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Observation of Crossover in the Dynamic Exponent z in Fe and Ni

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Time-differential perturbed angular correlations have been determined with use of the dynamical critical exponent z for Fe and Ni. In both systems, crossover from $z = 2.5$ to $z = 2.0$ is observed as $T \to T_C$. The value $z = 2.5$ ($d = 3$ Heisenberg model) has been previously observed in neutron-scattering experiments; the value $z = 2.0$ (order-parameter–nonconserving models), in hyperfine-interaction experiments. Thus, the existence of two distinct kinds of dynamic critical behavior, each corresponding to a different universality subclass, is confirmed.

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Spin fluctuations near the Curie temperature in ferromagnets have been studied via neutron scattering and hyperfine interaction methods. For the isotropic, three-dimensional systems Fe and Ni, neutron scattering yields dynamic exponent values of $z = 2.7(2)$ and $z = 2.46(25)$,[1,2] respectively. In contrast, hyperfine-interaction studies invariably produce $z = 2.0(2)$ for the same materials.[3-7]

In a recent Letter,[8] two of us addressed this apparent discrepancy by suggesting that the effective value of z crosses over from 2.5 to 2.0 as the wave vector of the contributing fluctuations becomes small. To justify our hypothesis, we noted that neutron scattering experiments depend predominately on "large" values of q, while hyperfine experiments probe the region near $q = 0$.

From a theoretical point of view, the value $z = 2.5$ is expected for the three-dimensional Heisenberg model (a system that has a conserved order parameter), while $z \cong 2.0$ is predicted for systems that contain appreciable order-parameter–nonconserving terms.[9] Our crossover hypothesis therefore demands identification of suitable spin-nonconserving perturbations, as well as, eventually, explicit calculation of crossover behavior.

From an experimental point of view, the best

test of our hypothesis would be direct observation of crossover in z. To this end, we report here two new experiments, one on Ni and one on Fe, both of which provide direct experimental evidence for the postulated crossover in z. As in our earlier work[3,4] the experiments are based on differential perturbed angular correlation (DPAC) measurements of nuclear relaxation times over a range of temperatures above T_C.

For $T > T_C$ in ferromagnets, critical spin fluctuations are described by the dynamic structure factor involving an inverse correlation length, κ:

$$S(\vec{q},\omega) = 2\pi\omega^{-1}(\vec{q})S(\vec{q})f_{\vec{q}/\kappa}[\omega/\omega(\vec{q})]. \quad (1)$$

Here, $\omega(\vec{q})$ is the energy linewidth, $S(\vec{q})$ is the static or equal-time correlation function, $f_{\vec{q}/\kappa}$ gives the energy line shape, and $\vec{q}$ and ω are the wave vector and energy of the fluctuations involved. All temperature dependence enters through the singular behavior of κ. Physically, $\omega^{-1}(\vec{q})$ is determined by the lifetime of fluctuations with wave vector $\vec{q}$. According to the dynamic-scaling hypothesis[9] the energy linewidth is a homogeneous function of q and κ:

$$\omega(\vec{q}) = q^z\Omega(\vec{q}/\kappa). \quad (2)$$

Here, z is the dynamic critical exponent on which our discussion is focused.

In neutron scattering experiments the exponent z may be found by observing $\omega(\vec{q})$ at T_C. Since $\kappa \to 0$ as $T \to T_C$, Eq. (2) becomes

$$\omega(\vec{q}) = q^z\Omega(\infty). \quad (3)$$

To find z it is thus sufficient to fit energy linewidths at different q to a power law. Because of the presence of a lattice Bragg peak at $q = 0$, neutron scattering experiments are resolution limited to a range of $q \geq 0.05$ Å^{-1}.

For hyperfine-interaction experiments, the exponent z is found by determining τ_i, the ith component of the spin-autocorrelation time, from measured values of the nuclear relaxation time (see below). By definition,[8]

$$\tau_i = C_0\int_{v_q} S(\vec{q},0)\,d^dq. \quad (4)$$

By use of Eq. (1), and the defining equations for the static exponents ν and η, it may be shown for lattice dimensionality $d = 3$ that[8]

$$\tau_i = C_1 t^{-w}, \quad w = \nu(z - 1 - \eta); \quad (5)$$

where t is the reduced temperature, $T/T_C - 1$, for $T > T_C$. Measured values of τ_i are fitted to Eq. (5), and, with use of known values of ν and η, z is deduced. As $t \to 0$, the integrand in Eq. (4) becomes sharply peaked near $q = 0$. Hence, hyperfine measurements of τ_i can probe the region near $q = 0$ that is inaccessible to neutron scattering. On the other hand, for large t, the integral in Eq. (4) is weighted substantially by large values of q.[8] It is this variation in the q weighting of τ_i that makes observation of crossover in z a possibility. Since past hyperfine-interaction experiments yielded z values that apply to "small" q, it was the goal of the new experiments to expand the range of q by expanding the reduced-temperature range to larger t.

As in earlier work,[3,4] ^{100}Pd was produced via ^{103}Rh$(p,4n)^{100}$Pd at the Harvard University cyclotron. After chemical separation of Pd from the natural Rh target, Ni and Fe source foils were made by electroplating from $(NH_4)_2SO_4$ solution, followed by diffusion *in vacuo* at 1370 K. The 3-mm-diam source foils were placed in a novel, double-heater vacuum oven especially designed for DPAC measurements at 1300 K. During measurement, the oven chamber was kept at 2×10^{-6} Torr, and exhibited a long-term temperature stability of 0.05 K. With the full heater current flowing, the measured magnetic field at the sample was $\cong 1$ Oe.

DPAC measurements were made with a standard four counter setup in which two pairs of spectra, $C_{ij}(\theta,\tilde{t})$, were recorded at counter angles of $\theta = \pi$ and $\theta = \pi/2$. $\tilde{t}$ is the time delay between emission of the cascade γ rays. Data were reduced by forming the ratio of background-corrected counting rates,

$$R(\tilde{t}) = \frac{[C_{13}(\pi,\tilde{t})C_{24}(\pi,\tilde{t})]^{1/2} - [C_{23}(\pi/2,\tilde{t})C_{14}(\pi/2,\tilde{t})]^{1/2}}{[C_{13}(\pi,\tilde{t})C_{24}(\pi,\tilde{t})]^{1/2} + [C_{23}(\pi/2,\tilde{t})C_{14}(\pi/2,\tilde{t})]^{1/2}}. \quad (6)$$

This reduction is particularly efficacious because it eliminates irrelevant variables such as single and coincidence counting efficiencies.[10] To minimize background correction problems at large $\tilde{t}$'s, spectra in which large $\tilde{t}$'s were important were taken when the sample sources were very weak. Background determinations were checked for self-consistency in each experiment.[11]

For ^{100}Rh nuclei with spin $\vec{I}$ imbedded in an Fe or Ni lattice we assume an isotropic hyperfine inter-

action of the form $H = A\vec{I}\cdot\vec{S}(\tilde{t})$. In this case, if the correlation times of the fluctuating host electron spins, $\vec{S}(\tilde{t})$, are the shortest time in the problem, and spin fluctuations are isotropic, then[4]

$$R(\tilde{t}) = (A_2/5)\exp(-\tilde{t}/\tau_r)(1+2\cos\omega_L\tilde{t}+2\cos 2\omega_L\tilde{t}). \quad (7)$$

The nuclear relaxation time, τ_r, is given by

$$1/\tau_r = 2\tau_i[\gamma H(0)/S]^2 S(S+1), \quad (8)$$

where $\gamma = \mu_I/I\hbar$ is the nuclear magnetogyric ratio, and $H(0)$ is the saturation hyperfine field. The Larmor frequency is given by $\omega_L = \gamma H(T)/I\hbar$. Above T_C, $\omega_L = 0$, so that,

$$R(t) = A_2\exp(-\tilde{t}/\tau_r). \quad (9)$$

To obtain the spin-autocorrelation time τ_i, data are fitted to Eq. (9), and the resulting values of τ_r are converted via Eq. (8).

The precise values of T_C were determined for the Ni and Fe samples by different methods. For Fe, the combined effects of small τ_r, large ω_L and limited instrumental time resolution prevented the observation of oscillatory signals below T_C or reliable tracking of τ_r close to T_C above the transition. To make the best of this difficult situation, we utilized the temperature dependence of the amplitude $R(0)$ defined by Eq. (7). For $T \gg T_C$, $R(0) = A_2$; for $T \ll T_C$, the precessional terms are attenuated and $R(0) = A_2/5$. Model calculations for the intermediate region show that $R(0)$ breaks away from A_2 at about 1 K *above* T_C of Fe. In this way we are able to estimate $T_C = 1042 \pm 2$ K from the data plotted in Fig. 1.

For Ni, no corresponding problems exist and T_C was determined by fitting a power law to τ_r for the reduced-temperature range $10^{-4} \leq t \leq 5 \times 10^{-3}$, as in previous work.[3,4] The resulting value was $T_C = 627.5 \pm 0.2$ K.

Figure 2 shows all available τ_i values as a function of reduced temperature for Ni and Fe, as deduced from DPAC experiments with the ^{100}Rh probe. Where the present data overlap previous measurements, with both DPAC and the Mössbauer effect[8] (and, thus, a variety of probe nu-

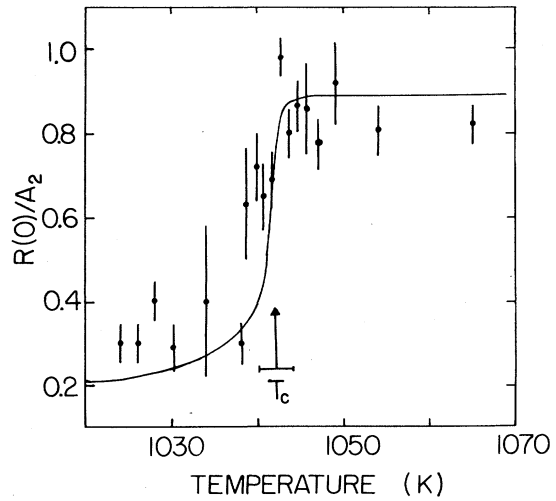

FIG. 1. Amplitude of the reduced DPAC signal near T_c for $Fe^{100}Rh$. The estimate for T_c shown was obtained by methods described in the text, and was used in calculating reduced temperatures for Fe given in Fig. 2.

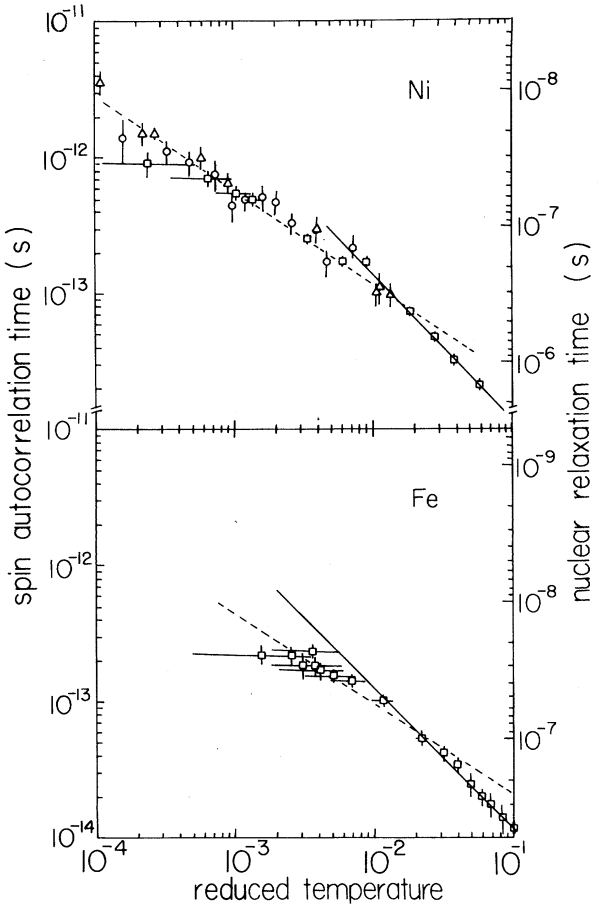

FIG. 2. Reduced temperature variation of the spin-autocorrelation time (left-hand scale) as deduced from measured nuclear relaxation (right-hand scale) via Eq. (8). For Ni (top graph) the round and triangular points were published previously (Refs. 3 and 4), and the square points are new. For Fe all data points are new. The straight lines were obtained from least squares fits to the temperature regions $t < 2\times10^{-2}$ and $t > 2\times10^{-2}$ and yield exponent values quoted in the text.

clei), agreement in the spin-correlation times is obtained. Power-law fits to the data were made for a variety of t ranges with results as follows: Far from T_C in each case, $w = 1.0(1)$, or $z = 2.5(2)$, in agreement with neutron experiments. Close to T_C, Ni yields $w = 0.70(3)$ or $z = 2.0(1)$ and Fe shows a bending toward lower values of w and z. When combined, the data on Fe and Ni clearly confirm crossover in z and thus quantitatively confirm our hypothesis.[12] We therefore have for the first time an unambiguous, consistent interpretation of both neutron-scattering and hyperfine-interaction results on Fe and Ni.

What is the cause of the observed crossover? First, we note that τ_i is the integral of a product: a weight function $S(\vec{q})f_{\vec{q}/K}(0)$ and a wave-vector–dependent relaxation time, $\omega^{-1}(\vec{q})$. As the critical point is approached, the weight function becomes sharply peaked at small q and the region of q space in which the dynamics are affected by the broken conservation law at $q = 0$ is expected to increase. The combination of these two effects may be responsible for the surprisingly sharp crossover observed.

One crossover mechanism which has been treated theoretically, at least for localized spins, is the dipole interaction. Here, the crossover temperature for $q = 0$ dynamics is the ratio of dipole to exchange strengths[13]:

$$t_x = [g^2\mu_B{}^2/a^3]/S(S+1)k_B T_C, \qquad (10)$$

where $g\mu_B$ is the electronic dipole moment, a is the nearest-neighbor distance, and k_B is Boltzmann's constant. It can be shown that Eq. (10) determines an upper bound on the crossover temperature for τ_i.[11] Evaluation of (10) for Ni and Fe leads to crossover temperatures of 2×10^{-5} and 2×10^{-4}, respectively. Since the observed crossover occurs at $t \cong 10^{-2}$ in both systems, it is tempting to conclude that the cause is not dipolar interactions. On the other hand, since Eq. (10) is derived for localized spins, it may not be quantitatively correct for the itinerant, metallic systems. Thus, while a definitive statement cannot be made, the data are suggestive of a mechanism other than the dipole interaction.

We have seen, in a single material, behavior that corresponds to two distinct dynamic universality classes. Far from T_C we see behavior predicted for pure Heisenberg systems. Close to T_C, we find behavior predicted for perturbations that do not conserve the order parameter. The sharp crossover, which may be somewhat unique to the localized nuclear probe, allows observation of both "asymptotic" exponent values.

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