Theory of Ultrasonic Spin Echoes*

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A theory of ultrasonic spin echoes valid for arbitrary spin is presented. Semiclassical vector-model equations are constructed for components of the dipole and quadrupole moment tensors which are excited by ultrasonic pulses in analogy with the well-known Bloch equations. The echo amplitudes are found to depend on the wave number of the pulse, the crystal length, spin density, sound velocity, the density of the medium as well as the magnitude of the appropriate macroscopic dipole or quadrupole moment. Our numerical estimates for a number of physical systems indicate that ultrasonic-spin-echo experiments are feasible for a variety of nuclear and electronic spin systems.

I. INTRODUCTION

Since the discovery of nuclear-magnetic-spin echoes by Hahn[1] in 1950, the spin-echo technique[2] has become a powerful tool for the study of relaxation phenomena in a large variety of physical and biological systems. It has long been known[3] that spin-echo phenomena can also be induced by acoustic pulses, and recently interesting experiments[4,5] on spin echoes induced by acoustic pulses or a mixture of acoustic and electromagnetic pulses have been reported. The purpose of the present paper is to present a theory of ultrasonic spin echoes valid for arbitrary spin which makes possible detailed physical interpretations of experimental observations.

It is well known that acoustic pulses usually couple to the nuclear or electronic spins via their quadrupole moments. Because of this, the usual Bloch equations cannot be applied to the spin systems excited by acoustic pulses. In fact if one writes down the Heisenberg equations of motion for the spin operator or the magnetization vector, one finds that it is coupled to the quadrupole moments. Proceeding in this way, one would obtain a hierarchy of equations linking all the multipole moments of the system. Such a complicated set of equations would make the physical interpretation of acoustic-spin-echo experiments very difficult if not impossible. We have found, however, that for spin-one systems at any temperature or for higher-spin systems in the high-temperature limit ($kT \gg \hbar\Omega$, where Ω is the Larmor frequency) the hierarchy of equations decouple in the usual resonant approximation. The decoupling is such that semiclassical vector models can be used to describe the effects of acoustic and/or electromagnetic pulses on the spin system.

We derive five sets of Bloch-like equations to describe the precession of suitably defined Bloch vectors which consist of components of the dipole and/or quadrupole moment tensors. Three additional damping or relaxation functions besides the usual ones corresponding to T_1 and T_2 can be defined which describe the relaxation of the $\Delta m = \pm 2$, ± 1, and 0 components of the quadrupole moment tensor. These additional relaxation functions, measurable in ultrasonic-spin-echo experiments, give a more complete characterization of the spin system under study. The Bloch-like equations greatly facilitate the interpretation of experimental observations and enhance the utility of the ultrasonic-spin-echo technique as a tool in solid-state physics.

In this paper we also compute the ultrasonic echo amplitudes for two-pulse sequences. They are found to depend on the wave number of the exciting pulses, the crystal length, spin density, sound velocity in the medium, and the density of the medium, as well as the magnitude of the appropriate components of the Bloch vectors. Numerical estimates of acoustic pulse widths and amplitudes necessary for 90° pulses and of the acoustic strain pulse height for echo pulses are given for the Fe^{++} electron-spin resonance in MgO : Fe^{++}, the In^{115} nuclear-spin resonance in InAs, and the Mn^{55} nuclear-spin resonance in antiferromagnetic $RbMnF_3$. Based on these estimates we believe that ultrasonic-spin-echo experiments are feasible for nuclear-spin as well as electronic-spin systems.

In Sec. II we give the essence of the derivation of the Bloch-like equations for ultrasonic exciting pulses where some of the lengthier calculations are deferred to the Appendix. In Sec. III we give the echo amplitudes for two-pulse sequences and in Sec. IV we present some detailed numerical examples.

II. BLOCH-LIKE EQUATIONS

The well-known Bloch equations for the motion of an effective magnetization $\vec{\mathfrak{M}}$ due to an effective magnetic field $\mathcal{K}$ can be written in the form

$$\frac{\partial \vec{\mathfrak{M}}}{\partial t} = \gamma \vec{\mathfrak{M}} \times \vec{\mathcal{K}}, \tag{1}$$

where γ is the effective gyromagnetic ratio. We shall show that, under the appropriate conditions, the equations of motion for spins driven by an acoustic field can be cast into the form of Eq. (1) with suitably defined magnetizations $\vec{\mathfrak{M}}$ and effective fields $\vec{\mathcal{H}}$. The appropriate conditions are that either the spin J of the magnetic system is equal to 1, or that the high-temperature limit is obtained. The high-temperature limit, for our purposes, is that $\hbar\Omega \ll kT$, where Ω is the Larmor frequency of the spins and T is the temperature.

While an electromagnetic field couples to spin tensors of the first rank (commonly called the dipole moments or components of the magnetization), acoustic fields usually couple to spin tensors of the second rank (commonly called quadrupole moments). We define these multipole operators in the following way:

$$A^1_{\pm 1} = \mp [\tfrac{2}{3} J(J+1)]^{-1/2} J_\pm , \quad (2a)$$

$$A^1_0 = [\tfrac{1}{3} J(J+1)]^{-1/2} J_z , \quad (2b)$$

$$A^2_{\pm 2} = [\tfrac{2}{15} J(J+1)(2J-1)(2J+3)]^{-1/2} J^2_\pm , \quad (2c)$$

$$A^2_{\pm 1} = \mp [\tfrac{2}{15} J(J+1)(2J-1)(2J+3)]^{-1/2} \{J_\pm , J_z\} , \quad (2d)$$

$$A^2_0 = [\tfrac{1}{45} J(J+1)(2J-1)(2J+3)]^{-1/2} \times [J^2_Z - \tfrac{1}{3} J(J+1)] , \quad (2e)$$

where J_i is the usual angular momentum operator with magnitude J, $J_\pm = J_X \pm iJ_y$, and the curly brackets in Eq. (2d) denote the anticommutator. Equations (2) define a set of irreducible tensor operators all of which are normalized so that the trace of $|A|^2$ is $2J+1$. These properties make them very easy to work with for our purposes. The operators are, of course, proportional to the usual dipole and quadrupole operators.

We shall consider an ensemble of nuclear or electronic spins with magnitude J and gyromagnetic ratio γ in a static external magnetic $\vec{H}_0$ along the z axis. These spins will be perturbed with either an electromagnetic or acoustic pulse whose width is small compared to the spin-relaxation times. For a standing-wave electromagnetic pulse the appropriate Hamiltonian is

$$H_{em} = -2\hbar\gamma H_1 J_x \cos\omega t , \quad (3)$$

where H_1 is an rf magnetic field. The spin-phonon coupling via quadrupolar or single-ion magnetostriction is described by the Hamiltonian

$$H_{ac} = -\hbar \sum_{m=-2}^{+2} \kappa_m e_m A^2_m \cos[\omega(t - x/v)] , \quad (4)$$

for an acoustic pulse of velocity v traveling in the x direction. In this equation e_m is the appropriate elastic strain and κ_m, which has the units of frequency, satisfies the relation

$$(\kappa_m e_m)^* = (-1)^m \kappa_{-m} e_{-m} .$$

The usual spin-phonon coupling can be cast into this form and in Sec. IV the κ_m will be expressed in terms of more familiar constants.

There is also a class of spin-phonon couplings where the acoustic wave couples to the dipole moments of the magnetization, A^1_m, and not to the quadrupole moments A^2_m. The magnetoelastic interaction with nuclear and electronic spins in magnetically ordered materials and the Alpher-Rubin interaction are examples of such couplings. These cases are described by Eq. (3) if H_1 is proportional to the strain and $\cos\omega t$ is replaced by $\cos[\omega(t - x/v)]$. The main body of the paper is not concerned with these couplings, but they will be analyzed in Sec. IV.

Since the derivation of the Bloch-like equations of motion is somewhat lengthy and complicated, the details of their derivation are given in the Appendix. We find that five sets of semiclassical Bloch vectors can be defined and that each set consists of the expectation values of the components of multipole operators A^μ_m. These sets of Bloch vectors satisfy the Bloch-like equations of motion, Eq. (1), which are more conveniently written

$$\begin{aligned} \frac{\partial \mathfrak{M}_x}{\partial t} &= \omega_0 \mathfrak{M}_y - \omega_1 \mathfrak{M}_z \sin(\omega t - \phi) , \\ \frac{\partial \mathfrak{M}_y}{\partial t} &= -\omega_0 \mathfrak{M}_x + \omega_1 \mathfrak{M}_z \cos(\omega t - \phi) , \\ \frac{\partial \mathfrak{M}_z}{\partial t} &= \omega_1 \mathfrak{M}_x \sin(\omega t - \phi) - \omega_1 \mathfrak{M}_y \cos(\omega t - \phi) , \end{aligned} \quad (5)$$

where ϕ is zero for standing-wave pulses and $\omega x/v$ for pulses traveling along the x axis with a velocity v. The five sets of Bloch vectors are conveniently summarized in Table I where the angular brackets $\langle\ \rangle$ denote the thermal average of the quantity enclosed.

Each of the five rows in this table gives a set of three $\mathfrak{M}_i$ as well as the ω_1 and ω_0 which are to be used in Eqs. (5). We have labeled the sets by the letters M and U to denote whether the driving field is electromagnetic (M) or acoustic (U). The first column in Table I gives the type of applied pulses which couples to the appropriate Bloch vectors. We note that a given type of pulse can couple to or excite more than one Bloch vector. For example a M pulse of a given frequency excites both the dipole Bloch vector and suitable components of the quadrupole operators as indicated in rows 1 and 2 of Table I. Similarly for an U pulse as indicated in rows 3 and 5 of Table I. Furthermore a given component of a dipole or quadrupole operator can couple to both types of pulses. As a consequence of this, ecno signals may be obtained by applying

TABLE I. The vector components of the effective magnetization $\vec{\mathfrak{M}}$ and the values of the effective ω_0 and ω_1 to be used in the Bloch equation given by Eq. (5). The cases are labeled by M or U according to whether the driving field is electromagnetic or acoustic. Case M_1 and M_2 are valid for arbitrary spin. Cases U_1 and U_2 are valid for $J=1$ or in the high-temperature limit. Case U_3 is valid only for $J=1$.

Type	$\mathfrak{M}_z$	$\mathfrak{M}_x$	$\mathfrak{M}_y$	ω_1	ω_0
M_1	$\langle A_0^1\rangle$	$\langle A_{-1}^1 - A_1^1\rangle/\sqrt{2}$	$i\langle A_1^1 + A_{-1}^1\rangle/\sqrt{2}$	γH_1	γH_0
M_2	$\langle A_0^2\rangle$	$\langle A_{-1}^2 - A_1^2\rangle/\sqrt{2}$	$i\langle A_1^2 + A_{-1}^2\rangle/\sqrt{2}$	$\sqrt{3}\,\gamma H_1$	γH_0
U_1	$\langle A_0^1\rangle$	$\langle A_{-1}^2 - A_1^2\rangle/\sqrt{2}$	$i\langle A_1^2 + A_{-1}^2\rangle/\sqrt{2}$	$[\frac{2}{3}J(J+1)]^{-1/2}\,\lvert\kappa_1 e_1\rvert$	γH_0
U_2	$\langle A_0^1\rangle$	$\langle A_2^2 + A_{-2}^2\rangle/\sqrt{2}$	$-i\langle A_2^2 - A_{-2}^2\rangle/\sqrt{2}$	$[\frac{1}{6}J(J+1)]^{-1/2}\,\lvert\kappa_2 e_2\rvert$	$2\gamma H_0$
U_3	$\langle A_0^2\rangle$	$\langle A_{-1}^1 - A_1^1\rangle/\sqrt{2}$	$i\langle A_1^1 + A_{-1}^1\rangle/\sqrt{2}$	$\frac{3}{2}\,\lvert\kappa_1 e_1\rvert$	γH_0

a large variety of pulse sequences which greatly enhances the flexibility of the technique as a tool in solid-state physics. The results of Table I together with the Bloch-like equations (5) enable us to calculate the precession of the various components of dipole or quadrupole operators driven by the applied pulses in exactly the same way as is usually done with the magnetization vector in an rf magnetic field. The fact that the acoustic pulses are traveling waves rather than standing waves can be taken into account and gives rise to some interesting new effects.[4] Detailed calculations on echo signals which arise from different applied pulse sequences will be given in Sec. III.

We conclude this section by summarizing the assumptions under which the results of Table I were derived. The pulse duration is assumed short in comparison with the spin-relaxation times and nonresonant terms are neglected. Within these assumptions the cases M_1 and M_2 are exact for arbitrary spin. The cases U_1, U_2, U_3 are exact for $J=1$. The cases U_1 and U_2 are also true if one makes the additional high-temperature approximation and neglects corrections of order $(\hbar\Omega/kT)$ compared to one. In this regard we wish to point out that in the high-temperature limit the length of the Bloch vectors in cases M_2 and U_3 are proportional to $(\hbar\Omega/kT)^2$ whereas those in the remaining cases are proportional to $\hbar\Omega/kT$.

III. ECHO SIGNALS

In Sec. II we discussed the free precession of the dipole and quadrupole tensors excited by magnetic M or acoustic U pulses. Since more than one set of Bloch-like vectors can be excited by either M or U pulses and a given component of the dipole or quadrupole tensor operator may couple to both M or U pulses, then it appears that echo signals will arise by applying two-pulse sequences in any possible combination, namely, MM, UU, UM, and MU. It will turn out that echo signals will arise in the first three cases, but not the last. In addition we find that there is a standing-wave echo for an MM pulse sequence, a traveling wave echo parallel to the applied acoustic pulses for a UU pulse sequence, and a traveling wave antiparallel to the applied acoustic pulse for a UM pulse sequence. No echo signal will arise if the two applied acoustic pulses travel in opposite directions. The traveling-wave acoustic echo amplitude is also found to depend on the wave number of the pulse, the crystal length, spin density, sound velocity and the density of the medium, as well as the magnitude of the appropriate macroscopic dipole or quadrupole moment as calculated from the Bloch-like equations (5).

Consider two interaction regions A and B in a crystal separated by a distance l. For an applied standing-wave pulse, the spins in both regions will be excited at the same time. On the other hand, for an applied traveling-wave pulse, there will be a time delay of $t_0 = l/v$ (where v is the velocity of the traveling wave) in the interaction time for the spins in the two regions. Because of this, the time separation between the two applied pulses may be different in the two regions A and B. As a result the time at which the spins in the two region rephase again will also be different. For a standing-wave echo signal to arise the spins in the two regions must rephase at the same time. For a traveling-wave echo signal to arise, the time at which rephasing is completed for the spins in the two regions must differ by t_0. In Table II we have listed all the possible sequences of two applied pulses with the interaction time and the rephasing time for the spins in the two separate regions. If a spin is pulsed first at time t_1 and at time t_2, the time of rephasing is $t_2 + (t_2 - t_1)$.

From Table II and using the criterion for echo signals to arise, one arrives at Table III which gives the direction and type of echo signals which can arise in a two-pulse sequence. We note that the conditions that we have imposed are analogous to the phase-matching conditions in optical photon-echo experiments. In arriving at Table III we have

TABLE II. The type of echo pulse obtained from various exciting pulse sequences where S denotes a standing-wave pulse and R and L denote traveling-wave pulses to the right and left, respectively. In each case the first row gives the time that the first pulse reaches spins in regions A and B. The second row gives the time that the second pulse reaches spins in regions A and B. The third row gives the times at which the spins in these regions rephase.

Case	Pulses	Interaction and rephasing time at region A	Interaction time and rephasing time at region B
S–S	S	0	0
	S	τ	τ
	S	2τ	2τ
	R	0	t_0
R–R	R	τ	$\tau+t_0$
	R	2τ	$2\tau+t_0$
	R	0	t_0
R–S	S	τ	τ
	L	2τ	$2\tau-t_0$
	S	0	0
S–R	R	τ	$\tau+t_0$
	None	2τ	$2\tau+2t_0$
	R	0	t_0
R–L	L	τ	$\tau-t_0$
	None	2τ	$2\tau-3t_0$

also assumed that macroscopic components of $\langle A^1_{\pm 1}\rangle$ will generate M pulses and those of $\langle A^2_{\pm m}\rangle$ primarily generate acoustic pulses.

In general, an applied electromagnetic pulse will include both M_1 and M_2 components, and an acoustic pulse at frequency $\omega_0=\gamma H_0$ will contain both U_1 and U_3 components while an acoustic pulse at frequency $\omega_0=2\gamma H_0$ will contain only the U_2 component and is decoupled from the U_1 and U_3 components. However since the effective magnetization $\vec{\mathfrak{M}}$ for M_1 and U_1 have no components in common with the effective magnetization $\vec{\mathfrak{M}}$ for M_2 and U_3, respectively, there is no interference between M_1 and M_2 components or between U_1 and U_3 components. They may therefore be treated independently. In addition, because of our assumption that macroscopic components of dipole moment generate M pulses and those of quadrupole moment generate U pulses, the M_2 component of the applied electromagnetic pulse will generate ultrasonic-echo signal while the M_1 component will generate electromagnetic echo signal and can therefore be easily distinguished experimentally. This also has the consequence that the U_3 component of an applied acoustic pulse will not generate echo signals of either electromagnetic or acoustic type in any two-pulse sequence.

Finally, the magnitude of the echo pulse should be estimated. From the Bloch-like equations (5) and the ω_1 from Table I, the values of $\mathfrak{M}_x$ and $\mathfrak{M}_y$ after a two-pulse sequence can be obtained. Further, the amplitude of the voltage from a magnetic echo pulse is known. By turning the Hamiltonian given by Eq. (4) into an energy density, the amplitude of the acoustic echo pulse can easily be calculated.

The contribution to the free-energy density from the spin-phonon Hamiltonian given by Eq. (4) is

$$F=-\hbar\Sigma_m\kappa_m n_s\langle A^2_m(x,t)\rangle e_m(x,t), \qquad (6)$$

where n_s is the density of spins and $\langle A^2_m\rangle$ is the thermal average of the appropriate multipole operator. Only the $m=\pm 2$ components or the $m=\pm 1$ components are excited in a given experiment and the other components can be neglected. Assuming that the effective magnetization $\mathfrak{M}_+$, where

$$\mathfrak{M}_+=(\mathfrak{M}_x+i\mathfrak{M}_y)/\sqrt{2},$$

describes a traveling square pulse of height $\mathfrak{M}_{max}$ and any width, one obtains that the maximum induced strain e_{max} is

$$e_{max}=\hbar n_s|\kappa_m\mathfrak{M}_{max}|ql/2\rho v^2. \qquad (7)$$

In this equation q is the wave number of the acoustic wave, l is the length of the sample, ρ is the mass density of the sample, and v is the acoustic velocity.

IV. DISCUSSION

In Secs. II and III of this paper we have shown how pulsed acoustic experiments can be analyzed in a manner analogous to pulsed electromagnetic experiments. This section is devoted to some numerical estimates of echo pulse amplitudes for particular systems.

The Hamiltonian describing the interaction of an electronic spin with acoustic phonons in a cubic lattice is usually written

$$H_{s\text{-}p}=\tfrac{3}{2}G_{11}\Sigma_i e_{i,i}[S_i^2-\tfrac{1}{3}S(S+1)] + G_{44}\Sigma_i e_{i,i+1}\{S_iS_{i+1}\}, \qquad (8)$$

TABLE III. Some two-pulse sequences and the type of echo pulse induced. M and U stand for electromagnetic and acoustic pulses, respectively, while S, R, and L stand for standing wave, traveling wave to the right, and traveling wave to the left, respectively.

Pulse 1	Pulse 2	Echo Pulse 3
M_1 (S)	M_1 (S)	M (S)
U_1 (R)	U_1 (R)	U (R)
U_2 (R)	U_2 (R)	U (R)
M_2 (S)	M_2 (S)	U (S)
U_1 (R)	M_2 (S)	U (L)

where G_{11} and G_{44} are spin-phonon coupling constants and e_{ij} is the elastic strain. The Latin subscripts denote Cartesian components along cubic directions in the crystal and the spin operators are denoted by S instead of J. Using Eqs. (2) this is easily put into the form of Eq. (4), yielding

$$\kappa_2 e_2 = [\tfrac{2}{15} S(S+1)(2S-1)(2S+3)]^{1/2} \times [\tfrac{3}{8} G_{11}(e_{xx} - e_{yy}) - \tfrac{1}{2} i G_{44} e_{xy}], \quad (9a)$$

$$\kappa_1 e_1 = -[\tfrac{2}{15} S(S+1)(2S-1)(2S+3)]^{1/2} \times \tfrac{1}{2} G_{44}(e_{xz} - i e_{yz}), \quad (9b)$$

$$\kappa_0 e_0 = [\tfrac{1}{45} S(S+1)(2S-1)(2S+3)]^{1/2} \times \tfrac{3}{2} G_{11}(e_{zz} - \tfrac{1}{2} e_{xx} - \tfrac{1}{2} e_{yy}). \quad (9c)$$

These questions are appropriate only if the external magnetic field is along a cubic axis. If this is not the case a further rotation must be made.

As an example of such a system consider the impurity ion Fe^{++} in MgO which has the largest spin-phonon coupling constant known. Consider probing the $\Delta m = 2$ transition by applying U_2 pulses of longitudinal waves down the cube x axis with the external magnetic field along the z axis. Using a value[6] of $G_{11} = 800\ \text{cm}^{-1}$ with this $S = 1$ system, one obtains from Table I that

$$\omega_1 = 1.1 \times 10^{14} e_{xx}\ \text{sec}^{-1},$$

where e_{xx} is the strain amplitude in the pulse. With a strain amplitude of 10^{-6}, a time of 1.4×10^{-8} sec is necessary for a 90° pulse. Further, using $\rho = 3.6\ \text{g/cm}^3$, $v = 10^6$ cm/sec, and a spin density of $n_s = 10^{18}\ \text{cm}^{-3}$, the maximum strain amplitude of the echo pulse by Eq. (7) is

$$e_{max} = 0.96 \times 10^{-8} M_+(ql).$$

At a frequency of 1 GHz the value of (ql) for a crystal 1 cm long is 6.3×10^3. Further, if the spins are wholly inverted, $\mathfrak{M}_+$ can be as large as $\langle A_0^1 \rangle / \sqrt{2}$ which is 3.3×10^{-3} for the $\Delta m = 2$ resonance at a frequency of 1 GHz and a temperature of 4.2 K. Thus an echo strain greater than 10^{-7} obtains.

The Hamiltonian describing the quadrupolar interaction of nuclear spins with acoustic phonon in a cubic lattice is usually written

$$H_Q = [eQ/2I(2I-1)]((S_{11} - S_{12}) \Sigma_i e_{i,i} \times [I_i^2 - \tfrac{1}{3} I(I+1)] + S_{44} \Sigma_i e_{i,i+1} \{I_i, I_{i+1}\}), \quad (10)$$

where the spin operators are denoted by I instead of J. Again, by comparing with Eq. (2), we obtain

$$\kappa_{+2} e_2 = (eQ)[\tfrac{2}{15}(I+1)(2I+3)/I(2I-1)]^{1/2} \times [\tfrac{1}{8}(S_{11} - S_{12})(e_{xx} - e_{yy}) - \tfrac{1}{4} i S_{44} e_{xy}], \quad (11a)$$

$$\kappa_{+1} e_1 = (eQ)[\tfrac{2}{15}(I+1)(2I+3/I(2I-1)]^{1/2} \times \tfrac{1}{4} S_{44}(e_{xz} - i e_{yz}), \quad (11b)$$

$$\kappa_0 e_0 = (eQ)[\tfrac{1}{45}(I+1)(2I+3)/(2I-1)]^{1/2} \times \tfrac{1}{4}(S_{11} - S_{12})(e_{zz} - \tfrac{1}{2} e_{xx} - \tfrac{1}{2} e_{yy}). \quad (11c)$$

These equations, like Eqs. (9), are appropriate only if the external magnetic field points along a cube edge.

As an example of such a nuclear-spin system consider the In^{115} nuclear spin in InAs and again consider probing the $\Delta m = 2$ transition by applying U_2 pulses of longitudinal waves down the cube x axis with the external field along the z axis. Using a value[7] of $Q(S_{11} - S_{12}) = 23.9 \times 10^{-9}$ statcoulombs cm^{-1} with this $I = \frac{9}{2}$ system yields

$$\omega_1 = 3.3 \times 10^8 e_{xx}\ \text{sec}^{-1}.$$

With a strain amplitude of 10^{-6}, a time of 4.7×10^{-3} sec is necessary for a 90° pulse. Using $\rho = 5.67$ g/cm^3, $v = 3.83 \times 10^5$ cm/sec, and $n_s = 1.72 \times 10^{22}\ \text{cm}^{-3}$, the maximum strain amplitude of the echo pulse is

$$e_{max} = 7.32 \times 10^{-9} \mathfrak{M}_+(ql).$$

At a field of 10^4 G the $\Delta m = 2$ resonance is at a frequency of 1.87×10^7 Hz and the value of $\langle A_0^1 \rangle$ is 3.06 $\times 10^{-4}$ at a temperature of 4.2 K. With a crystal 1 cm long this yields a maximum stress of 4.8×10^{-10}.

As noted in Sec. I, there are a number of systems where the effective spin-phonon coupling is to the dipole moments of the spin operators. The effective H_1 (or ω_1) in these cases depends on the strain and they can be treated as M_1 cases (see Table I) except that the pulses are traveling-wave pulses and the appropriate rows of Table II must be used with them. Since the values of H_1 and the components of the strains they involve depend on the particular problem under consideration and the external conditions, we shall not enumerate them here. The amplitude of the ultrasonic-spin-echo pulse can be calculated with trivial extensions of Eqs. (6) and (7). If a frequency κ_1 is defined in terms of the effective H_1 such that

$$\gamma H_1 J_x = \kappa_1 e \mathfrak{M}_x, \quad (12)$$

then Eq. (7) can be used with no alternation.

As an example of such a case consider the Mn^{55}

nuclear spin in antiferromagnetic $RbMnF_3$ which has the strongest known nuclear-spin-phonon coupling presents an almost ideal situation for pulsed acoustic experiment.[8] Our estimate of the ω_1 for coupling to the nuclear ω_{n-} mode at $T=4.2$ K is

$$\omega_1 \sim 10^{13} e ,$$

and the maximum echo strain amplitude is about 10^{-3} for a crystal 1 cm in length. Unfortunately also because of the strong coupling it is impossible to generate an acoustic signal strong enough so that it can be transmitted through a 1-cm sample under the above condition since the nuclear spins would absorb all of the acoustic energy. To overcome this we suggest the following: (i) Use a thin sample, (ii) use a sequence of pulses $U(R)$-$M(S)$ so that the echo pulse is $U(L)$, i.e., case R-S in Table II, or (iii) weaken the effective H_1 by means of reorienting the direction of the sublattice magnetization.

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APPENDIX

In this appendix we shall show that the Heisenberg equations of motion of the dipole and quadrupole operators under the Hamiltonian (3) and (4) can be cast into the form of Bloch-like equations (5). To do this we first evaluate the commutators among the irreducible spin tensor operators defined in Eqs. (2) for arbitrary spin J. First we have

$$[A_0^1, A_m^\mu] = [\tfrac{1}{3}J(J+1)]^{-1/2} m A_m^\mu , \tag{A1}$$

$$[A_{\pm 1}^1, A_m^\mu] = \mp [\tfrac{2}{3}J(J+1)]^{-1/2} \times [\mu(\mu+1) - m(m\pm 1)]^{1/2} A_{m\pm 1}^\mu , \tag{A2}$$

which can be easily deduced from the fact that A_m^μ are irreducible tensor operators. Next we need the commutators of A_m^2 among themselves which, in general, can be expressed as the sum of a third rank tensor and a first rank tensor except in the case of $J=1$ where the maximum rank of the spin tensor cannot exceed two. Thus we may write

$$[A_{\pm m}^2, A_{\mp m}^2] = a_m^J A_0^1 + b_m^J A_0^3 , \tag{A3}$$

$$[A_0^2, A_{\pm m}^2] = c_m^J A_{\pm m}^1 + d_m^J A_{\pm m}^3 , \tag{A4}$$

where b_m^1, d_m^1, and c_2^J are zero.

In the high-temperature limit $\langle A_0^3 \rangle$ is of order $(\hbar\Omega/kT)^3$ while $\langle A_0^1 \rangle$ is of order $(\hbar\Omega/kT)$, where $\langle A \rangle$ denotes the thermal average of A. Since we are ultimately concerned with constructing equations of motion for sets of $\langle A_m^\mu \rangle$'s, the third rank tensor term in Eq. (A3) is negligible in the high-temperature limit and will henceforth be neglected.

To determine the constants a_m^J and c_m^J we use the orthogonality properties of the irreducible tensors to obtain

$$\mathrm{Tr}[A_{\pm m}^2, A_{\mp m}^2] A_0^1 = (2J+1) a_m^J , \tag{A5}$$

$$\mathrm{Tr}[A_0^2, A_{\pm m}^2] A_{\mp m}^1 = (2J+1) c_m^J . \tag{A6}$$

A tedious but straightforward calculation then yields

$$a_1^J = -[\tfrac{1}{3}J(J+1)]^{-1/2}, \quad a_2^J = [\tfrac{1}{12}J(J+1)]^{-1/2} , \quad c_1^1 = -(\tfrac{3}{2})^{1/2} . \tag{A7}$$

Using these commutators, it is straightforward to derive the Heisenberg equations of motion for each case we are considering. An example will suffice. Consider the case U_1. The interaction Hamiltonian reads

$$H_I = \hbar\kappa_1 e_1 (A_1^2 - A_{-1}^2) \cos[\omega(t - x/v)] . \tag{A8}$$

The Heisenberg equations of motion for $A_{\pm 1}^2$, A_0^1 in the rotating wave approximation and using the commutators from (A1) and (A3) show that the time derivatives of the quantities $\langle A_{\pm 1}^2 \rangle$ and $\langle A_0^1 \rangle$ can be expressed in terms of these quantities themselves in the high-temperature limit. By appropriately scaling these quantities, one obtains the Bloch-like equation (5). This is in contrast to the case U_3 where the time derivatives of the quantities $\langle A_0^2 \rangle$, $\langle A_{\pm 1}^1 \rangle$ usually involve these quantities themselves together with $\langle A_{\pm 1}^3 \rangle$. The presence of these extra terms spoils the Bloch-like equations for this case except for $J=1$ since $d_m = 0$. Hence the case U_3 is only true for $J=1$. The case U_2 can be derived in analogous fashion as U_1.

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