Violation of "σH invariance" in optical orientation of spins in semiconductors

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A change in the polarization of luminescence with simultaneous reversal of the signs of the circular polarization of the exciting light and of the magnetic field, a phenomenon that imitates T-invariance violation, was observed in experiment and explained.

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In experiments on optical orientation of spins in semiconductors one usually specifies two axial vectors corresponding to the angular momentum (polarization) of the quanta of the exciting light and to the external magnetic field. In all studies performed to date, the invariance of the effect of optical polarization with respect to simultaneous reversal of the directions of these vectors has not been subject to any doubt. Indeed, such an operation is the equivalent of time reversal. The results presented below demonstrate that the question of time reversal in experiments on optical orientation is not a trivial one. These results are connected with a manifestation of a nonequilibrium character of the optical orientation under conditions of crystalline anisotropy.

In semiconductors of the GaAs type, the degree of circular polarization ρ of the recombination radiation is equal to the projection of the average spin of the electrons S on the direction of observation $\mathbf{n}$ ($\rho = \mathbf{S}\cdot\mathbf{n}$). In a transverse magnetic field $\mathbf{H}$, in the simplest case, which is equivalent to the Hanle effect in gases, the decrease of ρ with increasing $\mathbf{H}$ is described by the factor $[1+(\gamma_e TH)^2]^{-1}$, where γ_e is the gyromagnetic ratio for the electron and T is the lifetime of the spin orientation. Obviously, this effect is even with respect to a reversal of the magnetic field.

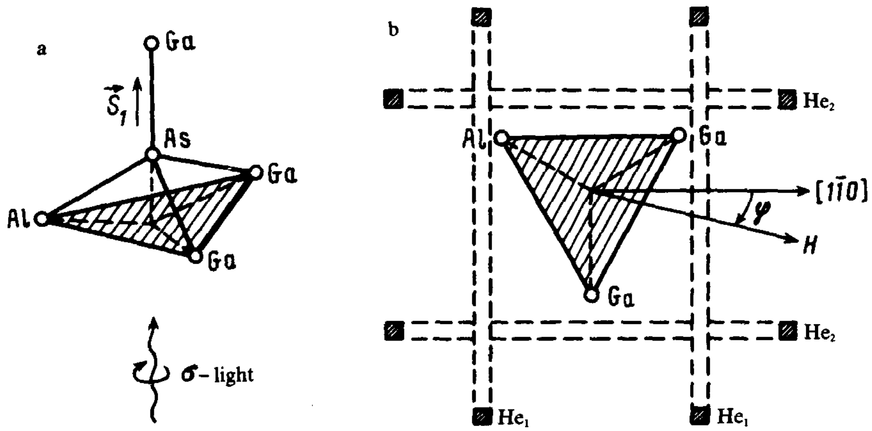

FIG. 1. Scheme of the experiment.

The real situation in semiconductors is more complicated, owing to the cooling of the nuclear spin system in the field of the optically oriented electrons and to the polarization of the nuclei in the field **H**.[1-3] The result is a hyperfine field $\mathbf{H}_N$ of the nuclei and the precession of the oriented electrons takes place in the combined field $\mathbf{H}+\mathbf{H}_N=\mathbf{h}$.

Local violation of cubic symmetry when some of the Ga atoms of the initial GaAs crystal are replaced by Al leads to anisotropy of the hyperfine field in $Al_xGa_{1-x}As$ crystals and to the appearance of nuclear quadrupole effects in the optical channel.[4-6]

We report below the results of an investigation of the anistropy and inversion effects in the case of optical pumping of an *n*-type $Al_{0.24}Ga_{0.76}As$ crystal. Figure 1 shows the scheme of the experiment. Circularly polarized light from an He-Ne laser is directed normal to the surface of the crystal along the $\langle 111 \rangle$ axis [Fig. 1(a)]. The value of ρ is measured for the recombination radiation in a direction close to this axis. The measurements are performed by the methods of two-channel counting of "left" and "right" protons. The field **H** is rotated with the aid of two pairs of Helmholtz coils (He_1 and He_2) in a plane perpendicular to the laser beam [Fig. 1(b)]. The direction of the vector **H** is determined by the angle ϕ, which is reckoned from the $\langle 1\bar{1}0 \rangle$. The earth's field is cancelled out. The figure shows the nearest neighbors of the As nucleus in the case when one of the Ga atoms is replaced by Al.

Figures 2(a) and 2(b) show the experimental plots of $\rho(\phi)$ for a field $H=2$ Oe. The measurements were made at 4.2 K. The asymmetry parameter $\eta(\mathbf{H})=[\rho(\mathbf{H})-\rho(-\mathbf{H})]/[\rho(\mathbf{H})+\rho(-\mathbf{H})]$ reaches a maximum $\eta(\mathbf{H}_M)\approx 15\%$ at three values of the angle ϕ that depend on the sign of σ.

If the time reversal operation is identified with the operation of simultaneous reversal of the signs of σ and **H**, then one should expect the pictures shown in Fig. 2(a) (σ^+ polarization) and 2(b) (σ^- polarization) to differ by a rotation through 180°. Experiment shows, however, that the effect of optical orientation is invariant with

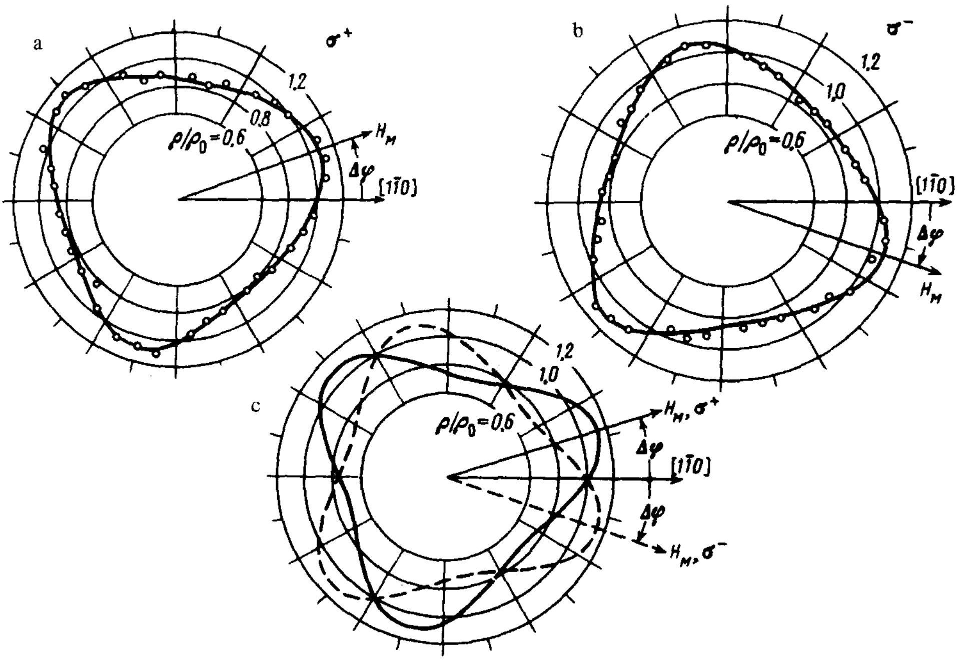

FIG. 2. Plot of $\rho(\phi)$ in polar coordinates [$\rho_0=\rho(\phi)$ at $\phi=0$]: a—experiment, excitation with σ^+ light; b—experiment, excitation with σ^- light; c—numerical computer calculation; the solid and dashed curves correspond to polaizations σ^+ and σ^-, respectively.

respect to simultaneous reversal of the signs of σ and **H** and of rotation of the crystal through an angle $2\Delta\phi+\pi=35+40^\circ+180^\circ$.

This "violation" of T-invariance is due to the essentially nonequilibrium character of the effect of optical orientation (recombination and spin relaxation make the orientation process irreversible in time).

In fact, from Bloch's equations for the average spin of electrons oriented in the presence of a magnetic field we have

$$S = \frac{T}{\tau} S_1 + (\gamma_e T)[S \times \mathbf{h}], \qquad (1)$$

where $\mathbf{S}_1$ is the average spin of the electron at the instant of its production, τ is the electron lifetime in the conduction band, and $\mathbf{h}=\mathbf{H}_N+\mathbf{H}$ is the effective magnetic field acting on the electron spin, and is the sum of the external field and the hyperfine field. The solution of Eq. (1) is of the form

$$S = \frac{T}{\tau}\,\frac{S_1 + (\gamma_e T)^2(S_1\mathbf{h})\,\mathbf{h} + (\gamma_e T)[S_1 \times \mathbf{h}]}{1+(\gamma_e T h)^2}\,. \qquad (2)$$

Whereas the first two terms in the numerator of (2) reverse sign when the signs of $\mathbf{S}_1$ and $\mathbf{h}$ are simultaneously reversed, the third term is even with respect to this operation, and this reflects the essential role of the irreversible dissipative processes in optical orientation.

We emphasize, however, that in the described experiment, as in most experiments on optical orientation in semiconductors, the quantity registered was the projection of the spin $\mathbf{S}$ on the pump direction $\mathbf{S}_1/S_1$

$$\rho = \frac{(\mathbf{S}\mathbf{S}_1)}{S_1} = \frac{T[S_1^2 + (\gamma_e T)^2(\mathbf{S}_1 h)^2]}{\tau S_1[1 + (\gamma_e T h)^2]} \tag{3}$$

which is directly independent of the value of the term $(\gamma_e T)[\mathbf{S}_1 \times \mathbf{h}]$.

The experimental manifestation of the considered effect is connected with the cubic (and not spherical) symmetry of the sample material. Thus, if we supplement the well-known expression for the nuclear-field components $H_{Ni} = h_N(\mathbf{S}\cdot\mathbf{h}')h'_i/(h'^2 + h_L^2)$[1] with the corresponding cubic invariant

$$H_{Ni} = h_N \frac{(\mathbf{S}\mathbf{h}')h'_i + b\, S_i\, h_i'^2}{h'^2 + h_L^2}, \tag{4}$$

then simultaneous solution of Eqs. (2) and (4) leads to the appearance of terms with different time parities in the dependence of ρ on $\mathbf{S}_1$ and $\mathbf{H}$. Just as in experiment, ϕ remains unchanged in this case if, simultaneously with the inversion of $\mathbf{S}_1$ and $\mathbf{H}$, the angle ϕ is changed by a fixed amount. In (4), h_N is the nuclear saturation field, h_L^2 is the square of the modulus of the local field, and $\mathbf{h}' = \mathbf{H} + h_e\mathbf{S}$ is the effective field acting on the nuclei and includes the field $h_e\mathbf{S}$ of the oriented electrons.

The proof that the anisotropy of $\mathbf{H}_N$ can be taken into account in the form (4) is beyond the scope of this brief communication. The coefficient b can, is particular, be determined by the quadrupole interaction, which manifests itself in a predominant alignment of the moments of the As nuclei along the four equivalent threefold axes.[4-6] In this particular case it is possible to obtain an expression practically analogous with (4) by introducing for these nuclei an anisotropic g factor ($g_{\parallel} \neq g_{\perp}$, where $g_{\parallel}$ and $g_{\perp}$ are the values of the g factor in directions parallel and perpendicular to the threefold axis corresponding to the given As nucleus). Figure 2(c) shows the result of a calculation performed for this case with the aid of a computer, for the following values of the parameters: $H = 2$ Oe, $h_N = -5$ kOe, $\gamma_e T = 0.4$ Oe^{-1}, $h_L = 0$, $g_{\perp}/g_{\parallel} = 0.8$, $h_e = 2.5$ Oe.

We note that the good agreement between the data of Figs. 2(a), 2(b), and 2(c) pertains only to the angular dependences. The absolute values of the calculated and the experimental values of ρ differ noticeably in this case. The possibility of an exact determination of the entire set of parameters by comparing the results of the experiment with the performed calculation requires therefore an additional analysis. However, the good agreement of the angular dependences demonstrate that allowance for dissipation of the angular momentum and for the anisotropy of the hyperfine field can explain fully all the observed inversion effects.

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