Determination of the sign of the g factor and observation of deformation of epitaxial films in the transverse effect of optical orientation in semiconductors

V. L. Vekua, R. I. Dzhioev, B. P. Zakharachenya, E. L. Ivchenko, and V. G. Fleĭsher

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences

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We investigate the transverse effect of optical orientation in semiconductors of cubic symmetry. The effect is connected with rotation of the vector of the total angular momentum of the free carriers in the transverse magnetic field, and is observed in a direction perpendicular to the direction of the magnetic field and to the direction of the light beam producing the spin orientation. This effect was used for a direct measurement of the sign of the g factor of the conduction electrons in the GaAs crystals, and also in the solid solutions GaP_xAs_{1-x} and $Al_xGa_{1-x}As$ grown in the form of epitaxial films on gallium arsenide substrates. It is shown that the study of the transverse optical-orientation effect makes it possible to investigate the optical anisotropy of thin semiconducting films. The experimental results offer evidence of the presence of uniaxial deformation in the investigated epitaxial films. In the case of GaP_xAs_{1-x}, as shown by calculation, this deformation amounts to 0.7×10^{-3}.

The method of optical orientation is presently being used with success to investigate semiconductor crystals[1-4]. The optical orientation was heretofore detected principally by means of the circular polarization of the photoluminescence, the observed effect being that of longitudinal optical orientation (along the beam of exciting light). We report here the observation of a transverse optical orientation effect. In this effect, the orientation is detected in a direction perpendicular to the direction of the exciting beam. It is produced by applying a magnetic field perpendicular to both of these directions. The degree of circular polarization of the luminescence in the perpendicular direction is in this case an odd function of the magnetic field intensity **H** as well as of the g-factor of the conduction electrons, so that the sign of the g-factor can be determined directly. This direct method of measuring the sign of the g-factor undoubtedly has a number of advantages.

If the recombination radiation is weakly absorbed in the crystal, then it is possible in the transverse effect to vary the optical path length of this radiation by displacing the exciting beam. This provides a convenient and sensitive method of investigating optical anisotropy of thin semiconductor films, due for example to internal or external stresses and electric fields.

1. DIRECT DETERMINATION OF THE SIGN OF THE G-FACTOR OF FREE CARRIERS

To determine the sign of the g-factor of the conduction electrons in crystals of the III-V group, one usually uses the theoretical formula obtained in[5] in the two-band approximation:

$$g_n = 2\left[1 - \frac{2}{3}\frac{|P|^2}{m_0}\frac{\Delta}{E_g(E_g+\Delta)}\right]; \qquad (1)$$

Here E_g is the width of the forbidden band, Δ is the spin-orbit splitting of the valence band, and m_0 is the mass of the free electron. The interband matrix element of the momentum operator $P = \langle S|\hat{p}_z|Z\rangle$ can be expressed, by starting from the two-band model, in terms of the effective mass of the conduction electrons[5].

An attempt to obtain a more accurate formula for the parameter P was made in[6] on the basis of a semiempirical model. In the case when $|g_n| > 1$, this method of determining the sign of g_n is subject to no doubt, and both methods yield identical results. In the case of gallium arsenide, however, the expression in the square brackets of (1) is close to zero and the indicated methods of estimating the parameter P lead to different signs of g_n (the g-factor is equal to 0.2 and -0.06 respectively at $E_g = 1.52$ eV, $\Delta = 0.34$ eV, and $m^* = 0.067m_0$). In addition, in this case it may turn out that the contribution of remote bands to the expression for g_n is not negligibly small. It was therefore of interest to determine the sign of the g-factor of the electrons experimentally, primarily in gallium arsenide.

Figure 1 shows a scheme of the experimental geometry. An exciting-light beam from a helium-neon laser of wavelength $\lambda = 6328$ Å was incident on the crystal in the z direction; the recombination radiation was extracted in the x direction; the magnetic field was applied parallel to the y axis.

In a magnetic field, the balance equations for the number of electrons n in the conduction band and for the vector of their resultant spin **s** take the form

$$\frac{n}{\tau_0} = w, \quad \frac{\mathbf{s}}{T_1} + [\mathbf{s}\omega_L] = \dot{\mathbf{s}}. \qquad (2)$$

Here $s = |n_{+1/2} - n_{-1/2}|$, $n_{\pm 1/2}$ is the number of electrons with spin projection $\pm 1/2$ on the direction of the predominant orientation of the electron spins, τ_0 is the lifetime inverse to the lifetime of the directional angular momentum $T_1^{-1} = \tau_0^{-1} + \tau_s^{-1}$, τ_s is the spin relaxation time, w and **s**/2 are the rates of generation of the

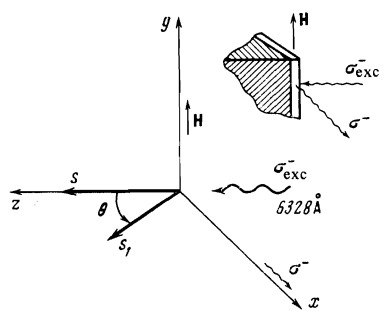

FIG. 1. Experimental geometry used to register the transverse optical-orientation effect. Excitation along the z axis, registration of the recombination radiation along the x axis; $s_1 = s[1 + (\omega_L T_1)^2]^{-1/2}$.

electrons and of the directional angular momentum of the electrons in the case of interband absorption of circularly polarized light, $\omega_L = g_n\mu_B H/\hbar$ is the frequency of the Larmor precession of the electron spins in the magnetic field, and μ_B is the Bohr magneton.

The solution of the vector equation (2) is

$$s = T_1(\dot{s} + T_1[\omega_L s]) / (1 + (\omega_L T_1)^2). \quad (3)$$

According to (3), in a transverse magnetic field the fector **s** is rotated through an angle $\theta = \tan^{-1}(g_n\mu_B H_y T_1/\hbar)$ and its modulus decreases by a factor $[1 + (\omega_L T_1)^2]^{1/2}$. Recognizing that the degree of circular polarization of the luminescence in the direction of **e** ($|\mathbf{e}| = 1$) in crystals of the III–V group is $\rho_\sigma = -0.5\mathbf{s}\cdot\mathbf{e}/n$[7], we obtain

$$\rho_\sigma^{(x)} = 0{,}5\mathscr{P}_0 \frac{T_1}{\tau_0} \frac{H/H_{1/2}}{1+(H/H_{1/2})^2} \operatorname{sign}(g_n H_y \sigma_{exc}), \quad (4)$$

where $\sigma_{exc} = \pm 1$ for right-hand and left-hand circular polarization of the exciting light, respectively, $H_{1/2} = \hbar/|g_n|\mu_B T_1$, and $\mathscr{P}_0 = \dot{s}/w$ is the degree of spin orientation of the electrons at the instant of their production. We note that in contrast to the longitudinal effect, when the degree of orientation decreases by a factor of 2 at $H = H_{1/2}$, in the transverse effect $\rho_\sigma^{(x)}$ reaches a maximum at this value of the magnetic-field intensity.

Figure 2 shows the experimental setup for the study of the transverse optical-orientation effect. The circular polarizer CP1, consisting of a linear polarizer and a quarter-wave mica phase plate, makes the emission of the helium-neon laser LG-36 circularly polarized. The sample is mounted on a cold finger and is placed in the gap of the magnet through an opening in the end cap. The measurements were made at 85°K. The use of a diaphragm ensures registration of the radiation from the crystal surface perpendicular to the x axis (see Fig. 1).

The circular polarization was detected with the aid of a quartz analyzer A[8] and a synchronous detector. The ratio of the signals with the circular polarizer CP2 removed and inserted determines the degree of polarization of the radiation. The spectral instrument MDR-1 separated a narrow quantum-energy interval near the maximum of the luminescence line corresponding to transitions to the acceptor levels.

Figure 3 shows the results of measurement of the field dependences of the transverse optical-orientation effect $\rho_\sigma^{(x)}(H)$ in a GaAs crystal (curve 1), and also in the solid solutions $GaP_{0.3}As_{0.7}$ and $Al_{0.3}Ga_{0.7}As$ (curves 2 and 3). The experimental values were obtained at a fixed position of the laser spot on the xy surface near the zy surface (Fig. 1), which constituted a cleavage surface. The solid curves on Fig. 3 were calculated from formula (4). The best agreement between the theoretical and experimental results is obtained by choosing $H_{1/2}$ close to the values obtained by observation of the photoluminescence along the exciting beam. In all three crystals, the g-factor was positive.

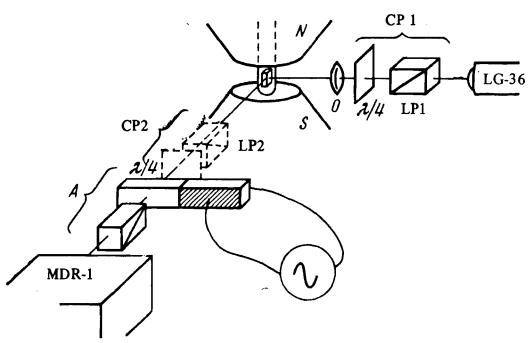

FIG. 2. Experimental setup: CP1 and CP2–circular polarizers, LG-36–helium-neon laser, A–quartz analyzer for the polarized light, MDR-1–spectrometer.

2. OBSERVATION OF DEFORMATION OF EPITAXIAL FILMS

To measure the sign of the g-factor of the electrons in GaAs-GaP and GaAs-AlAs solid solutions we used epitaxial films grown on GaAs single crystals. We observed that the degree and sign of the circular polarization of the luminescence in the transverse optical-orientation effect were dependent on the position of the laser beam on the film surface. This dependence

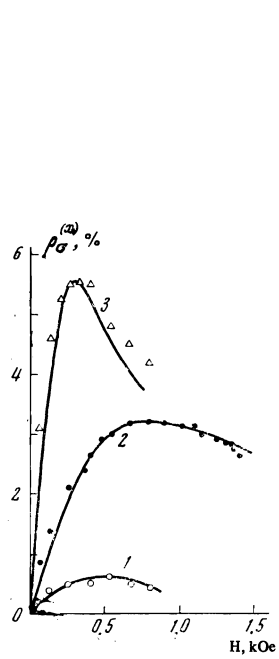

FIG. 3

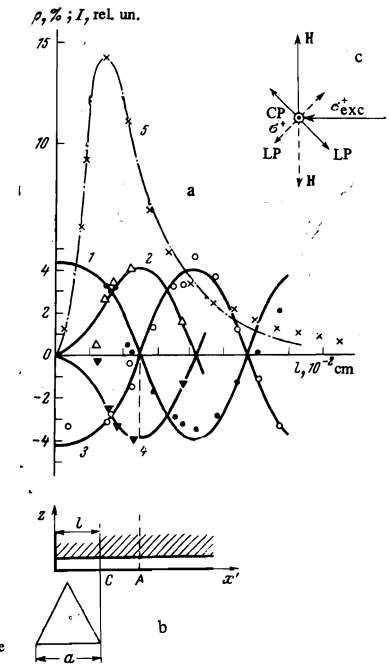

FIG. 4

FIG. 3. Experimental and theoretical plots of $\rho\sigma^{(x)}$ against the transverse magnetic field: 1–GaAs, acceptor concentration $N_A \approx 3 \times 10^{18}$ cm^{-3}; 2–$GaP_{0.3}As_{0.7}$. $N_A \approx 10^{18}$ cm^{-3}; 3–$Al_{0.3}Ga_{0.7}As$, $N_A \approx 10^{17}$ cm^{-3}. The curves were calculated from formula (4) with $H_{1/2}$ chosen equal to 0.5, 0.8, and 0.3 kOe, respectively.

FIG. 4. a) Variation of the intensity and of the polarization of the recombination radiation when the laser beam is moved along the x′ axis. ●–degree of circular polarization of the luminescence in the x direction for σ^+ polarization in the laser beam as a function of l in the case of $H_y > 0$; Δ–degree of linear polarization under the same conditions, ▼–the same for $H_y < 0$; X–variation of the recombination-radiation intensity with displacement of the laser beam. Curves 1–4 were calculated from formulas (6), and curve 5 from formula (5). b) Approximated distribution of the intensity over x with the laser beam focused on the surface of an expitaxial film; a–dimension of spot on this surface. c) Direction of oscillations of the electric vector of linearly polarized (LP) light at the point A for two opposite directions of the transverse magnetic field following excitation by circularly polarized (CP) light. The recombination radiation is directed from the plane of the figure towards the reader.

was obtained with the laser beam moving slowly from the yz surface into the interior of the crystal along the axis $x' = -x$ (Fig. 4b). The beam motion was effected by means of the objective O (Fig. 2), which moved perpendicular to this beam. The speed of the spot on the crystal surface was 300 μ/min.

The intensity distribution in the focused laser beam was measured with the aid of a slit collimator and a photodetector located directly behind the collimator. This distribution can be approximated by an isosceles triangle. The dimension of the base was 150–200 μ. We shall characterize the position of the laser beam by the coordinate l of the point C (Fig. 4b). The intensity I_L of the recombination radiation first increases as l increases from zero (curve 5 on Fig. 4a), and then decreases as a result of the increase of the absorption in the epitaxial film. At the same time, $\rho_\sigma^{(x)}(l)$ is an oscillating function of l. When the direction of the magnetic field is reversed, the phase of the circular-polarization signal is reversed (Fig. 4a).

We note here that h = 0 there are observed small signals corresponding to circular polarization. They appear to result from reflections of the recombination radiation propagating inside the epitaxial film. The experimental points were marked on Fig. 4a after subtracting the signals corresponding to H = 0 at the same values of l; this is permissible since $\rho_{H=0} \ll \rho_{H^{1/2}} < 0.06$.

The radiation was registered within the angle ±6°, corresponding to an angular divergence less than ±2° inside the crystal. Under these conditions, the observed reversal of the sign of the circular polarization cannot be connected with reflections from the internal surfaces. It is natural to assume that the oscillations of ρ_σ are connected with the optical anisotropy of the epitaxial film. If the film is birefringent, then the phase difference of the components of the light vector **E** will depend on the optical-path length in the film. One should observe maximum linear polarization at the points where the sign of $\rho_\sigma^{(x)}$ is reversed. The direction of the vector **E** is determined in this case by the orientation of the principal axes of the dielectric tensor κ_{ij}.

To measure the linear polarization and to determine the orientation of the vector **E**, a quarter-wave plate was placed in front of the quartz modulator. The linear polarization was then transformed into circular polarization and the phase of the signals at the output of the synchronous detector was determined by the orientation of the vector **E** relative to the axes of this plate. The maximum signal was obtained by rotating the plate about the optical axis. In this case the position of the plate corresponded to the orientation of the vector **E** at an angle 45° to the axes y and z, as shown in Fig. 4c for the point A. Reversal of the magnetic-field direction leads to rotation of the vector **E** through 90° (Fig. 4c). In Fig. 4a, the experimental points for the curves ρ_π, $_{Hy>0}(l)$ and ρ_π, $_{Hy<0}(l)$ were obtained for different values of l at a fixed orientation of the quarter-wave plate, corresponding to the maximum signal at the point A. Thus, the observed facts offer evidence of the presence of birefringence in the film ($n_y \neq n_z$).

We proceed to a quantitative description of the experimental results. Assume that the crystal surface element $dS = dx'dy$ is exposed to radiation of intensity $dI = j(x', y)dS$, where the function $j(x', y)$ describes the intensity distribution of the exciting light over the light spot. Assuming that the intensity of the luminescence from this section in a unit solid angle is $dI_L^{(0)} = r_L j(x', y)dS$, where r_L = const, and denoting by α the effective coefficient of recombination-radiation absorption in the interior of the crystal, we obtain an expression for the intensity of the emerging light per unit solid angle in the x direction:

$$I_L = r_L \int j(x', y) e^{-\alpha x'} dS. \tag{5}$$

Assuming that the epitaxial film has optical anisotropy, i.e., that the refractive indices n_y and n_z are not equal, we obtain the following expression for the degrees of circular polarization $\rho_\sigma^{(x)}$ and linear polarization $\rho_\pi^{(x)}$ at an angle ±45° to the axes y and z:

$$\rho_\sigma^{(x)} + i\rho_\pi^{(x)} = \rho_0 \int j(x', y) \exp[-(\alpha - i\beta)x'] dS \left(\int j(x', y) \exp(-\alpha x') dS \right)^{-1} \tag{6}$$

Here $\beta = 2\pi(n_y - n_z)/\lambda$; λ is the wavelength of the luminescence light in vacuum, and ρ_0 is the degree of circular polarization of the radiated photons. By specifying the intensity distribution $j(x', y)$ over the light spot, we can determine the dependence of $\rho_\sigma^{(x)}$ and $\rho_\pi^{(x)}$ on the position of the spot.

The solid curves in Fig. 4a, which are plots of I_L, $\rho_\sigma^{(x)}$, and $\rho_\pi^{(x)}$ against l in GaAs-GaP, were constructed for a triangular distribution of the pump-light intensity along the x axis and for a rectangular distribution along the y axis, with the following choice of the parameters: $\alpha = 60$ cm^{-1}, $|\beta| = 100$ cm^{-1}, $|n_y - n_z| = 10^{-3}$, and $a = 190$ μ. Inasmuch as sign $\rho_\sigma^{(x)}$ = sign $\rho_\pi^{(x)}$, in the case when the plate C is located to the left of the point A (see Fig. 4b), it follows from (6), in addition, that $\beta > 0$, i.e., $n_y > n_z$. We note that the g-factor was determined with the aid of formula (4) also at $l < x'_A$.

Measurements of the $\rho_\sigma^{(x)}(l)$ dependence at different values of H have shown that the position of the points $\rho_\sigma^{(x)}(l) = 0$ remains unchanged. This excludes the possibility of attributing the observed oscillations of $\rho_\sigma^{(x)}(l)$ to the Voigt effect. Nor can the observed effect be attributed to internal electric fields capable of causing optical anisotropy of the film, whether linear (the Pockels effect) or quadratic (the Kerr effect) in the field. Indeed, according to an estimate based on the formulas that determine the interband contribution to the anisotropy of the dielectric constant κ_{ij}[9, 10], the experimentally observed difference $n_y - n_z$ corresponds to an electric field intensity 10^5 V/cm, which cannot occur in strongly doped crystals in view of the screening of the field by the free carriers.

Let us examine, finally, the possibility of attributing the observed oscillations of $\rho_\sigma^{(x)}(l)$ to uniaxial deformation of the films. The difference between the dielectric-constant components κ_{yy} and κ_{zz} under uniaxial deformation is determined by the relation (see[11], Sec. 36)

$$\kappa_{yy} - \kappa_{zz} = \frac{e^2|P|^2\Phi}{\hbar^3\omega^2(E_g - \hbar\omega)^{1/2}} b(\varepsilon_{yy} - \varepsilon_{zz}),$$
$$\Phi = \frac{8}{5} \frac{\bar{m}_1\bar{m}_2}{\bar{m}_1^{1/2} + \bar{m}_2^{1/2}} + \frac{1}{10}(\bar{m}_1^{3/2} + \bar{m}_2^{3/2}), \tag{7}$$

where e is the electron charge, ω is the frequency

of the light, ϵ_{ij} is the deformation tensor, b is the constant of the deformation potential, $2\overline{m}_i^{-1} = m^{*-1} + m_{vi}^{-1}$ (i = 1, 2), and m_{vi} is the effective mass of the light and heavy holes. Assuming $\hbar\omega \approx 1.8$ eV, $E_g - \hbar\omega \approx 0.04$ eV, and $\kappa_{yy} - \kappa_{zz} \approx 2n\,(n_y - n_z)$, we obtain at $n_y - n_z = 10^{-3}$, according to (7), the value $2b(\epsilon_{yy} - \epsilon_{zz}) \approx 4$ meV. Expression (7) is valid if the following inequalities are satisfied:

$$2|b(\varepsilon_{yy}-\varepsilon_{zz})| \ll E_g - \hbar\omega \ll \Delta, E_g,$$

which is indeed the situation in our case, as can be readily seen. According to the measured temperature dependence of the elastic constants of doped GaAs[12], the ratio of the deformation-potential constants d and b is close to $\sqrt{3}$; in this case the product $2b(\epsilon_{yy} - \epsilon_{zz})$ determines the splitting of the light- and heavy-hole subbands in the center of the Brillouin zone (the point Γ) independently of the positions of the xyz axes relative to the principal symmetry axes of the crystal. Using the value b = 3 eV measured in[12], we get $\epsilon_{yy} - \epsilon_{zz} = 0.7 \times 10^{-3}$, corresponding to internal stresses on the order of 10^3 kgf/cm^2.

We note here that a similar picture of the oscillations of ρ was also observed in an epitaxial GaAlAs film, but was not observed in the case of a bulky GaAs crystal. The presence of deformation is conformed by measurement of the linear polarization along the axes y and z at H = 0 and of excitation by unpolarized light. Such polarization was observed, and the vector **E** was directed along the y axis on the long-wave edge of the luminescence line and along the z axis on the short-wave edge. This indicates a splitting of the fourfold-degenerate acceptor level under deformation, with the level ±3/2 turning out to be above the level ±1/2. These results will be reported on in greater detail in a separate publication.

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*$[S\omega_L] = S \times \omega_L$.

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