[33]S. Q. Wang, W. E. Evenson, and J. R. Schrieffer, Phys. Rev. Letters 23, 92 (1969); W. E. Evenson, J. R. Schrieffer, and S. Q. Wang, J. Appl. Phys. 41, 1199 (1970).

[34]M. Cyrot, Phys. Rev. Letters 25, 871 (1970).

[35]W. Langer, M. Plischke, and D. Mattis, Phys. Rev. Letters 23, 1448 (1969).

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Nonlinear Effects in the Critical Dynamics of Easy-Axis Ferro- and Antiferromagnets *

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Approximate kinetic equations have been derived for the critical dynamical variables of easy-axis ferro- and antiferromagnets. Particular attention is paid to nonlinear couplings and their influence on the dynamics of the long-wavelength fluctuations of the order parameter. The nonlinear nature of the problem is reflected in the inverse reduced-susceptibility matrix. When only the linear susceptibilities are retained the equations reduce to those of Schwabl and Michel. Including both linear and nonlinear susceptibilities leads to a number of new effects. Among these are the renormalization of the decay rates, a change below T_c in the relative weights of the two central peaks in the linear dynamical susceptibility of the antiferromagnetic order parameter, and the introduction of a high-frequency background in the order-parameter power spectrum. Provided the thermodynamic scaling laws are obeyed, these effects are essentially independent of temperature. An experimental test of the theory involving measurements of the width of the imaginary part of the dynamic susceptibility of the ferromagnetic order parameter at corresponding temperatures above and below T_c is proposed.

I. INTRODUCTION

According to the current interpretation,[1] insofar as the critical dynamics is concerned, magnetic systems undergoing second-order phase transitions fall into two categories. In the first category are the so-called conventional systems, which are characterized by the property that, as the critical point is approached from the high-temperature side, the fluctuations in the order parameter decay at a rate which, in the first approximation, is inversely proportional to the corresponding susceptibility. The unconventional or strong-coupled systems are those where the decay rate has a weaker temperature dependence. Easy-axis ferro- and antiferromagnets are in the first category, while isotropic and planar ferro- and antiferromagnets are in the second. (In applying these criteria it must be kept in mind that when the anisotropy is weak, e.g., MnF_2, fully conventional behavior may be realized only at temperatures very close to T_c; at higher temperatures the dynamics may resemble that of isotropic systems.[2])

The purpose of this paper is to examine in detail the critical dynamics of conventional systems, with particular emphasis on the nonlinear effects. The approach will be sufficiently general to encompass both ferro- and antiferromagnets at temperatures above and below the critical temperature. The starting point in the analysis is a set of kinetic equations for the critical dynamical variables which was obtained recently by Kawasaki.[3] As discussed by him the critical dynamical variables are those variables whose long-wavelength fluctuations decay very slowly near the critical point. Included in this set are the hydrodynamic variables as well as the order parameter, if the latter is not conserved. In addition, in a nonlinear theory one must also include products of these variables. For easy-axis ferromagnets the critical variables are combinations of the energy density and the magnetic-moment density along the preferred axis. If the Hamiltonian has the property of being invariant with respect to spin rotations about the preferred axis, then the order parameter is also a hydrodynamic variable. In the case of easy-axis antiferromagnets the critical dynamical variables are normally the energy density, the magnetization density (provided the system has the rotational symmetry mentioned above), and the staggered-moment density along the preferred axis. Since the uniform field susceptibility of an antiferromagnet remains finite at the critical point, the fluctuations in the magnetization do not behave anomalously. Furthermore, in the absence of an external field, which we will henceforth assume, there is no linear thermodynamic coupling between the magnetization and either the staggered magnetization or the energy. Although nonlinear

couplings are present (i.e., the magnetization can couple to a product involving one factor of the magnetization and two factors of the order parameter), we will nevertheless exclude the magnetization, thus limiting our critical dynamical variables to combinations of the order parameter and energy densities. In spite of not being able to give a rigorous justification for this step, we feel that it is a reasonable one. In effect we are assuming that the critical dynamics of the easy-axis ferro- and antiferromagnets are essentially the same, apart from those aspects which reflect the conservation of the order parameter. Thus, with this choice of critical dynamical variables, the easy-axis systems separate into two categories depending on whether the order-parameter density is also a hydrodynamic variable. We will refer to those systems where the order parameter is conserved as "ferromagnets," and those systems where it is not conserved as "antiferromagnets," although included among the latter are those ferromagnets not possessing rotational symmetry about the easy axis.

The easy-axis systems are distinguishable from the isotropic and planar systems by the fact that the time derivatives of the critical dynamical variables, expressed as commutators with the Hamiltonian, are not themselves critical variables. As a consequence fluctuations in the rate of change of the critical dynamical variables decay rapidly in time. (This property is made obvious by a direct evaluation of the relevant commutator, all terms of which involve at least one factor of the transverse-spin operators, which are not included in the set of critical variables.) Because of this property, the general kinetic equations for the critical dynamical variables developed in Ref. 3 reduce to a particularly simple form, since the first-moment frequency matrix defined by Eq. (2.6) of that paper is zero. This is a consequence of the fact that this matrix is in effect the projection of the rates of change of the critical variables on the space spanned by the variables themselves. Since the fluctuations in the rates of change decay quickly, the projection is taken to be zero.

Rather than write the equation in the form employed by Kawasaki, it is convenient to label the variables by a Greek and a Roman index. The Greek index specifies the particular symmetrized combination of products of the order parameter and energy densities, and the Roman index specifies the set of wave vectors characterizing the fluctuations. Thus $\alpha = 1$ is a fluctuation in the order-parameter density, $\alpha = 2$ a fluctuation in the energy density, $\alpha = 3$ the product of two fluctuations in the order-parameter density, $\alpha = 4$ the (symmetrized) product of the energy and order-parameter densities, $\alpha = 5$ the product of two fluctuations in the energy density, etc. If fluctuations in the order-parameter density having wave vector $\vec{q}$ are designated by $N_{\vec{q}}$ and fluctuations in the energy density by $E_{\vec{q}}$, then the critical dynamical variables have the form

$$A_{1n} = (N_{\vec{q}} - \langle N_{\vec{q}} \rangle) / [(N_{\vec{q}}, N_{\vec{q}}) - \beta | \langle N_{\vec{q}} \rangle |^2]^{1/2} , \quad (1)$$

$$A_{2n} = (E_{\vec{q}} - \langle E_{\vec{q}} \rangle) / [(E_{\vec{q}}, E_{\vec{q}}) - \beta | \langle E_{\vec{q}} \rangle |^2]^{1/2} , \quad (2)$$

$$A_{3n} = (N_{\vec{q}_1} N_{\vec{q}_2} - \langle N_{\vec{q}_1} N_{\vec{q}_2} \rangle) / [(N_{\vec{q}_1} N_{\vec{q}_2}, N_{\vec{q}_1} N_{\vec{q}_2}) - \beta | \langle N_{\vec{q}_1} N_{\vec{q}_2} \rangle |^2]^{1/2} , \text{ etc.}, \quad (3)$$

where we have subtracted the ensemble average to ensure $\langle A_{\alpha n} \rangle = 0$. The variables, as defined, are also normalized in the sense

$$(A_{\alpha n}, A_{\alpha n}) = 1 , \quad (4)$$

where (X, Y) denotes the inner product

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

in which $\mathcal{H}$ is the Hamiltonian and $\beta = 1/KT$.

With the vanishing of the first-moment matrix and the change in labeling, the equations equivalent to Eq. (2.21) of Ref. 3 take the form

$$\frac{dA_{\alpha m}}{dt} = - \sum_{\beta, n} \Gamma_{\alpha m, \beta n} A_{\beta n} , \quad (6)$$

where the matrix $\underline{\Gamma}$ is expressed as a product

$$\underline{\Gamma} = \underline{T}\,\underline{U} . \quad (7)$$

The matrix $\underline{T}$ is given by

$$T_{\alpha m, \beta n} = \int_0^\infty (\dot{A}_{\alpha m}(t), \dot{A}_{\beta n}) dt , \quad (8)$$

in which $\dot{A} = (i/\hbar)[\mathcal{H}, A]$. In writing $\underline{T}$ in this way explicit use has been made of the fact that the fluctuation in $\dot{A}$ decay in a microscopic time. The matrix $\underline{U}$ is the (normalized) inverse-susceptibility matrix defined by

$$(\underline{U}^{-1})_{\alpha m, \beta n} = (A_{\alpha m}, A_{\beta n}) . \quad (9)$$

The approximations that precede Eq. (6) have been discussed by Kawasaki.[3] In this paper, Eq. (6) is taken as the starting point and additional approximations are introduced which lead to further simplifications. As with many treatments of dynamical effects, the approximations are difficult to control. They are plausible, but precise estimates of the attendant errors are hard to obtain. This difficulty is partially offset by the fact that the theory, in its final form, makes a number of predictions which in principle can be tested by experiment. The extent to which these predictions are verified can perhaps be taken as evidence supporting the over-all success of the approach.

II. FORMALISM

The focus of attention in this section is on the

kinetic equations for the linear fluctuations $N_{\vec{q}}$ and $E_{\vec{q}}(\alpha = 1, 2)$ for $q \ll k_c$, where k_c is the inverse correlation length associated with the order-parameter susceptibility $(N_{\vec{q}}, N_{\vec{q}})$. If the nonlinear terms corresponding to $\alpha > 2$ are omitted from $\underline{T}$ and $\underline{U}^{-1}$ the equations take the simple form

$$\frac{dA_{1n}}{dt} = -\Gamma^R_{1n,1n}A_{1n} - \Gamma^R_{1n,2n}A_{2n} , \tag{10}$$

$$\frac{dA_{2n}}{dt} = -\Gamma^R_{2n,1n}A_{1n} - \Gamma_{2n,2n}A_{2n} , \tag{11}$$

where Γ^R is a 2×2 matrix obtained by leaving out all but the first two rows and columns of $\underline{T}$ and $\underline{U}^{-1}$. In writing these equations it has been recognized that $\Gamma^R_{\alpha n,\beta m} = 0$ for $n \neq m$, which is a consequence of translational invariance. Equations (10) and (11) are a set of coupled *linear* equations for the fluctuations in the order-parameter and energy densities. They resemble, and in fact are equivalent to, the dynamical equations of Schwabl and Michel.[4,5] Equations similar to these have also been used in the interpretation of recent neutron-scattering data from MnF_2.[6]

In the remainder of this section we will outline an approximate treatment of the nonlinear terms in the kinetic equations. The coupling to products of the fluctuations has two main effects. First, the decay rates $\Gamma^R_{\alpha n,\beta m}(\alpha, \beta = 1, 2)$ are renormalized. Second, transients are introduced which lead to high-frequency wings in the absorptive parts of the dynamic susceptibilities. In dealing with the nonlinear terms it is convenient to consider separately their effect on the matrices $\underline{U}$ and $\underline{T}$. We begin with $\underline{U}$. The basic approximation in the evaluation of $\underline{U}$ is equivalent to replacing all elements of $\underline{U}^{-1}$ of a given α, β by the value obtained in the limit as all the wave vectors approach zero, i.e.,

$$(\underline{U}^{-1})_{\alpha m,\beta n} \to (\underline{U}^{-1})_{\alpha 0,\beta 0} , \tag{12}$$

where

$$(\underline{U}^{-1})_{\alpha 0,\beta 0} = \lim_{\text{all } q\to 0} (A_{\alpha m}, A_{\beta n}) \quad (\alpha \neq \beta)$$
$$= 1 \qquad (\alpha = \beta) . \tag{13}$$

This approximation amounts to treating all the $A_{\alpha m}$ of a given α on an equal footing insofar as their contribution to $\underline{U}$ is concerned. It is suggested by the behavior of $(N_{\vec{q}}, N_{\vec{q}})$ which, in general, is rather insensitive to q when $q \ll k_c$ and can be approximated by its small-q limit, the isothermal thermodynamic susceptibility. (Strictly speaking, the limit $q \ll k_c$ applies only to $\beta = 1, 2$. The amplitudes A_{1m} and A_{2m} will also couple to products of fluctuations with individual factors having wave vectors $\approx k_c$. For these entries the approximation is somewhat more severe.)

Having made this approximation we can write the kinetic equations in the form

$$\frac{dA_{\alpha m}}{dt} = -\sum_{\substack{\beta,\gamma \\ n,p}} T_{\alpha m,\beta n} \frac{1}{N^{(1m)}_\beta} \hat{U}_{\beta\gamma} \frac{1}{N^{(1m)}_\gamma} A_{\gamma p} , \tag{6'}$$

where $N^{(1m)}_\beta$ is the number of modes of type β which are coupled to A_{1m}, that is to say, the number of modes of type β whose wave vectors sum to $\vec{q}$, the wave vector of A_{1m}.[3] The matrix $\hat{\underline{U}}$ is given by the inverse of the reduced-susceptibility matrix:

$$(\hat{\underline{U}}^{-1})_{\alpha\beta} = (\underline{U}^{-1})_{\alpha 0,\beta 0} , \tag{14}$$

where $(\underline{U}^{-1})_{\alpha 0,\beta 0}$ is given by Eq. (13). With respect to the terms associated with values of β or $\gamma > 2$, Eq. (6′) is in effect an averaged kinetic equation involving an averaged T matrix

$$\frac{1}{N^{(1m)}_\beta} \sum_n T_{\alpha m,\beta n}$$

and an averaged amplitude

$$\frac{1}{N^{(1m)}_\gamma} \sum_p A_{\gamma p} .$$

The simplification which results from this approximation occurs because the elements of the matrix $\hat{\underline{U}}^{-1}$ can all be expressed in terms of the second and higher derivatives of the free energy with respect to temperature and field conjugate to the order parameter, which are evaluated in the limit of zero conjugate field, i.e.,

$$(\hat{\underline{U}}^{-1})_{11} = 1 , \tag{15}$$

$$(\hat{\underline{U}}^{-1})_{12} = (\hat{\underline{U}}^{-1})_{21} = T_c \frac{\partial \langle N_0 \rangle}{\partial T_c} \Big/ (T_c C^*_H)^{1/2} (\chi_T)^{1/2} , \tag{16}$$

$$(\hat{\underline{U}}^{-1})_{22} = 1 , \tag{17}$$

$$(\hat{\underline{U}}^{-1})_{13} = \frac{\partial^2 \langle N_0 \rangle}{\partial H^{*2}} \Big/ \left(\frac{\partial \langle N_0 \rangle}{\partial H^*}\right)^{1/2} \left(\frac{\partial^3 \langle N_0 \rangle}{\partial H^{*3}}\right)^{1/2} , \text{ etc.} , \tag{18}$$

where C^*_H is the magnetic specific heat at constant conjugate field H^*; χ_T is the isothermal order-parameter susceptibility; T_c is the critical temperature. It should be noted that the reduced-susceptibility matrix still has an infinite number of rows and columns. Presumably the essential features of the theory are retained if $\hat{\underline{U}}^{-1}$ is approximated by a finite matrix obtained by omitting elements associated with high-order thermodynamic derivatives. This matter has not been investigated in detail, however.

At this point it is convenient to introduce the thermodynamic scaling hypothesis for the free energy.[7,8] It is postulated that the singular part of the free energy has the form[9]

$$f_{\text{sing}}(T,H^*)=\left(1-\frac{T_c}{T}\right)^{-\gamma+2\Delta} F\left(\frac{(1-T_c/T)^{\Delta}}{H^*}\right) \quad (T>T_c)\,, \quad (19)$$

where γ and Δ have the conventional interpretation as critical indices. With this form for the free energy we obtain the important result that the elements of the matrix $\underline{\hat{U}}$ approach a constant value in the limit $T\to T_c$ (apart from terms involving $\ln|1-T_c/T|$ which would arise from logarithmic singularities in the various susceptibilities). This property is a consequence of the fact that the elements of $\underline{\hat{U}}^{-1}$ are all of the type

$$\left.\frac{\partial^{l+m+n+p} f_{\text{sing}}}{\partial\epsilon^{l+m}\,\partial H^{*n+p}}\right|_{H^*=0} \Bigg/ \left(\left.\frac{\partial^{2l+2n} f_{\text{sing}}}{\partial\epsilon^{2l}\,\partial H^{*2n}}\right|_{H^*=0}\right)^{1/2}\left(\left.\frac{\partial^{2m+2p} f_{\text{sing}}}{\partial\epsilon^{2m}\partial H^{*2p}}\right|_{H^*=0}\right)^{1/2},$$

where $\epsilon=|1-T_c/T|$.

It is beyond the scope of this paper to discuss whether the scaling laws are exact for any particular system. The evidence that they are often a good approximation is strong, and when this is the case $\underline{\hat{U}}$ is at worst a slowly varying function of ϵ.[10]

Provided the free energy has the form displayed in Eq. (19), the temperature dependence of the critical slowing down of the fluctuations in the order parameter is determined by the matrix $\underline{T}$. In discussing this matrix it is necessary to consider separately the two classes of systems. We begin with antiferromagnets. Our main interest is the equation for A_{1m}; for $q\ll k_c$, the energy fluctuation A_{2m} has only a secondary role. The right-hand side of Eq. (6′) is seen to involve combinations of terms of the form

$$\sum_{\beta,n}\frac{1}{N_\beta^{(1m)}}\,T_{1m,\beta n}\hat{U}_{\beta\gamma}\,.$$

It is argued that near the critical point only the term $\beta=1$, $n=m$ makes an important contribution to the sum when $q\ll k_c$, that is

$$\sum_{\beta,n}\frac{1}{N_\beta^{(1m)}}\,T_{1m,\beta n}\hat{U}_{\beta\gamma}\approx T_{1m,1m}\hat{U}_{1\gamma}\,. \quad (20)$$

The justification for this approximation comes from an examination of the temperature and wave-vector dependence of the various terms. We have

$$T_{1m,1m}\sim\epsilon^{\gamma}q^0\,,$$

$$T_{1m,2m}\sim\epsilon^{\alpha/2+\gamma/2}\,q^2\,,$$

$$T_{1m,3m}\sim\epsilon^{\gamma+\Delta}\,q^0\quad(T<T_c),\ \text{etc.},$$

where the temperature dependence is assumed to arise solely from the normalization of the $A_{\beta n}$, since $\dot{A}_{\beta n}$ is not a critical dynamical variable. The dependence on q^2 in $T_{1m,2m}$ (but not in the other terms) reflects the fact that the energy density is also a hydrodynamic variable. An examination of the remaining terms in the series indicates that they are all of the form $\epsilon^s q^0$ with $s>\gamma$. Thus, for $\beta>2$, the $T_{1m,\beta n}$ vanish more rapidly with temperature than $T_{1m,1m}$, while at a fixed temperature $T_{1m,2m}$ is small compared with $T_{1m,1m}$ for $q\ll k_c$, even though it is vanishing less rapidly with temperature. It must be emphasized that these arguments are strictly qualitative and do not necessarily hold at fixed wave vectors for all values of ϵ. A more detailed justification which goes beyond plausibility can only come from an examination of specific cases.

Assuming the validity of the approximations reflected in Eq. (20) we can write the kinetic equation for the order parameter in the form

$$\frac{dA_{1m}}{dt}=-\Gamma_N(m)\sum_{\beta,n}\hat{U}_{1\beta}\,\frac{1}{N_\beta^{(1m)}}\,A_{\beta n} \quad \text{(antiferromagnet)}\,, \quad (21)$$

where

$$\Gamma_N(m)=T_{1m,1m}\,. \quad (22)$$

The analysis of the $\underline{T}$ matrix in the ferromagnetic problem is complicated slightly by the fact that both the order parameter and the energy density are hydrodynamic variables. As a consequence, the kinetic equations for A_{1m} and A_{2m} remain coupled in the small-q limit. Thus it is necessary to separate out both $\beta=1$ and $\beta=2$ in the ferromagnetic equivalent to Eq. (20):

$$\sum_{\beta,n}\frac{1}{N_\beta^{(1m)}}\,T_{1m,\beta n}\hat{U}_{\beta\gamma}\approx T_{1m,1m}\hat{U}_{1\gamma}+T_{1m,2m}\hat{U}_{2\gamma}\,, \quad (23)$$

$$\sum_{\beta,n}\frac{1}{N_\beta^{(1m)}}\,T_{2m,\beta n}\hat{U}_{\beta\gamma}\approx T_{2m,1m}\hat{U}_{1\gamma}+T_{2m,2m}\hat{U}_{2\gamma}\,. \quad (24)$$

The terms which are omitted on the right-hand side of (23) and (24) are all of order q^2 but vanish more rapidly with temperature in the limit $\epsilon\to 0$.

The kinetic equations which follow from these approximations take the form

$$\frac{dA_{1m}}{dt}=-\sum_{\beta,n}\left(T_{1m,1m}\hat{U}_{1\beta}+T_{1m,2m}\hat{U}_{2\beta}\right)\frac{1}{N_\beta^{(1m)}}\,A_{\beta n}\,, \quad (25)$$

$$\frac{dA_{2m}}{dt}=-\sum_{\beta,n}\left(T_{2m,1m}\hat{U}_{1\beta}+T_{2m,2m}\hat{U}_{2\beta}\right)\frac{1}{N_\beta^{(1m)}}\,A_{\beta n} \quad \text{(ferromagnet)}\,. \quad (26)$$

Equations (21), (25), and (26) are the principal results of this section. We postpone discussion of their significance until Sec. III.

III. DISCUSSION

In this section the kinetic equations obtained in Sec. II are discussed in detail. We begin with the

antiferromagnetic case, Eq. (21). It is convenient to rewrite this equation in terms of the Laplace transforms of the $A_{\alpha m}$:

$$\tilde{A}_{\alpha m}(S) = \int_0^\infty e^{-St} A_{\alpha m}(t)\, dt . \tag{27}$$

The transform of (21) takes the form

$$\begin{aligned} S\,\tilde{A}_{1m}(S) - A_{1m}(0) = &-\Gamma_N(m)\,\hat{U}_{11}\tilde{A}_{1m}(S) \\ &-\Gamma_N(m)\,\hat{U}_{12}\tilde{A}_{2m}(S) \\ &-\Gamma_N(m)\sum_{\substack{\beta>2\\ n}}\hat{U}_{1\beta}\frac{1}{N_\beta^{(1m)}}\tilde{A}_{\beta n}(S) . \end{aligned} \tag{28}$$

A nonlinear equation of this type can be solved by iteration. To obtain insight into the significance of the various terms we first consider the zeroth-order solution which is obtained by leaving out all terms on the right-hand side with $\beta > 2$. What remains is a set of equations involving $N_{\vec{q}}$ and $E_{\vec{q}}$. For small q ($\ll k_c$) the fluctuations in the energy density decay at a rate determined by the renormalized thermal-diffusion constant (apart from spin-lattice effects[11]). Writing this rate as $\bar{\Gamma}_E(m)$[12] we obtain the zeroth-order solution

$$\begin{aligned} \tilde{A}_{1m}(S)^0 + &\frac{A_{1m}(0) + (\hat{U}_{12}/\hat{U}_{11})A_{2m}(0)}{S+\bar{\Gamma}_N(m)} \\ &- \frac{(\hat{U}_{12}/\hat{U}_{11})A_{2m}(0)}{S+\bar{\Gamma}_E(m)} \end{aligned} \tag{29}$$

in the limit $\bar{\Gamma}_E(m) \ll \bar{\Gamma}_N(m)$. In this equation $\bar{\Gamma}_N(m)$ denotes the renormalized decay rate

$$\bar{\Gamma}_N(m) = \hat{U}_{11}\Gamma_N(m) . \tag{30}$$

By taking the scalar product of (28) with $A_{1m}(0)$ we obtain the zeroth-order approximation to the Laplace transform of the relaxation function $(A_{1m}(t), A_{1m})$. Writing this transform as

$$R_{1m}(S) = \int_0^\infty e^{-St}(A_{1m}(t),\, A_{1m}) , \tag{31}$$

we find

$$\begin{aligned} R_{1m}(S)^0 = &\frac{1+(\hat{U}_{12}/\hat{U}_{11})[(\underline{\hat{U}}^{-1})_{21}/(\underline{\hat{U}}^{-1})_{11}]}{S+\bar{\Gamma}_N(m)} \\ &- \frac{(\hat{U}_{12}/\hat{U}_{11})[(\underline{\hat{U}}^{-1})_{21}/(\underline{\hat{U}}^{-1})_{11}]}{S+\bar{\Gamma}_E(m)} , \end{aligned} \tag{32}$$

having made use of Eqs. (4) and (9).

Equation (32) is a generalization of the analogous expression obtained by solving Eqs. (10) and (11). It differs in that it involves matrix elements of $\underline{\hat{U}}$ rather than the truncated inverse reduced-susceptibility matrix obtained by leaving out all but the first two rows and columns of $\underline{\hat{U}}^{-1}$. The relaxation function associated with the truncated matrix has a transform which can be written[6]

$$R_{1m}(S)^R = \frac{\chi_S/\chi_T}{S+\Gamma_N(m)(\chi_T/\chi_S)} + \frac{1-\chi_S/\chi_T}{S+\Gamma_E(m)} , \tag{33}$$

where $\Gamma_E(m)$, the unrenormalized decay rate, is equal to $T_{2m,2m}$, and χ_S is the adiabatic order-parameter susceptibility, which is related to χ_T by

$$\frac{\chi_T-\chi_S}{\chi_T} = (\underline{\hat{U}}^{-1})_{12}(\underline{\hat{U}}^{-1})_{21} = T_c\left(\frac{\partial\langle N_0\rangle}{\partial T_c}\right)^2 \Big/ C_H^*\chi_T . \tag{34}$$

To obtain the first-order expression for the A_{1m} the nonlinear terms in Eq. (28) are evaluated using A_{1m}^0 and A_{2m}^0. The relaxation function which results from continuing the iteration gives rise to a linear dynamic order-parameter susceptibility $\chi(\vec{q}, \omega)$ which has the following characteristics. The function $\chi''(\vec{q}, \omega)/\omega$ has a narrow central peak of width $\bar{\Gamma}_E(m)$, a broader peak of width $\bar{\Gamma}_N(m)$, and a very broad background arising from the coupling to the $A_{\beta m}$ with $\beta > 2$. The relative weights of the $\bar{\Gamma}_N$ and $\bar{\Gamma}_E$ peaks are

$$1 + (\hat{U}_{12}/\hat{U}_{11})[(\underline{\hat{U}}^{-1})_{21}/(\underline{\hat{U}}^{-1})_{11}]$$

and

$$-(\hat{U}_{12}/\hat{U}_{11})[(\underline{\hat{U}}^{-1})_{21}/(\underline{\hat{U}}^{-1})_{11}] ,$$

respectively. It is apparent that the nonlinear terms, in addition to giving rise to a broad background in the power spectrum, also renormalize the decay rates of the order-parameter and energy fluctuations. Furthermore, they affect the relative weight of the two peaks which are present below T_c. Provided the static scaling laws are obeyed, these effects are essentially independent of temperature. Their importance depends on the relative magnitudes of the elements of $\underline{\hat{U}}^{-1}$. About these little can be said without specific expressions for the free energy, apart from the obvious remark that above T_c all entries of $\underline{\hat{U}}^{-1}$ involving expectation values of odd powers $N_{\vec{q}}$ vanish identically. As a consequence, $\hat{U}_{12}$, $\hat{U}_{13}$, $\hat{U}_{15}$, etc., are all equal to zero in the disordered state and the $\bar{\Gamma}_E$ peak is absent.

The analysis of the kinetic equations for ferromagnets proceeds in a similar fashion. In the first iteration the zeroth-order solution to (25) and (26) is used in the evaluation of the nonlinear terms. In solving the zeroth-order equations an important simplification occurs if $T_{1m,1m}$, $T_{1m,2m}$, and $T_{2m,2m}$ are such that

$$T_{1m,1m} \ll T_{2m,2m} , \tag{35a}$$

$$|T_{1m,2m}|^2 \ll T_{1m,1m}\,T_{2m,2m} . \tag{35b}$$

[A calculation of these terms in the random-phase approximation indicates

$$T_{1m,1m} \sim \epsilon^{\gamma} q^2 ,$$

$$T_{2m,2m} \sim \epsilon^{\alpha} q^2 ,$$

$$T_{1m,2m} \sim \epsilon^{\beta+\gamma/2+\alpha/2} q^2 \quad (T < T_c)$$

$$= 0 \qquad (T > T_0) ,$$

so that (35) may hold very close to T_c.] Assuming the applicability of (35) the zeroth-order expression for $R_{1m}(S)$ has the form

$$R_{1m}(S)^0 = 1/[S + \bar{\Delta}_N(m)] , \tag{36}$$

where $\bar{\Delta}_N(m)$ is a renormalized decay rate which is given by

$$\bar{\Delta}_N(m) = \hat{U}_{11} \left(1 - \frac{(\hat{U}_{12})^2}{\hat{U}_{11}\hat{U}_{22}}\right) \Delta_N(m) , \tag{37}$$

with $\Delta_N(m) = T_{1m,1m}$. This result is to be compared with the corresponding expression obtained from Eqs. (10) and (11):

$$R_{1m}(S)^R = 1/[S + \Delta_N(m)] . \tag{38}$$

Comparing (38) with (36) and (37) it is apparent that, just as in the antiferromagnetic case, the nonlinear couplings have renormalized the decay rates of the $N_{\vec{q}}$. In addition, they contribute high-frequency components to the power spectrum. There is, however, an important difference in the behavior of ferromagnets as opposed to antiferromagnets. For fixed temperature the central peak in $\omega^{-1}\times$(the absorptive part of the dynamic order-parameter susceptibility of the ferromagnet) has a width which goes to zero in the long-wavelength limit (apart from dipolar and spin-lattice effects). On the other hand, the nonlinear background, which arises primarily from couplings to products of fluctuations with wavelengths on the order of k_c^{-1}, does not shrink in width appreciably in this limit. The relative weight of the background does vanish as q^2, however. If the same limit is taken for antiferromagnets, the width of the thermal part of the spectrum also vanishes as q^2, but the width of the spin part and the width and relative weight of the nonlinear background change only slightly.

Most of these effects can be seen in the first-order approximation to $R_{1m}(S)$ above T_c:

$$R_{1m}(S)^1 = \frac{1}{S + \hat{U}_{11} T_{1m,1m}} - \frac{T_{1m,1m}}{S + \hat{U}_{11} T_{1m,1m}} \times \sum_{\substack{\beta>2 \\ n}} \frac{\hat{U}_{1\beta}(\hat{\underline{U}}^{-1})_{\beta 1}}{N_{\beta}^{(1m)}} \int_0^{\infty} e^{-St} \left(\frac{A_{\beta n}(t)^0}{A_{\beta n}(0)}\right) dt . \tag{39}$$

For the purposes of a qualitative analysis $A_{\beta n}(t)^0$ can be approximated by $A_{\beta n}(0)\, e^{-\Gamma_\beta t}$, where Γ_β is an effective decay rate which is approximately equal to the zeroth-order decay rate when the fluctuations making up $A_{\beta n}$ all have wave vectors close to k_c. The equation that results from making this approximation takes the form

$$R_{1m}(S)^1 = \frac{1}{S + \hat{U}_{11} T_{1m,1m}} - \sum_{\beta>2} \frac{\hat{U}_{1\beta}(\hat{\underline{U}}^{-1})_{\beta 1}}{\hat{U}_{11}(\hat{\underline{U}}^{-1})_{11}} \left(\frac{\hat{U}_{11} T_{1m,1m}}{\hat{U}_{11} T_{1m,1m} - \Gamma_\beta}\right) \times \left(\frac{1}{S + \Gamma_\beta} - \frac{1}{S + \hat{U}_{11} T_{1m,1m}}\right) . \tag{40}$$

For ferromagnets $\hat{U}_{11} T_{1m,1m}/\Gamma_\beta$ approaches zero in the small-q limit so that

$$R_{1m}(S)^1 \to \frac{1}{S + \hat{U}_{11} T_{1m,1m}} , \tag{41}$$

as $q \to 0$. In the case of antiferromagnets the ratio $\hat{U}_{11} T_{1m,1m}/\Gamma_\beta$ remains finite in this limit, so that the second term in Eq. (40) can not in general be ignored.

IV. SUMMARY

Approximate kinetic equations have been derived for the critical dynamical variables of easy-axis ferro- and antiferromagnets. Particular attention has been paid to nonlinear couplings and their influence on the dynamics of the long-wavelength fluctuations of the order parameter. As shown in Eqs. (21), (25), and (26), the nonlinear nature of the problem is reflected in the inverse reduced-susceptibility matrix $\hat{\underline{U}}$. When only the linear susceptibilities are retained the equations reduce to those of Schwabl and Michel.[4,5] Including both the linear and the nonlinear susceptibilities gives rise to a number of new effects. Among these are the renormalization of the decay rates [Eqs. (30) and (37)], a change below T_c in the relative weight of the two central peaks in the linear dynamical susceptibility for the antiferromagnetic order parameter [Eq. (32)], and the introduction of a high-frequency background in the order-parameter power spectrum. Provided the thermodynamic scaling laws are obeyed, $\hat{\underline{U}}$ is independent of temperature near T_c (apart from logarithmic terms), so that these effects are essentially temperature independent in the critical region. As for their magnitude, little can be said without detailed information about the free energy.

In spite of its generality the theory does make a number of predictions which, in principle, are capable of being checked. The one most accessible to experiment pertains to the ratio of the decay rates of the order-parameter fluctuations in ferromagnets at corresponding temperatures above and below T_c.

Provided Eq. (35) is satisfied[13] we may define the

following ratio:

$$W=\frac{[\text{expt width } \chi''\ (T=T_c-)]\chi_T(T_c-)}{[\text{expt width } \chi''\ (T=T_c+)]\chi_T\ (T_c+)}\ , \qquad (42)$$

where "expt width χ''" refers to the experimental width of $\omega^{-1}\times$(imaginary part of the dynamic order-parameter susceptibility), which could be measured in an inelastic neutron-scattering experiment and $T_c\pm=T_c\pm\Delta T$. If nonlinear effects are unimportant, $W=1$, whereas if they do play an important role, $W\neq1$. The difference arises because the reduced-susceptibility matrix has entries $(\hat{\underline{U}}^{-1})_{13}$, $(\hat{\underline{U}}^{-1})_{15}$, etc., which are identically equal to zero above T_c and which in general differ from zero below. Calculations carried out using estimates of the lowest-order nonlinear susceptibilities for the three-dimensional Ising model[9] suggest $W>1$. However, this result should be taken with some caution, since it is not clear whether (i) a model devoid of dynamics is capable of characterizing nonlinear dynamical effects, and (ii) an estimate of $\hat{\underline{U}}$ based on only a few entries in $\hat{\underline{U}}^{-1}$ is adequate.

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[1]K. Kawasaki, Progr. Theoret. Phys. (Kyoto) 40, 706 (1968).
[2]E. K. Riedel, J. Appl. Phys. 42, 1383 (1971).
[3]K. Kawasaki, Ann. Phys. (N. Y.) 61, 1 (1970).
[4]F. Schwabl and K. H. Michel, Phys. Rev. B 2, 189 (1970).
[5]K. H. Michel and F. Schwabl, Z. Physik 238, 264 (1970); 240, 354 (1970).
[6]D. L. Huber, Intern. J. Quantum Chem. 5, 667 (1971).
[7]L. P. Kadanoff, Physics 2, 263 (1966).
[8]B. Widom, J. Chem. Phys. 43, 3898 (1965).
[9]J. W. Essam and D. L. Hunter, J. Phys. C 1, 392 (1968).
[10]L. P. Kadanoff *et al.*, Rev. Mod. Phys. 39, 395 (1967).
[11]D. L. Huber, Phys. Rev. B 3, 836 (1971).
[12]$\overline{\Gamma}_E(m)$ is related to the unrenormalized decay rate $\Gamma_E(m)$ by

$$\overline{\Gamma}_E(m)=\hat{U}_{22}\left(1-\frac{(\hat{U}_{12})^2}{\hat{U}_{11}\hat{U}_{22}}\right)\Gamma_E(m).$$

[13]If Eq. (35) is satisfied, then the experimental width of $\chi''(\vec{q},\omega)/\omega$ is inversely proportional to χ_T. A less rapid variation with temperature indicates either a breakdown of the condition or that the system is not sufficiently close to T_c to be in the fully conventional regime.

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Density-of-States Spectra for the fcc Lattice with Variable Second-Neighbor Interactions*

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Using an analytic dispersion relation we have computed density-of-states spectra on an fcc lattice. The transition from the spectrum of the nearest-neighbor fcc case has been studied in detail as the strength of second-neighbor interactions increases and eventually produces the spectrum of the nearest-neighbor sc case. Graphical results for the magnitude of the group velocity have been used to improve the resolution of the singularities of the spectra enabling the changes in number, order, and degeneracies of these singularities to be followed as the second-neighbor interaction is varied. A simple algebraic analysis of the critical points of the dispersion relation gives a complete explanation of all features found in the spectra and reveals a singularity not previously found in three-dimensional spectra. The results can be applied to spin waves in ferromagnetic insulators as well as to electronic-energy bands in the tight-binding approximation and have relevance to a large number of phenomena in solid-state physics.

I. INTRODUCTION

Considerations of density-of-states spectra arise frequently in solid-state physics. They have been observed experimentally through electron spectroscopy for chemical analysis,[1] superconducting tunneling,[2] and incoherent neutron scattering[3]; may be computed[4] from theoretical models (often in harmonic approximations) whose force constants are sometimes obtained from inelastic neutron scattering[5] data for the dispersion; and are used to evaluate thermodynamic functions,[4] Debye temperatures, Fermi levels, etc.

The general features of such spectra include singularities which have been discussed and classified in a formal manner by van Hove[6] and Phillips[7] who include the effect of branch-crossover degeneracies. Despite this activity relatively little appears to be known about the density-of-states spectra for basic single-branch three-dimensional cases. One reason may be the problem of computing the spectra which has resulted in a concentra-