Giant exciton resonance in nonlinear optical activity

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A giant exciton resonance has been detected experimentally in the nonlinear optical activity of a gallium arsenide crystal.

1. We have studied the intensity-dependent change in the polarization structure of an intense, linearly polarized wave in a gallium arsenide crystal in the region of a single-photon exciton resonance ($\hbar\omega \sim 1.5$ eV). The angle of the nonlinear rotation of the polarization plane exceeds 15°, and the corresponding constant of the nonlinear

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optical activity reaches a value of 300 deg·cm/MW.

Nonlinear polarization self-effects of light in semiconductors have recently attracted considerable interest. In particular, the two-photon exciton resonance has been studied by nonlinear-optical-activity spectroscopy in gallium arsenide.[1] The nonlinear depolarization of light[2,3] and the self-rotation of the polarization ellipse[4] at a two-photon biexciton resonance have been observed in cuprous chloride crystals. The interest in research in this direction stems primarily from the new spectroscopic possibilities for studying the nonlinear susceptibilities of semiconductors and the promising outlook for the use of these effects in bistable optical devices of a new type for information processing, which code signals in terms of the polarization state of the light.

2. In crystals with the zinc blende symmetry ($\bar{4}3m$, gallium arsenide), a nonlinear, intensity-dependent change in the polarization structure of linearly polarized light could result only from a nonlinear gyrotropy and a nonlinear anisotropy (a nonlinear optical activity).[5] The possibility of observing large values of the specific constants for the self-effects of light near exciton and biexciton resonances in crystals was first pointed out in Ref. 6. The nonlinear optical activity associated with a nonlinear anisotropy due to an undulation of the band structure of the crystal arises near exciton absorption frequencies because of the resonant nature of the corresponding susceptibility. A nonlinear gyrotropy has features other than resonant effects. The spatial dispersion effects in optics, including nonlinear effects, are known to be determined by the small parameter a/λ, where λ is the wavelength of the light, and a is a scale length in the medium (the lattice constant in the nonresonant case). In the region of an exciton resonance, the Bohr radius of the exciton ($R_B = 150$ Å for GaAs) serves as a scale length,[1)] and the contribution of the nonlinear anisotropy to the polarization self-effect (the nonlinear rotation angle $\beta^{(2)}$) which is the dominant contribution in the nonresonant case, becomes comparable to or even smaller than the contribution from the spatial dispersion of the nonlinearity (the angle $\beta^{(1)}$): $\beta^{(1)}/\beta^{(2)} \simeq 16\pi na/\lambda \gtrsim 1$.

3. In the present experiments we studied gallium arsenide samples with thicknesses over the range 2–70 μm. The measurements were carried out at a crystal temperature of 100 K. The light source was a parametric generator using a $LiIO_3$ crystal (the pulse repetition frequency is ~ 2 Hz, the length of the single pulses is ~ 15 ps, and the spectral width of the light is ~ 2 meV).

Near the single-photon exciton resonance we detect an intensity-dependent depolarization of the light, which reaches 10% (Ref. 8). The light, incident on the crystal along the $\langle 001 \rangle$ direction, has a linear polarization at an angle $\varphi = \pi/8$ with the $\langle 100 \rangle$ axis. Here the "depolarization of the transmitted light" is to be understood as the ratio of the energies in the pulses for the components of the light at the exit from the crystal. These components are polarized perpendicular to and parallel to the original polarization direction. The depolarization increases at the resonances with increasing excitation level, up to $Q = 1$ pJ/μm^2. With a further increase in the excitation level, the degree of polarization falls off, the line broadens, and there is a complete degradation at excitation levels above 20 pJ/μm^2. The relative contribution of the rotation of the polarization plane (the principal axis of the ellipse) to the depolarized component in the test crystal is 70% at resonance (Fig. 1).

When the light transmitted through the crystal is observed with an analyzer

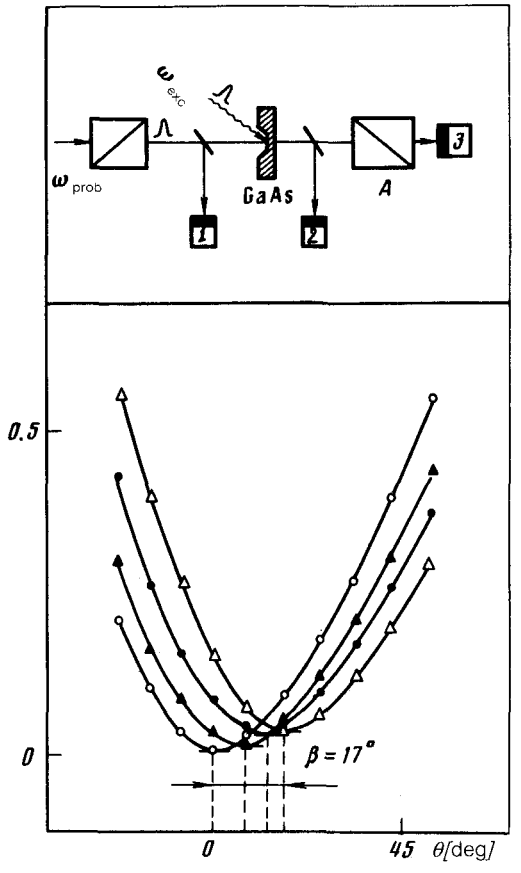

FIG. 1. Transmission of the system consisting of the polarizer, the sample, and the analyzer versus the relative position of the polarizer and the analyzer (the angle θ) for various excitation intensities: ●—0.4 pJ/μm^2; △—1.0 pJ/μm^2 (Ref. 7); ▲—6.0 pJ/μm^2; ○—"empty" polarimeter, without a sample. The inset is the optical arrangement in the experiments. *A*—analyzer; *P*—polarizer; 1,2,3—photodetectors.

oriented at an angle of 45° with respect to the polarizer, the transmission coefficient of the analyzer depends on only the angle, through which the polarization plane (the principal axis of the ellipse) is rotated in the nonlinear crystal. It does not depend on the magnitude of the induced eccentricity or on the relative size of the incoherent component in the light flux. It thus is possible to measure the frequency dispersions of the "pure" rotation of the polarization plane (Fig. 2). The angle of the nonlinear rotation exceeds 15°; the rotation is of a resonant nature; and the center of the line agrees in frequency with the maximum of the depolarization line. An important point is that the rotation angle differs in sign for different orientations of the crystal ($\varphi = \pi/8$ and $\varphi = -\pi/8$); the maximum values of the rotation angle differ for these two orientations. In experiments in which the light transmitted through the polarimeter was passed through a monochromator tuned in synchronization with the parametric light source (the slit width is $\sim$0.2 meV) yielded results identical to those found in experiments in which there was a direct detection of the light transmitted by the polarizer, the sample, and the analyzer.

4. In discussing possible mechanisms for the depolarization of the light which could serve as alternatives to the nonlinear optical activity we must bear in mind that

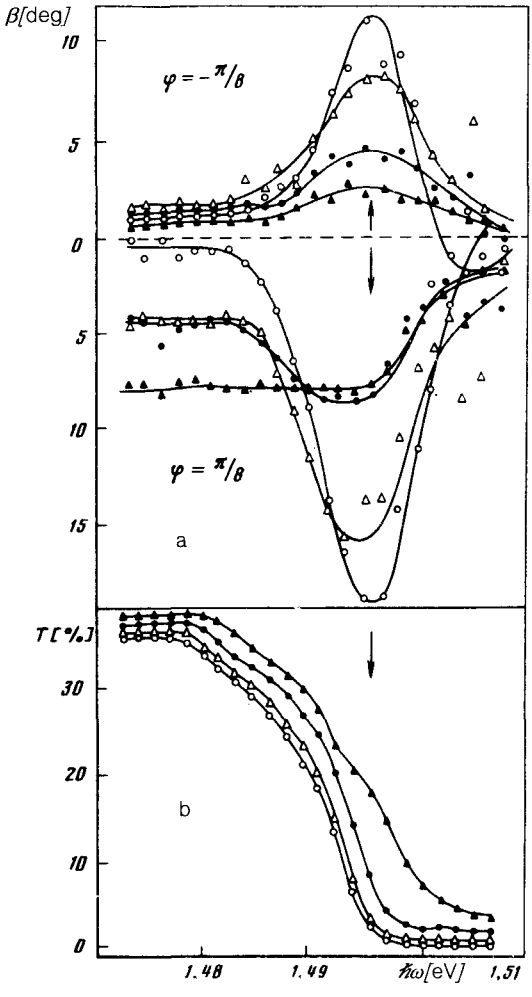

FIG. 2. a—The angle (β) of the nonlinear rotation; b—the transmission (T) of a sample 70 μm thick versus the energy of the excitation photon, $\hbar\omega$, for various excitation intensities [$\varphi = \sphericalangle(\mathbf{E};\langle 100\rangle)$]: ○—1.6 pJ/$\mu$m^2; △—3.0 pJ/$\mu$m^2; ●—5.0 pJ/$\mu$m^2; ▲—15 pJ/$\mu$m^2. The arrow is the frequency position of the exciton resonance.

the appearance of the light component polarized orthogonal to the pump light could not be a consequence of birefringence due to a second-order spatial dispersion (since the $\langle 001\rangle$ direction is the optic axis) or a multifrequency scattering (since the orthogonal component arises primarily as a result of a "pure" rotation of the polarization plane, which is of a nonlinear nature). The observed rotation of the polarization cannot be attributed to a self-rotation of the polarization ellipse[4] because (a) the light incident on the crystal is linearly polarized and (b) the sign of the rotation of the polarization plane depends on the relative orientation of the electric field of the probing wave and of the symmetry axis of the crystal, as is characteristic of nonlinear optical activity in crystals of this symmetry,[1] but not characteristic of the self-rotation of the polarization ellipse. The excitonic nature of the observed giant effect in the nonlinear optical activity is proved by (a) the frequency position of the maximum of the polarization resonance, which coincides with the absorption resonance in the 1s

state of a free exciton in GaAs, (b) the significant decrease in the self-rotation of the polarization as the sample temperature is raised to 300 K (this decrease stems from an increase in the rate of thermal dissociation of excitons), and (c) the degradation of the polarization resonance with increasing excitation intensity, because of the screening at high exciton densities[9] (Fig. 2a).

5. The experiments show that it is possible to degrade the exciton resonance not only by raising the intensity of the probing beam but also by arranging a two-photon excitation of free carriers in the crystal during nonlinear absorption of light with a wavelength of 1.06 μm ($\hbar\omega_{exc} = 1.12$ eV). A complete degradation of the exciton resonance at a 20-ps delay of the probing pulse ($\hbar\omega_{prob} = 1.497$ eV) with respect to the exciting pulse sets in when the intensity of the excitation at 1.06 μm is $Q^{exc} = 75$ pJ/μm^2 ($Q^{prob} = 1$ pJ/μm^2), corresponding to a free-carrier density $N \sim 5 \times 10^{17}$ cm^{-3}. A complete restoration of the exciton resonance, as a result of the recombination of electron-hole pairs, occurs 2.5 ns after the excitation.

6. The nonlinear rotation and the depolarization are observed with a signal-to-noise ratio of 1000–100 in relatively thick samples under conditions of a nonlinear brightening of the crystal (Fig. 2b). An exciton resonance could not be observed in the absorption spectra; i.e., the method of a polarization self-effect is exceedingly sensitive for the spectroscopy of excitons.

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[1)]We believe it would be very interesting to study the effect on the nonlinear optical activity of a "plasma" spatial dispersion, resulting from a coherent spatial transport of excitation by an exciton over a mean free path.[7]

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