Optical polarization of nuclei in silicon subjected to optical pumping in weak magnetic fields

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(Submitted 15 May 1978)
Zh. Eksp. Teor. Fiz. 75, 1743–1754 (November 1978)

An investigation was made of the optical polarization of the ^{29}Si nuclei in silicon containing deep and shallow impurity levels, and of the influence of a magnetic field on the polarization processes. The degree of the optical polarization of the nuclei in compensated silicon was determined as a function of the pump radiation intensity. The magnetic-field dependence of the degree of the optical polarization of the ^{29}Si nuclei in silicon containing phosphorus and gold impurity atoms was analyzed theoretically. The results of the theoretical calculations were compared with the experimental data.

PACS numbers: 78.20.Ls

In an earlier paper[1] we demonstrated that the processes of the dynamic polarization of the nuclei by optical pumping are much more effective in compensated silicon than in phosphorus-doped n-type silicon. For example, in the case of silicon doped with phosphorus and compensated with gold the degree of the optical polarization of the ^{29}Si nuclei was 30 times greater than the degree of the nuclear polarization attained in uncompensated silicon with the same phosphorus concentration. This increase in the degree of the optical polarization of the nuclei was due to a reduction in the photoelectron lifetime. However, gold impurity atoms not only reduced the photoelectron lifetime (and thus enhanced the effectiveness of the optical polarization of the nuclei), but could themselves act as the centers of relaxation and dynamic polarization of the lattice nuclei. It was found that optical pumping of phosphorus-and gold-doped silicon with circularly polarized light revealed that the degree of polarization of the ^{29}Si nuclei and the direction of the polarization relative to the direction of propagation of the pump radiation depended on a longitudinal magnetic field.[2]

We shall report an investigation of the processes of the dynamic polarization of the silicon nuclei by optical pumping, which are caused by the dipole–dipole interaction of these nuclei with electrons localized at deep-level impurity centers, and an investigation of the influence of an external magnetic field on the degree of the optical polarization of the nuclei. We shall report the results of an experimental study of the dynamic polarization of the ^{29}Si nuclei in optically pumped phosphorus-and gold-doped silicon subjected to various magnetic fields.

1. DYNAMIC POLARIZATION OF NUCLEI BY DIPOLE-DIPOLE INTERACTION WITH PARAMAGNETIC IMPURITIES

Illumination of a silicon crystal with circularly polarized light results in the excitation of conduction-band electrons with an average spin projection S_z^e which differs from the equilibrium value S_0. As shown in Ref. 3, the direct interaction of the ^{29}Si nuclei with optically oriented conduction-band electrons is negligibly small compared with the interaction of these nuclei with electrons localized at donor centers.

The oriented conduction-band electrons may be captured by donor levels. If this produces magnetic centers whose spin polarization differs from the equilibrium state, the interaction of the lattice nuclei with these centers may result in dynamic polarization of the former. If the impurity atoms have a deep level in the band gap, the hyperfine interaction of the lattice nuclei with electrons localized at this level may be dominated by the dipole–dipole interaction, in contrast to a shallow level when the wave function of a localized electron extends over many lattice constants and the contact interaction predominates.

The Hamiltonian of the dipole–dipole interaction of a nucleus whose spin is I with an electron whose spin is S can be written in the form[4]

$$\mathscr{H}_{dd}=\gamma_S\gamma_I\hbar^2r^{-3}(A+B+C+D+E+F), \tag{1}$$

where

$$\left.\begin{aligned}
A&=(1-3\cos^2\theta)I_zS_z;\\
B&=-\tfrac{1}{4}(1-3\cos^2\theta)(I_+S_-+I_-S_+);\\
C&=-\tfrac{3}{2}\sin\theta\cos\theta e^{-i\varphi}(I_zS_++I_+S_z);\\
D&=-\tfrac{3}{2}\sin\theta\cos\theta e^{i\varphi}(I_zS_-+I_-S_z);\\
E&=-\tfrac{3}{4}\sin^2\theta e^{-2i\varphi}I_+S_+;\\
F&=-\tfrac{3}{4}\sin^2\theta e^{2i\varphi}I_-S_-;
\end{aligned}\right\} \tag{2}$$

γ_S and γ_I are the gyromagnetic ratios of the electron and nucleus, respectively; r is the distance between the electron and nucleus; θ and φ are polar coordinates of the vector **r** joining the electron and nucleus; $I_\pm=I_x\pm iI_y$; $S_\pm=S_x\pm iS_y$.

Each term in Eq. (1) corresponds to the following transitions between states with the projections $I_z=m_I$ and $S_z=m_S$:

$$\left.\begin{aligned}
&A:\ \Delta m_S=0,\ \Delta m_I=0,\ \Delta(m_S+m_I)=0;\\
&B:\ \Delta m_S=\pm1,\ \Delta m_I=\mp1,\ \Delta(m_S+m_I)=0;\\
&C:\ \Delta m_S=+1,\ \Delta m_I=0,\ \Delta(m_S+m_I)=+1;\\
&\quad\ \Delta m_S=0,\ \Delta m_I=+1,\ \Delta(m_S+m_I)=+1;\\
&D:\ \Delta m_S=-1,\ \Delta m_I=0,\ \Delta(m_S+m_I)=-1;\\
&\quad\ \Delta m_S=0,\ \Delta m_I=-1,\ \Delta(m_S+m_I)=-1;\\
&E:\ \Delta m_S=+1,\ \Delta m_I=+1,\ \Delta(m_S+m_I)=+2;\\
&F:\ \Delta m_S=-1,\ \Delta m_I=-1,\ \Delta(m_S+m_I)=-2.
\end{aligned}\right\} \tag{3}$$

Thus, the Hamiltonian (1) contains operators linking the states $|m_I\rangle$ and $|m_I\pm1\rangle$. The interaction (1) be-

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tween the nucler and electron spins results in relaxation of the nuclear moments only if it is modulated in time. If this modulation is a random function of time, the nuclear relaxation rate is proportional to the spectral density $J(\omega)$ of this random function at a frequency ω corresponding to a transition between the nuclear states $|m_I\rangle$ and $|m_I \pm 1\rangle$.

The interaction of nuclei with fixed paramagnetic centers is modulated by the time dependence of the electron spin S. This modulation is due to the relaxation of the spins S, which is due to the interaction of electron spins with the lattice.[4,5]

It is clear from Eq. (3) that nuclear relaxation in the dipole–dipole interaction with electron spins may be due to the following processes:

1) reorientation of the electron and nuclear spins toward one another (transitions with $\Delta m_s = \pm 1. \Delta m_I = \mp 1$); this process it described by the term B containing the operator $I_+S_- + I_-S_+$;

2) rotation of the nuclear spin alone without a change in the electron spin ($\Delta m_I = \pm 1, \Delta m_s = 0$); it is due to to the terms I_+S_z and I_-S_z in C and D;

3) rotation of the electron and nuclear spins in one direction ($\Delta m_I = \pm 1, \Delta m_s = \pm 1$); this process corresponds to the operators I_+S_+ amd I_-S_- contained in E and F.

We can calculate the transition probabilities for each of these processes. We shall consider the case when $S = 1/2$ and $I = 1/2$ and we shall assume that the signs of the magnetic moments of the electron μ_S and nucleus μ_I are negative. This applies to silicon because the spin of the ^{29}Si nucleus is $I = 1/2$ and we have $\mu_I < 0$.

For the relaxation process 1), which is due to the operator $I_+S_- + I_-S_+$, we can introduce the probability $W[(+-) \to (-+)]$ of a transition from a state with the projections of the electron $m_S = +1/2$ and nuclear $m_I = -1/2$ spins to a state with $m_S = -1/2$ and $m_I = +1/2$, and the probability of the reverse transition $W[(-+) \to (+-)]$. Since these probabilities apply to relaxation transitions, it follows that[5]

$$\frac{W((+-)\to(-+))}{W((-+)\to(+-))} = \exp\left(\frac{\mu_S H_0}{kT} - \frac{\mu_I H_0}{kT}\right) = e^{\Delta-\delta}.$$

Here,

$$\Delta = \frac{\mu_S H_0}{kT} = -\frac{\gamma_S \hbar H_0}{kT}, \quad \delta = \frac{\mu_I H_0}{kT} = -\frac{\gamma_I \hbar H_0}{kT},$$

k is the Boltznamm constant, and T is the absolute temperature. We can show[6] that

$$W((+-)\to(-+)) = w_1 \exp\left(\frac{\Delta-\delta}{2}\right),$$
$$W((-+)\to(+-)) = w_1 \exp\left(-\frac{\Delta-\delta}{2}\right). \quad (4)$$

The quantity w_1 represents the probability of rotation of the nuclear spin ($\Delta m_I = \pm 1$) induced by an alternating magnetic field exerted on the nuclear spin by the electron spin $\mathbf{S}(t)$.

If we assume that the time dependence $\mathbf{S}(t)$ is described by the Bloch equations with the relaxation times $\tau_1 = \tau_2 = \tau_s$, and that the interaction of the spin S with the nuclei is a weak perturbation,[5] we find that the probability w_1 of the $(+-) \rightleftarrows (-+)$ transitions is

$$w_1 = \frac{1}{16}\frac{\gamma_S^2\gamma_I^2\hbar^2}{r^6}(1-3\cos^2\theta)^2 \int_{-\infty}^{\infty} \langle S_+(0)S_-(\tau)\rangle \exp(-i\omega_I\tau)\,d\tau$$
$$= \frac{1}{20}\frac{\gamma_S^2\gamma_I^2\hbar^2}{r^6}\frac{\tau_S}{1+(\omega_S-\omega_I)^2\tau_S^2}. \quad (5)$$

We have allowed here for the fact that[5]

$$\langle S_+(0)S_-(\tau)\rangle = {}^2/_3 S(S+1)\exp(i\omega_S\tau - \tau/\tau_S), \quad S = {}^1/_2,$$

and we have carried out averaging over the angles.

Similarly, in the case of w_2 and w_3 corresponding to the transitions $(\pm, +) \rightleftarrows (\pm, -)$ and $(++) \rightleftarrows (--)$, we obtain

$$w_2 = \frac{3}{20}\frac{\gamma_S^2\gamma_I^2\hbar^2}{r^6}\frac{\tau_S}{1+\omega_I^2\tau_S^2}, \quad (6)$$

$$w_3 = \frac{6}{20}\frac{\gamma_S^2\gamma_I^2\hbar^2}{r^6}\frac{\tau_S}{1+(\omega_S+\omega_I)^2\tau_S^2} \quad (7)$$

and

$$W((\pm,+)\to(\pm,-)) = w_2 e^{\delta/2}, \quad W((\pm,-)\to(\pm,+)) = w_2 e^{-\delta/2}; \quad (8)$$
$$W((++)\to(--)) = w_3 e^{(\Delta+\delta)/2}, \quad W((--)\to(++)) = w_3 e^{-(\Delta+\delta)/2}. \quad (9)$$

Using the probabilities (4), (8), and (9), we can derive the transport equations for the populations n_+ and n_- of the states with the nuclear spin projections $m_I = +1/2$ and $m_I = -1/2$. We shall assume that $\Delta \ll 1$ and $\delta \ll 1$ and retain only the first term in the expansion of the exponential function:

$$\begin{aligned} dn_+/dt = &-n_+N_-w_1(1-\Delta/2+\delta/2) + n_-N_+w_1(1+\Delta/2-\delta/2) \\ &-n_+(N_++N_-)w_2(1+\delta/2) + n_-(N_++N_-)(1-\delta/2)w_2 \\ &-n_+N_+w_3(1+\Delta/2+\delta/2) + n_-N_-w_3(1-\Delta/2-\delta/2). \end{aligned} \quad (10)$$

Introducing the electron and nuclear polarizations

$$P_e = (N_+-N_-)/(N_++N_-), \quad P_n = (n_+-n_-)/(n_++n_-)$$

and their equilibrium values in a magnetic field H_0 at a temperature T

$$P_{e0} = -\Delta/2, \quad P_{n0} = -\delta/2,$$

we find that the steady-state values P_e and P_n are easily obtained from Eq. (10):

$$P_n = P_{n0} - \xi(P_e - P_{e0}), \quad (11)$$
$$\xi = (w_3 - w_1)/(w_1 + 2w_2 + w_3). \quad (12)$$

Thus, it is clear from Eq. (11) that the magnitude and direction of the nuclear magnetization which appears on interaction with electrons, whose polarization is kept constant and different from the Boltzmann value, depends on the relationship between the probabilities w_1, w_2, and w_3. For example, if the interaction between electrons and nuclei is of the contact type, the Hamiltonian of this interaction contains only the operator $I_+S_- + I_-S_+$, corresponding to the reorientation of the nuclear and electron spins toward one another. The values of w_2 and w_3 then vanish and we find from Eq. (12) that $\xi = -1$. If the electron polarization is $P_e = 0$, which can be achieved by saturating an electron resonance with a strong rf field of frequency ω_S, the nuclei become dynamically polarized (Overhauser effect) and we find that $P_n \approx -P_{e0}$, where the direction of the nuclear polarization P_n is opposite to its equilibrium value P_{n0}.

If the magnetic hyperfine interaction between the electron and nuclear spins is of the dipole–dipole type and a sample is in a strong magnetic field H_0, i.e., if $\omega_s\tau_s \gg 1$, it then follows from Eqs. (5), (6), and (7) that the probabilities w_1 and w_3 are negligibly small compared with w_2, since in the case we have

$$w_1/w_2 \approx w_3/w_2 \approx (\gamma_I/\gamma_S)^2 \sim 10^{-6}.$$

The nuclear relaxation is then governed only by the process in which the nuclear spin is rotated without a change in the electron spin: $(\pm, +) \rightleftarrows (\pm, -)$, whereas the transitions $(+, -) \rightleftarrows (-, +)$ and $(+, +) \rightleftarrows (-, -)$, that produce the nuclear polarization, are "forbidden" in a strong magnetic field. In Eq. (12), we then have $\xi \approx 0$ and $P_n \approx P_{n0}$, i.e., the dynamic polarization of the nuclei is not observed when P_e deviates from the equilibrium value P_{e0}.

Under these conditions the dynamic nuclear polarization is attained by the method of saturation of the "forbidden" transitions by an rf field or by the solid effect.[5] The transition probabilities $W_{rf}[(+, +) \rightleftarrows (-, -)]$ or $W_{rf}[(+, -) \rightleftarrows (-, +)]$ may then be found, depending on the rf field intensity, to be considerably greater than the probability of the relaxation transitions $W[(\pm, +) \rightleftarrows (\pm, -)]$, which results in the dynamic nuclear polarization.

If an external magnetic field H_0 is so weak that $\omega_s\tau_s \lesssim 1$, the probabilities of the relaxation transitions w_1, w_2, and w_3 are comparable [see Eqs. (5)–(7)] and the dynamic polarization of the nuclei may appear as a result of nuclear relaxation due to the dipole–dipole interaction between the nuclei and electrons which are in a nonequilibrium state (this state may be created by optical pumping). The ratio of the probabilities w_1, w_2, and w_3 in the case of weak fields and very short times τ_S is $w_1:w_2:w_3 = 2:6:12$, which gives $\xi = 0.385$. It should be noted that when the modulation of the dipole–dipole interaction is due to random relative motion of particles with spin I and S characterized by a very short correlation time, we find that $w_1: w_2: w_3 = 2:3:12$ and $\xi = 0.5$ (Refs. 4 and 5).

Illumination of a silicon crystal containing paramagnetic impurity atoms with circularly polarized light in a weak magnetic field H_0 should result in strong polarization of electron spins: $|P_e| \gg |P_{e0}|$. Then, the degree of polarization of the silicon lattice nuclei is

$$P_n \simeq -\xi P_e. \tag{13}$$

If we assume that the electron polarization is independent of an external longitudinal magnetic field H_0, we find that the dependence of the nuclear degree of polarization P_n on H_0 is governed by the dependence of ξ on the magnetic field. This dependence can be found in Eqs. (5)-(7) and (12), where $\omega_s = \gamma_s H_0$ and $\omega_I = \gamma_I H_0$. Bearing in mind that $\omega_s \gg \omega_I$ (for silicon we have $\gamma_S/\gamma_I = 3310$) and that the inequality $\omega_I\tau_S \ll 1$ is

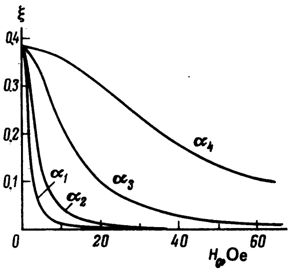

FIG. 1. Dependence of ξ on the magnetic field H_0. The values of α_1, α_2, α_3, and α_4 are associated with $\tau_S = 10^{-7}$, 5×10^{-8}, 2×10^{-8}, and 10^{-8} sec, respectively.

satisfied in weak magnetic fields, we obtain the following expression for ξ:

$$\xi = \frac{6(1+\omega_S^2\tau_S^2)^{-1} - (1+\omega_S^2\tau_S^2)^{-1}}{(1+\omega_S^2\tau_S^2)^{-1} + 6(1+\omega_I^2\tau_S^2)^{-1} + 6(1+\omega_S^2\tau_S^2)^{-1}} \approx 5(13+6\gamma_S^2\tau_S^2H_0^2)^{-1} \tag{14}$$

or

$$\xi = 5/(13+\alpha H_0^2), \tag{15}$$

where

$$\alpha = 6\gamma_S^2\tau_S^2. \tag{16}$$

Figure 1 shows the dependence of ξ on H_0 calculated from Eq. (14). We can see from Fig. 1 that in weak fields H_0 the value of ξ differs from zero and that it decreases on increase of H_0.

We have thus established that the dynamic nuclear polarization due to the dipole–dipole interaction with oriented electrons appears only in weak magnetic fields. The range of fields H_0 in which this polarization is possible depends on the spin relaxation time of electrons τ_S and, as can be seen from Fig. 1, it increases on reduction in τ_S.

2. OPTICAL POLARIZATION OF NUCLEI IN SILICON DOPED WITH DEEP- AND SHALLOW-LEVEL IMPURITIES

We shall now consider the ^{29}Si nuclei in silicon which interact with electrons localized at shallow donor impurities, for example, phosphorus atoms (this interaction is of the contact type), and with electrons localized at deep impurities, for example gold atoms (this interaction is of the dipole–dipole type).

The dependence of the steady-state degree of nuclear polarization P_n on an external magnetic field H_0 in which optical pumping takes place can be found using Eq. (11), which can be generalized to the case of the interaction between the ^{29}Si nuclei and various impurity centers:

$$P_n = P_{n0} - \xi_P f_P (P_e^P - P_{e0}^P) - \xi_{Au} f_{Au} (P_e^{Au} - P_{e0}^{Au}), \tag{17}$$

where P_e^P and P_e^{Au} are the degrees of polarization of the electrons localized at the phosphorus and gold atoms, respectively; P_{e0}^P and P_{e0}^{Au} are the equilibrium values of these polarizations; $f_P = T_1/T_{1P}$ and $f_{Au} = T_1/T_{1Au}$; T_{1P} and T_{1Au} are the relaxation times of the ^{29}Si nuclei due to the interactions with the electrons localized at the phosphorus and gold atoms, respectively; $T_1 = (1/T_{1P} + 1/T_{1Au})^{-1}$ is the total nuclear relaxation time. We find that $\xi_P = -1$, because the interaction of the

^{29}Si nuclei with the electrons localized at phosphorus atoms is of the contact type, whereas the quantity ξ_{Au} is given by Eq. (15).

The equilibrium nuclear and electron polarizations (P_{e0}^{P} and P_{e0}^{Au}) can be ignored in weak magnetic fields. Moreover, we shall assume that the polarizations of the electrons localized at the phosphorus and gold atoms as a result of illumination of a silicon crystal with circularly polarized light are equal: $P_e^P = P_e^{Au} = P_e$. We then find from Eq. (17) that

$$P_n = P_e(f_P - \xi_{Au} f_{Au}) = P_e \frac{T_1}{T_{1Au}} \left(\frac{T_{1Au}}{T_{1P}} - \xi_{Au} \right). \quad (18)$$

The spin-lattice relaxation time T_{1P} due to the interaction of the ^{29}Si nuclei with the electrons localized at the phosphorus atoms is usually several hours (this is found from the experimental data). The time T_{1Au} is much shorter and amounts to a few tens of minutes. Thus, in weak magnetic fields the ratio T_{1Au}/T_{1P} may be less than ξ_{Au}. It then follows from Eq. (18) that the sign of the nuclear polarization is opposite to that for a sample containing phosphorus atoms alone. An increase in the external magnetic field H_0 reduces the value of ξ_{Au} (Fig. 1) and in some field H_0 we find that $\xi_{Au} = T_{1Au}/T_{1P}$ so that the nuclear polarization P_n vanishes. A further increase in H_0 makes $T_{1Au}/T_{1P} - \xi_{Au}$ a positive quantity and the direction of the nuclear polarization is reversed.

In weak magnetic fields we must allow for the influence of a local magnetic field H_L of the ^{29}Si nuclei in a crystal (according to Ref. 7, $H_L = 0.176$ Oe for silicon). If the optical pumping takes place in a magnetic field H_0 comparable with H_L, the degree of nuclear polarization is proportional to $H_0^2/(H_0^2 + 3H_L^2)$ (Ref. 8).

It therefore follows that the degree of nuclear polarization is

$$P_n = P_e \frac{T_1}{T_{1Au}} \left(\frac{T_{1Au}}{T_{1P}} - \xi_{Au} \right) \frac{H_0^2}{H_0^2 + 3H_L^2} . \quad (19)$$

The experiments showed that the times T_1, T_{1P}, and T_{1Au} do not vary greatly with the magnetic field. Then, the dependence of the degree of nuclear polarization (produced by optical pumping of silicon doped with gold and phosphorus) on the magnetic field is given by

$$P \propto \left(\beta - \frac{5}{13 + \alpha H_0^2} \right) \frac{H_0^2}{H_0^2 + 3H_L^2}, \quad (20)$$

where $\beta = T_{1Au}/T_{1P}$ and $\xi_{Au} = 5/(13 + \alpha H_0^2)$ [see Eq. (15)].

We can see from Eq (19) that the magnitude and direction of the nuclear polarization depend on an external magnetic field H_0 in which optical pumping takes place and on the values of β and α, which are governed by the properties of the crystal. The quantity α depends on the spin-lattice relaxation time τ_S of the electrons localized at deep centers [see Eq. (16)] and β is governed by the ratio T_{1Au}/T_{1P}, which depends on the ratio of the phosphorus and gold concentrations in the silicon crystal.

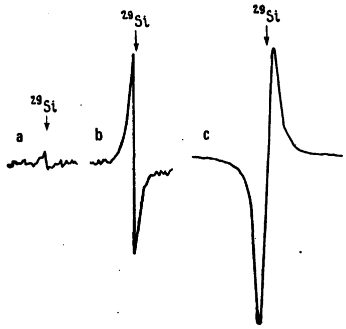

FIG. 2. Nuclear magnetic resonance signals of the ^{29}Si nuclei in Si:P:Au with $N(P) \approx 5 \times 10^{15}$ cm^{-3} and $N(Au) \approx 3 \times 10^{16}$ cm^{-3}: a) equilibrium signal in a magnetic field 4.9 kOe at $T = 300$°K; b) signal after illumination with circularly polarized light for 15 min in a field $H_0 = 50$ Oe at $T = 77$°K; c) signal after illumination with circularly polarized light for 15 min in a field 0.7 Oe at $T = 77$°K (in this case the instrumental gain is reduced by a factor of 10).

3. EXPERIMENTAL RESULTS AND DISCUSSION

A. Influence of an external magnetic field on the degree of optical polarization of the ^{29}Si nuclei in compensated silicon

Our experiments were carried out on silicon doped with phosphorus in concentrations $N(P)$ of a) $\approx 10^{16}$, b) 5×10^{15}, and c) 10^{15} cm^{-3}, and compensated with gold. The gold concentration was practically the same in all these samples: $\sim 3 \times 10^{16}$ cm^{-3}. Moreover, we studied silicon doped with phosphorus alone in a concentration $N(P) \approx 2 \times 10^{13}$ cm^{-3}. The experimental method used in the optical polarization of the ^{29}Si nuclei and in detecting this polarization by the NMR method was described in detail earlier.[2,3]

Illumination of the gold-and phosphorus-doped silicon with circularly polarized light resulted in the dynamic polarization of the silicon lattice nuclei. The direction of the nuclear polarization depended on the external magnetic field H_0 in which the optical pumping took place. This polarization was detected from a considerable increase in the NMR signal of ^{29}Si. Figure 2 shows the dynamic polarization signals of the ^{29}Si nuclei in silicon doped with gold and phosphorus, obtained under equilibrium conditions in a field 4.9 kOe (a) and after 15-min illumination with circularly polar-

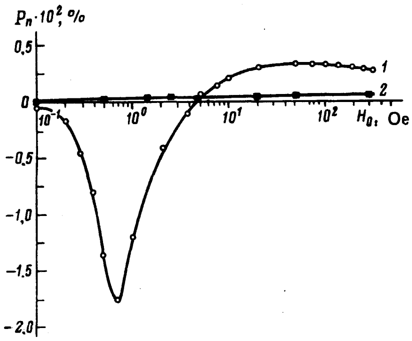

FIG. 3. Dependence of the degree of polarization P_n of the ^{29}Si nuclei on the magnetic field H_0 obtained for Si:P:Au (curve 1) and Si:P (curve 2) when pumped optically with circularly polarized light.

ized light in magnetic fields of 0.7 Oe (b) and 50 Oe (c). It is clear from Fig. 2 that the directions of the nuclear polarization produced by optical pumping in magnetic fields of 0.7 Oe and 50 Oe are opposite.

Figure 3 shows the dependence of the degree of polarization of the ^{29}Si nuclei in silicon with $N(P) \approx 5 \times 10^{15}$ cm^{-3} and $N(Au) \approx 3 \times 10^{16}$ cm^{-3} on the longitudinal magnetic field H_0 in which the optical pumping took place (curve 1), as well as the same dependence for silicon containing only phosphorus atoms (curve 2). It is clear from Fig. 3 (curve 1) that Si: P: Au exhibits inversion of the nuclear polarization on increase of H_0. The maximum absolute value of the optical polarization of the ^{29}Si nuclei is attained in a field of $H_0 = 0.7$ Oe and it is equal to 1.75×10^{-2}%, which corresponds to the equilibrium polarization in a field 6.65×10^5 Oe at $T = 77$°K. An increase in H_0 reduces the polarization P_n which vanishes at $H_0 = 4.5$ Oe. In fields $H_0 > 4.5$ Oe the nuclear polarization is reversed in direction, in spite of the fact that the sign of the circular polarization of the pump radiation remains unchanged.

When silicon doped with phosphorus alone is subjected to optical pumping in various magnetic fields H_0, the NMR signals are of one sign only (curve 2 in Fig. 3). The time taken to establish the maximum polarization of the nuclei, i.e., the spin–lattice relaxation time of the ^{29}Si nuclei in phosphorus-and gold-doped silicon subjected to optical pumping, varies from ~15 min in $H_0 = 0.1$ Oe to 45–60 min in $H_0 = 10$ Oe.

The change in the direction of the nuclear polarization on increase of the longitudinal magnetic field H_0 exhibited by Si: P: Au is due to two processes of the dynamic polarization of the ^{29}Si nuclei. One of them is the dynamic polarization of the nuclei due to the contact interaction with the electrons localized at the phosphorus atoms. We can see from Fig. 3 (curve 2) that in the case of silicon doped solely with phosphorus the nuclear polarization observed in all fields H_0 is of the same sign and its value does not vary greatly with H_0.

The second process causing the dynamic polarization of the ^{29}Si nuclei is the interaction of these nuclei with the electrons captured by the gold atoms. As pointed out earlier, gold has deep levels in silicon and in this case the dipole–dipole interaction predominates, which reverses the nuclear polarization direction. As shown above (Sec. 1), the dipole–dipole interaction polarizes dynamically the ^{29}Si nuclei only if optical pumping takes place in weak magnetic fields. The dependence of the degree of the optical polarization P_n of the ^{29}Si nuclei on H_0 is then governed by the dependence of ξ on H_0 [see Eq. (15) and Fig. 1]. This dependence shows that P_n due to this polarization mechanism decreases on increase of H_0.

The relative contributions of these two dynamic polarization mechanisms depend on the ratio of the nuclear relaxation times $\beta = T_{1Au}/T_{1P}$ [see Eq. (20)], which is determined by the relative concentrations of the phosphorus and gold atoms in a crystal. Figure 4 gives the dependence of the degree of the optical polarization $P_n/|P_{n\,max}|$ of the ^{29}Si nuclei on H_0 for various gold-doped silicon samples containing phosphorus in concentrations of 10^{16}, 5×10^{15}, and 10^{15} cm^{-3} (Figs. 4a, 4b, and 4c, respectively). The value of $|P_{n\,max}|$ for these samples is 3.8×10^{-3}, 1.75×10^{-2}, and 2×10^{-3}%, respectively. The continuous curves in Fig. 4 represent the dependences of P_n on H_0 calculated from Eq. (20). The best agreement between the experimental and theoretical curves is obtained for the following parameters α and β: a) $\alpha = 3.2$ rad$^2\cdot$Oe^{-2}, $\beta = 0.15$; b) $\alpha = 10$ rad$^2\cdot$Oe^{-2}, $\beta = 0.03$; c) $\alpha = 50$ rad$^2\cdot$Oe^{-2}, $\beta = 0$. It is clear from Fig. 4 that the proposed model of the dynamic polarization of the ^{29}Si nuclei by optical pumping of silicon containing phosphorus and gold atoms describes quite well the real nuclear dynamic polarization processes.

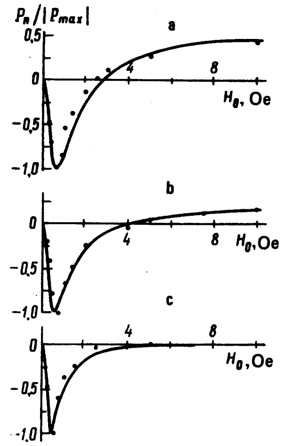

FIG. 4. Dependence of the degree of the optical polarization on H_0 for: a) Si:P:Au with $N(P) \approx 10^{16}$ cm^{-3}; b) Si:P:Au with $N(P) \approx 5 \times 10^{15}$ cm^{-3}; c) Si:P:Au with $N(P) \approx 10^{15}$ cm^{-3}. The gold concentration in all three samples was $N(Au) \approx 3 \times 10^{16}$ cm^{-3}. The continuous curves are the calculated dependences.

We investigated the dependence of the degree of the optical nuclear polarization P_n on the pump radiation intensity I_p reaching Si:P:Au samples. The dependences were recorded in magnetic fields corresponding to positive and negative values of P_n (Fig. 5). Curve 1 was obtained by optical pumping in a magnetic field $H_0 = 10$ Oe and curve 2 in a magnetic field $H_0 = 0.7$ Oe. It is clear from Fig. 5 that the degree of the optical polarization of the ^{29}Si nuclei in all the Si:Au:P samples in $H_0 = 10$ Oe increases linearly on increase of the

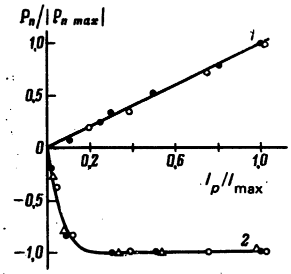

FIG. 5. Dependences of the degree of the optical polarization of the ^{29}Si nuclei on the intensity of the pump radiation reaching Si:P:Au samples: 1) optical pumping in a field 10 Oe; 2) optical pumping in a field 0.7 Oe; ○) $N(P) \approx 10^{16}$ cm^{-3}; ●) $N(P) \approx 5 \times 10^{15}$ cm^{-3}; △) $N(P) \approx 10^{15}$ cm^{-3}.

pump radiation intensity. On the other hand, the degree of the nuclear polarization corresponding to negative values of P_n and $H_0 = 0.7$ Oe increases on increase of I_p when the radiation intensity is low but it reaches saturation at $I_p = 0.21_{p\ max}$ and remains constant when the intensity is increased still further.

In weak magnetic fields ($H_0 = 0.7$ Oe) the dynamic polarization of the ^{29}Si nuclei is due to their interaction with the electrons localized at the gold atoms which produces paramagnetic Au^0 centers. Illumination of a crystal with circularly polarized light results in the spin orientation of the Au^0 centers. This may occur, for example, as a result of the capture of an oriented conduction-band electron by an Au^+ ion, formed as a result of the capture of a hole by an Au^0 atom under optical illumination conditions. The number of the Au^0 atoms is practically unaffected. Since the photon flux corresponding to $I_p = I_{p\ max}$ is $\sim 10^{18}$ cm$^{-2}\cdot$sec^{-1} and the concentration of gold in the investigated samples is $\sim 10^{16}$ cm^{-3}, it follows that practically all the neutral gold atoms participate in the process of nuclear polarization even at low pump radiation intensities. This results in saturation of P_n on increase of I_p (curve 2 in Fig. 5).

An increase in the magnetic field H_0 results in the dominance of the dynamic polarization process by the contact interaction between the ^{29}Si nuclei and electrons localized at the phosphorus atoms. The presence of neutral paramagnetic Au^0 atoms results in "leakage" of the nuclear polarization because the main spin-lattice relaxation mechanism of the ^{29}Si nuclei is the interaction with the Au^0 atoms. However, as shown above, this interaction does not result in nuclear polarization in magnetic fields exceeding 5 Oe.

The value of P_n in a strong magnetic field when $\xi_{Au} = 0$ can be found from Eq. (18);

$$P_n = \frac{T_1}{T_{1P}} P_e = \frac{1}{T_{1P}} \left(\frac{1}{T_{1P}} + \frac{1}{T_{1Au}} \right)^{-1} P_e.$$

If we bear in mind that $T_{1Au} \ll T_{1P}$ ($\beta = 0.03$), we obtain

$$P_n \approx (T_{1Au}/T_{1P}) P_e. \tag{21}$$

The value of T_{1Au} is governed by the concentration of the paramagnetic Au^0 atoms in a sample and is independent of the pump radiation intensity. The time T_{1P} depends on the degree of the electron occupancy of the donor levels of phosphorus.[3] Since the photoelectron lifetime in gold-doped silicon is short ($\sim 10^{-8}$–10^{-9} sec), the steady-state density of the conduction electrons during illumination is low ($\sim 10^9$–10^{10} cm^{-3}), so that the degree of the electron occupancy of the donor levels of phosphorus is also low. Under these conditions the value of $1/T_{1P}$ increases linearly on increase of the pump radiation intensity.[3,9] It follows from Eq. (21) that P_n also increases linearly with the pump radiation intensity. This is confirmed by the experimental results (curve 1 in Fig. 5). The basically different dependences of the optical polarization of the nuclei on the intensity of the pump radiation in the case of the positive and negative values of P_n also shows that the optical polarization of the nuclei in compensated silicon is due to two different centers.

B. Determination of the spin relaxation time of electron localized at deep impurity centers

The spin relaxation time of electrons localized at the gold impurity centers can be estimated from Eq. (16) using the above values of α for the investigated silicon samples and assuming that γ_S, for an electron localized at a gold atom, is close to the gyromagnetic ratio of a free electron $\gamma_S = 1.75 \times 10^7$ rad. Hz. Oe^{-1}. Consequently, we find that $\tau_S = 0.42 \times 10^{-7}$, 0.74×10^{-7}, and 1.65×10^{-7} sec for our samples a, b, and c, respectively. On the other hand, the time τ_S for the investigated samples can be determined using the dependence of the nuclear relaxation time T_1 on the magnetic field.

The spin-lattice relaxation of the ^{29}Si nuclei in strong magnetic fields is mainly due to their dipole-dipole interaction with electrons localized at gold atoms. It is known[4] that the nuclear spin-lattice relaxation time T_1 calculated allowing for the nuclear spin diffusion in the case of the dipole-dipole interaction in strong magnetic fields of the lattice nuclei with fixed paramagnetic impurities is proportional to $(1 + \omega_I^2 \tau_S^2)^{1/4}$.

It follows that the relaxation time of the ^{29}Si nuclei is independent of the magnetic field H_0 as long as $\omega_I \tau_S \ll 1$ and begins to increase with H_0 when $\omega_I \tau_S \gtrsim 1$. The time T_1 increases by a factor of $2^{1/4}$ for $\omega_I \tau_S = 1$. Having determined experimentally the magnetic field H_0' in which the time T_1 increases on increase of H_0 by a factor of $2^{1/4}$ relative to its value in weak magnetic fields, we can apply the condition

$$\tau_S = 1/\omega_I = 1/\gamma_I H_0' \tag{22}$$

($\gamma_I = 5.31 \times 10^3$ rad$\cdot$ Oe$^{-1}\cdot$ sec^{-1}) to find the value of τ_S.

Figure 6 shows the dependence of the spin—lattice relaxation time T_1 of the ^{29}Si nuclei on the magnetic field H_0 in Si:P:Au with the phosphorus concentration $N(P) \approx 10^{16}$ cm^{-3} (curve 1) or $N(P) = 5 \times 10^{15}$ cm^{-3} (curve 2). It is clear from Fig. 6 that in the case of the second sample the value of T_1 in a field $H_0 = 2.3$ kOe exceeds its value in weak magnetic fields by a factor of $2^{1/4}$. For this value of H_0' we find τ_S from Eq. (22), which gives $\tau_S = 0.8 \times 10^{-7}$ sec, in good agreement with the value determined above from the data in Fig. 4b.

It is clear from Fig. 6 (curve 1) that the relaxation time T_1 for the sample with $N(P) \approx 10^{16}$ cm^{-3} increases only slightly on application of $H_0 \approx 5$ kOe. Thus, for this sample the value of H_0 in which T_1 increases by a factor of $2^{1/4}$ is greater and the time τ_S is less than for the sample with $N(P) = 5 \times 10^{15}$ cm^{-3}. This is in

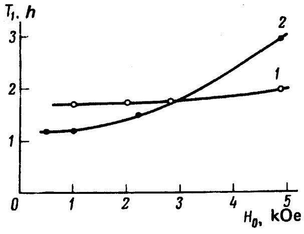

FIG. 6. Dependences of the spin—lattice relaxation time T_1 of the ^{29}Si nuclei on the magnetic field H_0 in Si:P:Au with $N(P) \approx 10^{16}$ cm^{-3} (curve 1) and $N(P) \approx 5 \times 10^{15}$ cm^{-3} (curve 2).

agreement with the values of τ_S obtained above for these samples.

It follows from the above results that the dynamic nuclear polarization is most effective in weak magnetic fields if it is due to the dipole–dipole interaction. This is associated with the fact that a reduction in the magnetic field increases the contribution made to the nuclear relaxation process by the transitions involving the rotation of the electron and nuclear spins in the same ($\Delta m_S = \pm 1$, $\Delta m_I = \pm 1$) and opposite ($\Delta m_S = \pm 1$, $\Delta m_I = \mp 1$) directions.

The range of magnetic fields in which nuclei are polarized dynamically as a result of the dipole–dipole interaction of nuclei with electrons trapped by deep levels is a function of the spin relaxation time of these electrons. This makes it possible to use optical pumping in weak magnetic fields in estimating the spin relaxation time of electrons localized at deep levels.

The authors express their gratitude to V. I. Perel' for a valuable discussion of the results and to R. A. Zhitnikov for his interest.

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Translated by A. Tybulewicz

Relaxation of longitudinal microwave magnetization in parametric excitation of spin waves in ferrites

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(Submitted 15 May 1978)
Zh. Eksp. Teor. Fiz. 75, 1755–1762 (November 1978)

We measured the relaxation frequency of the longitudinal macroscopic alternating magnetization produced in ferrites when spin waves are parametrically excited by parallel pumping. It is observed that this frequency always exceeds the spin-wave relaxation frequency determined from the threshold of the parametric excitation, and depends on many parameters such as the supercriticality, temperature, and sample diameter. To explain the experimental results, it is assumed that phase mismatch contributes to the damping of the longitudinal macroscopic magnetization. This mismatch is a result of the fact that the pump excites a packet of spin waves whose distribution over the eigenfrequencies has a finite half-width. The existing nonlinear theory of parametric excitation of spin waves in ferrites leads only to qualitative agreement with experiment, and great discrepancies appear in the quantitative estimates. The objects of the investigation were single-crystal spheres of yttrium iron garnet at a pump frequency 9370 MHz.

PACS numbers: 75.30.Ds, 75.50.Gg, 78.70.Gq

The most important parameter used in the analysis of parametric excitation of spin waves in ferrites is the relaxation frequency of parametrically excited spin waves (PSW) with wave vector $k-\gamma_k$.[1] Since the pump excites a PSW packet that is narrow in terms of k ($\Delta k \ll k$, Ref. 2), parametric spin-wave instability is described in the theory only by one characteristic time $T_k = 1/\gamma_k$, which determines, in particular, the threshold value of the microwave magnetic pump field h_{thr} at which spin-wave instability sets in.[3] However, the experiments described below show that a single time T_k cannot describe even all the macroscopic characteristics of parametrically regenerated ferrites.

We have measured the characteristic time $T_{kz} = 1/\gamma_{kz}$ of the free damping of longtudinal microwave magnetization $m_z(t)$ produced in the case of parallel pumping of spin waves in ferrites: $m_z(t) = m_z \exp(-2\gamma_{kz} t)$. The magnetization m_z is the projection of the vector of the microwave magnetization of the ferrite on the z axis, along which both the constant and alternating magnetic fields $\mathbf{H}_0$ and $\mathbf{h}$ are directed; m_z is given by[1]

$$m_z = \sum_k (V_k b_k b_{-k} \exp(2i\omega_k t) + \text{c.c.}), \quad (1)$$

where b_k is the complex amplitude of the spin wave with wave vector k, and ω_k is the PSW frequency and is equal to half the pump frequency ω_p. We note that it is precisely the appearance of m_z that makes possible absorp-

