

Imprisonment of polaritons

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The first experimental detection of a direct manifestation of polariton imprisonment in the emission spectrum of CdS crystals is reported.

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The term “imprisonment” first appeared in spectroscopy in the study of resonance radiation in gases.¹ A photon at the resonant frequency emitted by an atom at some arbitrary point in the volume, before becoming external radiation, undergoes multiple acts of absorption and subsequent emission. The impediment to the conversion of resonance radiation into external photons came to be described as the imprisonment of the resonance radiation by the volume of the gas. The imprisonment of resonance radiation is a phenomenon of great physical generality. It must be taken into account in studies of energy-transport and emission processes in practically any absorbing medium. Imprisonment can be observed not only for photons but for other elementary excitations as well. For example, the imprisonment of acoustic phonons in the levels of impurity centers has been detected and investigated in ruby (see, for example, Ref. 2). In the present study we have made the first experimental observation of an effect due to the imprisonment of polaritons.

We studied the emission spectrum of the exciton state $n = 1A$ of CdS crystals under excitation by the 476.5-nm line at $T = 2$ K in samples with different exciton lifetimes τ (τ was determined by the probability of nonradiative exciton loss^{3,4}). The polariton resonance luminescence spectra of the samples differed significantly both in the overall intensity and in the shape of the polariton luminescence lines⁴ (Fig. 1). Under identical excitation and recording conditions, but in samples with different values of τ , we measured the ratio I_r/I_{r-2LO} , where I_r is the intensity of the polariton emission with energy E_r at the center of the polariton luminescence band ($\lambda_r = 485.5$ nm), and I_{r-2LO} is the intensity of the emission arising after scattering of the E_r polaritons by $2LO$ phonons ($\lambda_{r-2LO} = 500.3$ nm) (Fig. 2). The measured ratio increased from values $\simeq 1:5$ in high-quality samples with large τ to values of the order of 150–200 in the lowest-quality samples with small τ .

In terms of the polariton model the emission arises as a result of the conversion of polaritons into external photons at the boundary of the crystal. In order to contribute to the observable radiation, a polariton starting from a point with coordinate x must reach the surface. The probability of this occurrence is proportional to $\sim e^{-\alpha(E)x}$, where $\alpha(E) = \Gamma(E)/v(E)$, $v(E)$ being the group velocity of the polariton and $\Gamma(E)$ being the probability that the polariton is scattered out of a state with a given energy and wave vector.⁵ As the resonance is approached from the low-energy side, $v(E)$ falls sharply, leading to an increase in $\alpha(E)$. As a result, the mean free path l of the polaritons is very small in the resonance region [$l(E) = 1/\alpha(E) \sim 10^{-5}$ cm; Ref. 6]. Here l is

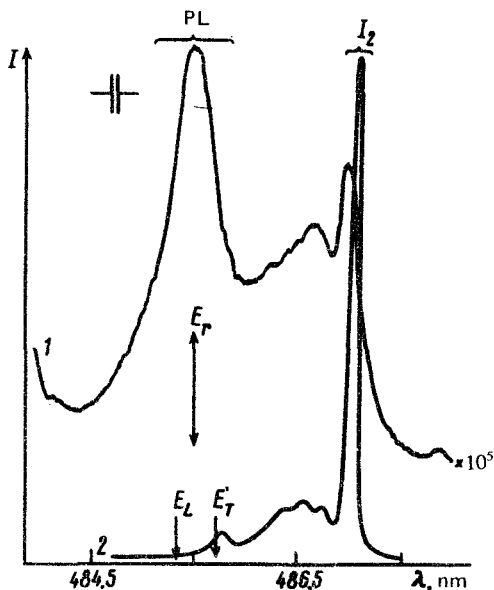


FIG. 1. Luminescence spectra of two CdS samples with different lifetimes at $T = 2$ K. Spectrum (1) corresponds to a sample with $\tau \approx 5 \times 10^{-12}$ sec; spectrum (2) to a sample with $\tau \approx 5 \times 10^{-9}$ sec (the value of τ was determined by the method of Ref. 3). The arrows E_L and E_T correspond to the bottoms of the longitudinal and transverse exciton bands.

limited by nonradiative loss processes and by the removal of polaritons to other points in the polariton band on account of scattering by phonons and elastic scattering by impurities (at liquid-helium temperatures the elastic scattering processes play an appreciable role^{7,8}).

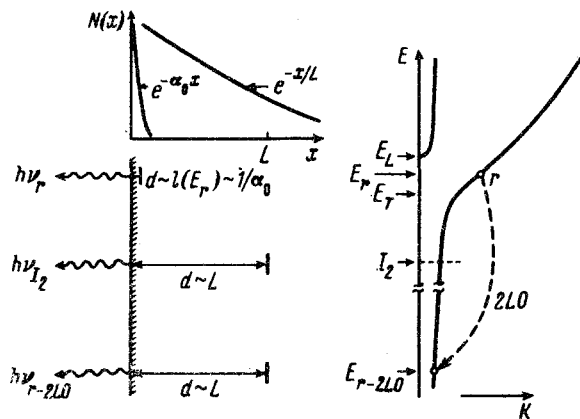


FIG. 2. Diagram explaining the effect of polariton imprisonment on the observable spectrum. On the left are a schematic of the polariton spatial distribution functions $N(x)$, which arise in samples with different values of τ , and a diagram illustrating the "effective emission depth" d of the corresponding parts of the luminescence spectrum.

Let $d(E)$ be the "effective emission depth," which specifies the thickness of the layer whose polaritons give the main contribution to the emission with $h\nu = E$. In the resonance region $d(E)$ is determined by the mean free path $l(E_r)$. For polaritons with energy $E_r - 2LO$ the mean free path is very large. In this case $d(E_r - 2LO)$ is determined not by $l(E_r - 2LO)$ but by the effective diffusion length L , which governs the spatial distribution of the polaritons. At small values of τ one has $L \simeq O$ and the spatial distribution of polaritons that arises in the crystal is determined by the absorption coefficient α_0 of the exciting light. For excitation above E_g one has⁶ $\alpha_0 \simeq \alpha(E_r)$. As a result, the majority of the polaritons arising in the sample are found in a layer whose thickness ($\simeq 1/\alpha_0$) is comparable to $l(E_r)$. Therefore, the values of d for polaritons with energies $E_r - 2LO$ and E_r are approximately the same. Of course, in this case the intensity of the resonance radiation is much greater than the intensity of the radiation involving phonons. As τ increases, there is sufficient time for the polaritons to diffuse deep into the crystal. According to our estimates, $L \simeq 10^{-3}$ cm in the perfect samples. In this case the polariton luminescence will again be due to polaritons from the surface layer. However, in contrast to the small- τ case, the polaritons in the surface layer constitute an insignificant fraction of the total number of polaritons. The majority of polaritons with energy E_r are unable to leave the crystal. They are "imprisoned" by the bulk of the material on account of multiple scattering processes. This also leads to an increase in the relative intensity of the less probable $2LO$ emission, since this emission is due to polariton states from a region whose thickness is of the order of L .

The imprisonment of polaritons is also manifested in the intensity ratio of the free and bound exciton emissions. Figure 1 shows, in addition to the polariton luminescence, the region of bound-exciton luminescence (486–487 nm). In the samples with large τ the luminescence spectra have a familiar form—the intensity of the bound-exciton emission is substantially higher than that of the polariton-luminescence line, and the most intense line is I_2 (an exciton bound to a neutral donor.⁹ The emission spectrum of samples with small τ are highly unusual. At a low overall intensity the polariton-luminescence line is the most intense line of the spectrum.

The main channel for the formation of bound excitons is the trapping of free excitons (polaritons) by capture centers.¹⁰ The relative intensity of the free and bound exciton emissions is therefore proportional to the number of capture centers. At first glance, it would be natural to assume that the largest relative intensity of the impurity line I_2 should be observed in the most defective samples. However, this assumption is valid if the volumes of the samples contributing to the polariton-luminescence and I_2 lines are the same. When the imprisonment of polaritons in the region of the polariton-luminescence line is taken into account, such an assumption is unjustified. As τ increases, $d(E_r)$ remains essentially unchanged, while the value of d for the I_2 line increases, since the values of the absorption coefficient for the I_2 line are substantially smaller than $\alpha(E_r)$.⁹ Consequently, when the imprisonment of polaritons is taken into account, the relative intensity of I_2 is governed not by the concentration of capture centers but by the total number of luminescence centers, which depends on the value of L .

In summary, we have given the first experimental demonstration of the imprisonment of polaritons by a crystal. Because of multiple scattering processes, the majority

of the polaritons in the resonance region are locked in the crystal and cannot reach the surface and be converted into external radiation. The radiative escape of polariton energy in this case occurs mainly on account of scattering by phonons out of the resonance region into states with small absorption coefficients or on account of the trapping of the polariton by impurity levels, followed by radiation.

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