Cooling of the spin system of semiconductor lattice nuclei in the field of electrons oriented by light

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Cooling of the spin system of semiconductor lattice nuclei in the field of electrons oriented by circularly polarized light is considered. As a result of cooling application of an external magnetic field induces a considerable nuclear polarization and an aditional nuclear field H_N acts on the electron spins. An equation is derived for the behavior of the z-component of the mean electron spin $\langle S_z \rangle$ in a transverse field H_x when cooling depends only on the oriented electron field. Characteristic features of the dependence of $\langle S_z \rangle$ on H_x are a narrow line near $H_x = 0$ and an additional peak in strong fields. Instability and hysteresis may arise in the case of a positive electron g-factor (g_e^*) . Experimental results for a p-Al_{0.26}Ge_{0.74}As crystal $(g_e^*>0)$ with an acceptor (Zn) concentration $\simeq 10^{18}$ cm⁻³ confirm the model under consideration. The effect of variable magnetic fields which alter the nuclear spin temperature down to values lying in the $10^{-3}-10^{-4}$ °K range occurs in experiments carried out at 77°K. The local nuclear field and electron field at the nuclei are found from the experimental data to be (4.2 ± 0.6) Oe and (5.4 ± 1.7) $\langle S_z \rangle$ Oe, respectively.

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1. INTRODUCTION

The absorption of circularly polarized light under conditions of optical orientations of the electrons is accompanied by dynamic polarization of the nuclei.^[1-3] In the presence of an external magnetic field $H > H_L$, where H_L is the local field of the nuclei, the dynamic polarization is well described within the framework of the Overhauser effect. The average spin $\langle I_z \rangle$ of the nuclei along the propagation direction of the exciting light is determined by the deviation of the average spin $\langle S_r \rangle$ of the photoexcited electrons from the thermodynamicequilibrium value $\langle S_T \rangle$. It was observed recently that optical orientation is accompanied by an appreciable nuclear polarization also in the case $H \le H_{L^*}^{[4,5]}$ In this range of fields, the Zeeman energy of a nucleus is commensurate with the energy of its spin-spin interaction with the neighboring nuclei. The change in the energy of the nuclear Zeeman pool (ZP) due to the hyperfine interaction with the electrons oriented by the light is accompanied by a change of the energy of the nuclear spinspin pool (SP). The change of the energy of the ZP is proportional to $\langle S \rangle$ H. Depending on the sign of this product, the energy of the SP increases or decreases as a result of its "thermal" contact with the ZP. With increasing energy of the SP, one can speak of cooling in the region of negative temperatures, since the energy spectrum of the SP is bounded from above. With decreasing energy of the SP, cooling takes place in the region of positive temperatures. Thus, under conditions of optical orientation in weak fields $H \leq H_L$, it is possible to cool the spin system of the nuclei, in analogy with dynamic cooling in strong field $(H \gg H_L)$ following radio-frequency saturation of the transitions near the resonance frequency.^[6]

The theory of the cooling of the spin system of lattice nuclei of a semiconductor in the case of optical orientation was developed by D'yakonov and Perel'.^[7] An interesting possibility of dynamic polarization of nuclei under conditions of optical orientation is cooling at a zero external magnetic field, when the change of the Zeeman energy of the nuclei can be obtained as a result of the field of optically-oriented electrons. Allowance for this field leads to a number of interesting features that manifest themselves in experiments on optical orientation in weak fields.

2. CHANGE OF ELECTRON-SPIN ORIENTATION IN A TRANSVERSE MAGNETIC FIELD (HANLE EFFECT) UPON COOLING OF THE SPIN SYSTEM OF THE LATTICE NUCLEI

The lowering of the spin temperature Θ of nuclei leads to an increase of the nuclear polarization along the external field *H*. If the polarization is much less than the limiting value and if the lattice consists of nuclei of one sort, then

$$\langle \mathbf{I} \rangle = (I+1) \mu_I \mathbf{H} / 3\Theta,$$
 (1)

where I is the spin of the nucleus, μ_I is its magnetic moment and $\langle I \rangle$ is the average value of the projection of the nuclear spin on the direction of the magnetic field, while Θ is expressed in energy units. The increase of the nuclear polarization is accompanied by an increase of the effective magnetic field $H_N = h_N \langle I \rangle$ acting on the electron spins. The value of h_N determines the maximum field $H_{N \max}$ at 100% polarization of the nuclei $(h_N = H_{N \max}/I)$.

If the light orients the electron spins along z, then the average value $\langle S_z \rangle$ of the projection of the electron spins along z will change as a result of the action of the combined field $H_1 = H + H_N$. In this case the field H_N can manifest itself by a change in the relaxation time τ_S (see^[2]) (in the presence of a component H_{Nz} along z), or else in a change in the transverse component of the magnetic field on account of H_{Nx} under conditions of the Hanle effect in an oblique field.^[8] In a pure transverse external field $(\langle S \rangle \perp H)$ it might seem that H_N cannot appear. Indeed, the nonequilibrium nuclear polarization produced as a result of the hyperfine interaction with the electrons oriented by the light is very small (see^[9]) and vanishes within a time $T_2 \approx 10^{-4}$ sec. Nor can cooling occur in a purely transverse field, since Θ is determined by the scalar product $\langle S \rangle \cdot H^{[7]}$:

$$\frac{1}{\Theta} = \frac{4I \langle \mathbf{S} \rangle \mathbf{H}}{\mu_I (H^2 + 3H_L^2)}.$$
 (2)

(The vanishing of $1/\Theta$ denotes that the temperature of the nuclear spin system does not differ from the lattice temperature, taken to be ∞ .) Nonetheless, strong manifestations of H_N are observed in experiments in a purely transverse geometry (see^[4,5]). To explain the observed effect it is necessary to assume that the light produces a longitudinal magnetic-field component. D'yakonov and Perel' pointed out the role of the effective field H_e of the electrons oriented by the light. The influence of this field in oblique geometry is considered in a paper by Paget.^[10] Assume that $H_e = h_e \langle S_e \rangle (h_e/2)$ is the field of the fully oriented electrons) is appreciable and let us calculate the function $S_{\epsilon}(H)$ under conditions when the spin system of the lattice nuclei is cooled in the field H_e . The experimental results presented below confirmed the presence of cooling and justifies the choice of the considered model.

The change of the average electron spin with time is described by the Bloch equation

$$\langle \dot{\mathbf{S}} \rangle = \frac{\mathbf{S}_0 - \langle \mathbf{S} \rangle}{\tau} - \frac{\langle \mathbf{S} \rangle}{\tau_s} + \gamma_* [\langle \mathbf{S} \rangle \times \mathbf{H}_1], \qquad (3)$$

where γ_e is the gyromagnetic ratio for the electrons, τ is the lifetime of the photoexcited electrons, and S_0 is the maximum attainable average spin ($|S_0| = 0.25$ in crystals of the GaAs type). Under stationary conditions we have

$$\langle \mathbf{S} \rangle = \frac{T}{\tau} \mathbf{S}_0 + \gamma_* T[\langle \mathbf{S} \rangle \times \mathbf{H}_1], \qquad (4)$$

where T is the total lifetime of the spin orientation and is determined by the equation $T^{-1} = \tau^{-1} + \tau_s^{-1}$.

Using (1) and (2), we express H_1 in the form

$$\mathbf{H}_{i} = \mathbf{H} + \frac{4I(I+1)h_{N}}{3} \frac{(\mathbf{H} + h_{e}\langle \mathbf{S} \rangle) \left(\langle \mathbf{S} \rangle \langle \mathbf{H} + h_{e}\langle \mathbf{S} \rangle\right)\right)}{(\mathbf{H} + h_{e}\langle \mathbf{S} \rangle)^{2} + 3H_{L}^{2}} .$$
 (5)

H in (2) has been replaced here by $\mathbf{H} + h_e \langle S \rangle$ to allow for the field of the oriented electrons at the nuclei. At H = 0we have $\langle S \rangle = \langle S_x \rangle_0 = TS_0/\tau$. The additional field, defined by the second term of (5), is oriented along the resultant of the fields H and $h_e \langle S \rangle$, and the inclination of the field \mathbf{H}_1 varies in a complicated manner in space when the value of \mathbf{H}_x varies. This notwithstanding, the precession of the vector $\langle S \rangle$ is determined only by the field component H_N directed along the external field, as much as $[\mathbf{S} \times \mathbf{H}_1] \sim [\mathbf{S} \times \mathbf{H}]$ (see (5)). At $H = H_x$, the field H_N influences only the precession of $\langle S \rangle$ about the x axis. Taking the vector product of (5) and $\langle S \rangle$, we obtain

$$[\langle \mathbf{S} \rangle \times \mathbf{H}_{1}] = [\langle \mathbf{S} \rangle \times \mathbf{H}] \left(1 + \frac{\langle \mathbf{S} \rangle^{2} h_{e} h_{N}'}{H^{2} + h_{e}^{2} \langle \mathbf{S} \rangle^{2} + 3H_{L}^{2}} \right).$$
(6)

Here $h'_N = 4I(I+1)h_N/3$.

At $H = H_x$, the cooling of the spin system of the nuclei in the field of the electrons oriented by the light leads to the appearance of an additional transverse field, equal to

$$H_{x}\langle S\rangle^{2}h_{e}h_{N}^{\prime\prime}/(H_{x}^{2}+h_{e}^{2}\langle S\rangle^{2}+3H_{L}^{2}).$$

This field is either added to the external field H_x or subtracted from it, depending on the sign of the product $h_c h'_N$.

The hyperfine interaction energy $\varepsilon_{I,S}$ can be expressed both in terms of the field H_e and in terms of the field H_N , with

$$\operatorname{sign} \varepsilon_{IS} = \operatorname{sign} [g_e^* \mu_B \langle S \rangle H_N] = \operatorname{sign} [-\gamma_I \hbar \langle I \rangle H_e].$$
(7)

Here g_e^* is the g factor of the electrons in the crystal, μ_B is the Bohr magneton, γ_I is the gyromagnetic ratio for nuclei, and \hbar is Planck's constant. Substituting the values of $H_e = h_e S$ and $H_N = h_N \langle I \rangle$ we see that at $\gamma_I > 0$ we have $h_e h'_N < 0$ if $g_e^* > 0$. If the g-factor is negative, then $h_e h'_N$ > 0. In the case of a positive g-factor, cooling takes place in the region of negative temperatures, and the additional field connected with the polarization of the nuclei is directed opposite to the external field. At g_e^* < 0 these fields are in the same direction.

Let us find an equation connecting $\langle S_s \rangle$ and H_x . Taking the scalar product of (4) and $\langle S \rangle$ and using (6), we find that

$$S_0 \langle S_z \rangle \langle S \rangle^2 = T/\tau$$

$$S_0 \langle S_y \rangle \langle S \rangle^2 = H\gamma_e \tau [1 + \langle S \rangle^2 h_e h_S'/(H^2 + h_e^2 \langle S \rangle^2 + 3H_L^2)].$$

Introducing the dimensionless parameters $\alpha = 3T^2 \gamma_e^2 H_L^2$ and $\beta = \langle S_x \rangle_0^2 h_e h'_N / 3H_L^2$, we obtain an equation that relates the dimensionless variables $\chi = \langle S_x \rangle / \langle S_x \rangle_0$ and $b = H_x^2 / 3H_L^2$:

$$(1-\chi)(1+b)^2 = \alpha b \chi (1+b+\beta \chi)^2.$$
 (8)

This equation describes the behavior of the z component in a transverse field H_x under conditions of optical cooling of the spin system of the nuclei in the field of electrons oriented by light. It is easily seen that in the absence of an electron field ($\beta = 0$) Eq. (8) goes over into the equation $\chi = 1/(1 + \alpha b)$, which corresponds to the usual Lorentz curve that describes the Hanle effect, $\langle S_e \rangle$ $= S_0 T/\tau (1 + \gamma_e^2 T^2 H^2)$, with half-width $H_{1/2}$ equal to $1/\gamma_e T$.

We see thus that the effect of cooling of the spin system of the nuclei in the field of the electrons is taken into account in (8) by the single parameter β . An important role is played by the spin of β . Figure 1a shows plots of $\chi(b^{1/2})$ for positive β at $\alpha = 0.01$. A characteristic feature of these plots is the presence of a narrow line in the region of weak fields $(H \leq H_L)$ and of an additional maximum in the region of strong fields. The depth of the minimum of χ increases with increasing β , and its position depends little on β . At the same time, the additional maximum shifts towards stronger fields with increasing β . The entire $\chi(b^{1/2})$ curve lies under



FIG. 1. Calculated plots of $\langle S_{g} \rangle$ against the transverse magnetic field H_x (the Hanle effect) in the dimensionless coordinates $\chi = \langle S_{g} \rangle / \langle S_{g} \rangle_0$ and $b^{1/2} = H_x / 3^{1/2} H_L$. The influence of the cooling of the lattice-nuclei spin system in the field of electrons oriented by light is characterized by the parameter β . At $\beta = 0$ there is no nuclear field at the electrons and the half-width of the $\chi(b^{1/2})$ curve is determined by the parameter α . a) $\beta \ge 0$, $\alpha = 0.01$. b) $\beta \le 0$. The solid lines correspond to $\alpha = 0.1$ and the dashed lines to $\beta = -40$, $\alpha = 0.25$ and 0.05.

the corresponding Lorentz curve. Interesting singularities are possessed also by the $\chi(b^{1/2})$ at $\beta < 0$. Thus, at the characteristic point $b_1 = \beta - 1$ we have $\chi = 1$. The field H_M corresponding to the maximum at the point b_1 + 1 is determined by the geometric mean $(h_e h'_N)^{1/2}$.

At definite parameter ratios, Eq. (8) has three real roots. It is easy to verify that at $\alpha(|\beta| - 1) > 4$, at the point b_1 , it is possible to have besides the solution $\chi = 1$ also values of $\langle S_z \rangle$ determined by the equation $\chi = \frac{1}{2} \pm \left[\frac{1}{4}\right]$ $-1/\alpha(|\beta|-1)^{1/2}$. This means that there are S-shaped sections that include instability regions. Experiments should then reveal jumplike changes of the polarization. The instability regions can arise in fields for which αb >3. Figure 1b shows plots of $\chi(b^{1/2})$ at fixed values β = 0, -10, -20 and α = 0.1. Curves for different values of α have been plotted for $\beta = -40$. As seen from the figures, with increasing the slope of the curve past the additional maxima increases first, after which the instability region appears. In experiment, the presence of this region should lead to jumplike changes of $\langle S_{s} \rangle$ at the boundaries of the regions, and of the dependence of $\langle S_{z} \rangle$ on the direction of variation of the external field H_{x} (hysteresis). The width of the hysteresis loop increases with increasing α . We note that the relatively weak electron field $h_e(S)$ influences strongly the form of the dependence of $\langle S_z \rangle$ at external fields $H_x \gg h_e$, if h'_N is large. At $\beta < 0$, the $\chi(b^{1/2})$ curve crosses the Lorentz curve at one point in the region of small b, and with increasing b it subsequently approaches this curve but remains above it.

In the experiment, the value of the field h'_N can be determined if the field *H* is inclined to the *x* axis at a small angle $\varphi \ll 1$. Equation (8) then goes over into

$$(1-\chi)(1+b)^2 = ab\chi(1+b+\beta\chi\pm\delta b''_{\mu}\phi)^2,$$
 (9)

which differs from (8) in the additional term $b^{1/2}$, where $\delta = h'_N \langle S_x \rangle_0 / \sqrt{3} H_L$. Different signs correspond to opposite directions of the external field H and are a reflection of the appearance of asymmetry in the dependence of $\langle S_x \rangle$ on H. The relative shift $|H_{M_1}| - |H_{M_2}|$ of the positions of the additional maxima and the dependence of $\langle S_x \rangle$ on H for opposite directions of H determines the value of the field h'_N :

$$\pm h_{s'} = (|H_{s_{1}}| - |H_{s_{2}}|) / \langle S_{z} \rangle_{0} \varphi.$$
(10)

At h'_N of the order of $10^4 - 10^5$ Oe, an appreciable asymmetry should appear already at very small angles φ .

The narrow line near H=0 under the condition $b \ll 1$ is described by the equation

$$b = (1-\chi)/\alpha\chi(1-\beta\chi)^2.$$
(11)

In the case of large β , when $\beta \chi \gg 1$, this line can be conveniently used to determine the values of the local fields, by measuring $H_{\rm M}$ and $H_{1/2}$

$$3H_{L^{2}} \approx (H_{\mathbf{x}^{2}}/H_{\gamma_{0}}) (\partial H/\partial \eta), \quad \eta = [(1-\chi)/\chi^{2}]^{\gamma_{0}}.$$
(12)

This formula is valid if $\partial H/\partial \eta = \text{const}$ for an appreciable fraction of the lines, starting with H = 0.

We present below experimental results that confirm the reality of the considered process of cooling of the spin system of lattice nuclei in the field of electrons oriented by light. A preliminary report of these results was published earlier.^[5]

3. EXPERIMENT

The simplest way to investigate the dependence of $\langle S_{r} \rangle$ on H_{r} is to measure the degree of the circular polarization ρ of the recombination radiation with participation of optically oriented electrons. For GaAs crystals and for solid solutions on its basis, the numerical value of ρ is equal to that of $\langle S_{s} \rangle$ if the investigated radiation is in the direction of the z axis. Figure 2 shows a simplified diagram of the experiment. A linearly polarized He-Ne laser beam passes through a quarterwave plate $\lambda/4$ and produces optical orientation in the sample. The crystal is oriented in such a way that the incident and reflected rays are superimposed. The luminescence is registered in a "reflection" geometry in a direction making a small angle with the z axis. The value of ρ is measured with a circular-polarization analyzer (PA), which comprises a quartz phase modulator and a linear polarizer. The necessary spectral range is chosen with the aid of a spectral instrument (SI). The radiation is registered with a cooled photomultiplier operating in the photon-counting regime. The quartz modulator is analogous to that described by Jasperson and Schnatterly.^[11] The piezoelectric quartz,



FIG. 2. Simplified diagram of experiment. $M_{1,2}$ —operating coils, $M_{3,4,5}$ —Helmholtz coils to cancel the earth's magnetic field, $L_{1,2}$ —focusing lenses, PA—polarization analyzer, $\lambda/4$ —quarter-wave plate, He-Ne—helium-neon laser, SI—spectral instrument, PM—photomultiplier, QO—quartz-modulator os-cillator, SS—switching system, SU—scaler unit, DPU—print-out unit.

exciting the longitudinal oscillations in a bar of fused quartz is connected in the negative-feedback circuit of the oscillator QO. The oscillator operates at a frequency 30.265 kHz. Adjacent half-cycles of the fusedquartz bar correspond to predominant transmission of photons having opposite signs of the circular polarization (σ^* and σ^-) through the polarization analyzer system. If the number of these photons is N^+ and N^- , respectively, then $\rho = (\pi/2.36)(N^* - N^-)/(N^* + N^-)$. The coefficient $\pi/2.36$ is determined by the form of the transmission function of the PA. The switching system (SS) ensures sequential switching, in synchronism with the quartz operation, of the counting channels of a twochannel scaler unit (SU) to the photomultiplier output. The digital printout unit (DPU) records the ratios $N^*/$ N^{-} . A more detailed description of the system for measuring ρ in the photon-count regime was published earlier.^[12]

The earth's magnetic field is cancelled out by three pairs of Helmholtz coils (M_3, M_4, M_5) . The axis of the coil pair M_1 is aligned with the x axis or is turned through an angle φ around the y axis. The coils M_2 are used to investigate the effect of an alternating magnetic field of low frequency on the spin system of the nuclei. Most measurements were performed at 77 °K.

The investigations were performed on $p-Al_xGa_{1-x}As$

FIG. 3. Hanle effect in transverse (curve 1) and inclined magnetic field $(2-\varphi=5^\circ)$, $3-\varphi=15^\circ$, $4-\varphi=25^\circ$), at a fixed sign of the circular polarization of the exciting light (σ_{μ}).



FIG. 4. Frequency dependences of the variation of the luminescence polarization following application of an additional alternating magnetic field H_{y} . at $H_{x} = 12.5$ Oe and 40 Oe. Excitation σ_{x} . The arrows show the direction of the variation of ρ_{x} .

crystals with constant composition gradient (+ 0.01 μ^{-1}) with x = 0.26 on the surface. The crystals were not compensated and the Zn acceptor concentration was ~ 10^{18} cm⁻³.

Figure 3 shows plots of $\chi \equiv \rho / \rho'$ against *H* for the angles $\varphi = 0^{\circ}$ (curve 1), 5° (2), 15° (3) and 25° (4) (ρ_0 is the value of ρ at H = 0 Oe). As seen from the figure, even at small values of φ the $\rho(H)$ curve becomes strongly asymmetrical. A narrow line is observed near H = 0, as well as additional maxima followed by steep drops. However, the heights of the additional maxima do not reach $\chi = 1$, probably because of the inhomogeneity of the field h_e (see below). The presented curves correspond to the case $\beta < 0$, which should be expected for a crystal with a positive g-factor. In accordance with the model developed above, elimination of the effective field of the nuclei at the electrons should be accompanied by an increase of ρ in the region of weak fields and by a decrease of ρ in the region of the additional maximum and directly beyond it. This is observed in experiment. The saturating low-frequency field acting along the yaxis, produces in ρ changes of opposite signs for different sections of the $\rho(H)$ curve. The fact that ρ decreases in the fall-out region past the additional maximum demonstrates that the field H_N decreases the depolarizing action of the external field, i.e., that these fields are oppositely directed.

Figure 4 shows the frequency dependence of the variation of following application of H_{y} , for two values of the field H_{x} .

The next fact that confirms the cooling of the nuclear spin system is the possibility of changing ρ in the case of synchronous modulation of the circular polarization of the exciting beam and a field H_s parallel to this beam. Turning to (2), we note that if the time-averaged value of $(\langle \mathbf{S} \cdot \mathbf{H} \rangle)$ is not equal to zero, then Θ decreases. Depending on the phase difference between the oscillations of S and H, the cooling takes place in the region of positive or negative temperatures. The polarization of the nuclei cooled in the oscillating magnetic field is determined by the thermodynamic-equilibrium value in the constant electric field H (see (1)). A more detailed analysis of the cooling in an oscillating magnetic field is given elsewhere.^[13] The lifetime of the photoexcited electrons is $< 10^{-7}$ sec and if the polarization of the excited light is modulated at a frequency ~ 30 kHz the value of $\langle S_z \rangle$ changes in phase with the variation of this po-





FIG. 5. Variations of ρ in the case of synchronous modulation of the circular polarization of the exciting light (σ_{\star}) and of the longitudinal magnetic field H_{g} at a frequency ~30 kHz, as functions of the phase shift ψ between σ_{\star} and $H_{g \star}$.

larization. To perform the experiment, the positions of the quartz modulator and of the $\lambda/4$ plate were interchanged (see Fig. 2), and the reference voltage from the quartz oscillator was applied through a phase shifter and an amplifier to the coils M₂. Figure 5 shows the variation of ρ with changing phase difference ψ between the oscillations of $\langle S_{z} \rangle_{\sim}$ and H_{z} , corresponding to variation of Θ (see (2) and^[13]).

A nuclear spin system can be cooled not only in an oscillating magnetic field but also in the oscillating field of optically-oriented electrons. This effect decreases with increasing modulation frequency of the excitinglight polarization, but at a frequency ~ 30 kHz it is still distinctly observed. Figure 6 shows a plot of $\rho(H)$ (curve 1) in the case of a sharply-focused laser beam with modulated polarization. The additional maximum is distinctly observable. It is obvious that this curve corresponds to smaller values of β than in the case of the curves shown in Fig. 3. With decreasing intensity of the exciting light, when the effective field of the electron decreases, the $\rho(H)$ curve becomes smoother. Smoothing is observed also when a low-frequency alternating field (not in synchronism with the polarization modulation) is turned on. This field heats the spin system of the nuclei. Curve 2 corresponds to turning on an additional magnetic field $H_{g_{n}}$ with amplitude 1 Oe and



FIG. 6. Hanle effect in a transverse magnetic field (H_x) in the case of modulation of the polarization of the exciting light (1). Curve 2 corresponds to superposition of an additional alternating (5 kHz) field H_{gx} . In the upper corner is shown the frequency dependence of ρ following application of H_{gx} and H_x = 37 Oe.



FIG. 7. Hanle effect in a transverse magnetic field at a fixed sign of the polarization of the exciting light. 1-T = 77 °K, 2-T = 100 °K. The arrows show the direction of the variation of ρ following application of the additional field H_{y} . The points towards which the arrows are directed determine the limits of the variation of ρ in the absence of influence of the nuclei. The Lorentz curve (3) corresponds to averaging within these limits.

frequency 5 kHz. The frequency dependence of the heating effect is shown in the upper part of curve 6 for a field $H_x = 37$ Oe. The results shown in Figs. 5 and 6 offer evidence of cooling of the spin system of the lattice nuclei in the oscillating field at relatively high modulation frequency, when the period of the variation of $\langle S \rangle$ is comparable with the time T_2 of the transverse nuclear relaxation ($10^{-4}-10^{-5}$ sec).

To exclude the influence of the nuclei and to realize the case $\beta = 0$ in the case of excitation by light of fixed circular polarization, it is possible to apply along y an alternating field H_{vx} that saturates the nuclear transitions. It is impossible to eliminate the influence of the nuclei completely. As a result, ρ is overestimated in the region of the additional maximum and underestimated in the region of the minimum. We can thus obtain the upper and lower bounds of the half-width $H_{1/2}$ of the pure "electronic" ($\beta = 0$) luminescence-depolarization curve in a transverse magnetic field. Curve 3 of Fig. 7 was obtained by averaging the measured values of $\rho(H_r)$ for the points marked by the arrows under conditions of saturation of the nuclear transitions. Curve 1 corresponds to $H_{yz} = 0$. The same figure shows a plot of $\rho(H_x)$ at ~ 100 °K (curve 2). It is seen that raising the temperature also smoothes out the $\rho(H_x)$ curve, i.e., eliminates the influence of the nuclei.

4. DISCUSSION OF RESULTS

All the obtained experimental dependences confirm the considered model. The characteristic form of the depolarization curves in a transverse field, including the narrow line near H=0, the additional maximum and the fall-off that follows it, the strong asymmetry in the case of small rotation of the field H, the opposite signs of ρ when an alternating field H_{y-} is turned on at different H_x —all these follow from the model with cooling of the nuclear spin system in the field of opticallyoriented electrons with positive g-factor. In the experiment, the ratio $\rho/\rho_0 = \chi = 1$ is not reached in the additional maximum (see Fig. 1b). This is probably due



FIG. 8. Narrow line (near $H_x = 0$) of the Hanle curve, plotted in coordinates H_x and $((1 - \chi)/\chi^3)^{1/2}$.

to the spatial inhomogeneity of the spin density in the excitation volume inasmuch as, first, the intensity in the laser beam is not uniformly distributed, and second, the diffusion of the oriented electrons leads to the appearance of a gradient of $\langle S_{z} \rangle$ directed normal to the surface of the crystal. This results in a spatial inhomogeneity of the parameter β , and the experimentally observed relations correspond to averaging of β over the luminescent body. In the calculation presented above we considered a lattice consisting of nuclei of one sort. In the experiments, we investigated a crystal containing four isotopes. Exact expressions for $\langle I \rangle$, Θ and H_L for this case are given in the paper of D'yakonov and Perel'.^[7] The parameters H_L , h_N , and h_e given below and characterizing the influence of the nuclear spin system correspond to averaging over the volume and over the nuclei of the isotopes Ga⁶⁹, Ga⁷¹, Al²⁷, and As⁷⁵.

Figure 8 shows a section of the narrow line near $H_x = 0$, plotted in the coordinates H_x and $\eta = [(1-\chi)/\chi^3]^{1/2}$. The fact that the plot is almost linear confirms the possibility of describing the narrow line by Eq. (11). Substituting in (12) the values of H_M , $H_{1/2}$, and $\partial H/\partial \eta$, determined from the data of Figs. 7 and 8, we get $H_L = (4.2 \pm 0.6)$ Oe. In this case $\alpha = (4.9 \pm 0.9) \cdot 10^{-2}$ and $\beta = 15 \pm 2$. By varying the degree of polarization of the exciting beam, its intensity, etc., we can vary the effective field of the electrons at the nuclei and consequently change β . Experiment reveals a shift of the additional maxima with changing intensity. A correlation between H_M and $\langle S_e \rangle_0$ is also observed.

Figure 9 shows plots of $\rho(H)$ at $\varphi = 2^{\circ}$ for two values of $\langle S_{z} \rangle_{0}$, demonstrating this correlation. The quantity $(h'_{N})_{1}$, determined from the asymmetry of the additional maxima (see (12)), amounted to $(1.5 \pm 0.3) \times 10^{4}$ Oe.

So far we have disregarded the influence of the additional relaxation channels of the nuclear spins, which leads to the appearance of a leakage factor^[7] and to a lowering of the maximum attainable value of $1/\Theta$. The leakage can be taken into account by introducing the parameters β and δ a factor k < 1. This is equivalent to replacing h'_N by kh'_N . The experimentally determined value of $(h'_N)_1$ agrees with kh'_N . In this case $kh'_N \approx 3 \times 10^3$ Oe. A comparison with the calculated maximum nuclear field in the GaAs crystal^[10] shows that k is of the order of 0.1. The field h_e of the optically-oriented electrons can be determined from the relation $h_e = 3H_L^2\beta/$ $kh'_N\langle S_x\rangle^2_{0^*}$. Using the obtained values of H_L , β , and kh'_N , we obtain $h_e = (5.4 \pm 1.7)$ Oe. The additional field of the nuclei $H_N = h_N\langle I_x \rangle$ is directed opposite to the external field H_x . At $H_x < H_M$ we have $|H_N| > |H_x|$ and at $H_x > H_M$ we have $|H_N| < |H_x|$. The spin temperature of the nuclei can be determined from the experimental values of the field H_N :

$$\Theta = \Theta_{\rm M} H_{\rm x}/H_{\rm N}, \quad \Theta_{\rm M} = h_{\rm N} (I+1) \mu_{\rm I}/3.$$

The quantity $\Theta_{\rm M}$ corresponds to the spin temperature at $H_x = H_{\rm M} = H_N$ and its value is $(1-2) \times 10^{-3}$ °K. We have assumed h_N equal to 3×10^4 Oe. Θ decreases by almost one order of magnitude at $H \approx H_L$. In z zero external field we can estimate $\Theta = \Theta_0$ from the formula

$$\Theta_0 \approx 3\mu_I H_L^2 / 4Ikh_e \langle S_z \rangle_0^2$$

which describes cooling in a field $h_e \langle S_z \rangle_0 \ll 3^{1/2} H_L$. The order of magnitude of Θ_0 is $10^{-4} \,^{\circ}$ K. The average spin of the nuclei at H = 0 is

$$\langle I_z \rangle_0 = 4Ik(I+1)h_e^2 \langle S_z \rangle_0^3/9H_L^2 \approx 3 \cdot 10^{-4}.$$

Thus, the spin system of the lattice nuclei becomes cooled by light in a zero external magnetic field, and an appreciable nuclear polarization is produced.

We note that the small value of the electron field H_e $= h_{o}(S) < 1$ Oe influences substantially the function $\rho(H)$ in strong external fields $H_x \gg 1$ Oe. The ratio of fields H_e and H_N depends on the ratio of the concentrations n_e and n_{nuc} of the oriented electrons and of the nuclei, with $h_e/kh_N = (n_e/n_{nuc}) (g^* \mu_B/\hbar \gamma_I)$. This ratio can be obtained by expressing the energy of the hyperfine interaction in terms of the field $H_e = h_e S$ and in terms of the field H_N = $kh_N \langle I \rangle$. Substituting the experimentally obtained values of h_e and kh_N , we obtain $n_{\rm nuc}/n_e \approx 2 \times 10^5$. For g^* we assumed a value + 0.5.^[14] Under stationary conditions we have $n_e = GT$, where G is the number of electrons generated per unit time and $T = \hbar/g^* \mu_B H_{1/2} \approx 0.7 \cdot 10^{-8}$ sec. If the power of the sharply-focused laser beam is 40 mW and the absorption coefficient is large (10^4 cm^{-1}) , n_e reaches a value 10¹⁶ cm⁻³. The concentration $n_{\rm nuc}$ of the oriented nuclei that contribute to the effective field H_N amounts to 2×10^{21} cm⁻³, which is smaller by one order of magnitude than the number of nuclei per unit



FIG. 9. Hanle effect (σ_z) at a small inclination of the magnetic field $(\varphi = 2^{\circ})$ for two values of $\langle S_z \rangle_0$.

volume. This result may be due to the spatial inhomogeneity of the spin density, due both to the distribution of the intensity of the exciting light in the semiconductor volume and to the possible localization of the oriented electrons.

We note the strong temperature dependence of the observed effect when the temperature T_0 is raised from 77 °K to ~ 100 °K. In this range, the electron spin-relaxation time is $\tau_S \sim T_0^{-n}$, where n = 2-3.^[14,15] If $\langle S_z \rangle_0$ is small, so that $\langle S_z \rangle_0 = 0.25T/\tau \approx 0.25\tau_S/\tau$, then β ~ $\langle S_z \rangle_0^2 \sim T_0^{-4} - T_0^{-6}$. The experimentally observed (Fig. 7) vanishing of the structure of the $\rho(H_x)$ curve when T_0 changes from 77 °K to ~ 100 °K can be attributed to the abrupt decrease of β . At $T_0 = 4.2$ °K, the plots of $\rho(H_x)$ do not differ qualitatively from those observed at 77 °K.

We have not considered here transient processes or a number of dynamic effects (excitation by intermittent light, application of a pulsed magnetic field, etc.) which indicate that the stationary values of the polarization take a long time to reach the steady state. A quantitative interpretation of these results calls for an additional analysis. In particular, it is necessary to explain the difference between the half-widths of the $\rho(H_x)$ curves when the semiconductor is excited with light with constant and alternating-sign circular polarization.

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¹G. Lampel, Phys. Rev. Lett. 20, 491 (1968).

²A. I. Ekimov and V. M. Safarov, Pis'ma Zh. Eksp. Teor. Fiz. **15**, 257 (1972) [JETP Lett. **15**, 179 (1972)]; V. L. Berkovits, A. I. Ekimov, and V. I. Safarov, Zh. Eksp. Teor. Fiz. 65, 346 (1973) [Sov. Phys. JETP 38, 169 (1974)].

- ³M. I. D'yakonov and V. I. Perel', Zh. Eksp. Teor. Fiz. 65, 362 (1973) [Sov. Phys. JETP **38**, 177 (1974)]; L. L. Buishvili, N. P. Georgadze, and A. I. Ugulava, Fiz. Tverd. Tela (Leningrad) **16**, 3043 (1974) [Sov. Phys. Solid State **16**, 1966 (1975)].
- ⁴V. G. Fleisher, V. L. Vekua, R. I. Dzhioev, and B. P. Zakharchenya, Pis'ma Zh. Eksp. Teor. Fiz. **21**, 547 (1975) [JETP Lett. **21**, 255 (1975)].
- ⁵V. A. Novikov and V. G. Fleisher, Pis'ma Zh. Tekh. Fiz. 1, 935 (1975) [Sov. Tech. Phys. Lett. 1, 404 (1975)].
- ⁶B. N. Provotorov, Zh. Eksp. Teor. Fiz. **41**, 1582 (1961)
 [Sov. Phys. JETP **14**, 1126 (1962)]; M. Goldman, Spin Temperature and NMR in Solids, Oxford, 1970; V. A. Atsarkin and M. I. Podak, Usp. Fiz. Nauk 107, 3 (1972) [Sov. Phys. Usp. **15**, 1 (1972)].
- ⁷M. I. D'yakonov and V. I. Perel', Zh. Eksp. Teor. Fiz. **68**, 1514 (1975) [Sov. Phys. JETP **41**, 759 (1975)].
- ⁸M. I. D'yakonov, V. I. Perel', V. L. Berkovits, and V. I. Safarov, Zh. Eksp. Teor. Fiz. **67**, 1912 (1974) [Sov. Phys. JETP **40**, 950 (1975)].
- ⁹B. P. Zakharchenya, M. E. Kompan, and V. G. Fleisher, Pis'ma Zh. Eksp. Teor. Fiz. **19**, 734 (1974) [JETP Lett. **19**, 378 (1974)].
- ¹⁰D. Paget, Thèse de 3e cycle, Orsay, 1975.
- ¹¹S. N. Jasperson and S. E. Schnatterly, Rev. Sci. Instrum. 40, 761 (1969).
- ¹²A. P. Balatsyuk, V. A. Novikov, and V. G. Fleisher, Prib. Tekh. Eksp. No. 1, 174 (1976).
- ¹³V. G. Fleisher, R. I. Dzhioev, and B. P. Zakharchenya, Pis'ma Zh. Eksp. Teor. Fiz. 23, 22 (1976) [JETP Lett. 23, 18 (1976)].
- ¹⁴A. H. Clark, R. D. Bernham, D. J. Chadi, and R. M. White, Phys. Rev. [B] **12**, 5758 (1975).
- ¹⁵D. Z. Garbuzov, A. I. Ekimov, and V. I. Safarov, Pis'ma Zh. Eksp. Teor. Fiz. **13**, 36 (1971) [JETP Lett. **13**, 24 (1971)].

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Magnetization precession in superfluid phases of He³

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Solutions of the Leggett equations are obtained which describe the motion of magnetization in both superfluid phases of He^3 located in a strong dc magnetic field. Rotation of magnetization by an ac magnetic field is described. The results are compared with available experimental data.

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1. INTRODUCTION

Progress in the understanding of the nature of the superfluid phases of He³ is due to a considerable degree to the study of the properties of these phases by the NMR method. The corresponding experiments lend themselves to a quantitative interpretation with the aid of the system of equations for spin dynamics in triplet pairing, obtained by Leggett^[11]:

$$\dot{\mathbf{S}} = \gamma [\mathbf{S} \times \mathbf{H}] + \mathbf{R}_{\mathcal{D}}(\mathbf{d}), \tag{1}$$

$$\mathbf{d} = [\mathbf{d}(\mathbf{n}) \times \gamma (\mathbf{H} - \gamma \mathbf{S}/\chi)].$$
⁽²⁾

Here **H** is the external magnetic field, **S** is the total spin of the considered amount of helium, χ is its magnetic susceptibility, γ is the gyromagnetic ratio for the He³ nuclei, and **d** is a vector in spin space and characterizes the spin structure of the wave function of the condensate. Its exact determination (see^[21], p. 367), will not be needed here. When the pairing is in the *p* state, as is the case in He³, **d** depends linearly on the

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