Particle-particle interactions in a dense polariton system and shift of the exciton term

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The transmission spectra of mixed exciton-polariton modes with energy below the exciton resonance of CdS are investigated at high levels of optical excitation, and the efficiency of polariton-polariton interaction processes is demonstrated. At maximum pumping an increase in the attenuation of polaritons is observed, along with a shift of the exciton term towards the violet.

INTRODUCTION

At this time, the linear crystal optics of excitonic polaritons is one of the most highly developed areas of exciton physics, in which the level of understanding has reached a point where in most respects there is not only qualitative but also quantitative agreement between theory and experiment. The processes that take place under weak excitation of the crystal are, as a rule, associated with spatial energy relaxation of the polaritons, in which the coordinates and quasimomenta of the latter can change significantly but their internal states usually do not change. Conversely, in those papers that undertake to discuss exciton resonances at high levels of excitation, changes in the spectra are as a rule associated with screening of the Coulomb interaction in the dense electron-hole system, i.e., with changes in the internal structure of the exciton.

In Ref. 1 it was asserted, apparently for the first time, that redistribution of the population of polariton states on the dispersion curve, caused by nonlinear polariton-polariton interaction processes, can lead to strong distortion of the polariton resonance transmission line, so that its visible broadening and disappearance from the spectrum might nevertheless be unrelated to any significant change in the internal structure of the exciton. In view of this assertion, the following question arises: how can the excitation levels at which effects of renormalization of the exciton ground state begin to appear in a dense electron-hole system be determined experimentally? This question is the topic of the present paper.

EXPERIMENTAL SETUP

The experiments were carried out on thin films of the crystal CdS, with thicknesses 0.7 μm and C axes in the plane of the film. The samples were placed in a helium cryostat with its temperature set at 6 K and maintained there with an accuracy of 0.1 K. The light source was tunable cumarin 152A dye laser with a line width around 0.5 nm, with a power per pulse of 1 kW, and pulse length 0.3 nsec. The tunable laser was pumped by a nitrogen laser with frequency 25 Hz. The rest of the experimental scheme was similar to that described in Ref. 1, and will therefore be described only briefly in what follows.

The radiation was focused onto the crystal into a spot with a diameter of 100 μm, using the standard excitation geometry for a mixed mode[2]: the electric vector of the incident electromagnetic wave and the C axis of the crystal were in the plane of incidence of the light, and the angle of incidence was 20°. The frequency of the laser radiation was chosen in such a way that the emission band fell in the resonance region of the mixed mode. The transmission signal was recorded using a DFS-24 spectrometer, an 18 ELUFM pulsed photomultiplier, and a C7-12 stroboscopic oscilloscope. In order to obtain the correct value of the transmission through the crystal a reference signal was simultaneously recorded, which was split off from the primary beam of the tunable laser. The digital accumulation of the spectra was effected with a multichannel analyzer.

POLARITON-POLARITON INTERACTION

When a crystal is excited by a tunable-laser pulses that fall within a resonance region of a mixed mode, a large number of polaritons are created with differing frequencies and wave vectors. So long as the intensity of the light is not too high, the propagation of these polaritons through the crystal can be treated as independent, and a sharply defined transmission line which corresponds to the mixed-mode polariton resonance should be observed in the light exiting the crystal. As the intensity of the light increases, polariton-polariton interactions set in and take place with conservation of energy and quasimomentum:

$$\hbar\omega_1+\hbar\omega_2=\hbar\omega_3+\hbar\omega_4,$$
$$\hbar\mathbf{k}_1+\hbar\mathbf{k}_2=\hbar\mathbf{k}_3+\hbar\mathbf{k}_4.$$

Polaritons *1* and *2* excite a virtual intermediate state with energy $\hbar\omega_1+\hbar\omega_2$ and wave vector $\mathbf{k}_1+\mathbf{k}_2$, which then decays into polaritons *3* and *4*. The same equations hold for the case where a real intermediate state is present as well, e.g., when biexcitons are excited with **E** polarized perpendicular to the C axis.[3]

Processes of this kind are conveniently investigated by using the following graphical construction (see Fig. 1). We mark on the polariton dispersion curve point *1* with coordinates (k_1, ω_1) corresponding to the first polariton that participates in the interaction. We then make this point the origin of a new auxiliary system of coordinates k' and ω' with axes parallel to the axes of the original system. Once again we plot the dispersion curve in this new system of coordinates for the polaritons and mark on it point *2* with coordinates (in the new system) corresponding to the energy and momentum of the second polariton. It is not difficult to see that the coordinates of this point $(\mathrm{k}_1+\mathrm{k}_2, \omega_1+\omega_2)$ will correspond to the momentum and energy of the virtual inter-

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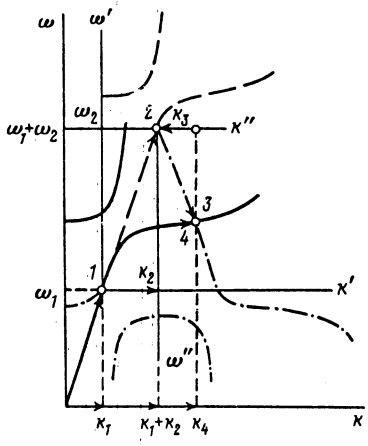

FIG. 1. "Algebra" of dispersion curves, which clearly shows the laws of conservation of energy and quasimomentum for the polariton-polariton interaction.

mediate state in the original system of coordinates. In order to find all possible variants of the decay, it is now sufficient to make the point *2* of the intermediate state the origin of a coordinate system k″, ω'' with axes antiparallel to the axes of the original system. The points of intersection of the polariton curve plotted in this "reversed" system of coordinates with the polariton curves of the original system then determine the energies and wave vectors of polaritons *3* and *4*. It is clear that, using this construction, we find only those types of decay that occur in the plane of incidence of the light, although we can obviously satisfy the conservation laws by considering other decays in which the wave vectors for polaritons *3* and *4* do not lie in the plane of incidence. In this case the "algebra of curves" should be replaced by an "algebra of surfaces."

From what was said above, it is clear that in the experimental situation investigated there are many possible different types of polariton-polariton interaction leading to redistribution of the polaritons along their dispersion curve. However, the interactions that are most probable are those that lead to the creation of polaritons on dispersion-curve segments characterized by a high density of states.

In Fig. 2a just one of these processes is shown: the interaction of two lower-branch polaritons (LBP), leading to intense population of the states in the vicinity of the "bottleneck," which causes the mixed-mode transmission line to "disappear."[1] This process can be suppressed quite effectively by retuning the center of the laser emission band to lie above the polariton resonance. In this case the transmission line can be observed at very high levels of excitation.

Figure 2b shows a scattering process involving the upper-branch polaritons (UBP). It is apparent from this figure that decay types are possible in which both particles created by the decay lie in a polariton-dispersion region with high density of states. However, the process in which "resonant" polaritons interact should have the highest probability, i.e., the process shown in Fig. 2c. The density of these polaritons in the crystal is always considerably higher than that of the "nonresonant" polaritons, since the group velocity near the resonance is significantly decreased; as a result, the spatial dimension of the optical pulse is "compressed." In addition to the processes listed above, a process in which LBP and UBP polaritons interact is also possible and can be analyzed analogously without difficulty.

We now examine briefly the processes of polariton-polariton interaction from the point of view of nonlinear optics. Processes of the type considered above, which are discussed within a single framework in terms of polariton-polariton interactions, are classified in different ways in traditional nonlinear optics. Thus, for the case where both the initial and final polaritons that participate in the elementary interaction event are "light-like," the interaction must be considered a four-photon process, i.e., hyper-Raman light scattering (scattering of light by light in the material); see Fig. 3a. This is the usual way to treat polariton-polariton interactions that involve the excitation of excitonic molecules.[3] If both the initial polaritons and one of the final polaritons is "light-like," while the other final polariton is "exciton-like," then the process is two-photon Raman scattering by the excitons (Fig. 3b). In the case where both final polaritons are "exciton-like," one should obviously speak of two-photon absorption (Fig. 3c).

Finally, the scattering of "resonance" polaritons generally has no nonlinear-optics analogs. This is because a description of the interaction of light with matter, in the language of nonlinear susceptibilities, becomes strictly

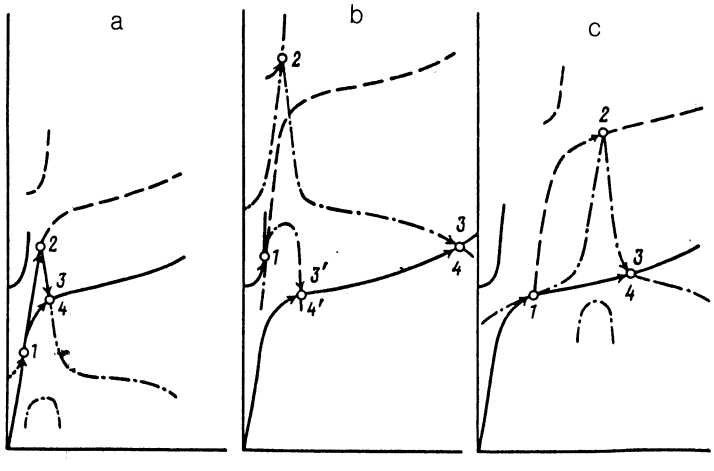

FIG. 2. Schematic illustration of the interaction processes: (a) LBP polaritons, (b) UBP polaritons, (c) "resonant" polaritons.

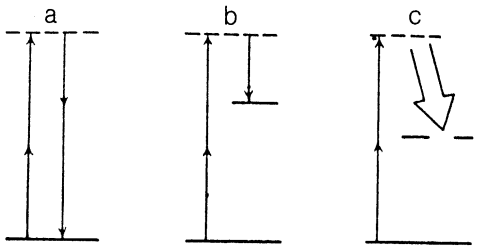

FIG. 3. Nonlinear-optic analogs of polariton-polariton interaction processes (a) hyper-Raman light scattering, (b) two-photon Raman scattering by excitons, (c) two-photon absorption.

speaking incorrect in the resonance region, because the polarization here can no longer be expanded as a power series in the field. For the "light-like" portions of the dispersion curve the nonlinear-optics approach is nevertheless fully applicable, although a rigorous investigation of this problem should obviously take into account the nonlocal character of th excitonic polarization, which is connected with spatial dispersion effects.

EXPERIMENTAL RESULTS AND DISCUSSION

The general character of the change, in the spectrum of a pulse passing through the crystal with increase of pumping, is shown in Fig. 4. As the excitation level increases a sublinear growth in the light intensity at the output of the crystal is observed with a pronounced tendency towards saturation. This leads to stabilization of the pulse amplitude from the photomultiplier on the screen of the oscilloscope, while the amplitude of the pulse in the reference beam undergoes considerable fluctuations. The crystal behaves like an "optical stabilitron," i.e., strong variations in the intensity of the input radiation lead to very small variations in the output signal. This leads to a smoothing of the spectral profile of the pulse; its maximum becomes flattened and the width of the flattened portion of the spectrum increases as the pumping increases. At the same time, a decrease is observed in the mixed-mode transmission line until it disappears completely from the spectrum. Figure 5 shows the transmission spectrum of the crystal at various pumping levels. Especially noteworthy is the considerable decrease in transmission over a wide region around the mixed-mode polariton resonance.

It is natural to associate the observed change in the transmission with the processes of polariton-polariton interaction discussed above. In fact, all of these processes depend quadratically on the pumping, and their probability should increase rapidly as the excitation level grows. The interaction of upper-branch polaritons is closest to two-photon absorption, and therefore one may hope to describe the change in transmission for the UBP in terms of a two-photon absorption coefficient. Figure 6 shows the dependence of the transmission for UBP on pumping, plotted using the spectra shown in Fig. 5. The curve in the figure was obtained by a best fit of the experimental points to the function (the factor 0.6 takes into account losses due to reflection from the crystal boundaries)

$$T=I_t/I_p=\frac{0.6}{1+\beta\, dI_p/\hbar\omega}$$

which is the solution to the equation that describes the change in intensity of the light within a medium with two-photon absorption. Here I_t and I_p are the intensities of the light that emerges from and is incident on the crystal, β is the two-photon absorption coefficient, d is the crystal thickness, and ω is the frequency of the light.

It is clear that this curve describes the experimentally

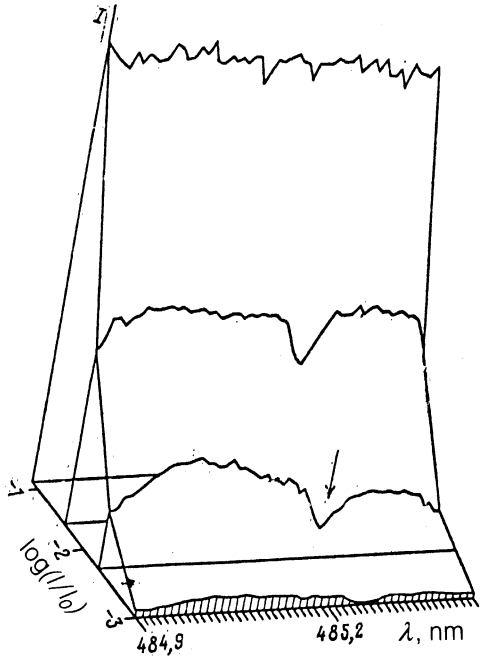

FIG. 4. General character of the change in the spectrum of light transmitted through the crystal as the excitation level increases. The arrow denotes the transmission line of the polariton mixed-mode resonance; $I_0 = 3$ to 5 MW/cm^2, λ is the wavelength.

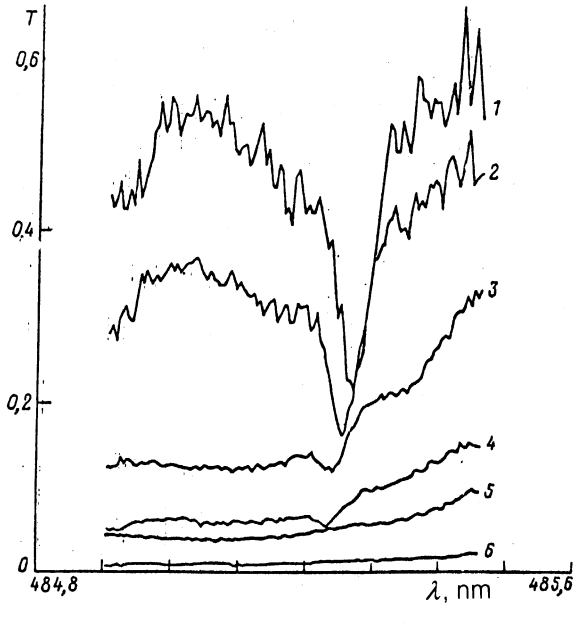

FIG. 5. Transmission spectrum of the crystal for various pumpings: (*1*)—0.002 I_0, (*2*)—0.013 I_0, (*3*)—0.05 I_0, (*4*)—0.07 I_0, (*5*)—0.28 I_0, (6)—I_0; here $I_0 = 3$ to 5 MW/cm^2. A significant decrease in transmission is evident over a wide range around the polariton mixed-mode resonance.

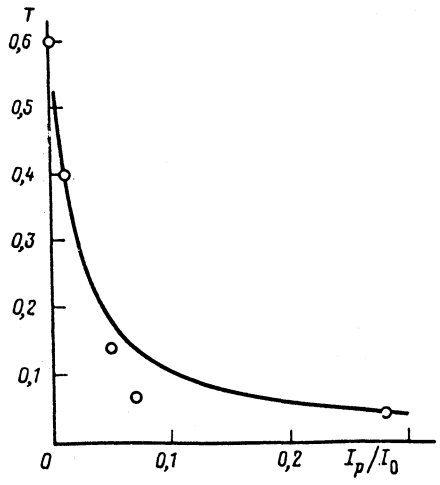

FIG. 6. Transmission in the UBP region as a function of excitation level. The curve is the result of a calculation with $\beta I_0 d/\hbar\omega = 48.3$, which gives the best fit to the experimental data; $I_0 = 3$ to 5 MW/cm^2.

observed dependence quite well; the above-noted tendency of the intensity of the light leaving the crystal to stabilize finds a natural explanation within the framework of the so-called limiting effect, i.e., the tendency of I_t to $0.6\,\hbar\omega/\beta d$ as $I_p \to \infty$. The two-photon absorption coefficient is determined by the imaginary part $\mathrm{Im}\chi^{(3)}$ of the nonlinear third-order susceptibility, which turns out to be on the order of 10^{-4} cm^3/erg, a value that exceeds $\mathrm{Im}\chi^{(3)}$ for interband transitions in CdS by nine orders of magnitude.[4] A further indication of the gigantic value of the nonlinearity is clearly the fact that the two-photon absorption significantly exceeds even the resonant one-photon absorption at the mixed-mode polariton resonance.

It is very significant that the polaritons remain "good" quasiparticles over practically the entire range of pumpings investigated (i.e., up to several MW/cm^2)—in other words, that the polariton interaction takes place with conservation of quasimomentum. A clear confirmation of this is a singularity in the transmission spectrum, which first appears on the violet side of the polariton resonance at roughly 100 kW/cm^2 and then is present in practically all of the spectra (see Fig. 7). The magnitude of this singularity is very sensitive to the poorly controllable conditions for observation and excitation (focusing, the position on the crystal, etc.); however, its spectral position is well reproduced from spectrum to spectrum. Under especially favorable conditions, it takes the form of a pronounced peak that is mirror-symmetric with respect to the transmission line for the mixed-mode polariton resonance (it is this case that is shown in Fig. 7). It is logical to associate this feature with interaction involving "resonance" polaritons, which, as stated above, should be the most efficient of all the polariton-polariton interaction processes. As a result of this process, the "exciton-like" portion of the polariton dispersion curve should be strongly populated (see Fig. 2c). Despite their "exciton-like" character, these polaritons have a rather low but nonetheless finite probability of passing through the crystal, converting at its rear face into two photons, and thus contributing to the radiation transmitted through the crystal. It is this type of polariton that takes part in the interference of additional waves observed in thin crystals above the polariton resonance.[5]

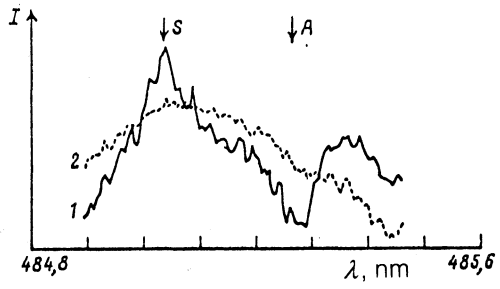

FIG. 7. Spectrum of radiation transmitted through the crystal for an excitation level around 100 kW/cm^2. The S peak, which appears as a result of the interaction of "resonance" polaritons, is clearly seen. The spectrum of light incident on the crystal is shown dashed. The arrow A denotes the position of the polariton mixed-mode resonance.

As was mentioned above, at sufficiently high pumping levels the spectrum of light transmitted through the crystal is characterized by a wide flat maximum, against which a complete disappearance of all singularities connected with the polariton resonance is observed (see Fig. 4) at a certain pump intensity. It is important to note that the disappearance of the singularities is not due to screening of the excitons or to a strong increrase in the role of inelastic scattering processes, but rather to the effects discussed above, i.e., limiting and population of states in the region of the polariton bottleneck.[1]

At the highest pumping levels, which according to estimates can reach 3 to 5 MW/cm^2, the line corresponding to the polariton mixed-mode resonance appears once more on the wide flat maximum in the spectrum of the light transmitted through the crystal. This line is strongly broadened compared to the case of weak pumping and rapidly shifts towards the violet end of the spectrum with increasing excitation level (see Fig. 8). The very fact that it appears in the

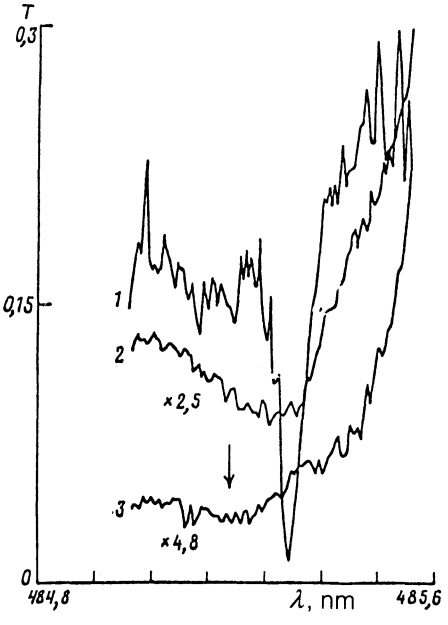

FIG. 8. Transmission spectrum of the mixed mode for maximum pumping: (*2*) 0.5 I_0, (*3*) I_0. For comparison we show the spectrum (*1*) for 0.05 I_0, where $I_0 = 3$ to 5 MW/cm^2. A distinct violet shift of the exciton term is seen.

spectrum implies a decrease in the efficiency of the limiting process, which in our view is connected with an increase in the polariton attenuation. The shift of the transmission line towards the high-energy side is obviously caused by the repulsive interaction between excitons at small distances.[6] Note that a rather small violet shift of the line can also be seen at small pumping (see Fig. 5); however, since its value is smaller than the width of the transmission line, in this case it is difficult to associate it unambiguously with the shift in the exciton term. It is for this reason that observation of the shift in the exciton term for the allowed polarization $\mathbf{E} \perp \mathbf{c}$, where the width of the polariton resonance is large, is accompanied by serious difficulties.

Thus, the results of this paper demonstrate the high efficiency of polariton-polariton interaction processes leading to gigantic values of the two-photon absorption in the region of the mixed-mode polariton resonance, and to a limitation of the radiation exiting the crystal; the range of pumping over which the polaritons may be considered "good" elementary excitations has been determined (up to 2 to 3 MW/cm^2), and an increase in the polariton attenuation was observed, accompanied by a shift of the exciton term toward the violet at pumps 3 to 5 MW/cm^2.

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