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Polaron effect in resonant Raman scattering from quantum wells in a high magnetic field: Decompensation of electron and hole contributions

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In the framework of a simple model we show that there is no compensation of electron and hole contributions to the intensity of one-phonon resonant Raman scattering in the range of parameters where the incoming or outgoing resonances are split by the Fröhlich electron-phonon interaction (magnetopolaron region). The difference in the penetration of electron and hole wave functions into the barrier, usually assumed to account for the nonzero value of the one-phonon scattering intensity, can be considered as a competing (or complementing) mechanism in the magnetopolaron range, depending on the parameters of the quantum-well structure.

The longstanding interest in the magnetopolaron effect (see, for example, Refs. 1–4) reflects both the difficulties in its theoretical treatment and its appearance in various experimental investigations. Among others, optical methods have been used to study this effect in semiconductor materials. The splitting of the electronic excitations in the range of resonant polaron coupling in a high magnetic field was observed in the spectra of absorption and reflection in bulk samples of TlBr.[5] Similar effects have been studied in the magnetoabsorption of bulk GaAs and CdTe (Refs. 6 and 7) and in InP by Raman spectroscopy.[8] Theoretical models of various types were applied to the difficult problem of the calculation of electronic excitation spectra in the presence of the polaron effect.[4,9,10] Inclusion of the polaron effect in calculations of the Raman efficiency leads to even stronger difficulties.

The polaron splitting of resonances in the two-phonon Raman scattering from a quantum well in a high magnetic field has been considered in Ref. 11. The case of one-phonon scattering via the states of uncorrelated electron-hole pairs has a particular feature which makes it different from multiphonon scattering: under conditions of negligible polaron coupling for Fröhlich interaction the amplitude of the process in the dipole approximation (i.e., when the wave vector of the phonon is zero) vanishes because of the compensation of the electron and hole contributions. One-phonon resonant Raman scattering in quantum wells was recently studied in Refs. 12 and 13 in the range of resonant polaron coupling between two Landau levels. It was assumed that the electron and hole contributions to the scattering amplitude differ only in sign and a nonzero value of the scattering efficiency is due to the different penetration of the electron and hole wave functions into the barrier. With this assumption, which is generally not correct, the electron contribution to the amplitude was investigated.

In this paper we use a simple model based on the effective-mass approximation to show that in the limit of infinite barriers (complete confinement of the electron and hole wave functions in the well) and in the range of resonant polaron coupling for electrons (*or* for holes) the electron and hole contributions to the amplitude *do not* compensate each other (they *do* without polaron coupling). The decompensation is induced by the polaron effect and takes place in the range of magnetic fields and laser frequencies securing the resonant coupling of the Landau levels via electron-phonon interaction and either incoming or outgoing resonance for optic transitions. Out of this range, however, the decompensation is negligible.

The efficiency of Raman scattering in a general form[14,15] is given by

$$d^2S/d\Omega\, d\omega_s = (\omega_s^3\omega_l/c^4)[n(\omega_s)/n(\omega_l)]e^*_{s\alpha}e_{s\beta}e_{l\gamma}e^*_{l\lambda}\mathbf{S}_{\alpha\gamma\beta\lambda}, \tag{1}$$

where Ω is the solid angle, c the velocity of light in vacuum, $n(\omega)$ the refractive index, $\mathbf{e}_l$ ($\mathbf{e}_s$) the polarization vector, ω_l (ω_s) the frequency for incident (scattered) radiation, and $\mathbf{S}_{\alpha\gamma\beta\lambda}$ the light-scattering tensor of fourth rank. The general version of the diagrammatic technique for calculating $\mathbf{S}_{\alpha\gamma\beta\lambda}$ has been developed in Refs. 14 and 15. We consider the scattering by optical phonons in a single quantum well with completely confined electron and hole wave functions. The nonrenormalized wave functions for electrons and holes in a high magnetic field at the Landau gauge are given by

$$\Psi_{n,N,k_y} = [\exp(ik_y y)/\sqrt{L_y}]u_n(x - x_{k_y})\varphi_N(z)v_0(\mathbf{r}), \tag{2}$$

where L_xL_y is the area of a quantum well and N (n) is the number of the size-quantized (Landau) level. The wave function of a one-dimensional oscillator in the Landau sublevel with quantum number n is given by

$$u_n(x) = (m_e\omega_e/\pi\hbar)^{1/4}(1/\sqrt{n})\exp\left(-m\omega_e/2\hbar x^2\right) \times H_n(x\sqrt{2m_e\omega_e/\hbar}) \tag{3}$$

with the Hermite polynomials $H_n(x)$, $\omega_{e(h)} = eH/m_{e(h)}c$ being the cyclotron frequency and $m_{e(h)}$ the effective mass of the electron and the hole. The wave functions for electron and hole states in the well are

$$\varphi_N(z) = \sqrt{2/a}\cos\pi Nz/a, \quad N = 1,3,5,..., \tag{4}$$

$$\varphi_N(z) = \sqrt{2/a}\sin\pi Nz/a, \quad N = 2,4,6,..., \tag{5}$$

where a is the well width and z lies within the interval $-a/2 \le z \le +a/2$. The position of the oscillator center for electrons and holes is equal to $x_{k_y} = \mp l_H^2 k_y$, respectively, $l_H = \sqrt{\hbar c/eH}$ being the magnetic length and $v_0(\mathbf{r})$ the Bloch function.

For the one-phonon Raman process in the dipole approximation (or in the backscattering configuration) the in-plane component of the phonon wave vector is equal to zero (as shown below), thus avoiding the question of the in-plane dispersion of the Raman phonon and contribution to the scattering amplitude from interface modes. We also assume the broadening of the phonon states to be negligible. Under these approximations the light-scattering tensor can be written in terms of the amplitude as

$$\mathbf{S}_{\alpha\gamma\beta\lambda} = \sum_{q_z} [A_{\alpha\gamma}(q_z)A^*_{\beta\lambda}(q_z)/V_0\hbar^2\omega_s^2\omega_l^2] \times\delta[\omega_l - \omega_s - \omega_{\rm LO}(q_z)], \quad (6)$$

where V_0 is the normalization volume and $A_{\alpha\gamma}(q_z)$ the one-phonon scattering amplitude as a function of the quantized wave vector $q_z = \pi m/a$ for confined LO phonons.

The two diagrams contributing to the one-phonon scattering amplitude are shown in Figs. 1(a) and 1(b). The full (open) circles correspond to the electron and hole interaction with confined LO phonons (photons). The dashed (wavy) lines are for photons (phonons) and the upper (lower) solid lines for electrons (holes). One must take into account the fact that the electron and the hole should be excited in the states with the same quantized subband index N and Landau levels n in both diagrams for scattering amplitude in Figs. 1(a) and 1(b). There is no constraint on the value of the z component of the Raman phonon wave vector.

To calculate the scattering amplitude we need the electron $G_e(N,n;\omega)$ and hole $G_h(N,n;\omega)$ Green functions for confined states which are given by

$$G_e(N,n;\omega) = [\omega - \omega_N^e - (n+\tfrac{1}{2})\omega_e - \Xi^e(N,n;\omega) + i\gamma_e/2]^{-1}, \quad (7)$$

$$G_h(N,n;\omega) = [\ \omega - \omega_N^h - (n+\tfrac{1}{2})\omega_h - \omega_g - \Xi^h(N,n;\omega) + i\gamma_h/2]^{-1}, \quad (8)$$

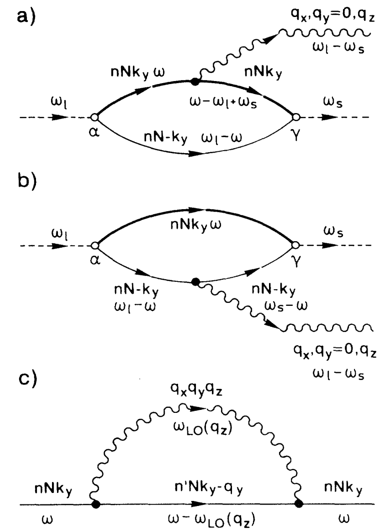

FIG. 1. The diagrams for electron (a) and hole (b) contributions into the amplitude of the one-phonon resonant Raman scattering from a quantum well in a high magnetic field. The bold lines for the electron Green functions take into account the renormalization with the self-energy diagram (c) shown at the bottom.

where ω_N^e (ω_N^h) is the size-quantized energy of the electron (hole) and $\hbar\omega_g$ the energy gap. The self-energy $\Xi(N,n;\omega)$ accounts for polaron coupling. Although only confined optical modes contribute to the one-phonon amplitude in the backscattering configuration, the interaction with interface modes can be important for the renormalization of the intermediate electronic states. Here we assume that the polaron coupling is only produced by confined phonons and neglect by interface phonons. This approximation should be quantitatively correct for thick layers (see Refs. 16 and 17).

To evaluate the expression for electron (hole) -LO-phonon vertices we take the Hamiltonian for Fröhlich electron-phonon interaction based on the hydrodynamic model for confined optical vibrations[18] in the form of[19]

$$H_{e\text{-ph}} = 2\Bigg\{ \sum_{\mathbf{q}_\perp, m=1,3,5,\ldots} \left[(-1)^{(m+1)/2}\sin\left(\frac{\pi m z}{a}\right) + \exp\left(-\frac{aq_\perp}{2}\right)\sinh(q_\perp z)\right] + \sum_{\mathbf{q}_\perp, m=2,4,6,\ldots} \left[(-1)^{m/2}\cos\left(\frac{\pi m z}{a}\right) - \exp\left(-\frac{aq_\perp}{2}\right)\cosh(q_\perp z)\right]\Bigg\}[C_\mathbf{q} b_\mathbf{q}\exp(i\mathbf{q}_\perp\mathbf{r}_\perp) + C^*_\mathbf{q} b^\dagger_\mathbf{q}\exp(-i\mathbf{q}_\perp\mathbf{r}_\perp)], \quad (9)$$

where

$$C_\mathbf{q} = \mp i\hbar\omega_{\rm LO}(4\pi\alpha_{e(h)}l^3_{pe(h)}/V_0)^{1/2}(1/l_{pe(h)}q), \quad l_{pe(h)} = \sqrt{\hbar/2m_{e(h)}\omega_{\rm LO}}, \quad (10)$$

$q = \sqrt{q_\perp^2 + (\pi m/a)^2}$, α is the Fröhlich electron-phonon coupling constant and $b^\dagger_\mathbf{q}$ ($b_\mathbf{q}$) the phonon creation (annihilation) operator. The upper (lower) sign corresponds to the electrons (holes). Although the values α and l_p are different for electrons and holes, the interaction $C_\mathbf{q}$ differs only in sign.

The matrix elements of the Hamiltonian of Eq. (9) evaluated between the wave functions of Eq. (2) are different from zero only for m-even confined modes because of the aforementioned selection rules for size-quantized levels number N which allow the excitation of electrons and holes with the same N in the interband optical transitions. For even m we obtain for transitions involving one confined LO phonon:

$$R^{mN}_{nn'}(k_y,q_x,q_y) = \langle N,n',k'_y|H_{e\text{-ph}}|N,n,k_y\rangle = \pm(4/\pi)C^*_\mathbf{q}\delta_{k_y,k'_y+q_y}\exp\left[\mp i l_H^2 q_x(k_y - q_y/2)\right]K_{n,n'}(\pm l_H q_y, -l_H q_x)\Phi(l_H q_\perp, m, N), \quad (11)$$

$$K_{n,n'}(\rho) = \sqrt{\min(n!,n'!)/\max(n!,n'!)}\, i^{|n-n'|}\exp(-\rho^2/4)(\rho/\sqrt{2})^{|n-n'|}\exp\left[i(\phi-\pi/2)(n-n')\right] L^{n-n'}_{\min(n,n')}\left(\rho^2/2\right), \quad (12)$$

where ρ is a two-dimensional in-plane wave vector written in polar coordinates $\rho = \sqrt{\rho_x^2+\rho_y^2}$, $\phi = \arctan(\rho_y/\rho_x)$, and $L^n_m(x)$ is the Laguerre polynomial. The function $K_{n,n'}(\rho)$ is independent of the Landau number for $n = n'$ and $\rho \to 0$. After integration over the z coordinate within the quantum well we found

$$\Phi(l_H q_\perp, m, N) = -\frac{\pi}{2}\left[\frac{1}{2}\delta_{m,2N} + \exp\left(-\frac{aq_\perp}{2}\right)\sinh\left(\frac{aq_\perp}{2}\right)\left(\frac{1}{aq_\perp/2} - \frac{aq_\perp/2}{\pi^2N^2+(aq_\perp/2)^2}\right)\right], \quad (13)$$

where $N = 1,2,3,\ldots$ and $m = 2,4,6,\ldots$.

The transverse component for the Raman phonon wave vector (see Fig. 1) must be equal to zero as follows from Eq. (11) (which gives $q_y = 0$ for $k_y = k'_y$) and from the sum over k_y applied to the same equation which gives $q_x = 0$. Taking into account that $K_{n,n}(0) = 1$ and evaluating the integral in Eq. (13) for $q_\perp = 0$ we find finally for the vertex of the electron (hole) -phonon interaction related to the Raman phonon

$$\sum_{k_y} R^{mN}_{nn}(k_y,0,0) = \mp C^*_{q_z}(L_xL_y/2\pi l_H^2)[\delta_{m,2N}+2] \quad (14)$$

with the upper (lower) sign for electrons (holes). Note that Eq. (14) is independent of n.

From now on we consider the range of laser frequencies which corresponds to either the incoming or the outgoing resonance for scattering via the state of an electron-hole pair with size-quantized level number N and Landau number n and neglect optical transitions to any other state in the well. It is also assumed that the strength of the magnetic field ensures resonant coupling of the electron state N,n with the electron state N,n' through electron-phonon interaction (magnetopolaron region). This implies that the following condition approximately holds:

$$\omega_{\rm LO} = (n-n')\omega_e. \quad (15)$$

The Green function for electrons is taken to be the solution of the Dyson equation with the self-energy $\Xi(N,n;\omega)$ calculated in the lowest approximation with the diagram shown in Fig. 1(c). For this calculation we restrict ourselves to the resonant electron transition $N,n \to N,n'$, assuming that the broadening of the electron state N,n' is determined by some other scattering mechanism which is approximated here by the constant value $\delta/2$,[10] a reasonable assumption provided the energy of the electron in the state N,n' is not enough for emission of LO phonons. Note that unequal effective masses guarantee the separation in magnetic field of polaron resonances for electrons and holes, allowing us to neglect the polaron effect for holes.

Evaluating the equation for the electron self-energy in the usual way we obtain

$$\Xi(N,n;\omega) = \sum_{q_z}\frac{\varepsilon(q_z)}{\omega-\omega_{\rm LO}(q_z)-\omega^e_N-(n'+\frac{1}{2})\omega_e+i\delta/2}, \quad (16)$$

$$\varepsilon(q_z) = \alpha\frac{32}{\pi^2}\frac{l_p}{a}[\hbar\omega_{\rm LO}(q_z)]^2 \times\frac{n}{n'}\int_0^\infty dx\frac{e^{-x}x^{n-n'}[L^{n-n'}_{n'}(x)]^2\Phi^2(x,m,N)}{x+q_z^2l_H^2}, \quad (17)$$

where $x = l_H^2q_\perp^2/2$. After substitution of Eqs. (16) and (17) into Eq. (7) the renormalized electron Green function [bold lines in Figs. 1(a) and 1(b)] reads

$$G(N,n;\omega) = (\omega-p)/[\omega-\omega_1(\varepsilon)][\omega-\omega_2(\varepsilon)], \quad (18)$$

where the two renormalized poles ω_1 and ω_2 corresponding to the new excitation branches are

$$\omega_{1,2}(\varepsilon) = (p+q)/2 \pm \sqrt{[(q-p)/2]^2+\varepsilon} \quad (19)$$

with

$$p = \omega_{\rm LO}(q_z)+\omega^e_N+(n'+\tfrac{1}{2})\omega_e - i\delta/2, \quad (20)$$

$$q = \omega^e_N+(n+\tfrac{1}{2})\omega_e - i\gamma_e/2. \quad (21)$$

We calculate the two contributions to the scattering amplitude using the renormalized Green functions:

$$A^e_{\alpha\gamma}(q_z) = -\left(\frac{e}{m_0}\right)^2\frac{L_xL_y}{2\pi\hbar l_H^2}\frac{C^*_{q_z}p_{cv\alpha}p^*_{cv\gamma}[\delta_{m,2N}+2][x-\omega_2(0)]}{[x-\omega_1(\varepsilon)][x-\omega_1(\varepsilon)-\omega_{\rm LO}(q_z)]}\frac{x-\omega_2(0)-\omega_{\rm LO}(q_z)}{[x-\omega_2(\varepsilon)][x-\omega_2(\varepsilon)-\omega_{\rm LO}(q_z)]}, \quad (22)$$

$$A^h_{\alpha\gamma}(q_z) = -\left(\frac{e}{m_0}\right)^2\frac{L_xL_y}{2\pi\hbar l_H^2}C^*_{q_z}p_{cv\alpha}p^*_{cv\gamma}[\delta_{m,2N}+2] \times\left(\frac{x-\omega_2(0)}{\omega_{\rm LO}(q_z)[x-\omega_1(\varepsilon)][x-\omega_2(\varepsilon)]} - \frac{x-\omega_2(0)-\omega_{\rm LO}(q_z)}{\omega_{\rm LO}(q_z)[x-\omega_1(\varepsilon)-\omega_{\rm LO}(q_z)][x-\omega_2(\varepsilon)-\omega_{\rm LO}(q_z)]}\right), \quad (23)$$

where

$$x = \omega_l-\omega_g-\omega^h_N-(n+\tfrac{1}{2})\omega_h+i\gamma_h/2. \quad (24)$$

It follows from Eqs. (22) and (23) that the electron and the hole contributions to the amplitude do not compensate each other exactly in the range of resonant coupling for Landau levels of the electron (or hole) states. Total compensation takes place only in the limit of $\varepsilon = 0$. Adding Eqs. (22) and (23) and using Eq. (1) we obtain for the efficiency of resonant scattering by confined optical phonon q_z via the state of the electron-hole pair with quantum numbers N,n,

$$S(q_z) = S_0(q_z)\tilde\varepsilon^2(q_z)/|\tilde x-\tilde\omega_1(\tilde\varepsilon)|^2|\tilde x-\tilde\omega_2(\tilde\varepsilon)|^2|\tilde x-\tilde\omega_1(\tilde\varepsilon)-1|^2|\tilde x-\tilde\omega_2(\tilde\varepsilon)-1|^2, \quad (25)$$

where

$$S_0(q_z) = (\omega_s/\omega_l)[n(\omega_s)/n(\omega_l)]|\mathbf{p}_{cv}\mathbf{e}_l|^2|\mathbf{p}_{cv}\mathbf{e}_s|^2(e^2/m_0c^2)^2 \times[4\omega_{0c}^2(\alpha l_p)/\omega_{\rm LO}^6(q_z)\pi^2m^2\hbar^8][\delta_{m,2N}+2]^2. \quad (26)$$

All variables with tildes are taken in units of $\omega_{\rm LO}(q_z)$.

To compare the total scattered intensity of Eq. (25) [diagrams (a) and (b) in Fig. 1] with the separate contribution of the electron part [diagram (a) in Fig. 1] we evaluate the value $t = S(q_z)/S^e(q_z)$ as a function of the Fröhlich constant α. Using Eq. (22) for calculating $S^e(q_z)$ we obtain

$$t \sim \varepsilon^2/|x-\omega_2(0)|^2|x-\omega_2(0)-\omega_{\rm LO}(q_z)|^2. \quad (27)$$

For split incoming resonance corresponding to the excitation of an electron in the vicinity of the anticrossing point [defined by $(n+\frac{1}{2})\omega_e = \omega_{\rm LO}(q_z) + (n'+\frac{1}{2})\omega_e$, i.e., $p \approx q$] we have $[x-\omega_2(0)] \sim \sqrt{\alpha}\omega_{\rm LO}$. Using as a rough estimation of Eq. (17) $\varepsilon \sim \alpha\omega_{\rm LO}^2$ we thus find $t \sim \alpha$. For laser frequencies and magnetic fields corresponding to the resonant excitation of the electron away from the anticrossing the value $[x-\omega_2(0)]$ rapidly increases; this leads to a strong decrease in t. The same arguments can be applied for outgoing resonance.

In Fig. 2 we show the results of calculations for the dimensionless cross section $S(q_z)/S_0(q_z)$ as function of the magnetic field for effective masses of electrons and heavy holes corresponding to GaAs $m_e = 0.068m_0$, $m_h = 0.49m_0$, for $\gamma_e = \gamma_h = \delta = 0.02\omega_{\rm LO}$ and $\varepsilon = 0.001\omega_{\rm LO}^2$. The set of curves illustrate the splitting of the incoming resonance for the electron-hole pair state in the Landau level $n = 2$ ($n' = 0$) at $(\omega_l - \omega_g - \omega_N^e - \omega_N^h) = 1.325, 1.375, 1.425, 1.475, 1.525\omega_{\rm LO}$ in the range of magnetic fields where $\omega_e \sim \omega_{\rm LO}/2$. Similar results can be obtained for the outgoing resonance, i.e., for laser frequencies one LO-phonon frequency higher than the one of Fig. 2.

To summarize, we have shown that the amplitude of the one-phonon resonant Raman scattering in quantum wells under a high magnetic field for Fröhlich interaction has a finite value in the range of resonant polaron coupling for electrons or for holes even in the case of infinite barriers and shows the split incoming and outgoing resonances as a function of laser frequency and magnetic field. The contribution to decompensation resulting from the different penetration of electron and hole wave functions into the barrier should be compared with decompensation induced by the polaron effect taking into account parameters of the quantum-well structure. For heavy holes in GaAs both effects add because of the smaller penetration of the hole wave functions into the barrier (the difference in the penetration for electrons and heavy holes is mainly determined by the masses whereas the difference in the barrier energy results in a smaller effect). In the case of light holes the penetration difference is dominated by barriers and the two contributions subtract.

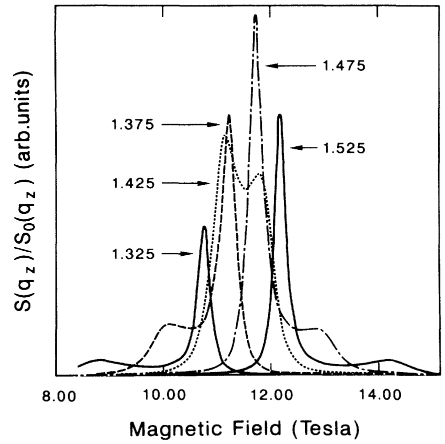

FIG. 2. The results of calculations for dimensionless value $S(q_z)/S_0(q_z)$ as function of the magnetic field in the range of polaron splitting of the incoming resonance for five values of the laser frequency (the numbers attached to the various curves represent the value of $\omega_l - \omega_g - \omega_N^e - \omega_N^h$ in units of $\omega_{\rm LO}$).

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