Intraband resonance Raman scattering in anisotropic quantum dots

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We have developed a theory of the one-phonon intraband resonance Raman scattering (IRR) in anisotropic quantum dots subjected to an arbitrarily directed magnetic field. The differential Raman cross section is obtained. The resonance structure of the Raman cross section is studied. It is shown that the quantum dot subjected in a magnetic field can be used as the detector of phonon modes. The interesting multiplet structure of the resonance peaks is studied.

I. INTRODUCTION

Theoretical and experimental studies of the Raman scattering and photoluminescence in quantum dots (QD) are of great interest because the understanding of the scattering mechanisms is of fundamental importance for the applications. Raman scattering can provide the direct information on the electronic structure, phonon spectrum, and optical properties of QDs. The most part of papers is devoted to studying the interband Raman scattering. However, we suppose that the intraband resonance Raman scattering is also of great interest because the distance between discrete levels in QDs can be done of order the optical phonon energy with help of the magnetic field. It is important to note that the optical phonon emission is known to play a dominant role in QDs among the scattering mechanisms present in polar semiconductors.

Modern nanotechnology enables one to fabricate quantum dots of different shapes. In the past years the significant interest has been given to quantum wells and QDs that are characterized by an asymmetric confining potential. In this work we present a theoretical study of the intraband resonance Raman scattering of an anisotropic quantum dot subjected to a uniform magnetic field arbitrarily directed with respect to the potential symmetry axes. The applied magnetic field gives us the possibility to change the distance between levels and to adjust the energy levels of QDs on the various phonon modes. The study of the different polarization for the incident and emitted radiation yields the additional information about the phonon spectrum. Note that the study of intraband Raman scattering lets us to obtain the simple analytic formulae for the cross-section in the case of anisotropic QDs.

Resonance intraband Raman scattering in our case can be qualitatively described in the following way: the absorption of quantum $\hbar \omega_i$ of the high-frequency field (laser pump), emission of optical phonon $\hbar \omega q$ (phonon $\hbar \omega s$) in an intermediate state and emission of photon $\hbar \omega_s$ (optical phonon $\hbar \omega q$) in the initial state (Figure 1). In this approach, the Raman cross section is calculated from third order time-dependent perturbation theory.

We model the semiconductor QD using the asymmetric parabolic confinement $V(x) = m^* (\Omega_1 x^2 + \Omega_2 y^2 + \Omega_2 z^2)/2$ (here $m^*$ is the electron effective mass, $\Omega_i$ ($i = x, y, z$) are the characteristic frequencies of the parabolic potential).

The spectrum of electrons in such dot placed in an arbitrarily directed magnetic field $\mathbf{B}$ with the vector potential $\mathbf{A} = (B_y x/2 - B_x y, 0, B_z x/2)$ has the form $\varepsilon_{nml} = \hbar \omega_1 (n + 1/2) + \hbar \omega_2 (m + 1/2) + \hbar \omega_3 (l + 1/2)$ ($n, m, l = 0, 1, 2, \ldots$), where hybrid frequencies $\omega_j$ ($j = 1, 2, 3$) are obtained from the sixth-order algebraic equation.

II. RAMAN CROSS SECTION

The differential resonance Raman cross section $d^2 \sigma / d \Omega d \omega_s$ of a volume $V$ per unit solid angle $d \Omega$ for incident radiation with the frequency $\omega_i$ and emitted radiation with the frequency $\omega_s$ is given by

$$
\frac{d^2 \sigma}{d \Omega d \omega_s} = \frac{V^2 \omega^2_\alpha n^3_i}{8\pi^3 c^3 \omega_i} W(\omega_s, \varepsilon_s) \tag{1}
$$

where $n_i$ ($n_s$) is the refractive index of the medium with frequency $\omega_i$ ($\omega_s$), $c$ is the velocity of light, $\varepsilon_s$ is the unit polarization vector of the emitted radiation. The transition rate is calculated according to

$$
W(\omega_s, \varepsilon_s) = \frac{2\pi}{\hbar} \sum_\alpha |W_{\alpha \alpha}|^2 \delta(\hbar \omega_i - \hbar \omega_s - \hbar \omega_q), \tag{2}
$$

where $\alpha = (n, m, l)$ are the quantum numbers of the initial electron states in QD.

We consider only resonance transitions. In this case the scattering amplitude probability for phonon emission in QDs is described by a sum of two terms

$$
W_{\alpha \alpha} = \sum_{\alpha', \alpha''} \langle \alpha | \hat{H}_R(\omega_i) | \alpha'' \rangle \langle \alpha'' | \hat{H}_L | \alpha \rangle \delta(\varepsilon_{\alpha'} - \varepsilon_{\alpha} - \hbar \omega_i) (\varepsilon_{\alpha''} - \varepsilon_{\alpha} - \hbar \omega_s) + \sum_{\alpha', \alpha''} \langle \alpha | \hat{H}_L | \alpha'' \rangle \langle \alpha'' | \hat{H}_R(\omega_s) | \alpha \rangle \delta(\varepsilon_{\alpha'} - \varepsilon_{\alpha} - \hbar \omega_i) (\varepsilon_{\alpha''} - \varepsilon_{\alpha} - \hbar \omega_q) \tag{3}
$$

FIG. 1. Transitions leading to resonant absorption.
The first term in Eq. (3) corresponds to the transitions depicted on Figure 1, the second term in Eq. (3) corresponds to the transitions depicted on Figure 1.

Here $H_L$ is the operator electron-phonon interaction

$$\hat{H}_L = \sum_q D_q C_q \exp(iq \mathbf{r}) + \text{c.c.}$$

where $D_q$ is the electron-phonon coupling constant.

The operator of the electron-photon coupling interaction can be expressed as

$$\hat{H}_R = \frac{e}{m^*} \sqrt{\frac{2\pi \hbar N}{\varepsilon \omega}} \mathbf{e} \mathbf{P},$$

where $N$ is the number of initial-state photons with frequency $\omega$, $\mathbf{e}$ is the polarization vector and $\mathbf{P} = \mathbf{p} - e A/c$ is the generalized momentum, $\varepsilon$ is the real part of the dielectric constant.

A direct calculation of the matrix elements of electron-photon and electron-phonon interactions is a complicated problem in our case. However, the method of canonic transformation of the phase space allows us to resolve this problem using only simple calculations from linear algebra.

In particular, in our preceding papers we used this method to study hybrid-19, hybrid-phonon20 and hybrid-impurity resonances in this system.

Using the results obtained in 19, we can write the matrix elements of the operator $\hat{H}_R$ in the following form

$$\langle n'm'l'|\hat{H}_R|nml\rangle = i e \hbar \sqrt{\frac{\pi N}{m^* \varepsilon \omega}} \times \left[ X_1 \sqrt{\frac{1}{1 + \delta_{n',n+1} \delta_{m',m} \delta_{l',l}}} + X_2 \sqrt{\frac{1}{m + \delta_{n,n} \delta_{m',m} \delta_{l',l}}} + X_3 \sqrt{1 + \delta_{n',n} \delta_{m',m} \delta_{l',l+1}} \right].$$

where the coefficients $X_i$ ($i = 1, 2, 3$) were found in 19.

We introduce the notation

$$J(n'm'n'l', n'm'l') = \left( \frac{n'm'n'l'}{n'm'l'} \right)^{1/2} (1 - n', - n'') \times \left[ 1 + \delta_{n',n+1} \delta_{m',m} \delta_{l',l} \right] \times \exp[i\varphi_1(n', - n'')] \times \exp[i\varphi_3(l', - l''')] \times g_1 l'' \times l'' \times \left( g_2^l \right) l'' \times \left( g_2^l \right) l'' \times \left( g_2^l \right) l'' \times \left( g_2^l \right) l'' .$$

Here $g_2 = \sqrt{\lambda_j^2 + \lambda_j^2} / \sqrt{2\lambda_j}$, tan $\varphi_j = \lambda_j / \lambda_j$, $\lambda_j = \sqrt{\hbar/m^* \omega_j}$ ($j = 1, 2, 3$) are the hybrid lengths, $L_n(x)$ are the generalised Laguerre polynomials, $\lambda_i = h(b_1 q_x + b_2 q_y + b_3 q_z)$ ($i = 1, 2, 3$), $\lambda_i = b_1 q_x + b_2 q_y + b_3 q_z$ ($i = 4, 5, 6$), $b_j$ are components of the matrix $[19, 20, 21]$

Using (7), we can write the matrix elements of the operator electron-phonon interaction as

$$\langle n'm'l'|\hat{H}_L|n''m''l''\rangle = \sum_q D_q \sqrt{N_q} \exp(-g^2/2) \times \exp[-(\kappa_1 \lambda_1 + \kappa_2 \lambda_2 + \kappa_3 \lambda_3) / 2] J(n'm'n'l', n'm'l') \times$$

where $N_q$ is the number of phonons with the wave vector $q$ and $g^2 = g_1^2 + g_2^2 + g_3^2$.

Substituting (6) and (7) into (3) after some cumbersome algebra it is possible to get analytic expression for $W_{a \alpha}$. We consider only the resonance Raman scattering.

In this case the frequency of the pump is equal to the distance between the levels of QD. For definiteness, assume that we pump the QD by the laser with frequency $\omega_i = \omega_1$. Then we need to keep only resonance terms in the formula for $W_{a \alpha}$. In this case for the first term in Eq. (3), we obtain

$$\sum_{\alpha', \alpha''} \frac{\langle \alpha|\hat{H}_R(\omega_i)|\alpha''\rangle \langle \alpha'\hat{H}_L|\alpha'\rangle \langle \alpha'\hat{H}_R(\omega_i)|\alpha\rangle}{(\varepsilon_{\alpha'} - \varepsilon_{\alpha} - \hbar \omega_i)(\varepsilon_{\alpha''} - \varepsilon_{\alpha} - \hbar \omega_i + \hbar \omega_q)} = \frac{-\pi e^2}{m^* \varepsilon} \sqrt{\frac{N_q}{\omega_i \omega_q}} \sum_q D_q \sqrt{N_q} \exp(-g^2/2) \times$$

$$\exp[-(\kappa_1 \lambda_1 + \kappa_2 \lambda_2 + \kappa_3 \lambda_3) / 2] \sqrt{n + \Gamma X_1^2} \omega_i - \omega_1 \times \left[ \sqrt{m + \Gamma X_1^2 J(nm + 1, n + 1m)} \right].$$

Here the indices $i$ and $s$ are related to the incident and emitted photons, respectively.

The second term in (3) has the following form

$$\sum_{\alpha', \alpha''} \frac{\langle \alpha|\hat{H}_L|\alpha''\rangle \langle \alpha'\hat{H}_R(\omega_i)|\alpha'\rangle \langle \alpha'\hat{H}_L|\alpha\rangle}{(\varepsilon_{\alpha'} - \varepsilon_{\alpha} - \hbar \omega_i)(\varepsilon_{\alpha''} - \varepsilon_{\alpha} - \hbar \omega_i + \hbar \omega_q)} = \frac{-\pi e^2}{m^* \varepsilon} \sqrt{\frac{N_q}{\omega_i \omega_q}} \sum_q D_q \sqrt{N_q} \exp(-g^2/2) \times$$

$$\exp[-(\kappa_1 \lambda_1 + \kappa_2 \lambda_2 + \kappa_3 \lambda_3) / 2] \sqrt{n + \Gamma X_1^2} \omega_i - \omega_1 \times \left[ \sqrt{m + \Gamma X_1^2 J(nm + 1, n + 1m)} \right].$$

Now we need to sum these terms to get $W_{a \alpha}$. Taking into account the conversation law $\hbar \omega_i = \hbar \omega_q + \hbar \omega_s$ we can transform denominators in Eq. (9) and Eq. (10). In this case we get for $W_{a \alpha}$ the following formula

$$W_{a \alpha} = \frac{-\pi e^2}{m^* \varepsilon} \sqrt{\frac{N_q}{\omega_i \omega_q}} \sum_q D_q \sqrt{N_q} \exp(-g^2/2) \times$$

$$\exp[-(\kappa_1 \lambda_1 + \kappa_2 \lambda_2 + \kappa_3 \lambda_3) / 2] \sqrt{n + \Gamma X_1^2} \omega_i - \omega_1 \times \left[ \sqrt{m + \Gamma J(nm + 1, n + 1m)} \right]$$
laser and changing, for example, the magnitude of the magnetic field). Hence, using the tunable cross sections dispersion and if the frequency of the pump is equal to \( \omega_{2} \), we get for the first difference
\[
\sqrt{m+1}J(nm+1, n+1ml) - \sqrt{m}J(nml, n+1ml-1) = -\exp(i\varphi_{1}) \exp(-i\varphi_{2}) g_{1}g_{2}L_{n}^{1}(g_{1}^{2})L_{m}(g_{2}^{2})L_{l}(g_{3}^{2})
\]
(12)

The second difference in Eq. (11) is calculated in analogy with Eq. (12). Using the obtained estimation, we get the following formula for the square of the scattering amplitude probability
\[
|W_{ao}|^{2} = \frac{\pi^{2}e^{2}}{m^{2}} N_{s}(N_{s}+1) \sum_{q} |D_{q}|^{2} N_{q} \exp(-g_{1}^{2})
\]
\[
\times [L_{n}^{1}(g_{1}^{2})^{2]}L_{m}(g_{2}^{2})^{2]}L_{l}(g_{3}^{2})^{2} \frac{(n+1)(X_{l}^{2})^{2}}{(\omega_{1}-\omega_{l})^{2} + \Gamma^{2}}
\]
\[
\times \left| \frac{g_{2}X^{S}_{p}}{\omega_{2}-\omega_{s}} \exp(-i\varphi_{2}) + \frac{g_{3}X^{S}_{p}}{\omega_{3}-\omega_{s}} \exp(-i\varphi_{3}) \right|^{2}.
\]
(13)

Then we can write the final formula for the Raman cross-section taking into account the smearing of the hybrid levels by collisions
\[
\frac{d^{2}\sigma}{d\Omega d\omega_{s}} = V \omega_{2}^{2} n_{s} e^{4} N_{s}(N_{s}+1) \sum_{q} |D_{q}|^{2} N_{q} \exp(-g_{2}^{2})
\]
\[
\times g_{1}^{2} L_{n}^{1}(g_{1}^{2})^{2]}L_{m}(g_{2}^{2})^{2]}L_{l}(g_{3}^{2})^{2} \frac{(n+1)(X_{l}^{2})^{2}}{(\omega_{1}-\omega_{l})^{2} + \Gamma^{2}}
\]
\[
\times \left| \frac{g_{2}X^{S}_{p}}{\omega_{2}-\omega_{s}} \exp(-i\varphi_{2}) + \frac{g_{3}X^{S}_{p}}{\omega_{3}-\omega_{s}} \exp(-i\varphi_{3}) \right|^{2}
\]
\[
\times \delta(\omega_{1} - \omega_{s} - \omega_{q}),
\]
(14)

where \( \Gamma \) is the lifetime broadening.

III. RESULTS AND DISCUSSIONS

Equation (14) clearly shows that if one ignores the optical phonons dispersion and if the frequency of the pump is equal to \( \omega_{1} \) then we have the input resonance on the frequency \( \omega_{1} \) and the output resonance on the frequencies \( \omega_{2} \) and \( \omega_{3} \). Note that it is forbidden transitions with simultaneous changing more than one quantum numbers in the case of absorption (emission) of photon due to the selection rules. The possible transitions is shown on the Figure 2.

It is important to note that the hybrid frequency \( \omega_{k} \) \((k = 1, 2, 3)\) is determined by the magnitude and the direction of the magnetic field (i.e. they can be tuned with the help of the magnetic field). Hence, using the tunable laser and changing, for example, the magnitude of the magnetic field we can register phonon modes (with frequencies \( \omega_{q} = \omega_{1} - \omega_{2} \) and \( \omega_{q} = \omega_{1} - \omega_{3} \)) in quantum dots as series resonance peaks in the dependance of the Raman cross section on the magnetic field. The frequency of the phonon mode can be determined from the dependence of the magnetic field on the hybrid frequencies.

Let us now to study effects arising due to the dispersion of the phonons. Replacing the sum over the phonon wave vector by the integral in Equation (11) and assuming a parabolic dispersion law for long-wave phonons \( \omega_{q} = \omega_{0}(1 - \omega_{0}^{-2}V_{s}^{2}q^{2}) \), where \( \omega_{0} \) is the optical-phonon threshold frequency and \( V_{s} \) is the speed of sound, we can easily evaluate the integral with respect to \( |q| \) thanks to the presence of a delta function \( \delta(\omega_{1} - \omega_{s} - \omega_{q}) \).

Converting to spherical coordinates we obtain the following equation for the Raman cross section
\[
\frac{d^{2}\sigma}{d\Omega d\omega_{s}} = \frac{V \omega_{2}^{2} n_{s} e^{4} N_{s}(N_{s}+1)N_{0}^{2} \omega_{0}^{3/2}}{8\hbar^{4}V_{s}^{3}n_{s}e^{2}m^{2} \sqrt{\Delta\omega_{0}}}
\]
Then we can rewrite Equation (15) as

\[ \times \int_{0}^{2\pi} \int_{0}^{\pi} \sin \theta d\theta |D_{q}|^2 \exp(-y^2) y_1^2 \]

\[ \times [L_n(y_1^2)]^2[L_m(y_2^2)]^2[L_l(y_3^2)]^2 \frac{(n + 1)(X_1)^2}{(\omega_1 - \omega_i)^2 + \Gamma^2} \]

\[ \times \left| \frac{y_2 X_2^S}{\omega_2 - \omega_s - i\Gamma} \exp(-i\varphi_2) + \frac{y_3 X_3^S}{\omega_3 - \omega_s - i\Gamma} \exp(-i\varphi_3) \right|^2 \],

Here we replace \( N_q \) by the Plank distribution function \( N_0 \), \( y_j \) can be obtained from \( q_j \) if we write the vector \( q \) in the spherical coordinates, \( D_q \) depends on the electron-phonon interaction and \( \Delta \omega_0 = \omega_1 - \omega_s - \omega_0 \).

Let us consider, first of all, the polarization potential scattering (PO phonons). In this case the electron-phonon coupling constant

\[ |D_q|^2 = \frac{2\sqrt{2\pi \hbar^2 \Omega \omega_0^3/2}}{\sqrt{\sigma^3 q^2}}. \] (16)

Then we can rewrite Equation (15) as

\[ \frac{d^2\sigma}{d\Omega d\omega_s} = \frac{\sqrt{2\pi V \omega_2 n_s n_i^2 e^4 N_s(N_s + 1) \Omega \omega_0^2}}{4V_s^2 c^3 m^{3/2} \varepsilon^2 |\Delta \omega_0|^{3/2}} \int_{0}^{2\pi} \int_{0}^{\pi} \sin \theta d\theta |D_{q}|^2 \exp(-y^2) y_1^2 \]

\[ \times [L_n(y_1^2)]^2[L_m(y_2^2)]^2[L_l(y_3^2)]^2 \frac{(n + 1)(X_1)^2}{(\omega_1 - \omega_i)^2 + \Gamma^2} \]

\[ \times \left| \frac{y_2 X_2^S}{\omega_2 - \omega_s - i\Gamma} \exp(-i\varphi_2) + \frac{y_3 X_3^S}{\omega_3 - \omega_s - i\Gamma} \exp(-i\varphi_3) \right|^2 , \] (17)

The Raman cross section depends on the polarization both the input signal and output one. Let us consider the case when the polarization vector of the incident and emitted fields are perpendicular to the magnetic field. In this case the hybrid frequencies are determined by the formulae \( \omega_{1,2} = [\sqrt{\Omega_0^2 + \omega_i^2} \pm \sqrt{\omega_s^2 - \omega_i^2} + \omega_0^2]/2, \omega_3 = \Omega_s \), and the values of \( y_j \) \((j = 1, 2, 3)\) have the following form

\[ y_j = \frac{l_j}{\sqrt{2}} \left[ \frac{\omega_0 |\Delta \omega_0|}{V_s} \right]^{\sin \theta} \sqrt{\frac{\omega_s^2 \Omega_s^2 + (\omega_s^2 - \omega_i^2)^2}{\omega_i^2}} \times \left[ (\omega_s^2 - \omega_i^2)^2 \sin^2 \varphi + \omega_s^2 \omega_i^2 \cos^2 \varphi \right]^{1/2} \]

(18)

Note that in this case \( X_1^j = X_3^S \) \((j = 1, 2, 3)\). Equation (17) clearly shows that the Raman cross section has singularities at the points where \( \Delta \omega_0 = 0 \). On the Figure 3 it is shown the dependence of the Raman cross section on the magnetic field (here we taken into account the finite phonon relaxation time).

The different situation takes place in the case of deformation potential scattering (DO phonons). The Raman cross section of deformation potential scattering connected with one of polarization potential scattering by the following estimation

\[ \frac{d^2\sigma_{PO}}{d\Omega d\omega_s} = \frac{m V_s^2}{2\hbar |\Delta \omega_0|} \frac{d^2\sigma_{DO}}{d\Omega d\omega_s} \] (19)

It is important to note that in the points where \( |\Delta \omega_0| = 0 \) the Raman cross section is equal to zero in contradiction to the case of PO-phonons. In the case of DO-phonons the cross section has the complex doublet structure. The width of the resonance curve is enough small (of order 1 Oe) in this situation. In the most simple case of transitions from the ground state \( n = m = l = 0 \) the resonance curve consists of two symmetrically positioned sharp peaks to the left and right of the point \( \Delta \omega_0 = 0 \) (Fig. 4). In the case of transitions from the state \( n = m = l = 1 \) each of the doublet peaks splits
up into two ones (Fig. 5). If we take into account the finite phonon relaxation time in QDs, the resonance curve doesn’t change its form in the difference from the polarization scattering but its minimum displaces in the point where $\Delta \omega + \tau^{-1} = 0$ (here $\tau$ is the relaxation time).

In conclusion, we have investigated theoretically the resonance Raman scattering in the anisotropic quantum dots in the presence of arbitrarily directed magnetic field and polarization vector. Raman scattering lets us to detect phonon modes in QD using the tunable laser and changing magnetic field. If we ignore optical phonon dispersion, we have a resonance peak corresponding to the emission of optical phonon mode. The interesting doublet structure of peaks arises if one takes into account the dispersion of long-wave optical phonons in the case of deformation scattering. In this case the resonances let us to observe the threshold frequency of optical phonons. In the resonance point the cross section is equal to zero but in a small neighborhood of this point cross section has symmetrically positioned (to the left and right) peaks. The number of peaks depends on the initial quantum state. We hope that our calculations can further stimulate more experimental measurements on Raman intensities of semiconductor QDs.

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