Interference emission of polariton states in crystals with spatial dispersion

A. G. Abdukadyrov, M. I. Sazhin, A. V. Sel'kin, and N. Kh. Yuldashev
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

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The formation of the polariton luminescence spectra of a crystal is analyzed in the case in which the conditions for the applicability of the Boltzmann kinetic equation do not hold. A dissipative attenuation due to the elastic scattering of exciton polaritons leads to an interference of the emitting states.

The polariton luminescence mechanism leads to a correct description of many characteristic features of the low-temperature emission spectra of crystals.[1] A description of this type usually takes an approach involving some solution or other of the boltzmann kinetic equation for the polariton distribution function. In taking this approach one is tacitly assuming that the necessary conditions for the use of the kinetic equation are satisfied:

$$|\mathrm{Re}\,\mathbf{k}_\beta| \gg \alpha_\beta \sim |\mathrm{Im}\,\mathbf{k}_\beta|, \quad |\mathrm{Re}\,(\mathbf{k}_{\beta'} - \mathbf{k}_{\beta''})| \gg \alpha_{\beta'}, \quad \alpha_{\beta''}, \tag{1}$$

where β, $\beta' \neq \beta''$ are the indices of the dispersion branches of the polaritons, and $\mathbf{k}_{\beta(\beta',\beta'')}$ and $\alpha_{\beta(\beta',\beta'')}$ are the wave vectors and absorption coefficients, respectively. The first of these inequalities means that the polariton mean free path α_β^{-1} must be greater than the wavelength $\lambda_\beta = 2\pi/|\mathrm{Re}\mathbf{k}_\beta|$. The second inequality corresponds to the condition that the interference of the polariton states of different dispersion branches is ignored. An interference of this sort arises if the dissipative attenuation of an exciton, Γ, is nonzero. This interference would be manifested as additional (interference) energy fluxes in a medium with a spatial dispersion.[2]

In this letter we are reporting an experimental and theoretical study of the low-temperature ($T = 2$K) polariton luminescence of mixed modes of a uniaxial CdS crystal (the $A_{n=1}$ exciton) under conditions such that inequalities (1) do not hold. The inset at the top of Fig. 1(a) shows the geometry in which the emission is detected. The optic axis of the crystal, the **C** axis, lies in the xy plane, which is the plane of the emitting face ($\mathbf{C}\|\mathbf{e}_x$). The emission is detected in the xz plane in the optical polarization $\mathbf{E}\perp[\mathbf{C}\mathbf{k}_0]$, at an emission angle φ. Here $\mathbf{k}_0$ is the wave vector of the photon emitted into the external medium. Figure 1(b) shows dispersion curves of the mixed $M1$ and $M2$ modes which are emitted at an angle φ in the case $\Gamma = 0$. Near the resonant frequency of a transverse exciton, ω_0, these curves are described by

$$\omega = \omega_L + \frac{\hbar k_x^2}{2M_\parallel} + \frac{\hbar(\mathbf{k}^2 - k_x^2)}{2M_\perp} - \frac{\epsilon_b \tilde{\omega}_{LT}}{c\mathbf{k}/\omega_0)^2 - \epsilon_b}, \tag{2}$$

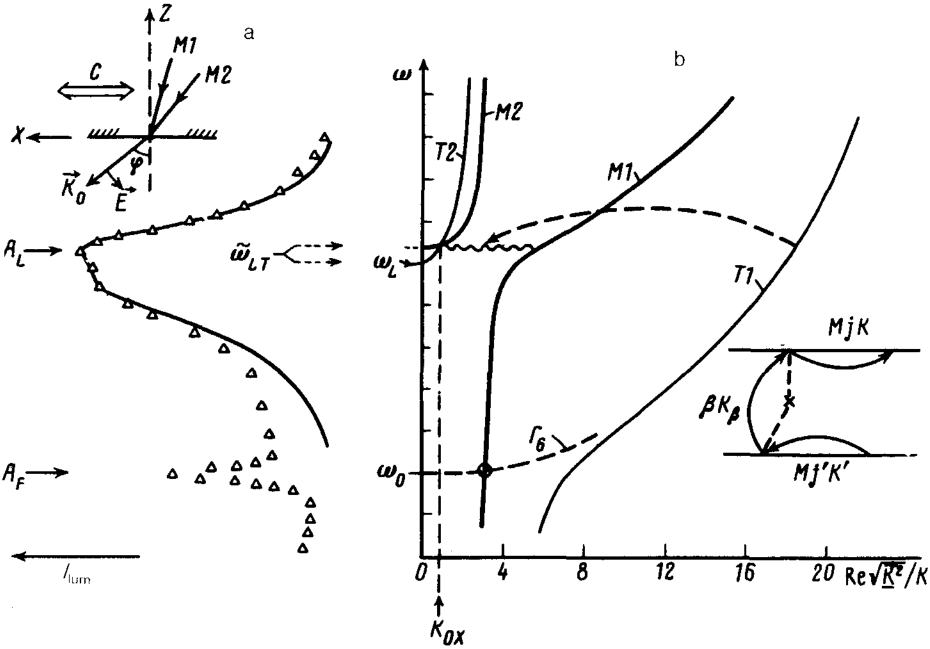

FIG. 1. a—Comparison of experimental and theoretical spectra of the mixed-mode emission from a CdS crystal ($T = 2$ K) near the $A_{n=1}$ exciton resonance; b—theoretical dispersion curves of $M1$ and $M2$ mixed modes and $T1$ and $T1$ transverse modes for an emission angle $\varphi = 67.5°$. The inset in part a shows the emission geometry; the inset in part b shows diagrams for the Green's function which describe the emission process.

where $\omega_L = \omega_0 + \omega_{LT}, M_\perp$ and $M_\parallel$ are the effective masses of the exciton in the propagation directions $\mathbf{k}\perp\mathbf{C}$ and $\mathbf{k}\|\mathbf{C}$, respectively, ϵ_b is the background dielectric constant, $\tilde{\omega}_{LT} = \omega_{LT}\sin^2\varphi/\epsilon_b^2$ is the effective longitudinal-transverse splitting, ω_{LT} is the longitudinal-transverse splitting of an exciton state in the polarization $\mathbf{E}\perp\mathbf{C}$, and the quantity ω_{LT} characterizes the dispersion of the transverse modes $T1$ and $T2$ in Fig. 1(b), appearing in an expression like (2), in which $\tilde{\omega}_{LT}$ must be replaced by ω_{LT}, and ω_L by ω_0.

If $\Gamma \neq 0$, conditions (1) may be violated in certain parts of the spectrum. This comment applies primarily to regions with $\mathrm{Re}\mathbf{k}_{M2} \to 0$, where the values of $|\mathrm{Re}\mathbf{k}_{M1}|$ and $|\mathrm{Re}\mathbf{k}_{M2}|$ are approximately equal to each other, i.e., near the frequency ω_L at small emission angles φ. However, conditions (1) may remain valid for transverse $T1$ modes and mixed modes of type $M1$ with large values of $|\mathbf{k}_{M1}|$. For such modes, one can use a distribution function, and the process by which the mixed modes are emitted should be treated as a quasielastic scattering of $T1$ and $M1$ polaritons (with large values of $|\mathbf{k}_{M1}|$) into $M1$ and $M2$ emitting states, with an immediate emission from these states. Since the emission line of the mixed modes is formed in a relatively narrow spectral interval, we write the distribution function of the scattered polaritons

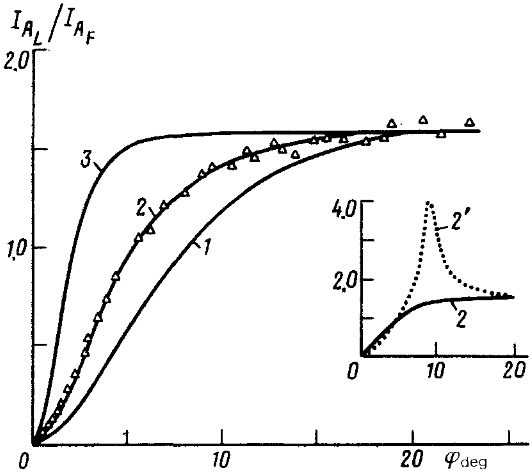

FIG. 2. Ratio of the maximum intensities of the A_L and A_F emission lines versus the emission angle φ. The triangles are experimental points. Theoretical curves 1–3 incorporate the interference of the modes for the polariton distribution depths L given in the text proper. The inset compares the results calculated with (2) and without (2′) the interference component.

in the form $f_{\beta\mathbf{k}}(\omega,z) = f_{\beta\mathbf{k}}(\omega)\exp(-z/L)$, with a z dependence determined by the effective depth of the distribution, L.

We calculate the intensity of the external emission, I, by the Keldysh diagram technique. The inset at the right in Fig. 1(b) shows diagrams for Green's functions renormalized to incorporate the light-exciton, exciton-lattice, and other interactions. The interference component of the external emission corresponds to a Green's function with $j \neq j'$. The spectral intensity of the luminescence is determined by the amplitude coefficients for transmission from the crystal into the external medium, which are calculated in the dead-layer model with Maxwellian boundary conditions and Pekar auxiliary boundaries.

The theoretical and experimental results are compared in Fig. 1(a) (the triangles are experimental, and the solid curve is theoretical) for the emission angle $\varphi = 67.5°$. There is a good agreement between theory and experiment near the frequency of a longitudinal exciton, ω_L. The calculation reproduces the slight doublet structure, which is formed both as a result of the individual $M1$ and $M2$ components and as a result of their interference. The reason for the discrepancy between the experimental data and the theoretical predictions in the long-wave part of the spectrum is that the theory ignores the frequency dependence of the distribution function and the component of the emission from states of the Γ_6 exciton (the emission line A_F).

The experimental points (the triangles) in Fig. 2 show the ratio of the maxima of the spectral intensities of the A_L and A_F emission lines (I_{A_L}/I_{A_F}) versus the emission angle φ in the geometry of mixed-mode emission (Fig. 1). Since I_{A_F} depends only weakly on φ, this ratio should be thought of as a normalized φ dependence of the intensity of the emission in the $M1$ and $M2$ modes. In choosing values of the theoretical parameters, we must allow for the circumstance that all the parameters except L are fixed by other, independent experimental data.[3,4] For this reason, the only adjustable parameter of the theory is the distribution depth L. The theoretical curves in Fig. 2 correspond to the values $L = 0.3$, 0.8, and 2.0 μm (curves 1, 2, and 3); the experimental data agree with curve 2. The interference component of the $M1$ and $M2$ modes is very important, as is shown by the inset in Fig. 2: Curve 2′ was calculated without consideration of the interference and thus differs radically from curve 2, which incorporates this interference and agrees well with the experimental data.

That value of the angle φ at which curve 2′ has its maximum corresponds to the maximum interference effect. The values of Γ and φ are related to each other in such a way that dispersion relation (2), which incorporates the parameter $\Gamma(\omega \to \omega + i\Gamma/2)$ at a fixed value of $k_x = k_0 \sin\varphi$, has a multiple-root solution for $\mathbf{k}^2$.

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