Critical-Point Anomalies in the Electron-Paramagnetic-Resonance Linewidth and in the Zero-Field Relaxation Time of Antiferromagnets

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A theory is presented for the anomalies in the electron-paramagnetic-resonance (EPR) linewidth and zero-field relaxation time of antiferromagnets. The analysis applies to the paramagnetic state immediately above the Néel point in systems where the dominant spin-spin interaction is the isotropic exchange coupling. It is assumed that the dipolar coupling is the principal source of anisotropy. The EPR linewidth and the relaxation rates for fluctuations in the total magnetization are separated into critical and noncritical parts; the latter are approximated by their values in the high-temperature limit. The anomalous increases in the linewidths and the relaxation rates are shown to arise from processes in which a fluctuation in the total magnetization decays into two fluctuations of the staggered magnetization via the dipolar coupling. The predictions of the theory are compared with linewidth measurements in $RbMnF_3$, MnF_2, MnO, and MnS.

I. INTRODUCTION

In a recent paper[1] (hereafter referred to as I) the author discussed spin-spin relaxation in ferromagnets in the vicinity of the Curie point. Particular attention was paid to the role played by the dipolar interaction and its influence on the decay of the fluctuations in the total magnetization. In this paper the analysis begun in I is extended to cover relaxation effects in antiferromagnets. The objective is to study the decay rates for the fluctuations in the total magnetization in the paramagnetic region above the Néel point. In addition to zero-field relaxation we will also consider the related problem of the electron-paramagnetic-resonance (EPR) linewidth.

Our main interest is in antiferromagnetic insulators where the dominant interaction is the Heisenberg exchange coupling $J\vec{S}_i\cdot\vec{S}_j$. We specifically rule out other than dipolar anisotropy, a restriction which limits the applicability of the analysis to S-state systems such as MnF_2. It should be noted that critical relaxation effects associated with single-ion anisotropy have been discussed by Tomita and Kawasaki from a somewhat different point of view.[2] Also, Fedders has examined the dipolar contribution to the EPR linewidth using an approach rather similar to ours.[3] However, his analysis was restricted to the high-temperature limit, whereas our primary concern is the behavior near the critical point.

The remainder of the paper is divided into three parts. First, the zero-field relaxation is investigated. Second, a general study is made of the linewidth problem. Finally, the published data on the EPR linewidths in $RbMnF_3$, MnF_2, MnO, and MnS are interpreted according to the theory developed in the preceding sections.

II. ZERO-FIELD RELAXATION

The starting point for the calculation of the relaxation time is an equation given previously in I. In the exponential approximation, which is appropriate to the exchange-narrowing limit, the time dependence of the relaxation function characterizing the z component of the total spin takes the form e^{-t/T_2}, where T_2 is given by I, Eq. (11):

$$\frac{1}{T_2}=KTg^6\mu_B^6\hbar^{-2}\chi_T^{-1}N^{-2}\times\sum_{\vec{q}}\{[U_{xx}(\vec{q})-U_{yy}(\vec{q})]^2+4U_{xy}(\vec{q})^2+U_{xz}(\vec{q})^2+U_{yz}(\vec{q})^2\}\times\int_0^\infty dt\,(S(\vec{q},t),\ S(\vec{q}))^2\,. \tag{1}$$

In this equation K is Boltzmann's constant, T is the temperature, g is the electronic g factor, μ_B is the Bohr magneton, and χ_T is the uniform field susceptibility. The $U_{\alpha\beta}(\vec{q})$ are related to the Fourier transform of the spatial part of the dipolar interaction:

$$U_{\alpha\beta}(\vec{q})=\sum_j e^{-i\vec{q}\cdot\vec{r}_{ij}}[3(\vec{r}_{ij})_\alpha(\vec{r}_{ij})_\beta-\delta_{\alpha\beta}r_{ij}^2]\,r_{ij}^{-5}\,, \tag{2}$$

where the sum is over the N spins in the lattice. The symbol $(S(\vec{q},t),\ S(\vec{q}))$ denotes a relaxation function defined by

$$(S(\vec{q},t),\ S(\vec{q}))=\int_0^{1/KT}d\lambda\,\langle e^{(\lambda+it/\hbar)\mathcal{H}}\times S_\alpha(\vec{q})\,e^{-(\lambda+it/\hbar)\mathcal{H}}S_\alpha(-\vec{q})\rangle-(1/KT)\langle S_\alpha(\vec{q})\rangle\langle S_\alpha(-\vec{q})\rangle\,, \tag{3}$$

in which

$$S_\alpha(\vec{q}) = \sum_j e^{-i\vec{q}\cdot\vec{r}_j} S_{\alpha j} , \quad (4)$$

with $\alpha = x$, y, or z. In Eq. (3) $\mathcal{H}$ is the Hamiltonian and the brackets denote an ensemble average.

It is important to keep in mind that Eq. (1) is in itself an approximate expression which is obtained by factorizing a four-spin correlation function.[1] As with many calculations of this type, it is difficult to estimate the error associated with the factorization. From a comparison with experiment (to be discussed below) it appears that the approximation preserves the essential features of the exact expression in that it leads to results which compare favorably with experiment both in their magnitude and temperature dependence.

A second point meriting comment is that Eq. (1) is appropriate only for those temperatures where the important contributions to the sum over wave vectors $\vec{q}$ come from regions of the Brillouin zone where the relaxation function $(S_\alpha(\vec{q}, t), S_\alpha(\vec{q}))$ is approximately the same for each of the spin components. For cubic systems this is not an important restriction. However, in systems of lower symmetry, like MnF_2, in the limit as T approaches T_N (The Néel temperature), Eq. (1) must ultimately be replaced by a more general expression which incorporates the anisotropy of the relaxation function, an effect which of course arises from the dipolar interaction itself.

Equation (1) is suitable for both ferromagnets and antiferromagnets. The difference in the behavior of the two types of systems with respect to zero-field relaxation has its origin in both the behavior of χ_T and the region of the zone associated with the critical-point anomalies. The susceptibility of the ferromagnet diverges near the critical temperature. Moreover, the important contributions to the $\vec{q}$ sum come from wave vectors near the center of the zone. In antiferromagnets, on the other hand, χ_T is finite at T_N, and the important contributions come from wave vectors near $\vec{K}_0$, the superlattice vector for the ordered state. Because of the latter property, it is important to single out the contribution from the $\vec{K}_0$ region. We do this by introducing the function $\Delta(1/T_2)$, defined by

$$\begin{aligned}\Delta(1/T_2) = {} & KTg^6 \mu_B^6 \hbar^{-2} \chi_T^{-1} N^{-2} \\ & \times \{[U_{xx}(\vec{K}_0) - U_{yy}(\vec{K}_0)]^2 + 4U_{xy}(\vec{K}_0)^2 \\ & \times U_{xz}(\vec{K}_0)^2 + U_{yz}(\vec{K}_0)^2\} \\ & \times \sum_{\vec{q}}{}' \int_0^\infty dt\, (S(\vec{q}+\vec{K}_0, t), S(\vec{q}+\vec{K}_0))^2 . \end{aligned} \quad (5)$$

The symbol $\sum'$ indicates that the summation is restricted to the interval $|\vec{q}|\xi \lesssim 1$, where ξ is the correlation length associated with the staggered susceptibility $\chi(\vec{q})^*$, which is defined by

$$\chi(\vec{q})^* = g^2 \mu_B^2 (S(\vec{K}_0 + \vec{q}), S(\vec{K}_0 + \vec{q})) . \quad (6)$$

The reason for introducing $\Delta(1/T_2)$ is that near T_N we have the *approximate* equality

$$1/T_2(T) \approx \Delta(1/T_2) + 1/T_2(\infty) , \quad (7)$$

where $1/T_2(\infty)$ is the relaxation time for the high-temperature limit, where ξ is taken to be zero. The approximate nature of Eq. (7) must be stressed. The equation amounts to asserting that the wave vectors near $\vec{K}_0$ are the source of the anomalous increase in $1/T_2$ near T_N; the contribution from the remainder of the zone varies more slowly with temperature. The justification for this approximation will be discussed in Sec. IV A in connection with the EPR linewidth in $RbMnF_3$, where $\Delta(1/T_2)$ as well as the anomalous part of the linewidth is equal to zero. There it is shown that approximating the noncritical part of the linewidth by the infinite-temperature limit introduces errors of no more than 15% for $(T - T_N)/T_N \gtrsim 0.1$, a level of accuracy we assume holds for other systems as well.

Since we will in effect be using empirical values for $1/T_2(\infty)$ our main interest is in $\Delta(1/T_2)$. As noted, $\Delta(1/T_2)$ characterizes the relaxation rate for fluctuations along the z direction. In the investigation of the anisotropy of the relaxation rate it is helpful to introduce the principal values and principal axes of the tensor $U_{\alpha\beta}(\vec{K}_0)$. After making the principal-axis transformation, $U_{\alpha\beta}(\vec{K}_0)$ takes the form

$$\begin{pmatrix} U_a & 0 & 0 \\ 0 & U_b & 0 \\ 0 & 0 & U_c \end{pmatrix} .$$

With reference to the principal axes the relaxation rate for fluctuations along a direction characterized by cosines γ_a, γ_b, γ_c can be written

$$\begin{aligned}\Delta(1/T_2) = A[& (1-\gamma_a^2) U_a^2 + (1-\gamma_b^2) U_b^2 + (1-\gamma_c^2) U_c^2 \\ & - 2U_a U_b \gamma_c^2 - 2U_b U_c \gamma_a^2 - 2U_a U_c \gamma_b^2] , \end{aligned} \quad (8)$$

where A is given by

$$\begin{aligned} A = {} & KTg^6 \mu_B^6 \hbar^{-2} \chi_T^{-1} N^{-2} \\ & \times \sum_{\vec{q}}{}' \int_0^\infty dt\, (S(\vec{q}+\vec{K}_0, t), S(\vec{q}+\vec{K}_0))^2 . \end{aligned} \quad (9)$$

Equation (8) is seen to have the effect of separating $\Delta(1/T_2)$ into two factors—an angular part which reflects the symmetry of the magnetic lattice and an isotropic but temperature-dependent part which incorporates the spin dynamics. In the language of mode-mode-coupling theory Eq. (8) characterizes a decay process where a fluctuation in the total magnetization decays into two fluctuations of the staggered magnetization via the dipolar interaction.

In Eq. (9) the dominant temperature dependence

is connected with the relaxation function. Experimental[4] and theoretical studies[5-7] carried out near T_N have shown that the relaxation function is adequately approximated by

$$(S(\vec{q}+\vec{K}_0, t),\ S(\vec{q}+\vec{K}_0)) \simeq \frac{\chi(0)^*}{g^2\mu_B^2}\ \frac{e^{-\Gamma_q t}}{1+q^2\xi^2}\ , \quad (10)$$

where Γ_q is of the form

$$\Gamma_q = \xi^{-3/2} f(q\xi)\ , \quad (11)$$

with $f(0)$ being finite. Equations (10) and (11), it should be noted, are appropriate only for those temperatures where the dipolar contribution to $(S(\vec{q}+\vec{K}_0, t),\ S(\vec{q}+\vec{K}_0))$ can be neglected. Inserting (10) and (11) into (9) leads to the result

$$A \propto (T/\chi_T)\xi^{5/2} \quad (12)$$

for the temperature dependence of $\Delta(1/T_2)$. The exponent $\frac{5}{2}$ is in agreement with the value reported by Kawasaki for nondipolar anisotropy.[6] It differs, however, from $\frac{3}{2}$, the value associated with the anomalous part of $1/T_2$ in ferromagnets.[1,6] (Using a somewhat different approach Kawasaki has also obtained values $\frac{1}{2}$ and $\frac{3}{2}$ for the exponents characterizing the EPR linewidth in ferro- and antiferromagnets, respectively.[8])

Equations (8) and (9) have a form identical to what would have been obtained with a single-ion anisotropy of the form

$$\tfrac{1}{2}g^2\mu_B^2\,(U_a S_a^2 + U_b S_b^2 + U_c S_c^2)\ ,$$

where $S_{a,b,c}$ refer to spin components along the principal axes. It must be emphasized that this equivalence applies only to $\Delta(1/T_2)$. Differences in the range of the two types of interactions cause the equivalence to break down at arbitrary points in the Brillouin zone. As a consequence, $1/T_2 - \Delta(1/T_2)$ calculated with the effective single-ion anisotropy will not, in general, be the same as that obtained with the true dipolar interaction.

III. EPR LINEWIDTH

The EPR linewidth is a measure of the relaxation rate for spin fluctuations perpendicular to the static field. The analysis of the linewidth is complicated by the effect of the field on the spin dynamics. In discussing this problem it is convenient to distinguish between extrinsic and intrinsic effects. The extrinsic effects are accounted for by inserting factors of $e^{\pm ig\mu_B H_0 t/\hbar}$ in the relaxation functions associated with spin components perpendicular to the static field. In addition, the linewidth has also to be evaluated with the appropriate exponential factors included, all other parameters remaining unchanged. The intrinsic effects are those which can not be accounted for by this procedure. Of primary importance among intrinsic effects is the influence of the field on the decay of the fluctuations of the staggered magnetization. Since relatively little is known about the intrinsic effects, we will not consider them in any detail except to point out where they appear to come into play. Fortunately, the extrinsic effects are far easier to deal with.

The calculation of the EPR linewidth can be carried out along the lines of the zero-field analysis. We make a separation similar to that outlined in Sec. II by writing the linewidth $\gamma(T)$ as

$$\gamma(T) \approx \Delta\gamma(T) + \gamma(\infty)\ , \quad (13)$$

where $\gamma(\infty)$ is the limiting value at high temperatures. Apart from intrinsic effects the field dependence of $\Delta\gamma(T)$ depends on the relative magnitudes of $g\mu_B H_0/\hbar$ and Γ_0, the decay rate for the superlattice point. We consider two limiting cases. In the long-correlation-time limit, $g\mu_B H_0/\hbar\Gamma_0 \gg 1$, only the secular part of the dipolar interaction contributes to the width.[9] In this case we have

$$\Delta\gamma(T) = A\,[U_{zz}(\vec{K}_0) - \tfrac{1}{2}U_{xx}(\vec{K}_0) - \tfrac{1}{2}U_{yy}(\vec{K}_0)]^2\ , \quad g\mu_B H_0/\hbar\Gamma_0 \gg 1\ , \quad (14)$$

where A is given by Eq. (9). The z axis is along the static field.

In the short-correlation-time limit, $g\mu_B H_0/\hbar\Gamma_0 \ll 1$, both the secular and nonsecular terms contribute to the broadening. The resulting linewidth is then given by the angular average of the zero-field rate in the plane perpendicular to $\vec{H}_0$. (The use of the angular average for the linewidth is equivalent to replacing the decay rate of the resonant component of the transverse magnetization by its time average over a Larmor period, an approximation which appears to be satisfactory for frequencies near the center of the line.)

The appropriateness of the short- or long-correlation-time limit must be decided for each individual system. It should be noted, however, that the linewidths in the two limits often have rather different angular dependences. As a consequence it may be possible to infer the correct limit from angular data alone.

IV. COMPARISON WITH EXPERIMENT

A. $RbMnF_3$

Measurements of the critical behavior of the EPR linewidth in $RbMnF_3$ have been reported by Gupta and Seehra.[10] They find that the linewidth narrows slightly as the critical point is approached from the high-temperature side. Relative to the high-temperature limit the width at $(T - T_N)/T_N = 0.1$ has decreased by 15% for microwave frequencies on the order of 9.2 GHz. For frequencies on the order of 24 GHz the decrease is much less, amounting to about 2% at the same temperature.

An examination of the relevant neutron data (Ref.

4) shows that the condition $g\mu_B H_0/\hbar\Gamma_0 \ll 1$ is satisfied for both frequencies for $(T-N)/T_N > 0.01$. Thus in this range the short-correlation-time limit is appropriate so that γ is given by the angular average of $1/T_2$. Because the magnetic lattice is simple cubic, the $U_{\alpha\beta}(\vec{K}_0)$ are all zero,[11] with the result that $\Delta(1/T_2)$ and $\Delta\gamma$ vanish identically. Thus the linewidth is predicted to be nondivergent, in agreement with experiment.

The slight narrowing reported in Ref. 10, which has been attributed to decay processes involving spin fluctuations near the center of the zone,[12] is a direct measure of the accuracy of the approximations embodied in Eq. (7). In the absence of accurate zero-field data we use the x-band result, 15%, as the estimated error for $(T-T_N)/T_N \gtrsim 0.1$. Although the narrowing becomes less pronounced for temperatures greater than $1.1T_N$, the increase in accuracy is only apparent since the approximations reflected in Eqs. (10) and (11) begin to break down in this region.

B. MnF_2

Measurements of the EPR linewidth in MnF_2 near T_N have been reported by Seehra and Castner[13] and Seehra.[14] It is found that the linewidth increases as the critical point is approached from the high-temperature side. The width for $\vec{H}_0$ parallel to the c axis is greater than the corresponding width with $\vec{H}_0$ perpendicular to $\vec{c}$. Below $(T-T_N)/T_N = 0.15$ the linewidth becomes field dependent.[14] Above this temperature the following behavior is observed:

$$\gamma(T)-\gamma(\infty) = R_{\parallel}(T-T_N)^{-1.20}, \quad \vec{H}_0 \parallel \vec{c} \tag{15}$$

$$\gamma(T)-\gamma(\infty) = R_{\perp}(T-T_{\perp})^{-1.17}, \quad \vec{H}_0 \perp \vec{c} \tag{16}$$

$$T_N - T_{\perp} = 1.2\,^{\circ}\mathrm{K}. \tag{17}$$

The results of extensive measurements of the parameters characterizing the relaxation functions in MnF_2 have been published recently by Schulhof *et al.*[15] For $(T-T_N)/T_N < 0.15$, it is found that the relaxation functions for fluctuations with wave vectors near $\vec{K}_0$ become anisotropic with the critical behavior limited to spin fluctuations parallel to the c axis. From the discussion in the preceding sections it is apparent that Eqs. (8) and (14) apply only outside this region, that is, $(T-T_N)/T_N \gtrsim 0.15$. Data on Γ_0, when extrapolated to temperatures greater than $1.15T_N$, indicate $g\mu_B H_0/\hbar\Gamma_0 \ll 1$ for realistic values of the field. Thus the short-correlation-time limit is again appropriate.

For MnF_2, which has a body-centered tetragonal lattice, the principal axes of $U_{\alpha\beta}(\vec{K}_0)$ coincide with the a, b, and c axes. Relative to these axes we have $U_a = U_b = U_{\perp}$ and $U_c = U_{\parallel}$. Thus the zero-field relaxation rate for spin fluctuations along a direction making an angle α with respect to the c axis takes the form

$$\Delta(1/T_2) = A(U_{\parallel}-U_{\perp})^2 \sin^2\alpha, \tag{18}$$

where A is given by Eq. (9). As noted, the anomalous part of the linewidth is obtained by averaging $\Delta(1/T_2)$ over directions in the plane perpendicular to $\vec{H}_0$. The result of this averaging can be written

$$\Delta\gamma = A(U_{\parallel}-U_{\perp})^2(1-\tfrac{1}{2}\sin^2\theta), \tag{19}$$

where θ is the angle between the external field and the c axis. From (19) it is evident that $\Delta\gamma(\vec{H}_0 \parallel \vec{c})/\Delta\gamma(\vec{H}_0 \perp \vec{c}) = 2$, which is in agreement with the experimental value at 77 °K (see note added in proof). It is worth pointing out that, were the long-correlation-time limit appropriate, the ratio would be 4 instead of 2.

Values for ξ, the correlation length, are also reported in Ref. 15. It is found that

$$\xi \propto (T-T_N)^{-0.63}, \quad (T-T_N)/T_N \leq 0.15. \tag{20}$$

Assuming this dependence to hold at higher temperatures we would have

$$A \propto (T/\chi_T)(T-T_N)^{-1.6}. \tag{21}$$

Since $\xi^2 \propto \chi(0)$* (approximately) it can be argued that away from T_N the critical exponent for ξ should approach the molecular field value $-\frac{1}{2}$ in which case we would have

$$A \propto (T/\chi_T)(T-T_N)^{-1.25}. \tag{22}$$

When allowance is made for the temperature variation of (T/χ_T), Eqs. (21) and (22) are found to bracket the data. It should be noted, however, that both (21) and (22) become less accurate with increasing $T-T_N$ and should not be taken seriously for $T \geq 2T_N$.

A further test of the theory comes from a numerical evaluation of $\Delta\gamma$ itself. This is carried out in the Appendix with the result

$$\Delta\gamma = 640 \pm 380\ \mathrm{Oe} \quad (\vec{H}_0 \parallel \vec{c},\ T = 77\,^{\circ}\mathrm{K}), \tag{23}$$

$$\Delta\gamma = 320 \pm 190\ \mathrm{Oe} \quad (\vec{H}_0 \perp \vec{c},\ T = 77\,^{\circ}\mathrm{K}). \tag{24}$$

These numbers, whose uncertainty reflects the uncertainties in the values of the empirical parameters characterizing the relaxation function, are to be compared with the experimental[16] values

$$\Delta\gamma = 180\ \mathrm{Oe} \quad (\vec{H}_0 \parallel \vec{c},\ T = 77\,^{\circ}\mathrm{K}), \tag{25}$$

$$\Delta\gamma = 110\ \mathrm{Oe} \quad (\vec{H}_0 \perp \vec{c},\ T = 77\,^{\circ}\mathrm{K}). \tag{26}$$

It is apparent that the agreement, while not precise, is at least semiquantitative and could be improved by a more realistic choice for the upper limit of the integral in Eq. (A1).

As has been pointed out, for $(T-T_N)/T_N < 0.15$ the linewidth becomes field dependent. The ques-

tion then arises as to whether the field dependence is an extrinsic or an intrinsic effect. We argue for the latter. Were the field dependence extrinsic it would come from the freezing out of the nonsecular terms in the dipolar interaction which accompanies the breakdown of the condition $g\mu_B H_0/\hbar\Gamma_0 \ll 1$. Since Γ_0 decreases as $\xi^{-3/2}$, such a freezing out might occur very close to T_N. Were this the cause of the field dependence there would be a change in the linewidth from a form described by Eq. (19) to a form characteristic of Eq. (14). However, both equations yield the same result for $\vec{H}_0$ parallel to the c axis, so there should be no extrinsic field dependence for fields in this direction, a result in contradiction to what is actually observed.

Our final comment on MnF_2 concerns the anisotropy in the relaxation function. In the linewidth measurements the anisotropy appears to be masked by the field dependence. Such is not the case for the zero-field time. To account for the anisotropy in the calculation of the relaxation time for the fluctuations perpendicular to the c axis, the integral in Eq. (9) must be replaced by

$$\int_0^\infty dt\, (S_\parallel(\vec{q}+\vec{K}_0, t),\ S_\parallel(\vec{q}+\vec{K}_0)) \times (S_\perp(\vec{q}+\vec{K}_0, t),\ S_\perp(\vec{q}+\vec{K}_0)) ,$$

where $\parallel$ and $\perp$ refer to spin components parallel and perpendicular to the c axis. An evaluation of $\Delta(1/T_2)$ using the modified expression for A leads to the conclusion that $\Delta(1/T_2)$ approaches a finite value in the limit as T approaches T_N, since $\Gamma_{\perp 0}$ and $\xi_\perp$ remain finite.

C. MnO and MnS

EPR linewidth measurements in MnO and MnS have been reported by Battles.[17] These compounds have an fcc magnetic lattice and undergo an ordering of the second kind, type A, where the spins are confined to (111) planes.[18] The superlattice vector is of the form $(\pi/a,\ \pi/a,\ \pi/a)$.[19] According to Fig. 2 of Ref. 17 the linewidths have divergent components which vary as $T/(T-T_N)$ for $T \geq T_N + 10\,^\circ$K.

The absence of neutron data comparable to those available for MnF_2 precludes quantitative analyses of these systems. [We note, however, that the observed temperature dependence is not very much different from that given by Eq. (22).] However, the most interesting aspect of the problem is not the rate of divergence but rather the existence of a divergence to begin with. As has been noted, the simple- (and body-centered-) cubic lattices show no divergence, $\Delta(1/T_2)$ and $\Delta\gamma$ being identically equal to zero.

The divergence in the fcc system arises because the tensor $U_{\alpha\beta}(\vec{K}_0)$ has the form

$$\begin{pmatrix} 0 & U & U \\ U & 0 & U \\ U & U & 0 \end{pmatrix}$$

in a coordinate system whose axes coincide with the [100] directions of the cubic lattice.[11] The principal-axis transformation reduces the above matrix to the form

$$\begin{pmatrix} -U & 0 & 0 \\ 0 & -U & 0 \\ 0 & 0 & 2U \end{pmatrix} ,$$

with c axis along one of the equivalent [111] directions. The zero-field relaxation time thus has the angular dependence

$$\Delta(1/T_2) = 9U^2(1-\gamma_c^2)A \tag{27}$$

for spin fluctuations along a direction making an angle $\cos^{-1}\gamma_c$ with respect to the closest [111] axis.

As noted, the EPR linewidth in the short-correlation-time limit is obtained by averaging $1/T_2$ over directions in the plane perpendicular to $\vec{H}_0$. The general analysis is complicated and will not be dealt with here. We consider only the cases where $\vec{H}_0$ is along the [100] and [112] directions. After some calculation we find

$$\Delta\gamma = 4.1U^2A \quad (\text{short-correlation time},\ \vec{H}_0 \parallel [100]) , \tag{28}$$

$$\Delta\gamma = 2.0U^2A \quad (\text{short-correlation time},\ \vec{H}_0 \parallel [112]) . \tag{29}$$

The nonzero value for $\Delta\gamma$ with $\vec{H}_0 \parallel [100]$ is to be compared with the corresponding result for long-correlation times:

$$\Delta\gamma = 0 \quad (\text{long-correlation time},\ \vec{H}_0 \parallel [100]) . \tag{30}$$

Since the experimental results indicate a divergence we conclude that the short-correlation-time limit is appropriate, at least for $T - T_N \gtrsim 10\,^\circ$K. We should mention, however, that the angular dependence of $\Delta\gamma$, as reflected in Eqs. (28) and (29), is stronger than what has been reported for MnS, where the ratio $\Delta\gamma[100]/\Delta\gamma[112]$ is slightly less than unity.[17]

V. FINAL COMMENTS

In the preceding sections we have outlined a theory for the critical anomalies in the zero-field relaxation rate and EPR linewidth in antiferromagnets. The essential feature of the theory is the association of the anomalous increase in these quantities with a process in which a fluctuation in the total magnetization decays into two fluctuations in the staggered magnetization via the dipolar cou-

pling. The increase occurs because near T_N both the correlation length and the lifetime of the fluctuations in the staggered magnetization increase rapidly with decreasing temperature. In this respect the behavior of $1/T_2$ in antiferromagnets and ferromagnets is similar, the principal difference being that in the latter system the decay is into fluctuations with wave vectors near the center of the zone.[1]

It has been noted that the decay process is strictly forbidden in an isotropic system since the total spin is a constant of the motion. This suggests that we may treat the relaxation in the spirit of perturbation theory, an approach followed in the derivation of Eq. (1) where it was assumed that the influence of the dipolar interaction on $(S(\vec{q}, t), S(\vec{q}))$ could be neglected. At the end of Sec. IV B it was pointed that this approximation cannot be made very close to T_N, where the anisotropic terms in the Hamiltonian have a pronounced effect on the dynamics of the staggered magnetization. When the anisotropy is allowed for, both $\Delta(1/T_2)$ and $\Delta\gamma$ remain finite at the critical point.

A second aspect of the theory which received some comment is that its applicability is limited to the critical region above the Néel point. At high temperatures the mode-mode-coupling approach breaks down and the linewidth is no longer dominated by the critical fluctuations. It was estimated that this happens at temperatures $T \gtrsim 2T_N$.

A third point is that the theory also applies, with obvious modifications, to the case of nondipolar anisotropy provided the anisotropy is sufficiently weak so as to maintain the exchange-narrowing limit. When this is not the case it is necessary to go beyond the exponential approximation for $(S(0, t), S(0))$, a point discussed in some detail in Ref. 2. (In easy-axis systems the exponential approximation is valid as long as $1/T_2 \ll \Gamma_{\perp 0}$, where $\Gamma_{\perp 0}$ is the decay rate for fluctuations perpendicular to the easy axis.)

Finally, we should like to emphasize that additional work is needed to refine the quantitative aspects of the theory. More must be learned about the temperature dependence, angular dependence, and magnitude of the contributions from the noncritical modes. Additional experimental work is also called for. It would be particularly useful to have accurate measurements of the zero-field relaxation time to avoid the complications of the intrinsic field dependence. A direct confirmation of the angular dependence of $\Delta(1/T_2)$ would be valuable at this point.

Note added in proof. Measurements of $\Delta\gamma$ in MnF_2 have been reported by Seehra (Phys. Rev., following paper) which confirm the angular dependence predicted by Eq. (19). An interesting feature of the data is that the ratio $\Delta\gamma(\vec{H}_0 \| \vec{c})/\Delta\gamma(\vec{H}_0 \perp \vec{c})$ remains equal to 2 down to within half a degree of the Néel point. This behavior is a consequence of the fact that in tetragonal symmetry (and $g\mu_B H_0/\hbar\Gamma_0 \ll 1$) we have

$$\Delta\gamma(\vec{H}_0 \| \vec{c}) = \Delta(1/T_2(\perp)) ,$$

while

$$\Delta\gamma(\vec{H}_0 \perp \vec{c}) = \tfrac{1}{2}[\Delta(1/T_2(\|)) + \Delta(1/T_2(\perp))] ,$$

where the parentheses associated with $1/T_2$ indicate directions relative to the c axis. Since $\Delta(1/T_2(\|)) = 0$ [Eq. (18)] we obtain the result

$$\Delta\gamma(\vec{H}_0 \| \vec{c})/\Delta\gamma(\vec{H}_0 \perp \vec{c}) = 2 ,$$

which is valid even at temperatures where the $(S_\alpha(\vec{K}_0, t), S_\alpha(\vec{K}_0))$ are anisotropic. This analysis of course neglects intrinsic field effects, which appear to be unimportant at χ-band frequencies for $T - T_N \geq \frac{1}{2}$ °K.

APPENDIX

In this Appendix we outline a numerical calculation of the anomalous part of the linewidth in MnF_2. In this analysis we make full use of the experimental information about the relaxation functions which was reported in Ref. 15. Assuming an exponential decay for the relaxation function as in Eq. (10) and extending the upper limit on the q sum to infinity leads directly to the result

$$\Delta\gamma = \frac{KTg^2\mu_B^2\chi(0)^{*2}(U_\| - U_\perp)^2(1 - \frac{1}{2}\sin^2\theta)}{4\pi^2\hbar^2(c/av)\chi_T N} \times \int_0^\infty \frac{\hat{q}^2 d\hat{q}}{(1+\hat{q}^2\xi^2)^2\Gamma_{\hat{q}}} , \quad \text{(A1)}$$

where $\hat{q}^2 = q_x^2 + q_y^2 + (c/a)^2 q_z^2$, c and a denote lattice constants, and v is the volume per spin. We evaluate this expression at $T = (T_N + 10)$ °K, which is approximately the lower limit of the isotropic regime. At this temperature we have[15]

$$\chi(0)^*/\chi_c = 13 \pm 3 , \quad \text{(A2)}$$

where χ_c is the susceptibility of the noninteracting lattice evaluated at T_N. Also, it is found that[15]

$$\xi = 7.3 \pm 0.9 \text{ Å} \quad \text{(A3)}$$

and

$$\Gamma_{\hat{q}} = 0.35[1 + 0.38(\hat{q}\xi)^2 + \cdots] \text{ meV} \quad \text{(A4)}$$

at the same temperature. Values of $U_\| - U_\perp$ can be inferred from the work of Keffer.[20] According to Ref. 20 we have

$$(U_\| - U_\perp)^2 = 50/v^2 . \quad \text{(A5)}$$

With $v = 39$ Å^3, $a/c = 1.5$, $T_N = 67.5$ °K, χ_T/χ_c

$= 0.39$, and $g = 2$ it is found that

$$\Delta\gamma = 640 \pm 380 \text{ Oe} \quad (\vec{H}_0 \| \vec{c},\ T = 77\,^\circ\text{K}), \qquad \text{(A6)}$$

$$\Delta\gamma = 320 \pm 190 \text{ Oe} \quad (\vec{H}_0 \perp \vec{c},\ T = 77\,^\circ\text{K}), \qquad \text{(A7)}$$

which are the results quoted in the main text.

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Role of Anisotropy in the Critical-Point Anomaly in EPR Linewidth of MnF_2 †

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EPR linewidth measurements in MnF_2 are reported near the Néel temperature T_N as a function of orientation and temperature. The angular dependence of the temperature-dependent part of the linewidth for $T \lesssim T_N + 10\,^\circ$K follows the curve $(1 - \frac{1}{2}\sin^2\theta)$, in agreement with the prediction of a recent theory by Huber ($\theta = 0$ for magnetic field $\vec{H} \| \vec{c}$ axis). The observed breakdown of the Huber theory for $T > T_N + 10\,^\circ$K is briefly discussed.

I. INTRODUCTION

Recent experimental studies of the electron-paramagnetic-resonance (EPR) linewidth in antiferromagnets have shown that the anisotropy plays a fundamental role in the nature of the anomaly observed near the Néel temperature T_N. For example, in the uniaxial antiferromagnet MnF_2,[1] the EPR line broadens as $T \to T_N^+$, whereas in the cubic antiferromagnets $RbMnF_3$,[2] $KMnF_3$,[3] and $KMn:MgF_3$[3] the EPR line narrows slightly in the same temperature range. Earlier attempts to quantitatively explain the EPR line broadening in MnF_2 were only partially successful.[4] In the preceding paper[5] (hereafter referred to as I) Huber has presented a theory which clearly brings out the role played by the anisotropy in the critical-point anomalies in the EPR linewidth in antiferromagnets. In this paper we present the first experimental evidence which quantitatively verifies several aspects of the Huber theory.

II. REVIEW OF HUBER'S THEORY

In the calculations given in I it is assumed that the dominant spin-spin interaction is the isotropic exchange coupling of the Heisenberg type and that the dipole-dipole interaction is the source of anisotropy. Manganese fluoride fits such a Hamiltonian to an excellent approximation. For our purposes, the starting point is an equation for the zero-field relaxation rate $1/T_2$ derived by Huber using the random-phase approximation (RPA). This equation is

$$1/T_2 = KTg^6\mu_B^6\hbar^{-2}\chi^{-1}N^{-2}\sum_{\vec{q}}\{[U_{xx}(\vec{q}) - U_{yy}(\vec{q})]^2 + 4U_{xy}^2(\vec{q}) + U_{xz}^2(\vec{q}) + U_{yz}^2(\vec{q})\}\int_0^\infty dt\,(S(\vec{q},t), S(\vec{q}))^2\,, \qquad (1)$$