Optical orientation in a system of electrons and lattice nuclei in semiconductors. Theory

M. I. D'yakonov and V. I. Perel'

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

(Submitted February 20, 1973)
Zh. Eksp. Teor. Fiz. **65**, 362–375 (July 1973)

Interaction between optically oriented electrons and lattice nuclei in semiconductors is considered. Possible mechanisms of spin relaxation of electrons localized on shallow donors and also the effect of a longitudinal magnetic field on this relaxation are considered. Dynamic polarization of nuclei by electrons leads to the appearance of an effective magnetic field which acts on the electron spin and is due to hyperfine interaction. By enhancing or weakening the effect of the external magnetic field on the electron spin relaxation, this field affects the stationary degree of their orientation. Resonant alteration of electron orientation under nuclear-magnetic-resonance conditions caused by variation of the magnitude and direction of the nuclear effective magnetic field strength is also considered. The theory describes satisfactorily the main experimental results on optical orientation of electron and nuclear spins.

1. INTRODUCTION

During the optical orientation of electrons in semiconductors[1,2], dynamic polarization of the lattice nuclei[1] occurs, which affects in turn the orientation of the electrons and, because of this, can be detected by optical methods[3]. In our previous paper[4] we considered the hyperfine interaction of optically oriented electrons captured by impurity centers with the nuclei of these centers. Under such conditions the magnetic field created at an electron by the randomly oriented spin of the nucleus leads to the depolarization of the electron. An external magnetic field parallel to the orienting light beam decreases the rate of depolarization by disrupting the coupling between the spins of the electrons and nuclei. The depolarizing action of the nuclei also depends on the degree of order of the nuclear spins. Such order arises because of the dynamic polarization of the nuclei by the electrons. Any action on the nuclear spins (e.g., NMR) therefore leads to a change in the degree of orientation of the electrons.

In the present paper we consider the situation corresponding to the experiments of Ekimov and Safarov[3,5], in which the optical orientation was effected in a $Ga_xAl_{1-x}As$ solid solution, in which all the nuclei of the host lattice possess spin. Under the conditions of these experiments electrons thrown into the conduction band by the circularly polarized light are apparently rapidly captured by shallow donor centers. The region of localization of an electron encompasses a large number ($\sim 10^5$) of lattice nuclei. This situation is essentially different from the conditions considered previously[4], in which each electron interacts with only one nucleus. Indeed, unpolarized nuclei now create only a weak fluctuational magnetic field at the electron. On dynamic polarization of the nuclei, however, a strong field appears, which can reach several kOe. It is this field which is the main reason for the influence of the nuclear polarization on the electron spin.

In the presence of an external magnetic field, the action of the nuclear field on the electron spin is weakened or strengthened. Even λ if the hyperfine interaction is not the main mechanism of the electron spin relaxation, but the spin relaxation time depends on the magnetic field, on polarization of the nuclei this time will be lengthened or shortened, depending on whether the mean spin of the nucleus and the magnetic field are parallel or antiparallel. For polarization of the nuclei by the electrons the mean electron and nuclear spins are found to be parallel. Thus, the degree of polarization of the electrons will depend on the relative orientation of the external magnetic field and the mean electron spin. This asymmetry is in fact observed[5]. Under NMR conditions the magnitude and direction of the nuclear field change, and this leads to the resonance change in the orientation of the electrons that was observed by Ekimov and Safarov[3,5] using the degree of polarization of the luminescence.

In Secs. 2 and 3 we consider the possible mechanisms of spin relaxation of electrons localized at shallow donors, and also the effect of this relaxation of a longitudinal magnetic field. An increase of the spin orientation in an external magnetic field, indicating the existence of such an effect, has been observed experimentally[5,6].

Any spin relaxation mechanism can be interpreted as the action of random local magnetic fields acting on the electron spin. The nature of the effect of an external longitudinal magnetic field on the relaxation depends essentially on the value of the correlation time characterizing the rate of change of the local fields. If the correlation time is long compared with the period of precession of the electron spin in the local field, an external field will slow down the spin relaxation considerably as soon as it becomes greater than the local field. In the opposite case, dynamic averaging of the local fields occurs and there will be a noticeable slowing-down of the spin relaxation in the external field only when the period of precession of the spin in this field is comparable with the correlation time.

The correlation time of the local field is, in any case, not longer than the time of an electron jump from one donor to another, Both the estimates and the experimental data of[5] are evidence that dynamic averaging of the local fields does in fact occur. Only this case is considered in the present paper.

In Sec. 4 and 5, we consider the dynamic polarization of nuclei by oriented electrons, leading to the appearance of an additional effective magnetic field acting on the electron spin. The stationary orientation of the electrons is calculated as a function of the external magnetic field. In Sec. 6 the change in the degree of orientation during nuclear magnetic resonance is discussed.

The main experimental results of[5] are well described on the basis of the picture proposed here of optical orientation in a system of interesting electrons and nuclear spins.

2. THE LOCAL MAGNETIC FIELDS ACTING ON THE ELECTRON SPIN

A characteristic of the spin relaxation of localized electrons is that the orientation of the spins of such electrons is very sensitive to the local magnetic fields in the crystal. We shall discuss two possible reasons for the existence of such fields: the hyperfine interaction with the lattice nuclei and the exchange interaction with the holes bound to the acceptors in p-type semiconductors. We shall estimate the magnitude of the local fields due to these interactions.

Owing to the hyperfine interaction with the lattice nuclei, a localized electron spin is acted upon by the effective magnetic field

$$\mathbf{H}_c = (\mu_0 g)^{-1} v_0 \sum_n |\psi(\mathbf{r}_n)|^2 A_n \mathbf{I}_n, \qquad (1)$$

where μ_0 is the Bohr magneton, g is the g-factor, v_0 is the volume of a unit cell, $\psi(r_n)$ is the value of the "effective-mass method" wavefunction (describing the state of the electron at the donor) at the site of the n-th nucleus, I_n is the spin of this nucleus, and A_n is the constant of the hyperfine interaction of the electron with the n-th nucleus; this constant takes the same value for nuclei of the same type occupying equivalent positions in the lattice. If the nuclear spins are not ordered, we find for the mean square effective field

$$\overline{H_c^2} = (\mu_0 g)^{-2} \frac{v_0}{8\pi a^3} \sum_\alpha A_\alpha^2 I_\alpha (I_\alpha + 1). \qquad (2)$$

Here the summation is performed over the nuclei in one unit cell, and a is the donor radius. In calculating (2) we have taken the electron wavefunction in the form

$$\psi(r) = (\pi a^3)^{-1/2} e^{-r/a}. \qquad (3)$$

For the following, the value of the precession frequency of the electron spin in the random field of the nuclei, $\omega_c = \mu_0 g(\overline{H_c^2})^{1/2}/\hbar$, is important. For a shallow donor in gallium arsenide, an estimate gives $\omega_c \sim 5 \times 10^8$ sec^{-1}. For $2 > g > 0.5$ [7,8], this corresponds to a magnetic field between 30 and 100 Oe¹⁾.

We now proceed to an estimate of the effective magnetic fields due to the exchange interaction of a localized electron with the holes at the acceptors. This interaction has the form[9]

$$V_{\text{exch}} = \Delta \mathbf{J} \sigma,$$

where J is the angular-momentum operator of the hole, σ are the Pauli matrices, and Δ is a constant depending on the overlap of the electron and hole wavefunctions. If the acceptor radius is much smaller than the donor radius, the effective magnetic field acting on the electron spin and due to the exchange interaction with an acceptor situated at a distance r can be estimated as follows:

$$\mathbf{H}_c = \frac{\Delta_{\text{ex}}}{2\mu_0 g} e^{-2r/a} \mathbf{J}, \qquad (4)$$

where Δ_{ex} is the magnitude of the exchange splitting in the exciton. The mean square field can be obtained by averaging over the sitting of the nearest acceptor. For $N_A a^3 \ll 1$, where N_A is the concentration of acceptors, we obtain (for $J = 3/2$)

$$\overline{H_c^2} = \frac{15\pi}{128} \frac{N_A a^3}{(\mu_0 g)^2} \Delta_{\text{ex}}^2. \qquad (5)$$

For most semiconductors, the quantity Δ_{ex} is not known. Roughly estimating $\Delta_{\text{ex}} \sim 10^{-5} - 10^{-4}$ eV for an acceptor concentration of $N_A = 10^{16}$ cm^{-3}, we find $\omega_c \sim 10^9 - 10^{10}$ sec^{-1}.

Thus, the local magnetic fields due to the holes are somewhat greater than the fields of the nuclei. However, the spin relaxation of the electrons depends not only on the magnitude of the random field, but also on its rate of change. If we denote the characteristic time of variation of the effective magnetic field by γ^{-1}, the spin relaxation time will be of the order of $\tau_s \sim \gamma/\omega_c^2$ (dynamic averaging—it is assumed that $\omega_c/\gamma \ll 1$).

If ω_c is determined by the hyperfine interaction, in view of the slow change of the spin state of the nuclei it can be assumed that the variation of the effective field is associated with electron hops between donors, and γ^{-1} is the time an electron spends at one donor²⁾. But in the case of exchange interaction with the holes there is also another reason for the variation of the effective magnetic field, viz., the spin relaxation of the holes. Because of the very strong spin-orbit coupling in the valence band, the spin relaxation time of the holes can be so short that it is this time which will determine the quantity γ^{-1} in this case. The electron spin relaxation due to the exchange interaction with the holes will then be strongly suppressed.

Along with the factors considered above, there may also be other causes of spin relaxation of the localized electrons. If, e.g., the electron spends an appreciable fraction of its time in excited states with non-zero orbital angular momentum, the spin-orbit interaction may be such a cause. Paramagnetic impurities can also play a role, if their concentration is sufficiently large. In all cases, the important parameters determining the relaxation are the magnitude of the effective local magnetic field H_c and the correlation time γ^{-1} characterizing the rate of change of this field with time.

3. SPIN RELAXATION OF THE ELECTRONS AND ITS DEPENDENCE ON THE MAGNETIC FIELD

We consider the longitudinal spin relaxation of electrons in a magnetic field $H_c(t)$ varying randomly in time, in the presence of a constant external field H_0. The probability per unit time of a spin flip is given by the expression[11]

$$W = \frac{1}{2} \left(\frac{\mu_0 g}{\hbar}\right)^2 \operatorname{Re} \int_0^\infty \exp(-i\Omega_e \tau) \langle H_{c-}(0) H_{c+}(\tau) \rangle \, d\tau, \qquad (6)$$

where $\Omega_e = \mu_0 g H_0/\hbar$ is the precession frequency of the electron spin in the external field (which is assumed to be along the z-axis), $H_{c-} = H_{cx} - iH_{cy}$, and the angular brackets denote averaging over all realizations of the random field.

In the following, we shall use the simplest model for the field correlation function, putting

$$\langle H_{c-}(0) H_{c+}(\tau) \rangle = \overline{H_{c\perp}^2} \exp(-\gamma\tau - i\Omega'\tau), \qquad (7)$$

where $\overline{H_{c\perp}^2} = \overline{2/3\, H_c^2}$, $\overline{H_c^2}$ is the mean square of the random field, and γ is the inverse correlation time of the random field. The factor $\exp(-i\Omega'\tau)$ takes into account the regular variation of the random field in time, due to the precession of the spins of the holes and nuclei or of the orbital angular momenta in the external magnetic field ($\Omega' > 0$ if the direction of this precession is opposite to the direction of precession of a free-electron spin).

Substituting (7) into the expression (6) and introducing the electron spin relaxation time $\tau_S = (2W)^{-1}$, we find

$$\frac{1}{\tau_s}=\frac{2}{3}\frac{\omega_c^2\gamma}{\gamma^2+(\Omega_e+\Omega')^2}, \tag{8}$$

$$\omega_c^2=(\mu_0 g/\hbar)^2\overline{H_c^2}. \tag{9}$$

If the relaxation occurs at the nuclei, then $\Omega' \ll \Omega_e$. It can be seen from expression (8) that the spin relaxation time begins to depend on the external magnetic field when the period of precession of the spin in this field becomes of the order of or smaller than the characteristic correlation time of the random field. For $\Omega_e = \Omega' = 0$, we have $\tau_S^{-1} = \frac{2}{3}\omega_c^2\gamma^{-1}$, which reflects the dynamic averaging of the local fields. Formula (8) is valid of $\gamma\tau_S \gg 1$, i.e., if the spin relaxation during the correlation time is insignificant. This restriction is associated with the use of the perturbation-theory formula (6).

The simple dependence $\tau_S(H_0)$ given by formula (8) is based on the assumption (7). In fact, this dependence can be more complicated, since the values of γ are different for donors with different environments, and τ_S^{-1} is determined by the average of the expression (8) over a certain distribution of values of γ. Formula (8) was derived as if the random fields were classical. The same result is also obtained for τ_S in a quantum-mechanical treatment. The quantum generalization of the method used here is given in the following section in an application to the spin relaxation of the nuclei.

4. EFFECT OF THE ELECTRONS ON THE SPIN STATE OF THE NUCLEI

We shall find the change in the spin state of the nuclei due to the hyperfine interaction with the electrons, taking into account the possible spin orientation of the electrons. We write the Hamiltonian of the interaction of a nucleus with the electrons in the form

$$V=-\mu_I\hat{\mathbf{I}}\hat{\mathbf{H}}_s, \tag{10}$$

where μ_I and $\hat{\mathbf{I}}$ are the magnetic moment and spin operator of the nucleus, and

$$\hat{\mathbf{H}}_s=-\frac{16\pi}{3I}\mu_0\sum_n\delta(\mathbf{r}_n-\mathbf{R})\hat{\mathbf{S}}_n \tag{11}$$

is the operator of the magnetic field due to the electrons and acting on the nucleus; r_n and $\hat{S}_n$ are the coordinates and spin of the n-the electron, and R is the position of the nucleus under consideration.

The probability $W_{\mu,\mu-1}$ of a transition of the nucleus from the Zeeman sublevel μ to the sublevel $\mu-1$ per unit time is given by an expression analogous to (6):

$$W_{\mu,\mu-1}=\frac{\mu_I^2}{2\hbar^2}(I+\mu)(I-\mu+1)\,\mathrm{Re}\int_0^\infty d\tau\langle\hat{H}_{s-}(0)\hat{H}_{s+}(\tau)\rangle. \tag{12}$$

Here we are explicitly taking into account the quantum character of the field $\hat{H}_S$, by taking $\hat{H}_S(\tau)$ to be the operator $\hat{H}_S$ in the interaction picture. We have neglected the Zeeman energy of the nucleus in the magnetic field, assuming that the important values of τ in the integral (12) are those such that $\omega_0\tau \ll 1$, where ω_0 is the nuclear spin precession frequency. The angular brackets denote averaging over the states of the electron subsystem.

Taking (11) into account, we can write the integral occurring in formula (12) in the form

$$\mathrm{Re}\int_0^\infty d\tau\langle\hat{H}_{s-}(0)\hat{H}_{s+}(\tau)\rangle=\left(\frac{16\pi\mu_0}{3I}\right)^2\left(\frac{1}{2}-S\right)K_1(\mathbf{R},\Omega_e), \tag{13}$$

where S is the mean value of the projection of the electron spin along the direction of the magnetic field, and

$$K_1(\mathbf{R},\omega)=\mathrm{Re}\int_0 d\tau e^{i\omega\tau}\langle\hat{\rho}(\mathbf{R},0)\hat{\rho}(\mathbf{R},\tau)\rangle. \tag{14}$$

Here $\hat{\rho}(R, \tau)$ is the operator, in the interaction picture, of the electron density

$$\rho(\mathbf{R})=\sum_n\delta(\mathbf{r}_n-\mathbf{R})$$

at the given nucleus. The quantity K_1 can be expressed in terms of the fluctuation spectrum of the electron density at the nucleus. We introduce the function

$$K(\mathbf{R},\omega)=\frac{1}{2}\,\mathrm{Re}\int_0^\infty d\tau\, e^{i\omega\tau}\langle\hat{\rho}(\mathbf{R},0)\hat{\rho}(\mathbf{R},\tau)+\hat{\rho}(\mathbf{R},\tau)\hat{\rho}(\mathbf{R},0)\rangle. \tag{15}$$

Then[12],

$$K_1(\mathbf{R},\omega)=\exp\left(-\frac{\hbar\omega}{2T}\right)\left[\mathrm{ch}\left(\frac{\hbar\omega}{2T}\right)\right]^{-1}K(\mathbf{R},\omega),$$

where T is the temperature in energy units. Finally, we find a general formula for the transition probability:

$$W_{\mu,\mu-1}=\left(\frac{8\pi\mu_0\mu_I}{3I\hbar}\right)^2(I+\mu)(I-\mu+1)K(\mathbf{R},\Omega_e)(1+2S_T)(1-2S). \tag{16}$$

Here $S_T = -\frac{1}{2}\tanh(\hbar\Omega_e/2T)$ is the thermodynamic equilibrium value of the projection of the electron spin along the direction of the magnetic field.

In an analogous way, we find for the probability of the reverse transition

$$W_{\mu-1,\mu}=\left(\frac{8\pi\mu_0\mu_I}{3I\hbar}\right)^2(I+\mu)(I-\mu+1)K(\mathbf{R},\Omega_e)(1-2S_T)(1+2S). \tag{17}$$

The probabilities of the direct and reverse transitions differ by the factors $(1 \pm 2S_T)(1 \pm 2S)$. This difference exists if the spin orientation of the electrons is not the thermodynamic equilibrium orientation, and leads in this case to orientation of the nuclear spins.

The condition for the applicability of formulas (16) and (17) is that the probability of a change of nuclear spin during the correlation time of the electron density be small compared with unity. These formulas are valid irrespective of the nature of the orbital states of the electrons.

If the electrons are localized at donors we can assume, as in formula (7), that

$$\frac{1}{2}\langle\hat{\rho}(\mathbf{R},0)\hat{\rho}(\mathbf{R},\tau)+\hat{\rho}(\mathbf{R},\tau)\hat{\rho}(\mathbf{R},0)\rangle=F|u(\mathbf{R})|^4v_0^2\psi^4(r)\exp(-\gamma_e\tau),$$

$$K(\mathbf{R},\omega)=F|u(\mathbf{R})|^4v_0^2\psi^4(r)\gamma_e(\gamma_e^2+\omega^2)^{-1}. \tag{18}$$

Here F is the degree of occupation of the donor, u(R) is the Bloch amplitude at the site of the given nucleus, normalized in the volume v_0 of the unit cell, $\psi(r)$ is the "effective-mass method" wave-function of the electron at the donor (formula (3)), r is the distance from the nucleus to the nearest donor center, and γ_e^{-1} is the time that the electron stays at one donor, which coincides with the correlation time, introduced in Sec.

3, of the random nuclear field acting on the electron spin.

Using formula (18), we find

$$W_{\mu,\mu\pm1}=(I\mp\mu)(I\pm\mu+1)F\frac{\omega^2(r)\gamma_e}{\gamma_e^2+\Omega_e^2}(1\mp2S_T)(1\pm2S),\tag{19}$$

$$\omega(r)=\frac{A}{2\hbar}\frac{v_0}{\pi a^3}e^{-2r/a},\tag{20}$$

where $\omega(r)$ is the precession frequency of the nuclear spin in the magnetic field created by the localized-electron spin as a result of the contact hyperfine interaction, and A is the hyperfine interaction constant.

The change in the populations of the Zeeman nuclear sublevels as a result of the interaction with the electrons is described by the equations

$$\frac{d\Phi_{\mu\mu}}{dt}=-(W_{\mu,\mu+1}+W_{\mu,\mu-1})\Phi_{\mu\mu}+W_{\mu+1,\mu}\Phi_{\mu+1,\mu+1}+W_{\mu-1,\mu}\Phi_{\mu-1,\mu-1},\tag{21}$$

where $\Phi_{\mu\mu}$ is a diagonal element of the spin density matrix of the nuclei (the population of sublevel μ). If the other factors influencing the nuclear spin are unimportant, the stationary distribution of the populations $\Phi_{\mu\mu}$ can be obtained by equating the right-hand side of Eq. (21) to zero. Using formulas (16) and (17), we find that, in the stationary state,

$$\Phi_{\mu\mu}=Z^{-1}e^{\mu x/I},\tag{22}$$

$$x=I\left(\frac{\hbar\Omega_e}{T}+\ln\frac{1+2S}{1-2S}\right),\tag{23}$$

where $Z=\sum\limits_{\mu}\exp(\mu x/I)$ is a normalization factor. The mean value $\langle I_z\rangle$ of the projection of the nuclear spin along the direction of the magnetic field is

$$\langle I_z\rangle=IB_I(x),\tag{24}$$

where $B_I(x)$ is the Brillouin function[13]. If we introduce the electron spin temperature T_S, the second term in formula (23) can be written as $-\hbar\Omega_e/T_S$.

Formula (24) describes the well-known dynamic polarization of nuclei due to a difference between the spin temperature of the electrons and the lattice temperature. Under conditions of optical orientation of the electrons, the first term in (23) is usually small compared with the second. Figure 1 shows the dependence of the stationary degree of orientation of the nuclei on the degree of orientation of the electrons, as obtained from formulas (23) and (24) with neglect of the quantity $\hbar\Omega_e/T$.

We shall now obtain an equation describing the change of nuclear orientation with time. Multiplying Eq. (21) by μ and summing, we obtain

$$\frac{d\langle I_z\rangle}{dt}=\sum_{\mu}\Phi_{\mu\mu}(W_{\mu,\mu+1}-W_{\mu,\mu-1}).$$

Using expressions (16) and (17) for W, we can rewrite this equation in the form

$$\frac{d\langle I_z\rangle}{dt}=-\frac{1}{T_{1e}(r)}\left(\langle I_z\rangle-\langle I_\perp^2\rangle\,\mathrm{th}\frac{x}{2I}\right),\tag{25}$$

where $I_\perp^2=I_x^2+I_y^2$, and the longitudinal nuclear spin relaxation time $T_{1e}(r)$ due to the interaction with the electrons is given by the formula

$$\frac{1}{T_{1e}(r)}=2\left(\frac{8\pi\mu_0\mu_I}{3I\hbar}\right)^2K(R,\Omega_e)(1-4SS_T).\tag{26}$$

If we assume that the electrons are localized at the donors and use the approximation (18), then

$$\frac{1}{T_{1e}(r)}=2F\frac{\omega^2(r)\gamma_e}{\gamma_e^2+\Omega_e^2}(1-4SS_T).\tag{27}$$

In particular, for $S=0$, the formula (27) gives the longitudinal relaxation time of the nuclei positioned near the donor, due to the unoriented electrons situated at this donor. We shall estimate T_{1e} in zero magnetic field for the nuclei lying in the region of localization of the electron ($r\ll a$). Putting $\gamma_e\sim10^{10}\ \mathrm{sec}^{-1}$ and $\omega(0)\sim10^6\ \mathrm{sec}^{-1}$, for $F=10^{-3}$ we obtain the value $T_{1e}(0)\sim5$ sec.

We shall make several comments concerning Eq. (25). The quantity $\langle I_\perp^2\rangle$, which itself varies in the orientation process, appears in this equation. While the degree of polarization of the nuclei remains small, we can put $I_\perp^2=\frac{2}{3}I(I+1)$. For an arbitrary degree of polarization of the nuclei, we can express $\langle I_\perp^2\rangle$ in terms of $\langle I_z\rangle$ by using the concept of a nuclear spin temperature[11].

In Eq. (25) the diffusion of the nuclear magnetization as a result of the spin-spin interaction of the nuclei has not been taken into account. Neglect of the diffusion is justified if the time T_{1e} is much shorter than the time of diffusion over the donor radius a. The diffusion coefficient $D\sim10^{-13}\ \mathrm{cm^2sec^{-1}}$[11], so that the diffusion time $a^2/D\sim10$ sec (for a shallow donor in GaAs, $a\sim10^{-6}$ cm). For $T_{1e}(0)\gg a^2/D$, the diffusion will lead to rapid equalization of the degree of orientation in the region of the donor. The stationary degree of orientation of the nuclei will then depend essentially on their rate of spin relaxation at the paramagnetic centers (which, in particular, can be neutral acceptors). If the donor concentration is so great that the orientation has time to diffuse over the distance between donors in the time $T_{1e}(0)$, a uniform distribution of nuclear spins is established. In this case, the quantity $T_{1e}^{-1}(r)$ in Eq. (25) must be averaged over all positions of nuclei of a given type and a term responsible for the relaxation at the paramagnetic impurities must be added to the right-hand side of this equation. Then, putting $\hbar\Omega_e/T\ll S$ and assuming the nuclear polarization to be small, we obtain

$$\frac{d\langle I_z\rangle}{dt}=-\frac{1}{T_{1e}}\left[\langle I_z\rangle-\frac{4}{3}SI(I+1)\right]-\frac{1}{T_1'}\langle I_z\rangle,\tag{28}$$

Here,

$$\frac{1}{T_{1e}}=N_D\int\frac{d^3\mathbf{r}}{T_{1e}(r)}=\frac{\pi}{8}\frac{N_Da^3}{T_{1e}(0)}.\tag{29}$$

Here T_1' is the nuclear spin relaxation time due to spin diffusion to the paramagnetic centers[11]. This time, generally speaking, can depend on the intensity of the exciting light, since the electrons thrown into the conduction band can create new paramagnetic centers by being captured at deep traps. In addition, the electrons can influence the spin relaxation of the existing centers, thereby altering their effectiveness as a source of spin relaxation of the nuclei.

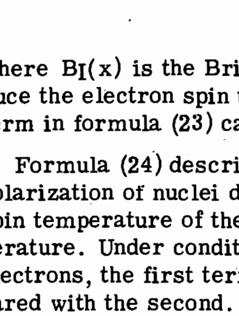

FIG. 1. Limiting nuclear polarization as a function of the degree of orientation of the electrons (2S) according to formula (24) with $\hbar\Omega_e\ll T$: 1–I = ½; 2–I = 3/2.

As can be seen from Eq. (28), the total longitudinal relaxation time of the nuclear spins is

$$T_1 = (1/T_{1e} + 1/T_1')^{-1},$$

and the stationary degree of orientation is given by the formula

$$\langle I_z\rangle = \tfrac{4}{3}I(I+1)(T_1/T_{1e})S, \qquad (30)$$

which is applicable so long as $\langle I_z\rangle \ll 1$.

5. EFFECT OF THE POLARIZED NUCLEI ON THE SPIN ORIENTATION OF THE ELECTRONS

The polarization of the nuclei arising as a result of their interaction with the oriented electrons changes, in its turn, the spin orientation of the electrons. This change can be due to two factors. Firstly, commensurately with the appearance of the nuclear polarization, the magnitude of the random (transverse) magnetic field created by the nuclear spins as a result of the hyperfine interaction decreases. Therefore, the electron spin relaxation due to this interaction will decrease. If the contact interaction (10) is the only cause of longitudinal nuclear relaxation, the transfer of spin from the electron system to the nuclear system ceases after the stationary nuclear orientation (24) is reached. This is connected with the fact that the interaction (10) conserves the total spin of the electrons and nuclei, so that if the nuclear spin has attained its stationary value spin relaxation of the electrons at the nuclei cannot proceed.

The second factor causing a change in the electron spin relaxation on polarization of the nuclei is the appearance in this case of a regular longitudinal magnetic field arising from the oriented nuclei and acting on the electron spins. According to the results of Sec. 3, this field, when combined with the external field, will change the rate of spin relaxation of the electrons even in those cases when the hyperfine interaction is not the direct cause of this relaxation. If all the nuclei in the region occupied by the electron at the donor were completely oriented, the effective magnetic field H_N would be of order $AI/\mu_0 g$. This field can be of order 10^4 Oe[3)]. Therefore, even for negligible polarization of the nuclei, the effective field H_N can be found to be important.

We shall consider the role of this field in more detail, assuming the first of the factors enumerated above to be unimportant (i.e., assuming that the polarization of the nuclei is small ($\langle I_z\rangle \ll I$) or that the electron spin relaxes mainly away from the nuclei). Allowance for the effect of the regular effective field of the polarized nuclei on the electron spin relaxation reduces in this case to replacing the quantity Ω_e in formula (8) (and also in formula (27)) by the quantity $\mu_0 g\,|\mathbf{H}_0 + \mathbf{H}_N|$, where H_0 is the external magnetic field. Then the spin relaxation time τ_s will be determined by the formula

$$\frac{1}{\tau_s} = \frac{2}{3}\frac{\omega_c^2}{\gamma}\left[1+\left(\frac{\mathbf{H}_0+\eta\mathbf{H}_N}{H_\gamma}\right)^2\right]^{-1},$$
$$H_\gamma = \gamma[\mu_0(g+g')]^{-1}, \quad \eta = g/(g+g'), \quad g' = \Omega'/\mu_0 H_0. \qquad (31)$$

The regular nuclear field H_N is determined by averaging the expression (1):

$$\mathbf{H}_N = (\mu_0 g)^{-1} v_0 \sum_n |\psi(\mathbf{r}_n)|^2 A_n \langle \mathbf{I}_n\rangle. \qquad (32)$$

For relaxation at the nuclei, we can take $g' = 0$; if the relaxation is due to the holes, g' is the g-factor of a hole; if the relaxation occurs because of the spin-orbit interaction, then $g' = 2m_0/m$, where m_0 is the free-electron mass and m is the effective mass.

In the stationary state the preferred orientation of the nuclear spins coincides with the electron spin orientation, so that the direction of H_N is determined by the sign of the circular polarization of the exciting light. Thus, in a magnetic field the rate of spin relaxation of the electrons and, consequently, their stationary degree of orientation depend on the sign of the circular polarization of the light.

The stationary value of S (the mean z-component of the electron spin) will be greater when the preferred spin orientation of the electrons thrown into the conduction band coincides with the direction of the external magnetic field H_0. The expression for S has the form

$$S = S_0(1 + \tau/\tau_s)^{-1}, \qquad (33)$$

where S_0 is the initial value of S, which is determined by the degree of polarization of the light, the selection rules, and the relaxation processes which can occur during the time of thermalization of the electrons[2], and τ is the electron lifetime.

In the case under consideration, formula (33) is in fact an equation for S, since the nuclear field H_N appearing in the expression (31) for τ_s depends on the degree of orientation of the electrons. This dependence can be established easily in two opposite limiting cases.

1. The polarization of the nuclei is maximum. This case can be realized if the time of diffusion of a nuclear spin over the donor radius is greater than the time T_{1e} occurring in formula (25). In this case, the polarization of the nuclei is given by formula (24). Substituting (24) into (32), we obtain

$$H_N = (\mu_0 g)^{-1} \sum_\alpha A_\alpha I_\alpha B_{I_\alpha}(x), \qquad (34)$$

where the summation is taken over all the nuclei in the unit cell. The formulas (31)–(34) determine the stationary value of S as a function of the external magnetic field.

For small values of S and $S_T \ll S$, the Brillouin function in formula (34) can be replaced by the first term of its expansion in powers of S:

$$B_I(x) = \tfrac{4}{3}S(I+1).$$

As can be seen from Fig. 1, this expansion gives an error of less than 10% up to $S = 0.2$. Finally, Eq. (33) for S takes in this case the form

$$S = S_0 \Big/ \left\{1 + \frac{\tau}{\tau_s(0)}\left[1+\left(\frac{H_0+\beta S}{H_\gamma}\right)^2\right]^{-1}\right\},$$
$$1/\tau_s(0) = 2\omega_c^2(3\gamma)^{-1}, \qquad (35)$$

where $\tau_s(0)$ is the electron spin relaxation time in the absence of the magnetic field (i.e., of both the external and nuclear fields),

$$\beta = 4[3\mu_0(g+g')]^{-1} \sum_\alpha A_\alpha I_\alpha (I_\alpha + 1). \qquad (36)$$

Figure 2 shows the dependences $S(H_0)$ following from formula (35), for S_0 parallel to H_0 and for S_0 antiparallel to H_0.

We recall that when the nuclei attained their maximum polarization (24), the relaxation of the electron spin at the nuclei ceases. Therefore, in the case under consideration the time τ_s is determined by other relaxation mechanisms.

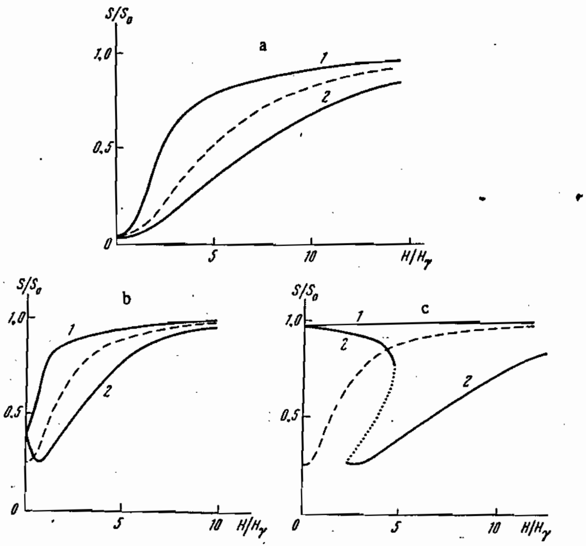

FIG. 2. Dependence, obtained from formula (35), of the electron spin orientation on the external longitudinal magnetic field H_0. The dashed curves are constructed with neglect of the nuclear field ($\beta = 0$). The curves 1 are for S_0 parallel to H_0, and the curves 2 for S_0 antiparallel to H_0. a—$\tau/\tau_s(0) = 24$, $\beta S_0/H_\gamma = 5$; b—$\tau/\tau_s(0) = 3$, $\beta S_0/H_\gamma = 2.5$; c—$\tau/\tau_s(0) = 3$, $\beta S_0/H_\gamma = 10$. In the case (c) with S_0 antiparallel to H_0, in a certain range of fields Eq. (35) has three solutions, of which one corresponds to an unstable state (the dotted curve).

2. The nuclear polarization is much smaller than the maximum. In this case, according to formula (30), the orientation of the nuclei is proportional to that of the electrons. Then,

$$H_N = 4(3\mu_0 g)^{-1} \sum_\alpha A_\alpha I_\alpha (I_\alpha + 1) \left(\frac{T_1}{T_{1e}}\right)_\alpha S \qquad (37)$$

and, as before, the equation for S can be written in the form (35), but with the expression for β (36) replaced by an analogous expression in which there is an additional factor $(T_1/T_{1e})_\alpha$ in the summand. In this case, the quantity β can itself depend on the magnetic field. The dependences $S(H_0)$ obtained in experiment[5] are close to that depicted in Fig. 2a.

The polarization of the nuclei by the electrons proceeds fairly slowly. According to the model proposed here, immediately after the exciting circularly polarized light is switched on, the electron spin orientation takes a value corresponding to the dashed curve in Fig. 2. Then, commensurately with the polarization of the nuclei, which proceeds with a characteristic time T_1, the electron orientation should tend to a value corresponding to the upper or lower curve of Fig. 2, depending on the relative orientation of S_0 and H_0. In the experiments of[5], inertial effects with characteristic time of order 10 sec were observed in the establishment of the luminescence polarization. However, the reasons for the essential difference between these effects in the parallel and antiparallel orientations of S_0 and H_0 remain unclear.

6. EFFECT OF NUCLEAR MAGNETIC RESONANCE ON THE ELECTRON SPIN ORIENTATION

During the action of a radio-frequency (RF) field on the spins of the polarized nuclei, the effective magnetic field acting on the electron spins changes in both magnitude and direction. This can explain the experimentally observed[3,5] change (reflecting the change of orientation of the electrons under NMR conditions) of the luminescence polarization. Typical NMR frequencies are small compared with the inverse electron lifetime τ^{-1}, so that even in resonance conditions the effective regular field of the nuclei acts as a static field on the electron spin.

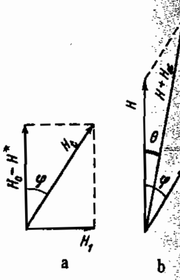

FIG. 3. a—The magnetic field H_t acting on the nuclei in the rotating coordinate frame; b—the combined magnetic field $H + H_\alpha$ acting on an electron in the case $H_\alpha \parallel H_t$.

In NMR conditions, the nuclear field H_N is no longer parallel to S_0. The combined field $H_0 + H_N$ will thus make a certain angle θ with S_0. In sufficiently high fields, when

$$\mu_0 g |\mathbf{H}_0 + \mathbf{H}_N| \gg \hbar(1/\tau + 1/\tau_s),$$

in stationary conditions only the component of the electron spin along the direction of the combined field is conserved. This component will be expressed by formula (33), with the quantity S_0 replaced by $S_0 \cos\theta$. In order to obtain the desired stationary value S of the component of the spin along the direction of S_0, we must introduce a further factor $\cos\theta$. Thus,

$$S = S_0 \cos^2\theta (1 + \tau/\tau_s)^{-1}, \qquad (38)$$

$$\cos^2\theta = \frac{(H + H_{\alpha\parallel})^2}{(H + H_{\alpha\parallel})^2 + H_{\alpha\perp}^2}. \qquad (39)$$

Here H is the sum of the external field and the fields of all the nuclei for which the RF field is not equal to the resonance field; H_α is the field of the nuclei (of type α) which are in NMR conditions; $H_{\alpha\parallel}$ and $H_{\alpha\perp}$ are the components of H_α along and perpendicular to the external field H_0.

We shall consider the nature of the variation of the nuclear field H_α under NMR conditions. Since the field H_α is proportional to the mean spin $\langle I_\alpha \rangle$ of the nuclei of type α, we can make use of the well-known picture of the behavior of a nuclear spin in solids[11]. This behavior is usually described by means of the concept of the spin temperature in the rotating coordinate frame[11,15].

Let the coordinate frame rotate about the direction of the constant magnetic field H_0 (the z axis) with the frequency ω of the RF field. In this frame, the nuclei are acted upon by a static magnetic field H_t with components $H_0 - H^*$ along the z axis and H_1 in the xy plane, where $H^* = -\hbar\omega I_\alpha/\mu_\alpha$ and H_1 is the amplitude of the RF field (cf. Fig. 3a). On passage through the resonance, the direction of the field H_t is rotated through 180°. In the following we shall assume that the characteristic time of variation of the field H_t is much longer than the transverse relaxation time T_2. In addition, we shall confine ourselves to treating the case of a strong RF field, much greater than the local field determining the width of the unsaturated NMR line. Then the character of the variation of the field H_α is easily described[11] in two limiting cases.

1. Adiabatically fast passage (the time of passage through the resonance is shorter than the longitudinal relaxation time T_1). In this case, the mean spin $\langle I_\alpha \rangle$,

and with it the field H_α, remains unchanged in magnitude and rotates together with the field H_t, remaining all the time parallel or antiparallel to it, depending on the initial orientation. If we assume for definiteness that the quantity $\omega - \omega_0$ changes from positive to negative values on passage through the resonance, then

$$H_{\alpha\parallel}=\frac{\omega-\omega_0}{((\omega-\omega_0)^2+\Omega_1^2)^{1/2}}H_{\alpha 0},\quad H_{\alpha\perp}=\frac{\Omega_1}{((\omega-\omega_0)^2+\Omega_1^2)^{1/2}}H_{\alpha 0},\qquad (40)$$

where ω_0 is the resonance frequency for the type of nucleus under consideration in the field H_0; $\Omega_1 = \mu_\alpha H_1/\hbar I_\alpha$; $H_{\alpha 0}$ is the component of the nuclear field along the direction of H_0 before passage through the resonance and far from resonance.

2. Slow passage. In this case, a stationary value of the nuclear field, independent of the initial conditions, is established for each value of the field H_t. The projection of the vector H_α on the direction of the field H_t equals the quantity $H_{\alpha 0} \cos\varphi$ (φ is the angle between H_0 and H_t), and the projections of the vector H_α on a plane perpendicular to H_t are equal to zero. Then,

$$H_{\alpha\parallel}=\frac{(\omega-\omega_0)^2}{(\omega-\omega_0)^2+\Omega_1^2}H_{\alpha 0},\quad H_{\alpha\perp}=\frac{\Omega_1(\omega-\omega_0)}{(\omega-\omega_0)^2+\Omega_1^2}H_{\alpha 0}.\qquad (41)$$

We can assume that the value of $H_{\alpha 0}$ in formulas (41) is the same as in formulas (40), if the mean electron spin (which determines the pumping for the nuclear spin) changes little during the NMR.

The formulas (38)—(41) describe the effect of NMR on the spin orientation of the electrons. This effect is associated with the variation of the combined field $H + H_\alpha$ during NMR (Fig. 3b) and is determined by two factors. First, the RF field leads to the appearance of a nuclear-field component perpendicular to S_0, and this component reduces the electron orientation. Thus, the factor $\cos^2\theta$ in formula (38) changes. For small values of H_α, we can put $\cos^2\theta = 1 - H_{\alpha\perp}^2/H^2$. For fast passage, the dependence of the quantity $H_{\alpha\perp}^2$ on the resonance detuning is described by a Lorentzian contour. A characteristic feature of slow passage is the vanishing of $H_{\alpha\perp}$ at resonance.

Secondly, the change in the magnitude of the combined field during NMR leads to a change of the time τ_s: If, e.g., S_0 and H_0 are antiparallel, then before the passage the nuclear field is opposite in direction to the external field. During fast passage, the field H_α is rotated and the combined field increases monotonically from the value $H - H_{\alpha 0}$ to the value $H + H_{\alpha 0}$, leading to an increase of τ_s and to an increase of spin orientation. During slow passage, the combined field and, consequently, τ_s first increase, reaching a maximum at resonance (where $H_\alpha = 0$), and then decrease to their original values.

The nature of the change of electron orientation during NMR, as obtained from the above treatment, is in complete agreement with the results of the experiment of[5].

Above, we have been interested in the projection S of the mean electron spin along the direction of H_0. We remark that the perpendicular component which appears in NMR conditions rotates with the RF-field frequency ω. This should lead to oscillations of the luminescence polarization in observations at an angle to H_0.

We are grateful to A. I. Ekimov and B. I. Safarov for communicating the experimental data before publication, and for useful discussions.

---

[1] In a paper of Haraldson and Ribbing [8] it was remarked that the large EPR line-width in gallium arsenide (~100 Oe) may be due, at least partially, to the hyperfine interaction of the electron at the donor with the lattice nuclei.

[2] This time is somewhat shorter than the hopping time determining the hopping conductivity. The experimental data on the hopping conductivity in GaAs (cf., e.g., the survey by Shklovskiĭ [10]) make it possible to estimate the magnitude of γ: $\gamma \sim 10^{10}-10^{11}$ sec^{-1} for a donor concentration $N_D = 10^{16}$ cm^{-3} and temperature 4°K.

[3] We considered earlier [14] the effects due to this field which arise on dynamic polarization of the nuclei.

[1] G. Lampel, Phys. Rev. Lett. 20, 491 (1968).
[2] R. R. Parsons, Phys. Rev. Lett. 23, 1152 (1969); A. I. Ekimov and V. I. Safarov, ZhETF Pis. Red. 12, 293 (1970) [JETP Lett. 12, 198 (1970); B. P. Zakharchenya, V. G. Fleĭsher, R. I. Dzhioev, Yu. P. Veshchunov and I. B. Rusanov, ZhETF Pis. Red. 13, 195 (1971) [JETP Lett. 13, 137 (1971)]; M. I. D'yakonov and V. I. Perel', Zh. Eksp. Teor. Fiz. 60, 1954 (1971) [Sov. Phys.-JETP 33, 1053 (1971)].
[3] A. I. Ekimov and V. I. Safarov, ZhETF Pis. Red. 15, 257, 453 (1972) [JETP Lett. 15, 179, 319 (1972)].
[4] M. I. D'yakonov and V. I. Perel', Zh. Eksp. Teor. Fiz. 63, 1883 (1972) [Sov. Phys.-JETP 36, 995 (1973)].
[5] V. L. Berkovits, A. I. Ekimov, and V. I. Safarov, Zh. Eksp. Teor. Fiz. 65, 346 (1973) [this issue, p. 169].
[6] R. R. Parsons, Can. J. Phys. 49, 1850 (1971).
[7] W. Duncan and E. E. Schneider, Phys. Lett. 7, 23 (1963).
[8] S. Haraldson and C.-G. Ribbing, J. Phys. Chem. Sol. 30, 2419 (1969).
[9] G. L. Bir and G. E. Pikus, Simmetriya i deformatsionnye effekty v poluprovodnikakh (Symmetry and Deformation Effects in Semiconductors) Nauka, M., 1972, p. 562; G. I. Pikus and G. L. Bir, Zh. Eksp. Teor. Fiz. 60, 195 (1971) [Sov. Phys.-JETP 33, 108 (1971)].
[10] B. I. Shklovskiĭ, Fiz. Tekh. Poluprov. 6, 1197 (1972) [Sov. Phys.-Semiconductors 6, 1053 (1973)].
[11] A. Abragam, The Principles of Nuclear Magnetism, Oxford University Press, 1961 (Russ. transl., IIL, M., 1963).
[12] L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Fizmatgiz, M., 1959 [English translation published by Pergamon Press, Oxford, 1960].
[13] C. Kittel, Introduction to Solid State Physics, Wiley N. Y., 1956 (Russ. transl., Fizmatgiz, M., 1963, p. 246).
[14] M. I. D'yakonov and V. I. Perel', ZhETF Pis. Red. 16, 563 (1972) [JETP Lett. 16, 398 (1972)].
[15] A. G. Redfield, Phys. Rev. 98, 1787 (1955).

Translated by P. J. Shepherd
40