Resonant photovoltaic effect in the NMR of nuclei in a semiconductor lattice

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The first experimental observation of NMR signals in the photocurrent due to the asymmetry of the scattering of optically oriented electrons in a III-V semiconductor is reported. The results of photovoltaic measurements are compared with data found from optical experiments.

The optical cooling of the nuclear spin system of a semiconductor during the orientation of electrons by light results in a nuclear polarization along the direction of the external magnetic field[1] **H**. The change in this polarization at NMR changes the effective hyperfine field $\mathbf{H}_N = a\beta\mathbf{H}$ (β is the inverse spin temperature of the nuclei, and a is a constant) exerted by the nuclei on the electron spins. The NMR can thus be detected indirectly in any effect in which the dependence of the average electron spin **S** on the magnetic field $\mathbf{H} + \mathbf{H}_N$ is manifested. In particular, an NMR can be detected successfully in a semiconductor by an optical method in experiments on the magnetic depolarization of recombination radiation.[2]

In a recently discovered surface photovoltaic effect due to an optical orientation

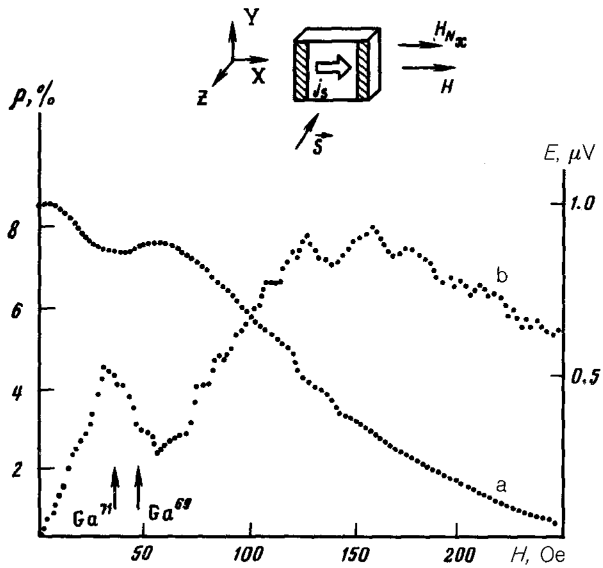

FIG. 1. a—depolarization of luminescence in a transverse magnetic field; b—the photovoltaic effect which arises at the surface of the crystal versus the magnetic field. The signal buildup times are 4 min for curve a and 2 h for curve b.

of spins,[3] there is a coupling of the momentum of electrons with their spin[4] which gives rise to a current along the surface of the semiconductor. In the same effect one might see evidence of resonant changes in the field $\mathbf{H}_N$ since the photocurrent depends on the orientation of the electrons.

It should be noted, however, that in the optical detection of NMR one "uses" electrons which recombine through donor centers and which interact with only strongly polarized lattice nuclei in the immediate vicinity of these centers. In a photovoltaic effect, in contrast, the conduction electrons which carry the surface photocurrent "sense" a considerably weaker field of the nuclei, averaged over the entire region of the crystal near the surface. Nevertheless, the results presented below show that even under the conditions of the photovoltaic effect there are resonant changes in the surface photocurrent during an NMR involving lattice nuclei.

The experimental geometry is shown in the inset in Fig. 1. The current of the asymmetric scattering along the surface of the semiconductor satisfies $j_S \propto [\vec{\nabla} s \times \mathbf{S}]_X$ and is proportional to[3] S_Y. Here $\mathbf{S}$ is the average spin of the electrons, and $\vec{\nabla} s$ is the gradient of the spin density which arises from the spin diffusion into the interior of the crystal.

To eliminate photovoltaic effects which are not associated with the spin orientation of electrons, we modulate the circular polarization of the light with the help of quartz modulators working at the frequencies 30 and 50 kHz. An effective polarization of the nuclei in field $H > H_L$ ($\mathbf{H}_L$ is the local field of the nuclei) is possible only during a resonant cooling of the nuclear spin system in a field of electrons, $\mathbf{H}_e \propto \mathbf{S}$, which is oscillating at a frequency ω_0 close to the NMR frequency.[5]

The resonant cooling is accompanied by the appearance of a field $\mathbf{H}_N$, despite the rapid change in the polarization direction of the electrons. Experimentally, the constant component of this field, H_{N_X} (along $\mathbf{H}$) is manifested; this component depends on H in accordance with the dispersion law

$$H_{N_X} = h_N \frac{(H_0 - H)S^2 \sqrt{H_e^2 + H_L^2}}{(H_0 - H)^2 + H_e^2 + H_L^2}, \tag{1}$$

where $H_0 = \omega_0/\gamma_I$, γ_I is the nuclear gyromagnetic ratio, and h_N is a constant. This field affects the average electron spin $\mathbf{S}$. During optical detection of the NMR and also in the photovoltaic effect, different projections of $\mathbf{S}$ are manifested. In a magnetic field $\mathbf{H} + \mathbf{H}_N$ these projections are described by

$$S_Z = S_0 / [1 + (H + H_{N_X})^2], \tag{2}$$

$$S_Y = S_Z(H + H_{N_X}). \tag{3}$$

Here S_0 is the average spin of the electrons in the absence of a magnetic field, and here and below the magnetic field is expressed in units of $\mu_e T_S/\hbar$, where μ_e is the magnetic moment, and T_S the duration of the spin orientation, of the electrons.

The projection S_Z is proportional to ρ, the degree of circular polarization of the recombination radiation, and S_Y determines the asymmetric scattering current j_S. The most favorable conditions for observing this current hold near the temperature of liquid nitrogen. To observe the effect of nuclear polarization on the photovoltaic effect, we selected the crystal $Ga_{0.73}Al_{0.27}As$ doped with Zn ($N_{Zn} = 10^{18}$ cm^{-3}), in which the nuclear field H_N is manifested in optical measurements at temperatures below 90 K. Figure 1 shows the experimental results obtained with this crystal at 77 K and at an exciting-light wavelength of 632.8 nm. To improve the signal-to-noise ratio, we used a signal-buildup technique. Curve *a* shows the behavior of the depolarization of the luminescence in a transverse magnetic field, while curve *b* shows the signal of the photovoltaic effect which arises in the crystal. In both cases we observe structural features near resonant values of the field H (at the arrows) which are caused by the resonant cooling of the spin system of the lattice nuclei. These structural features have the characteristic shape of dispersion curves with a center at $H_0 = \omega_0/\gamma_I$. As the frequency ω_0 is varied, the position of the dispersion curve changes correspondingly. We restrict the analysis below to the behavior $\rho(H)$ and $j_S(L)$ at fields corresponding to the NMR of the lattice nuclei. Figure 2 shows the results of more-accurate measurements in this region. The solid curves were calculated from (1)–(3) with allowance for the field H_{N_X} produced by the Ga^{69} and Ga^{71} nuclei. Although all the lattice nuclei in the GaAlAs crystal have a magnetic moment, only the nuclear field of the Ga isotopes is manifested in resonant cooling. The effect of the other lattice nuclei is weakened significantly by a quadrupole interaction.[6] The NMR lines of the isotopes Ga^{69} and Ga^{71} on the curves of $\rho(H)$ and $j_S(H)$ in Figs. 1 and 2 (a and b) merge. The primary reason for this effect is that the superposition of two closely spaced NMR lines having the shape of dispersion curves is an unfavorable circumstance for a visual separation of these lines; a second factor is the broadening of the NMR lines caused by

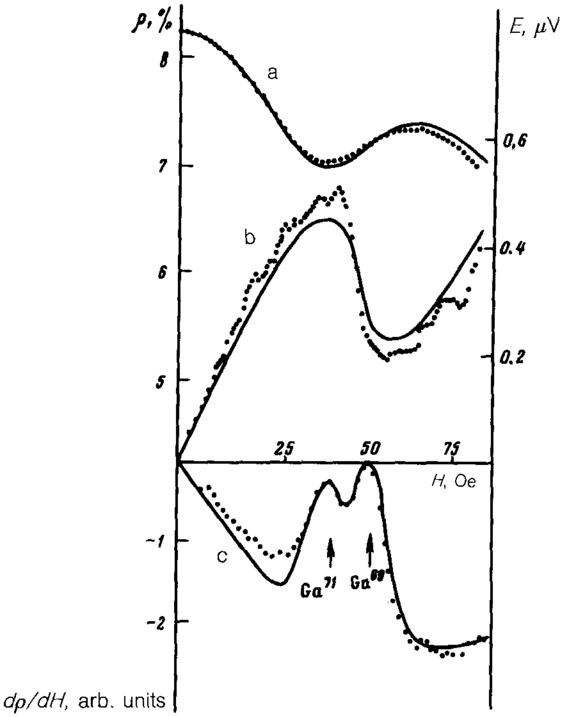

FIG. 2. a—depolarization of luminescence; b—photovoltaic signal; c—the derivative $d\rho/dH$ versus the magnetic field. The solid lines show results calculated from Eqs. (1)–(3) for the following parameter values: a,b—$h_N(\mathrm{Ca}^{69}) = 2.63$ kOe, $h_N(\mathrm{Ca}^{71}) = 2.25$ kOe, $H_e^2 + H_L^2 = 352$ Oe^2; c—$h_N(\mathrm{Ca}^{69}) = 917$ Oe, $h_N(\mathrm{Ca}^{71}) = 792$ Oe, $H_e^2 + H_L^2 = 69$ Oe^2.

the oscillating field of the electrons, H_e, which plays the role of an alternating magnetic field.

The ample signal-to-noise ratio in optical measurements made it possible to carry out experiments with a shallow modulation of the degree of circular polarization of the exciting light; specifically, the modulation was considerably less than would be required for observation of the photovoltaic effect. The average electron spin **S** and, correspondingly, the electron field $\mathbf{H}_e$ and the half-width of the NMR lines are smaller. Furthermore, a slight modulation of the field H was used to measure the derivative $d\rho/dH$, so that it became possible to further improve the conditions for resolving the NMR lines of the isotopes Ga^{69} and Ga^{71} (Fig. 2c). The solid line in Fig. 2c was found by differentiating Eq. 2 with respect to H. It can be seen from this figure that there is a good agreement betweeen the theoretical and experimental results at $H > 25$ Oe. The discrepancy at $H < 25$ Oe is due to a nonresonant cooling of the nuclear spin system in the oscillating electron field, which is manifested in weak fields[7] but which was ignored in the calculation of the $d\rho/dH$ curve.

The observation of a resonant change in the photocurrent at the NMR frequency is evidence of an effective spin exchange between conduction electrons and trapped electrons, with the result that their average spins rapidly become the same. The change in **S** of the trapped electrons, which interact far more effectively with the lattice nuclei, causes the corresponding change in the average spin of the conduction electrons.

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