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Ultrasonic Attenuation in the Heisenberg Paramagnet*

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Two models which describe the interaction between the spin system and the lattice are presented for examining the propagation of sound waves in ferromagnetic insulators. In particular, expressions for the ultrasonic attenuation coefficient at high temperatures and near the Curie point are obtained in terms of time-dependent correlation functions. The attenuation coefficient is found to be proportional to the square of the phonon frequency and to increase rapidly in the vicinity of the Curie point.

I. INTRODUCTION

WE shall study in this paper the ultrasonic attenuation of sound waves propagating in a Heisenberg paramagnet for temperatures near and above the transition point (a ferromagnetic insulator in the paramagnetic state). The thermal fluctuations of the spins become large near the transition point (critical fluctuations)[1] and may produce an appreciable scattering of the phonons whenever a significant coupling between the spins and the phonons exists. The ultrasonic attenuation due to the spin-phonon interaction is a mechanism with which we may probe systems exhibiting second-order phase transitions. We shall show that the part of the four-spin correlation function which gives the large distance behavior of the system contains a description of the ultrasonic attenuation in magnetic systems. However, this part of the four-spin correlation function is the most difficult part to compute because it refers to a situation in which the behavior of the system is dominated by many collisions. In the language of perturbation theory, the theory of ultrasonic attenuation in the Heisenberg paramagnet requires extensive resummation of diagrams or equivalently integral equations.

Other authors have examined the ultrasonic attenuation near the magnetic critical point.[2,3] Their theories differ most profoundly in the treatment of the four-spin correlations. Tani and Mori[2] eliminate the time integral of this correlation function with the aid of the continued fraction representation.[4] They then consider only the temperature dependence of those correlations which are diagonal in the phonon wave vector and use the high-temperature values of the nondiagonal correlations for all temperatures. They conclude that the ultrasonic attenuation is proportional to $\omega^2/(T-T_c)$ for temperatures near the critical point T_c and for a spin-phonon interaction which is linear in the phonon-field operator. The quantity ω is the angular frequency of the sound wave; that is to say, $\omega=c|\mathbf{q}|$, where c is the speed of sound, $\mathbf{q}$ is the phonon wave vector ($qd\ll1$), and d is the distance between lattice sites. Papoular considers, for the ferromagnetic case, an interaction Hamiltonian which is quadratic in both the spin and the phonon variables.[3] He examines only the equal time correlations, expresses the four spin correlations as a product of equal time pair correlations, and computes the contribution due to the quasielastic scattering of phonons by the spin system to the ultrasonic attenuation.[3] His results predict that the ultrasonic attenuation is proportional to $\omega^3/(T-T_c)^{1/2}$ for a spin-phonon interaction which is quadratic in the phonon-field operators. The few experimental measurements[5,6] are not sufficient to resolve the above theoretical discrepancies. An analysis of the data of Neighbours *et al.*[5] reveals that the ultrasonic attenuation in the antiferromagnet $\mathrm{MnF_2}$ is approximately proportional to ω near the transition point. However, even in the paramagnetic state the antiferromagnet differs substantially from the ferromagnet near the transition point and consequently their results give us little additional understanding of the ferromagnetic problem. The preliminary experiments of Lüthi[6] on the ferromagnetic metal Gd show that the ultrasonic attenuation is proportional to ω^2 near the transition point. This result is encouraging even though care must be exercised in applying such results to ferromagnetic insulators.

Bearing in mind that a better theory for the Heisenberg spin system will hopefully be developed, we shall formulate the problem in terms of the exact four-spin correlation functions. We shall assume that the spin-phonon interaction is sufficiently weak and thereby shall perform the calculation by computing the "polari-

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[1] H. S. Bennett, Ann. Phys. (N.Y.) **39**, 127 (1966).

[2] K. Tani and H. Mori, Phys. Letters **19**, 627 (1966).

[3] M. M. Papoular, Compt. Rend. **258**, 4446 (1964).

[4] H. Mori, Progr. Theoret. Phys. (Kyoto) **34**, 399 (1965).

[5] J. R. Neighbours *et al.*, Phys. Rev. Letters **11**, 125 (1963).

[6] B. Lüthi (private communication).

zation operator" for the phonon propagator to lowest order in the spin-phonon interaction.[7]

We shall employ two simple models to characterize the interactions between the spin system and the lattice. One model describes the volume magnetostriction which obtains from an expansion of the exchange integral in powers of the ionic displacements, and the other model describes the magnetostriction associated with a single ion.

As other authors have done, we shall introduce an approximation for the four-spin correlation function. We shall factorize the four-spin correlation function into terms containing products of time-dependent and of time-independent pair correlation functions. Reference 8 contains a discussion of the spin-diffusion coefficient based upon the same approximation for the four-spin correlation function. We find that those terms which contain products of time-independent (equal time) pair correlations do not contribute to the acoustic attenuation or to the spin diffusion. Because the differences among the various theoretical calculations of these quantities arise mostly from the approximate solution of the spin problem, we include in Appendix B a discussion of the physics contained in the factorization procedure. We shall conclude that the ultrasonic attenuation in the ferromagnetic insulator in the paramagnetic state is proportional to $\omega^2 F(\chi,D)$, where F is a function of the static susceptibility $\chi(\beta)$ and the spin diffusion coefficient $D(\beta)$. The parameter β is the inverse temperature measured in energy units, i.e., $\beta=(1/kT)$, where k is Boltzmann's constant.

Until a more rigorous treatment of the three-dimensional Heisenberg model is developed for temperatures near the transition point, the present calculations must be considered preliminary and the present results must be viewed with caution for those regions where the approximations are questionable.

II. FORMULATION

A. Hamiltonians

The total Hamiltonian,

$$H=H_{\text{spin}}+H_{\text{phonon}}+H_{\text{int}}{}^{V}+H_{\text{int}}{}^{S}, \tag{1}$$

contains the physical description of the ferromagnetic insulator. For the spin part of the crystal, we shall adopt the Heisenberg model of magnetism which attributes a localized spin to each site of the crystal lattice,

$$H_{\text{spin}}=-\tfrac{1}{2}\sum_{\alpha,\alpha'} J(\boldsymbol{\alpha}-\boldsymbol{\alpha}')\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}',t). \tag{2}$$

The spin operator in the Heisenberg representation for the localized spin at site $\boldsymbol{\alpha}$ and at time t is represented by $\mathbf{S}(\boldsymbol{\alpha},t)$. The exchange interaction $J(\boldsymbol{\alpha}-\boldsymbol{\alpha}')$ describes the interaction between the spin at site $\boldsymbol{\alpha}$ and the spin at site $\boldsymbol{\alpha}'$. We shall use the convention that $J(0)=0$ so that we may extend the double summation over all lattice sites, including the term $\boldsymbol{\alpha}=\boldsymbol{\alpha}'$. We shall treat in this paper only the ferromagnetic case for which $J(\boldsymbol{\alpha}-\boldsymbol{\alpha}')>0$.

The spin operators obey the equal-time commutation relations

$$[S_x(\boldsymbol{\alpha},t),S_y(\boldsymbol{\alpha}',t)]=i\delta_{\alpha,\alpha'}S_z(\boldsymbol{\alpha},t), \text{ etc.}, \tag{3}$$

and

$$[S_x(\boldsymbol{\alpha},t),S_x(\boldsymbol{\alpha}',t)]=0, \text{ etc.}, \tag{4}$$

and satisfy the subsidiary condition,

$$\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha},t)=S(S+1), \tag{5}$$

where S is the spin quantum number.

For the phonon part of the crystal, we shall use the harmonic approximation to treat the ionic interaction,

$$H_{\text{phonon}}=\sum_{\alpha}\frac{P^2(\boldsymbol{\alpha},t)}{2M}+\tfrac{1}{2}\sum_{\alpha,\alpha'}U_i(\boldsymbol{\alpha},t)C_{ij}(\boldsymbol{\alpha},\boldsymbol{\alpha}')U_j(\boldsymbol{\alpha}',t). \tag{6}$$

The momentum $P_i(\boldsymbol{\alpha},t)=M\partial U_i(\boldsymbol{\alpha},t)/\partial t$ is conjugate to the ionic displacement $U_i(\boldsymbol{\alpha},t)$, and M is the mass of the ion. We obtain the tensor C_{ij} from the potential energy of the ions V, namely,

$$C_{ij}(\boldsymbol{\alpha},\boldsymbol{\alpha}')=\frac{\partial}{\partial U_i(\boldsymbol{\alpha},t)}\frac{\partial}{\partial U_j(\boldsymbol{\alpha}',t)}V\bigg|_{(U)=0}, \tag{7}$$

where i and j refer to the Cartesian vector components; $i, j=x, y,$ or z. The dynamical variables for the phonon Hamiltonian satisfy the commutation relations,

$$[U_i(\boldsymbol{\alpha},t),P_j(\boldsymbol{\alpha}',t)]=i\hbar\delta_{i,j}\delta_{\alpha,\alpha'}. \tag{8}$$

We express the interaction between the spin and the phonons as the sum of two terms,

$$H_{\text{int}}=H_{\text{int}}{}^{V}+H_{\text{int}}{}^{S}. \tag{9}$$

The first term $H_{\text{int}}{}^{V}$ describes the volume magnetostriction, while the second term $H_{\text{int}}{}^{S}$ describes the magnetostriction associated with a single ion. We assume that the electrons are tightly bound to their ion cores and move rigidly with the ions as the ions oscillate about their equilibrium positions $\boldsymbol{\alpha}$. Thus, the exchange integral depends upon the instantaneous positions of the ions. We also assume that the ionic displacements are small compared to the lattice spacing. We then obtain the volume magnetostriction Hamiltonian by expanding the exchange integral about the equilibrium sites $\boldsymbol{\alpha}$ in powers of the displacements $U_i(\boldsymbol{\alpha},t)$,

$$H_{\text{int}}{}^{V}=-\tfrac{1}{2}\sum_{\alpha,\alpha'}\{U_i(\boldsymbol{\alpha},t)-U_i(\boldsymbol{\alpha}',t)\}(\nabla_{\alpha})_i J(\boldsymbol{\alpha}-\boldsymbol{\alpha}')\times\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}',t). \tag{10}$$

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[7] E. Pytte, Ann. Phys. (N. Y.) 32, 377 (1965). In this paper the "polarization operator" is computed rigorously in the low-temperature region where the Holstein-Primakoff approximation is valid. For all other temperature regions, the spin system is treated in the random-phase approximation.

[8] H. S. Bennett and P. C. Martin, Phys. Rev. 138, A608 (1965).

We consider the gradient of the exchange integral, which determines the strength of the interaction, as an experimental parameter and accordingly introduce the coupling function

$$Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')=(\boldsymbol{\nabla}_\alpha)_i J(\boldsymbol{\alpha}-\boldsymbol{\alpha}'). \tag{11}$$

For a lattice with cubic symmetry, the single ion magnetostriction Hamiltonian assumes the form

$$\begin{aligned}H_{\text{int}}{}^S=G_{11}\sum_\alpha\{&\epsilon_{xx}(\boldsymbol{\alpha},t)[S_x{}^2(\boldsymbol{\alpha},t)-\tfrac{1}{2}(S_y{}^2(\boldsymbol{\alpha},t)+S_z{}^2(\boldsymbol{\alpha},t))]\\&+\epsilon_{yy}(\boldsymbol{\alpha},t)[S_y{}^2(\boldsymbol{\alpha},t)-\tfrac{1}{2}(S_z{}^2(\boldsymbol{\alpha},t)+S_x{}^2(\boldsymbol{\alpha},t))]\\&+\epsilon_{zz}(\boldsymbol{\alpha},t)[S_z{}^2(\boldsymbol{\alpha},t)-\tfrac{1}{2}(S_x{}^2(\boldsymbol{\alpha},t)+S_y{}^2(\boldsymbol{\alpha},t))]\}\qquad(12)\\&+G_{44}\sum_\alpha\{\epsilon_{xy}(\boldsymbol{\alpha},t)[S_x(\boldsymbol{\alpha},t)S_y(\boldsymbol{\alpha},t)+S_y(\boldsymbol{\alpha},t)S_x(\boldsymbol{\alpha},t)]\\&+\epsilon_{yz}(\boldsymbol{\alpha},t)[S_y(\boldsymbol{\alpha},t)S_z(\boldsymbol{\alpha},t)+S_z(\boldsymbol{\alpha},t)S_y(\boldsymbol{\alpha},t)]\\&+\epsilon_{zx}(\boldsymbol{\alpha},t)[S_z(\boldsymbol{\alpha},t)S_x(\boldsymbol{\alpha},t)+S_x(\boldsymbol{\alpha},t)S_z(\boldsymbol{\alpha},t)]\}.\end{aligned}$$

We may express the strain components $\epsilon_{ij}(\boldsymbol{\alpha},t)$ in terms of the Fourier transform of the phonon displacement operator $U_i(\mathbf{q},t)$,

$$U_i(\mathbf{q},t)=\sum_\alpha e^{-i\mathbf{q}\cdot\alpha}U_i(\boldsymbol{\alpha},t); \tag{13}$$

namely,

$$\epsilon_{ij}(\boldsymbol{\alpha},t)=\frac{i}{2}\sum_{\mathbf{q}}{}'\{q_iU_j(\mathbf{q},t)+q_jU_i(\mathbf{q},t)\}e^{i\mathbf{q}\cdot\alpha}. \tag{14}$$

The prime indicates that we sum over only those wave vectors $\mathbf{q}$ which lie in the first Brillouin zone. The quantities G_{11} and G_{44} are temperature-independent coupling constants. We may also write Eq. (12) in a more compact way,

$$H_{\text{int}}{}^S=i\sum_\alpha\sum_{\mathbf{q}}{}'G_{ij}q_jU_i(\mathbf{q},t)F_{ij}[S(\boldsymbol{\alpha},t)]e^{i\mathbf{q}\cdot\alpha}. \tag{15}$$

The form (15) is also valid for more general crystal structures. We obtain G_{ij} and F_{ij} by comparing Eq. (15) with Eqs. (12) and (14). A special case arises for the single-ion magnetostriction when $S=\frac{1}{2}$ and we shall restrict our present calculation of this term to the cases for which $S>\frac{1}{2}$.

We obtain the equations of motion for any of the operators U_i, P_i, and S_i, which are all in the Heisenberg representation,

$$i\hbar(\partial A/\partial t)=[A,H], \tag{16}$$

by using the commutation relations for the respective operators. The operator A is any member of the set $\{A\}=U_i$, P_i, and S_i.

B. Correlation Functions

When the system is in thermodynamic equilibrium, we may compute the thermal expectation value of an operator X by using the canonical ensemble,

$$\langle X\rangle=\text{Tr}(e^{-\beta H}X)/\text{Tr}(e^{-\beta H}). \tag{17}$$

Because the operators U_i and P_i commute with all the spin operators S_i, we take the trace over the direct product space of the infinite dimensional space associated with the lattice operators U_i and P_i and the finite dimensional space associated with the spins S_i at each site.

We shall discuss the acoustic properties of the system in terms of the time-ordered lattice displacement correlation function (phonon Green's function),

$$D_{ij}(\boldsymbol{\alpha},t;\boldsymbol{\alpha}',t')=-i\langle(U_i(\boldsymbol{\alpha},t)U_j(\boldsymbol{\alpha}',t'))_+\rangle, \tag{18}$$

where $(\cdots)_+$ denotes the Wick time-ordering operation. When we restrict all times to the interval $0\leq t\leq -i\hbar\beta$, the correlation function (18) satisfies the time boundary condition,

$$D_{ij}(\boldsymbol{\alpha},t;\boldsymbol{\alpha}',t')=D_{ij}(\boldsymbol{\alpha},t+i\hbar\beta;\boldsymbol{\alpha}',t'). \tag{19}$$

The time periodic condition (19) and the invariance of the equilibrium system under translation through a lattice vector and under time translation allow us to express functions in terms of Fourier series representations:

$$\begin{aligned}&D_{ij}(\boldsymbol{\alpha},t;\boldsymbol{\alpha}',t')\\&\quad=\frac{1}{N}\frac{1}{(-i\hbar\beta)}\sum_{\mathbf{q}}{}'\sum_\nu e^{i\mathbf{q}\cdot(\alpha-\alpha')-i\omega_\nu(t-t')}D_{ij}(\mathbf{q};\omega_\nu),\end{aligned} \tag{20}$$

where $\mathbf{q}$ is the wave vector, N is the total number of sites in the lattice, $\omega_\nu=i(\pi\nu/\hbar\beta)$, and ν includes zero and all positive and negative even integers.

We assume that either the volume or the single-ion magnetostrictive interaction dominates for a given solid and we neglect any cross terms between these two types of spin-phonon interactions. We consider first the volume magnetostrictive interaction (10). The commutation relation (8) and the operator equation of motion (16) lead to an equation of motion for the phonon function,

$$\begin{aligned}-\frac{\partial^2}{\partial t^2}D_{ij}(\boldsymbol{\alpha},t;\boldsymbol{\alpha}',t')-\frac{1}{M}\sum_{\alpha''}C_{ik}(\boldsymbol{\alpha},\boldsymbol{\alpha}'')D_{kj}(\boldsymbol{\alpha}'',t;\boldsymbol{\alpha}',t')\\=\frac{1}{M}\delta_{ij}\delta_{\alpha,\alpha'}\delta(t-t')+\frac{i}{M}\sum_{\alpha''}Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}'')\\\times\langle(\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}'',t)U_j(\boldsymbol{\alpha}',t'))_+\rangle.\end{aligned} \tag{21}$$

We show in Appendix A that the last term of Eq. (21) becomes, in lowest-order perturbation theory,

$$\begin{aligned}-\frac{i}{2M}\sum_{\alpha'',\alpha_1,\alpha_2}Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}'')Q_k(\boldsymbol{\alpha}_1-\boldsymbol{\alpha}_2)\\\times\int_0^{-i\hbar\beta}d\bar{t}\langle(\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}'',t)\mathbf{S}(\boldsymbol{\alpha}_1,\bar{t})\cdot\mathbf{S}(\boldsymbol{\alpha}_2,\bar{t}))_+\rangle\\\times[D_{kj}(\boldsymbol{\alpha}_1,\bar{t};\boldsymbol{\alpha}',t')-D_{kj}(\boldsymbol{\alpha}_2,\bar{t};\boldsymbol{\alpha}',t')],\end{aligned} \tag{22}$$

where we now evaluate the thermal expectation values in the absence of the spin-phonon interaction. In addition, the phonon function contains a trace over only the states associated with H_{phonon} and the spin function contains a trace over only the states associated with H_{spin}.

The Fourier space transform of the equation of motion for the phonon function then becomes to the lowest order in the coupling $Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')$,

$$-\frac{\partial^2}{\partial t^2}D_{ij}(\mathbf{q},\,t-t')-\frac{1}{M}C_{ik}(\mathbf{q})D_{kj}(\mathbf{q},\,t-t')=\frac{1}{M}\delta_{ij}\delta(t-t')$$

$$\times\int_0^{-i\hbar\beta}d\bar{t}P_{ik}{}^V(\mathbf{q},\,t-\bar{t})D_{kj}(\mathbf{q},\,\bar{t}-t'),\quad(23)$$

where the kernel in Eq. (23) is

$$P_{ij}{}^V(\mathbf{q},\,t-\bar{t})=-\frac{i}{4M}\sum_{\alpha,\alpha',\alpha_1,\alpha_2}Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')Q_j(\boldsymbol{\alpha}_1-\boldsymbol{\alpha}_2)$$

$$\times(e^{-i\mathbf{q}\cdot\alpha}-e^{-i\mathbf{q}\cdot\alpha'})(e^{i\mathbf{q}\cdot\alpha_1}-e^{i\mathbf{q}\cdot\alpha_2})$$

$$\times\langle(\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}',t)\mathbf{S}(\boldsymbol{\alpha}_1,\bar{t})\cdot\mathbf{S}(\boldsymbol{\alpha}_2,\bar{t}))_+\rangle.\quad(24)$$

The corresponding kernel for the single-ion magnetostrictive interaction (12) has the form

$$P_{ij}{}^S(\mathbf{q},\,t-\bar{t})=-\frac{i}{M}q_lq_mG_{il}G_{jm}\sum_{\alpha,\alpha_1}e^{-i\mathbf{q}\cdot(\alpha-\alpha_1)}$$

$$\times\langle(F_{il}(S(\boldsymbol{\alpha},t))F_{jm}(S(\boldsymbol{\alpha}_1,\bar{t})))_+\rangle.\quad(25)$$

In both cases the kernels ("polarization operators") depend on four-spin correlation functions.

Solving the secular equation which results when $P_{ij}=0$ in Eq. (23) gives us the force constants $C_{ij}(\mathbf{q})$ in terms of the eigenfrequencies $\omega(\lambda,\mathbf{q})$ and the polarization vectors $e_i(\lambda,\mathbf{q})$,

$$\frac{1}{M}C_{ij}(\mathbf{q})=\sum_\lambda\omega^2(\lambda,\mathbf{q})e_i(\lambda,\mathbf{q})e_j(\lambda,\mathbf{q}).\quad(26)$$

Inserting Eq. (26) into Eq. (23) we express the Fourier series coefficient $D_{ij}(\mathbf{q};\omega_\nu)$, which appears in Eq. (20), in terms of the eigenfrequencies, the polarization vectors, and the Fourier series coefficients $P_{ij}(\mathbf{q};\omega_\nu)$ of either kernel (24) or kernel (25);

$$D_{ij}(\mathbf{q},\omega_\nu)$$

$$=\frac{1}{M}\sum_\lambda\frac{e_i(\lambda,\mathbf{q})e_j(\lambda,\mathbf{q})}{\omega_\nu{}^2-\omega^2(\lambda,\mathbf{q})-e_k(\lambda,\mathbf{q})P_{kl}(\mathbf{q};\omega_\nu)e_l(\lambda,\mathbf{q})}.\quad(27)$$

We obtain in this way a relation between the Fourier series coefficient of the phonon function D_{ij} and the kernel P_{ij} on the set of points ω_ν. In addition, the function $P_{ij}(\mathbf{q};z)$ and the function $D_{ij}(\mathbf{q};z)$ are analytic functions of complex z except on the real axis.[9] When we approach the real axis from above, we may define the real and the imaginary parts of P_{ij} by the relation[10]

$$\lim_{\epsilon\to0^+}P_{ij}(\mathbf{q};\omega+i\epsilon)=\mathrm{Re}\{P_{ij}(\mathbf{q},\omega)\}$$

$$-i\,\mathrm{Im}\{P_{ij}(\mathbf{q},\omega)\}.\quad(28)$$

The singularities of $D_{ij}(\mathbf{q};z)$ contain a description of the dominant excitation modes in the system. In particular, the poles of Eq. (27) occur to lowest order in the spin-phonon coupling at the points,

$$\bar{\omega}(\lambda,\mathbf{q})=\omega(\lambda,\mathbf{q})+\mathrm{Re}\left\{\frac{e_iP_{ij}e_j}{2\omega(\lambda,\mathbf{q})}\right\}-i\,\mathrm{Im}\left\{\frac{e_iP_{ij}e_j}{2\omega(\lambda,\mathbf{q})}\right\}.\quad(29)$$

Thus, the phonons interacting with the spin-system experience a frequency shift,

$$\Delta\omega(\lambda,\mathbf{q})=\mathrm{Re}\left\{\frac{e_i(\lambda,\mathbf{q})P_{ij}[\mathbf{q};\omega(\lambda,\mathbf{q})]e_j(\lambda,\mathbf{q})}{2\omega(\lambda,\mathbf{q})}\right\}\quad(30)$$

and a damping

$$\Gamma(\lambda,\mathbf{q})=\mathrm{Im}\left\{\frac{e_i(\lambda,\mathbf{q})P_{ij}[\mathbf{q};\omega(\lambda,\mathbf{q})]e_j(\lambda,\mathbf{q})}{2\omega(\lambda,\mathbf{q})}\right\}.\quad(31)$$

By convention, the acoustic attenuation coefficient has the form

$$\alpha(\lambda,\mathbf{q})=\{\Gamma(\lambda,\mathbf{q})/c(\lambda)\},\quad(32)$$

where $c(\lambda)$ is the speed of sound for a polarization of the type λ.

C. Approximate Spin-Correlation Functions

The kernel $P_{ij}(\mathbf{q},\,t-\bar{t})$ depends upon the four-spin correlation function,

$$f_4(\boldsymbol{\alpha},\boldsymbol{\alpha}',\boldsymbol{\alpha}_1,\boldsymbol{\alpha}_2;t-\bar{t})$$

$$=\langle(S_i(\boldsymbol{\alpha},t)S_j(\boldsymbol{\alpha}',t)S_k(\boldsymbol{\alpha}_1,\bar{t})S_l(\boldsymbol{\alpha}_2,\bar{t}))_+\rangle.\quad(33)$$

We note that $\boldsymbol{\alpha}=\boldsymbol{\alpha}'$ and $\boldsymbol{\alpha}_1=\boldsymbol{\alpha}_2$ for the single-ion magnetostriction and that $i=j$ and $k=l$ for the volume magnetostrictive interaction. Because we know the exact solution to the three-dimensional Heisenberg Hamiltonian for only zero and infinite temperatures, we must resort to approximation procedures for finite temperatures.[11] Following the procedure presented in Sec. III of Ref. 8, we approximate the spin function (33) by the sum of all possible factorizations in terms of lower-order correlation functions. Since we restrict

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[9] L. Kadanoff and P. C. Martin, Ann. Phys. (N.Y.) **24**, 419 (1963).

[10] We have the relation

$$\lim_{\epsilon\to0^+}\frac{1}{\omega+i\epsilon}=P\frac{1}{\omega}\mp i\pi\delta(\omega),$$

where P here represents the principal value.

[11] M. Wortis, Phys. Rev. **138**, A1126 (1965); G. S. Rushbrooke and D. S. Wood, Proc. Phys. Soc. (London) **68A**, 1161 (1955).

the present calculations to the paramagnetic region in the absence of any external magnetic field, the quantity $\langle S_i(\boldsymbol{\alpha},t)\rangle$ is equal to zero and we therefore approximate the four-spin correlation function as a sum of products of all possible pair correlations:

$$\begin{aligned}
&\langle(S_i(\boldsymbol{\alpha},t)S_j(\boldsymbol{\alpha}',t)S_k(\boldsymbol{\alpha}_1,\bar{t})S_l(\boldsymbol{\alpha}_2,\bar{t}))_+\rangle \\
&\quad\approx\langle S_i(\boldsymbol{\alpha},t)S_j(\boldsymbol{\alpha}',t)\rangle\langle S_k(\boldsymbol{\alpha}_1,\bar{t})S_l(\boldsymbol{\alpha}_2,\bar{t})\rangle \\
&\quad+\langle(S_i(\boldsymbol{\alpha},t)S_k(\boldsymbol{\alpha}_1,\bar{t}))_+\rangle\langle(S_j(\boldsymbol{\alpha}',t)S_l(\boldsymbol{\alpha}_2,\bar{t}))_+\rangle \\
&\quad+\langle(S_i(\boldsymbol{\alpha},t)S_l(\boldsymbol{\alpha}_2,\bar{t}))_+\rangle\langle(S_j(\boldsymbol{\alpha}',t)S_k(\boldsymbol{\alpha}_1,\bar{t}))_+\rangle. \qquad (34)
\end{aligned}$$

When we substitute (34) into the expressions (24) and (25) for the kernel P_{ij}, that portion of the sum containing all the equal-time correlations is zero and only terms arising from the last two terms of factorization (34) remain. Furthermore, the correlation function $\langle(S_i(\boldsymbol{\alpha},t)S_j(\boldsymbol{\alpha}',t'))_+\rangle$ for the no external-field paramagnetic region is zero unless $i=j$. We have for an isotropic Heisenberg Hamiltonian,

$$\langle(S_i(\boldsymbol{\alpha},t)S_j(\boldsymbol{\alpha}',t'))_+\rangle=\delta_{i,j}\bar{M}_2(\boldsymbol{\alpha},t;\,\boldsymbol{\alpha}',t'), \qquad (35)$$

where, for example,

$$\bar{M}_2(\boldsymbol{\alpha},t;\,\boldsymbol{\alpha}',t')=\langle(S_z(\boldsymbol{\alpha},t)S_z(\boldsymbol{\alpha}',t'))_+\rangle. \qquad (36)$$

In Appendix B, we show that the factorization (34) is exact at $t=\bar{t}$ and $\beta J=0$ and discuss the physics which the factorization contains. The factorization is an approximation to the exact four-spin function for finite temperatures and $t\neq\bar{t}$ and therefore is always subject to criticism. However, we wish to emphasize that the factorization maintains the correct symmetry properties of the kernel with respect to the frequency ω and the wave vector $\mathbf{q}$. Hence, we shall be able to answer rigorously the question as to whether the attenuation coefficient is an even or an odd function of ω and of $\mathbf{q}$.

III. ATTENUATION COEFFICIENTS

Continuing our analysis, we substitute the factorization (34) into the expression for the kernel $P_{ij}(\mathbf{q},\,t-\bar{t})$ and compute the Fourier coefficients $P_{ij}(\mathbf{q};\omega_\nu)$. By introducing the effective coupling function $\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})$, we write the expressions for $P_{ij}(\mathbf{q};\,\omega_\nu)$ in the general form

$$e_i(\lambda,\mathbf{q})P_{ij}(\mathbf{q};\omega_\nu)e_j(\lambda,\mathbf{q})=\frac{(-i)}{MN(-i\hbar\beta)}\sum_{\mathbf{k}}{}'\sum_{\nu'}\gamma_\lambda{}^2(\mathbf{k},\mathbf{q}) \times\bar{M}_2(\mathbf{k};\omega_{\nu'})\bar{M}_2(\mathbf{k}-\mathbf{q};\omega_{\nu'}-\omega_\nu), \qquad (37)$$

where $\bar{M}_2(\mathbf{k};\omega_\nu)$ is the Fourier series coefficient for the spin pair-correlation function (36) and where the details of the coupling function depend upon which of the two spin phonon interactions we use.

It is convenient to express the coefficient $\bar{M}_2(\mathbf{k};\omega_\nu)$ in terms of its spectral weight function,[8]

$$\bar{M}_2(\mathbf{k};\omega_\nu)=2\int_{-\infty}^{+\infty}\frac{d\omega'}{2\pi}\frac{\chi''(\mathbf{k},\omega')}{(\omega'-\omega_\nu)}. \qquad (38)$$

Then, as usual, using the fact that

$$\frac{1}{\beta\hbar}\sum_{\nu}\frac{1}{\omega-\omega_\nu}=n(\omega)+(\tfrac{1}{2}), \qquad (39)$$

where $n(\omega)=[\exp(\beta\hbar\omega)-1]^{-1}$, we perform the summation over ν' in Eq. (37):

$$\begin{aligned}
&e_i(\lambda,\mathbf{q})P_{ij}(\mathbf{q},\omega_\nu)e_j(\lambda,\mathbf{q}) \\
&\quad=-\frac{1}{MN}\sum_{\mathbf{k}}{}'\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\int_{-\infty}^{+\infty}\frac{d\omega''}{\pi}\chi''(\mathbf{k},\omega') \\
&\qquad\times\chi''(\mathbf{k}-\mathbf{q},\,\omega'')\frac{[n(\omega')-n(\omega'')]}{(\omega'-\omega''-\omega_\nu)}. \qquad (40)
\end{aligned}$$

Applying the prescription (28) to Eq. (40) yields the imaginary part of $e_i(\lambda,\mathbf{q})P_{ij}(\mathbf{q},\,\omega+i\epsilon)e_j(\lambda,\mathbf{q})$:

$$\begin{aligned}
&\mathrm{Im}\{e_i(\lambda,\mathbf{q})P_{ij}(\mathbf{q},\omega)e_j(\lambda,\mathbf{q})\} \\
&\quad=-\frac{1}{MN}\sum_{\mathbf{k}}{}'\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\chi''(\mathbf{k},\omega')\chi''(\mathbf{k}-\mathbf{q},\,\omega'-\omega) \\
&\qquad\times[n(\omega')-n(\omega'-\omega)]. \qquad (41)
\end{aligned}$$

Reference 8 contains an evaluation of the integral appearing in Eq. (41) for the region $\beta\hbar\omega\ll1$. The present calculations also satisfy the condition $\beta\hbar\omega\ll1$. We restrict the calculations to the paramagnetic region $\beta\leq\beta_c$ and consider the wave vector to approach zero. The energy $\hbar\omega$ of the phonon is $\hbar cq$ and the condition $\beta_c\hbar cq\ll1$ is valid for sufficiently small q. Because the density of states factor of Eq. (41) has the limit

$$n(\omega')-n(\omega'-\omega)\approx-\beta\hbar\omega[n(\omega')]^2e^{\beta\hbar\omega'} \qquad (42)$$

for small enough ω, the low-frequency behavior of the spectral weight function contributes most significantly to the frequency integral. Following Ref. 8, we use the low frequency, small wave vector form of the spectral weight function:

$$\chi''(\mathbf{q},\omega)\approx\chi(\mathbf{q},0)Dq^2\omega/[\omega^2+(Dq^2)^2], \qquad (43)$$

where D is the temperature-dependent diffusion coefficient and $\chi(\mathbf{q},0)$ is the wave vector-dependent susceptibility

$$\chi(\mathbf{q},0)=\int_{-\infty}^{+\infty}\frac{d\omega}{\pi}\frac{\chi''(\mathbf{q},\omega)}{\omega}. \qquad (44)$$

We shall use the effective field [random-phase approximation (RPA)] form for the $\mathbf{q}$ and temperature dependence of $\chi(\mathbf{q},0)$,

$$\chi(\mathbf{q},0;\,\mathrm{RPA})=\chi(0,0)/[1+\chi(0,0)j(\mathbf{q})]. \qquad (45)$$

We write the interaction transform in the form

$$j(\mathbf{q})=I(0)-I(\mathbf{q}), \qquad (46)$$

where

$$I(\mathbf{q})=\sum_{\alpha} e^{i\mathbf{q}\cdot\alpha}J(\alpha)\,, \tag{47}$$

and where $\chi(0,0)\equiv\chi$ is the static susceptibility. At high temperatures the expression (45) is exact. The spectral weight function (43) contains the phenomenological assumption that a diffusion equation for the spin magnetization characterizes the long time, $t\gg\tau=(\hbar/J)$, large distance, $r\gg d$, behavior of the system.

Inserting the low-frequency spectral function (43) into Eq. (41) produces the result

$$\operatorname{Im}\{e_i(\lambda,\mathbf{q})P_{ij}(\mathbf{q},\omega)e_j(\lambda,\mathbf{q})\}$$

$$=-\frac{\chi^2\omega}{\beta\hbar MN}\sum_{\mathbf{k}}{}'\,\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})\left\{\frac{(D_2+D_1)[\omega^2+(D_2-D_1)^2]}{[\omega^2+D_2{}^2-D_1{}^2]^2+4D_1{}^2\omega^2}\right.$$

$$\left.\times[1+\chi j(\mathbf{k})]^{-1}[1+\chi j(\mathbf{k}-\mathbf{q})]^{-1}\right\}, \tag{48}$$

where $D_2=D(\mathbf{k}-\mathbf{q})^2$ and $D_1=Dk^2$. Because only small wave vector acoustic phonons propagate easily in a lattice, we shall evaluate the summation in the limit of small q. Acoustic phonons typically have wave vectors $q\sim10^{-5}q_D$, where q_D is the Debye wave vector, $q_D\approx d^{-1}$.

Before we may proceed further, we require the form of the coupling $\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})$ and the exchange transform $j(\mathbf{k})$. We limit the discussion to the case of a simple cubic lattice having only nearest-neighbor exchange interactions. Also, since we are investigating the low frequency, small wave vector results of Eq. (48), we need the above two functions only in the limit of small wave vectors. The exchange transform for the above case becomes

$$j(\mathbf{k})=I(0)-I(\mathbf{k})=2J\sum_{i}(1-\cos k_id)\,, \tag{49}$$

where $i=x$, y, and z, and where d is the lattice constant. For small values of k we write

$$j(\mathbf{k})=Jd^2k^2\,. \tag{50}$$

Comparing Eq. (24) and Eq. (34) gives us the effective coupling coefficient for the volume magnetostrictive interaction,

$$\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})=-(\tfrac{3}{2})\{(e_i(\lambda,\mathbf{q})[Q_i(\mathbf{k})-Q_i(\mathbf{k}-\mathbf{q})])$$

$$\times([Q_j(\mathbf{k})-Q_j(\mathbf{k}-\mathbf{q})]e_j(\lambda,\mathbf{q}))\}\,. \tag{51}$$

Definition (11) yields $Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')=-Q_i(\boldsymbol{\alpha}'-\boldsymbol{\alpha})$ and the Fourier transform $Q_i(\mathbf{k})$ becomes

$$\mathbf{Q}(\mathbf{k})=\sum_{\boldsymbol{\alpha}} e^{i\mathbf{q}\cdot\alpha}\mathbf{Q}(\alpha)=2iQ\sum_{j}\hat{\jmath}\,\sin k_jd\,, \tag{52}$$

where Q is a parameter determining the strength of the interaction and $\hat{\jmath}$ is a unit vector directed along one of the Cartesian axes.

The effective coupling coefficient for a longitudinal phonon propagating along one of the crystal axes, $\mathbf{e}=(\mathbf{q}/q)=\hat{z}$, reduces in the small q limit to the expression

$$\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})\approx6Q^2d^2q^2\cos^2k\mu d\,, \tag{53}$$

where μ is the cosine of the angle between $\mathbf{k}$ and $\mathbf{q}=q\hat{z}$. The effective coupling coefficient for the single-ion interaction assumes under the same conditions as Eq. (53) the form

$$\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})\approx3G_{11}{}^2q^2\,. \tag{54}$$

In order to evaluate the summation in expression (48) we first introduce a simplification concerning the lattice structure. We replace the discrete summation over the first Brillouin zone by an integral over the Debye zone;

$$\lim_{N\to\infty}\frac{1}{N}\sum_{\mathbf{k}}{}'(\cdots)\to\frac{v}{(2\pi)^3}\int_0^{q_D}q^2dq\int_0^{2\pi}d\phi\int_{-1}^{+1}d\mu(\cdots)\,, \tag{55}$$

where $\mu=\cos\theta$ and where $q_D=(6\pi^2/v)^{1/3}$ and $v=d^3$ for a simple cubic lattice. Next we write that part of the integrand of Eq. (48) between the curly brackets in terms of functions which are even and odd in μ; i.e.,

$$\{\cdots\}(\mu)=\tfrac{1}{2}[\{\cdots\}(\mu)+\{\cdots\}(-\mu)]$$

$$+\tfrac{1}{2}[\{\cdots\}(\mu)-\{\cdots\}(-\mu)] \tag{56}$$

$$=F_e(\mu)+F_0(\mu)\,,$$

where $F_e(\mu)=F_e(-\mu)$ and $F_0(\mu)=-F_0(-\mu)$. Since the coupling $\gamma_\lambda{}^2$ is an even function of μ, only the term $F_e(\mu)$ contributes. The dominant contribution to the integral of Eq. (48) comes from the region of small k and thus for convenience in performing the angular integration over μ we approximate the $\cos^2k\mu d$ factor of Eq. (53) by 1. The additional restraints that

$$k_D{}^2d^2\gg q^2d^2\,, \tag{57}$$

$$\omega^2\gg D^2q^4\,, \tag{58}$$

and

$$(1/\chi J)\gg q^2d^2\,, \tag{59}$$

allow us to perform more readily the integration over k. The results are, when β approaches zero,

$$\operatorname{Im}\{e_iP_{ij}e_j\}\approx-\frac{\gamma_\lambda{}^2(0,\mathbf{q})\omega d^3q_D(\chi J)^2}{4\pi^2\hbar\beta J^2DM} \tag{60}$$

and when β is near β_c,

$$\operatorname{Im}\{e_iP_{ij}e_j\}\approx-\frac{\gamma_\lambda{}^2(0,\mathbf{q})\omega d^2(\chi J)^{3/2}}{16\pi\hbar\beta J^2DM}\,. \tag{61}$$

Inserting Eq. (60) and Eq. (61) into Eq. (32) leads to the respective expressions for the attenuation coefficient: when β approaches zero,

$$\alpha(\mathbf{q})\approx\frac{\gamma_\lambda{}^2(0,\mathbf{q})d^2(q_Dd)(\chi J)^2}{8\pi^2\hbar\beta J^2DMc} \tag{62}$$

and when β is near β_c,

$$\alpha(\mathbf{q})\approx\frac{\gamma_\lambda{}^2(0,\mathbf{q})d^2(\chi J)^{3/2}}{32\pi\hbar\beta J^2DMc}, \tag{63}$$

where for a simple cubic lattice $q_Dd\approx3.9$, where

$$\gamma_\lambda{}^2(0,\mathbf{q})\approx6Q^2d^2q^2 \tag{64}$$

for the volume magnetostrictive interaction and

$$\gamma_\lambda{}^2(0,\mathbf{q})=3G_{11}{}^2q^2 \tag{65}$$

for the point ion magnetostrictive interaction, and where $\omega(\mathbf{q})=cq$.

IV. RESULTS AND CONCLUSIONS

The results (62) and (63) tell us that the attenuation coefficient is a function of the static susceptibility $\chi(\beta)$ and of the spin diffusion $D(\beta)$ and that it is proportional to an even power of the wave vector, namely $q^2=[\omega^2(\mathbf{q})/c^2]$.

We now return to the restraints (57) to (59) and demonstrate that experimental conditions satisfying them exist. The ratio (q/q_D) is typically about 10^{-5} and this satisfies inequality (57). Because $q_D\approx(1/d)\approx(10^{+8}/\text{cm})$ and $c\approx(10^5\text{ cm/sec})$ for many ferromagnets, we have $q\approx(10^3/\text{cm})$ and $\omega=cq\approx(10^8/\text{sec})$. The diffusion coefficient attains a maximum value at $\beta=0$ and has the value for a simple cubic lattice,[8]

$$D(\beta=0)\approx[4.14\times10^{-5}BT_c/\{S(S+1)\}^{1/2}]\ \text{cm}^2\ \text{sec}^{-1}\ {}^\circ\text{K}^{-1}, \tag{66}$$

where we have used $\beta_cJ\approx[3/S(S+1)]$, where $10^2\ {}^\circ\text{K}\lesssim T_c\lesssim10^3\ {}^\circ\text{K}$ for many ferromagnets, and where B depends upon the method[12,13] used to compute D and has the values $0.19\lesssim B\lesssim0.33$. Thus, inequality (58) becomes

$$c^2\gg D^2q^2, \tag{67}$$

and the above estimates for c and D meet condition (67).

Finally, condition (59) requires that

$$\chi J\ll10^{10}. \tag{68}$$

In the high-temperature limit the susceptibility goes to zero linearly with β,

$$\chi\underset{\beta\to0}{\longrightarrow}\frac{S(S+1)\beta}{3}. \tag{69}$$

The limit (69) is exact. The susceptibility becomes infinite at the critical point and most treatments of the susceptibility predict that it will behave sufficiently close to the critical point according to the form

$$\chi j=A[\beta/(\beta_c-\beta)]^\gamma, \tag{70}$$

where A varies slowly with β and is usually about $1.0\gtrsim A\gtrsim0.01$ and where $1\leq\gamma\lesssim2$. The Weiss molecular field gives $\gamma=1$; the RPA models give $\gamma=2$; and the Padé approximant method gives a value which is indistinguishable from $\gamma\approx(4/3)$. The experiments thus far appear to agree most closely with the Padé-approximant method because for most ferromagnets it is found that $1.2\lesssim\gamma\lesssim1.6$. The limit (69) readily satisfies inequality (59) via inequality (68) at high temperatures. However, Eq. (70) does not satisfy inequality (59) for β sufficiently near β_c; i.e.,

$$A\times10^{-10}\gtrsim[(\beta_c/\beta)-1]^\gamma. \tag{71}$$

But the latter does not concern us because from Appendix B we argue that the factorization (34) becomes much less reliable whenever $[(\beta_c/\beta)-1]\lesssim10^{-2}$. Temperatures as close to the transition point as $[(\beta_c/\beta)-1]\approx10^{-4}$ readily meet inequality (68), and such temperatures are outside the expected region of validity of our theory. Finally the above values for the speed of sound and the wave vector satisfy the condition following Eq. (41), namely, $\beta_c\hbar cq\ll1$, provided $T_c\gg10^{-3}\ {}^\circ\text{K}$.

In order to determine the temperature dependence of the attenuation coefficient we require the temperature dependence of $\chi(\beta)$ and $D(\beta)$. Equations (69) and (70) give us $\chi(\beta)$. Dimensional arguments alone determine the form of $D(\beta)$ at high temperatures, and the different evaluations all have the form

$$D(\beta J=0)=B(Jd^2/\hbar)[S(S+1)]^{1/2}, \tag{72}$$

where B varies as indicated in Eq. (66). In the neighborhood of the Curie point, the nonlinear integral equation method (nl) and the sum-rule moment method (sm) both predict that the diffusion coefficient approaches zero as β approaches β_c; i.e.,

$$\frac{D_{nl}(\beta\to\beta_c)}{D_{nl}(\beta=0)}=\frac{B_{nl}}{S(S+1)(\chi J)^{1/4}} \tag{73}$$

and

$$\frac{D_{sm}(\beta\to\beta_c)}{D_{sm}(\beta=0)}=\frac{B_{sm}}{S(S+1)(\chi J)}, \tag{74}$$

where $B_{nl}\approx2.38$ and $B_{\text{sm}}\approx1.15$. It is interesting to notice that if one uses the sum-rule moment form of the diffusion coefficient and assumes that the molecular-field theory gives the static susceptibility correctly, then one obtains the expression for the spectral weight $\chi''(\mathbf{q},\omega)$ originally assumed by Van Hove.[14]

We conclude from these considerations that the ultrasonic attenuation coefficient goes to zero linearly with β in the high-temperature limit and that the coefficient increases rapidly in the neighborhood of the Curie

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[12] P. G. de Gennes, J. Phys. Chem. Solids 4, 223 (1958).

[13] H. Mori and K. Kawasaki, Progr. Theoret. Phys. (Kyoto)

[14] L. Van Hove, Phys. Rev. **95**, 1374 (1954).

TABLE I. The small wave vector limit of the effective coupling $\gamma_\lambda^2(\mathbf{k},\mathbf{q})$ which results from Eq. (34). The direction of propagation is $\mathbf{q}=q(\sin\theta\cos\varphi\hat{x}+\sin\theta\sin\varphi\hat{y}+\cos\theta\hat{z})$, the longitudinal mode polarization vector is $\mathbf{e}_L=\mathbf{q}/q$, and a transverse mode polarization vector is $\mathbf{e}_T=-\sin\varphi\hat{x}+\cos\varphi\hat{y}$, where q, θ, and φ are the spherical coordinates with one of the crystal axes as the z axis.

Interaction and mode	$\lim_{\substack{k\to 0\\ q\to 0}} \gamma_\lambda^2(\mathbf{k},\mathbf{q})$
Eq. (10) Longitudinal	$6Q^2d^2q^2$
Eq. (10) Transverse	$(\frac{3}{2})Q^2d^6q^2\sin^2\theta\sin^2\varphi\cos^2\varphi(k_x-k_y)^2$
Eq. (12) Longitudinal	$[3G_{11}^2\{\cos^2\theta(\cos^2\theta-\sin^2\theta)+\sin^4\theta(\cos^4\varphi-\sin^2\varphi\cos^2\varphi+\sin^4\varphi)\}+4G_{44}^2\sin^2\theta(\cos^2\theta+\sin^2\theta\cos^2\varphi\sin^2\varphi)]q^2$
Eq. (12) Transverse	$[9G_{11}^2\sin^2\theta\sin^2\varphi\cos^2\varphi+G_{44}^2\{\cos^2\theta+\sin^2\theta(\cos^2\varphi-\sin^2\varphi)^2\}]q^2$

TABLE II. Temperature dependence of the attenuation predicted by Eq. (34). The temperature factors are $F_0=[(\chi J)^2/\beta D]$, $F_c=[(\chi J)^{3/2}/\beta D]$, and $F_c^t=(1/\beta D)$. The respective attenuation coefficients are directly proportional to these temperature factors.

Interaction and mode	Temperature factor	
	$\beta\to 0$	$\beta\to\beta_c$
Eq. (10) Longitudinal	F_0	F_c
Eq. (10) Transverse	F_0	F_c^t
Eq. (12) Longitudinal	F_0	F_c
Eq. (12) Transverse	F_0	F_c

point according to the manner in which $(\chi J)^\eta$ increases. The exponent η depends upon the method which one chooses to compute the diffusion coefficient and has the bounds $(\frac{3}{2})\lesssim\eta\lesssim(\frac{5}{2})$.

We call attention to the fact that treating the spin system in the RPA leads to no attenuation of the phonons in the neighborhood of the Curie point,[7] for both $\beta\gtrsim\beta_c$ and $\beta\lesssim\beta_c$. We also want to emphasize that all estimates of the behavior of D near β_c are as yet bery unreliable.[15]

We may easily extend our present calculation to include the attenuation of transverse modes and the propagation of the sound in arbitrary directions relative to the crystal axes. For the volume magnetostriction, Eq. (10), the attenuation of a longitudinal mode is independent of the direction of propagation. The attenuation of a transverse mode is zero for sound waves propagating along the crystal axes. When the transverse mode propagates off the crystal axes, the attenuation depends upon the angles that the propagation vector and the polarization vector make with the crystal axes. Equation (51) gives us these angular dependences. In addition, the attenuation of an off-axis transverse mode exhibits a different temperature dependence in the neighborhood of the Curie point from that given by Eq. (63). Equation (51) for a transverse mode (off-axis propagation) gives a $\mathbf{k}$ dependence of the effective coupling $\gamma_\lambda^2(\mathbf{k},\mathbf{q})$ which is different from that obtained for a longitudinal mode. When we perform the $\mathbf{k}$ integration, the singular factor $(\chi J)^{3/2}$ does not occur and the increase in the attenuation now arises from the $(1/D)$ factor. The explicit temperature dependence of the attenuation depends on the methods by which the diffusion coefficient D and the static susceptibility χ are calculated, as we have indicated in Eqs. (70), (73), and (74).

For the single-ion magnetostriction interaction (12), which is the dominant interaction in most solids, both the frequency and the temperature dependence of the attenuation are independent of the relative direction of the propagation vector and the polarization vector and of the directions of these latter vectors relative to the crystal axes. The effective coupling is, however, different for the different cases. For a transverse mode propagating along a crystal axis, the effective coupling $3G_{11}^2$ in Eq. (65) is replaced by G_{44}^2. For both longitudinal and transverse modes propagating in an off-axis direction, the effective coupling becomes a linear combination of G_{11}^2 and G_{44}^2, for which the coefficients depend on the angles which the propagation vector and the polarization vector make with the crystal axes. We obtain the explicit angle dependence in a straightforward manner from Eq. (25). Tables I and II contain a summary of the contents of the above two paragraphs.

As a final point, we mention that if we remove the restriction that the volume magnetostriction be invariant under rotations and consider a more general interaction term,

$$\sum_{\alpha,\alpha'}[U_i(\alpha,t)-U_i(\alpha',t)]Q_{ijk}(\alpha,\alpha')S_j(\alpha,t)S_k(\alpha',t),\quad(75)$$

then we shall obtain the same frequency and temperature results because the Fourier coefficient $P_{ij}(\mathbf{q},\omega_\nu)$ will still have the form (37) and the $\mathbf{k}$ and $\mathbf{q}$ dependence of the effective coupling $\gamma_\lambda^2(\mathbf{k},\mathbf{q})$ will remain the same.

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APPENDIX A: PERTURBATION THEORY

We want to evaluate the spin-displacement correlation function,

$$\langle(U_j(\alpha',t')\mathbf{S}(\alpha,t)\cdot\mathbf{S}(\alpha'',t))_+\rangle,\quad(A1)$$

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[15] J. Kocinski, J. Phys. Chem. Solids 25, 211 (1964). This work derives an expression for D which is finite at $\beta=\beta_c$. However, this calculation uses the constant coupling approximation which ignores the fact that the values of the molecular field differ from site to site. This neglect leads to serious error, particularly in the neighborhood of the Curie point.

to lowest order in the spin-phonon interaction, H_{int}. We refer to Eq. (17) and consider the implicit numerator of correlation (A1),

$$N_j=\mathrm{Tr}[e^{-\beta H}(U_j(\boldsymbol{\alpha}',t')\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}'',t))_+]. \tag{A2}$$

The operator equation,

$$U(-i\hbar\beta)=U_0(-i\hbar\beta)U_I(-i\hbar\beta), \tag{A3}$$

relates the operator $U(-i\hbar\beta)=e^{-\beta H}$ to the corresponding operator in the absence of the interaction $U_0(-i\hbar\beta)=e^{-\beta H_0}$, where

$$U_I(-i\hbar\beta)=\left(\exp\left[(-i/\hbar)\int_0^{-i\hbar\beta}d\bar{t}H_I(\bar{t})\right]_+\right). \tag{A4}$$

This relation is obtained by solving the equation of motion for $U_0^{-1}(t)U(t)$ in the usual way[16] and then by performing the analytic continuation to $t=-i\hbar\beta$. Here $H_I(t)$ is the interaction Hamiltonian in the interaction representation,

$$H_I(t)=U_I(t)H_{\mathrm{int}}U_I^{-1}(t). \tag{A5}$$

By expanding the operator $U_I(-i\hbar\beta)$,

$$U_I(-i\hbar\beta)\approx 1-(i/\hbar)\int_0^{-i\hbar\beta}d\bar{t}H_I(\bar{t})+\cdots, \tag{A6}$$

we shall be able to compute the quantity N_j to lowest order in the spin-phonon interaction. The expression (A6) yields

$$N_j\approx\mathrm{Tr}\left[e^{-\beta H_0}\left(\left\{1-(i/\hbar)\int_0^{-i\hbar\beta}d\bar{t}H_I(\bar{t})+\cdots\right\}\right.\right.$$
$$\left.\left.\times U_j(\boldsymbol{\alpha}',t')\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}'',t)\right)_+\right], \tag{A7}$$

where we now take the trace over states which are eigenfunctions of H_0. Since H_{phonon} is invariant under a uniform translation of the lattice, any term with an odd number of displacement operators is zero. Thus, to the lowest order in the interaction H_{int} we find,

$$N_j=\frac{i}{2}\sum_{\alpha_1,\alpha_2}Q_k(\boldsymbol{\alpha}_1-\boldsymbol{\alpha}_2)\,\mathrm{Tr}\left[e^{-\beta H_0}\right.$$
$$\times\int_0^{-i\hbar\beta}d\bar{t}\left(\{U_k(\boldsymbol{\alpha}_1,\bar{t})-U_k(\boldsymbol{\alpha}_2,\bar{t})\}\mathbf{S}(\boldsymbol{\alpha}_1,\bar{t})\cdot\mathbf{S}(\boldsymbol{\alpha}_2,\bar{t})\right.$$
$$\left.\left.\times\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}'',t)U_j(\boldsymbol{\alpha}',t')\right)_+\right]. \tag{A8}$$

Then using the finite temperature generalization of the Wick's theorem[17] to contract the phonon operators, we obtain

$$\langle(U_j(\boldsymbol{\alpha}',t')\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}'',t))_+\rangle\approx-\tfrac{1}{2}\sum_{\alpha_1,\alpha_2'}Q_k(\boldsymbol{\alpha}_1-\boldsymbol{\alpha}_2)$$
$$\times\int_0^{-i\hbar\beta}d\bar{t}\langle(\mathbf{S}(\boldsymbol{\alpha}_1,\bar{t})\cdot\mathbf{S}(\boldsymbol{\alpha}_2,\bar{t})\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}'',t))\rangle$$
$$\times[D_{kj}(\boldsymbol{\alpha}_1,\bar{t};\alpha't')-D_{kj}(\boldsymbol{\alpha}_2,\bar{t};\alpha't')]. \tag{A9}$$

The term (22) in the text then follows from Eq.(A9).

APPENDIX B: FOUR-SPIN CORRELATIONS

Since we do not know the exact solution for finite temperatures to the three-dimensional Heisenberg model, we have introduced the factorization (34).

In this Appendix, we shall discuss the extent to which this factorization is justified.

We may make three rigorous statements concerning the factorization. First, the factorization is unique in the sense that all other possible factorizations give no contribution in the paramagnetic region to the expression for $P_{ij}(\mathbf{q},t-\bar{t})$, Eq. (24) and Eq. (25). Second, the factorization yields an exact expression for $P_{ij}(\mathbf{q},t-\bar{t})$ at $\beta J=0$ and at $t=\bar{t}$. In the limit of $\beta J=0$, the spin state $n_i(\boldsymbol{\alpha}_i)$ at site $\boldsymbol{\alpha}_i$ becomes independent of the spin state $n_{i'}(\boldsymbol{\alpha}_{i'})$ at the site $\boldsymbol{\alpha}_{i'}$, $\boldsymbol{\alpha}_{i'}\neq\boldsymbol{\alpha}_{i'}$, and the trace over all spin states reduces to the product of traces over all spin states at each site,

$$\lim_{\beta J\to 0}\mathrm{Tr}_N[\cdots]=\prod_{i=1}^{N}\mathrm{Tr}_{\alpha 1}[\cdots]; \tag{B1}$$

i.e., we have an N product spin space. We then explicitly evaluate[18] all traces appearing in the factorization (34) and find that at $\beta J=0$ and $t=\bar{t}$ the left-hand side equals the right-hand side. Third, the extension of the factorization (34) to the ferromagnetic region leads to an exact expression for $P_{ij}(\mathbf{q},t-\bar{t})$ at $\beta J=\infty$ and for any $t-\bar{t}$. When $\beta J=\infty$, the spins at all sites are in the ground state and the trace reduces to one diagonal matrix element. We then use the time evolution of operators in the Heisenberg representation

$$S_i(\boldsymbol{\alpha},t)=[\exp(i/\hbar)H_{\mathrm{spin}}(t-t')]$$
$$\times S_i(\boldsymbol{\alpha},t')[\exp(-i/\hbar)H_{\mathrm{spin}}(t-t')] \tag{B2}$$

to prove the third statement for which the ground state is an eigenfunction of H_{spin}.

A calculation of the specific heat gives us additional insight about the physics which the $t=\bar{t}$ factorization describes. We express the specific heat at constant volume in terms of the four spin correlations and lower-

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[16] S. Schweber, *An Introduction to Relativistic Quantum Field Theory* (Harper & Row Publishers Inc., New York, 1961), Chap. II.

[17] C. Bloch and C. de Dominicis, Nucl. Phys. **10**, 509 (1959).

[18] J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).

order correlations;

$$\frac{4C_v}{k\beta^2}=\sum_{\boldsymbol{\alpha},\boldsymbol{\alpha}'}\sum_{\boldsymbol{\alpha}_1,\boldsymbol{\alpha}_2} J(\boldsymbol{\alpha}-\boldsymbol{\alpha}')J(\boldsymbol{\alpha}_1-\boldsymbol{\alpha}_2)\times\{\langle(\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}',t))(\mathbf{S}(\boldsymbol{\alpha}_1,t)\cdot\mathbf{S}(\boldsymbol{\alpha}_2,t))\rangle-\langle\mathbf{S}(\boldsymbol{\alpha},t)\cdot\mathbf{S}(\boldsymbol{\alpha}',t)\rangle\langle\mathbf{S}(\boldsymbol{\alpha}_1,t)\cdot\mathbf{S}(\boldsymbol{\alpha}_2,t)\rangle\}. \quad \text{(B3)}$$

In the $\beta J\rightarrow 0$ limit, we evaluate Eq. (B3) exactly and then evaluate it with the four-spin correlations replaced by the factorization (34). We find that the resulting expressions for the specific heat $(C_v/Nk\beta^2)$ are the same.

Previous work[1] shows that as we approach the transition region the correlations of greater number of spins become successively more important. For example, whenever the temperature fails to satisfy the condition $[(\beta_c/\beta)-1]\gg 0.01$, then the correlations of three spins become comparable in magnitude to the correlation of two spins. We conclude from this that the factorization (34) is much less reliable near the transition point. But even though the factorization is suspect near the transition point, it is mathematically well defined for all temperatures such that $\beta<\beta_c$ and provides us with an interpolation to the transition region. In particular, using the factorization (34), the spectral weight function (43), and the RPA approximation for $\chi(\mathbf{q},0)$, we calculate the specific heat in the vicinity of the transition point for a simple cubic lattice. The result is that the specific heat diverges as the square root of the static susceptibility:

$$\frac{C_v}{Nk}\xrightarrow[\beta\rightarrow\beta_c-]{}\frac{27}{2\pi^2}(\chi J)^{1/2}. \quad \text{(B4)}$$

If one believes that the exact specific heat has a logarithmic divergence and that the static susceptibility diverges as $(T-T_c)^{-\gamma}$ where $2>\gamma>1$, then the critical fluctuations described by the insertion of form (43) into the factorization (34) are too large because they lead to a divergence which is more singular than a logarithmic divergence.

Since we require a knowledge of $\mathrm{Im}(e_iP_{ij}e_j)$ for small wave vectors q and low frequencies ω, we want the behavior of the four-spin correlation function, $f_4(\boldsymbol{\alpha},\boldsymbol{\alpha}',\boldsymbol{\alpha}_1,\boldsymbol{\alpha}_2;t-\bar{t})$, for times $|t-\bar{t}|\gg\tau$ and for distances $|\boldsymbol{\alpha}-\boldsymbol{\alpha}_1|$, $|\boldsymbol{\alpha}-\boldsymbol{\alpha}_2|$, $|\boldsymbol{\alpha}'-\boldsymbol{\alpha}_1|$, and $|\boldsymbol{\alpha}'-\boldsymbol{\alpha}_2|$ all much greater than the lattice spacing d. In addition, we may assert that the coupling $Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')$ is a short-ranged function; i.e., $Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')$ is very small for $|\boldsymbol{\alpha}-\boldsymbol{\alpha}'|>d$. From this assertion we argue that those terms of the kernel (24) for which $\boldsymbol{\alpha}$ is near $\boldsymbol{\alpha}'$ and for which $\boldsymbol{\alpha}_1$ is near $\boldsymbol{\alpha}_2$ dominate. The latter is compatible with the previous conditions. But thus far we have discussed only the $t=\bar{t}$ aspects of the factorization and we may question whether the preceding discussion is relevant to the attenuation coefficient; particularly since only intuitive statements are available for the $t\neq\bar{t}$ case at finite temperatures.

The factorization is a statement that a given function of t and $\bar{t}$ behaves as the sum of a time-independent function $(t=\bar{t})$ and of products of two time-dependent functions. The pair-correlation form (43) states that for the paramagnetic region the self-diffusion of the spins dominates over all other possible mechanisms at large distances and long times $(q\rightarrow 0$ and $\omega\rightarrow 0)$. Hence, in effect, the factorization (34) and the form (43) permit us to obtain information about that part of the large-distance long-time behavior of the four-spin correlations which arises from the large-distance long-time behavior of the pair correlations, whenever the system is sufficiently far from the transition region.

We may strengthen our intuitive discussion for $t\neq\bar{t}$ by examining a simple solution to the diffusion equation for the magnetization $m(\mathbf{r},t)$,

$$\partial m(\mathbf{r},t)/\partial t=D\nabla^2 m(\mathbf{r},t), \quad \text{(B5)}$$

with the $t=0$ time boundary condition that

$$m(\mathbf{r},0)=\begin{cases} M \text{ if } \mathbf{r}=0\\ 0 \text{ if } \mathbf{r}\neq 0\end{cases};$$

namely,

$$m(\mathbf{r},t)=M(\pi/Dt)^{3/2}\exp[-(r^2/4Dt)] \quad \text{(B6)}$$

for $t\gg\tau$ and $r\gg d$. From Eq. (B6) we obtain the hydrodynamic domain and the condition that $|m(\mathbf{r},t)/M|\ll 1$, whenever t is great enough for fixed r or whenever r is great enough for fixed t. If we assume that the higher-order correlations $\bar{M}_2$ and f_4 exhibit diffusion properties which are similar to those described by Eq. (B5), then we may extend the suggestions of Eq. (B6) to these functions. That is to say, for high enough temperatures, the spin at site $\boldsymbol{\alpha}$ at time t is very weakly correlated with the spin at site $\boldsymbol{\alpha}$ at time $\bar{t}$, whenever $|t-\bar{t}|\gg\tau=(\hbar/J)$ and the spin at site $\boldsymbol{\alpha}$ at time t is very weakly correlated with the spin at site $\boldsymbol{\alpha}_1$ at time $\bar{t}$ whenever $|\boldsymbol{\alpha}-\boldsymbol{\alpha}_1|\gg c_1$, for fixed $|t-\bar{t}|$ where c_1 is the correlation length. A modified RPA method predicts[1] that when $t=\bar{t}$, the correlation length behaves as $(1+\chi J)^{1/2}d$.

Bearing in mind the additional intuitive statements of the preceding paragraph, we re-examine the four-spin function f_4 and approximation (34) to it. We argue that the hydrodynamic domain $(q\rightarrow 0$ and $\omega\rightarrow 0)$ obtains whenever all the unbarred variables are far from the barred or numbered ones and that any approximation to f_4 must maintain this feature. Observe that the hydrodynamic domain of the last two terms of the factorization (34) corresponds to the hydrodynamic domain of the four spin function f_4, but that the first term fails to meet this criterion. However, because the contributions due to the first term sum to zero in all calculations of the specific heat (a $t=\bar{t}$ property) and of the diffusion coefficient and the attenuation coefficient ($t\neq\bar{t}$ properties), the first term of the factorization leads to no consequence for the above cases and we may ignore its failure to describe the hydrodynamic domain.