Theoretical treatment of the resonant hyper-Raman scattering in A²B⁶ semiconductors

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Abstract. In this work the theoretical treatment of the hyper-Raman scattering of light by LO-phonons is given for A²B⁶ semiconductors of the wurtzite structure, taking into account the influence of the complex top valence band which consists of the three closely-lying subbands.

1. Introduction

The hyper-Raman scattering (HRS) of light by phonons was observed in a number of semiconductors under two-photon excitation near the absorption edge [1-12]. In publications [13-17], devoted to theoretical investigations of the resonant HRS, the scattering process was considered within the framework of the model taking into account the hydrogen-like Wannier excitons as intermediate virtual states. This model enables one to explain many features observed in the HRS. In particular, an abrupt increase in the hyper-Raman intensity near the two-photon resonance with the lowest exciton state was reported to be found under experimental investigations of the resonant HRS by longitudinal optical (LO) phonons in a CdS crystal [11]. This phenomenon was explained by a scattering mechanism including two-photon dipole transitions to excitonic states of the s-type [16].

In this work the resonant HRS in direct-band hexagonal semiconductor crystals A²B⁶ is considered, taking into account the influence of the complicated top valence band structure.

2. Basic formulas

From a microscopic point of view the Stokes resonant HRS process can be described as follows: 1) absorption of two incident photons; 2) a creation of a phonon; 3) emission of a scattered photon [13,15]. In the initial and final states an electronic system of a semiconductor is assumed to be in the ground state. And intermediate virtual states are the excitons formed by a transition of an electron from the degenerate (or almost degenerate) valence band v to the excitonic level E_λ near the degenerate conduction band c. The wave functions of the relative electron-hole motion $\tilde{\chi}^{(\lambda)}_{j_e j_h}(\boldsymbol{r})$ are assumed to satisfy a system of the effective mass approximation equations [18-20]. Here the indices $j_e=1,2,\ldots s_c$ and $j_h=1,2,\ldots,s_v$ denote the states in the conduction band c and the valence band v, and s_c and s_v are the appropriate degeneracy multiplicities. Owing to small magnitudes of photon wave vectors the HRS process is supposed to occur in the center of a zone of Brillouin. Hence in this paper the excitons with the wave vector K=0 are considered.

In hexagonal semiconductors A²B⁶ the top valence band is known to be split into three closely-lying doubly degenerate subbands Γ_9, Γ_7 and Γ_7 by the crystal structure and the spin-orbit interaction

[21,22]. The transitions from them to the excitonic levels near the lowest doubly degenerate conduction band Γ_7 lead to the three hydrogen-like series A, B and C in the absorption spectra. If the scalar mass $\bar{\mu}_{j_e j_h}$ is introduced, then for $\tilde{\chi}_{j_e j_h}^{(\lambda)}(r)$ in these crystals the equation system can be written as

$$\left(\frac{1}{2} \frac{1}{\bar{\mu}_{j_e j_h}} \left(\hat{p}_x^2 + \hat{p}_y^2 + \hat{p}_z^2 \right) + U_{eh}(r) \right) \tilde{\chi}_{j_e j_h}^{(\lambda)}(r) + \sum_{j_h'=1}^{s_v} \hat{V}_{j_h' j_h} \tilde{\chi}_{j_e j_h'}^{(\lambda)}(r) = \left(E_\lambda - \varepsilon_{j_e j_h} \right) \tilde{\chi}_{j_e j_h}^{(\lambda)}(r) \qquad (1)$$

where $\hat{p}_\alpha = -i\hbar \, d/dr_\alpha$, $U_{eh}(r)$ is the Coulomb energy of the electron-hole interaction, $\varepsilon_{j_e j_h}$ is the energy gap. The operator $\hat{V}_{j_h' j_h}$ has a form

$$\hat{V}_{j_h' j_h} = \delta_{j_h' j_h} \frac{1}{2} \left\{ \left(\frac{1}{\mu_{j_e j_h}^\perp} - \frac{1}{\bar{\mu}_{j_e j_h}} \right) \left(\hat{p}_x^2 + \hat{p}_y^2 \right) + \left(\frac{1}{\mu_{j_e j_h}^{||}} - \frac{1}{\bar{\mu}_{j_e j_h}} \right) \hat{p}_z^2 \right\} + \left(\delta_{j_h' j_h} - 1 \right) D_{j_h' j_h}^{\beta\alpha} \hat{p}_\alpha \hat{p}_\beta . \qquad (2)$$

Here the effective mass parameters $D_{jj'}^{\alpha\beta}$ are defined by [18,20]

$$D_{jj'}^{\alpha\beta} = \frac{1}{2m} \delta_{jj'} \delta_{\alpha\beta} + \frac{1}{m^2} \sum_n \frac{\pi_{jn}^\alpha \pi_{nj'}^\beta}{\varepsilon_{jn}} \qquad (3)$$

where $\boldsymbol{\pi}_{jn}$ is the interband matrix element of the momentum operator.

The band structure of the considered crystals is mainly known [22]. For them the dipole transitions from the top valence band v and the lowest conduction band c to the higher-lying conduction band are possible. Besides, the selection rules allow the dipole transitions between the top and lower-lying valence bands. Thus, one can determine the nonzero matrix elements of the dipole transitions between these bands. This enables one to find the nonzero parameters $D_{jj'}^{\alpha\beta}$.

If the anisotropy influence on $U_{eh}(r)$ and $\mu_{j_e j_h}$ and the terms associating equations for different subbands in Eq. (1) are neglected ($\hat{V}_{j_h' j_h} \approx 0$), the equation system is reduced to the individual hydrogen-like equations [18,19] for which the wave functions $\chi_{j_e j_h}^{(\xi \ell m)}(r)$ and energies are well known [23]. Disregarded terms can be taken into account with the use of the perturbation theory.

Thus, considering $\hat{V}_{j_h' j_h}$ in the equation system (1) as perturbation, the first-order corrections for the lowest discrete energy levels $A_{n=1}$ and $B_{n=1}$ were found

$$E_{A_{n=1}}^{(1)} = \frac{2}{3} R_A \left\{ 2 \left(\frac{\bar{\mu}_A}{\mu_A^\perp} - 1 \right) + \left(\frac{\bar{\mu}_A}{\mu_A^{||}} - 1 \right) \right\} \qquad (4)$$

$$E_{B_{n=1}}^{(1)} = \frac{2}{3} R_B \left\{ 2 \left(\frac{\bar{\mu}_B}{\mu_B^\perp} - 1 \right) + \left(\frac{\bar{\mu}_B}{\mu_B^{||}} - 1 \right) \right\} \qquad (5)$$

where $\mu_{A(B)}$ and $R_{A(B)}$ are the effective mass and the exciton Rydberg for the A (B) excitonic series. Besides, the wave functions in the first approximation were obtained for these states. But appropriate expressions are not presented in this work because they too cumbersome.

3. The hyper-Raman scattering
In order to estimate the role of the first order corrections obtained the HRS by LO-phonons under two-photon excitation near the $B_{n=1}$ level was considered for the scattering geometry $z(xxz)y$.

As it is known, the two-photon dipole transition to s-exciton state can be described with the use of the three-band as well as two-band models, and their contributions are opposite in sign [15,24]. In accordance with the two-band model absorption of an photon is accompanied by the weakly-forbidden dipole transition from the ground state to an exciton of the p-type, another photon causes the intraband dipole transition between excitons [24,25]. In this work allowance was made for the dipole transitions between excitonic states of the different series too. The mechanism of the intraband Fröhlich exciton-lattice interaction is considered in the zero phonon wave vector approximation. Under calculations the dominant scattering mechanism including the two-photon transition to the $B_{n=1}$ exciton state was only taking into account. The Green's function approach was used under summation over intermediate excitonic states [15,26].

Taking into account the published data [22,27], the rough estimations of the frequency dependence of the resonant HRS cross section were performed for a CdS crystal. For comparison figure 1 demonstrates the frequency dependence of hyper-Raman cross section calculated for the wave functions and the energy in a first approximation and with regard for the dipole transitions between excitons relating to different subbands (1) and for unperturbed wave functions (2). As can be seen from the figure, the inclusion of these transitions and the first order correction can give rise to some increase in the cross section.
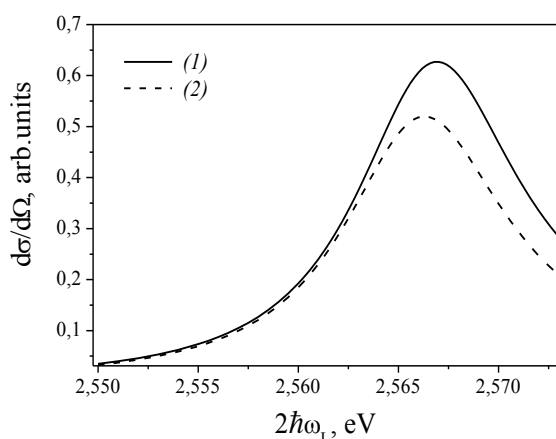

Figure 1. The HRS cross section calculated for the scattering mechanism including the two-photon transition to the $B_{n=1}$ exciton state in CdS as a function of the doubled energy of incident photons.

4. Conclusion

Taking into account the influence of anisotropy and the complicated structure of the top valence band with the use of the perturbation theory, for the hexagonal semiconductors A^2B^6 the wave functions and the energies in the first approximation were obtained for the $A_{n=1}$ and $B_{n=1}$ excitonic states. The HRS by LO-phonons near the two-photon resonance with the $B_{n=1}$ exciton level was considered for a CdS crystal. Rough estimations performed have shown that allowance made for the first order correction and the dipole transitions between the excitons of the different series do not lead to considerable change of the frequency dependence of the HRS cross section.

Acknowledgments

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