

## Section 5. Physics

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### EXCITON-TWO-PHONON RABMAN SCATTERING OF LIGHT IN A QUANTUM WELL

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after Muhammad ibn Musa al-Khwarizmi

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#### Abstract

The theory of exciton-two-phonon resonant Raman scattering of light in a quantum well has been developed. It is shown that in the case of exciton-two-phonon Raman scattering of light, in which two-dimensional excitons appear as intermediate states, it leads to a sharp increase in scattering (in  $(\alpha_0^{-3} / \ln^2 \alpha_0)$  times (where  $\alpha_0$  – dimensionless Fröhlich constant of interaction of two-dimensional excitons with LO phonons,  $\alpha_0 \ll 1$ ) compared to the mechanism of electron-hole pairs. The amplification is due to the fact that in a quantum well, in which the energies of the electron and hole are dimensionally quantized, the process of direct creation (or direct annihilation) of an exciton and the actual emission of a second phonon by the exciton are possible. The scattering tensor at the maximum of the second phonon repetition peak is  $\alpha_0^{-1}$ .

**Keywords:** *exciton-two-phonon, resonant Raman scattering, quantum well, two-dimensional excitons, intermediate states, direct creation, phonon, scattering tensor, low dimension, electron and hole, for their potential applications in microelectronic device technology*

#### Introduction

In recent years a great deal of interest has been devoted to the study and the engineering of high-quality devices of very low dimension, essentially quantum-well, wires, or quantum-dots (QDs) semiconductors (Xiao Z., Zhu J., He F., 1995). Because of their low-dimensionality of these systems exhibit many new physical effects (Mukhopadhyay

S., 1995) which are extremely interesting from the point of view of fundamental physics and also for their potential applications in microelectronic device technology.

Consequently much effort has lately gone into understanding and exploring the physical properties of these systems both theoretically and experimentally. These studies have been performed with the proposal of understanding

the fascinating novel phenomena and of fabricating devices with new functions or to improve the performance of the existing devices (Wen Fang X. (2001). Excitons play a dominant role in their physical properties; therefore, their stability is important for possible devices requiring this characteristic (Moussaouy A. El., Bria D., Nougauoi A., Charrou R., Bouhassoune M., 2003). In semiconductor quantum wells, the electron–phonon interaction is usually much less.

As is known, multiphonon resonant Raman scattering (MRRS) is observed during monochromatic irradiation of some polar semiconductors in the fundamental absorption region (Xiao Z., Zhu J., He F., 1995; Mukhopadhyay S., 1995; Sood A.K., Mendez, J., Cardona M. & Ploog K., 1985; Meynadres M.H., Finkman E., Sturge M.D., Warlock J.M. & Tamargo M.C., 1987). The method (MRRS) has been intensively used in recent years to obtain information about both vibrational modes and electronic states and features of electron-phonon and exciton-phonon interactions in systems of reduced dimensionality (heterostructures, quantum wells, wires and points). Secondary lines glow (phonon repetitions) is observed at frequencies  $\omega_s = \omega_l - N\omega_{LO}$ , where  $\omega_l$  is the frequency of the exciting light;  $\omega_{LO}$  – frequency of bulk longitudinal (optical) phonons;  $N$  is the scattering order, i.e. those. number of  $LO$  phonons backgrounds emitted during scattering.

Theoretical studies of MRRS processes in a bulk semiconductor have shown that two types of processes contribute to the scattering cross sections: scattering through intermediate states of free electron-hole (EDH) (Goltsev A.V., Lang I.G., Pavlov S.T. Bryzhina M. F., 1983) and through excitonic states (Korovin L.I., Pavlov S. T. & Eshpulatov B. E., 1990).

Scattering with the participation of free EHPs in two-dimensional systems was studied in (Korovin L.I., Pavlov S. T. & Eshpulatov B. E., 1991), where it was shown that the MRRS cross section is enhanced in  $\alpha_0^{-1}$  times compared to the three-dimensional case. For phonon repetitions  $N \geq 2$ .

This article examines two-phonon resonant Raman scattering (RRS) in a single quantum well in the case where the intermediate states are two-dimensional excitons.

### Statement of the problem and necessary relationships

Let us consider a single quantum well with infinite potential walls located between the  $z = 0$  and  $z = d$  planes. Let us further assume that the relation

$$d \ll r_0, \quad (1)$$

$r_0 = \varepsilon \hbar^2 / \mu e^2$  – exciton Bohr radius,  $\varepsilon$  – dielectric constant of the quantum well material,  $e$  – electron charge,  $\mu$  – reduced effective mass.

Inequality (1) ensures the two-dimensionality of the exciton. We will assume that the exciton, emitting phonons, remains in the  $1S$  state all the time.

In the case of the second phonon repetition, the scattering tensor is determined by the expression (Korovin L.I., Pavlov S. T. & Eshpulatov B. E., 1988).

$$S_{\beta\gamma\beta'\gamma'} = S_{\beta\gamma\beta'\gamma'}^{(0)} (S_1 + S_2), \quad (2)$$

Where

$$S_{\beta\gamma\beta'\gamma'}^{(0)} = \left( \pi \hbar^6 \omega_s^2 \omega_l^2 \right)^{-1} (\hbar \omega_{LO})^4 (2d / \pi r_0)^4 \times \\ \times J_\beta J_\gamma J_{\beta'} J_{\gamma'} \delta(\omega_l - \omega_s - 2\omega_{LO}). \quad (3)$$

Scalar functions  $S_1$  and  $S_2$  represent one- and two-fold sums over quantum numbers of size quantization

$$S_1 = \alpha_0^2 \sum_{n=0}^{\infty} \int K dK I^2(K, n) \left[ (1+a_h)^{-3/2} - (1+a_e)^{-3/2} \right]^4 \times \\ \times |G(n, n, K, \omega_l - \omega_{LO})|^2 |G(n, n, 0, \omega_l)|^2 |G(n, n, 0, \omega_s)|^2, \quad (4)$$

Where

$$\alpha_0 = 2\hbar\omega_{LO} (\varepsilon_\infty^{-1} - \varepsilon^{-1}); a_{e(h)} = (\alpha_0 m_{e(h)} / 4m_e) K, \\ \omega_s = \omega_l - 2\omega_{LO} \\ G(n, n', \kappa, \omega) = \\ = [\omega - \omega(n, n', \kappa) - (i/2)\gamma(n, n', \kappa)]^{-1} \quad (5)$$

Green's function The expression for the Green's function (5) includes the function  $\gamma(n_e n_h, \mathbf{K}) = \tau^{-1}(n_e n_h, \mathbf{K})$ , where  $\tau(n_e n_h, \mathbf{K})$  is the lifetime of an exciton in the state  $(n_e n_h, \mathbf{K})$  The function  $\gamma$ , which has the meaning of a mass operator, is not calculated further; it is only assumed that

$$(\gamma / \omega_{LO}) \ll 1. \quad (6)$$

If we calculate  $\gamma$  to first order from the coupling constant  $\alpha_0$ , then it is obvious that  $\gamma \sim \alpha_0$ .

$$S_2 = \alpha_0^2 \sum_n \int_0^\infty K dK I^2(K, n, n') \left\{ \left| G(n, n', K, \omega_l - \omega_{LO}) \chi(n, n', \omega_l) \right|^2 \times \right. \\ \left. \times \chi(n, n', \omega_s) + \left| G(n', n, K, \omega_l - \omega_{LO}) \right|^2 \chi(n', n, \omega_l) \chi(n', n, \omega_s) \right\}, \quad (7)$$

$$\alpha_{h(e)} = (\alpha_0 m_{h(e)} / 4m_e) K, \quad \omega_s = \omega_l - 2\omega_{LO}.$$

Functions look like

$$\chi(n, n', \omega), I(K, n) u I(K, n, n') \quad \chi(n, n', \omega) = (1 + \alpha_h)^{-3} |G(n, n, 0, \omega)|^2 + \\ + (1 + \alpha_e)^{-3} |G(n', n', 0, \omega)|^2, \quad (8)$$

$$I(K, n) = \left( \frac{2}{x} + \frac{1}{b_n^2 + x^2} \right) \left\{ 1 - \frac{2b_n^2 [1 - \exp(-x)]}{x(b_n^2 + x^2)(2b_n^2 + 3x^2)} \right\}, \quad b_n = 2\pi n, \quad (9)$$

$$I(K, n, n') = \left[ \pi^2 (n - n')^2 + x^2 \right]^{-1} + \left[ \pi^2 (n + n')^2 + x^2 \right]^{-1}, \quad x = Kd. \quad (10)$$

in  $S_1$  and  $S_2$  the Green's function  $G(n, n, 0, \omega_l)$  corresponds to the direct production of an exciton, and  $G(n, u, 0, \omega_s)$  corresponds to its direct annihilation.

These processes can only take place if  $K=0$  (the small impulse of the light wave is neglected).

$$|G(n, n, 0, \omega)|^2 = \left[ (\omega - \omega'_g - n^2 \omega_\mu)^2 + \gamma^2 \right]^{-1}, \quad \omega_\mu = \omega_{0e} + \omega_{0h} - \omega'_g = \omega_g - \Delta\omega. \quad (11)$$

The Green's function  $G(n, n', K, \omega_l - \omega_{LO})$  corresponds to the emission of a phonon by an exciton both for the case of scattering in

the same zone ( $n = n'$ ) and for the case of transition to another zone ( $n \neq n'$ ). Its square modulus is equal to

$$|G(n, n', K, \omega_l - \omega_{LO})|^2 = \left[ \left( \omega_l - \omega_{LO} - \omega'_g - \omega_{0e} n^2 - \omega_{0h} n'^2 - \frac{\hbar K^2}{2m_e} \right)^2 + \gamma^2 \right]^{-1}. \quad (12)$$

### Frequency dependence of the scattering tensor

Let us first consider the case of scattering in the same zone, described by the function  $S_1$ . At frequencies  $\omega_l < \omega'_g + n^2 \omega_\mu$  all Green's functions are non-resonant (no real transitions) and  $S_1 \sim \alpha_0^2$ , which corresponds to background scattering.

At the frequency  $\omega \omega_l^{(1)} = \omega'_g + n^2 \omega_\mu$  real direct production of an exciton becomes possible, and at this frequency  $|G(n, n, 0, \omega)|^2 \sim \gamma^2$ . Since  $\gamma \sim \alpha_0$ , then  $S_1 \sim \alpha_0^0$ . At frequency  $\omega_l^{(1)}$  there is a peak that is  $\alpha_0^{-2}$  times higher than the background. In the frequency domain background. In the frequency domain

$\omega' + n^2 \omega_\mu < \omega_l < \omega'_g - n^2 \omega_\mu + \omega_{LO}$  (13) all Green's functions are non-resonant and  $S_1 \sim \gamma^2$ . Starting from frequency

$$\omega_l^{(2)} > \omega'_g - n^2 \omega_\mu + \omega_{LO} \quad (14)$$

becomes resonant  $G(n, n, K, \omega_l - \omega_{LO})$  (real phonon emission is possible). Therefore, with sufficient accuracy we can assume that

$$|G(n, n, K, \omega_l - \omega_{LO})|^2 = \\ = \frac{2\pi}{\gamma} \delta \left( \omega_l - \omega_{LO} - \omega'_g + n^2 \omega_\mu - \frac{\hbar K^2}{2m_e} \right). \quad (15)$$

If the parameter is  $\gamma / \omega_{LO} \ll 1$ , then when integrating over the variable  $K$ , the contribution of the pole of the function  $G(n, n, K, \omega_l - \omega_{LO})$  becomes dominant. Therefore, with sufficient accuracy we can assume that

$$|G(n, n, K, \omega_l - \omega_{LO})|^2 = \\ = \frac{2\pi}{\gamma} \delta \left( \omega_l - \omega_{LO} - \omega'_g + n^2 \omega_\mu - \frac{\hbar K^2}{2m_e} \right). \quad (15)$$

Then in the frequency domain (13)

$$S_1 = \frac{2\pi a_0^2 m_e}{\gamma \hbar} \sum_n I^2(K_0, n) \left[ (1+a_h)^{-3/2} - (1+a_e)^{-3/2} \right]^4 \times |G(n, n, 0, \omega_l)|^2 |G(n, n, 0, \omega_s)|^2 \quad (16)$$

$$K_0 = \sqrt{2m_e / \hbar} \sqrt{\omega_l - \omega_{LO} - \omega'_g + n^2 \omega_m},$$

those  $S_1 \sim \alpha_0$ . If  $\omega_l = \omega_l^{(3)} = \omega'_g + n^2 \omega_{LO} + 2\omega_{LO}$  (frequency corresponding to direct annihilation), then  $|G(n, n, 0)|^2 \sim \gamma^2$  and  $S_1 \sim \alpha_0^{-1}$ .

Thus, the frequency dependence  $S_1(\omega_l)$  has two peaks: weaker  $S_1(\omega_l) \sim \alpha_0^0$ , corresponding to real direct exciton production;

$$S_1 = \frac{2\pi a_0^2}{\gamma} \left( \frac{3\alpha_0}{8} \right)^4 \frac{2m_e^3}{\hbar^3} \left( \frac{\omega_h - \omega_e}{\omega_h + \omega_e} \right)^4 \sum_n (\omega_l - \omega_{LO} - \omega'_g + n^2 \omega_\mu)^2 \times |G(n, n, 0, \omega_l)|^2 |G(n, n, 0, \omega_s)|^2, \quad (17)$$

$$\omega_l \geq \omega'_g + \omega_{LO} + n^2 \omega_\mu.$$

The frequency dependence of  $S_2$  differs from the frequency dependence of  $S_1$  in that at frequency  $\omega_l^{(1)} S_2 \sim \alpha_0^{-1}$ , while  $S_1 \sim \alpha_0^0$ . The fact is that the function  $|G(n, n, K, \omega_l - \omega_{LO})|^2$  can be approximated by the  $\delta$  – function at frequencies

$$\omega_l^{(4)} = \omega'_g + \omega_c n^2 + \omega_v n'^2 + \omega_{LO}. \quad (18)$$

$$S_2 = \frac{2\pi a_0^2 m_e}{\gamma \hbar} \sum_{n>n'} \left[ I^2(K_{01, n, n'}) c(n, n', \omega_l) c(n, n', \omega_s) + I^2(K_{02, n', n}) c(n, n', \omega_l) c(n', n, \omega_s) \right], \quad (19)$$

Where

$$K_{01} = \sqrt{2m_e / \hbar} \sqrt{\omega_l - \omega_{LO} - \omega'_g - \omega_{0e} n^2 - \omega_{0h} n'^2}, \quad K_{02} = \sqrt{2m_e / \hbar} \sqrt{\omega_l - \omega_{LO} - \omega'_g - \omega_{0e} n'^2 - \omega_{0h} n^2}. \quad (20)$$

he part of formula (19), which depends on  $K_{02}$ , is valid in a wider frequency range  $\omega_l^{(5)} = \omega'_g + \omega_{0e} n'^2 + \omega_{0h} n^2$ . approaching  $\alpha_0 K_{01(2)} \ll 1$

strong peak  $S_1(\omega_l^{(3)}) \sim \alpha_0^{-1}$ , corresponding to real direct annihilation;

In the case of equal effective masses ( $m_e = m_h$ )  $S_1$  becomes zero.

In the limiting case  $\alpha_0 K_0 \ll 1$  (usually  $\alpha_0 \cong 10^{-5} \div 10^{-6} \text{ cm}$ ,  $K_0 \cong 10^3 \div 10^4 \text{ cm}^{-1}$  formula (16) is simplified and takes the form

This frequency interval includes the frequency  $\omega_l^{(1)}$ , at which  $|G(n, n', 0, \omega_l^{(1)})| \sim \gamma^{-2}$ , if the condition  $(n = n') \omega(n^2 - n'^2) \omega_v > \omega_{LO}$  real transition between bands  $nm$  and  $nm'$  with emission of LO phonon. Replacing  $|G(n, n', K, \omega_l - \omega_{LO})|^2$  in (7) with a  $\delta$  – function and integrating over  $K$ , for  $S_2$  we obtain the expression

$$I(K_0, n, n') \rightarrow (2/\pi^2) \left[ (n^2 + n'^2) / (n^2 - n'^2)^2 \right]$$

$$I(K_0, n, n') \rightarrow (2/\pi^2) \left[ (n^2 + n'^2) / (n^2 - n'^2)^2 \right]$$

and from (7), (8) we obtain

$$S_2 = \frac{16\alpha_0^2 m_e}{\gamma \pi^3 \hbar} \sum_{n>n'} \frac{(n^2 + n'^2)^2}{(n^2 - n'^2)^2} |G(n, n, 0, \omega_l)|^2 |G(n, n, 0, \omega_s)|^2, \quad (21)$$

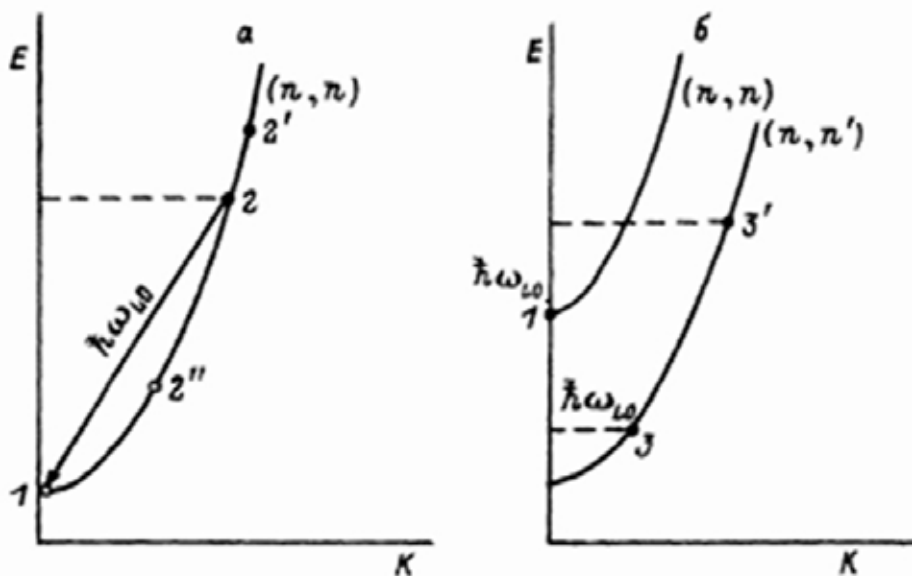
$$\omega_l \geq \omega'_g + \omega_{LO} + \omega_{0e} n^2 + \omega_{0h} n'^2.$$

#### 4. Discussion of the results obtained

When deriving formula (17), terms containing  $|G(n', n', 0, \omega_s)|^2$  are omitted, since the maxima of these functions are located outside the frequency interval  $\omega_l^{(5)}$ . Formulas (16) and (17) are valid in the vicinity of the maximum of the Green's function, which lead

to large values of  $S_1$  and  $S_2 \sim \alpha_0^{-1}$ . Function  $S_1$  contains one such maximum at  $\omega = \omega_l^{(3)}$ , corresponding to direct annihilation of excitons. Function  $S_2$  has two strong maxima: one at the frequency  $\omega_l = \omega_l^{(1)}$  (direct exciton production) and the other coinciding with the strong maximum of function  $S_1$ . Exciton transitions for the cases  $n \neq n'$  are shown in (Fig. 1).

**Figure 1.** Scheme of exciton transitions in the case of taking into account only one (a) and two (б) excitonic zones



1 – exciton energy after direct production or before indirect annihilation; 2, 2', 2'' and 3' – after indirect birth; 3 – before indirect annihilation.

$E$  is the energy of the exciton band,  $K$  is the modulus of the exciton wave vector.

The considered excitonic mechanism of two-phonon RRS leads to a sharp increase in the scattering cross section (scattering tensor  $S_{bg'b'g'} \sim a_0^{-1}$  at the resonant frequencies of the exciting light) compared to electron-hole pairs as intermediate states ( $S_{bg'b'g'} \sim a_0^2 \ln^2 a_0$ ).  $a_0^{-3} / \ln^2 a_0 a_0$ . Thus, there is an increase in scattering by  $a_0^{-3} / \ln^2 a_0 a_0$  times. From this we can conclude that in a quasi-two-dimensional electron system, the mechanism of two-phonon RRS is predominant. This conclusion seems justified specifically for two-phonon scattering, when the exciton appears only in the act of indirect creation (or indirect annihilation) and single emission of a  $LO$  - phonon. phonons, the question of the relationship between the contribution of the exciton mechanism and the EHP mechanism to scattering becomes more complicated. This is due to the fact that when a  $LO$  - phonon is emitted by a hot exciton, it

can go into the EHP state and then phonons will be emitted by the electron and hole. Without exploring the relative roles of the two scattering mechanisms in this paper. We only note that the dependence of the scattering tensor on the coupling constant  $\alpha_0$  in the case of MFRRS with a purely exciton mechanism. Remains the same as in the case of two-phonon RRS, since the appearance of an additional coupling constant in the numerator during the transition from  $N$  to  $N + 1$  emitted phonons will be compensated by the appearance of the constant  $\gamma \sim a_0$  in the denominator, which comes from the process of real emission of a phonon by an exciton .

The excitonic scattering mechanism in a bulk semiconductor is  $a_0^{-2}$  times weaker than scattering in a quasi-two-dimensional system. The enhancement of two-phonon scattering compared to the bulk case is explained by the fact that in a quantum well in the frequency range corresponding to direct production or direct annihilation of an exciton, real phonon emission is possible, while in a bulk semiconductor two-phonon scattering consists of two indirect processes – creation and annihilation exciton.

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