Optical observation of multiphoton NMR of absorption in semiconducting crystals

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(Submitted July 13, 1977)
Pis'ma Zh. Eksp. Teor. Fiz. **26**, No. 5, 377–381 (5 September 1977)

We have observed, for the first time, multiphoton (up to 5 photons) NMR absorption in a solid, in an investigation, by the optical-orientation method, of quadrupole-perturbed NMR transitions of As^{75} in mixed GaAs–AlAs crystals.

PACS numbers: 76.60.Gv

A rigorous analysis of the Schrödinger equation in the theory of magnetic resonance shows that when a spin wave is acted upon by an oscillating field H_1 which is not rotating but is linearly polarized, it is possible to have, besides the fundamental resonance at the frequency ω_0, additional resonances at the multiple frequencies $\omega=\omega_0/n$; in the general case n is an arbitrary integer.[1] This corresponds to the transitions with simultaneous absorption of "n" RF photons.[2] These resonances, however, are much weaker than the fundamental one. Estimates show that for $n=3$, for example,[3] this attenuation is by a factor of the order of $(\gamma H_1/\omega_0)^2$, where γ is the gyromagnetic ratio. At realistically attainable values of H_1 (several gausses), such transitions can obviously be observed only for resonance at low frequencies, and this poses considerable difficulties when ordinary NMR methods are used.

New possibilities for the investigation of resonances in weak fields are provided by optical means based on the use of the method of optical orientation in semiconductors.[4–7] An appreciable nuclear magnetization is attained here on account of the dynamic polarization of the lattice nuclei by the spin-oriented nonequilibrium photoelectrons. In addition, because of the hyperfine interaction, the polarized nuclei exert a strong reaction on the spin orientation of the electrons, which can be easily detected optically. This makes it possible, in particular, by following the electron orientation with the aid of the polarization of the recombination luminescence, to detect optically magnetic resonance of nuclei of a semiconductor lattice.[4,5]

In the present study we have observed, by an optical method, multiphoton absorption on the quadrupole-perturbed transition of As^{75} (spin 3/2) in the solid solutions $Ga_{0.9}Al_{0.1}As$ at 4.2 K. In the geometry wherein the external field H_0 is perpendicular to the exciting light beam, and no ordinary NMR transitions are observed,[6] we have observed two lines whose frequencies were dependent on the crystal orientation relative to H_0. These lines are interpreted as transitions $|+3/2\rangle - |-3/2\rangle$ of the As^{75} nucleus in the presence of a strong quadrupole perturbation along the $\langle 111\rangle$ crystallographic direction. The quadrupole perturbation splits the fourfold-degenerate level of As^{75} into two. An additional doublet splitting of the levels takes place in a weak external magnetic field. The Zeeman splitting of the quadrupole level $|\pm 3/2\rangle$ depends

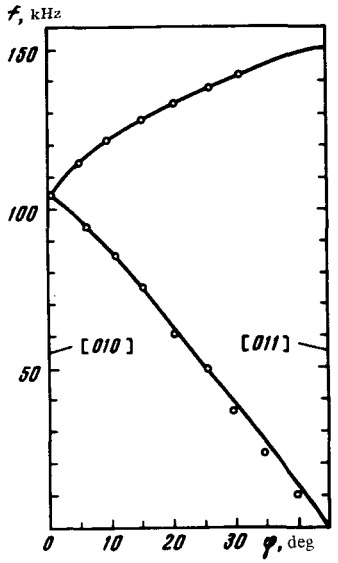

FIG. 1. Position of quadrupole-perturbed resonances $|+3/2\rangle - |-3/2\rangle$ of the As^{75} nucleus as a function of the angle θ between the directions of the magnetic field and the [010] axis, when the crystal is rotated in the (100) plane. $H_0 = 82$ G. Solid line—calculation.

on the angle θ between H_0 and the axes of the electric field gradient[8] (in our case, between H_0 and the $\langle 111 \rangle$ axis):

$$\omega = 3\gamma H_0 \cos\theta. \tag{1}$$

The crystal has four nonequivalent $\langle 111 \rangle$ directions. In our experiments, the external magnetic field and the RF field perpendicular to it were oriented in the (100) plane, which coincided with the plane of the surface of the sample. For this orientation of H_0, the angle θ is different for the two pairs of $\langle 111 \rangle$ directions. The experiment revealed in fact two lines. Figure 1 shows the dependence of the line frequency on the crystal orientation in the case of rotation in the (100) plane. The solid lines in Fig. 1 are the results of a calculation based on formula (1) for As^{75}. The splendid qualitative and quantitative agreement uniquely identifies the lines as transitions between the levels $|+3/2\rangle - |-3/2\rangle$ of the As^{75} nucleus with strong quadrupole perturbation along $\langle 111 \rangle$. This perturbation can be due, for example, to replacement of one of the four gallium atoms adjacent to the arsenic in the solid solutions by aluminum. These transitions will be considered in detail in a separate paper.[1)]

The intensities of the observed lines varied with the crystal orientation—the lines were made relatively stronger by shifts to lower frequencies. Thus, in this system, even in the presence of an appreciable external field (~ 100 G), when all the remaining NMR transitions of the nuclei of the host lattice are in the region of sufficiently high frequencies, a suitable choice of the crystal orientation permits observation of an intense isolated line at low frequencies. Figure 2 shows a typical optically registered spectrum—i.e., the dependence of the polarization of the crystal luminescence on the frequency of the applied RF field. In the presented frequency region of low RF power (curve a) one can see the two above-described lines of the quadrupole-perturbed transition of the As^{75} nucleus. With increasing RF intensity (curves b and c),

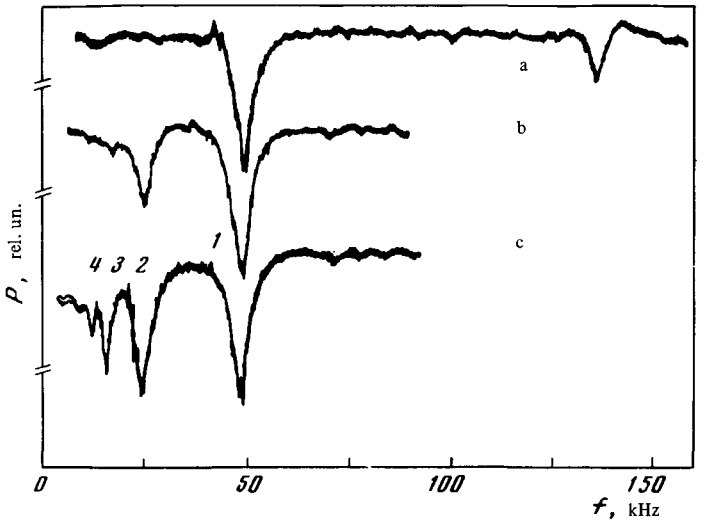

FIG. 2. Dependence of the degree of circular polarization of the luminescence of a $Ga_{0.9}Al_{0.1}As$ crystal on the frequency of the alternating RF field. $H_0 = 82$ G. $\phi = 27°$. The spectra were obtained for different RF-field powers increasing from "a" to "c."

the low-frequency line reaches saturation rapidly, and additional lines appear on its low-frequency side. The frequencies of these lines are smaller by a factor two, three, four··· than the fundamental frequency, and these additional resonances are naturally interpreted as multiphoton transitions, i.e., transitions with simultaneous absorption of two, three, ··· RF quanta. When the orientation of the crystal is changed (or the external field is changed), the additional resonances are also shifted together with the fundamental one; their frequencies remain at all times multiples of the frequency of the fundamental resonance. That is to say, the ratio $\omega_n/\omega_0 = n$ is preserved.

One might assume that the observed resonances are single-photon transitions due to multiple (2ω, 3ω, ···) harmonics of the generator. However, by placing in the supply circuit of the RF coil a tunable narrow-band filter it could be unequivocally determined that the observed resonances are due precisely to the fundamental frequency of the generator. Thus, for example, a filter that suppresses the frequencies ω_n decreased strongly precisely this resonance in the spectrum, leaving the intensities of the remaining ones practically unchanged.

For ordinary NMR transitions, an analysis of the selection rules shows that at $H_1 \perp H_0$ only odd resonances should be observed (absorption of an odd number of RF quanta), and the even ones can appear if a component H_1 parallel to H_0 is present.[1] These simple selection rules, however, cannot be directly applied to the more complicated case of quadrupole-perturbed transitions investigated by us, where it appears that it is already necessary to take into account the relative orientations of H_1, H_0, and the electric field gradient.

The authors thank D. Z. Garbuzov for supplying the epitaxial GaAlAs layers for our measurements and M. I. D'yakonov for certain discussions.

[1)]We have learned that similar transitions were observed by B. P. Zakharchenya, V. A. Novikov, and V.G. Fleisher [see JETP Letters **26**, 316 (1977)].

[1]J. Winter, C.R. Acad. Sci. **241**, 375 (1955).
[2]J. Margerie and J. Brossel, C.R. Acad. Sci. **241**, 373 (1955).
[3]A. Abragam, Principles of Nuclear Magnetism, Oxford, 1961, Chap. 2.
[4]A.I. Ekimov and V.I. Safarov, Pis'ma Zh. Eksp. Teor. Fiz. **15**, 453 (1972) [JETP Lett. **15**, 319 (1972)].
[5]V.L. Berkovits, A.I. Ekimov, and V.I. Safarov, Zh. Eksp. Teor. Fiz. **65**, 346 (1973) [Sov. Phys. JETP **38**, 169 (1973)].
[6]M.I. D'yakonov, V.I. Perel', V.L. Berkovits, and V.I. Safarov, Zh. Eksp. Teor. Fiz. **67**, 1912 (1974) [Sov. Phys. **40**, 950 (1975)].
[7]V.A. Novikov and V.G. Fleĭsher, Zh. Eksp. Teor. Fiz. **71**, 778 (1976) [Sov. Phys. JETP **44**, 410 (1976)].
[8]A. Abragam, Principles of Nuclear Magnetism, Oxford, 1961, Chap. 7.