

Accordingly, we must replace  $S_n(k, \Omega)$  in (13) by  $S_{n-2}(k, \Omega)$ , and use  $\omega_q$  in place of  $\omega_{\alpha, \beta}(q)$ . The  $\omega_q$  spectrum is not symmetrical about  $q=0$  (the point  $2p_F$ ). In place of the resonance points  $\pm q_n/2$  we have the points  $-k_n$  and  $q_n - k_n$ , so that

$$\omega_{-k_n} = \omega_{q_n - k_n}.$$

Outside the term repulsion region  $q \approx q_0$  we can put

$$\omega_q \approx \begin{cases} \omega_\beta(q) & q < 0 \\ \omega_\alpha(q) & q > 0 \end{cases},$$

$|q| \gg \xi_0^{-1}$ , so that the transitions are between the  $\alpha$  and  $\beta$  terms. In place of (14) we get at  $\bar{\omega}\tau_{n-2} \gg 1$

$$\begin{aligned} \gamma_V(\omega) &= \pi^2 (n-2)^2 C_n^2 \left(\frac{m}{m^*}\right)^3 \frac{\Delta^4}{T} \frac{v_F}{s(q_n - k_n) - s(-k_n)} \\ &\times \frac{1}{\omega^2 (-k_n) \text{sh}^2[\omega(-k_n)/T]}. \end{aligned} \quad (\text{A.6})$$

Formulas (14) and (A.6) give relations that agree qualitatively. At a temperature  $T > \bar{\omega}$  we must consider the case  $\bar{\omega}\tau_{n-2} < 1$ , i.e.,

$$\bar{\omega} < \pi(n-2)^2 (u/v_F) T.$$

In accord with the discussion at the end of Sec. 3 we obtain in place of (A.6)

$$\gamma_V \approx \pi^2 (n-2)^2 C_n^2 \left(\frac{m}{m^*}\right)^3 \left(\frac{\Delta}{\bar{\omega}}\right)^4 T \tau_{n-2} v_F \bar{Q}, \quad (\text{A.7})$$

where

$$\bar{Q} = \bar{\omega}^2 \int \frac{dq}{\omega_q^4} \sim Q,$$

$Q$  is the reciprocal-lattice vector. In (A.7) we must take

into account all values of  $n$ , and the optimal is  $n=3$ . We obtain the following estimate:

$$\gamma_V \approx 10^{-4} v_F Q \approx (1-10) \text{ cm}^{-1} \quad (\text{A.8})$$

independently of the temperature. Activation of the intramolecular oscillations can result in an increase of (A.8) with increasing temperature.

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## Optical-orientation anisotropy produced in semiconductors by quadrupole splitting of the spin levels of the lattice nuclei

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The onset of crystal anisotropy of the magnetic depolarization of recombination radiation under optical-orientation conditions is considered theoretically. The influence of the anisotropy of the nuclear field on the behavior of the average spin of the excited electrons is analyzed on the basis of general considerations. The concrete model chosen to describe this anisotropy is quadrupole splitting of the nuclear spin levels. Calculations for an external magnetic field much stronger than the local field produced at the nucleus by the neighboring nuclei agree with the experimental data in the corresponding region of magnetic-field values.

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It was recently observed that the crystal anisotropy exerts a substantial influence on the optical orientation of electrons in semiconductors of the gallium-arsenide type.<sup>1-5</sup> The shape of the plot of the magnetic depolarization of the recombination radiation (the Hanle curve) turned out to be strongly dependent on the orientation of the external magnetic field relative to the crystal axes. The crystal anisotropy manifests itself particularly strongly in the hysteresis observed by Novikov and Fleisher<sup>1,4</sup> when the exciting-light beam is directed

along the [001] axis and the magnetic field is located in the (001) plane of the crystal. In this geometry, the hysteresis exists in a narrow region of angles near a field direction along the [110] axis.

The anisotropy of the Hanle curve was interpreted as a manifestation of the quadrupole splitting of the spin levels of the lattice nuclei, which influences the dynamic polarization of the nuclei by oriented electrons. It is known that dynamic polarization of the nuclei leads to

the appearance of an effective magnetic field, which in turn influences the degree of orientation of the electrons and by the same token the polarization of the recombination radiation.<sup>6,7</sup> This interpretation is directly confirmed by optical detection of nuclear magnetic resonance,<sup>2-4</sup> which has shown that the arsenic nuclei in the solid solution Ga<sub>x</sub>Al<sub>1-x</sub>As undergo a strong quadrupole interaction whose symmetry axis is directed along one of the {111} axes of the crystal. It was suggested<sup>1-5</sup> that a possible cause of the quadrupole splitting of the levels is a local cubic-symmetry distortion due to partial replacement of the gallium atoms in the GaAs lattice by aluminum.

We calculate in this paper the dynamic polarization of the lattice nuclei by oriented electrons in the presence of quadrupole splitting of the spin levels of the nuclei, and consider also the shape of the Hanle curve under these conditions. In Sec. 1 we analyze the influence of the nuclear-field anisotropy on the shape of the Hanle curve on the basis of general symmetry considerations. In Secs. 2 and 3 we consider the influence of quadrupole interaction on the dynamic polarization of the nuclei. In Sec. 4 we present the results of numerical calculation of the shape of the Hanle curve, with account taken of the quadrupole interaction and of the "electron field." The results of the calculations in the corresponding region of magnetic-field values agree with the experimental data.<sup>1,4</sup>

## 1. ANISOTROPY OF THE NUCLEAR FIELD AND THE HANLE EFFECT

The polarization of nuclei under conditions of optical orientation depends, generally speaking, on the orientation of the average spin of the electrons and of the external magnetic field relative to the crystal axes. From symmetry considerations we can write down the following general expression for the nuclear spin acting on the electron spin:

$$H_{N\alpha} = \sum_{\beta} a_{\alpha\beta} S_{\beta}, \quad (1)$$

where  $a_{\alpha\beta}$  is a tensor that depends on the magnitude and direction of the external magnetic field  $\mathbf{H}$  and satisfies the necessary symmetry requirements.

We assume hereafter also that this tensor is invariant to time reversal, an assumption valid for the model considered below. Then all the  $a_{\alpha\beta}$  components are even functions of the magnetic field. For a cubic crystal, in the coordinate frame of the principal crystallographic axes, the tensor  $a_{\alpha\beta}$  satisfies relations of the type

$$\begin{aligned} a_{xx}(H_x, H_y, H_z) &= a_{xx}(H_x, H_x, H_y), \\ a_{xx}(H_x, H_y, H_z) &= a_{yy}(H_y, H_x, H_z), \end{aligned}$$

$$a_{xy}(H_x, H_y, H_z) = a_{yx}(H_y, H_x, H_z), \quad a_{yz}(H_x, H_y, H_z) = a_{zx}(H_x, H_z, H_y). \quad (2)$$

The diagonal components are even here to the change of sign of any field projection, while for the off-diagonal components we have

$$\begin{aligned} a_{xx}(H_x, H_y, H_z) &= -a_{xx}(-H_x, H_y, H_z) = -a_{xx}(H_x, H_y, -H_z) \\ &= a_{xx}(H_x, -H_y, H_z), \end{aligned} \quad (3)$$

etc.

We consider the influence of the effective magnetic field of the polarized nuclei on the Hanle effect in the case when the excited beam is directed along the [001] axis and the external magnetic field lies in the (001) plane. In this case we have for the nuclear field

$$H_{Nx} = a_{xx}S_x + a_{xy}S_y, \quad H_{Ny} = a_{yx}S_x + a_{yy}S_y, \quad H_{Nz} = a_{zz}S_z. \quad (4)$$

At  $H_z$  the components  $a_{xx}$ ,  $a_{yy}$ ,  $a_{zz}$ , and  $a_{zz}$  are equal to zero by virtue of relations (3).

The stationary value of the average spin  $\mathbf{S}$  of the electrons under optical orientation conditions is given by

$$\frac{\mu_0 g}{\hbar} [(H + H_N) \times \mathbf{S}] = \frac{\mathbf{S} - \mathbf{S}_0}{\tau}, \quad (5)$$

where  $\mu_0$  is the Bohr magneton,  $g$  is the  $g$ -factor of the electron,  $\mathbf{S}_0$  is the average electron spin produced by the light in the absence of a magnetic field (in our case  $\mathbf{S}_0$  is directed along the  $z$  axis,  $S_{0z} = S_0$ ), and  $\tau$  is the characteristic time of orientation loss and is determined by the recombination and spin relaxation. Besides the external magnetic field  $\mathbf{H}$ , Eq. (5) takes into account also the nuclear field  $\mathbf{H}_N$ , which depends in turn on  $\mathbf{S}$  in accordance with (4).

From the system (4) and (5) we obtain expressions for  $S_x$  and  $S_y$  in terms of  $S_z$ :

$$\begin{aligned} s_x &= \frac{h_y + (\alpha_{xy}h_y - \alpha_{yy}h_x)s_z}{1 + \varepsilon s_z - \alpha s_z^2} \omega s_z, \\ s_y &= \frac{-h_x + (\alpha_{yx}h_x - \alpha_{xx}h_y)s_z}{1 + \varepsilon s_z - \alpha s_z^2} \omega s_z, \end{aligned} \quad (6)$$

and an equation for  $s_z$ :

$$s_z = \left[ 1 + \omega^2 \frac{1 + 2\delta s_z + \gamma^2 s_z^2}{(1 + \varepsilon s_z - \alpha s_z^2)^2} \right]^{-1}. \quad (7)$$

In formulas (6) and (7) we have introduced the notation

$$\begin{aligned} s &= \mathbf{S}/S_0, \quad \alpha_{xx} = (a_{xx} - a_{zz})S_0/\mathcal{H}, \quad \alpha_{yy} = (a_{yy} - a_{zz})S_0/\mathcal{H}, \\ \alpha_{xy} &= a_{xy}S_0/\mathcal{H}, \quad \alpha_{yx} = a_{yx}S_0/\mathcal{H}, \quad \omega = H/\mathcal{H}, \end{aligned}$$

where  $\mathcal{H} = \hbar(\mu_0 g \tau)^{-1}$  is the characteristic magnetic field corresponding to the half-width of the usual Hanle curve,  $\mathbf{h}$  is a unit vector in the direction of the external field  $\mathbf{H}$ ,

$$\begin{aligned} \alpha &= \alpha_{xy}\alpha_{yx} - \alpha_{xx}\alpha_{yy}, \quad \varepsilon = \alpha_{xy} - \alpha_{yx}, \\ \gamma^2 &= (h_y\alpha_{xy} - h_x\alpha_{yx})^2 + (h_y\alpha_{xx} - h_x\alpha_{yy})^2, \\ \delta &= h_y(h_y\alpha_{xy} - h_x\alpha_{yx}) + h_x(h_y\alpha_{xx} - h_x\alpha_{yy}). \end{aligned} \quad (8)$$

The experimentally determined degree of circular polarization of the luminescence is proportional to  $S_x$ , and its dependence on the magnetic field is determined by Eq. (7). The coefficients  $\alpha_{ik}$  depend on the direction of the external magnetic field. Generally speaking, these coefficients, meaning also  $\alpha$ ,  $\varepsilon$ ,  $\gamma$ , and  $\delta$ , can depend also on  $H$ . In weak-weak magnetic fields this dependence can be attributed to the local field of nuclear spins, and in strong fields to the  $H$ -dependence of the spin-lattice relaxation of the nuclei and electrons or of the rate of dynamic polarization of the nuclei by the electrons. One can expect, however, the existence of a sufficiently large field interval in which this dependence is practically nonexistent. In this interval, the dependence of  $s_x$  on  $H$  (i.e., on  $\omega$ ) can be obtained directly from (7), by solving (7) with respect to  $\omega$ :

$$\omega^2 = \frac{1 - s_z}{s_z} \frac{(1 + \varepsilon s_z - \alpha s_z^2)^2}{(1 + 2\delta s_z + \gamma^2 s_z^2)}. \quad (9)$$

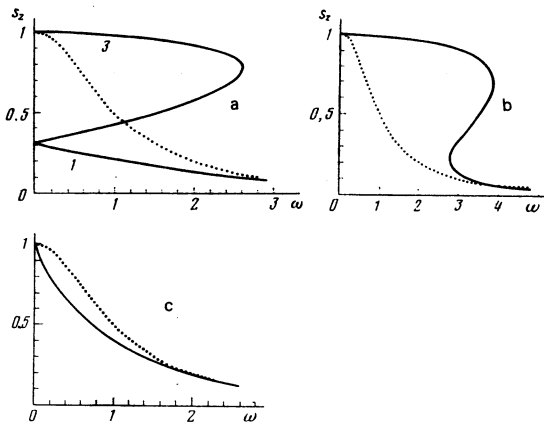


FIG. 1. Shape of Hanle curve at various values of the parameters:  $\varepsilon = \delta = 0$ , a)  $\alpha = 10$ ,  $\gamma^2 = 0.2$ ; b)  $\alpha = 10$ ,  $\gamma^2 = 0$ ; c)  $\alpha = 1$ ,  $\gamma^2 = 0$ . The dotted curves show the usual Hanle curve  $\alpha = \gamma = \varepsilon = \delta = 0$ ,  $\omega = H/\mathcal{H}$  is the magnetic field measured in units of the half-width of the Hanle curve.

We consider now the dependence of the shape of the Hanle curve  $s_x(\omega)$  on the direction of the magnetic field relative to the crystallographic axes. If the field  $\mathbf{H}$  is directed along the [100] axis ( $h_y = h_z = 0$ ), then it follows from the symmetry properties (2) and (3) that  $\alpha_{yy} = \alpha_{xx} = \alpha_{yx} = 0$ , so that in (9) we have  $\alpha = \varepsilon = \delta = \gamma = 0$ . Thus, formula (9) yields in this case the usual Hanle curve  $s_x = (1 + \omega^2)^{-1}$ . The reason is that the nuclear field is directed along the average spin of the electron and therefore does not come into play.

Let now the field  $\mathbf{H}$  be directed along the [110] axis ( $h_x = h_y = 2^{-1/2}$ ). Then  $\alpha_{xy} = \alpha_{yx}$  and  $\alpha_{xx} = \alpha_{yy}$ , so that  $\varepsilon = \delta = 0$ . In this case the function  $s_x(\omega)$  is complicated (see Fig. 1). The most interesting circumstance is the possible ambiguous dependence of the degree of polarization on the magnetic field (hysteresis). At  $\alpha > 1$  the inhomogeneity exists already in a zero magnetic field,<sup>1)</sup> when Eq. (9) leads, besides the solution  $s_x = 1$ , to a solution  $s_x = \alpha^{-1/2}$ . Hysteresis appears also at sufficiently large absolute values of the negative  $\alpha$  (see Fig. 2).

In the three stationary states in the hysteresis region the magnitude and direction of the nuclear field are substantially different. In particular, in the case  $\alpha > 1$  (Fig. 1a) the nuclear-field component  $\mathbf{A} = \mathbf{H}_N - a_{zz}\mathbf{S}$  perpendicular to the average electron spin has a positive projection on the direction of the external field  $\mathbf{H}$  and therefore enhances the action of the latter. On the line 3, the projection of the vector  $\mathbf{A}$  on the direction of  $\mathbf{H}$  is negative and exceeds  $\mathbf{H}$ .

At an arbitrary direction of  $\mathbf{H}$  in the (001) plane, other than the symmetrical direction, the Hanle effect is described by the general expression (9). Since the signs of the coefficients  $\varepsilon$  and  $\delta$  in this expression depend on the sign of  $S_0$  the shape of the Hanle curve turns out to be sensitive to reversal of the sign of the circular polarization of the exciting light. From expressions (8) and from the symmetry properties (2) and (3) it follows that the reversal of the sign of the circular polarization is equivalent to a reflection of the direction of the magnetic field relative to the [110] or [100] axis.

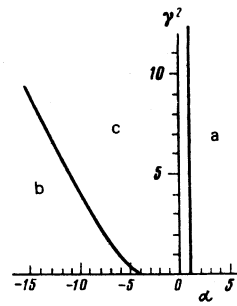


FIG. 2. Ranges of the parameters  $\alpha$  and  $\gamma^2$ , corresponding to different shapes of the Hanle curves. Regions a and b correspond to Hanle curves with hysteresis (Figs. 1a and 1b, respectively), and region c corresponds to curves without hysteresis (Fig. 1c).

## 2. EFFECT OF QUADRUPOLE INTERACTION ON THE DYNAMIC POLARIZATION OF NUCLEI BY ELECTRONS

We consider a nucleus with spin  $\frac{3}{2}$ , with a quadrupole moment, and situated in an inhomogeneous electric field, and assume that the quadrupole interaction has a symmetry axis. We designate the unit vector along this axis by  $\mathbf{n}$ . Assume, in addition, that a weak magnetic field  $\mathbf{H}$  is present, such that the Zeeman splitting is much less than the quadrupole splitting. In this case the energy levels of the nuclei and the corresponding wave functions are given by<sup>6</sup>

$$\begin{aligned} E_{\pm m} &= \Delta \pm \frac{3}{2} \hbar \Omega \cos \theta, & \Psi_{\pm m} &= \Phi_{\pm m}, \\ E_{\pm} &= \pm \frac{1}{2} \hbar \Omega (\cos^2 \theta + 4 \sin^2 \theta)^{1/2}, \\ \Psi_{+} &= \Phi_{+} \cos \chi + \Phi_{-} \sin \chi, & \Psi_{-} &= -\Phi_{+} \sin \chi + \Phi_{-} \cos \chi. \end{aligned} \quad (10)$$

Here  $\Delta$  is the energy of the quadrupole splitting,  $\hbar \Omega = (\frac{3}{2}) \mu_I H$ ,  $\mu_I$  is the magnetic moment of the nucleus,  $\theta$  is the angle between the direction of the magnetic field  $\mathbf{H}$  and the vector  $\mathbf{n}$ ,  $\Phi_m$  are the wave functions describing a state with a spin projection  $m$  on the  $\mathbf{n}$  direction, and  $\tan(2\chi) = 2 \tan \theta$ .

Using formulas (10) for the wave functions, we can obtain the following expression for the average nuclear spin in terms of the level populations.

$$\mathbf{I} = \frac{3}{2} \mathbf{n} p + \lambda \left[ 2\mathbf{h} - \frac{3}{2} \mathbf{n} (\mathbf{h} \cdot \mathbf{n}) \right] q, \quad \lambda = [4 - 3(\mathbf{h} \cdot \mathbf{n})^2]^{-1/2}, \quad (11)$$

where  $\mathbf{h}$  is a unit vector along  $\mathbf{H}$ ,  $p$  is the difference between the populations of the levels  $+\frac{3}{2}$  and  $-\frac{3}{2}$ , and  $q$  is the difference between the populations of the levels  $+$  and  $-$ . It is seen that the vector  $\mathbf{I}$  lies in the plane defined by the vectors  $\mathbf{n}$  and  $\mathbf{h}$ .

We consider now the dynamic polarization of the nuclei by oriented electrons. We assume a contact interaction of the nuclear and electron spins. Then, calculating in the usual manner the probabilities of the transitions between the spin levels of the nucleus, we obtain

$$\begin{aligned} W\left(\pm; \mp \frac{3}{2}\right) &= W\left(\pm \frac{3}{2}; \mp\right) = \frac{3}{2T_{1e}} \sin^2 \chi [1 \pm 2S_n], \\ W\left(\pm; \pm \frac{3}{2}\right) &= W\left(\mp \frac{3}{2}; \mp\right) = \frac{3}{2T_{1e}} \cos^2 \chi [1 \mp 2S_n], \\ W(\pm; \mp) &= \frac{3}{2T_{1e}} \frac{1}{6} \{8 - 3 \sin^2 2\chi \pm 16(S_n) \cos 2\chi \pm 8\lambda [S_h - (S_n)(\mathbf{h} \cdot \mathbf{n})]\} \end{aligned} \quad (12)$$

Here  $T_{1e}$  is the time of relaxation of the nuclear spin on the electrons in the absence of the quadrupole splitting.

Using formulas (2) and assuming the population differences  $p$  and  $q$  to be small, we obtain for these quantities the following system of equations:

$$\begin{aligned} \frac{dp}{dt} &= \frac{3}{2T_{1e}} [q \cos 2\chi - p + 2S_n] - \frac{1}{T_1} p, \\ \frac{dq}{dt} &= \frac{3}{2T_{1e}} \left\{ p \cos 2\chi + \left[ \sin^2 2\chi - \frac{41}{3} \right] q \right. \\ &\left. + \frac{2}{3} S_n \cos 2\chi + \frac{4}{3} \lambda [S_h - (S_n)(h_n)] \right\} - \frac{1}{T_1} q. \end{aligned} \quad (13)$$

We have added to the right-hand sides of these equations terms that describe spin-lattice relaxation not due to the electrons (leakage). The time  $T_1$  characterizes this relaxation. The relaxation terms that describe the leakage are expressed in the simplest form, which is valid, for example for isotropic quadrupole relaxation.<sup>2)</sup>

We assume henceforth that  $T_{1e} \gg T_1$  (large leakage). Under these conditions the stationary solutions of Eqs. (13) take the form

$$p = 3fS_n, \quad q = f\lambda [-(S_n)(h_n) + 2S_h], \quad (14)$$

where  $f = T_1/T_{1e}$  is the leakage factor.

Formulas (11) and (14) yield the solution of the problem of dynamic polarization of the nuclei when account is taken of the quadrupole splitting in the case of strong leakage.

In a cubic crystal, the quadrupole splitting can be due to local symmetry violation due to impurities, defects, etc. In the solid solutions  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  ( $x \approx 0.25$ ), in which quadrupole-interaction effects were observed,<sup>1-5</sup> the gradients of the local electric fields at the As nuclei may be due to replacement of one or several of the nearest gallium atoms by aluminum atoms. If only one of the four nearest neighbors is replaced by aluminum, then the quadrupole-interaction axis is one of the principal diagonals of the unit cube. Bearing this situation in mind, we consider a model with four types of nuclei, for each of which the quadrupole-interaction is one of the four threefold axes  $\{111\}$ . Then the average spin of these nuclei is

$$\mathbf{I} = \frac{1}{4} \sum_{i=1}^4 \left\{ \frac{3}{2} n_i p_i + \lambda_i \left[ 2h - \frac{3}{2} n_i(n_i h) \right] q_i \right\}, \quad (15)$$

where  $\mathbf{n}_i$  is a unit vector along the  $i$ -th threefold axis, while  $p_i$  and  $q_i$  are expressed in terms of  $\mathbf{n}_i$  by formulas (14).

The expression for the nuclear field (which is proportional to the vector  $\mathbf{I}$ ) can be written in the form (1), with

$$\begin{aligned} a_{xx} &= \frac{1}{12} H_N^{(1)} f (12 + 3d_{xx} + 8\lambda h_x^2 - 10b_{xx}), \\ a_{xy} &= \frac{1}{12} H_N^{(1)} f (3d_{xy} + 8\lambda h_x h_y - 4b_{yx} - 6b_{xy}), \end{aligned} \quad (16)$$

where

$$\Lambda = \sum_{i=1}^4 \lambda_i^2$$

is a cubic invariant that depends on the direction of the magnetic field,  $d_{\alpha\beta}$  and  $b_{\alpha\beta}$  are defined by the formulas

$$d_{\alpha\beta} = \sum_{i=1}^4 \lambda_i^2 (h_{ni})^2 n_{i\alpha} n_{i\beta}, \quad b_{\alpha\beta} = \sum_{i=1}^4 \lambda_i^2 (h_{ni}) n_{i\alpha} h_{i\beta}, \quad (17)$$

and  $H_N^{(1)}$  is the nuclear field corresponding to the case

when all the considered nuclei are fully polarized in one direction. The expressions for the other components of the tensor  $a_{\alpha\beta}$  are similar to (16).

If the external magnetic field lies in the (001) plane, then

$$d_{xx} = \frac{1}{27} \Lambda (3 + 4h_x^2 h_y^2), \quad d_{xy} = \frac{8}{27} \Lambda (h_x h_y), \quad b_{\alpha\beta} = \frac{1}{9} \Lambda h_{\alpha} h_{\beta} (5 - 2h_{\alpha}^2),$$

and the invariant  $\Lambda$  is expressed in terms of the angle  $\varphi$  between the direction  $\mathbf{h}$  and the  $x$  axis as follows:

$$\Lambda = \frac{24}{17 + \cos 4\varphi}. \quad (18)$$

The expressions for the parameters that determine in accord with formulas (9) the shape of the Hanle curve are

$$\alpha = \xi_i^2 \Lambda^2 6(7 - \sin^2 2\varphi) \sin^2 2\varphi, \quad \gamma^2 = \xi_i^2 \Lambda^2 (1 + 3 \sin^2 2\varphi) \sin^2 2\varphi, \quad (19)$$

$$e = \delta = \frac{1}{2} \xi_i \Lambda \sin 4\varphi, \quad \xi_i = H_N^{(1)} f_i S_i / 54 \mathcal{H}. \quad (20)$$

We note that these expressions were obtained under the assumption that the field  $\mathbf{H}_N$  is due entirely to nuclei with large quadrupole splitting. It can be assumed that in a real situation there exist also a large number of nuclei for which the quadrupole splitting is small. (In the case of solid solutions these are the nuclei As, which are not nearest neighbors of Al, as well as nuclei of Ga and of Al itself.) The contribution from these nuclei to the field  $\mathbf{H}_N$  is described by the usual expression  $\mathbf{H}'_N = H_N^{(2)} f_2 (\mathbf{S} \cdot \mathbf{h}) \mathbf{h}$ . Allowance for this field merely increases the parameter  $\alpha$  by the amount

$$\Delta\alpha = 2\xi_i \xi_2 \Lambda \sin 2\varphi, \quad (21)$$

where

$$\xi_2 = H_N^{(2)} f_2 S_2 / \mathcal{H}. \quad (22)$$

The distortion of the shape of the Hanle curve by the nuclear field is connected in this model exclusively with the difference of the populations of the levels  $\pm(q_i \neq 0)$ , since the nuclear field connected with the difference of the populations on the levels  $\pm\frac{3}{2}$ , and is always directed along  $\mathbf{S}$ . In fact, according to (14) the sum  $\sum p_i \mathbf{n}_i \sim \mathbf{S}$  makes no contribution to the coefficients (19).

### 3. ANTICROSSING OF SPIN LEVELS OF NUCLEI

The results of the preceding section are generally speaking not valid when the magnetic field is directed along one of the  $[110]$  axes. In fact, if the magnetic field is perpendicular to the quadrupole-interaction axis ( $\vartheta = \pi/2$ ), then the levels  $\pm\frac{3}{2}$  coincide in the zeroth approximation in the parameter  $\mu_0 H / \Delta$  [see formula (10)]. However, when the weak interaction between these levels is taken into account, these levels do not coincide and the so-called level anticrossing takes place (Fig. 3). The dependence of the level splitting on the angle  $\vartheta$  is given by the usual expression

$$\delta E = ((\delta E_0)^2 + 4|V|^2)^{1/2}, \quad \delta E_0 = 3\hbar\Omega \cos \vartheta, \quad (23)$$

where  $V$  is the matrix element of the interaction with the magnetic field between the states  $\pm\frac{3}{2}$ . This element is due to an admixture of  $\pm\frac{1}{2}$  states to the  $\pm\frac{3}{2}$  states. This admixture can be due to the weak inhomogeneity of the quadrupole interaction or to the magnetic field itself in the higher-order approximations in the parameter  $\mu_0 H / \Delta$ . In the former case  $V \propto H$  and in the latter  $V \propto H^2$ .

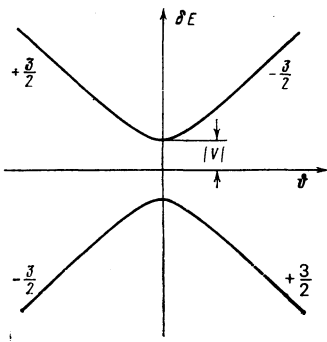


FIG. 3. Dependence of level energy on  $\vartheta$  in the anticrossing region. At sufficiently large angles  $\vartheta$  the wave functions of the stationary states are, with high accuracy, functions of  $\varphi_{+3/2}$  and  $\varphi_{-3/2}$ , and are their superpositions in the anticrossing region.

In the region where  $\delta E_0 \sim V$ , a mixing of the states  $\pm \frac{3}{2}$  and  $-\frac{3}{2}$  takes place, and at  $\delta E_0 = 0$  ( $\vartheta = \pi/2$ ) the correct wave functions are the half-sum and half-difference of the states  $\pm \frac{3}{2}$ . In these stationary states the angular momentum of the nucleus is zero, so that optical orientation of the nuclei is impossible in these states. Thus, in a narrow region of angles near  $\vartheta = \pi/2$  the degree of orientation of the nucleus acquires a singularity. If the magnetic field lies in the (001) plane, such singularities can appear whenever the direction of  $\mathbf{H}$  is close to one of the twofold axes [110]. This excludes a contribution to the nuclear field from the states  $\pm \frac{3}{2}$  for nuclei whose quadrupole interaction axes coincide with the axes  $[1\bar{1}1]$  and  $[\bar{1}11]$ . At the same time the states  $\pm \frac{3}{2}$  of the remaining nuclei with quadrupole axes  $[111]$  and  $[1\bar{1}\bar{1}]$  will produce a nuclear field that influences the electron spin. We recall that in the absence of level crossing the combined field of the nuclei in the  $\pm \frac{3}{2}$  is directed along the average spin of the electrons and therefore does not influence the Hanle effect.

Let us examine in greater detail the dynamic polarization of the nucleus under conditions of level anticrossing. The wave functions  $\psi_1$  and  $\psi_2$  of the stationary states  $\pm \frac{3}{2}$  are

$$\begin{aligned} \psi_1 &= \varphi_{+3/2} \cos \alpha + \varphi_{-3/2} \sin \alpha, \\ \psi_2 &= -\varphi_{+3/2} \sin \alpha + \varphi_{-3/2} \cos \alpha, \end{aligned} \quad (24)$$

$$\cos 2\alpha = \frac{\delta E_0}{((\delta E_0)^2 + 4|V|^2)^{1/2}}.$$

The contribution  $\delta \mathbf{I}$  of these states to the angular momentum is expressed in the following manner in terms of their population difference  $p$ :

$$\delta \mathbf{I} = \frac{1}{2} n \bar{p} \cos 2\alpha, \quad (25)$$

where  $\mathbf{n}$ , as before, is a unit vector along the quadrupole-interaction axis.

A conclusion similar to that made in Sec. 3 leads to the expression  $\bar{p} = 3f\mathbf{S} \cdot \mathbf{n} \cos 2\alpha$ . Far from the anticrossing region, at  $\delta E_0 \gg |V|$ , this formula goes over into the first formula of (14). We then have for  $\delta \mathbf{I}$

$$\delta \mathbf{I} = \frac{9}{2} f (\mathbf{S} \mathbf{n}) \frac{(\delta E_0)^2}{(\delta E_0)^2 + 4|V|^2}. \quad (26)$$

From (26) and from expression (23) for  $\delta E_0$  we see that

$\delta \mathbf{I}$  vanishes at  $\vartheta = \pi/2$ .

The level anticrossing causes the coefficients  $a_{ik}$  given by (16) to acquire additional resonant terms that exist at those external magnetic field directions at which the field is perpendicular to one or two quadrupole interaction axes:

$$\delta a_{\alpha\beta} = -\frac{3}{4} H_N^{(1)} f \sum_{i=1}^4 n_{i\alpha} n_{i\beta} \frac{1}{1+b^2(\mathbf{h}\mathbf{n}_i)^2}, \quad (27)$$

where  $b = 3\hbar\Omega/(2|V|)$ .

If the external field is close in direction to [110], then we can put  $\varphi = \pi/4$  in expressions (16)–(22), which vary slowly with change of angle  $\varphi$ . When the increment (27) is taken into account, we obtain the following expressions for the parameters  $\alpha$  and  $\gamma^2$ , which determine the shape of the Hanle curve near the direction  $\varphi = \pi/4$ :

$$\alpha = 3\xi_1(1+9R)[27\xi_1(1+R)+\xi_2], \quad \gamma^2 = \xi_1^2 \Lambda^2 [1+(9R)^2]. \quad (28)$$

Here  $R = [1+2b^2(\varphi - \pi/4)^2/3]^{-1}$ . The parameters  $\varepsilon$  and  $\delta$  are equal to zero at the assumed accuracy. At  $b \gg 1$  the coefficients  $\alpha$  and  $\gamma$  increase sharply in a small region of the angles  $|\varphi - \pi/4| \sim 1/b$ . In this region, the parameter  $\alpha$  increases by one order of magnitude. (We recall the hysteresis of the Hanle curve at  $\alpha > 1$ .)

The solid curve of Fig. 1a was constructed with the aid of formulas (9) and (28) at parameter values  $(\frac{3}{2})\xi_1 = 0.025$  and  $\xi_2 = 20$  for a field directed along [110] (in this case  $\alpha \approx 10$  and  $\gamma^2 \approx 0.2$ ). These parameter values correspond, for example, to the conditions  $\mathcal{H} = 50$  Oe,  $S_0 = 0.05$ ,  $H_N^{(1)} f \approx 900$  Oe, and  $H_N^{(2)} f_2 = 20000$  Oe. If the level anticrossing is not taken into account, the same values of the parameters  $\xi_1$  and  $\xi_2$  correspond to  $\alpha = 1$  and  $\gamma^2 \approx 0$ . The corresponding curve is shown in Fig. 1c.

Experiments<sup>1,4</sup> have revealed hysteresis phenomena in a narrow region near  $\varphi = \pi/4$ . It can be assumed that these phenomena are connected with the considered anticrossing of the levels  $\pm \frac{3}{2}$ . We note that the anticrossing effect is due to the same interaction  $V$  as the experimentally observed<sup>2,3</sup> nuclear magnetic resonance on the transitions between the levels  $\pm \frac{3}{2}$ . In the absence of this interaction, the transitions between the levels  $\pm \frac{3}{2}$  and  $-\frac{3}{2}$  are forbidden. Anticrossing is in fact "resonance at zero frequency."

#### 4. EFFECT OF ELECTRON FIELD. DISCUSSION OF RESULTS

It is known<sup>10,11</sup> that under optical orientation the polarization of nuclei in semiconductors is greatly influenced by the effective fields produced by the electrons on the nuclei of the lattice (the electron field). Novikov and Fleisher have shown that allowance for the electron field leads to good agreement between the theoretical shape of the Hanle curve and the experimental data<sup>11</sup> (see also Ref. 12). Since they did not take into account the influence of quadrupole effects, their calculation pertains in fact to a magnetic field directed along the  $\{111\}$  axis. (It was shown above that at this field direction no quadrupole effects appear.)

The electron field can be taken into account in our

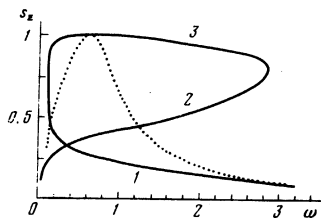


FIG. 4. Hanle-curve shape calculated with allowance for the level anticrossing and for the electric field. Solid curve—external magnetic field directed along the [110] axis, dotted—along the [100] axis. The stable solutions correspond to branches 1 and 2.

model by replacing  $\mathbf{H}$  with  $\mathbf{H} - H_e \mathbf{S}$ , where  $H_e/2$  is the electron field at 100% electron orientation. Even a weak electron field (compared with the external field) leads to substantial deformation of the Hanle curve. This is due to the following circumstance. In the absence of the electron field, as seen from formulas (4), the  $S_x$  component does not lead to the appearance of a nuclear field perpendicular to the pump direction. In the presence of an electron field this is not the case, and a self-consistent situation is possible wherein the electron field is directed practically along the  $z$  axis, and the nuclear-field component located in the  $xy$  plane almost cancels out the action of the external field on the electron spin. This component is of the order of  $a_x S_x \sim H_N f h_x h_z S_x$ . Recognizing that  $h_x \approx H_x/H$  and  $h_z \approx H_e S_0/H$  we find that this cancellation is possible at  $H \sim (H_N f H_e S_0)^{1/2}$ , i.e., when the external magnetic field greatly exceeds the electron field. A similar result was obtained by Novikov and Fleisher.<sup>11</sup>

Figure 4 shows the Hanle curve for two directions of the external magnetic field (along the axes [100] and [110]), calculated with account taken of the electron field and of the level anticrossing. We used in the calculation Eq. (5) and the expression obtained above for the nuclear field, with  $\mathbf{H}$  replaced by  $\mathbf{H} - H_e \mathbf{S}$ . The parameters  $\xi_1$ ,  $\xi_2$ , and  $S_0$  were assumed to be the same as for the curves of Fig. 1, and  $H_e S_0 = 1$  Oe. A comparison of the curves calculated for  $\mathbf{H}$  directed along the [110] axis with allowance for the electron field (solid curve in Fig. 4) and without this field (Fig. 1) shows that the electron field has practically no effect on the shape of the Hanle curve in the region  $\omega \geq 1$ . The curves of Fig. 4 are close in form to the curves obtained in the experiments of Ref. 4, if it is assumed that the branches 1 and 2 are realized in the experiment in the hysteresis region. An analysis of the stability of the solutions existing in this region, carried out with the aid of the time-dependent equations (13), confirms this assumption (the branch 3 turns out to be unstable). We recall that the results of the present paper pertain to the region  $H_L \ll H$  and therefore do not describe the experimentally observed abrupt change of the polarization at  $H < H_L$ .

Figure 5 shows the Hanle curves for two magnetic-

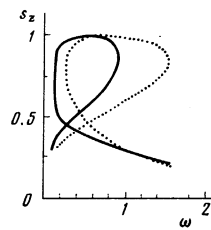


FIG. 5. Shape of Hanle curve in the vicinity of the [110] axis. The solid curve corresponds to the angle  $\varphi = 40^\circ$  and the dotted one to  $\varphi = 50^\circ$ .

field directions making angles  $\pm 5^\circ$  with the [110] direction ( $\varphi = 40^\circ$  and  $50^\circ$ ). In the calculation we used the value  $b = 30$ , and the remaining parameters were the same as for Fig. 4. It is seen that the angular dependence of the polarization is asymmetrical relative to the [110] direction. Reversal of the sign of  $S_0$  reflects the angular diagram relative to the [110] axis and relative to the [100] axis. A similar result was obtained in experiment and was interpreted on the basis of the phenomenological analysis of Merkulov and Fleisher.<sup>5</sup>

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- <sup>1</sup>As noted above, in weak fields of the order of the local field  $H_L$  of the nuclear spins the coefficients  $\alpha$  and  $\gamma$  depend on  $H$ . A zero magnetic field must therefore be taken here to mean a field satisfying the condition  $H_L \ll H \ll \mathcal{H}$ .
- <sup>2</sup>It is known that in III-V compounds at liquid-nitrogen temperatures and higher the nuclear spin-lattice relaxation is due to quadrupole interaction with phonons.<sup>9</sup>
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