Quadrupole effects and singularities of NMR of optically oriented nuclei in semiconducting solid solutions

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(Submitted July 6, 1977)
Pis'ma Zh. Eksp. Teor. Fiz. **26**, No. 4, 316–319 (20 August 1977)

We have observed resonant transitions of optically oriented nuclei, with frequencies that vary sinusoidally, when the crystal is rotated, between zero and thrice the ordinary NMR frequency. The resonances are due to violation of the cubic symmetry when gallium is replaced by aluminum in the solid solution GaAlAs.

PACS numbers: 76.60.Gv

The numerous papers on optical orientation of electron and nuclear spins in semiconductors do not contain as yet any reports of the manifestation of the crystal-field anisotropy. In recent investigations of optical orientation in GaAlAs solid solutions we have observed a number of strong anisotropic effects. Study of NMR on optically oriented nuclei has shown that nuclear quadrupole interaction appears in the optical channel. The NMR was registered optically by observing the change of the degree ρ of the luminescence circular polarization following application of a weak alternating field at the resonant frequencies. In the investigated crystals, ρ is numerically equal to the projection $\langle S_z \rangle$ of the average electron spin on the observation direction. When nuclear polarization appears, the electron spins are acted upon by the effective magnetic field H_N of the nuclei.[1,2] Using conditions under which this influence turns out to be strong, we can register NMR with high sensitivity by varying the field at the resonance frequencies.

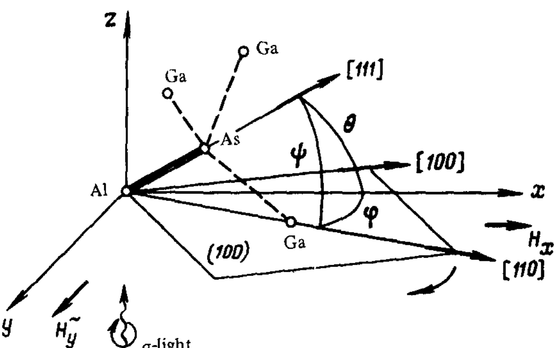

FIG. 1. Experimental geometry. The case is shown when one Ga atom closest to the As atom is replaced by Al. The notation is explained in the text.

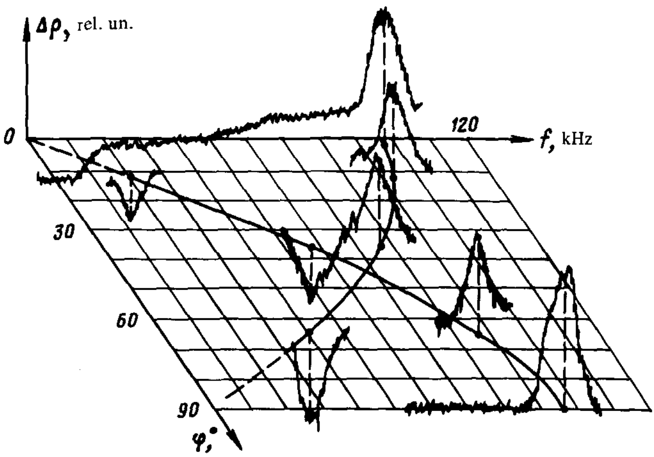

FIG. 2. Changes $\Delta\rho$ of the degree of circular polarization of the luminescence as functions of the angle ϕ and of the frequency f of the alternating field $H_y^{\sim}$. The constant field $H_x = 57$ Oe. The temperature is 77 K.

Figure 1 shows the geometry of the experiment. The exciting circularly polarized (σ) beam of an He-Ne laser propagates along the z axis perpendicular to the crystal surface [the (100) plane]. A constant magnetic field H_x is applied along the x axis. An alternating magnetic field $H_y^{\sim}$ acts along the y axis. The crystal is rotated around the z axis and the NMR spectra are registered for different angles ϕ between the [110] axis and the x axis. The measurements were made on p-$Al_{0.24}Ga_{0.76}As$ crystals at 77 K.

The electron spins precess in the combined field $\mathbf{H}_x + \mathbf{H}_N$. The field $\mathbf{H}_N$ is varied at the NMR frequencies and with it the rate of electron-spin precession and the projection $\langle S_z \rangle$. Changes ($\Delta\rho$) are then observed in the circular polarization of the luminescence, as shown in Fig. 2. It must be noted that the structure of the spectrum depends strongly on the field H_x and on the crystal orientation. Figure 2 shows lines whose motion was reliably traced in a wide range of variation of the fields H_x and the angles ϕ. In individual regions of H_x and ϕ, additional details of the spectrum are observed, including both broad smeared lines and well-resolved ones. We confine ourselves in this communication to the behavior of the individual resonance lines shown in Fig. 2. The frequencies of these resonances vary linearly with the field H_x. Attention must be called to the good discrimination of the resonance lines when the NMR is optically recorded in a relatively weak field (several dozen oersteds). NMR spectra of semiconductors were heretofore registered optically in fields exceeding a kilooersted.

The NMR was optically detected directly by means of the dependence of $\langle S_z \rangle$ on $\mathbf{H}_N$. The function $\langle S_z \rangle$ is very complicated[3] that depends on the field H_x on the light intensity, and on the crystal orientation. We cannot dwell here on the forms and signs of the resonance signals. We note only that the NMR spectra of Fig. 2 contain low-frequency and high-frequency branches, with the low-frequency branch corresponding to a decrease of ρ (to negative $\Delta\rho$), and the high-

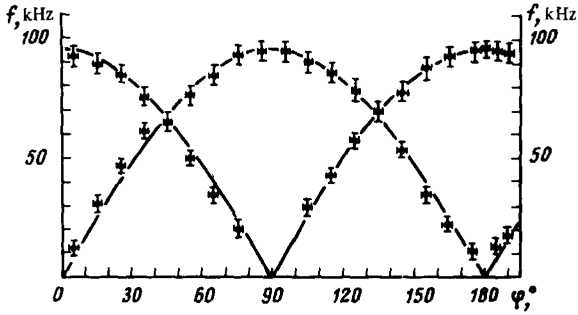

FIG. 3. Angular dependences of the resonant frequencies for the As^{75} nuclei. $H_x = 57$ Oe. Temperature 77 K.

frequency branch to positive $\Delta\rho$. In weaker fields H_x, positive $\Delta\rho$ are observed also for the low-frequency branch. The dependence of the NMR signal on the amplitude of the field $H_y^{\sim}$ turns out to be quite complicated. A strong nonlinearity is observed in weak fields $H_y^{\sim}$. In stronger $H_y^{\sim}$ (on the order of one oersted and higher) a smoothing of the NMR spectra and a reversal of the sign of $\Delta\rho$ are observed in a certain range of the fields H_x. The frequency dependences cited by two of us[3] are then observed.

Figure 3 shows the experimental resonant frequencies as functions of the angle ϕ. The solid lines are segments of sinusoids.

The cause of such angular dependences is partial replacement of gallium by aluminum in the GaAlAs solid solutions. The cubic symmetry of the unit cells is then distributed, and noticeable field gradients and associate quadrupole effects appear. In the investigated compounds, on the average, one of the four Ga atoms closest to As^{75} was replaced by Al. The Ga—Al and As—Al atomic bonds are oriented along the body diagonals of a cube. Thus, field gradients acting on the As^{75} nuclei appear in these directions in various cells.

The maximal frequencies on Fig. 3 correspond to directions of the field H_x along the diagonals of the cube faces. These frequencies differ from the value $3\gamma_I H_x$ (γ_I is the gyromagnetic ratio for the As^{75} nuclei) by a factor ≈ 0.82 corresponding to the value of $\cos\psi$ for ψ equal to the angle between the axes [110] and [111]. Inclination of the magnetic field relative to the z axis in a way that the angle between the field and the [111] axis decreases is accompanied, as expected, by an additional increase of the resonance frequency.

Our results thus attest to the presence of resonant transitions between levels split in an external field, with magnetic quantum numbers $\pm 3/2$. This splitting is much less than the quadrupole splitting between the levels 1/2 and 3/2, so that the nuclear Zeeman energy enters in the spin Hamiltonian in the form of a term proportional to $H\cos\theta$, where θ is the angle between the direction of the crystal-field gradient and the external magnetic field. Under conditions when the electrons are optically oriented, the hyperfine interaction produces either predominantly $+1/2 \to +3/2$ transitions or $-1/2 \to -3/2$ transitions, depending on the sign of the circular polarization of the exciting light. The predominant population of one of the 3/2 levels causes the field $\mathbf{H}_N$ to have to component that acts on the electron spin, on top of the component due to cooling of the nuclear spin system in the electron field.[3] The equalization of the populations in the slow passage through the resonances alters the field $\mathbf{H}_N$ and consequently the value of ρ. Transitions between the $\pm 3/2$ levels become possible because of the admixture of states with quantum number 1/2 at $\theta \neq 0$. The two frequency branches on Fig. 3 are connected with the contributions of two pairs of cube body diagonals located in mutually perpendicular planes. The resonant frequencies change with changing ϕ like $3\gamma_I H_x \cos\psi \cos\phi$ for one pair and like $3\gamma_I H_x \cos\psi \sin\phi$ for the other.

Analogous resonant transitions are observed also for the isotopes Ga^{69} and Ga^{71}, but they are less pronounced and more difficult to trace, probably because of the smaller dipole moment of these nuclei, the larger interatomic Ga—Al distance, and the fact that each Ga nucleus has several nearest neighbors replaced by Al.

Just as this communication was submitted, we learned that analogous transitions had been observed by V. L. Berkovits and V. I. Safarov.

The authors thank M. I. D'yakonov, V. I. Perel', and I. A. Merkulov for discussions.

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