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Optically induced nuclear magnetic resonance in semiconductors

V. K. Kalevich

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad (Submitted May 28, 1986)

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A magnetic resonance of the lattice nuclei in a semiconductor induced by light modulated at the NMR frequency was observed for the first time. It was found that a comparison of the NMR signals formed as a result of modulation of light and due to interaction with an rf field made it possible to determine the fields of optically polarized electrons at the individual isotopes in the lattice.

Optical orientation of electrons in a semiconductor subjected to a magnetic field is accompanied by the polarization of the lattice nuclei. 1-3 This effect is a consequence of lowering of the temperature of the nuclear spin system because of the contact with nonequilibrium photoelectron spins." The application of an alternating external magentic field to a cooled nuclear spin system increases its temperature and this is manifested by a reduction in the nuclear polarization (nuclear magnetic resonance). 1-3,5 It has been shown theoretically that an increase in the temperature of the nuclear spin systems can result also from the application of an alternating field of the hyperfine interaction H, created by polarized electrons at the nuclei. Such heating has been observed 7 in a weak external static magnetic field H ~ HL (here, HL is the local field at the nuclei amounting to v1 Oe). We shall report the first observation fo an NMR as a result of heating of the Zeeman reservoir of the nuclear spin system by an oscillating field of electrons in the presence of a strong magnetic field H >> HL. We shall also show that a comparison of the NMR signals induced by an alternating field of electrons and by an rf field makes it possible to determine the electron fields at the individual isotopes in the attice. We shall demosntrate that an electron field oscillating at a high frequency can be generated by modulating either the degree of circular polarization or the intensity of the exciting light.

 In GaAs-type semiconductors the electron field is formed mainly by the s electrons localized at shallow donors and in the presence of nuclei of me kind this field is described by3

$$H_{\epsilon}(r) = FSb_{\epsilon} \exp\left(-2r/a_{R}\right). \tag{1}$$

Here, F = n_d/N_d is the factor representing the occupancy of the donors by electrons (Nd is the mncentration of the donor centers and nd is the concentration of nonionized donors); S is the average electron spin; aB is the Bohr radius of electrons; r is the distance from the donor centers; be/2 is the field of totally polarized electrons in the case when F = 1 and r = 0.

It follows from Eq. (1) that the field He is proportional to F and S. In turn, the value of s is governed by the degree of circular polarization F, whereas F at a fixed temperature of a crysal is determined by the intensity I of the incident light. Moreover, the spin S may depend also on this intensity.2,8

It follows that the component of the electron field oscillating at a frequency Ω can be generated quite simply by modulating at this frequency either the degree of circular polarization or the intensity of the exciting light beam.

In our experiments such modulation of light was achieved by the linear electrooptic effect in a KDP crystal⁹ forming a part of a wide-band ML-3 modulator.

Our experiments were carried out on GaAs $(N_{Ge} \approx 4\cdot10^{16}~cm^{-3})$ and $Al_{0.26}Ga_{0.74}As$ $(N_{Zn} \sim 10^{18}~cm^{-3})$ crystals exhibiting p-type conduction and subjected to a magnetic field H, directed at an angle α to a beam of exciting light characterized by a circular polarization \mathscr{P} . In this experimental geometry the magnetic depolarization curve of the luminescence emitted by a crystal exhibited an additional maximum corresponding to the compensation by the external field H of the effective magnetic field Hs, created at electrons by the polarized nculei. Selection of the angle a can shift this maximum toward higher fields H ~ 1 kOe, as reported in Ref. 10 (continuous curve in Fig. 1).

The action of a magnetic field alternating at the NMR frequency and oriented at right-angles to the static field H. reduces the polarization of the nuclei and, consequently, reduces the field H_N . This reduction in the nuclear field alters greatly the value of p, if the field H is selected so that the derivative dp/dH is large. 11

By way of example, the dashed curve in Fig. 1 shows the resonances of $\rho(H)$ for a GaAs crystal obtained under the influence of an rf field. The NMR spectrum of this crystal recorded, in the course of slow variation of frequency of an rf field of amplitude $2H_1 = 0.02$ Oe, in a field H = 392 Oe is shown in Fig. 2a.

The NMR spectra obtained for a GaAs crystal as a result of modulation of the polarization and intensity of light with modulation depths m, and m_I amounting to 2.1% $m_{\mathcal{P}} = (\mathcal{P}_{max} - \mathcal{P}_{min})/(\mathcal{P}_{max} + \mathcal{P}_{min})$, $m_I = (I_{max} - I_{min})/(I_{max} + (min))$ are plotted in

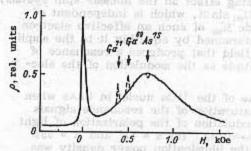


FIG. 1. Curves representing the depolarization of the luminescence emitted by a GaAs (N_{Ge} \approx 4·10¹⁶ cm⁻³) crystal in a magnetic field inclined at α = 77°; T = 1.9 K, S₀ = 0.068. The dashed curve was obtained in the presence of an rf field of frequency 509 kHz and amplitude 2H1 = 0.04 Oe.

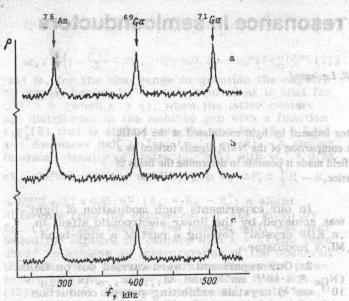


FIG. 2. Nuclear magnetic resonance spectra of a GaAs crystal obtained in the course of slow variation of the rf field frequency (a), modulation of the polarization of light (b), and modulation of the intensity of light (c). H = 392 Oe, a = 77°, T = 1.9 K, a) $2H_1 = 0.02 \text{ Oe}$; b) $m_{e} = 0.021 \text{ c}$ $m_{I} = 0.021$.

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Figs. 2b and 2c, respectively. The presence, as in the case of the spectrum in Fig. 2a representing the effects of the interaction with an rf field, of resonance signals due to all three isotopes in the crystal lattice and the fact that the signal/noise ratio is at least 10 shows that the resonance depolarization of the nuclear spin system by the oscillating electron field is very effective.

It is worth noting that modulation of light may be accompanied by the cooling of the nuclear spin system in an alternating electron field and by an increase in the nuclear polarization if the modulation frequency is close to the NMR frequency (resonance However, estimats indicate that when cooling). 12 the depth of modulation is of the order of ~1%, this effect is negligible.

3. According to Eq. (1), the electron field decreaes exponentially on increase in the distance r from the center of a donor. Its component oscillates in time and is transverse to the field H is represented by Hel(r)singt and responsible for the excitation fo the NMR, and it is also strongly inhomogeneous in the vicinity of a donor. However, in an analysis of the experimental results this component may be replaced by an equivalent (in respect of the depolarizing effect on the nuclear spin system) effective field Heisingt, which is independent of r. The amplitude Hel of such an effective electron field can be determined by equating it to the amplitude of the rf field that produces a resonance of the same magnitude as the modulation of the electron field.

In the case of the 71 Ga nuclei in GaAs when there was no saturation of the resonance signals representing modulation of the polarization of light with a depth $m_p=0.012$ for $\alpha=77^{\circ}$ and H=392Oe, and when the illumination power density was $W \approx 20 \text{ W/cm}^2$, we found that the field was $H_{e\perp} =$ (6 ± 3)·10⁻³ Oe.

In the experiment involving modulation of the intensity of light, which was carried out under the same conditions as the polarization modulation experiment, it was found that for $m_{J}=m_{J'}=0.012$ the field experienced by the 71 Ga nuclei in GaAs was Hel-(6 ± 2)·10⁻³ Oe. It should be pointed out that this field agreed, within the limits of the experimental error, with field Hel deduced by modulating the polarization.

The greatest interest lies in the determination, from the known values of the fields Hel, of the occupancy F of the donors by electrons and of the field be, because the product of these two quantities he = Fbe is a coefficient of proportionality between the electron field He and the average spin of electrons S [see Eq. (1)].

A comparison of the fields Hel found by modulation of the polarization and intensity I of the incident light can be used to estimate the factor F if the field He depends only on I via this factor. However, in the case of F for the investigated crystals an additional analysis was necessary because they exhibited a strong dependence of S and I, which in the case of p-type crystals cannot yet be explained in a satisfactory manner.

The coefficient he can be determined by modulation of the polarization of light. It can be found if the field He is divided by the corresponding amplitude $S_{\perp} = m_F S_{\perp}$, where S_{\perp} and S_{\perp} are the trnasverse components of the oscillatory and constant (in time) parts of the spin S. In practice, one measures the projection of S_Z (ρ = S_Z) of the spin S along the z axis, which is selected to be the direction of the exciting light beam. Applying the Bloch equation to the spin S in the combined field $(H+H_N)$ (Ref. 13), we readily obtain an an expression which relates S_{\perp} to S_Z : $S_{\perp}=S_0$. $[(S_Z/\bar{S}_0)-\cos^2\alpha]^{1/2}$, where $S_0=S_Z(H=0)$, We then find that

$$h_s = H_{s\perp}/S_{\perp} = H_{s\perp}/m_{\tilde{g}}S_0\sqrt{(S_s/S_0) - \cos^2 a}$$
 (2)

Substituting into Eq. (2) the values $S_Z/S_0 = 0.24$, $S_0 = 0.068$, $m_F = 0.012$ and the inclination of the field H amounting to $\alpha = 77^{\circ}$, used in the measurements of the field $H_{\rm el}$, we find finally that in the case of the $^{71}{
m Ga}$ nuclei in a GaAs crystal the field is Hel = 17 ± 8 Oe. We can similarly show that in the case of an Al $_{0.26}$ Ga $_{0.78}$ As crystal at T = 77 K for W \approx 20 W/cm² the field at the ⁷¹Ga nuclei is H_{el} = 59 ± 29 Oe.

It therefore follows that under optical orientation conditions a resonance depolarization of the nuclear spin system in a semiconductor may be due to modulated light. A comparison of the NMR signals induced optically and by an rf field makes it possible to determine the electron field at the individual isotopes in the crystal lattice.

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