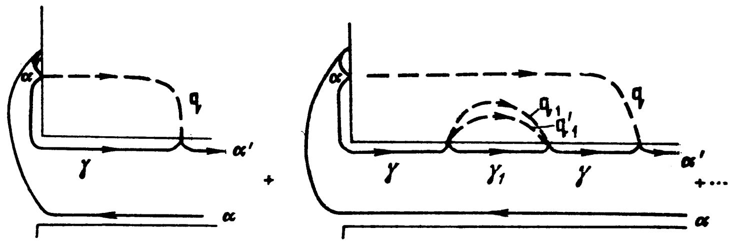

$$C_{jj'}(\mathbf{q},\mathbf{q}')=\frac{iC\hbar\omega_{TO(m)}}{|\mathbf{q}+\mathbf{q}'|V}\Gamma_{jj'}(\mathbf{q},\mathbf{q}')(1-\delta_{j1}\delta_{j'1}),$$

$$\Gamma_{jj'}(\mathbf{q},\mathbf{q}')=\left\{\frac{(q+q')_z}{|\mathbf{q}+\mathbf{q}'|}e_{\mathbf{q}j}^{x}e_{\mathbf{q}'j'}^{y}+\text{c.p.}\right\},$$

where $\mathbf{q}$ and $\mathbf{q}'$ are the wave vectors of optical polarization phonons, α and α are the quantum numbers of electrons in a magnetic field, $b_{\mathbf{q}j}$ and a_α are the phonon and electron second-quantization operators, respectively, V is the normalization volume, $J_{\alpha\alpha'}(\mathbf{q}+\mathbf{q}')$ is a matrix element of the $\exp\{i(\mathbf{q}+\mathbf{q}',\mathbf{r})\}$ operator calculated using the wave functions of electrons in a magnetic field,[15,16] j and j' are the numbers of the vibration branches ($j=1$ refer to LO phonons, whereas $j=2$, 3 refer to TO phonons), A, D, and C are constants which need to be determined, and c.p. stands for cyclic permutation.

The phonon polarization vectors are of the form

$$\mathbf{e}_{\mathbf{q}_1}=(e_{\mathbf{q}}^{x},e_{\mathbf{q}}^{y},e_{\mathbf{q}}^{z}),$$
$$e_{\mathbf{q}}^{\mu}=q^{\mu}/|\mathbf{q}|,\ \mu=x,y,z, \tag{4}$$
$$\mathbf{e}_{\mathbf{q}_2}=\left(\frac{e_{\mathbf{q}}^{y}}{[(e_{\mathbf{q}}^{x})^2+(e_{\mathbf{q}}^{y})^2]^{1/2}},-\frac{e_{\mathbf{q}}^{x}}{[(e_{\mathbf{q}}^{x})^2+(e_{\mathbf{q}}^{y})^2]^{1/2}},0\right),$$
$$\mathbf{e}_{\mathbf{q}_3}=\left(\frac{e_{\mathbf{q}}^{z}e_{\mathbf{q}}^{x}}{[(e_{\mathbf{q}}^{x})^2+(e_{\mathbf{q}}^{y})^2]^{1/2}},\frac{e_{\mathbf{q}}^{y}e_{\mathbf{q}}^{z}}{[(e_{\mathbf{q}}^{x})^2+(e_{\mathbf{q}}^{y})^2]^{1/2}},-[(e_{\mathbf{q}}^{x})^2+(e_{\mathbf{q}}^{y})^2]^{1/2}\right).$$

The absorption coefficient can be calculated using the Kubo formula for the complex electrical conductivity[17] and the diagram technique of Konstantinov and Perel'[18] when the density matrix $f_{\alpha\alpha'}^{\nu}$ is found from the quantum kinetic equation

$$(S+i\omega_{\alpha'\alpha})f_{\alpha\alpha'}^{\nu}=F_{\alpha\alpha'}^{\nu}+\sum_{\beta\beta'}f_{\beta\beta'}^{\nu}W_{\beta\alpha}^{\beta'\alpha'}, \tag{5}$$

where $S=-\omega+\delta$, $\delta\to 0$; $\omega_{\alpha'\alpha}=\omega_{\alpha'}-\omega_\alpha$ (ω is the frequency of the absorbed radiation), $F_{\alpha\alpha'}^{\nu}$ and $W_{\beta}^{\beta'\alpha'}$ are the angular and horizontal irreducible parts of Eq. (5).

it is shown in Ref. 10 that the angular parts of Eq. (5) are important; an allowance for this circumstance and a detailed analysis of the perturbation theory series for complex electrical conductivity demonstrates that the infinite series of graphs shown in Fig. 1 is an essential part of the problem. The CPR line profile is determined by the function W which, as a result of summation of an infinite series in Fig. 1, results in renormalization of the electron line under the external phonon line.[10] In this case W has the form shown in Fig. 2:

$$W=\sum_{\gamma_1\mathbf{q}_1\mathbf{q}_1'jj'}\frac{(i\hbar)^2|C_{ij'}(\mathbf{q}_1,\mathbf{q}_1')|\,|J_{\gamma\gamma_1}(\mathbf{q}_1+\mathbf{q}_1')|^2}{[S+i(\omega_{\gamma_1\alpha}+\omega_{LO(\Gamma)}+p\omega_{jO(m)}+q\omega_{j'O(m)})]}. \tag{6}$$

If we carry out the necessary integration with respect to the phonon momenta and retain in Eq. (6) only the resonance terms, which corresponds to $n_{\gamma_1}=0$ in W, we find that for a combination of $2LO(\Gamma)$ phonons, we have

$$\lambda=\frac{\Omega_3-3\omega_{LO(\Gamma)}}{\Omega},\quad B=\frac{0{,}32A^2\omega_{LO(\Gamma)}^2}{32\cdot 2^{1/2}\pi\Omega^2Ra^3},$$
$$\lambda=\frac{\Omega_3-2\omega_{LO(\Gamma)}-\omega_{TO(\Gamma)}}{\Omega}, \tag{7}$$
$$B=\frac{0{,}28D^2\omega_{LO(\Gamma)}\omega_{TO(\Gamma)}}{32\cdot 2^{1/2}\pi\Omega^2Ra^3}$$

whereas for a combination of $LO(\Gamma)+TO(\Gamma)$ phonons, we obtain

$$W=-\frac{BB_0\Omega}{(\varepsilon+\lambda)^{1/2}},$$

and for a combination of $2TO(\Gamma)$ phonons, we find that

$$\lambda=\frac{\Omega_3-\omega_{LO(\Gamma)}-2\omega_{TO(\Gamma)}}{\Omega},$$
$$B=\frac{0{,}8C^2\omega_{TO(\Gamma)}^2}{32\cdot 2^{1/2}\pi\Omega^2Ra^3},$$

where $\varepsilon=(\omega-\Omega_3)/\Omega$ is the dimensionless energy of an electron measured from the $N=3$ Landau level, $\Omega_3=(\varepsilon_3-\varepsilon_0)/\hbar$ is the separation between the $n=3$ and $n=0$ Landau levels, a is a lattice constant, and R is the magnetic length;

$$B_0=[1+2\hbar\Omega/\varepsilon_g]^{1/4}.$$

We can show[11] that for each of the three phonon combinations $2LO(\Gamma)$, $LO(\Gamma)+TO(\Gamma)$, and $2TO(\Gamma)$ the spectrum of the electron–phonon system consists of two branches (an electron level with the quantum Landau number $n=3$ and a level with $n=0$ plus two optical phonons from the corresponding phonon combination participating in the pinning process).

The electrical conductivity is now described by

$$\sigma^{\mu\nu}(\omega)=\sum_{\alpha\alpha'}j_{\alpha\alpha'}^{\mu}f_{\alpha\alpha'}^{\nu},$$

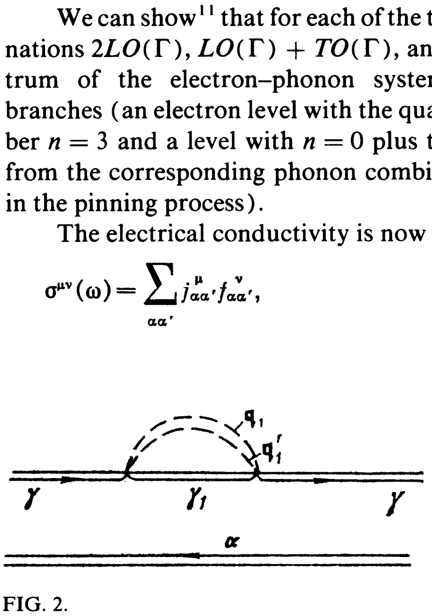

FIG. 2.

where $j^{\mu}_{\alpha\alpha'}$ is a matrix element of the current density. If we use Figs. 1 and 2, we find that the electrical conductivity becomes

$$\sigma^{\mu\nu}(\omega) = \sum_{\alpha\alpha'\gamma q} \frac{(i\hbar)^{-1} j^{\mu}_{\alpha\alpha'} j^{\nu}_{\alpha'\alpha} |C_q|^2 |J_{\alpha'\gamma}(q)|^2 N_\alpha}{[S+i(\omega_{\gamma\alpha}+\omega_0)-W]\hbar\omega_{\alpha'\alpha}\hbar(\omega_{\gamma\alpha}+\omega_0)(S+i\omega_{\alpha'\alpha})}, \quad (8)$$

where N_α is the electron distribution function and C_q is the electron–phonon interaction constant of the Frohlich Hamiltonian.[12]

The absorption coefficient of external radiation $K(\omega)$, related to the diagonal part of the electrical conductivity given by Eq. (8) by the expression

$$K(\omega) = \frac{2\pi}{cn_0} \operatorname{Re} \sigma^{\mu\nu}(\omega)$$

(n_0 is the refractive index and c is the velocity of light) is obtained from Eq. (8) by integrating the resonant denominator with respect to q_z and k_z. At low temperatures we can replace N_α by a step and the absorption coefficient is then given by

$$\frac{K(\omega)}{K_0} = \begin{cases} 0, & \varepsilon<0,\ b<0,\ |b|>x_0^2/B_0^2, \quad (9) \\ \ln \dfrac{[b+(x_0/B_0)^2]^{1/2}+x_0/B_0}{|[b+(x_0/B_0)^2]^{1/2}-x_0/B_0|}, & \varepsilon^*<\varepsilon<0, \quad (10) \end{cases}$$

$$\frac{K(\omega)}{K_0} = \begin{cases} \dfrac{f(\varepsilon)x_0/B_0}{[(\varepsilon-(BB_0)^{2/3}/2)^2+{}^3/_4(BB_0)^{4/3}]^{1/2}}, & \varepsilon>0, \quad \dfrac{x_0}{B^{1/3}B_0^{4/3}}<1, \quad (11) \\ \ln \dfrac{(2x_0/B_0)^2}{(\varepsilon+(BB_0)^{2/3}/2)^2+{}^3/_4(BB_0)^{4/3}}, & \varepsilon>0, \quad \dfrac{x_0}{B^{1/3}B_0^{4/3}}>1, \quad (12) \end{cases}$$

where $b = \varepsilon + iBB_0/\varepsilon^{1/2}$, $x_0 = 2^{1/2}\pi^2 R^3 N$ is the dimensionless Fermi momentum, N is the density of electrons in the $n=0$ Landau band, $\Omega_1 = (\varepsilon_1 - \varepsilon_0)/\hbar$, $\Omega_3 = (\varepsilon_3 - \varepsilon_0)/\hbar$, and

$$K_0 = l^2\hbar\Omega/[cn_0 2^{1/2}\pi^2 m^2 R^4 \Omega_1(\Omega_1+\omega_{LO(\Gamma)})(\Omega_3-\Omega_1)].$$

In the range where $\varepsilon<0$ and $b<0$, but $|b| \leqslant x_0^2/B_0^2$, we find that $K(\omega)=0$ in Eq. (9).

Beginning from the frequencies satisfying the condition $|b| \leqslant x_0^2/B_0^2$, the photon energy becomes sufficient to transfer an electron from the $n=0$ Landau level to the $n=3$ Landau band and to create a phonon. In the range $\varepsilon^* < \varepsilon < 0$, where ε^* is the real root of the equation $b + x_0^2/B_0^2$, we find that Eq. (10) is valid and it has a maximum at $\varepsilon = -(BB_0)^{2/3}$. In this case, we obtain

$$\varepsilon^* \approx -(x_0/B_0)^2, \quad x_0/B^{1/3}B_0^{4/3}>1,$$
$$\varepsilon^* \approx -(BB_0)^{2/3} - {}^2/_3(x_0/B_0)^2, \quad x_0/B^{1/3}B_0^{4/3}<1.$$

For $\varepsilon>0$ and $x_0/B^{1/3}B^{4/3}<1$, then Eq. (11) with a maximum at $\varepsilon = 2(BB_0)^{2/3}$ applies; however, for $x_0/B^{1/3}B_0^{4/3}>1$, then Eq. (12) is valid and it has a maximum at $\varepsilon = 0.5(BB_0)^{2/3}$.

We now estimate the constants A, D, and C governing the strength of the two-phonon interaction, we do this by a comparison with the experimental data reported in Refs. 3, 4, 7, and 8 for n-type InSb. Pinning has been observed for the $2LO(\Gamma)$ combination of phonons when the condition $n=3$, $p=2$, and $q=0$ is satisfied in Eq. (1). In the case of a $LO(\Gamma)+TO(\Gamma)$phonon combination there is no pinning and this is characterized by $n=3$, $p=1$, and $q=1$ in Eq. (1); pinning does appear in the case of $2TO(\Gamma)$ phonons for $n=3$, $p=0$, and $q=2$. The total splitting energy in pinning is $\hbar\Delta_{\rm exp} = 0.2$–$0.8$ meV in all cases. Measurements reported in these papers were carried out on samples with an electron density $N = 1.6\times10^{16}$ cm^{-3}, which corresponds to the case when $x_0/(B^{1/3}B_0^{4/3})>1$. It then follows from Eqs. (10) and (12) that the theoretical splitting under pinning conditions is

$$\hbar\Delta_{\rm theor} = 1.5(BB_0)^{2/3}\hbar\Omega. \quad (13)$$

In the case of a combination of $2LO(\Gamma)$ phonons we can determine the splitting energy if $\hbar\Omega$ in Eq. (13) is found from $\Omega_3 = 2\omega_{LO(\Gamma)}$. It follows from this equation that pinning should be observed in magnetic fields of intensity $H = 23.8$ kOe. The experiments give $H = 24$ kOe. A comparison of the experimental and theoretical values of the splitting gives the constant $A = (2.4 \pm 0.8)\times10^{-14}$ cm^2. In the case of the $LO(\Gamma)+TO(\Gamma)$phonon combination the solution of the equation $\Omega_3 = \omega_{LO(\Gamma)} + \omega_{TO(\Gamma)}$ gives $H = 23$ kOe for the magnetic field. Pinning is observed in experiments in a field $H = 23.5$ kOe. In this case the constant is $D = (2.6 \pm 0.9)\times10^{-14}$ cm^2. For the $2TO(\Gamma)$ phonon combination the magnetic resonance field deduced from the equation $\Omega_3 = 2\omega_{TO(\Gamma)}$ is $H = 22$ kOe. The experimental value is $H = 22.2$ kOe. The constant C deduced from the experimental splitting under pinning conditions is then $(1.6 \pm 0.6)\times10^{-14}$ cm^2.

Note that

$$\Omega_3 = \frac{\varepsilon_g}{2\hbar}[(1+14\hbar\Omega/\varepsilon_g)^{1/2} - (1+2\hbar\Omega/\varepsilon_g)^{1/2}],$$

which corresponds to the selection $f_1(\varepsilon) = 1 f_4(\varepsilon) = 0$ in Eq. (2). The correctness of this selection is supported by the very good agreement between the theoretical and experimental values of the magnetic resonance fields. We determined the constants assuming the following phonon energies: $\hbar\omega_{LO(\Gamma)} = 24.4$ meV and $\hbar\omega_{TO(\Gamma)} = 22.8$ meV (Refs. 19 and 20).

We shall conclude by noting that the two-phonon interaction gives rise to effects comparable in magnitude with the effects due to the one-phonon interaction.[21,22]

It would be interesting to find the interaction constants for the two-phonon Hamiltonian theoretically since the experimental values are already known. However, this complex task is outside the scope of the present work.

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