Exciton polarization self-effect of picosecond light pulses in a composite GaAs/$Al_x Ga_{1-x}$As superlattice

S. A. Akhmanov, N. I. Zheludev, C. N. Ironside,[1)] Z. M. Kostov, and S. V. Popov

M. V. Lomonosov Moscow State University;[1)] University of Glasgow

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A nonlinear depolarization of picosecond light pulses has been observed near an exciton absorption resonance in a composite GaAs/$Al_x Ga_{1-x}$As superlattice.

1. In this letter we are reporting the results of an experimental study in which a strong, nonlinear depolarization of light near a single-photon exciton resonance was detected for the first time in a composite GaAs/$Al_x Ga_{1-x}$As superlattice. Measurements of the dispersion of the nonlinear depolarization carried out at room temperature and liquid-nitrogen temperature clearly demonstrate that this depolarization is of an exciton nature. The magnitude of the effect is at least 100 times that in bulk GaAs under dynamic-bleaching conditions[1,2] at comparable intensities and comparable crystal thicknesses.

2. A resonant depolarization of light near the line of a two-photon biexciton absorption resonance was first observed in cuprous chloride (Ref. 3; see also Refs. 4

and 5). A nonlinear exciton polarization resonance has been detected in gallium arsenide, where an essentially "pure" rotation of the polarization plane was observed in a "transmission" arrangement[1,2] and a "reflection" arrangement.[6] The sign (direction) of the rotation was related to the orientation of the crystallographic axes of the sample with respect to the electric vector of the wave. Near the exciton resonance, the induced gyrotropy in a transmission arrangement is huge, even at the scale of the natural optical activity. The specific self-rotation constant reaches 1000 deg/mm at an applied intensity on the order of 10 MW/cm^2 at resonance.

Because of the quantum-size effects, which increase the binding energy of free excitons and which "accentuate" the exciton resonances in the linear and nonlinear absorption spectra in composite superlattices based on gallium arsenide, the quantum superstructures are obvious candidates for a study of polarization nonlinearities.

3. We studied the intensity-dependent depolarization of picosecond light pulses, which initially had a linear polarization, as they propagated through a composite superlattice along the ⟨001⟩ direction, which is normal to the plane of the structure. The superstructure contained 20 GaAs/$Al_xGa_{1-x}As$ layer pairs ($x = 0.3$), with equal well and barrier thicknesses of 97 ± 3 Å. The superstructure was synthesized by electronO-beam epitaxy. The optical element was a diaphragm 2 mm in diameter, with transition layer and substrate etched away. The measurements were taken at sample temperatures of 150 and 300 K.

As the light source we used a parametric generator based on a $LiIO_3$ crystal pumped by a single-pulse Nd:YAG laser with frequency doubling. The length of the pulse from the parametric generator was ~20 ps; the spectral width of the light was ~2 meV; and the pulse repetition frequency was 2 Hz. The sample was positioned between crossed high-quality Glan prisms. The residual transmission coefficient of the polarizer-sample-analyzer system was 10^{-4} in the transparency region of the superlattice. We take the "unknown degree of depolarization of the light" to be the ratio of the pulse energies for the light components at the exit from the crystal which are polarized perpendicular to and parallel to the original direction. We are thus not distinguishing among the induced rotation, the self-induced ellipticity, and the incoherent depolarization. However, the crystal orientation was chosen on the basis of the requirements imposed on the experimental geometry by the condition for effective observation of a nonlinear optical activity.[5] The light was incident along a ⟨001⟩ direction and has a linear polarization which made an angle of 22.5° with the angle ⟨100⟩ axis. In this orientation the self-induced ellipticity and the self-induced polarization rotation, which are related to the spatial dispersion of the nonlinearity and the anisotropy of the nonlinear absorption, should be seen simultaneously. The energy of the light pulses which excited the superstructure was measured by a photometric detector. The energy of the pulses in the reference and signal channels of the polarimeter was measured by cooled photomultipliers. The optical layout and the other experimental details were simllar to those described in Ref. 7.

4. The results are summarized in Fig. 1 and Fig. 2a. In the depolarization spectra we see a clearly defined line, which corresponds to an exciton absorption in this structure. We can clearly see a splitting of the peak into two isolated lines, separated by an energy interval of 10 meV. At 150 K the resonant depolarization increases with the

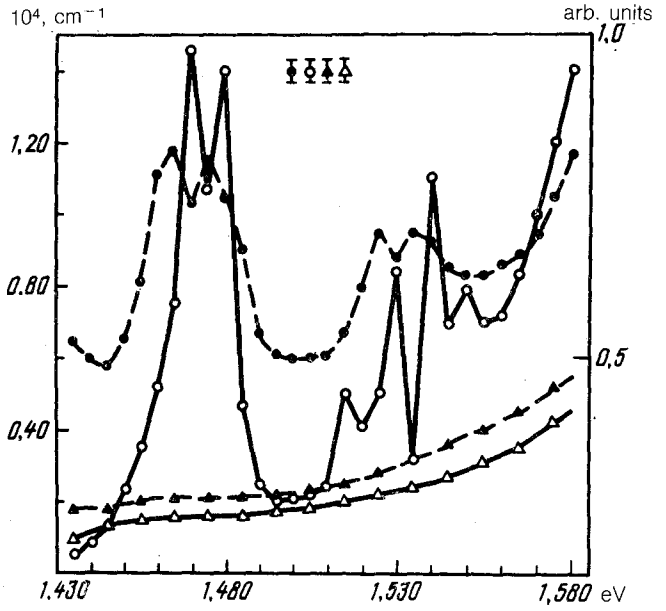

FIG. 1. Degree of depolarization of the light (○, △; arbitrary units) and absorption coefficient (dashed line; ●, ▲, units of reciprocal centimeters) at an excitation level of 10^{-2} pJ/μm^2 (●,○) or 1 pJ/μm^2 (▲ and △) as functions of the excitation photon energy (in electron volts). The sample temperature was 300 K.

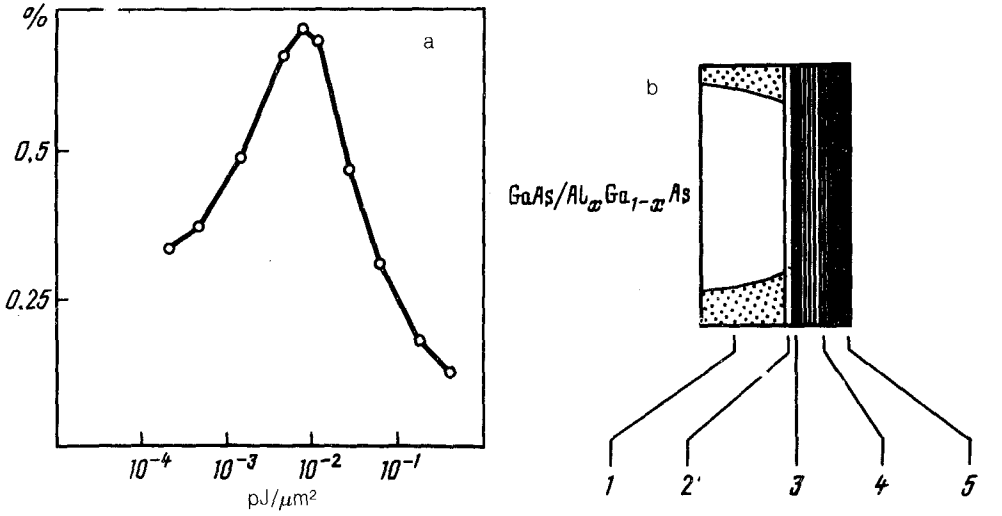

FIG. 2. a: Degree of depolarization (the relative energy of the depolarized component) at the maximum of the one-photon exciton resonance at 150 K. The excitation energy, in picojoules per square micron, is plotted along the abscissa. b: Architecture of the composite GaAs/$Al_xGa_{1-x}As$ superstructure. 1—Substrate, $x = 0$; 2—transition layer, $x = 0.05$–0.6; 3,5—protective layers, $x = 0.16$–0.17; 4—20 pairs of layers of the superstructure with $(x = 0)/(x = 0.3)$, with a total thickness of 0.40 μm ($N_p = 10^{16}$ cm^{-3}).

pump to 10^{-2} pJ/μm^2, reaching 0.78. With a further increase in the excitation, the exciton depolarization line begins to degrade, and it fades away completely at an excitation level on the order of 1 pJ/μm . The number density of the photoinduced free excitons can be estimated from $N = Qk(\hbar\omega)^{-1}R$, where k is the absorption coefficient in the resonant region, Q is the energy density of the exciting light, R is a loss factor associated with the reflection, and $\hbar\omega$ is the photon energy of the absorbed light. For 1 pJ/μm^2 this expression yields $N = 2\times10^{18}$ cm^{-3}. At photon energies 60 meV above the resonant value, the depolarization stems from an entrainment of interband transitions. As the excitation intensity is raised, the spectral edge of the interband absorption and of the interband depolarization shift into the "blue" region because of a dynamic filling of the conduction band by photoinduced carriers.

Despite the difference between the microscopic symmetry of the superlattice and that of the single crystal, the direction normal to the plane of the superlattice (the direction which we studied) retains the symmetry of an inversion fourfold rotation axis. This circumstance means that the nature of the depolarization exciton resonance may be associated with both a latent anisotropy of the cubic nonlinear susceptibility and a nonlocal nature of the nonlinear response.[7] According to the results of a recent study by Dubenskaya *et al.*,[8] an anisotropy appears in the cubic susceptibility of gallium arsenide near the resonance of a one-photon interband (exciton) absorption because of a difference between the matrix elements for transitions from the bands of heavy and light holes to the conduction band (or exciton band). Carrying out elementary calculations of the spatial dispersion of the nonlinearity is a more-complex task, requiring data on the matrix elements of the magnetic dipole and quadrupole transitions. A distinctive feature of a superlattice should be manifested here, in the existence of several length scales of the nonlocality: the lattice constant of the crystal lattice, the diameter of an exciton, and the period of the superstructure.

5. The strong nonlinear polarization self-effect resonance observed in a composite superlattice is the most promising entity for (a) studying polarization multistability and instability (the "disruptive" polarization instability) in cubic crystals with an anisotropic nonlinear refraction and an anisotropic nonlinear absorption with external optical feedback[9,10] and (b) for developing new types of elements for controlling light with light, in particular, polarization modulators.[2]

[1]N. I. Zheludev, Z. M. Kostov, A. A. Karasev, and M. S. Nunuparov, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 578 (1986) [JETP Lett. **43**, 747 (1986)].
[2]Z. M. Kostov and N. I. Zheludev, Opt. Quantum Electron. **20**, 30 (1988).
[3]N. Itoh and T. J. Katohno, J. Phys. Soc. Jpn. **51**, 707 (1982).
[4]N. I. Zhludev, J. Phys. Soc. Jpn. **54**, 2778 (1985).
[5]M. Kuwata and M. Nagasava, J. Phys. Soc. Jpn. **56**, 39 (1987).
[6]Z. M. Kostov and N. I. Zheludev, Opt. Lett. **13**, 640 (1988).
[7]S. A. Akhmanov, N. I. Zheludev, and R. S. Zadoyan, Zh. Eksp. Teor. Fiz. **91**, 984 (1986) [Sov. Phys. JETP **64**, 579 (1986)].
[8]M. G. Dubenskaya, T. M. Il'inova, and A. A. Fortygin, in: Proceedings of the Thirteenth International Conference on Coherent and Nonlinear Optics (Minsk, 6–9 September 1988), Part II, p. 126.
[9]S. A. Akhmanov and N. I. Zheludev, in: *Nonlinear Phenomena in Solids* (ed. M. Borisov), World Scientific, Singapore, 1985, p. 233.
[10]J. Ymoto and K. Otsuka, Phys. Rev. Lett. **54**, 1806 (1985).

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