PHYSICAL REVIEW B VOLUME 11, NUMBER 9 1 MAY 1975

Interaction of nuclear spins with phonons in a dense paramagnetic insulator

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(Received 27 August 1974)

A theory of high-temperature nuclear acoustic resonance (NAR) in a dense paramagnetic insulator is presented. First, by the use of a previously described diagrammatic technique, an equation of motion is derived and solved for the nuclear quadrupolar spin-correlation function in a dense paramagnet. From this function, the $T = \infty$ NAR line shape is calculated and found to be Lorentzian. The NAR linewidths resulting from this calculation are found to be within observable range in many cases. Expressions for the acoustic attenuation due to both the dynamic nuclear quadrupolar interaction and the phonon modulation of the hyperfine interaction are then derived in the high-temperature limit by a calculation of the phonon self-energy due to these processes. By the use of the calculated nuclear quadrupolar correlation function and previously determined electron and nuclear functions for the correlation functions which occur in the expressions for the attenuation, it is shown that the attenuation due to the second process has no resonant part, while the resonant part of the first process is of such a magnitude that the observation of room-temperature NAR in Rb^{85} and Rb^{87} in $RbMnF_3$ and isomorphic compounds might be possible.

I. INTRODUCTION

The observation of nuclear acoustic resonance (NAR) in dense magnetic insulators has thus far been limited to temperatures well below the magnetic transition temperature.[1–10] Measurement of NAR in the nuclei of both partially magnetic and magnetic ions in the paramagnetic phase of such substances ought to be possible if the nuclei have sufficiently narrow acoustic resonance line shapes. If one assumes that the major nuclear-spin–phonon interaction is the dynamic nuclear quadrupolar interaction,[11,12] the possibility of such an observation is limited to nuclei with spin I greater than $\frac{1}{2}$ and to nuclei whose quadrupole moments are large enough to enable the resonant acoustic attenuation to be within the range of detection by present ultrasonic techniques. Since there exist nuclei in paramagnetic insulators which meet these criteria, it would be useful to have some knowledge of the linewidths and acoustic attenuation one would expect for such systems.

The purpose of the present paper is to theoretically examine the question of high-temperature NAR in dense magnetic insulators. In particular, the acoustic attenuation due to the dynamic quadrupolar nuclear-spin–phonon coupling mechanism[11,12] and due to phonon modulation of the hyperfine interaction[13] will be calculated for such a system. The latter mechanism will be included for completeness but will be shown to contribute no resonant attenuation at these temperatures. This paper contains two separate calculations; that of the NAR line shape and that of the magnitude of the acoustic attenuation. These calculations have been separated for clarity and for logical presentation.

The acoustic attenuation line shapes due to the dynamic quadrupolar spin-phonon coupling mechanism[11,12] are proportional to nuclear quadrupole spin correlation functions and observation of NAR is a direct measurement of these functions. In order to obtain quantitative predictions for the NAR line shapes and linewidths in various nuclei due to this mechanism, these functions will be calculated from first principles to lowest order in A/J, where A is a hyperfine energy and J is an exchange constant. Integral equations for these functions will be generated by a generalization of the $T=\infty$ diagrammatic technique used in a previous paper[14] (henceforth referred to as I) to obtain NMR line shapes and linewidths in magnetic insulators. Solutions to order A/J will then be obtained by the use of the microscopic self-consistent results of Ref. 15 for the electron-spin correlation function which occurs in the nuclear-function equations. From the solutions for the quadrupolar functions, NAR linewidths will be calculated for various nuclei in various magnetic insulators and some of them will be shown to be within observable range.

The method used to obtain the acoustic attenuation is based upon a calculation of the phonon self-energy and is discussed in several places.[16–22] From the theoretical expression for the acoustic attenuation, the maximum resonant attenuation for NAR will be calculated at room temperature for the Mn^{55}, Rb^{85}, and Rb^{87} nuclei in $RbMnF_3$. The infinite-temperature NAR line shapes and linewidths which were calculated from the results for the nuclear quadrupole-quadrupole spin correlation function will be used in order to obtain numerical values for the resonant attenuation. It is hoped that these predictions of NAR line shapes, linewidths, and acoustic-attenuation magnitude might stimulate some experimental interest in searching

for NAR in magnetic insulators at high temperatures.

For the purposes of this paper, high temperature will be taken to mean temperatures much greater than the magnetic transition temperature of the electronic spins in the system, or equivalently, temperatures where the infinite-temperature approximation for the dynamical spin-spin correlation functions is valid. In the context of this paper, the term "resonant" attenuation will be taken to mean that contribution to the acoustic attenuation which depends upon an externally applied magnetic field.

Section II is devoted to a discussion of the various interactions which occur in the physical model to be used and to a discussion of the notation that will be used throughout the paper. Section III contains a brief derivation and solution of the integral equations for the nuclear quadrupolar functions by the method of I. Predictions for the NAR linewidths of various nuclei in various paramagnets are also included in Sec. III. In Sec. IV the formalism for calculating the attenuation is briefly outlined and the approximations used in the calculation are discussed. Section V contains the results of the calculation and a discussion of the implications that these results have upon the possibility of the observation of NAR in a dense paramagnet at room temperature. The implications that they have for NAR in $RbMnF_3$ are particularly emphasized. Some defects of the theory and suggestions for overcoming them are also discussed in Sec. V.

The new results of this paper are the diagrammatic derivation of equations of motion for the nuclear quadrupolar spin correlation functions and the first-principles solution of these equations, the prediction of NAR linewidths for nuclei in several paramagnets, the derivation for paramagnetic insulators of the high-temperature acoustic attenuation due to both the dynamic quadrupolar interaction[11,12] and to the phonon modulation of the hyperfine interaction,[13] the use of microscopically determined correlation functions for the line shapes, the proof that the phonon modulation of the hyperfine interaction does not contribute to the resonant attenuation at $T=\infty$, and estimates of the maximum attenuation due to the dynamic quadrupolar interaction for the Mn^{55}, Rb^{85}, and Rb^{87} nuclei in $RbMnF_3$ which indicate that NAR might be observable in the latter two nuclei.

II. PHYSICAL MODEL

It is assumed that the Hamiltonian for the dense magnetic insulator can be written as a sum of electron-spin, phonon, nuclear-spin, electron-spin–phonon, and nuclear-spin–phonon parts:

$$H = H_S + H_P + H_I + H_{SP} + H_{IP} \,. \tag{1}$$

The electron-spin part H_S is assumed to be describable by an isotropic Heisenberg paramagnet and therefore has the form shown in Eq. (1) of I and in Eq. (18) of Ref. 15. Since the dynamics of the nuclear spins are of primary interest in the present paper, the explicit form of this interaction will not be shown here. Also, since the dynamics of the electron spins affect the nuclear spins only through the presence of the electron-spin–spin correlation functions in the equations of motion for the nuclear-spin–spin correlation function,[14] and since the effects of an external magnetic field and the electron-spin dipole-dipole interactions are only small perturbations on the Heisenberg interactions,[14,15,22] these two effects will be neglected in the following discussion.

Following Ref. 20, it is assumed that since the effect of interest is the change in the phonon spectrum due to spin-phonon interactions, it is sufficient to use the harmonic approximation for the phonons. In terms of phonon normal coordinates, this has the form[16,20]

$$H_P = \frac{1}{2}\sum_{\vec{q},\lambda} [\dot{Q}(\vec{q},\lambda,t)\dot{Q}(-\vec{q},\lambda,t) + \omega^2(\vec{q},\lambda)Q(\vec{q},\lambda,t)Q(-\vec{q},\lambda,t)] \,, \tag{2a}$$

where $\vec{q}$ is a wave vector in the first Brillouin zone, λ specifies the phonon branch, and $\omega(\vec{q},\lambda)$ is the harmonic frequency of the $(\vec{q},\lambda)$ phonon mode. The normal coordinate $Q(\vec{q},\lambda,t)$ and its canonical momentum $\dot{Q}(\vec{q},\lambda,t)$ satisfy the usual commutation relations:

$$[\dot{Q}(\vec{q},\lambda,t),\ Q(\vec{q}',\lambda',t)] = -i\hbar\,\delta(\lambda,\lambda')\delta(\vec{q}+\vec{q}') \,, \tag{2b}$$

$$[Q(\vec{q},\lambda,t),\ Q(\vec{q}',\lambda',t)] = [\dot{Q}(\vec{q},\lambda,t),\ \dot{Q}(\vec{q}',\lambda',t)] = 0 \,. \tag{2c}$$

For a crystal with cubic symmetry, the Hamiltonian describing the nuclear-spin system is assumed to have the form

$$H_I = -\hbar\gamma H_0 \sum_{\vec{l}} I_z(\vec{l},t) - \sum_{\vec{l},\vec{l}'} \{[A_\parallel(\vec{l}-\vec{l}') - A_\perp(\vec{l}-\vec{l}')]\, I_z(\vec{l},t)S_z(\vec{l}',t) + A_\perp(\vec{l}-\vec{l}')\vec{I}(\vec{l},t)\cdot\vec{S}(\vec{l}',t)\} \,, \tag{3}$$

where $\vec{S}(\vec{l},t)$ and $\vec{I}(\vec{l},t)$ are the electron- and nuclear-spin operators at site $\vec{l}$ in the Heisenberg representation, H_0 is the external magnetic field which has been taken to be in the z direction, the subscript z on the spin operators means the z Cartesian component, γ is the nuclear gyromagnetic ratio, and $A_\parallel(\vec{l}-\vec{l}')$ and $A_\perp(\vec{l}-\vec{l}')$ are the interaction energies, parallel and perpendicular to the magnetic field direction, between a nuclear spin at site $\vec{l}$ and an electron spin at site $\vec{l}'$. Following I, it is assumed that the Hamiltonian of Eq.

(3) has a negligible effect on the dynamics of the electron spins so that the electron-spin–spin correlation function which was calculated in Ref. 15 can be used for the electron function which will occur in the equations for the nuclear quadrupolar functions discussed below. In addition, the effect of the nuclear dipole-dipole interaction is assumed to be small enough to be neglected.

The spin-phonon interactions to be considered in this paper are taken to be linear in the strains or displacements and quadratic in the spin operators. The electron-spin–phonon interactions H_{SP} are discussed in several places[17–22] and are also assumed to have negligible effect on the dynamics of the nuclear spins. The nuclear-spin–phonon interactions will be taken to consist of two parts:

$$H_{IP} = H_{IP}^{Q} + H_{IP}^{A} , \tag{4}$$

where H_{IP}^{Q} is the contribution due to the dynamic nuclear quadrupolar interaction[11,12] and H_{IP}^{A} is the contribution due to phonon modulation of the hyperfine interaction.[13] The dynamic nuclear quadrupolar interaction may be written[11,12]

$$H_{IP}^{Q} = \sum_{\vec{l}} \vec{\vec{Q}}(\vec{l}) : \vec{\nabla}\vec{E}(\vec{l}) , \tag{5a}$$

where $\vec{\vec{Q}}(\vec{l})$ is the nuclear electric-quadrupole-moment tensor at site $\vec{l}$ and $\vec{\nabla}\vec{E}(\vec{l})$ is the crystalline electric-field-gradient tensor at the same site. In the static case, all of the field-gradient components in Eq. (5a) vanish for a crystal with cubic symmetry. However, in the presence of an external acoustic wave, the cubic symmetry is destroyed and one may expand the field-gradient tensor in powers of the induced-strain tensor. If one keeps only terms linear in the strains, such an expansion takes the form

$$(\vec{\nabla}\vec{E})_{ij} = S_{ijkl}\epsilon_{kl} , \tag{5b}$$

where ϵ_{kl} is the induced-strain tensor, S_{ijkl} is the gradient elastic tensor which has been discussed elsewhere[11,12,23–25] and repeated Cartesian indices are summed over. For a crystal with cubic symmetry, there are two nonvanishing components of the gradient elastic tensor which are denoted in the Voigt notation as S_{11}-S_{12} and S_{44}.[24,25]

If one expands the strain tensor in phonon normal coordinates in the usual manner,[16,20–22] expresses the quadrupole moment tensor in terms of its equivalent nuclear-spin operators,[11,26] and substitutes the result into Eqs. (5a) and (5b), it is shown in detail in Appendix A that the nuclear-spin–phonon Hamiltonian H_{IP}^{Q} can be written

$$H_{IP}^{Q} = \frac{ieQ\eta_I}{2(MN)^{1/2}} \sum_{\vec{l},\vec{q},\lambda} |\vec{q}|\, e^{i\vec{q}\cdot\vec{l}} A_{2m}(\vec{l}, t) \times W_{-m}(\vec{q}, \lambda) Q(\vec{q}, \lambda, t) , \tag{6}$$

where e is the electronic charge, Q is the nuclear quadrupole moment, m is summed from -2 to 2, the $A_{2m}(\vec{l}, t)$ are the irreducible-tensor nuclear-spin operators defined in Eq. (3) of I, M is the mass of the displaced ion, N is the number of lattice sites,

$$\eta_I = [(I+1)(2I+3)/30I(2I-1)]^{1/2} ,$$

and the $W_{-m}(\vec{q}, \lambda)$ are functions that depend upon the directions of polarization and propagation of the phonons and upon the components of the gradient elastic tensor. These functions are shown explicitly in Appendix A.

In order to obtain the form of the nuclear-spin–phonon interaction H_{IP}^{A} that has its origin in the phonon modulation of the hyperfine interaction, one begins with the hyperfine part of the interaction given by Eq. (3) and expands the hyperfine eneggies $A_{\parallel}(\vec{l}-\vec{l}')$ and $A_{\perp}(\vec{l}-\vec{l}')$ in a power series in the displacements from equilibrium.[13] If one keeps only terms which are linear in the displacements and expands those displacements in phonon normal coordinates,[16] it is shown in Appendix B that H_{IP}^{A} may be written

$$H_{IP}^{A} = \frac{1}{(MN)^{1/2}} \sum_{\vec{l},\vec{l}',\vec{q},\lambda} (e^{i\vec{q}\cdot\vec{l}'} - e^{i\vec{q}\cdot\vec{l}}) \times Q(\vec{q}, \lambda, t) F(\vec{q}, \lambda, \vec{l}, \vec{l}', t) , \tag{7}$$

where $F(\vec{q}, \lambda, \vec{l}, \vec{l}', t)$ is a linear function of the nuclear spin at site $\vec{l}$, the electronic spin at site $\vec{l}$, the spatial derivatives of the hyperfine energies of Eq. (3), and the phonon polarization direction of the $(\vec{q}, \lambda)$ phonon mode. The explicit form of $F(\vec{q}, \lambda, \vec{l}, \vec{l}', t)$ is shown in Appendix B.

III. NUCLEAR QUADRUPOLAR CORRELATION FUNCTION

A. Derivation of the equations of motion

The notation that will be used here for the spin correlation functions is exactly the same as that used in Ref. 15 and in I and is discussed at length in those references. In particular, at $T=\infty$, these functions are defined as

$$G_{\alpha\beta}^{J}(\vec{l}, \vec{l}', t-t') = \langle A_{\alpha}^{J}(\vec{l}, t) A_{\beta}^{J\dagger}(\vec{l}', t') \rangle \Theta(t-t') , \tag{8}$$

where the A_{α}^{J} are the irreducible-tensor spin operators defined in Eq. (3) of I and J can be either I for the nuclear spin or S for the electron spin. The translational invariance of the system in time and the invariance of the crystal lattice under translations through a lattice vector enable one to Fourier transform Eq. (8) by the prescription given in Eq. (5) of I to obtain $G_{\alpha\beta}^{J}(\vec{q}, \omega)$. This Fourier-transformed function with $J=I$ and $\alpha=\beta=(2, m)$ will be calculated in this section and will be shown in Sec. IV to be proportional to the acoustic attenuation due to the dynamic quadrupolar interaction.[11,12]

The method which will be used here to derive the

equations of motion for the quadrupolar correlation functions is a generalization of that used in I for the dipolar functions and is thoroughly discussed there. The detailed arguments of the derivation will therefore not be discussed here; the reader is instead referred to Sec. II of I, where the discussion is sufficiently general as to be applicable, with appropriate modifications, to the quadrupolar as well as the dipolar functions.

The first step is to specialize the correlation function $G^J_{\alpha\beta}$ to the case of interest, $J=I$ and $\alpha=\beta=(2,m)$, i.e., the nuclear quadrupolar function. In what follows, this function will, where possible, be denoted simply as G^I_α, while the electron dipolar spin correlation function will be denoted as G^S_α. Following I, the nuclear quadrupolar function is now expressed in terms of a mass operator or self-energy function Σ^I_α. This representation takes the form

$$i\frac{\partial}{\partial t}G^I_\alpha(\vec{q},t)-\int d\bar{t}\,\Sigma^I_\alpha(\vec{q},t-\bar{t})G^I_\alpha(\vec{q},\bar{t})=i\delta(t). \qquad (9)$$

In order to derive an equation for the self-energy functional Σ^I_α, it is convenient to consider the nuclear-spin system described by Eq. (3) with $H_0=0$. It is shown in Appendix A of I that the presence of a magnetic field does not affect the NAR line shapes and linewidths. This result will follow directly if one goes through the arguments of Appendix A of I with the replacement $1m\to 2m$. It is shown in Sec. V of this paper, however, that the presence of a magnetic field affects the position in frequency at which the NAR line will occur as well as the angular dependence of the acoustic attenuation. So for the purpose of obtaining the line shapes themselves, magnetic field effects will be neglected. For the evaluation of the magnitude of the attenuation discussed in the following sections, however, these effects will be included.

For the nuclear-spin system described by the $H_0=0$ version of Eq. (3), the criteria for the existence of a nuclear-spin self-energy Σ^I_α which can be written as a functional of both G^I_α and G^S_α are discussed in detail in Sec. II of I and carry over directly to the quadrupolar-function case. Integral equations for the quadrupolar correlation functions will therefore be obtained by forming the Reiter-type[27] moment diagrams and by resumming or renormalizing these diagrams by the method of Ref. 15. The moment diagrams must, of course, be generalized here to include the nuclear quadrupolar operators $A^I_{2,m}$ given in Eq. (3) of I. The graphical representation of these operators is shown in Fig. 1.

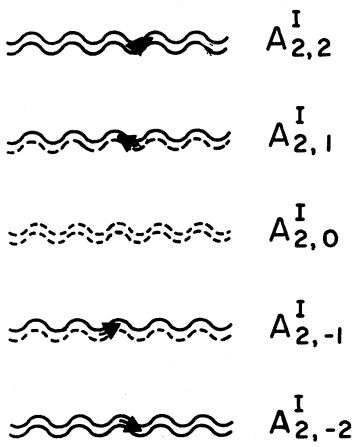

FIG. 1. Diagrammatic representation of the nuclear quadrupolar tensor spin operators at a given lattice site and a given time.

The basic vertices at $T=\infty$ for the quadrupolar functions are obtained from matrix elements of the Liouville operator exactly as in I. Following I, the discussion will be limited to only these basic vertices since they give all moments exactly to order A/J. The only basic vertices which contribute for the Hamiltonian of Eq. (3) are shown in Fig. 2 along with the Fourier transforms of their analytic expressions. The reduced hyperfine energies $\alpha_\parallel(\vec{q})$ and $\alpha_\perp(\vec{q})$ which occur in that figure are defined in Eq. (7) of I. The smooth lines in these vertices are the lines for the electron spins, defined in Fig. 1 of I, and will be assumed to be known functions.

An expression for the spin self-energy Σ^I_α can now be obtained by putting together the vertices of Fig. 2 and by following the rules and procedures described in Ref. 15 and in I. The rest of this section shall be concerned only with the lowest-order or "bubble" approximation to the self-energy. The "bubble" diagrams are the diagrams which can be formed from just two of the vertices of Fig. 2. In the absence of an external magnetic field there are three independent quadrupolar correlation functions with labels $\alpha=(2,\pm 2)$, $\alpha=(2,\pm 1)$, and $\alpha=(2,0)$, and for isotropic exchange all electron dipolar correlation functions $\alpha=(1,m)$ are equal. Thus the abbreviations

$$G^I_2\equiv G^I_{2,\pm 2},\quad G^I_1\equiv G^I_{2,\pm 1},\quad G^I_0\equiv G^I_{2,0},\quad \text{and } G^S\equiv G^S_\alpha$$

shall be made in what follows with a similar notation for the spin self-energy functions. The "bubble" approximation to the nuclear quadrupolar spin self-energies is therefore

$$\Sigma^I_2(\vec{q},t)=\frac{2}{N}\sum_{\vec{q}'}G^S(\vec{q}',t)\{2[\alpha_\parallel(\vec{q}')]^2G^I_2(\vec{q}-\vec{q}',t)+[\alpha_\perp(\vec{q}')]^2G^I_1(\vec{q}-\vec{q}',t)\}, \qquad (10a)$$

$$\Sigma_1^I(\vec{q},t)=\frac{1}{N}\sum_{\vec{q}'} G^S(\vec{q}',t)\{2[\alpha_\perp(\vec{q}')]^2G_2^I(\vec{q}-\vec{q}',t)+[\alpha_\parallel(\vec{q}')]^2[G_1^I(\vec{q}-\vec{q}',t)+3G_0^I(\vec{q}-\vec{q}',t)]\}\,, \tag{10b}$$

$$\Sigma_0^I(\vec{q},t)=\frac{6}{N}\sum_{\vec{q}'}[\alpha_\perp(\vec{q}')]^2G^S(\vec{q}',t)G_1^I(\vec{q}-\vec{q}',t)\,. \tag{10c}$$

These equations reproduce the frequency moments of $G_\alpha^I(\vec{q},t)$ exactly to lowest order in A/J. Equations (9) and (10) form a set of nonlinear integral equations for the nuclear quadrupolar correlation functions. These sets of equations are closed if it is assumed that the electron dipole-dipole correlation function is known. In Sec. II B, the solutions to Eqs. (10) will be obtained and their implications for NAR experiments in dense paramagnets will be discussed.

B. Results for the quadrupolar functions–NAR line shapes

In order to solve the integral equations for the nuclear quadrupole-quadrupole correlation functions which were derived in subsection *A* and thus obtain NAR line shapes and linewidths, one must proceed in exactly the same manner as was outlined in Sec. III of I for the dipolar functions. In particular, these nuclear spin correlation functions and their corresponding self-energy functions are first expressed in terms of spectral representations, as given in Eqs. (9)–(11) of I, where α in those equations can now mean 2, 1 or 0. When Eqs. (10) are Fourier transformed and the imaginary parts are taken, the following equations are obtained for the quadrupolar functions $\Gamma_\alpha^I(\vec{q},\omega)$:

$$\Gamma_2^I(\vec{q},\omega)=\frac{2}{N}\sum_{\vec{q}'}\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}g^S(\vec{q}',\omega')\{2[\alpha_\parallel(\vec{q}')]^2g_2^I(\vec{q}-\vec{q}',\omega-\omega')+[\alpha_\perp(\vec{q}')]^2g_1^I(\vec{q}-\vec{q}',\omega-\omega')\}\,, \tag{11a}$$

$$\Gamma_1^I(\vec{q},\omega)=\frac{1}{N}\sum_{\vec{q}'}\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}g^S(\vec{q}',\omega')\{2[\alpha_\perp(\vec{q}')]^2g_2^I(\vec{q}-\vec{q}',\omega-\omega')+[\alpha_\parallel(\vec{q}')]^2 \times[g_1^I(\vec{q}-\vec{q}',\omega-\omega')+3g_0^I(\vec{q}-\vec{q}',\omega-\omega')]\}\,, \tag{11b}$$

$$\Gamma_0^I(\vec{q},\omega)=\frac{6}{N}\sum_{\vec{q}'}[\alpha_\perp(\vec{q}')]^2\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}g^S(\vec{q}',\omega')g_1^I(\vec{q}-\vec{q}',\omega-\omega')\,. \tag{11c}$$

Equation (11) of I with $\alpha=2,1,0$, and Eqs. (11) above are the nonlinear integral equations that will be discussed here.

Again following the treatment of the dipolar functions in I, only the nuclear autocorrelation function will be kept and thus the wave-vector dependence of the quadrupole-quadrupole function will be neglected. This wave-vector independence can again be confirmed for low frequencies by computing moments. Furthermore, only the case of isotropic hyperfine coupling will be considered for this function. The reason for this is that the anisotropic case does not appear to be relevant for discussing NAR line shapes in any of the common nuclei in dense paramagnetic insulators. In particular, the study of the quadrupolar functions is limited to nuclei with spin $I>\frac{1}{2}$, and the observation of NAR requires nuclei with large quadrupole moments Q.[11,12] As far as can be determined, in the common paramagnetic insulators with cubic structure, the only nuclei with $I>\frac{1}{2}$ and with large Q have isotropic rather than anisotropic hyperfine coupling.

The specialization to isotropic hyperfine coupling means that the hyperfine energy is described by Eq. (14) of I and that the three quadrupolar correlation functions are equal. Equations (11) therefore reduce to one independent equation. In what follows the nuclear quadrupolar spectral function

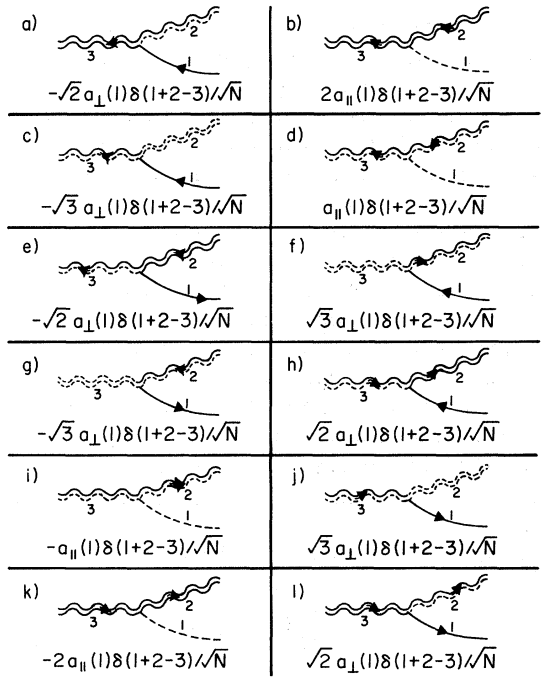

FIG. 2. Basic vertices which contribute to the nuclear quadrupolar self-energy for the electron-spin–nuclear-spin interaction of Eq. (3). The operators are at wave vectors $\vec{q}$ and the vertex is at time t. The reduced hyperfine energies defined in Eq. (7) of I act at the vertices. The abbreviations $3=\vec{q}$, $1=\vec{q}_1$, $2=q_2$ have been used.

will be denoted as g_Q with a similar notation being assumed for the linewidth function. In terms of the dimensionless variables defined in Sec. III of I, the wave-vector-independent equation for the quadrupolar correlation functions is therefore

$$\tilde{\Gamma}_Q(y) = \frac{6a^2}{V^2} \int_{-\infty}^{\infty} \frac{dy'}{\pi} \beta(y') \tilde{g}_Q(y - y') , \tag{12}$$

where $\beta(y)$ is defined and has been calculated in Sec. II of I with typical $\beta(y)$ shown in Fig. 4 of that reference. Of course, $\tilde{g}_Q$ can be related to $\tilde{\Gamma}_Q$ by Eqs. (16b) and (16c) of I, where now $\alpha = Q$ in those equations. The true solutions for Γ_Q and g_Q are given by Eqs. (18a) and (18b) of I and from those equations and Eq. (16a) of that reference, it can easily be seen that the frequency-dependent quadrupolar linewidths take an exchange-narrowed hyperfine-broadened form, which is a result similar to that found in I for the linewidths of the dipolar functions.

The same reasoning that led to the solution for the dipolar functions in Sec. II of I applies here as well. In particular, perturbation theory in A/J is valid here so as a first approximation for $\tilde{g}_Q(y)$ one can use Eq. (24) of I with $\alpha = Q$. Further, over the range of y for which $\tilde{g}_Q(y)$ is of appreciable magnitude, $\tilde{\Gamma}_Q(y)$ is independent of y. Equation (12) therefore gives

$$\tilde{\Gamma}_Q(y) \simeq \tilde{\Gamma}_Q(0) \simeq (6a^2/V^2)\beta(0) . \tag{13}$$

When substituted into Eqs. (16b) and (16c) of I, Eq. (13) clearly shows that to lowest order in A/J the NAR line-shape function $\tilde{g}_Q(y)$ (nuclear spectral function) is Lorentzian. As in the case of NMR line shapes in I, this Lorentzian behavior has been shown to follow directly from the equations of motion and the fact that $A/J \ll 1$.

Predictions for the high-temperature NAR linewidths of various nuclei in various paramagnetic insulators can be obtained from Eq. (13) by the use of the experimental hyperfine and exchange energies and the results obtained in I for the function $\beta(y)$. These linewidths have been computed for several nuclei in several substances and the results are shown in Table I. The quadrupole moments Q of the various nuclei are also listed in that table.

From the numbers in Table I, one can conclude that there are several nuclei in paramagnetic insulators which have linewidths small enough and quadrupole moments large enough to possibly make room-temperature NAR observable. Among these are Rb^{85} and Rb^{87} in $RbMnF_3$ and in $RbCoF_3$. Of course, the only definitive method of determining whether NAR would really be observable in these nuclei would be to perform the linewidth calculation described above *plus* a calculation of the resonant acoustic attenuation for them due to the dynamic nuclear quadrupolar interaction.[11,12] The latter calculation will be discussed in the following sections. It is hoped, however, that the linewidth calculations presented here will stimulate some experimental interest in looking for high-temperature NAR in paramagnetic insulators.

IV. CALCULATION OF THE ACOUSTIC ATTENUATION

The method that will be used to calculate the acoustic attenuation due to the spin-phonon processes described by H_{IP}^Q and H_{IP}^A of Sec. II is the same as that used in Refs. 18–22, so it will only be briefly discussed here. One can conveniently discuss the acoustical properties of the system by the use of the phonon Green's function[16]

$$D(\vec{q}, \lambda, t - t') = (1/\hbar)\langle (Q(\vec{q}, \lambda, t) Q(-\vec{q}, \lambda, t'))_+ \rangle , \tag{14}$$

where the angular brackets $\langle \cdots \rangle$ denote an average in the canonical ensemble and the circular brackets $(\cdots)_+$ denote the Wick time-ordering operation. $D(t)$ is Fourier transformed according to the usual prescription[16]

$$D(\omega_\nu) = \int_0^{-i\hbar\beta} dt\, e^{i\omega_\nu t} D(t) , \tag{15}$$

where $\omega_\nu = \pi\nu/i\hbar\beta$, $\beta = 1/kT$, and ν is an even integer. By the use of Eqs. (2), (6), (7), (14), and the Heisenberg equation of motion

$$i\hbar \frac{\partial X}{\partial t}(t) = [X(t),\ H_P + H_{IP}] , \tag{16}$$

one can obtain an exact equation of motion for the phonon Green's function. This equation is

$$\left(\frac{\partial^2}{\partial t^2} + \omega^2(\vec{q}, \lambda)\right) D(\vec{q}, \lambda, t - t') - \delta(t - t')$$

$$= \frac{ieQ\eta_I |\vec{q}|}{2(MN)^{1/2}} \sum_{\vec{l}} e^{-i\vec{q}\cdot\vec{l}} W_{-m}(-\vec{q}, \lambda) \langle (A_{2m}(\vec{l}, t) Q(-\vec{q}, \lambda, t'))_+ \rangle$$

$$+ \frac{1}{(MN)^{1/2}} \sum_{\vec{l}, \vec{l}'} (e^{-i\vec{q}\cdot\vec{l}} - e^{-i\vec{q}\cdot\vec{l}'}) \langle (F(\vec{q}, \lambda, \vec{l}, \vec{l}', t) Q(-\vec{q}, \lambda, t'))_+ \rangle , \tag{17}$$

Following Refs. 18–22, the terms on the right-hand side of Eq. (17) can be expanded in powers of $H_{IP} = H_{IP}^Q + H_{IP}^A$. Since H_{IP} is linear in the normal coordinate $Q(\vec{q}, \lambda, t)$ and since the thermal average of an odd number of such coordinates vanishes, only odd powers of H_{IP} will contribute to the expansion. When such

an expansion is made, Eq. (17) can be rewritten

$$\left(\frac{\partial^2}{\partial t^2}+\omega^2(\vec{q},\lambda)\right)D(\vec{q},\lambda,t-t')=\delta(t-t')+\int_0^{-i\hbar\beta}d\bar{t}\;\Pi(\vec{q},\lambda,t-\bar{t})D(\vec{q},\lambda,\bar{t}-t')\;. \tag{18a}$$

Upon Fourier transforming and rearranging, Eq. (18a) becomes

$$D(\vec{q},\lambda,\omega_\nu)=[-\omega_\nu^2+\omega^2(\vec{q},\lambda)-\Pi(\vec{q},\lambda,\omega_\nu)]^{-1}\;, \tag{18b}$$

where $\Pi(\vec{q},\lambda,\omega_\nu)$ is the phonon self-energy. If only the lowest-order terms in the expansion are kept, the self-energy can be written

$$\Pi(\vec{q},\lambda,\omega_\nu)=\Pi^Q(\vec{q},\lambda,\omega_\nu)+\Pi^A(\vec{q},\lambda,\omega_\nu)\;, \tag{19}$$

where Π^Q is the contribution from H_{IP}^Q and corresponds to the lowest-order expansion of the first term on the right-hand side of Eq. (17), and Π^A is the contribution from H_{IP}^A and corresponds to the lowest-order expansion of the second term on the right-hand side of Eq. (17). In general, there will also be contributions from H_{IP}^A in the first term on the right-hand side of that equation and from H_{IP}^Q in the second term. However, since such contributions will depend on correlation functions with odd numbers of nuclear or electronic spins, these contributions vanish in the high-temperature limit.

Explicitly, the two contributions to the phonon self-energy take the form

$$\Pi^Q(\vec{q},\lambda,\omega_\nu)=\frac{1}{4}\frac{e^2Q^2}{M}\eta_I^2|\vec{q}|^2\times(-1)^m W_{-m}(-\vec{q},\lambda)W_m(\vec{q},\lambda)\mathcal{G}_{2m}^I(\vec{q},\omega_\nu) \tag{20a}$$

and

$$\Pi^A(\vec{q},\lambda,\omega_\nu)=\frac{1}{\beta}\sum_{\nu'}\frac{1}{MN}\sum_{\vec{q}'}(-1)^m L_m(\vec{q},\vec{q}',\lambda)\times\mathcal{G}_{1m}^I(\vec{q}',\omega_{\nu'})\mathcal{G}_{1,-m}^S(\vec{q}-\vec{q}',\omega_\nu-\omega_{\nu'})\;, \tag{20b}$$

where m is summed from -2 to 2 in Eq. (20a) and from -1 to 1 in Eq. (20b) and $L_m(\vec{q},\vec{q}',\lambda)$ is a function that depends upon the phonon propagation and polarization directions and upon the spatial Fourier transform of products of spatial derivatives of the hyperfine energies of Eq. (3). The form of this function is shown in Appendix C. The functions $\mathcal{G}_{2m}^I(\vec{q},\omega_\nu)$, $\mathcal{G}_{1m}^I(\vec{q},\omega_\nu)$, and $\mathcal{G}_{1m}^S(\vec{q},\omega_\nu)$ are, respectively, the Fourier transforms of the nuclear quadrupole-quadrupole spin correlation function, the nuclear dipole-dipole spin correlation function, and the electronic dipole-dipole spin correlation func-

TABLE I. NAR linewidths at $T=\infty$.

Substance: Nucleus	Hyperfine constant $A(10^{-4}\ \mathrm{cm}^{-1})$	Theoretical linewidth (Gauss)	Quadrupole moment[j] $(10^{-24}\ \mathrm{cm}^2)$
$RbMnF_3$: Mn^{55}	-90.8[a]	1323.0	0.5
Rb^{85}	-0.0366 ± 0.0025[b]	1.03	0.31
Rb^{87}	-0.1210 ± 0.0073[b]	5.53	0.15
$KMnF_3$: Mn^{55}	-91.0[c]	1189.5	0.5
K^{39}	-0.0084[d]	0.167	• • •
MnO: Mn^{55}	-81.5 ± 2.2[e]	944.3	0.5
O^{17}	2.46 ± 0.05[f]	24.9	-4×10^{-3}
αMnS: Mn^{55}	-71.0 ± 5.3[e]	760.8	0.5
S^{33}	1.65 ± 0.04[f]	25.8	-6.4×10^{-2}
αMnSe: Mn^{55}	-67.0 ± 4.5[e]	782.2	0.5
$KCoF_3$: Co^{59}	-98.0[g]	305.1	0.5
CoO: Co^{59}	-98.0[g]	326.8	0.5
O^{17}	3.1[h]	12.9	-4×10^{-3}
$RbCoF_3$: Co^{59}	-98.0[g]	339.7	0.5
Rb^{87}	-6.8[i]	37.2	0.15
Rb^{85}	-2.06[i]	6.93	0.31
$TlMnF_3$: Mn^{55}	-90.8[a]	1249.8	0.5

[a]Reference 28.
[b]Reference 29.
[c]Reference 28, measured in $KMgF_3$.
[d]Reference 30.
[e]Reference 31.
[f]Reference 32.
[g]Reference 33.
[h]Reference 34.
[i]Reference 35.
[j]Reference 36.

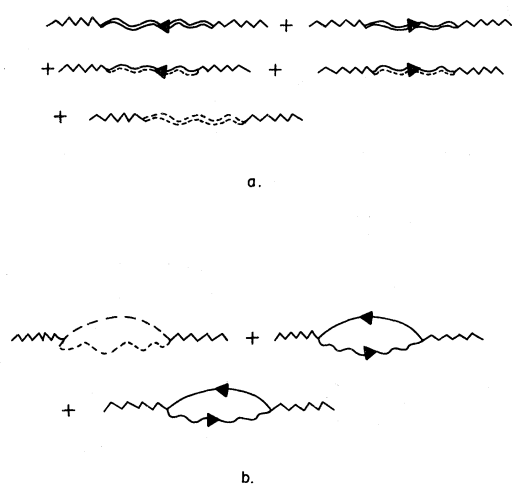

FIG. 3. (a) Diagrammatic representation of the phonon self-energy due to the dynamic nuclear quadrupolar interaction and described by Eq. (20a). (b) The diagrammatic representation of the phonon self-energy due to the phonon modulation of the hyperfine interaction and described by Eq. (20b). The spin lines have the meaning shown in Fig. 1 of I and Fig. 1 of the present paper, and the zig-zag lines are phonon lines.

tion. These functions are related to the correlation functions discussed in I and in Sec. III of this paper by the fluctuation-dissipation theorem.[37] In order to obtain Eq. (20b) it was necessary to assume that mixed electron-spin–nuclear-spin correlation functions like

$$\langle (I_z(\vec{1}, t) S_z(\vec{1}_2, t) I_z(\vec{1}_3, t') S_z(\vec{1}_4, t'))_+ \rangle$$

could be factored into the product of a nuclear-spin correlation function and an electronic-spin correlation function. Such a factorization ought to be valid in the high-temperature limit being considered here. Also, in both Eqs. (20a) and (20b) only spin-conserving spin correlation functions have been kept [i.e., keep only $\langle (I_+ I_-)_+ \rangle$, $\langle (I_z I_z)_+ \rangle$, $\langle (I_+^2 I_-^2)_+ \rangle$, $\langle (S_+ S_-)_+ \rangle$, etc.].

Equations (20a) and (20b) can be interpreted diagrammatically in a manner similar to the diagrammatic interpretation of the phonon self-energy discussed in Refs. 21 and 22. The diagrams corresponding to Eq. (20a) are shown in Fig. 3(a) and those corresponding to Eq. (20b) are shown in Fig. 3(b). In those figures, the diagrammatic notation which was introduced in I and in Sec. III above is used for the electronic- and nuclear-spin lines and the zig-zag lines are phonon lines.

In order to obtain the acoustic attenuation from Eqs. (20a) and (20b) one uses the prescription[16,21]

$$\alpha(\vec{q}, \lambda, \omega) = \mathrm{Im}\{\Pi(\vec{q}, \lambda, \omega + i\epsilon)/2 V_\lambda \omega(\vec{q}, \lambda)\}, \quad (21)$$

where ϵ is a positive infinitesimal, ω is a real frequency, and V_λ is the velocity of sound for the $(\vec{q}, \lambda)$ phonon mode.

In the present paper, only cubic crystal structures will be considered. It is then convenient, following Refs. 20–22 to define two coordinate systems; one which coincides with the crystal axes and one which has its z direction coinciding with the direction of the external magnetic field $\vec{H}_0$. In general, the direction of the external field is specified with respect to the crystalline axes by the spherical angles (ϑ, ϕ). The transformation which takes spin operators from one coordinate system to the other is discussed in detail in Refs. 20 and 22 so it will not be shown explicitly here. After making such a coordinate transformation on Eqs. (20a) and (20b), using the fluctuation dissipation theorem[37] to relate the correlation functions in those equations to those calculated in I, in Sec. III above, and in Refs. 15 and 22, and using the prescription shown in Eq. (21), the contributions of Π^Q and Π^A to the acoustic attenuation take the form

$$\alpha^Q(\vec{q}, \lambda, \omega) = \frac{\beta\omega |\vec{q}|^2 e^2 \eta_I^2 Q^2}{8 M V_\lambda \omega(\vec{q}, \lambda)} U_m(-\vec{q}, \lambda) U_{-m}(\vec{q}, \lambda) g_{2m}^I(\omega) \quad (22a)$$

and

$$\alpha^A(\vec{q}, \lambda, \omega) = \frac{\beta\omega}{2 M V_\lambda \omega(\vec{q}, \lambda)} \frac{1}{N} \sum_{\vec{q}'} K_{mm'}(\vec{q}, \vec{q}', \lambda) \times \int \frac{d\omega'}{\pi} g_{1m}^I(\omega') g_{1m'}^S(\vec{q} - \vec{q}', \omega - \omega'), \quad (22b)$$

where $\beta = 1/kT$, $U_m(\vec{q}, \lambda)$, and $K_{mm'}(\vec{q}, \vec{q}', \lambda)$ are related to $W_m(\vec{q}, \lambda)$ and $L_m(\vec{q}, \vec{q}', \lambda)$ by the coordinate transformation discussed above, $g_{2m}^I(\omega)$, $g_{1m}^I(\omega)$, and $g_{1m}^S(\vec{q}, \omega)$ are spectral functions[14,15,22] corresponding to the correlation functions of Eq. (20), in Eq. (22a) m is summed from -2 to 2, in Eq. (22b), m, m' are summed from -1 to 1, and the fact, shown in I, that $g_{1m}^I(\omega)$ and $g_{2m}^I(\omega)$ are independent of $\vec{q}$ has been taken into account. The function $g_{1m}^I(\omega)$ has been discussed in I, the function $g_{2m}^I(\omega)$ has been discussed in Sec. III, and the function $g_{1m}^S(\vec{q}, \omega)$ has been discussed in Refs. 15 and 22 and has been shown in these references to be independent of m (i.e., independent of the external magnetic field).

V. RESULTS AND CONCLUSIONS

As was stated earlier, the term "resonant" acoustic attenuation is taken to mean that portion of the attenuation which depends upon the external magnetic field $\vec{H}_0$. In the following discussion, only the resonant contributions to Eqs. (22a) and (22b) are of interest. By taking into account the fact that $g_{1m}^S(\vec{q}, \omega)$ in Eq. (22b) is independent of m, by explicitly looking at the form for $K_{mm'}(\vec{q}, \vec{q}', \lambda)$ in that equation, and by doing the sum on m and m' in $\alpha^A(\vec{q}, \lambda, \omega)$, it can be shown that each term in this sum is proportional to the linewidth function $\Gamma_{1m}^I(\vec{q}, \omega)$ which is defined in Eq. (A1a) of Appendix

TABLE II. Attenuation angular dependences for some typical phonon modes (cubic crystal). The z-axis is taken along a cubic crystalline axis, and $\tilde{S} \equiv \frac{1}{2}(S_{11} - S_{12})$.

Mode	$\lvert U_2(\vec{q}, \lambda)\rvert^2$	$\lvert U_1(\vec{q}, \lambda)\rvert^2$
$\vec{q} \parallel \hat{e} \parallel [001]$	$\frac{3}{8}\tilde{S}^2 \sin^4\vartheta$	$6\tilde{S}^2 \sin^2\vartheta\cos^2\vartheta$
$\vec{q} \parallel \hat{e} \parallel [110]$	$\frac{1}{4}[S_{44}\sin 2\phi(1+\cos^2\vartheta) + \frac{3}{4}\tilde{S}\sin^2\vartheta]^2$ $+S_{44}^2\cos^2 2\phi\cos^2\vartheta$	$\frac{1}{4}\sin^2 2\vartheta[S_{44}\sin 2\phi + \frac{3}{4}\tilde{S}]^2$ $+S_{44}^2\sin^2\vartheta\cos^2 2\phi$
$\vec{q} \parallel \hat{e} \parallel [111]$	$S_{44}^2[\cos\vartheta\cos 2\phi + \sin\vartheta(\cos\phi + \sin\phi)]^2$ $+\frac{1}{4}S_{44}^2[(1+\cos^2\vartheta)\sin 2\phi$ $+\sin 2\vartheta(\sin\phi - \cos\phi)]^2$	$\frac{1}{4}S_{44}^2[\cos 2\phi\ \sin 2\vartheta + 2(\cos\phi + \sin\phi)]^2$ $+\frac{1}{4}S_{44}^2[2(\sin\phi - \cos\phi)\cos 2\vartheta$ $-\sin 2\phi\ \sin 2\vartheta]^2$
$\vec{q} \parallel [001]$, $\hat{e} \parallel [\cos\psi,\ \sin\psi,\ 0]$	$\frac{1}{16}S_{44}^2[\sin^2(\phi - \psi) + \cos^2\vartheta\cos^2(\phi - \psi)]^2$	$\frac{1}{16}S_{44}^2[\cos^2\vartheta\sin^2(\phi - \psi)$ $+\cos^2 2\vartheta\cos^2(\phi - \psi)]^2$

A of I. It is shown in that appendix that this linewidth function is independent of the external magnetic field. Therefore, $\alpha^A(\vec{q}, \lambda, \omega)$ is independent of field and does not contribute to the resonant acoustic attenuation. In other words, the acoustic attenuation resulting from phonon modulation of the hyperfine interaction has no resonant part, at least in the high-temperature limit. Physically, this statement implies that phonon modulation of the hyperfine interaction will not induce transitions between the energy levels of the nuclear-spin system. The function $\alpha^A(\vec{q}, \lambda, \omega)$ therefore only contributes to the background attenuation and will be dropped in the following discussion.

The field dependence of the function $\alpha^Q(\vec{q}, \lambda, \omega)$, defined by Eq. (22a) can be explicitly seen if the form of the spectral function $g_{2m}^I(\omega)$ which occurs in that equation is examined. From Eq. (A2) of Appendix A of I with $1m \rightarrow 2m$, this function has the form

$$g_{2m}^I(\omega) = \Gamma_Q(\omega)/\{[\omega - m\omega_0 - \Pi_Q(\omega)]^2 + [\Gamma_Q(\omega)]^2\}, \quad (23a)$$

where $\omega_0 = \gamma H_0$. The fact that the functions $\Gamma_Q(\omega)$ and $\Pi_Q(\omega)$ are independent of the external field and thus of m has been assumed and can be proven by arguments exactly like those for the dipolar case in Appendix A of I. If, in that appendix, one lets $1m \rightarrow 2m$, all of the arguments there can be shown to apply in the quadrupolar case as well. The functions $\Pi_Q(\omega)$ and $\Gamma_Q(\omega)$ have been calculated in Sec. III for the case of isotropic hyperfine coupling. Also, from the field-independent properties of these functions and from the comparison of Eq. (23a) with Eqs. (16c) and (18b) of I, it can be seen that

$$g_{2m}^I(\omega) = g_Q(\omega - m\omega_0)\ . \quad (23b)$$

The function $g_Q(\omega)$ is the nuclear quadrupole-quadrupole spin spectral function for isotropic hyperfine coupling and has been calculated in Sec. III to lowest order in A/J. This calculation showed that this function is Lorentzian in shape and that this fact can be obtained without any *ad hoc* shape assumptions. Further, Eq. (23b) shows that the shape of $g_{2m}^I(\omega)$ is again Lorentzian, but with the center of the line shifted by $m\omega_0$. Thus, the effect of an external magnetic field is merely to shift the center of the line without changing the line shape.

Since only acoustic phonons are of interest here, the approximation $\omega(\vec{q}, \lambda) = V_\lambda |\vec{q}|$ may be made in Eq. (22a). Also, since only resonant terms are of interest, $\omega = \omega(\vec{q}, \lambda)$. When these facts are taken into account and only the resonant terms in the sum over m are kept in that equation, the result is

$$\alpha^Q(\vec{q}, \lambda, \omega) = \frac{\omega^2 e^2 Q^2 \eta_I^2}{8MV_\lambda^3 kT}\{|U_2(\vec{q}, \lambda)|^2[g_{22}^I(\omega) + g_{2,-2}^I(\omega)] + |U_1(\vec{q}, \lambda)|^2[g_{21}^I(\omega) + g_{2,-1}^I(\omega)]\}\ . \quad (24)$$

The functions $|U_2(\vec{q}, \lambda)|^2$ and $|U_1(\vec{q}, \lambda)|^2$ are dependent upon the angles (ϑ, ϕ) as well as on the phonon mode. They are tabulated in Table II for several typical phonon modes. Similar functions have been calculated by Fedders[19] for APR in cubic crystals and the expressions in Table II can be obtained by the replacement $G_{11},\ G_{44} \rightarrow \frac{1}{2}(S_{11} - S_{12}),\ S_{44}$ in his results. In the table the shorthand notation $\tilde{S} \equiv \frac{1}{2}(S_{11} - S_{12})$ has been used.

In order to obtain an order of magnitude estimate of the maximum resonant acoustic attenuation given by Eq. (24), only the longitudinal mode $\vec{q} \parallel \hat{e} \parallel [001]$ will be considered and the angles (ϑ, ϕ) will be taken as $(\frac{1}{2}\pi, 0)$. Also, only the resonance at $\omega = 2\omega_0$ will be explicitly considered. Other phonon modes and resonances will have a maximum attenuation which differs from this number by a constant of the order unity and perhaps by a replacement of $\frac{1}{2}(S_{11} - S_{12})$ by S_{44}. The maximum attenuation in this

TABLE III. Estimates of maximum resonant attenuation for Mn^{55}, Rb^{85}, Rb^{87} in $RbMnF_3$.

Nucleus	Resonant attenuation (cm^{-1})
Mn^{55}	7.23×10^{-8}
Rb^{85}	6.26×10^{-5}
Rb^{87}	5.21×10^{-5}

case is

$$\alpha^Q(2\omega_0)=\frac{3\omega_0^2e^2Q^2(S_{11}-S_{12})^2(I+1)(2I+3)g_{22}^I(2\omega_0)}{40MV^3kTI(2I-1)}\ . \tag{25a}$$

It should be noted in passing that the attenuation in Eq. (25a) displays all of the basic properties that are expected for a resonant acoustic attenuation at high temperature: it is proportional to ω_0^2, T^{-1}, and $(MV^3)^{-1}$.

From Eq. (23b) combined with Eq. (16c) of I, it can be seen that

$$g_{22}^I(2\omega_0)=1/\Gamma_Q(0)\ . \tag{25b}$$

$\Gamma_Q(0)$ is the NAR linewidth and has been tabulated for various nuclei in various substances in Table I. Once a particular substance and nucleus have been specified, all other quantities that occur in Eq. (25a) are known numerically from other sources except the gradient-elastic-tensor component $S_{11}-S_{12}$. Since this quantity has not been measured experimentally, at least for the nuclei listed in Table I, in order to obtain a numerical estimate of the maximum attenuation, it is necessary to postulate a model which will enable this constant to be calculated.

Taylor and Bloembergen[23] have proposed a point-charge model for approximately calculating the gradient-elastic-tensor components. Within this model, the component $S_{11}-S_{12}$ for a cubic lattice takes the form

$$S_{11}-S_{12}=11.8\ e^*(1-\gamma_\infty)/a^3\ , \tag{26}$$

where e^* is the effective ionic charge, a is the lattice spacing, and γ_∞ is the Sternheimer antishielding factor.[38–46] Sundfors[25] has improved upon a purely ionic point-charge model for the gradient-elastic-tensor components by including covalent effects but such an improvement will not be considered here and the component $S_{11}-S_{12}$ will be taken as given by Eq. (26). The model described by that equation is at least useful to enable one to make an order-of-magnitude estimate for the resonant attenuation.

The calculation will now be specialized even further, to the evaluation of the maximum acoustic attenuation for NAR in the Mn^{55}, Rb^{85}, and Rb^{87} nuclei in $RbMnF_3$. The reasons for considering only $RbMnF_3$ are that there will be less than an order of magnitude difference between the maximum attenuation for the Mn^{55} nucleus in the other manganese compounds listed in Table I and that for $RbMnF_3$, the quadrupole moments of the nonmagnetic nuclei in those compounds are smaller by two orders of magnitude than those of the Rb^{85}, Rb^{87}, and Mn^{55} nuclei, and the Sternheimer antishielding factor for the cobalt ion does not appear to be available in the literature.

The values for the maximum acoustic attenuation for Mn^{55}, Rb^{85}, and Rb^{87} in $RbMnF_3$ have been calculated for a frequency of 10 MHz and a temperature of 300 K using the linewidths and quadrupole moments which are listed in Table I. The results are listed in Table III. In order to calculate these numbers, the velocity of sound has been taken to be $V=5.22\times10^5$ cm/sec[47] and in order to calculate the gradient-elastic-tensor components by Eq. (26) the parameters $e^*=e$, $a=4.235$ Å,[47] $\gamma_\infty(Mn^{2+})=11.37$,[45] and $\gamma_\infty(Rb^+)=-70.7$[45] have been used.

From the values of the attenuation listed in Table III, one can immediately conclude that the observation of room-temperature NAR in the Mn^{55} nucleus in $RbMnF_3$ is probably impossible by present ultrasonic techniques.[48,49] On the other hand, the possibility of the observation of this phenomenon in the rubidium nuclei in that substance looks promising.[48,49] Unfortunately, however, since the NAR linewidths for these nuclei are so narrow, there is a possibility that this maximum absorption might be decreased due to the static field gradients which always occur in a real (as opposed to an ideal) crystal.[50] In addition, if one were to include covalent as well as ionic effects in the calculation of the gradient-elastic-tensor components, the numbers in Table III could change by several per cent.[25] The ideal situation with regard to the gradient-elastic-tensor components would be if one could obtain experimental values for $S_{11}-S_{12}$ which were obtained by a non-NAR technique.[23,51–54] The proper conclusion from these facts is that, under ideal experimental conditions, room-temperature NAR in Rb^{85} and Rb^{87} might be observable by present ultrasonic techniques in $RbMnF_3$ or perhaps in an isomorphic rubidium compound.

Of course, as the temperature is lowered, while still remaining above the magnetic transition temperature, the attenuation is enhanced due to the T^{-1} dependence. At the same time, however, the high-temperature limit for the attenuation and the infinite-temperature limit for the spin correlation functions become less accurate and a more general temperature dependence for these functions must be taken into account.

To summarize, two different kinds of calculations were carried out in this paper. The first

was the first-principles calculation of the NAR line shape by the derivation and solution of the equation of motion for the quadrupole-quadrupole nuclear-spin correlation function. The derivation of the equation was carried out by an extension of the diagrammatic technique of I and the solution for the NAR line shape, $g_Q(\omega)$, was found to be Lorentzian, Numerical values for NAR linewidths for various nuclei in various paramagnetic insulators were obtained from the line-shape results by the use of the electron-spin correlation function which was microscopically determined in Ref. 15 and these linewidths, shown in Table I, are within observable range in many cases.

In the second calculation, expressions for the acoustic attenuation due to both the dynamic quadrupolar interaction[11,12] and the phonon modulation of the hyperfine interaction were derived in the high-temperature limit by a calculation of the phonon self-energy[17–22] due to these processes. The resulting attenuation depends upon the nuclear-spin and electron-spin correlation functions which have been calculated previously. By use of the results for these functions, obtained in I, Ref. 15, and in Sec. III of the present paper, it was shown that the attenuation due to the phonon modulation of the hyperfine interaction has no resonant part and that the NAR line shape due to the dynamic quadrupolar interaction is given by the Lorentzian function $g_Q(\omega)$ of Sec. III with the center of the line displaced by a multiple of ω_0. In the case of the dynamic quadrupolar nuclear-spin–phonon interaction, the angular dependences for the resonances at $\omega = \pm\omega_0$ and $\omega = \pm 2\omega_0$ have been calculated for various modes and are shown in Table II. Also, for this case, the maximum resonant attenuation due to NAR in the Mn^{55}, Rb^{85}, and Rb^{87} nuclei in $RbMnF_3$ has been estimated by assuming a point-charge model to calculate the gradient-elastic-tensor components. The results of this estimate are shown in Table III and indicate that the Rb^{85} and Rb^{87} nuclei in $RbMnF_3$, or perhaps in an isomorphic compound, are, under ideal experimental conditions, good possibilities for the observation of NAR at room temperature.

In conclusion, two possible complimentary experiments are suggested by the above results. The first of these is a non-NAR type of experiment for the determination of the gradient-elastic-tensor components of the Rb^{85} and Rb^{87} nuclei in $RbMnF_3$, which might improve the estimate of the resonant attenuation by giving better values for these quantities. The second of these is a room-temperature NAR experiment on these nuclei in that substance.

APPENDIX A: FORM OF THE DYNAMIC NUCLEAR QUADRUPOLAR INTERATION

Beginning with Eq. (5a), one expands the field gradient tensor in terms of the strain as is shown in Eq. (5b). Then the Hamiltonian H_{IP}^{Q} takes the form

$$H_{IP}^{Q} = \sum_{\vec{l}} S_{ijkm} Q_{ij}(\vec{l}) \epsilon_{km}(\vec{l}) , \tag{A1}$$

where $Q_{ij}(\vec{l})$ is the ij Cartesian component of the quadrupole moment tensor at site $\vec{l}$, S_{ijkm} is the gradient elastic tensor, and $\epsilon_{km}(\vec{l})$ is the strain tensor at site $\vec{l}$. For a lattice with cubic symmetry, the only nonzero components of S_{ijkm} are $S_{11} - S_{12}$ and S_{44} (in Voigt notation).[24,25] The strain tensor can be expressed in terms of derivatives of the components of the displacement $\vec{U}$ as[55]

$$\epsilon_{ij} = \frac{1}{2}\left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i}\right) . \tag{A2}$$

and the displacement may be expanded in phonon normal coordinates as[16]

$$U_i(\vec{l}) = \frac{1}{(MN)^{1/2}} \sum_{\vec{q},\lambda} e_i(\vec{q},\lambda)\, e^{i\vec{q}\cdot\vec{l}}\, Q(\vec{q},\lambda) , \tag{A3}$$

where $e_i(\vec{q},\lambda)$ is the ith component of the phonon polarization vector and $Q(\vec{q},\lambda)$ is the phonon normal coordinate. The quadrupole moment tensor is most conveniently discussed in terms of its spherical tensor components and these are easily expressed in terms of their equivalent spherical tensor nuclear spin operators as[11,26]

$$Q_m(\vec{l}) = eQ\eta_I A_{2m}(\vec{l}) . \tag{A4}$$

Here, e is the electronic charge, Q is the nuclear quadrupole moment,

$$\eta_I = [(I+1)(2I+3)/30I(2I-1)]^{1/2} ,$$

and $A_{2m}(\vec{l})$ with $-2 \le m \le 2$ is the irreducible-tensor nuclear-spin operator defined in Eq. (3) of I.

Combining Eqs. (A1)–(A4) after some lengthy algebra, one obtains

$$H_{IP}^{Q} = \frac{ieQ\eta_I}{2(MN)^{1/2}} \sum_{\vec{l},\vec{q},\lambda} |\vec{q}|\, e^{i\vec{q}\cdot\vec{l}} A_{2m}(\vec{l},t) W_{-m}(\vec{q},\lambda) Q(\vec{q},\lambda,t), \tag{A5}$$

where the $W_{-m}(\vec{q},\lambda)$ have the form

$$\begin{aligned}
W_{\mp 2}(\vec{q},\lambda) &= [\tilde{S}(e_x q_x - e_y q_y) \pm 4iS_{44}(e_x q_y + e_y q_x)]/4|\vec{q}| , \\
W_{\mp 1}(\vec{q},\lambda) &= \pm S_{44}[e_z(q_x \pm iq_y) + (e_x \pm ie_y)q_z]/|\vec{q}| , \\
W_0(\vec{q},\lambda) &= \sqrt{3}\,\tilde{S}[2e_z q_z - e_x q_x - e_y q_y]/2|\vec{q}| .
\end{aligned} \tag{A6}$$

Here e_i is a shorthand notation for $e_i(\vec{q},\lambda)$, and $\tilde{S} \equiv \frac{1}{2}(S_{11} - S_{12})$.

APPENDIX B: FORM OF THE PHONON MODULATION OF THE HYPERFINE INTERATION

Beginning with Eq. (3), the hyperfine energies are expanded in a power series in the displacements $\vec{U}$ from equilibrium[13] and only the lowest-order terms are kept

$$A(\vec{l}-\vec{l}') = A(\vec{l}-\vec{l}')|_{eq} + [\vec{U}(\vec{l}) - \vec{U}(\vec{l}')] \cdot \nabla_{\vec{l}} A(\vec{l}-\vec{l}') . \tag{B1}$$

Here A means either $A_\parallel$ or $A_\perp$ and $|_{eq}$ means evaluation at equilibrium. By dropping the first term, expanding the quantities $\vec{U}(\vec{l})$ in Eq. (B1) in phonon normal coordinates as in Eq. (A2), and substituting that result into Eq. (3), the spin-phonon interaction can be seen to take the form

$$H_{IP}^{A} = \frac{1}{(MN)^{1/2}} \sum_{\vec{l},\vec{l}',\vec{q},\lambda} (e^{i\vec{q}\cdot\vec{l}'} - e^{i\vec{q}\cdot\vec{l}}) \times Q(\vec{q},\lambda,t) F(\vec{q},\lambda,\vec{l},\vec{l}',t) , \quad \text{(B2)}$$

where

$$F(\vec{q},\lambda,\vec{l},\vec{l}',t) = \hat{e}(\vec{q},\lambda)\cdot\{\nabla_{\vec{l}} A_\parallel(\vec{l}-\vec{l}') I_z(\vec{l}) S_z(\vec{l}') + \nabla_{\vec{l}} A_\perp(\vec{l}-\vec{l}')[I_x(\vec{l})S_x(\vec{l}') + I_y(\vec{l})S_y(\vec{l}')]\} . \quad \text{(B3)}$$

APPENDIX C: FORM OF THE FUNCTION $L_m(\vec{q},\vec{q}',\lambda)$

Beginning with Eqs. (17) and (19) and the results of Appendix B, some straightforward algebraic manipulation shows that the function $L_m(\vec{q},\vec{q}',\lambda)$ in Eq. (20b) has the form

$$L_0(\vec{q},\vec{q}',\lambda) = \tfrac{1}{6} I(I+1)S(S+1) \times\{\hat{e}(\vec{q},\lambda)\cdot[\vec{B}_\parallel(\vec{q}-\vec{q}')+\vec{B}_\parallel(\vec{q}')]\}^2 ,$$

$$L_{\pm1}(\vec{q},\vec{q}',\lambda) = \tfrac{1}{6} I(I+1)S(S+1) \times\{\hat{e}(\vec{q},\lambda)\cdot[\vec{B}_\perp(\vec{q}-\vec{q}')+\vec{B}_\perp(\vec{q}')]\}^2 . \quad \text{(C1)}$$

Here, $\hat{e}(\vec{q},\lambda)$ is the phonon polarization vector and

$$\vec{B}(\vec{q}) = \sum_{\vec{l}} e^{-i\vec{q}\cdot\vec{l}} \nabla_{\vec{l}} A(\vec{l}) = \vec{q} A(\vec{q}) . \quad \text{(C2)}$$

In Eq. (C2) B and A have the subscript $\parallel$ or $\perp$.

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