Nuclear Magnetization in the Presence of Ultrasonic Excitation*

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(Received September 30, 1957; revised manuscript received November 12, 1957)

The field gradient at the site of a nucleus resulting from a relative displacement of surrounding charge is expanded in a Taylor series. The resulting expression is used to calculate the transition probability induced by this displacement for transitions of quadrupolar nuclei between $\Delta m = \pm 2$ spin levels in a sodium chloride-type lattice. The equilibrium nuclear magnetization in the presence of ultrasonic and thermal lattice vibrations is derived on the assumption of fast mutual spin-flip processes. The derivation is also based on a strain-free lattice.

I. INTRODUCTION

SEVERAL experiments[1-3] have demonstrated that nuclear-spin transitions in solids can be induced by ultrasonic waves introduced at the frequency of either the $\Delta m = \pm 1$ transition frequency or the $\Delta m = \pm 2$ transition frequency. The sound wave is coupled to the nuclear spin through the interaction of the quadrupole moment of the nucleus with the electric field gradient generated by the ultrasonic waves. Van Kranendonk[4] and Das, Roy, and Ghosh Roy[5] have treated thermal relaxation in crystals and it is of some interest to examine the relaxation induced by ultrasonic waves. Chang[6] has calculated the thermal transition probabilities for chlorine in sodium chlorate.

In this paper we derive the effect of ultrasonic waves on the nuclear magnetization for the case where the nuclei reside in an environment where the electric field has cubic symmetry, i.e., the splitting of the spin levels is determined solely by the interaction of the magnetic moment with the applied external field. We ignore the effect of strains which cause the local electric fields to depart from cubic symmetry and consequently bring about quadrupole broadening of the spin levels.[7,8] The derivation is directly applicable to such cases as sodium nuclei in sodium chloride where the line width is quite close to the value calculated from Van Vleck's formula.[9] The results can be used with some modification for nuclei with quadrupole moments larger than that of sodium.

The derivation is divided into two parts. Part II contains the derivation for the ultrasonic transition probability and part III is devoted to derivation of the nuclear magnetization in the presence of ultrasonic and thermal lattice vibrations.

II. ULTRASONIC TRANSITION PROBABILITY

Consider a system of nuclei which occupy the lattice sites of a crystal. Let each nucleus have a spin I and nuclear quadrupole moment Q. The nuclear quadrupole moment interacts with the electric field gradient produced by neighboring charges, and we treat the quadrupolar interaction as a perturbation of the Zeeman energy. A sound wave in the crystal modulates the quadrupolar coupling, and the quadrupolar interaction couples the spin system to the crystalline lattice. We compute the probability per unit time that the sound wave will produce a transition of a nuclear spin between its Zeeman levels.

Electrostatic Potential

Let N be a nucleus and q_x an external charge. Suppose the sound wave displaces the nucleus N by $\mathbf{S}^{(0)}$ and the charge q_α by $\mathbf{S}^{(\alpha)}$. (See Fig. 1.) Here $\rho_n(\mathbf{r})$ is the nuclear charge density at a position $\mathbf{r}_n$ from the center of mass of the nucleus. We assume that after a displacement of the nucleus the nuclear charge distribution remains unchanged with respect to the nuclear center of mass.
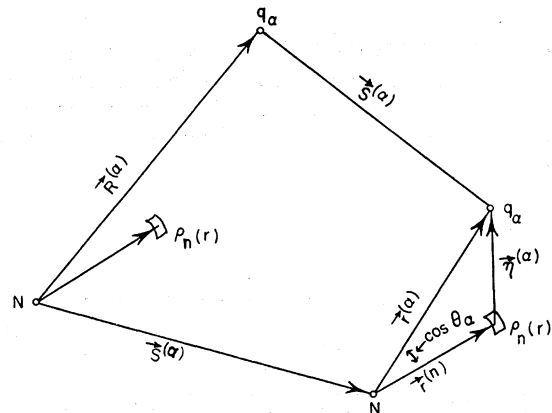

FIG. 1. The location and relative displacements of nuclear and electronic charge used in calculating the transition probabilities resulting from these displacements.

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After displacement, the electrostatic potential at $\mathbf{r}_n$ due to the charge q_α is

$$V_\alpha = q_\alpha/\eta^{(\alpha)}. \tag{1}$$

We expand $1/\eta^{(\alpha)}$ as a function of $r^{(\alpha)}$, $r^{(n)}$, and $\cos\theta_\alpha$; and the quadrupolar term of the expansion is $[(r^{(n)})^2/(r^{(\alpha)})^2]P_2(\cos\theta_\alpha)$, where $P_2(\cos\theta_\alpha)$ is the Legendre polynomial of order two. We write the Cartesian components of the vectors in Fig. 1 as follows:

$$\mathbf{r}^{(n)} = (x_i^{(n)}),$$
$$\mathbf{R}^{(\alpha)} = (X_i^{(\alpha)}),$$
$$\mathbf{S}^{(\alpha)} = (x_i^{(\alpha)}),$$
$$\mathbf{S}^{(0)} = (x_i^{(0)}), \quad i = (1, 2, 3). \tag{2}$$

Then

$$\mathbf{r}^{(\alpha)} = \mathbf{R}^{(\alpha)} + \mathbf{S}^{(\alpha)} - \mathbf{S}^{(0)},$$

or

$$\mathbf{r}_i^{(\alpha)} = (X^{(\alpha)} + x^{(\alpha)} - x^{(0)})_i. \tag{3}$$

Using Eq. (2) and Eq. (3) and writing

$$\cos\theta_\alpha = \frac{\mathbf{r}^{(\alpha)} \cdot \mathbf{r}^{(n)}}{(r^{(\alpha)})(r^{(n)})},$$

we find for the quadrupolar term of the electrostatic potential

$$V_\alpha = \frac{q_\alpha}{2(r^{(\alpha)})^5}[3X_i^{(e)}x_j^{(n)}\delta^{ij}X_l^{(e)}x_m^{(n)}\delta^{lm}$$
$$- (r^{(n)})^2(r^{(e)})^2]. \tag{4a}$$

In Eq. (4), $X_i^{(e)} \equiv (X^{(\alpha)} + x^{(\alpha)} - x^{(0)})_i$, where (e) refers to a charge outside the nucleus. Summation over indices j and m gives

$$V_\alpha = \frac{q_\alpha}{2(r^{(\alpha)})^5} \sum_{i,l=1}^{3} [3X_i^{(e)}X_l^{(e)}x_i^{(n)}x_l^{(n)}$$
$$- (r^{(n)})^2(r^{(e)})^2]. \tag{4b}$$

We separate the coordinates $x^{(n)}$ and $X^{(e)}$, and find

$$V_\alpha = \frac{q_\alpha}{6(r^{(\alpha)})^5} \sum_{i,l=1}^{3} [3x_i^{(n)}x_l^{(n)} - (r^{(n)})^2\delta^{il}]$$
$$\times [3X_i^{(e)}X_l^{(e)} - (r^{(\alpha)})^2\delta^{il}]. \tag{5}$$

Each of the square brackets may be considered as the component of a tensor. The potential V_α becomes

$$V_\alpha = \frac{q_\alpha}{6(r^{(\alpha)})^5}\mathbf{A}:\mathbf{B}, \tag{6}$$

where $\mathbf{A}$ and $\mathbf{B}$ are second-rank tensors. Consider the tensor $\mathbf{A}$:

$$A_{il} = 3x_i^{(n)}x_l^{(n)} - (r^{(n)})^2\delta^{il}.$$

The tensor is symmetric; and there are five independent components, since it has zero trace. Each component is a homogeneous quadratic function of x, y, z. We shall choose for a new basis the five linearly independent

functions $r^2Y_2{}^m$, where $Y_2{}^m$ are the unnormalized spherical harmonics of degree two. Then the five independent components, which are sufficient to define the tensor, become in Cartesian coordinates

$$A_0 = \tfrac{1}{2}(3z^2 - r^2) = r^2P_2(\cos\theta) = r^2Y_2{}^0, \tag{7a}$$

$$A_{\pm 1} = 3z(x \pm iy) = r^2P_2{}^{\pm 1}(\cos\theta)e^{\pm i\phi} = r^2Y_2{}^{\pm 1}, \tag{7b}$$

$$A_{\pm 2} = 3(x \pm iy)^2 = r^2P_2{}^{\pm 2}(\cos\theta)e^{\pm 2i\phi} = r^2Y_2{}^{\pm 2}. \tag{7c}$$

We drop the subscripts and write the Cartesian coordinates explicitly. We also drop the superscripts and keep in mind that the A's refer to nuclear coordinates. We may treat the tensor $\mathbf{B}$ exactly as we have treated $\mathbf{A}$ and reduce $\mathbf{B}$ to five linearly independent components analogous to the A's of Eqs. (7).

Using the A_i of Eqs. (7) and the analogous B_i, we expand the potential V_α in terms of the reduced tensors. The explicit expression for V_α is

$$V_\alpha = \frac{q_\alpha}{(r^{(\alpha)})^5}\Big(A_0B_0 + \tfrac{1}{6}A_{+1}B_{-1} + \tfrac{1}{6}A_{-1}B_{+1}$$
$$+ \frac{1}{24}A_{+2}B_{-2} + \frac{1}{24}A_{-2}B_{+2}\Big)$$

or

$$V_\alpha = \frac{q_\alpha}{(r^{(\alpha)})^5} \sum_{i=-2}^{2} C_iA_iB_{-i}, \tag{8}$$

where the C_i are numerical constants.

Quadrupolar Interaction Energy

One finds the contribution from the charge q_α to the nuclear quadrupolar interaction energy by multiplying V_α with the nuclear charge density $\rho_n(r)$ and integrating over the region occupied by the central nucleus:

$$H_\alpha = \int V_\alpha\rho_n(r)d\tau_n,$$

where H_α is the contribution to the Hamiltonian from the charge q_α. Substitution of V_α from Eq. (8) yields

$$H_\alpha = \frac{q_\alpha}{(r^{(\alpha)})^5} \sum_{i=-2}^{2} C_iB_{-i}\int \rho_n(r)A_id\tau_n. \tag{9}$$

Next we introduce the components of the nuclear quadrupole moment, which we define as

$$Q_i \equiv \int \rho_n(r)A_id\tau_n, \quad (i = 0, \pm 1, \pm 2). \tag{10}$$

The A_i are functions of the Cartesian coordinates of an element of the nuclear charge. We transform the A_i from functions of Cartesian coordinate operators to functions of the components of the nuclear-spin operator. (Ramsey[10] gives a detailed discussion of the

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[10] Norman F. Ramsey, *Nuclear Moments* (John Wiley and Sons, Inc., New York, 1953), p. 16.

transformation.) We choose a representation in which both the operator I^2 and the operator I_z are diagonal. Then in operator form

$$Q_0 = C\{\tfrac{1}{2}[3I_z^2 - I(I+1)]\}, \tag{11a}$$

$$Q_{\pm 1} = \tfrac{3}{2}C[I_z(I_x \pm iI_y) + (I_x \pm iI_y)I_z], \tag{11b,c}$$

$$Q_{\pm 2} = 3C[I_x \pm iI_y]^2, \tag{11d,e}$$

where $C = eQ/[I(2I-1)]$. The scalar "Q" is conventionally called "the nuclear quadrupole moment."

Using Eqs. (11), we write the Hamiltonian, H_α, as

$$H_\alpha = \frac{q_\alpha}{(r^{(\alpha)})^5} \sum_{i=-2}^{2} C_i Q_i B_{-i}^{(\alpha)}. \tag{12}$$

The $B_i^{(\alpha)}$ are functions of the relative displacement of the charge q_α with respect to the displacement of the central nucleus. To find the total contribution to the quadrupolar energy from all external charges, we must sum over all charges q_α. Hence, the total quadrupolar interaction energy H' becomes

$$H' = \sum_\alpha H_\alpha = \sum_\alpha \frac{q_\alpha}{(r^{(\alpha)})^5} \sum_{i=-2}^{2} C_i Q_i B_{-i}^{(\alpha)}. \tag{13}$$

We are interested in the physical situation which exists when a sound wave displaces the nuclei and modulates the quadrupolar interaction energy; consequently, we expand the $B_i^{(\alpha)}/(r^{(\alpha)})^5$ in terms of the relative displacement of the charge q_α with respect to the central nucleus. For convenience, we shall make several changes of notation [see Fig. 1 and Eq. (2)]. Let

$$\boldsymbol{\xi}^{(\alpha)} = \mathbf{S}^{(\alpha)} - \mathbf{S}^{(0)} = (\xi_i^{(\alpha)}),$$

$$\mathbf{R}_\alpha = \mathbf{r}^{(\alpha)} = (X_i^{(\alpha)} + \xi_i^{(\alpha)}), \quad (i = 1, 2, 3).$$

It follows from Fig. 1 that $\mathbf{R}_\alpha = \mathbf{R}^{(\alpha)}$ when $\boldsymbol{\xi} = 0$. The explicit expressions for the $B_i^{(\alpha)}$ are:

$$B_0^{(\alpha)} = \tfrac{1}{2}[3(X_3^{(\alpha)} + \xi_3^{(\alpha)}) - (R_\alpha)^2],$$

$$B_{\pm 1}^{(\alpha)} = 3(X_3^{(\alpha)} + \xi_3^{(\alpha)})[(X_1^{(\alpha)} + \xi_1^{(\alpha)}) \pm i(X_2^{(\alpha)} + \xi_2^{(\alpha)})],$$

$$B_{\pm 2}^{(\alpha)} = 3[(X_1^{(\alpha)} + \xi_1^{(\alpha)}) \pm i(X_2^{(\alpha)} + \xi_2^{(\alpha)})]^2.$$

The general form for the expansion of $B_i^{(\alpha)}/R_\alpha^5$ is

$$\frac{B_i^{(\alpha)}}{R_\alpha^5} = \frac{B_i^{(\alpha)}}{R_\alpha^5}\bigg|_{\xi=0} + \sum_{k=1}^{3} \frac{\partial}{\partial \xi_k^{(\alpha)}}\left(\frac{B_i^{(\alpha)}}{R_\alpha^5}\right)\bigg|_{\xi=0} \xi_k^{(\alpha)}$$

$$+ \tfrac{1}{2} \sum_{k,l=1}^{3} \frac{\partial^2}{\partial \xi_k^{(\alpha)} \partial \xi_l^{(\alpha)}}\left(\frac{B_i^{(\alpha)}}{R_\alpha^5}\right)\bigg|_{\xi=0} \xi_k^{(\alpha)} \xi_l^{(\alpha)} + \cdots, \tag{14}$$

where $i = 0, \pm 1, \pm 2$.

If the quantum number m labels the allowed values of the nuclear spin operator I_z, we consider only processes for which $\Delta m = \pm 2$. Consequently, the matrix elements of the operator H' which are of interest are:

$$(Im|H'|I\, m+2)$$

$$= (Im|Q_{-2}|I\, m+2) \sum_\alpha \frac{q_\alpha}{R_\alpha^5} C_2 B_{+2}^{(\alpha)}, \tag{15a}$$

$$(Im|H'|I\, m-2)$$

$$= (Im|Q_{+2}|I\, m-2) \sum_\alpha \frac{q_\alpha}{R_\alpha^5} C_2 B_{-2}^{(\alpha)}. \tag{15b}$$

Now

$$(Im|Q_{+2}|I\, m-2) = \frac{3eQ}{I(2I-1)}$$

$$\times [(I-m+2)(I+m-1)(I-m+1)(I+m)]^{\frac{1}{2}}$$

and

$$(Im|Q_{-2}|I\, m+2) = \frac{3eQ}{I(2I-1)}$$

$$\times [(I+m+2)(I-m-1)(I+m+1)(I-m)]^{\frac{1}{2}}.$$

Next we need the expansion of B_{+2}^α/R_α^5 and B_{-2}^α/R_α^5 in terms of the relative displacement of the charge q_α. Since the direct process interests us, we need examine only the linear terms of the expansion (see Van Kranendonk[4]). The coefficient of $\xi_k^{(\alpha)}$ is

$$\frac{\partial}{\partial \xi_k^{(\alpha)}}\left(\frac{B_{\pm 2}^\alpha}{R_\alpha^5}\right)\bigg|_{\xi=0} = \frac{1}{(R^{(\alpha)})^5}(X_1^{(\alpha)} \pm iX_2^{(\alpha)})$$

$$\times \left[\frac{-15}{(R^{(\alpha)})^2}(X_1^{(\alpha)} \pm iX_2^{(\alpha)})X_k^{(\alpha)} + 6(\delta_{1k} \pm i\delta_{2k})\right]. \tag{16}$$

We treat lattices of the sodium-chloride type and assume that the six nearest neighbors of a specific nucleus are point charges. If the axes of the Cartesian coordinate system coincide with the cubic axes of the crystal, then the coordinates of the six charges (the particular nucleus which we are considering is at the origin) are:

$$\begin{aligned}
\alpha &= 1, & (X_1^{(1)}, X_2^{(1)}, X_3^{(1)}) &= (a, 0, 0); \\
\alpha &= 2, & (X_1^{(2)}, X_2^{(2)}, X_3^{(2)}) &= (0, a, 0); \\
\alpha &= 3, & (X_1^{(3)}, X_2^{(3)}, X_3^{(3)}) &= (0, 0, a); \\
\alpha &= -1, & (X_1^{(-1)}, X_2^{(-1)}, X_3^{(-1)}) &= (-a, 0, 0); \\
\alpha &= -2, & (X_1^{(-2)}, X_2^{(-2)}, X_3^{(-2)}) &= (0, -a, 0); \\
\alpha &= -3, & (X_1^{(-3)}, X_2^{(-3)}, X_3^{(-3)}) &= (0, 0, -a).
\end{aligned} \tag{17}$$

In Eq. (17), a is the equilibrium lattice constant. For $\alpha = 1$, substitution of Eq. (17) into Eq. (16) gives

$$\frac{\partial}{\partial \xi_k^{(1)}}\left(\frac{B_{+2}^{(1)}}{R_1^5}\right)\bigg|_{\xi=0} = \frac{1}{a^4}\left[-\frac{15}{a}X_k^{(1)} + 6(\delta_{1k} + i\delta_{2k})\right]. \tag{18}$$

Multiply Eq. (18) by $\xi_k^{(1)}$, the k-component of the relative displacement, and sum over the index k:

$$\sum_{k=1}^{3} \frac{\partial}{\partial \xi_k^{(1)}}\left(\frac{B_{+2}^{(1)}}{R_1^5}\right)\bigg|_{\xi=0} \xi_k^{(1)} = -\frac{9}{a^4}\xi_1^{(1)} + \frac{6i}{a^4}\xi_2^{(1)}.$$

Therefore, if we retain only linear terms, the expansion for $B_{+2}^{(1)}/R_1{}^5$ is

$$\frac{B_{+2}^{(1)}}{R_1{}^5} \cong -\frac{9}{a^4}\xi_1^{(1)} + \frac{6i}{a^4}\xi_2^{(1)}.$$

Similarly, we find the linear terms for all the $B_{\pm2}^{(\alpha)}/R_\alpha{}^5$; and the results are:

$$\frac{B_{+2}^{(1)}}{R_1{}^5} \cong -\frac{9}{a^4}\xi_1^{(1)} + \frac{6i}{a^4}\xi_2^{(1)},$$

$$\frac{B_{+2}^{(-1)}}{R_{-1}{}^5} \cong \frac{9}{a^4}\xi_1^{(-1)} - \frac{6i}{a^4}\xi_2^{(-1)}, \tag{19a}$$

$$\frac{B_{+2}^{(2)}}{R_2{}^5} \cong \frac{9}{a^4}\xi_2^{(2)} + \frac{6i}{a^4}\xi_1^{(2)},$$

$$\frac{B_{+2}^{(-2)}}{R_{-2}{}^5} \cong -\frac{9}{a^4}\xi_2^{(-2)} - \frac{6i}{a^4}\xi_1^{(-2)}, \tag{19b}$$

$$\frac{B_{+2}^{(3)}}{R_3{}^5} \cong 0, \quad \frac{B_{+2}^{(-3)}}{R_{-3}{}^5} \cong 0. \tag{19c}$$

Presence of a Sound Wave

Consider a unidirectional standing sound wave propagated in the direction of the x axis. The displacement of a lattice point is

$$S = A \cos(kx - \delta) \cos\omega t,$$

where $k = 2\pi/\lambda$, with λ the wavelength of the sound wave, A is the amplitude of the sound wave, and δ is an arbitrary phase factor. For the nucleus we find

$$S_1^{(0)} = A \cos\delta \cos\omega t, \tag{20}$$

and for the charge q_1

$$S_1^{(1)} = A \cos(ka - \delta) \cos\omega t. \tag{21}$$

Since the product ka is small for a direct process, we make the approximation

$$\cos(ka - \delta) = \cos\delta + ka \sin\delta. \tag{22}$$

Combining Eqs. (20), (21), and (22), we have

$$\xi_1^{(1)} = Aka \sin\delta \cos\omega t; \tag{23}$$

also

$$\xi_2^{(1)} = \xi_3^{(1)} = 0. \tag{24}$$

Computation of the relative displacements of the five other charges shows that

$$\xi_1^{(2)} = \xi_2^{(2)} = \xi_3^{(2)} = 0, \qquad \xi_2^{(-1)} = \xi_3^{(-1)} = 0,$$

$$\xi_1^{(3)} = \xi_2^{(3)} = \xi_3^{(3)} = 0, \qquad \xi_1^{(-2)} = \xi_2^{(-2)} = \xi_3^{(-2)} = 0, \tag{25}$$

$$\xi_1^{(-1)} = -Aka \sin\delta \cos\omega t, \quad \xi_1^{(-3)} = \xi_2^{(-3)} = \xi_3^{(-3)} = 0.$$

Substitution of the relative displacements in Eqs. (19a, b, c) yields

$$\frac{B_{+2}^{(1)}}{R_1{}^5} \cong -\frac{9}{a^4}Aka \sin\delta \cos\omega t, \tag{26a}$$

$$\frac{B_{+2}^{(-1)}}{R_{-1}{}^5} \cong -\frac{9}{a^4}Aka \sin\delta \cos\omega t, \tag{26b}$$

$$\frac{B_{+2}^{(2)}}{R_2{}^5} = \frac{B_{+2}^{(-2)}}{R_{-2}{}^5} = \frac{B_{+2}^{(3)}}{R_3{}^5} = \frac{B_{+2}^{(-3)}}{R_{-3}{}^5} = 0. \tag{26c}$$

Next we expand Eq. (15a):

$$(Im|Q_{-2}|I\ m+2) \sum_\alpha \frac{q_\alpha}{R_\alpha{}^5}C_2 B_{+2}^{(\alpha)} = \frac{3eQ}{I(2I-1)}$$

$$\times [(I+m+2)(I-m-1)(I+m+1)(I-m)]^{\frac{1}{2}}C_2$$

$$\times \left\{ \frac{q_1 B_{+2}^{(1)}}{R_1{}^5} + \frac{q_2 B_{+2}^{(2)}}{R_2{}^5} + \frac{q_3 B_{+2}^{(3)}}{R_3{}^5} + \frac{q_{-1} B_{+2}^{(-1)}}{R_{-1}{}^5} \right.$$

$$\left. + \frac{q_{-2} B_{+2}^{(-2)}}{R_{-1}{}^5} + \frac{q_{-3} B_{+2}^{(-3)}}{R_{-3}{}^5} \right\}.$$

Let $q_\alpha = \gamma e$ for all α, where γ is a constant and e is the electronic charge. Substitution of Eqs. (26a, b, c) in the expanded form of Eq. (15a) determines the matrix element $(Im|H'|I\ m+2)$:

$$(Im|H'|I\ m+2) = -\frac{e^2 Q\gamma}{8I(2I-1)}$$

$$\times [(I+m+2)(I-m-1)(I+m+1)(I-m)]^{\frac{1}{2}} \frac{18}{a^3}$$

$$\times Ak \sin\delta \cos\omega t. \tag{27}$$

If we recall that $B_{-2}^{(\alpha)} = (B_{+2}^{(\alpha)})^*$, then from Eq. (15b) we find the matrix element $(Im|H'|I\ m-2)$:

$$(Im|H'|I\ m-2) = -\frac{e^2 Q\gamma}{8I(2I-1)}$$

$$\times [(I-m+2)(I+m-1)(I-m+1)(I+m)]^{\frac{1}{2}} \frac{18}{a^3}$$

$$\times Ak \sin\delta \cos\omega t. \tag{28}$$

Transition Probability per Unit Time

If the nucleus is initially in the state m, one finds from first-order perturbation theory the probability that in the time t the nucleus will make a transition to one of the states k is

$$W_{km} = \frac{|H'^0|^2}{\hbar^2}g(\omega)\frac{\pi}{2}t. \tag{29}$$

Here $H_{km}'^0$ is time-independent, and $g(\omega)$ is the nor-

malized shape function of the nuclear resonance line. If we average Eq. (29) over the frequency spread $\delta\omega$ of the resonance line, we find the average transition probability per unit time:

$$W = \left(\frac{W_{km}}{t}\right)_{Av} = \frac{|H_{km}'^0|^2}{4\hbar^2\delta\nu}. \tag{30}$$

If we substitute the value of the matrix element $H_{km}'^0$ from Eq. (27) or Eq. (28) into Eq. (30), we find the probability per unit time for a $\Delta m = -2$ transition is

$$W = \frac{81}{64\hbar^2\delta\nu} \frac{e^4 Q^2 \gamma^2}{I^2(2I-1)^2}(I+m+2)(I-m-1)$$
$$\times (I+m+1)(I-m)\frac{A^2 k^2}{a^6}\sin^2\delta, \tag{31}$$

and the probability per unit time for a $\Delta m = +2$ transition is

$$W = \frac{81}{64\hbar^2\delta\nu} \frac{e^4 Q^2 \gamma^2}{I^2(2I-1)^2}(I-m+2)(I+m-1)$$
$$\times (I-m+1)(I+m)\frac{A^2 k^2}{a^6}\sin^2\delta. \tag{32}$$

III. NUCLEAR MAGNETIZATION

In the presence of acoustic vibrations at the frequency of the spin transitions the nuclei approach an equilibrium magnetization which is less than the magnetization when only the thermal vibrations are present.[2] It is the purpose of the following development to exhibit the dependence of the magnetization of the nuclei on the acoustic energy density. It is assumed that the mutual spin-flip mechanism brings the spin system to a spin temperature in the time of the order of T_2, the transverse relaxation time. Since T_2 is of the order of several hundred microseconds, whereas thermal and ultrasonic transition times are of the order of seconds, the spin system has a well-defined spin temperature during the acoustic radiation of the crystal. However, the region in which the spin temperature is uniform is limited by the following considerations. The ultrasonic transition probability varies as $\sin^2\delta$, where δ is the phase of the standing unidirectional acoustic wave. In order that the energy possessed by the spin system distribute itself uniformly throughout the spin system it would be necessary for the spin conduction to be large compared to $1/T_1$. However, since spin conduction takes place by a random walk process it would be necessary for $T_2(\lambda/a)^2 \ll T_1$, where λ is the ultrasonic wavelength and a is the lattice parameter. $T_2(\lambda/a)^2$, however, is at least several orders of magnitude larger than T_1 for the nuclei we have concerned ourselves with. We shall therefore ignore spin conduction and limit ourselves to a development of the magnetization in a small region of the crystal where

δ is constant and then integrate over the volume of the sample to get the expression for the total magnetization of the sample. We assume that quadrupolar effects do not sensibly increase the mutual spin-flip time.[8]

Let $n(m)d\tau$ and $n(m-1)d\tau$ be the number of nuclear spins in volume $d\tau$ with magnetic quantum number (m) and $(m-1)$, respectively. If $nd\tau$ is the total number of spins in $d\tau$ and I is the nuclear spin, the difference between the populations of two adjacent energy levels is

$$[n(m)-n(m-1)]d\tau \cong \frac{nd\tau}{2I+1}\frac{\hbar\omega}{kT_s} = \frac{nd\tau}{2I+1}\Delta_s, \tag{33}$$

where $\omega = \gamma H_0$ is the Larmor frequency of precession. To obtain Eq. (33) we assume that $\hbar\omega \ll kT_s$, and T_s is the temperature of the spin system.

If w_{m-1}^m is the probability per unit time for a thermally induced transition of a spin from the state (m) to the state $(m-1)$, the time rate-of-change of the population density $n(m)$ due to transitions between the two levels is

$$\frac{d}{dt}[n(m)] = -n(m)w_{m-1}^m$$
$$+n(m-1)w_{m-1}^m(1+\Delta_l), \tag{34a}$$

and for $n(m-1)$

$$\frac{d}{dt}n(m-1) = -n(m-1)w_{m-1}^m(1+\Delta_l)$$
$$+n(m)w_{m-1}^m. \tag{34b}$$

In Eqs. (34a, b), $\Delta_l = \hbar\omega/kT_l$, where T_l is the temperature of the lattice. From Eqs. (34a, b), we have

$$\frac{d}{dt}[n(m)-n(m-1)] = 2n(m-1)w_{m-1}^m(1+\Delta_l)$$
$$-2n(m)w_{m-1}^m. \tag{35a}$$

If we retain only terms which are linear in Δ,

$$\Delta_l n(m-1) \cong \Delta_l[n/(2I+1)];$$

and we may write for Eq. (35):

$$\frac{d}{dt}[n(m)-n(m-1)] = -2w_{m-1}^m[n(m)-n(m-1)]$$
$$+2\Delta_l\left(\frac{n}{2I+1}\right)w_{m-1}^m. \tag{35b}$$

The z-component of the macroscopic magnetization which arises from the nuclear spin system in $d\tau$ is

$$m_z d\tau = \gamma\hbar d\tau \sum_{m=-I}^{I} m n(m). \tag{36}$$

For half-integral spin, Eq. (36) becomes

$$m_z = \gamma\hbar\frac{2n}{2I+1}\left(\frac{\hbar\omega}{kT_s}\right)[I^2+(I-1)^2+\cdots+(\tfrac{1}{2})^2], \tag{37}$$

where we have used Eq. (33). Collecting terms in Eq. (37), we find

$$m_z = \gamma \hbar n \left(\frac{\hbar\omega}{kT_s}\right)\left(\frac{I}{3}\right)(I+1). \qquad (38)$$

For $\Delta m = \pm 1$ transitions we may write

$$\frac{dm_z}{dt} = \gamma\hbar \frac{d}{dt}[n(m) - n(m-1)].$$

Then it follows from Eq. (33) and Eq. (35a) that

$$\frac{d}{dt}m_z = 2\gamma\hbar \frac{n}{2I+1} w_{m-1}{}^m (\Delta_l - \Delta_s). \qquad (39)$$

Using Eq. (38), we may write Eq. (39) as

$$\frac{d}{dt}m_z = \frac{6}{I(I+1)(2I+1)} w_{m-1}{}^m (m_{z0} - m_z) \qquad (40)$$

where $m_{z0} = \frac{1}{3}\gamma\hbar I(I+1)n\Delta_l$ is the value of m_z when the spin system is in thermal equilibrium with the lattice. To find the total change in m_z due to thermal processes with $\Delta m = \pm 1$, we must sum Eq. (40) over (m):

$$\frac{d}{dt}m_z\bigg|_{\Delta m = \pm 1} = \frac{6(m_{z0} - m_z)}{I(I+1)(2I+1)} \sum_{m=1}^{-I+1} w_{m-1}{}^m. \qquad (41)$$

If we assume that the quadrupolar interaction couples the nuclear spin system to the crystalline lattice,

$$w_{m-1}{}^m \propto |(I\ m-1|Q_{-1}|Im)|^2. \qquad (42)$$

Since

$$(I\ m-1|Q_{-1}|Im) = \frac{3eQ}{2I(2I-1)}(2m-1)$$
$$\times [(I+m)(I-m+1)]^{\frac{1}{2}}, \qquad (42)$$

we obtain

$$w_{m-1}{}^m \propto \frac{9e^2Q^2}{4I^2(2I-1)^2}(2m-1)^2(I+m)(I-m+1).$$

The transition probability $w_{m-1}{}^m$ also depends upon the lattice coordinates, and we may write

$$w_{m-1}{}^m = \frac{9e^2Q^2}{4I^2(2I-1)^2}(2m-1)^2(I+m)$$
$$\times (I-m+1)F(l). \qquad (43)$$

In Eq. (43), $F(l)$ is a function of the lattice coordinates only. With the value of $w_{m-1}{}^m$ from Eq. (43), we find for Eq. (41)

$$\frac{d}{dt}m_z\bigg|_{\Delta m = \pm 1} = \frac{6(m_{z0} - m_z)}{I(I+1)(2I+1)} \frac{9e^2Q^2}{4I^2(2I-1)^2}F(l)$$
$$\times \sum_{m=I}^{-I}(2m-1)^2(I+m)(I-m+1). \qquad (44)$$

The quadrupolar coupling between the nuclear spin system and the crystalline lattice also produces $\Delta m = \pm 2$ transitions; hence, we compute the change of m_z produced by these transitions. The population difference between the states (m) and $(m-2)$ is

$$[n(m) - n(m-2)] = \frac{2n}{2I+1}\left(\frac{\hbar\omega}{kT_s}\right) = \frac{2n}{2I+1}\Delta_s. \qquad (45)$$

The time rate-of-change of the population difference is

$$\frac{d}{dt}[n(m) - n(m-2)] = -2w_{m-2}{}^m[n(m) - n(m-2)]$$
$$+ \frac{4n}{2I+1}w_{m-2}{}^m \Delta_l. \qquad (46)$$

In Eq. (46) we have used the approximation

$$\Delta_l n(m-2) \cong \frac{n}{2I+1}\Delta_l.$$

Since $dm_z/dt = 2\gamma\hbar(d/dt)[n(m) - n(m-2)]$ for $\Delta m = \pm 2$ transitions, we find, by the same method as we used to find Eq. (41),

$$\frac{dm_z}{dt} = \frac{24(m_{z0} - m_z)}{I(I+1)(2I+1)} \sum_{m=I}^{-I+2} w_{m-2}{}^m. \qquad (47)$$

The transition probabilities $w_{m-2}{}^m$ are

$$w_{m-2}{}^m = \frac{9e^2Q^2}{I^2(2I-1)^2}(I+m)(I-m+1)(I+m-1)$$
$$\times (I-m+2)G(l), \qquad (48)$$

where $G(l)$ is a function of the lattice coordinates only. Substitution of the $w_{m-2}{}^m$ from Eq. (48) into Eq. (47) yields

$$\frac{dm_z}{dt} = \frac{24(m_{z0} - m_z)}{I(I+1)(2I+1)} \frac{9e^2Q^2}{I^2(2I-1)^2}G(l)$$
$$\times \sum_{m=I}^{-I+2}(I+m)(I-m+1)(I+m-1)(I-m+2). \qquad (49)$$

Finally, we consider the time rate-of-change of m_z which is caused by the sound wave in the crystal. We shall choose the frequency of the sound wave in such a way that it produces $\Delta m = \pm 2$ transitions. The change with time of the population density difference, $[n(m) - n(m-2)]$, is

$$\frac{d}{dt}[n(m) - n(m-2)] = 2n(m-2)W_{m-2}{}^m$$
$$- 2n(m)W_{m-2}{}^m. \qquad (50)$$

In Eq. (50), $W_{m-2}{}^m$ is the probability per unit time

that the ultrasonic energy produces a $\Delta m = \pm 2$ transition. It follows from Eq. (41) that

$$\frac{dm_z}{dt}\bigg|_{\text{ultrasonic}} = \frac{-24}{I(I+1)(2I+1)} m_z \sum_{m=I}^{-I+2} W_{m-2}{}^m. \quad (51)$$

The ultrasonic transition probabilities are given by Eq. (31) of part II:

$$W_{m-2}{}^m = \alpha(I-m+2)(I+m-1)(I-m+1)(I+m), \quad (52)$$

where

$$\alpha = \frac{81}{64} \frac{e^4 Q^2 \gamma^2 A^2 k^2}{I^2(2I-1)^2 \hbar^2 a^6 \delta \nu} \sin^2\delta.$$

We express the total time rate-of-change of m_z as

$$\frac{dm_z}{dt} = \frac{dm_z}{dt}\bigg|_{\Delta m = \pm 1} + \frac{dm_z}{dt}\bigg|_{\Delta m = \pm 2} + \frac{dm_z}{dt}\bigg|_{\text{ultrasonic}}. \quad (53)$$

The first two terms on the right side of Eq. (53) are given by Eq. (41) and Eq. (47), respectively. Instead of evaluating the sums over the transition probabilities, we shall introduce two relaxation times T' and T'' which we define as follows:

$$\frac{1}{T'} = \frac{6}{I(I+1)(2I+1)} \sum_{m=I}^{-I} w_{m-1}{}^m,$$

$$\frac{1}{T''} = \frac{24}{I(I+1)(2I+1)} \sum_{m=I}^{-I+2} w_{m-2}{}^m.$$

Introduction of the relaxation times enable us to write Eq. (41) and Eq. (47) as

$$\frac{dm_z}{dt}\bigg|_{\Delta m = \pm 1} = \frac{m_{z0} - m_z}{T'}, \quad (54a)$$

and

$$\frac{dm_z}{dt}\bigg|_{\Delta m = \pm 2} = \frac{m_{z0} - m_z}{T''}, \quad (54b)$$

respectively. We also define an ultrasonic relaxation time by

$$\frac{1}{T_u} \equiv \frac{24}{I(I+1)(2I+1)} \sum_{m=I}^{-I+2} W_{m-2}{}^m. \quad (54c)$$

Using Eqs. (54a, b, c), we may write Eq. (53) as

$$\frac{dm_z}{dt} = \frac{m_{z0} - m_z}{T_1} - \frac{m_z}{T_u}, \quad (55)$$

where

$$1/T_1 = 1/T' + 1/T'',$$

and T_1 is the observed spin-lattice relaxation time. One can compute the ultrasonic relaxation time from Eqs. (54c) and (52).

At constant ultrasonic excitation for a period of time much longer than T_1, the magnetization density is constant in a region where the phase δ is constant. That is, $dm_z/dt = 0$. From this we get:

$$m_z = \frac{m_{z0}}{1 + (T_1/T_u)} = \frac{m_{z0}}{1 + \beta \sin^2\delta}, \quad (56)$$

where

$$\beta = \frac{T_1}{\sin^2\delta} \frac{24}{I(I+1)(2I+1)} \sum_{m=I}^{-I+2} W_{m-2}{}^m,$$

and the W are given by Eq. (52).

In order to find the total magnetization, we must integrate Eq. (56) over the volume of the crystal. In the case where we have a standing wave in the x direction in a uniform cylinder, the phase δ is given by $2\pi x/\lambda$, where λ is the ultrasonic wavelength. The total magnetization M_z is given by

$$M_z = \int m_z d\tau = m_{z0} \int \frac{A\,dx}{1 + \beta \sin^2(2\pi x/\lambda)}$$

$$= M_{z0}/(1+\beta)^{\frac{1}{2}}, \quad (57)$$

where A is the cross-sectional area of the uniform cylinder and M_{z0} is the total magnetization of the nuclei in the absence of ultrasonic radiation.

IV. DISCUSSION

The above calculations have been used in several experiments, one of which is reported in this issue.[11] We have also examined the dependence of the ultrasonic transition probability on direction for sodium nuclei in sodium chloride.[12] We used nearest and next-nearest neighbor interaction in conjunction with Eq. (16) and found that γ has no directional anisotropy to within 25%.

V. ACKNOWLEDGMENTS

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[11] See Jennings, Tanttila, and Kraus, Phys. Rev. **109**, 1059 (1958), following paper.

[12] These measurements are the basis of a Master's thesis to be submitted by D. A. Jennings to the Graduate School of Michigan State University.