Appearance of surface and bulk polaritons in luminescence spectra of ZnTe crystals

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It is shown that the low-temperature luminescence spectra of ZnTe crystals in the region of the ground state have a complex structure which is due to the appearance of surface polaritons, together with bulk polaritons.

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In recent years studies have shown that the structure of low-temperature spectra of GaAs,[1] ZnTe, $Zn_xCd_{1-x}Te$,[2] CuCl,[3] ZnSe,[4] and CdTe[5] crystals in the region of the exciton ground state ($n = 1$) is formed by definite parts of both (upper and lower) branches of bulk polaritons. Moreover, it is well known that the crystal surface, on which the conversion of bulk polaritons to photons also naturally occurs, plays an important role in the formation of exciton-polariton luminescence spectra. Since there are also surface polariton states on the crystal surface it is possible, in principle, to expect the simultaneous appearance of bulk and surface polaritons in the luminescence spectra. Until now, surface polaritons have been successfully observed only by the method of total interior disruption (TID).

In this work experimental results are given for the first time concerning the observation of surface polaritons in exciton-polariton luminescence spectra.

Low-temperature reflection and luminescence spectra were studied for ZnTe crystals grown from the gaseous phase, in the region of the exciton ground state ($n = 1$). The measurements were carried by a photographic method on a diffraction spectrograph with a dispersion 7.2 Å/mm. The crystals were excited by an argon-ion (Ar^+) laser with an output beam power of about 50 MW.

Figure 1a shows a reflection spectrogram for the ZnTe crystals studied at $T = 4.2$ K in the region of the fundamental absorption edge. The structure of the reflection spectrum with a sharply defined minimum at $\lambda = 5206$ Å and a weak minimum at $\lambda = 5212$ Å is due to the formation of excitons in the ground state ($n = 1$). It should be noted that for the observed shape of the reflection band, in the absence of the so-

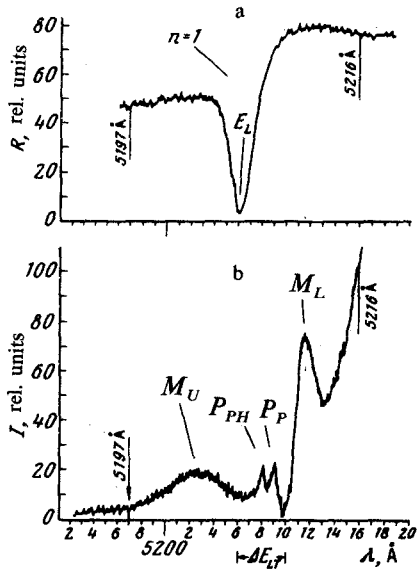

FIG. 1. Spectra: a-reflection and b-emission of ZnTe crystals in the region of the exciton ground state ($n = 1$) at $T = 4.2$ K.

called "spike," its minimum corresponds to the energy of the longitudinal exciton E_L.[1]

Figure 1b shows the luminescence spectrogram for the same samples in the exciton resonance region at $T = 4.2$ K and a low density of excitation by the argon-ion laser. Evidently, four emission bands denoted by M_U, P_{PH}, P_P, and M_L are observed in the given region. Their maximum intensities occur respectively at $\lambda = 5202$ Å, $\lambda = 5208$ Å, $\lambda = 5209$ Å, and $\lambda = 5211$ Å. In Ref. 2 it is shown that the M_U band is due to emission of the upper polariton branch (UPB), and the M_L band to the lower polariton branch (LPB) from the region of its "narrow-neck," i.e., the aforementiond bands represent emission from definite regions of the dispersion branches of bulk polaritons. The energy gap between the short-wavelength edge of the LPB emission ($\lambda = 5210$ Å) and E_L characterizes the value of the longitudinal transverse splitting ΔE_{LT} of the exciton ground state ($n = 1$) which for ZnTe crystals is 1.5 MeV. Therefore, it can be seen from Fig. 1b that the emission of bulk polaritons occurs outside the region ΔE_{LT}.

The P_{PH} and P_P bands occur within the ΔE_{LT} region and cannot be attributed to the emission of bulk polaritons. Actually, on the one hand this region is characterized by total internal reflection for bulk polariton emission. On the other hand, bulk polariton emission comprises only those regions of their dispersion branches for which there is, along with the exciton, also a photon component. However, within the ΔE_{LT} region the LPB polaritons are in fact pure excitons. Photons or photon-like bulk-branch polaritons with energies corresponding to the ΔE_{LT} region do not occur in the crystal. Consequently, the observation of bulk polariton luminescence in this region is fundamentally impossible.

Meanwhile, the doublet structure of the luminescence spectra for ZnTe crystals

within the ΔE_{LT} region is well explained by taking the surface polariton states into consideration, despite the fact that their appearance in ordinary luminescence spectra is forbidden by the law of the conservation of momentum. Actually, it has been shown theoretically in Ref. 6 that such a situation occurs only for an ideally-plane crystal edge and by neglecting scattering processes of surface polaritons by acoustic phonons. In this same work it has been shown that by taking these factors into account the given exclusion is removed and it is possible to observe the emission of surface polaritons in exciton-polariton luminescence spectra in the form of a band doublet within the region ΔE_{LT}. The following fact is important in explaining the reasons for the origin of the said doublet structure of the surface polariton luminescence spectrum. The probability for the scattering of surface polaritions by acoustic phonons is a maximum near $E = E_S$, where E_S is the maximum energy of the surface polaritons, since their exciton component is large in this region. For energies $E \gtrsim E_T$ (where E_T is the energy of the LPB bulk polaritons), the surface polaritons are basically photons. Thus, they interact weakly with the phonons and cannot contribute to the luminescence spectra. However, near E_T the probability for the scattering of surface polaritons by surface irregularities is a maximum. Inasmuch as the structure of the luminescence spectrum is the product of the density of surface polariton states and their scattering cross section, the difference in the scattering cross sections for irregularities and acoustic phonons leads to the formation of a doublet structure in the luminescence spectrum for surface polaritons.

In conclusion, we note that on increasing the temperature to 20–30 K a redistribution of the intensities between the P_{PH} and P_P bands is observed. The intensity of the P_{PH} band increases somewhat in comparison with P_P. This type of temperature evolution of the emission doublet for surface polaritons has been predicted theoretically in Ref. 6. However, this question along with an analysis of the temperature changes in the emission spectra of bulk polaritons will be considered in more detail separately.

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