

## HYPERFINE INTERACTION IN OPTICAL ORIENTATION OF ELECTRONS IN SEMICONDUCTORS

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Hyperfine interaction between optically oriented electrons captured by impurity centers and nuclei of the centers is considered. It is shown that this interaction decreases the stationary degree of orientation of the electrons and results in polarization of the nuclei. The stationary polarization of the electrons and nuclei is calculated as a function of excitation light intensity and the effect of longitudinal and transverse magnetic fields or of a varying field at nuclear resonance frequency on the polarization is considered.

### 1. INTRODUCTION

THE spin orientation of electrons in the case of interband absorption of circularly polarized light<sup>[1-5]</sup> is an effective means of investigating the energy spectrum and the spin interactions in semiconductors. Even in his first study, devoted to optical orientation in silicon, Lampel<sup>[1]</sup> observed an appreciable polarization of <sup>29</sup>Si nuclei, due to their interaction with the electrons oriented by the light. Ekimov and Safarov<sup>[6,7]</sup> have observed that the resultant polarization of the nuclei exerts in turn an appreciable influence on the electron orientation. The most convincing proof of this influence is the decrease of degree of orientation of electrons under conditions of nuclear resonance on the nuclei Ga, Al and As, observed in the solid solution p-Ga<sub>x</sub>Al<sub>1-x</sub>As<sup>[7]</sup>. The degree of orientation of the electrons was measured by determining the circular polarization of the recombination radiation. Ekimov and Safarov observed<sup>[6]</sup> an abrupt decrease of the electron orientation in a perpendicular magnetic field on the order of several Oe, whereas further depolarization of the electrons requires much stronger fields (the width of the Hanle line is of the order of 100 Oe). The decrease of the electron orientation in a weak transverse field, just as in nuclear resonance, is due to depolarization of the nuclei.

In the solid solutions Ga<sub>x</sub>Al<sub>1-x</sub>As at 4°K, the degree of orientation of the electrons following stationary excitation is of the order of several per cent. It can be assumed that the smallness of the orientation is due to the interaction of the spins of the electrons and the nuclei. If the nuclei are not polarized, they produce a random magnetic field in which the electrons become depolarized. Such an interpretation is confirmed by the strong increase of the electron orientation, observed by Ekimov and Safarov following application of a magnetic field parallel to the orienting beam. In an external magnetic field, the coupling between the spins of the electrons and the nuclei is broken, and the depolarizing action of the nuclei becomes weaker. This may be the reason for a certain increase in the degree of polarization observed by Parsons<sup>[8]</sup> in GaSb in a longitudinal field.

The depolarization of the electrons in the hyperfine

interaction is accompanied by orientation of the nuclei, which in turn decreases their influence on the electron spin. This is precisely the reason for the decrease of the electron orientation when the nuclear spins become disordered (in nuclear resonance or in a weak perpendicular magnetic field). This causes also the increase of the electron orientation observed in<sup>[6]</sup> in a zero field with increasing intensity of the exciting light.

It is clear that the hyperfine interaction is significant only if the electron is localized to one degree or another, for otherwise the field of the nuclei average out. Indeed, at 77°K the electron orientation in the solid solution p-Ga<sub>x</sub>Al<sub>1-x</sub>As is much higher than at 4°K<sup>[3]</sup>. Veshchunov, Dzhiyev, Zakharchenya, and Fleisher<sup>[9]</sup> have found that in this material the degree of orientation is low at temperatures below 30°K, and then increases sharply. This indicates that at low temperatures the recombination radiation proceeds from donor levels. The sharp increase of the polarization is due to thermal ionization of the donors and to the dynamic averaging of the nuclear magnetic field acting on the nonlocalized electron in the conduction band. The appreciable degree of orientation observed in strongly doped GaAs<sup>[10]</sup> is probably due to the fact that the donor states are merged in this case with the conduction band.

We consider in this paper optical orientation under conditions when the hyperfine interaction of the electrons and nuclei is significant. It is assumed that the electron thrown into the conduction band by circularly polarized light is captured by an impurity atom whose nucleus has spin. As a result of the hyperfine interaction, the electron orientation is decreased during the time of stay on the impurity center, and the nucleus becomes polarized. The nuclear-relaxation time is assumed to be large in comparison with the time of stay of the electron at the center. As a result of the competition between the polarizing action of the captured electrons and the nuclear relaxation, which occurs mainly in the intervals between the captures, a certain stationary polarization of the nuclei and of the electrons is established. We calculate this stationary polarization as a function of the intensity of the exciting light and consider the influence exerted on it by longitudinal and transverse magnetic fields and of an alternating field at the nuclear-resonance frequency. The theory contains a

qualitatively correct description of all the experimental results listed above.

It should be noted, however, that the theory presented here is applicable, strictly speaking, only to the situation when the electron localized at the center interacts with only one nucleus. In the experiments of Ekimov and Safarov, the electrons interacted with the nuclei of the main lattice of the crystal. The electron at a shallow donor level is localized in a region that includes a large number of such nuclei. For a quantitative description of the experimental results of [6,7], it is therefore necessary to generalize the theory to include the case when the electron interacts simultaneously with many nuclei. In addition, under the conditions of these experiments, an important role is probably played by jumps of the electron from one center to another, which occur during the lifetime. As shown in the present paper, during the time that the electron stays on one center, the hyperfine interaction decreases the degree of orientation by not more than a factor of three. The additional decrease is apparently due to the jumps.

**2. CHANGE OF SPIN STATE DURING THE TIME THAT THE ELECTRON STAYS AT THE IMPURITY CENTER**

We consider an electron captured by an impurity center at the instant  $t = 0$ . We assume that at this instant the electron is oriented and has an average spin  $S_0$ . The spin direction is parallel to the direction  $n$  of the orienting light beam. The nucleus of the impurity center, with angular momentum  $I$ , has a certain arbitrary spin state. As a result of the hyperfine interaction and the interaction with the external magnetic field, the spin states of both the electron and of the nucleus are altered. We are interested in these states at the instant  $t$  when the electron leaves the center for some reason (for example, by recombining with a hole).

The spin part of the wave function of the electron and nucleus can be written in the form

$$\Psi = \sum_{m\mu} C_{m\mu}(t) \varphi_m \chi_{\mu}, \tag{1}$$

where  $\varphi_m$  describes the state of the electron with spin projection  $m$  on the direction of the external magnetic field ( $m = \pm 1/2$ ), and  $\chi_{\mu}$  describes the state of the nucleus ( $-I \leq \mu \leq I$ ). The dependence of the coefficient  $C_{m\mu}$  on the time can be easily obtained by reexpanding the wave function (1) in terms of states with definite energy and projection  $M$  of the total angular momentum on the direction of the magnetic field. For a given  $M$ , the energy can assume two values,  $E_M^{\pm}$  (with the exception of the case  $M = \pm(I + 1/2)$ , when there is only one value  $E_M^{\pm}$ ); the corresponding wave functions are designated  $\psi_M^{\pm}$  (the  $\pm$  symbols correspond to the total angular momentum  $I \pm 1/2$  in a zero field), and

$$\Psi = \sum_M [a_M^+(t) \psi_M^+ + a_M^-(t) \psi_M^-], \tag{2}$$

where  $a_M^{\pm}(t) = a_M^{\pm}(0) \exp(-iE_M^{\pm}t)$ .

The wave functions  $\psi_M^{\pm}$  and the energy values  $E_M^{\pm}$  are obtained by diagonalizing the Hamiltonian

$$\mathcal{H} = AIS + \mu_0 gHS,$$

where  $A$  is the hyperfine structure constant,  $\mu_0$  is the Bohr magneton,  $g$  is the g-factor, and  $H$  is the magnetic field. The corresponding formulas are [11]

$$E_M^{\pm} = -1/4A \pm 1/2A(I + 1/2)(1 + 4Mx / (2I + 1) + x^2)^{1/2}, \tag{3}$$

$$\begin{aligned} \psi_M^+ &= \alpha_M \varphi_{1/2} \chi_{M-1/2} + \beta_M \varphi_{-1/2} \chi_{M+1/2}, \\ \psi_M^- &= -\beta_M \varphi_{1/2} \chi_{M-1/2} + \alpha_M \varphi_{-1/2} \chi_{M+1/2}, \end{aligned} \tag{4}$$

where

$$x = 2\mu_0 gHA^{-1}(2I + 1)^{-1}, \tag{5}$$

$$\alpha_M = \left[ \frac{2E_M^+ + A(M + 1/2) + \mu_0 gH}{4E_M^+ + A} \right]^{1/2}, \quad \beta_M = (1 - \alpha_M^2)^{1/2}. \tag{6}$$

Substituting (4) in (2) and comparing with (1), we can express the coefficients  $C_{m\mu}(t)$  in terms of  $a_M^{\pm}$  and thus obtain their time dependence:

$$\begin{aligned} C_{1/2, M-1/2}(t) &= e^{iAt/4} [u_M C_{1/2, M-1/2} + v_M C_{-1/2, M+1/2}], \\ C_{-1/2, M+1/2}(t) &= e^{iAt/4} [-v_M C_{1/2, M-1/2} + u_M C_{-1/2, M+1/2}]. \end{aligned} \tag{7}$$

Here  $C_{m\mu}$  are the values of the coefficients  $C_{m\mu}(t)$  at  $t = 0$ , and the quantities  $u_M$  and  $v_M$  are given by

$$\begin{aligned} u_M &= \cos \omega_M t + i(\beta_M^2 - \alpha_M^2) \sin \omega_M t, \\ v_M &= -2i\alpha_M \beta_M \sin \omega_M t; \end{aligned} \tag{8}$$

$$\omega_M = \frac{\omega_h}{2} \left( 1 + \frac{4M}{2I + 1} x + x^2 \right)^{1/2}, \quad \omega_h = \frac{A(I + 1/2)}{\hbar}. \tag{9}$$

We note that the square root in (9), just as in the Breit-Wigner formula (3), should be taken to mean  $1 - x$  (including  $x > 1$ ) at  $M = I - 1/2$  and  $x > 0$ .

The evolution of the spin density matrix  $\rho_{m\mu, m'\mu'}$  of the electron + nucleus system can be easily obtained with the aid of (7):

$$\rho_{m\mu, m'\mu'}(t) = C_{m\mu}(t) C_{m'\mu'}^*(t). \tag{10}$$

It must be borne in mind here that at  $t = 0$ , i.e., at the instant when the electron is captured by the impurity center, the density matrix of the system is a product of the electron density matrix  $f_{mm'}$  and the nuclear density matrix  $\Phi_{\mu\mu'}$ .

**3. ORIENTATIONS OF ELECTRONS AND NUCLEI IN THE ABSENCE OF A MAGNETIC FIELD**

In the absence of a magnetic field, the quantity  $2\omega_M$  does not depend on the index  $M$  and is equal to the total hyperfine splitting  $\omega_h$ . We choose the quantization axis along the initial direction of the electron spin  $S_0$ . The projection  $S_n$  of the electron spin at the instant  $t$  on the initial spin direction can be calculated with the aid of the density matrix (10):

$$S_n(t) = 1/2 \sum_{\mu} (\rho_{1/2\mu, 1/2\mu} - \rho_{-1/2\mu, -1/2\mu}). \tag{11}$$

We use formulas (7) and (10) to express  $S_n(t)$  in terms of the initial spin density matrix  $\Phi_{\mu\mu'}$  of the nucleus:

$$S_n(t) = S_0 + 1/2 \sum_{\mu} |v_{\mu+1/2}|^2 [(\Phi_{\mu+1, \mu+1} - \Phi_{\mu\mu}) - 2S_0(\Phi_{\mu+1, \mu+1} + \Phi_{\mu\mu})]. \tag{12}$$

Using (6) and (8) with  $H = 0$ , we obtain

$$S_n(t) = S_0 + (I + 1/2)^{-2} \sin^2(\omega_h t / 2) [\langle I_n \rangle - 2S_0 \langle I_n^2 \rangle], \tag{13}$$

where  $I_n^2 = I(I + 1) - I_n^2$  is the square of the projection of the nuclear spin on a plane perpendicular to  $S_0$ , and the

angle brackets denote averaging over the initial state of the nucleus.

The polarization of the recombination radiation is determined by the value of the electron spin averaged over the time of its stay at the impurity center:

$$\bar{S}_n = \frac{1}{\tau} \int_0^\tau e^{-t/\tau} S_n(t) dt, \tag{14}$$

where  $\tau$  is the average lifetime of the electron at the center. From (13) and (14) we obtain

$$\bar{S}_n = S_0 + \frac{1}{2} \frac{(\omega_n \tau)^2}{1 + (\omega_n \tau)^2} \frac{\langle I_n \rangle - 2S_0 \langle I_{\perp}^2 \rangle}{(I + 1/2)^2}. \tag{15}$$

We see that the change of the electron spin consists of two parts. The first part is proportional to the average spin of the nucleus and describes the orientation of the electron by the nucleus. This effect has a pure quantum origin and is smaller the larger the nuclear spin  $I$ . The second part describes the vanishing of the electron orientation and is proportional to the mean-squared transverse component of the nuclear spin.

This second effect admits of a simple classical interpretation, in which the role of the nucleus reduces to the production of a magnetic field acting on the electron spin and equal to  $AI/\mu_0 g$ . Indeed, if the direction of  $S_0$  does not coincide at the initial instant with the direction of this field, then the electron spin will precess about it with frequency  $\omega_n$ . At  $\omega_n \tau \gg 1$ , the value of  $S_n$  averaged over the precession period is  $S_n \cos^2 \theta$ , where  $\theta$  is the angle between  $S_0$  and  $I$  (in the classical limit  $I \gg 1/2$ , so that the change of the nuclear spin direction can be neglected). Averaging over the initial state of the nuclei, we obtain

$$\bar{S}_n = S_0 - S_0 \langle I_{\perp}^2 \rangle / I^2, \tag{16}$$

which agrees with (15) if we neglect in the latter the term with  $\langle I_n \rangle$  and assume  $\omega_n \tau \gg 1$  and  $I \gg 1/2$ . In the case of a random distribution of the nuclear spins, formula (16) yields  $\bar{S}_n = S_0/3$ . In this case  $\langle I_n \rangle = 0$ ,  $\langle I_{\perp}^2 \rangle = 2I(I + 1)/3$ , and from the exact formula (15) we get

$$\bar{S}_n = S_0 \left[ 1 - \frac{(\omega_n \tau)^2}{1 + (\omega_n \tau)^2} \frac{2I(I + 1)}{3(I + 1/2)^2} \right]. \tag{17}$$

According to (17), if  $\omega_n \tau \gg 1$  then  $\bar{S}_n = S_0/2$  at  $I = 1/2$  and  $\bar{S}_n \rightarrow S_0/3$  in the classical limit  $I \gg 1$ .

To find  $S_n$  in the general case it is necessary to know the spin state of the nuclei. This state is determined by two factors: by the action of the electrons captured by the centers, and by nuclear relaxation. Let us find the change of the nuclear density matrix

$$\Delta \Phi_{\mu\mu'} = \sum_m \rho_{m\mu, m\mu'}(t) - \Phi_{\mu\mu'}$$

during the time when one electron stays at the center. During this time the nuclear relaxation, in view of its slowness, can be disregarded. From (10) we obtain for the diagonal elements

$$\begin{aligned} \Delta \Phi_{\mu\mu} = & \frac{1}{2} |v_{\mu+1/2}|^2 [(1 - 2S_0)\Phi_{\mu+1, \mu+1} - (1 + 2S_0)\Phi_{\mu\mu}] \\ & + \frac{1}{2} |v_{\mu-1/2}|^2 [(1 + 2S_0)\Phi_{\mu-1, \mu-1} - (1 - 2S_0)\Phi_{\mu\mu}]. \end{aligned} \tag{18}$$

We have averaged here over the time of the stay of the electron at the center. This averaging is denoted by a superior bar,

$$|v_{\mu}|^2 = \frac{1}{2} \frac{(\omega_n \tau)^2}{1 + (\omega_n \tau)^2} \frac{(I + 1/2)^2 - M^2}{(I + 1/2)^2}. \tag{19}$$

We can now write a kinetic equation describing the change of the nuclear density matrix over a time that is long in comparison with the time of stay of one electron at the center:

$$d\Phi_{\mu\mu} / dt = f \Delta \bar{\Phi}_{\mu\mu} - (\hat{v}\Phi)_{\mu\mu}, \tag{20}$$

where  $f$  is the number of electrons captured by one center per unit time, and  $\Delta \bar{\Phi}_{\mu\mu}$  is determined by expression (18), in which the matrix  $\Phi$  must be replaced by  $\bar{\Phi}(t)$ , since the change of the state of the nuclei during the time of stay of one electron at the center is small ( $f\tau \ll 1$ ). The second term in the right-hand side of (20) describes nuclear relaxation. The first term in (20) describes transitions between the spin levels of the nuclei as the result of hyperfine interaction with captured electrons. If there were no electron orientation ( $S_0 = 0$ ), then this interaction would lead to an equalization of the populations of the spin sublevels of the nuclei, i.e., to their additional spin relaxation. In this case the quantity  $(1/2)f|v_{\mu+1/2}|^2$  has the meaning of the probability of the transition  $\mu \rightleftharpoons \mu + 1$ .

As is well known, it is convenient to describe the spin state not by means of a density matrix, but by means of average values of the polarization moments of different orders (orientation, alignment, etc.). The relaxation times of these moments, generally speaking, are different. Let us find these times for relaxation on captured electrons. Multiplying the first term of (20) by  $\mu$  and summing over  $\mu$ , we obtain (at  $S_0 = 0$ )

$$f \sum_{\mu} \mu \Delta \bar{\Phi}_{\mu\mu} = -v_{e1} \langle I_n \rangle,$$

where

$$v_{e1} = \frac{2f}{(2I + 1)^2} \frac{(\omega_n \tau)^2}{1 + (\omega_n \tau)^2} \tag{21}$$

is the reciprocal time of decay of the orientation (of the average nuclear spin) as a result of the hyperfine interaction with captured non-oriented electrons. Analogously, at  $S_0 = 0$  we obtain for the second moment

$$f \sum_{\mu} [\mu^2 - I(I + 1)/3] \Delta \bar{\Phi}_{\mu\mu} = -v_{e2} \langle I_n^2 - I(I + 1)/3 \rangle,$$

where  $v_{e2}$  is the reciprocal time of decay of the alignment, i.e., of the quantity  $\langle I_n^2 - I(I + 1)/3 \rangle$ . Calculation yields  $v_{e2} = 3v_{e1}$ .

Equations (20) can be solved for an arbitrary nuclear spin if the character of the nuclear relaxation is known. We present the solution for the simplest case  $I = 1/2$ . In this case there is no alignment and (20) can be reduced to an equation for the single quantity  $\langle I_n \rangle$  by introducing a certain "dark" time of nuclear relaxation  $\nu_1$ :

$$d\langle I_n \rangle / dt = -(\nu_{e1} + \nu_1) \langle I_n \rangle + \nu_{e1} S_0. \tag{22}$$

Under stationary conditions we obtain

$$\langle I_n \rangle = S_0 \nu_{e1} / (\nu_{e1} + \nu_1). \tag{23}$$

We recall that the frequency  $f$  of the captures, and consequently also  $\nu_{e1}$ , is proportional to the intensity of the exciting light. Thus, at low intensities  $\langle I_n \rangle$  increases linearly and at high intensity it saturates at the level  $S_0$ . Substituting the obtained value of  $\langle I_n \rangle$  in formula (15) and using the fact that  $\langle I_{\perp}^2 \rangle = 1/2$  at  $I = 1/2$ , we get

$$\bar{S}_n = S_0 \left( 1 - \frac{\nu_{e1}}{f} \frac{\nu_1}{\nu_{e1} + \nu_1} \right). \quad (24)$$

with increasing light intensity, the electronic orientation increases and tends to  $S_0$  when  $\nu_{e1} \gg \nu_1$ .

At arbitrary nuclear spin, the electron orientation at low intensities is given by formula (17), and at high intensities when the dark nuclear relaxation is insignificant,  $\bar{S}_n \rightarrow S_0$ . We note that since the electron orientation depends on the spin state of the nuclei, the nuclear relaxation time should come into play in the transient processes that are connected, for example, with the change of intensity or of the degree of polarization of the exciting light.

#### 4. ORIENTATION OF ELECTRONS AND NUCLEI IN LONGITUDINAL MAGNETIC FIELDS

A magnetic field directed along the initial electron spin  $S_0$  should increase the stationary orientation of the electrons and decrease the stationary orientation of the nuclei. The reason for both effects lies in the fact that the longitudinal magnetic field weakens the coupling between the electron and nuclear spins. A parameter characterizing the influence of the field is the quantity defined by formula (5).

Expressions for the electron spin and for the change of the nucleon density matrix are given as before by formulas (12) and (18). Now, however, we have in place of formula (19) for  $|\bar{v}_M|^2$

$$|\bar{v}_M|^2 = \frac{(I + 1/2)^2 - M^2}{2(I + 1/2)^2} \frac{(\omega_n \tau)^2}{1 + (\omega_n \tau)^2 [1 + 4Mx/(2I + 1) + x^2]}. \quad (25)$$

At low light intensities, when the nuclei are not oriented, we can obtain an explicit expression for the magnetic-field dependence of the degree of orientation of the electrons. To this end, we put  $\Phi_{\mu\mu} = 2I + 1^{-1}$  in (12). We then obtain

$$\bar{S}_n = S_0 \left[ 1 - (I + 1/2)^{-1} \sum_M |\bar{v}_M|^2 \right]. \quad (26)$$

In particular, at  $I = 1/2$

$$\bar{S}_n = S_0 \left[ 1 - \frac{1}{2} \frac{(\omega_n \tau)^2}{1 + (\omega_n \tau)^2 + (\mu_0 g H \tau / \hbar)^2} \right]. \quad (27)$$

Figure 1 shows plots of  $\bar{S}_n(H)$  calculated from (26) for the case  $I = 3/2$ .

With increasing intensity of the exciting light, the spin distribution of the nuclei becomes ordered and this, in turn, influences the electron orientation. In the simplest case  $I = 1/2$ , at arbitrary intensities, formulas (23) and (24) are valid but with a value of  $\nu_{e1}$  that depends on the magnetic field and is given not by (21), but by

$$\nu_{e1} = \frac{f}{2} \frac{(\omega_n \tau)^2}{1 + (\omega_n \tau)^2 + (\mu_0 g H \tau / \hbar)^2}. \quad (28)$$

Thus, in a longitudinal magnetic field, to obtain the same nuclear orientation and at  $h = 0$ , it is necessary

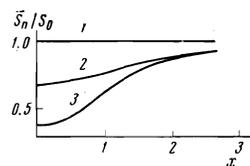


FIG. 1. Dependence of the stationary electron orientation on the longitudinal magnetic field at low intensity of the exciting light for  $I = 3/2$ . The values of the parameter  $\omega_n \tau$  are zero (1), 1 (2), and  $\infty$  (3).

to have a larger light intensity. The electron orientation, to the contrary, increases with increasing longitudinal magnetic field (see formulas (24) and (28)).

For nuclei with angular momenta  $I > 1/2$ , the general character of the dependence of the electronic and nuclear orientations on the magnetic field and on the intensity of the exciting light remains the same. There appears, however, a qualitatively new effect, namely the dependence of the orientation on the sign of the magnetic field, i.e., on whether the magnetic field is directed parallel or antiparallel to the initial electron spin  $S_0$ . This asymmetry is most strongly pronounced in intermediate fields and is connected with the fact that the quantities  $|\bar{v}_M|^2$  (which determine, according to (18), the probabilities of the transitions between the spin sublevels of the nucleus), are not even functions of the magnetic field (see formula (25)).

It is curious, however, that if the populations of all the nuclear spin sublevels relax in equal fashion (i.e., if the last term in (20) reduces to  $-\nu[\Phi_{\mu\mu} - (2I + 1)^{-1}]$ ) then it can be shown that there is no asymmetry in the electron orientation. At the same time, even in this case the stationary state of the nuclei depends on the sign of the magnetic field (relative to  $S_0$ ). Therefore, if the states of the nuclei does not have time to change following a reversal of the sign of the magnetic field, then the electron orientation changes and will then tend to a stationary value with the nuclear-relaxation time.

On the other hand, if the spin relaxation of the nuclei is not described by a single constant (for example, if the orientation and the alignment relax differently), as is usually the case, then the stationary orientation of the electrons depends on the sign of the magnetic field. The corresponding formulas can be obtained with the aid of (12), (20), and (25) if the character of the nuclear relaxation is known.

#### 5. DEPOLARIZATION IN A TRANSVERSE MAGNETIC FIELD (HANLE EFFECT)

A magnetic field transverse to  $S_0$  flips the spins of the electrons and nuclei and therefore decreases the stationary degree of orientation. The character of the influence of the transverse magnetic field is significantly different in the three field-variation intervals. In the weakest fields, the main effect is connected with the influence of the magnetic field on the nuclear spins, mainly in those time intervals when there is no electron at the center. When the nuclear spin precession period becomes smaller than the nuclear-relaxation time, the orientation and the alignment of the nuclei vanish, and the stationary value of the electron spin decreases to the value corresponding to a random distribution of the nuclear spins (formula (17)). We assume that the nuclear-relaxation time is so long that the spins of the nuclei are completely disoriented even in a field that does not affect as yet the electrons directly ( $\mu_0 g H \ll \hbar \omega_h$ ,  $\hbar \tau^{-1}$ ). With further increase of the magnetic field, the electron spins will turn. So long as  $\mu_0 g H \ll \hbar \omega_h$ , the two hyperfine states with total angular momentum  $I \pm 1/2$  will correspond to g-factors  $\pm g(2I + 1)^{-1}$ , so that in this region the hyperfine interaction decreases the depolarizing action of the transverse field. Finally, at  $\mu_0 g H \gg \hbar \omega_h$ , the coupling be-

tween the electron and nuclear spins breaks and the depolarization proceeds in the usual manner.

We consider first the case when the intensity of the exciting light is so low that there is no polarization of the nuclei. Then the spin intensity matrix of the nuclei is given by  $\Phi_{\mu\mu'} = \delta_{\mu\mu'}(2I+1)^{-1}$ . We shall assume as before that the quantization axis is chosen along the magnetic field. The initial direction  $n$  of the average electron spin  $S_0$  is taken to be the  $x$  axis. Then the average value of the electron spin projection on this direction at the instant  $t$  is given by the formula

$$\bar{S}_n(t) = \frac{1}{2} \sum_{\mu} [\rho_{\mu\mu, -\nu\mu} + \rho_{-\nu\mu, \nu\mu}]. \quad (29)$$

Using formulas (10) and (7) and the assumption that the nuclear spins are randomly distributed, we obtain from (29)

$$\bar{S}_n = S_0(2I+1)^{-1} \text{Re} \sum_{\mu} \overline{u_{\mu+1/2} u_{\mu-1/2}}, \quad (30)$$

where the superior bar, as before, denotes averaging over the time of stay of the electron at the impurity center (see (14)). Substituting the expression for  $u_M$  from (8), we finally get

$$\bar{S}_n = \frac{S_0}{2(2I+1)} \sum_{\mu} \left[ \frac{1+B_{\mu}}{1+(\tau\Delta_{\mu}^+)^2} + \frac{1-B_{\mu}}{1+(\tau\Delta_{\mu}^-)^2} \right], \quad (31)$$

where  $\Delta_{\mu}^{\pm} = \omega_{\mu} \pm 1/2 \pm \omega_{\mu} - 1/2$ ,

$$B_{\mu} = \frac{\omega_h^2 [\mu + 1/2 + x(I + 1/2)] [\mu - 1/2 + x(I + 1/2)]}{(2I+1)^2 \omega_{\mu+1/2} \omega_{\mu-1/2}}. \quad (32)$$

We recall that  $x = \mu_0 g H / (\hbar \omega_h)$ , and the frequencies  $\omega_M$  depend on the magnetic field in accordance with (9). At  $H = 0$ , Eq. (31) coincides with (17). We see that the form of the  $\bar{S}_n(H)$  plot (the Hanle line) is in general very complicated and is determined by the parameter  $\omega_h \tau$ . At  $\omega_h \tau \ll 1$ , the hyperfine interaction is negligible and the magnetic field influences the orientation only when  $x \gg 1$ . Then  $\omega_M = \mu_0 g H / 2\hbar$ ,  $B_{\mu} = 1$ , and formula (31) yields the usual relation

$$\bar{S}_n = S_0 [1 + (\mu_0 g H \tau / \hbar)^2]^{-1}. \quad (33)$$

On the other hand, if  $\omega_h \tau \gg 1$ , then the orientation vanishes already at magnetic fields such that  $x \ll 1$ . In this case we obtain from (31)

$$\bar{S}_n = \bar{S}_n(0) \left[ 1 + \left( \frac{\mu_0 g H \tau}{\hbar(2I+1)} \right)^2 \right]^{-1} \quad (34)$$

where  $\bar{S}_n(0)$  is determined by formula (17) at  $\omega_h \tau \gg 1$ . Thus, in this case the Hanle line again turns out to have a Lorentz shape, but with a  $g$ -factor decreased by a factor  $2I+1$ . Figure 2 shows a plot of  $\bar{S}_n(H)$  for a nuclear spin  $I = 3/2$ .

Formula (31) was derived under the assumption that the nuclei are not polarized. If the intensity of the ex-

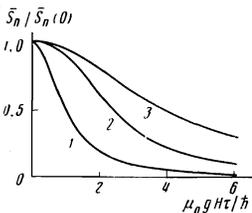


FIG. 2. Dependence of the stationary orientation of the electrons on the transverse magnetic field in the case of a random distribution of the nuclear spins at  $I = 3/2$ . The values of the parameter  $\omega_h \tau$  are zero (1), 2 (2), and  $\infty$  (3).

citing light is so high that the nuclei become noticeably polarized in the absence of a field, then this polarization vanishes in a weak magnetic field with values determined by the nuclear  $g$ -factor and by the nuclear spin-relaxation time. Accordingly, the electronic orientation decreases from the value (15) to the value (17), i.e., by an amount

$$\Delta S = \frac{1}{2} \frac{(\omega_h \tau)^2 \langle I_n \rangle + 2S_0 \langle I_n^2 \rangle - I(I+1)/3}{1 + (\omega_h \tau)^2 (I + 1/2)^2}, \quad (35)$$

which is determined by the orientation and alignment of the nuclei in a zero magnetic field. We present a formula describing the spike on the Hanle line in a weak field at  $I = 1/2$ :

$$\bar{S}_n = S_0 \left[ 1 - \frac{\nu_{e1}}{f} + \frac{\nu_{e1}}{f} \frac{\nu_{e1}}{\nu_{e1} + \nu_1} \frac{1}{1 + (\mu_N g_N H T / \hbar)^2} \right], \quad (36)$$

where  $\mu_N g_N H / \hbar$  is the nuclear-spin precession frequency in a magnetic field, the effective relaxation time  $T$  depends on the intensity and on the degree of polarization of the exciting light and is determined by the formula

$$T = [(\nu_{e1} + \nu_1)^2 + 4S_0^2 \nu_{e1}^2 (\omega_h \tau)^{-2}]^{-1/2},$$

and  $\nu_{e1}$  is given by formula (21) at  $I = 1/2$ .

At  $H = 0$ , formula (36) coincides with formula (24). We see that the width of the spike is determined not only by the dark relaxation of the nuclear spin, but also by the relaxation due to the captured electrons, so that the spike broadens with increasing exciting-light intensity. A spike on the Hanle line in weak magnetic fields was observed in [6].

## 6. OPTICAL ORIENTATION UNDER NUCLEAR RESONANCE CONDITIONS

We have seen above that optically oriented electrons polarize nuclei and as a result the electron-spin relaxation due to the hyperfine interaction decreases. Under nuclear-resonance conditions, the alternating magnetic field strives to equalize the populations of the spin levels of the nuclei, and consequently to eliminate the polarization of the nuclei. The electronic orientation therefore decreases under these conditions. This effect was observed in [7].

Nuclear resonance can be described with the aid of an equation for the off-diagonal density matrix of the nuclei  $\Phi_{\mu\mu'}$ :

$$\frac{d\Phi_{\mu\mu'}}{dt} = f \Delta \Phi_{\mu\mu'} - (\hat{V} \Phi)_{\mu\mu'} - \frac{i}{\hbar} [\hat{V}, \Phi]_{\mu\mu'}, \quad (37)$$

which is a natural generalization of (20). Here  $\hat{V} = -\mu_N g_N (\mathbf{H} + \mathbf{H}_1) \mathbf{I}$  is the operator of the interaction of the nuclear spin with the magnetic field, where  $\mathbf{H}$  is the constant field directed along  $S_0$  (the  $z$  axis) and  $\mathbf{H}_1$  is an alternating field perpendicular to the constant field ( $H_{1x} = H_1 \cos \omega t$ ,  $H_{1y} = -H_1 \sin \omega t$ ).

The expression for  $\Delta \Phi_{\mu\mu'}$  can be derived in analogy with the derivation of (18). We present here this expression for the simplest case  $I = 1/2$ :

$$f \Delta \Phi_{\frac{1}{2}, \frac{1}{2}} = -f \Delta \Phi_{\frac{1}{2}, -\frac{1}{2}} = -\frac{1}{2} \nu_{e1} (\Phi_{\frac{1}{2}, \frac{1}{2}} - \Phi_{-\frac{1}{2}, -\frac{1}{2}}) + \nu_{e1} S_0, \quad (38)$$

$$\overline{f \Delta \Phi_{\frac{1}{2}, -\frac{1}{2}}} = -g \Phi_{\frac{1}{2}, -\frac{1}{2}},$$

where  $\nu_{e1}$  is given by (28) and

$$g = f \left[ 1 - \exp \left( \frac{-i \omega_h x t}{2} \right) \left( \cos \omega_0 t + \frac{i x}{(1+x^2)^{1/2}} \sin \omega_0 t \right) \right]$$

$$\times \left( \cos \frac{\omega_n t}{2} - 2i S_0 \sin \frac{\omega_n t}{2} \right). \quad (39)$$

From (37) and (38) we obtain an expression for the change in the nuclear orientation as a result of the alternating field:

$$\langle \Delta I_n \rangle = - \langle I_n \rangle \frac{\Omega_1^2 T_1 T_2}{1 + \Omega_1^2 T_1 T_2 + \delta^2 T_2^2}, \quad (40)$$

where  $\langle I_n \rangle = \nu_{e1} \mathbf{T}_1 \mathbf{S}_0$  is the stationary nuclear orientation in the absence of an alternating field (see formula (23)),

$$\Omega_1 = \mu_N g_N H_1 / \hbar, \quad T_1 = (\nu_{e1} + \nu_1)^{-1}, \quad T_2 = (\bar{g}' + \nu)^{-1}, \\ \delta = \omega - \mu_N g_N H / \hbar + \bar{g}'',$$

$\bar{g}'$  and  $\bar{g}''$  are the real and imaginary parts of  $\bar{g}$ ,  $\nu_1^{-1}$  and  $\nu^{-1}$  are the longitudinal and transverse times of the dark relaxation of the nuclear spin.

The change of the nuclear orientation causes a change in the orientation of the electrons, since at  $I = 1/2$

$$\bar{S}_n = S_0(1 - \nu_{e1}/f) + (\nu_{e1}/f) \langle I_n \rangle.$$

We present expressions for the quantities  $\bar{g}'$  and  $\bar{g}''$ , which determine the broadening and the shift of the resonance as a result of the interaction of the nuclear spins with the electron spins for the case of weak and strong magnetic fields. In a weak field ( $x \ll 1$ ,  $\mu_0 g H \tau / \hbar \ll 1$ ) we have

$$\bar{g}' = \nu_{e1}, \quad \bar{g}'' = f S_0 \omega_n \tau [1 + (\omega_n \tau)^2]^{-1}.$$

In a strong field ( $x \gg 1$ )

$$\bar{g}' = f(\omega_n \tau)^2 [4 + (\omega_n \tau)^2]^{-1}, \quad \bar{g}'' = 4f S_0 \omega_n \tau [4 + (\omega_n \tau)^2]^{-1}.$$

We see that the broadening and the shift of the resonance line are proportional to the capture frequency  $f$ , i.e., to the intensity of the exciting light. The shift of the resonance frequency reverses sign when the sign of

the electron orientation changes, i.e., when the circular polarization of the exciting light changes sign.

Under resonance conditions, changes take place not only in the orientation but also in the alignment of nuclei with spin  $I \geq 3/2$ , as well as in other higher polarization moments. Since all the polarization moments of the nuclei influence the orientation of an electron in a magnetic field, the resonance line shape can be very complicated in the general case.

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