Nonequilibrium Bose condensation of polaritons into a macro-occupied mode in HgI_2 and PbI_2 crystals

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Intense laser light induces a change in the low-temperature luminescence spectra of HgI_2 and 4H-PbI_2 crystals. This change has been studied. Its nature indicates nonequilibrium Bose condensation processes, manifested in an active absorption of polaritons by a macro-occupied mode outside a condensate.

Several theoretical studies[1–3] have been carried out on the propagation of an intense polariton wave or of a macro-occupied polariton mode (MPM) in a resonance region of a crystal below the bottom of the exciton band. The active absorption of anti-Stokes polaritons induced by the MPM has been attributed to a nonequilibrium Bose condensation in these studies. In the present letter we report the first experimental observation of this type of Bose condensation of polaritons (in HgI_2 and 4H-PbI_2 crystals).

The experimental procedure is essentially one of measuring the changes in the luminescence spectrum of a crystal which is excited in its exciton absorption region by a dye laser (laser 1). These changes occur in the presence of light from laser 2 in the transparency region below the bottom of the exciton band. The two tunable lasers (1 and 2) are identical in structure, have a spectral linewidth of 0.4–0.7 meV, and are simultaneously pumped by the beam from an N_2 laser with a power of 500 kW in a pulse 8 ns long, with a repetition frequency of 10 Hz. The beams from lasers 1 and 2 are focused on the same point on the surface of the crystal. The temperature here can be varied over the range 4.2–60 K. The light pulses from lasers 1 and 2 are synchronized by equalizing the optical path lengths. The luminescence spectra are recorded with a PGS-2 diffraction spectrograph.

The measurements are carried out in two steps. In the first step, the temperature of the crystals is 4.2 K. The experimental layout is shown in the insets in Figs. 1 and 2. The photon energy of laser 1, $\hbar\omega$, is held constant during the measurements; for HgI_2, it is 2.350 eV at an excitation power density $I = 100$ kW/cm^2. The broad luminescence band which arises here provides probe light with a continuous spectrum over the interval 2.314–2.325 eV below the level of a transverse exciton, $\hbar\omega_T = 2.3348$ eV (1 in Fig. 1). The MPM is produced by the light from laser 2, with $I_0 = 100$ kW/cm^2, in the transparency region of the crystal on the long-wavelength side of the luminescence band. Curves 2 and 3 in Fig. 1 are two spectra recorded—in contrast with curve 1 in Fig. 1—in the presence of MPMs with various values of $\hbar\omega_0$. A particularly noteworthy point is that in the presence of an MPM a narrow and pronounced ($\sim$50%) reabsorption dip appears in the probe luminescence spectrum at an energy $\hbar\omega_r$ on the anti-Stokes region of $\hbar\omega_0$. As $\hbar\omega_0$ is tuned from 2.300 to 2.310 eV, this dip

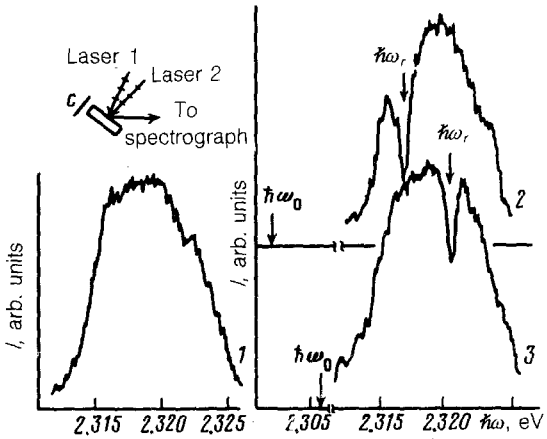

FIG. 1. Luminescence spectra of HgI_2 crystals at 4.2 K. 1—Without a macro-occupied polariton mode; 2—$\hbar\omega_0 = 2.3020$ eV; 3—$\hbar\omega_0 = 2.3060$ eV.

shifts in a linear fashion, without any change in shape, in the same direction, remaining within the luminescence band, following the change in $\hbar\omega_0$. The interval $\hbar\omega_r - \hbar\omega_0$ remains constant in the process, equal to 14.6 meV, i.e., to the energy of an optical phonon of symmetry A_{1g}.

The results found for the 4H-PbI_2 crystals ($\hbar\omega_T = 2.5079$ eV) are similar. Here we have $I = I_0 = 100$ kW/cm^2 and $\hbar\omega = 2.528$ eV; the value of $\hbar\omega_0$ was varied over the range 2.479–2.483 eV. Figure 2 shows three luminescence spectra for two values of $\hbar\omega_0$ (curves 2 and 3) and without the use of laser 2 (curve 1). Again in this case, the interval $\hbar\omega_r - \hbar\omega_0$ is independent of $\hbar\omega_0$, but it is equal to 11.8 meV, i.e., to the energy of the most Raman-active phonon, of symmetry A_1. The widths of the dips are 0.5–0.7 meV for HgI_2 and 0.4 meV for 4H-PbI_2 and do not exceed the spectral line width of laser 2. As the excitation power is raised to 200 kW/cm^2 (HgI_2) or to 400 kW/cm^2 (4H-PbI_2), we observe no changes in the widths of the dips.

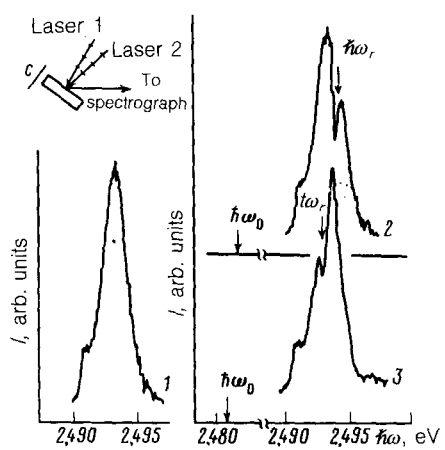

FIG. 2. Luminescence spectra of 4H-PbI_2 crystals at 4.2 K. 1—Without a macro-occupied polariton mode; 2—$\hbar\omega_0 = 2.4817$ eV; 3—$\hbar\omega_0 = 2.4810$ eV.

The particular features of the reabsorption bands observed here can be interpreted quite well on the basis of the theory of Refs. 1–3. Specifically, the phonoriton structural change in the spectra of polaritons and optical phonons in the anti-Stokes region of the MPM would, even in its early stage, involve an increase in the probability for induced Raman scattering of polaritons out of the anti-Stokes region into the MPM. It was shown in Refs. 1–3 that this absorption of anti-Stokes polaritons by an MPM is one possible mechanism for the occurrence of a nonequilibrium Bose condensation of polaritons.

Analyzing Figs. 1 and 2 again, and making use of the results of Refs. 1–3, we see clearly that the narrow dips which appear in the presence of an MPM correspond to those induced transitions of polaritons into an MPM involving optical phonons which were examined in those other papers. The observed pumping of a certain fraction of the polaritons out of the dip region of the probe luminescence band into the MPM means that these polaritons have a nonequilibrium Bose condensation.[1)]

In addition to causing the dips, the nonequilibrium Bose condensation may also cause a substantial change in the very nature of the polariton luminescence spectrum. We studied this possibility in the HgI_2 crystals. We excited them with the light from laser 1 in the exciton absorption region, at $\hbar\omega = 2.338$ eV with $I = I_0 = 100$ kW/cm^2. To achieve a polariton luminescence of a thermalized nature, we raised the temperature to 55 K. The single luminescence band which appears under these conditions is a phonon repetition of thermalized polaritons of a nonlinear polariton wave involving an A_{1g} phonon with $\hbar\Omega_0 = 14.6$ meV (curve 10 in Fig. 3). In this case, $\hbar\omega_0$ was scanned near this phonon repetition. The region of induced transitions of anti-Stokes polaritons into an MPM is therefore close to the "bottleneck" for the nonlinear polariton wave,

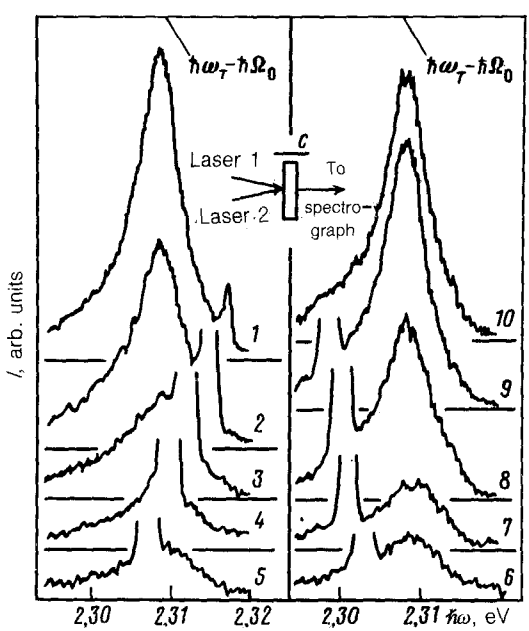

FIG. 3. Polariton luminescence spectra of HgI_2 crystals at 55 K for various values of $\hbar\omega_0$ (eV): 1—2.3167; 2—2.3146; 3—2.3188; 4—2.3096; 5—2.3071; 6—2.3025; 7—2.3009; 8—2.2997; 9—2.2982; 10—without a macro-occupied polariton mode. $\hbar\omega_T - \hbar\Omega_0 = 2.309$ eV.

where the density of polaritons is at a maximum. Figure 3 illustrates the transformation of the luminescence spectra as $\hbar\omega_0$ is tuned from 2.298 to 2.316 eV. While the position of the MPM satisfies the condition $\hbar\omega_0 \approx \hbar\omega_T - \hbar\Omega_0$, we observe a sharp decrease in the intensity of the luminescence band. We believe that this decrease reflects a decrease in the density of bottleneck polaritons due to induced transitions to the MPM, i.e., due to nonequilibrium Bose condensation. Blocking the beam from laser 2, i.e., eliminating the MPM, leads to a restoration of the intensity of the phonon repetition of the polaritons of the nonlinear polariton wave to its initial level (10 in Fig. 3).

We note in conclusion that only the initial stage of the polariton structural change occurred in our experiments. In this stage, the width of the region with the change in structure, Δ_{p-k_0}, is small in comparison with the laser linewidth. Under these conditions, the broadening of the induced absorption band with increasing I_0, which was predicted in Refs. 1–3, could not be detected experimentally.

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[1)]Goto *et al.*[4] at one point associated the reabsorption dip in the luminescence spectrum with an equilibrium Bose condensation of excitons, but they later changed their minds.

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[4]T. Goto, T. Anzai, and M. Ueta, J. Phys. Soc. Jpn. **35**, 940 (1973).
[5]T. Anzai, T. Goto, and M. Ueta, J. Phys. Soc. Jpn. **38**, 774 (1975).

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