Optical bistability related to excitons in an uncooled semiconductor

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A resonant optical nonlinearity involving excitons has been discovered in room-temperature semiconductors. It has been found that Fabry-Perot etalons made of multilayer GaSe single crystals exhibit a bistable behavior when the excitons are resonantly excited by intense light pulses.

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Bistable semiconductor optical elements are attracting increasing interest. An optical bistability arises in Fabry-Perot resonators filled with a nonlinear medium when subjected to an intense light beam which alters the refractive index of the medium and thus the optical path length of the resonator. The transmission of the resonator becomes nonlinear, and regions of differential amplification, saturation, and hysteresis arise.[1–3] The advantages of semiconductors stem from their large nonlinearities. The nonlinearities are particularly large in the case of the resonant excitation of exci-

tons. In this letter we report the observation of a resonant nonlinearty involving excitons in uncooled GaSe semicondutors in the form of Fabry-Perot resonators. An optical bistability at room temperature has been detected previously only in composite GaAs–GaAlAs etalons in the form of superlattices.[4]

We selected GaSe crystal for this attempt to fabricate bistable optical elements operating at room temperature because of the pronounced nonlinearity near the resonant exciton line and also because of the large exciton binding energy (E_{ex}) in this material [$E_{ex} = 20$ meV (Ref. 5); $E_{ex} \sim kT$ at $T = 300$ K]. We should also point out the simplicity of fabricating resonators of suitable size through cleavage from a multilayer GaSe crystal bar and the circumstance that the exciton level lies in the range of the output frequencies of common dye lasers.

In the present experiments we used resonators cleaved from bars of hexagonal ϵ-GaSe crystals grown by the Bridgman method. The measurements were taken with the samples at room temperature. The samples were bombarded with pulses from an LZhI-402 laser, whose active medium is a solution of the dye rhodamine *B* (the wavelength can be tuned over the interval 610–635 nm; the output linewidth is about 12 Å; and the length of the output pulse is 120 ns). The resonator mirrors are formed by the natural parallel faces of the samples (the reflectance for normal incidence in the transparency region is 22%). The optic axis of the crystals, the resonator axis (normal to the plane of the layer), and the axis of the laser beam were all coincident. In this case the

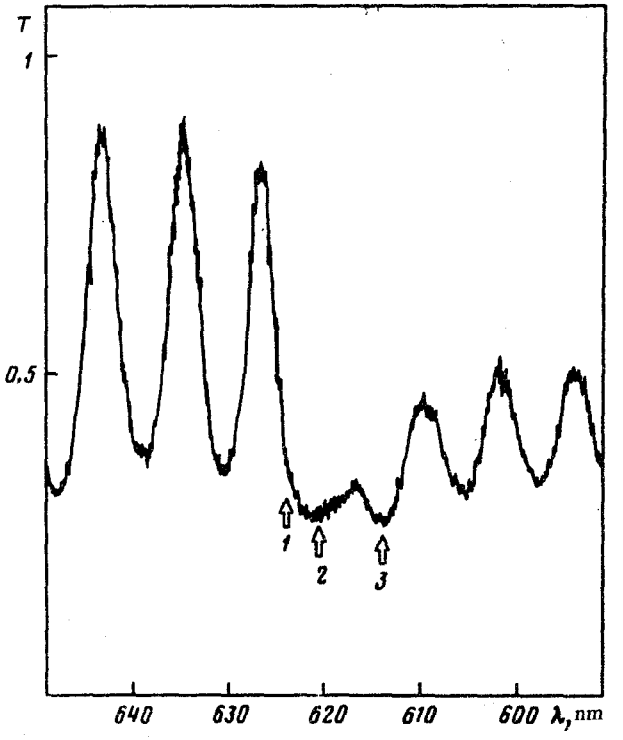

FIG. 1. Transmission spectrum of a 8-μm-thick GaSe crystal.

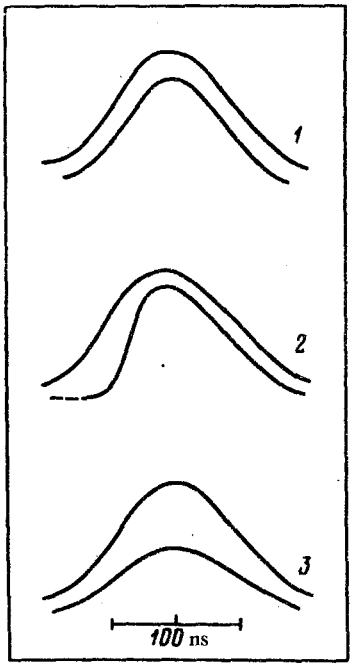

FIG. 2. Oscilloscope traces of the laser pulse, of the pulse incident on the etalon, and of the pulse transmitted through the etalon. The laser wavelengths are: 1—624 nm; 2—620.7 nm; 3—614 nm (see the arrows in Fig. 1).

laser beam was polarized perpendicular to the optic axis, and the exciton transition was partially allowed by the spin-orbit interaction; the exciton absorption coefficient was $\sim\simeq 10^3$ cm^{-1}. Because of the latter circumstance it was possible to work with resonators of suitable length, ~ 10 μm, with a short relaxation time, $\sim 10^{-13}$ s. The incident beam and that transmitted through the sample were detected with coaxial photocells connected to a dual-trace oscilloscope. The two measurement channels were identical.

Figure 2 shows oscilloscope traces of the incident laser pulse and of the pulse transmitted through a resonator consisting of a GaSe crystal 8 μm thick for the cases in which the wavelength lies in the transparency region, in the exciton absorption region, and in the interband absorption region (the transmission spectrum of the sample is shown in Fig. 1). Figure 3 shows the results of an analysis of these oscilloscope traces. We note that as the laser wavelength is tuned, while its intensity is held constant, the nature of the transmission changes: (1) A nonlinear transmission arises only in the case of a resonant exciton transition. (2) The curve of the nonlinear transmission exhibits hysteresis and a region of differential amplification (the light intensity at the exit from the sample varies to a greater extent than that at the entrance to the sample). (3) The absorption is linear when the laser frequency is tuned away from the exciton resonance line (the transparency region and the region of interband transitions). The light intensity at which the room-temperature bistable element is switched (Fig. 3) is

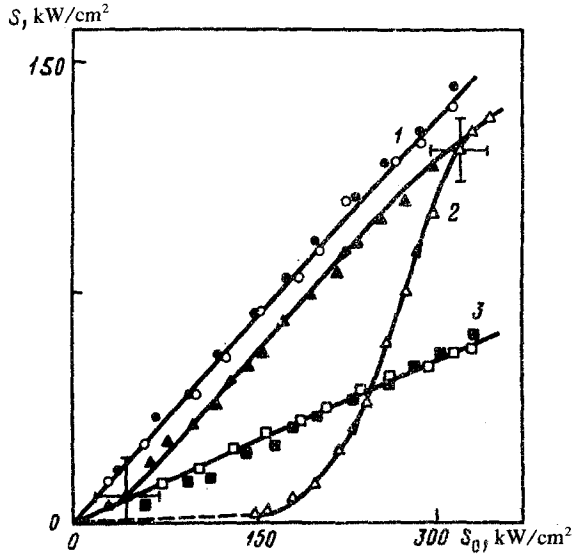

FIG. 3. Intensity of the light transmitted by the etalon vs the intensity of the incident light.

approximately equal to the values found for GaSe etalons cooled to liquid-nitrogen temperature.

Thermal effects are apparently not governing in this bistability, since the bistable behavior is not seen when the laser output is tuned to the region of the strong absorption due to interband transitions (line 3 in Fig. 3). Additional experiments will make it possible to determine the mechanism for the observed optical bistability of GaSe crystals at room temperature. The bistable behavior of the resonators might result from many effects: screening of excitons,[6,7] a shift of an exciton level during pronounced excitation,[8] nonequilibrium scattering by phonon,[9] etc. The lack of a delay in these events, the short time required for the establishment of the field in the thin semiconductor resonators, and the possibility of reducing the recovery time for the exciton absorption after excitation (by adding impurities to the crystal, for example) raise the hope that it will be possible to fabricate bistable optical elements with picosecond switching times at room temperature.

In conclusion we wish to call attention to the promising possibility of using quasi-two-dimensional systems, in which the exciton binding energy is higher, to fabricate bistable optical elements. An increase in the exciton binding energy weakens the thermal ionization of the excitons and reduces the relative contribution of the absorption due to interband transitions at the resonant excitation frequency. The use of intercalated multilayer semiconductors[10] for this purpose may be of some interest.

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