Direct observation of a nuclear spin resonance in an oscillating electric field

I. I. Sadykov and E. P. Khaĭmovich
Physicotechnical Institute, Kazan Academy of Sciences of the USSR

(Submitted 15 April 1981; resubmitted 14 August 1981)
Pis'ma Zh. Eksp. Teor. Fiz. **34**, No. 8, 441–444 (20 October 1981)

The very small variation of dielectric losses ($\tan\delta \gtrsim 10^{-9}$) caused by resonance energy absorption by the nuclear spin system has been measured for the first time. The effect is called nuclear electric resonance (NER). The R_{14} constants, which connect the applied electric field with the field gradient of the nuclei of a GaAs crystal induced by this field, are determined.

PACS numbers: 77.85. + x, 77.40. + i, 76.90. + d

The effect of electric fields on Zeeman nuclear levels has been studied in Refs. 1–3. Since the electric field does not have a direct effect on nuclei, the authors of Ref. 1 assumed that the field E_k, as a result of displacing the lattice charges, produces a gradient V_{ij} at the nucleus, which interacts in turn with the quadrupole moment Q_N of the nucleus.

Two methods have been used so far to observe this effect: by splitting the NMR line by a static field[1,3] and by saturating the NMR signal by a resonance-frequency variable electric field.[2]

In this letter we are reporting the first observation of direct absorption of electric-field energy by the nuclear spin system at transition frequencies $\Delta m = \pm 2$. We shall call this effect nuclear electric resonance (NER). To observe NER, we used a nuclear acoustic resonance (NAR) spectrometer, which was developed elsewhere[4] and refined by us. The refinement involved the replacement of an acoustic cavity by an electric resonant LCR circuit (Fig. 1). A $0.3 \times 1.6 \times 1.6$-cm, high-resistance ($5 \times 10^7$ Ω cm) GaAs crystal sample to be studied was placed between two plates of

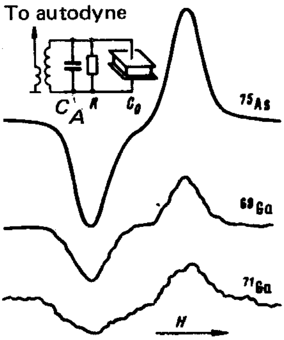

FIG. 1. Traces of nuclear electric resonance signals in GaAs and the diagram for the connection of the electric curcuit with the sample to the autodyne. $C_0 = 7.2$-pf capacitor with the sample, $C_A = 3.2$-pf supplementary capacitor of the assembly, $Q_C = 1100$, and $\nu = 16.68$ MHz. The integration constant is 10 sec.

a condenser C_0.

The field E_C was applied along the piezoelectrically inactive [001] axis of the crystal. To increase the sensitivity of this method, we immersed the circuit with the sample into a helium cryostat. Figure 1 is a trace of the NER signals from ^{75}As, ^{69}Ga, and ^{71}Ga nuclei recorded at 16.68-MHz electric field frequency in magnetic fields of 11.44, 18.15, and 6.42 kHz, respectively. The magnetic field was modulated by a square-wave current with a 6.6-Hz frequency and a modulation amplitude that exceeded the NER line width by a factor of 2–3. To interpret the results obtained by us, we shall use the phenomenological approach developed in Ref. 1.

In the linear approximation in E_C we have

$$V_{ij} = \sum_k R_{ijk} E_C + \sum_{l\,m} \sum_k S_{ijlm} d_{lmk} E_C . \quad (1)$$

The gradient V_{ij} is determined primarily by the first term that contains a tensor of rank three R_{ijk}. The second term takes into account the inverse piezoelectric effect which is proportional to the piezoelectric modulus d_{lmk} and to the coupling tensor S_{ijlm}. The contribution of this term to the GaAs sample under investigation is $\leqslant 1\%$ and can be neglected. We note that the R_{ijk} and d_{lmk} tensors are nonvanishing only for crystals without a center of inversion. At high field intensities the terms nonlinear in E_C should be included in (1). Our estimates and the results of the experiment[3] show, however, that the effect of quadratic terms (electrostriction effect[5]) is negligible in fields $\leqslant 2000$ V/cm. We shall therefore use $V_{ij} = \Sigma_k R_{ijk} E_k(t)$ in the subsequent calculations, where $E_C(t)$ is the oscillating field.

The dielectric properties of the material are characterized by the loss tangent, $\tan\delta$. We shall therefore express the NER absorption in terms of $\tan\delta_N$. We see that

$$\tan\delta_N = \frac{P_N(C_o + C_A)}{P_E\, Q_E C_o} , \quad (2)$$

TABLE I.

Isotopes	$\tan\delta_N$, 10^{-9}	$\Delta\nu$, kHz	Q_N, 10^{-24}cm^2	R_{14}, 10^{16} cm^{-1}		
					Ref. 1	Ref. 2
^{75}As	93 ±6	4.34 ±0.15	0.30	1.56	1.55	2.0
^{69}Ga	18 ±1.3	3.85 ±0.13	0.19	1.35	1.05	1.5
^{71}Ga	3.5 ±0.3	5.0 ±0.17	0.12	1.28	0.9	1.5

where P_N and P_E is the power absorbed by the spin system and the electric LCR circuit, and Q_E is the circuit's quality factor. A quantum-mechanical calculation of P_N with allowance for (1) is analogous to the calculation of the absorption power of ultrasonic energy during a nuclear acoustic resonance.[6] Thus we have

$$\tan\delta_N = \frac{[R_{ijk}K(\theta)]^2}{4\epsilon_0}\ \frac{N\nu(\pi e Q_N)^2 g(\nu)}{kT}\ F(J, m). \qquad (3)$$

Here ϵ_0 is the dielectric constant, N is the number of spins per unit volume, ν is the frequency, $g(\nu)$ is the shape function, $F(J, m)$ is a function that is determined by the nuclear spin and by the transition $\Delta m = \pm 1, \pm 2$, and $K(\theta)$ determines the orientational dependence of NER. It follows from the symmetry properties of the GaAs crystal that there is only one, nonvanishing element of the R_{14} tensor (in Vogt notation).

Table I gives the values of $\tan\delta_N$, the NER line width at half intensity, and the values of Q_N used in the calculations. For a comparison, the values of R_{14} determined by using other methods are given. The ratio P_N/P_E in the Eq. (2) was measured from the amplitude A_s of the NER signals and from the amplitude A_C of the

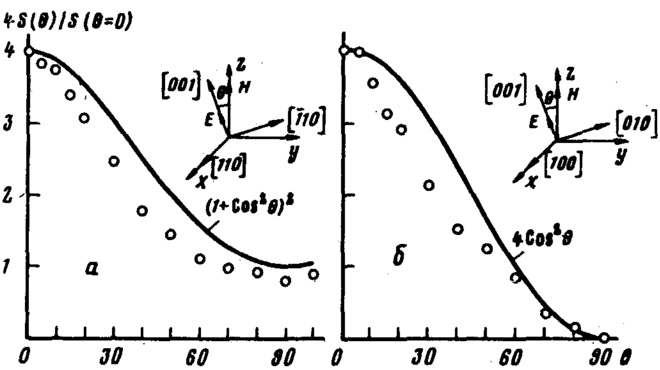

FIG. 2. Experimental points and theoretical dependences of the area under the curve for the NER signal on the angle θ for two orientations of the crystal axes and for the electric field (E) and magnetic field (H) with respect to the laboratory coordinate system.

calibrator's signal, with allowance for the equivalent resistance R_e of the LCR circuit and the equivalent resistance ΔR_C of the calibrator. The indicated parameters are related by the relation[4]

$$\frac{\Delta R_E}{R_E} = \frac{P_N}{P_E} = \frac{A_S}{A_C} \frac{R_E}{\Delta R_C}.$$

The measurement accuracy of R_{14} is determined primarily by a generally large error of Q_N.

This method was also used to determine the angular dependence of the signal cross section $S(\theta)$ (see Fig. 2). The experimental data are found to be in qualitative agreement with the theoretical dependence. This agreement, moreover, is better in the direct NER than in the saturation method.[2] The high measurement accuracy and the fact that the low rf power ($< 10^{-6}$ W) supplied to the sample completely eliminates the nonlinear effects are two important advantages of the proposed method.

The authors wish to thank V. K. Nurmukhametov for furnishing the sample.

1. D. Gill and N. Bloembergen, Phys. Rev. 129, 2398 (1963).
2. E. Brun, R. J. Mahler, H. Mahon, and W. L. Pierce, Phys. Rev. 129, 1965 (1963).
3. K. A. Dumas, J. F. Soest, and A. Sher, Phys. Rev. B 20, 4406 (1979).
4. I. I. Sadykov, Prib. Tekh. Eksp. No. 1, 129 (1978); No. 6, 92 (1978).
5. I. S. Zheludev, Osnovy segneto élektrichestva (Principles of Ferroelectricity), Atomizdat, Moscow, 1973.
6. D. I. Bolef, Physical Acoustics (W. P. Mason, ed.) 4A, Academic, NY, 1967.

Translated by S. J. Amoretty
Edited by Robert T. Beyer