ACOUSTIC FREE INDUCTION IN A QUADRUPOLAR SPIN SYSTEM IN A CUBIC CRYSTAL

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ABSTRACT. The bulk magnetisation vectors excited by an acoustic pulse at magnetic reasonance has been calculated in a cubic crystal, where each lattice site is assumed to be occupied by a nucleus of spin 1 (one). The Hamiltonian has been cast in a general form, for any interaction of the form $(\vec{I}.\vec{D}.\vec{I})$, and calculation has been done by setting up the Rabi-Bloch matrices, and neglecting, for simplicity, the spin-spin interactions. It is found that with a longitudinal excitation in one of the cubic axes, the transverse components of the average macroscopic moment vectors $<M_x>_{av}$ and $<M_y>_{av}$, are practically zero, being contributed by nuclei located at thin layers (of width equal to half wavelength of the acoustic waves) at the two ends of the sample. The results are identical with those of Kessel (Kessel, 1962) who analysed the above situation by a first order time-dependent perturbation method.

INTRODUCTION

The absorption of acoustic energy at magnetic reasonance in solids, when the reasonance is excited by an impressed acoustic field [Kastler (1952), Altshulen (1955), Jacobsen and Stevens (1963)], has been demonstrated both in pulsed [Jacobsen, Shiren and Tucker (1959), Tucker (1961), Shiren (1962), and Guermeur, Joffrin, Levelut and Penne (1964)] and CW [Proctor and Tantilla (1956), Proctor and Robinson (1956), Menes an..Bolef (1958), Bolef and Menes (1959), and Bolef, de Klerk and Gosser (1962)] experiments. In such experiments one particular lattice mode is strongly excited, and the energy in tha lattice mode is transmitted to the spin system via the spin-phonon interaction. There must be some relaxation mechanism (other than the so-called direct process) that will maintain the Boltzman population excess in the spin system. And since the excitation is strong, effectively equalising the upward and the downward transition probabilities of a spin, there will be a net absorption of energy from the excited lattice mode. The amount of the absorption is a measure of the direct spin-phonon interaction that couples the particular lattice mode with the magnetic spin system. This fact, together with the absence of "penetration depth" effect in metals leads to several interesting possibilities that can be explored by acoustic magnetic resonance experiments. Already, quite a few experiments have been done yielding important results [Bolef and Menes (1961), Shiren (1962), and Guermeur, Joffrin, Levelut and Penne (1965)]

However, there has not been any induction experiment in this field, so far. The possibility of such an expeirment, using pulse technique, was analysed by Kessel (1962) a few years back. Although the method used in the analysis is sufficient for tackling the situation with simplifying assumptions, i.e. the neglect of spin-spin interaction, it is felt that a density matrix treatment of the phenomenon will be more convenient in more general causes*. We describe such a treatment which is similar to Lowe and Norberg's (1957) analysis of free induction in electromagnetically excited NMR. In our analysis we shall consider a cubic crystal, having lattice sites occupied by nuclei of spin 1 (one). The spin system will thus be coupled to the excited lattice mode by nuclear quadrupole interaction.

ACOUSTICALLY INDUCED NUCLEAR MAGNETIC INDUCTION IN A CUBIC CRYSTAL

(A) Spin-phonon interaction Hamiltonian in a quadrupolar spin system in an acoustically excited cubic crystal : Let us write the interaction term in the form,

$$H = -\hbar(\vec{I}.\vec{\vec{D}}.\vec{I}) \qquad \ldots (1)$$

where $\vec{I}$ is the nuclear spin vector, and $\vec{\vec{D}}$ is a tensor of rank two in three dimension, containing the lattice coordinates. In a rectangular coordinate system (X, Y, Z), where Z is the axis of quantisation of the spin system (that is, the direction of the D.C. magnetic field), it will be seen that the tensor, D, is symmetric for the particular spin-lattice system. Expanding (1), we can thus write :

$$\begin{aligned} H' = -\hbar[&\tfrac{1}{2}(D_{xx}+D_{yy})\vec{I}^2+\tfrac{1}{2}(2D_{zz}+D_{xx}-D_{yy})I_z^2+ \\ &+\tfrac{1}{4}(D_{xx}-D_{yy})(I_+^2+I_-^2)+D_{xz}(I_xI_z+I_zI_x)+ \\ &+D_{yz}(I_yI_z+I_zI_y)+D_{xy}(I_xI_y+I_yI_x)] \end{aligned} \qquad \ldots (2)$$

Dropping the terms that couple states with $\Delta m = 0$, we have :

$$\begin{aligned} H' = -\hbar\,[&\tfrac{1}{4}(D_{xx}-D_{yy})(I_+^2+I_-^2)+D_{xz}(I_xI_z+I_zI_x)+D_{yz}(I_yI_z+I_zI_y)+ \\ &+D_{xy}(I_xI_y+I_yI_x) \end{aligned} \qquad \ldots (3)$$

We may note that the density-matrix method is effictively equivalent to the ordinary time-dependent perturbation method, upto certain degree of approximation. However, the former is different from the latter when the actual calculation is concerned. In the time-dependent perturbation method, one expands the time-dependent wave function in a complete set of time-independent basis functions, and solves for the expansion coefficients a_m, a_n etc. from a set of coupled differential equations. One popular way of doing this is to reduce these differential equations to a set of coupled algebraic equations by making use of Laplace Transforms. In general, it is difficult to obtain these transforms. However, in density-matrix method one avoids this difficulty by calculating the products ($a_n\, a_m$) which are more directly useful for calculating expectation values.

The components of D are functions of the local strain at the nuclear sites, and one can expand D in terms of the strain components e_{ij} in a Taylors series. Thus introducing the spin-phonon coupling constant G, we can write (in Voigt notation) :

$$D_j = D_j^{(0)} + \sum_i G_{ji} e_i + \text{higher order terms.} \quad i, j = 1, 2, 3, \ldots, 6.$$

where

$$G_{ji} = \frac{\partial}{\partial e_i} (D_j)_{e_i \to 0} \qquad \ldots (4)$$

In order to define G uniquely, the (4) is written in the coordinate system (X_0, Y_0, Z_0) that coincides with the crystallographic axes. This also enables one to simplify the expansion (4) by using symmetry arguments. For example, in cubic symmetry, we have the expansion:

$$D_{x_0x_0} = \frac{G_{11}}{2} (2e_{x_0x_0} - e_{y_0y_0} - e_{z_0z_0}), \qquad D_{x_0y_0} = G_{44} e_{x_0y_0}$$

$$D_{y_0y_0} = \frac{G_{11}}{2} (2e_{y_0y_0} - e_{z_0z_0} - e_{x_0x_0}), \qquad D_{y_0z_0} = G_{44} e_{y_0z_0}$$

$$D_{z_0z_0} = \frac{G_{11}}{2} (2e_{z_0z_0} - e_{x_0x_0} - e_{y_0y_0}), \qquad D_{z_0z_0} = G_{44} e_{x_0z_0} \qquad \ldots (5)$$

The components of D, used in the Hamiltonian (3) can then be obtained by using the following usual transformation relations :

$$D_{ii} = \sum_{(\nu\mu)} \alpha_{i\nu} \alpha_{i\mu} D_{\nu\mu}.$$

$$D_{ij} = \sum_{(\nu\mu)} (a_{i\nu} a_{j\mu} + \alpha_{j\nu} a_{i\mu}) D_{\nu\mu} \qquad \ldots (6)$$

where

$$(\nu\mu) \equiv (x_0x_0), (y_0y_0), (z_0z_0), (x_0y_0), (y_0z_0), \text{ and } (x_0z_0).$$

and the coefficients a are given in table I.

TABLE I

Transformation coefficients. '$a_{i\nu}$', for two Cartesian coordinate systems, i and ν, having the same origin and their mutual orientation being specified by the Eulerian angles θ, ϕ and ψ.

i \ ν	x_0	y_0	z_0
x	$\cos\psi \cos\phi \cos\theta + \sin\psi \sin\phi$	$\cos\psi \sin\phi \cos\theta - \sin\psi \cos\phi$	$\cos\psi \sin\theta$
y	$\sin\psi \cos\phi \cos\theta - \cos\psi \sin\phi$	$\sin\psi \sin\phi \cos\theta + \cos\psi \cos\phi$	$\sin\psi \sin\theta$
z	$-\cos\phi \sin\theta$	$-\sin\phi \sin\theta$	$\cos\theta$

We will now consider a special case. Let us assume that a longittudinal acoustic wave of circular frequency ω has been impressed in the crystal with the propagation vector in the crystallographic (001) direction. We also assume that the laboratory Y-direction and the crystal (010) directions coincide, and the crystal has been rotated about the Y-axis such that the propagation vector $\vec{k}$ (still in the crystal (001) direction) makes an angle θ with the D.C. magnetic field, ie. the Z-direction. This longitudinal wave will generate a standing wave pattern, and an axially symmetric strain field (time-dependent) in the crystal, the axis of symmetry being in the direction of $\vec{k}$. Let us also choose the laboratory X—axis such that it coincides with the projection of $\vec{K}$ on the $X-Y$ plane. In such a case, we see that the only strain component is $e_{z_0z_0}$, where the direction z_0 is the crystal (001) and thus from (5), we get :

$$D_{z_0z_0} = G_{11} e_{z_0z_0}$$

$$D_{x_0x_0} = D_{y_0y_0} = -\frac{G_{11}}{2} e_{z_0z_0} \qquad \ldots (7)$$

$$D_{x_0y_0} = D_{y_0z_0} = D_{z_0x_0} = 0$$

Transforming (7) into the (XYZ) system (using (6), and the table I, with $\phi = \psi = 0$),

$$D_{zz} = \frac{G_{11}}{2} e_{z_0z_0}(3\cos^2\theta - 1), \qquad D_{xz} = \frac{3}{2} G_{11} e_{z_0z_0} \sin\theta\cos\theta$$

$$D_{xx} = \frac{G_{11}}{2} e_{z_0z_0}(3\sin^2\theta - 1), \qquad D_{xy} = D_{yz} = 0$$

$$D_{yy} = -\frac{G_{11}}{2} e_{z_0z_0} \qquad \ldots (8)$$

The Hamiltonian (3) then becomes :

$$H' = -\hbar\left[\frac{3}{8} G_{11} e_{z_0z_0}(I_+^2 + I_-^2)\sin^2\theta + \frac{3}{4} G_{11} e_{z_0z_0}(I_xI_z + I_zI_x)\sin 2\theta\right] \qquad \ldots (9)$$

The above formulation holds for any interaction of the form (1). Nuclear quadrupolar interaction is also of this form and the Hamiltonian (9) is applicalbe to this case. The constants G_{11} etc. in (9) can be measured by suitable experiments.

However, to obtain the theoretical expression for G_{11}, one has to consider the particular interaction, in this case nuclear quadrupolar interaction, explicitly.

Writing

$$H' = -\hbar(\vec{\vec{Q}}) : (\vec{\vec{\nabla E}})$$

where $\vec{\vec{Q}}$ is the nuclear quadrupole moment tensor, and $(\vec{\vec{\nabla E}})$, the electric field gradient tensor at the nuclear site, one obtains in this special case.

$$G_{11} = Ae\frac{\partial}{\partial ez_0z_0}(q)]_{ez_0z_0 \to 0} \quad \ldots (10)$$

where

$$A = \frac{eQ}{2I(2I-1)}$$

and

$$eq = \sum_j e_j(3\cos^2\theta_j - 1)r_j^{-3}$$

Q is the scalar quadrupole moment of the nucleus, and 'eq' is the axial electric field gradient with the symmetry axis along the z_0-axis. In the definition of 'eq', j refers to the jth. charge-point external to the nucleus, and $\vec{r_j}$ is the vector joining this charge-point with the origin (the center of the nucleus), and θ_j is the angle between $\vec{r_j}$ and the symmetry axis.

(b) Total Hamiltonian for the spin-lattice system : The total Hamiltonian can thus be written as

$$H = H_0 + H_1 + H_2 + H'(t) \quad \ldots (11)$$

H_0 is the Zeeman Hamiltonian for the spin system, H_1 describes the magnetic dipole interaction between the spins, and H_2 stands for all other terms in the total Hamiltonian for the statistic spinlattice system. $H'(t)$ is given by the expression (9). If we neglect those parts of H_1 and H_2 that couples states with $\Delta m \neq 0$, we are left with the following Hamiltonian :

$$H = H_0 + H'(t) \quad \ldots (12)$$

where

$$H_0 = -\hbar\sum_n \omega_0 I z_n$$

and

$$H'(t) = -\hbar\sum_n [2\omega_n^{(1)}(I_{x_n}I_{z_n} + I_{z_n}I_{x_n}) + \omega_n^{(2)}(I^2{}_{+n} + I^2{}_{-n})] \cos\omega t$$

with

$$\left.\begin{aligned} \omega_0 &= \gamma H_0 \\ \omega^{(1)} &= \tfrac{3}{8}\, G_{11}\, ez_0z_0(0) \sin 2\theta \\ \omega^{(2)} &= \tfrac{3}{8}\, G_{11} ez_0z_0(0) \sin^2\theta \end{aligned}\right\} \quad \ldots (13)$$

It has been assumed that $ez_0z_0 = ex_0z_0(0)\cos wt$, and γ is the gyromagnetic ratio for the nucleus. We shall be concerned with the cases where $H'(t)$ can be treated as a small perturbation.

(C) Rabi-Bloch matrices for the system : The Schrodinger equation is :

$$i\hbar \frac{\partial}{\partial t} \psi(t) = H(t)\psi(t) \qquad \ldots \quad (14)$$

If we write $\psi(t) = \exp\left(-\frac{it}{\hbar} H(0)\right) \psi'(t)$, the equation (14) transforms into

$$\exp\left(\frac{it}{\hbar} H(0)\right) H'(t) \exp\left(-\frac{it}{\hbar} H(0)\right) \psi'(t) = i\hbar \frac{\partial}{\partial t} \psi'(t) \qquad \ldots \quad (15)$$

We now assume that the acoustic wave is impressed only for a duration t_ω, in the form of a square pulse of carrier frequency ω, and that t_ω is sufficiently small so that those terms in H_1 and H_2 which do not commute with $H'(t)$ or $H(0)$ have expectation values much less than 1 when multiplied by $t_\omega/\hbar$. Their effect upon $H'(t)$ may then be ignored. Thus retaining only the terms H_0 in $H(0)$ the equation (15) is approximated as :

$$\exp\left(-i\omega_0 t \sum_n I_{z_n}\right) H'(t) \exp\left(i\omega_0 t \sum_n I_{z_n}\right) \psi'(t) = i\hbar \frac{\partial}{\partial t} \psi'(t). \qquad \ldots \quad (16)$$

Using the transformation properties of the spin operators under the rotation about the Z-axis, the equation (16) reduces to

$$iL(t)\, \psi'(t) = \frac{\partial}{\partial t} \psi'(t) \qquad \ldots \quad (17)$$

where

$$L(t) = \Sigma \left[2\omega_n^{(1)}\{(I_{x_n}I_{z_n} + I_{z_n}I_{x_n}) \cos_0 \omega_0 t + (I_{y_n}I_{z_n} + I_{z_n}I_{y_n}) \sin \omega_0 t\}\right.$$

$$\left. + 2\omega_n^{(2)}\{(I_{x_n}^2 - I_{y_n}^2) \cos 2\omega_0 t + (I_{x_n}I_{y_n} + I_{y_n}I_{x_n}) \sin 2\omega_0 t\}\right] \cos \omega t \qquad \ldots \quad (18)$$

The solution of (17) is

$$\psi'(t_\omega) = \exp\left\{ i \int_0^{t_\omega} L(t)dt \right\} \psi'(0) \qquad \ldots \quad (19)$$

which reduces to (neglecting small terms containing t_ω),

$$\psi(t_\omega) = R_{\omega=\omega_0}\psi(0), \quad \text{when} \quad \omega = \omega_0$$

and

$$\psi(t_\omega) = R_{\omega=2\omega_0}\psi(0), \quad \text{when} \quad \omega = 2\omega_0 \qquad (20)$$

where

$$R_{\omega=\omega_0} = \exp\left(i\theta_1 \sum_n I_{z_n}\right) \exp\left\{i \sum_n \theta_{2n}(I_{y_n}I_{x_n}+I_{z_n}I_{x_n})\right\}$$

$$R_{\omega=2\omega_0} = \exp\left(i\theta_1 \sum_n I_{z_n}\right) \exp\left\{i \sum_n \theta_{3n}(I_{x_n}{}^2-I_{y_n}{}^2)\right\}$$

$$\theta_1 = \omega_0 t, \quad \theta_2 = \omega^{(1)}t_\omega \text{ and } \theta_3 = \omega^{(2)}t_\omega. \qquad \ldots \quad (21)$$

In a representation in which I_z is diagonal, the matrices $R_{\omega=\omega_0}$ and $R_{\omega=2\omega_0}$ have been evaluated. The results for spin $I = 1$ are given in tables II and III.

(D) Computation of $<M_x>_{av}$, $<M_y>_{av}$ and $<M_z>_{av}$: We know that if we assume that the solution of the time-dependent Schrodinger equation :

$$i\hbar \frac{\partial}{\partial t} \psi_n(t) = H_n(t)\psi_n(t)$$

is of the form

$$\psi_n(t) = R_n(t)\, \psi_n(0)$$

then

$$<I_{x_n}> = \int \psi_n{}^+(0)R_n{}^+(t)I_{x_n}R_n(t)\, \psi_n(0)d\tau \qquad (22)$$

Expanding

$$\psi_n(0) = \sum_p a_{p_n}(0)u_{p_n}$$

we have

$$<I_{x_n}> = \text{Trace } \{\rho_n(0)R_n{}^+(t)I_{x_n}R_n(t)\} \qquad (23)$$

where

$$<u_{q_n}|\rho_n(0)|u_{p_n}> = a_{p_n}{}^*(0)\ a_{q_n}(0) \qquad \ldots \quad (24)$$

For a system of N spins, if we neglect the mutual interactions between the spins, we obtain

$$<\vec{M}>_{av} = \sum_n^{1,2,..N} \text{Trace} \left\{ \frac{\exp(\zeta I_{z_n})R_n{}^+\gamma\hbar(i\vec{I}_{x_n}+j\vec{I}_{y_n})+k\vec{I}_{z_n})R_n}{\text{Trace } \{\exp(\zeta I_{z_n})\}} \right\} \qquad \ldots \quad (25)$$

where $\exp(\zeta I_{z_n})/\text{Trace } \{\exp(\zeta I_{z_n})\}$ has been taken as equal to $\rho_n(0)$. $\zeta = \gamma\hbar H_0/kT$, T being the temperature of the sample and k, the Boltzman factor. The spin

system is here assumed to be at thermal equilibrium before the application of the acoustic pulse.

We the R matrices as given in tables II and III, and using the expression (25) we get the following expressions for the induced bulk nuclear magnetisation at the end of the acoustic pulse.

TABLE II

The matrix, $R_{\omega=\omega 0}$, for spin $I = 1$, in a representation in which I_z is diagonal. The elements are $<q|K|p>$, and $S_g\theta_2 \equiv$ the sign of the angle θ_2.

q \ p	1	0	−1
1	$\exp(i\theta_1)(1+\cos\theta_2)/2$	$S_g\theta_2(i\exp(i\theta_1)\sin\theta_2)/\sqrt{2}$	$\exp(i\theta_1)(1-\cos\theta_2)/2$
0	$S_g\theta_2(i\sin\theta_2)/\sqrt{2}$	$\cos\theta_2$	$S_g\theta_2(-i\sin\theta_2)/\sqrt{2}$
−1	$\exp(i\theta_1)(1-\cos\theta_2)/2$	$S_g\theta_2(-i\exp(-i\theta_1)\sin\theta_2)/\sqrt{2}$	$\exp(-i\theta_1)(1+\cos\theta_2)/2$

TABLE III

The matrix $R_{\omega=2\omega 0}$, for spin $I = 1$, in a representation in which I_z is diagonal. The elements are $<q|R|p>$, and $S_g\theta_3 \equiv$ the sign of the angle θ_3.

q \ p	1	0	−1
1	$\exp(i\theta_1)\cos\theta_3$	0	$Sg\ \theta_3(i\exp(i\theta_1)\sin\theta_3)$
0	0	1	0
−1	$S_g\theta_3(i\exp(-i\theta_1)\sin\theta_3)$	0	$\exp(-i\theta_1)\cos\theta_3$

Case I. $\omega = \omega_0$

$$<M_x>_{av} = \gamma\hbar \frac{\cosh \zeta - 1}{1+2\cosh \zeta} \sin \theta_1 \sum_n^{1,2,..,N} S_g(\theta 2_n) \sin 2\theta 2_n$$

$$<M_y>_{av} = \gamma\hbar \frac{\cosh \zeta - 1}{1+2\cosh \zeta} \cos \theta_1 \sum^{1,2,..,N} S_g(\theta 2_n) \sin 2\theta 2_n$$

$$<M_z>_{av} = \gamma\hbar \frac{\sinh \zeta}{1+2\cosh \zeta} \sum^{1,2,..,N} \cos \theta 2_n \qquad (26)$$

Case II. $\omega = 2\omega_0$

$$<M_x>_{av} = 0$$

$$<M_y>_{av} = 0$$

$$<M_z>_{av} = \gamma\hbar \frac{2\sinh \zeta}{1+2\cosh \zeta} \sum^{1,2,..,N} \cos 2\theta 3_n \qquad (27)$$

After the withdrawal of the pulse, the time-dependence of $<M_x>_{av}$ and $<M_y>_{av}$ will be given as usual by the Fourier transform of the steady NMR line shape.

Refering to (26), and from the definitions of θ_2 as given in (21) and (13), we see that

$$\sum_n S_g(\theta 2_n) \sin 2\theta 2_n = \sum_n S_g\{a.e^{(n)}{}_{z_0 z_0}(0)\} \sin\left\{\frac{a}{2} e^{(n)}{}_{z_0 z_0}(0)\right\}, \qquad \ldots \quad (28)$$

where

$$a = \{\tfrac{3}{8}\ G_{11} \sin 2\theta\} t_\omega\ .$$

$e^{(n)}{}_{z_0 z_0}(0)$, for a standing wave pattern where end surfaces of the sample are at antinodes, may be written as

$$e^{(n)}{}_{z_0 z_0}(0) = 2KA_0 \sin KZ_n.$$

and hence we see from (28) that the main contribution to the summation will come from the two layers at the ends of the sample, layers having a thickness "l" given by

$$kl = \pi/2 \text{ or } l = \lambda/4.$$

As an example, for experiment at 10 mc/s, $l \cong 10^{-2}$ cm. and an insignificantly small number of nuclei are taking effective part in contributing to $<M_x>_{av}$ and $<M_y>_{av}$.

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REFERENCES

Altshuler, S. A., 1955, *Soviet Phys. JETP.* **1**, 37.

Barnes, D. J., 1963, *Nature*, **200**, 253.

Bolef, D. I., and Menes, M., 1959, *Phys. Rev.* **114**, 1441.

Bolef, D. I., de Klerk, J. and Gosser, R. B., 1962, *Rev. Sci.Instruments.* **33**, 631.

Bolef, D. I. and Menes, M., 1961, *J. Phys. Chem. Solids.* **19**, 79.

Guermeur, R., Joffrin, J., Levelut, A. and Penne, J., 1964, *Phys. Rev. Letters.* **13**, 107.

Guermeur, R., Joffrin, J., Levelut, A, and Penne, J., 1965, *Phys. Rev. Letters.* **15**, 203.

Jacobsen, E. H. and Stevens, K. W. H. 1963, *Phys. Rev.*, **129**, 2063.

Jacobsen, E. H., Shiren, N. S. and Tucker, E. B., 1959, *Phys. Rev. Letters.* **3**, 81.

Kastler, A., 1952, *Experientia.* **8**, 1.

Kossel, A. R., 1962, *Soviet Phys. JETP.* **14**, 895.

Lowe, I. J., and Norberg, R. E., 1957, *Phys. Rev.* **107**, 46.

Menes, M. and Bolef, D. I., 1958, *Phys. Rev.* **109**, 218.

Proctor, W. G. and Tantilla, W. *H.*, 1956, *Phys. Rev.* **101**, 1757.

Protector, W. G. and Robinson, W., 1956, *Phys. Rev.* **104**, 1344.

Shiren, N. S., 1961, *Phys. Rev. Letters.* **4**, 168.

Also, (1962), Bulletin Du Groupement d' informations mutuelles, AMPERE, Compte Rendu, du 11° colloque. Eindhoven 2-6 Juillet.

————, 1962, *Phys. Rev.* **128**, 2103.

Tucker, E. B., 1961, *Phys. Rev. Letters.* **6**, 183.