Hydrodynamics of easy-axis antiferromagnets in a magnetic field: Tricritical behavior*

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A linear hydrodynamic theory for the spin dynamics of easy-axis antiferromagnets in an applied magnetic field is outlined, with particular emphasis on the behavior along the second-order line including the region near a tricritical point. Expressions are obtained for the ω,q-dependent spin susceptibilities. In the paramamagnetic phase, the dynamic staggered susceptibility is characterized by a single peak at $\omega = 0$. In the antiferromagnetic phase, at long wavelengths, a three-peak structure is found: two narrow peaks centered at $\omega = 0$ with widths proportional to q^2, which are associated with the coupled magnetization-energy fluctuations and a broad peak, also at $\omega = 0$, which characterizes the adiabatic, isomagnetic decay of the staggered moment. The relative weight of the broad peak is equal to the ratio of the adiabatic staggered susceptibility at constant magnetization to the isothermal staggered susceptibility at constant field. The coupling of the magnetization-energy fluctuations also affects the frequency dependence of the uniform field susceptibility. The temperature dependence of various parameters in the theory is discussed in light of static scaling laws and a brief comparison is made with the hydrodynamics of antiferromagnets in and near a spin-flop phase.

I. INTRODUCTION

In recent years considerable attention has been paid to phenomena connected with so-called tricritical points. The term tricritical, introduced by Griffiths,[1] refers to a point in an appropriate thermodynamic phase space which is the intersection of three lines of second-order transitions. In addition to the familiar He^3-He^4 mixtures, Griffiths also called attention to the fact that tricritical points may be found in certain antiferromagnets which undergo metamagnetic transitions; e.g., dysprosium aluminum garnet (DAG), $FeCl_2$, $Ni(NO_3)\cdot 2H_2O$.

Up to now the emphasis in tricritical work has largely been on obtaining an understanding of the thermodynamic features of the transition, as distinct from the dynamical characteristics. Since an understanding of the dynamics must rest inevitably upon thermodynamic foundations, the lack of attention paid to the dynamics is understandable. However, in the past few years considerable progress has been made in tricritical thermodynamics through scaling law,[2] renormalization group,[3,4] and numerical studies.[5,6] In view of this, it seems appropriate to begin to develop corresponding theories of tricritical dynamics.

To our knowledge, the only work on tricritical dynamics which has appeared in the literature is that of Kawasaki and Gunton[7] who formulated a mode coupling theory for He^3-He^4 mixtures. In this paper we outline a theory for the dynamics of uniaxial antiferromagnets which show tricritical behavior. In spite of the similarities in the tricritical thermodynamics, dynamical phenomena in antiferromagnets differ significantly from dynamical phenomena in the mixtures. As a consequence, our approach and results are altogether different from those of Ref. 7.

Briefly, our work is an extension of previous studies of the critical dynamics of uniaxial easy-axis antiferromagnets[8-10] to the situation where there is a magnetic field along the easy axis. In the analysis, particular emphasis is placed on the behavior near a tricritical point. It is a quasi-hydrodynamic theory which carries with it the understanding that it characterizes only the long-wavelength low-frequency response of the system. The regions of applicability of the theory are difficult to determine precisely; however, we expect the theory to be limited to frequencies $\omega \ll \min \times (KT, \omega_{ex})$, where ω_{ex} is a characteristic exchange frequency, and where wavelengths are much greater than the lattice spacing. Also, the theory is phenomenological in spirit. That is, expressions obtained for the dynamical susceptibilities involve both thermodynamic functions, which can be calculated provided the thermodynamic equation of state is known, and dissipative parameters characterizing the relaxation of the fluctuations in the thermodynamic variables. Although formal expressions for the relaxation rates are given, reliable quantitative estimates appear to be impossible to obtain at this time. Thus the usefulness of the theory, in addition to giving a qualitative picture of the dynamics, lies in providing functional forms which can be fit to experimental data in order to determine empirical values of the parameters.

Hydrodynamic theories such as ours probably

will be most useful in interpreting the results of inelastic-neutron-scattering experiments. Unfortunately, scattering studies of tricritical behavior in antiferromagnets have so far been either difficult to interpret (DAG)[11] or else hampered by experimental difficulties ($FeCl_2$).[12] As a consequence there are as yet no data to which we can apply our theory. However, it is hoped that the availability of a hydrodynamic theory will stimulate experiments to probe tricritical dynamics.

II. PRELIMINARY ANALYSIS

We begin the development of the hydrodynamic theory with a discussion of the systems to which it may be applicable. As anticipated in the Introduction, we shall be concerned with antiferromagnets which in zero field undergo second-order transitions from a paramagnetic to an antiferromagnetic state. We assume that the anisotropy is such that the system has an easy axis of staggered magnetization, which is to say that in the ordered state in zero field the electronic moments are aligned either parallel or antiparallel to a particular crystallographic axis, which we refer to as the c axis. We also postulate that the (internal) magnetic field, when present, is also along the c axis. (Hereafter, magnetic field will be taken to mean internal magnetic field.)

It should be noted that all uniaxial easy-axis antiferromagnets do not display tricritical behavior. Many antiferromagnets show evidence of a low-temperature spin-flop phase in high magnetic fields. For reasons which will become apparent, our theory does not apply to antiferromagnets in or near (in the sense of phase space) the spin-flop phase.[13]

A further assumption pertains to the rotational symmetry of the Hamiltonian. We postulate that the dominant terms in the spin Hamiltonian are invariant with respect to rotations about the c axis. This assumption, which is appropriate for systems like $FeCl_2$, has the consequence that the longitudinal component of the total spin, or magnetization, is an approximate constant of the motion. This symmetry leads to a significant simplification in the analysis of the kinetic equations in the long wavelength limit. However, certain aspects of the analysis are independent of this assumption, as will be made clear below. In addition to the rotational symmetry, we assume that the spin system is weakly coupled to the crystal lattice so that we may speak of a local spin temperature distinct from the lattice temperature.

As mentioned in the Introduction, the theory outlined here represents an extension of earlier theories of the critical hydrodynamics of uniaxial antiferromagnets[8-10] to situations where a magnetic field is present. Because of this it is worthwhile to review the findings of Refs. 8–10. This we do now.

In formulating a hydrodynamic theory it is necessary to identify the appropriate hydrodynamic variables. In the case of uniaxial easy-axis antiferromagnets with the aforementioned rotational symmetry, these are the longitudinal (i.e., parallel to the c axis) component of the magnetization, and the energy density, both of which, being approximately conserved, have long wavelength fluctuations which are slowly decaying in time. In addition, near the Néel point, the longitudinal component of the staggered magnetization undergoes critical slowing down with the consequence that it, too, must be counted among the hydrodynamic variables.

The hydrodynamic equations lead to expressions for the dynamic susceptibilities, $\chi_{AA}(q,\omega)$, functions of "wave vector" q and frequency ω, which are associated with the variable A. In zero field the magnetization of an antiferromagnet is zero both above and below the Néel temperature T_N. As a consequence, the fluctuations in the longitudinal component of the magnetization $M(q)$, are decoupled from the fluctuations in the energy density $E(q)$, and the staggered magnetization $N(q)$. [In the case of $M(q)$ and $E(q)$, q is a measure of the distance from the center of the Brillouin zone. In the case of $N(q)$, it is a measure of the distance from the antiferromagnetic superlattice point.] In the hydrodynamic regime the imaginary part of the corresponding dynamic susceptibility has a Lorentzian form:

$$\frac{\chi''_{MM}(q,\omega)}{\omega\chi_{MM}(q)} = \frac{D_M q^2}{\omega^2 + (D_M q^2)^2}, \quad T \gtrless T_N \tag{1}$$

where D_M is identified with the spin diffusion constant. [Here and in subsequent usage q^2 will stand for the more general form $a_1 q_x^2 + a_2 q_y^2 + a_3 q_z^2$.]

In contrast, when $T < T_N$ the fluctuations in the staggered magnetization are coupled to the fluctuations in the energy density since $\partial N/\partial T \neq 0$, N denoting the staggered magnetization. This coupling leads to a two-component expression for $\chi''_{NN}(q,\omega)$:

$$\frac{\chi''_{NN}(q,\omega)}{\omega\chi_{NN}(q)} = \left(\frac{\chi_S^*}{\chi_T^*}\right)\frac{\Gamma_N^*}{\omega^2 + (\Gamma_N^*)^2} + \left(1 - \frac{\chi_S^*}{\chi_T^*}\right)\frac{D_E q^2}{\omega^2 + (D_E q^2)^2}, \tag{2}$$

where χ_T^* is the isothermal staggered susceptibility, χ_S^* is the adiabatic staggered susceptibility, Γ_N^* denotes the adiabatic decay rate of the staggered magnetization, and D_E is the thermal diffusion constant for the magnetic lattice. Above T_N,

$\chi_S^* = \chi_T^*$ so that only the first term of (2) is present.

Equation (2) is seen to describe a decay process characterized by two time constants, fast adiabatic decay at the rate Γ_N^* followed by the slow diffusive decay of the induced energy density or, equivalently, temperature fluctuations. Experimental evidence supporting this equation comes from inelastic-neutron-scattering studies of MnF_2 (Ref. 14) and FeF_2.[15] However, it should be mentioned that below T_N only the slow relaxing component of χ''_{NN} has been observed; it is believed that the adiabatic part is obscured by the background.

A magnetic field along the c axis gives rise to a magnetization M which is also a function of the staggered field H^* and the temperature. As a consequence $M(q)$ becomes coupled to $N(q)$ in the ordered state in addition to being coupled to $E(q)$ both above and below the transition. Because of this coupling $\chi''_{MM}/\omega\chi_{MM}$ becomes the sum of two Lorentzians each having a width proportional to q^2, which characterize the evolution of the coupled $M(q)$-$E(q)$ modes.

Similarly, in the ordered state $\chi''_{NN}/\omega\chi_{NN}$ involves three Lorentzians, a fast relaxing term with relative weight equal to the ratio of the adiabatic staggered susceptibility at constant magnetization to the isothermal staggered susceptibility at constant field, and two slow relaxing terms with widths proportional to q^2 which are associated with the coupled $M(q)$-$E(q)$ modes. In the paramagnetic state the diffusive terms are absent so that the fast relaxing part has unit relative weight. This form is seen to be a generalization of the zero field result, Eq. (2), which comes about because below T_N the staggered magnetization is coupled to two conserved variables, the longitudinal component of the magnetization and the energy density.

In Sec. III we develop a mathematical description of the hydrodynamics in the field corresponding to the physical picture outlined above. In Sec. IV we present the results of the analysis of a simple model which yields analytic expressions for χ''_{NN} and χ''_{MM}. In Sec. V we comment on the temperature dependence of χ'' with reference to the second-order line and the tricritical point. Finally, a brief comparison is made with the hydrodynamics associated with the spin-flop phase.

III. KINETIC EQUATIONS

In order to develop a mathematical description of tricritical spin dynamics we make use of the kinetic equations proposed by Mori.[16] In the Mori formalism a set of appropriate dynamical variables is singled out along with a corresponding inner product. Identifying the variables with the elements of a column vector $\vec{A}(t)$ Mori showed that $\vec{A}(t)$ satisfies the exact equation of motion

$$\vec{A}' - i\underline{\tilde{\omega}} \cdot \vec{A} + \int_0^t ds\, \underline{\tilde{\phi}}(t-s) \cdot \vec{A}(s) = \vec{f}(t), \tag{3}$$

where

$$\vec{A}' = \frac{d\vec{A}}{dt}. \tag{4}$$

The symbol $\underline{\tilde{\omega}}$ denotes the frequency matrix

$$i(\underline{\tilde{\omega}})_{ij} = \sum_k (\vec{A}', \vec{A}^\dagger)_{ik} ((\vec{A}, \vec{A}^\dagger)^{-1})_{kj}, \tag{5}$$

in which $(\vec{A}, \vec{A}^\dagger)^{-1}$ is the inverse of the generalized susceptibility matrix defined by

$$(\vec{A}, \vec{A}^\dagger)_{ij} = (A_i, A_j^\dagger), \tag{6}$$

where the dagger implies Hermitian conjugate. The symbol (a, b) will be used to denote the inner product:

$$(a, b) = \int_0^\beta d\lambda \langle e^{\lambda \mathcal{H}} a e^{-\lambda \mathcal{H}} b\rangle - \beta \langle a\rangle \langle b\rangle, \tag{7}$$

where $\mathcal{H}$ is the Hamiltonian, $\beta = 1/KT$, and the brackets denote a statistical average.

If a projection operator $\mathcal{P}$ onto the space spanned by the dynamical variables is defined by

$$\mathcal{P}\vec{G} = (\vec{G}, \vec{A}^\dagger) \cdot (\vec{A}, \vec{A}^\dagger)^{-1} \cdot \vec{A}, \tag{8}$$

where $\vec{G}$ is an arbitrary vector, and $(\vec{G}, \vec{A}^\dagger)$ is defined in a manner analogous to $(\vec{A}, \vec{A}^\dagger)$, then $\vec{f}(t)$, the "random force," can be written

$$\vec{f}(t) = \exp[it(1-\mathcal{P})\mathcal{L}](1-\mathcal{P})\vec{A}', \tag{9}$$

in which $\mathcal{L}$ is the Liouville operator associated with the Hamiltonian $\mathcal{H}$. The vector $\vec{f}(t)$ can be shown to be orthogonal to $\vec{A}$ in the sense

$$(\vec{f}(t), \vec{A}^\dagger) = 0. \tag{10}$$

Also, in Eq. (3) the symbol $\underline{\tilde{\phi}}(t)$ denotes a matrix defined by

$$\underline{\tilde{\phi}}(t) = (\vec{f}, \vec{f}(-t)^\dagger) \cdot (\vec{A}, \vec{A}^\dagger)^{-1}. \tag{11}$$

In applying the Mori formalism it is necessary to identify the appropriate dynamical variables. The analysis of Sec. II suggests $M(q)$, $E(q)$, and $N(q)$ as the appropriate set for uniaxial antiferromagnets. Two comments are appropriate here. First, by omitting products such as $E(q_1)M(q_2)$, $M(q_1)M(q_2)$ we are in effect formulating a linear theory of tricritical dynamics. Since the linear theory appears to be a satisfactory first approximation for easy-axis magnets in zero field, including nonlinear effects at this level of analysis,

seems unwarranted. The second comment pertains to the absence of the transverse components of the magnetization and staggered magnetization. We argue for their omission on the following grounds. The postulated absence of rotational symmetry about axes perpendicular to the c axis has the consequence that fluctuations in the transverse components of the total spin decay rapidly on a hydrodynamic time scale. Similarly, it is argued that since it is postulated that the system is not in or near a spin-flop phase, the transverse staggered static susceptibilities do not show divergent behavior. As a result, fluctuations in the transverse components of the staggered magnetization do not undergo thermodynamic slowing down and hence are not to be included among the hydrodynamic variables. (Note that this argument does not rule out nonhydrodynamic transverse spin waves.) In addition, by making use of the identity[17]

$$(A_i'(q),\, A_j(q)^\dagger) = i\langle [A_i(q),\, A_j(q)^\dagger]\rangle\,, \tag{12}$$

it can be shown that with our choice of variables the frequency matrix $\underline{\omega}$ is equal to zero.

In applying Eq. (3) to the variables $N(q)$, $M(q)$, and $E(q)$ it is convenient to replace the variables by their normalized counterparts

$$A_1(q) = N(q)/(N(q),\, N(q)^\dagger)^{1/2}\,, \tag{13}$$

$$A_2(q) = M(q)/(M(q),\, M(q)^\dagger)^{1/2}\,, \tag{14}$$

$$A_3(q) = E(q)/(E(q),\, E(q)^\dagger)^{1/2}\,. \tag{15}$$

Equation (3) with $\underline{\omega}=0$ and $\vec{A}$, as given by Eqs. (13)–(15) are the fundamental equations in the theory. By taking the inner product of $\vec{A}(t)$ with $\vec{A}^\dagger$ we obtain the relaxation matrix $(\vec{A}(t),\, \vec{A}^\dagger)$ which, in turn, can be related to the imaginary part of the dynamic susceptibility through the equation[18]

$$\chi''_{A_i A_i}(q,\omega)/\omega\chi_{A_i A_i}(q) = \mathrm{Re}\int_0^\infty e^{-i\omega t}(\vec{A}(t),\, \vec{A}^\dagger)_{ii}\,dt\,, \tag{16}$$

where Re denotes real part.

Since $(\vec{f}(t),\, \vec{A}^\dagger)=0$ it is evident that $\vec{f}(t)$ contributes to $(\vec{A}(t),\, \vec{A}^\dagger)$ only through $\vec{\phi}(t)$. Because $\vec{\phi}$ cannot be calculated exactly, further approximations are required. These fall into two categories. The first pertains to the matrix $(\vec{A},\, \vec{A}^\dagger)$. The diagonal elements of $(\vec{A},\, \vec{A}^\dagger)$ are equal to 1 since the A_i are normalized. We approximate the off-diagonal elements by their limiting values as $q\to 0$. Strictly speaking, this limits the applicability of the theory to wavelengths which are much greater than any of the correlation lengths associated with the static susceptibilities $\chi_{A_i A_j}(q)$. Such an approximation is compatible with the hydrodynamic character of the theory, and can always be relaxed should it prove necessary in applications. As a result of the approximation, we can write the off-diagonal elements of $(\vec{A},\, \vec{A}^\dagger)$ in terms of thermodynamic functions. We have

$$(\vec{A},\, \vec{A}^\dagger)_{ii} = 1,\quad (\vec{A},\, \vec{A}^\dagger)_{ij} = (\vec{A},\, \vec{A}^\dagger)_{ji}\,, \tag{17}$$

$$(\vec{A},\, \vec{A}^\dagger)_{12} = \left(\frac{\partial N}{\partial H}\right)_T \Big/ (\chi_T^* \chi_T)^{1/2}\,, \tag{18}$$

$$(\vec{A},\, \vec{A}^\dagger)_{13} = T^{1/2}\left(\frac{\partial N}{\partial T}\right)_H \Big/ (\chi_T^* C_H)^{1/2}\,, \tag{19}$$

$$(\vec{A},\, \vec{A}^\dagger)_{23} = T^{1/2}\left(\frac{\partial M}{\partial T}\right)_H \Big/ (\chi_T C_H)^{1/2}\,, \tag{20}$$

where χ_T is the isothermal uniform field susceptibility, χ_T^* is the isothermal staggered susceptibility at constant field, and C_H is the specific heat at constant field. It is also understood that $\partial N/\partial H$, $\partial N/\partial T$, $\partial M/\partial T$, C_H, and χ_T are to be evaluated at fixed staggered field.

The second approximation pertains to the matrix $(\vec{f},\vec{f}(-t)^\dagger)$. By construction, $\vec{f}$ is orthogonal to $\vec{A}$. Hence if $\vec{A}$ includes all the hydrodynamic variables, then $(\vec{f},\vec{f}(-t)^\dagger)$ will decay rapidly in comparison with $(\vec{A}(t),\vec{A}^\dagger)$. This suggests that we approximate the elements of $(\vec{f},\vec{f}(-t)^\dagger)$ by terms of the form const $\times\,\delta(t)$ with the consequence that Eq. (3) takes the form

$$\vec{A}' = -\,\underline{\Gamma}\cdot(\vec{A},\vec{A}^\dagger)^{-1}\cdot\vec{A}\,, \tag{21}$$

having omitted $\vec{f}(t)$. The Hermitian matrix $\underline{\Gamma}$ is related to $(\vec{f},\vec{f}(-t)^\dagger)$ by

$$\underline{\Gamma} = \int_0^\infty dt\,(\vec{f},\vec{f}(-t)^\dagger)\,. \tag{22}$$

Equation (21) leads to expressions for $\chi''/\omega\chi$ which are essentially Lorentzian in form with frequency-independent widths. This approximation, also, can be relaxed in a straightforward way by allowing for a finite decay time for $(\vec{f},\vec{f}(-t)^\dagger)$. However, because of the success in fitting the zero field data to Lorentzian forms[14,15] such an elaboration does not appear to be required at this time.

Although independent of time, the matrix $\underline{\Gamma}$ is a function of q. Since $M(q=0)$ and $E(q=0)$ are constant apart from basal plane anisotropy and spin-lattice effects, the corresponding elements of $\underline{\Gamma}$, Γ_{2j}, and Γ_{3j}, $j=1,2,3$, must vanish as $q\to 0$. However, we expect Γ_{11} to remain finite in the same limit since $N(q=0)$ does not commute with the Hamiltonian.

IV. MODEL CALCULATIONS

A. Analysis

The calculation of the relaxation functions $(A_i(t), A_j^\dagger)$ from Eq. (21) presents no particular difficulties. However, instead of presenting the results in full generality we choose to make the further approximation of neglecting the off-diagonal elements of $\vec{\vec{\Gamma}}$. Such an approximation simplifies the algebra while preserving the essential features of the physics. Thus we take $\vec{\vec{\Gamma}}$ to be of the form

$$\vec{\vec{\Gamma}} = \begin{pmatrix} \Gamma_N & 0 & 0 \\ 0 & D_M q^2 & 0 \\ 0 & 0 & D_E q^2 \end{pmatrix} . \tag{23}$$

Noting that Γ_{22} and Γ_{33} are zero when $q=0$ (Sec. III), we have assumed that Γ_{22} and Γ_{33} vanish as q^2. The constants D_M and D_E are identified with the (bare) spin and energy diffusion constants, respectively, while Γ_N is the isothermal, constant field decay rate of the staggered magnetization. We assume that Γ_N, D_M, and D_E are q independent, an approximation in keeping with our treatment of $(\vec{A}, \vec{A}^\dagger)$. It should be remembered, however, that all three parameters are functions of temperature, a point we return to in Sec. V. In addition, should it be necessary, the effects of basal plane anisotropy and the spin-lattice coupling can be included by adding $1/T_2$ to $D_M q^2$ and $1/T_1$ to $D_E q^2$, where T_2 and T_1 are spin-spin and spin-lattice relaxation times, respectively.

Our primary interest is in the susceptibilities $\chi_{NN}(q,\omega)$ and $\chi_{MM}(q,\omega)$ associated with $N(q)$ and $M(q)$, respectively. In the paramagnetic phase $N(q)$ is decoupled from $M(q)$ and $E(q)$. Making use of Eqs. (16), (21), and (23) we find

$$\chi''_{NN}(q,\omega)/\omega\chi_{NN}(q) = \Gamma_N/[\omega^2 + (\Gamma_N)^2] , \tag{24}$$

a result which is actually independent of the treatment of the off-diagonal elements of $\vec{\vec{\Gamma}}$.

In the antiferromagnetic phase the dynamics is more complicated since, as noted in Sec. II, $N(q)$ couples to both $M(q)$ and $E(q)$. A significant simplification results if it is further assumed that q is sufficiently small so that $D_E q^2$, $D_M q^2 \ll \Gamma_N$. In this limit we have

$$\begin{aligned} \frac{\chi''_{NN}(q,\omega)}{\omega\chi_{NN}(q)} = {} & \left(\frac{1}{V_{11}}\right)\frac{\underline{\Gamma}_N}{\omega^2 + (\underline{\Gamma}_N)^2} \\ & - \left(\frac{V_{12}}{V_{11}}\right) \mathrm{Re}\left(\frac{U_{12}(i\omega + \underline{D}_E q^2) + U_{23}U_{13}\underline{D}_M q^2}{(i\omega + s_+)(i\omega + s_-)}\right) \\ & - \left(\frac{V_{13}}{V_{11}}\right) \mathrm{Re}\left(\frac{U_{13}(i\omega + \underline{D}_M q^2) + U_{23}U_{12}\underline{D}_E q^2}{(i\omega + s_+)(i\omega + s_-)}\right) , \end{aligned} \tag{25}$$

where the matrices $\vec{\vec{U}}$ and $\vec{\vec{V}}$ are defined by

$$\vec{\vec{U}} = (\vec{A}, \vec{A}^\dagger) , \tag{26}$$

$$\vec{\vec{V}} = \vec{\vec{U}}^{-1} = (\vec{A}, \vec{A}^\dagger)^{-1} . \tag{27}$$

Also we have

$$\underline{\Gamma}_N = V_{11}\Gamma_N , \tag{28}$$

$$\underline{D}_M = (1 - U_{23}^2)^{-1} D_M , \tag{29}$$

$$\underline{D}_E = (1 - U_{23}^2)^{-1} D_E . \tag{30}$$

The $s_\pm$ denote the decay rates of the coupled $M(q)$-$E(q)$ fluctuations and are given by

$$s_\pm = (q^2/2)\{\underline{D}_M + \underline{D}_E \pm [(\underline{D}_M - \underline{D}_E)^2 + 4U_{23}^2\underline{D}_M\underline{D}_E]^{1/2}\} . \tag{31}$$

Because of the uniform field, $M(q)$ and $E(q)$ are coupled in both the paramagnetic and antiferromagnetic phases. In the antiferromagnetic phase, when $\Gamma_N \gg D_M q^2$, $D_E q^2$, we obtain the result

$$\frac{\chi''_{MM}(q,\omega)}{\omega\chi_{MM}(q)} = \mathrm{Re}\left(\frac{i\omega + U_{23}^2\underline{D}_M q^2 + \underline{D}_E q^2}{(i\omega + s_+)(i\omega + s_-)}\right) , \tag{32}$$

where the symbols have the same meaning as in Eq. (25). Equation (32) also applies to the paramagnetic state independent of the relative magnitudes of Γ_N, $D_E q^2$, and $D_M q^2$.

B. Discussion

Equations (24), (25), and (32) are the principal results of Sec. IV A. Equation (24) is seen to have the same form as Eq. (2) when the latter is applied to the paramagnetic phase. In order to establish that (25) and (32) reduce to results obtained previously when the uniform field is equal to zero we note that $H=0$ implies $M=0$ so that $U_{23}=0$. As a consequence, Eq. (32) becomes identical to Eq. (1). Likewise $\partial N/\partial H$, which is equal to $\partial M/\partial H^*$, also vanishes when $H=0$ with the consequence that $U_{12}=0$. Hence in this limit $V_{11} = 1/(1 - U_{13}^2)$, $V_{12}=0$, and $V_{13} = -U_{13}/(1 - U_{13}^2)$, where U_{13}^2 can be written

$$U_{13}^2 = T\left(\frac{\partial N}{\partial T}\right)^2 \Big/ \chi_T^* C_H = (\chi_T^* - \chi_S^*)/\chi_T^* . \tag{33}$$

With the help of (33) it is readily established that (25) reduces to (2) for $H=0$, $T < T_N$. In general, when $H \neq 0$ all elements of $\vec{\vec{U}}$ will be nonzero in the antiferromagnetic phase.

The physical interpretation of Eqs. (24), (25), and (32) is similar to that outlined in the discussion in Sec. II. In the paramagnetic state the fluctuations in N are decoupled from the fluctuations in M and E. As a consequence, χ''_{NN} has the simple Lorentzian form shown in Eq. (24). In the antiferromagnetic state N couples to M and E

which in turn are coupled to each other. This coupling shows up in the form of Eq. (25). The second and third terms characterize the diffusive decay of the induced $M(q)$-$E(q)$ fluctuations. The coupling between the magnetization and energy density, which also is evident in Eq. (32), arises from the nonzero value of U_{23}^2. From Eq. (20) the parameter U_{23}^2 is seen to be equal to $(\chi_T - \chi_S)/\chi_T$, where χ_S is the adiabatic uniform field susceptibility at constant staggered field.

Since $\vec{\vec{V}} \cdot \vec{\vec{U}} = \vec{\vec{1}}$, the relative weight of the fast relaxing term in Eq. (25) is equal to $1/V_{11}$. In order to interpret this result we introduce the free energy $dF = -SdT - MdH - N\,dH^*$ which is a function of the intensive variables $H^*(=x_1)$, $H(=x_2)$, and $T(=x_3)$. We also define the elements of a matrix $\vec{\vec{\mathfrak{F}}}$ by

$$\mathfrak{F}_{ij} = -\left(\frac{\partial^2 F}{\partial x_i \partial x_j}\right)_{x_{k\neq i,j}} . \tag{34}$$

In terms of $\mathfrak{F}$, V_{11} can be written

$$V_{11} = \mathfrak{F}_{11}(\vec{\vec{\mathfrak{F}}}^{-1})_{11} . \tag{35}$$

Using standard thermodynamic arguments it can be shown that elements of the matrix $\vec{\vec{\mathfrak{F}}}^{-1}$ can be expressed as derivatives of the internal energy W with respect to the conjugate extensive variables $X_1 = N$, $X_2 = M$, and $X_3 = S$. The relation is[19]

$$(\vec{\vec{\mathfrak{F}}}^{-1})_{ij} = \left(\frac{\partial^2 W}{\partial X_i\, \partial X_j}\right)_{X_{k\neq i,j}} , \tag{36}$$

where

$$dW = X_1\,dx_1 + X_2\,dx_2 + X_3\,dx_3 , \tag{37}$$

and W and F are connected by the equation

$$F = W - x_1 X_1 - x_2 X_2 - x_3 X_3 . \tag{38}$$

Making use of (34) and (36) we obtain the result

$$V_{11} = \left(\frac{\partial N}{\partial H^*}\right)_{T,H} \Big/ \left(\frac{\partial N}{\partial H^*}\right)_{S,M} , \tag{39}$$

with the consequence that the relative weight of the fast relaxing part is equal to the ratio of the adiabatic staggered susceptibility at constant magnetization to the isothermal staggered susceptibility at constant field. It should be noted that relative weight is independent of the off-diagonal elements of $\vec{\vec{\Gamma}}$ as long as $\Gamma_{11} \gg \Gamma_{ij}$ $(ij \neq 11)$.

Another feature of Eqs. (25) and (32) is the renormalization of the decay rates. In particular, Γ_N is renormalized by the factor V_{11}. Since thermodynamic stability requires[19] $U_{ij}^2 < 1$ $(i \neq j)$ and $\det \vec{\vec{U}} \neq 0$, it follows that $V_{11} \geq 1$ so that $\underline{\Gamma}_N > \Gamma_N$. Likewise D_M and D_E are renormalized by the factor $(1 - U_{23}^2)^{-1}$ which is equal to χ_T/χ_S. Since $\chi_T \geq \chi_S$ we have $\underline{D}_M(\underline{D}_E) \geq D_M(D_E)$ as well.

Another aspect of our results is that the evolution of the coupled $M(q)$-$E(q)$ modes is independent of the dynamics of the staggered magnetization when $\Gamma_N \gg D_M q^2$, $D_E q^2$. This is apparent in Eq. (31) where the decay rates of the coupled modes are seen to depend on thermodynamic functions evaluated at constant staggered field as opposed to constant staggered magnetization.

V. TEMPERATURE DEPENDENCE

In the studies described in Secs. III and IV, no specific mention was made of the temperature dependence of the various parameters in the expressions for χ''. The analysis was based largely on the symmetry properties of the Hamiltonian. As a consequence the applicability of Eq. (32) is not limited to temperatures and fields near the transition line. Moreover, the presence of the second and third terms on the right-hand side of Eq. (25) is a general feature of the hydrodynamic region independent of the phase transition. [The Lorentzian form for the first term in (25) is probably appropriate only when there is critical slowing down of the staggered moment. When this is not the case the frequency dependence of this term may be more complicated; however the relative weight will still be given by $(\partial N/\partial H^*)_{S,M}/(\partial N/\partial H^*)_{T,H}$.]

Since the primary focus in this paper is on the dynamics near a tricritical point, we will limit our comments on the temperature dependence to the region near the transition curve with particular emphasis on the behavior along the second-order line including the tricritical point, where critical slowing down is expected to occur. The temperature dependence of $\chi''/\omega\chi$ arises from two types of parameters, the U_{ij} (and V_{ij}) and the decay rates Γ_{ij}. We consider the former first.

Although detailed statements can be made only for systems where the equation of state is known, considerable information can be obtained by making use of the static scaling laws for the critical exponents.[20] Along the second-order line we expect singular behavior in the specific heat and the longitudinal staggered susceptibility. In addition, Riedel has shown that there are induced critical fluctuations in the magnetization such that $M - M_c(H) \propto d_c^{1-\alpha'}$ and $\partial M/\partial H \propto d_c^{-\alpha'}$, where α' is the specific-heat exponent and d_c is a measure of the distance from the second-order line in the physical plane $H^* = 0$.[2] Because of this we obtain the important result that if scaling holds then the nonzero off-diagonal elements of $\vec{\vec{U}}$ are all characterized by a critical exponent equal to zero. To see this we associate the exponent γ' with χ_T^*,

$\frac{1}{2}\Delta' + \alpha' - 1$ with $\partial N/\partial H$, and $-\beta + 1$ with $\partial N/\partial T$. We then have (for $\alpha' \geq 0$)

$$U_{12}^2 = \left(\frac{\partial N}{\partial H}\right)^2 \Big/ \chi_T \chi_T^* \propto d_c^{\,-\Delta - 2(\alpha'-1)+\gamma'+\alpha'}, \quad (40)$$

$$U_{13}^2 = T\left(\frac{\partial N}{\partial T}\right)^2 \Big/ \chi_T^* C_H \propto d_c^{\,2(\beta-1)+\gamma'+\alpha'}, \quad (41)$$

$$U_{23}^2 = T\left(\frac{\partial M}{\partial T}\right)^2 \Big/ \chi_T C_H \propto d_c^{\,0}. \quad (42)$$

If the static scaling laws[20] $\Delta' = 2(\gamma' + \beta)$, $\gamma' + 2\beta + \alpha' - 2 = 0$ are obeyed, then the critical exponents of the U_{ij} are equal to zero. Thus near the tricritical point the U_{ij} are constant to within logarithmic corrections.

As a consequence of Eqs. (40)–(42) we anticipate that near the tricritical point in the antiferromagnetic phase all elements of $(\vec{A}, \vec{A}^\dagger)$ (or $\vec{U}$) will be significant, which carries with it the implication that $N(q)$, $M(q)$, and $E(q)$ remain coupled. However, if the system is moved along the second-order line in a direction away from the tricritical point we expect the parameters characterizing the coupling of $M(q)$ to $N(q)$ and $E(q)$ to become smaller, finally disappearing in the zero field limit. Similar behavior is predicted for $(\vec{A}, \vec{A}^\dagger)_{23}$ in the paramagnetic state so that $M(q)$ and $E(q)$ are strongly coupled near the tricritical point with the coupling becoming weaker as the system is moved toward zero field.

The temperature dependence of the decay rates Γ_{ij} is a more difficult question. Experimental evidence[15] as well as theoretical arguments by Kawasaki[21] and Riedel[22] suggest that in the case of easy-axis magnets in zero field the conventional theory of critical slowing down should apply close to the critical point, at least as a first approximation.[23] Were this to be the case in finite field we would expect that the Γ_{ij} varied with temperature as $[\chi_{A_iA_i}(0)\chi_{A_jA_j}(0)]^{-1/2}$ in the critical region. In particular we would have $\Gamma_N \propto (\chi_T^*)^{-1}$, $D_M \propto \chi_T^{-1}$, and $D_E \propto C_H^{-1}$. Although this behavior is plausible in light of our current knowledge of spin dynamics, a different temperature dependence cannot be ruled out at this point. Another aspect of the temperature dependence concerns the critical slowing down of the longitudinal fluctuations in the magnetization. Should this happen $M(q)$ becomes a hydrodynamic variable independent of the rotational symmetry about the c axis.

Our final comment concerns a comparison with the hydrodynamics of the spin-flop phase. In the flop phase the staggered moment is perpendicular to the c axis. If, in addition, there is rotational symmetry about the c axis, then the flop state has associated with it a weakly damped hydrodynamic spin-wave mode. The frequency of this mode is determined by the eigenvalues of a frequency matrix involving $N_x(q)$, $N_y(q)$, $E(q)$, and $M_z(q)$ $(z = c)$ and is given by[8]

$$\omega_q = g\mu_B N\left[\chi_{yy}^*(q)\left(\frac{\partial M}{\partial H}\right)_{S,H^*}\right]^{-1/2}, \quad (43)$$

where the staggered moment is taken to be parallel to the x axis. In (43) g is the g factor, μ_B is the Bohr magneton, and χ_{yy}^* denotes the transverse staggered susceptibility. Because of the rotational symmetry we have $\chi_{yy}^* \propto q^{-2}$ so that $\omega_q \propto q$ at long wavelengths.[8] The hydrodynamics of the flop phase is thus significantly different from the hydrodynamics in the antiferromagnetic phase, where $\vec{N}$ is parallel to the c axis. In this configuration only relaxational modes are expected.

In the paramagnetic phase, cross-over behavior in the critical dynamics is anticipated near the point of intersection of the second-order antiferromagnetic–paramagnetic (AFM-P) and spin-flop–paramagnetic (SF-P) lines. Near the (SF-P) line we expect behavior characteristic of a planar antiferromagnet; near the AFM-P line easy-axis behavior is expected. In the immediate vicinity of the point of intersection intermediate behavior is plausible.[24] This will be discussed in a separate publication.[25]

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