

Effect of the surface and size of specimens on polariton kinetics and polariton trapping

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The effect of the finite dimensions of specimens on polariton kinetics and polariton trapping is investigated experimentally for the first time. It is found that the lifetime of the polaritons decreases in thin specimens. An experimental estimate of the diffusion length of polaritons ($L \sim 5\text{--}7 \mu\text{m}$) is obtained for high-quality CdS specimens.

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The study of the properties of the surface and near-surface regions, which have a considerable influence on different properties of semiconductors, is attracting the close attention of researchers. The influence of the surface increases in small specimens. In this work, we investigated the influence of the specimen thickness on the kinetics and trapping of polaritons.

The emission spectra of the exciton state $n = 1A$ of CdS crystals excited with the 476.5-nm laser line and at $T = 2K$ in specimens with different thicknesses (d) were investigated under the same conditions of excitation and recording. The experiments were performed on perfect plates, grown out of the gas phase without special doping. In all, about 40 specimens were studied. In the region $d \lesssim 2 \mu\text{m}$, we used specimens in which interference of additional waves was previously observed.¹ From this set, specimens of different thicknesses with longest polariton lifetimes (τ) were chosen. The specimens were selected on the basis of the form of the 2LO luminescence band, arising after scattering of polaritons by two LO phonons from the resonance region. As is well known,² the form of this band reflects the polariton energy distribution function $N(E)$. In turn, $N(E)$ depends strongly on τ .^{3,4} The larger τ , the larger the degree of thermalization of polaritons (thermal equilibrium with the lattice at liquid helium temperatures is not completely established due to the characteristics of the energy relaxation of polaritons⁵). As a characterization of the degree of thermalization and, therefore, of the lifetime τ , we determined the quantity T^* , corresponding to the effective temperature that is established within a time τ for the distribution $N(E)$. The values of T^* were determined from the form of the short-wavelength part of the 2LO luminescence band (a more detailed analysis of the function $N(E)$ and the determination of T^* will be published later). Figure 1 shows the results of measurements only for specimens in which the values of T^* were minimum.

As d is decreased, the quantity T^* increases, which corresponds to a decrease of the polariton lifetime. This is evidently related to an increase in the effect of near-surface regions on the overall kinetics of polaritons. In the near-surface region, τ can be much shorter than in the bulk for two reasons. First, near the surface there is a large concentration of defects, which shorten the lifetime of polaritons compared with

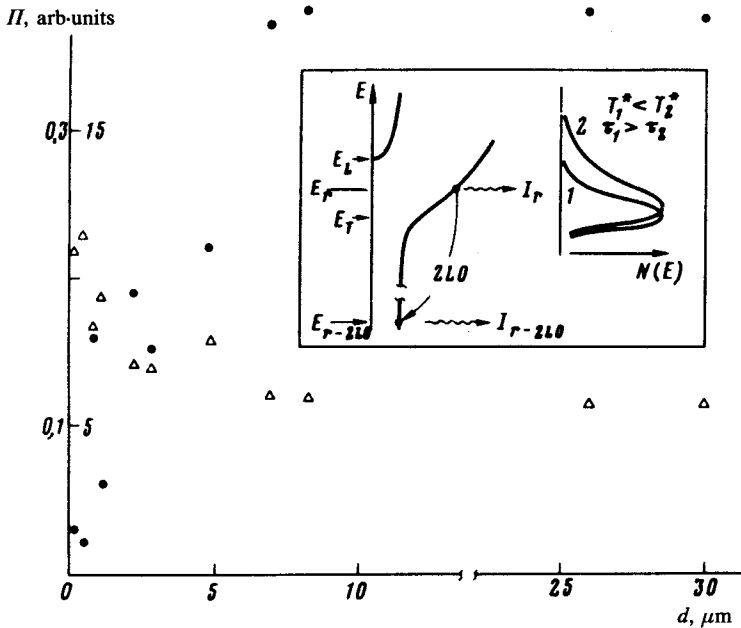


FIG. 1. Dependence of the effective polariton temperature $T^*(\Delta)$ and the ratio $\Pi(\bullet)$ on specimen thickness. The polariton dispersion curve is shown in the insert and the parts of the curve determining the measured intensities of polariton radiation are indicated (E_L and E_T are the energies of the bottom of the longitudinal and transverse exciton bands). The polariton energy distribution for two specimens with different thicknesses ($d_1 > d_2$) is shown schematically in the insert.

bulk polaritons (this explanation was used in Ref. 6 to interpret the results). Second, the near-surface electric field, which leads to band bending near the boundary of the semiconductor, can have a large effect on τ . The presence of an electric field can lead⁷ to a dependence of the exciton lifetime on the coordinates and a considerable decrease in τ near the surface. In addition, the near-surface field can pull excitons out of the bulk toward the surface,⁸ which can also decrease τ . In principle, the decrease in τ in thin specimens could be due to the increase in the probability of radiative annihilation of polaritons,⁹ However, the intensity of resonance emission of polaritons in this case should have increased appreciably compared with the emission of excitons bound on impurities, which was not observed in the experiment.

The thickness of the specimens will also affect polariton trapping. As demonstrated in Ref. 10, the degree of polariton trapping can be characterized by the quantity $\Pi = I_{r-2LO}/I_r$, where I_r is the intensity of radiation of polaritons with energy E_r at the center of the polariton resonance in luminescence line ($\lambda_r = 485.5$ nm), while I_{r-2LO} is the intensity of radiation arising after scattering of polaritons by 2LO phonons ($\lambda_{r-2LO} = 500.3$ nm) (Fig. 1). For large d , the ratio Π does not depend on the specimen thickness. It begins to decrease with 5-7 μm . For minimum d , the ratio Π decreases by more than an order of magnitude compared with thick specimens.

In thick specimens with large τ , as a result if the spatial relaxation, within a lifetime, polaritons occupy a volume determined by the diffusion length L ; in this case,

they accumulate as a result of energy relaxation in the resonance region.^{3,4} In the resonance region the coefficient of absorption is very large and, correspondingly, the mean free path of a polariton (l) is very small ($\sim 10^{-5}$ cm). As a result, only an insignificant part of the polaritons from the near-surface region with thickness of the order of l can leave the crystal in the form of radiation I_r . Most of the polaritons in the resonance region are trapped in the bulk of the specimen by phonon and impurity scattering and cannot approach the surface in order to be transformed into radiation. Radiative escape of polariton energy accumulated in the bulk occurs in this case primarily as a result of capture of polaritons in fine impurity levels followed by emission.¹⁰ The escape of 2LO luminescence radiation occurs without losses from any point within the crystal, since the coefficient of absorption in this region is close to zero. As a result, the intensity I_{r-2LO} is proportional to the number of polaritons accumulated in the entire volume. Thus the quantity I is proportional to the ratio of the volume occupied by the polaritons to the volume contributing to the resonance radiation. In thick crystals, the volume occupied by polaritons is characterized by the quantity L and, as long as $d > L$, it is determined only by diffusion of polaritons. As d is decreased, the thickness of the specimen becomes comparable to the diffusion length L . At this point, a further increase in d leads to the fact that the volume occupied by the polaritons is no longer determined by diffusion but by the thickness of the specimen. This is evidently the reason for the observed decrease in the magnitude of I in thin specimens. The specimen thicknesses for which I begins to decrease can be used to estimate the magnitude of L for ideal CdS specimens at liquid-helium temperatures, as a result of which we obtain $L \cong 5 \div 7 \mu\text{m}$.

Thus, we have demonstrated in this work, the considerable influence of the size of the specimen on the kinetics of polaritons.¹¹ We have also demonstrated the possibility of obtaining valuable information on the kinetics of characteristic excitations in the bulk and near the surface by studying a set of specimens with different thicknesses.

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¹¹The influence of the specimen size on the emission kinetics and emission spectrum was also noted previously, for example, in luminophors¹¹ and molecular crystals.¹²

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