Magnetopolaron-induced increase of the efficiency in two-LO-phonon Raman scattering from quantum wells

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The magnetopolaron wave function and energy for resonantly coupled electronic and vibrational excitations in a quantum well are derived and used to evaluate the efficiency of Raman scattering by two LO phonons. A strong increase of the scattering intensity along with the splitting of Raman peaks is predicted in the magnetopolaron regime. [S0163-1829(96)03848-9]

I. INTRODUCTION

Raman scattering from semiconducting low-dimensional structures provides information on the spectrum of electronic and lattice excitations[1] and is used along with many other techniques for sample characterization. Over the past years the process of one-LO-phonon Raman scattering in quantum wells has attracted considerable attention from both experimentalists and theorists.[2–4] However, less attention was paid to the multiphonon processes because of difficulties in the measurement and the interpretation of the data. To our knowledge, there are only a few investigations of this kind.[5–14]

In this paper we develop a theoretical model for the magnetopolaron effect (Johnson-Larsen effect[15–17]) in two-phonon Raman scattering from semiconductor quantum-well structures. The Johnson-Larsen effect appears when the energy separation of a pair of Landau levels becomes close to the energy of an optical phonon

$$n\hbar\omega_{eH}\simeq\hbar\omega_{\mathrm{LO}}, \tag{1}$$

where $\omega_{eH}=|e|H/(m_e c)$, n is an integer, e is the charge of the electron, H is the magnetic field, m_e is the electron effective mass, and c is the speed of light. An initial investigation of the effect by Johnson and Larsen[15–17] was performed for bulk semiconductors. Korovin and Pavlov[18] have shown that in this case the magnetopolaron splitting is $\sim\alpha^{3/2}\hbar\omega_{\mathrm{LO}}$, where α is the Fröhlich[19] electron-phonon coupling constant (for a review see also Ref. 20). This estimate changes to $\sim\alpha^{1/2}\hbar\omega_{\mathrm{LO}}$ in the case of quantum wells.[21] The magnetopolaron effect in two-dimensional and quasi-two-dimensional systems was also investigated in Refs. 22–27.

The appearance of magnetopolaron effects in Raman scattering has been studied theoretically in Refs. 9–11 and 28–32. The corresponding splitting of a one-phonon Raman peak in bulk semiconductors was investigated in Refs. 28 and 29, both for Fröhlich and deformation potential interactions. An initial theoretical approach to two- and three-LO-phonon Raman scattering from quantum wells in the magnetopolaron regime was discussed in Refs. 9–11 under the approximation of infinitely high barriers and heavy holes $m_h\gg m_e$. The dipole-forbidden one-LO-phonon process was studied in Refs. 30 and 32.

In this paper we derive analytical expressions for the magnetopolaron wave function and apply the results to calculating the scattering efficiency of Raman scattering by two LO phonons. We show that the resonant coupling between pure electronic and mixed electron-phonon states results in a strong increase of the scattering intensity and a splitting of the Raman peaks. The paper is organized as follows. The model is given in Sec. II. The polaron wave function and energy are calculated in Sec. III for the case $\omega_{eH}\sim\omega_{\mathrm{LO}}$ and the results are used to calculate the two phonon scattering efficiency in Sec. IV. Section V is devoted to the discussion and the main conclusions of the work.

II. MODEL

Let us consider a semiconductor quantum well (QW) structure of type I with energy gap E_g and barriers ΔE_c and ΔE_v for electrons and holes, respectively. The magnetic field is directed perpendicular to the QW plane (in the z direction) and the corresponding vector potential is taken as

$$\mathbf{A}=\mathbf{A}(0,xH,0). \tag{2}$$

The width of the barriers is assumed to be large enough to neglect tunneling of electrons and holes outside the well. The laser frequency will be taken within the interval where only the discrete part of the QW electron and hole excitation spec-

trum is relevant for intermediate states in the two LO-phonon Raman scattering. The electron wave function is then given by

$$\Psi^e_{n,k_y,m}(x,y,z)=\Phi_n(x+a_H^2k_y)\frac{1}{\sqrt{L_y}}e^{ik_yy}\varphi^e_m(z), \quad (3)$$

where

$$\Phi_n(x)=\frac{1}{\sqrt{\pi^{1/2}2^n n!\,a_H}}H_n(x/a_H)\exp\left(-\frac{x^2}{2a_H^2}\right),$$

$$a_H=\sqrt{\frac{c\hbar}{eH}},$$

$H_n(t)$ is a Hermite polynomial, and $\varphi^e_m(z)$ the normalized electron wave function of the mth discrete state[33] in a one-dimensional QW, which can be written as

$$\varphi^e_m(z)=C_m\exp(\kappa_m z)\times\begin{cases}\cos(k_md/2), & m=1,3,5,\ldots\\ -\sin(k_md/2), & m=2,4,6,\ldots\end{cases}$$

for $z\leqslant 0$,

$$\varphi^e_m(z)=C_m\times\begin{cases}\cos[k_m(z-d/2)], & m=1,3,5,\ldots\\ \sin[k_m(z-d/2)], & m=2,4,6,\ldots\end{cases}$$

for $0\leqslant z\leqslant d$ and,

$$\varphi^e_m(z)=C_m\exp[-\kappa_m(z-d)]\times\begin{cases}\cos(k_md/2), & m=1,3,5,\ldots\\ \sin(k_md/2), & m=2,4,6,\ldots\end{cases} \quad (4)$$

for $z\geqslant d$, where d is the QW width,

$$C_m^2=\frac{2\kappa_m}{1+\kappa_m d\pm\cos(k_md)\pm(\kappa_m/k_m)\sin(k_md)}$$

is the normalization constant with upper (lower) sign for the odd (even) levels,

$$k_m=\sqrt{2m_e\varepsilon^e_m/\hbar^2},\quad \kappa_m=\sqrt{Q_e^2-k_m^2},\quad Q_e=\sqrt{2m_e\Delta E_c/\hbar^2},$$

and k_m for the mth level is found from the equations

$$\cot t=\frac{t}{\sqrt{\beta_e^2-t^2}},\quad \tan t=-\frac{t}{\sqrt{\beta_e^2-t^2}}$$

for odd and even states, respectively; $t=kd/2$ and $\beta_e=Q_ed/2$. There are no states for $t=0$, while the level $m=1$ always exists independent of the width and depth of the QW. The existence of all other levels is subject to the condition $k_m<Q_e$. The electron energy calculated at the bottom of each QW band is

$$E^e_{n,m}=(n+1/2)\hbar\omega_{eH}+\varepsilon^e_m. \quad (5)$$

Similarly, the wave function of the hole is

$$\Psi^h_{n,k_y,m}(x,y,z)=\Phi_n(x-a_H^2k_y)\frac{1}{\sqrt{L_y}}e^{ik_yy}\varphi^h_m(z), \quad (6)$$

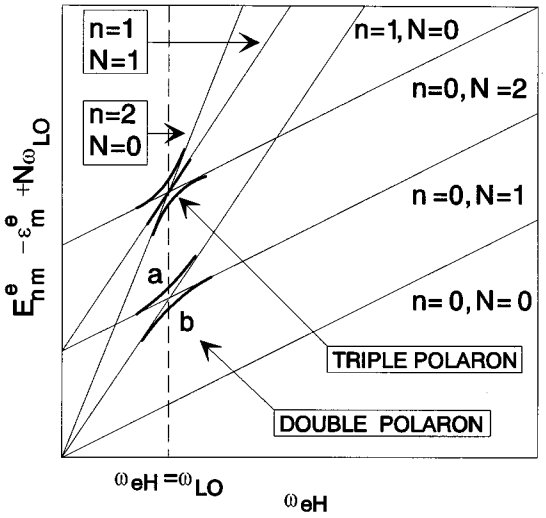

FIG. 1. Schematic representation of the energy states of the electron and phonon subsystems shown as a function of magnetic field (normalized to the cyclotron frequency of the conduction electrons). Double and triple polaron splitting appears in the vicinity of points where either two or three branches cross at $\omega_{eH}=\omega_{\rm LO}$, respectively. The splitting at $\omega_{eH}=\omega_{\rm LO}/2$ is not shown in the figure.

where $\varphi^h_m(z)$ can be obtained from the $\varphi^e_m(z)$ through the substitution $(m_e,\Delta E_c)\rightarrow(m_h,\Delta E_v)$. The energy of the hole is

$$E^h_{n,m}=E_g+(n+1/2)\hbar\omega_{hH}+\varepsilon^h_m,\quad \omega_{hH}=\frac{eH}{m_hc}. \quad (7)$$

III. POLARON WAVE FUNCTION

Let us assume that the magnetic-field strength lies in the interval where $\omega_{eH}\simeq\omega_{\rm LO}$. We do not take into account the phonon dispersion and broadening and we assume that the energy separation of different phonon modes is larger than the polaron splitting so that in the theory we only have to consider one phonon mode at a time. The energy separation of size quantized levels in the electronic subsystem is assumed to be larger than both $\hbar\omega_{\rm LO}$ and $\hbar\omega_{eH}$, which allows us to consider only one specific QW level. The energy spectrum of the system consisting of (i) one electron in the Landau level n corresponding to some size quantized state m and (ii) N LO phonons is shown in Fig. 1, where the origin of electron energies is taken to be ε^e_m. The electron-phonon interaction results in the splitting of degenerate states at all crossing points and double, triple, and higher polarons appear in the energy spectrum. In this paper we are interested mainly in the effects related to the degeneracy of the two states $(n=1,\ N=0)$ and $(n=0,\ N=1)$ at the magnetic field $\omega_{eH}=\omega_{\rm LO}$. For the polaron wave function we use the ansatz

$$\Theta(x,y,z,b)|0\rangle=\sum_{k_y}a_0(k_y)\Psi_{1,k_y}(x,y,z)|0\rangle + \sum_{k_y,\nu}a_1(k_y,\nu)\Psi_{0,k_y}(x,y,z)b^\dagger_\nu|0\rangle, \quad (8)$$

where the superscripts (subscripts) e (m) of the electron wave functions have been omitted, $b_\nu^\dagger$ (b_ν) is the creation (destruction) operator for the phonon ν, and $|0\rangle$ is the phonon vacuum. The first and the second terms in the wave function of Eq. (8) correspond to the states $N=0$ and $N=1$, respectively. The index ν labels the phonon states and consists of the quantum numbers $\mathbf{q}_\perp$ and j, where $\mathbf{q}_\perp$ is the in-plane wave-vector component and j corresponds to the phonon size quantization. In the limit of three-dimensional phonons the index j has to be substituted by the wave-vector component q_z. The approximation of dispersionless phonons is well justified for the quasi-two-dimensional modes of GaAs–AlAs structures when $d>50$ Å. Our approach is valid for wide enough QW's, where interface phonon effects are not important in the polaron regime. Their role in the magnetopolaron effect in narrow QW's has been studied in Ref. 24.

Let us substitute Eq. (8) into the Schrödinger equation

$$H\Theta = E\Theta, \tag{9}$$

where

$$H = H_0 + V^e, \quad H_0 = H_e + H_{\rm ph}, \tag{10}$$

H_e ($H_{\rm ph}$) is the Hamiltonian of electrons (phonons), and V^e is the electron-phonon Fröhlich interaction. It is easy to see that

$$\begin{aligned} H_e\Psi_{0,k_y} &= [\varepsilon_m^e + (1/2)\hbar\omega_{eH}]\Psi_{0,k_y}, \\ H_e\Psi_{1,k_y} &= [\varepsilon_m^e + (3/2)\hbar\omega_{eH}]\Psi_{1,k_y}, \\ H_{\rm ph}|0\rangle &= 0, \quad H_{\rm ph}b_\nu^\dagger|0\rangle = \hbar\omega_{\rm LO}b_\nu^\dagger|0\rangle. \end{aligned} \tag{11}$$

From here on, the electron energy is calculated with the energy ε_m^e as the origin and the notation $\Sigma_0 \equiv \hbar\omega_{eH}/2$, $\Sigma_1 \equiv 3\hbar\omega_{eH}/2$ is used.

We take the interaction V^e in a general form

$$V^e = \sum_\nu \left[C_\nu^e(\mathbf{r}_\perp, z)b_\nu + C_\nu^{e*}(\mathbf{r}_\perp, z)b_\nu^\dagger\right], \tag{12}$$

where

$$C_\nu(\mathbf{r}_\perp, z) = C_q e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}\xi_\nu(z),$$

with $\xi_\nu(z) = \exp(iq_z z)$ for three-dimensional and $\xi_\nu(z) = \eta_{q_\perp, j}(z)$ for quasi-two-dimensional phonons. For Fröhlich interaction[19] with three-dimensional phonons

$$\begin{aligned} C_q &= -i\hbar\omega_{\rm LO}\left(\frac{4\pi\alpha l^3}{V_0}\right)^{1/2}\frac{1}{ql}, \quad l = \sqrt{\frac{\hbar}{2m_e\omega_{\rm LO}}}, \\ \alpha &= \frac{e^2}{2\hbar\omega_{\rm LO}l}(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}), \end{aligned} \tag{13}$$

where V_0 is the normalization volume and ε_0 (ε_∞) the static (high-frequency) dielectric constant. The actual form of the function $\eta_{q_\perp, i}(z)$ can be found in Refs. 34–36.

To calculate the double polaron wave function (see Fig. 1) and the corresponding energy splitting, we take into account only the coupling between two states: ($n=0$, $N=1$) and ($n=1$, $N=0$). All other possible transitions lead to corrections of higher order in α when $\omega_{eH} \simeq \omega_{\rm LO}$.[22,23] Therefore

$$V^e b_\nu^\dagger|0\rangle \simeq C_\nu(\mathbf{r}_\perp, z)|0\rangle \tag{14}$$

and

$$V^e|0\rangle = \sum_\nu C_\nu^*(\mathbf{r}_\perp, z)b_\nu^\dagger|0\rangle. \tag{15}$$

The derivation of the polaron wave function and energy is formally the same for three- and quasi-two-dimensional phonons. Using Eqs. (8), (11), (14), and (15), the Schrödinger equation may be written as

$$\begin{aligned} &(E-\Sigma_1)\sum_{k_y} a_0(k_y)\Psi_{1,k_y}|0\rangle + (E-\Sigma_0-\hbar\omega_{\rm LO}) \\ &\times\sum_{k_y}\Psi_{0,k_y}\sum_\nu a_1(k_y,\nu)b_\nu^\dagger|0\rangle \\ &-\sum_{k_y} a_0(k_y)\Psi_{1,k_y}\sum_\nu C_\nu^*(\mathbf{r}_\perp, z)b_\nu^\dagger|0\rangle \\ &-\sum_{k_y}\Psi_{0,k_y}\sum_\nu a_1(k_y,\nu)C_\nu(\mathbf{r}_\perp, z)|0\rangle \\ &= 0. \end{aligned} \tag{16}$$

Let us multiply the above equation on the left-hand side by $\langle 0|$ and $\langle 0|b_{\nu'}$. Making use of the orthonormality of the phonon states, we arrive at the system of equations

$$\begin{aligned} &(E-\Sigma_1)\sum_{k_y} a_0(k_y)\Psi_{1,k_y} - \sum_{k_y}\Psi_{0,k_y} \\ &\times\sum_\nu a_1(k_y,\nu)C_\nu(\mathbf{r}_\perp, z) = 0, \\ &(E-\Sigma_0-\hbar\omega_{\rm LO})\sum_{k_y} a_1(k_y,\nu)\Psi_{0,k_y} - \sum_{k_y} a_0(k_y) \\ &\times\Psi_{1,k_y}C_\nu^*(\mathbf{r}_\perp, z) = 0. \end{aligned} \tag{17}$$

In order to eliminate the dependence on the electron coordinates, we multiply the first equation by $\Psi_{1,k_y'}^*$ and the second by $\Psi_{0,k_y'}^*$ and integrate over $\mathbf{r}$. The following matrix element appears

$$\begin{aligned} &\int d^3r\Psi_{0,k_y'}^* C_\nu^*\Psi_{1,k_y} \\ &= \delta_{k_y', k_y-q_y}\exp\left(\frac{i}{2}a_H^2 q_x(k_y+k_y')\right)U_{10}^*(\nu), \end{aligned} \tag{18}$$

where

$$U_{10}^{*}(\nu)\equiv U^{*}(\nu)=\mathcal{M}^{*}(\nu)C_q^{*}K_{10}(a_Hq_y,-a_Hq_x),$$

$$K_{nm}(\mathbf{p})=K_{nm}(p_x,p_y)$$

$$=\left[\frac{\min(n!,m!)}{\max(n!,m!)}\right]^{1/2} i^{|n-m|}\left(\frac{p}{\sqrt{2}}\right)^{|n-m|}\exp\left(-\frac{p^2}{4}\right)$$

$$\times\exp[i(\phi-\pi/2)(n-m)]L_{\min(n,m)}^{|n-m|}(p^2/2), \qquad (19)$$

and

$$\mathcal{M}_{2D}(q_\perp,j)=\int_{-\infty}^{\infty}dz[\varphi_m(z)]^2\eta_{q_\perp,j}(z),$$

$$\mathcal{M}_{3D}(q_z)=\int_{-\infty}^{\infty}dz[\varphi_m(z)]^2\exp(iq_zz) \qquad (20)$$

for two- and three-dimensional phonons, respectively; $\mathbf{p}$ is a two-dimensional vector, $p=\sqrt{p_x^2+p_y^2}$, $\phi=\arctan(p_y/p_x)$, and $L_m^n(t)$ is the Laguerre polynomial. Using Eq. (18) we arrive at the system of equations

$$(E-\Sigma_1)a_0(k_y)-\sum_\nu a_1(k_y-q_y,\nu)$$

$$\times\exp[-ia_H^2q_x(k_y-q_y/2)]U(\nu)=0,$$

$$(E-\Sigma_0-\hbar\omega_{\rm LO})a_1(k_y,\nu)-a_0(k_y+q_y)$$

$$\times\exp[ia_H^2q_x(k_y+q_y/2)]U^{*}(\nu)=0. \qquad (21)$$

Finally, substitution of the coefficient $a_1(k_y,\nu)$ from the second equation in Eq. (21) into the first one results in the equation for the polaron energy

$$E-\Sigma_1-\frac{A^2}{E-\Sigma_0-\hbar\omega_{\rm LO}}=0, \qquad (22)$$

where

$$A^2=\sum_\nu|U(\nu)|^2. \qquad (23)$$

Equation (22) has two solutions

$$E_{a,b}=\frac{\Sigma_1+\Sigma_0+\hbar\omega_{\rm LO}}{2}\pm\sqrt{\left(\frac{\Sigma_1-\Sigma_0-\hbar\omega_{\rm LO}}{2}\right)^2+A^2}, \qquad (24)$$

where the indices a and b correspond to the upper and lower signs, respectively (see Fig. 1). Using the result of Eq. (24), we find the polaron wave function

$$\Theta_p|0\rangle=\sum_{k_y}a_{0p}(k_y)\left[\Psi_{1,k_y}+\sum_\nu\frac{\exp[ia_H^2q_x(k_y-q_y/2)]}{E_p-\Sigma_0-\hbar\omega_{\rm LO}}\right.$$

$$\left.\times U^{*}(\nu)\Psi_{0,k_y-q_y}b_\nu^{\dagger}\right]|0\rangle, \qquad (25)$$

where p is a or b. Direct calculation shows that wave functions of the two branches are orthogonal $\int d^3r\Theta_b^{*}\Theta_a=0$. From the normalization condition $\int d^3r\Theta_p^{*}\Theta_p=1$ we find that

$$\sum_{k_y}|a_{0p}(k_y)|^2=\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\rm LO})^2}\right]^{-1}. \qquad (26)$$

Let us choose the coefficient $a_{0p}(k_y)$ as

$$a_{0p}(k_y)=\delta_{k_y,k_y'}\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\rm LO})^2}\right]^{-1/2}, \qquad (27)$$

so that the polaron wave functions are characterized by the quantum number k_y. Substitution of Eq. (27) in Eq. (25) results in the final expression for the polaron wave functions

$$\Theta_{pk_y}|0\rangle=\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\rm LO})^2}\right]^{-1/2}$$

$$\times\left[\Psi_{1,k_y}+\sum_\nu\frac{\exp[ia_H^2q_x(k_y-q_y/2)]}{E_p-\Sigma_0-\hbar\omega_{\rm LO}}\right.$$

$$\left.\times U^{*}(\nu)\Psi_{0,k_y-q_y}b_\nu^{\dagger}\right]|0\rangle, \qquad (28)$$

which are orthonormal $\int d^3r\Theta_{p',k_y'}^{*}\Theta_{p,k_y}=\delta_{p,p'}\delta_{k_y,k_y'}$. Under conditions of exact resonance $\omega_{eH}=\omega_{\rm LO}$ we have $E_{a,b}^{\rm res}=\Sigma_1\pm A$ and the splitting $\Delta E_{\rm res}=E_a-E_b=2A$ is proportional to $\sqrt{\alpha}$, a characteristic feature of the quasi-two-dimensional systems, as has been shown by Korovin *et al.* in Ref. 21 (see also Refs. 22 and 23). According to the calculations of Ref. 24, the magnetopolaron splitting in GaAs-AlAs QW's is $\sim 0.2\hbar\omega_{\rm LO}$. In the limit $k_y=0$ the wave function of Eq. (28) is reduced to the one in Eq. (15) of Ref. 23, where the two-phonon term has also been taken into account. Note that the two-phonon contribution is not important for the calculation of the magnetopolaron splitting to the lowest order in α.

We proceed to estimate the magnetopolaron splitting $\Delta E=2A$ for Fröhlich interaction with three-dimensional LO phonons. Using Eqs. (19) and (13) we obtain

$$\Delta E_{3D}=2\hbar\omega_{\rm LO}\sqrt{\frac{\alpha l}{\pi}}\left[\int_0^{\infty}dx x\exp(-x)\right.$$

$$\left.\times\int_{-\infty}^{\infty}dq_z\frac{|\mathcal{M}_{3D}(q_z)|^2}{2x+a_H^2q_z^2}\right]^{1/2}, \qquad (29)$$

where $x=a_H^2q_\perp^2/2$. For quasi-two-dimensional phonons

$$\Delta E_{2D}=2\left[\frac{S}{(2\pi)^2}\int d\mathbf{q}_\perp\exp(-a_H^2q_\perp^2/2)\frac{a_H^2q_\perp^2}{2}\right.$$

$$\left.\times\sum_j|C_q|^2|\mathcal{M}_{2D}(q_\perp,j)|^2\right]^{1/2}, \qquad (30)$$

where S is the normalization area. The polaron wave function under conditions of exact resonance can be written as

$$\Theta_{pk_y}(\omega_{eH}=\omega_{\rm LO})|0\rangle$$

$$=\frac{1}{\sqrt{2}}\left[\Psi_{1,k_y}\pm\frac{1}{A}\sum_{\nu}\exp[ia_H^2q_x(k_y-q_y/2)]\right.$$

$$\left.\times U^*(\nu)\Psi_{0,k_y-q_y}b_\nu^\dagger\right]|0\rangle. \quad (31)$$

Since both A and U^* are of order $\sqrt{\alpha}$, the two component $(n=1,\ N=0)$ and $(n=0,\ N=1)$ are equally represented in the polaron state.

Finally, we give below the expressions for the magnetopolaron energy and wave function far from the resonance where

$$A\ll\hbar|\omega_{eH}-\omega_{\rm LO}|. \quad (32)$$

The results are different for lower (left) and higher (right) magnetic fields (see Fig. 1). In particular,

$$E_{a,r}=E_{b,l}\simeq\frac{3}{2}\hbar\omega_{eH}+\frac{A^2}{\hbar(\omega_{eH}-\omega_{\rm LO})},$$

$$\Theta_{ak_y,r}|0\rangle=\Theta_{bk_y,l}|0\rangle$$

$$\simeq\left[\Psi_{1,k_y}+\sum_{\nu}\frac{\exp[ia_H^2q_x(k_y-q_y/2)]}{\hbar(\omega_{eH}-\omega_{\rm LO})}\right.$$

$$\left.\times U^*(\nu)\Psi_{0,k_y-q_y}b_\nu^\dagger\right]|0\rangle; \quad (33)$$

$$E_{a,l}=E_{b,r}\simeq\frac{1}{2}\hbar\omega_{eH}+\hbar\omega_{\rm LO}+\frac{A^2}{\hbar(\omega_{\rm LO}-\omega_{eH})},$$

$$\Theta_{ak_y,l}|0\rangle=\Theta_{bk_y,r}|0\rangle$$

$$\simeq\left[\frac{A\Psi_{1,k_y}}{\hbar(\omega_{\rm LO}-\omega_{eH})}+\frac{1}{A}\sum_{\nu}\exp[ia_H^2q_x(k_y-q_y/2)]\right.$$

$$\left.\times U^*(\nu)\Psi_{0,k_y-q_y}b_\nu^\dagger\right]|0\rangle. \quad (34)$$

Equations (34) coincide with the results of the lowest order perturbation theory when only $(n=0,\ N=1)\rightarrow(n=1,\ N=0)$ transitions are taken into account. The states $\Theta_{ak_y,r}$ and $\Theta_{bk_y,l}$ are formed mainly by the state $n=1,\ N=0$ with a small admixture of states $n=0,\ N=1$, whereas the opposite is true for the states $\Theta_{ak_y,l}$ and $\Theta_{bk_y,r}$. The energy corrections are of first order in α and corrections to the wave functions are of the order $\sqrt{\alpha}$. However, these results are not accurate[23] since all corrections to the wave functions and energy of the same order in α related to transitions into other states of the system have been neglected.

IV. TWO-PHONON RAMAN SCATTERING

In order to calculate the scattering efficiency we use the connection between the light-scattering tensor[37,38] $S_{\alpha\gamma\beta\lambda}$ and the normalized probability $\overline{W_s}$ per one incident photon l to emit the quantum s of scattered light:

$$e_{s\alpha}^*e_{s\beta}e_{l\gamma}e_{l\lambda}^*S_{\alpha\gamma\beta\lambda}=\frac{V_0c^2n_ln_s}{(2\pi)^3u_lu_s\omega_s\omega_l}\overline{W_s}, \quad (35)$$

where ω_l, $\mathbf{e}_l$, n_l, and u_l (ω_s, $\mathbf{e}_s$, n_s, and u_s) are the frequency, polarization, refractive index, and group velocity of incident (scattered) light. Using Fermi's golden rule,

$$\overline{W_s}=\frac{2\pi}{\hbar}\sum_f|\langle f|\mathcal{H}|i\rangle|^2\delta(E_i-E_f), \quad (36)$$

$$\langle f|\mathcal{H}|i\rangle=\sum_{\beta,\gamma}\frac{\langle f|U_s|\gamma\rangle\langle\gamma|V|\beta\rangle\langle\beta|U_l|i\rangle}{(E_i-E_\beta)(E_i-E_\gamma)}. \quad (37)$$

In Eq. (37), U_l (U_s) corresponds to the interaction of electronic subsystem with incident (scattered) light. The ground-state wave function of the crystal can be written as

$$\Psi_i=\delta(\mathbf{r}_1-\mathbf{r}_2)|0\rangle, \quad (38)$$

where $\mathbf{r}_1$ ($\mathbf{r}_2$) is a coordinate of electron (hole). Similarly, the wave function of the final state in the process of Raman scattering by two LO phonons is

$$\Psi_f=\delta(\mathbf{r}_1-\mathbf{r}_2)b_{\nu_1}^\dagger b_{\nu_2}^\dagger|0\rangle. \quad (39)$$

The matrix elements of U_l and U_s in Eq. (37) include the constants M_l and M_s:

$$M_{l(s)}=-\left(\frac{\pi\hbar^{1/2}}{V_0}\right)\frac{e}{m_0}(\mathbf{e}_{l(s)}\mathbf{p}_{cv})\left(\frac{u_{l(s)}}{cn_{l(s)}\omega_{l(s)}}\right)^{1/2}, \quad (40)$$

where $\mathbf{p}_{cv}$ is the interband matrix element of the momentum operator. The interaction of the electronic subsystem with phonons $V=V^e+V^h$ has been defined in Eq. (12), where one has to substitute $C_q^h=-C_q^e$ for holes.

Since we are interested only in the process of resonant two-LO-phonon Raman scattering involving a double magnetopolaron (see Fig. 1), a constrain on the incident light frequency ω_l has to be imposed so that the direct creation of the real triple and higher polaron states will be impossible. In the vicinity of $\omega_{eH}=\omega_{\rm LO}$ this results in the condition

$$\hbar\omega_l<\hbar\omega_{l0}=E_g+\varepsilon^e+\varepsilon^h+(5/2)\hbar\omega_{eH}+(5/2)\hbar\omega_{hH}. \quad (41)$$

It will be shown below that both incoming and outgoing resonances in two-LO-phonon processes are formed with the participation of only double polaron states when $\omega_l<\omega_{l0}$. We also assume that $m_h>m_e$.

All calculations below are carried out in terms of magnetopolaron states. However, the selection of important contributions to the scattering efficiency seems to be easier when the scattering process is considered on the basis of bare electronic excitations. In Fig. 2 we show four diagrams for the scattering amplitude that give the main contribution for $\omega_{eH}\simeq\omega_{\rm LO}$. In the diagrams, solid lines above (below) the dash-dotted line correspond to electrons (holes), dashed lines to phonons, and dotted lines represent photons. Only the Landau number of electronic excitations are shown. Vertical sections 1, 2, and 3 correspond to the three intermediate states. At first, let us neglect the polaron effect. This is the same as neglecting the dressing of the electronic propagators $n=1$, which occurs through the resonant transitions into the

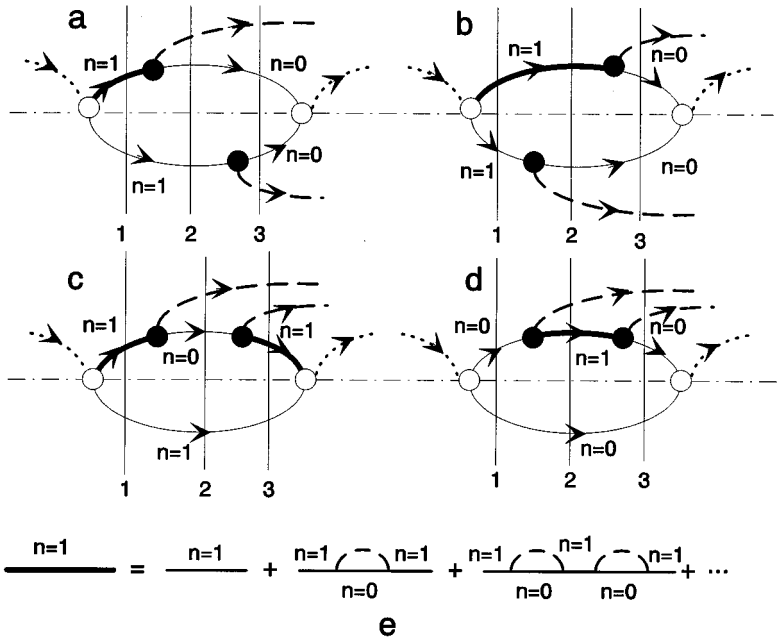

FIG. 2. Diagrams for two-LO-phonon Raman scattering amplitude in the basis of bare excitations. The vertical sections 1, 2, and 3 indicate intermediate states. The diagrams are selected in such a way that all of them describe contributions with two real intermediate states: 1 and 2 for processes with direct creation (a and c) and 2 and 3 for processes with direct recombination (b and d). The polaron effect is taken into account through dressing (e) the electron lines $n=1$ by resonant transitions to the state $n=0$ with emission of LO phonons.

state $n=0$ with emission of LO phonons (see e in Fig. 2). For *bare* electronic propagators it is easy to see that for $\omega_{eH}=\omega_{\mathrm{LO}}$ the states 1 and 2 in diagram a of Fig. 2 can be simultaneously real while the state 3 is then virtual, which corresponds to the doubly resonant process with indirect recombination. For the process b in Fig. 2, the states 2 and 3 can be real while the state 1 virtual, corresponding to the process with indirect creation. Similarly, the diagrams of c and d of Fig. 2 show the doubly resonant processes of incoming and outgoing character, respectively. Thus we have selected all of the most important contributions. Now taking into account the infinite series of the diagrams in e in Fig. 2 with the help of the Dyson equation, one can calculate the scattering amplitude for two-LO-phonon Raman scattering in the polaron regime. This approach has been used in Refs. 9–11, 28, 29, and 32. Note that in this case all vertices of interaction with light (empty circles) and phonons (filled circles) are calculated with the wave functions of bare electrons and holes from Eqs. (3) and (6), respectively.

Another equivalent way to do the calculation is to work in the polaron basis, which is assumed in Eq. (37). Let us define the intermediate states β and γ in Eq. (37). All important contributions to the scattering amplitude are shown in Fig. 3. The double solid lines in the diagrams represent polaron propagators. The empty squares correspond to the interaction with light. Note that creation of a polaron and a hole through the absorption of the incident light quantum occurs via coupling to the pure electronic component of the polaron wave function in Eq. (28). However, the process of polaron-hole recombination can take place in two different ways: depend-

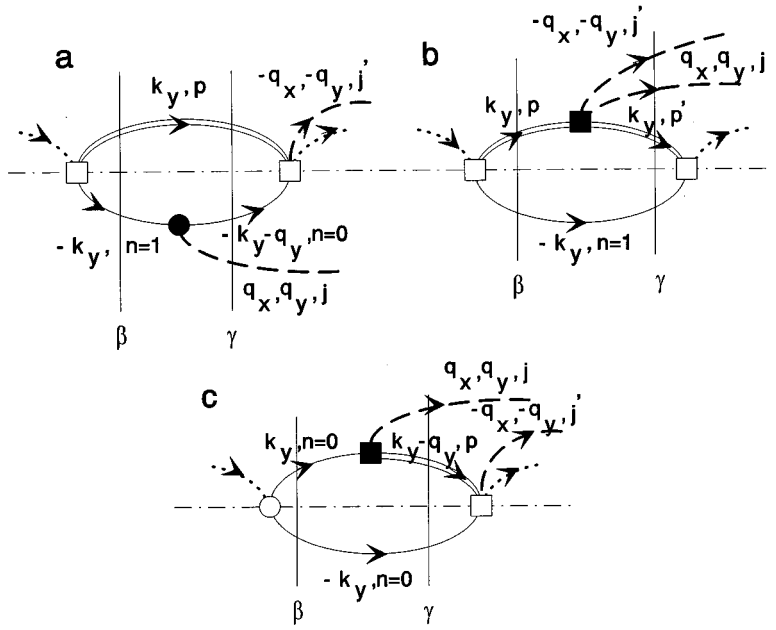

FIG. 3. Same as in Fig. 2, but represented in the polaron basis. Empty and filled squares correspond to the vertices of interaction with light and phonons, respectively. The corresponding expressions have to be evaluated with the wave functions of a double polaron shown by the double lines. There are two intermediate states β and γ instead of the three states in Fig. 2.

ing on the electronic component of the polaron wave function that couples to the hole, the recombination results in a scattered photon alone or accompanied by one phonon, as shown in (b) and in (a) and (c), respectively, of Fig. 3.

It is worth noting that the electron-phonon interaction can be separated into two parts: resonant and nonresonant. We call the interaction resonant if it leads to *real* transitions of the electron with emission of a LO phonon. For $\omega_{eH}=\omega_{\mathrm{LO}}$ this means that the resonant part of interaction is represented by matrix elements $\langle n-1|C_{\nu}^{e*}|n\rangle$ from Eq. (12), where n is the Landau number. This part has been taken into account exactly in the calculation of the polaron states. The rest of the interaction (nonresonant) can result in the polaron-polaron transitions with emission of LO phonons. In the diagrams of Fig. 3, the vertices of the nonresonant polaron-phonon interaction are shown as filled squares. Note that it is precisely the nonresonant character of all transitions with phonons involving polaron states that allows us to calculate the scattering amplitude via perturbation theory, whereas the infinite series of contributions has to be taken into account for resonant transitions of e in Fig. 2.

We proceed to calculate the scattering amplitude taking into account the three contributions of Fig. 3, which are equivalent to the four diagrams of Fig. 2 with the dressed lines representing the electronic state $n=1$. Let us consider each contribution separately.

(i) Diagram a in Fig. 3 is equivalent to the sum of the two diagrams a and b and in Fig. 2. In the polaron basis the state β in Eq. (37) consists of the polaron (p,k_y) and the hole (n',k_y'), and the matrix element of transition $(i\rightarrow\beta)$ is given by

$$\langle\beta|U_l|i\rangle=M_l\Pi\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^2}\right]^{-1/2}\delta_{k_y',-k_y}\delta_{n',1}, \tag{42}$$

where

$$\Pi=\int_{-\infty}^{\infty}dz\,\varphi^e(z)\,\varphi^h(z)$$

and $\varphi(z)$ has been defined in Eq. (4). The transition $\beta\rightarrow\gamma$ consists in the emission of a phonon $\nu\equiv(q_x,q_y,j)$ by the hole that ends in the state (n'',k_y'),

$$\langle\gamma|V|\beta\rangle=P_{\nu}^{h*}K_{n',n''}(-a_Hq_y,-a_Hq_x)\times\exp[-ia_Hq_x(k_y'-q_y/2)]\,\delta_{k_y'',k_y'-q_y}, \tag{43}$$

where

$$P_{\nu}^{h(e)}=C_q^{h(e)}\mathcal{M}^{h(e)}(\nu). \tag{44}$$

Finally, the transition $\gamma\rightarrow f$ may be represented as

$$\langle f|U_s|\gamma\rangle=\langle f|U_s|\gamma\rangle_1+\langle f|U_s|\gamma\rangle_2, \tag{45}$$

where

$$\langle f|U_s|\gamma\rangle_1=\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^2}\right]^{-1/2}\times\frac{M_s\Pi\,\delta_{-k_y'',k_y-q_{1y}}\delta_{n'',0}\delta_{\nu\nu_2}}{E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}}}P_{\nu_1}^{e*}K_{10}(a_Hq_{1y},-a_Hq_{1x})\times\exp[ia_H^2q_{1x}(k_y-q_{1y}/2)], \tag{46}$$

and $\langle f|U_s|\gamma\rangle_2$ differs from $\langle f|U_s|\gamma\rangle_1$ by the substitution $\nu_2\rightarrow\nu_1$. Evaluating sums over $\beta\equiv(p,k_y,n',k_y')$ and $\gamma\equiv(n'',k_y',\nu)$ in Eq. (37), we find

$$\langle f|\mathcal{H}|i\rangle_{\mathrm{I}}=\mathcal{H}_{\mathrm{I}1}+\mathcal{H}_{\mathrm{I}2}, \tag{47}$$

where

$$\mathcal{H}_{\mathrm{I}1}=\mathcal{B}(\omega_l,H)M_lM_s\Pi^2\frac{L_xL_y}{2\pi a_H^2}P_{\nu_1}^{e*}K_{10}(a_Hq_{1y},-a_Hq_{1x})\times P_{\nu_2}^{h*}K_{10}(-a_Hq_{2y},-a_Hq_{2x})\,\delta_{\mathbf{q}_{1\perp}+\mathbf{q}_{2\perp},0}, \tag{48}$$

$$\mathcal{B}(\omega_l,H)=\sum_p\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^2}\right]^{-1}\times[\hbar\omega_l-E_g-\varepsilon^e-\varepsilon^h-E_p-(3/2)\hbar\omega_{hH}]^{-1}\times(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^{-1}[\hbar\omega_l-E_g-\varepsilon^e-\varepsilon^h-E_p-(1/2)\hbar\omega_{hH}-\hbar\omega_{\mathrm{LO}}]^{-1}, \tag{49}$$

and $\mathcal{H}_{I2}$ differs from $\mathcal{H}_{I1}$ by the substitution $\nu_1\leftrightarrow\nu_2$. We have used also the relation

$$\sum_{k_y}\exp[ia_H^2k_y(q_{1x}+q_{2x})]=\frac{L_xL_y}{2\pi a_H^2}\delta_{q_{1x}+q_{2x},0}. \tag{50}$$

Note that $\mathcal{B}(\omega_l,H)$ contains two energy denominators depending on the incident light frequency ω_l. They correspond to the incoming and outgoing resonances, each one being split into two peaks. The factor $(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^{-1}\sim A^{-1}$ for $\omega_{eH}\sim\omega_{\mathrm{LO}}$ leads to an increase of the scattering efficiency.

All the selection rules that follow from the matrix elements of Eqs. (42), (43), and (46) have been taken into account when drawing diagram a of Fig. 3. The two contributions in Eq. (47) correspond to the exchange of all three quantum numbers $(q_x,q_y,j)\leftrightarrow(-q_x,-q_y,j')$ between the two phonons in the diagram. Such substitution is necessary in order to take into account the interference effects in the scattering efficiency. Thus the three diagrams of Fig. 3 have to be complemented by three additional diagrams with exchanged phonon indices.

(ii) This is the case of the diagrams in b in Fig. 3 and c in Fig. 2. In this case both phonons in a scattering process are emitted by the polaron. The state β is the same as in case (i). However, in the state γ there are two phonons, a polaron, and a hole.

The final expressions for case (ii) becomes

$$\langle f|\mathcal{H}|i\rangle_{\mathrm{II}}=\mathcal{H}_{\mathrm{II}1}+\mathcal{H}_{\mathrm{II}2}, \tag{51}$$

where

$$\mathcal{H}_{\mathrm{II}1}=Z(\omega_l,H)M_lM_s\Pi^2\frac{L_xL_y}{2\pi a_H^2}P^{e*}_{\nu_1}K_{10}(a_Hq_{1y},-a_Hq_{1x})\times P^{e*}_{\nu_2}K_{01}(a_Hq_{2y},-a_Hq_{2x})\delta_{\mathbf{q}_{1\perp}+\mathbf{q}_{2\perp},0}\times\sum_{p'}\left[1+\frac{A^2}{(E_{p'}-\Sigma_0-\hbar\omega_{\mathrm{LO}})^2}\right]^{-1}\times[\hbar\omega_l-E_g-\varepsilon^e-\varepsilon^h-E_{p'}-(3/2)\hbar\omega_{hH}-2\hbar\omega_{\mathrm{LO}}]^{-1},\tag{52}$$

$$Z(\omega_l,H)=\sum_p\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^2}\right]^{-1}\times(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^{-1}\times[\hbar\omega_l-E_g-\varepsilon^e-\varepsilon^h-E_p-(3/2)\hbar\omega_{hH}]^{-1},\tag{53}$$

and $\mathcal{H}_{\mathrm{II}2}$ can be obtained from $\mathcal{H}_{\mathrm{II}1}$ through the exchange of the phonon indices. The last energy denominator in Eq. (53) describes an incoming resonance. It is clear from the last denominator in Eq. (52) that outgoing resonance from this contribution is impossible when $\omega_l<\omega_{l0}$. We can then substitute $E_{p'}\rightarrow(3/2)\hbar\omega_{eH}$ and use the fact that $\Sigma_{p'}[\ ]^{-1}=1$.

(iii) In this case state β consists of the electron and the hole in the Landau level $n=0$ with the opposite sign of k_y, as shown in *c* Fig. 3. In state γ there is a phonon emitted by the electron, a polaron, and a hole. This contribution corresponds to diagram *d* in Fig. 2. We find

$$\langle f|\mathcal{H}|i\rangle_{\mathrm{III}}=\mathcal{H}_{\mathrm{III}1}+\mathcal{H}_{\mathrm{III}2},\tag{54}$$

$$\mathcal{H}_{\mathrm{III}1}=X(\omega_l,H)M_lM_s\Pi^2\frac{L_xL_y}{2\pi a_H^2}P^{e*}_{\nu_1}K_{01}(a_Hq_{1y},-a_Hq_{1x})\times P^{e*}_{\nu_2}K_{10}(a_Hq_{2y},-a_Hq_{2x})\delta_{\mathbf{q}_{1\perp}+\mathbf{q}_{2\perp},0}\times[\hbar\omega_l-E_g-\varepsilon^e-\varepsilon^h-(1/2)\hbar\omega_{eH}-(1/2)\hbar\omega_{hH}]^{-1},\tag{55}$$

$$X(\omega_l,H)=\sum_p\left[1+\frac{A^2}{(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^2}\right]^{-1}\times(E_p-\Sigma_0-\hbar\omega_{\mathrm{LO}})^{-1}\times[\hbar\omega_l-E_g-\varepsilon^e-\varepsilon^h-E_p-(1/2)\hbar\omega_{hH}-\hbar\omega_{\mathrm{LO}}]^{-1}.\tag{56}$$

The last energy denominator corresponds to the outgoing resonance. The total matrix element for transition into the final state f with phonons ν_1 and ν_2 is

$$\langle f|\mathcal{H}|i\rangle=\langle f|\mathcal{H}_{\mathrm{I}}|i\rangle+\langle f|\mathcal{H}_{\mathrm{II}}|i\rangle+\langle f|\mathcal{H}_{\mathrm{III}}|i\rangle.\tag{57}$$

Let us substitute Eq. (57) into Eq. (36). The sum over final states in Eq. (36) runs over all combinations of two phonons $\Sigma_f\cdots=\underline{\Sigma}_{\{\nu_1,\nu_2\}}\cdots=\frac{1}{2}\Sigma_{\nu_1,\nu_2}\cdots$. To calculate the probability W_s we use the following properties of the function $K_{mn}(p_x,p_y)$:

$$K_{mn}(p_x,p_y)=K_{nm}(-p_x,p_y),$$

$$K^*_{nm}(p_x,p_y)=K_{nm}(p_x,-p_y).$$

Finally, with the help of Eq. (35), we find

$$e^*_{s\alpha}e_{s\beta}e_{s\gamma}e^*_{s\lambda}S_{\alpha\gamma\beta\lambda}=2\mathcal{K}\delta(\hbar\omega_l-\hbar\omega_s-2\hbar\omega_{\mathrm{LO}})\sum_{\mathbf{q}_\perp}B^2_{01}(q_\perp)\times\sum_{j,j'}|[\mathcal{A}(\omega_l,H)+\mathcal{C}(\omega_l,H)]\times P^e_{\mathbf{q}_\perp,j}P^e_{-\mathbf{q}_\perp,j'}+\mathcal{B}(\omega_l,H)\times(P^e_{\mathbf{q}_\perp,j}P^h_{-\mathbf{q}_\perp,j'}+P^e_{-\mathbf{q}_\perp,j'}P^h_{\mathbf{q}_\perp,j})/2|^2,\tag{58}$$

where

$$\mathcal{K}=\frac{\Pi^4}{(2\pi)^2}\frac{S^2}{V_0a_H^4}\frac{\hbar e^4}{m_0^4}\frac{|\mathbf{e}_l\mathbf{p}_{cv}|^2|\mathbf{e}_s\mathbf{p}_{cv}|^2}{\omega_l^2\omega_s^2},$$

$$B_{01}(q_\perp)=|K_{01}(-a_Hq_y,a_Hq_x)|^2=\frac{a_H^2q_\perp^2}{2}\exp\left(-\frac{a_H^2q_\perp^2}{2}\right),$$

$$\mathcal{A}(\omega_l,H)=(\hbar\omega_l-\widetilde{E}_{gH}-\hbar\omega_{\mu H}-2\hbar\omega_{\mathrm{LO}})^{-1}Z(\omega_l,H),$$

$$\mathcal{C}(\omega_l,H)=(\hbar\omega_l-\widetilde{E}_{gH})^{-1}X(\omega_l,H),\tag{59}$$

$$\widetilde{E}_{gH}=E_g+\varepsilon^e+\varepsilon^h+(1/2)\hbar\omega_{\mu H},$$

$$\omega_{\mu H}=\omega_{eH}+\omega_{hH}=\frac{eH}{\mu c},\quad \mu=\frac{m_em_h}{m_e+m_h}.$$

The results of the sums in $Z(\omega_l,H)$, $X(\omega_l,H)$, and $\mathcal{B}(\omega_l,H)$ are

$$Z(\omega_l,H)=[(\hbar\omega_l-\hbar\Omega_{\mathrm{in}})^2-(\Delta/2)^2]^{-1},$$

$$(\omega_l,H)=[(\hbar\omega_l-\hbar\Omega_{\mathrm{out}})^2-(\Delta/2)^2]^{-1},$$

$$\mathcal{B}(\omega_l,H)=(2\hbar\omega_l-\hbar\Omega_{\mathrm{in}}-\hbar\Omega_{\mathrm{out}})\times Z(\omega_l,H)X(\omega_l,H),\tag{60}$$

where

$$\hbar\Omega_{\mathrm{in}}=\widetilde{E}_{gH}+\hbar\omega_{hH}+\hbar(\omega_{eH}+\omega_{\mathrm{LO}})/2,$$

$$\hbar\Omega_{\mathrm{out}}=\widetilde{E}_{gH}+\hbar\omega_{\mathrm{LO}}+\hbar(\omega_{eH}+\omega_{\mathrm{LO}})/2,$$

$$\Delta=2\sqrt{\left(\frac{\hbar\omega_{eH}-\hbar\omega_{\mathrm{LO}}}{2}\right)^2+A^2}.\tag{61}$$

Note that Ω_{in} and Ω_{out} correspond to the incoming and outgoing resonances in the limit of zero magnetopolaron splitting.

V. DISCUSSION AND CONCLUSIONS

The magnetopolaron effect results in (i) a strong increase of the scattering intensity and (ii) a splitting of the incoming and outgoing resonances for two-LO-phonon Raman scattering into two doublets. The first doublet appears around the frequency $\Omega_{\rm in}$ at $\hbar\omega_{R\ {\rm in}\pm}=\hbar\Omega_{\rm in}\pm(\Delta/2)$ and corresponds to the incoming resonance when the energy of the initial state is equal to the energy of the first intermediate state β with a hole $n=1$ and a double polaron in an upper a or lower b energy branch (see Fig. 1). The second doublet at $\hbar\omega_{R\ {\rm out}\pm}=\hbar\Omega_{\rm out}\pm(\Delta/2)$ is close to a frequency $\Omega_{\rm out}$ and corresponds to the outgoing resonance when the initial state energy equals the energy of the second intermediate state γ consisting of the hole $n=0$, the upper or lower polarons, and the LO phonon. Since we assumed $m_h>m_e$, the difference

$$\Omega_{\rm out}-\Omega_{\rm in}=\omega_{\rm LO}-\omega_{hH} \tag{62}$$

is positive, but $\Omega_{\rm out}<\omega_{l0}$, where ω_{l0} corresponds to the direct creation of a triple polaron (see Fig. 1) as defined in Eq. (41). In Refs. 9–11 the approximation $m_h\to\infty$ has been used. In this case $\omega_{hH}=0$ and $\Omega_{\rm out}=\omega_{l0}$, which means that one cannot neglect the contribution of the triple polaron.

Our result in Eq. (58) is valid for $\omega_{eH}\simeq\omega_{\rm LO}$, $\omega_l\simeq\Omega_{\rm in}$, and $\omega_l\simeq\Omega_{\rm out}$. First let us consider the range

$$|\omega_l-\Omega_{\rm in}|\sim A,\quad |\omega_{eH}-\omega_{\rm LO}|\sim A. \tag{63}$$

Substituting $\mathcal{A}$, $\mathcal{C}$, and $\mathcal{B}$ in Eq. (58), we find that close to the incoming resonance

$$\begin{aligned}e^*_{s\alpha}e_{s\beta}e_{s\gamma}e^*_{s\lambda}S_{\alpha\gamma\beta\lambda}\simeq 2\mathcal{K}Z^2(\omega_l,H)\,\delta(\hbar\omega_l-\hbar\omega_s&\\ -2\hbar\omega_{\rm LO})\sum_{\mathbf{q}_\perp}B^2_{01}(q_\perp)&\\ \times\sum_{j,j'}\left|\frac{P^e_{\mathbf{q}_\perp,j}P^e_{-\mathbf{q}_\perp,j'}}{\hbar\omega_l-\widetilde{E}_{gH}-\hbar\omega_{\mu H}-2\hbar\omega_{\rm LO}}\right.&\\ \left.+\frac{(P^e_{\mathbf{q}_\perp,j}P^h_{-\mathbf{q}_\perp,j'}+P^e_{-\mathbf{q}_\perp,j}P^h_{-\mathbf{q}_\perp,j'})/2}{\hbar\omega_l-\widetilde{E}_{gH}-2\hbar\omega_{\rm LO}}\right|^2.&\end{aligned} \tag{64}$$

The first and the second term in vertical bars correspond to the processes where both phonons are emitted by the electron and where one phonon is emitted by the electron and another by the hole, respectively. The energy denominators in vertical bars describe virtual intermediate states for corresponding processes and cannot be equal to zero for laser frequency close to the incoming resonance. In order to analyze the resonance behavior of the scattering efficiency we introduce finite lifetimes for resonant states. Then the function $Z^2(\omega_l,H)$ from Eqs. (60) and (64) is transformed into

$$\begin{aligned}Z^2(\omega_l,H)=|\hbar\omega_l-\hbar\omega_{R\ {\rm in}+}-i\hbar\Gamma_{{\rm in}a}/2|^{-2}&\\ \times|\hbar\omega_l-\hbar\omega_{R\ {\rm in}-}-i\hbar\Gamma_{{\rm in}b}/2|^{-2}.&\end{aligned}$$

For both components in the doublet we have

$$Z^2(\omega_{R\ {\rm in}\pm},H)=\left(\frac{\hbar\Gamma}{2}\right)^{-2}\left[\Delta^2+\left(\frac{\hbar\Gamma}{2}\right)^2\right]^{-1}, \tag{65}$$

where $\Delta\sim\sqrt{\alpha}$. Far from the points of anticrossing the polaron effect is not important and conditions for double resonance cannot be realized. Only single resonance is possible and instead of $Z^2(\omega_l,H)$ a factor

$$\left(\frac{\hbar\Gamma}{2}\right)^{-2}(\Delta E)^{-2}$$

appears in the scattering efficiency where ΔE is the energy detuning. Thus the polaron effect corresponding to the conditions of a double resonance leads to the increase of the scattering efficiency by the factor $(\Delta E/\Delta)^2$ when $\Delta\gg(\hbar\Gamma/2)$. Since $\Delta\sim\sqrt{\alpha}$, the scattering efficiency in the magnetopolaron regime is of order α and of order α^2 otherwise.

Note that the result of Eq. (65) has a general character and can be applied to all cases of double resonance where the crossing of energy branches results in the splitting of a double resonance peak. Neglecting the splitting *leads to an incorrect estimate* for scattering efficiency $\sim(\hbar\Gamma/2)^{-4}$, which follows from the simultaneous vanishing of the two out of three energy denominators corresponding to the intermediate states. This estimate is immediately obtained if the resonant coupling between the states $(n=1,\ N=0)$ and $(n=0,\ N=1)$ leading to the thick dressed lines for electrons (see Fig. 2) is incorrectly neglected *under the resonant conditions* $\omega_{eH}=\omega_{\rm LO}$.

In a similar way, close to the outgoing resonance

$$|\omega_l-\Omega_{\rm out}|\sim A,\quad |\omega_{eH}-\omega_{\rm LO}|\sim A, \tag{66}$$

one finds

$$\begin{aligned}e^*_{s\alpha}e_{s\beta}e_{s\gamma}e^*_{s\lambda}S_{\alpha\gamma\beta\lambda}&\\ =2\mathcal{K}X^2(\omega_l,H)\,\delta(\hbar\omega_l-\hbar\omega_s-2\hbar\omega_{\rm LO})&\\ \times\sum_{\mathbf{q}_\perp}B^2_{01}(q_\perp)\sum_{j,j'}\left|\frac{P^e_{\mathbf{q}_\perp,j}P^e_{-\mathbf{q}_\perp,j'}}{\hbar\omega_l-\widetilde{E}_{gH}}\right.&\\ \left.+\frac{(P^e_{\mathbf{q}_\perp,j}P^h_{-\mathbf{q}_\perp,j'}+P^e_{-\mathbf{q}_\perp,j}P^h_{\mathbf{q}_\perp,j'})/2}{\hbar\omega_l-\widetilde{E}_{gH}-\hbar\omega_{\mu H}}\right|^2,&\end{aligned} \tag{67}$$

where

$$\begin{aligned}X^2(\omega_l,H)=|\hbar\omega_l-\hbar\omega_{R\ {\rm out}+}-i\hbar\Gamma_{\rm out}a/2|^{-2}&\\ \times|\hbar\omega_l-\hbar\omega_{R\ {\rm out}-}-i\hbar\Gamma_{{\rm out}b}/2|^{-2}.&\end{aligned}$$

On the right-hand side of Eqs. (64) and (67) one can easily separate the contributions of processes of type (i), where one phonon is emitted by the electron and another by the hole, and types (ii) and (iii), where both phonons are emitted by the electron. For $\omega_{eH}=\omega_{\rm LO}$ the energy detuning in the only virtual state for process (i) is $\hbar\omega_{\rm LO}-\omega_{hH}$, whereas for (ii) and (iii) it is $2\hbar\omega_{\rm LO}$ for both incoming and outgoing resonances. Thus the processes of type (i) are more effective in the Raman scattering and should give the main contribution for the case involving the light holes in GaAs.

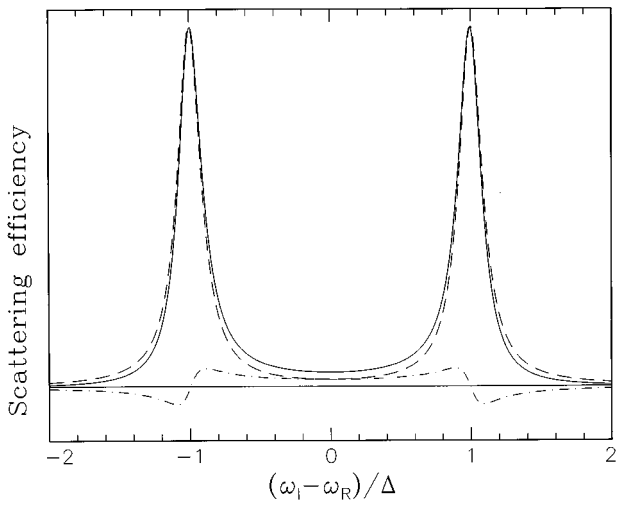

FIG. 4. Magnetopolaron-induced splitting of the incoming or the outgoing resonance profile in two-LO-phonon Raman scattering. The dashed curve corresponds to the direct contribution from lower and higher polaron branches, whereas the dash-dotted line shows their interference. The solid curve is the total scattering efficiency. A phenomenological broadening of 0.1Δ has been used in the calculation. Far from the magnetopolaron resonance both peaks should be reduced by the factor $\sim(\Delta E/\Delta)^2$, where ΔE is the energy detuning and Δ the magnetopolaron splitting.

In Fig. 4 we show the results of calculations of the resonant profile for two-LO-phonon Raman scattering in the polaron regime when $\omega_{eH}=\omega_{\mathrm{LO}}$. The profile is the same for both incoming and outgoing resonances showing the double polaron splitting of the peak. The dashed line shows direct contributions from the two polaron branches, whereas the dash-dotted lines represent their interference. The sum is shown as a solid line. We have used a phenomenological broadening of 0.1Δ in the calculations. Note that exactly in the middle between two peaks the interference contribution is equal to the direct one, whereas it is not important close to both resonances.

Double resonance conditions cannot be satisfied in the case of one-LO-phonon Raman scattering via the intraband Fröhlich interaction because of the Landau number conservation in interaction with light.[28–30] Although there is a magnetopolaron splitting of the Raman peak, there is no increase in the scattering efficiency. Nevertheless, double resonance in a one-LO-phonon Raman scattering can be realized for interband transitions via deformation potential or in the electric field.[28–30]

Now we discuss qualitatively the role of higher polarons (see Fig. 1) in Raman scattering. At $\omega_l=\omega_{l0}$ the direct creation of a triple polaron becomes possible. Triple polarons appear as a result of strong interaction between the states $(n=2,\ N=0)$, $(n=1,\ N=1)$, and $(n=0,\ N=2)$ when $\omega_{eH}\simeq\omega_{\mathrm{LO}}$. Under such conditions a three-LO-phonon Raman scattering might be triply resonant. In terms of bare excitations this means that three out of four intermediate states in the scattering process can be real. Using the basis of magnetopolaron intermediate states, one can say that the process takes place via real states of a triple polaron. The scattering efficiency is proportional to α and the triple resonance splits into three peaks.[9–11] All higher-order Raman processes can be described qualitatively in a similar way. This means that under resonant conditions with respect to the incident light frequency and the magnetic-field strength the scattering efficiency of N-LO-phonon Raman scattering is of the order of α for any N. One can say that N out of $N+1$ intermediate states in the scattering process are real and only one state is virtual. However, it is more correct to say that all real intermediate states represent the real state of a magnetopolaron with multiple splitting. This qualitative picture may break down for higher-order processes when the Landau levels are not equidistant, e.g., because of nonparabolicity effects. Obviously, this is not the case for two-LO-phonon process where only the energy separation of one pair of Landau levels is important, which can be tuned by the magnetic-field strength.

Finally, we analyze qualitatively the role of phonon dispersion in the growth direction. For narrow QW's ($d<50$ Å in GaAs) the characteristic separation $\Delta\omega_j$ of quantized phonon modes may be larger than their broadening. Then each quantized phonon mode results in a double magnetopolaron at the magnetic field strength to be found from the condition $\omega_{eH}=\omega_j$. Then one has to distinguish between two phonons in the final state, which can be of a different frequency. To test double polaron states, we need at least one of these phonons to be in resonance with the electron cyclotron frequency. This is easy to achieve by tuning the magnetic field. Another phonon can be either from the same or from some other size quantized mode. All equations obtained above are directly applicable to the case when both phonons correspond to the same branch. Then we have to substitute $\omega_{\mathrm{LO}}\rightarrow\omega_j$ and $\Sigma_\nu\rightarrow\Sigma\mathbf{q}_\perp$ in all equations. Let us assume that the magnetopolaron splitting $2A_j=2\Sigma\mathbf{q}_\perp|U(j,\mathbf{q}_\perp)|^2\ll\Delta\omega_j$. We consider the incident light frequency $\omega_l\simeq\Omega_{j\ \mathrm{in}}$ and $\omega_l\simeq\Omega_{j\ \mathrm{out}}$, where

$$\hbar\Omega_{j\ \mathrm{in}}=\widetilde{E}_{gH}+\hbar\omega_{hH}+(\hbar\omega_{eH}+\hbar\omega_j)/2,$$

$$\hbar\Omega_{j\ \mathrm{out}}=\widetilde{E}_{gH}+\hbar\omega_j+(\hbar\omega_{eH}+\hbar\omega_j)/2.$$

The two peaks in the resonant profile split into two doublets at $\omega_l=\Omega_{j\ \mathrm{in}}\pm(\Delta/2)$ and $\omega_l=\Omega_{j\ \mathrm{out}}\pm(\Delta/2)$, where

$$\Delta_j=2\sqrt{\left(\frac{\hbar\omega_{eH}-\hbar\omega_j}{2}\right)^2+A_j^2}.$$

The generalization to the case of two different phonons in the final state is rather straightforward and consists of eliminating one of the two contributions to each of three matrix elements in Eq. (57). The exchange of the phonon indices in Eqs. (48), (52), and (55) does not lead to equivalent contributions anymore. One of the two contributions in each pair is always nonresonant and can be neglected. The other limit $2A_j\gg\Delta\omega_j$ is more complicated and we leave its analysis for future work.

To summarize, the wave function and energy spectrum of a double polaron has been derived for resonantly coupled electron and phonon subsystems of a quantum well. The magnetic-field range both in resonance and far from resonance has been analyzed. In the nonresonant range of the

magnetic-field strength the results coincide with those of lowest-order perturbation theory. The scattering efficiency for a two-LO-phonon Raman scattering has been calculated in the vicinity of incoming and outgoing resonance with a double polaron. The splitting of the peaks in the Raman profile is accompanied by a strong increase in the scattering efficiency related to the doubly resonant character of the scattering process under conditions of a resonant polaron coupling.

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