Anomalous Polaritonic Luminescence from Rare-Gas Solids

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Polaritonic phenomena interconnected with excitonic spatial dispersion were extensively explored theoretically and experimentally. The exciton-photon interaction leads to the formation of polaritonic states energetically positioned at both sides of the initial exciton. In a large ideal crystal of cubic symmetry, where the interval of the longitudinal-transverse splitting does not contain excitonic levels, the polaritonic dispersion branches lie beyond this interval at both sides of its boundaries. On the contrary, in a crystalline grain comparable or less in size than the wavelength in the substance, the interval of the longitudinal-transverse splitting is filled in continuously by excitonic states intercepting a significant part of the oscillator strength of the excitonic transition. In the previous experiments with polycrystalline samples of solid Kr and Xe the formation of the lower polaritonic state was traced by the red shift of the luminescence spectrum relative to the bottom E_1 of the lowest excitonic band (E_1^{Kr}=10.14 eV;E_1^{Xe}=8.36 eV).

In the present report we explore the new crystal growing technique, which allowed to obtain the solid Kr and Xe samples with essentially improved crystallographic properties and to resolve the internal structure of the luminescence bands at the edge of exciton absorption.The photoluminescence experiments were carried out at the SUPERLUMI experimental station at HASYLAB, DESY, Hamburg. Unlike previous works, where the observed red polaritonic shift was small commensurably with a weak inelastic polariton-photon scattering, a large polaritonic shift of luminescence is not due to energy dissipation, the energy conservation law being met due to equal probabilities for opposite-sign energy shifts. Such effect is possible if the crystalline grains are comparable in size with light wavelength, which provides the filling in the interval of the longitudinal-transverse splitting by excitons with sufficient oscillator strength. And the sample structure must be perfect enough to lowering the exciton scattering rate with respect to the rate of the polariton formation through exciton-photon coupling.

For the first time the excitation spectra of free-exciton luminescence band were recorded simultaneously below E_1 and within the interval of the longitudinal-transverse splitting. The luminescence of non-equilibrium polaritons was observed both within the longitudinal-transverse splitting interval and at photoexcitation below the bottom of the excitonic band. The excitation spectrum below E_1 is determined by competition of two processes. The first one is the creation of excitons by photons with energy E_1 at the Lorenz tail of excitonic absorption. The second process is a competing absorption related to the direct formation of two-site excitonic polarons (self-trapped excitons). Both excitation spectra of polaritonic luminescence below E1 and within the longitudinal-transverse splitting interval show high sensitivity to crystal quality of the samples.