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Critical spin dynamics in EuO†

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Mode-mode coupling equations have been solved for the decay rates of the hydrodynamic spin fluctuations in the vicinity of the critical point in EuO. Dipolar effects are included. With no adjustable parameters calculated values of the spin-wave linewidth and the paramagnetic spin diffusion constant are in reasonable agreement with available experimental data for $|1 - T/T_c| \gtrsim 0.05$.

Over the past few years a series of measurements probing the critical behavior of the prototype Heisenberg ferromagnet EuO have been carried out using neutron-scattering techniques.[1-4] Recently, data have been obtained which yield values for the spin-wave linewidths in the hydrodynamic region below T_c,[3] and the spin diffusion constant in the paramagnetic phase.[4] The purpose of this paper is to report the results of theoretical calculations of both of these quantities which are in substantial agreement with experiment. The calculations, which have been carried out self-consistently in the lowest-order mode-coupling formalism,[5,6] make full use of the experimental results for the static parameters and the spin-wave frequency and hence contain no adjustable parameters. We include the effects of the dipolar interaction in an approximation which is appropriate some distance from the critical point, but breaks down very close to T_c where there is crossover from quasi-isotropic to dipolar behavior in the dynamics as well as in the statics.[7]

We begin with the calculation for the ordered phase. The dynamic transverse and longitudinal susceptibilities are postulated to have the hydrodynamic form

$$\chi_{\parallel}(q,\omega) = \omega\chi_{\parallel}(q)\Gamma(q)/[\omega^2 + \Gamma(q)^2], \tag{1}$$

$$\chi_{+-}(q,\omega) = \omega\chi_{\perp}(q)\Lambda(q)/\{[\omega - \omega(q)]^2 + \Lambda(q)^2\}. \tag{2}$$

In these equations, $\chi_{\parallel}(q)$ and $\chi_{\perp}(q)$, the static susceptibilities per spin in units of $g^2\mu_B^2$, are approximated by

$$\chi_{\parallel}(q) = (2Ja^2)^{-1}/(\kappa_1^2 + q^2), \tag{3}$$

$$\chi_{\perp}(q) = \chi_{xx}(q) + \chi_{yy}(q) = 2S\sigma/\omega(q), \tag{4}$$

where J $(=J_{\mathrm{nn}} + J_{\mathrm{nnn}})$ is the total exchange integral, S is the spin, σ is the reduced magnetization, a is the lattice constant, κ_1 is the inverse correlation length, and $\omega(q)$ is the spin-wave frequency. Several points should be mentioned in connection with Eqs. (1)–(4). First, we have postulated a central peak in $\chi_{\parallel}(q,\omega)$. Although there is as yet no experimental evidence for such a peak in EuO we argue it would be hard to detect [because of the magnitude of $\chi_{\parallel}(q)$ relative to $\chi_{\perp}(q)$] in the region where the theory is expected to be applicable ($q/\kappa_1 \lesssim 1$, $T<68$K). Second, the functional form for $\chi_{+-}(q,\omega)$ is consistent with the spin-wave peak dominating the spectral weight, as expected in the hydrodynamic regime. Third, the Lorentzian form for $\chi_{\parallel}(q)$ has been used in Ref. 2 in a fit to the data at finite q. It should be noted that the form assumed for $\chi_{\parallel}$ is not appropriate for a strictly isotropic system since it remains finite in the $q \to 0$ limit, whereas a divergence $\sim 1/q$ is expected.[8] However, experiments on the weakly anisotropic antiferromagnet MnF_2 (where the comparison is with the longitudinal staggered susceptibility)[9] as well as on EuO suggest that Eq. (3) is a reasonable approximation even when the anisotropy is small. Fourth, the functional form for $\chi_{\perp}(q)$ is consistent with the hydrodynamic expression for the spin-wave frequency $\omega(q) = 2S\sigma/\chi_{\perp}(q)$.[10]

The essential approximation in the treatment of the dipolar interaction below T_c is the neglect of the nonsecular terms (i.e., those which do not commute with the longitudinal component of the total spin). In the mode coupling approach such an approximation is justified as long as the spin-wave modes do not overlap the central mode. In this approximation we have

$$\omega(\vec{q}) = \mathfrak{D}q^2 + 2\pi g^2\mu_B^2 S\sigma(q_x^2 + q_y^2)/q^2 v, \tag{5}$$

where v is the volume per spin. The value assumed for the spin-wave dispersion constant $\mathfrak{D}$ $(=2JS\sigma a^2)$, is in agreement with the experimental data in EuO.[4] Even at low temperatures, Eq. (5) is incorrect in the long-wavelength limit. We can compensate for this by multiplying the calculated values of the spin-wave linewidths by the ratio of the exact to approximate [i.e., Eq. (5)] harmonic spin-wave frequencies, as suggested by detailed balancing symmetry.[11]

In the mode coupling analysis we make several approximations which by now are more or less standard[5,6]: (a) a random-phase-approximation decoupling of the static four spin functions; (b) exponential time dependence of the two spin functions; (c) we have neglected the real part of the spin wave self-energy, which contributes a small frequency shift; (d) the upper limits in the integrals over wave vector are chosen sufficiently large so as to have a negligible effect on the solutions. In addition, in the evaluation of the terms associated with the dipolar interaction we replace combinations of components of the Fourier transform of the dipolar tensor by their equivalent angular averages.

In terms of the scaled variables $\vec{x}=\vec{q}/\kappa_1$, $\vec{y}=\vec{k}/\kappa_1$, $\tilde{\Gamma}=\Gamma/\mathfrak{D}\kappa_1^2$, $\tilde{\Lambda}=\Lambda/\mathfrak{D}\kappa_1^2$, and $\tilde{\omega}=\omega/\mathfrak{D}\kappa_1^2$ the mode coupling equations take the form

$$\tilde{\Gamma}(x)=\alpha(1+x^2)\int d\vec{y}\left(\frac{[y^2-(\vec{x}-\vec{y})^2]^2[\tilde{\Lambda}(y)+\tilde{\Lambda}(|\vec{x}-\vec{y}|)]}{(\phi^2+y^2)[\phi^2+(\vec{x}-\vec{y})^2]}\{[\tilde{\Lambda}(y)+\tilde{\Lambda}(|\vec{x}-\vec{y}|)]^2+[\tilde{\omega}(y)-\tilde{\omega}(|\vec{x}-\vec{y}|)]^2\}^{-1}\right), \tag{6}$$

$$\tilde{\Lambda}(x)=\alpha(\phi^2+x^2)\int d\vec{y}\left(\frac{[y^2-(\vec{x}-\vec{y})^2]^2+\frac{9}{5}\phi^4}{(\phi^2+y^2)[1+(\vec{x}-\vec{y})^2]}[\tilde{\Lambda}(y)+\tilde{\Gamma}(|\vec{x}-\vec{y}|)]\{[\tilde{\Lambda}(y)+\tilde{\Gamma}(\vec{x}-\vec{y})]^2+[\tilde{\omega}(x)-\tilde{\omega}(y)]^2\}^{-1}\right), \tag{7}$$

with

$$\alpha=k_B T_c\,\kappa_1 a/64\pi^3 J S^2\sigma^2. \tag{8}$$

Since κ_1 varies with temperature approximately as σ^2 the parameter α is at most weakly temperature dependent. In the case of EuO we obtain $\alpha\approx 0.023$. Also, we have

$$\phi=(2\pi g^2\mu_B^2/3vJ)^{1/2}/\kappa_1 a. \tag{9}$$

For modes $q\lesssim\kappa_1$, ϕ is a measure of the importance of the dipolar interaction relative to the isotropic exchange interaction.

Since the secular part of the dipolar interaction does not commute with the transverse components of the total spin we take the solutions to the mode coupling equations to be of the form

$$\Gamma(q)=\mathfrak{D}\kappa_1^2\gamma(q/\kappa_1)^2, \tag{10}$$

$$\Lambda(q)=\mathfrak{D}\kappa_1^2[\lambda_0+\lambda_1(q/\kappa_1)^2+\lambda_2(q/\kappa_1)^4]. \tag{11}$$

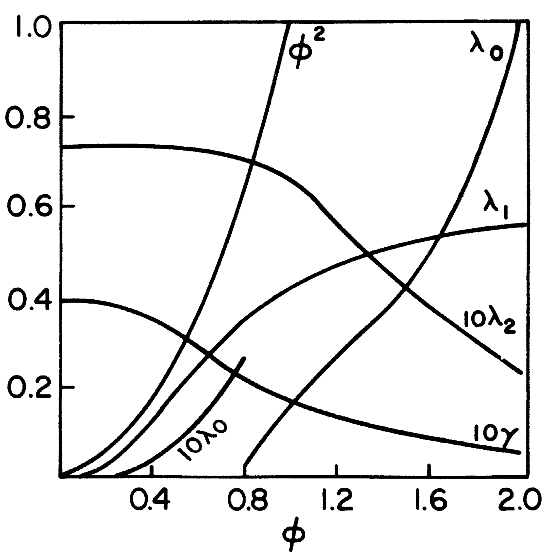

FIG. 1. γ, λ_0, λ_1, and λ_2 vs ϕ for EuO. ϕ^2 is plotted for comparison.

Our calculated values of $\gamma, \lambda_0, \lambda_1, \lambda_2$ are shown as functions of ϕ in Fig. 1 for the range $0\leqslant\phi\leqslant 2$. It should be noted that λ_0 and λ_1 vanish in the $\phi=0$ limit whereas λ_2 and γ remain finite. Thus our results are consistent with dynamic scaling for a purely isotropic system with a dynamic exponent $z=\frac{5}{2}$.[12] As ϕ increases beyond 1 the approximate treatment of the dipolar interaction begins to break down, and the theory becomes qualitatively incorrect.

In Figs. 2 and 3 we have compared the predictions of the theory against the measured values

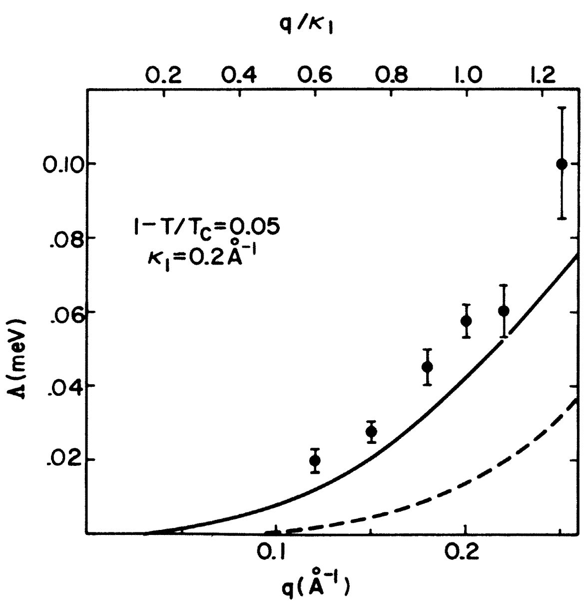

FIG. 2. Angular average of the spin wave linewidth $\Lambda(q)$ vs q for $1-T/T_c=0.05$. The solid curve is with the dipolar interaction; the broken curve is for the isotropic magnet ($\phi=0$). The upper scale shows values of the ratio q/κ_1. The data are from Ref. 3. The curves here and in Figs. 3 and 5 were calculated with $J=0.0626$ meV, $\kappa_1 a=3.28|T/T_c-1|^{0.68}$ $(T>T_c)$, and $\kappa_1 a=7.87\times|1-T/T_c|^{0.68}$ $(T<T_c)$.

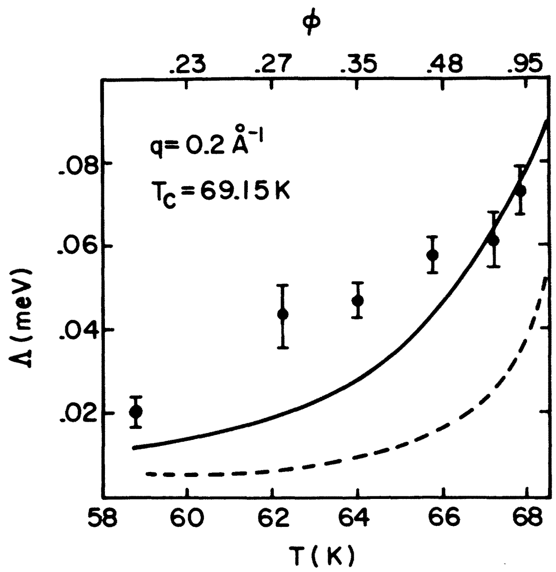

FIG. 3. Angular average of the spin-wave linewidth $\Lambda(q)$ vs T for $q=0.2$ Å^{-1}. The solid curve is with the dipolar interaction; the broken curve is for the isotropic magnet. The upper scale shows the corresponding values of ϕ. The data are from Ref. 3.

of the spin wave linewidths, both as a function of wave vector at fixed temperature (Fig. 2) and as a function of temperature at fixed wave vector (Fig. 3). The results with the dipolar interaction are shown as solid curves. The broken curves are calculated with $\phi=0$. The data which have been limited to the region of quantitative applicability of the theory, $q/\kappa_1 \lesssim 1$, $\phi<1$, are seen to be in rather good agreement with the theory.

The analysis of the spin dynamics above T_c proceeds in a similar fashion. However in this case it is necessary to include both the secular and the nonsecular parts of the dipolar interaction.[13] The functional form for the dynamic susceptibility is identical to Eq. (1). The static susceptibility $\chi(q)$ is taken to be $(1/2Ja^2)/(\kappa_1^2+q^2)$ with the appropriate experimental correlation length. The coefficient $(1/2Ja^2)$ here and in Eq. (3) is chosen so that at T_c, $\chi_{\parallel}(q)=\frac{1}{2}\chi_{\perp}(q)=\chi(q)$ in the absence of anisotropy. The mode-coupling equation becomes

$$\tilde{\Gamma}(x)=\beta(1+x^2)\times\int d\vec{y}\,\frac{[y^2-(\vec{x}-\vec{y})^2]^2+6\phi^4}{(1+y^2)[1+(\vec{x}-\vec{y})^2][\tilde{\Gamma}(y)+\tilde{\Gamma}(|\vec{x}-\vec{y}|)]}, \qquad (12)$$

where $\vec{x}=\vec{q}/\kappa_1$, $\vec{y}=\vec{k}/\kappa_1$, $\tilde{\Gamma}=\Gamma/J(\kappa_1 a)^{5/2}$, $\beta=k_B T_c/16\pi^3 J$, and ϕ is defined by Eq. (9). Equation (12) has been solved with the ansatz $\tilde{\Gamma}(x)=\lambda_0'+\lambda_1' x^2$. The results obtained for λ_0', which is proportional to the zero-field spin-spin relaxation rate, and λ_1', the scaled spin diffusion constant, are shown in Fig. 4. The parameter λ_0' vanishes as ϕ^4, whereas λ_1' remains finite in the limit as ϕ approaches zero. Our results are thus consistent with dynamic scaling with $z=\frac{5}{2}$ for the isotropic system and a spin-spin relaxation rate which varies as $\kappa_1^{-3/2}$ for small ϕ.[13]

Numerical values for the paramagnetic spin

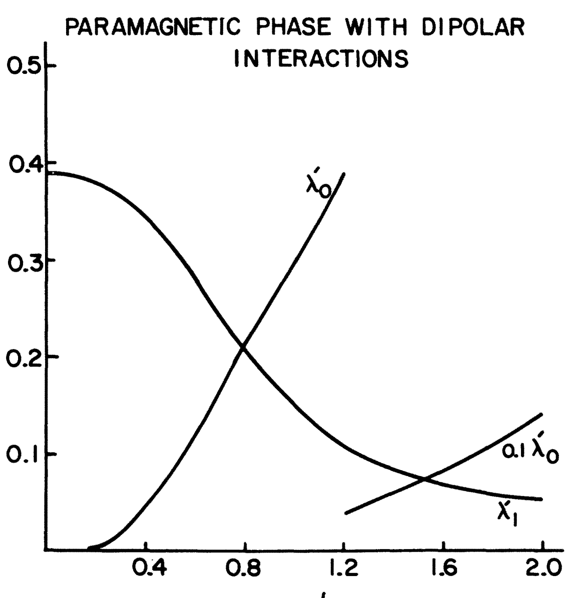

FIG. 4. λ_0' and λ_1' vs ϕ. The curves were calculated with $\beta=0.023$. Curves corresponding to other values of β (e.g., $\beta=0.192$ in EuO) can be obtained by multiplying λ_0' and λ_1' by the ratio $(\beta/0.023)^{1/2}$.

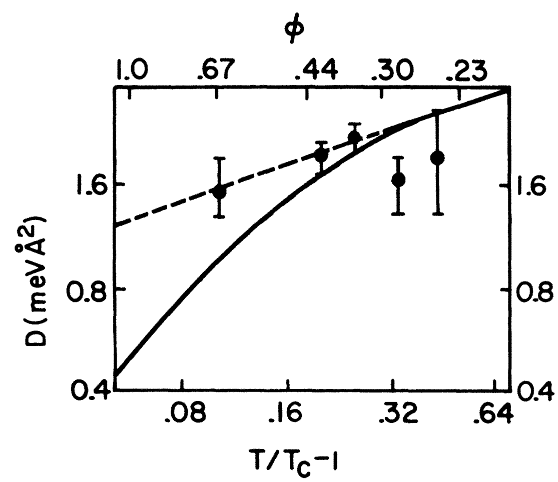

FIG. 5. Log-log plot of the paramagnetic spin diffusion constant D vs T/T_c-1. The solid curve is with the dipolar interaction; the broken curve is for the isotropic magnet. The upper scale shows the corresponding values of ϕ. The data are from Ref. 4.

diffusion constant D are shown in Fig. 5 along with the predictions of the isotropic theory[14,15] which seem to be in slightly (but probably not significantly) better agreement with the data. As with the analysis below T_c the theory breaks down for ϕ [defined by Eq. (9)] >1. Because of the difference in the magnitudes of κ_1, below T_c, ϕ equals 1 when $1 - T/T_c = 0.015$, whereas above T_c, ϕ equals 1 at $T/T_c - 1 = 0.06$. It is worth pointing out that above T_c the condition $\phi = 1$ is equivalent to $g^2\mu_B^2\chi(0) = 3/4\pi$, which is approximately the same as the boundary between isotropic and dipolar dynamics inferred in Ref. 7.

We have also investigated the critical dynamics at T_c. In this case the dipolar interaction modifies the wave-vector dependence of $\Gamma(q)$. We find the leading corrections to be of the form

$$\Gamma(q) = A q^{5/2}[1 + a_1(q_c/q)^2 + O((q_c/q)^4)], \qquad (13)$$

where A is a parameter characteristic of the isotropic system, $q_c = (2\pi g^2\mu_B^2/3vJa^2)^{1/2}$, and a_1 is a dimensionless constant. The value for A given by mode coupling theory[16] exceeds the experimental value by about 60%.[4] However, the theory does predict a value for a_1 which is in better agreement with experiment, where the data can be adequately fit with $a_1 = 1$.[4]

We believe our calculations establish that mode-coupling theory provides a reasonably quantitative description of the critical dynamics of a cubic ferromagnet with both exchange and dipolar interactions. There is one possible exception to this in the characterization of the longitudinal dynamics in the ordered phase. Assuming diffusive behavior for the longitudinal mode the theory, where applicable, predicts $\Gamma(q)/\omega(q) \lesssim 0.04$. As was mentioned a longitudinal mode of any form has not yet been detected in EuO or in Fe.[17] However, a central peak below T_c has been reported for the cubic ferromagnet CoS_2, a system which appears to be intermediate between EuO and Fe in the relative importance of the dipolar interaction.[18] The theory outlined in this paper can be applied to these other systems provided the susceptibilities have the form displayed in Eqs. (1)–(4). To obtain quantitative estimates for the relaxation rates information must be available about the spin wave dispersion, the saturation magnetization, and the inverse correlation lengths.[19]

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[19]Calculations of $\Lambda(q)$ for iron, with $\alpha \approx 0.03$ and $\phi \approx 0$, give results which are within a factor of 2 of the spin-wave linewidth data reported in Ref. 17.