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

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Optical orientation of nonequilibrium electrons in $Ga_xAl_{1-x}As$ crystals is investigated at 4.2 °K. Spin orientation of electrons is detected by the polarization of the donor-acceptor recombination radiation. It is shown that the spin-relaxation time and the degree of orientation of the electrons captured by the donor significantly increase if an external longitudinal magnetic field is applied. Dynamic polarization of the nuclei by electrons induces an effective magnetic field due to hyperfine interaction, which acts on the electron spin. It is shown that the nuclear field weakens or intensifies the effect of the external field (depending on their mutual orientation) and thus affects the electron spin orientation and polarization of recombination luminescence. Nuclear magnetization relaxation processes and the longitudinal nuclear relaxation time are determined for nonstationary conditions on the basis of variation of luminescence polarization. Variations in luminescence polarization under resonance conditions for nuclei of the crystal proper lattice, which are due to variation in the magnitude and direction of the nuclear field, are investigated. The line shape of optically detected nuclear resonance on adiabatically rapid passage, slow passage, or saturation of resonance is analyzed in detail. The experimental data are in good agreement with the theory.

1. INTRODUCTION

The method of optical orientation of the spins of non-equilibrium electrons has recently begun to be widely applied in the study of semiconductors[1-5]. In these experiments, spin-oriented electrons are excited from the valence band to the conduction band as a result of absorption of circularly polarized light. Since the oriented electrons produced by the light also emit polarized light on recombination, a study of the polarization of the recombination luminescence makes it possible to determine the degree of orientation of the nonequilibrium electrons. The luminescence polarization measured in these experiments reflects the stationary orientation of the electrons which develops in the lifetime of a nonequilibrium electron as a consequence of the different spin relaxation processes and spin interactions. Therefore, the method of optical orientation can be applied successfully to the study of all these processes.

Of interest is the case when the electrons interact effectively with the nuclei. In the very first paper devoted to optical orientation in semiconductors[1] it was discovered that, as a result of the hyperfine interaction, the oriented electrons induce strong dynamic polarization of the nuclei of the proper crystal lattice. Experiments have been performed on silicon and the increase in the polarization of the ^{29}Si nuclei on optical pumping of polarized electrons was detected by radiospectroscopic methods (from the increase in the nuclear magnetic resonance signal).

Later, it was shown by two of the authors of the present article[6,7] that the resulting nuclear magnetization exerts, in its turn, a strong influence on the orientation of the electrons. Thus, the possibility arises of detecting the change of nuclear polarization by a purely optical method, by observing the orientation of the electrons. In particular, for crystals of $Ga_xAl_{1-x}As$ solid solutions, it has been possible[6,7], using the variation of the luminescence polarization, to register the change of the nuclear magnetization under conditions of nuclear magnetic resonance at all the nuclei of the proper lattice: ^{75}As, ^{69}Ga, ^{71}Ga and ^{27}Al.

Up to the present time there have been two theoreti-

cal papers, by D'yakonov and Perel'[8,9], in which electron-nuclear interaction processes during optical pumping of polarized electrons are considered. In the first of these[8], a particular model was considered—the interaction of an oriented electron, captured by an impurity center (donor), with the nucleus of this center. Within the framework of this model it was already possible to give a qualitatively correct description of the phenomena observed in experiment[6,7]. However, the second model[9], which takes into account the interaction between an oriented electron captured by a donor and the host-lattice nuclei lying in the region of localization of the electron, corresponds more exactly to the experimental conditions.

In this paper, an experimental study of electron-nuclear interaction processes in semiconductors is performed on the basis of the theoretical ideas developed by D'yakonov and Perel'[9]. In particular, the effect of a longitudinal (parallel to the orientation of the spins) external magnetic field on the electron spin relaxation is studied. The transition processes establishing the nuclear magnetization when the nuclei are polarized by the oriented electrons are studied by an optical method. The line-shape of the optically detected nuclear resonance is analyzed in detail.

2. EXPERIMENTAL METHOD

Crystals of $Ga_xAl_{1-x}As$ were used for the investigation, the composition ($x < 0.3$) being chosen such that the band gap was somewhat less than the energy of a quantum of radiation from a He-Ne laser. To establish a preferred orientation of the spins of the nonequilibrium electrons, linearly polarized (π) radiation from the laser was transformed by means of a $\lambda/4$-plate into circularly polarized radiation with right (σ^+) or left (σ^-) rotation. Numerically, the preferred spin orientation obtained can be characterized by the average spin S of the electrons:

$$S = \frac{1}{2}\frac{n_+ - n_-}{n_+ + n_-}, \qquad (1)$$

where $\frac{1}{2}$ is the spin of one electron and $n_\pm$ is the number of electrons with spin orientation along or opposite

to the direction of propagation of the exciting light. The initial orientation of the electrons produced by the light is determined by the ratio of the probabilities of interband transitions to states with $m = +\frac{1}{2}$ and $m = -\frac{1}{2}$ in the conduction band. For crystals with band-structure of the GaAs type (the mixed crystals studied, with $x < 0.3$, have an analogous band scheme), the selection rules give[4] $S = 0$ for excitation by π-polarized light and $S_0 = \pm 0.25$ for excitation by $\sigma^{\pm}$ circularly polarized light (the sign $\pm$ for S_0 means that, on changing the polarization of the exciting light from σ^+ to σ^-, the direction of the preferred orientation of the spins is reversed).

During the lifetime of the electrons before recombination, the orientation of their spins can change as a result of different spin relaxation processes. The stationary value of S in this case will be determined by the ratio of the lifetime τ of the electron and its spin relaxation time τ_S:

$$S = \frac{S_0}{1 + \tau/\tau_s}. \qquad (2)$$

The luminescence polarization will reflect this stationary orientation of the electrons. The luminescence polarization is characterized by the degree of polarization ρ, which is defined as

$$\rho = \frac{J_+ - J_-}{J_+ + J_-}, \qquad (3)$$

where $J_{\pm}$ is the luminescence intensity in the σ^+ and σ^- polarizations. The selection rules for the reverse interband transitions (in emission) give a simple relation connecting the degree of polarization of the luminescence and the average spin of the electrons[4]:

$$S = \rho. \qquad (4)$$

Thus, a measurement of the luminescence polarization makes it possible to determine immediately the average spin of the oriented electrons. Observed variations of the degree of polarization of the luminescence reflect changes in the lifetime and spin relaxation time of the nonequilibrium electrons[3].

Thus, the optical orientation experiment consists in studying the polarization of the recombination luminescence of the crystal when it is excited by circularly polarized light. To measure ρ we used a special modulator—a rotating $\lambda/4$ plate. In passing through the $\lambda/4$ plate, the circularly polarized light is transformed into linearly polarized light, with mutually perpendicular directions for the σ^+ and σ^- polarizations. Placed behind the $\lambda/4$ plate was a polaroid which, on rotation of the plate, separated out the σ^+ and σ^- polarized light alternately. If the luminescence radiation is partly polarized ($J_+ \neq J_-$), the light that has passed through the polaroid will be modulated in intensity with an amplitude equal to $J_+ - J_-$. The phase of the signal is reversed, in accordance with the change of sign of S, when the polarization of the exciting light changes from σ^+ to σ^-; the signal vanishes for excitation by π-polarized light.

In addition to this modulator, working at a frequency of 73 Hz, we also used an ordinary disk chopper with frequency 600 Hz, which fully modulated the light. Thus, two alternating signals were distinguished at the photomultiplier: one at frequency 73 Hz, proportional to $J_+ - J_-$, and the second at frequency 600 Hz, proportional to $J_+ + J_-$. By measuring these signals after separate selective amplification and synchronous detection, it was possible to determine the degree of polarization of

the luminescence (cf. (3)). A block diagram of the setup is shown in Fig. 1.

For the investigation under nuclear magnetic resonance (NMR) conditions the sample was placed in a magnetic field, longitudinal with respect to the direction of propagation of the exciting light[1]. An alternating radio-frequency (RF) field was created by means of a coil. To detect the resonance, scanning of both the RF-field frequency and the static field intensity H_0 was used.

The crystals investigated were layers of solid solutions, grown by the method of liquid epitaxy. The samples were of the p-type, with hole concentration $N_p \sim 10^{16}\,cm^{-3}$ at room temperature. All the experiments were performed at the temperature of liquid helium.

3. EXPERIMENTAL RESULTS AND DISCUSSION

In the weakly alloyed crystals studied, as in all p-type samples, the dominant band in the luminescence spectrum at $4.2°K$ is that due to transitions to the acceptor level. The earlier investigations[11] of the shift in the band maximum with increasing excitation intensity and temperature, and also of the temperature dependence of the luminescence intensity and of the lifetime of the nonequilibrium electrons showed that, at $4.2°K$, the band is due to emission transitions from donor to acceptor. In a measurement of the Hanle effect in the given crystal[11], it was found that a nonequilibrium electron captured by a donor has a fairly long lifetime: $\sim 10^{-8}$ sec.[2] However, in a study of the optical orientation[6], it was found that, in ordinary conditions, the spin orientation of the nonequilibrium electrons at the donors is small: $S = 1\%$. This shows that the spins of the electrons captured by the donor relax very rapidly, with $\tau_S \approx 10^{-9}$ sec.

For the spin relaxation of electrons captured by a donor, D'yakonov and Perel'[9] considered a mechanism due to the action of random local magnetic fields. These local fields arise as a result of the exchange interaction of the electron with the holes surrounding it or as a consequence of the hyperfine interaction with the nuclear magnetic moments of the proper lattice that lie in the region of localization of the electron. From the estimates of D'yakonov and Perel', these fields can attain a magnitude of the order of several hundred gauss. However, the local field acting on the electron spin does not remain constant in time, but varies as a result of, e.g., the spin relaxation of the holes or electron hops from donor to donor by tunneling[3].

A. Effect of an external logitudinal magnetic field

According to D'yakonov and Perel'[9], the application of a sufficiently strong external magnetic field directed along the orientation of the spins (i.e., along the direction of propagation of the exciting light) should lead to a substantial slowing-down of the spin relaxation due to the local fields. Thus, in a longitudinal field, the time τ_S should increase, and this, in its turn, according to (2), will lead to an increase in the magnitude of the average spin S of the nonequilibrium electrons.

We have traced the variation of the stationary orientation of the nonequilibrium electrons in an external longitudinal magnetic field up to intensities $H_0 = 20$ kG. The resulting dependence $S(H_0)$ is shown in Fig. 2. It can be seen that the orientation of the electrons is in-
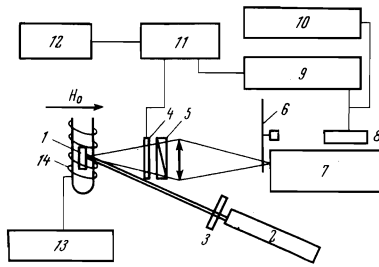
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FIG. 1. Block diagram of the experimental setup. 1—Sample; 2—He-Ne laser; 3—λ/4-plate; 4—rotating λ/4-plate; 5—polaroid; 6—disk modulator; 7—DFS-12 spectrograph; 8—photomultiplier; 9,10—selective amplifiers, tuned to 73 and 600 Hz respectively; 11—synchronous detector; 12—automatic recorder; 13—radio-frequency generator; 14—induction coil.
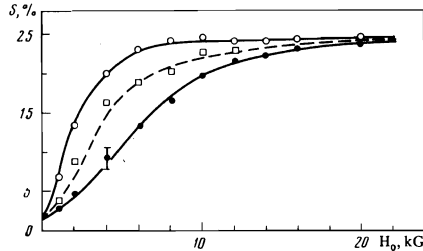

FIG. 2. Dependence of the spin orientation of the electrons on the intensity of the external longitudinal magnetic field H_0. The points O and ● correspond to the stationary state of orientation on excitation by σ^+ and σ^- polarized light (with a constant orientation of H_0). The values of the points □ are obtained from the transition processes establishing the stationary orientation (see the text). All the lines are drawn in accordance with the theoretical dependence $S(H_0)$ (formulas (2) and (6)) with the choice $\tau/\tau_S(0) = 24$ and $H_\gamma = 600$ G. The dashed line corresponds to the case $H_N = 0$. The solid lines (for the case $H_N \neq 0$) are obtained assuming $H_N(G) = 168$.

deed found to be strongly dependent on H_0, and the magnitude of the average spin of the electrons increases from 0.01 to 0.25 on increase of H_0 from 0 to 20 kG. This strong increase of S up to the maximum values shows that the localized-electron spin relaxation mechanism considered in[9], arising from the action of random magnetic fields, is the decisive mechanism in the case that we have investigated.

From the dependence $S(H_0)$ shown in Fig. 2, it can be seen that, in intermediate fields $0 < H_0 < 20$ kG, the orientation of the electrons differs strongly for the two (σ^+ and σ^-) polarizations of the exciting light, for a constant orientation of the external field H_0. The σ^+ and σ^- exciting light creates in the crystal oriented electrons with opposite spin directions (along or against the direction of propagation of the exciting light), and the observed asymmetry implies that the magnitude of the average spin of the oriented electrons depends strongly on whether the spin of the electrons is oriented along or against the direction of the external field.

This explanation is also confirmed by the fact that completely analogous differences in the values of S were also observed using the same polarization of the exciting light but reversing the direction of the external magnetic field.

The observed asymmetry can be explained[9] if we take into account that, because of the hyperfine interaction, the oriented electrons produced by the light strongly polarize the nuclei. Then, as a result of the same hyperfine interaction, the polarized nuclei[9] create at the electron a certain regular magnetic field H_N, the in-

tensity and orientation of which are determined by the polarization of the nuclei:

$$H_N \sim \sum_n |\psi(r_n)|^2 A_n \langle I_n \rangle, \qquad (5)$$

where $\psi(r_n)$ is the value of the electron wavefunction at the site of the n-th nucleus, $\langle I_n \rangle$ is the average value of the nuclear spin, due to the polarization of the nuclei, and A_n is the hyperfine interaction constant, which is the same for nuclei of the same type. The direction of this field is determined by the direction of the preferred orientation of the nuclear spins and clearly coincides with the orientation of the electron spins.

As a result, for one of the polarizations of the exciting light, when the spin orientation of the electrons produced by the light is parallel to the external field ($S \uparrow\uparrow H_0$), the nuclear field adds to the external field and the electron finds itself in an effective field greater than the external field. For the other polarization of the exciting light, when the electron spins are antiparallel to the external field ($S \uparrow\downarrow H_0$), the effective field at the electron is smaller than the external field. Since the electron spin relaxation rate τ_S^{-1} depends on the magnitude of the magnetic field acting on the electron, according to (2) the magnitude of the average electron spin S will differ for the cases described above: in Fig. 2, the upper curve corresponds to electrons in the effective field $H_{eff} = H_0 + H_N$, and the lower curve to electrons in the field $H_{eff} = H_0 - H_N$.

This interpretation of the observed dependence $S(H_0)$ is confirmed by investigations of the transition processes establishing the stationary state after the exciting light is switched on. If the nuclei are unpolarized[4], then, immediately after the circularly polarized exciting light is switched on, the oriented electrons produced by the light are in a field $H_{eff} = H_0$, since for depolarized nuclei $\langle I \rangle = .0$ and $H_N = 0$. As nuclear magnetization is gradually created as a result of polarization of the nuclei by the electrons, a nuclear field H_N appears and increases, and the effective field at the electron will be changed. Since the electron spin relaxation rate and, correspondingly, the magnitude of the average electron spin depend on H_{eff}, the observed luminescence polarization should reflect these changes of H_N and H_{eff}.

Experimentally, these processes are observed directly from the change in time of the signal $J_+ - J_-$ after the circularly polarized exciting light is switched on. The signal was recorded directly on the roll of an
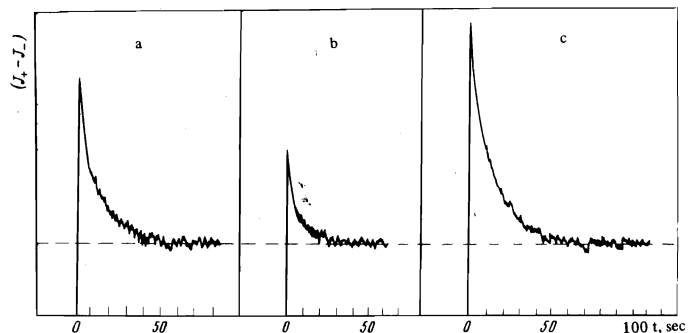

FIG. 3. Establishment of the stationary signal $(J_+ - J_-)$ when σ-polarized light is switched on. The geometry is $S\uparrow\downarrow H_0$, and $H_0 = 3.6$ kG. a—The nuclei are preliminarily polarized; b—before the light is switched on, passage through the resonance of the ^{71}Ga nuclei was effected in darkness; c—passage through the resonances of all the nuclei of the proper lattice was effected in darkness.

automatic recorder. The investigations were carried out in the antiparallel geometry S $\uparrow\downarrow$ H$_0$, since in the other case (the parallel geometry S $\uparrow\uparrow$ H$_0$) the transition processes developed much more rapidly and it was difficult to register them with the technique applied (the time-constant of the instrument was ~0.5 sec).

Figure 3 shows the observed variation of the signal J$_+$ − J$_-$ when σ-polarized light is switched on. As can be seen, at the initial moment the electron orientation is large [(J$_+$ − J$_-$) ~ ρ ~ S, cf. (3), (4)], and then falls fairly slowly (in the present case, with a time-constant of 20 sec) to its stationary value. For the geometry S $\uparrow\downarrow$ H$_0$, this behavior of S obviously corresponds to a decrease of the effective field at the electron, from the value H$_{eff}$ = H$_0$ at the initial time to the stationary value H$_{eff}$ = H$_0$ − H$_N$. The time for the polarization to fall characterizes the time for establishment of the stationary nuclear magnetization.

The small squares in Fig. 2 mark the electron average-spin values corresponding to the maximum "kick" J$_+$ − J$_-$ in such transition processes, for different values of the external field H$_0$. The curve obtained is, obviously, the dependence of the degree of orientation of the electrons on the effective field H$_{eff}$ = H$_0$ (for depolarized nuclei, H$_N$ = 0). It can be seen that these points fall between the stationary values, which are determined by the electron orientation in the fields H$_{eff}$ = H$_0$ ± H$_{N,st}$.

The stationary nuclear magnetization (and, consequently, the field H$_N$) can also be changed significantly by applying an alternating RF field under NMR conditions. The corresponding changes of the field H$_N$ (and of the field H$_{eff}$) can also be detected by observing the transition processes. The largest change of H$_N$ is attained in adiabatically fast passage through the resonance[13], which reverses the nuclear magnetization (i.e., the direction of the field H$_N$ is reversed).

The experiment was set up in the following way. For a given value of the field H$_0$, the stationary polarization was established on excitation by circularly polarized light. The exciting light was then switched off and passage through the resonance was effected in darkness using scanning of the RF-field frequency. The instantaneous value of the degree of polarization of the luminescence when the exciting light was again switched on reflected the change of H$_N$ (and, correspondingly, of H$_{eff}$) attained. Figure 3 shows the observed increase, for the geometry S $\uparrow\downarrow$ H$_0$, of the degree of polarization of the luminescence in the case of passage through the resonance of one nucleus (^{71}Ga) (Fig. 3b) and through the resonances of all four nuclei of the proper lattice: ^{75}As, ^{69}Ga, ^{71}Ga and ^{27}Al (Fig. 3c). The latter case clearly corresponds to a change of the effective field from H$_{eff}$ = H$_0$ − H$_N$ to H$_{eff}$ = H$_0$ + H$_N$. The decrease then observed of the signal J$_+$ − J$_-$ to its stationary value is due to the relaxation of the field H$_N$ to its stationary value. The experiments performed clearly demonstrate the effect of the nuclear field on the orientation of the electrons produced by the light.

According to D'yakonov and Perel'[9], the dependence of the electron spin relaxation time on the field (which also determines the dependence S(H$_0$)) in the presence of nuclear polarization is determined by the formula

$$\tau_s(H_0) = \tau_s(0)\left[1 + \left(\frac{H_0 \pm \eta N_N}{H_\gamma}\right)^2\right]. \tag{6}$$

Here, H$_\gamma$ = a(gμ$_0$)$^{-1}$γ, where g is the g-factor of the electron, μ$_0$ is the Bohr magneton, γ is the frequency of variation of the local field (in the simplest case, the jump frequency of the electron from donor to donor), H$_N$ is the nuclear field and a and η are coefficients (0.1 < a, η < 1) depending on the specific mechanism of spin relaxation of the localized electron. The curves of Fig. 2 are obtained theoretically using this formula and substituting it into (2). The dashed curve is derived neglecting the polarization of the nuclei, i.e., H$_N$ = 0 and H$_{eff}$ = H$_0$. Good agreement with the experimental values obtained in the study of the transition processes was attained with the choice H$_\gamma$ = 600 G, which corresponds to γ^{-1} ~ 10^{-10} sec. The solid curves are derived with allowance for the polarization of the nuclei (H$_N$ ≠ 0); it was assumed that H$_N$ is proportional to S. Agreement with the experimental points was achieved by choosing the maximum value of the nuclear field H$_N$ = 4000 G (for S = 0.25), i.e., a field of the same order as that given by the theoretical estimates[9].

B. Kinetics of the transition processes

In the transition phenomena described above, the duration of the process of establishing the stationary state is determined by the time T$_{1e}$ of the longitudinal nuclear relaxation due to the interaction of the nuclei with the nonequilibrium electrons. The time T$_{1e}$ can be measured experimentally. If the changes of ρ in the transition processes are small compared with the stationary value ρ$_{st}$ of the polarization and the nuclear field is much smaller in magnitude than the external field (H$_N$ ≪ H$_0$), then, as can be seen from formulas (2) and (6), we can assume that the quantity ρ(t) − ρ$_{st}$ is proportional to H$_N$, i.e., the changes in the luminescence polarization observed in the study of the transition processes directly reflect changes in the magnitude of the nuclear field H$_N$.

Figure 4 shows the changes (obtained from curves analogous to Fig. 3a) in time of the quantity ρ(t) − ρ$_{st}$, for several values of the external field H$_0$. In the chosen semilogarithmic scale the observed dependences are close to the linear ones that should be observed in the case of the exponential law for the variation of the nuclear field:

$$H_N \sim 1 - \exp(-t/T_{1e}).$$

The slope of the straight lines in Fig. 4 characterizes the time T$_{1e}$ in the given field H$_0$.

In Fig. 4, it can be seen clearly that the nuclear relaxation time increases substantially with increase of the external field intensity H$_0$. According to D'yakonov
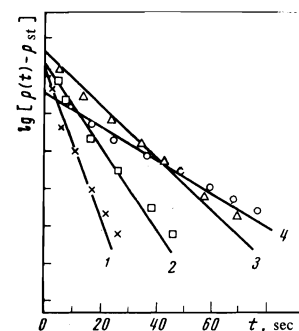

FIG. 4. Variation in time of the luminescence polarization after σ-exciting light is switched on. S$\uparrow\downarrow$H$_0$ and H$_0$ = 3.6 kG. The nuclei were previously depolarized. 1—H$_0$ = 2 kG, 2—4 kG, 3—6 kG, and 4—10 kG.

and Perel'[9], the longitudinal nuclear spin relaxation time T_{1e} due to the interaction with the electrons is given by the formula

$$T_{1e}(H_0) \sim T_{1e}(0)\,[1 \pm (\Omega_e / \gamma)^2], \qquad (7)$$

where $\Omega_e = g\mu H_0$ is the precession frequency of the electron spin in the external field H_0 and γ is the frequency of variation of the local field.

Formula (7) gives a good description of the experimentally observed dependence of the time T_{1e} (measured from the curves of Fig. 4) on the magnitude of the external field H_0 (Fig. 5). From the dependence $T_{1e}(H_0)$ shown in Fig. 5, one can determine the time γ^{-1} of variation of the local field, which is found to be equal to $\sim 3 \times 10^{-11}$ sec, and the nuclear relaxation time in zero field ($T_{1e} = 14$ sec).

The longitudinal nuclear relaxation time T_{1e} can be measured also in an investigation of the processes of decay (vanishing) of the nuclear magnetization. If, e.g., the polarization of the exciting light is changed to linear after the stationary state is established, the unoriented electrons produced by the π-polarized light will induce the depolarization of the nuclei. The nuclear depolarization time was measured experimentally in the following way: after establishment of the stationary orientation during σ excitation, the polarization of the exciting light was changed to linear for a certain time and then σ-polarized light was switched on again. If noticeable depolarization of the nuclei occurs during the period of irradiation by π-polarized light, this will induce a corresponding change of H_N and H_{eff}. Therefore, when σ-polarized light is switched on a second time, the degree of polarization of the luminescence at the initial moment will differ from the stationary value ρ_{st} (and will then relax to ρ_{st}). The magnitude of the deviation is determined by the extent of the change of H_N and, consequently, will increase with increase in the time of irradiation by π-polarized light, tending to a certain maximum value corresponding to complete depolarization of the nuclei ($H_N \rightarrow 0$).

Figure 6 shows the superposition, for the antiparallel geometry $S \uparrow\!\!\downarrow H_0$, of the resulting transition processes corresponding to different periods of irradiation by π-polarized light. It can be seen that the readings increased monotonically, and from their envelope, which satisfies the dependence

$$\rho - \rho_{st} \sim 1 - \exp(-t/T_{1c}),$$

we can determine the time T_{1e}, which in the present case is 19 sec.

The nuclear depolarization time in darkness (in the absence of nonequilibrium electrons) T_1 can be measured completely analogously. In this case, instead of changing the polarization, we simply switch off the exciting light for a certain time and record the change of ρ relative to its stationary value when the light is switched on again. The longitudinal nuclear relaxation time T_1 measured in this way in the absence of photoelectrons produced by the light was found to be fairly large and amounted to ~ 35 min in a field $H_0 = 3.6$ kG.

If the magnetic field is switched simultaneously each time the light is switched off and then switched on again, it is possible to trace by an analogous method the decay in a small field (and even in zero field) of the nuclear magnetization created during the irradiation of the crystal by σ-polarized light in the large field H_0. The
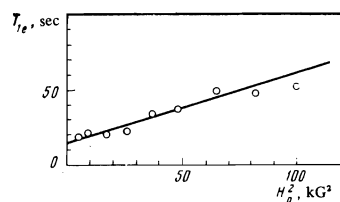

FIG. 5. Dependence of the longitudinal nuclear relaxation time T_{1e} on the intensity of the external longitudinal field H_0. $S \uparrow\!\!\downarrow H_0$.
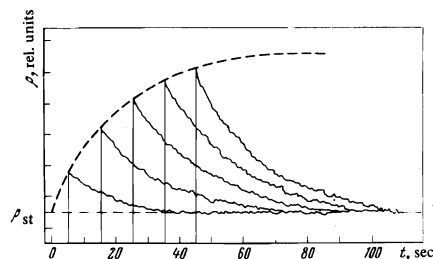

FIG. 6. The transition processes establishing the stationary luminescence polarization when σ-exciting light is switched on; each process corresponds to a different period of preliminary irradiation of the crystal by π-polarized light. $S \uparrow\!\!\downarrow H_0$. The curves $\rho(t)$ are spaced out along the time axis in accordance with the duration of the irradiation by π-polarized light. $H_0 = 3.6$ kG.

measurements showed that the longitudinal nuclear relaxation time T_1 decreases sharply in weak fields. Thus, for $H_0 = 0$ the time $T_1 \approx 16$ sec. With increasing H_0, the time increased rapidly, and in a field $H_0 = 100$ G, $T_1 \approx 60$ sec, while in a field $H_0 = 1000$ G, $T_1 \approx 10$ min.

C. Spin orientation of the electrons under conditions of resonance at the nuclei of the proper lattice

The model considered above for the effect of the nuclear field on the spin orientation of an electron receives further confirmation in the study of the phenomena occurring during NMR at the nuclei of the proper lattice. As already reported previously[6,7], the change of polarization of the nuclei under NMR conditions leads to a decrease of the degree of orientation of the electrons, the optically observed[6] (from the change in the luminescence polarization) NMR line having a doublet structure. Further investigations have established that the shapes of the observed lines depend strongly on the experimental conditions for detection of the resonance: the scanning rate, the power of the RF field, etc. Figure 7 shows one of the optically recorded NMR spectra, obtained in the antiparallel geometry $S \uparrow\!\!\downarrow H_0$ with rapid scanning of the RF-field frequency. Resonances of all the magnetic nuclei of the proper lattice of the $Ga_xAl_{1-x}As$ solid solutions investigated appear in the spectrum. As can be seen from Fig. 7, both a decrease and an increase of the degree of polarization of the luminescence are observed in the region of the resonances, and the lines have the form of the derivatives even though no modulation of the external field H_0 or RF-field frequency was applied in the detection.

The observed changes of ρ can be explained if we consider the behavior in NMR conditions of the effective field H_{eff}, which is the vector sum of the external field H_0 and the fields created by the different types of nuclei: H_α, H_β, H_K and H_η. Away from resonance, all these vectors are collinear (Fig. 8a)—this is ensured by the experimental conditions, since the exciting light propagates along the external field and the spins of the
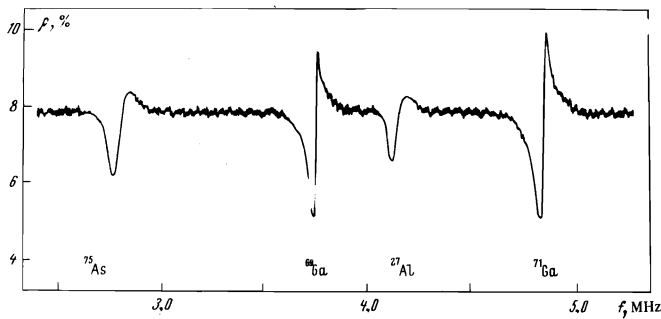
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FIG. 7. NMR spectrum of a crystal of $Ga_{0.7}Al_{0.3}As$, detected by the change of luminescence polarization. $S\uparrow\downarrow H_0$ and $H_0 = 3.6$ kG. The scanning of ω was performed from lower to higher frequencies.
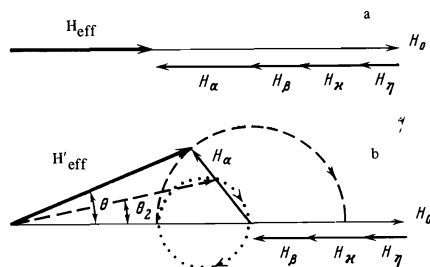

FIG. 8. Diagram of the variation of the effective field H_{eff} in NMR conditions. a—Away from the resonance; b—in the resonance region of one of the nuclei. The dashed line describes the motion of the tip of the nuclear magnetization vector during adiabatically fast passage through the resonance. The dotted line shows the same for slow passage, when the stationary state is established for each value of ω.

electrons produced by the light (and of the nuclei polarized by these electrons) are collinear with H_0. But under resonance conditions for any of the nuclei (e.g., those of type α), because of the appearance of a transverse component of magnetization for these nuclei the corresponding field will no longer be collinear with H_0 but will be oriented at a certain angle to H_0. Correspondingly, the resultant field H_{eff} will also be turned through a certain angle θ relative to its original direction (Fig. 8b). As a result, the oriented electrons will lie in a field H'_{eff} directed at an angle θ relative to their spin orientation. In this case, it can be shown easily[9] that for sufficiently large intensities H_{eff} the magnitude of the average electron spin decreases to the value $S' = S\cos^2\theta$. Moreover, under resonance conditions a change in the absolute magnitude of the field H_{eff} also occurs, corresponding to the rotation of the field H_α (and its possible decrease in magnitude) (Fig. 8b). This induces a corresponding change in the spin relaxation rate and in the magnitude of the average electron spin (cf. subsection A). For the antiparallel geometry $S\uparrow\downarrow H_0$ and resonance at one of the nuclei, the field H_{eff} can only increase in magnitude (Fig. 8b), and under resonance conditions this should give rise to a corresponding increase of S (and ρ).

Thus, for the antiparallel geometry $S\uparrow\downarrow H_0$ under resonance conditions, a rotation of the effective field H_{eff} will cause a decrease of the average spin S of the electrons, while a change in the absolute magnitude of H_{eff} will give rise to an increase of the average spin. The pattern actually observed for the change of the luminescence polarization will be determined by the combined effect of both factors.

Using these considerations and knowing the behavior of the field H_α in resonance conditions, we can give a

qualitative explanation of all the observed changes of the luminescence polarization[5]. The nature of the change of the nuclear field H_α in NMR conditions has been considered by D'yakonov and Perel'[9] on the basis of the concept of the existence of a spin temperature in the rotating coordinate frame[13].

The spectrum of Fig. 7 was obtained with fairly rapid scanning of the RF-field frequency ω, during which the condition for adiabatically fast passage was fulfilled[13]: the time of passage through the resonance was short compared with the longitudinal nuclear relaxation time T_{1e}. In this case, during the passage through the resonance the nuclear magnetization vector does not have time to change in absolute magnitude and, during the scanning of the frequency ω, the field H_α is simply rotated together with the field H_t acting in the rotating coordinate frame. The dashed line in Fig. 8b describes the motion of the tip of the vector H_α (and, correspondingly, of the tip of the vector H_{eff}) during adiabatically fast passage through the resonance.

The decrease of ρ observed in the spectra of Fig. 7 as the resonance frequency ω_0 is approached is due, clearly, to the rotation of H_{eff} and the increase of the angle θ. In the region of exact resonance ($\omega = \omega_0$), the angle θ is a maximum, and the maximum reduction in the degree of polarization (which is proportional to $\cos^2\theta$) corresponds to this angle. After passage through the resonance frequency, the angle θ decreases as we move away from ω_0 and the degree of polarization increases. Far from resonance, H_{eff} again becomes longitudinal (collinear with H_0), but its absolute magnitude is now greater than its original magnitude by $2H_\alpha$ as a result of the reversal of the magnetization of the nuclei α (as a result of the passage through the resonance, the vector H_α is rotated through 180°). Corresponding to this increase of H_{eff}, the degree of polarization increases, after passage through the resonance, to a value greater than the original value. The field H_α then relaxes to the original state, with the longitudinal relaxation time T_{1e}; correspondingly, the degree of polarization of the luminescence returns to the stationary value ρ_{st}, with the same time T_{1e}[6].

A completely different line shape is obtained in the case of slow passage, when, for each value of the frequency (or external field intensity) in the region of the resonance, the stationary state has time to be established. Experimentally, this is conveniently done point by point: for fixed values of the frequency ω (or H_0), the change of ρ when the RF field is switched on and off is investigated. Figure 9 shows the observed changes of ρ at high RF power in the case of resonance at the ^{71}Ga nuclei. A similar picture was also obtained for the resonances at all the other nuclei.

The observed changes of ρ can also be explained by considering the behavior of H_α and H_{eff} in these conditions. Immediately after the RF field is switched on (at time t_1), the nuclear field is rotated through a certain angle, i.e., is oriented along the field H_t acting in the rotating coordinate frame. Corresponding to this are a rotation of H_{eff} through an angle θ and a decrease of the degree of polarization to a value ρ_1 proportional to $\cos^2\theta$. Then, in a time approximately equal to the longitudinal nuclear relaxation time T_{1e}, the absolute magnitude of H_α decreases to a value determined by the saturation of the resonance. Correspondingly, the angle θ decreases to the value θ_2 (cf.
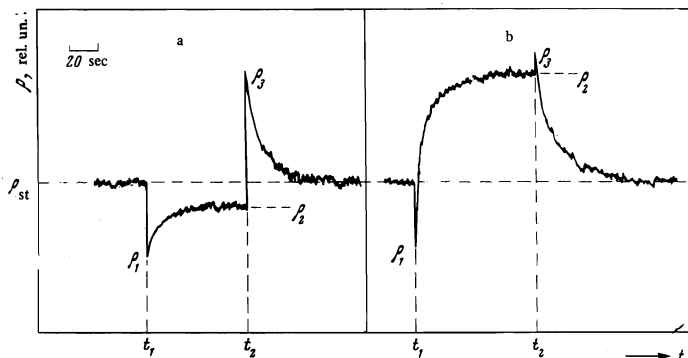
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FIG. 9. Change of the luminescence polarization on switching on (at time t_1) and switching off (at time t_2) of the RF field in the region of resonance at the nuclei ^{71}Ga. S↑↓H$_0$ and H$_0$ = 3.6 kG. a—$\omega \neq \omega_0$; b—$\omega \approx \omega_0$.

Fig. 8b), and the degree of polarization with the RF field switched on increases, with the time T_{1e}, to the stationary value $\rho_2 \sim \cos^2 \theta_2$ determined by the stationary value of $|H_\alpha|$ in the presence of the RF power. When the RF power is switched off (at time t_2), the field H_α (and H_{eff}) will take the original direction (collinear with H_0). But since H_α has decreased in magnitude, H_{eff} will take a value somewhat greater than its initial value, (in accordance with the decrease of $|H_\alpha|$). Therefore, after the RF power is switched off, the degree of polarization increases to a value ρ_3 greater than the initial value and then returns to the stationary value ρ_{st}, with a characteristic time T_{1e} and commensurately with the relaxation of the nuclear field H_α to its original stationary value (with the RF power switched off).

Thus, the quantity $\rho_3 - \rho_{st}$ is determined entirely by the change in the magnitude of H_α brought about by the saturation of the resonance, while the quantity $\rho_2 - \rho_{st}$ depends in addition on the angle of rotation of H_α in the presence of the saturating RF field. As can be seen from Fig. 9, these quantities vary with the resonance detuning $\omega - \omega_0$. Figure 10 shows the dependences, obtained from the data of Fig. 9, of ρ_2 and ρ_3 on the quantity $\omega - \omega_0$ (in this case, scanning of the external field H_0 was used).

The observed nontrivial dependences arise from the nature of the behavior of H_α in a strong RF field. According to the theory of[9], in these conditions

$$|H_\alpha| = (H_{\alpha\perp}^2 + H_{\alpha\parallel}^2)^{1/2} = \frac{|\omega - \omega_0|}{[(\omega - \omega_0)^2 + \Omega_1^2]^{1/2}} H_{\alpha 0} \qquad (8)$$

where $H_{\alpha\parallel}$ and $H_{\alpha\perp}$ are the components of the field H_α along and perpendicular to H_0, $H_{\alpha 0}$ are the values of the nuclear field away from the resonance, and Ω_1 = $g\mu_0 H_1$, where H_1 is the amplitude of the RF field. In Fig 8b, the dotted curve shows the motion of the tip of the vector H_α (and, correspondingly, of the tip of the vector H_{eff}) during slow passage through the resonance.
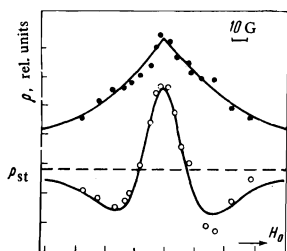
The curve for ρ_3 in Fig. 10 directly reflects the change of $|H_\alpha|$ on passage through the resonance: the magnitude of the deviation $\rho_3 - \rho_{st}$ is proportional to the change $|H_\alpha| - H_{\alpha 0}$ (cf. above). The solid line for ρ_3 in Fig. 10 was drawn in accordance with formula (8) with the appropriate choice of value of Ω_1 and of the quantity $\rho_3 - \rho_{st}$ for $\omega = \omega_0$. As can be seen, the theoretical curve, which has a characteristic discontinuity in the region of $\omega = \omega_0$, is in good agreement with the experimental points.

The curve for ρ_2 has a more complicated form. The decrease of the degree of polarization ($\rho_2 < \rho_{st}$) for $\omega - \omega_0 > 0$ is obviously due to the rotation of H_α, i.e., to the appearance of a transverse component $H_{\alpha\perp}$. However, as the resonance is approached, it can be seen from (8) that the magnitude of $|H_\alpha|$ decreases, and $|H_\alpha| \to 0$ as $\omega \to \omega_0$. Correspondingly, $H_{\alpha\perp} \to 0$, and the angle θ_2 decreases, causing an increase of ρ_2. In the region of exact resonance $\omega = \omega_0$, complete saturation sets in: $|H_\alpha| = 0$. Correspondingly, $H_{\alpha\perp} = 0$ and $\theta_2 = 0$, and the vector H_{eff} is again collinear with H_0 (Fig. 8b). However, the absolute value of H_{eff} is greater than its original value by an amount $|H_\alpha|$ (the field H_α has vanished). Therefore, when $\omega = \omega_0$, the degree of polarization ρ_2 increases to values greater than ρ_{st} even in the presence of the RF power. Inasmuch as complete depolarization of the nuclei occurs at $\omega = \omega_0$, the field H_{eff} is in no way changed immediately after the RF power is switched off. In fact, in the experiment at $\omega = \omega_0$, the values of ρ_2 and ρ_3 are almost the same (Figs. 9b and 10)[7]. On further scanning of the frequency a completely analogous picture was observed with increasing $|\omega - \omega_0|$ (Fig. 10).

The lineshapes shown in Fig. 10 were observed only at very high RF powers, when considerable saturation of the resonance was attained. With decreasing RF power, the lineshapes changed: in the curve for ρ_2, the positive spike at $\omega = \omega_0$ decreased and then disappeared completely, so that at low RF powers the curve for ρ_2 (corresponding to the record of the spectrum during slow scanning) had the usual bell-shaped form (Fig. 11). In the curve for ρ_3 the discontinuity at $\omega = \omega_0$ also disappeared, and the curve took an almost Lorentzian shape (Fig. 11). In addition, as the RF power decreased the resonance lines became noticeably narrower (cf. curves 1, 2 and 3 in Fig. 11). Figure 12 shows the dependence obtained for the half-width of the ^{71}Ga line on the RF power. Extrapolation to zero value
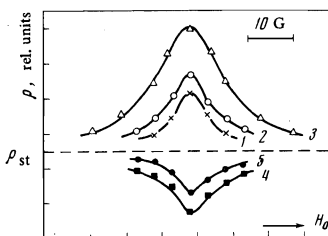

FIG. 11

FIG. 12

FIG. 10. Change of the luminescence polarization in the region of resonance at the ^{71}Ga nuclei at high RF power. S↑↓H$_0$ and H$_0$ = 3.6 kG. The points ● correspond to the quantity ρ_3 and ○ to the quantity ρ_2 (cf. Fig. 9.). The solid line for ρ_3 is drawn using formula (8) with Ω_1 = 70 G.

FIG. 11. The NMR line shape for the ^{71}Ga nuclei at low values of the RF power (obtained from data analogous to those shown in Fig. 9). S↑↓H$_0$ and H$_0$ = 3.6 kG. The curves 1, 2, and 3 show the dependence $\rho_3(\omega)$, curve 4 is $\rho_1(\omega)$ and curve 5 is $\rho_2(\omega)$. Curves 1, 2, and 3 are obtained for different RF-power values (increasing from 1 to 3). The curves 2, 4, and 5 all correspond to the same RF power.

FIG. 12. Dependence of the half-width (ΔH) of the nuclear resonance line of the nuclei ^{71}Ga on the RF power (H$_1$); S↑↓H$_0$ and H$_0$ = 3.6 kG.
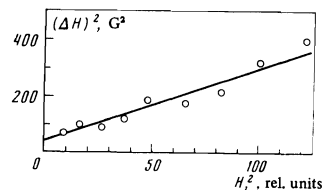
of the RF field gives a half-width of 7 ± 2 G for the line. For ^{27}Al the half-width of the line was of the same order. For ^{69}Ga and ^{75}As the lines were somewhat broader (of the order of $10-15$ G), which could be due to quadrupolar broadening arising from the fact that the environment of the nuclei in the solid solutions is not perfectly cubic.

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[1]In this case, additional polarization of the luminescence appears [10], due to the thermalization of the electrons over the spin sublevels split by the magnetic field (orientation of the spins by the magnetic field). However, this correction will be the same (in magnitude and sign) for σ^+, π and σ^- excitation. It is therefore easy to distinguish the contribution to the luminescence polarization from the purely optical orientation of the electrons: for this, for $\sigma^{\pm}$ excitation the degree of polarization of the luminescence was measured not with respect to zero but with respect to the signal $J_+ - J_-$ for π excitation. The values of ρ given below were obtained by this method.

[2]For comparison, we note that, in the given crystals at $77°$K, and in strongly alloyed crystals even at $4.2°$K, the lifetime of a nonequilibrium electron amounts only to $\sim 10^{-10}$ sec [11].

[3]Estimates for the hopping conductivity show [12] that, for the actual concentration of uncontrolled donors ($\sim 10^{16}$ cm^{-3}) in the crystals studied, the time for such a jump amounts to $\sim 10^{-10}$ sec.

[4]As will be shown below in subsection B, depolarization of the nuclei can be effected either by exposing the crystal to linearly polarized (or else unpolarized) light, or by keeping it in a field $H_0 = 0$ for some time.

[5]In principle, a quantitative description can also be given, if the dependence $\rho(H)$ and the absolute values of all the nuclear fields acting on the electron (H_α, H_β, H_χ and H_η) are known.

[6]The adiabatic character of the passage was also manifested in the fact that, irrespective of the direction of the scanning—from low to high frequencies or vice versa—, the decrease of ρ was always observed as the resonance frequency ω_0 was approached, and the increase of ρ after passage through ω_0. As a result, the form of the spectrum recorded changed on change of direction of the scanning: the minima and maxi-

ma of the resonance lines in Fig. 7 changed places.

[7]The difference observed in Fig. 10 between ρ_2 and ρ_3 in the region $\omega = \omega_0$ is obviously due to the fact that complete saturation of the resonance was not attained in this case.

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