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Dynamical crossover to dipolar behavior in isotropic ferromagnets at and above T_c

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Apparent discrepancies between neutron spin echo and neutron scattering experiments in the paramagnetic phase of isotropic ferromagnets are explained. Dipolar forces play an important role in the scaling region and cause, especially at T_c, a crossover in the shape function of the dynamical correlation from a non-Lorentzian shape (nonexponential in time) at large values of the wave vector to a Lorentzian shape (exponential) at small values, even in a region where the effective critical exponent of the energy linewidth is given by $z_{\text{eff}}=2.5$. This explains the experimental situation in EuO. For $T>T_c$, the inclusion of the shape crossover improves the agreement between the theoretical and the measured linewidth in Fe. Predictions are made for the longitudinal relaxation function at T_c.

Recent neutron scattering experiments[1-5] provided evidence of a non-Lorentzian shape function for the dynamical correlation function in isotropic Heisenberg ferromagnets. Agreement with dynamical critical theory was established.[6-8] However, in neutron spin-echo measurements in EuO (Ref. 9) a Lorentzian line shape was found, and explanations by asymptotic theories within the Heisenberg model failed.[10] On the other hand, the energy width $\Gamma_{\mathbf{q}}$ of both experiments turned out to be in agreement with the prediction of renormalization-group (RG) theory for isotropic Heisenberg ferromagnets ($\Gamma_{\mathbf{q}}\sim q^{2.5}$).[4,9]

Though the exchange interaction is much stronger in isotropic ferromagnets, dipolar interactions are also present, and RG theory predicts that, in the presence of those interactions, the true asymptotics are described by the dipolar fixed point rather than by the isotropic Heisenberg fixed point.[11] This would mean that the asymptotic dynamics is that of a relaxation model with a nonconserved order parameter with the critical exponent $z=2$ (we neglect $\eta\sim 0.05$ in the following), instead of a model with mode-coupling terms and a conserved order parameter with $z=2.5$. At T_c, for decreasing wave vector q, a dynamical crossover between these two cases had been expected at the wave vector $\mathbf{q}_d$ where the static crossover takes place. This was not found in experiments. Recently, mode-coupling theory was applied to this problem[12] and the linewidth was calculated, assuming a Lorentzian shape, leading to the result that the dynamic crossover in the *linewidth* sets in at wave vectors one order of magnitude *smaller* than expected and in agreement with experiments. It is the aim of this paper to show that the crossover in the *line shape* sets in a *larger q* values (of the order of the static value) for which the linewidth $\Gamma_{\mathbf{q}}$ still behaves like $q^{2.5}$. For that purpose we solve the mode-coupling equations without the Lorentzian approximation. We also consider the situation at temperatures above T_c.

We start from the mode coupling equations for a ferromagnet described by the Hamiltonian

$$\mathcal{H}=\sum_{i,j,\alpha}J_{ij}S_i^\alpha S_j^\alpha+\mathcal{H}_d\,. \tag{1}$$

The short-range interaction part is given by the exchange couplings J_{ij}, and the sum runs over the sites i,j of the spins $\mathbf{S}_i$ and $\alpha=1,2,3$. $\mathcal{H}_d$ contains the isotropic dipolar interaction of the spins. The Fourier-transformed spin $\mathbf{S}_{\mathbf{q}}$ is decomposed into a longitudinal and a transverse part with respect to the wave vector $\mathbf{q}$. Then by standard techniques the equations for the longitudinal and transverse relaxation functions $F^{L,T}$ have been derived.[12] In the asymptotic region these functions satisfy the scaling laws with λ arbitrary

$$F^{L,T}(\mathbf{q},\kappa,g,t)=F^{L,T}(\lambda\mathbf{q},\lambda\kappa,\lambda^2 g,\lambda^{-2.5}t)\,, \tag{2}$$

where the arguments are wave vector, inverse correlation length, dipolar strength, and time. Introducing scaling variables $u=tAq^{5/2}$, $x=\kappa/q$, and $y=\sqrt{g}/q$ we find for $F^\alpha(1,x,y,u)=f^\alpha(x,y,u)$

$$\frac{d}{du}f^\alpha(x,y,u)=-\int_0^u du'k^\alpha(x,y,u-u')f^\alpha(x,y,u')\,. \tag{3}$$

The kernel $k^\alpha(x,y,u)$ reads

$$k^\alpha(x,y,u)=\frac{2\pi^2}{\chi^\alpha(x,y)}\int_0^\infty d\rho\int_{-1}^1 d\eta[v^\alpha_{\beta\gamma}\rho_-^{-2}\chi^\beta(x/\rho,y/\rho)\chi^\gamma(x/\rho_-,y/\rho_-)f^\beta(\rho^{2.5}u,x/\rho,y/\rho)f^\gamma(\rho_-^{2.5}u,x/\rho_-,y/\rho_-)]\,, \tag{4}$$

with $\rho_-=(\rho^2-2\rho\eta+1)^{0.5}$ and the mode-coupling vertices[12]

$$v^L_{TT}=2\eta^2(\rho\eta-\tfrac{1}{2})^2,\quad v^T_{LL}=2(1-\eta^2)\left[\rho\eta-\tfrac{1}{2}+\frac{y^2}{2}\right]^2,\quad v^T_{TT}=(1-\eta^2)\left[1+\frac{1}{2\rho_-^2}\right](\rho\eta-\tfrac{1}{2})^2\,,$$
$$v^T_{LL}=(1-\eta^2)\frac{1}{2\rho_-^2}(\rho\eta-\tfrac{1}{2})^2,\quad v^T_{LT}=\left[2-\left[1+\frac{1}{\rho_-^2}\right](1-\eta^2)\right]\left[\rho\eta-\tfrac{1}{2}+\frac{y^2}{2}\right]^2, \tag{5}$$

and the scaled static susceptibilities $\chi^a(x,y)$. The nonuniversal time scale A is related to the exchange couplings. Within the spirit of the asymptotic theory it may be determined by comparison with, say, the energy width at some q vector (e.g., the large-q region). In the case of EuO, it turns out that it suffices to take the value $A=1.3$ meV Å$^{2.5}$ known from measured exchange couplings in order to reach satisfactory agreement. Choosing a value for A that fits Γ_q to the experimental linewidth alters A by about 10%. The constant g is determined by the ratio of dipolar to exchange interaction. [$g=0.59$ (EuO), 0.02 (Fe); q and κ are measured in lattice units $a=5.14$ Å (EuO), 2.87 Å (Fe); and the temperature dependence of κ is taken from experiment.] Equations (3) have already been solved, approximating the shape of $f^{L,T}(x,y,u)$ by an exponential, leading to a set of self-consistent equations for the relaxation rates $\gamma^{L,T}(x,y)$. In Ref. 13, an approximate solution of the mode-coupling equations was suggested, by using $\exp(-u/\gamma^{L,T})$ with the self-consistently determined $\gamma^{L,T}$ on the right-hand side and performing the time integrations. This amounts to the first iteration of Eqs. (3). The deviation in the shape from an exponential (Lorentzian) is overestimated by this approximation. Therefore, we have iterated Eqs. (3) until self-consistency was achieved.

The neutron scattering cross section in general contains both the longitudinal and the transverse relaxation function. Under the condition of unpolarized incoming neutrons and scattering near the reciprocal lattice vector $\tau=0$ only the transverse fluctuations are measured. This is the case in the experiments in EuO or Fe mentioned above. The scattering cross section is given by

$$S^T(q,\kappa,g,\omega)\sim\chi^T(q,\kappa,g)F^T(q,\kappa,g,\omega)\,, \tag{6}$$

where, for the static transverse susceptibility χ^T, we use the Ornstein-Zernike form $\chi^T=(\kappa^2+q^2)^{-1}$.

Let us first consider the case $T=T_c$. In Fig. 1, $f^T(0,y,s)$ is shown for various values of the scaling variable y. For small values of y (large q or small g) the critical shape of the isotropic ferromagnet with short-range interaction is recovered.[14] With increasing y, the shape crosses over to a pure exponential as one expects from RG theory considerations. However, this crossover takes place at y values of the order of unity (well within the experimentally accessible region in EuO and similar to the statics). In remarkable contrast to the shape, the ω linewidth

$$\Gamma_q^T=A\gamma^T(0)q^{2.5}\Omega^T(y),\quad \Omega^T(y)=\gamma^T(y)/\gamma^T(0)$$

changes its behavior from a constant $\Omega(y)$ (that means $\Gamma_q^T\sim q^{2.5}$) to an $\Omega^T(y)\sim y^{0.5}$ behavior (that means $\Gamma_q^T\sim q^2$) at y values an order of magnitude smaller than in the statics; this was first noticed in Ref. 12. So far, those small y values have not been reached experimentally. This explains why at large-q vectors in EuO a non-Lorentzian shape has been measured,[4] but in spin-echo experiment at small q vectors an exponential has been found,[9] while both experiments found the same effective critical exponent $z_{\mathrm{eff}}=2.5$ for the ω width and the relaxation rate, respectively. In Fig. 2, we show the result of our calculation for EuO. Agreement with both width and relaxation function is achieved.

Just as in the pure short-range interaction case,[14] the deviation from the pure exponential is accompanied by the development of a strongly damped oscillation in the relaxation function. Whether this is a real effect or an artifact of the mode-coupling theory cannot be decided. In the case of pure short-range interactions the corresponding function calculated in RG theory does not show this oscillation.[8]

It has turned out that constant energy scans in neutron scattering are a very sensitive tool to investigate the shape of the dynamical correlation functions. The position of the peak q_0 and the width of the maximum in the scattering intensity depend strongly on the shape of the correla-

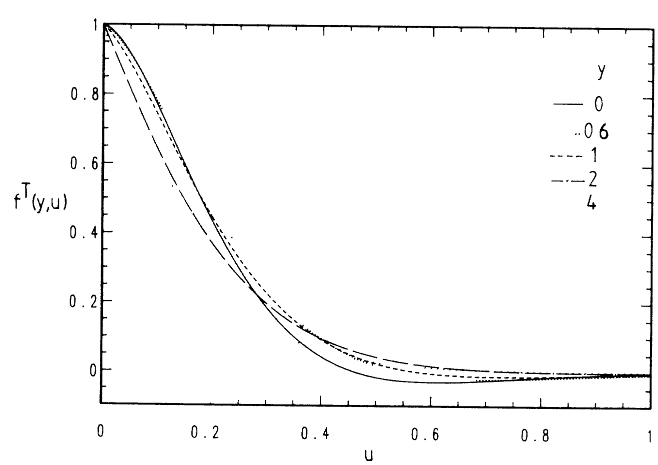

FIG. 1. The transverse spin-relaxation function $f^T(y,u)$ at T_c in scaled time $u=Aq^{2.5}t$ and inverse wave vector $y=g^{0.5}/q$, showing the shape crossover to a pure exponential decay for $y>1$.

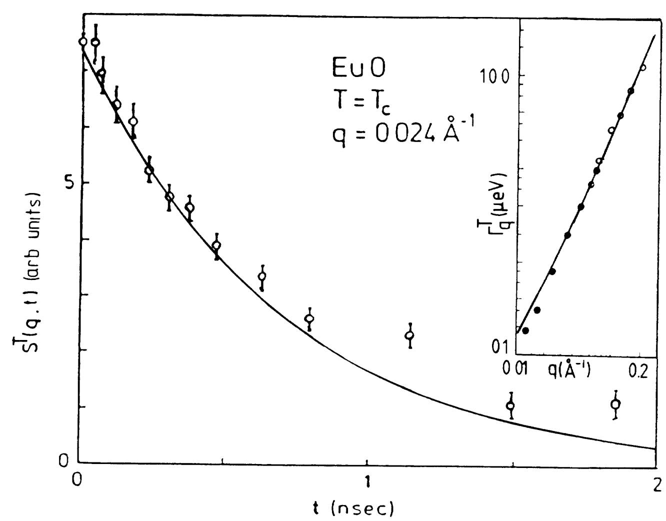

FIG. 2. Comparison between the transverse relaxation function calculated from Eqs. (3) and measurements of Mezei in EuO at T_c (Ref. 9). The nonuniversal parameter $A=1.3$ meV Å$^{-2.5}$. The inset shows the linewidth of quasielastic scattering (dots, Ref. 9; circles, Ref. 4).

tion function.[6,15] In the pure short-range interaction case for any finite energy ω, the peak position q_0 is finite and nonzero because the scattering intensity vanishes for q tending to zero.[8,14] When one includes dipolar forces, the intensity stays finite at $q=0$ and is proportional to ω^{-2}. Therefore, the peak position q_0 moves to zero for small but finite ω (we do not consider the position and the width of the peak as a well-defined quantity in this crossover energy region). In Fig. 3, we show some typical constant ω scans (the nonuniversal parameters are those of EuO). The shape of these curves for large wave vectors is the same as in the case of a pure short-range interaction, and they decay like $q^{-4.5}$, because of the scaling law for the relaxation function, Eq. (2), and because $f^T(0,\infty,s=0)$ is finite. In the region where the peak is well defined, no substantial changes in the relaxation between q_0 and ω, in comparison to the case without dipolar forces, appear.

We now turn to the results for $T>T_c$. There, two scaling variables, $x=\kappa/q$ and y, have to be considered. It turns out, by inspection of Eqs. (3)-(5) that the natural variables[12] in that case are $y/x=g^{0.5}/\kappa=tg\phi$ and $(x^2+y^2)^{0.5}=r$. The iteration of Eqs. (3) can be done for fixed ϕ, i.e., separately for each temperature $T>T_c$. For ϕ different from $\pi/2$, one may display the results as functions of x only, since one has $x=r(1+tg^2\phi)^{-1}$. However, one has to keep in mind that the physical quantities, considered as functions of x alone, do not scale in x. In Fig. 4 we compare the normalized ω linewidth $\Omega^T(x,y)$ with the measurements in Fe.[16] An improvement is obtained with respect to the theoretical results of Ref. 12 (compare with Fig. 3 there), but especially for large values of the temperature, deviations remain. Those may be due to background effects or other forces not included in our theory.

An important physical quantity, showing new interesting features, is the longitudinal relaxation function. At $\phi=0$, it is equal to the transverse relaxation function, and crosses over to a Lorentzian (exponential) as x is increased, and one enters the hydrodynamical region. A different crossover in shape is expected at $T_c(\phi=\pi/2)$.

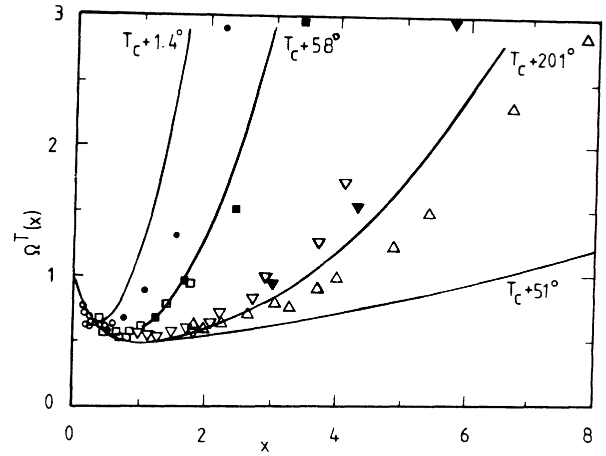

FIG. 4. Comparison of the normalized transverse width with measurements at different temperatures by Mezei in Fe (Ref. 16). $\Omega^T(x)=\gamma^T(x,y)/\gamma^T(0,0)$. The symbols are at $T_c+51°$, $T_c+21.6°$, $T_c+20.1°$, $T_c+5.8°$, $T_c+5.5°$, $T_c+1.4°$, and $T_c+1.1°$. The curve at $T=T_c$ is calculated without dipolar forces (Ref. 14).

For small values of y (large q, short-range case) again transverse and longitudinal relaxation are equal. However, as one increases y (lowers q) the shape of the longitudinal relaxation function *does not* cross over to a Lorentzian (exponential), but remains similar to the one in the short-range case (see Fig. 5). We remark that at T_c and for $q\rightarrow 0$ the longitudinal components of the spin become uncritical secondary variables with a constant relaxation rate. This crossover is also seen in the linewidth Γ_q^L. The $q^{2.5}$ behavior at small y crosses over to the q-independent behavior at larger y values, as expected from statics, and is qualitatively similar to the results of Ref. 12.

A semiphenomenological model that does not take into account dipolar forces has been suggested in Ref. 17, in order to explain the spin-echo experiments in EuO. We have applied mode-coupling theory including dipolar

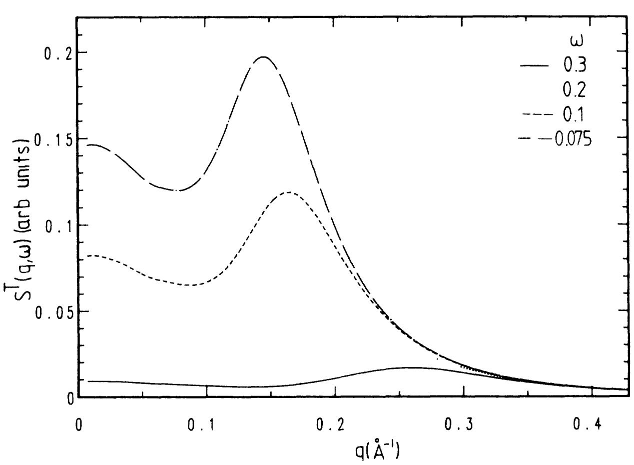

FIG. 3. Typical constant energy scans showing the transversal scattering intensity as function of wave vector q for different scaled energies. The value of A is that for EuO.

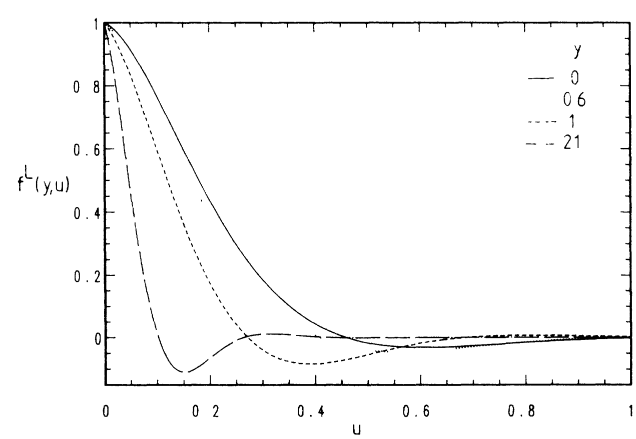

FIG. 5. The longitudinal spin-relaxation function $F^L(y,u)$ at T_c calculated from Eqs. (3). Variables are the same as in Fig. 1. No crossover to an exponential for large y is seen.

forces[12] to neutron scattering experiments, and found an explanation for apparent contradictions[18] regarding the shape of the dynamical correlations. These can be understood as asymptotic crossover phenomena between the conserved Heisenberg dynamics and the nonconserved dipolar dynamics. However, an important point is to recognize that different physical quantities (e.g., linewidth and shape) may have different crossover points.

Note added in proof: The shape crossover at T_c was also considered in a recent paper by Frey, Schwabl, and Thoma [Phys. Lett. A **129**, 343 (1988)].

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