Electrically Induced Nuclear Quadrupole Spin Transitions in a GaAs Single Crystal*†

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The direct induction of nuclear $E2$ spin transitions in a gallium arsenide single crystal by application of an external oscillatory electric field has been previously reported by some of the authors. This paper gives the results of a further investigation of the same phenomenon. Theoretical expressions are given for the equilibrium nuclear magnetization in a crystalline lattice under the combined influence of a static magnetic field, externally induced electric field gradients, and thermal spin-lattice interactions, for various relative orientations of the applied fields and the crystalline symmetry axes. The theoretical predictions were tested for the three nuclides Ga^{69}, Ga^{71}, and As^{75} by using pulsed nuclear magnetic resonance techniques to sample the equilibrium magnetization in a GaAs crystal at 77°K under the influence of a uniform, externally applied, radio-frequency, electric field. A description of the experimental apparatus is given. The dependence of the quadrupolar saturation on both the electric field amplitude and the crystal orientation was measured. The angular dependence of the saturation was found to be in reasonable agreement with the theory.

I. INTRODUCTION

A SYSTEM of nuclear spins with associated $M1$ and $E2$ moments, imbedded in a crystalline lattice and placed in a static magnetic field, has definite orientational energy levels. If the charge environment of such nuclei generates time-varying electric field gradients at the nuclear sites, there will be, in general, time-varying torques exerted on the nuclei. If this time variation contains the right frequency components, transitions may be induced between the various Zeeman levels of the nuclei. It is, in fact, the random relative motion of the lattice charges due to thermal lattice vibrations which in many cases induces nuclear spin transitions through quadrupolar interactions, leading to relaxation of the nuclear spin system.

In 1956 Proctor and Robinson[1] observed externally induced nuclear spin transitions caused by nuclear electric quadrupoles interacting with ultrasonically excited electric field gradients in a NaCl lattice. In 1961 Bloembergen[2] suggested that in a lattice where local inversion symmetry is lacking, one should be able either to cause a static first-order $E2$ shift or induce first-order $E2$ spin transitions by application of external electric fields, although Gutowski and Williams[3] in 1957 had tried without success to observe a static shift of the Cl^{35} quadrupole resonance frequency in $NaClO_3$, where local inversion symmetry about the Cl nuclei is lacking. Kushida and Saiki[4] then observed a static induced shift of the Br^{81} $E2$ resonance in $NaBrO_3$ under the influence of an external electric field, and, at about the same time, Armstrong, Bloembergen, and Gill[5] observed a similar effect on the Cl^{35} $E2$ resonance in $KClO_3$ and $NaClO_3$. This was followed by the observation of an induced static electric splitting of the Ga^{71} nuclear magnetic resonance (NMR) line in a GaAs single crystal by Gill and Bloembergen.[6] Brun *et al.*[7] then observed externally induced $\Delta m=\pm 2$ transitions of the Ga^{69}, Ga^{71}, and As^{75} nuclear spins in a GaAs crystal, by direct application of an oscillatory electric field at twice the Larmor frequency.

Gallium arsenide is a nearly ideal substance for such experiments. The point symmetry is tetrahedral, giving a first-order shift in the crystalline electric field gradient components with an applied electric field, but no zero-order gradient. GaAs single crystals are available with high electrical resistivity, in contrast to most of the other III-V compounds, which have the same structure. Furthermore, all three nuclear species in GaAs are easily observable, have the same spin ($I=3/2$), and moderately large $E2$ moments.

II. THEORY

Equilibrium Magnetization

In order to observe the effect of induced $E2$ transitions by sampling the nuclear magnetization of the crystal, a static external magnetic field H_0 is applied in the z direction. Before any perturbation is turned on, the magnetization of the sample, due to one species of nuclei, is

$$M_z=M_0=\chi_0 H_0=N\gamma^2 I(I+1)\hbar^2 H_0/3kT,$$

where N is the number of spins per unit volume, T is the temperature, and γ is the magnetogyric ratio. If we now turn on a perturbing E field, at a frequency $\omega=2\gamma H_0$ (twice the Larmor frequency), inducing $\Delta m=\pm 2$ transitions, and furthermore assume that a

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[1] W. G. Proctor and W. A. Robinson, Phys. Rev. **104**, 1344 (1956).
[2] N. Bloembergen, Science **133**, 1363 (1961).
[3] H. S. Gutowski and G. A. Williams, Phys. Rev. **105**, 464 (1957).
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[6] D. Gill and N. Bloembergen, Bull. Am. Phys. Soc. **7**, 84 (1962).
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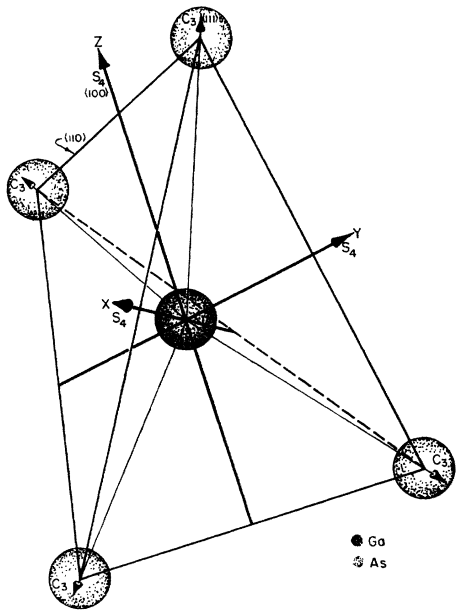

FIG. 1. Symmetry axes of the tetrahedron.

Boltzmann distribution of the Zeeman-level populations is continually maintained by spin-spin interactions, the equilibrium magnetization M_z under the combined influence of the perturbing field and the thermal spin-lattice interactions, for $I=3/2$, is given by[8]

$$M_z=\frac{M_0}{1+(8/5)WT_1},$$

where T_1 is the spin-lattice relaxation time, and W is the average induced $E2$ transition probability per unit time under the influence of a sinusoidal perturbation. Time-dependent first-order perturbation theory gives for W:

$$W=\pi|H_{mn}{}^{0'}|^2g(\omega)/2\hbar^2,$$

where $H_{mn}{}^{0'}$ is the amplitude of the perturbation matrix element between states n and m, and $g(\omega)$ is the normalized density of available transitions between the initial and final states; i.e., the relative number of possible transitions in a frequency interval $d\omega$ is $g(\omega)d\omega$ and $\int_{-\infty}^{+\infty}g(\omega)d\omega=1$. We assume $g(\omega)$ to be the Gaussian distribution:

$$g(\omega)=\frac{1}{\pi^{1/2}\delta\omega}\exp\left\{-\left[\frac{\omega-\omega_{mn}}{\delta\omega}\right]^2\right\},$$

where $\delta\omega$ is the half-width for a decrease by a factor $1/e$. Evaluating W at the center of the quadrupole resonance, we get

$$W_{\max}=\pi^{1/2}|H_{mn}{}^{0'}|^2/2\hbar^2\delta\omega,$$

which gives for the equilibrium magnetization:

$$M_z=M_0\left[1+\frac{4\pi^{1/2}|H_{mn}{}^{0'}|^2T_1}{5\hbar^2\delta\omega}\right]^{-1}.$$

Quadrupole Matrix Elements

The interaction energy U of a charge distribution ρ with an external electric potential $V(x^i)$ is

$$U=V(0)\int\rho dv+\left(\frac{\partial V}{\partial x^i}\right)_0\int\rho x^i dv+\frac{1}{2}\left(\frac{\partial^2V}{\partial x^i\partial x^j}\right)_0\int\rho x^ix^jdv+\cdots$$

(summation over $i=1, 2, 3$ is implied for repeated indices). We are interested in the last term above, which is the quadrupole energy. By writing the amplitudes of the sinusoidally time-varying field gradient components as $V_{ij}{}^0$ and properly grouping the terms in the summation $V_{ij}{}^0x^ix^j$, we have for the amplitude of the quadrupole perturbation Hamiltonian:

$$H^{0'}=-\frac{1}{2}\int_{\text{nucleus}}\rho[\tfrac{1}{4}(V_{xx}{}^0-V_{yy}{}^0-2iV_{xy}{}^0)Y_{22}+\tfrac{1}{4}(V_{xx}{}^0-V_{yy}{}^0+2iV_{xy}{}^0)Y_{2,-2}-\tfrac{1}{2}(V_{xz}{}^0-iV_{yz}{}^0)Y_{21}+\tfrac{1}{2}(V_{xz}{}^0+iV_{yz}{}^0)Y_{2,-1}+\tfrac{1}{6}(3/2)^{\frac{1}{2}}(2V_{zz}{}^0-V_{xx}{}^0-V_{yy}{}^0)Y_{20}]dv,$$

where the Y_{ij} are defined, for example, by Pake and Feenberg.[9] For $I=3/2$, this yields the following matrix elements for $\Delta m=\pm2$ transitions:

$$|H_{m,m\pm2}{}^{0'}|^2=e^2Q^2[(V_{xx}{}^0-V_{yy}{}^0)^2+4(V_{xy}{}^0)^2]/48$$

where Q is the electric quadrupole moment of the nucleus. This gives for the magnetization:

$$M_z=M_0\left\{1+\frac{\pi^{1/2}e^2Q^2T_1}{60\hbar^2\delta\omega}[(V_{xx}{}^0-V_{yy}{}^0)^2+4(V_{xy}{}^0)^2]\right\}^{-1}.$$

Field Gradient Components

We expand the quantities V_{ij} as a power series in the applied electric field:

$$V_{ij}=(V_{ij})_{E=0}+C_{ij,k}E_k+\cdots,$$

where $C_{ij,k}=(\partial V_{ij}/\partial E_k)_{E=0}$. The quantities V_{ij}, since they are derivatives of a scalar with respect to homogeneous coordinates, are the components of a second-rank tensor. We presently see that, for crystals having

[8] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961), p. 141.

[9] E. Feenberg and G. E. Pake, *Notes on the Quantum Theory of Angular Momentum* (Stanford University Press, Stanford, 1959), Chap. 4.

the zincblende structure, symmetry requires the $(V_{ij})_{E=0}$ to all be zero. Thus, since the tensor V_{ij} is the contraction of the set of quantities $C_{ij,k}$ with the vector E_k, $C_{ij,k}$ must itself be a third-rank tensor and transform accordingly. In order to find out the form of the tensor $C_{ij,k}$ we must consider the symmetry properties of the particular crystal lattice in which we are interested.

The zincblence lattice consists of two interlocked face-centered cubic lattices, one displaced relative to the other along a body diagonal a distance one-quarter of its length. Each fcc lattice consists wholly of one kind of nucleus, and each nucleus finds itself at the center of a regular tetrahedron with nuclei of the other type at the vertices. Figure 1 shows a typical nucleus and its four nearest neighbors, with the symmetry axes of the tetrahedron. The three S_4 rotation-reflection axes of order four are equivalent and mutually orthogonal, and we choose them as a basis. The S_4 axes of the tetrahedron are parallel to the $\langle 100\rangle$ cubic axes. The complete set of transformations under which the tetrahedron remains invariant is the symmetry group T_d. Under any transformation from this group, therefore, any tensor which describes a crystal with tetrahedral symmetry must also be invariant. Using this fact and the equivalence of the coordinates, one can immediately show that all off-diagonal components of the $(V_{ij})_{E=0}$ must be zero (and Laplace's equation requires the diagonal terms to be zero also) and that the only nonzero components of $C_{ij,k}$ are those for which i, j, and k are all different. To first order in the electric field we have

$$V_{ij}=C_{ij,k}E_k,$$

where the tensor $C_{ij,k}$ is, for our choice of coordinates:

$$C_{ij,k}=\begin{array}{c} \\ xx \\ yy \\ zz \\ xy \\ xz \\ yz \end{array}\begin{array}{c} \begin{array}{ccc} E_x & E_y & E_z \end{array} \\ \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{bmatrix} \end{array}\beta.$$

If the static magnetic field H_0 is parallel to the [001] axis, the only nonzero component of the field gradient which appears in the expression for the equilibrium magnetization is $V_{xy}=\beta E_z$, and we get

$$M_z=M_0\left[1+\frac{\pi^{1/2}e^2Q^2T_1\beta^2E_z^{02}}{15\hbar^2\delta\omega}\right]^{-1},$$

where E_z^0 is the amplitude of the z component of the applied electric field. A measurement of M_z/M_0 for known Q, T_1, $\delta\omega$, and E_z^0 determines β^2 and hence the tensor $C_{ij,k}$.

Having found the explicit form of $C_{ij,k}$ in the S_4 coordinate system, we may now determine the components of this tensor after an arbitrary rotation of the coordinates, using the general tensor transformation:

$$C_{ij,k'}=C_{lm,n}\frac{\partial x^{i'}}{\partial x^l}\frac{\partial x^{j'}}{\partial x^m}\frac{\partial x^{k'}}{\partial x^n}.$$

It is very easily shown that

$$C_{ij,k'}=\beta\epsilon_{lmn}\frac{\partial x^{i'}}{\partial x^l}\frac{\partial x^{j'}}{\partial x^m}\frac{\partial x^{k'}}{\partial x^n},$$

where ϵ_{lmn} is equal to one for l, m, n all different, and is zero otherwise. The primes refer to the rotated coordinates, and the unprimed coordinates are the S_4 axes.

Suppose now that we rotate the crystal lattice relative to the initial coordinate system, which remains fixed relative to the laboratory and H_0. The applied electric field direction, however, is fixed relative to the crystalline axes and rotates with the lattice. For the specific rotations we shall consider, we assume the electric field parallel to the $\langle 100\rangle$ axis which is initially along z. First we rotate the crystal through an angle θ about one of the $\langle 100\rangle$ axes normal to H_0, say x. Under this transformation, $C_{ij,k}$ becomes

$$C_{ij,k'}=\begin{bmatrix} 0 & 0 & 0 \\ -\sin 2\theta & 0 & 0 \\ \sin 2\theta & 0 & 0 \\ 0 & -\sin 2\theta & \cos 2\theta \\ 0 & \cos 2\theta & \sin 2\theta \\ \cos 2\theta & 0 & 0 \end{bmatrix}\beta,$$

and the magnetization is

$$M_z=M_0\left[1+\frac{\pi^{1/2}e^2Q^2T_1\beta^2E^{02}\cos^2\theta}{15\hbar^2\delta\omega}\right]^{-1}.$$

Next, we consider the effect of a rotation about a $\langle 110\rangle$ axis. We first align a $\langle 110\rangle$ axis with x, by performing a 45° rotation about z, and then rotate the crystal through an arbitrary angle θ about x. The results are

$$C_{ij,k}''=\begin{bmatrix} 0 & \sin\theta & -\cos\theta \\ 0 & -3\sin\theta\cos^2\theta & \cos\theta(1-3\sin^2\theta) \\ 0 & \sin\theta(3\cos^2\theta-1) & 3\sin^2\theta\cos\theta \\ \sin\theta & 0 & 0 \\ -\cos\theta & 0 & 0 \\ 0 & \cos\theta(1-3\sin^2\theta) & \sin\theta(3\cos^2\theta-1) \end{bmatrix}\beta,$$

and the magnetization is

$$M_z=M_0\left[1+\frac{\pi^{1/2}e^2Q^2T_1\beta^2E^{02}}{60\hbar^2\delta\omega}(1+\cos^2\theta)^2\right]^{-1}.$$

Note that we might also have determined the orientational dependence of M_z by directly transforming the second-rank tensor V_{ij}.

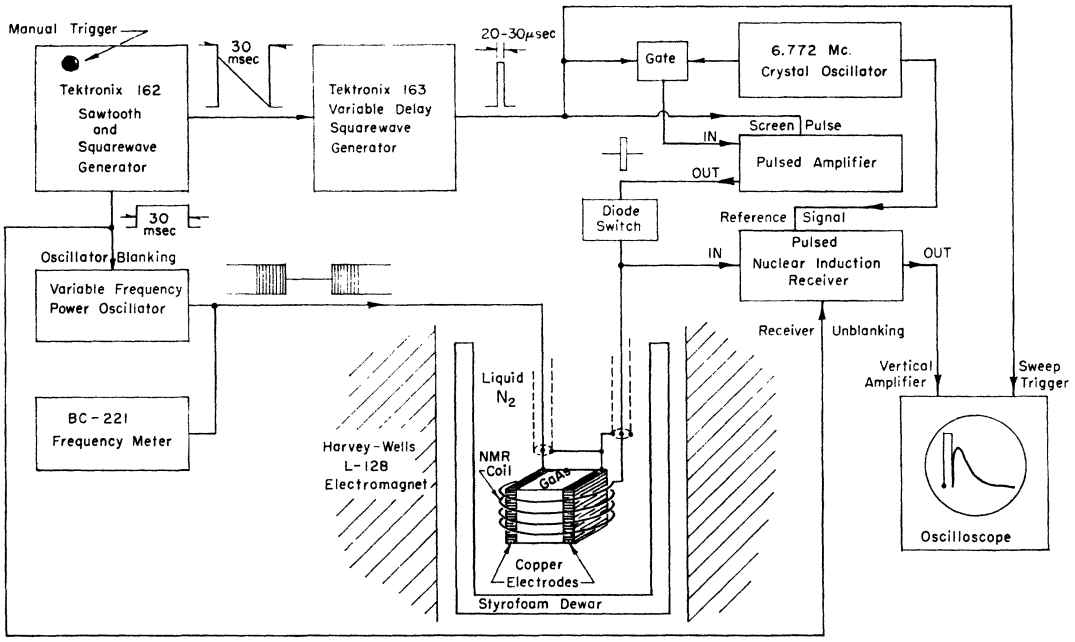

FIG. 2. Block diagram of experimental apparatus.

III. EXPERIMENTAL APPARATUS

Figure 2 is a block diagram of the experimental setup used to electrically induce $E2$ nuclear spin transitions in a GaAs single crystal, and to detect the presence of the transitions by sampling the nuclear magnetization of the crystal in a static magnetic field. The apparatus is essentially the same as that used by Brun *et al.*,[7] except that the sample geometry and sample holder were both improved to allow measurements of the orientation dependence, and to allow the applied electric field in the sample to be determined.

A $\frac{1}{2}$-in. cube with two {100} faces and four {110} faces was cut with a diamond saw from a large, high-resistivity, single crystal of GaAs, kindly supplied by the Monsanto Chemical Company. The crystal was oriented for cutting with the aid of a goniometer, utilizing optical reflections from cleavage planes, known to be {110} faces, which were exposed in small chips along the edge of the ingot. The crystal was then mounted on a polystyrene holder with copper electrodes covering the {100} faces. Special care was taken to ensure that no air gap existed between the copper and the crystal. Copper wire wound around the polystyrene holder served as both the transmitter and receiver coil for the pulsed nuclear induction apparatus. Because the sample holder was made large enough to allow the crystal to be rotated freely about a ⟨100⟩ axis without disturbing the coil, the filling factor was only about 0.2, but the signal-to-noise ratio was approximately 150 under optimum conditions.

The sample holder was immersed in liquid nitrogen contained in a styrofoam Dewar, specially made to fit in the 2-in. gap between the 12-in.-diameter pole faces of the electromagnet. A large protractor, rigidly fastened to the upper end of the rod which supported the sample holder in the magnet, served to indicate the orientation of the crystal relative to the magnetic field direction. Short lengths of RG-62 coaxial cable connected the copper electrodes to a radio-frequency power oscillator, hereafter designated ($E2$), which generated the electric field for inducing $E2$ spin transitions, and the NMR coil to a pulsed nuclear induction receiver and transmitter, hereafter designated ($M1$).

The power oscillator ($E2$), receiver, and pulsed power transmitter ($M1$) used in the early stage of the experiment were developed along more or less conventional lines and are described in detail by Pierce.[10] The transmitter ($M1$) consisted of a crystal oscillator running continually, and a gated power amplifier. The crystal oscillator and gate were adapted from a design of Blume.[11] The transmitter was coupled to the tuned NMR coil through a self-acting diode switch, consisting of a pair of back-biased FD-300 diodes. The very low junction capacitance, high forward conductance, and extremely high back resistance of these diodes results in a very efficient coupling-decoupling device.

The receiver, with a gain of about 10^4, was linear to within 1% over the range of signal amplitudes encountered in this experiment. It allowed either coherent or incoherent detection of the NMR signals. With an input Q of approximately 100, the total recovery time of the system following a transmitter pulse was about 50 μsec.

A fast-acting relay kept the input of the receiver shorted to ground except when the power oscillator ($E2$) was turned off, to avoid continuous saturation.

[10] W. L. Pierce, Ph.D. thesis, University of Colorado, 1962 (unpublished).
[11] R. J. Blume, Rev. Sci. Instr. 32, 554 (1961).

Using this system, the accuracy of the measurements was limited by the instability of the frequency of the pulsed power oscillator ($E2$), which amounted to about 500 cps. Therefore, in order to obtain more reliable data, a somewhat modified system was set up, where the power oscillator ($E2$) was replaced by a gated power amplifier fed from a General Radio signal generator.

IV. EXPERIMENTAL PROCEDURE

After the sample was cooled to 77°K and properly oriented, the nuclear resonance signal of one of the isotopes was zero-beat against the crystal oscillator reference signal by adjusting H_0. The width of the output pulse of the NMR transmitter ($M1$) was adjusted to yield maximum signal amplitude, indicating a 90° pulse for the magnetization. The power oscillator ($E2$) was turned on, the rf voltage between the copper electrodes was set to the desired value, and the frequency adjusted to twice the Larmor frequency, with the aid of a BC-221 frequency meter.

The sequence of events which resulted in obtaining a single nuclear induction decay on the oscilloscope was as follows: The Tektronix 162 waveform generator was manually triggered, and generated one linear sawtooth and one rectangular pulse, both of 30 msec length. The rectangular pulse turned off the power oscillator ($E2$) and opened the receiver blanking relay. The sawtooth went directly to the trigger input of a Tektronix 163 pulse generator. Its trigger level was set to generate a single rectangular pulse 5 msec after the leading edge of the sawtooth. This pulse simultaneously triggered the horizontal sweep of the oscilloscope, opened the rf gate between the crystal oscillator and the power amplifier in the NMR transmitter ($M1$), and turned on the power amplifier. The power amplifier then delivered a rectangular rf pulse to the sample coil, causing the 90° rotation of the nuclear magnetization of one of the isotopes in the GaAs crystal. About 50 μsec after the end of the rf pulse, the receiver recovered and the nuclear induction signal appeared on the oscilloscope, and then decayed with a time constant of 200 μsec. The maximum amplitude A, which is proportional to M_z, was measured and recorded.

V. EXPERIMENTAL RESULTS

The objectives of these experiments were to investigate the angular dependence of the induced quadrupolar saturation, as a test of the validity of the simple theoretical model developed above, and to measure the interaction parameter β. The determination of β requires a measurement of T_1 and $\delta\omega$, as well as M_z/M_0. All the measurements were made at 77°K. The relaxation data were obtained by completely saturating the nuclear spin system with a rapid series of magnetic 90° pulses, and then sampling the magnetization with a single 90° pulse at a known time after the last saturating pulse. The results were:

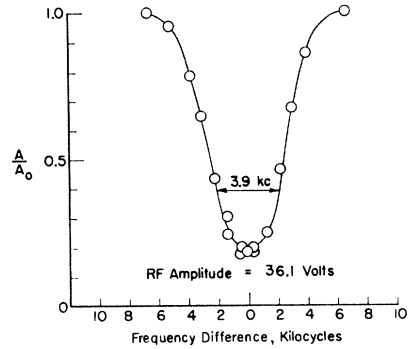

FIG. 3. Resonance curve for Ga^{69}.

$$T_1(As^{75}) = 5.1\pm0.3 \text{ sec},$$
$$T_1(Ga^{69}) = 15.5\pm0.6 \text{ sec},$$
$$T_1(Ga^{71}) = 36.4\pm1.9 \text{ sec}.$$

These values agree reasonably well with those obtained by Brun *et al.*[7] using a different GaAs crystal, and quite well with those obtained by Mieher.[12]

The quadrupolar linewidths $\delta\omega$ were obtained from the resonance curves where A/A_0 is plotted vs the frequency of the applied rf electric field. Figure 3 is a typical example of such a quadrupolar resonance curve. It should be noted that since it is the function $W(\omega)$, in the expression $M_z/M_0=[1+(8/5)WT_1]^{-1}$, which is assumed to be Gaussian, some care must be taken in determining $\delta\omega$ from the resonance curves. If A is the observed signal amplitude at a frequency ω of the rf electric field, then $A/A_0=[1+a\pi^{1/2}\delta\omega g(\omega)]^{-1}$ where a is a constant, and $A_{\min}/A_0=[1+a]^{-1}$. Therefore, the amplitude at which $g(\omega)$ is down to $g_{\max}/e$ is given by

$$\frac{A^*}{A_0}=\frac{1}{1+a/e}=\left[1+\left(\frac{A_0}{A_{\min}}-1\right)\frac{1}{e}\right]^{-1},$$

and it is at A^* that $\delta\omega$ must be measured. The quadrupolar line widths of the three isotopes are of the same order of magnitude, but depend on the orientation of the sample relative to the static magnetic field. For E and H_0 parallel to the same $\langle 100\rangle$ axis, the following

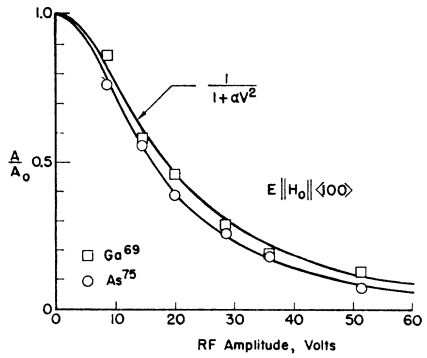

FIG. 4. Saturation vs rf amplitude.

[12] R. Mieher, Phys. Rev. 125, 1537 (1962).

widths were measured:

$$\delta\omega(\mathrm{As}^{75}) = 9.1\pm1.3 \text{ kc/sec},$$
$$\delta\omega(\mathrm{Ga}^{69}) = 12.0\pm2.0 \text{ kc/sec},$$
$$\delta\omega(\mathrm{Ga}^{71}) = 14.5\pm2.0 \text{ kc/sec}.$$

The interaction parameter β was determined from the saturation curve given in Fig. 4. The saturation, expressed by A/A_0, is plotted versus the amplitude of the applied rf voltage for both the As^{75} and Ga^{69}, with E and H_0 parallel to the same $\langle 100\rangle$ axis. A similar curve was obtained for Ga^{71}. If normalized to the same line width, the experimental points all fall on one curve, which proves the quadrupolar nature of the interaction involved in this kind of process. The rf voltage was measured with a calibrated Hewlett-Packard VTVM, and the theoretical curve $A/A_0=[1+\alpha V^2]^{-1}$ was fitted to the experimental points. The parameter α was found to be

$$\alpha(\mathrm{As}^{75}) = (3.8\pm0.2)\times10^{-3} \text{ V}^{-2}$$
$$\alpha(\mathrm{Ga}^{69}) = (2.9\pm0.3)\times10^{-3} \text{ V}^{-2}$$
$$\alpha(\mathrm{Ga}^{71}) = (2.4\pm0.7)\times10^{-3} \text{ V}^{-2}.$$

Using for the electric quadrupole moments the values $Q(\mathrm{As}^{75})=0.3\times10^{-24}$ cm^2, $Q(\mathrm{Ga}^{69})=0.23\times10^{-24}$ cm^2, and $Q(\mathrm{Ga}^{71})=0.15\times10^{-24}$ cm^2, and converting the values of the rf voltage into field strength by dividing by the thickness of the crystal between the $\langle 100\rangle$ faces, which was 1.23 cm, the interaction parameter for the various nuclei becomes

$$\beta(\mathrm{As}^{75}) = (2.0\pm0.2)\times10^{10} \text{ cm}^{-1},$$
$$\beta(\mathrm{Ga}^{69}) = (1.5\pm0.2)\times10^{10} \text{ cm}^{-1},$$
$$\beta(\mathrm{Ga}^{71}) = (1.5\pm0.2)\times10^{10} \text{ cm}^{-1}.$$

The errors assigned to β do not account for the uncertainties in the values of the electric quadrupole moments. The dynamic interaction parameters which were determined in this experiment agree reasonably well with the static values $\beta(\mathrm{As}^{75})=(1.55\pm0.35)\times10^{10}$ cm^{-1}, $\beta(\mathrm{Ga}^{69})=(1.05\pm0.15)\times10^{10}$ cm^{-1}, and $\beta(\mathrm{Ga}^{71})=(0.95\pm0.1)\times10^{10}$ cm^{-1}, found by Gill and Bloembergen[13] using the same electric quadrupole moments.

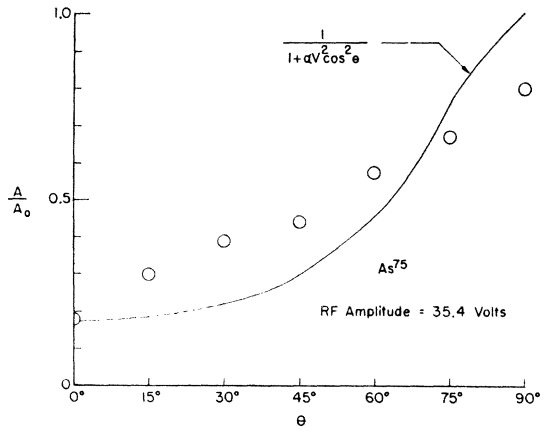

FIG. 5. Saturation vs rotation about $\langle 100\rangle$.

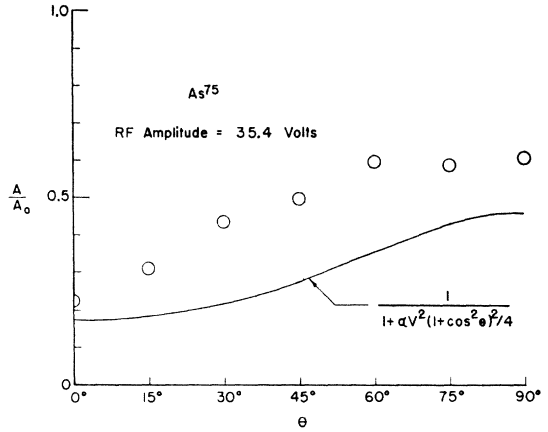

FIG. 6. Saturation vs rotation about $\langle 110\rangle$.

Finally, the angular dependence of the As^{75} saturation was measured as a function of the orientation of the crystal relative to the static magnetic field. The amplitude of the rf electric field and its direction relative to the sample were kept constant, the direction being parallel to a $\langle 100\rangle$ axis.

The experimental results are compared with the theoretical predictions for rotation about a $\langle 100\rangle$ axis in Fig. 5, and for rotation about a $\langle 110\rangle$ axis in Fig. 6. The agreement is fair. The deviations are due primarily to the variation with orientation of the quadrupolar line width.

From the good agreement of the static and dynamic measurements of the interaction parameter, as well as from the angular dependence data, we conclude that the piezoelectric part of the quadrupolar interaction (see Armstrong, Gill, and Bloembergen[5]) is of minor importance in the reported experiment, since sound waves due to piezoelectric excitation of the crystal would greatly alter the measured quantities. In order to check this, the angular dependence of ultrasonic excitation at the same frequency was measured. An x-cut transducer was placed on a $\{100\}$ face to induce $\Delta m=\pm2$ transitions. The crystal was rotated about a $\langle 100\rangle$ axis perpendicular to the H_0 field. Within experimental accuracy, no angular dependence of the saturation was observed, which indicates that if there was any ultrasonic angular dependence, it was destroyed by scattering in this crystal.

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[13] D. Gill and N. Bloembergen (private communication).