First-order spatial dispersion in hot gallium arsenide electron-hole plasma: picosecond-resolution polarization diagnostics

N. I. Zheludev and D. Yu. Parashchuk
M. V. Lomonosov State University, Moscow

(Submitted 10 May 1990)
Pis'ma Zh. Eksp. Teor. Fiz. **52**, No. 1, 683–686 (10 July 1990)

A new effect has been observed: the induction of an optical gyrotropy in a nonequilibrium electron-hole plasma of optically pumped gallium arsenide. The effect was detected in a "pump-probe" arrangement in which a laser beam with $\lambda = 532$ nm was reflected from a [001] surface.

In this letter we are reporting an experimental study in which a time-varying gyrotropy of a GaAs single crystal was observed in "reflection" experiments for the first time. The crystal was pumped by picosecond light pulses. The rotation of the polarization azimuthal angle was 10^{-5} rad at a hot-carrier density $\sim 10^{20}\,\text{cm}^{-3}$. The lifetime of the induced gyrotropy was ~ 300 ps.

Reflection nonlinear polarimetry has already proved its effectiveness for studying exciton states[1,2] and isotropic electron-hole plasmas in semiconductors.[3] For diagnostics of the nonlocal hot-carrier response in GaAs, we used a reflection micropolarimetry technique with a picosecond time resolution. We used a pump-probe principle: The [001] crystallographic surface of the GaAs was pumped by a linearly polarized optical pump pulse, which altered the conditions for the reflection of the probe beam, which had the same initial polarization and wavelength. In other words, the polarization azimuthal angle rotated. The pump beam and the probe beams propagated approximately along the normal to the surface. The pulsed light in the probe system (from a quasi-cw YAG laser) and that in the pump system (more intense by a factor of 15) had the following parameter values: a wavelength of 532 nm, a pulse length of 60 ps (in packets of 20 pulses), a packet repetition frequency of 1.3 kHz, and a pump spot area of $2\times 10^{-4}\,\text{cm}^2$ on the sample. The probe-beam polarimeter was formed by Glan and Cotton prisms with an extinction coefficient $\xi = 1.2\times 10^{-6}$. The measurements were carried out by synchronous-detection apparatus; the pump beam was chopped at a frequency of 218 Hz. The sensitivity of the polarimeter at the averaging time $T = 20$ s was 10^{-6} rad. We studied single-crystal plates, both sides of which were optically polished and treated with a bromine-methanol etchant.

The following results were established: (a) The angle through which the polarization plane of the reflected probe beam is rotated increases monotonically with increasing pump intensity (Fig. 1a). (b) A rotation of the crystal through 90° around the $\langle 001\rangle$ direction or a rotation of the crystal through 180° around the $\langle 110\rangle$ direction, i.e., a change in the pumped surface, leads to a change in the sign of the rotation (Fig. 2). (c) The gyrotropy relaxes over a time on the order of 300 ps. This time was determined by varying the optical delay θ between the pump beam and the probe beam

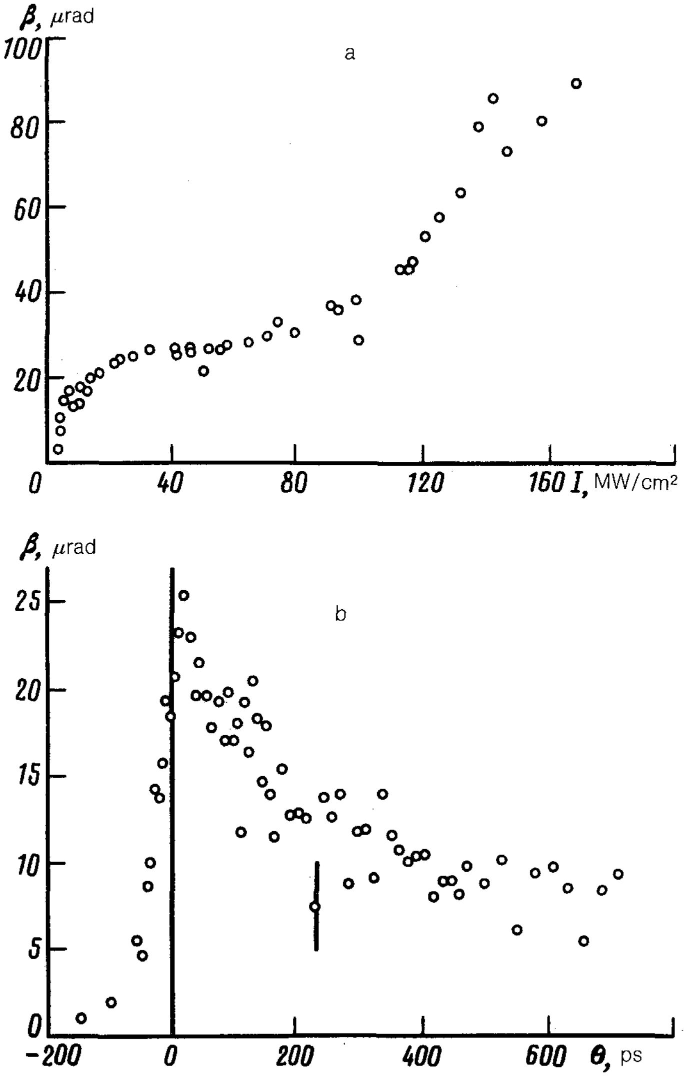

FIG. 1. The angle (β) through which the polarization of the probe beam is induced to rotate versus (a) the pump intensity I_p ($\theta = 0$) and (b) the delay (θ) of the probe beam with respect to the pump ($I_p = 80$ MW/cm^2).

(Fig. 1b). (d) The orientation dependence of the induced-rotation effect remains the same in nature over the entire intensity range studied and also as we vary the delay between the probe and the pump pulses over the lifetime of the induced gyrotropy.

The minor term in the nonlinear polarization, which is responsible for the observed induced gyrotropy of GaAs, can be written in the form

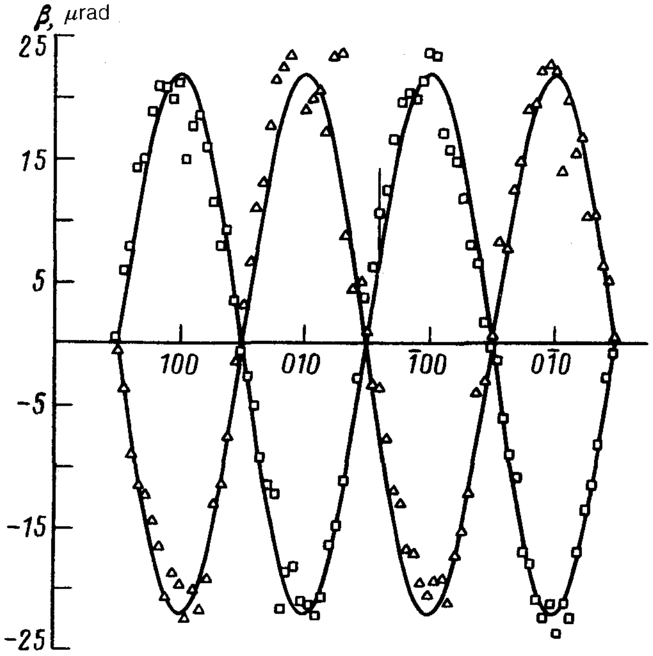

FIG. 2. The angle (β) through which the polarization of the probe beam is induced to rotate versus the orientation of the azimuthal polarization of the pump with respect to the symmetry axes of the crystal. □, △—Experimental points corresponding to the pumping of opposite faces of the crystal ($I_p = 50$ MW/cm^2).

$P_i^{nl} = k_m \gamma_{ijklm}^{(3)} E_j^s E_k^p (E_l^p)^*$, where k is the wave vector of the probing wave, and E^p and E^s are the fields of the pump and probe pulses. The symmetry of the effect which results from a nonlinear response of this type corresponds completely to the experimental results. For example, as the crystal is rotated 90° around the fourfold rotation-inversion axis (such a rotation is equivalent to the sequential use of a fourfold rotation inversion and an inversion), the rotation angle should change sign, since the operation of inversion, while not affecting the actual tensor $\gamma^{(3)}$ of the material, does change the sign of the wave vector **k**. This sign inversion in crystals of symmetry $\bar{4}3m$ distinguishes any optical effect due to a first-order spatial dispersion in a fundamental way from effects which are unrelated to the direction of the wave vector.Correspondingly, a rotation of the crystal through 180° around the ⟨110⟩ direction should also be accompanied by a change in the sign of the induced gyrotropy.

To explain the observed lifetime of the gyrotropy will require a microscopic analysis. The number density of photoinduced free carriers at a pump intensity ~50 MW/cm^2 is estimated to be 5×10^{20} cm^{-3}, and the particular reflection properties of the surface are determined primarily by plasma effects.[3] As a result of the optical pumping and the redistribution of charge into high-lying indirect valleys, the effective-mass tensor acquires components of such a nature that the field of the probe beam gives rise to circular currents (the effects of the first-order spatial dispersion are being taken into account). The result is to change the polarization of the reflected signal (cf. the appearance of a transverse mass during carrier heating by a static electric field[4]).

The induced gyrotropy relaxes as a result of recombination, diffusion, and the redistribution of electrons into the central valley of the conduction band with an isotropic effective mass.

[1] M. J. Kuwata, J. Lumin. **38**, 247 (1987).
[2] Z. M. Kostov and N. I. Zheludev, Opt. Lett. **13**, 640 (1988).
[3] D. H. Auston and C. V. Shank, Phys. Rev. Lett. **32**, 1120 (1974).
[4] E. M. Conwell, *High Field Transport in Semiconductors,* Academic, New York, 1967.

Translated by D. Parsons