Anomalous Ultrasonic Attenuation above the Magnetic Critical Point*†

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The mode-mode coupling theory of Kadanoff and Swift is used to describe the rise in the ultrasonic attenuation as a material approaches its magnetic transition. Near the transition, dynamic fluctuations in the magnetic order cause increased absorption of the sound. Predictions are made of the increased attenuation in various models of magnetic systems. A comparison with experiment is carried out.

I. INTRODUCTION

IN several experiments[1–4] involving the propagation of sound waves through magnetic materials near their critical point, an anomaly was observed in the attenuation coefficient. For longitudinal sound waves above the magnetic transition, this anomaly was observed to have the form

$$\Delta\alpha_l(q,\epsilon)=Bq^2\epsilon^{-y}, \tag{1}$$

where B is a constant, q is the wave vector of the sound wave, $\epsilon\equiv(T-T_c)/T_c$, and the critical index y was found to vary quite widely from material to material. It was found to vary from zero for EuO (no divergence as $T\rightarrow T_c$) to 1.37 for Dy.

Many attempts have been made to explain this phenomenon theoretically, but these have only met with limited success. These calculations have generally either been based on the continued-fraction method of Mori[5–7] or on a Green's-function approach.[8] The principal source of difficulty has been the determination of the temperature dependence of the four-spin correlation function that arises during the calculation. The employment of a factorization approximation for this four-spin correlation function apparently overestimates the effect of correlations and thus leads to too large a value for y. In this work we estimate the four-spin correlation function by the use of the mode-mode coupling theory of Kadanoff and Swift.[9] We obtain qualitative agreement with a calculation of Kawasaki[10] in which the four-spin correlation function is estimated by the continued-fraction method. In Kawasaki's work, only the isotropic Heisenberg ferromagnet and antiferromagnet are considered, but his calculation could be extended to the systems we discuss here. His work gives only the q^2 dependence and the exponent y in Eq. (1); it does not provide for an estimate of the coefficient B.

The important elements in the Kadanoff-Swift theory are the operators that correspond to conserved quantities and to other quantities that exhibit a "slowing down" as the critical point is approached. These are the number density operator $n_{\rm op}(\mathbf{r})$, the energy density operator $\mathcal{E}_{\rm op}(\mathbf{r})$, and the momentum density operator $\mathbf{g}(\mathbf{r})$, which were used in Ref. 9. In addition to these, in either an isotropic system or a uniaxial system we also have a spin density operator $\sigma_{\rm op}(\mathbf{r})$, which corresponds to the local magnetization density. In a ferromagnetic system, $\sigma_{\rm op}(\mathbf{r})$ corresponds to the magnetic order parameter and exhibits critical behavior near the magnetic transition. For an antiferromagnetic system the order parameter is the sublattice magnetization which is proportional to the operator $\sigma_{\rm op}{}^s(\mathbf{r})$. Although $\sigma_{\rm op}{}^s(\mathbf{r})$ does not correspond to a conserved quantity, it exhibits critical behavior near T_c, and it will turn out to be the important operator in an antiferromagnetic system. For convenience in what follows, we shall refer to the set of conserved quantities and those exhibiting a critical slowing down as "conserved."

According to the theory, the most important modes damping the sound are in the hydrodynamic regime. The modes are then described by local equilibrium states in which the equilibrium parameters of the system are slowly varying functions of space and time. These states are formed from linear combinations of the operators corresponding to "conserved" quantities acting upon a typical equilibrium state $|\,\rangle$. We use these operators in the form of their Fourier transforms

$$a_i(\mathbf{q})=\int(dr)e^{-i\mathbf{q}\cdot\mathbf{r}}a_i(\mathbf{r}), \tag{2}$$

where the $a_i(\mathbf{r})$ represent the density of the "conserved" quantities. These operators are chosen such that the local equilibrium states

$$|\,i,\mathbf{q}\rangle=a_i(\mathbf{q})|\,\rangle, \tag{3a}$$

$$\langle i,\mathbf{q}\,|=\langle\,|\,a_i(-\mathbf{q}) \tag{3b}$$

are orthonormal, i.e.,

$$\langle i,\mathbf{q}\,|\,j,\mathbf{q}'\rangle=\delta_{ij}(2\pi)^3\delta(\mathbf{q}-\mathbf{q}'). \tag{4}$$

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[10] K. Kawasaki, Phys. Letters **26A**, 543 (1968).

Following Ref. 9, we have the following linearly independent operators:

$$a_i(q)=\{[k_B\rho C_p(\mathbf{q})]^{1/2}T(q)\}^{-1}\times[\mathcal{E}_{\rm op}(\mathbf{q})-(\langle\mathcal{E}+p\rangle/\langle n\rangle)n_{\rm op}(\mathbf{q})]\equiv S_{\rm op}(\mathbf{q})/[k_B\rho C_p(\mathbf{q})]^{1/2}, \quad (5a)$$

$$a_2(\mathbf{q})=\frac{(\rho\beta)^{1/2}}{\langle n\rangle}c(\mathbf{q})n_{\rm op}(\mathbf{q})+\left[\frac{1}{k_B\rho}\left(\frac{1}{C_V(\mathbf{q})}-\frac{1}{C_p(\mathbf{q})}\right)\right]^{1/2}S_{\rm op}(\mathbf{q}), \quad (5b)$$

$$a_3(\mathbf{q})=(\beta/\rho)^{1/2}g_{\rm op}{}^x(\mathbf{q}), \quad (5c)$$

$$a_4(\mathbf{q})=(\beta/\rho)^{1/2}g_{\rm op}{}^y(\mathbf{q}), \quad (5d)$$

$$a_5(\mathbf{q})=(\beta/\rho)^{1/2}g_{\rm op}{}^z(\mathbf{q}), \quad (5e)$$

where ρ is the density of the material, $\beta\equiv 1/k_BT$, C_p and C_V are the specific heats at constant pressure and volume, $S_{\rm op}$ is the entropy density operator, and $c(\mathbf{q})$ is the longitudinal sound velocity. In addition to these we have for the ferromagnetic system

$$a_6(\mathbf{q})=\sigma_{\rm op}(\mathbf{q})/[\chi(\mathbf{q})/\beta\mu_0{}^2]^{1/2}, \quad (5f)$$

where $\mu_0\sigma(\mathbf{r})$ is the magnetic moment associated with $\sigma(\mathbf{r})$, and $\chi(\mathbf{q})$ is the qth Fourier component of the susceptibility $\chi(\mathbf{r}-\mathbf{r}')$ determined by the variation of $\sigma(\mathbf{r})$ due to the change in the applied field $H(\mathbf{r}')$ parallel to the direction of $\sigma(\mathbf{r})$.

For the antiferromagnetic system, the normalized operator corresponding to a fluctuation in the order parameter is

$$a_6{}^s(\mathbf{q})=\sigma_{\rm op}{}^s(\mathbf{q})/[\chi_s(\mathbf{q})/\beta\mu_0{}^2]^{1/2}, \quad (6)$$

where $\chi_s(\mathbf{q})$ is the qth Fourier component of the staggered susceptibility. For an antiferromagnet,

$$a_6{}^s(\mathbf{q})=a_6(\mathbf{q}+\mathbf{K}_0),$$

where $\mathbf{K}_0$ is the reciprocal-lattice vector that defines the superlattice structure. However, we shall find it convenient to use the special operator $a_6{}^s$ for an antiferromagnetic system. Note that only above T_c do the operators a_6 and $a_6{}^s$ correspond to fluctuations in the magnetic order parameter, and also only above T_c are they automatically orthogonal to a_1 and a_2.

In Sec. II we calculate the anomalous attenuation coefficient. We shall show that its temperature divergence is determined essentially by the relaxation rate of a fluctuation in the magnetic order parameter. In Sec. III we point out that this relaxation rate can be determined experimentally, directly from neutron scattering data or indirectly from the nuclear-magnetic-resonance (NMR) linewidth. In Sec. IV we give the calculated order-parameter relaxation rates for various model systems. This will give us predictions for the exponent y for these models. In Sec. V we discuss the experimental picture for various real materials and see how it compares with our calculations. Finally, in Sec. VI we summarize our results.

II. DERIVATION OF ANOMALOUS SOUND ATTENUATION COEFFICIENT

For a longitudinal sound wave directed along the x axis of the system and having a damping rate small compared to the sound-wave frequency, the sound attenuation coefficient is given by[9]

$$\alpha_l(\mathbf{q},s)=[8.69/2c(\mathbf{q})]\times\{\langle|a_2(-\mathbf{q})\mathfrak{L}P(s-P\mathfrak{L}P)^{-1}P\mathfrak{L}a_2(\mathbf{q})|\rangle+\langle|a_3(-\mathbf{q})\mathfrak{L}P(s-P\mathfrak{L}P)^{-1}P\mathfrak{L}a_3(\mathbf{q})|\rangle\}, \quad (7)$$

where we are to take the real part of the right-hand side of Eq. (7). In Eq. (7), $\mathfrak{L}$ is the Liouville operator for the system,

$$P=1-\sum_j a_j(\mathbf{q})|\rangle\langle|a_j(\mathbf{q})$$

is the projection operator which rejects the local equilibrium states, and the factor $8.69/c(q)$ is used to convert the attenuation coefficient to dB/cm in order to facilitate comparison with experiment. We next make use of the fact that

$$\mathfrak{L}|\rangle=\langle|\mathfrak{L}=0$$

to write the sound attenuation coefficient as

$$\alpha_l(\mathbf{q},s)=[8.69/2c(\mathbf{q})]\times\{\langle|[\mathfrak{L},a_2(-\mathbf{q})]P(P\mathfrak{L}P-s)^{-1}P[\mathfrak{L},a_2(\mathbf{q})]|\rangle+\langle|[\mathfrak{L},a_3(-\mathbf{q})]P(P\mathfrak{L}P-s)^{-1}\times P[\mathfrak{L},a_3(\mathbf{q})]|\rangle\}. \quad (8)$$

From the definitions of the conserved quantities[9] we have

$$[\mathfrak{L},a_2(\mathbf{q})]=iqj_2{}^x(\mathbf{q}), \quad (9a)$$

$$[\mathfrak{L},a_3(\mathbf{q})]=iq(\beta/\rho)^{1/2}\tau^{xx}(\mathbf{q}), \quad (9b)$$

where $\mathbf{j}_2$ is the current corresponding to the conserved quantity a_2, and τ^{ij} is the usual stress tensor. In the first term on the right-hand side of Eq. (8), the current in $j_2{}^x$ corresponding to particle flow is removed by the projection operators. This is because the particle flow is proportional to the momentum density. The first term is hence proportional to the thermal conductivity of the system. In particular, we find that

$$\langle|[\mathfrak{L},a_2(-\mathbf{q})]P(P\mathfrak{L}P-s)^{-1}P[\mathfrak{L},a_2(\mathbf{q})]|\rangle=q^2\lambda(\mathbf{q},s)\rho^{-1}\{[C_V(\mathbf{q})]^{-1}-[C_p(\mathbf{q})]^{-1}\}, \quad (10)$$

where λ is the thermal conductivity. Scaling-law arguments[11,12] show that the thermal conductivity is nondivergent at the magnetic critical point. If this is the case, then the quantity in Eq. (10) can have at best a constant part and hence could not contribute to a divergent sound attenuation coefficient. However,

[11] K. Kawasaki, Progr. Theoret. Phys. (Kyoto) **40**, 706 (1968).
[12] D. L. Huber, Solid State Commun. **6**, 685 (1968).

Wagner[13] claims that the part of the thermal conductivity corresponding to energy transport through the spin system, while nondivergent for the isotropic ferromagnet, is divergent for the isotropic antiferromagnet. In particular, he finds that $\lambda_s \sim \epsilon^{-1/3}$ for this system. If such is the case, then near T_c when C_p becomes significantly larger than C_V, the quantity in Eq. (10) would exhibit a divergence for the isotropic antiferromagnet. Wagner finds this result by the application of the dynamic scaling techniques used by Ferrell *et al.*[14] in their calculation of the thermal conductivity in liquid helium. We regard the direct application of this procedure to the antiferromagnet as questionable and also point out that experimental work[15–17] gives no indication of a divergent part to the thermal conductivity for antiferromagnetic systems.

We also have

$$\langle|[\mathcal{L}, a_3(-\mathbf{q})]P(P\mathcal{L}P-s)^{-1}P[\mathcal{L},a_3(\mathbf{q})]|\rangle = \beta q^2\rho^{-1}\langle|\tau^{xx}(-\mathbf{q})P(P\mathcal{L}P-s)^{-1}P\tau^{xx}(\mathbf{q})|\rangle. \quad (11)$$

It is possible in some systems for $\tau^{xx}(\mathbf{q})$ to be proportional to the energy density. In this case the action of the projection operators in Eq. (11) would be to give a result of zero. For such a system there would be no anomalous sound attenuation as $T \to T_c^+$. In any event the projection operator will eliminate the part of $\tau^{xx}(\mathbf{q})$ that is proportional to the energy density. We will not take this into account at the present, but will discuss it in more detail in Sec. V.

We can represent

$$X \equiv (P\mathcal{L}P-s)^{-1}$$

in terms of states involving multiple transport processes. From Ref. 9 the part of X with wave vector $\mathbf{q}$ is given by

$$X_{\mathbf{q}} = \frac{1}{2!}\sum_{\nu,\nu'}\int\frac{d\mathbf{q}'}{(2\pi)^3} \times\frac{a_\nu(\mathbf{q}')a_{\nu'}(\mathbf{q}-\mathbf{q}')|\rangle\langle|a_\nu(-\mathbf{q}')a_{\nu'}(\mathbf{q}'-\mathbf{q})}{s_\nu(\mathbf{q}')+s_{\nu'}(\mathbf{q}-\mathbf{q}')-s} + \frac{1}{3!}\sum_{\nu,\nu',\nu''}\int\frac{(d\mathbf{q}')(d\mathbf{q}'')}{(2\pi)^6} \times\frac{a_{\nu'}(\mathbf{q}')a_{\nu''}(\mathbf{q}'')a_\nu(\mathbf{q}-\mathbf{q}'-\mathbf{q}'')|\rangle}{s_\nu(\mathbf{q}-\mathbf{q}'-\mathbf{q}'')+s_{\nu'}(\mathbf{q}')+s_{\nu''}(\mathbf{q}'')-s} \times\langle|a_{\nu'}(-\mathbf{q}')a_{\nu''}(-\mathbf{q}'')a_\nu(\mathbf{q}'+\mathbf{q}''-\mathbf{q})+\cdots, \quad (12)$$

where $s_\nu(\mathbf{q})$ is the eigenvalue of the state $|\nu,\mathbf{q}\rangle$ [i.e., $\mathcal{L}|\nu,\mathbf{q}\rangle = s_\nu(\mathbf{q})|\nu,\mathbf{q}\rangle$]. In what follows we shall ignore the terms in $X_{\mathbf{q}}$ involving more than two transport processes.

[13] H. Wagner (unpublished report).
[14] R. A. Ferrell, N. Menyhard, H. Schmidt, F. Schwabl, and P. Szepfaluzy, Phys. Rev. Letters **18**, 891 (1967).
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[17] R. H. Donaldson and D. T. Edmonds, Phys. Letters **2**, 130 (1962).

Most of the possible intermediate states will contribute to the "background" attenuation. We shall concern ourselves only with those states that contribute a diverging term as the magnetic critical point is approached. The dominant term leading to the anomalous sound attenuation as $T \to T_c^+$ arises from the intermediate state involving two fluctuations in the magnetic order parameter. Physically, this corresponds to the absorption of a phonon of wave vector $\mathbf{q}$ and the creation of two "spin fluctuations." This gives us

$$\Delta\alpha_l(\mathbf{q},s) = \frac{2.17q^2}{c(\mathbf{q})\rho k_B T} \times\int\frac{d\mathbf{q}'}{(2\pi)^3}\frac{|\langle|\tau^{xx}(-\mathbf{q})Pa_6(\mathbf{q}')a_6(\mathbf{q}-\mathbf{q}')|\rangle|^2}{s_\sigma(\mathbf{q}')+s_\sigma(\mathbf{q}-\mathbf{q}')-s} \quad (13)$$

for a ferromagnetic system. For an antiferromagnetic system the a_6 operators would be replaced by a_6^s operators and s_σ would be interpreted as the relaxation rate of a fluctuation in the magnetic order parameter σ^s.

We now turn to estimating the various quantities on the right-hand side of Eq. (13). We shall evaluate Eq. (13) in the region where the sound-wave frequency $|s|$ is much smaller than the frequency associated with a typical spin fluctuation. We also take $q \ll \xi^{-1}$, where ξ is the characteristic length[18] describing fluctuations in the magnetic order parameter. The dominant contribution to the integral then comes from the region where $q' \lesssim \xi^{-1}$. This means that we can take

$$\int\frac{d\mathbf{q}'}{(2\pi)^3} \to \xi^{-3}. \quad (14)$$

At the relevant wave vector $q' \sim \xi^{-1}$, the frequency denominator is

$$s_\sigma(\mathbf{q}')+s_\sigma(\mathbf{q}-\mathbf{q}')-s|_{q'\sim\xi^{-1}} \approx 2s_\sigma^*, \quad (15)$$

where

$$s_\sigma^* \equiv s_\sigma(\mathbf{q}')|_{q'\sim\xi^{-1}}.$$

The next step in our estimate is to evaluate

$$|M_{\mathbf{q},\mathbf{q}}|^2 \equiv |\langle|\tau^{xx}(-\mathbf{q})Pa_6(\mathbf{q}')a_6(\mathbf{q}-\mathbf{q}')|\rangle|^2 \quad (16)$$

in the $q, q' \to 0$ limit.

The diagonal component of the stress tensor is essentially the pressure p, and so we can write

$$\lim_{q\to0,q'\to0}|M_{\mathbf{q},\mathbf{q}'}|^2 = \lim_{q\to0,q'\to0}|\langle|p(-\mathbf{q})[1-\sum_j a_j(\mathbf{q})]|\rangle \times\langle|a_j(-\mathbf{q})]a_6(\mathbf{q}')a_6(\mathbf{q}-\mathbf{q}')|\rangle|^2 \approx \lim_{q\to0,q'\to0}|\langle|p(-\mathbf{q})a_2(\mathbf{q})|\rangle \times\langle|a_2(-\mathbf{q})a_6(\mathbf{q}')a_6(\mathbf{q}-\mathbf{q}')|\rangle|^2, \quad (17)$$

where the last step in Eq. (17) follows, since only the

[18] L. P. Kadanoff, W. Götze, D. Hamblen, R. Hecht, E. A. S. Lewis, V. V. Palciauskas, M. Rayl, and J. Swift, Rev. Mod. Phys. **39**, 395 (1967).

term in the projection operator involving the state $a_2(\mathbf{q})|\,\rangle$ gives rise to a divergence in the matrix element. In the zero wave-vector limit, the operators in Eq. (17) can be replaced by thermodynamic derivatives.[19] In particular,[9]

$$a_2(q\to 0)=(\rho k_B T)^{1/2}\frac{c(q)}{\langle n\rangle}\frac{\partial}{\partial\mu}\bigg|_{S/N}, \tag{18a}$$

$$a_6(q\to 0)=\left(\frac{k_B T}{\chi(q)}\right)^{1/2}\frac{\partial}{\partial H}\bigg|_{T}. \tag{18b}$$

Hence, in Eq. (17) what we basically have to evaluate is

$$\frac{\partial\chi}{\partial\mu}\bigg|_{S/N}=\frac{C_\chi}{C_\mu}\frac{\partial\chi}{\partial\mu}\bigg|_{T}, \tag{19}$$

where C_χ is the specific heat at constant susceptibility and C_μ is the specific heat at constant chemical potential. Use of the Gibbs-Duheim relationship[20] then gives us

$$\frac{\partial\chi}{\partial\mu}\bigg|_{T}=\langle n\rangle\frac{\partial\chi}{\partial p}\bigg|_{T}. \tag{20}$$

Similarly,

$$\frac{\partial p}{\partial\mu}\bigg|_{S/N}=\frac{C_p}{C_\mu}\langle n\rangle. \tag{21}$$

We thus find that

$$\lim_{q\to 0,\,q'\to 0}|M_{q,q'}|^2\approx(\rho k_B T)^2 c^4\frac{C_p^2C_\chi^2}{C_\mu^4}\left(\frac{1}{\chi}\frac{\partial\chi}{\partial p}\bigg|_{T}\right)^2. \tag{22}$$

Taking $\chi=\chi_0\epsilon^{-\gamma}$, we can rewrite Eq. (22) as

$$\lim_{q\to 0,\,q'\to 0}|M_{q,q'}|^2\approx(\rho k_B T\gamma)^2 c^4\frac{C_p^2C_\chi^2}{C_\mu^4}\left(\frac{1}{T_c}\frac{dT_c}{dp}\right)^2\epsilon^{-2}. \tag{23}$$

At this point we substitute Eqs. (14), (15), and (23) into Eq. (13) and obtain for T near T_c

$$\Delta\alpha_l(q)\approx 1.09q^2\gamma^2c^3\rho k_B T\frac{C_p^2C_\chi^2}{C_\mu^4}\left(\frac{1}{T_c}\frac{dT_c}{dp}\right)^2\frac{\xi^{-3}\epsilon^{-2}}{s_\sigma^*}. \tag{24}$$

Relatively far from the magnetic critical point we expect the specific heats in Eq. (24) to be essentially the same (since they will be dominated by the nonmagnetic lattice contribution). In this region, $C_p^2C_\chi^2/C_\mu^4$ will be of order unity. But very near T_c, provided that the coupling between the spins and the lattice is sufficiently weak that the specific heats may be considered as the sum of a magnetic part and a nonmagnetic part, we expect

$$C_p^2C_\chi^2/C_\mu^4\sim\epsilon^{2\alpha}, \tag{25}$$

where α is the critical index that describes the divergence of the magnetic portion of C_p in zero applied field. We will assume that we are in the region where Eq. (25) holds. We also write $\xi=\xi_0\epsilon^{-\nu}$ and make use of the scaling relationship[18] $3\nu=2-\alpha$ to obtain

$$\Delta\alpha_l(q)\sim\frac{q^2c^3\rho k_B T_c\gamma^2}{\xi_0^3}\left(\frac{1}{T_c}\frac{dT_c}{dp}\right)^2\frac{\epsilon^{+\alpha}}{s_\sigma^*}. \tag{26}$$

Equation (26) is our basic result. Note that the dominant temperature dependence of $\Delta\alpha_l$ is determined by the characteristic frequency s_σ^*.

The same considerations hold for an antiferromagnetic system for which we also obtain Eq. (26), with s_σ^* being interpreted as the relaxation rate of $\sigma^s(\xi^{-1})$.

From Eq. (26) we see that our calculation gives $\Delta\alpha_l(q)$ proportional to q^2. This is a consequence of our taking

$$|s|=\omega=cq\ll s_\sigma^*$$

in our evaluation of Eq. (13). Actually $s=-i\omega$, and taking the real part of the right-hand side of Eq. (13), we obtain

$$\Delta\alpha_l(q,\omega)\approx\frac{2.17q^2}{c(q)\rho k_B T}\int\frac{dq'}{(2\pi)^3}\times\frac{|M_{q,q'}|^2[s_\sigma(\mathbf{q}')+s_\sigma(\mathbf{q}-\mathbf{q}')]}{[s_\sigma(\mathbf{q}')+s_\sigma(\mathbf{q}-\mathbf{q}')]^2+\omega^2}. \tag{27}$$

For $\omega\gtrsim s_\sigma^*$, we can no longer ignore the ω^2 in the denominator of the integrand. Also, if $q\gg\xi^{-1}$, then the magnetic coherence length no longer sets the only scale of lengths in the problem. When either of these situations occur, we can no longer estimate the integral in the manner we did before, and Eq. (26) will not hold.

Because of the importance of the relaxation rate s_σ^* in Eq. (26) and because of the difficulty of determining it for realistic physical models, we shall point out two experimental methods of determining it before we go on to the results of model calculations.

III. EXPERIMENTAL DETERMINATION OF s_σ^*

It is well known[21] that if the scattering vector $\mathbf{K}$ is not parallel to the direction of magnetic order, there is a peak in the inelastic scattering cross section when neutrons are scattered from magnetic materials. This peak corresponds to the scattering of neutrons from dynamic spin fluctuations. For $K\lesssim\xi^{-1}$, s_σ^* determines the width of this scattering peak, and so a measurement of the peak width as $T\to T_c$ gives a direct measurement of the temperature dependence of s_σ^*. This conclusion is equally valid for both ferromagnetic and antiferromagnetic systems.

[19] See, e.g., L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley Publishing Co., Inc., Reading, Mass., 1960).

[20] See, e.g., H. B. Callen, *Thermodynamics* (John Wiley & Sons, Inc., New York, 1966).

[21] A review of the theory of the critical magnetic scattering of neutrons has been given by P.-G. de Gennes, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1964).

Also, Heller[22] has shown that in magnetic systems near T_c, the NMR linewidth can be attributed to fluctuations in the local hyperfine fields seen by the nuclei. The result for the NMR linewidth can be expressed as

$$\delta\nu_c=\frac{1}{\pi\sqrt{3}}\left(\frac{A}{\hbar}\right)^2\sum_{\mathbf{q}}\left(\frac{\langle|S_c(\mathbf{q})S_c(-\mathbf{q})|\rangle}{s^c(\mathbf{q})}+\frac{1}{2}\frac{\langle|S_a(\mathbf{q})S_a(-\mathbf{q})|\rangle}{s^a(\mathbf{q})}+\frac{1}{2}\frac{\langle|S_{a'}(\mathbf{q})S_{a'}(-\mathbf{q})|\rangle}{s^{a'}(\mathbf{q})}\right), \quad (28)$$

where the interaction between the nucleus and the electron spin is taken as

$$\mathcal{H}_{\rm int}=A\mathbf{I}\cdot\mathbf{S}, \quad (29)$$

and the average local field seen by the nucleus is taken to lie along the c axis. If the c axis corresponds to the direction of the order parameter in space, then

$$\langle|S_c(\mathbf{q})S_c(-\mathbf{q})|\rangle\sim\chi(q),$$

which diverges like $\epsilon^{-\gamma}$ as $q\to 0$ and $T\to T_c$. The dominant contribution to Eq. (28) as $T\to T_c$ occurs for $q\lesssim\xi^{-1}$, and so, estimating the sum over wave vectors in the same manner as before, we obtain

$$\delta\nu_c\sim\xi^{-3}\epsilon^{-\gamma}/s_\sigma{}^*\sim\epsilon^{2-\gamma-\alpha}/s_\sigma{}^*. \quad (30)$$

Hence, if $s_\sigma{}^*\sim\epsilon^x$ for a given material, then we expect the following to hold as $T\to T_c{}^+$:

(a) The anomalous longitudinal sound attenuation coefficient should behave like

$$\Delta\alpha_l\sim\epsilon^{+\alpha-x}. \quad (31a)$$

(b) The width of the neutron scattering peak in the limit of zero momentum transfer should behave like

$$\text{peak width}\sim\epsilon^x, \quad (31b)$$

provided that the momentum transfer vector is not parallel to the direction of the order parameter.

(c) The NMR linewidth should behave like

$$\delta\nu\sim\epsilon^{2-\gamma-\alpha-x}. \quad (31c)$$

Equations (31) provide for relations between experimentally measurable quantities, and hence give a check of the mode-mode coupling procedure that does not depend upon the model used to evaluate $s_\sigma{}^*$.

IV. PREDICTED RESULTS FOR VARIOUS MODEL SYSTEMS

A. Heisenberg Ferromagnets

The Hamiltonian for the Heisenberg ferromagnet may be written as

$$\mathcal{H}=-\sum_{\langle ij\rangle}\left[K_{ij}(S_i{}^xS_j{}^x+S_i{}^yS_j{}^y)+J_{ij}S_i{}^zS_j{}^z\right], \quad (32)$$

where the K_{ij} and the J_{ij} are such that it is energetically favorable for neighboring spins to be aligned. We distinguish among three different cases:

Case 1: $K=J$. This case corresponds to an isotropic system, and the anomalous spin fluctuation relaxes via a diffusive mechanism, i.e.,

$$s_\sigma(\mathbf{q})=Dq^2. \quad (33)$$

According to Kawasaki[23] and to Halperin and Hohenberg,[24] $D\sim\xi^{-1/2+\eta/2}$, where η is the parameter measuring the deviation of the spin-spin correlation function from the Ornstein-Zernike form. Kadanoff *et al.*[18] give $\eta=0.07\pm0.07$. We thus take

$$s_\sigma{}^*\sim\xi^{-5/2+\eta/2}\sim\epsilon^{5/2\nu-\nu\eta/2}, \quad (34)$$

and so we would predict

$$\Delta\alpha_l\sim\epsilon^{-5/3+11\alpha/6+\eta/3-\alpha\eta/6} \quad (35)$$

for this system.

Case 2: $K>J$. This corresponds to a planar model where the order parameter lies in the xy plane. For this case Kawasaki[25] has shown that

$$s_\sigma{}^*\sim\xi^{-3/2}\sim\epsilon^{3/2\nu}, \quad (36)$$

which means that we expect

$$\Delta\alpha_l\sim\epsilon^{-1+3\alpha/2} \quad (37)$$

for this system.

Case 3: $J>K$. This corresponds to a model exhibiting uniaxial anisotropy. No completely satisfactory theory exists that gives $s_\sigma(q)$ for this particular case. However, if $J\gg K$, there are no compelling reasons for doubting the validity of the conventional theory[26,27] for the critical slowing down of the system. The conventional theory predicts that

$$s_\sigma{}^*\sim 1/\chi\sim\epsilon^\gamma. \quad (38)$$

A specific model in which the uniaxial anisotropy arises from spin-orbit coupling is considered in Sec. IV C. The result is in accord with Eq. (38). Studies have also been made on a particularly anisotropic system—the kinetic Ising model. The results are somewhat ambiguous. Kawasaki and Yamada[23,28] find that in the local equilibrium approximation, Eq. (38) holds for the relaxation rate for fluctuation in the order parameter. However, Suzuki *et al.*[29] find that the relaxation rate approaches zero like

$$s_\sigma{}^*\sim\epsilon^\Delta \quad (\Delta=2.00\pm0.05) \quad (39)$$

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[29] M. Suzuki, H. Ikari, and R. Kubo, J. Phys. Soc. Japan Suppl. **26**, 1969.

TABLE I. Expected critical behavior of anomalous ultrasonic attenuation, width of neutron scattering peak, and NMR linewidth for some idealized model systems.

Model system	$\Delta\alpha_l$[a]	Peak width	NMR linewidth
Isotropic Heisenberg ferromagnet	$\sim\epsilon^{-5/3+11\alpha/6+\eta/3-\alpha\eta/6}$	$\sim\epsilon^{5/3-5\alpha/6-\eta/3+\alpha\eta/6}$	$\sim\epsilon^{1/3-\gamma-\alpha/6+\eta/3-\alpha\eta/6}$
Planar Heisenberg ferromagnet	$\sim\epsilon^{-1+3\alpha/2}$	$\sim\epsilon^{1-\alpha/2}$	$\sim\epsilon^{1-\gamma-\alpha/2}$
Uniaxial Heisenberg ferromagnet	$\sim\epsilon^{-\gamma+\alpha}$	$\sim\epsilon^{\gamma}$	$\sim\epsilon^{2-2\gamma-\alpha}$
Isotropic Heisenberg ferromagnet	$\sim\epsilon^{-1+3\alpha/2}$	$\sim\epsilon^{1-\alpha/2}$	$\sim\epsilon^{1-\gamma-\alpha/2}$
Planar Heisenberg ferromagnet	$\sim\epsilon^{-1+3\alpha/2}$	$\sim\epsilon^{1-\alpha/2}$	$\sim\epsilon^{1-\gamma-\alpha/2}$
Uniaxial Heisenberg ferromagnet	$\sim\epsilon^{-\gamma+\alpha}$	$\sim\epsilon^{\gamma}$	$\sim\epsilon^{2-2\gamma-\alpha}$
Spin-orbit coupling model	$\sim\epsilon^{-\gamma+\alpha}$	$\sim\epsilon^{\gamma}$	$\sim\epsilon^{2-2\gamma-\alpha}$

[a] We expect $\Delta\alpha_l$ to be proportional to q^2 for all of these model systems.

for a two-dimensional Ising system. This is an indication that the critical index describing the relaxation of a spin fluctuation may be different from that of the static susceptibility, which is 7/4 for a two-dimensional Ising system.[18]

We shall take Eq. (38) to hold for a uniaxial material, and thus we expect that for such systems

$$\Delta\alpha_l\sim\epsilon^{-\gamma+\alpha}. \tag{40}$$

B. Heisenberg Antiferromagnets

The Hamiltonian for the Heisenberg antiferromagnet is given by Eq. (32), except that the K_{ij} and the J_{ij} are such that it is energetically favorable for neighboring spins to be aligned antiparallel. We again distinguish among three different cases:

Case 1: $K=J$. This case corresponds to the isotropic Heisenberg antiferromagnet. A fluctuation in the sublattice magnetization relaxes according to [23,24]

$$s_\sigma{}^*\sim\xi^{-3/2}\sim\epsilon^{3/2\nu}, \tag{41}$$

and so we expect

$$\Delta\alpha_l\sim\epsilon^{-1+3\alpha/2}. \tag{42}$$

Case 2: $|K|>|J|$. For this system the order parameter lies in the xy plane. Kawasaki[30] claims that the result for this case is the same as in case 1, i.e.,

$$s_\sigma{}^*\sim\xi^{-3/2}\sim\epsilon^{3/2\nu}. \tag{43}$$

If this is the case, we would expect

$$\Delta\alpha_l\sim\epsilon^{-1+3\alpha/2}. \tag{44}$$

Case 3: $|J|>|K|$. Like the uniaxial ferromagnet, there is no completely satisfactory theory for this case. However, for very strong anisotropy it is reasonable to expect the conventional theory[22,31] to hold. This gives us

$$s_\sigma{}^*\sim 1/\chi_s\sim\epsilon^{\gamma}, \tag{45}$$

and so we expect

$$\Delta\alpha_l\sim\epsilon^{-\gamma+\alpha} \tag{46}$$

for the single-axis antiferromagnet.

C. System with Weak Spin-Orbit Coupling

We next consider a system in which the spin-orbit coupling is sufficiently weak that the orbital angular momentum and the spin angular momentum are individually good quantum numbers, but in which this interaction is the dominant mechanism through which a fluctuation in the order parameter relaxes.

In the mode-mode coupling formalism[9] this relaxation rate may be written as

$$s_\sigma(\mathbf{q},s)=-\langle|a_6(-\mathbf{q})\mathfrak{L}P(P\mathfrak{L}P-s)^{-1}P\mathfrak{L}a_6(\mathbf{q})|\rangle \tag{47}$$

for a ferromagnetic system (for an antiferromagnetic system we replace the a_6 operators with $a_6{}^s$ operators). We write the spin-orbit coupling term as

$$\mathfrak{IC}_{\rm so}=\Lambda\sum_{\mathbf{R}}\mathbf{L}(\mathbf{R})\cdot\mathbf{S}(\mathbf{R}), \tag{48}$$

where $\mathbf{L}$ is the orbital angular momentum of the ion and the sum is over all the magnetic ions. In terms of this Hamiltonian we can rewrite Eq. (47) as

$$\begin{aligned}s_\sigma(\mathbf{q},s)=&\frac{-1}{\hbar^2\chi(\mathbf{q})/\beta\mu_0{}^2}\sum_{\mathbf{R},\mathbf{R}'}e^{i\mathbf{q}\cdot\mathbf{R}-i\mathbf{q}\cdot\mathbf{R}'}\\&\times\langle|[\sigma(\mathbf{R}),\mathfrak{IC}_{\rm so}]P(P\mathfrak{L}P-s)^{-1}P[\sigma(\mathbf{R}'),\mathfrak{IC}_{\rm so}]|\rangle\\=&\frac{\Lambda^2}{\chi(\mathbf{q})/\beta\mu_0{}^2}\sum_{\mathbf{R},\mathbf{R}'}e^{i\mathbf{q}\cdot\mathbf{R}-i\mathbf{q}\cdot\mathbf{R}'}\langle|[\mathbf{L}(\mathbf{R})\times\mathbf{S}(\mathbf{R})]_z\\&\times P(P\mathfrak{L}P-s)^{-1}P[\mathbf{L}(\mathbf{R}')\times\mathbf{S}(\mathbf{R}')]_z|\rangle,\end{aligned} \tag{49}$$

where we have taken the z component of $\mathbf{S}$ to correspond to the order parameter σ.

$\mathbf{L}(\mathbf{R})$ is a highly localized function of position, and this eliminates any long-range coupling between the spins in the matrix element that could give rise to critical behavior as $T\to T_c$. We also expect that $(P\mathfrak{L}P-s)^{-1}$ will essentially be a time characterizing the orbital electronic relaxation. This also should not exhibit critical behavior. The entire matrix element will be noncritical as $T\to T_c$. Hence, as far as its critical behavior is concerned,

$$s_\sigma(q)\sim 1/\chi(q), \tag{50}$$

which gives us

$$s_\sigma{}^*\sim 1/\chi\sim\epsilon^{\gamma} \tag{51}$$

for this system.

The sound attenuation coefficient will then behave like

$$\Delta\alpha_l\sim\epsilon^{-\gamma+\alpha}. \tag{52}$$

[30] K. Kawasaki (private communication).
[31] T. Moriya, Progr. Theoret. Phys. (Kyoto) 28, 371 (1962).

TABLE II. Experimental results for anomalous ultrasonic attenuation in some magnetic insulators.

Material	Anisotropy factor	Apparent classification	q^2 behavior	y	ϵ range	Ref.
EuO	4×10^{-4}	Isotropic ferromagnet	No anomalous attenuation above T_c			4, 33
$RbMnF_3$	5×10^{-6}	Isotropic antiferromagnet	Yes	0.32	2×10^{-4}–4×10^{-2}	3, 34
			?	0.7 –1[a]	4×10^{-2}– ?	33, 34
MnF_2	3×10^{-2}	Borderline between isotropic and single-axis antiferromagnet	No	0.14	10^{-4}–4×10^{-2}	35
			Yes	0.2	3×10^{-4}–6×10^{-2}	32
			Yes	0.4 –0.5	3×10^{-4}–3×10^{-2}	36, 37
			?	1.6[a]	6×10^{-2}– ?	34
CoO	See text for discussion of structure		No	0.35	2×10^{-4}–2×10^{-3}	38
			No	2.6	2×10^{-3}– 10^{-2}	38

[a] These exponents are very uncertain because of the small values of the anomalous attenuation involved.

D. Summary

In this section we have given the theoretical order-parameter relaxation rates for various models of magnetic systems. We note that except for the spin-orbit coupling model, any interaction between the spin system and the lattice system was neglected in determining the order-parameter relaxation rate. Once the relaxation rate s_σ^* is determined, we have predictions as to the critical behavior of the anomalous ultrasonic attenuation coefficient, the width of the neutron scattering peak in the $K\rightarrow 0$ limit, and the NMR linewidth. We summarize these results in Table I.

V. DISCUSSION OF SPECIFIC MATERIALS

In this section we shall compare our theory with experimental measurements. As calculations of s_σ^* have been made only for localized spin models, we shall restrict ourselves to materials for which this is a good approximation. In particular, we shall discuss several insulators and rare-earth metals.

A. Insulating Magnetic Materials

A common feature of the magnetic insulators is the presence of nonmagnetic anions as well as the magnetic cations. Interaction between these two systems can give rise to other relaxation processes besides these discussed in Sec. IV. In Eq. (26), s_σ^* is the actual relaxation rate of a typical spin fluctuation. If there are various independent relaxation processes, s_σ^* is the sum of the rates from all relaxation processes. These processes are governed by different dynamics and are affected differently by the onset of the phase transition, i.e., they will be characterized by different powers of ϵ. If one term in s_σ^* is much larger than all the others, then its behavior as $T\rightarrow T_c$ will essentially determine the exponent y in Eq. (1). As the critical point is approached, if this particular term in s_σ^* is going to zero at a faster rate than other terms, it may be that at some value of ϵ another term will begin to dominate s_σ^*, and we will see a region where a different value of y appears. We thus have the possibility of the exponent y changing to smaller values as the critical point is approached. It is interesting to note that in the magnetic insulators discussed below, where anomalous sound attenuation is present, there is some experimental evidence that more than one region of behavior exists for $\Delta\alpha_l(q)$.

In Table II[32–38] we give the experimental results for $\Delta\alpha_l(q)$ for EuO, $RbMnF_3$, MnF_2, and CoO. The anisotropy factor is defined as

$$A=[|J(\mathbf{q}=0)|-|K(\mathbf{q}=0)|]/|J(q=0)|\,, \quad (53)$$

where J and K are defined through Eq. (32). The values[32] of A given in Table II are meant to give the reader a qualitative feeling for how anisotropic a material is.

1. *Europium Oxide*

EuO is a ferromagnetic insulator with Eu^{+2} ions forming an fcc lattice. The low anisotropic factor indicates that it is probably well represented as an isotropic Heisenberg ferromagnet. Experiments[4,33] in which longitudinal sound waves are propagated along the [110] and [100] axes indicate that there is no anomalous attenuation above T_c, but that there is a very sharp rise in the sound attenuation as we pass below T_c.

In an isotropic system the diagonal component of the stress tensor is given by

$$\tau^{ii}=-\langle\delta\mathcal{H}/\delta V\rangle. \quad (54)$$

In the limit of weak coupling between the spin system and the lattice system the magnetic portions of the operators are what enter in Eq. (11). Lüthi and Pollina[4] have pointed out that for an isotropic Heisenberg system in which the exchange coupling is of a strictly nearest-neighbor type, the magnetic portion of the stress tensor and the magnetic portion of the energy density will both be proportional to the same linear combination of nearest-neighbor operators. Hence, for

[32] B. Lüthi, P. Papon, and R. J. Pollina, in Proceedings of the Fourteenth Annual Conference on Magnetism (unpublished).
[33] B. Lüthi and R. J. Pollina, Bull. Am. Phys. Soc. **14**, 418 (1968).
[34] B. Lüthi (private communication).
[35] A. Ikushima Phys. Letters **29A**, 364 (1969).
[36] J. R. Neighbors and R. W. Moss, Phys. Rev. **173**, 542 (1968).
[37] R. G. Evans, Phys. Letters **27A**, 451 (1968).
[38] A. Ikushima, Bull. Am. Phys. Soc. **14**, 418 (1969); Phys. Letters **29A**, 417 (1969).

such a system,

$$\tau_m{}^{ii} \propto \mathcal{H}_m, \tag{55}$$

and the projection operators in Eq. (11) act to give a zero result, and we would expect no anomalous sound attenuation. From Eqs. (7) and (10), any magnetic contribution to the sound attenuation would be due to the magnetic contribution to the thermal conductivity and would be nondivergent. This has also been noted in Ref. 4. This is believed to be the explanation for the lack of anomalous attenuation above T_c in EuO. There is also additional experimental evidence to support this.

In EuO it is observed[39] that the magnetic specific heat C_m and the magnetic part of the differential thermal expansion coefficient β_m are strictly proportional to each other at temperatures above T_c. Since

$$C_m = \frac{\partial}{\partial T}\langle \mathcal{H}_m \rangle \tag{56a}$$

and

$$\beta_m = -\tfrac{1}{3} K_T V \frac{\partial}{\partial V}\frac{\partial}{\partial T}\langle \mathcal{H}_m \rangle, \tag{56b}$$

where K_T is the isothermal compressibility of the system, we can take this observation to imply

$$\langle \mathcal{H}_m \rangle \propto \frac{\partial \langle \mathcal{H}_m \rangle}{\partial V} \propto \tau_m{}^{ii}.$$

Hence, independent of any specific model of the magnetic Hamiltonian for EuO, the experimental work indicates that we should expect no anomalous sound attenuation in this material.

2. *Rubidium Manganese Fluoride*

$RbMnF_3$ is an insulating antiferromagnetic having the perovskite structure with the Mn^{+2} ions lying on a simple cubic lattice. The extremely small anisotropy factor indicates that the material should be well represented as an isotropic Heisenberg antiferromagnet. We would then expect the sound attenuation coefficient to behave like

$$\Delta\alpha_l \sim q^2 \epsilon^{-1+3\alpha/2} \sim q^2 \epsilon^{-1}, \tag{57}$$

where the last follows because measurements[40] indicate $\alpha \approx 0$ for $RbMnF_3$. Experimentally,[33,34] there appear to be two regions of behavior, with the region further away from T_c roughly agreeing with Eq. (57) and the region nearer T_c having a weaker divergence. The change-over occurs at $\epsilon \approx 4\times 10^{-2}$. If this change in behavior is attributable to the behavior of $s_\sigma{}^*$, then it should be observed in other phenomena as indicated in Sec. III.

The $K \to 0$ neutron scattering peak has been determined for this material.[41,42] In Ref. 41 there is an indication that the peak width departs from an ϵ^1 behavior as $T \to T_c{}^+$. In the temperature range studied by Golding,[3] where $\Delta\alpha_l \sim q^2\epsilon^{-0.32}$, the behavior of the $K \to 0$ scattering peak can be approximately taken as

$$\text{peak width} \sim \epsilon^{0.4}. \tag{58}$$

This is in better agreement with the sound attenuation work in this temperature region. However, there are problems in taking the $K \to 0$ limit particularly very near T_c, and so this observation is by no means conclusive. More recent measurements of the peak width[42] give

$$\text{peak width} \sim \epsilon^1 \tag{59}$$

for $\epsilon \gtrsim 2.4\times 10^{-2}$, and the width seems to extrapolate to zero at T_c if this power law is maintained. It would be useful if precise measurements could be made of the peak width for ϵ in the range 10^{-3}–10^{-4}, for these would clearly resolve the matter. Let us consider possible relaxation mechanisms in $RbMnF_3$. In the "linear" region, $s_\sigma{}^*$ is dominated by a process in which the order in the sublattice magnetization relaxes by the spin-exchange process described in Sec. IV B. We can estimate the relaxation rate in this region as $(k_B T_c/\hbar)\epsilon^1$. As $T \to T_c$, the contribution of this decay process becomes less important.

We next note that the F^{19} nucleus has a large nuclear moment and has a 100% isotopic abundance in nature. When the spin fluctuation region relaxes so slowly by the spin-exchange process that it would exist for times comparable to the precession time of the F^{19} nucleus in the effective field set up by the ordered moments, there can be effective coupling between these two systems. We would expect the relaxation rate due to this coupling to be proportional to the precessional frequency of the F^{19} nucleus. Below T_c,[43]

$$\nu_{19}(T) = \nu_{19}(0) M_s(T)/M_s(0) = \nu_{19}(0)\epsilon^\beta. \tag{60}$$

In the scaling idea[18] there is a symmetry in the order of magnitude of fluctuations under the interchange $T_c - T \to -(T_c - T)$, and so we take Eq. (60) to hold above T_c as well when the effective field seen by the F^{19} is due to a fluctuation of the magnetic order parameter. Near T_c we thus expect a region where $s_\sigma{}^* \sim \epsilon^\beta$, and in this region

$$\Delta\alpha_l \sim q^2 \epsilon^{-\beta}, \tag{61}$$

which gives the right temperature dependence. However, estimates of $\nu_{19}(0)$ clearly indicate that this effect is much too small to account for the position of the change-over at $\epsilon \approx 4\times 10^{-2}$. We are unable to find a relaxation mechanism that correctly describes the behavior of $RbMnF_3$.

We also note that if the contribution given in Eq. (10) is divergent, as suggested by Wagner,[13] this could also account for the changing of y to a smaller value as $T \to T_c{}^+$. In $RbMnF_3$ most of the spin-system energy

[39] B. E. Argyle, N. Miyata, and T. D. Schultz, Phys. Rev. **160**, 413 (1967).
[40] D. T. Teaney, V. L. Moruzzi, and B. E. Argyle, J. Appl. Phys. **37**, 1122 (1968).
[41] R. Nathans, F. Menzinger, and S. J. Pickart, J. Appl. Phys. **39**, 1237 (1968).
[42] J. M. Hastins (private communication).
[43] P. Heller and G. B. Benedek, Phys. Rev. Letters **8**, 428 (1962).

TABLE III. Experimental results of anomalous ultrasonic attenuation in some rare-earth metals.

Material	Anisotropy factor	y	ϵ range	B Theoret.	B Expt.	Ref.
Gd	5×10^{-5}	1.2	10^{-3}–10^{-1}	7.6×10^{-9}	6.6×10^{-9}	1, 2, 32
Ho	-7×10^{-2}	1.0[a]	3×10^{-4}–10^{-1}	3.4×10^{-9}	5.2×10^{-9}	2, 32
Tb	-0.4	1.24[a]	7×10^{-3}–10^{-1}	4.7×10^{-9}	1.2×10^{-9}	2, 32
Dy	-0.3	1.37[a]	3×10^{-3}–10^{-1}	3.0×10^{-9}	1.2×10^{-9}	2, 32

[a] These experimental measurements are for the transition from the paramagnetic state to the spiral spin structure.

is due to nearest-neighbor exchange, and if the exchange constants do not vary drastically with small changes in the lattice parameter, the projection operators in Eq. (11) could drastically reduce the calculated value of B given in Eq. (26). This could still be the dominant effect relatively far from T_c where $1/C_V-1/C_p\approx0$, and this would give $y=1$. However, nearer T_c when $1/C_V-1/C_p\approx A|\ln\epsilon|/C_V^2$ (taking $C_p=C_V+A|\ln\epsilon|$), the contribution of Eq. (10) could dominate, thus giving rise to a smaller value of y.

3. *Manganese Fluoride*

MnF_2 is an insulating antiferromagnet having the rutile structure. The Mn^{+2} ions form a body-centered tetragonal unit cell. The direction of easy magnetization is along the c axis. Its anisotropy factor is such that it is borderline as to whether it should be compared with an isotropic or a single-axis antiferromagnet. It seems to us that because of its structure it is more likely that the single-axis model is a better representation. This choice is consistent with the NMR linewidth measurements on this material,[22] which give the NMR linewidth diverging like $\sim\epsilon^{-0.6}$. This gives $s_\sigma^*\sim\epsilon^{1.2-1.3}$, depending on our choice of α and γ. (For MnF_2,[18] $\gamma\sim\frac{4}{3}$ and $\alpha\lesssim0.16$.) The NMR linewidth measurements are in the region $2\times10^{-2}\gtrsim\epsilon\gtrsim5\times10^{-4}$. In this same temperature region,[32,35–37] $\Delta\alpha_l$ appears to diverge at a much slower rate than we would expect from $s_\sigma^*\sim\epsilon^{1.2-1.3}$. It may be that this low value of y is due to another relaxation process taking over, but it is strange that it would not show up in the NMR work. There are indications[34] that for $\epsilon\gtrsim6\times10^{-2}$, $\Delta\alpha_l$ diverges at a faster rate than it does closer to T_c. Probably the same mechanism giving rise to the odd behavior of y for $RbMnF_3$ is in operation here, and so the remarks made in Sec. V A *2* may be relevant for this material also. Ikushima[35] indicates that the q^2 behavior may not hold either for this material. He finds $\Delta\alpha_l$ going like $q^{1.6-1.8}$. If we are out of the region where the q^2 behavior is found, then we no longer expect our theory to hold.

4. *Cobalt Oxide*

CoO is an insulating antiferromagnet with the Co^{+2} ions forming an fcc lattice. Below T_c the ordered structure consists of the spin sites in (111) planes being ferromagnetically ordered with the moments in adjacent planes being oppositely directed. The actual direction[44] of the magnetic moments lies close to the $[\bar{1}\bar{1}7]$ direction. We hesitate to characterize this structure by any of the models in Sec. IV.

For longitudinal sound waves propagating in the [100] direction there were found to be two distinct regions[38] of behavior for $\Delta\alpha_l$. The wave-vector dependence was observed to go as q^1 rather than as q^2. There was also found to be strong attenuation of shear waves propagating in this material, and the anomalous behavior for the shear waves was similar to that of the longitudinal waves. The presence of the q^1 behavior indicates that this material lies outside the region of validity for our theory.

B. Rare-Earth Metals

The rare-earth metals crystallize in a hcp form. Their magnetic properties are due to the deep-lying $4f$ electrons on the ion cores. The ions are in a $+3$ valence state and interact with each other through the conduction electrons by means of a Ruderman-Kittel[45] type of interaction. This gives rise to effective exchange constants that are long-ranged and have an oscillatory nature, and hence is why these materials exhibit such intricate magnetic structures.[46,47] Strong spin-orbit coupling is present in these materials, and the ion spin and orbital angular momentum are tightly coupled to form a state of definite total angular momentum. In general, besides the kinds of anisotropy described by Eq. (32) there is also anisotropy within the basal planes of these materials. It is not completely clear how this anisotropy will affect the relaxation rate s_σ^*. If it is strong enough to pin the spin system and prevent a uniform precessional mode from existing, it is reasonable to suppose that the conventional theory[27,28] of spin relaxation will hold, which means that

$$s_\sigma^*\sim1/\chi\sim\epsilon^\gamma. \tag{62}$$

Because of the long-range nature of the exchange interaction in the rare-earth metals, the projection operators in Eq. (11) will not greatly reduce the value of B given in Eq. (26). Hence, even if the contribution of Eq. (10)

[44] W. L. Roth, Phys. Rev. **110**, 1333 (1958); **111**, 772 (1958).
[45] M. A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954).
[46] D. H. Martin, *Magnetism in Solids* (The MIT Press, Cambridge, Mass., 1967).
[47] B. R. Cooper, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic Press Inc., New York, 1968).

is divergent, it will not be observed, and Eq. (26) will give a good estimate for $\Delta\alpha_l$.

In Table III we give the experimental results for $\Delta\alpha_l$ for Gd, Ho, Tb, and Dy. The attenuation coefficient is for sound propagating along the c axis of these materials. In all cases $\Delta\alpha_l$ is proportional to q^2. The anisotropy factor is defined through Eq. (53). In these materials only one region of behavior of the critical exponent y is observed. We can thus assume that the dominant contribution to s_σ^* scales like an energy characterizing the magnetic transition, i.e.,

$$s_\sigma^*|_{\epsilon=1}\approx k_B T_c/\hbar. \tag{63}$$

Using Eq. (63) in Eq. (26), we can estimate the temperature-independent coefficient B as

$$B\approx\frac{c^3\rho\hbar\gamma}{\xi_0^3}\left(\frac{1}{T_c}\frac{dT_c}{dp}\right)^2. \tag{64}$$

In Table III we compare our predicted coefficient in Eq. (64) with the coefficients determined in Ref. 2 at $\epsilon=0.01$ and $\nu=50$ MHz. In evaluating Eq. (64) we use experimental measurements of dT_c/dp from Ref. 48 and the sound velocity measurements from Ref. 2. We also use the result of a mean-field-theory calculation for a hcp structure having only a nearest-neighbor interaction to estimate $\xi_0\approx a/\sqrt{6}$, where a is the nearest-neighbor distance. There is a good order-of-magnitude agreement between theory and experiment, which is all we can hope for due to the crudeness of our estimate of B. This is still very encouraging, since it indicates that the correct attenuation mechanism has been considered.

1. Gadolinium

At $T_c\approx292$°K, Gd orders into a ferromagnetic state with the net moment lying along the c axis. The anisotropy factor in Table III corresponds to the region near T_c, and because of its low value it appears that Gd should be considered as an isotropic ferromagnet. As the temperature lowers, the anisotropy factor passes through zero and changes sign. The material remains a ferromagnet, but the orientation of the net moment migrates from the c axis to an easy direction in the basal plane.[46,47]

For an isotropic ferromagnet we expect

$$\Delta\alpha_l\sim q^2\epsilon^{-5/3+11\alpha/6+\eta/3-\alpha\eta/6}, \tag{65}$$

and experimentally[1] for Gd

$$\Delta\alpha_l\sim q^2\epsilon^{-1.2\pm0.1}. \tag{66}$$

It is doubtful that Eqs. (65) and (66) can be reconciled.

In measuring the magnetic susceptibility of single-crystal Gd, Graham[49] found a significant difference between the cases when the applied field was parallel to the c axis and when it was perpendicular to it. His results indicate more uniaxial anisotropy than given by the factor in Table III. If we were to consider Gd as a uniaxial system, we would obtain

$$\Delta\alpha_l\sim q^2\epsilon^{-\gamma+\alpha}, \tag{67}$$

which is in better agreement with Eq. (66) (Graham[49] gives $\gamma\approx\frac{4}{3}$ and Kadanoff *et al.*[18] give $\alpha\lesssim0.16$).

Although a free Gd^{+3} ion is an S-state ion, in a crystal the ground state may have some admixing of higher momentum states. The weak spin-orbit coupling model described in Sec. IV C may hence have some relevance for Gd. It is difficult to know how to classify this material.

2. Holmium

At $T_N\approx132$°K, Ho orders into a spiral spin structure. In this structure the spins lie in planes perpendicular to the c axis with the spins in a given plane being ferromagnetically aligned. The alignment direction within the basal plane changes by a fixed angle between adjacent planes, and so the spin direction spirals in a circle as we move along the c axis. The turn angle between layers varies with temperature. At $T_c\approx19°$ the spin direction tips out of the basal plane and has components both parallel and perpendicular to the c axis.[46,47] The component perpendicular to the c axis continues to rotate as in the spiral spin structure but now there is a net moment along the c axis.

The anisotropy factor in Table III is such that it is not clear whether Ho should be considered as an isotropic antiferromagnet—especially in view of the difficulty with Gd that appears to arise from a smaller anisotropy factor. Also, the spiral spin structure of Ho is nothing like the "normal" structure of an isotropic Heisenberg antiferromagnet (the magnetic structure of $RbMnF_3$). The anisotropy in Ho is planar, so perhaps a good representation of the system is the planar model in Sec. IV A. This gives

$$\Delta\alpha_l\sim q^2\epsilon^{-1+3\alpha/2}, \tag{68}$$

exactly the same as for the isotropic Heisenberg antiferromagnet. If $\alpha\approx0$, then

$$\Delta\alpha_l\sim q^2\epsilon^{-1}, \tag{69}$$

which is what is observed for Ho.

3. Terbium

At $T_N\approx228$°K, Tb orders into the spiral spin structure described in Sec. V B 2. Then at $T_c\approx222$°K, the spiral ordering changes into a ferromagnetic ordering in which the net magnetization is along one of the easy directions in the basal plane. An increase in the sound attenuation is observed as each critical temperature is approached. The relatively high critical temperature for the transition to the ferromagnetic state seems to indicate a rather strong anisotropy within the basal plane. Assuming that this is strong enough to pin the

[48] D. B. McWhan and A. L. Stevens, Phys. Rev. **154**, 438 (1967).
[49] C. D. Graham, J. Appl. Phys. **36**, 1135 (1965).

spin system, Eq. (62) indicates that

$$\Delta\alpha_l \sim q^2\epsilon^{-\gamma+\alpha}, \tag{70}$$

which closely agrees with the experimental result[2] of

$$\Delta\alpha_l \sim q^2\epsilon^{-1.24\pm0.1} \tag{71}$$

for typical values of γ and α.

4. *Dysprosium*

At $T_N \approx 177°$K, Dy orders into the spiral spin structure described in Sec. V B *2*. Then at $T_c \approx 85°$K, the ordering becomes ferromagnetic with the direction of magnetization lying along one of the easy directions in the basal plane (just as for Tb). The sizes of the two critical temperatures seem to indicate that the relative strength of the anisotropy within the basal plane is not as great for Dy as it is for Tb. Agreement with the experimental result[2] of

$$\Delta\alpha_l \sim q^2\epsilon^{-1.37\pm0.1} \tag{72}$$

can be obtained if this anisotropy is large enough to pin the spins such that Eq. (62) holds.

We also note that there is significant attenuation of shear waves in this material[2] with

$$\Delta\alpha_s \sim q^2\epsilon^{-0.8\pm0.1}. \tag{73}$$

This would have to arise from terms in the Hamiltonian other than the volume magnetostriction term which our calculation uses in a phenomenological way.

VI. SUMMARY

In this paper we have presented a calculation of the anomalous attenuation of long-wavelength longitudinal sound waves as the magnetic critical point is approached. In this calculation we assumed a homogeneous model in which the specific details of the lattice were ignored. There were found to be two contributions to the anomalous sound attenuation. The first involves a coupling of the phonons to the energy density of the spin system, and it gives a contribution going as the thermal conductivity. This is nondivergent for a ferromagnetic system, but its status for an antiferromagnetic system is unclear. The other attenuation mechanism is the absorption of a phonon and the emission of two spin fluctuations. We found that its contribution to the anomalous attenuation coefficient could be written as

$$\Delta\alpha_l = Bq^2\epsilon^{\alpha-x}, \tag{74}$$

where B is essentially temperature-independent, and the relaxation rate of a typical spin fluctuation is taken as

$$s_\sigma^* \sim \epsilon^x \tag{75}$$

as $T \to T_c^+$. We then pointed out two ways of determining x experimentally, but unfortunately no given material has been adequately studied by neutron scattering or NMR in the temperature region where the sound attenuation measurements were made. Such a study would check the critical exponents given in Sec. III C.

We also gave the results of calculations of s_σ^* for various model systems and indicated how we expected the anomalous attenuation to behave for these systems. In making comparison between our theory and experimental measurements on real systems, we ran into difficulty because of the complexity of real materials. Except in one instance (i.e., CoO), the q^2 behavior in Eq. (74) was observed to hold. Materials in which nonmagnetic ions as well as magnetic ions were present were observed to have different values for x in different temperature regions. This may be due to more than one relaxation mechanism contributing to s_σ^*. However, if the thermal conductivity contains a divergent part for antiferromagnetic systems, then this could also account for the observed behavior. More work will be necessary in order to clarify this point. For the rare-earth metals we were able to estimate the size of the coefficient B and obtain reasonable agreement with the experimental results. The one material in which there was no anomalous attenuation above T_c could be explained within the framework of our theory.

We conclude that our calculation appears to have some validity for most of the materials considered in this work, but we note that there is still much work to be done in order to explain the existing experimental results completely.

Note added in proof. Lüthi *et al.*[50] have discovered that the Gd sample discussed in Refs. 1, 2, and 32 was unusually anisotropic. Hence, it is reasonable to treat this particular specimen as a single-axis anisotropic material. Attenuation measurements were then made on an isotropic specimen of Gd with a critical exponent $y \approx 1.6$ being found. This is in excellent agreement with Eq. (65).

[50] B. Lüthi, T. J. Moran, and R. J. Pollina, J. Phys. Chem. Solids (to be published).