Resonant Raman scattering of light in the region of the ground state

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We obtained for the first time good agreement between the observed and theoretical frequency dependences of the cross sections of single- and two-phonon scattering in the region of the exciton ground state. It is shown that the asymmetry of the cross sections is due to the dispersion of the exciton band $n = 1$. We investigate the influence of absorption and of the exciton lifetime on the scattering cross section.

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The study of resonant Raman scattering of light in crystals is the subject of many papers (see, e.g., [1,2]). However, a comparison of experiment with theory is quite difficult, especially in the region of the ground state of the exciton, first because of the insufficient number of experimental points, which can usually be obtained with the aid of a set of exciting lines, and second because of the uncertainty introduced as a rule by absorption of the incident and scattered light. The results of such a comparison, recently obtained in [3,4] for single-photon scattering, are in our opinion far from complete in the frequency region where the cross section varies rapidly and nonmonotonically.

In this paper, using the method of scattering in mixed crystals, [5] we have investigated in detail the frequency dependence of the cross sections of single- and two-photon scattering by LO phonons in the region of the ground state of the exciton and propose a theoretical explanation of the observed effects. The experiments were performed on $Zn_xCd_{1-x}Te$ crystals in $X(YY)\bar{X}$ geometry at $T = 77\,^\circ K$, and the excitation was with an He-Ne laser. The strong dependence of the width of the forbidden-band of $Zn_xCd_{1-x}Te$ on x makes it possible to vary almost smoothly the frequency of the excitonic transition relative to the frequency of the exciting light.

Figure 1a shows the results of measurements of the two-phonon scattering line intensity. Attention is called to the fact that this dependence is patently asymmetrical—the resonance with the scattered light ($\Delta = 2$) is much more strongly pronounced than the resonance with the exciting light ($\Delta = 0$). A theoretical calculation of the amplitude of this process was carried out in the two-band approximation with allowance for the dispersion of the exciton bands, and of the dependence of the intraband matrix elements of the Fröhlich electron-phonon interaction on the phonon wave vector. The amplitude takes the form

$$A_q^{II}(\omega) \sim \sum_{pp_1p_2p'} K_{pp'}(0;\omega) K_{p_1-q^{(j)};p_2-q^{(j)}}(q;\omega-\Omega_q) \times [K_{p_2p'}(0;\omega-2\Omega_q) - K_{p_2-q;p'}(0;\omega-2\Omega_q)], \quad (1)$$

where

$$K_{pp'}(q,\omega) = \sum_{n=1}^{\infty} \frac{\psi_n(p)\psi_n(p')}{\omega - \epsilon_g + E_n - \frac{q^2}{2M} + i\Gamma} + \frac{1}{\pi a} \int \frac{k\,dk\,\psi_k(p)\psi_k(p')}{\omega - \epsilon_g - \frac{k^2}{2\mu} - \frac{q^2}{2M} + i\Gamma}. \quad (2)$$

Here ω is the frequency of the exciting light, Ω_q is the frequency of the emitted phonon, ϵ_g is the width of the forbidden band, R is the Rydberg constant for the exciton, n is the number of the exciton band, m_j are the masses of the electron and hole, Γ is the width of the exciton level, $q^{(j)} = (M/m_j)q$, $1/\mu = 1/m_1 + 1/m_2$, and $M = m_1 + m_2$. The matrix elements of the electron-phonon and electromagnetic interactions are taken at the center of the band and are not written out in (1). The calculations show that in the frequency region of interest to us the largest and most rapidly varying term of the amplitude is the term diagonal in n with $n = 1$. With accuracy sufficient for comparison with experiment, we can confine outselves to this term for the calculation of the cross section. The amplitude then takes the form

$$A_q^{II}(\omega) \sim \frac{v_0}{\pi a^3} \frac{1}{(qa)^2} \frac{1}{R^3} \times \left[\frac{(e^{iq^{(1)}r} - e^{iq^{(2)}r})_{11} (e^{iq^{(1)}r} - e^{iq^{(2)}r})_{11}}{(\delta - 1 - i\gamma)(\delta_1 - 1 - i\gamma)(\delta' - 1 - i\gamma)} \right], \quad (3)$$

where

$$\delta = \frac{\omega - \epsilon_g}{R}; \quad \delta' = \frac{\omega - \epsilon_g - 2\Omega_q}{R}; \quad \delta_1 = \frac{\omega - \epsilon_g - \frac{q^2}{2M} - \Omega_q}{R}.$$

a is the Bohr radius of the exciton and v_0 is the volume of the unit cell.

Investigations of the single-phonon scattering process show that resonance with the scattered light predominates in this case. The theory of first-order scattering cannot explain this fact. To explain the observed frequency dependence of the intensity of the single-phonon line we have proposed that in this case we are dealing with a second-order process. This assumption is based on the fact that in mixed crystals the concentration fluctuations lead to irregular static displacement of the atoms, which can be resolved into all possible types of zero-frequency displacements. To calculate the cross section we have assumed that the exciton is scattered most effectively by optical displacements of the LO type.

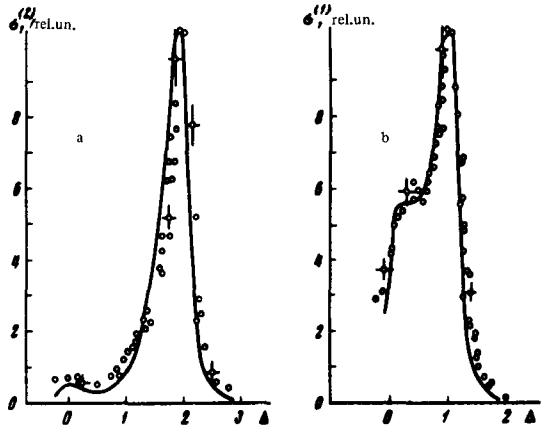

FIG. 1. Frequency dependence of the cross sections of two-phonon (a) and single-phonon (b) scattering; $\Delta = (\omega - \omega_g - R)/\Omega_0$; points-experimental data, solid curves-theory.

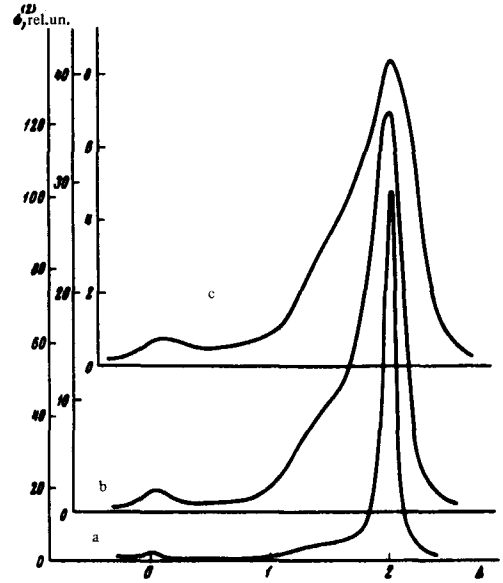

FIG. 2. Plots of $\delta^{(2)}(\Delta)$ at different values of Γ (theory): a—$\Gamma = 0.05\Omega_0$; b) $\Gamma = 0.15\Omega_0$; c) $\Gamma = 0.25\Omega_0$.

When considering the influence of the absorption on the intensity of the scattering it is necessary to take into account a fundamental feature of solid solutions, namely the smearing of the interband absorption edge as a result of fluctuations of the composition. An estimate of the characteristic energy according to[6], $\bar{\epsilon} = \Delta\epsilon_g c^2 (\Delta\epsilon_g m_1 d^2/\hbar^2)^3$ ($\Delta\epsilon_g$ is the difference between the widths of the forbidden bands of the mixed crystals, m_1 is the electron mass, d is the lattice constant, and c is the concentration) leads to a value $\bar{\epsilon} \approx 0.05$ eV. This is in good agreement with the energy $1/s = (2.5 - 5.0) \times 10^{-2}$ eV which determines the scale of variation of the absorption coefficient near the edge of the band. These estimates give grounds for assuming that in the solid solutions investigated by us the exciton line appears against a high-power background of intraband absorption, which changes little over the energy of two phonons ($\Omega_0 = 0.024$ eV). The scattering length is determined in this case by the large (but slowly varying) reciprocal of the absorption coefficient of the fluctuation background. In accordance with[8], the correction for absorption has in this case a negligible effect on the frequency dependence of the scattering cross section. The theoretical cross sections $\sigma^{(1)}$ and $\sigma^{(2)}$, plotted without corrections for absorption, are shown in Fig. 1 for the values $m_2/m_1 = 3$ and $\Gamma = 0.22\ \Omega_0$. For the crystals that are being mixed, m_2/m_1 is equal to 4 and 1 for CdTe and ZnTe, respectively; the value of $\Gamma(T = 77\,^\circ\text{K})$ obtained from reflection is $0.25\Omega_0$; an estimate of the fluctuation-induced $\Gamma = \Delta\epsilon_g c^{1/2} (Rmd^2/\hbar^2)^{3/4}$ yields $\Gamma = 0.2\Omega_0$. Both $\sigma^{(1)}$ and $\sigma^{(2)}$ are very sensitive to variation of Γ (Fig. 2). In particular, Γ influences the asymmetry, i.e., the ratio of $\sigma^{(1)}(1)/\sigma^{(1)}(0)$ and $\sigma^{(2)}(2)/\sigma^{(2)}(0)$. On the other hand, the asymmetry is also sensitive to the absorption, and an increase in the slope of the absorption curve leads to a decrease of the asymmetry. This has enabled us to analyze the accuracy of our assumption that the absorption coefficient is independent of the frequency. Since the half-widths of the maxima also depend on Γ (Fig. 2), it follows that ϵ cannot exceed $(1.0 - 0.5) \times 10^{-1}$ eV. At these values of $\bar{\epsilon}$, the theoretical $\sigma^{(1)}(\Delta)$ and $\sigma^{(2)}(\Delta)$ curves constructed with and without allowance for absorption are in equally good agreement with experiment, and the difference between the values of Γ lies within the limits of its measurement accuracy.

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