Breakdown of Dynamic Scaling Analysis in Isotropic Ferromagnets

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(Received 5 December 1983)

The predictions of dynamic scaling theory are shown to be in disagreement with the observed wave-vector and temperature dependence of the inelastic neutron scattering from iron and nickel above T_c, outside the region of small wave vectors. This breakdown becomes particularly severe when the characteristic energy of the fluctuations becomes comparable to kT, which occurs at considerably smaller reduced wave vectors for metallic ferromagnets such as Fe, Co, and Ni than for Heisenberg insulators such as EuO.

PACS numbers: 75.40.Bw, 61.12.Fy, 64.60.Ht, 75.50.-y

Early inelastic neutron-scattering experiments on paramagnetic iron[1] and nickel[2] at small wave vectors q were found to be in general agreement with dynamic scaling theory,[3] with subsequent theoretical developments[4] resolving[5] most remaining difficulties. At larger wave vectors the scattering above the Curie temperature T_C was found[6,7] to evolve from the small-q regime in a continuous fashion into a very energy-dispersive "ridge" of intensity, whose width in q was considerably narrower than expected on the basis of hydrodynamic (spin-diffusion) theory. Shirane and co-workers[8-10] have recently confirmed this general experimental picture,[11] but came to the conclusion[9,12] that spin diffusion provides a "full explanation" for all the neutron data.[6-13] In this note we show that this conclusion is incorrect, and that in fact the diffusive form of dynamic scaling theory fails when the characteristic energy E_c of the fluctuations becomes comparable with kT, as expected.[3] We use this physical criterion to show that the region in the Brillouin zone where diffusion dominates the spin dynamics is much smaller for the band ferromagnets Fe, Co,[14] and Ni than for Heisenberg insulators such as EuO.[15]

We start with the general form for the scattering function $S(q,E)$ which governs the wave-vector and energy dependence of the neutron scattering:[16]

$$S(q,E)=\chi(q)F(q,E)\left[\frac{E/kT}{1-\exp(-E/kT)}\right], \quad (1)$$

where the system is assumed isotropic above T_c, and we drop overall multiplicative constants for simplicity. At small (q,E) the wave-vector-dependent susceptibility $\chi(q)$ is usually taken to be[1,2,5,12,14,15] of the Ornstein-Zernike form while the spectral weight function $F(q,E)$ is taken to be a simple Lorentzian in energy.[17] Thus

$$S(q,E)=\frac{1}{\kappa^2+q^2}\,\frac{1}{\Gamma(q)\{1+[E/\Gamma(q)]^2\}}\times\left[\frac{E/kT}{1-\exp(-E/kT)}\right]. \quad (2)$$

If $\Gamma(q)\ll kT$ then the thermal factor (in brackets) is close to unity over the energy range of interest, and its variation with energy can be neglected. However, this is not an appropriate assumption in the present case.[18] Dynamic scaling theory[3] introduces[19] a characteristic energy E_c [$E_c=\Gamma(q)$ in this case] by which the dynamics of the system are scaled, in analogy with the role the correlation range ξ ($\xi=1/\kappa$) plays in providing a characteristic length scale for the statics. For an isotropic ferromagnet the characteristic energy should scale as[3]

$$\Gamma(q)=Aq^{5/2}f(\kappa/q), \quad (3)$$

where A is a materials-dependent constant and $f(\kappa/q)$ is the scaling function. Both A and $f(x)$ must be obtained from microscopic theory, or measured experimentally. However, the limiting forms for $f(x)$ are known: For $x=\kappa/q\to 0$, $f(x)\to 1$, while for $x\to\infty$, $f(x)\propto x^{1/2}$. Note in particular that at $T=T_c$, $\kappa=0$ and thus $f(x)\equiv 1$. Consequently if $\Gamma(q)$ is known at T_c then Eq. (3) scales the characteristic energy for all κ/q above T_c through the function $f(\kappa/q)$. Equations (2) and (3) have been reported to provide a good quantitative description of the spin dynamics at small (q,E) for Fe,[1,5] Ni,[2,5] Co,[14] and EuO.[15] The experimentally determined values of A are given in Table I.

Outside the small-wave-vector region direct measurements of the energy dependence of $F(q,E)$

TABLE I. Comparison of parameters for the isotropic ferromagnets.

	A (meV-Å$^{5/2}$)	T_c (meV)	a (Å)	Γ(Z.B.) (meV)	A/kT_c (Å$^{5/2}$)	q_c (Å)
Fe [a]	130	89.8	2.88	914	1.45	0.25
Co [b]	300	119.6	3.61	1199	2.51	0.25
Ni [c]	350	54.4	3.54	1469	6.43	0.17
EuO [d]	3.3	5.9	5.13	5.5	0.56	0.67

[a]Ref. 1.
[b]Ref. 14.
[c]Ref. 2.
[d]Ref. 15.

are difficult since the scattering in Fe and Ni is highly dispersive (i.e., the absolute values of A are large). Thus experimentally it was advantageous[6,7] to make measurements at fixed energy transfer E as a function of q (so-called constant-E scans), whereby a peak position $q_0(E)$ and a width $q_w(E)$ were obtained directly from the data at each energy. These experimentally determined values then can be compared with theoretical predictions. For Eqs. (2) and (3) the calculations can be done analytically at $T = T_c$ since the scaling function is unity and thus $\Gamma(q) = Aq^{5/2}$. Taking the derivative of Eq. (2) with respect to q yields the theoretical relation

$$E = 3Aq_0^{5/2}. \tag{4}$$

Comparing with the measurements[6,7] at larger q then *uniquely* determines the required value of A. There are systematic discrepancies between Eq. (4) and the data,[6,7] but a best-fit representation yields values of $A \sim 142$ meV-Å$^{5/2}$ for Ni and $A \approx 62$ meV-Å$^{5/2}$ for Fe. These values are less than half the values obtained from the analysis at small q (Table I). Thus Eqs. (2) and (3) cannot explain the observed peak positions unless A is assumed to be strongly q dependent. Such an assumption contradicts the scaling hypotheses.

A full comparison of theory and experiment is shown in Fig. 1, where we have plotted the peak positions. It is important to note that for the theory both of these quantities are controlled by the single parameter A. The utility of this particular method of comparison is that the parameter A in fact drops out of the problem so that we obtain a *unique* theoretical prediction, which is a straight line with slope 1.573. For iron (solid circles) we see that there are discrepancies even at quite modest values of q and E, and the disagreement becomes larger with increasing q. Similar results are obtained for Ni (open circles); a complete analysis for nickel will be given elsewhere. At smaller q the data are expected to fall on the theoretical curve, but unfortunately there are no constant-E measurements in this regime. Clearly this is a region where further work would be desirable.

To calculate the temperature dependence of the scattering we need to know the temperature dependence of κ as well as the scaling function $f(\kappa/q)$ in Eq. (3). For this purpose we used the measured[1,2,5] temperature dependence of κ, along with the calculated scaling function of Rêsibois and Piette[4] which is in good agreement with the small-q data.[5] The detailed results clearly depend on the particular choices of param-

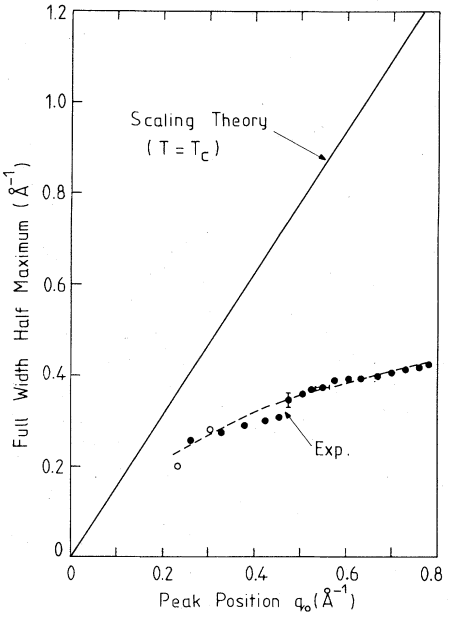

FIG. 1. Comparison of the predicted (solid curve) widths and positions in q with the experimentally determined values for iron (Ref. 7) (solid circles) and nickel (Ref. 6) (open circles). All adjustable parameters drop out of the theory, so that a unique and unambiguous comparison can be made.

eters and functions, but there are two theoretical results of general validity which disagree in a qualitative manner with the experimental observations under consideration. The first result is that the calculated peak positions $q_0(E)$ are strongly dependent on temperature; typically the values of $q_0(E)$ increase by $\sim 40\%$ in going from T_c to $1.4T_c$ for both nickel and iron. Experimentally *no* temperature dependence is observed in either system. The second result is that for increasing T above T_c the calculated widths $q_w(E)$ initially *decrease* as a result of the deep minimum in the scaling function $f(\kappa/q)$. This minimum was an essential theoretical property needed to understand the small-q data,[5] but was neglected in the recent work.[8-10,12,20] Experimentally for iron[7] the widths above T_c are not sensitive to temperature, while for nickel[6] they monotonically *increase* as the temperature passes through T_c.

Thus our overall conclusion is that the agreement between the theory as given by Eqs. (2) and (3) and the measurements at larger q is rather poor, with the discrepancies becoming systematically larger with increasing wave vector. This failure of the theory is expected[3] since the dynamic scaling assumptions should be valid only for macroscopic distances and times. In particular for Eq. (3) to be valid we require the characteristic energy Γ to be small compared to any other characteristic energy in the problem, such as kT. The dynamics are related to the spatial coordinates through relations such as Eqs. (3) and (4), so that if the parameters have been measured we can then utilize these relations to compare equivalent regions in q over which the scaling assumptions should be valid. For this purpose we define[21] q_c to be the characteristic wave vector at which the dispersion relation reaches kT:

$$q_c \approx (kT_c/3A)^{2/5}. \quad (5)$$

The values (in reduced units) are given in the table, where we see that the characteristic wave vectors are much smaller for the 3d magnets. Thus spin diffusion can be expected to describe the dynamics properly over a more restricted fraction of the Brillouin zone for the (high-T_c) metallic ferromagnets relative to the (low-T_c) Heisenberg insulators such as EuO, as found experimentally.[15] This difference in behavior is due to the larger values of A/kT_c for the 3d magnets, which ratio may be useful as a criterion for distinguishing "itinerant" from "localized" spin systems. Another way to view the same basic behavior is to compare the characteristic energies predicted by Eq. (3) at the zone boundaries (Z.B.). For the band ferromagnets we find (Table I) that Γ(Z.B.) is of the order of band energies (in electronvolts), while for EuO we find $\Gamma(\text{Z.B.}) \sim kT_c$. This result emphasizes the fact that the microscopic physics (contained in A and kT_c) underlying the magnetism in these two classes of systems is of course different. It also implies that the sharp "ridge" of scattering above T_c observed in iron and nickel is intimately related to the band properties of these itinerant magnets.

One important assumption in the above analysis is the Lorentzian form for the spectral weight function $F(q, E)$. Scaling theory does not specify F, but it does demand that the shape be the same for any particular value of κ/q. In particular at T_c, $F(q, E/E_c)$ must be the same function for all q, so that any observed change in shape would also indicate a breakdown of the scaling assumptions. Substantial deviations from the Lorentzian form have in fact already been observed near T_c for the 3d magnets[1,2,14] at quite small q, while for EuO[22] and EuS[23] the data reveal a qualitative change in the shape of $F(q, E)$ at large q. In the latter case the maximum in $F(q, E)$ shifts to finite energy halfway to the zone boundary, with relatively well defined spin waves persisting above T_c for $q \gtrsim q_c$. If we use Eq. (5) to "scale" this observed behavior to the 3d ferromagnets, we might then expect structure to develop in the spectral weight function at considerably smaller reduced wave vectors, as reported originally.[6,7] The details of the shape of $F(q, E)$ for iron and nickel, however, have not been satisfactorily resolved experimentally.[11]

Finally we comment on the shape of the spectral weight function in the (small-q) scaling region. At T_c, the theoretical prediction for Fig. 1 will *always* be a straight line passing through the origin, with only the slope of the line depending on the detailed shape of $F(q, E)$. This is a general result which follows from the fact that at T_c the correlation length drops out of the problem,[19] so that dimensionally the only way to construct a length is through the quantity energy/A. Thus scaling demands that Eqs. (3), (4), and (5) all have the same form at T_c. The q widths must also obey an equation of the same form, which then necessitates a linear relationship for the theory. Certainly a spectral function different from a Lorentzian, which gives a smaller slope, would be in better agreement with the data in

Fig. 1 (presumably at the expense of the small-q data). It is clear, however, that the data in this region of q disagree with the scaling predictions in a fundamental way.

I would like to thank R. Pynn and D. Givord for helpful discussions and assistance. I would also like to thank G. Shirane for communicating his results prior to publication. This work was supported in part by the National Science Foundation through Grant No. DMR 79-00908.

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[21]Where Eq. (2) "fails" is certainly a matter of choice (and accuracy of data). We need $\Gamma \ll kT$; Eq. (4) chooses $\Gamma = kT_c/3$ as a "boundary." Of course any particular choice for the cutoff will not affect the *inter*-comparison of the different systems.

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