Picosecond time-resolved photon echo in system of intrinsic excitations of a medium (mixed $CdSe_xS_{1-x}$ crystals)

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A picosecond photon echo due to intrinsic excitations of a medium (excitons trapped at potential fluctuations in mixed $CdSe_xS_{1-x}$ crystals) has been demonstrated unambiguously, with time resolution, for the first time.

Research on photon echoes is a powerful method for studying the phase relaxation of optical excitations.[1] The possibilities of using photon echoes for optical data processing are also being examined.[2] The echo pulse is excited as a result of the restoration of a macroscopic, coherent, nonlinear polarization in a system of two-level absorbers with inhomogeneous broadening after the application of temporally separated exciting pulses of length $\Delta t < T_2$, where T_2 is the transverse relaxation time. This relaxation time is a measure of the irreversible dephasing of an individual absorber (see Refs. 1 and 3–5, for example, for more details). The inset in Fig. 1 shows the layout for measuring stimulated photon echoes. Coherent exciting pulses are applied to the sample; pulse 1 is applied at the time $t = 0$, and pulse 2 is applied after a delay

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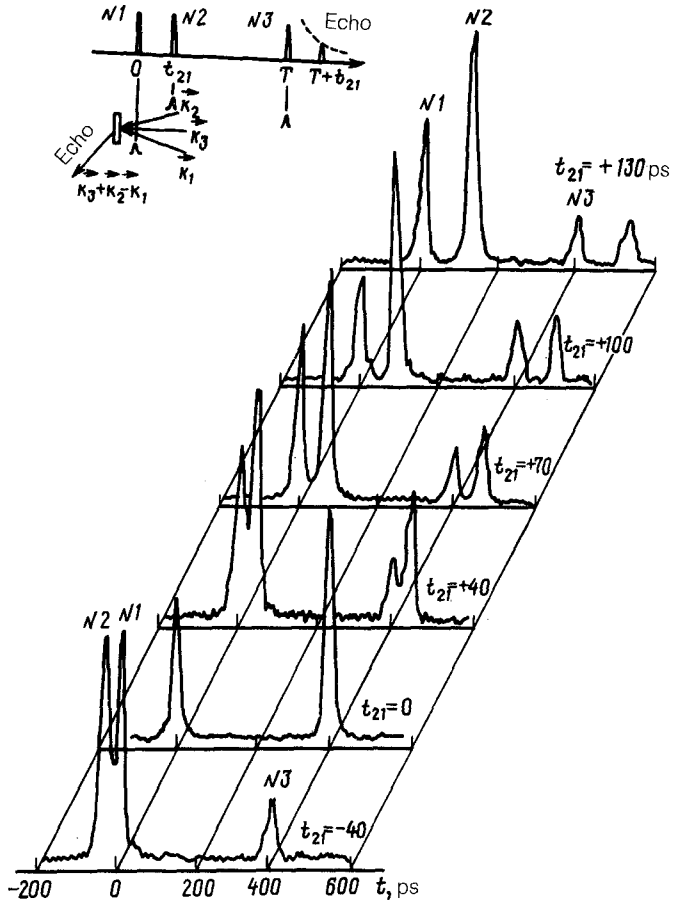

FIG. 1. Time-resolved photon echo (the fourth pulse in the train; see the inset) for various delays (t_{21}) between exciting pulses 1 and 2. The sample was made of $CdSe_{0.6}S_{0.4}$; the temperature was 10 K; the delay of the probe pulse (3) was fixed at $T = 400$ ps; the energy of the exciting photon was $h\nu_{ex} = 2.0505$ eV; and the excitation intensity was $I_{ex} = 0.05$ mW. The curves have been normalized to a common height of the highest pulse.

t_{21}. In addition, a probe pulse, 3, is applied at $t = T$. The echo pulse is generated at the time $t = T + t_{21}$, in the matching direction for the wave vectors, $\mathbf{k}_3 + \mathbf{k}_2 - \mathbf{k}_1$. The two-beam arrangement for spontaneous photon echoes corresponds to the degenerate case $\mathbf{k}_3 = \mathbf{k}_2$, $T = t_{21}$. The peak intensity satisfies $I_{echo} \sim \exp(-4t_{21}/T_2)$, so one can determine T_2 from measurements of the dependence $I_{echo}(t_{21})$ as the delay t_{21} is varied.

The photon echo was first demonstrated experimentally through a direct oscilloscopic study, for a system of impurity centers in ruby.[6] That method was subsequently used to study other extrinsic excitations in various matrices, with times T_2 in the nanosecond and microsecond ranges (see the bibliographies in Refs. 1, 5, and 7). The

phase relaxation of intrinsic electronic excitations in semiconductor crystals occurs more rapidly. In bulk crystals of GaAs (Ref. 8) and CdSe (Ref. 9) and in GaAs/GaAlAs quantum-well structures,[10,11] values of T_2 on the order of a few picoseconds have been found for free excitons near liquid-helium temperature. Values on the order of a few femtoseconds have been found for free carriers in bulk GaAs at 300 K (Ref. 7). Those measurements were carried out by an indirect method: A time-integrated signal selected by angle was recorded in a layout for spontaneous photon echoes. That approach is less reliable. In particular, it does not permit a clear distinction from the case of homogeneous broadening, in which the signal representing the macroscopic coherent response is generated in the matching direction immediately after pulse 2 and decays in accordance with[4] $\exp(-2t_{21}/T_2)$.

In the present letter we are reporting the first unambiguous observation—with a time resolution in the picosecond range—of the photon echo in a system of intrinsic excitations of a medium. In particular, we studied the mixed semiconducting crystals $CdSe_xS_{1-x}$ whose optical properties near liquid-helium temperature are determined by excitons trapped at fluctuations of the crystal potential.[12] The samples were high-quality leaves, mo more than 50 μm thick, grown from the gas phase. The experiments were carried out at $\approx$10 K in both the geometry for spontaneous photon echoes and the geometry for stimulated photon echoes. The exciting light was produced by a tunable laser using the dye rhodamine 6G, which was synchronously pumped by the second harmonic of an Nd:YAG laser oscillator and which generated pulses 7 ns long at a repetition frequency of 500 kHz. For detection we used a high-speed image-converter camera in a regime of synchronous scanning with a charge-coupled-device target, an image intensifier, and a computer-based data acquisition system (the dynamic range was $\gtrsim 10^3$:1, and the time resolution was 20 ps).

Figure 1 shows image-converter traces recorded in the configuration for a stimulated photon echo with an excitation photon energy $h\nu_{ex} = 2.0505$ eV, which corresponds to the resonant excitation of trapped excitons in the $CdSe_{0.6}S_{0.4}$ sample. The curves for various delays t_{21} correspond completely to the pattern expected (see the inset). Remarkably, at $t_{21} < 0$ there is no echo signal in the direction $\mathbf{k}_3 + \mathbf{k}_2 - \mathbf{k}_1$. The traces of pulses 1, 2, and 3 are the result of a scattering of the corresponding beams in the given direction, since they were nominally blocked beyond the exit window of the cryostat. This scattering could influence the results of measurements of the photon echo by an indirect method, while in our case the time resolution makes it possible to distinguish "clean" signals and to observe intensity saturation of the echo pulse at excitation levels I_{ex} above a critical level I_{cr} (Fig. 2). At the same time, we observe a bleaching for a beam 2. As the photon energy $h\nu_{ex}$ is reduced, the critical level I_{cr} becomes lower.

The values of T_2 estimated from these experiments were hundreds of picoseconds (400 ps for the conditions in Fig. 1). These times are significantly longer than the values which have been reported for binary semiconductor crystals. We observe a smooth increase in T_2 with decreasing $h\nu_{ex}$.

The experimental data can be explained at a qualitative level by the model of exciton trapping. For example, the evident reason for the dephasing in comparison with GaAs and CdSe is that for trapped excitons, in contrast with free excitons,

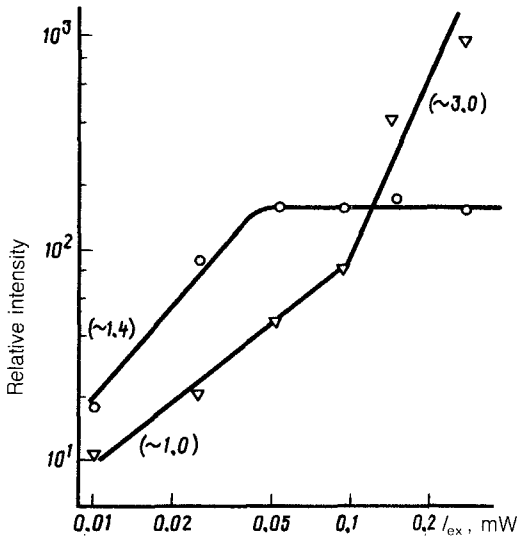

FIG. 2. Intensity of the echo pulse (circles) and intensity of pulse 2 after transmission through the sample (triangles) versus the excitation level in the configuration for spontaneous photon echoes ($CdSe_{0.6}S_{0.4}$ sample, 10 K, excitation energy $h\nu_{ex} = 2.0526$ eV, $t_{21} = 100$ ps). The approximate slope is shown in parentheses.

interparticle dephasing collisions are not important. In addition, they are not observed in the luminescence or optical amplification spectra of strongly excited $CdSe_xS_{1-x}$ crystals.[13] Furthermore, the saturation of the photon-echo signal and the bleaching of beam 2 are attributed to a saturation of resonant states of trapped excitons. Their density decreases smoothly with decreasing $h\nu$; i.e., the level of I_{cr} should also decrease, just as is seen experimentally.

Possible dephasing processes for trapped excitons are radiative recombination[12,13] and hops (accompanied by the emission of phonons) to states with a greater trapping depth. The same processes determine the luminescence kinetics of $CdSe_xS_{1-x}$ at liquid-helium temperatures, for which the characteristic times[14] are on the same order of magnitude as the values found for T_2. A decrease in the hopping probability for states with a deeper trapping level[12] explains the observed increase in T_2 with decreasing $h\nu_{ex}$.

These results do not contradict the theoretical predictions[15] of an effect of disorder and Anderson trapping on phase relaxation in semiconductors. A more detailed analysis goes beyond the scope of this letter and will be the subject of subsequent publications.

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