RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 43, NUMBER 10 1 APRIL 1991

Dynamics of longitudinal and transverse fluctuations above T_C in EuS

P. Böni
Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

D. Görlitz and J. Kötzler
Institut für Angewandte Physik, Universität Hamburg, 2000 Hamburg 36, Federal Republic of Germany

J. L. Martínez
Institut Laue Langevin, 38042 Grenoble CEDEX, France
(Received 10 December 1990)

Using inelastic scattering of polarized neutrons near a finite Bragg reflection, the dynamics of longitudinal $\delta \mathbf{S}(\mathbf{q}) \parallel \mathbf{q}$ and transverse $\delta \mathbf{S}(\mathbf{q}) \perp \mathbf{q}$ spin fluctuations have been measured. The relaxation rates Γ_α follow rather precisely the dipolar dynamic-scaling function calculated recently by mode-coupling theory based on the Lorentzian approximation. The predicted longitudinal line shape near T_C disagrees with our data.

I. DIPOLAR-INFLUENCED FLUCTUATIONS

In all Heisenberg ferromagnets, the dipole-dipole interaction lifts the degeneracy between magnetization modes propagating along and transverse to the spin direction $\mathbf{S}(\mathbf{q})$. Below the Curie temperature T_C, one of the well-known consequences of the long-range dipolar fields is to lower the spin-wave energies with increasing alignment of $\mathbf{q}$ towards the spontaneous magnetization $\langle \mathbf{S} \rangle$. Above T_C, the dipolar anisotropy was detected recently in the static susceptibility of the magnetization fluctuations in EuS and EuO by means of polarized neutron scattering.[1] Here the dipolar fields prevent the susceptibility of the long-wavelength, longitudinal fluctuations, $\delta \mathbf{S}_l(\mathbf{q}) \parallel \mathbf{q}$, from criticality while the transverse fluctuations drive the ferromagnetic transition, with the result:

$$\chi_\alpha(\mathbf{q},T) = \frac{q_d^2}{\kappa^2(T) + q^2 + \delta_{\alpha,l} q_d^2}, \quad \alpha = l, t \,, \qquad (1)$$

where $\mathbf{q}$ is the momentum transfer with respect to the nearest reciprocal lattice vector $\boldsymbol{\tau}$.

Equation (1) defines a region of dipolar anisotropic behavior around the critical point, $\kappa^2(T) + q^2 \leq q_d^2$, as illustrated in the inset of Fig. 1. Physically, the characteristic dipolar wave number q_d can be inferred, e.g., from the relation between the inverse correlation length κ and the homogeneous internal susceptibility, $\chi_t(0,T) = [q_d/\kappa(T)]^2$ or by equating the dipolar width of the spin-wave band to the spin-wave energy at q_d, $\mu M_S = D q_d^2$.

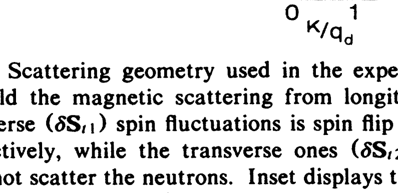

FIG. 1. Scattering geometry used in the experiment. In a vertical field the magnetic scattering from longitudinal ($\delta \mathbf{S}_l$) and transverse ($\delta \mathbf{S}_{t1}$) spin fluctuations is spin flip and non-spin flip, respectively, while the transverse ones ($\delta \mathbf{S}_{t2}$) parallel to $\mathbf{Q} \approx \boldsymbol{\tau}$ do not scatter the neutrons. Inset displays the regimes of isotropic (I) and dipolar (D) static critical behavior.

One of the most significant features related to this dipolar crossover has been observed in the relaxation rate of the homogeneous magnetization in many ferromagnets. As reviewed recently,[2] $\Gamma(q=0, T \geq T_C)$ changes from a critical speeding up in the isotropic (Heisenberg) region to the thermodynamic slowing down in the dipolar limit when passing the static crossover, $\chi_t = 1$, given by Eq. (1). This crossover for $q = 0$ and $\kappa > 0$ is in contrast to the absence of any dipolar effects in the relaxation rate of the transverse magnetization modes at T_C of, e.g., Fe (Ref. 3) and EuO,[4] where $\Gamma_t(q, T_C)$ maintained the isotropic behavior, at least down to the lowest accessible wave numbers, $q \leq q_d/5$. This puzzling situation was clarified by Frey and Schwabl,[5] who numerically evaluated the mode-coupling (MC) equations for the Heisenberg ferromagnet with dipolar interaction. Using their result, the measured crossover of the homogeneous relaxation from isotropic to dipolar behavior, e.g., on EuS, could quantitatively be explained.[2] Moreover, Frey and Schwabl predicted the dynamic dipolar crossover of the transverse relaxation at T_C to be located at much smaller wave numbers, $q \lesssim q_d/10$, which have not yet been probed to date. This shift of the dynamic crossover from the "static" one is via the mode-mode coupling intimately related to the dynamics of the longitudinal fluctuations in the dipolar regime.

In the present Rapid Communication, we report measurements of the linewidth and the line shape of the longitudinal and transverse fluctuations at and above T_C. It is our goal to provide a check of the numerical MC approach which predicts relaxation rates and line shapes for the longitudinal fluctuations being distinctly different from the transverse ones. For this purpose, we have chosen the ferromagnet EuS ($T_C = 16.56$ K), in which dipolar effects are rather strong giving rise to a large dipolar wave number, $q_d = 0.22$ Å^{-1}.[1] On the other hand, since spin-orbit coupling and hence cubic anisotropy can be neglected, this material represents the model Heisenberg ferromagnet with dipolar interaction considered in the MC theory. In fact, this model is confirmed by the excellent agreement between experiment and MC results on the homogeneous relaxation of EuS.[1] Furthermore, previous constant momentum and constant energy scans of the transverse fluctuations in EuS revealed deviations from the isotropic critical dynamics, which tentatively were related to dipolar effects.[6]

II. EXPERIMENT

In a first step we explain the procedure for measuring the paramagnetic fluctuations in an isotropic ferromagnet. The magnetic cross section for neutrons is given by[7]

$$\frac{d\sigma}{d\Omega} \propto \langle \delta S_\perp(-\mathbf{Q}) \delta S_\perp(\mathbf{Q}) \rangle , \tag{2}$$

where $\delta S_\perp(\mathbf{Q})$ are the spin components transverse to the scattering vector $\mathbf{Q} = \boldsymbol{\tau} + \mathbf{q}$. Equation (2) tells us that only the transverse susceptibility χ_t can be determined in the forward direction, where $\boldsymbol{\tau} = 0$. In order to measure the longitudinal fluctuations δS_l one has to go to a finite Bragg peak $\boldsymbol{\tau}$ with $\mathbf{q}$ perpendicular to $\mathbf{Q} \simeq \boldsymbol{\tau}$. Unfortunately, there is still no discrimination between the transverse δS_t and the longitudinal fluctuations δS_l (see Fig. 1).

The separation of the longitudinal from the transverse susceptibility can be achieved by the use of polarized neutrons, namely, if the spin fluctuations are parallel to the polarization $\mathbf{P}$ of the neutrons, then the scattered neutrons maintain their spin eigenstate. In Fig. 1 we show one typical scattering geometry used in our experiment for measuring the paramagnetic fluctuations: A small vertical guide field $H \simeq 5$ G at the sample position directs $\mathbf{P} \perp \mathbf{Q}$ and $\mathbf{P} \perp \delta \mathbf{S}_l$. Therefore, the spin-flip cross section is directly proportional to the longitudinal susceptibility $\chi_l(\mathbf{q})$. The non-spin-flip cross section is, on the other hand, proportional to the transverse susceptibility. Obviously, the situation is reversed for a horizontal guide field $\mathbf{H} \parallel \mathbf{q}$. Then the non-spin-flip cross section is proportional to $\chi_l(\mathbf{q})$.

The neutron-scattering experiments were conducted on the triple-axis spectrometer IN12 located in the neutron guide hall at the Institut Laue Langevin. The incident neutrons were polarized by means of a bender which was mounted after the pyrolytic graphite [002] monochromator. In order to remove higher-order neutrons a Be filter was placed in between. The scattered neutrons were analyzed with respect to energy and polarization using a Heusler crystal set for the (111) reflection. The final neutron energy E_f was kept fixed at 3.13 meV. A spin flipper placed after the sample enabled us to measure the spin-flip and non-spin-flip cross sections separately. The overall flipping ratio of the instrument was of the order of 47. The effective collimations of 40′-50′-50′-50′ between reactor and detector yielded an overall energy resolution of typically 0.04 meV [half width at half maximum (HWHM)] for the present experiment, which is well suited for measuring longitudinal widths. It is, however, rather coarse for measuring the transverse fluctuations.

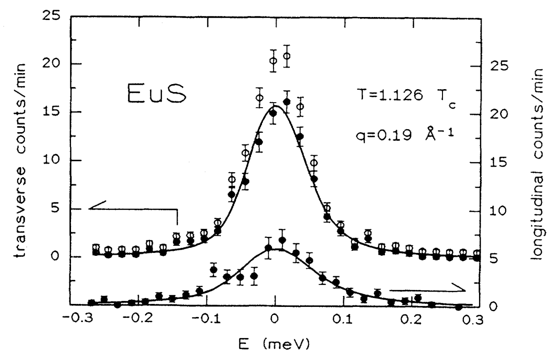

FIG. 2. Constant-$\mathbf{q}$ scans of the transverse and longitudinal scattering at 1.12 T_C, where the open circles label raw data. Solid circles show the same data after subtraction of the elastic incoherent intensity and a background of 0.5 cts/min. Solid lines represent fits to Lorentzians folded with the spectrometer resolution ($\Delta E = 40$ μeV).

The measurements were carried out on an enriched sample ^{153}EuS that was composed of roughly 40 fragments of the multiplatelet EuS sample assembled originally by Bohn *et al.*[8] The original sample was not useful because of the huge incoherent scattering from the glue and the coarse mosaic $\eta_s \gtrsim 3°$ that prevented any meaningful experiments near a Bragg peak, because (i) intermixing of χ_l and χ_t, and (ii) smearing out of $\mathbf{q}$. Therefore, we have removed as much glue as possible from the fragments and have realigned them on a new sample holder. The final mosaic of our sample was $\eta_s = 0.8°$ that translates into an effective q resolution (HWHM) $\Delta q = 0.015 \ll q_d \simeq 0.22$ Å^{-1}. The sample was mounted on the cold finger of a pumped He (orange) cryostat. The temperature was stable within 0.01 K during the duration of the experiment. The Curie temperature was determined by measuring the critical scattering near the Bragg peak and turned out to be $T_C = 16.33 \pm 0.03$ K.

Figure 2 shows that the spin-flip cross sections measured at $T = 1.12$ T_C in a vertical ($\mathbf{H} \perp \mathbf{q}$) and a horizontal field ($\mathbf{H} \parallel \mathbf{q}$) are rather different. First of all, χ_l is smaller than χ_t because the longitudinal fluctuations do not diverge near T_C. This observation is in qualitative agreement with previous work by Kötzler *et al.*[1] Second, the (longitudinal) energy width Γ_l is larger than Γ_t, as expected on the basis of MC theory.[5] Similar measurements have been performed at other temperatures as well. By measuring the non-spin-flip cross sections in a horizontal and vertical field we confirmed the internal consistency of our results.

III. RESULTS

In order to compare our data with the results of the MC theory,[5] which assumed the Lorentzian approximation, we parametrized the magnetic cross sections

$$S_\alpha(\mathbf{q},\omega) = 2k_B T \chi_\alpha(\mathbf{q},T) F_\alpha(\mathbf{q},\omega) \tag{3}$$

in terms of the Lorentzian spectral weight function

$$F_\alpha(\mathbf{q},\omega)=\frac{1}{\pi}\frac{\Gamma_\alpha}{(\hbar\omega)^2+\Gamma_\alpha^2}\ . \tag{3a}$$

$S_\alpha(\mathbf{q},\omega)$ was convoluted with the four-dimensional resolution function of the IN12, and for each $\mathbf{q}$ the normalization constant χ_α and linewidth Γ_α were determined by using a nonlinear least-squares-fitting routine (solid lines in Fig. 2). The parameterizations ensure that the dispersion of the magnetic scattering is properly taken into account.

The transverse widths [Fig. 3(a)] at 1.006 T_C increase with increasing q according to the dynamical-scaling prediction for an isotropic Heisenberg ferromagnet, $\Gamma_t = Aq^{2.5}$.[9] This dependence is consistent with previous unpolarized beam measurements[10] at T_C, where $A=2.1 \pm 0.3$ meV Å$^{2.5}$. The present Γ_t are smaller because the data have been taken slightly above T_C due to technical reasons. The q dependence of Γ_l demonstrates nicely that the longitudinal fluctuations are significantly suppressed for $q < q_d$.

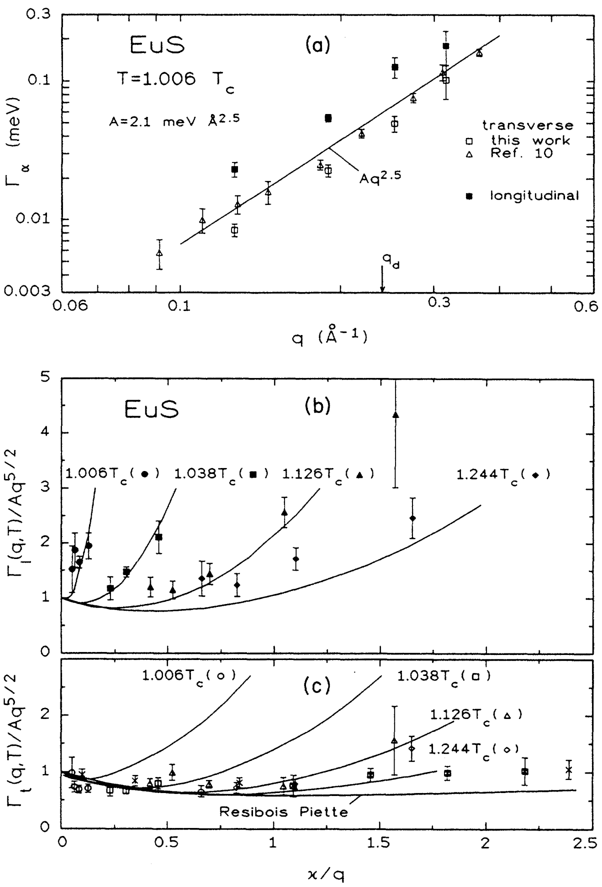

FIG. 3. (a) Linewidths close to T_C. (b) Comparison of the longitudinal linewidth of Lorentzian fits normalized to $Aq^{5/2}$ with $A=2.1$ meV Å$^{5/2}$ (Ref. 9) to the dipolar dynamic-scaling function predicted by mode coupling calculations (Ref. 5) for $q_d=0.245$ Å^{-1} for EuS. (c) Same as (b) for the transverse widths. Also indicated are previous data taken from Ref. 6 for $T/T_C=1.01$, 1.06, 1.21, 2.00 (x from left to right) and from Ref. 18 for $T=1.78$ T_C (⊠). The Résibois Piette dynamic-scaling function (Ref. 19) depicts the isotropic limit.

To provide a direct comparison with the mode-coupling results,[5] all fitted Lorentzian widths were normalized to the (hypothetical) linewidth of the isotropic system at T_C as given above, and depicted in Figs. 3(b) and 3(c) against the scaled inverse correlation length κ/q, using $\kappa=0.55$ Å^{-1} $(T/T_C-1)^{0.69}$.[11] Also included are data points Γ_t from previous unpolarized beam measurements. The solid lines indicate dynamic scaling functions of the MC theory[5] which depend only on the dipolar scaling variable $q_d/\kappa(T)$ with $q_d=0.245$ Å^{-1}. This value emerged from our data analysis and is in good agreement with previous measurements.[1] This representation does not involve any free parameter and obviously, within experimental error, theory and experiment are in excellent agreement. This is true, in particular, with regard to the longitudinal data, for which our experimental conditions were optimized, because Γ_l has not been measured in any isotropic system before.

As another important feature we discuss the spectral weight function $F_\alpha(q,\omega)$ in more detail. We recall that both the above analysis and the MC evaluation of the linewidths[5] were based on the supposition of Lorentzian shapes corresponding to exponential decays of the longitudinal and transverse modes. At T_C of EuO, using high-resolution neutron-spin echo, Mezei and co-workers[4,12] reported exponential behavior of the transverse modes for small wave numbers $q < q_d$. On the same material, a non-Lorentzian shape appeared for $q > q_d$,[12,13] which was consistent with a renormalization-group result for the Heisenberg ferromagnet without dipolar interaction.[14]

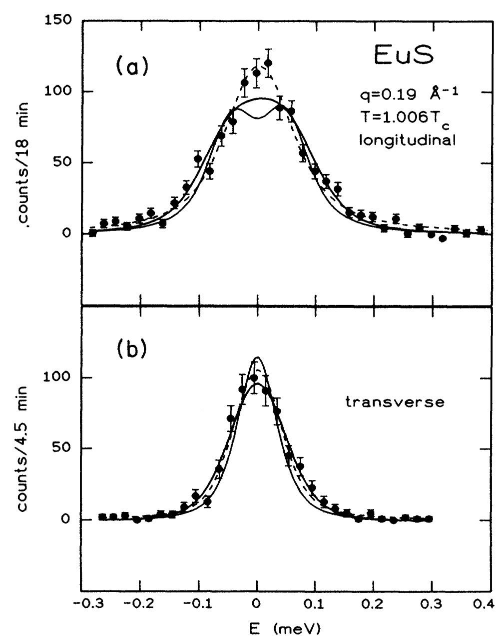

FIG. 4. Constant-$\mathbf{q}$ scans probing paramagnetic fluctuations with $q=0.19$ Å^{-1} very close to T_C. Data were corrected as in Fig. 2 and fitted alternatively to (a) longitudinal and (b) transverse spectral weight functions of MC theory (thick lines) and to Lorentzians (dashed lines). Thin lines depict the unconvoluted MC data.

More recently, the line-shape problem has been attacked also by the MC approach including the dipolar force.[15] At T_C the longitudinal shape is predicted to become more Gaussian at short times, followed by damped oscillatory behavior. Away from T_C the calculated longitudinal and transverse relaxation functions look rather similar, exhibiting a Gaussian, followed by an exponential decay at long times. This latter scenario is compatible with our data. For $T \gtrsim 1.038\ T_C$ all our longitudinal data can equally well be fitted by either a Lorentzian or Gaussian spectral weight function, whereas the transverse data are slightly better approximated by a Lorentzian, in particular at higher temperature, as in EuO.[16] We emphasize that the linewidth is only affected by the line shape to a minor extent because the latter is only an integral characterization of the profile.

In order to test this challenging MC prediction very close to T_C, we have measured the longitudinal spectral weight function for $q = 0.19$ Å^{-1} with improved statistics. In Fig. 4 we compare our data at $1.006\ T_C$ with corresponding shape functions based on MC for either the longitudinal or the transverse fluctuations. The transverse data [Fig. 4(b)] agree equally well with the predicted transverse line shape and the Lorentzian profile. In contrast, Fig. 4(a) shows that the longitudinal data are incompatible with the predicted longitudinal line shape, which is too square shaped near $E = 0$. Additional χ^2 analysis reveal that neither a pure Lorentzian nor a pure Gaussian are appropriate parametrizations of the data. This observation indicates that the line shapes for the longitudinal and the transverse fluctuations are different very close to T_C, however, less different than predicted by MC theory. In this context it might be interesting to note that the transverse shape function of MC theory describes the longitudinal data at $1.006\ T_C$ and $q = 0.19$ Å^{-1} best, although there is no theoretical justification for performing such a comparison.

In conclusion, the linewidths of the longitudinal spectral weight functions of EuS are in good agreement with the MC work[5] for the Heisenberg ferromagnet with dipolar interaction. This comparison is based on the Lorentzian assumption for both the evaluation of the measured neutron cross section and the numerical solution of the MC equations. Our line-shape analysis, very close T_C indicates that the calculated longitudinal spectral weight function does not agree with our data. For definite statements concerning the line shape, we propose to solve the problem by an experiment designed for energy resolutions $\Delta E \ll \Gamma_\alpha$ at T_C.

Perhaps the present results can also motivate future experimental and theoretical work on the critical behavior below T_C of dipolar Heisenberg ferromagnets, being considered as a "giant" step.[17] There, the presence of the order parameter $\langle \mathbf{S} \rangle$, requires separate investigations of fluctuations, perpendicular (spin waves) and parallel to $\langle \mathbf{S} \rangle$, both of which are split by the dipolar interaction into longitudinal and transverse modes.

ACKNOWLEDGMENTS

We thank H. G. Bohn for the loan of the isotopically enriched ^{153}EuS single crystals and E. Frey and F. Schwabl for supplying us with numerical results for Fig. 4. We are indebted to M. Koch and R. Thut of the Laboratory for Neutron Scattering, Eidgenössische Technische Hochschule Zürich at the Paul Scherrer Institute for the skillful construction of the sample holder.

[1] J. Kötzler, F. Mezei, D. Görlitz, and B. Farago, Europhys. Lett. **1**, 675 (1986).
[2] J. Kötzler, Phys. Rev. B **38**, 12027 (1988).
[3] F. Mezei, Phys. Rev. Lett. **49**, 1096 (1982).
[4] F. Mezei, Physica B **136**, 417 (1986).
[5] E. Frey and F. Schwabl, Z. Phys. B **71**, 355 (1988).
[6] P. Böni, G. Shirane, H. G. Bohn, and W. Zinn, J. Appl. Phys. **63**, 3089 (1988).
[7] W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Clarendon, Oxford, 1971), p. 111.
[8] H. G. Bohn, W. Zinn, B. Dorner, and A. Kollmar, Phys. Rev. B **22**, 5447 (1980).
[9] B. I. Halperin and P. C. Hohenberg, Phys. Rev. **177**, 952 (1969).
[10] P. Böni, G. Shirane, H. G. Bohn, and W. Zinn, J. Appl. Phys. **61**, 3397 (1987).
[11] L. Passell, O. W. Dietrich, and J. Als-Nielsen, Phys. Rev. B **14**, 4808 (1976).
[12] F. Mezei, B. Farago, S. M. Hayden, and W. G. Stirling, Physica B **156 & 157**, 226 (1989).
[13] P. Böni, M. E. Chen, and G. Shirane, Phys. Rev. B **35**, 8449 (1987).
[14] R. Folk and H. Iro, Phys. Rev. B **32**, 1880 (1985); **34**, 6571 (1986).
[15] E. Frey, F. Schwabl, and S. Thoma, Phys. Rev. B **40**, 7199 (1989).
[16] P. Böni and G. Shirane, Phys. Rev. B **33**, 3012 (1986).
[17] B. Heinrich and A. S. Arrott, J. Appl. Phys. **57**, 3709 (1985).
[18] L. Rebelsky and P. Böni (unpublished).
[19] P. Résibois and C. Piette, Phys. Rev. Lett. **24**, 514 (1970).