ON THE THEORY OF THE SPIN ECHO

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A theoretical discussion is given of the effect of two resonance pulses—an acoustic and an electromagnetic one, and also of two acoustic pulses—on the nuclear spin system in a cubic crystal. Expressions describing the effect of the acoustic pulse on the free precession signal due to the electromagnetic pulse and expressions for the spin echo signals are obtained.

1. At the present time a large amount of varied information on the internal structure of many materials and on processes which occur in them is obtained from experiments involving spin echo and free nuclear precession produced by resonant electromagnetic pulses. An examination of similar phenomena involving acoustic pulses appears to be of interest.

It has been shown previously [1,2] that in cubic crystals an acoustic pulse of Larmor frequency $\omega_0 = \gamma H_0$ applied at $t = 0$, and an acoustic pulse of frequency $\omega = 2\omega_0$ applied at $t = \tau$, give rise to a spin echo signal at $t = 2\tau$. A single acoustic pulse does not give rise to a free precession signal [2] and alters the z component of magnetization from M_0 to $M_0 \cos\beta$ or $M_0 \cos 2\gamma$ [3] (β and γ will be defined later in terms of parameters describing the substance and the pulse).

In order to carry out an experiment with two acoustic pulses it is necessary to introduce into the crystal sound at the Larmor frequency and at double the Larmor frequency, or to reduce to one-half the magnitude of the magnetic field within a time $\tau < T_1$ (T_1 is the longitudinal relaxation time). These difficulties can be avoided if one electromagnetic and one acoustic pulse is applied to the substance.

Since the z component of the magnetization does not precess, its response to an acoustic pulse could be determined, for example, from the magnitude of the free precession signal from the second electromagnetic pulse. It is, therefore, of interest to study the results of applying an acoustic and an electromagnetic pulse to a nuclear spin-system. In the present article we examine the theory of this phenomenon for the case $I = 1$.

2. We consider a cubic crystal containing nuclei of spin $I = 1$ and of quadrupole moment Q placed in a magnetic field H_0. We follow the method of calculation presented in [2].

If acoustic oscillations are introduced into the crystal along the C_4 symmetry axis longitudinal acoustic standing waves are set up in the crystal.[2] The operator for the interaction of the ultrasound with the nucleus giving rise to resonance transitions in the Zeeman spectrum is given by [2]

$$\hat{h}(t) = -\hbar\,[2\omega_1(\hat{I}_x\hat{I}_z + \hat{I}_z\hat{I}_x) + \omega_2(\hat{I}_+ + \hat{I}_-)]\cos\omega t,$$

$$\omega_1 = \frac{3e^2Qq_1Ak}{8I(2I-1)\hbar}\sin\mathbf{kR}\sin 2\theta,$$

$$\omega_2 = \frac{3e^2Qq_1Ak}{8I(2I-1)\hbar}\sin\mathbf{kR}\sin^2\theta, \qquad (1)$$

where 2A and $\mathbf{k}$ are the oscillation amplitude and the propagation vector lying in the xy plane and making an angle θ with respect to the z axis chosen parallel to H_0, and eq_1 is the electric field gradient with respect to a displacement between the particles resulting from the oscillations about the equilibrium distance between them; $\mathbf{R}$ is the position vector of the nucleus in the coordinate system fixed in the crystal. We neglect the time required to set up the standing waves in the crystal.

For a pulse with the oscillating magnetic field $2H_1 \cos\omega t$ directed parallel to the x axis, the operator for the interaction with the nucleus is, as is well known,

$$\hat{h}(t) = -\hbar\omega^1(\hat{I}_+ + \hat{I}_-)\cos\omega t, \qquad \omega^1 = \gamma H_1. \qquad (2)$$

After the pulse has been applied the wave function for the nuclear spin can be represented in the form

$$\Psi(t) = \sum_m C_m(t_\omega)\, e^{-iE_m t/\hbar}\chi_m, \qquad (3)$$

where $C_m(t)$ is determined by the Schrödinger equation with the Hamiltonian $\hat{\mathcal{H}}_0 + \hat{h}(t)$, with the operator $\hat{h}(t)$ given in the form (1) or (2), while $\hat{\mathcal{H}}_0$, E_m and χ_m are respectively the Hamiltonian, its eigenvalue, and its eigenfunction in the absence of a pulse.

In the case of an acoustic pulse of frequency $\omega = \omega_0 = \gamma H_0$ and of duration $t_\omega \gg \omega^{-1}$ we obtain* for $C_m(t)$

$$C_{\pm 1}(t) = C^0_{\pm 1}\cos^2\frac{|\omega_1|t}{2} + C^0_{\mp 1}\sin^2\frac{|\omega_1|t}{2} \pm \frac{i}{\sqrt{2}}\,\mathrm{sign}\,\omega_1\, C^0_0 \sin|\omega_1|t,$$

$$C_0(t) = C^0_0\cos|\omega_1|t + \frac{i}{\sqrt{2}}\,\mathrm{sign}\,\omega_1\,(C^0_1 - C^0_{-1})\sin|\omega_1|t, \quad (4)$$

where $C^0_m = C_m(t = 0)$.

For an acoustic pulse with $\omega = 2\omega_0$ and $t_\omega \gg \omega^{-1}$ we have

$$C_{\pm 1}(t) = C^0_{\pm}\cos|\omega_2|t + i\,\mathrm{sign}\,\omega_2\, C^0_{\mp 1}\sin|\omega_2|t, \quad C_0(t) = C^0_0. \quad (5)$$

For an electromagnetic pulse we have

$$C_{\pm 1}(t) = C^0_{\pm 1}\cos^2\frac{\omega^1 t}{2} - C^0_{\mp 1}\sin^2\frac{\omega^1 t}{2} + \frac{i}{\sqrt{2}}C^0_0\sin\omega^1 t,$$

$$C_0(t) = C^0_0\cos\omega^1 t + \frac{i}{\sqrt{2}}(C^0_1 + C^0_{-1})\sin\omega^1 t. \quad (6)$$

With the aid of (3) — (6) we can evaluate the expectation values for the components of the nuclear spin. When two pulses are applied this procedure is carried out twice, with the nuclear state just before the second pulse being given by formula (3) with $t = \tau$.

In order to obtain the resultant magnetization it is necessary to add the expectation values of the components of the spin of all the nuclei in the sample. In order to do this, we shall average over the nuclear position vector, treating $\mathbf{k}\cdot\mathbf{R}$ as arbitrary, and over the frequency distribution, which we assume to be Gaussian.[5] Since at $t = 0$ the nuclei were in thermal equilibrium we shall make use of Boltzman statistics for the initial distribution of the N nuclei of the sample.

The results of such a program of calculations for different combinations of pulses are given in the table where

$$\alpha = \omega^1 t_\omega, \quad \beta = |\omega_1| t_\omega, \quad \gamma = |\omega_2| t_\omega,$$

$$f(p) = \exp[-i\omega_0 p - p^2/2T_2^{*2}],$$

a bar over an expression denotes averaging over the nuclear coordinates, and T_2^{*-2} is the mean square deviation of the frequency from γH_0.

The average value of the x and y components of the spin of an individual nucleus after an acoustic pulse of frequency ω_0 is proportional to

First pulse	Second pulse	$M^+ = M_x + iM_y$ in units of $2N\gamma\hbar^2\omega_0/3kT$
ac., $\Delta m = 1$	e. m.	$i\sin\alpha\,\overline{\cos\beta}\,f(t-\tau)$
ac., $\Delta m = 1$	ac., $\Delta m = 1$	0
ac., $\Delta m = 1$	ac., $\Delta m = 2$	$\mathrm{sign}(\omega_1\omega_2)\,\overline{\sin\beta\sin\gamma}\,f(t-2\tau)$
ac., $\Delta m = 2$	e.m.	$i\sin\alpha\,\overline{\cos 2\gamma}\,f(t-\tau)$
ac., $\Delta m = 2$	ac., $\Delta m = 1$	$\mathrm{sign}(\omega_1\omega_2)\,\overline{\sin\beta\sin 2\gamma}\,\{\overline{\cos^2(\beta/2)}\,f(t+\tau) + \overline{\sin^2(\beta/2)}\,f(t-3\tau)\}$
ac., $\Delta m = 2$	ac., $\Delta m = 2$	0
e. m.	ac., $\Delta m = 1$	$i\frac{1}{2}\sin\alpha\,\{\overline{(\cos 2\beta + \cos\beta)}\,f(t) + \overline{(\cos 2\beta - \cos\beta)}\,f(t-2\tau)\}$
e. m.	ac., $\Delta m = 2$	$i\sin\alpha\,\overline{\cos\gamma}\,f(t)$

$\mathrm{sign}\,\omega_1\cdot\sin|\omega_1|t_\omega$. In averaging over the coordinates, this expression vanishes if the dimension of the sample parallel to the direction of propagation of sound is an even multiple n of half a wavelength. If n is odd, then the total magnetization is determined by the nuclei situated in a layer half a wavelength thick in the direction perpendicular to the direction of propagation of sound.

For a sample of linear dimensions ~ 1 cm and for a frequency $\nu \sim 10^7$ cps this layer will contain ~ 1% of all the nuclei. Moreover, as a result of averaging over the initial conditions it turns out that the total magnetization in the x and y directions is proportional to $(\hbar\omega_0/kT)^2$, i.e., it is smaller by a factor $(\hbar\omega_0/kT)$ than the magnetization due to the electromagnetic pulse $(\hbar\omega_0/kT) \sim 10^{-6}$ at $T \sim 300°$ K and $H_0 \sim 10$ kilogauss. Therefore, it is impossible to observe the free precession signal due to an acoustic pulse.

3. It should be noted that the results shown in the table have been obtained by neglecting terms proportional to $(\hbar\omega_0/kT)^2$. Also we have not taken into account here processes associated with longitudinal relaxation, for, as is well known, in a solid the nuclear longitudinal relaxation times satisfy $T_1 \gg T_2^*$.

From the table it can be seen that in those cases when the electromagnetic pulse follows the acoustic pulse only a free precession signal appears following the second pulse. The ratio of this signal to the free precession signal due to a single electromagnetic pulse is equal to $P = \cos|\omega_{1,2}|t_\omega$. This can be easily understood if we take into account the fact that a single acoustic pulse does not give rise to a free precession signal and alters the value of the z-component of the magnetization from M_0 to $M_0\cos|\omega_{1,2}|t_\omega$ and that, moreover, the free precession signal following the electromagnetic pulse is proportional to the magnetization existing before it was applied.

*I take this opportunity to note that the expression sign $\omega_{1,2}$ has been omitted for the corresponding formulas of [2]. Apart from the sign, however, this does not affect the results of [2].

In those cases when the acoustic pulse follows the electromagnetic one a free precession signal from the first electromagnetic pulse is present and, therefore, a spin echo signal can be produced. In this case, as expected, no free precession signal is produced by the second pulse.

The ratio of the spin echo signal due to the electromagnetic and the acoustic pulses giving rise to the $\Delta m = 1$ transitions, to the echo signal due to two electromagnetic pulses [5] is given by

$$P' = \overline{(\cos 2\beta - \cos\beta)} / 2 \sin^2 \frac{\alpha}{2}.$$

A measurement of the values of P and P′ provides a method for studying the changes in the crystalline electric fields at nuclear sites as a result of the application of ultrasound.

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