Phonoriton modification of the polariton dispersion curve in CdS

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Experiments have been carried out on CdS semiconductors ($T = 5$ K) in a region of a small exciton-absorption coefficient in an intense pump light wave. The transmission coefficient is observed to decrease at a frequency higher than the pump frequency by an amount equivalent to an optical phonon. This change is probably due to a phonoriton modification of the dispersion curve of polaritons under the influence of the intense pump.

The renormalization of the dispersion curve of exciton polaritons in semiconductors as a result of illumination with intense light pulses has been discussed extensively in many papers from both the theoretical and experimental standpoints. There are a variety of mechanisms for changes in the spectra of elementary excitations: strong exciton-exciton interactions and exciton-biexciton interactions.[1–3] A renormalization of this type has been observed experimentally in CuCl and CdS crystals.[4–7]

Our purpose in the present study was to search in the transmission spectra of CdS crystals ($T = 5$ K) in the presence of intense light pulses for evidence of structural changes (a splitting) in the dispersion curve of polaritons which would be a consequence of the formation of phonoritons in them.[8–10] The creation of a new elementary excitation (Fig. 1) in a direct-gap semiconductor is possible if there is a strong exciton-photon and polariton-phonon interaction in the presence of an intense polariton wave $\mathbf{k}_0$.

In order to legitimately introduce the concept of a phonoriton elementary excitation and to observe it experimentally, we need to satisfy the following relations[9]:

$$\Omega_c \gtrsim \Omega_{\mathbf{p}-\mathbf{k}}, \tag{1}$$

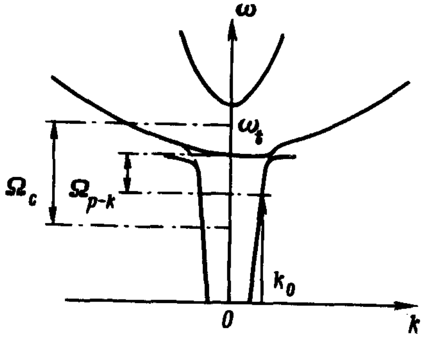

FIG. 1. Dispersion curve of the phonoritons.

$$[2\Delta(\mathbf{p}-\mathbf{k})\Omega_{\mathbf{p}-\mathbf{k}}]^{1/2} \gtrsim \gamma(\mathbf{p}) + \gamma^A_{\mathrm{ph}}(\mathbf{p}-\mathbf{k}), \tag{2}$$

$$\Delta(\mathbf{p}-\mathbf{k}) > \gamma_{\mathrm{pulse}}(\mathbf{k}), \tag{3}$$

where $\Omega_c = (2\omega_{LT}\omega_T)^{1/2}$ and $\Omega_{\mathbf{p}} - {}_{\mathbf{k}}$ are the nutation frequency and the frequency of a longitudinal optical phonon, respectively; $\hbar\omega_{LT}$ is the longitudinal-transverse splitting; $\hbar\omega_T$ is the energy of the exciton level; $\gamma(\mathbf{p}) = 1/\tau$ is the reciprocal lifetime of the scattered exciton $\mathbf{p}$; $\gamma^A_{\mathrm{ph}}(\mathbf{p}-\mathbf{k}) = 1/\tau^A_{\mathrm{ph}}$ is the reciprocal lifetime of the scattered phonon $\mathbf{p}-\mathbf{k}$, which is determined by the lattice anharmonicity of the crystal; $\Delta(\mathbf{p}-\mathbf{k}) \approx 2(N_0 m_{\mathbf{p}-\mathbf{k}})^{1/2}$ is a scale value of the gap that forms; $m_{\mathbf{p}-\mathbf{k}} = V|M_{\mathbf{p}-\mathbf{k}}|^2$, $M_{\mathbf{p}-\mathbf{k}}$ is the magnitude of the matrix element of the exciton-phonon interaction; N_0 is the density of the exciton component of the polariton pump wave; and $\gamma_{\mathrm{pulse}}(\mathbf{k})$ is the spectral width of the initial polariton wave $\mathbf{k}$. Relations (1)–(3) determine (a) where the frequency of the initial polariton wave and that of its anti-Stokes component should lie in the exciton spectrum and (b) the intensity and width which this wave should have.

According to estimates in Refs. 9 and 10, in CdS with $N_0 = 10^{17}$ cm^{-3} we should have $\Delta^{\max} \approx 0.3$ meV for longitudinal optical phonons. The actual frequency width of the absorption line may be substantially greater than the phonoriton splitting.

In the present experiments we studied high-quality CdS wafer single crystals 1–5 μm thick. The transmission spectra of the samples found during pulsed probing illumination during simultaneous pumping by intense laser pulses were recorded by the experimental apparatus shown schematically in Fig. 2. The pulse length is $\tau^{\mathrm{pump}}_{\mathrm{pulse}} = \tau^{\mathrm{prob}}_{\mathrm{pulse}} = 10^{-8}$ s; the repetition frequency is $\nu = 12.5$ Hz; the spectral widths are $\Delta\gamma^{\mathrm{pump}} \lesssim 0.15$ Å and $\Delta\lambda^{\mathrm{prob}} \sim 150$ Å; and the maximum power densities are $S^{\mathrm{pump}} \sim 50$ MW/cm^2 (spot diameter of 150 μm) and $S^{\mathrm{prob}} \ll S^{\mathrm{pump}}$.

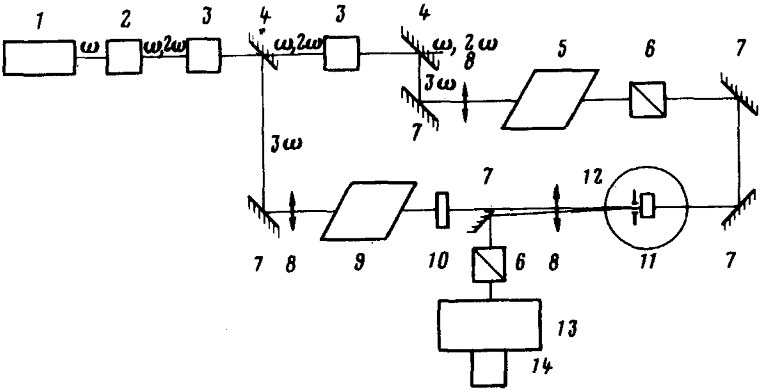

FIG. 2. The experimental arrangement. 1—Nd:YAG laser; 2, 3—frequency converters (CDA and KDP crystals); 4—selective mirror; 5—laser attachment using the dye coumarin 30 for the probing beam; 6—Glan prism; 7—mirrors with $R = 100\%$; 8—focusing lenses; 9—laser attachment using the dye coumarin 30 for the pump beam; 10—optical filters; 11—CdS crystal in a helium cryostat; 12—diaphragm (150 μm in diameter); 13—MDR-23 monochromator with a diffraction grating ($b = 1200$ lines/mm); 14—OSA-500.

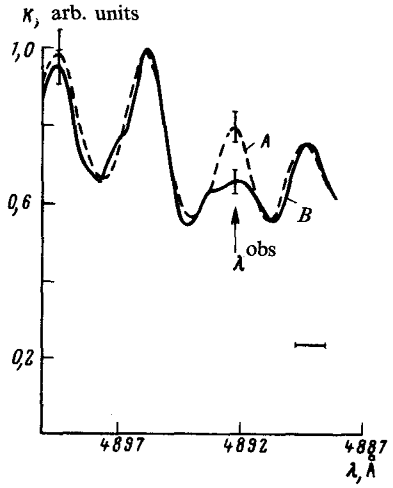

FIG. 3. Transmission spectra of a CdS sample ($T = 5$ K) with a thickness $d = 5\ \mu$m in an intense pump wave (A) and in its absence (B).

The spectra are measured in a backscattering geometry, which maximizes the phonoriton splitting, with $\mathbf{E}^{\text{pump}} \perp \mathbf{C}, \mathbf{E}^{\text{prob}} \perp \mathbf{C}, \mathbf{k}^{\text{pump}} \perp \mathbf{C}, \mathbf{k}^{\text{prob}} \perp \mathbf{C}$ (**E** and **k** are the polarization and wave vector of the light field, and **C** is the optic axis of the crystal).

Figure 3 shows the transmission spectra of a CdS sample ($T = 5$ K) with a thickness $d \approx 5\ \mu$m with and without pumping. The sample is pumped with intense pulses with a small spectral width at energies ≈ 50 meV below the exciton energy (for CdS, $\Omega_c = 100$ meV), where the exciton absorption coefficient is $\alpha \sim 25\ \text{cm}^{-1}$. At frequencies lying above the pump frequency by an amount equivalent to an optical phonon ($\hbar\Omega_{\mathbf{p}-\mathbf{k}} = 38$ meV) we observe a significant decrease in the transmission coefficient (K). In this case (Fig. 3) we find $\lambda^{\text{obs}} = 4892$ Å. We observe no changes elsewhere in the transmission spectrum. When the wavelength of the pump light is changed, the region of the induced decrease in K follows the change in the pump frequency. A deformation of the transmission spectrum is detected at the highest power densities of the pump radiation.

In an intense light wave, phonoritons probably form in CdS crystals, giving rise to a splitting of the polariton dispersion curve at energies $\hbar\omega^{\text{pump}} + \hbar\Omega_{\mathbf{p}-\mathbf{k}}$. In this frequency region the transmission coefficient should decrease significantly. Changes of this sort in K at wavelengths λ^{obs} were apparently observed in the present experiments. Unfortunately, the spectrum of the gap that forms could not be measured accurately because of the limited spectral resolution of the apparatus in these experiments ($\Delta\lambda^{\text{res}} = 1$Å).

In summary, we can apparently say that these experiments provide the first observation of the appearance of phonoritons—elementary excitations of a new type—in the transmission spectra of high-quality CdS crystals in an intense pump wave.

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