Singularity of polariton radiation of ZnTe crystals at high excitation levels

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We investigated the emission spectra of ZnTe crystals ($T = 4.2$ and $1.6\,^\circ$K) excited by an argon laser. The results are interpreted in terms of polariton luminescence of the lower and upper polariton branches. The appearance of a narrow peak—"needle" (half-width $\approx 10^{-5}$ eV) for the emission of the upper polariton branch, at $T = 1.6\,^\circ$K and at an excitation density 4×10^{21} photons/cm^2 sec, is attributed to the possibility of attaining Bose–Einstein condensation of the excitons on the upper polariton branch in a state with $k \approx 0$.

If crystals are laser-excited in the region of the essential exciton-photon mixing, one can expect to observe singularities due to the high excitation level in the spectra of their polariton luminescence. No such effects have been observed so far for polaritons. We discuss in this communication the results of investigations of the emission spectra of ZnTe crystals strongly excited near resonance.

The spectra were measured photographically using a diffraction spectrograph with naturally cleaved surfaces of ZnTe crystals grown by the sublimation method. The luminescence of the investigated crystals was excited by an argon laser with emission wave vectors $k_1 = 1.29\times10^5$ cm^{-1} (4880 Å) and $k_2 = 1.22\times10^5$ cm^{-1} (5145 Å) close to the value $k = 1.21\times10^5$ cm^{-1} required for the excitation of polaritons in ZnTe crystals at $T = 4.2$ and $1.6\,^\circ$K in the region of the fundamental ($n = 1$) exciton band. The shape of the reflection curve of the latter reveals singularities typical of spatial-dispersions effects, namely the absence of a pronounced maximum and a sharply pronounced minimum (Fig. 1a).

Figure 1b shows a microphotograph in the exciton-resonance region.[1)] At $T = 4.2\,^\circ$K and at an excitation density $P_{exc} = 4\times10^{21}$ photons/cm^2 sec, the spectrum consists of three bands M_l', M_l'', and M_u, the maxima of which correspond to $\lambda = 5208$ Å, $\lambda = 5206$ Å, and $\lambda = 5202$ Å. Comparison of this spectrum with the reflection spectrum (obtained during the time of laser action) has shown that the maximum of the M_l' band corresponds to the energy of the transverse exciton (E_T), while M_l'' corresponds to the minimum of the reflection curve. The band M_l, on the other hand, is shifted somewhat towards the short-wave side of the state of the longitudinal exciton (E_L) which occurs in the region of the minimum of the reflection curve in this case.[1]

Such a structure in the resonance region agrees with the assumption that it is of polariton origin. Indeed, the band M_l' can be ascribed to the emission of the lower polariton branch (LPB) in the "bottleneck" region, while M_l'' can be ascribed to the singularity in the passage of the polaritons from the crystal to the vacuum in the vicinity of the minimum of the reflection curve.[2,3] The band M_u (half-width $\approx 3\times10^{-3}$ eV) can be interpreted as the emission of the upper polariton branch (UPB), in analogy with the case of GaAs.[1]

Lowering the temperature to $1.6\,^\circ$K at the same value of P_{exc} leads to an appreciable change in the shape of the emission band of only the UPB (Fig. 1c). Instead of the M_u band there appears at $\lambda = 5201$ Å a narrow peak—"needle" (half-width $\approx 10^{-4}$ eV, indicated by an arrow in Fig. 1c)—and a weak background on the long- and short-wave sides of the peak. Lowering of P_{exc} to 10^{19} photons/cm^2 sec leads at $T = 1.6\,^\circ$K to a vanishing of the M_l' and M_l'' and of the "needle" from the spectrum, while the band M_u itself is distinctly observed. Thus, the shape of the UPB emission band exhibits a distinct dependence on the temperature and on the excitation density. It should be noted that the intensity of the "needle" is lower than that of the M_u band. This indicates that its onset, in all probability, is not connected with effects of amplification or of the stimulated emission, it being quite doubtful whether these effects are possible at all in this region. Indeed, there are no photons of this frequency in the crystal, since the corresponding polaritons are converted into light only on the surface, as they leave the crystal. Consequently, there should be no amplification of the photons with this frequency in the crystal in the case of the polariton mechanism.

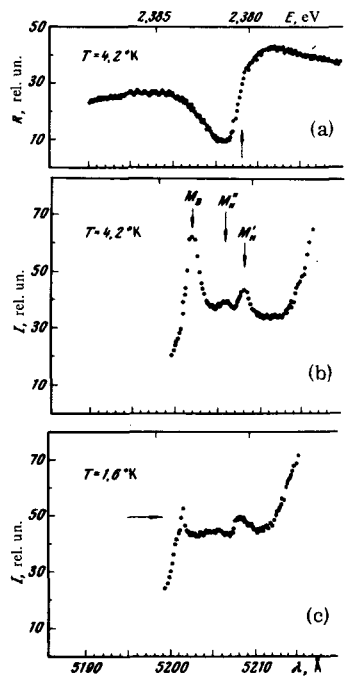

FIG. 1.

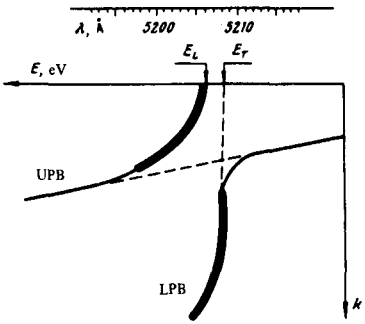

FIG. 2.

At the same time, the indicated singularities for the M_u band can be attributed, in our opinion, to Bose-Einstein condensation of the exciton (B-ECE) on the UPB in the state with $k \approx 0$. The feasibility in principle of condensation on this branch was noted by Knox.[4] Indeed, the B-ECE as a macroscopic filling of the lowest energy state near $k = 0$ is impossible on the LPB, since the polaritons of this branch are already actually transverse photons in this region, and the condensation would take place outside the crystal. For the UPB the situation is different, its polaritons in the region $k \approx 0$ are essentially excitonlike, and their Bose condensation at high density is possible.

Thus, assuming Bose condensation on the UPB, the needle-shaped peak can be attributed to the coherence of the recombining excitons of the Bose condensate. Its intensity is proportional to the concentration of the excitons in the condensate, which is certainly less than produced concentration of the excitonlike polaritons on the UPB. This causes the observed ratio of the intensities of the "needle" and of the M_u band.

The background against which the "needle" is observed can be attributed to the appearance of collective excitations of the phonon type in the high-density exciton system. It follows from the results that the critical temperature (T_{cr}) for the B-ECE on the UPB is not less than 1.6° for the employed P_{exc}. In this case, i.e., when $T_{cr} \neq 0$, the emission due to the collective excitations of the phonon type in the high-density exciton system should be present, according to[5], on both the long- and the short-wave sides of the "needle". It must be emphasized that the use of the results of a calculation of the shape of the emission band in Bose condensation, developed for excitons,[5,6] is justified in our case by the essentially excitonlike character of the UPB polaritons in the region $k \approx 0$ (see Fig. 2). The momentum conservation law is satisfied in the discussed case when account is taken of the crystal surface.

An estimate of the critical concentration (n_{cr}) for the B-ECE on the UPB from the critical temperature yields $\approx 6 \times 10^{15}$ cm^{-3}, and an estimate of the produced exciton density in the crystal at the employed P_{exc}, without allowance for the polariton character of the excitations, yields $\approx 3 \times 10^{16}$ cm^{-3}. These differences between the concentrations can be attributed in principle to the fact that in the second estimate of the concentration no account was taken of the presence of two branches and of the possible scattering of the excitations between them.

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[1)]While retaining the term "exciton," we use it in the sense of the exciton limit of the polariton branches (the corresponding regions of the polariton branches are shown thicker in Fig. 2).

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