PHYSICAL REVIEW B VOLUME 34, NUMBER 9 1 NOVEMBER 1986

Temperature dependence of paramagnetic neutron scattering from Heisenberg ferromagnets above T_c

R. Folk and H. Iro
Institut für Theoretische Physik, Universität Linz, A-4040 Linz-Auhof, Austria
(Received 14 May 1986)

The temperature dependence of the constant-energy scans for neutron scattering by isotropic ferromagnets is studied. The weak temperature dependence of the maxima of these scans is explained, and the temperature dependence of the width is predicted. In the case of a uniaxial ferromagnet the effects of dynamic Fisher-Langer corrections are calculated. In the constant-wave-vector scans a slight enhancement of the scattering near $\omega=0$ is found.

Recently, considerable progress has been made in the experimental study of the paramagnetic scattering of neutrons near T_c by isotropic ferromagnets. In particular, constant-energy measurements have turned out to be an important tool to gain more information about the dynamic scattering function.

Whereas constant-wave-vector measurements (at least for sufficiently small q), show a peak at zero energy transfer, in constant-energy scans a peak at finite wave vector q_0 appears. It has now become clear that this peak, which has been ascribed to spin-wave-like excitations in the paramagnetic phase, is explained naturally by the features of the correlation function.[1-3] The occurrence of the peak is a consequence of the conservation of the order parameter. Moreover, the position of the peak q_0 and its width Δq depend sensitively on the shape of the scattering function.[4] At T_c the shape is expected to be non-Lorentzian and agreement of the results of renormalization-group (RG) theory with experiments was recently demonstrated.[3] However, in the case of constant-wave-vector measurements in Ni at $q \gtrsim 0.3$ Å^{-1} controversial experimental results exist. One group reported a peak at finite energy transfer,[5] taken as evidence of spin-wave-like excitations above T_c, whereas no such evidence was found by Shirane and co-workers,[2,6] as one expects from the dynamical critical theory.

Above T_c the structure factor is strongly temperature dependent; thus by simple arguments (see, e.g., Ref. 7) one would expect a temperature dependence of the same kind in the peak position q_0 and the full width at half maximum Δq. However, various measurements at different temperatures in Fe,[8] Ni,[6,9,10] and Pd_2MnSn (Ref. 11) show that the peak position is nearly temperature independent. For the width, a flat[8] or monotonically increasing[10] temperature dependence has been reported. It is the aim of this Rapid Communication to solve this puzzle by applying the results of RG theory to this problem. It turns out that the weak temperature dependence in q_0 originates from the compensation of two effects contributing to the temperature dependence of q_0. A change in the constant-q scans may be caused by the variation of the width *and the change of the shape.* In particular, in addition to the crossover in the width, there is a crossover in shape, from the non-Lorentzian form at T_c to a Lorentzian, with increasing temperature.[12] In the temperature dependence of q_0 the effects of change in width and in shape compensate, whereas for the width Δq of the constant-energy scans these two effects go in the same direction. Therefore, we predict an increase of the width Δq further away from T_c.

We also consider the case of uniaxial ferromagnets, whose dynamics is described by a pure diffusive model without mode-coupling terms (model B of Ref. 13). The peak position q_0 and the width Δq are weakly decreasing functions of temperature. Within this more tractable model we studied the interesting question of the contributions of the next-to-leading terms to the dynamic correlation function,[14] known in statics as the Fisher-Langer corrections. We find a small increase in the constant-energy scans at small ω, which may be most conveniently seen in the intensity taken as function of temperature, quite analogous to critical statics.[15,16] Those corrections are not noticeable in q_0 and Δq.

The neutron scattering intensity is given, up to a thermal factor, by the dynamic magnetization correlation function $C(q,\omega,\xi)$,

$$C(q,\omega,\xi)\delta^{\alpha\beta}=\int dt\int d^3x\, e^{-i(\mathbf{q}\cdot\mathbf{x}-\omega t)} \times\langle S^\alpha(\mathbf{x},t)S^\beta(0,0)\rangle \ , \qquad (1)$$

where ξ is the correlation length [$\xi=\xi_0(T/T_c-1)^{-\nu}$ with $\nu\simeq 0.67$]. Renormalization-group theory shows[13] that $C(q,\omega,\xi)$ at criticality (i.e., for sufficient small arguments) is a homogeneous function and can be written in the following form:

$$C(q,\omega,\xi)=\chi(q,\xi)\frac{1}{\omega_c(q,\xi)}\phi\left(\frac{\omega}{\omega_c(q,\xi)},q\xi\right) \ . \qquad (2a)$$

Here one has separated out the static susceptibility $\chi(q,\xi)=q^{-2+\eta}X(q\xi)$ and introduced a characteristic frequency $\omega_c(q,\xi)$ which is also a homogeneous function

$$\omega_c(q,\xi)=Aq^z\Omega(q\xi) \ , \qquad (2b)$$

with the dynamic critical exponent z. We take ω_c to be the half-width of the correlation function. Because of our definitions the shape function ϕ has to fulfill the following relations:

$$\int_{-\infty}^{+\infty} ds\, \phi(s,x) = 2\pi \ , \tag{3a}$$

$$\phi(1,x) = \tfrac{1}{2}\phi(0,x) \ . \tag{3b}$$

First we discuss two limiting regions for (2), namely, the critical region $q\xi \gg 1$ and the hydrodynamic region $q\xi \ll 1$. From dynamic scaling theory one can make general statements about the behavior of the dynamic quantities Ω and ϕ. To obtain explicit expressions for Ω and ϕ one has to resort to perturbational calculations.

The half-width crosses over from $\omega_c \sim q^z$ for $q\xi \gg 1$ to $\omega_c \sim q^2$ for $q\xi \ll 1$, which implies for $\Omega(x)$ the limiting behavior $\Omega(x) = 1$ as $x \to \infty$ and $\Omega(x) \sim x^{-(z-2)}$ for $x \to 0$. For isotropic ferromagnets ($z = \frac{5}{2}$, taking $\eta = 0$) we use the following simple interpolation between these two limits:

$$\Omega(x) = 0.43 \frac{1 + x^{-2}}{x^{-3/2} + 0.43} \ . \tag{4}$$

This choice is (i) in numerical agreement with the mode-coupling result of Résibois and Piette,[17] (ii) compatible with the RG results of Ref. 18, and (iii) agrees with measurements.[19] Recently, the next correction to 1 in the $x \to \infty$ limit has been discussed;[20] one could also include such corrections in (4). However, the only property needed in our further considerations is that $x\,\Omega'(x) \to 0$ for $x \to \infty$. For uniaxial ferromagnets ($z = 4$, $\eta = 0$) the van Hove theory yields

$$\Omega(x) = 1 + \frac{1}{x^2} \ . \tag{5}$$

Concerning the variation of the shape of $\phi(s,x)$ with x one has in the limit of $x \to \infty$,

$$\phi(s,\infty) = 2\,\mathrm{Re} \frac{1}{is + \alpha[1 + i(\beta s/\alpha)]^{(z-4)/z}} \ , \tag{6}$$

with $\alpha = 0.78$ and $\beta = 0.46$.[3,21] In the opposite limit, $x \to 0$, $\phi(s,x)$ is a Lorentzian,

$$\phi(s,0) = 2 \frac{1}{1 + s^2} \ . \tag{7}$$

The explicit dependence of ϕ on x, describing the crossover from Eqs. (6) to (7), is not known. Therefore we shall consider below both forms in the whole region of x values. In the case of uniaxial ferromagnets there is no crossover in shape, if we neglect higher-order effects in the static fourth-order coupling, and (7) applies in the whole region of x values.

For the isotropic as well as the uniaxial ferromagnet we take the Ornstein-Zernike form for $\chi(q,\xi)$.

We are now in a position to calculate the maximum q_0 in the constant-energy scans. Because of the scaling properties one easily sees that q_0 obeys the scaling law

$$q_0 = \xi^{-1} Q(\xi^z \omega / A) \ , \tag{8}$$

where the scaling function Q depends on the explicit expressions for $\Omega(x)$ and $\phi(s,x)$. Since q_0 is finite at T_c we find the limiting behavior in the critical region $Q(y) \sim y^{1/z}$ and therefore

$$q_0 = b \left[\frac{\omega}{A} \right]^{1/z} , \tag{9}$$

with $b = 0.90$ and $b = 0.87$ for isotropic and uniaxial ferromagnets, respectively. In the hydrodynamic region we get $Q \sim y^{1/2}$ and

$$q_0 = c \left[\frac{\omega}{A} \right]^{1/2} \xi^{(z/2)-1} \ , \tag{10}$$

with $c = 1.53$ and $c = 1.0$ for the isotropic and uniaxial case, respectively. Though the shape of ϕ is the same in both cases the differing values for c result from the different amplitude in the limiting behavior of Eqs. (4) and (5) for $x \to 0$.

Similar arguments hold for the half-width of the constant-energy scans and one again finds the scaling behavior

$$\Delta q = \xi^{-1} P(\xi^z \omega / A) \ . \tag{11}$$

The scaling function P has the same power-law behavior as Q in the critical and hydrodynamic limit. Analogous to Eq. (9) we have

$$\Delta q = b' \left[\frac{\omega}{A} \right]^{1/z} , \tag{12}$$

with $b' = 0.68$ and $b' = 0.65$, respectively, and analogous to Eq. (10)

$$\Delta q = c' \left[\frac{\omega}{A} \right]^{1/2} \xi^{(z/2)-1} \ , \tag{13}$$

with $c' = 2.2$ and $c' = 1.4$, respectively. In order to evaluate the scaling functions Q and P between these two limits for the isotropic ferromagnet one needs the shape function ϕ. To cope with the fact that the crossover in the shape function ϕ is not known we calculate Q and P for both forms (6) and (7) in the whole region of x. This leads to the two curves for the maximum and for the width, shown in Figs. 1(a) and 1(b), respectively. Each of the curves is asymptotic to the true crossover functions Q and P in the appropriate limit. It is natural to expect that between these limits, Q and P should lie between the curves shown. This leads in the case of q_0 [Fig. 1(a)] to a weak temperature dependence. For Δq, however, the crossover from the critical to the Lorentzian shape enhances the temperature dependence [Fig. 1(b)].

To compare these predictions we have plotted experimental data for the peak position of Ni (Refs. 6, 9, and 22) and Pd_2MnSn (Ref. 11) into Fig. 1(a). They lie well within the crossover region; however, the error bars and the scatter of the data are too large to make more definite statements about the crossover function. An analogous comparison was made for the width in Fe (Ref. 8) and in Ni.[10] To judge this comparison one should keep in mind that the value of the width depends essentially on the background subtracted. We also like to remark that the values of the relative temperature distance from T_c enter sensitively via y into these plots. Some scatter of the data

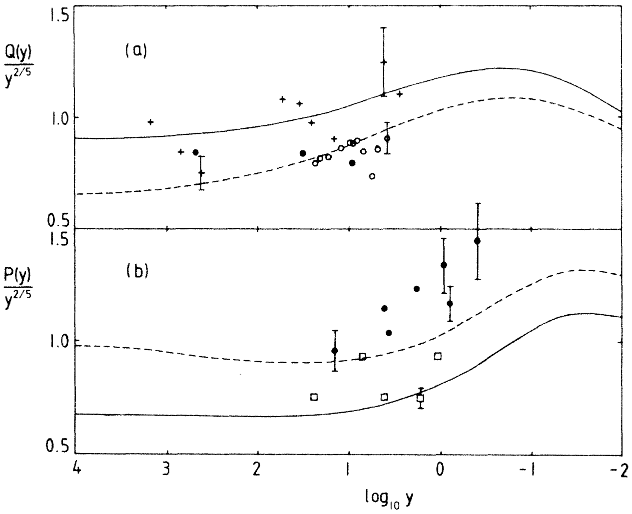

FIG. 1. For the constant-energy scans of isotropic ferromagnets we plot as functions of the scaling variable $y=(\omega/A)\xi^{5/2}$: (a) The reduced peak position $q_0(\omega/A)^{-2/5}=Q(y)/y^{2/5}$ for both shape functions ϕ. The solid line corresponds to the critical shape, Eq. (6), and the dashed line to the hydrodynamic shape, Eq. (7). (b) The reduced width $\Delta q(\omega/A)^{-2/5}=P(y)/y^{2/5}$, again for both shapes. The experimental data in (a) are taken for Ni from Ref. 9 (full circles) and from Refs. 6 and 22 (open circles), for Pd_2MnSn from Ref. 11 (crosses). In (b) they are taken from Ref. 10 for Ni (full circles) and from Ref. 8 for Fe (squares). The error bars result only from the experimental uncertainties for q_0 and Δq, given in the references referred to. In both figures there should be a crossover in the data from the solid line for large y to the dashed line for small y. Data belonging to the asymptotic region should lie on one universal curve.

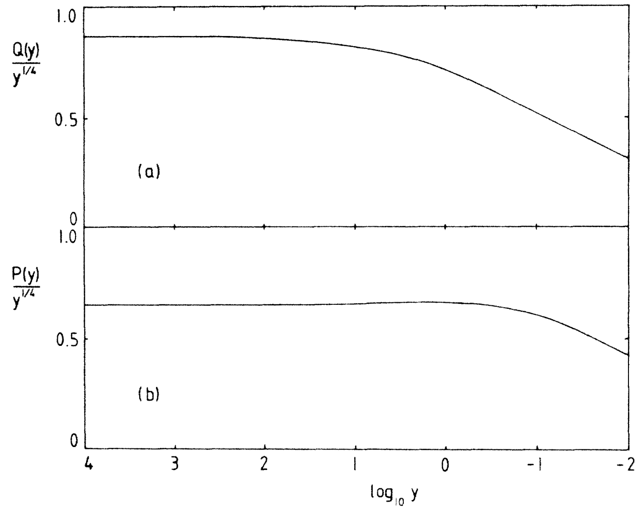

FIG. 2. For the constant-energy scans of uniaxial ferromagnets, we plot as functions of the scaling variable $y=(\omega/A)\xi^4$: (a) The reduced peak position $q_0(\omega/A)^{-1/4}=Q(y)/y^{1/4}$. (b) The reduced width $\Delta q(\omega/A)^{-1/4}=P(y)/y^{1/4}$.

"at T_c" in Fig. 6 of Ref. 22 may be attributed to a slight temperature dependence of q_0 and should be distinguished from background effects at larger q and ω values. Such background effects may also be the reason for deviations from the universal crossover function, that is, data whose ω, q, or ξ values are outside the asymptotic region are not expected to lie on the universal curve.

For the uniaxial ferromagnet the crossover functions Q and P are shown in Figs. 2(a) and 2(b). For that case we study also the contributions from the next-to-leading terms to the asymptotic behavior. Those can be included in the correlation function by a method introduced for the static case by Nelson,[23] which has been extended to relaxational and diffusive dynamics in Ref. 14. Here we discuss the case of purely diffusive dynamics only. We find[24] $[y=(\omega/A)\xi^4]$,

$$C(q,\omega,\xi)=\frac{2\xi^6}{x^2}\left\{\frac{y^2}{x^4}+[x^2+(1+p)F^{1/\nu-2}-pF^{(1-\alpha)/\nu-2}]^2\right\}^{-1}, \quad (14)$$

with $p=(2\nu-1)/\alpha$, where $\nu=0.63$, the specific-heat exponent $\alpha=0.11$ (Ref. 25), and

$$F(x,y)=\left\{\frac{y^2}{x^4}+(1+x^2)^2\right\}^{-1/4}. \quad (15)$$

The calculation of the maximum q_0 and the width Δq with Eq. (14) leads to the result already shown in Fig. 2 for the Lorentzian case. No effects of these Fisher-Langer corrections can be found; however, for the intensity in the constant-q scans near $\omega=0$ we find a slight enhancement of about 1%. Plotting the normalized scattering intensity as a function of temperature shown in Fig. 3, we obtain a small peak, similar to that found in the statics.[15,16] A general feature of these curves is a decreasing peak position

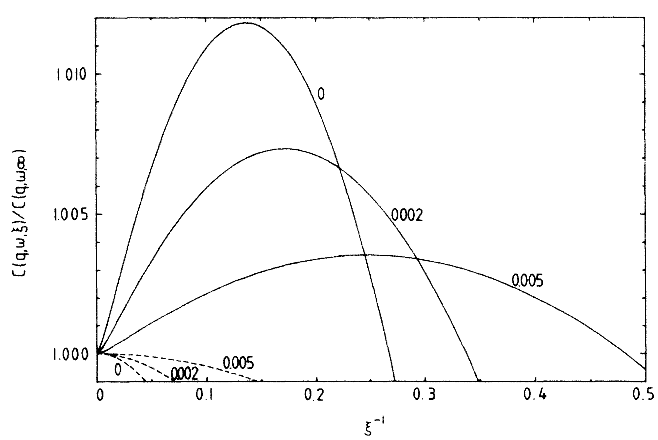

FIG. 3. The normalized structure function $C(q,\omega,\xi)/C(q,\omega,\infty)$ for uniaxial ferromagnets including the dynamic Fisher-Langer corrections (solid lines) plotted for $q=0.2$ and $\omega=0$, 0.002, 0.005 vs ξ^{-1} [see Eq. (14)]. The dashed lines show the structure function without Fisher-Langer corrections.

and width for decreasing q and ω. For smaller q values the height of the peak decreases faster with increasing ω.

Finally, we believe that our results should encourage further efforts on the experimental and theoretical determination of the crossover of the scaling function for the peak position and for the width of the constant-energy scans. For that purpose, improved experimental data are needed. In particular, more temperature-dependent measurements would be advantageous. On the theoretical side, the crossover in the shape function should be calculated.

This work was supported by the Fonds zur Förderung wissenschaftlicher Forschung (Wien, Austria).

[1]G. Shirane, J. Magn. Magn. Mater. **45**, 33 (1984).

[2]J. L. Martinez, P. Böni, and G. Shirane, Phys. Rev. B **32**, 7037 (1985).

[3]R. Folk and H. Iro, Phys. Rev. B **32**, 1880 (1985).

[4]J. P. Wicksted, P. Böni, and G. Shirane, Phys. Rev. B **30**, 3655 (1984).

[5]H. A. Mook and J. W. Lynn, J. Appl. Phys. **57**, 3006 (1985); J. W. Lynn and H. A. Mook, J. Magn. Magn. Mater. **54–57**, 1169 (1986).

[6]O. Steinsvoll, C. F. Majkrzak, G. Shirane, and J. P. Wicksted, Phys. Rev. B **30**, 2377 (1984).

[7]J. W. Lynn, Phys. Rev. Lett. **52**, 775 (1984).

[8]J. W. Lynn, Phys. Rev. B **11**, 2624 (1975); **28**, 6550 (1983).

[9]H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. **30**, 556 (1973).

[10]J. W. Lynn and H. A. Mook, Phys. Rev. B **23**, 198 (1981).

[11]G. Shirane, Y. J. Uemura, J. P. Wicksted, Y. Endoh, and Y. Ishikawa, Phys. Rev. B **31**, 1227 (1985).

[12]J. Hubbard, J. Phys. C **4**, 53 (1971).

[13]P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).

[14]R. Dengler, H. Iro, and F. Schwabl, Phys. Lett. **111A**, 121 (1985); **115A**, 459(E) (1986).

[15]M. E. Fisher and A. Aharony, Phys. Rev. B **10**, 2818 (1974).

[16]J. Als-Nielsen, Phys. Rev. **185**, 664 (1969).

[17]P. Résibois and C. Piette, Phys. Rev. Lett. **24**, 514 (1970).

[18]S. Ma and G. F. Mazenko, Phys. Rev. B **11**, 4077 (1975).

[19]G. Parette and R. Kahn, J. Phys. (Paris) **32**, 31 (1971).

[20]R. A. Ferrell and J. K. Bhattacharjee, J. Stat. Phys. **41**, 899 (1985).

[21]J. K. Bhattacharjee and R. A. Ferrell, Phys. Rev. B **24**, 6480 (1981).

[22]P. Böni and G. Shirane, J. Appl. Phys. **57**, 3012 (1985).

[23]D. R. Nelson, Phys. Rev. B **14**, 1123 (1976).

[24]The correlation function, Eq. (14), is obtained by the method described in Ref. 14, but with a different compromise condition:

$$\left(\frac{\omega}{Aq^2}e^{2l^*}\right)^2+[(qe^{l^*})^2+(\xi^{-1}e^{l^*})^2]^2=1$$

leading to the correct static limit.

[25]J. C. Le Guillou and J. Zinn-Justin, Phys. Rev. Lett. **39**, 95 (1977).