Sound propagation in magnets and its application to a planar ferromagnetic chain

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The velocity shift and the attenuation of ultrasonic waves in magnetic insulators are studied theoretically. By the treatment of the volume and single-ion magnetostrictive interactions on an equal footing, interesting interference effects between these two spin-phonon coupling mechanisms are possible. The results are applied to a planar ferromagnetic chain at low temperatures. The attenuation coefficient is found to depend linearly on the temperature and quadratically on the frequency. The velocity shift is also linear in the temperature but does not depend on the frequency.

I. INTRODUCTION

The spin-phonon interaction in magnetic systems and its influence on the propagation of ultrasonic waves through these materials have been of considerable interest for quite some time. Experimental and theoretical studies have been made on three-dimensional magnets at low temperatures[1,2] as well as near magnetic critical points.[3,4] A growing interest in recent years has been on quasi-lower-dimensional magnets[5] such as the antiferromagnetic chain compounds $CsNiCl_3$ and $RbNiCl_3$.[6] By studying the velocity shift and the attenuation of ultrasonic waves in these magnets, valuable information regarding the static as well as dynamic properties of the spins can be obtained.

There are two basic mechanisms through which acoustic waves can couple to the spins in a magnetic insulator. The first is the volume magnetostrictive interaction which is due to the modulation of the exchange integral among spins by lattice vibrations. The second is the single-ion magnetostrictive interaction which arises because the crystal-field environment surrounding the magnetic ions is perturbed by the phonons. There is experimental evidence that both mechanisms can be of comparable importance,[3,4] and so they shall be treated on an equal footing in this work. The possibility of coupling to the spin energy density, however, will not be considered here.[7]

By treating these two mechanisms on an equal footing here, we find interesting interference effects between these two types of couplings. These features are absent from earlier theories which considered only volume magnetostrictive interaction, or the results were computed separately for these mechanisms. Our expression for the ultrasonic attenuation coefficient in the weak spin-phonon coupling limit agrees with Tani and Mori;[8] however, our calculation for the corresponding frequency shift (or velocity shift) reveals an additional term that was missing from their work. Their result for the velocity shift has been used subsequently in other studies.

In Sec. II we write a spin-phonon Hamiltonian which consists of a volume and a single-ion magnetostriction interaction. The projection operator method of Mori[9] is used in Sec. III to derive expressions for the renormalized phonon frequency and its associated width. In the weak spin-phonon limit, the evaluation of these quantities in fact involves the spin system only. In Sec. IV we apply our results to planar ferromagnets at low temperatures. Detailed results for the attenuation coefficient and velocity shift are computed in Sec. V for one-dimensional (1D) spin systems with the planar ferromagnetic chain $CsNiF_3$ in mind.[10] General rules for computing static susceptibilities as well as dynamic relaxation functions involving an arbitrary number of boson or fermion operators are discussed in the Appendix within the free-particle model.

A brief account of some of the results here has been reported.[11] The present paper serves to fill in some of the details of the calculations.

II. SPIN-PHONON INTERACTIONS

We consider a localized spin system which can be described by a Hamiltonian of the form

$$\mathscr{H}_s = -\sum_{m,n} J_{m,n}\vec{S}_m\cdot\vec{S}_n + A\sum_n (S_n^z)^2 , \tag{2.1}$$

where J_{mn} and A are, respectively, the exchange integral and the single-ion anisotropy constant. Here, we will treat only the ferromagnetic case for which $J_{mn}>0$. Later, in Sec. IV we analyze in detail the case where $A>0$, so that Eq. (2.1) describes a planar ferromagnet.

Assuming that the electrons of the magnetic ions are bound rigidly to the ions as the ions vibrate about their equilibrium sites, the exchange integral between spins is then a function of the instantaneous positions of the ions. Also, if the amplitude of vibrations is small compared to the lattice spacings, we can expand the exchange integral up to the first power in the displacements $\vec{\eta}_m$ about the equilibrium sites $\vec{R}_m$ to obtain

$$-\sum_{m,n} J(\vec{r}_m-\vec{r}_n)\vec{S}_m\cdot\vec{S}_n = -\sum_{m,n}\left[J_{m,n}+\frac{\partial J}{\partial\vec{R}_m}\cdot\vec{\eta}_m+\frac{\partial J}{\partial\vec{R}_n}\cdot\vec{\eta}_n\right]\vec{S}_m\cdot\vec{S}_n . \tag{2.2}$$

Using the fact that $\partial J_{mn}/\partial\vec{R}_n = -\partial J_{mn}/\partial\vec{R}_m$, and transforming the displacements to phonon operators $b_\lambda(\vec{k})$ in the form

$$\vec{\eta}_m=(2MN)^{-1/2}\sum_{\vec{k},\lambda}\omega_\lambda^{-1/2}(\vec{k})[b_\lambda(\vec{k})+b_\lambda^\dagger(-\vec{k})]\times\hat{e}_\lambda(\vec{k})e^{-i\vec{k}\cdot\vec{R}_m}, \quad (2.3)$$

the volume magnetoelastic interaction can be written as

$$\mathscr{H}_{s\text{-ph}}^V=-\sum_{\vec{k},\lambda}\sum_{m,n}I_{mn}(\vec{k};\lambda)\omega_\lambda^{-1/2}(\vec{k})\times[b_\lambda(\vec{k})+b_\lambda^\dagger(-\vec{k})]e^{-\vec{k}\cdot\vec{R}_m}\vec{S}_m\cdot\vec{S}_n\,, \quad (2.4)$$

where

$$I_{mn}(\vec{k};\lambda)\equiv(2MN)^{-1/2}\hat{e}_\lambda(\vec{k})\cdot\frac{\partial J_{mn}}{\partial\vec{R}_m}(1-e^{i\vec{k}\cdot\vec{R}_{mn}})\,. \quad (2.5)$$

For simplicity, we take the single-ion magnetostriction Hamiltonian to have the form

$$\mathscr{H}_{s\text{-ph}}^s=V_s\sum_m\epsilon_{ij}(m)(S_m^z)^2\,. \quad (2.6)$$

The strain components $\epsilon_{ij}(m)$ can be expressed in terms of the displacements and then in phonon operators as follows:

$$\epsilon_{ij}(m)=\frac{-i}{2}\sum_{\vec{k}}[k_i\eta_j(\vec{k})+k_j\eta_i(\vec{k})]e^{-i\vec{k}\cdot\vec{R}_m}$$

$$=\frac{-i}{2(2MN)^{1/2}}\sum_{\vec{k},\lambda}\frac{[k_ie_j(\vec{k})+k_je_{\lambda i}(\vec{k})]}{\omega_\lambda^{1/2}(\vec{k})}\times[b_\lambda(\vec{k})+b_\lambda^\dagger(-\vec{k})]e^{-i\vec{k}\cdot\vec{R}_m}\,. \quad (2.7)$$

Thus we can rewrite the single-ion magnetostriction Hamiltonian as

$$\mathscr{H}_{s\text{-ph}}^s=-\sum_{\vec{k},\lambda}\sum_m K_{ij}(\vec{k})\omega_\lambda^{-1/2}(\vec{k})\times[b_\lambda(\vec{k})+b_\lambda^\dagger(-\vec{k})]e^{-i\vec{k}\cdot\vec{R}_m}(S_m^z)^2\,, \quad (2.8)$$

where

$$K_{ij}(\vec{k})\equiv\frac{iV_s}{2}\left[\frac{\hbar}{2MN}\right]^{1/2}[k_ie_{\lambda j}(\vec{k})+k_je_{\lambda i}(\vec{k})]\,.$$

By combining Eqs (2.5) and (2.8), the total spin-phonon Hamiltonian can be written as

$$\mathscr{H}_{s\text{-ph}}=-\sum_{\vec{k},\lambda}\omega_\lambda^{-1/2}(\vec{k})[b_\lambda(\vec{k})+b_\lambda^\dagger(-\vec{k})]\times\left[\sum_{m,n}I_{mn}(\vec{k};\lambda)e^{-i\vec{k}\cdot\vec{R}_m}\vec{S}_m\cdot\vec{S}_n+\sum_m K_{ij}(\vec{k})e^{-i\vec{k}\cdot\vec{R}_m}(S_m^z)^2\right]. \quad (2.9)$$

Strictly speaking, in the single-ion magnetoelastic Hamiltonian, all terms which are allowed by the symmetry of the lattice have to be included.[12] However, here, for simplicity we employ the Hamiltonian as given in Eq. (2.1), and only consider $\mathscr{H}_{s\text{-ph}}^s$ of the form of Eq. (2.8). Moreover, we have kept only the terms which are linear in the phonon operators in both the volume and single-ion magnetoelastic interactions.

III. RENORMALIZED PHONON FREQUENCY AND THE ASSOCIATED WIDTH

A. Mori's Formalism

The attenuation and the velocity shift of acoustic waves in a solid are related, respectively, to the phonon lifetime and the renormalized phonon frequency. The quantities will be derived in this section with the help of projection-operator techniques and certain methods we developed previously.[13]

In the presence of lattice vibrations, the Hamiltonian of the total system can be written as

$$\mathscr{H}=\mathscr{H}_{\text{ph}}+\mathscr{H}_s+\mathscr{H}_{s\text{-ph}}\,, \quad (3.1)$$

where $\mathscr{H}_{\text{ph}}$ is the Hamiltonian for the harmonic phonons, which takes the form

$$\mathscr{H}_{\text{ph}}=\sum_{\vec{k},\lambda}\omega_\lambda(\vec{k})b_\lambda^\dagger(\vec{k})b_\lambda(\vec{k})\,, \quad (3.2)$$

$\mathscr{H}_s$ is the Hamiltonian for the spin system which is given by Eq. (2.1), and $\mathscr{H}_{s\text{-ph}}$ denotes the spin-phonon interaction of the form written in Eq. (2.9). From Eq. (3.1) the equation of motion for the phonon is

$$\dot{b}_\lambda(\vec{k})=-i\omega_\lambda(\vec{k})b_\lambda(\vec{k})+\omega_\lambda^{-1/2}(\vec{k})f_\lambda(\vec{k})\,, \quad (3.3)$$

where $f_\lambda(\vec{k})$ is Hermitian and is defined as

$$f_\lambda(\vec{k})\equiv\sum_{m,n}I_{mn}(\vec{k};\lambda)e^{-i\vec{k}\cdot\vec{R}_m}\vec{S}_m\cdot\vec{S}_n+\sum_m K_{ij}(\vec{k})e^{-i\vec{k}\cdot\vec{R}_m}(S_m^z)^2\,. \quad (3.4)$$

Treating $f_\lambda(\vec{k})$ as a single entity, we see that the equation of motion for the phonons, Eq. (3.3), is linear in the operators $b_\lambda(\vec{k})$ and $f_\lambda(\vec{k})$. As shown in Ref. 13, this allows us to obtain an exact relation connecting the static correlation functions $(b_\lambda(\vec{k}),b_\lambda(-\vec{k}))$ and $(f_\lambda(\vec{k}),f_\lambda(k))$. Throughout this paper the inner product (A,B) is the correlation function defined as

$$(A,B)\equiv\int_0^B d\lambda\langle e^{\lambda\mathscr{H}}Ae^{-\lambda\mathscr{H}}B\rangle-\beta\langle A\rangle\langle B\rangle\,. \quad (3.5)$$

Using Eq. (3.3), we take the inner product on the right-hand side with $b_\lambda(-\vec{k})$ to obtain

$$(\dot{b}_\lambda(\vec{k}),b_\lambda(-\vec{k}))=-i\omega_\lambda(\vec{k})(b_\lambda(\vec{k}),b_\lambda(-k))+\omega_\lambda^{-1/2}(\vec{k})(f_\lambda(\vec{k}),b_\lambda(-\vec{k}))\,. \quad (3.6)$$

Employing the operator identity[14]

$$(\dot{A},B)=-i\langle[A,B]\rangle \tag{3.7}$$

together with the boson commutation relations, we obtain

$$(b_\lambda(\vec{k}),b_\lambda(-\vec{k}))=\omega_\lambda^{-1}(\vec{k})-i\omega_\lambda^{-3/2}(\vec{k})(f_\lambda(\vec{k}),b_\lambda(-\vec{k})) . \tag{3.8}$$

Similarly, using the adjoint of Eq. (3.3) and taking the inner product on the left-hand side with $f_\lambda(\vec{k})$, we find

$$(f_\lambda(\vec{k}),b_\lambda(-\vec{k}))=i\omega^{-3/2}(\vec{k})(f_\lambda(\vec{k}),f_\lambda(-\vec{k})) . \tag{3.9}$$

In obtaining Eq. (3.9), we have used the fact that $(A,\dot{B})=-(\dot{A},B)$, and

$$[f_\lambda(\vec{k}),b_{\lambda'}(\vec{k}')]=0 . \tag{3.10}$$

Substituting Eq. (3.9) in Eq. (3.8) yields the exact relation

$$(b_\lambda(\vec{k}),b_\lambda(-\vec{k}))=\omega_\lambda^{-1}(\vec{k})+\omega_\lambda^{-3}(\vec{k})(f_\lambda(\vec{k}),f_\lambda(-\vec{k})) . \tag{3.11}$$

On the right-hand side of Eq. (3.11), the first term is the phonon static correlation function in the absence of spin-phonon interactions, and the second term arises from interactions with the spin system. Since $f_\lambda(\vec{k})$ is linear in the magnetoelastic coupling constants, the second term is of second order in J' and V_s. We should also point out that this term has been omitted by Tani and Mori,[8] and therefore their results can only be correct to $\mathscr{O}(f^2)$ (i.e., to second order in the magnetoelastic coupling constants).

Next we will calculate the phonon relaxation function $\phi(\vec{k},\omega)$ defined as

$$\Phi(\vec{k},\omega)=\int_0^\infty dt\, e^{-i\omega t}(b_\lambda(\vec{k},t),b_\lambda(\vec{k},0)) . \tag{3.12}$$

From $\Phi(\vec{k},\omega)$ one can then identify the renormalized phonon frequency and the corresponding phonon lifetime. Employing the projection operator formalism with $b_\lambda(\vec{k})$ as the dynamic variable, we can write an exact equation for $\Phi(\vec{k},\omega)$:[9]

$$\{-i\omega-[(\dot{b}_\lambda(\vec{k}),b_\lambda(-\vec{k}))-\gamma(\vec{k},\omega)]/(b_\lambda(\vec{k}),b_\lambda(-\vec{k}))\}\Phi(\vec{k},\omega)=(b_\lambda(\vec{k}),b_\lambda(-\vec{k})) \tag{3.13}$$

where

$$\gamma(\vec{k},\omega)=\int_0^\infty dt\, e^{-i\omega t}(\exp[it(1-\mathscr{P})\mathscr{L}](1-\mathscr{P})\dot{b}_\lambda(\vec{k}),(1-\mathscr{P})\dot{b}_\lambda(-\vec{k})) . \tag{3.14}$$

In Eq. (3.14), $\mathscr{P}$ is the projection operator defined such that for any operator A

$$\mathscr{P}A=(b_\lambda(\vec{k}),b_\lambda(-\vec{k}))^{-1}(A,b_\lambda(-\vec{k}))b_\lambda(\vec{k}) , \tag{3.15}$$

and $\mathscr{L}$ is the full Liouville operator for the system. With the help of Eqs. (3.15), (3.9), and (3.3) we can write $(1-\mathscr{P})\dot{b}_\lambda$, which appears in Eq. (3.14), as

$$(1-\mathscr{P})\dot{b}_\lambda(\vec{k})=\frac{f_\lambda(\vec{k})}{\omega_\lambda^{1/2}(\vec{k})}+\frac{i(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))}{\omega_\lambda^2(\vec{k})(b_\lambda(\vec{k}),b_\lambda(-\vec{k}))}b_\lambda(\vec{k}) . \tag{3.16}$$

Using Eqs. (3.13) and (3.11), we can write $\Phi(\vec{k},\omega)$ in the form

$$\Phi(\vec{k},\omega)=[1+\omega_\lambda^{-2}(\vec{k})(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))]\Bigg/\left[\frac{\omega_\lambda(\vec{k})\gamma(\vec{k},\omega)}{1+\omega_\lambda^{-2}(\vec{k})(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))}+i\left[\omega-\frac{\omega_\lambda(\vec{k})}{1+\omega_\lambda^{-2}(\vec{k})(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))}\right]\right] . \tag{3.17}$$

From this equation we can identify the renormalized phonon frequency

$$\omega_{\rm ph}(\vec{k})=\frac{\omega_\lambda(\vec{k})[1-\gamma''(\vec{k},\omega)]}{1+\omega_\lambda^{-2}(\vec{k})(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))} , \tag{3.18}$$

and the corresponding width

$$\Gamma_{\rm ph}=\left.\frac{\omega_\lambda(\vec{k})\gamma'(\vec{k},\omega)}{1+\omega_\lambda^{-2}(\vec{k})(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))}\right|_{\omega=\omega_{\rm ph}} , \tag{3.19}$$

where $\gamma'(\vec{k},\omega)$ and $\gamma''(\vec{k},\omega)$ are, respectively, the real and imaginary parts of $\gamma(\vec{k},\omega)$. From $\omega_{\rm ph}$ and $\Gamma_{\rm ph}$ we can obtain, respectively, the velocity shift and the attenuation coefficient of acoustic waves.

B. Weak spin-phonon coupling limit

Although the calculations in Sec. III A are formally exact, the usefulness of the resulting expressions, i.e., Eqs. (3.18) and (3.19), is limited by the fact that they involve correlation functions, $(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))$ and $\gamma(\vec{k},\omega)$, which are to be evaluated with the full Hamiltonian, Eq. (3.17). Moreover, in $\gamma(\vec{k},\omega)$ the time development of the random force $f_\lambda(\vec{k})$ is governed by $(1-\mathscr{P})\mathscr{L}$, which makes the computation of expressions (3.18) and (3.19) extremely difficult.

However, the spin-phonon coupling is often sufficiently weak so that we only need to calculate $(f_\lambda(\vec{k}),f_\lambda(-\vec{k}))$ to the first nonvanishing order in the magnetoelastic coupling constants, i.e., $\mathscr{O}(f^2)$. In this limit our problem completely separates into a spin and a phonon part.

We see that in Eq. (3.16) the second term on the right-hand side is at least of $\mathscr{O}(f^2)$, and therefore can be dropped in calculating $\gamma(\vec{k},\omega)$ in Eq. (3.14). Thus to $\mathscr{O}(f^2)$, $\gamma(\vec{k},\omega)$ can be written

$$\gamma(\vec{k},\omega)=\omega_\lambda^{-1}(\vec{k})\int_0^\infty dt\, e^{-i\omega t}(\exp[it(1-\mathscr{P})\mathscr{L}]\times f_\lambda(\vec{k}),f_\lambda(-\vec{k}))\ . \quad (3.20)$$

Also, to $\mathscr{O}(f^2)$ the propagator $\exp[it(1-\mathscr{P})\mathscr{L}]$ can be replaced by $\exp(it\mathscr{L}_s)$, where $\mathscr{L}_s$ is the Liouville operator for the spin system only. And in the evaluation of the thermal averages, we only need to use the weight factor $\exp(-\beta\mathscr{H}_s)$. Hence $\gamma(\vec{k},\omega)$ can now be expressed in the form

$$\gamma(\vec{k},\omega)=\omega_\lambda^{-1}(\vec{k})\int_0^\infty dt\, e^{-i\omega t}(f_\lambda(\vec{k},t),f_\lambda(-\vec{k},0))^s \equiv\omega_\lambda^{-1}(\vec{k})(f_\lambda,f_\lambda)^s_{\vec{k},\omega}\ , \quad (3.21)$$

where the superscript s means that the quantity is to be evaluated with the spin Hamiltonian $\mathscr{H}_s$ only. From Eqs. (3.18) and (3.21) the renormalized phonon frequency to $\mathscr{O}(f^2)$ is now given by

$$\omega_{\rm ph}=\omega_\lambda(\vec{k})\left[1-\frac{(f_\lambda,f_\lambda)^s_{\vec{k}}}{\omega_\lambda^2(\vec{k})}+\frac{\mathrm{Im}(f_\lambda,f_\lambda)^s_{\vec{k},\omega}}{\omega_\lambda(\vec{k})}\right]\ , \quad (3.22)$$

and the corresponding width becomes

$$\Gamma_{\rm ph}=\mathrm{Re}(f_\lambda,f_\lambda)^s_{\vec{k},\omega}\,|_{\omega=\omega_\lambda(\vec{k})}\ . \quad (3.23)$$

Thus the phonons no longer appear in the problem, and all quantities now are to be evaluated with the spin Hamiltonian only. The superscript s will henceforth be dropped.

From Eq. (3.22), the relative velocity shift is given by

$$\frac{\Delta v_{\rm ph}}{v_{\rm ph}}=\frac{(f_\lambda,f_\lambda)_{\vec{k}}}{\omega_\lambda^2(\vec{k})}+\frac{\mathrm{Im}(f_\lambda,f_\lambda)_{\vec{k},\omega}}{\omega_\lambda(\vec{k})}\ , \quad (3.24)$$

where $v_{\rm ph}$ denotes the bare phonon velocity. The attenuation coefficient is obtained from Eq. (3.23) as

$$\alpha_{\rm ph}=\Gamma_{\rm ph}/v_{\rm ph}=\mathrm{Re}(f_\lambda,f_\lambda)_{k,\omega_\lambda(k)}/v_{\rm ph}\ . \quad (3.25)$$

In the work of Tani and Mori[8] the equation for the relative velocity shift does not contain the term $\omega_\lambda^{-2}(\vec{k})(f_\lambda,f_\lambda)_{\vec{k}}$. This can be traced back to the fact that they have neglected terms of $\mathscr{O}(f^2)$ in the phonon static correlation function. Their result has subsequently been used in other studies.[5]

IV. PLANAR FERROMAGNETS AT LOW TEMPERATURES

We will now only consider the phonon branch of interest and drop the index λ. From Sec. III we see that in the weak spin-phonon limit the problem has been reduced to the calculation of a static and a dynamic correlation function, namely $(f,f)_{\vec{k}}$ and $(f,f)_{\vec{k},\omega}$ of the pure spin system. Since $f_{\vec{k}}$ contains two spin variables, these are four-spin correlation functions. In general the calculations are rather difficult and quite often one must resort to some kind of decoupling scheme to break up these correlation functions. In this paper, however, we are only interested in ultrasonic properties in the low-temperature region, where the spin system can be described by an appropriate noninteracting magnon theory. In this case, these static and dynamic correlation functions can of course be computed exactly.

Here, we will focus our attention on planar ferromagnetic systems which can be described by the spin Hamiltonian as given in Eq. (2.1) with J and A both positive. The X-Y plane is therefore the easy plane. Even though our results so far are independent of the number of dimensions, eventually we will apply our results to the quasi-1D planar ferromagnets, a prototype of which is the compound $\mathrm{CsNiF_3}$.[10]

A magnon theory for planar ferromagnets with an easy X-Y plane has been developed by Villain[15] in terms of the conjugated variables $\varphi_m\equiv i\partial/\partial S_m^z$ and S_m^z, which satisfy the commutation relation

$$[\phi_m,S_m^z]=i\delta_{mn}\ , \quad (4.1)$$

and others are equal to 0. The transformation of spin operators to these variables can be accomplished using Villain's prescription

$$S_m^+=e^{i\varphi_m}[(S+\tfrac{1}{2})^2-(S_m^z+\tfrac{1}{2})^2]^{1/2}\ ,$$
$$S_m^-=[(S+\tfrac{1}{2})^2-(S_m^z+\tfrac{1}{2})^2]e^{-i\varphi_m}\ . \quad (4.2)$$

The spin Hamiltonian can be written in the harmonic approximation as

$$\mathscr{H}_s=\tfrac{1}{2}\sum_{\vec{q}}(a_{\vec{q}}S^z_{\vec{q}}S^z_{-\vec{q}}+b_{\vec{q}}\varphi_{\vec{q}}\varphi_{\vec{q}})\ , \quad (4.3)$$

where

$$\varphi_{\vec{q}}\equiv N^{-1/2}\sum_m\varphi_m e^{i\vec{q}\cdot\vec{R}_m}\ ,$$
$$S^z_{\vec{q}}\equiv N^{-1/2}\sum_m S^z_m e^{i\vec{q}\cdot\vec{R}_m}\ , \quad (4.4)$$

$$a_{\vec{q}}\equiv 2(A+J_0-J_{\vec{q}})\ ,$$
$$b_{\vec{q}}\equiv 2S^2(J_0-J_{\vec{q}})\ , \quad (4.5)$$

and

$$J_{\vec{q}}\equiv\sum_n J_{mn}e^{i\vec{q}\cdot\vec{R}_{mn}}\ . \quad (4.6)$$

Similarly, Eq. (3.4) can be converted into the form

$$f(\vec{k})=\sum_{\vec{q}}[L(\vec{k},\vec{q})+K_{ij}(\vec{k})]S^z_{\vec{q}}S^z_{\vec{k}-\vec{q}}+S^2L(\vec{k},\vec{q})\ , \quad (4.7a)$$

where

$$L(\vec{k},\vec{q})=I(\vec{k},\vec{q})-I(\vec{k},0) \quad (4.7b)$$

and

$$I(\vec{k},\vec{q}) \equiv \sum_m I_{mn}(\vec{k}) e^{-i\vec{q}\cdot\vec{R}_{mn}} . \tag{4.7c}$$

Next, using the magnon operators,

$$c_{\vec{q}} = \frac{1}{\sqrt{2}} \left[\frac{b_{\vec{q}}}{a_{\vec{q}}} \right]^{1/4} \varphi_{\vec{q}} + \frac{i}{\sqrt{2}} \left[\frac{a_{\vec{q}}}{b_{\vec{q}}} \right]^{1/4} S^z_{\vec{q}} , \tag{4.8}$$

which satisfy the boson commutation relations

$$[c_{\vec{q}}, c^\dagger_{\vec{q}'}] = \delta(\vec{q}-\vec{q}') , \tag{4.9}$$

and others equal to 0, $\mathscr{H}_s$ becomes

$$\mathscr{H}_s = \sum_{\vec{q}} \epsilon_{\vec{q}} (c^\dagger_{\vec{q}} c_{\vec{q}} + \tfrac{1}{2}) , \tag{4.10}$$

where $\epsilon_{\vec{q}}$ is the magnon dispersion relation given by

$$\epsilon_{\vec{q}} = (a_{\vec{q}} b_{\vec{q}})^{1/2} . \tag{4.11}$$

By using Eq. (4.8), $f(\vec{k})$ can be expressed in terms of the magnon operators. Then, in order to obtain $(f,f)_{\vec{k}}$ and $(f,f)_{\vec{k},\omega}$, we need to evaluate some generalized static susceptibilities and relaxation functions involving four magnon operators. General rules for calculating such quantities can be found in the Appendix.

By combining Eqs. (4.7) and (4.8) with Eqs. (A4) and (A5), we find after some straightforward calculations the following results:

$$(f,f)_{\vec{k},\omega} = \frac{-i\hbar}{4MN} \sum_{\vec{q}} \left[\frac{b_{\vec{q}} b_{\vec{k}-\vec{q}}}{a_{\vec{q}} a_{\vec{k}-\vec{q}}} \right]^{1/2} \left[\left[\frac{n_{\vec{k}-\vec{q}} + n_{\vec{q}} + 1}{\epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} \right] \left[\frac{1}{\omega - i\delta + \epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} + \frac{1}{\omega - i\delta - \epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} \right] P_+ \right.$$
$$\left. + \left[\frac{n_{\vec{k}-\vec{q}} - n_{\vec{q}}}{\epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} \right] \left[\frac{1}{-\omega - i\delta + \epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} + \frac{1}{\omega + i\delta - \epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} \right] P_- \right] , \tag{4.12}$$

$$(f,f)_{\vec{k}} = \frac{\hbar}{4MN} \sum_{\vec{q}} \left[\frac{b_{\vec{q}} b_{\vec{k}-\vec{q}}}{a_{\vec{q}} a_{\vec{k}-\vec{q}}} \right]^{1/2} \left[\left[\frac{n_{\vec{k}-\vec{q}} + n_{\vec{q}} + 1}{\epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} \right] P_+ + \left[\frac{n_{\vec{k}-\vec{q}} - n_{\vec{q}}}{\epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} \right] P_- \right] , \tag{4.13}$$

where

$$P_\pm = P_\pm(\vec{k},\vec{q}) \equiv \frac{2MN}{\hbar} \left| S^2 \left[\frac{a_{\vec{q}} a_{\vec{k}-\vec{q}}}{b_{\vec{q}} b_{\vec{k}-\vec{q}}} \right]^{1/2} L(\vec{k},\vec{q}) \pm [L(\vec{k},\vec{q}) + K_{ij}(\vec{k})] \right|^2 . \tag{4.14}$$

In deriving Eqs. (4.12) and (4.13), various terms have been grouped together so that from the resulting expressions it can be easily seen that $\mathrm{Re}(f,f)_{\vec{k},\omega}$ and $(f,f)_{\vec{k}}$ are both positive definite quantities, as they should be. From Eq. (3.25) the attenuation coefficient can be written as

$$\alpha_{\mathrm{ph}} = \frac{\hbar}{4MN} \sum_{\vec{q}} \left[\frac{b_{\vec{q}} b_{\vec{k}-\vec{q}}}{a_{\vec{q}} a_{\vec{k}-\vec{q}}} \right]^{1/2} \left[\left[\frac{n_{\vec{k}-\vec{q}} + n_{\vec{q}} + 1}{\epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} \right] [\delta(\omega_{\vec{k}} + \epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}) + \delta(\omega_{\vec{k}} - \epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}})] P_+ \right.$$
$$\left. + \left[\frac{n_{\vec{k}-\vec{q}} - n_{\vec{q}}}{\epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} \right] [\delta(\omega_{\vec{k}} + \epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}) + \delta(\omega_{\vec{k}} - \epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}})] P_- \right] , \tag{4.15}$$

and from Eq. (3.24) the relative velocity shift takes the form

$$\frac{\Delta v}{v_{\mathrm{ph}}} = \frac{\hbar}{4MN\omega^2_{\vec{k}}} \sum_{\vec{q}} \left[\frac{b_{\vec{q}} b_{\vec{k}-\vec{q}}}{a_{\vec{q}} a_{\vec{k}-\vec{q}}} \right]^{1/2} \left\{ \left[\frac{n_{\vec{k}-\vec{q}} + n_{\vec{q}} + 1}{\epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} \right] \left[\left[1 - \frac{\omega_{\vec{k}}}{\omega_{\vec{k}} + \epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} \right] + \left[1 - \frac{\omega_{\vec{k}}}{\omega_{\vec{k}} - \epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} \right] \right] P_+ \right.$$
$$\left. + \left[\frac{n_{\vec{k}-\vec{q}} - n_{\vec{q}}}{\epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} \right] \left[\left[1 - \frac{\omega_{\vec{k}}}{\omega_{\vec{k}} + \epsilon_{\vec{q}} - \epsilon_{\vec{k}-\vec{q}}} \right] + \left[1 - \frac{\omega_{\vec{k}}}{\omega_{\vec{k}} - \epsilon_{\vec{q}} + \epsilon_{\vec{k}-\vec{q}}} \right] \right] P_- \right\} . \tag{4.16}$$

There are three points that deserve comment at this stage. First, as we have pointed out in Sec. III, the contribution to the velocity shift in previous theories comes only from the imaginary part of $(f,f)_{\vec{k},\omega}$. The contribution from the term $(f,f)_{\vec{k}}$ was omitted. Consequently according to their calculation Eq. (4.16) would appear with the term $1-\omega_{\vec{k}}/(\omega_{\vec{k}}+\epsilon_{\vec{q}}+\epsilon_{\vec{k}-\vec{q}})$ replaced by $-\omega_{\vec{k}}/(\omega_{\vec{k}}+\epsilon_{\vec{q}}+\epsilon_{\vec{k}+\vec{q}})$ and similarly for the other terms. It is difficult to determine *a priori* the size of the omitted terms. Later we will evaluate this expression explicitly for the planar ferromagnetic chain and we find that, at least in this case, the omitted terms can in fact be larger than the ones considered by these earlier studies.

The second point is that in most previous studies either the volume magnetoelastic coupling or the single-ion magnetostriction was considered, but seldom both. The reason might be that for a given solid one of these mechanisms often dominates over the other. Bennett and Pytte,[16] and later Ghatak,[2] studied both types of interactions for ferromagnets. Unfortunately, calculations were carried out separately for the two mechanisms; consequently, cross interaction terms of the form as given by Eq. (4.14) were absent in their work. However, from the experimental works of Lüthi and co-workers,[3,4] it is known that these mechanisms can sometimes be comparable in strength. When this is the case, interferences between the two types of spin-phonon interactions can be significant.

Third, in earlier studies of ultrasonic attenuation in magnets, scattering processes are computed separately, and authors often just select processes which correspond to phonon annihilation. These scattering events of course have positive contributions to the attenuation coefficient. However, there are also processes in which phonons are created with the destruction of two magnons. These scattering terms thus have negative contributions to the attenuation and must be included. Here, we include all scattering processes consistently to $\mathscr{O}(f^2)$.

V. APPLICATION TO PLANAR FERROMAGNETIC CHAINS

Hereafter, we will concentrate on the one-dimensional (1D) planar ferromagnets.[10] The magnon energy is

$$\epsilon_q=2S\left[4J\sin^2\frac{qa}{2}\left[A+4J\sin^2\frac{qa}{2}\right]\right]^{1/2}, \tag{5.1}$$

which in the long-wavelength limit becomes $\hbar v_s q$, where the magnon velocity v_s is given by $2Sa(JA)^{1/2}\hbar$. The energy ϵ_{k-q} assumes the form

$$\epsilon_{k-q}\equiv 2S\left\{4J\sin^2\left[(kc-q)\frac{a}{2}\right]\times\left[A+4J\sin^2\left[(kc-q)\frac{a}{2}\right]\right]\right\}^{1/2}, \tag{5.2}$$

where c is the cosine of the angle between the phonon wave vector $\vec{k}$ and the chain. Owing to the 1D nature of the magnetic part of the system, q is the magnon wave vector along the chain direction, and $\vec{k}$ appears only in the combination kc. As will be seen later, this leads to some unique behavior in the ultrasonic properties at a certain angle between the chain and the direction of propagation of the acoustic waves. From Eqs. (4.7b) and (2.5), $L(\vec{k},q)$ is found to be given by

$$L(\vec{k},q)=-8i\left[\frac{\hbar}{2MN}\right]^{1/2}\hat{e}\cdot\hat{c}\times J'\sin\frac{kca}{2}\sin\frac{qa}{2}\sin\left[(kc-q)\frac{a}{2}\right], \tag{5.3}$$

where $\hat{c}$ is a unit vector in the chain direction and $J'\equiv\partial J(r)/\partial r$.

Since we are considering ultrasonic waves of frequency in the (1–100)-MHz range, therefore in general, we have $ka\approx 10^{-5}-10^{-6}$. Owing to the presence of the Bose factors in Eqs. (4.15) and (4.16), the dominant contributions to α_{ph} and Δv_{ph} come from scattering with long-wavelength magnons. Thus the long-wavelength approximation has been frequently used in calculations. Here, at first, we will also make this approximation; however, a more careful analysis will follow next. In this limit we have $n_q\cong k_BT/\epsilon_q$ and $(n_{k-q}-n_q)/(\epsilon_q-\epsilon_{k-q})\cong k_BT/(\epsilon_q\epsilon_{k-q})$, thus the expressions for α_{ph} and $\Delta v_{\text{ph}}/v_{\text{ph}}$ can be simplified as follows:

$$\alpha_{\text{ph}}=\frac{ak_BT}{64A^2v_{\text{ph}}v_sM}\left[P_+\int dq\,\delta(\omega_{\vec{k}}-\epsilon_q-\epsilon_{k-q})+P_-\int dq[\delta(\omega_{\vec{k}}+\epsilon_q-\epsilon_{k-q})+\delta(\omega_{\vec{k}}-\epsilon_q+\epsilon_{k-q})]\right], \tag{5.4}$$

$$\frac{\Delta v_{\text{ph}}}{v_{\text{ph}}}=\frac{k_BT}{16A^2M\omega_{\vec{k}}^2}[P_+(I_1+I_2)+P_-(I_3+I_4)], \tag{5.5}$$

where

$$I_{1,2}\equiv 1-\frac{\omega_{\vec{k}}a}{2\pi}\int_{-\pi/a}^{\pi/a}dq(\omega_{\vec{k}}\pm\epsilon_q\pm\epsilon_{k-q})^{-1},$$
$$I_{3,4}\equiv 1-\frac{\omega_{\vec{k}}a}{2\pi}\int_{-\pi/a}^{\pi/a}dq(\omega_{\vec{k}}\pm\epsilon_q\mp\epsilon_{k-q})^{-1}, \tag{5.6}$$

and

$$P_\pm=\left[\frac{4SAa^2}{\hbar v_s}\hat{e}\cdot\hat{c}J'kca\mp\frac{V_s}{2}(k_ie_j+k_je_i)\right]^2. \tag{5.7}$$

The δ function $\delta(\omega_{\vec{k}}+\epsilon_q+\epsilon_{k-q})$ has been omitted in Eq. (4.15), since the argument of this δ function can never be satisfied for positive frequencies. Thus from Eqs. (5.4) and (5.5) we see that α_{ph} and Δv_{ph} both have a linear temperature dependence. Moreover, since $P_\pm\sim k^2$, α_{ph} is quadratic in the phonon frequency. The frequency dependence of Δv_{ph} will be determined later.

Next we will make careful study of the ultrasonic attenuation coefficient. For this purpose we will take a closer look at the δ functions than previous theories have done. For each δ function we need to find all q values which will make the argument of the δ function vanish.

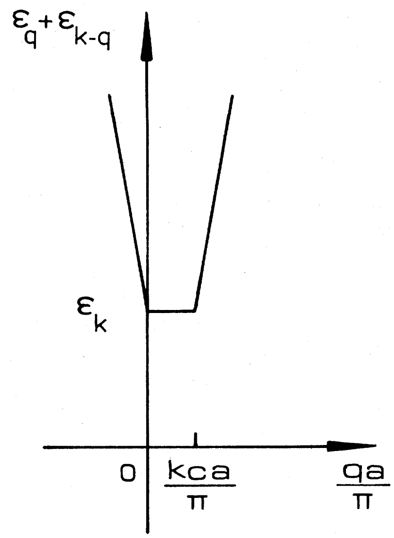

FIG. 1. $\epsilon_q+\epsilon_{k-q}$ as a function of q when qa is small. Without loss of generality, we have taken kc to be positive.

Actually, this can be accomplished by solving a quartic equation for each δ function. However, to do so would require a tremendous amount of algebra, and physical insight could be lost in the heavy computation. Our analysis below is simple and yet very accurate and quite physically revealing.

For the δ function $\delta(\omega_k-\epsilon_q-\epsilon_{k-q})$ we want to know if there are solutions to the equation

$$\omega_{\vec{k}}-\epsilon_q-\epsilon_{k-q}=0 \; . \tag{5.8}$$

Now, since $ka \approx 10^{-5}$–10^{-6}, we can write $\epsilon_q+\epsilon_{k-q}\cong 2\epsilon_q$ with extremely good accuracy except when q is comparable to or less than kc. Moreover, $v_{\rm ph}ka/v_s$ is practically always $\ll 1$. Thus the solutions to Eq. (5.8), if they exist, must always lie in the small-qa region. From Fig. 1, where the quantity $\epsilon_q+\epsilon_{k-q}$ is plotted as a function of q in the small-qa region, it is easy to see that this δ function has no contribution to $\alpha_{\rm ph}$ if $v_c c > v_{\rm ph}$. When $v_{\rm ph} > v_s c$ there are always two solutions. The one which is larger than kc is given by $q=\frac{1}{2}k(c+v_{\rm ph}/v_s)$, and the one which is negative is given by $q=\frac{1}{2}k(c-v_{\rm ph}/v_s)$.

Next we consider the two remaining δ functions $\delta(\omega_{\vec{k}}\mp\epsilon_q\pm\epsilon_{k-q})$, and look for solutions to the equations

$$\omega_{\vec{k}}\mp(\epsilon_q-\epsilon_{k-q})=0 \; . \tag{5.9}$$

The behavior of $\epsilon_q-\epsilon_{k-q}$ as a function of qa is depicted in Fig. 2. For $qa \gg kca$ we can write

$$\epsilon_q-\epsilon_{k-q}\cong k\left[\frac{\partial\epsilon_k}{\partial k}\right]_{k=0} = \frac{4Jkca\,\sin qa}{\{1-[A/(A+8J\sin^2 qa/2)]^2\}^{1/2}} \tag{5.10}$$

with extremely good accuracy. Equation (5.10) in general

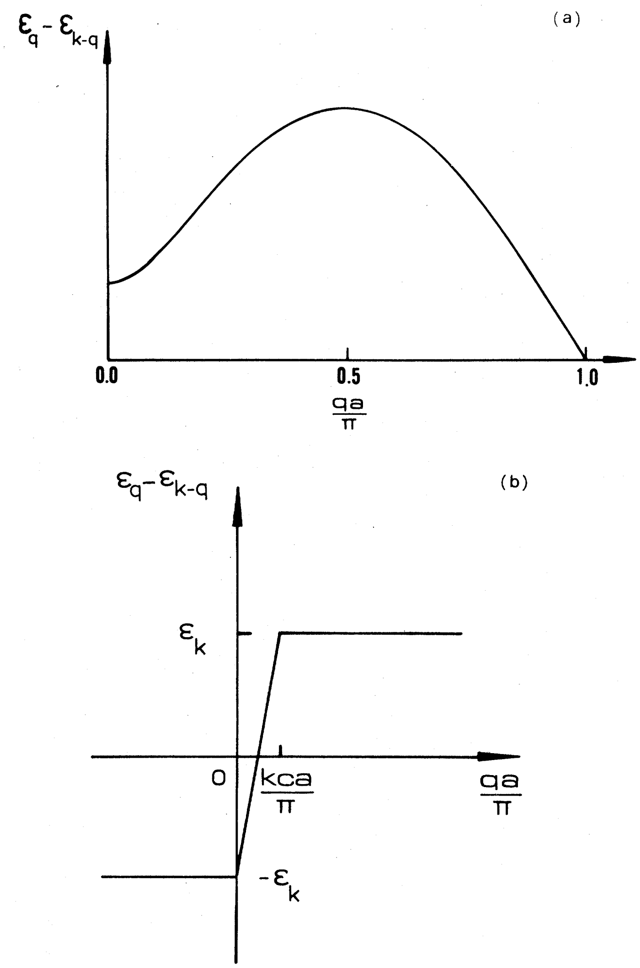

FIG. 2. (a) $\epsilon_q-\epsilon_{k-q}$ as a function of q. This quantity is extremely close to being a symmetric function of q except for q comparable to or less than kc. See (b) for the behavior in this region. (b) $\epsilon_q-\epsilon_{k-q}$ as a function of q in the region where q is comparable to or less than kc.

attains its maximum at some $q_{\max}$ where $q_{\max}a$ is not small. However, for

$$v_{\rm ph} > \left.\frac{4Ja\,\sin qa}{\{1-[A/(A+8J\sin^2 qa/2)]^2\}^{1/2}}\right|_{q=q_{\max}} \tag{5.11}$$

these δ functions do not contribute to $\alpha_{\rm ph}$. Otherwise, there are always two solutions to Eq. (5.9), one on each side of $q_{\max}$. Since at low temperature only long-wavelength magnons are significantly excited, the solutions closest to the zone center will therefore contribute most to the attenuation and velocity shift. Moreover, when $v_{\rm ph} < v_s c$ the solutions near the zone center in fact lie in the small-qa region and are given by $\frac{1}{2}k(c\pm v_{\rm ph}/v_s)$.

Scattering processes corresponding to the δ functions $\delta(\omega_{\vec{k}}\mp\epsilon_q\pm\epsilon_{k-q})$ are those in which a phonon and a magnon are annihilated and a magnon is created (or the reverse process). The physical reasons why a criterion exists (i.e., $v_{\rm ph} < v_{\max}c$) which must be met before these processes can occur is quite simple if we note that in contrast to the approach using the approximate magnon dispersion $\epsilon_q = v_s \mid q \mid$, there is an upper bound to the amount of energy a magnon can have. Thus if the phonon has too high a velocity, then these processes simply cannot take place because of energy conservation.

The curves in Figs. 1 and 2 have discontinuous slopes, and these can be traced back to the discontinuity in the slope of the magnon dispersion ϵ_q at $q=0$. It is easily seen that these curves join smoothly at $q=0$ and at kc. So if we neglect the small contributions to $\alpha_{\rm ph}$ from the large-qa solutions, Eq. (4.4) can be reduced to the simpler form

$$\alpha_{\rm ph} = \frac{ak_B T}{32A^2 v_{\rm ph} v_s M}[\Theta(v_{\rm ph} - v_s c)P_+ + \Theta(v_s c - v_{\rm ph})P_-] , \tag{5.12}$$

where $\Theta(x)$ is the unit step function.

The attenuation coefficient as given in Eq. (5.12) takes on different values depending on the relative magnitude between $v_{\rm ph}$ and $v_s c$. This difference is small if either the single-ion or the volume magnetostrictive interaction dominates, but can be significant if both coupling mechanisms have comparable strengths. For $v_{\rm ph} > V_s c$ the scattering processes that contribute to $\alpha_{\rm ph}$ are those where a phonon is annihilated while two magnons are simultaneously created (and the reverse processes). For $v_{\rm ph} < v_s c$ the relevant processes are those where a magnon absorbs a phonon and turns into another magnon (and the reverse processes). If it happens that $v_{\rm ph} = v_s c$, then the conditions specified by the three δ functions, i.e., Eqs. (5.8) and (5.9), are satisfied identically for all q in the small-a region, and so all long-wavelength magnons contribute to $\alpha_{\rm ph}$. The interaction between the phonons and magnons is so strong that our assumption of a weak spin-phonon coupling will no longer be true.

Next, we consider the velocity shift which can be obtained from Eq. (5.5) by performing the integrals of Eq. (5.6). By using the long-wavelength form for the magnon dispersion relation, all these integrals can be evaluated analytically. Dropping terms which are small because $ka < 10^{-5}$, we find $I_1 + I_2 \cong 2$ and $I_3 + I_4 \cong 2[1-(v_{\rm ph}/v_s c)^2]^{-1}$. Thus the velocity shift can be written as

$$\frac{\Delta v_{\rm ph}}{v_{\rm ph}} = \frac{k_B T}{8A^2 M\omega_{\vec{k}}^2}\left[P_+ + \frac{P_-}{1-(v_{\rm ph}/v_s c)^2}\right] . \tag{5.13}$$

The relative velocity shift therefore has a linear dependence on T and because $P_\pm \propto k^2$ it is independent of the sound frequency. Of course, the behavior of $\Delta v_{\rm ph}/v_{\rm ph}$, when $v_{\rm ph}$ happens to be extremely close to $v_s c$ is not expected to be given accurately by Eq. (5.13). Nevertheless, the incipient anomaly for $v_{\rm ph} \approx v_s c$ should certainly exist. For $v_s c > v_{\rm ph}$ we can anticipate a slower ultrasound velocity, and for $v_s c$ less than and sufficiently close to $v_{\rm ph}$ the P_- term in Eq. (5.13) can become sufficiently negative so as to cause the phonons to *speed* up. Note that the processes responsible for this P_- term are those where a magnon absorbs a phonon and changes into another magnon (and the reverse process). When $v_s c \approx v_{\rm ph}$ such scattering processes become extremely probable even though the spin-phonon coupling may be rather weak.

VI. CONCLUSIONS AND DISCUSSIONS

We have certainly chosen a rather simple form for the single-ion magnetostrictive interaction.[12] However, extensions of our calculations here to include more appropriate form for $\mathscr{H}^s_{s\text{-ph}}$ are quite straightforward. Basically, the same type of four-spin correlation function must be evaluated and the form for K_{ij} must then be remodified.

Detailed results for the ultrasonic attenuation coefficient and the velocity shift have been derived here for a planar ferromagnetic chain. An excellent candidate here is of course $CsNiF_3$, whose magnetic[10] as well as vibrational properties[17,18] have been very well studied. We hope that our work here will stimulate some experimental studies into the ultrasonic behavior of this system.[22]

For $CsNiF_3$, Dorner and Steiner[17] found that v_{11}, v_{33}, and v_{44}, are all larger than $v_{\rm ph}$ at $T=85$ K, and these phonon velocities depend very little on the temperature. Thus only the P_+ term in Eq. (5.12) contributes to $\alpha_{\rm ph}$ in this case, as we have pointed out before.[11] One can also do the calculation in the presence of a symmetry-breaking magnetic field. Results for $\alpha_{\rm ph}$ were presented earlier[11] and will not be discussed here any further.

ACKNOWLEDGMENTS

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APPENDIX: GENERAL RULES FOR CALCULATING FREE-PARTICLE RELAXATION FUNCTIONS AND GENERALIZED STATIC SUSCEPTIBILITIES

In calculating the dynamic responses of physical systems one is often faced with the problem of evaluating free-particle relaxation functions and generalized static susceptibilities involving a number of second quantized operators. General rules for writing such quantities will be outlined here for bosons as well as for fermions.[19,20]

Consider the free-particle relaxation function which, by definition, can be written

$$\begin{aligned}((a_1 a_2 \cdots)^\dagger, a_\alpha a_\beta \cdots)_z &\equiv \int_0^\infty dt\, e^{izt}([a_1(t)a_2(t)\cdots]^\dagger, a_\alpha(0)a_\beta(0)\cdots)\\ &= \int_0^\infty dt\, e^{izt}\int_0^\beta d\lambda \langle [a_1(t-i\lambda)a_2(t-i\lambda)\cdots]^\dagger a_\alpha a_\beta \cdots\rangle - \frac{i\beta}{z}\langle a_1 a_2\cdots\rangle^* \langle a_\alpha a_\beta \cdots\rangle ,\end{aligned} \tag{A1}$$

where the a's can be creation or annihilation operators. Moreover, they can all be bosons or fermions. Their time dependences are very simple, namely

$$a_k(t)=e^{-i\tilde{\epsilon}_k t}a_k \; , \tag{A2}$$

where ϵ_k is the single-particle energy and

$$\tilde{\epsilon}_k \equiv \begin{cases} -\epsilon_k, & \text{if } a_k \text{ is a creation operator} \\ +\epsilon_k, & \text{if } a_k \text{ is an annihilation operator} \; . \end{cases} \tag{A3}$$

The integrals in Eq. (A1) can then be evaluated straightforwardly to give

$$((a_1a_2\cdots)^\dagger, a_\alpha a_\beta \cdots)_z = i\left[\frac{1}{\hbar}\sum_{i=1}\tilde{\epsilon}_i-\omega+i\delta\right]^{-1} \times ((a_1a_2\cdots)^\dagger, a_\alpha a_\beta\cdots) \; , \tag{A4}$$

where the generalized static susceptibility $((a_1a_2\cdots)^\dagger, a_\alpha a_\beta\cdots)$ is given by

$$((a_1a_2\cdots)^\dagger, a_\alpha a_\beta\cdots) = \frac{e^{\beta\sum_i\tilde{\epsilon}_i}-1}{\sum_i\tilde{\epsilon}_i} \times \langle (a_1a_2\cdots)^\dagger a_\alpha a_\beta\cdots\rangle - \beta\langle a_1a_2\cdots\rangle^*\langle a_\alpha a_\beta\cdots\rangle \tag{A5}$$

In Eq. (A5) the thermal averages of a number of creation and annihilation operators can easily be written for bosons or fermions by using the Wick's theorem of Gaudin.[21]

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