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Shape crossover in the correlation function of isotropic ferromagnets above T_c

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We calculate numerically the asymptotic universal spin-relaxation function for an isotropic ferromagnet with short-range interaction by solving the mode-coupling equations for $T \geq T_c$. There is a crossover from a non-Lorentzian shape at T_c to a Lorentzian in the hydrodynamic region at $T > T_c$. We apply these results to the constant-energy neutron-scattering cross section and calculate the universal functions for the peak position and width in the scattering intensity as a function of wave vector and correlation length.

I. INTRODUCTION

Much progress has been made in recent years regarding the determination of the shape of the spin-correlation function by neutron scattering. Thus a direct, detailed, and quantitative comparison between theory and experiment is possible. In several isotropic ferrogmagnets deviations from the Lorentzian shape have been found in constant-momentum measurements, and especially in constant energy measurements (see, e.g., Ref. 1). This was to be expected on theoretical grounds, since renormalization-group (RG) calculations at T_c,[2,3] as well as mode-coupling calculations,[4,5] definitely lead to a non-Lorentzian shape. A comparison of the RG results at T_c with experiments was initiated in Ref. 6 and the importance of the correct shape in the interpretation of constant-energy scans was stressed. Further measurements on Ni,[7] EuO,[8] and Fe_3Si (Ref. 9) provided evidence for the shape proposed by RG theory.

The investigations of Ref. 6 were then extended to temperatures above T_c.[10] The distinct behavior of the peak position and the width in the intensity of the constant-energy measurements could be explained by the interplay between the shape crossover from the critical shape at T_c to the Lorentzian one in the hydrodynamic region further away and the change in the half-width of the scattering function. However, only the limiting behavior of these quantities could be given since the explicit shape crossover was not known. Very recently, in a RG calculation in one-loop order, this shape crossover was calculated.[11,12] However, the theoretical expressions are subject to some theoretical uncertainties. First there are ambiguities in the exponentiation procedure, which are necessary in order to fulfill exactly known scaling laws, and second, there is the restriction to one-loop order. Therefore it seems to be worthwhile to study the shape crossover and those universal crossover functions measured in the constant-energy scans by mode-coupling theory as well.

In this paper we are interested in the behavior of the asymptotic scattering function for isotropic ferromagnets with *short-range* interaction only (this implies a constant dynamic exponent $z = 2.5$). The application of our result is restricted to those regions in temperature and wave vector (the size of the region depends on the physical system) where this type of interaction dominates over the dipolar interaction. If, on the other hand, dipolar interactions become important and RG theory shows that this should be the case ultimately at criticality, another type of crossover (namely a change of the universality class) is to be expected. Then the relevant dynamics should be described by a relaxational-like model for a nonconserved order parameter (with $z = 2$) and the shape is essentially given by a Lorentzian. This type of crossover to dipolar dynamics without taking into account the shape crossover has been considered in Ref. 13. A similar crossover at very small values of momentum at T_c to a pure relaxational dynamics has been proposed in Ref. 14, in a model which takes into account the coupling of the magnetic moments to the conduction electrons in the case of an itinerant magnetic system. On the other hand, in Refs. 15 and 16 the irrelevance of this coupling to the asymptotic critical behavior was found. However, going to the background where critical fluctuations are less important, still another type of crossover sets in. In this region nonuniversal features such as the specific form of the short-range interaction, the lattice symmetry, etc., become important.

II. MODE-COUPLING EQUATIONS

As usual one starts from the Heisenberg model in order to describe the critical properties of isotropic ferromagnets

$$\mathcal{H} = -\sum_{\mathbf{x},\mathbf{x}'} J(\mathbf{x}-\mathbf{x}')\mathbf{S}_{\mathbf{x}}\cdot\mathbf{S}_{\mathbf{x}'} , \quad (1)$$

where $\mathbf{x}$ and $\mathbf{x}'$ denote the location of the three-component spin operators $\mathbf{S}_{\mathbf{x}}$, and $J_{(\mathbf{x}-\mathbf{x}')}$ is the short-ranged interaction given by exchange couplings. We are interested in the neutron-scattering cross section

$$S(\mathbf{q},\omega) = \text{const}\frac{\hbar\omega}{1-e^{-\beta\hbar\omega}}\chi_{\mathbf{q}}F(\mathbf{q},\omega) = \text{const}\,C(\mathbf{q},\omega) \quad (2)$$

with $\chi_{\mathbf{q}}$ the static susceptibility, $F(\mathbf{q},\omega)$ the Fourier transform of the spin-relaxation function $F(\mathbf{q},t)$ [with

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$F(\mathbf{q},-t)=F(\mathbf{q},t)$], and $C(\mathbf{q},\omega)$ the spin-spin correlation function [for $\beta\hbar\omega \ll 1$ one simply has $\chi_q F(\mathbf{q},\omega) \sim C(\mathbf{q},\omega)$]. Within mode-coupling theory the following equation for $F(\mathbf{q},t)$ is derived[5,16,17] in the critical regime:

$$\dot{F}(\mathbf{q},t)=-\sigma^2\int_0^t dt' K(\mathbf{q},t-t')F(\mathbf{q},t') \tag{3}$$

with the kernel

$$K(\mathbf{q},t)=(\kappa^2+q^2)\int d^3q' \frac{q^2-2\mathbf{q}\cdot\mathbf{q}'}{q'^2+\kappa^2}F(\mathbf{q}',t) \times F(\mathbf{q}-\mathbf{q}',t) \; . \tag{4}$$

Here the Ornstein-Zernike form for the static susceptibility has already been introduced (neglecting the small exponent $\eta=0.05$). The temperature dependence enters the equation only via the inverse correlation length κ, which goes to zero at the critical temperature T_c as $\kappa=\kappa_0[(T-T_c)/T_c]^{0.67}$. The constants σ and κ_0 are the only nonuniversal parameters; they have to be taken from experiment. The solution of Eq. (3) fulfills dynamical scaling with the dynamical critical exponent $z=2.5$, derived also in RG theory. This means that F and K are homogeneous functions,

$$F(\mathbf{q},t,\kappa)=F(\lambda\mathbf{q},\lambda^{-2.5}t,\lambda\kappa) \; , \tag{5a}$$

$$F(\mathbf{q},\omega,\kappa)=\lambda^{2.5}F(\lambda\mathbf{q},\lambda^{2.5}\omega,\lambda\kappa) \; , \tag{5b}$$

$$K(\mathbf{q},t,\kappa)=\lambda^{-5}K(\lambda\mathbf{q},\lambda^{-2.5}t,\lambda\kappa) \; . \tag{5c}$$

In the RG theory the dynamical shape function is calculated from the model equation of Ma and Mazenko[18] in a systematic expansion around the critical dimension $d=6$, whereas the mode-coupling equations are solved directly in $d=3$. Therefore, one may obtain different scaling functions in both theories. However, by generalizing the mode-coupling equations to d dimensions and then expanding around the dimension $d=6$ one sees the equivalence to the RG theory.[19]

In order to solve Eq. (3) it is useful, in light of Eqs. (5), to introduce scaling variables $u=\sigma q^{2.5}t$, $\eta=\kappa/q$, and the Fourier transform of $F(\mathbf{q},t)\equiv f(u,\eta)$ with respect to the scaling variable u [note $F(\mathbf{q},\omega)=(1/\sigma q^{2.5})f(s,\eta)$, $s=\omega/\sigma q^{2.5}$]. Inserting this in Eqs. (3) and (4) leads to

$$f(u,\eta)=\int_{-\infty}^{\infty}\frac{ds}{2\pi}[is+k(s,\eta)]^{-1}\exp(isu) \; , \tag{6a}$$

$$k(s,\eta)=8\pi(1+\eta^2)\int_0^\infty dx\, x^4\int_0^1 d\zeta\,\zeta^2\int_0^\infty du\,\exp(-isu)[(x_+^2+\eta^2)(x_-^2+\eta^2)]^{-1}f(ux_+^5,\eta x_+^{-1})f(ux_-^5,\eta x_-^{-1}) \tag{6b}$$

with

$$x_\pm^2=x^2\pm x\zeta+\tfrac{1}{4} \; .$$

III. RESULTS

We have solved the mode-coupling equation by iterating (6a) and (6b) starting from a Lorentzian. Since in each iteration step the function $f(u,\eta)$ has to be known in the whole (u,η) plane, an analytic expression has been fitted in every iteration step to the $f(u,\eta)$ calculated from (6a) for some lattice in the (u,η) plane and then inserted in (6b). This is the reason why we did not perform the u integration in (6b) explicitly, replacing it by an s integration, because otherwise two functions, $\mathrm{Re}k(s,\eta)$ and $\mathrm{Im}k(s,\eta)$, have to be fitted. It also turns out that it is useful to symmetrize the wave-vector dependence under the integration. The result for $f(u,\eta)$ and its Fourier transform $f(s,\eta)$ is shown in Figs. 1(a) and 1(b) and 2(a) and 2(b), respectively.

At T_c our results are identical to those Ref. 4 within the numerical accuracy [note the identification of our functions $ik(s)\sim m(\nu)$, $f(s)\sim -\mathrm{Im}g(\nu)$, and $s\sim\nu$ with those of Ref. 4, apart from two scaling factors]. Recently a similar result has been found at T_c in Refs. 20 and 21, where the q' integration in Eq. (4) was obtained by the standard method of asymptotic analysis. In Ref. 21 also a nonasymptotic theory, which combines large- and short-time behavior was presented. This leads to another type of shape crossover at T_c not treated here. In contradiction to the RG result a strongly overdamped oscillation appears in the time-dependent spin-relaxation function at T_c. This oscillation, however, does not lead to an observable structure in the Fourier transform apart from a flatter decrease at small s than in RG theory as can be seen from Fig. 2. As one increases η to the hydrodynamic regime $\eta>1$, the negative region practically disappears and a pure exponential is obtained.

Since the solution has to obey dynamical scaling with $z=2.5$ one can check at T_c ($\eta=0$) the asymptotic behavior for large s, namely $f(s,0)\sim s^{-2.6}$. In the limit $\eta\to\infty$ the behavior has to be $f(s,\infty)\sim s^{-2}$. For general values of η we have fitted the large-s behavior of the shape function by a power law in order to demonstrate the crossover from the critical to the Lorentzian decay of the shape (see Fig. 3). The deviation of 4% from the exact value at T_c is due to the inaccuracy of $f(s,\eta)$ at very small absolute values. This can be compared with the decay rate of the heuristic correlation function introduced to analyze the scattering cross section in Fe (Ref. 1) in the entire paramagnetic region.

The energy half-width $\omega_c(\mathbf{q},\kappa)$ is investigated in constant momentum scans. It can be read off from our result for $f(s,\eta)$. It is also a homogeneous function, so we write

$$\omega_c(\mathbf{q},\kappa)=7.5\sigma q^{2.5}\Omega(\eta) \ . \tag{7}$$

In Fig. 4 we compare our complete result for $\Omega(\eta)$ with the approximation of Resibois and Piette.[22] Their curve $\Omega(\eta)$ can be obtained from Eq. (6) by iterating with an s-independent kernel $k(\eta)$, which implies a Lorentzian shape for all η. We note that the difference between the two curves (solid and dashed) of Fig. 4 in the hydrodynamic regime results from dividing both half-widths by their value at T_c (this is the usual presentation of the experimental data). The data for Co (Ref. 23) and Fe (Ref. 24) indeed seem to be more compatible with the steeper descent in the small η region. However, we want to remark that only the data points at large q should be taken for a comparison with our theoretical curve since at small-q values dipolar effects set in.[13]

Constant-energy scans have turned out to be a much more sensitive tool for testing the shape than constant

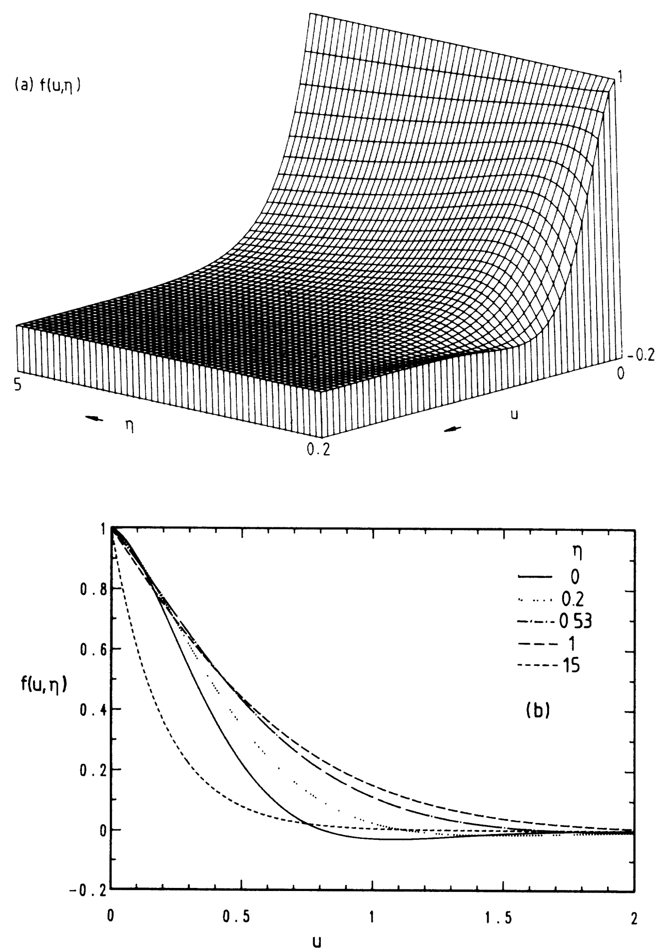

FIG. 1. The spin-relaxation function $f(u,\eta)$ in scaling variables $u=\sigma q^{2.5}t$ and $\eta=\kappa/q$: (a) in the whole (u,η) plane, (b) cuts through (a) showing the shape crossover to a pure exponential decay for $\eta>1$. From Eq. (3) we have $f(0,\eta)=1$.

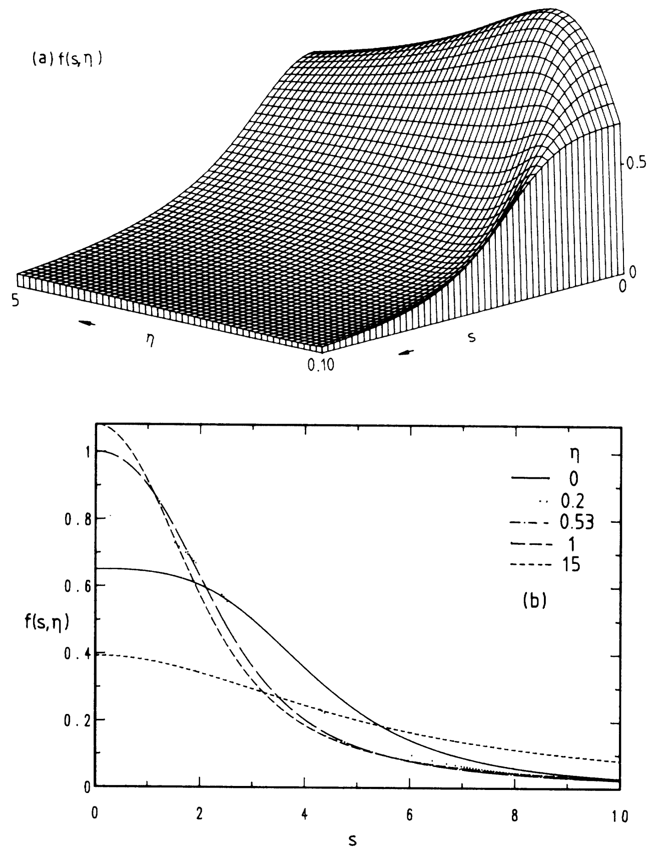

FIG. 2. The Fourier transform of the spin-relaxation function $f(s,\eta)$ in scaling variables $s=\omega/\sigma q^{2.5}$ and $\eta=\kappa/q$: (a) in the whole (s,η) plane, (b) cuts through (a) showing the shape crossover to a Lorentzian for $\eta>1$.

wave-vector scans. A characteristic feature is the appearance of a maximum in intensity[25] at a finite wave vector q_0 with a width Δq. Together with the information on the linewidth ω_c one gets an overall picture of the scattering function in the entire $(\mathbf{q},\omega)$ plane at different temperatures $T(\kappa)$. We display $S(\mathbf{q},\omega,\kappa)$ in form of constant in-

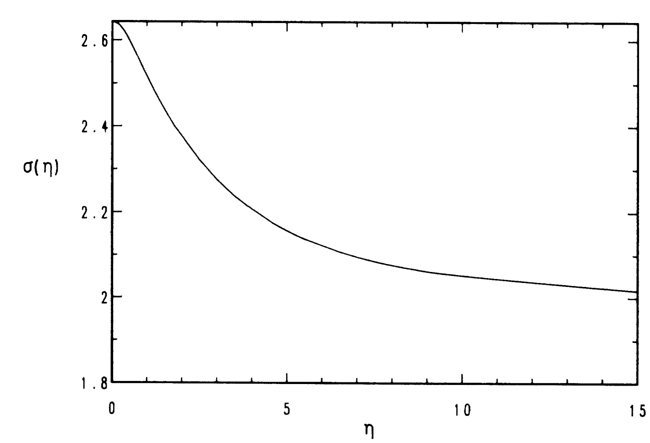

FIG. 3. Power-law behavior of the shape function $f(s,\eta)\sim s^{-\sigma(\eta)}$ for large scaling arguments as a function of η.

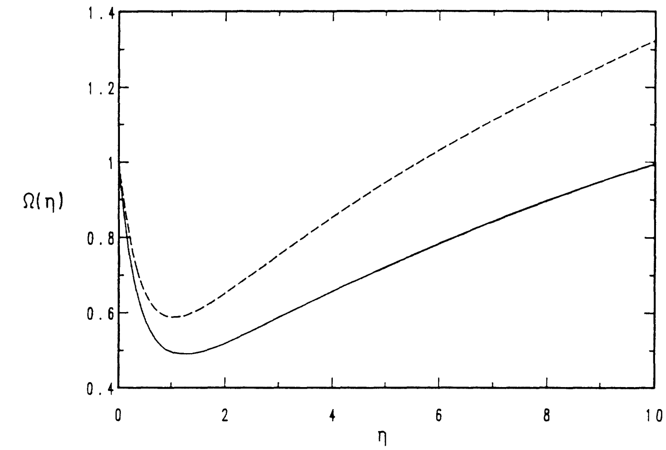

FIG. 4. Energy width at half-height of the relaxation function. Solid line: result of Eqs. 6(a) and 6(b). Dashed line: result of the Lorentzian approximation (Ref. 23). Both curves are normalized to 1 at T_c.

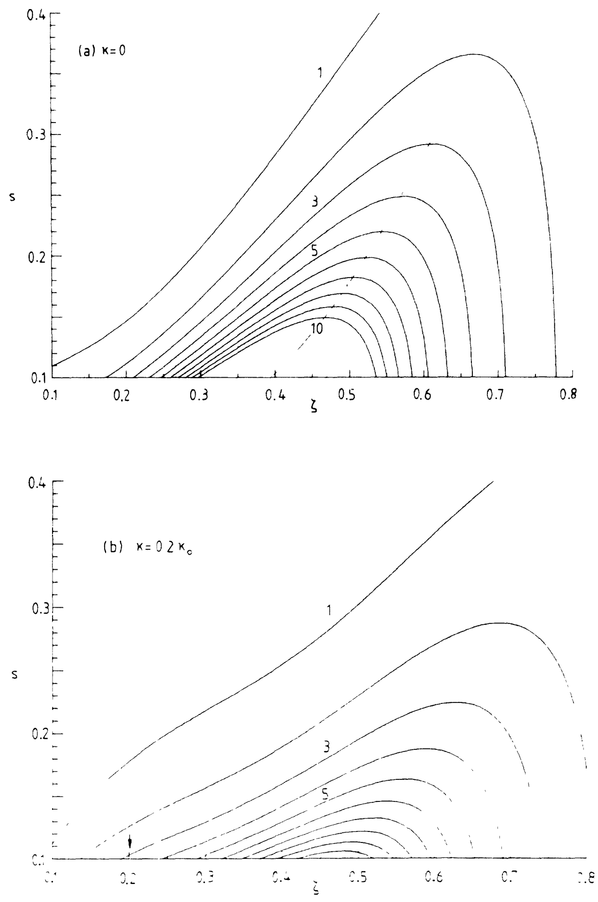

FIG. 5. Equal intensity contours of the scattering cross section at (a) T_c and (b) $\kappa=0.2\kappa_0$, $\zeta=q/\kappa_0$, and $s=\omega/\sigma q^{2.5}$. The dashed line in (a) gives the "dispersion" $s=\zeta_0^{2.5}$ between the peak position ζ_0 and the energy s in the constant-energy scans. The arrow in (b) marks the point $\eta=1$ separating the critical ($\eta<1$) from the hydrodynamical ($\eta>1$) region.

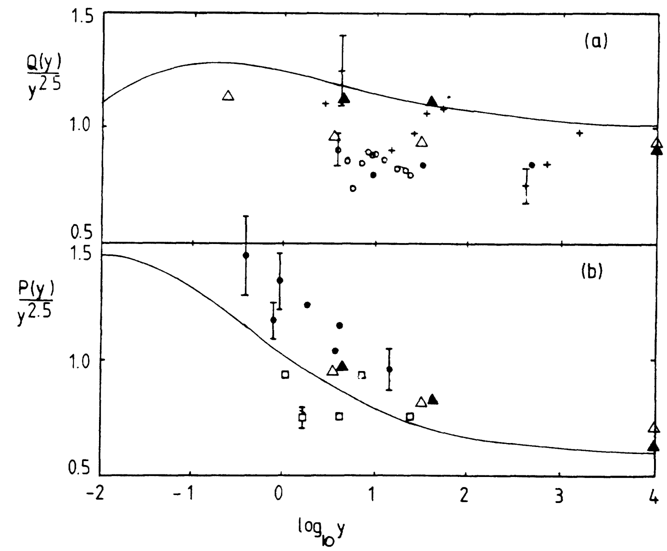

FIG. 6. Cuts at constant-energy transfer in Fig. 5 lead to the reduced peak position q_0 and width Δq. Shown in (a) are the universal scaling function Q: $q_0(\omega/\sigma)^{-0.4}\equiv Q(y)y^{-0.4}$, $y=\omega/\sigma\kappa^{2.5}$, and in (b) the universal scaling function P: $\Delta q(\omega/\sigma)^{-0.4}\equiv P(y)y^{-0.4}$. Large y means $T\rightarrow T_c$. The experimental data in (a) are taken for Ni from Refs. 25 (solid circles) and 26 and 27 (open circles), for Pd_2MnSn from Ref. 28 (crosses), and for EuS from Ref. 29 (open and solid triangles). In (b) they are taken for Ni from Ref. 30 (solid circles), for Fe from Ref. 31 (squares), and for EuS from Ref. 29 (open and solid triangles).

tensity lines [Figs. 5(a) and 5(b)] for $\kappa=0$ at T_c and $\kappa=0.2$. These lines are qualitatively the same as in RG theory (see Fig. 2 of Ref. 11). Ultimately all intensity lines meet at the origin $s=0,\zeta=0$. At nonzero κ the shape of the contour lines crossover [see arrow in Fig. 5(b)] from the hydrodynamical form (steep increase with ζ) to the critical form (flat increase with ζ). This leads to the structure seen at $\eta=1$. The maxima of the constant ω cuts lead to the "dispersion" $q_0=q_0(\omega,\kappa)$ (displayed as dashed lines in Fig. 5) and the width $\Delta q=\Delta q(\omega,\kappa)$. Both q_0 and Δq are homogeneous functions,[10] and the whole set of curves for different ω and κ should collapse to the universal scaling functions $Q(y)$ and $P(y)$, respectively. These are plotted in Fig. 6, and compared with available experimental data. The slight differences at T_c compared to the result of RG theory[6,12] are mainly due to the different shapes at T_c and can be considered as uncertainties of the respective theoretical expressions.

IV. CONCLUSION

We have presented within the mode-coupling theory the asymptotic spin-relaxation function applicable in that region of wave vector and temperature where the short-range interaction is dominant. There our results can be checked by neutron scattering, either in constant q or ω scans or with spin-echo methods.[32] By the last-mentioned method the time-dependent relaxation function can be measured directly. Suitable candidates may be the itinerant ferromagnets Ni or Fe. But even in the localized ferromagnets EuO and EuS at not too small momentum values our results may be applicable. Both neutron-scattering methods mentioned have been applied in EuO at T_c. At very small wave vectors ($q=0.024$ Å^{-1}) an exponential decay for $F(q,t)$ [a Lorentzian for $F(q,\omega)$] was found,[33] whereas at larger wave vectors ($q\sim 0.1$ Å^{-1}) the critical shape for $F(q,\omega)$ was found.[10] One may attribute this behavior to a crossover in the shape due to the dipolar interaction; this will be treated in a future publication. A crossover in the linewidth ω_c has been considered in Ref. 13 (under the assumption of a Lorentzian for the shape) and it was found that the dynamical crossover sets in at much lower q values than expected from the statics. The possibility of a dynamical crossover at a q vector different from the one in the static crossover was already stressed in Ref. 34. Very recently measurements that can be compared with the scaling functions Q and P have been made in EuS,[29] which however is an even more strongly dipolar ferromagnet than EuO. Whereas the qualitative behavior is as expected (see Fig. 6), it was claimed that deviations from the scaling property in the peak position were found (open and solid triangles in Fig. 6). These deviations also have been attributed to the dipolar interactions.

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