Neutron-Scattering Observations of Extreme Critical Slowing Down in a $d=3$ Random-Field System

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Extreme critical slowing down of the order-parameter fluctuations in a $d=3$ random-field Ising-model system is observed in quasielastic-neutron-scattering studies of $Fe_{0.46}Zn_{0.54}F_2$. This is manifest in the logarithmic time dependence of $I(\mathbf{q})$ in the $30-10^4$-sec time domain at, and near, $T_c(H)$ in zero-field-cooled measurements. Contrary to a report by others, $T_{eq}(H) > T_c(H)$ and no evidence for a first-order phase transition is found in this virtually gradient-free crystal.

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The lower critical dimension of the random-field Ising model (RFIM) is generally believed to be $d_l=2$,[1,2] although differing points of view remain as to the nature of the phase transition for $d > d_l$; i.e., whether[3,4] or not[5-7] it is continuous, and if so, what are the critical exponents and amplitudes? Of most current interest are recent theoretical[8,9] and indirect experimental [ac susceptibility[10] $\chi(\omega)$ and Faraday rotation[11]] insights pointing to extraordinary critical slowing down near the phase transition at $d=3$. Now, for the first time, such effects have been observed directly on the *order-parameter* fluctuations through the time dependence of the scattering intensity and the field scaling of the dynamic rounding in quasielastic-neutron-scattering experiments at the antiferromagnetic Bragg point.

The neutron-scattering data were obtained at the Oak Ridge National Laboratory. The crystal of $Fe_{0.46}Zn_{0.54}F_2$ and the experimental configuration are identical to those used for the $H=0$ study of random-exchange critical scattering,[12] except for the superconducting solenoid. The measured concentration gradient, as determined by optical birefringence,[13] is so small that the spread in transition temperatures at $H=0$ is only $\delta T_N=0.016$ K, with $T_N=35.667$ K.

The quasielastic scattering $I(\mathbf{q})$ is proportional to the correlation function $S(\mathbf{q})$, with appropriate resolution corrections. Previous studies[3] of the $d=3$ RFIM slightly *above*, but not too close to $T_c(H)$, showed the sum of a Lorentzian (L) plus Lorentzian squared (LSQ) to be an adequate approximation to $S(\mathbf{q})$; i.e., $S(q)=A/(\kappa^2+q^2)+B/(\kappa^2+q^2)^2$, where $\kappa=\xi^{-1}$ is the inverse correlation length. Figure 1 shows five $I(q)$ scans all after zero-field cooling (ZFC) to $H=3.0$ T, for temperatures T above and below $T_c(H)$. The three scans at $T=32.34$ K and above show no Bragg peak and are thus above $T_c(H)$. They have rather large-intensity tails in the critical scattering at large $q > \kappa$, whose amplitude varies only slowly with T. As $T_c(H)$ is approached from above,

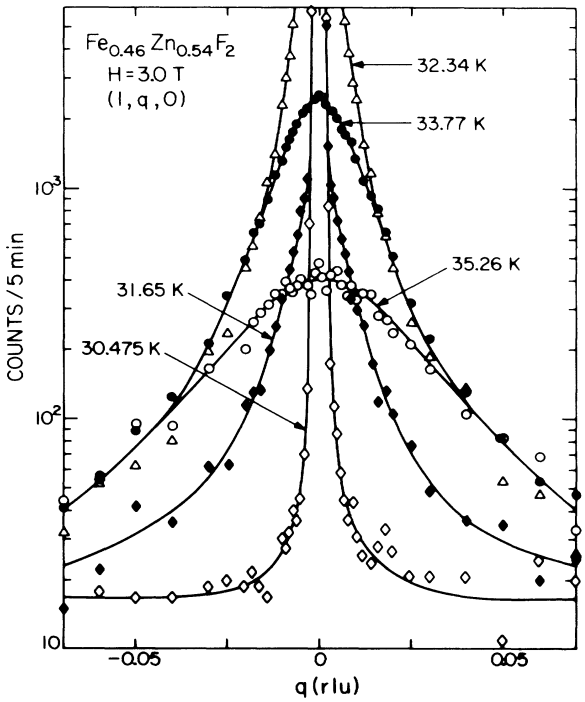

FIG. 1. Logarithm of the scattering intensity $I(q)$ vs q for transverse scans $(1,q,0)$ for five temperatures at $H=3.0$ T after a ZFC procedure. The scans at $T=32.34$, 33.77, and 35.26 K are all above $T_c(H)$ and are well described by a L+LSQ line shape. The $T=31.65$ and 30.475 K scans below $T_c(H)$ clearly show Bragg scattering for $|q| < 0.004$ and cannot be described by a L+LSQ line shape for $|q| > 0.004$.

the growing intensity at small q and decreasing peak widths indicate a divergence of ξ and the staggered susceptibility χ_s. By way of contrast, the scans at $T=30.475$ and 31.65 K show a resolution-limited, nearly Gaussian Bragg peak at $q=0$, in addition to the diffuse tails for larger q, indicating these scans are definitely below $T_c(H)\simeq 32.115$ K; a value independently determined by use of the capacitance method.[14] It is remarkable that the wing intensity for $T<T_c(H)$ is very much weaker than for $T>T_c(H)$, at comparable $|T-T_c(H)|$, and for $q>\kappa$ is strongly dependent on T, unlike the case for $T>T_c(H)$. Because they are much too narrow in the central region, the line profiles in the immediate vicinity above, to well below $T_c(H)$, *cannot* be fitted by a phenomenological L+LSQ line shape used in previous studies. These newly observed features preclude detailed interpretation of the critical behavior (i.e., critical exponent determination) in this region without a deeper theoretical understanding of $S(q)$ itself.

In spite of the uncertainties in $S(q)$, much can be learned by investigating the intensities near the Bragg point. We have carried out measurements of $I(1,0,0)$ and $I(1,-0.004,0)$ vs T following ZFC and FC procedures at three fields: 1.5, 1.9, and 3.0 T near $T_c(H)$. These are plotted in Fig. 2. For reference purposes, Fig. 3 shows $T_c(H)$ and $T_{eq}(H)$ as measured by the capacitance method,[10] after a mean-field correction was made. We first discuss the ZFC scattering results. The Bragg scattering at (1,0,0) is severely extinction limited below $T_c(H)$. Nevertheless, as T increases, the intensity decreases slightly because of the T dependence of the sublattice magnetization. In the region of $T_c(H)$, a peak arises from the critical scattering. Its maximum occurs for $T>T_c(H)$, because the critical scattering for $T>T_c(H)$ is much larger than for $T<T_c(H)$ and is rounded by the dynamics to be discussed below. Where Bragg scattering is completely excluded, as in $I(1,-0.004,0)$, it is clearly seen that the intensity falls more rapidly below $T_c(H)$ than it does above.

To establish the boundary $T_{eq}(H)$ above which no hysteresis is seen,[14] we have also examined the FC intensity at the same values of q and H (see Fig. 2). At (1,0,0) the FC, domain-induced, elastic scattering is not extinction limited below $T_c(H)$ and, hence, lies above the corresponding ZFC intensity. *Above* $T_c(H)$, the ZFC and FC scattering intensities, at both values of q, join with each other at $T_{eq}(H)$. The values so obtained for $T_{eq}(H)$ at all fields agree with the capacitance determination[14] of $T_{eq}(H)$ indicated by arrows in Fig. 2, and shown in Fig. 3. Clearly, $T_{eq}(H)$ lies *above* $T_c(H)$.

Why is there no divergence in the $q=0$ critical scattering intensity at $T_c(H)$ in the ZFC experiment? Since $\delta T_c(H)$ is only 0.016 K [see Fig. 2(c)], the extreme slowing down in the vicinity of $T_c(H)$ must cause the divergence to be impeded, even on the time scale of the neutron experiment. Evidence for such unusual dynamical effects is seen in the inset of Fig. 4, where $I(1,0,0)$ vs T is shown for $H=1.9$ T. At each T, a measurement of 3-min duration was made; but for the

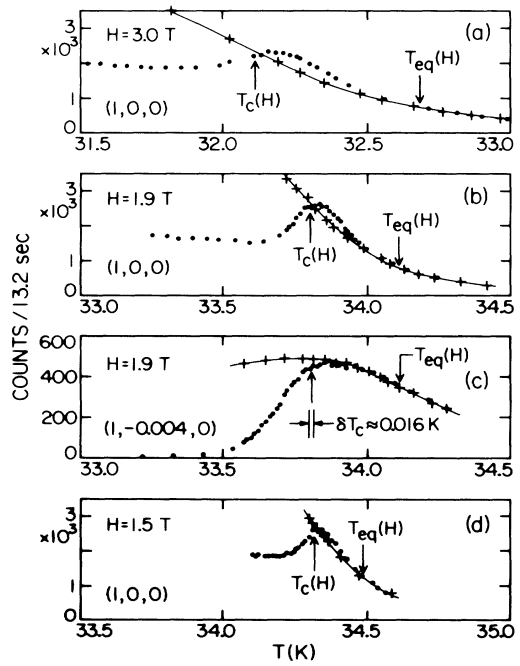

FIG. 2. Scattering intensity $I(q)$ vs T in the vicinity of $T_c(H)$, following (filled circles) ZFC and (crosses) FC procedures. The lines are guides to the eye. The arrows indicate the values of $T_c(H)$ and $T_{eq}(H)$ determined by capacitance measurements (see Fig. 3). [$T_c(1.5\text{ T})=34.315$ K; $T_c(1.9\text{ T})=33.805$ K; and $T_c(3.0\text{ T})=32.115$ K.]

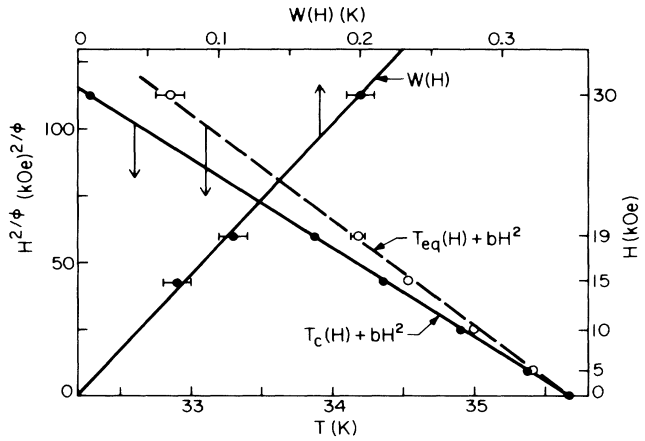

FIG. 3. $T_c(H)$, and "equilibrium temperature," $T_{eq}(H)$, (with a mean-field correction) vs $H^{2/\phi}$, as independently determined from capacitance measurements (Ref. 14). The effective width $W(H)$ of $I(1,0,0)$ vs $H^{2/\phi}$ is also shown. $T_c(H)$, $T_{eq}(H)$, and $W(H)$ all obey RFIM crossover scaling. The $d=3$ random exchange to RFIM crossover exponent $\phi=1.42\pm0.03$ (Ref. 12).

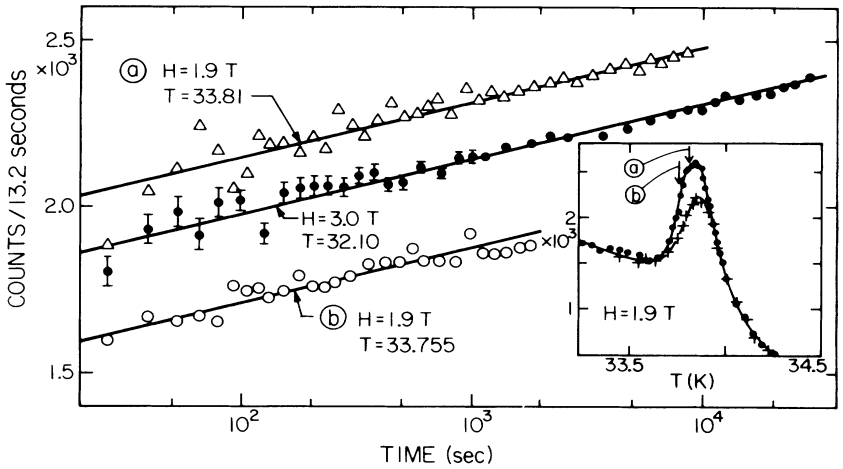

FIG. 4. Inset: $I(1,0,0)$ vs T at $H=1.9$ T for two different scan rates. $I(1,0,0)$ at $T=33.81$ K and $T=33.755$ K at $H=1.9$ T, and $T=32.10$ K at $H=3.0$ T vs the time t following a special ZFC procedure on a semilogarithmic scale. The two values of temperature at $H=1.9$ T are as indicated in the inset.

dashed curve only approximately half the points were sampled, thereby doubling the scan rate. The amplitude is reduced for the faster scan, but only in the vicinity of $T_c(H)$; *no* change is seen outside this region either above or below $T_c(H)$. This indicates that the time evolution appears only in the critical scattering, and is even slower than the measurement times of typically 3 min per point.

To directly measure the dynamic time scales while minimizing thermal equilibration effects, we moved as quickly as possible after ZFC, at a fixed T, to a final H that brought us close to $T_c(H)$. Immediately, $I(1,0,0)$ was recorded for fixed intervals of 13.2 sec. This experiment was performed at $H=1.9$ T near the ZFC peak at $T=33.81$ K and below the peak at $T=33.755$ K, and at the ZFC peak at $T=32.10$ for $H=3.0$ T. A plot of $I(1,0,0)$ vs $\ln t$ shown in Fig. 4 indicates approximately $\ln t$ behavior in all three cases. Small deviations from $\ln t$ behavior, possibly associated with instrumental equilibration effects, were observed in the initial 30 sec. A corresponding FC procedure was followed in which T was fixed at the value for which the ZFC peak occurs at $H=1.9$ T. Except for a weak initial time dependence ($t \leq 100$ sec), $I(1,0,0)$ remained virtually constant.

The contrast between the FC and ZFC results is striking. The absence of time evolution in $I(1,0,0)$ following FC shows that the effect is not in domain relaxation, but is consistent with the suppression of critical fluctuations as is the absence of a sharp peak in the temperature derivative of the capacitance[14] or birefringence[15] at the same field ($H=1.9$ T). However, the behavior of the ZFC $I(1,0,0)$ in Fig. 4 suggests that, as time evolved, the curves shown in the inset would have exhibited ever increasing amplitude as $T \rightarrow T_c(H)$. Hence, we expect $I(1,0,0)$ would approach a divergence at $T=T_c(H)$ and an appropriate static limit for $T \neq T_c(H)$, were it not for extreme slowing down of the critical fluctuations.

Independent evidence for the dynamic effects is found in the field scaling of the widths of the peak in $I(1,0,0)$ vs T. As is seen in Fig. 2, the apparent "rounding" increases with increasing field. One may quantify the rounding by taking the temperature separation $W(H)$ of the inflection points of $I(1,0,0)$ vs T at each of the fields. In Fig. 3, we plot $W(H)$ vs $H^{2/\phi}$ and find it to be linear within experimental error. The value of $W(H)$, at a given H, is comparable in magnitude to the ac $\chi(\omega)$ width[10,11] measured on the *same* time scale.[16] We show below that the $H^{2/\phi}$ dependence is consistent with dynamic scaling.

The observation of unusual time-dependent critical scattering in a $d=3$ RFIM system is in accord with recent theories[8,9] of activated dynamics, and the findings of the $\chi(\omega)$[10] and Faraday rotation[11] studies. Fisher[9] suggests that the peak amplitude χ_p of the *order-parameter* susceptibility scales with frequency ω as $\chi_p(\omega) \propto [\ln\omega]^{\bar{\gamma}/\bar{\nu}\theta}$, where θ is the "violation of hyperscaling" exponent. It is generally believed that $\bar{\nu}=1$,[3-5] but there is no experimental and theoretical agreement on $\bar{\gamma}$ and θ, although the ratio $\bar{\gamma}/\theta$ is of order unity. Because of this uncertainty and that associated with the relation between the observed line profile and χ_s, we assume $I_p(1,0,0) \propto \ln\omega \propto \ln\tau$, where $\tau=\omega^{-1}$ is the "measuring time." Hence, the observed $\ln t$ behavior of the experimental $I(1,0,0)$ vs t is in reasonable accord with the activated dynamic-scaling hypothesis. In conventional dynamic scaling, $\chi_p(\omega) \propto \omega^{-\bar{\gamma}/z\bar{\nu}}$ and would approach $\ln\tau$ behavior in the very large-z limit. Indeed, this interpretation of the ac $\chi(\omega)$[10] yielded $z \simeq 14$.

Because of the same uncertainties connected with $\chi_p(\omega)$, we cannot construct a dynamic reduced rounding temperature $t^*(\omega)$ for $I(1,0,0)$ in the manner used for the ac $\chi(\omega)$.[10] Nevertheless, we take the observed width $W(H)$ of $I(1,0,0)$ vs T, as shown in Fig. 3, as an

effective $t^*(\omega)$. In the region of crossover from random exchange to RFIM behavior, the dominant H dependence of $t^*(\omega)$ in Fisher's theory is $H^{2/\phi}$. The observed $W(H)$ vs H is thus in accord with his prediction in the crossover region, as it would be as well for conventional dynamic scaling in the large-z limit. That the H dependence of the rounding is consistent with either dynamic-scaling approach confirms its dynamic origin.

What relation does the present work have to the finding of a "temporal phase transition" by Birgeneau *et al.*[6] in the weakly anisotropic, RFIM system $Mn_{0.75}Zn_{0.25}F_2$? From an analysis of the neutron scattering in a crystal of considerably larger (and uncertain) concentration gradient, they deduced that the transition is first order, $T_c(H) = T_{eq}(H)$, and a virtual second-order phase transition exists, close to, but definitely *below* $T_c(H)$. It is clear from our study on the virtually gradient-free $Fe_{0.46}Zn_{0.54}F_2$ crystal that (1) $T_{eq}(H)$ lies well above $T_c(H)$, in complete agreement with capacitance[14] and birefringence[15] studies; (2) the $I(1, -0.004, 0)$ peak is rounded by the unusual dynamics and is not gradient induced; its asymmetry reflects the large difference in the critical scattering intensities above and below $T_c(H)$; and (3) no evidence exists for a first-order transition. We believe that in a study of a higher-quality $Mn_xZn_{1-x}F_2$ crystal similar conclusions would be obtained.

The new direct evidence for extreme critical slowing down of the fluctuations in the antiferromagnetic order parameter suggests further study of the q, as well as ω, dependence of the relaxation. We would expect a strong q dependence in the RFIM unlike the spin-glass problem where *no* q dependence is expected or observed. However, because of the extreme slowing down, even with techniques such as the neutron spin-echo method, it may be difficult to obtain sufficient resolution very close to $T_c(H)$ in RFIM systems.

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