

Kinetics of polariton luminescence

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(Submitted 2 August 1982)

Pis'ma Zh. Eksp. Teor. Fiz. **36**, No. 6, 196–198 (20 September 1982)

A manifestation of exciton kinetics has been studied experimentally in the shape of the polariton luminescence spectra for the first time. The spectral shape depends on the exciton lifetime. The basic factors determining the shape are identified.

PACS numbers: 71.36. + c, 71.35. + z

In the polariton model, exciton emission is regarded as the result of the conversion of a polariton into external photons at a boundary of the crystal. The dispersion curve for transverse polaritons near a prominent exciton resonance consists of a lower branch and an upper branch (1 and 2, respectively, in the inset in Fig. 1). According to Tait and Weiher,¹ the intensity of the emission at the energy E can be described by

$$I(E) = \sum_{P, f, i} \frac{\omega_{fi}^P(E) T_f(E) \rho_f(E) \Delta \Omega_f \Delta E}{D \alpha_f(E) + 1} \sim \sum_{P, f, i} \frac{|H_{fi}^P|^2 \rho_f(E_i) F(E_i) T_f(E) E^2 \Delta \Omega_{ex} \Delta E}{(D \alpha_f(E) + 1) V_f(E)}, \quad (1)$$

where $\omega_{fi}^P(E)$ and H_{fi}^P are the probability and matrix element for a (P -type) transition to the "emitting" state f (1,2) from other points (i) on the polariton branches; $\alpha(E)$, $V(E)$, and $T(E)$ are the absorption coefficient, the group velocity, and the transmission for a polariton at the boundary of the crystal; $\Delta \Omega_f$ and $\Delta \Omega_{ex}$ are the interior and exterior solid angles; D is the effective diffusion length; $F(E)$ is the probability that the state with the energy E is filled; and $\rho(E)$ is the polariton state density. Expression (1) describes the dependence of the polariton luminescence on the spatial distribution of the polaritons [through the factor $D\alpha(E)$] and their energy distribution. These distributions are determined by the particular features of the exciton kinetics.^{2,3} Despite the extensive research on polariton luminescence (see the review by Brodin *et al.*⁴), the effect of the exciton kinetics on the shape of the polariton luminescence spectrum has not been studied adequately. In this letter we show that the kinetics significantly determines the shape and nature of the polariton luminescence.

We have studied the polariton luminescence spectra of the $n = LA$ state of CdS and ZnSe crystals in samples with various exciton lifetimes (τ) at $T = 2$ K. The changes in the spectra in samples with different values of τ are identical for the CdS and ZnSe crystals, so we will report the results for CdS crystals only. The polariton luminescence spectra of these crystals were studied in the geometry $E \perp C$, $K \perp C$ with excitation by a laser line at 476.5 nm. Study of a large number of samples, with a range of quality, revealed a variety of spectral shapes; the most typical are shown in Fig. 1. It

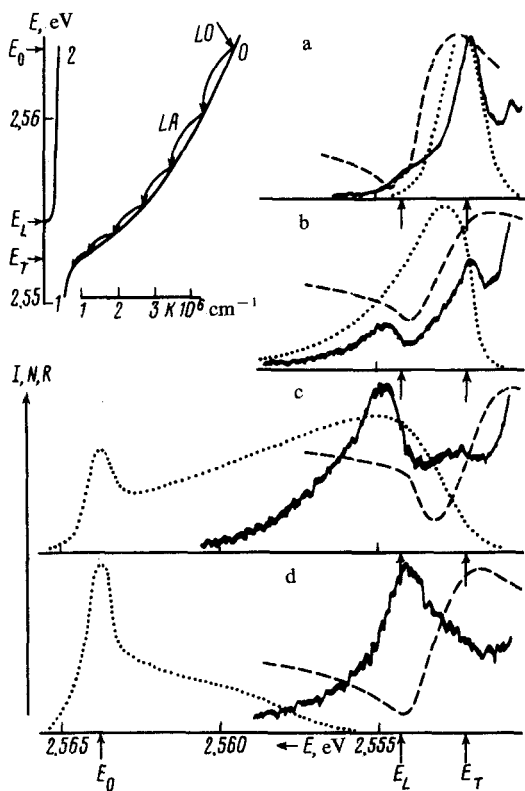


FIG. 1. Resonance-luminescence spectra in CdS crystals, in samples of various types, with excitation by 476.5-nm laser line at $T = 2$ K. The dashed curves are the reflection spectra $R(E)$. E_L and E_T are the energies of the bottoms of the longitudinal- and the transverse-exciton bands.

turns out that there is a correlation between the spectral shape and the lifetime τ . The inset in Fig. 1 is a schematic diagram of the events which occur in the CdS crystals upon excitation by the 476.5-nm line.³ Scattering of the exciting light by an LO phonon gives rise to polaritons at point 0 on branch 1. The subsequent scattering by LA phonons populates other points on the lower branch. The shape of the steady-state polariton energy distribution which results, $N(E)$, depends strongly on τ , which is in turn determined by radiationless transitions.³ The dotted curves in Fig. 1 are the functions $N(E)$ for these samples, found from the spectral shape of the two-LO-phonon emission of polaritons from various points on the lower branch.³

We divided the examples into three groups on the basis of their quality. In the most defective samples (group III) the lifetime was $\tau \lesssim \tau_a$ ($\tau_a \sim 5 \times 10^{-12}$ s is the lifetime with respect to the scattering of polaritons by LA phonons from point 0; Ref. 3). In these samples, most of the polaritons which arise do not manage to reach the resonance region and instead perish in the immediate vicinity of point 0 (Fig. 1d). In the samples of group II, τ is on the order of tens or hundreds of picoseconds.³ Near the resonance, these samples have a significant fraction of the total number of polaritons

(Figs. 1b and 1c). In the highest-quality samples we have^{3,5} $\tau \sim 1-2 \times 10^{-9}$ s, and the polaritons, which can be scattered by *LA* phonons many times, accumulate near the energy E_T (Fig. 1a).

Tait and Weiher¹ suggested that the polaritons are at thermal equilibrium with the lattice. At $T = 2$ K, this is not the case even for the highest-quality samples.^{5,6} To analyze the shape of the polariton luminescence spectrum, we can use expression (1) along with the experimental function $N(E)$, noting that $N(E) = \rho(E)F(E)$. A transition to the state F in this case can be caused both by events involving *LA* phonons¹ and by elastic scattering by defects, which play a significant role at low temperatures.^{7,8} As was shown in Ref. 1, processes involving *LA* phonons may be regarded as quasielastic. As a result, expression (1) may be rewritten as

$$I(E) = \sum_{f, i} \frac{T_f(E)N(E)E^2 \Delta\Omega_{ex} \Delta E}{(Da_f(E) + 1)V_f(E)} \left[|H_{fi}^{LA}|^2 + |H_{fi}^{elas}|^2 \right]. \quad (2)$$

We have used (2) for a quantitative analysis of the spectral shape. The coefficients $T(E)$ were calculated from the equations of Ref. 9; damping effects were ignored. In this case, only the lower branch contributes to the polariton luminescence below E_L . The calculated spectra give a good description of the observed polariton luminescence. The results of these calculations and their analysis will be published separately; here we will simply offer a qualitative explanation for the variety of forms taken by the luminescence. In the crystals of group III, τ is very short, and diffusion does not occur. The luminescence spectrum calculated from Eq. (2) with $D = 0$ is an isolated line with a maximum determined by the maximum of $T_1(E)$. This result is in agreement with the spectral shape of the group III samples (Fig. 1d, in which the polariton luminescence is seen as lines whose maxima coincide with minima in the reflection spectra [at $E < E_L$, we have $T_1(E) = 1 - R(E)$; Refs. 1 and 10]. A quantitative analysis shows that the contributions of branches 1 and 2 are roughly equal for polaritons of the same energy with $D = 0$. This result is somewhat unusual, since it has been assumed that only the upper branch contributes to the emission at $E > E_L$ (Refs. 1 and 11). An increase in τ leads to a diffusion of polaritons into the sample, as a result of which the factor $1/Da_f(E) + 1$, which depends on the spectral behavior of $a_f(E)$, begins to affect the spectral shape. Near the resonance, $a_1(E)$ increases sharply, from a value on the order of $10^2-10^3 \text{ cm}^{-1}$ below E_T to a value on the order of $10^4-10^5 \text{ cm}^{-1}$ above E_L . The coefficient $a_2(E)$ is large ($10^4-10^5 \text{ cm}^{-1}$) only in the immediate vicinity of E_L ($E - E_L \sim 0.1-0.2 \text{ meV}$). As E increases, $a_2(E)$ decreases rapidly and becomes smaller than $a_1(E)$. Diffusion should therefore reduce the relative importance of the polariton states of branch 1 above E_T and the importance of the upper branch in the immediate vicinity of E_L . In the samples of group II, in which the polariton states with $E > E_L$ are substantially populated, the luminescence acquires a distinctive shape with two maxima because of diffusion. One maximum results from the longest-wave of the filled polariton states of branch 1, from the E_T region; the other lies above E_L and is caused by the upper branch (Figs. 1b and 1c). A similar spectral shape has been observed for GaAs crystals.¹¹ The change in $N(E)$ with increasing τ increases the population of the lower-energy polariton states and increases the relative intensity of

the short-wave maximum (Fig. 1b). In the group I crystals, the function $N(E)$ is the factor which primarily determines the nature of the luminescence, imposing a boundary on the region of polariton states which can contribute to the emission (Fig. 1a). As Gross *et al.*¹⁰ have mentioned, the polariton luminescence in such samples is caused exclusively by states of the lower branch; the short-wave knee of the line is a consequence of the $T_1(E)$ structure. Absorption effects the shape of the primary maximum in this case.

In summary, in these experiments it has been possible, for the first time, to observe several polariton luminescence spectra in samples with various values of τ and to show that the various spectral shapes can be explained in a consistent way in terms of differences in the spatial and energy distributions of the polaritons. It appears that the spectral shapes shown here largely exhaust the possibilities for the polariton luminescence of semiconducting crystals.

We wish to thank S. A. Permogorov for many useful discussions.

¹W. C. Tait and R. L. Weiher, Phys. Rev. **178**, 1404 (1969).

²V. V. Travnikov and V. V. Krivolapchuk, Fiz. Tverd. Tela (Leningrad) **24**, 961 (1982) [Sov. Phys. Solid State **24**, 547 (1982)].

³S. A. Permogorov and V. V. Travnikov, Fiz. Tverd. Tela (Leningrad) **22**, 2651 (1980) [Sov. Phys. Solid State **22**, 1547 (1980)].

⁴M. S. Brodin, S. V. Marisova, and E. N. Myasnikov, Ukr. Fiz. Zh. **27**, 801 (1982).

⁵P. Wiesner and U. Heim, Phys. Rev. **B 11**, 3071 (1975).

⁶H. Sumi, Phys. Soc. Jpn. **41**, 526 (1976).

⁷A. A. Gogolin and E. I. Rashba, Solid State Commun. **19**, 1177 (1976).

⁸A. B. Pevtsov, S. A. Permogorov, and A. V. Sel'kin, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 419 (1981) [JETP Lett. **33**, 402 (1981)].

⁹A. V. Sel'kin, Candidate's Dissertation, Leningrad, 1974.

¹⁰E. Gross, S. Permogorov, V. Travnikov, and A. Selkin, Solid State Commun. **10**, 1071 (1972).

¹¹D. D. Sell, S. E. Stokowski, R. Dingle, and J. F. D'Iorio, Phys. Rev. **B 7**, 4568 (1973).

Translated by Dave Parsons

Edited by S. J. Amoretti