where

$$k_1^2=4\eta[(3+\eta\cos^2x)+3i\Gamma](3+\eta+3i\Gamma)^{-2}. \quad (A2)$$

The complete elliptic integral of the first kind can be represented in series form by

$$K(k)=\frac{1}{2}\frac{\pi}{2}\sum_n a_n^2k^{2n}, \quad |k|<1, \quad (A3)$$

where

$$a_n=\frac{1\times3\times5\times\cdots\times(2n-1)}{2\times4\times6\times\cdots\times2n}, \quad (A4)$$

$$a_0=1.$$

Only the first two terms of the series (A3) is necessary for four-place accuracy. There is no difficulty in extending the series to as many terms as needed.

Substituting the first two terms of Eq. (A3) into Eq. (A1), integrating the complex function with respect to x, and rearranging terms, we find that

$$I_2=I_2^a+I_2^b+I_2^c, \quad (A5)$$

where

$$I_2^a=\frac{3}{3+\eta}\frac{1}{Q}\sum_{R=1}^{\infty}\frac{y^2}{y^2+R^2}, \quad (A6)$$

$$I_2^b=(3\pi^2)^{-1}\eta Q\sum_{R=1}^{\infty}[(y^2+R^2)^{-1}-2R^2(y^2+R^2)^{-2}], \quad (A7)$$

$$I_2^c=-(2\pi)^{-1}(3\pi)^{-3}\eta^2(3+\eta)Q^2$$
$$\times\sum_{R=1}^{\infty}[(y^2+R^2)^{-2}-4R^2(y^2+R^2)^{-3}], \quad (A8)$$

and

$$y=(3\pi)^{-1}(3+\eta)Q. \quad (A9)$$

The sums in Eqs. (A6)–(A8) are evaluated by the use of the following Laplace transforms

$$\frac{1}{y^2+R^2}=\mathcal{L}\left[\frac{1}{y}\sin yt\right], \quad (A10)$$

$$\frac{R^2}{(y^2+R^2)^2}=\mathcal{L}\left\{\left(\frac{1}{2y}\right)[\sin yt+yt\cos yt]\right\}, \quad (A11)$$

$$\frac{R^2}{(y^2+R^2)^3}=\mathcal{L}\left\{\frac{1}{8y^3}[(1+y^2t^2)\sin yt-yt\cos yt]\right\}. \quad (A12)$$

Here the definition of the one-dimensional Laplace transform is used.[9] From Eq. (A10) one finds, for example, that the first sum in Eq. (A7) becomes

$$\sum_{R=1}^{\infty}\frac{1}{R^2+y^2}=\frac{1}{y}\int_0^{\infty}\frac{\sin yt}{e^t-1}. \quad (A13)$$

The other sums in I_2^a and I_2^c are obtained in a similar way. Integrating Eq. (A13) and its counter parts for the other sums, one easily obtains Eq. (20).

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Frequency Shifts of Acoustic Phonons in Heisenberg Paramagnets. III

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The propagation of sound waves in ferromagnetic and antiferromagnetic insulators is studied within the framework of two models which describe the interaction between the spin system and the lattice. Expressions for the frequency shifts (phonon renormalizations) at high temperatures and near the transition temperatures are obtained in terms of time-dependent correlation functions. The frequency shifts for long-wavelength phonons are found to be negative, to increase rapidly in the vicinity of the transition temperature, and to be less singular than the attenuation coefficients. The ratio of the frequency shift to the unperturbed phonon frequency is shown to be independent of the phonon frequency for long wavelengths. These results agree qualitatively with present experiments.

I. INTRODUCTION

RESEARCHERS[1–3] have measured recently the frequency-shift ratio (velocity-shift ratio) of acoustic-longitudinal phonons propagating in the magnetic insulators, EuO, $RbMnF_3$, and MnF_2. The maximum frequency shift occurs near the transition temperature T_c and is negative. It arises from the phonons interacting with the critical fluctuations of the spin system near the transition temperature. The experimental ratio of $\Delta\omega=\Delta cq$, divided by the unperturbed frequency $\omega=cq$, $\Delta\omega/\omega=\Delta c/c$, is independent of the phonon wave vector $\mathbf{q}$ for sufficiently small wave vectors[1–3] ($q<10^{-3}$ cm^{-1}). We denote the unperturbed speed of sound by c and the shift in the speed of sound by Δc.

We list here the recent experimental results. A log-

[1] B. Golding, Phys. Rev. Letters **20**, 5 (1968); and (private communication).
[2] B. Luthi (private communication).
[3] R. Moss and R. Leisure (private communication).

arithmic dependence upon the reduced temperature $\epsilon=(|T-T_c|/T_c)$ reproduces fairly well the frequency-shift ratio measured in the ferromagnet EuO,[2]

$$\Delta\omega/\omega\sim-\omega^0\ln\epsilon,$$

as ϵ approaches zero. The quantity ω^0 is a frequency-independent constant. A power law behavior approximates the frequency-shift ratio measured in the antiferromagnet MnF_2,[3]

$$\Delta\omega/\omega\sim-\omega^0\epsilon^{-\xi}.$$

The exponent ξ is about 0.4 ± 0.1. Luthi[2] reports that the antiferromagnetic metal Tb data do not fit a power law $\epsilon^{-\xi}$ for T near T_N (the Néel temperature) and for $\xi\neq0$; but that perhaps they would better fit a logarithmic dependence upon ϵ, i.e., when $\xi=0$. Experimental plots of $\Delta\omega/\omega$ as a function of temperature for the antiferromagnet $RbMnF_3$ exist.[1] However, an analysis of these data in terms of power laws ($\xi\neq0$) or logarithms ($\xi=0$) does not exist.

We cite two reasons why these results are preliminary, rather than definitive.[1] First, it is difficult to determine the experimental background corrections for Δc. Second, there are the additional uncertainties associated with dispersion (the variation of c with increasing ω). Because these two questions remain unsolved quantitatively, we consider at present that the experimental results outlined above suggest only qualitative trends.

In this paper, we calculate the frequency shift of acoustic phonons interacting with the localized spins in isotropic Heisenberg magnets. We have shown in Ref. 4 that the phonons interacting with the spin system experience an angular frequency shift $\Delta\omega(\lambda,\mathbf{q})$ which is given to lowest order in the spin-phonon coupling by the expression

$$\Delta\omega(\lambda,\mathbf{q})=\mathrm{Re}\left[\lim_{\delta\to0}\left(\frac{e_i(\lambda,\mathbf{q})P_{ij}[\mathbf{q};\omega(\lambda,\mathbf{q})+i\delta]e_j(\lambda,\mathbf{q})}{2\omega(\lambda,\mathbf{q})}\right)\right]. \quad (1)$$

The subscripts i and j refer to the components of the Cartesian coordinates. We obtain the phonon eigenfrequencies $\omega(\lambda,\mathbf{q})$ and the polarization vectors $\mathbf{e}(\lambda,\mathbf{q})$ by solving the secular equation for the unperturbed phonon part of the crystal Hamiltonian. The acoustic-phonon eigenfrequency $\omega(\lambda,\mathbf{q})$ becomes $\omega(\lambda,\mathbf{q})=c(\lambda)q$ in the small wave vector $\mathbf{q}$ limit. We denote the magnitude of the wave vector $\mathbf{q}$ by q and the speed of the acoustic phonon having polarization λ by $c(\lambda)$. The space-time Fourier transform of the polarization kernel, $P_{ij}(\mathbf{q};\omega)$, depends upon four-spin correlation functions. We refer the reader to Sec. II of Ref. 4 for a development of the above equation and for a discussion of the approximations used to compute $\Delta\omega(\lambda,\mathbf{q})$. The formalism is valid only for the paramagnetic region and only for those regions in which $|\Delta\omega(\lambda,\mathbf{q})|/\omega(\lambda,\mathbf{q})\ll1$. Approximating the four-spin correlation functions by the sum of all possible factorizations in terms of lower-order correlation functions, we find from Eq. (40) of Ref. 4 that the (angular) frequency shift of an acoustic phonon due to its interacting with the spin system becomes

$$\Delta\omega(\lambda,\mathbf{q})=\frac{1}{2\omega(\lambda,\mathbf{q})MN}\sum_{\mathbf{k}}{}'\gamma_\lambda^2(\mathbf{k},\mathbf{q})\times P\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\int_{-\infty}^{+\infty}\frac{d\omega''}{\pi}\chi''(\mathbf{k},\omega')\chi''(\mathbf{k}-\mathbf{q},\omega'')\times\left[\frac{n(\omega')-n(\omega'')}{(\omega'-\omega''-\omega)}\right]. \quad (2)$$

The mass of the magnetic ions is M; the total number of lattice sites is N; the quantity P represents the principal value of the frequency integrals; the effective spin-phonon coupling function is $\gamma_\lambda^2(\mathbf{k},\mathbf{q})$; the spectral weight function for the longitudinal-pair-correlation function is $\chi''(\mathbf{k},\omega)$; and $n(\omega)=[e^{(\beta\hbar\omega)}-1]^{-1}$. The parameter β is the inverse temperature measured in energy units, i.e., $\beta=1/kT$, where k is Boltzmann's constant.

II. CALCULATION OF $\Delta\omega(\lambda,q)$

We restrict the calculations to the paramagnetic region $\beta\leq\beta_c$, where $(k\beta_c)$ is the inverse transition temperature. We consider the phonon wave vector to approach zero. The energy $\hbar\omega(\lambda,\mathbf{q})$ of the unperturbed phonon is $\hbar c(\lambda)q$, and the condition $\beta_c\hbar c(\lambda)q\ll1$ is valid for sufficiently small q. The low-frequency behavior of the spectral weight functions contributes most significantly to the double-frequency integrations.[4,5] We may verify this by observing that the factor $(\omega'-\omega''-\omega)^{-1}$ is largest for values of ω' near $\omega''+\omega$ and that the density-of-states factor for absorption and emission of the eigenmodes associated with the spin system $n(\omega''+\omega)-n(\omega'')$ has the limit $-\beta\hbar\omega[n(\omega'')]^2e^{(\beta\hbar\omega'')}$ for small frequencies ω. Following Ref. 6, we use the low-frequency form of the spectral weight function

$$\chi''(\mathbf{q},\omega)=\chi(\mathbf{q},0)\omega\Gamma(\mathbf{q},0)/[\omega^2+\Gamma^2(\mathbf{q},0)]. \quad (3)$$

The static susceptibility $\chi(\mathbf{q},0)$ satisfies the sum rule

$$\chi(\mathbf{q},0)=\pi^{-1}\int_{-\infty}^{+\infty}d\omega\omega^{-1}\chi''(\mathbf{q},\omega), \quad (4)$$

and $\Gamma(\mathbf{q},\omega=0)$ is the static diffusivity.[6]

We insert Eq. (3) into Eq. (2). Because the low-frequency behavior contributes most significantly, we approximate $n(\omega')-n(\omega'')$ by the expression $(\omega''-\omega')/\beta\hbar\omega'\omega''$. This is valid for $\beta\hbar\omega'\ll1$ and for $\beta\hbar\omega''\ll1$. We then obtain the expression for the angular frequency

[4] H. S. Bennett and E. Pytte, Phys. Rev. **155**, 553 (1967).

[5] E. Pytte and H. S. Bennett, Phys. Rev. **164**, 712 (1967).

[6] H. S. Bennett, Phys. Rev. **174**, 629 (1968).

shift, namely,

$$\Delta\omega(\lambda,\mathbf{q})=-\frac{1}{2M\beta\hbar\omega(\lambda,\mathbf{q})N}\sum_{\mathbf{k}}{}'\gamma_\lambda^2(\mathbf{k},\mathbf{q})\times\chi(\mathbf{k},0)\chi(\mathbf{k}-\mathbf{q},0)\,, \quad (5)$$

where $\omega(\lambda,\mathbf{q})=c(\lambda)q$. We observe that the angular frequency shift does not depend explicitly upon the spin-diffusion coefficient

$$D=\lim_{\mathbf{q}\to 0} q^{-2}\Gamma(\mathbf{q},0)$$

for ferromagnets and

$$\Lambda=\lim_{\mathbf{q}\to \mathbf{K}_0} \Gamma(\mathbf{q},0)$$

for antiferromagnets. The vector $\mathbf{K}_0$ is one-half a reciprocal-lattice vector. We contrast this with the ultrasonic attenuation, which is inversely proportional to the respective diffusion coefficients.

Because only small wave-vector acoustic phonons propagate easily in a lattice, we shall evaluate the summation in Eq. (5) in the limit of small q. Acoustic waves typically have wave vectors $q\sim 10^{-5}\,q_0$, where q_0 is the Debye wave vector and is of the order of the inverse of the lattice constant d, $q_0\sim d^{-1}$. Using the modified random-phase approximation[6] (RPA) for the static susceptibility $\chi(\mathbf{k},0)$, we have for the ferromagnet

$$\chi_F(\mathbf{k},0)\approx\{F[a^2+F^{-1}i(\mathbf{k})]\}^{-1}\,, \quad (6)$$

and for the antiferromagnet

$$\chi_A(\mathbf{k},0)\approx\{A[b^2-A^{-1}i(\mathbf{k}-\mathbf{K}_0)]\}^{-1}\,, \quad (7)$$

where

$$a^2=(\chi F)^{-1},\quad \chi=\chi_F(0,0)\,,$$
$$b^2=(\chi' A)^{-1},\quad \chi'=\chi_A(\mathbf{K}_0,0)\,.$$

The ferromagnetic nearest-neighbor exchange integral is F and the antiferromagnetic nearest-neighbor exchange integral is $-A$. We denote the ferromagnetic static susceptibility by $\chi_F(\mathbf{k},0)$ and the antiferromagnetic static susceptibility by $\chi_A(\mathbf{k},0)$. The function $i(\mathbf{k})$ is $I(0)-I(\mathbf{k})$. The lattice transform of the exchange interaction $I(\mathbf{q})$ becomes for a simple cubic lattice with only nearest-neighbor exchange interactions,

$$I(\mathbf{q})=2J(\cos k_x d+\cos k_y d+\cos k_z d)\,,$$

where $J=F>0$ for the ferromagnets and $J=-A<0$ for the antiferromagnets.

TABLE I. The effective coupling S_λ^2 for the small wave-vector limit. 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. The quantity Q is the spatial gradient of the exchange integral, and G_{11} and G_{44} are the temperature-independent coupling constants for the point-ion magnetostrictive interaction. The equations to which we refer in this table are those of Ref. 4.

Interaction and mode	S_λ^2
Eq. (10) longitudinal	$6Q^2d^2$
Eq. (10) transverse	$[9(6\pi^2)^{4/3}/35]Q^2d^2\sin^2\theta\sin^2\varphi\cos^2\varphi$
Eq. (12) longitudinal	$\{3G_{11}^2[\cos^2\theta(\cos^2\theta-\sin^2\theta)+\sin^4\theta(\cos^4\varphi-\sin^2\varphi\cos^2\varphi+\sin^4\varphi)]+4G_{44}^2\sin^2\theta(\cos^2\theta+\sin^2\theta\cos^2\varphi\sin^2\varphi)\}$
Eq. (12) transverse	$\{9G_{11}^2\sin^2\theta\sin^2\varphi\cos^2\varphi+G_{44}^2[\cos^2\theta+\sin^2\theta(\cos^2\varphi-\sin^2\varphi)^2]\}$

TABLE II. Temperature dependence of the angular frequency shift predicted by Eq. (5). The temperature factor for the ferromagnet is $f_F(\lambda,\beta)$ and $F_0=[(\chi F)^2/\beta]$, $F_c=[(\chi F)^{1/2}/(8\pi\beta)]$, and $F_c{}^t=4/(15\beta)$. The equations to which we refer in this table are those of Ref. 4.

Interaction and mode	Temperature factor $f_F(\lambda,\beta)$	
	$\beta\to 0$	$\beta\to\beta_c$
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

When we evaluate the lattice summation in Eq. (5) for the ferromagnet, the dominant contribution arises from the small $\mathbf{k}$ values, and we may use the small wave-vector limit of $\chi_F(\mathbf{k},0)$. Since $\chi_A(\mathbf{k},0)$ for the antiferromagnet has the form (7), the dominant contribution to the lattice summation arises from those values of $\mathbf{k}$ near the point $\mathbf{k}=\mathbf{K}_0$. We write $\mathbf{k}'=\mathbf{k}-\mathbf{K}_0$ and displace the origin for the summation over the first Brillouin zone. We may use then the small $\mathbf{k}'=\mathbf{k}-\mathbf{K}_0$ limit of $\chi_A(\mathbf{k}'+\mathbf{K}_0,\,0)$. In order to perform the resulting lattice summations for a cubic lattice, we assume that the first Brillouin zone contains many points. This is equivalent to stating that N, the number of lattice sites, is extremely large. To effect a further simplification, we approximate the polyhedron for the first Brillouin zone by a sphere in wave-vector space, i.e.,

$$\lim_{N\to\infty} N^{-1}\sum_{\mathbf{k}}{}'(\cdots)\approx\frac{d^3}{(2\pi)^3}\int_0^{q_0}k^2dk\times\int_0^{2\pi}d\varphi\int_{-\pi}^{+\pi}\sin\theta\,d\theta(\cdots)\,,$$

where the identity $N^{-1}\sum_{\mathbf{k}}{}'=1$ determines

$$q_0=(6\pi^2)^{1/3}d^{-1}.$$

We find that

$$\Delta\omega(\lambda,\mathbf{q})=-[S_\lambda^2q/2Mc(\lambda)F^2]f_F(\lambda;\beta) \quad (8)$$

for the ferromagnets and

$$\Delta\omega(\lambda,\mathbf{q})=-[S_\lambda^2q/2Mc(\lambda)A^2]f_A(\lambda;\beta) \quad (9)$$

for the antiferromagnets. We tabulate the wave-vector-independent coupling coefficients S_λ^2 and the tempera-

TABLE III. Temperature dependence of the angular frequency shift predicted by Eq. (5). The temperature factor for the antiferromagnet is $f_A(\lambda,\beta)$, and $A_0=[(\chi'A)^2/\beta]$, $A_c=[(\chi'A)^{1/2}/8\pi\beta]$, and $A_c^t=4/(15\beta)$. The equations to which we refer in this table are those of Ref. 4.

Interaction and mode	Temperature factor $f_A(\lambda,\beta)$	
	$\beta\to 0$	$\beta\to\beta_c$
Eq. (10) longitudinal	A_0	A_c
Eq. (10) transverse	A_0	A_c^t
Eq. (12) longitudinal	A_0	A_c
Eq. (12) transverse	A_0	A_c

ture factors f_F and f_A in Tables I, II, and III, respectively.

III. RESULTS AND CONCLUSIONS

Expressions (8) and (9) are valid only for $(|\Delta\omega|/cq)\ll 1$. Because the decoupling procedure is not valid for values of $\epsilon\ll 10^{-2}$, the expressions (8) and (9) also become more suspect the closer we approach the critical temperature. We see from Tables II and III that the angular frequency shift in a ferromagnet behaves with temperature in a manner similar to that for the antiferromagnet. The static susceptibilities χ and χ' have within the context of static-scaling-law descriptions the same temperature dependence near T_c.

We know from Eq. (B4) of Ref. 4 that our theory overestimates the critical fluctuations because it predicts that the specific heats behave for β near β_c as the square roots of the static susceptibilities behave; namely,

$$C_v(F)\sim(\chi F)^{1/2} \quad\text{and}\quad C_v(A)\sim(\chi'A)^{1/2}.$$

Following the suggestions and results of the heuristic improvement for the ultrasonic attenuation coefficients,[7] we should not be surprised if replacing the temperature factors F_c and A_c by

$$F_c=[C_v(F)/8\pi\beta] \quad\text{and}\quad A_c=[C_v(A)/8\pi\beta]$$

gives better agreement with experiment.

In order to compare our results with experiment, we note that the velocity-shift ratio $\Delta c/c$ equals the angular-frequency-shift ratio $\Delta\omega/cq\sim\Delta c/c$. Our theory predicts that the velocity-shift ratio for longitudinal acoustic phonons in a magnetic insulator dominated by the volume magnetostrictive interaction has the form (when T is near T_c)

$$[\Delta\omega(\text{long.})/c(\text{long.})q]\sim-\omega^0\epsilon^{-\xi}, \qquad (10)$$

where ϵ is defined in Sec. I. The exponent ξ has the upper and lower bounds $0.66\geqslant\xi\geqslant 0$. The upper bound $\xi=0.66$ obtains from the scaling law result that $\chi F=\chi'A\sim\epsilon^{-4/3}$. The lower bound $\xi=0.0$ obtains from the heuristic suggestion in Ref. 7 that

$$(\chi F)^{1/2}\sim c_v(F)\sim\ln\epsilon+a$$

and

$$(\chi'A)^{1/2}\sim c_v(A)\sim\ln\epsilon+b,$$

where a and b are constants and where $C_v(F)$ and $C_v(A)$ are the respective specific heats. The range $0.66\geqslant\xi\geqslant 0.0$ agrees qualitatively with the known experimental data.[1–3]

The ratio of the angular frequency shift to the damping coefficient Γ, $|\Delta\omega|/\Gamma$, gives us an additional expression containing the diffusion coefficients. We expect that this ratio will be influenced to a lesser extent by the approximations used to treat the four spin-correlation functions, then either the frequency shift $\Delta\omega$ or the attenuation coefficient $\alpha=\Gamma/c$. For example, when β is near β_c and when we employ the longitudinal-volume-magnetostrictive interaction, we have

$$|\Delta\omega_F|/\Gamma_F=2D/(\chi F)qd^2c \qquad (11)$$

and

$$|\Delta\omega_A|/\Gamma_A=4\Lambda/qc. \qquad (12)$$

Expressions (11) and (12) are meaningful only when they are much less than unity.

We see from Eq. (12) that our theory states that the antiferromagnetic ratio $|\Delta\omega_A|/\Gamma_A$ depends upon temperature in the same manner that the diffusion coefficient Λ depends upon temperature. This suggests either a test of the validity of the theory if both Λ and $|\Delta\omega_A|/\Gamma_A$ are known independently, or a way to determine the temperature dependence of Λ near T_c from a knowledge of the ratio $|\Delta\omega_A|/\Gamma_A$.

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[7] H. S. Bennett, Phys. Rev. 181, 978 (1969).