Ultrasonic Attenuation in the Heisenberg Paramagnet. II. Antiferromagnets

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The propagation of sound waves in antiferromagnetic insulators is studied within the framework of two models which describe the interaction between the spin system and the lattice. In particular, expressions for the ultrasonic attenuation coefficient near the Néel 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, to increase rapidly in the vicinity of the Néel point, and to be less singular than the attenuation coefficient for ferromagnetic insulators.

I. INTRODUCTION

In this paper we extend the theory described in a previous publication[1] on ultrasonic attenuation in ferromagnetic isulators to antiferromagnetic insulators. In Ref. 1, we introduced two different interactions between the lattice and the Heisenberg spin system (a volume magnetostrictive interaction and a general single-ion magnetostrictive interaction) and then computed the ultrasonic attenuation coefficient at high temperatures and near the Curie point. The formal apsects for the paramagnetic state of the Heisenberg ferromagnet and of the Heisenberg antiferromagnet have much in common, and we briefly report here the procedure, assumptions, and results predicted by the theory of Ref. 1 for the antiferromagnet. We find that our antiferromagnetic results differ from our ferromagnetic results and from previous work on the antiferromagnet[2,3] and agree qualitatively with the recent experimental work on the antiferromagnetic insulator MnF_2.[4] In addition, the results of our theory agree with the observed fact that the attenuation coefficient for the ferromagnet near the critical temperature at constant sound frequency, $\omega=cq$, is more singular than that for the antiferromagnet.[4,5]

In the present discussion, we find that the antiferromagnetic attenuation coefficient is proportional to the phonon frequency squared and increases rapidly in the vicinity of the Néel point for both types of interactions. Papoular[3] obtains the result that the attenuation is linear in the phonon frequency. However, he considers only the absorption of phonons by the spin system and neglects the fact that the spin system may also emit phonons. Tani and Mori[2] use a volume magnetostrictive interaction and predict that the attenuation coefficient varies as the phonon frequency squared. This result agrees with the present calculation However, they find the temperature dependence of the attenuation coefficient to be the same for both the ferromagnet and the antiferromagnet and this disagrees with the above experiments[4,5] and the conclusions of this paper.

II. FORMALISM

We shall compute that portion of the ultrasonic attenuation which arises from the coupling between the spin system and the lattice. We assume that as a result of the critical fluctuations of the spin system the spin phonon interaction provides the dominant attenuating mechanism near the transition region. Other mechanisms will be present and will compete with the above for temperatures sufficiently far from the transition region.

We may use most of the formalism of Sec. II of Ref. 1 to calculate the ultrasonic attenuation coefficient for the antiferromagnetic insulator in the paramagnetic state. The major change is that the exchange interaction $J(\mathbf{a}-\mathbf{a}')$ between nearest-neighbor spins at sites $\mathbf{a}$ and $\mathbf{a}'$ dominates over all other more distant pairings and is negative. Thus, combining Eqs. (31), (32), and (41) of Ref. 1, we obtain the attenuation coefficient for a phonon having wave vector $\mathbf{q}$, polarization $\mathbf{e}(\lambda, \mathbf{q})$, and frequency $\omega(\lambda, \mathbf{q})=c(\lambda)\,|\,\mathbf{q}\,|$; namely,

$$\alpha(\lambda,\mathbf{q})=-[2M\,c(\lambda)\omega]^{-1}N^{-1}\sum_{\mathbf{k}}{}'\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})\times\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\chi''(\mathbf{k},\omega')\chi''(\mathbf{k}-\mathbf{q},\omega'-\omega)\times\{n(\omega')-n(\omega'-\omega)\}. \quad (1)$$

Here, $\gamma_\lambda{}^2(\mathbf{k},\mathbf{q})$ is an effective magnetoelastic coupling function, $\omega=\omega(\lambda,\mathbf{q})$, M is the ion mass, $n(\omega)=[\exp(\beta\hbar\omega)-1]^{-1}$, and $\chi''(\mathbf{q},\omega)$ is the spectral weight function for the spin pair correlation functons, i.e.,

$$2\hbar\chi''(\mathbf{k},\omega)=\sum_{\mathbf{a}}\int_{-\infty}^{+\infty}dt\exp(-i\omega t+i\mathbf{k}\cdot\mathbf{a})\times\langle[S_z(\mathbf{a},t),\,S_z(0,0)]\rangle.$$

[1] H. S. Bennett and E. Pytte, Phys. Rev. **155**, 553 (1967).
[2] K. Tani and H. Mori, Phys. Letters **19**, 627 (1966).
[3] M. Papoular, Compt. Rend. **258**, 5598 (1964).
[4] R. W. Moss (private communication).
[5] B. Lüthi, presented at the 1967 International Conference on Magnetism, Boston (unpublished).

We shall evaluate the integral in Eq. (1) for the region $\beta\hbar\omega \ll 1$ and $\beta \lesssim \beta_N$ where β_N is the inverse Néel temperature expressed in energy units, $\beta_N = (1/kT_N)$. The condition $\beta_N\hbar\omega \ll 1$ is valid for the frequencies employed in most experiments, i.e.,

$$\omega \ll (1.38/1.05)\, T_N \times 10^{11} (\text{deg sec})^{-1}. \quad (2)$$

Because the density-of-states factor in Eq. (1) has the limit

$$n(\omega') - n(\omega' - \omega) \approx -\beta\hbar\omega [n(\omega')]^2 \exp(\beta\hbar\omega')$$

for $\beta\hbar\omega \ll 1$, the low-frequency behavior of the spectral weight function contributes most significantly to the frequency integral. We therefore use the low-frequency representation of the spectral weight function,[6-8]

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

where $\chi(\mathbf{q}, 0)$ is the wave-vector-dependent susceptibility,

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

The function $\Gamma(\mathbf{q}, \omega)$ is a real function and is identical to the function $q^2 D(\mathbf{q}, \omega) \equiv \Gamma(\mathbf{q}, \omega)$ discussed in Ref. 6. We may interpret $D(\mathbf{q}, \omega)$ to be a generalized spectral diffusion function. The spectral form (3) implies that a region in $\mathbf{q}$ space exists such that for sufficiently low frequencies the system exhibits a hydrodynamic limit (attains local thermodynamic equilibrium). The connection between microscopic theory and the hydrodynamic domain is contained in the functions $\Gamma(\mathbf{q}, \omega)$ and $\chi(\mathbf{q}, 0)$. We show in Appendix A that a necessary condition for the validity of Eq. (3) for a given wave vector $\mathbf{q}$ is

$$1 \gg \left| \lim_{\omega \to 0} P \int_{-\infty}^{+\infty} \frac{d\omega'}{\pi} \frac{\Gamma(\mathbf{q}, \omega')}{(\omega'^2 - \omega^2)} \right|. \quad (5)$$

If one fails to demonstrate that inequality (5) is satisfied, then one properly should consider form (3) as a phenomenological construct.

As we do in Ref. 1, we use the effective field random-phase approximation results for the $\mathbf{q}$ dependent susceptibility. The random-phase approximation (RPA) susceptibility $\chi(\mathbf{q}, 0)$ for the antiferromagnet is

$$\chi(\mathbf{q}, 0; \text{RPA}) = \frac{\chi(\mathbf{K}_0, 0)}{1 + \chi(\mathbf{K}_0, 0)\{I(0) + I(\mathbf{q})\}}, \quad (6)$$

where $I(\mathbf{q})$ is the lattice transform of the exchange interaction

$$I(\mathbf{q}) = \sum_{\mathbf{a}} \exp(i\mathbf{q}\cdot\mathbf{a})\, J(\mathbf{a}). \quad (7)$$

The wave vector $\mathbf{K}_0$ is that value of $\mathbf{q}$ for which $I(\mathbf{q})$ has its maximum value. For a simple cubic lattice with nearest-neighbor interactions only, we have $\mathbf{K}_0 = (\pi/d)(1, 1, 1)$, and $I(\mathbf{K}_0) = zI$, where d is the lattice constant, z is the number of nearest neighbors, and I is the magnitude of the exchange integral. We also have the relation $I(\mathbf{q} + \mathbf{K}_0) = -I(\mathbf{q})$. The function $\chi(\mathbf{K}_0, 0) \equiv \chi'(\beta)$ for the antiferromagnet behaves in the same manner as the static susceptibility $\chi(0, 0) \equiv \chi(\beta)$ does for the ferromagnet. They both become infinite at the critical temperature; e.g.,

$$\chi' I \approx A(\beta_N/\beta_N - \beta)^{\gamma}, \quad (8)$$

where γ is the same for both antiferromagnets and ferromagnets according to present theories.

Inserting the low-frequency spectral function (3) into Eq. (1) produces the result

$$\alpha(\lambda, \mathbf{q}) \approx -[2\beta Mc(\lambda) N]^{-1} \sum_{\mathbf{k}}{}' \gamma_\lambda^2(\mathbf{k}, \mathbf{q})\, \chi(\mathbf{k}, 0) \times \chi(\mathbf{k} - \mathbf{q}, 0) \frac{(\Gamma_2 + \Gamma_1)\{\omega^2 + (\Gamma_2 - \Gamma_1)^2\}}{\{\omega^2 + \Gamma_2^2 - \Gamma_1^2\}^2 + 4\Gamma_1^2\omega^2}, \quad (9)$$

where $\Gamma_1 \equiv \Gamma(\mathbf{k}, 0)$ and $\Gamma_2 \equiv \Gamma(\mathbf{k} - \mathbf{q}, 0)$.

TABLE I. The small wave-vector limit ($\mathbf{k} \to 0$) of the effective coupling $\gamma_\lambda^2(\mathbf{k} - \mathbf{K}_0, \mathbf{q})$. The direction of propagation is $\mathbf{q} = q \times (\sin\theta\cos\varphi\hat{x} + \sin\theta\sin\varphi\hat{y} + \cos\theta\hat{z})$, 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	$\lim_{k\to 0, q\to 0} \gamma_\lambda^2(\mathbf{k} - \mathbf{K}_0, \mathbf{q})$
Equation (10)[a]	$6Q^2d^2q^2$
Longitudinal	
Equation (10)[a]	$\frac{3}{2}Q^2d^6q^2 \sin^2\theta \sin^2\varphi \cos^2\varphi (k_x^2 - k_y^2)^2$
Transverse	
Equation (12)[a]	$[3G_{11}^2 \cos^2\theta\, (\cos^2\theta - \sin^2\theta)$
Longitudinal	$+ \sin^4\theta\, (\cos^4\varphi - \sin^2\varphi\cos^2\varphi + \sin^4\varphi)$
	$+ 4G_{44}^2 \sin^2\theta\, (\cos^2\theta$
	$+ \sin^2\theta \cos^2\varphi \sin^2\varphi)]q^2$
Equation (12)[a]	$[9G_{11}^2 \sin^2\theta \sin^2\varphi \cos^2\varphi$
Transverse	$+ G_{44}^2 \cos^2\theta + \sin^2\theta\, (\cos^2\varphi - \sin^2\varphi)^2]q^2$

[a] The equations to which we refer in this table are those of Ref. 1.

III. RESULTS AND CONCLUSIONS

Because only small wave-vector acoustic phonons propagate easily in a lattice, we shall evaluate the summation in Eq. (9) in the limit of small $\mathbf{q}$. Acoustic waves typically have wave vectors $q \sim 10^{-5} q_0$, where q_0 is the Debye wave vector $q_0 \sim d^{-1}$. When we evaluate the k summation in Eq. (9) for the ferromagnet, the dominant contribution arises from small $\mathbf{k}$ values and we may use the small wave-vector limit of the spectral

[6] H. S. Bennett and P. C. Martin, Phys. Rev. **138**, A608 (1959).
[7] T. Moriya, Progr. Theoret. Phys. (Kyoto) **28**, 371 (1962).
[8] P. G. De Gennes and J. Villain, J. Phys. Chem. Solids **13**, 10 (1960).

TABLE II. Temperature dependence of the attenuation predicted by Eq. (9). The temperature factors are $A_0=[(\chi' I)^2/\beta\Lambda]$, $A_N=[(\chi' I)^{1/2}/\beta\Lambda]$, and $A_N{}^t=(1/\beta\Lambda)$. The respective attenuation coefficients are directly proportional to these temperature factors.

Interaction and mode	Temperature factor $\beta\to 0$	Temperature factor $\beta\to\beta_N$
Equation (10)[a] Longitudinal	A_0	A_N
Equation (10)[a] Transverse	A_0	$A_N{}^t$
Equation (12)[a] Longitudinal	A_0	A_N
Equation (12)[a] Transverse	A_0	A_N

[a] The equations to which we refer in this table are those of Ref. 1.

function (3). The small $\mathbf{q}$ limit of the function $\Gamma(\mathbf{q},\omega)$ has the form[6,7,9]

$$\Gamma(\mathbf{q},0)\underset{q\to 0}{\longrightarrow}Dq^2, \tag{10}$$

and this corresponds to a diffusion equation description for the magnetization. The quantity D is the spin diffusion coefficient. But, because $\chi(\mathbf{q},0)$ for the antiferromagnet has the form (6), which differs from that for the ferromagnet, the dominant contribution to the $\mathbf{k}$ summation in Eq. (9) arises from those values of $\mathbf{k}$ near the point $\mathbf{k}=\mathbf{K}_0$. We therefore displace the origin for the summation over the first Brillouin zone by $\mathbf{K}_0$ and expand the integrand in powers of $\mathbf{k}'=\mathbf{k}-\mathbf{K}_0$, about $\mathbf{k}=\mathbf{K}_0$.

Current theories[7,8] predict that as $\mathbf{k}$ approaches $\mathbf{K}_0$ the function $\Gamma(\mathbf{k},0)$ approaches a temperature-dependent coefficient which is wave-number-independent, i.e.,

$$\Gamma(\mathbf{k},0)\underset{\mathbf{k}\to\mathbf{K}_0}{\longrightarrow}\Lambda(\beta)+(\mathbf{k}-\mathbf{K}_0)^2\theta(\beta). \tag{11}$$

and that both the spin diffusion coefficient $D(\beta)$ [6,7] for the ferromagnet and the function $\Lambda(\beta)$ [7] for the antiferromagnet are zero at the critical point. The behavior of $\Lambda(\beta)$ for β near β_N depends on the method of calculation. Moriya's theory gives us the form

$$\Lambda(\beta)\underset{\beta\to\beta_N}{\longrightarrow}B(S)\,(I/\hbar)[\chi' I]^{-1}, \tag{12}$$

where $B(S)$ is a function of the spin quantum number S and is of the order of unity. However, we emphasize that all determinations of the $D(\beta)$ and $\Lambda(\beta)$ near the critical point are as yet very unreliable and that even nonzero values for D and Λ have not been definitely ruled out.

We evaluate Eq. (9) for a simple cubic lattice and for a longitudinal mode propagating along one of the crystal axes. When β is near β_N, the volume magnetostriction [Eq. (10) of Ref. 1] yields

$$\alpha_A(\mathbf{q})\approx[3Q^2d^2(\chi' I)^{1/2}/32\pi\beta I^2 Mc\Lambda]q^2, \tag{13}$$

and the single-ion magnetostriction [Eq. (12) of Ref. 1] yields the same relation as Eq. (13). We refer the reader to Ref. 1 for the definitions of these coupling constants.

Continuing our discussion, we compare expression (13) with the corresponding expression for the ferromagnet [Eqs. (63), (64), and (65) of Ref. 1],

$$\alpha_F(\mathbf{q})\approx[6Q^2d^4(\chi J)^{3/2}/32\pi\beta J^2 McD]q^2. \tag{14}$$

The above theories[7,8] predict that Λ and D have the same temperature dependence near the critical point. We conclude from this that ultrasomic attenuation in an antiferromagnet is less singular than that for a ferromagnet.

However, for temperatures sufficiently close to the critical point, the approximation involved in factorizing the four-spin correlation function becomes invalid and one may argue that the behavior of the attenuation coefficient predicted by Eq. (13) for the antiferromagnet and by Eq. (14) for the ferromagnet are in both cases too singular. Using the above approximations to treat the four-spin correlation function and the two-spin correlation function, we find that the specific heat varies respectively as $(\chi' I)^{1/2}$ and $(\chi J)^{1/2}$ for the antiferromagnet and ferromagnet near the critical point. If one believes that the specific heat should have a logarithmic singularity, then our approximations overestimate the critical fluctuations. Even though Eqs. (13) and (14) predict attenuation coefficients which may be too singular, we still expect $\alpha(\mathbf{q})$ to be more singular than the specific heat and to be of the form

$$\alpha\approx C[\beta_c/(\beta_c-\beta)]^\gamma$$

for $\beta\leq\beta_c$ and where γ for the antiferromagnet is smaller than it is for the ferromagnet.

Because the propagating phonons have very small q values and since we consider here only the attenuation due to the critical fluctuations, we expect that the attenuation should be larger in the ferromagnet for which the critical fluctuations occur for $\mathbf{q}\sim 0$ than in the antiferromagnet for which they occur for $\mathbf{q}\approx\mathbf{K}_0$. We also note that the expression for the ultrasonic attenuation in the antiferromagnet, Eq. (13), is more singular than the specific heat calculated in the same approximation, only to the extent that $(1/\Lambda)$ may be singular.

Finally, in Tables I and II we present the results of different configurations for both longitudinal and transverse sound waves.

APPENDIX A: SPECTRAL REPRESENTATIONS AND THE HYDRODYNAMIC LIMIT

We present in this appendix a few remarks on the spectral representation and the hydrodynamic limit (the limit in which all properties and excitation modes

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

of the system vary sufficiently slowly in time and space to insure local thermodynamic equilibrium).

Because $\omega\chi''(\mathbf{q},\omega)$ is an even function of ω and positive, we may construct the spectral representation [Eq. (2) of Ref. 7] for the longitudinal spin correlations. When $\beta<\beta_c$ and when no external fields are present,

$$\chi''(\mathbf{q},\omega)=\chi(\mathbf{q},0)\omega\Gamma(\mathbf{q},\omega)\Big/\left[\omega^2\left\{1-P\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\frac{\Gamma(\mathbf{q},\omega)}{\omega'^2-\omega^2}\right\}^2+\Gamma^2(\mathbf{q},\omega)\right], \tag{A1}$$

where we have suppressed the zz subscripts on χ'' and where P indicates a principal value integral. Representation (A1) serves merely to transfer our ignorance about the singular function $\chi''(\mathbf{q},\omega)$ to a hopefully better behaved function $\Gamma(\mathbf{q},\omega)$. We emphasize that representation (A1) is by no means a unique representation and that even for the paramagnetic state it may not be the appropriate representation in which to treat paramagnetic collective modes, if the latter should occur.

One condition for the existance of a hydrodynamic domain is that a sufficient number of interactions must occur in order to have local thermodynamic equilibrium. This means we want to examine the low-frequency behavior (i.e., the behavior for periods long compared to $\hbar/I$ or $\hbar/J$). The low-frequency representation, Eq. (3), obtains whenever

$$1\gg\left|\lim_{\omega\to 0}P\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\frac{\Gamma(\mathbf{q},\omega')}{\omega'^2-\omega^2}\right|. \tag{A2}$$

The fact that $\chi(\mathbf{q},0;\text{RPA})$ for the ferromagnet attains its greatest value at $\mathbf{q}=0$,

$$\chi_F(\mathbf{q},0;\text{RPA})=\chi[1+\chi\{J(0)-J(\mathbf{q})\}]^{-1}, \tag{A3}$$

suggests naturally to us the identity $\Gamma(\mathbf{q},\omega)=D(\mathbf{q},\omega)q^2$ for dominant diffusion mode. Condition (A2) then requires that for sufficiently small $\mathbf{q}$ (large distances) the principal value integral,

$$\lim_{\omega\to 0}\left|P\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\frac{D(\mathbf{q},\omega')}{\omega'^2-\omega^2}\right|,$$

must be finite. For the antiferromagnet, Eq. (6) attains its greatest value at $\mathbf{q}=\mathbf{K}_0$ and the low-frequency form (3) obtains whenever inequality (5) is satisfied. In order to explain this difference, we recall that the ferromagnetic Hamiltonian strives to have the direction of nearest-neighbor spins parallel (i.e., strives to maintain a smooth variation from one lattice site to the next), while the antiferromagnetic Hamiltonian strives to have a given spin point in the opposite direction to its nearest neighbors. This means that such quantities as the antiferromagnet's magnetization must be multiplied by an appropriate phase factor $\exp(i\mathbf{K}_0\cdot\mathbf{r})$ before they will appear to vary smoothly as a function of $\mathbf{r}$.

We have been examining the paramagnetic low-frequency (long-time) behavior of $\chi''(\mathbf{q},\omega)$. We now do not make explicitly any statements about the frequency ω and consider the wave-vector dependence of $\chi''(\mathbf{q},\omega)$. For simplicity, we shall discuss the ferromagnet. The application to the antiferromagnet follows the same procedure as contained in the discussion following Eq. (10) and in the first part of this appendix [i.e., replace $\mathbf{q}$ with the $(\mathbf{q}-\mathbf{K}_0)$].

We describe the spatial extent of the pair correlation functions, $\langle S_z(\mathbf{a},t)\,S_z(\mathbf{a}',t')\rangle$, for temperatures above the critical point by the correlation length c. A modified RPA method predicts that when $t=t'$, the correlation length behaves as $(1+\chi J)^{1/2}d$ for the ferromagnet. The spatial extent of any local magnetization is also of the order of c.

Brout[10] suggests that collective modes (e.g., spin waves) may exist above the critical point. If any collective modes do exist, then we expect them to be associated with the local magnetization and to have a spatial extent which is of the order of c. Those wave vectors $\mathbf{k}$ for which collective modes may exist should satisfy the inequality

$$kc\gg 1.$$

Because the form (3) does not admit the possibility of high-frequency paramagnetic collective modes, it is not compatible with the existance of such modes. We therefore expect at best that if a microscopic theory were to yield $\chi''(\mathbf{k},\omega)$ for all $\mathbf{k}$ and ω, then the form (A1) would obtain in some asymptotic sense only for the region $kc\ll 1$ and $\omega\tau\ll 1$ for the ferromagnet and for the region $|\mathbf{k}-\mathbf{K}_0|\,c\ll 1$ and $\omega\tau\ll 1$ for the antiferromagnet.

[10] R. Brout, Phys. Letters **24A**, 117 (1967).