# Spin Dynamics near the Magnetic Percolation Threshold

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An inelastic neutron-scattering study of spin relaxation in the dilute, two-dimensional Ising antiferromagnet $Rb_2Co_cMg_{1-c}F_4$ has been carried out for $c$ very close to the magnetic percolation threshold $c_p = 0.593$. The dynamical exponent $z$ relating the order-parameter relaxation rate to the magnetic correlation length is found to be $2.4^{+0.2}_{-0.1}$. It is argued that $z$ has this anomalously high value because of the fractal geometry of percolating networks.

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The static behavior of randomly diluted systems near their percolation thresholds is well established. Quantities such as the resistivity and pair connectedness lengths vanish according to known power laws, much as susceptibilities and correlation lengths diverge at ordinary, thermally driven phase transitions.[1] Very recently, theoretical attention has shifted to dynamical behavior on percolation networks. The problem which has been studied so far is that of random walks or, equivalently, diffusion.[2] Because of the restricted number of paths connecting two points on a percolating cluster, a random walker's travel times increase more rapidly with distance than they do on a regular lattice. Indeed, several authors[2] have shown that Fick's law, which states that the mean square distance $\langle r^2(t)\rangle$ traveled by a particle scales linearly with the number of time steps $t$, must be generalized to

$$\langle r^2(t)\rangle \sim t^{2/(2+\theta)}. \qquad (1)$$

As expected, the parameter $\theta$ is positive, and can be expressed in terms of the fractal and spectral dimensions of the percolating network. It is also quite large: For two- and three-dimensional percolation, $\theta \cong 0.8$ and 1.5, respectively.[2]

In this paper, we consider the dynamics of coupled spins on a percolating network. This problem is especially attractive because there are many well characterized magnetic dilution series, and the neutron-scattering technique gives direct access to the time- and distance-dependent correlation functions. We report here on an inelastic neutron-scattering study of the longitudinal spin fluctuations in a percolating Ising antiferromagnet, $Rb_2Co_cMg_{1-c}F_4$. The principle result is that the dynamical exponent $z\nu$, which relates the order-parameter relaxation time to the reduced temperature, is much larger than the static susceptibility exponent $\gamma$. In contrast, both experiments[3] on pure $Rb_2CoF_4$ and conventional theory[4] for nonrandom lattices give $z\nu = \gamma$.

We carried out most of our measurements with a triple-axis spectrometer at the cold neutron source of the Brookhaven high-flux beam reactor. This instrument has a double-crystal monochromator, and energy scans are performed by varying the incident neutron energy $E_i$ with the final energy $E_f$ fixed at 2.5 meV. The measured quasielastic energy resolution was 37 $\mu$eV, full width at half maximum (FWHM). We have also collected some data using an ordinary triple-axis spectrometer at a thermal beam. Here, spectra were taken by varying the final energy $E_f$ with $E_i$ fixed at 3.5 meV; the resolution was 64 $\mu$eV (FWHM).

As far as its magnetic properties are concerned, $Rb_2CoF_4$ is a stack of planes containing magnetic $Co^{2+}$ ions arranged on square lattices[3] with spacing $a = 4.12$ Å. The single-ion anisotropy is Ising-like. The intraplanar interactions are much stronger than the interplanar couplings. Consequently, the magnetic behavior is two dimensional, with a Néel point at 103 K and critical exponents as predicted by Onsager.

In the isostructural dilution series $Rb_2Co_c$-$Mg_{1-c}F_4$, grown by methods described elsewhere,[5] nonmagnetic $Mg^{2+}$ ions are randomly substituted for the magnetic $Co^{2+}$ ions. To facilitate comparison between static and dynamical properties, we used one of the samples for which Cowley and coworkers[1] have measured the instantaneous spin correlations. This sample is a single crystal, with $c = 0.58$, very close to the nearest-neighbor percolation threshold ($c = c_p = 0.593$) for a square lattice. We mounted it in a Displex cryostat with the ($h0l$) zone in the plane of the spectrometer. In this notation, ($00l$) is parallel to the Ising spin ($z$) axis; also, for $T = 50$ K, $a^* = 1.086$ Å$^{-1}$ and $c^* = 0.460$ Å$^{-1}$. The present experiment consists of measuring the inelastic scattering at the point $\vec{Q}_0 = (1, 0, 0.4)$, which lies on a line [$\vec{Q} = (1, 0, Q_z)$] in reciprocal space where the Bragg condition for two-dimensional antiferromagnetic ordering is satisfied. Thus, we are probing the fluctuations in the staggered magnetization, which has zero mean at all temperatures because $c$ is slightly below $c_p$.

Figure 1 shows inelastic spectra obtained for several temperatures with $E_f = 2.5$ meV. The important feature of these data is that the spectra narrow substantially over the relatively small temperature range between 75 and 45 K. Indeed, at 75 K [Fig. 1(a)] the spectrum has a full width which is two to three times the full width of the resolution function, while at $T = 45$ K [Fig. 1(c)], the profile is clearly resolution limited. Because the inelastic neutron-scattering cross section is proportional to the Fourier transform $F(\vec{Q}, \omega)$ of the spin-relaxation function, the narrowing of the experimental spectra signifies increasing relaxation times. Note that our energy resolution corresponds to time scales of order $10^{-9}$ sec.

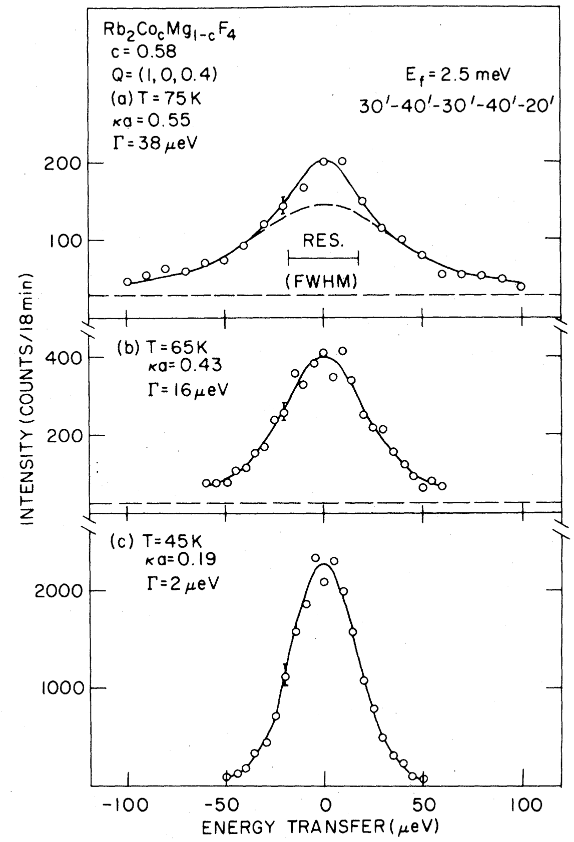


FIG. 1. Constant-$Q$ inelastic spectra, all collected for the collimations and final neutron energy indicated in (a). The dashed horizontal lines represent the constant background, the solid lines correspond to the profile (2) computed using the indicated values of Γ, and the dashed curve in (a) corresponds to the profile (2) calculated without the elastic background term.

To analyze our data, we assume that a single exponential decay rate Γ characterizes the spin relaxation observed at $\vec{Q}_0$. This implies that $F(\vec{Q}_0, \omega)$ is a Lorentzian of width Γ. We therefore fit the experimental spectra by the following form, where we correct for the finite experimental resolution, represented by Δ, by performing a convolution integral:

$$S(\omega_0) = \frac{1}{(2\pi)^{1/2}\Delta} \int_{-\infty}^{\infty} d\omega \exp\left[-\frac{1}{2}\left(\frac{\omega}{\Delta}\right)^2\left\{\frac{1}{\pi}\frac{A\Gamma}{\Gamma^2 + (\omega-\omega_0)^2} + B\delta(\omega-\omega_0)\right\}\right] + C. \quad (2)$$

The constant $C$ represents a flat, energy- and temperature-independent background determined at the lowest temperature; it corresponds to the dashed horizontal lines in Figs. 1(a) and 1(b). $B$ is the amplitude of the resolution-limited peak at $\omega = 0$ produced by incoherent scattering from the sample. Our procedure was to vary, at each temperature, only Γ and $A$ to obtain the best fit of Eq. (2) to the data. For the high-resolution spectra ($E_f = 2.5$ meV), such as those shown in Fig. 1, we fixed, for all $T$, $\Delta = 15.8$ μeV, $B = 130$ counts · μeV/min, and $C = 1.8$ counts/min. The solid line passing through the data in Fig. 1 are cross sections computed according to (2) for fitted values of Γ and $A$. For comparison, we show in Fig. 1(a) a dashed curve corresponding to the cross section (2) calculated *without* the term proportional to $B$.

We now wish to relate the dynamical quantity Γ to the instantaneous spin correlations in our sample. In general, for $c$ below the percolation threshold $c_p$, all spins reside in finite clusters, with a typical pair connectedness length $\xi_g$ which diverges as $c$ approaches $c_p$. Previous work[1,5] has established that the *magnetic* correlation length $\xi$ is the harmonic mean of $\xi_g$, which depends only on $c$, and a second length $\xi_T$, which depends only on $T$:

$$\xi^{-1} = \xi_T^{-1} + \xi_g^{-1}. \quad (3)$$

Because magnetic order in the clusters propagates through essentially one-dimensional paths, $\xi_T$

$\sim \xi_{1D}^{\nu_T}$, where $\xi_{1D}$ is the correlation length of the pure *one-dimensional* magnet with exchange interactions of the same strength as in the dilute system. Figure 2 shows the fitted values of $\Gamma/kT$ plotted against the inverse correlation lengths $\kappa a = a\xi^{-1}$ computed from Eq. (3) with $a\xi_g^{-1} = 0.004$ and $\nu_T = 1.33$, values found by Cowley and co-workers[1] to account for the static behavior in the same sample. We present only the results for temperatures larger than 50 K, where the fitted values of $\Gamma$ are comparable to or exceed one quarter of the resolution function's FWHM, and can therefore be considered reliable. The straight line corresponds to the power law which best describes the data, namely

$$\Gamma/kT = \mathscr{A}(\kappa a)^z \tag{4}$$

with $\mathscr{A} = 0.023 \pm 0.002$ and $z = 2.4 \pm 0.1$. The errors quoted here are statistical; conservatively taking into account possible errors in the background

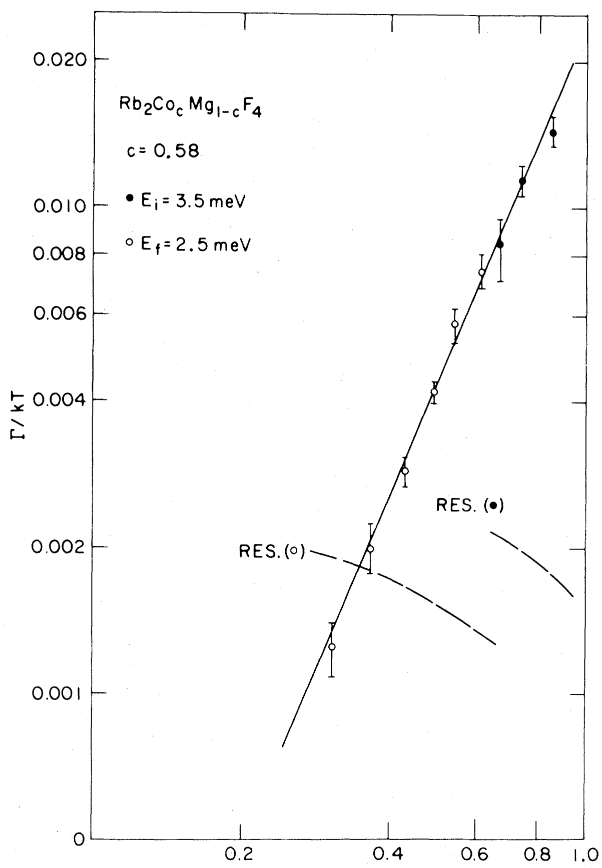


FIG. 2. Normalized relaxation rate $\Gamma/kT$ plotted against reduced inverse correlation length $\kappa a$. At the dashed curves, the FWHM of the resolution function exceeds $\Gamma$ by a factor of 4. The open and filled circles correspond to the two different spectrometers described in text.

parameters $B$ and $C$, we conclude that $z = 2.4^{+0.2}_{-0.1}$. We have chosen to use Eq. (4), rather than, for example, the form $\Gamma = \mathscr{A}(\kappa a)^z$, because it is consistent with both the conventional theory of the kinetic Ising model[4] (see below), and the freezing of classical paramagnets at $T = 0$.

Our measurements have been performed in the temperature range where the simple form (3) describes the static behavior. Nevertheless, it is possible that the dynamical critical regime is much smaller than the static critical region,[4] with the result that the $z$ which we find is not necessarily a *critical* exponent. In any case, because $\kappa >> \xi_g^{-1}$, this $z$ should also characterize the dependence of $\Gamma$ on $\xi_T$ exactly at percolation ($c = c_p$), at least for $T \gtrsim 50$ K.

To place our result for $z$ in context, recall that according to the conventional theory of the kinetic Ising model,[4] relaxation times are proportional to the static susceptibility $\chi$, or

$$\Gamma \sim \chi^{-1} \sim kT\kappa^{2-\eta}. \tag{5}$$

For the Ising model on a regular, two-dimensional lattice, experiment[3] is in agreement with (5). For our dilute system, $\eta \cong 0.33$,[1] and so $z = 2 - \eta \cong 1.67$, which is very far from the value $z \cong 2.4$ found in this experiment. There are several possible reasons for the failure of Eq. (5). The first is that the approximations which lead to (5) are valid only for spatial dimensionalities $d$ larger than 4.[4] The second and more important reason is that a percolating network is *not* a regular lattice, but an object with fractal and spectral dimensionalities between 1 and $d$. Indeed, a large value of $z$ is consistent with the notion that in a system as ramified as a percolating network, disturbances require a longer time to propagate between two points than they do on a regular lattice. The anomalous diffusion[2] on fractal lattices has the same physical origin. Equation (1) essentially means that when we study diffusion on length scales $l$, a real unit of time on a fractal lattice is equivalent to $l^{-\theta}$ units of time on a normal, Euclidean lattice. We now assume that this is also the case for spin relaxation in kinetic Ising models. Setting $l^{-1} = \kappa$, and using the conventional result (5), we obtain $\Gamma/\kappa^{\theta} \sim \chi^{-1}$. Thus, the dynamical exponent $z$ for spin relaxation is $2 + \theta - \eta$. For the two-dimensional percolation problem, $\theta \cong 0.8$[2] and $\eta \cong 0.33$,[1] which leads to $z \simeq 2.47$. Remarkably, this result, based on the conventional theory of the kinetic Ising model and the assumption that the fractal nature of the percolating network influences the time scales for diffusion and spin relaxation in the same way, is consistent with our experimental value, $z = 2.4^{+0.2}_{-0.1}$.

We have found an anomalously high dynamical exponent $z$ for a two-dimensional antiferromagnet near percolation and have argued that this is due to the unusual geometry of percolating networks. In the future, we plan to test the validity of (4) for a larger range of correlation lengths, and for other samples with $c < c_p$. In addition, the dynamical scaling predictions that $\Gamma(q) \sim q^z$ and $F(q=0,\omega) \sim \omega^{-(2-\eta+z)/z}$ should be checked. Studying the kinetic Ising model on percolating networks is a challenge for theorists. A useful first step would be to establish a theory for the dynamics of this model on a fractal lattice, and thus to determine the validity of our conjecture that $z = 2 + \theta - \eta$.

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