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Hydrodynamic modes in the spin-flop phase of an antiferromagnet*

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A linear theory of the hydrodynamic modes in the spin-flop phase of an antiferromagnet is outlined. The theory applies to systems with weak anisotropy in the basal plane. A variety of dynamical correlation functions are calculated in the adiabatic limit. The theory is used to interpret recent nuclear-magnetic-resonance experiments in MnF_2 by King and Rohrer which show evidence of a low-lying electronic mode near the spin-flop–paramagnetic phase boundary. The mode is identified as a zone-center magnon which is coupled to the nuclear spins through off-diagonal components of the dynamic electronic susceptibility tensor.

I. INTRODUCTION

In a recent paper we have outlined a quasihydrodynamic theory for the dynamics of a uniaxial antiferromagnet in a longitudinal external field.[1] The theory applied to the paramagnetic and antiferromagnetic phases where the magnetization, the staggered magnetization (when different from zero), and the field were collinear. In this paper we extend the analysis to the spin-flop phase. In this phase the staggered magnetization is perpendicular to the magnetization and the applied field. An important stimulus for this work comes from recent measurements carried out on MnF_2 by King and Rohrer.[2] Using nuclear-magnetic-resonance (NMR) techniques they were able to obtain indirect information about the frequency and linewidth of a low-lying electronic mode. We will show that our approach provides a natural interpretation of their experiment, leading to the identification of the mode as a hydrodynamic spin wave at the center of the Brillouin zone. In addition to the interpretation of the NMR experiments we obtain expressions for the dynamical correlation functions characterizing the various hydrodynamic variables. It should be emphasized that the present theory includes only linear terms in the hydrodynamic equations. Nonlinear interactions among the variables are expected to be important near the phase boundaries but probably can be neglected otherwise. Furthermore, even near the boundaries they should not affect the qualitative features of many of our results.

Insofar as its hydrodynamic behavior is concerned, a uniaxial antiferromagnet in the flop phase closely resembles a planar ferromagnet in a longitudinal field. There is thus some overlap of our work with earlier studies of the hydrodynamics of planar magnets.[3] Most of these, however, were limited to zero field. To our knowledge previous investigations of the hydrodynamics of a planar magnet in a finite field have not involved calculations of the correlation functions, which is an important feature of this work.

As in Ref. 1 our analysis is based on kinetic equations of the form proposed by Mori.[4] With x defining the direction of the average staggered moment and z the direction of the applied field, the relevant dynamical variables are the transverse components of the staggered magnetization, $N_x(q)$ and $N_y(q)$ (q denotes wave vector in a Fourier transform over lattice sites), and the longitudinal component of the direct magnetization, $M_z(q)$. In addition we must also consider the energy density $E(q)$. We do not include the momentum density of the magnons since we assume that the temperature is sufficiently high so that umklapp processes bring about rapid relaxation of the quasimomentum. We postulate that the spin Hamiltonian has approximate rotational symmetry about the direction of the field so that $M_z(0)$ is close to being a constant of the motion. As will be discussed below this appears to be a very good approximation in MnF_2.

II. THEORY

In developing the theory it will be convenient to work with the normalized variables A_i defined by

$$A_1 = N_x/(N_x, N_x)^{1/2} , \quad (1)$$

$$A_2 = N_y/(N_y, N_y)^{1/2} , \quad (2)$$

$$A_3 = M_z/(M_z, M_z)^{1/2} , \quad (3)$$

and

$$A_4 = E/(E, E)^{1/2} , \quad (4)$$

where we have suppressed the dependence on q. The symbol (a, b) denotes a "susceptibility" inner product defined by

$$(a, b) = \int_0^\beta \langle e^{\lambda H} a e^{-\lambda H} b^\dagger \rangle \, d\lambda - \beta \langle a \rangle \langle b^\dagger \rangle , \quad (5)$$

where $\beta = 1/kT$, T being the temperature and the dagger denotes adjoint. The symbol H is the Hamiltonian and the brackets refer to a thermal average.

In the Mori formalism the A_i are elements of a column vector $\vec{A}$ which obeys the equation

$$\frac{d\vec{A}(t)}{dt} = i\,\vec{\omega}\cdot\vec{A}(t) - \int_0^t ds\,\vec{\phi}(t-s)\cdot\vec{A}(s) + \vec{f}(t). \tag{6}$$

In this equation $\vec{\omega}$ is a frequency matrix with elements

$$\vec{\omega}_{ij} = -i\sum_k \left(\frac{dA_i}{dt}, A_k\right) V_{kj}, \tag{7}$$

where $\vec{V}$ is the inverse of the generalized susceptibility matrix $\vec{U}$ defined by

$$U_{ij} = U_{ji} = (A_i, A_j). \tag{8}$$

Also, $\vec{f}(t)$ designates a random force which is orthogonal to $\vec{A}$ in the sense[4]

$$(f_i(t), A_j(t')) \equiv 0, \tag{9}$$

while $\vec{\phi}$ is a matrix defined by

$$[\vec{\phi}(t)]_{ij} = \sum_k (f_i, f_k(-t))V_{kj}. \tag{10}$$

Unlike the paramagnetic and antiferromagnetic phases,[1] spin hydrodynamics in the flop phase is characterized by finite values for certain of the elements of $\vec{\omega}$. To see this we make use of the identity ($\hbar = 1$)

$$\left(\frac{dA_i}{dt}, A_j\right) = -i\langle[A_i, A_j^\dagger]\rangle. \tag{11}$$

Since we have

$$[N_x(q), N_y(q)^\dagger] = ig\mu_B M_z(0),$$
$$[N_y(q), M_z^\dagger(q)^\dagger] = ig\mu_B N_x(0),$$

where g is the electronic g factor and μ_B is the Bohr magneton, we obtain the equation

$$i\vec{\omega} = \begin{bmatrix} 0 & \alpha_1 & 0 & 0 \\ -\alpha_1 V_{11} + \alpha_2 V_{13} & 0 & -\alpha_1 V_{13} + \alpha_2 V_{33} & -\alpha_1 V_{14} + \alpha_2 V_{34} \\ 0 & -\alpha_2 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \tag{12}$$

where α_1 and α_2 are defined by

$$\alpha_1 = g\mu_B\langle M_z(0)\rangle/[(N_y,N_y)(N_x,N_x)]^{1/2}, \tag{13}$$

$$\alpha_2 = g\mu_B\langle N_x(0)\rangle/[(N_y,N_y)(M_z,M_z)]^{1/2}. \tag{14}$$

We have also made explicit use of the result $V_{i2} = V_{2i} = \delta_{i2}$. This follows from the equation

$$U_{i2} = U_{2i} = \delta_{i2}, \tag{15}$$

which is a consequence of the fact that y and $-y$ are equivalent directions.

In order to obtain a proper characterization of the dynamics it is necessary to take into account the relaxation terms in the kinetic equation. An approximate treatment of the dampling involves replacing the damping terms on the right-hand side of Eq. (6) by the vector $-\vec{\Gamma}\cdot\vec{V}\cdot\vec{A}$, where $\vec{\Gamma}$ is a diagonal matrix.[1] The elements of $\vec{\Gamma}$, Γ_{N_x}, Γ_{N_y}, Γ_{M_z}, and Γ_E, characterize the intrinsic decay rates of the corresponding fluctuations and are expressed formally by an integral of the form[4]

$$\int_0^\infty dt\,(f_i, f_i(-t)).$$

It should be noted that the diagonal form for $\vec{\Gamma}$ is in itself an approximation. More general $\vec{\Gamma}$ are allowed with both diagonal and off-diagonal elements. However, in view of the uncertainties about the relative sizes of the elements a more complicated form for the damping matrix appears to be an unnecessary refinement at this stage of the development of the theory.

In the analysis we make the conventional assumption that Γ_{N_x} is large in comparison with α_1, α_2, Γ_{N_y}, Γ_{M_z}, and Γ_E.[3] Since these parameters characterize the spin-wave frequency and linewidth and the decay rate of the energy fluctuations this assumption is equivalent to postulating that $\Gamma_{N_x}^{-1}$ is the smallest characteristic time in the problem. Although it is usually not possible to obtain reliable theoretical estimates of the elements of $\vec{\Gamma}$, the consequences of this hypothesis can be tested experimentally, at least in principle.

The relative magnitude of Γ_E is also a matter of importance. If Γ_E is greater than the spin-wave frequency, the spin waves are isothermal modes whereas if the opposite holds the modes are adiabatic. As will be discussed below, there is indirect evidence in the case of MnF_2 which suggests that the response at $q = 0$ is adiabatic. The wave-vector dependence of the decay rates Γ_{N_y} and Γ_{M_z} reflects the symmetry of the Hamiltonian. If there is perfect symmetry Γ_{N_y} and Γ_{M_z} vary as q^2, the latter because $M_z(0)$ is a constant of the motion, the former because $\Gamma_{N_y} \propto (N_y, N_y)^{-1}$ and (N_y, N_y) varies as q^{-2} for small q. In the absence of exact rotational symmetry a variation of the form $a_i + b_i q^2$ is predicted for both Γ_{N_y} and Γ_{M_z}.

Omitting the noise term and utilizing the matrix $\overleftrightarrow{\Gamma}$ to characterize the damping, we obtain an approximate kinetic equation of the form

$$\frac{d}{dt}\vec{A}(t) = i\overleftrightarrow{\omega}\cdot\vec{A}(t) - \overleftrightarrow{\Gamma}\cdot\overleftrightarrow{V}\cdot\vec{A}(t). \tag{16}$$

Equation (16), the principal result of this section, is the starting point for the analysis outlined in Secs. III and IV.

III. DYNAMIC SUSCEPTIBILITIES AND CORRELATION FUNCTIONS

In order to obtain expressions for the dynamic susceptibilities we make use of the equation[5]

$$\chi_{A_iA_j}(\omega) = \chi_{A_iA_j} - i\omega\int_0^\infty e^{-i\omega t}(A_i(t), A_j)\,dt, \tag{17}$$

where we have identified (A_i, A_j) with the static susceptibility $\chi_{A_iA_j}$. The integral can be computed by taking the one-sided Fourier transform of (16) and expressing the transformed variables in terms of the A_k, viz.,

$$\vec{A}(\omega) = \int_0^\infty e^{-i\omega t}\,\vec{A}(t)\,dt = (i\omega\overleftrightarrow{I} - i\overleftrightarrow{\omega} + \overleftrightarrow{\Gamma}\cdot\overleftrightarrow{V})^{-1}\cdot\vec{A}, \tag{18}$$

where $\overleftrightarrow{I}$ denotes the unit matrix. Equation (17) then becomes

$$\chi_{A_iA_j}(\omega) = \chi_{A_iA_j} - i\omega([(i\omega\overleftrightarrow{I} - i\overleftrightarrow{\omega} + \overleftrightarrow{\Gamma}\cdot\overleftrightarrow{V})^{-1}\cdot\vec{A}]_i, A_j). \tag{19}$$

Equation (19) is appropriate for all values of the parameters. As was mentioned, in the interpretation of the data on MnF_2 we are particularly interested in the behavior when $\Gamma_{N_x} \gg$ spin-wave frequency $\gg \Gamma_E$. As a consequence we will limit our detailed analysis to this regime. In this section we calculate the diagonal elements of the dynamic susceptibility tensor. These elements are conveniently represented in terms of the line-shape functions $f_{A_i}(\omega)$ defined by

$$f_{A_i}(\omega) = \chi''_{A_iA_i}(\omega)/\pi\omega\chi_{A_iA_i}. \tag{20}$$

As a result of the Kramers-Kronig relations the f_{A_i} have the normalization property

$$\int_{-\infty}^{\infty} d\omega f_{A_i}(\omega) = 1. \tag{21}$$

After some straightforward but tedious calculations we obtain the results

$$f_{A_1}(\omega) = \frac{1}{\pi}\left[\frac{\Gamma_{N_x}}{\omega^2 + (\Gamma_{N_x}V_{11})^2} + \frac{1}{2}(1 - V_{11}^{-1} - U_{14}^2)\left(\frac{\Delta}{(\omega-\omega_0)^2+\Delta^2} + \frac{\Delta}{(\omega+\omega_0)^2+\Delta^2}\right) + \frac{U_{14}^2\Gamma_E}{\omega^2+\Gamma_E^2}\right], \tag{22}$$

$$f_{A_2}(\omega) = \frac{1}{2\pi}\left(\frac{\Delta}{(\omega-\omega_0)^2+\Delta^2} + \frac{\Delta}{(\omega+\omega_0)^2+\Delta^2}\right), \tag{23}$$

$$f_{A_3}(\omega) = \frac{1}{\pi}\left[\tfrac{1}{2}(1 - U_{34}^2)\left(\frac{\Delta}{(\omega-\omega_0)^2+\Delta^2} + \frac{\Delta}{(\omega+\omega_0)^2+\Delta^2}\right) + \frac{U_{34}^2\Gamma_E}{\omega^2+\Gamma_E^2}\right], \tag{24}$$

$$f_{A_4}(\omega) = \frac{1}{\pi}\,\frac{\Gamma_E}{\omega^2+\Gamma_E^2}. \tag{25}$$

In these equations ω_0, the hydrodynamic spin-wave frequency, is given by

$$\omega_0^2 = (\alpha_2^2/V_{11})(V_{11}V_{33} - V_{13}^2), \tag{26}$$

and the spin-wave damping Δ by

$$\Delta = \tfrac{1}{2}[\Gamma_{N_y} + \Gamma_{M_z}V_{11}^{-1}(V_{11}V_{33} - V_{13}^2)], \tag{27}$$

for $\Delta \ll \omega_0$, which follows from the assumption that the anisotropy in the basal plane is small.

In order to interpret these equations we make use of the result that in the small-q limit the matrix $\overleftrightarrow{U}$ can be written in terms of the derivatives of the free energy.[1] We have

$$\overleftrightarrow{U} = \overleftrightarrow{V}^{-1} = \overleftrightarrow{C}\cdot\overleftrightarrow{\mathfrak{F}}\cdot\overleftrightarrow{C}, \tag{28}$$

where

$$C_{ij} = \delta_{ij}\mathfrak{F}_{ii}^{-1/2}, \tag{29}$$

and

$$\mathfrak{F}_{ij} = -\frac{\partial^2 F}{\partial x_i \partial x_j}. \tag{30}$$

In (30) we have $x_1 = H_x^*$, $x_2 = H_y^*$, $x_3 = H_z$, and $x_4 = T$ with H^* denoting the staggered field and H_z the uniform field, all thermodynamic derivatives being evaluated with $H^* = 0$. (Strictly speaking, when there is rotational symmetry about the z axis $\chi_{N_x N_x}$ as well as $\chi_{N_y N_y}$ diverges as $q \to 0$. Since all real magnetic systems are anisotropic to some degree their susceptibilities are finite in the small-q limit.)

From Eqs. (28)–(30) we have

$$V_{ij} = C_{ii}^{-1} (\overleftrightarrow{\mathfrak{F}}^{-1})_{ij} C_{jj}^{-1} . \tag{31}$$

Using standard thermodynamic analysis[6] we can express the matrix $\overleftrightarrow{\mathfrak{F}}^{-1}$ in terms of the second derivatives of the internal energy with respect to the extensive thermodynamic variables conjugate to H_x^*, H_y^*, H_z, and T. We have

$$(\mathfrak{F}^{-1})_{ij} = \frac{\partial^2 W}{\partial X_i \partial X_j} , \tag{32}$$

where $X_1 = \overline{N}_x$, $X_2 = \overline{N}_y$, $X_3 = \overline{M}_z$, and $X_4 = S$ (S being the entropy), while the two thermodynamic energies are related by

$$F = W - \sum_i x_i X_i . \tag{33}$$

From Eqs. (28)–(32) we have

$$V_{11} = \left(\frac{\partial \overline{N}_x}{\partial H_x^*}\right)_{H_z, T} \bigg/ \left(\frac{\partial \overline{N}_x}{\partial H_x^*}\right)_{\overline{M}_z, S} , \tag{34}$$

$$U_{14}^2 = 1 - \left(\frac{\partial \overline{N}_x}{\partial H_x^*}\right)_{H_z, S} \bigg/ \left(\frac{\partial \overline{N}_x}{\partial H_x^*}\right)_{H_z, T} , \tag{35}$$

$$U_{34}^2 = 1 - \left(\frac{\partial \overline{M}_z}{\partial H_z}\right)_{H_x^*, S} \bigg/ \left(\frac{\partial \overline{M}_z}{\partial H_z}\right)_{H_x^*, T} , \tag{36}$$

and

$$V_{11}^{-1}(V_{11}V_{33} - V_{13}^2) = \left(\frac{\partial \overline{M}_z}{\partial H_z}\right)_{H_x^*, T} \bigg/ \left(\frac{\partial \overline{M}_z}{\partial H_z}\right)_{H_x^*, S} . \tag{37}$$

As a consequence of (37) Eq. (26) can be written

$$\omega_0^2 = (g\mu_B \overline{N}_x)^2 / (N_y, N_y) \left(\frac{\partial \overline{M}_z}{\partial H_z}\right)_{H_x^*, S} , \tag{38}$$

a result analogous to that obtained for the planar magnet.[7]

Thus when $\Gamma_E \ll \omega_0$ the square of the spin-wave frequency is inversely proportional to the adiabatic direct susceptibility at fixed staggered field, whereas in the opposite limit it is inversely proportional to the isothermal direct susceptibility. Likewise the term involving Γ_{M_z} in the equation for Δ is renormalized by the ratio of isothermal to adiabatic direct susceptibilities whereas were the response isothermal there would be no renormalization factor.

Equation (22) is seen to contain three types of terms: a fast relaxation term with relative weight V_{11}^{-1} and linewidth $V_{11}\Gamma_{N_x}$, a thermal decay[7,8] term with width Γ_E and relative weight U_{14}^2, and a spin-wave term of relative weight $1 - V_{11}^{-1} - U_{14}^2$. The fast term characterizes the decay of the spin fluctuations at constant magnetization and entropy, the spin-wave part a decay at constant staggered field and entropy, and the thermal part a decay at constant staggered and uniform field.

Equation (23) contains only spin-wave terms. The thermal term is absent because $\overline{N}_y$ and hence $\partial \overline{N}_y / \partial T$ are equal to zero. On the other hand, $\partial \overline{M}_z / \partial T$ is in general different from zero. As a consequence there is a thermal term in f_{A_3} of relative weight U_{34}^2. Finally, Eq. (25) shows that the energy fluctuations relax at a rate which is independent of the dynamics of N_x, N_y, and M_z, provided Γ_E is the smallest characteristic frequency.

Equations (22)–(24) are potentially useful in the interpretation of inelastic-neutron-scattering data. They are expected to be valid throughout the spin-flop region as long as $\Gamma_{N_x} V_{11} \gg \omega_0 \gg \Delta, \Gamma_E$. Near the spin-flop–paramagnetic phase boundary nonlinear terms in the hydrodynamic equations become important. Nonlinear processes may affect the relative weights of the different terms in (22) and (24). In addition they will influence the temperature dependence of the decay rates, which would follow the conventional theory of critical slowing down were the linear theory exact.

IV. NMR STUDIES IN MnF_2

In the work described in Ref. 2 the ^{19}F NMR in MnF_2 was investigated. In a configuration with the oscillating field parallel to the z axis a strong resonance was observed in the flop state which was characterized by a phase that underwent a continuous change through nearly 2π as the temperature was lowered below T_c. The amplitude showed a broad maximum about 1 K below the transition temperature, dropping rapidly to zero at T_c. The authors interpret their results in terms of a resonant enhancement model in which the frequency of a low-lying electronic mode coupling to the rf field passes from above the nuclear resonance frequency ω_n to below ω_n as the transition is approached from the low-temperature side. By using the Bloch equations and the NMR enhancement factor given by molecular-field theory, they were able to infer both the frequency and the linewidth of the electronic mode.

In this section we will outline an interpretation of this experiment based on the hydrodynamic model developed in the preceding sections. We will show that the electronic mode seen indirectly in the NMR experiment can be identifed with a hydrodynamic spin wave whose frequency is given by Eq. (38).

The coupling of the nuclear spins to the electronic system is through the hyperfine interaction. If we neglect the "back reaction" of the nuclear system on the electronic spins we can view the hyperfine coupling as an effective field acting on the nuclei. It is the rf component of the hyperfine field perpendicular to the direction of the total (external plus hyperfine) static field which is responsible for the enhanced absorption. Immediately below T_c, as in the paramagnetic phase, the total static field at the nuclear sites is parallel to the z axis. As a consequence it is the oscillating component of the hyperfine field perpendicular to the z axis which is induced by an rf field parallel to the z axis that causes the absorption near the phase boundary.

In terms of the variables $\vec{N}$ and $\vec{M}$ the transverse hyperfine field at a nucleus coupling isotropically to spins on the A sublattice is proportional to $\vec{M}+\vec{N}$, whereas were the coupling to the B sublattice it would be proportional to $\vec{M}-\vec{N}$. Of the components of $\vec{M}_\perp$ and $\vec{N}_\perp$ only N_x has a finite thermal average in the flop phase. As a consequence all the static off-diagonal susceptibilities of the form $\chi_{N\perp Mz}$ and $\chi_{M\perp Mz}$ vanish except χ_{NxMz}. Hence the effective transverse rf field which is induced by a spatially uniform oscillating field of frequency ω_n along the z axis is determined largely by the $q=0$ limit of the off-diagonal electronic susceptibility $\chi_{NxMz}(\omega_n)$.

We can obtain an approximate expression for $\chi_{NxMz}(\omega_n)$ from Eq. (19) evaluated in the limit $\Gamma_{N_x} \gg \omega_0, \Gamma_E, \Delta$. The magnitude of Γ_E relative to ω_0 and Δ is somewhat of an open question. Estimates of ω_0 and Δ given in Ref. 2 are $\omega_0 \lesssim \Delta \simeq 2-6$ GHz. To our knowledge there have been no direct measurements of Γ_E^{-1} at $q=0$ in MnF_2, which we identify with the electronic spin-lattice relaxation time T_1. However, indirect information about the relaxation time can be obtained from ultrasonic attenuation data.[9] For MnF_2 these lead to the estimate $T_1 \approx 3\times 10^{-9}$ sec for $T\approx T_c, H_z=0$.[10] Adopting this value for the relaxation time in the flop phase we obtain $\max(\omega_0,\Delta)T_1 \geq 40$. As a consequence the response of the system at $q=0, \omega\approx\omega_0$ appears to be adiabatic, so that the limit $\Gamma_E \ll \omega_0, \Delta$ is probably justified.

In the low-frequency regime $\omega_n \ll \Gamma_{N_x} V_{11}$ we obtain from Eq. (19) the expression

$$\chi_{N_xM_z}(\omega_n)=\left(\frac{\partial \bar{N}_x}{\partial H_z}\right)_{H_x^*,T}\left(1-\frac{\omega_n(\omega_n^2-\omega_0^2U_{14}U_{34}U_{13}^{-1})}{(\omega_n-i/T_1)(\omega_n-\omega_0-i\Delta)(\omega_n+\omega_0-i\Delta)}\right), \tag{39}$$

assuming $\Delta \ll \omega_0$. For $\omega_n \approx \omega_0 \gg 1/T_1$ Eq. (39) reduces to

$$\begin{aligned}\chi_{N_xM_z}(\omega_n)&=-\left(\frac{\partial \bar{N}_x}{\partial H_z}\right)_{H_x^*,T}\frac{\omega_0(1-U_{14}U_{34}U_{13}^{-1})}{2(\omega_n-\omega_0-i\Delta)},\\&=-\left(\frac{\partial \bar{N}_x}{\partial H_z}\right)_{H_x^*,S}\frac{\frac{1}{2}\omega_0}{(\omega_n-\omega_0-i\Delta)},\end{aligned} \tag{40}$$

the last step following from the thermodynamic identity

$$1-U_{14}U_{34}U_{13}^{-1}=\left(\frac{\partial \bar{N}_x}{\partial H_z}\right)_{H_x^*,S}\Big/\left(\frac{\partial \bar{N}_x}{\partial H_z}\right)_{H_x^*,T}. \tag{41}$$

Equation (40) indicates that the enhancement factor for the NMR is proportional to the off-diagonal adiabatic electronic susceptibility. That it is the adiabatic rather than the isothermal susceptibility is a consequence of the assumption $\Gamma_E \ll \omega_0$. Were the inequality to go the other way, i.e., $\Gamma_E \gg \omega_0$, the enhancement would be proportional to $(\partial\bar{N}_x/\partial H_z)_{H_x^*,T}$ while the spin-wave frequency and damping would be given by the isothermal counterparts of (27) and (38):

$$\omega_0^2=(g\mu_B\bar{N}_x)^2\Big/(N_y,N_y)\left(\frac{\partial \bar{M}_z}{\partial H_z}\right)_{H_x^*,T}, \tag{42}$$

$$\Delta=\tfrac{1}{2}(\Gamma_{N_y}+\Gamma_{M_z}). \tag{43}$$

Also of interest is the phase lag of the electronic system ϕ_e defined by the ratio

$$\tan\phi_e=\chi'/\chi''=\Delta/(\omega_0-\omega_n), \tag{44}$$

which is the form hypothesized in Ref. 2.

Strictly speaking, when the spin-wave mode is overdamped, as occurs very close to T_c in MnF_2, Eqs. (40) and (44) have to be modified to include contributions from both the resonance and antiresonance terms. In the case of (40) the modification takes the form

$$\chi_{N_xM_z}(\omega_n)=\left(\frac{\partial \bar{N}_x}{\partial H_z}\right)_{H_x^*,S}\times\frac{\Gamma_{Mz}^R(i\omega_n+\Gamma_{Ny})+\omega_0^2}{\omega_0^2-\omega_n^2+i\omega_n(\Gamma_{N_y}+\Gamma_{M_z}^R)+\Gamma_{N_y}\Gamma_{M_z}^R}, \tag{45}$$

where

$$\Gamma^R_{M_z} = \Gamma_{M_z}\left(\frac{\partial \overline{M}_z}{\partial H_z}\right)_{H_x^*,T} \Big/ \left(\frac{\partial \overline{M}_z}{\partial H_z}\right)_{H_x^*,S} . \tag{46}$$

From (46) it is evident that in the limit $\omega_0 \to 0$ the dependence on Γ_{N_y} disappears leaving

$$\chi_{N_xM_z}(\omega_n) = \left(\frac{\partial \overline{N}_x}{\partial H_z}\right)_{H_x^*,S} \frac{\Gamma_{M_z}{}^R}{\Gamma^R_{M_z} + i\omega_n} , \tag{47}$$

which is the Debye approximation to a nonresonant susceptibility.

When $\omega_n \approx \omega_0$ one has also to consider the y component of the rf hyperfine field, which is determined by $\chi_{N_yM_z}(\omega_n)$. From Eq. (19) we obtain the result

$$\chi_{N_yM_z}(\omega_n) = \frac{ig\mu_B \overline{N}_x \omega_n^2}{(\omega_n - i/T_1)(\omega_n - \omega_0 - i\Delta)(\omega_n + \omega_0 - i\Delta)} . \tag{48}$$

The off-diagonal susceptibility $\chi_{N_yM_z}(\omega_n)$ vanishes in the static limit. On the other hand, when $\omega_n \approx \omega_0$ $\chi_{N_yM_z}(\omega_n)$ and $\chi_{N_xM_z}(\omega_n)$ behave similarly. With Eqs. (40) and (48) we have

$$\frac{\chi_{N_xM_z}(\omega_n)}{\chi_{N_yM_z}(\omega_n)} \approx \frac{i(\partial \overline{N}_x/\partial H_z)_{H_x^*,S}}{[(N_y,N_y)(\partial \overline{M}_z/\partial H_z)_{H_x^*,S}]^{1/2}} , \tag{49}$$

for $\omega_n \approx \omega_0$. In the overdamped case we find

$$\frac{\chi_{N_xM_z}(\omega_n)}{\chi_{N_yM_z}(\omega_n)} \approx \frac{i(\partial \overline{N}_x/\partial H_z)_{H_x^*,S}[\omega_0^2 + \Gamma^R_{M_z}(i\omega_n + \Gamma_{N_y})]}{g\mu_B \overline{N}_x \omega_n} , \tag{50}$$

when $\omega_n \gg 1/T_1$.

It is beyond the scope of this paper to consider in detail the temperature dependence of the various parameters appearing in the expressions for the susceptibility. However, we point out that theory (and experiment) indicates that $\partial \overline{N}_x/\partial H_z$ is large near the phase boundary and zero in the paramagnetic phase. The interpretation of the temperature dependence of the frequency and linewidth is made complicated by the fact that the small but finite value of ω_0 at $q = 0$ is indicative of a weak anisotropy in the basal plane. As a consequence the characteristic frequency of the $q = 0$ mode in the critical region will not have the scaling properties of the characteristic frequency of a hydrodynamic mode in a system with perfect rotational symmetry about the z axis, where $\omega_0 \propto q$ at long wavelengths.

In the interpretation of the data given in Ref. 2 ω_0 extrapolates to zero at T_c, whereas Δ remains finite. From Eqs. (27) and (38) it is evident that such behavior is consistent with $(\partial \overline{M}_z/\partial H_z)_{H_x^*,S}$ and (N_y, N_y) remaining finite as $T \to T_c$. General thermodynamic arguments[11] indicate that $(\partial \overline{M}_z/\partial H_z)_{H_x^*,S}$ is finite along a line of antiferromagnetic critical points, although singularities are possible at isolated points, e.g., the bicritical point. In contrast $(\partial \overline{M}_z/\partial H_z)_{H_x^*,T}$ has a specific-heat-like singularity in a finite field.[12] As mentioned, the finite value of (N_y, N_y) is a consequence of the anisotropy in the basal plane. Thus it appears that the ratio of $\chi_{N_xM_z}(\omega_n)$ to $\chi_{N_yM_z}(\omega_n)$ [Eqs. (49) and (50)] is likely to be much greater than 1 so that the y component of the effective rf field can probably be neglected as a first approximation.

Our final comment concerns the question of adiabatic versus isothermal response. It is our opinion that a definite answer to this question requires additional experiments. A possible approach involves the measurement of $\chi_{M_zM_z}(\omega_n)$ at low frequencies.[13] According to Eq. (24) if the adiabatic limit is appropriate there will be a thermal part in $\chi_{M_zM_z}$ of relative weight $U_{34}{}^2$ and width $1/T_1$. On the other hand if the isothermal limit is appropriate this term will be absent.

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