Inelastic scattering of upper-branch polaritons and their Bose–Einstein condensation

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The experimental manifestations of the interaction of the upper-branch polaritons, as well as the condition necessary for their appearance, are discussed.

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When a large excitation density is produced on the upper polariton branch (UPB) one can expect an interaction of the inelastic-scattering type between the polaritons. This paper reports the first experimental proof of this effect and discusses the conditions under which interactions between the upper-branch polaritons are possible.

The emission and reflection spectra of ZnTe and $Zn_xCd_{1-x}Te$ single crystals grown from the gas phase were investigated at $T=1.6$ °K. The investigations were made mainly with an argon (Ar^+) laser having a quantum energy close to the energies of the exciton resonances of the given crystals.

Figure 1a shows a microgram of a ZnTe crystal emission spectrum obtained at $T=1.6$ °K and at an Ar^+-laser excitation density $R_{exc}^{Ar^+}\approx 5\times10^{21}$ phot/cm^2 sec. The band I_1 ($\lambda=5221$ Å) is due to radiative recombination of the excitons localized on the neutral acceptor, the band M_L' ($\lambda=5208$ Å) is due to the emission of the lower polarization branch (LPB) in the "bottleneck" region.[1] Located between them is a narrow band P_U ($\lambda=5216$ Å), which is revealed by excitation with a mercury lamp or a nitrogen (N_2) laser with a quantum energy 1.5 eV larger than the widths of the forbidden bands of the investigated crystals. When an N_2 laser is used, there is likewise no emission-spectrum structure consisting of the bands M_L', M_L'', and M_U in the immediate region of the exciton resonance. The latter circumstance confirms the polariton character assumed for these bands in[1] and indicates furthermore that the bands P_U, M_L', M_L'', and M_U are of similar nature, which appears to be due to the peculiarities of excitation by an Ar^+ laser. It is also obvious that the presence of the indicated four bands cannot be attributed to the appearance of additional exciton-localization centers with increasing R_{exc}, since the N_2-laser-induced $R_{exc}^{N_2}\approx10^{23}$ phot/cm^3 sec exceeds $R_{exc}^{Ar^+}$ by more than one order of magnitude.

The P_U band possesses the following properties: 1) Its energy position has mirror symmetry, with respect to the band Q ($\lambda=5186$ Å), relative to the "needle" that serves as the optical attribute of Bose-Einstein condensation (BEC) on the UPB.[1] The band Q, on the other hand, is due to exciton emission from the excited ($n=2$) state. 2) The emission intensity depends quadratically on the excitation level, thus indicating the character of the processes that cause its appearance.

The foregoing aggregate of the properties of the P_U band indicates that if a large excitation density is produced on the UPB, then an interaction of the inelastic-scattering type takes place between them. One UPB polariton, whose energy is increased by the inelastic scattering, goes over to an excited ($n=2$) state. The other UPB polariton, with an appropriately decreased energy (≈6 meV), manifests itself on leaving the crystal in the form of photons having the energy of the P_U band. Thus, the inelastic scattering of the UPB polaritons, and also the properties of the P_U band, are analogous in many respects to their counterparts in the LPB. However, the half-width of the P_U band ($\approx10^{-4}$ eV) is much smaller than the corresponding half-widths of the P bands in inelastic scattering of the LPB polaritons ($\approx10^{-2}$ eV) in II-VI semiconductors. This appears to be a peculiarity of the inelastic scattering by UPB and is due to the BEC of its polaritons. Indeed, in the case of BEC, a macroscopically large number of the UPB polaritons is in the lowest energy state with $k\approx0$. Consequently the kinetic energy of their initial and final states, which causes the short- and long-wave tails of

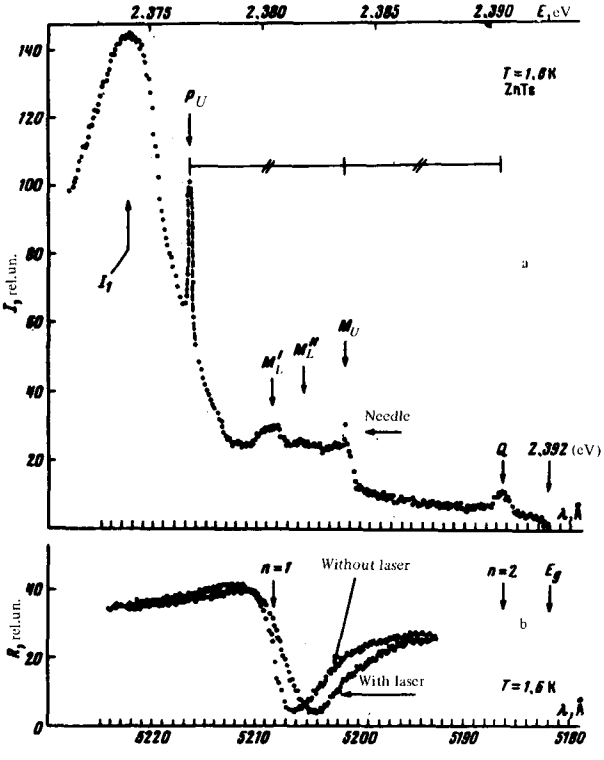

FIG. 1. Micrograms: a) emission spectrum of the ZnTe crystal at 1.6 °K and $R_{exc}^{Ar^+}\approx5\times10^{21}$ phot/cm^2 sec; b) reflection spectrum obtained for $T=1.6$ °K in the presence of laser excitation with $R_{exc}^{Ar^+}\approx10^{21}$ phot/cm^2 sec.

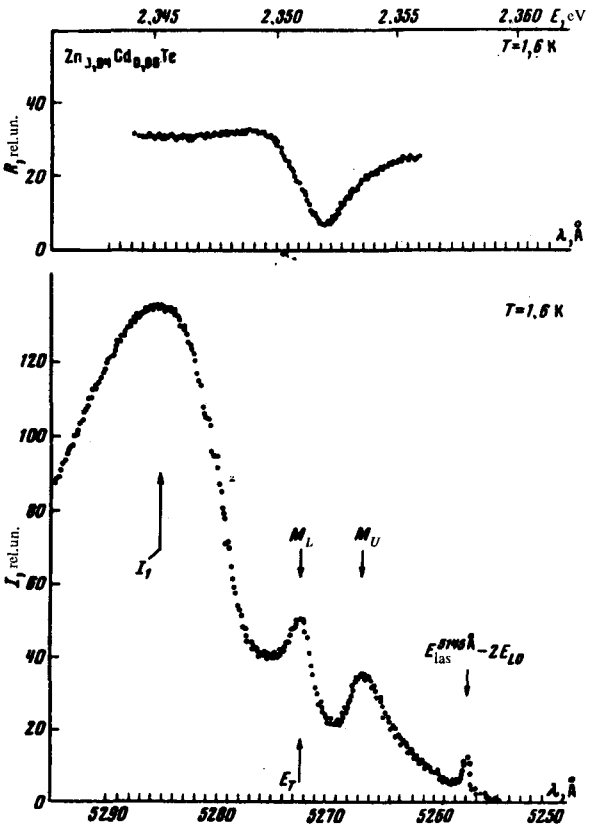

FIG. 2. Micrograms of the reflection and emission spectra of a $Zn_{0.94}Cd_{0.06}Te$ crystal at $T = 1.6$ °K and $R_{exc}^{Ar^+} \approx 5 \times 10^{21}$ phot/cm² sec.

the P bands, tends to zero. These tails are therefore missing from the P_U band, which takes the form of a narrow peak ("needle"). This conclusion is arrived at also from the following consideration: The presence of BEC for UPB polaritons is tantamount to the condition that their temperature is zero. In this case, as shown theoretically by Khadzhi for the inelastic scattering of excitons without allowance for the retardation effect, the shape of the P band tends to that of a δ function with negligible half-width.[2]

The short-wave shift of the reflection spectrum (and accordingly of the absorption spectrum) during the action of laser excitation at $T = 1.6$ °K (Fig. 1b) means that the repulsion forces predominate in the investigated system.[3,4]

Thus, the optical properties of the semiconductors in the case of BEC, i.e., the shift of the absorption band towards higher energies and the accompanying inelastic scattering, which were first predicted by Keldysh without allowance for the exciton-photon interaction,[3] actually exist also when the UPB is used to attain the condensation. Analogous statements are valid also with respect to the results of Moskalenko,[4] Buttner,[7] Hanamura,[8] and Khadzhi.[2]

For the effects of the interaction of the UPB polaritons to appear, including their inelastic scattering and BEC, it is necessary in my opinion to have the following:

1. $\nu_{exc\text{-}phot} > \nu_{exc\text{-}phon}$, where $\nu_{exc\text{-}phot}$ and $\nu_{exc\text{-}phon}$ are the frequencies of the exciton-photon and exciton-phonon collisions, respectively. This relation can apparently be satisfied mostly in the case of weak exciton-phonon interaction. The weakness of the latter in ZnTe crystals follows in part from the absence of phonon satellites of the M_L' bands. On the other hand, the exciton-phonon interaction constant g for the ZnTe crystals is equal to 0.238,[5] much less than unity, i.e., according to Toysawa,[6] the exciton-phonon interaction is actually weak. In this case the investigation must be carried out at $T < E_{LT}/k$, where E_{LT} is the longitudinal-transverse splitting and k is Boltzmann's constant.

2. $\nu_{exc\text{-}phot} > \nu_{exc\text{-}exc}$, where $\nu_{exc\text{-}exc}$ is the frequency of the exciton-exciton collisions. For ZnTe crystals, E_{LT} is of the order of 1.2 meV, and for $R_{exc}^{Ar^+} \approx 10^{21}$ phot/cm² sec we have $\nu_{exc\text{-}phot} \sim 10^{12}$ sec^{-1} and $\nu_{exc\text{-}exc} \sim 10^{11}$ sec^{-1}.

3. If conditions 1 and 2 are satisfied, the difference between the energies of the laser quanta (E_{las}) of the longitudinal exciton (E_L) should correspond to, and perhaps be a multiple of, the energy of the longitudinal optical (LO) phonon (E_{LO}).

The statements made in the last paragraph are proved by the following experimental facts: For ZnTe crystals we have $E_{las}^{5145\,Å} - E_L = 26$ meV, which is close to E_{LO}. In this case, UPB polaritons are produced with an overwhelming probability in the excitation process. The probability of polariton scattering by LO photons is large. Consequently, after the UPB polaritons lose rapidly their excess excitation energy as a result of intrabranch scattering by the LO phonons, their density in the region $k \approx 0$ becomes large. The latter are coherent in a certain sense, since their energy spread (and in part their k spread) is determined in many respects by the energy spread of the laser quanta, which is quite small. This seems to favor their BEC at a concentration higher than critical.

This process of polariton scattering by LO phonons should not be set in correspondence with resonant Raman scattering. Under these conditions the two effects are identical.

Under analogous experimental conditions, i.e., $T = 1.6$ °K and $R_{exc}^{Ar^+} \approx 5 \times 10^{21}$ phot/cm² sec, UPB emission is observed also for $Zn_{0.94}Cd_{0.06}Te$ crystal (band M_U in Fig. 2). There is, however, no interaction between its polaritons. In this case $E_{las}^{5145\,Å} - E_L = 2.2E_{LO}$, and the population of the region $k \approx 0$ of the UPB proceeds after these polaritons lose rapidly the excess excitation energy on two LO phonons as a result of interaction with acoustic phonons, which greatly decreases the rate of their energy relaxation. Since the group velocity of the UPB polaritons is large, the majority of the polaritons manages to leave the crystal before the interaction with the acoustic phonons takes place. Consequently it is less likely that a large density of the UPB polaritons will be produced in the $k \approx 0$ region with participation of acoustic phonons in the process of intrabranch scattering.

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