Dipolar Interactions in the Critical Dynamics of Heisenberg Ferromagnets

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It is shown that in contrast to previous assumptions the real magnetic dipole interaction has to be considered as a possible source of deviations of the dynamical exponent z from $\frac{5}{2}$ as observed recently on Fe and Ni. While the dipolar crossover temperatures agree with experiment, at T_c the dynamic crossover around the dipolar wave vector q_d as predicted by current theories is absent. It is argued that this effect is related to a hitherto overlooked dynamical decoupling between transverse and noncritical longitudinal fluctuations if q exceeds the inverse correlation range κ.

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Using advanced neutron techniques Mezei[1] extended earlier investigations of the critical dynamics in iron[2] down to energy and momentum transfers of $\hbar\omega_q = 1\ \mu$eV and $q \approx 0.006$ Å^{-1}. At the Curie temperature the relaxation rate Γ_q continued to slow down proportionally to q^z, with $z = \frac{5}{2}$, in accordance with dynamical scaling for Heisenberg ferromagnets,[3] whereas above T_c strong deviations from the established dynamical scaling law $\Gamma_q = q^z \Omega(\kappa/q)$ [2,3] occurred (κ is the inverse correlation length). Some indication of a breakdown of pure Heisenberg behavior was seen before in hyperfine experiments on Fe and Ni about reduced temperatures of $t = T/T_c - 1 \approx 0.008$ and was attributed to spin-nonconserving forces.[4] While dipolar interactions were assumed to be too weak to account for this effect, it was proposed that one consider spin-lattice, pseudodipolar couplings[2] and thermodynamic random fields.[1] It is the purpose of this Letter to present quantitative arguments suggesting that the real magnetic dipole-dipole interaction may well be the origin of the spin-nonconserving force and to discuss new consequences following from the experimental data available around the dipolar crossovers of Fe and EuO.

Dynamic crossover to dipolar critical ($z = 2$) behavior was first seen in studies of the relaxation rate of the magnetization, $\Gamma_{q=0}$, of nonmetalic cubic ferromagnets like $CdCr_2Se_4$ and EuO,[5] and could quantitatively be explained with a mode-coupling theory.[6,7] Accordingly, the crossover is related to the anisotropic $\vec{q}$-dependent susceptibility (SI units, Fisher exponent $\eta = 0$)

$$[\chi_{\vec{q}}^{\alpha}(t)]^{-1} = q_d^{-2}[\kappa^2(t) + q^2] + (q^{\alpha}/q)^2, \qquad (1)$$

in which the longitudinal component, $\vec{S}^{\alpha} = S\vec{e}^{\alpha} \parallel \vec{q}$, no longer grows to criticality if κ and q become smaller than the dipolar wave vector q_d. The numerical value of q_d can be obtained from the $q \to 0$ limit of Eq. (1), $[\chi_0^{\alpha}]^{-1} = (\kappa/q_d)^2 + N^{\alpha}$ (N^{α} is the demagnetization coefficient); using the experimental results on the internal homogeneous susceptibility $\chi_0 \equiv (q_d/\kappa)^2 = 0.0017 t^{-1.33}$ [8] and $\kappa = (1.10\ \text{Å}^{-1}) t^{0.67}$ [9] I find for Fe $q_d = 0.045$ Å^{-1} just where Γ_q exhibits strong effects by spin-nonconserving forces (see Fig. 2 of Ref. 1). Equation (1) also allows us to estimate the dipolar crossover temperature t_d from $\kappa(t_d) = q_d$, i.e., where $\chi_0(t_d) = 1$,[10] which for Fe takes the value $t_d = 0.008$ in excellent agreement with the value observed in the hyperfine experiments.[4] For Ni, using $\chi_0 = 0.00069 t^{-1.31}$ [11] I find $t_d = 0.0043$ consistent with the crossover reported in Ref. 4. These results for q_d and t_d are about one order of magnitude larger than those derived previously from an unjustified expression, $t_d = G_d/4\pi\sqrt{2}S(S+1)T_c$ with the dipolar coupling constant $G_d = (g\mu_B)^2/\mu_0 k_B v_{spin}$, which yielded $t_d = 0.0004$ for Fe and Ni.[4] On this basis the real dipolar interaction was excluded as the origin of the spin-nonconserving forces. As a more realistic estimate for the dipolar crossover in terms of G_d and T_c I propose $t_d = [S(S+1)G_d/3T_c]^{3/4}$ [12] giving, e.g., 0.009 for Fe and 0.004 for Ni.

To date the effect of dipolar anisotropy on the order-parameter correlation function

$$C_{\vec{q}}^{\alpha}(\omega) = \chi_{\vec{q}}^{\alpha} f^{\alpha}(\omega/\Gamma_{\vec{q}}^{\alpha})/\Gamma_{\vec{q}}^{\alpha} \qquad (2)$$

has been observed directly only in $C_{q=0}(\omega)$, where it leads to crossover from $z = -\frac{3}{2}$ to $z = 2$.[5] Hyperfine methods probe the integral $\int C_{\vec{q}}(\omega = 0) d^3q$ so that only an effective value of the dynamic critical exponent z is obtained, which samples the region of small q.[4] Diffuse inelastic neutron scattering measures the correlations between spin components perpendicular to the scattering vector $\vec{Q}$,[13]

$$d^2\sigma/d\Omega\, d\omega \propto \sum_{\alpha} [1 - (Q^{\alpha}/Q)^2] C_{\vec{q}}^{\alpha}(\omega). \qquad (3)$$

Consequently, one has to distinguish between small-angle measurements, where $\vec{q}=\vec{Q}$, and scattering around a Bragg position with $\vec{q}=\vec{Q}-\vec{\tau}_B$. The latter, detecting a mixture between longitudinal and transverse correlations, was used in the classical investigations on Fe[2] and Ni,[14] but since the minimum q values were more than a factor of 2 larger than q_d the dipolar anisotropy played no role. The same applies to the more recent (small-angle) work on Co.[15]

Neutron measurements extending to $q<q_d$ were performed on EuO[16] and Fe[1,17] by the small-angle technique, which according to Eq. (3), only detects the transverse correlation function $C_{\vec{q}}^{\perp}(\omega)$. For both examples, the static susceptibility $\chi_{\vec{q}}^{\perp}$ obeyed the classical Ornstein-Zernike law to below $q=q_d$ as expected from Eq. (1). The behavior of the corresponding relaxation rates $\Gamma_{\vec{q}}^{\perp}$ will be discussed with use of Fig. 1 where the regions of Heisenberg-isotropic (I) and dipolar-anisotropic (D, $z=2$) dynamics are separated by $\kappa^2+q^2=q_d^2$ [see Eq. (1)]. In I, the dipolar-induced spin relaxation with $z=-\frac{3}{2}$ dominates over the spin diffusion at small q ($<q_d^2/\kappa$, region I_r).[6,18] In the hydrodynamic ($q<\kappa, I_h$) and critical ($q>\kappa, I_c$) parts of the isotropic region, dynamical scaling with $z=\frac{5}{2}$ was shown to be valid for the EuO data.[16] Also on EuO, conventional slowing down, $z=2$,[6,18,19] was confirmed in the hydrodynamic dipolar region ($\boldsymbol{D}_h$).[20] For Fe, $z=\frac{5}{2}$ scaling turned out to be valid in I_c and in the upper part of I_h ($q>q_d$), whereas at smaller q significant deviations from the Resibois-Piette function for $\Omega(q/\kappa)$ were observed (see Fig. 3 of Ref. 1 and Ref. 21). It was recently argued that strong, but short-ranged pseudodipolar forces in Fe might be responsible for these deviations arising at larger κ.[22]

Here I want to focus on the critical dipolar region, $\boldsymbol{D}_c$, where for both Fe and EuO, a very interesting effect occurs: $z=\frac{5}{2}$ remains valid; i.e., at $q>\kappa$ *the dipolar anisotropy does not influence the relaxation rate of the transverse fluctuations.* This clear disagreement with current theoretical predictions is illustrated in Fig. 2 where for the extreme case, $T=T_c$, $\Gamma_{\vec{q}}^{\perp}(\kappa=0)$ normalized to the mode-coupling result, $Aq^{2.5}$ (valid for $q>q_d$), is plotted against q/q_d. As can be seen, no indication appears for either the crossover to $\Gamma_q \sim q^2$ predicted by the mode-coupling calculations[6,18,19] or the factor of a dipolar correction to scaling, $\frac{1}{2}[1+(q_d/q+q/q_d)\arctan(q_d/q)]$, proposed by Dietrich, Als-Nielsen, and Passell.[16] Though the latter factor was claimed to explain

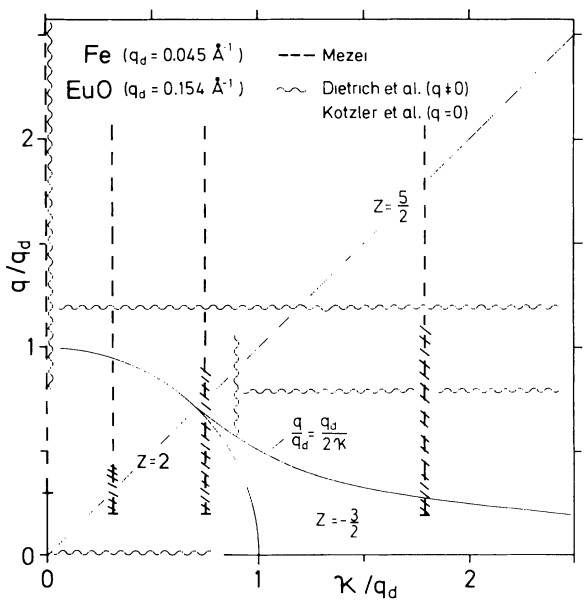

FIG. 1. κ-q plane (normalized to dipolar wave vector q_d) with regions of different dynamical critical exponent z for cubic Heisenberg ferromagnets with dipolar interaction according to mode-coupling theory (Refs. 6 and 18). Dashed and wavy lines indicate the experimental scans at constant q or $\kappa(T)$ on Fe (Ref. 1) and EuO (Ref. 16). Hatched parts designate violation of the $z=\frac{5}{2}$ dynamical scaling for Fe.

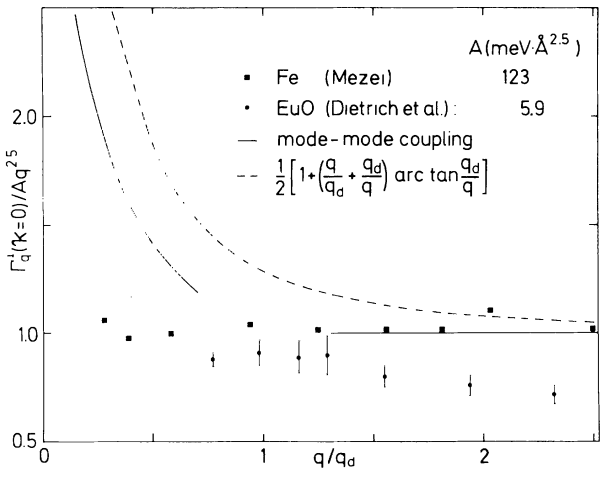

FIG. 2. Relaxation rate of transverse fluctuations at T_c around the dipolar wave vector q_d for Fe (q_d = 0.045 Å^{-1}) and EuO (q_d = 0.154 Å^{-1}). Comparison with available theories shows substantial discrepancies below $q=q_d$: mode-mode-coupling calculations yield $\Gamma_q = Aq_d^{1/2}q^2$ with $A=(J_2kT_c/2\pi^2v_{spin})^{1/2}$ (Refs. 6 and 23); the dashed line corresponds to a phenomenological dipolar correction to dynamical scaling (Refs. 16 and 23).

the EuO data,[16] Fig. 2 clearly shows that just around $q=q_d$, where the correction should be significant, the $\Gamma_{\vec{q}}^{\perp}$ of EuO strictly obeys the $z=\frac{5}{2}$ law. Therefore, it is more likely that the slight deviations from $z=\frac{5}{2}$ at $q>1.5q_d$ are not related to the dipolar forces but are of noncritical origin due to the rather large q_d of EuO amounting to 20% of the first Brillouin zone.

In conclusion, I have pointed out that in contrast to earlier estimates, the dipolar interaction should influence the critical dynamics investigated in the regions of low q and κ ($<q_d$) for cubic ferromagnets. My numerical values for q_d and t_d agree with experiments probing the hydrodynamic region. However, in the critical dipolar region, $\kappa<q<q_d$, the relaxation rates of the fluctuation in the transverse magnetization appear to be completely unaffected by the dipolar anisotropy, i.e., by the longitudinal fluctuations, which is in clear contradiction to current theories. Presumably, this phenomenologically proven decoupling for $q\gg\kappa$ between the critical transverse and the noncritical longitudinal fluctuations is also the reason why the pseudodipolar coupling in Fe exerts no effect on $\Gamma_{\vec{q}}^{\perp}$. As a result of the short range of this interaction (r_{pd}), it should influence $\Gamma_{\vec{q}}^{\perp}$ only when κ exceeds a characteristic pseudodipolar wave vector, $q_{pd}\approx r_{pd}^{-1}$, as seen for instance at $q=0$ on Ni.[24] Since the pseudodipolar force has the same symmetry as the dipolar one, I expect $\Gamma_{\vec{q}}^{\perp}$ to be unchanged in the critical region for $q\approx q_{pd}\gg\kappa$. There in fact dynamical scaling with $z=\frac{5}{2}$ continued to be valid.[1]

In order to investigate the so far unknown dynamics of the longitudinal fluctuations I propose to extend the measurements of the scattering function, $S_{\vec{q}}^{\alpha}(\omega)$, around a Bragg reflection into the dipolar region, $(\kappa^2+q^2)<q_d^2$. There it would be interesting to follow the behavior of $S_{\vec{q}}^{\parallel}$ and $S_{\vec{q}}^{\perp}$ from the apparently decoupled limit, $q>\kappa$, to the hydrodynamic dipolar region, $q<\kappa$, where the noncritical longitudinal fluctuations, e.g., give rise to the observed conventional slowing down.[5] Moreover, this problem might challenge the theorists to develop a coherent picture of dipolar anisotropic dynamics operative in all ferromagnets, not only above T_c but also in the ordered phase.

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