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Temperature dependence of the optical spectrum of the excitonic biphonon resonance

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Optical spectrum of the Raman scattering by one acoustical and one optical phonons in a quantum well — the spectrum of the excitonic biphonon resonance - has been experimentally investigated. The scattering spectra in two linear polarizations have been studied in their dependence on temperature, while the monochromatic optical excitation was linearly polarized. The co-polarized (with the excitation beam) spectrum was found to weaken, upon heating, homogeneously for all its components. In the cross-polarized spectrum, the effect of temperature was more pronounced yet inhomogeneous in the components: the high-energy side was found to be more sensitive to the heating.

Keywords: Raman scattering, spectroscopy, exciton.

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Inelastic light scattering in optical (LO) phonons in crystals was first observed by Mandelstam and Landsberg in 1928 [1] Later they involved Gross into their experimental research [2–4], who found in 1930 a fine structure of Rayleigh line caused by light scattering in acoustic phonons [5,6]. Theoretical predictions related to such scattering were published even earlier — in 1922 by Brillouin [7], and in 1926 by Mandelstam [8], and the effect then acquired its own name: Mandelstam–Brillouin scattering (or simply Brillouin scattering). At the same time light scattering in optical phonons is usually related to a wider class of phenomena, which are called Raman scattering for historical reasons.

Technically difficult experiments for inelastic light scattering gained a new momentum with appearance of the powerful laser sources of monochromatic light. In the beginning of 1970s Permogorov and Travnikov found lines of multiple light scattering in LO-phonons in crystals of A_2B_6 semiconductors. They related the observed broadening of these LO-replicas of incident radiation frequency with additional scattering in acoustic phonons [9,10]. Then several messages appeared about detection of a fine structure of LO-repetitions, which the authors to some extent associated with the combined processes involving one optical and strictly one acoustic phonons [11–13]. As for the conditions of spectra excitation and observation of clearly marked dispersion of acoustic satellites, the understanding was gradually achieved that the scattering mechanism was due to polaritons [14].

In the recent article [15] the optical spectrum with the permitted central LO-replica and its four acoustic satellites was first found for a quantum well. Obviously, the above mechanism is not possible here, since there are no polariton states oriented along the axis of the structure growth. On the other hand, the resonance behavior also differs from the case of volume crystals: the resonance is narrow, within the exciton contour, and Stokes shift of all components in the optical spectrum is fixed. Paper [15] shows that in the quantum well the optical spectrum of scattering is formed by the process with three resonant denominators involving $2s$-exiton state. This process, resulting in narrow acoustic satellites LO±LA, LO±TA of the central peak, will be called exciton biphonon resonance (EBR) for brevity.

This paper continues the experimental research of the EBR optical spectrum properties. The behavior of the EBR

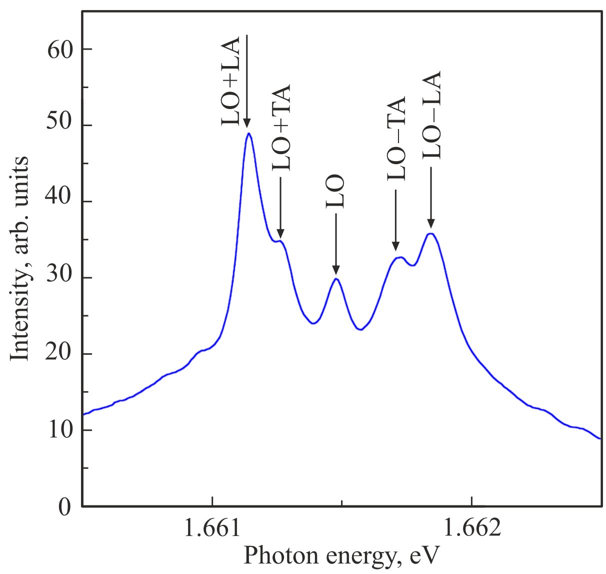

Figure 1. Optical spectrum of EBR in the area of maximum of the exciton main state line ($1s$ $1e1hh$) in the quantum well. $T = 8$ K. EBR line interpretation is given according to [15]. Excitation — 20.88 meV above the line marked as LO.

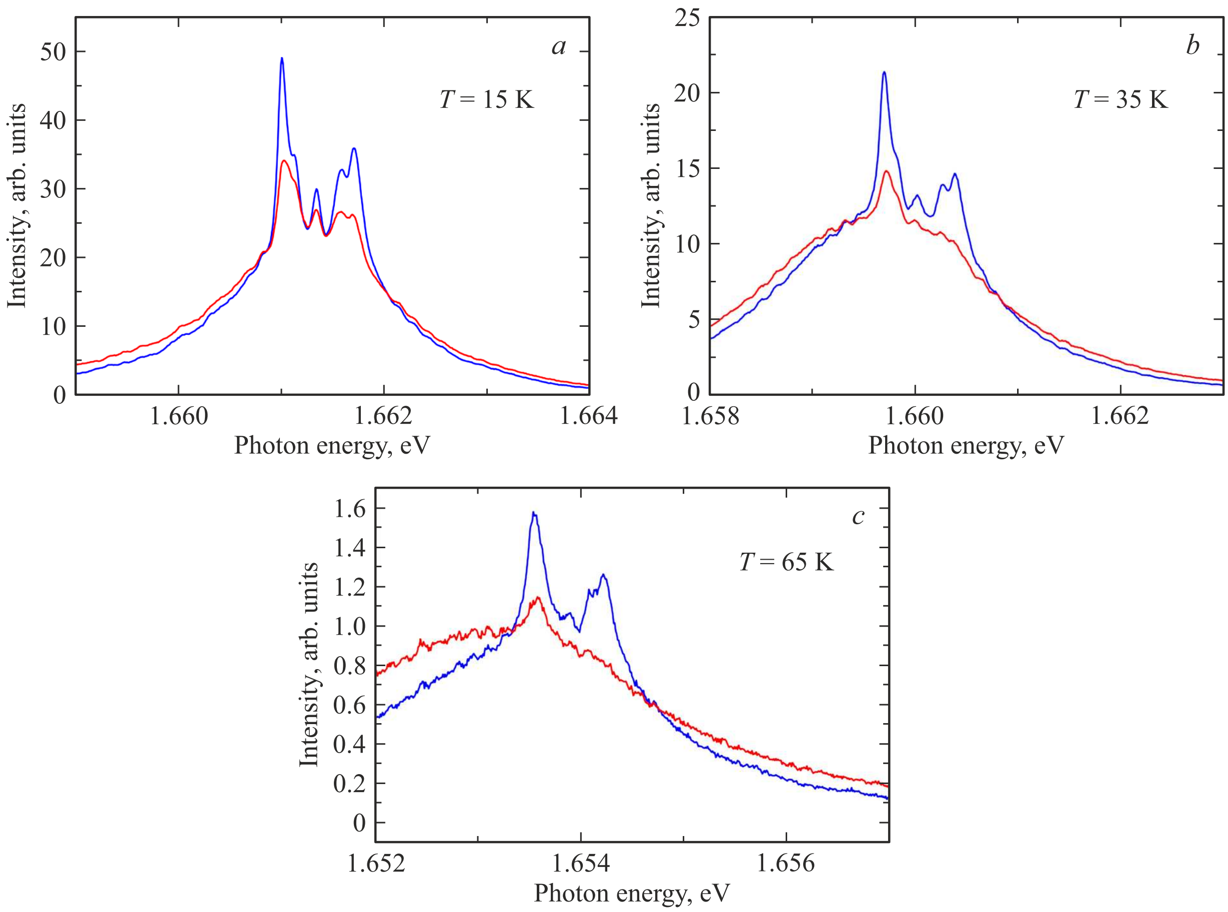

Figure 2. Temperature evolution of polarized EBR spectra. Blue is for the intensity of luminescence, polarized as excitation, red — of cross-polarized luminescence. The energy of the exciting quanta at each temperature is selected so that the EBR spectrum is near the maximum of the luminescence line.

lines in the range from helium to nitrogen temperatures has been studied for the first time. It was found that in the crossed polarization the shortwave acoustic satellites weaken and disappear with growth of temperature.

The specimen for the research contained an isolated quantum well (001)-CdTe, selectively doped with rhenium. See the description of the specimen and the method in [15]. The specimen was placed in a cold conduit of a helium-transport cryostat. Excitation via a tunable laser and scattering registration were done through a microlens. Spectral resolution determined using the observed semi-width of Rayleigh scattering line at the distance from the resonant cavities was less than $30\,\mu$eV. Note that the study that we conducted earlier for various types of spin flop combination scattering in quantum wells based on CdTe [16,17] showed that high-resolution spectra contain information that is not available for the spectral resolution values usually achieved in the experiment [18,19].

Fig. 1 shows the spectrum of secondary luminescence of the quantum well (001)-CdTe/(Cd,Mg)Te with width of 18 monolayers (around 6 nm) when excited approximately by energy of optical phonon above the main exciton state $1s\ 1e1hh$. EBR spectrum prevails in the radiation — five lines with width of around $100\,\mu$eV each, occupying in general the interval of around 1 meV. Note that the light of photoluminescence, which is usually the main optical response of the quantum well to excitation, for the shown spectral interval plays a small part: the contour of the exciton luminescence is seen as the smoothly changing background (with intensity of around 10 units), since its width ($\sim 3-4$ meV) closes the spectral window of the detector. „The central" line marked as LO, corresponds to the scattering with the optical phonon emission. Four of its symmetrically located satellites respond to scattering by emission of one optical and emission/absorption of one acoustic phonon (transverse or longitudinal). It is interesting that the intensity in the central line is lower than in the side ones. The reason for this consists in the difference of the mechanisms forming the central line and the EBR satellites [15].

Fig. 2 presents the polarized EBR spectra at several temperatures in the range from 15 to 65 K. In general, as the temperature increases, there is a shift of the main exciton state $1s$ towards the smaller energies as a result of the total temperature narrowing of the prohibited zone (Varshni law). One can also note the trend of increased contribution of luminescence compared to EBR contribution. While considering separately a series of copolarized spectra, we come to the conclusion that they are hardly affected by temperature. One might as well state the decrease in the contribution of the central line compared to the actual EBR lines. The cross-polarized part depends on temperature much stronger. Here two groups of the lines behave in a different way. EBR lines LO+LA, LO+TA, lying below the spectrum middle by their energy, weaken as the temperature increases much faster than the same lines in the copolarized spectrum: if at 15 K the ratio is approximately 2 : 1 in favor of the copolarized spectrum lines, at 65 K it is already closer to 4 : 1. But the behavior of the pair of lines LO−LA, LO−TA located above the middle of the EBR spectrum changes more drastically. These lines nearly disappear in the cross-polarized spectrum already at 35 K.

The reasons for the strong temperature dependence of the cross-polarized EBR spectrum, especially its high-energy side, are not currently clear. It occurs that high-energy EBR lines are sort of lying „at the anti-Stokes side" by acoustic phonons, i. e. the absorption of the acoustic phonon must take place in the optical transition, so that the occupation factor would manifest itself — increased concentration of thermal phonons. However, in this case it would be logical to expect the increased contribution of high-energy satellites compared to the low-energy ones with temperature growth regardless of the polarization. The experiment demonstrates a totally different effect in the cross-polarized spectrum and absence of any pronounced effect — in the copolarized one. Further development of the EBR theory is necessary with account of polarization.

Therefore, we studied the temperature behavior of the EBR spectrum in the range from helium to nitrogen temperatures with the resolution by linear polarization (under linearly polarized excitation). It was found that the EBR spectrum copolarized with the exciting beam gradually decreases when heated by intensity (not only by absolute value, but even in comparison with the photoluminescence), and also evenly for all EBR lines. However, in the cross-polarized spectrum the temperature effect is expressed stronger and unevenly in the lines. High-energy satellites LO−LA, LO−TA, which may hardly be seen in the spectrum already at 35 K, are especially sensitive to heating. To understand the obtained results, further development of the EBR theory is required.

Conflict of interest

The author declares that they have no conflict of interest.

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