Activated Dynamics in the Two-Dimensional Ising Spin-Glass $Rb_2Cu_{1-x}Co_xF_4$

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Activated dynamics are established in $Rb_2Cu_{0.782}Co_{0.218}F_4$, which is a model compound for the two-dimensional Ising spin-glass with random nearest-neighbor bonds. Cole-Cole analysis of the ac complex susceptibility reveals an extremely wide distribution of relaxation times, and a divergence, extracted to cover sixteen decades, of the median relaxation time τ_c towards $T_c=0$ K according to $\ln(\tau_c/\tau_0) \propto T^{-1-\psi\nu}$, with $\psi\nu=2.2\pm0.2$. Activated dynamic scaling of the out-of-phase susceptibility is found to hold over a wide range of temperatures and frequencies. Estimates for the critical exponents γ, ν, and ψ are given.

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The dynamics of random magnetic systems has very recently become a subject of considerable interest. Particularly for spin-glasses (SG),[1,2] theoretical arguments[3-6] have proposed that at low temperatures the dynamics is governed by thermally activated processes over high-energy barriers. In the case of a two-dimensional ($d=2$) SG with Ising interactions, for which there exists ample evidence for a *zero* critical transition temperature,[1,2,7] activated dynamics implies a critical behavior quite different from ordinary critical slowing down. The basic idea is that the typical barrier height B amounts to $B \sim L^{\psi}$ $(0 \le \psi \le d-1)$ for excitations up to length scale $L \sim \xi$, with $\xi \propto T^{-\nu}$ the critical correlation length.[4,6] For the characteristic relaxation time τ of the system this leads to

$$\ln\left(\frac{\tau}{\tau_0}\right) = \frac{B}{T} \propto \frac{\xi^{\psi}}{T} \propto \frac{1}{T^{1+\psi\nu}}, \tag{1}$$

where τ_0 is the microscopic single-spin-relaxation time. Activated dynamics thus involves a substantial faster divergence of τ than according to a conventional power law. The randomness further causes the barrier heights to be distributed with a typical spread ΔB of again $\sim L^{\psi}$, resulting in an extremely broad distribution of relaxation times. Because of the decisive role of the dimensionality, it is crucial that an experimental study of the critical dynamics be performed in a real $d=2$ SG. Superlattices of thin SG films and nonmagnetic layers have recently been found to exhibit crossover from $d=3$ to $d=2$,[8,9] but presently are too far from the $d=2$ limit to draw quantitative conclusions.

In this Letter, we present the first experimental verification of activated dynamics in a genuine $d=2$ system. This is accomplished in two entirely distinct ways: by analyzing the complex susceptibility, first, with a Cole-Cole approach and, second, with dynamic scaling. The former analysis allows one to extract relaxation times at various temperatures, while dynamic scaling rests on a more general basis. The system studied, $Rb_2Cu_{1-x}Co_x$- F_4, has been demonstrated to be a nearly ideal realization of the $d=2$ Ising SG with random nearest-neighbor bonds.[7] It is a random mixture of the archetypal square-lattice antiferromagnet Rb_2CoF_4 (effective spin $S=\frac{1}{2}$, $T_N=103.0$ K), having the layered K_2NiF_4 structure, and the isostructural ferromagnet Rb_2CuF_4 ($S=\frac{1}{2}$, $T_c=6.05$ K). $Rb_2Cu_{1-x}Co_xF_4$ exhibits SG behavior for $0.18<x<0.40$. The present single crystal has $x=0.218$.

The in-phase and out-of-phase susceptibilities $\chi'(\omega,T)$ and $\chi''(\omega,T)$ were collected by use of conventional mutual-inductance methods[10] at a set of discrete temperatures and at frequencies ranging from 0.3 Hz to 50 kHz with the driving field along the tetragonal crystal axis. All data were corrected for demagnetization. The results are presented in Fig. 1 versus the temperature. At first glance, χ' resembles the result typical for a $d=3$ SG, viz., it shows a clear maximum at a frequency-dependent temperature T_f. However, a number of remarkable distinctions for a $d=3$ SG are the following: (i) The maximum $\chi'(\omega,T_f)$ is strongly dependent on the frequency, varying by a factor of about 3 in our frequency range; (ii) the susceptibility deviates from being isothermal at temperatures as high as $2T_f$; (iii) whereas at higher temperatures χ'' shows the usual increase with increasing frequency, χ'' decreases with ω below approximately 4 K.

In order to deduce the relaxation times, the frequency dependence of the susceptibility has been analyzed using the Cole-Cole method.[11] This path has previously been followed with some success in examining the dynamics of various $d=3$ SG, including the "canonical" SG

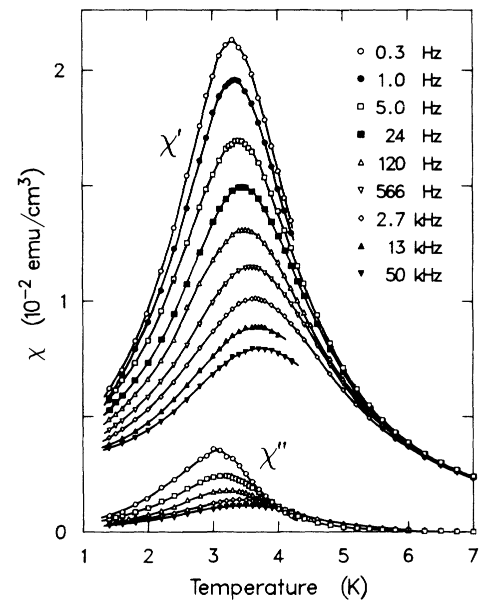

FIG. 1. In-phase (χ') and out-of-phase (χ'') linear susceptibilities for $Rb_2Cu_{0.782}Co_{0.218}F_4$ vs the temperature. Data were taken at frequencies $\omega/2\pi$ ranging from 0.3 Hz to 50 kHz. Solid lines are guides to the eye. For clarity χ'' is presented only for selected frequencies.

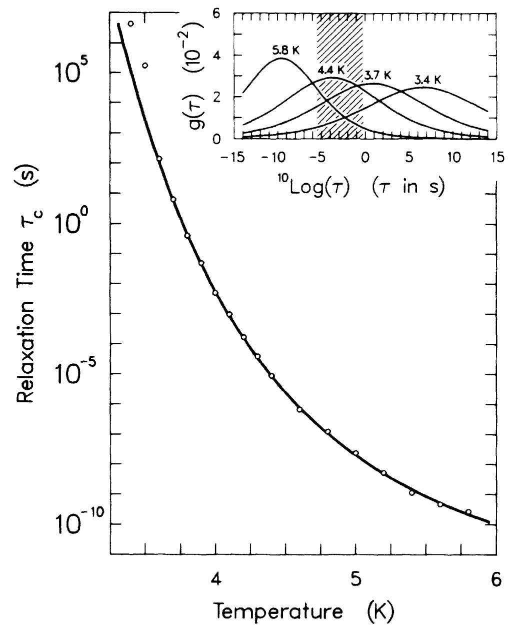

FIG. 2. Relaxation time τ_c vs temperature. Solid line is a fit of Eq. (1). Inset: distribution of relaxation times $g(\tau)$ vs the relaxation time τ. Shaded area denotes the time window of the present experiment.

$Cu_{1-x}Mn_x$ and $Eu_xSr_{1-x}S$.[12-14] In our case such an approach is particularly justified on the grounds of the extremely broad distribution $g(\tau)$ of relaxation times.[15] The Cole-Cole equation for the complex susceptibility may be written

$$\chi(\omega)=\chi_S+\frac{\chi_0-\chi_S}{1+(i\omega\tau_c)^{1-\alpha}}, \tag{2}$$

in which χ_0 and χ_S are the isothermal and adiabatic susceptibilities, respectively, and τ_c is the median relaxation time. The parameter α determines the width of the distribution, such that $\alpha=1$ corresponds to an infinitely wide distribution, while for $\alpha=0$ Eq. (2) reverts to the Debye equation appropriate for relaxation with a single time constant. Equation (2), separated into its real and imaginary parts, was, at constant temperatures, fitted to the susceptibility data of Fig. 1 with χ_S, χ_0, α, and τ_c as adjustable parameters. Excellent agreement was achieved. The resultant χ_S was found to equal zero within the experimental accuracy in all cases. Furthermore, χ_0 rises monotonically with decreasing temperature. Below 3.4 K, however, the error bars of χ_0 and thus τ_c rise excessively. The parameter α varies slowly and almost linearly with temperature from 0.848 ± 0.005 at 6.00 K to 0.902 ± 0.002 at 3.40 K. At temperatures so low that $\omega\tau_c\gg1$, the frequency dependence of $\chi''(\omega)$ reduces to $\omega^{-(1-\alpha)}$, yielding, for example, $\alpha=0.92\pm0.01$ at 2.00 K. (In the case of constant $\Delta B/B$, α should increase with decreasing temperature.) These truly high values give evidence of an extremely broad distribution of relaxation times persisting throughout the entire temperature range studied.

The temperature dependence of the relaxation time τ_c, which constitutes a principal result of our analysis of the complex susceptibility, is shown in Fig. 2. Clearly, the dynamic response slows down by as many as sixteen decades in time within the range of temperatures considered. This dramatic divergence is excellently described in terms of activated dynamics. The solid line in Fig. 2, then, represents a fit of $\tau_c=\tau_0\exp(b/T)^{1+\psi\nu}$, appropriate to a system with a zero critical temperature T_c [cf. Eq. (1)]. Note that the data points become rapidly more uncertain beyond the upper bound of the experimental time window (~1 s). The fit ($\chi^2\approx1$) yields $\psi\nu=2.2\pm0.2$,[16] $\tau_0=(2\pm1)\times10^{-13}$ s, and $b=10.8\pm0.6$ K. The finite value for $\psi\nu$ excludes a simple Arrhenius description, which would correspond to $\psi\nu=0$. The result for τ_0, which conforms to an exchange energy of order 40 K, is of quite reasonable magnitude, and so is the result for b, which is a measure of the temperature-dependent barrier height B via $B=b^{1+\psi\nu}/T^{\psi\nu}$ [cf. Eq. (1)].

It is of considerable interest to point out that the data in Fig. 2 are also at variance with power-law dynamics as well as a finite T_c. A fit of a power law diverging at $T_c=0$, i.e., $\tau_c\propto T^{-z\nu}$, entirely fails ($\chi^2\approx40$), and if pursued yields unphysical values for $z\nu$ of order 80. A finite T_c of the SG transition in $Rb_2Cu_{1-x}Co_xF_4$ could be argued for because of residual interlayer coupling. Then, the dynamics would be governed by ordinary critical slowing down, i.e., $\tau_c \propto (T-T_c)^{-z\nu}$. Although such a power law yields fits of reasonable quality ($\chi^2=1-4$), the resultant $z\nu=15\pm1$ is unacceptably large, while the $T_c=3.26\pm0.07$ K found is incompatible with $T_f=2.97\pm0.02$ K measured at a time of 90 s.[7] Forcing T_c to lower values drives $z\nu$ to values of over 20, and deteriorates the fit. In the case of activated dynamics, a finite T_c markedly worsens the fit. Setting $\tau_0=2\times10^{-13}$ s, we find $\chi^2=5$ for $T_c=1.0$ K, gradually increasing to $\chi^2=90$ for $T_c=3.0$ K. The inadequacy of this fitting function also rules out a Vogel-Fulcher description, which has $\psi\nu=0$ and a finite T_c in our notation. In conclusion, therefore, the temperature dependence of the relaxation time is excellently, and uniquely, accounted for in terms of activated dynamics with a vanishing critical temperature.

With knowledge of the median relaxation time τ_c and the width parameter α, it is straightforward to calculate the distribution of relaxation times associated with Eq. (2).[14] In the inset of Fig. 2 some representative distributions are shown. The salient feature here is that the profiles $g(\tau)$, which on a logarithmic scale are symmetric about τ_c, turn out to be extremely broad, and remain so up to temperatures substantially above T_f. In fact, at 5.8 K the distribution spans as many as ten decades in time (full width at half maximum), slowly spreading by another five decades when the temperature is reduced to 3.4 K. Meanwhile, the distribution is seen to shift as a whole towards larger times over a distance significantly beyond its width. The temperature dependence of $g(\tau)$ observed in $Rb_2Cu_{0.782}Co_{0.218}F_4$ is exemplary for the spectrum of relaxation times in the $d=2$ Ising SG. Indeed, it sharply contrasts with the results available for a $d=3$ SG above T_c.[12-14]

An alternative analysis of the data, which extends to lower temperatures and yields additional information on the critical exponents, is based on scaling of the susceptibility in the frequency domain according to activated dynamics. Contrary to the Cole-Cole analysis, scaling does not require any assumptions on the precise form of the distribution of relaxation times. For random systems controlled by a zero-temperature fixed point the dynamic spin-correlation function may quite generally be expressed in a scaling form with argument $\ln(\omega\tau_0)/\xi^{\psi}$,[6] and accordingly $\chi''(\omega,T)$ may be cast into the activated-dynamic-scaling form

$$\chi'' T^{-p} = \mathcal{F}[-\ln(\omega\tau_0)T^q]. \tag{3}$$

Here, $\mathcal{F}$ is a scaling function, while $p\equiv -1-\upsilon(2-\eta-\psi)=-1-\gamma+\psi\nu$, $q\equiv\psi\nu$, and τ_0 are scaling parameters; γ and η are the usual critical exponents.

In Fig. 3, we present a scaling plot of $\chi''(\omega,T)$ as obtained by optimizing p, q, and τ_0 such as to achieve maximal coincidence on a universal curve. In this plot all $\chi''(\omega,T)$ data have been included. The frequency $\omega/2\pi$ thus runs from 0.3 to 50000 Hz, while T varies from 1 to 7 K. Scaling is found to be satisfactorily obeyed for $p=-3.0\pm0.5$, $q=2.2\pm0.3$, and $\tau_0=10^{-13\pm1}$ s, apart from minor deviations near the maximum of the scaling function, which corresponds to $\omega\tau_c\approx1$ with the τ_c of the pertinent temperature. The result for $q=\psi\nu$ and τ_0 are in agreement with the corresponding results of the Cole-Cole analysis, which is most gratifying in that it demonstrates the consistency of the two approaches. Combination of the results for p and q yields $\gamma=4.2\pm0.6$ for the critical exponent of the order-parameter susceptibility, which compares well with the result $\gamma=4.5\pm0.2$ deduced from the static nonlinear susceptibility.[7] An important conclusion deduced from Fig. 3 is that activated dynamic scaling in $Rb_2Cu_{0.782}Co_{0.218}F_4$ holds over the entire regime of temperatures despite the freezing of the system into a nonequilibrium state. At low temperatures the system is obviously not left to relax for a time exceeding the longest relaxation time present in the spectrum, but only for the time necessary to take a data point (of order 10^2 s). Yet, the latter time still substantially exceeds the time scale ω^{-1} at which the fluctuations are probed by the susceptibility, and therefore the susceptibility may be expected to closely reflect true equilibrium.

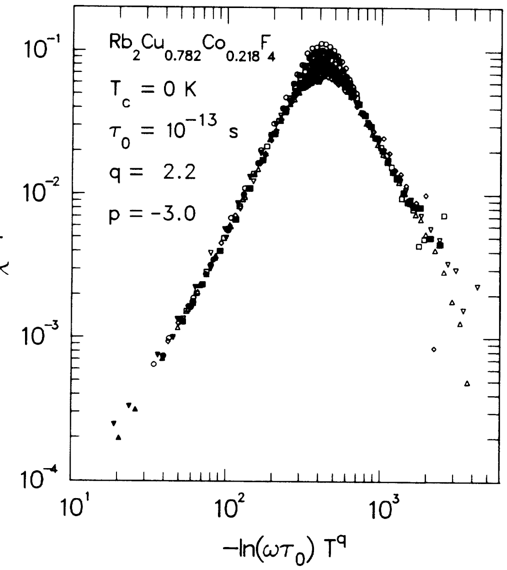

FIG. 3. Activated-dynamic-scaling plot of the $\chi''(\omega,T)$ data for temperatures ranging from 1 to 7 K and frequencies ranging from 0.3 Hz to 50 kHz. Symbols correspond to the frequencies in Fig. 1.

We finally derive a complete set of critical exponents. The results $\gamma=4.2\pm0.6$ favors a comparison of the present system with the $\pm J$ model for the $d=2$ Ising SG, for which estimates of γ range from 4.1 to 5.3, rather than the Gaussian model, which leads to $\gamma\approx7$.[17] Indeed, $Rb_2Cu_{1-x}Co_xF_4$ is expected to be in the $\pm J$ universality class because, first, the distribution of interactions is discrete, and, second, the number of spins subject to exchange fields balancing to zero is nonvanishing.[7] From the scaling relation $\gamma = \nu(2-\eta)$ we then deduce $\nu = 2.3 \pm 0.4$ upon adopting for η a small positive value ($\eta = 0.2 \pm 0.2$), as is appropriate for the $\pm J$ SG.[5] Further, by use of our result $\psi\nu = 2.2 \pm 0.2$, there follows $\psi = 0.9 \pm 0.2$, which is compatible with the upper limit $\psi \le d-1$.[4,6]

In summary, activated dynamics has been established in the model $d=2$ Ising short-range SG $Rb_2Cu_{0.782}$-$Co_{0.218}F_4$. This result has been accomplished by measuring the complex susceptibility, and analyzing the data with two independent methods, viz., Cole-Cole analysis and a scaling treatment. The two methods yield identical values for the relevant parameters. The results verify recent theoretical predictions, and the critical exponents are found to be in conformity with those of the $d=2$ $\pm J$ Ising SG.

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[15]Cole-Cole analysis implies symmetry of $g(\tau)$ vs $\log\tau$. In the experimental data justification for this is found in the symmetry of $\chi''(\omega)$ vs $\log\omega$.

[16]A naive method to determine $\psi\nu$ would be to assume that $T_f(\omega)$, defined by the maximum of χ' vs T, goes as $T_f(\omega) \propto [-\ln(\omega\tau_0)]^{-1/(1+\psi\nu)}$ [cf. Eq. (1)]. While this in the present case appears to be consistent with activated dynamics as opposed to power-law dynamics, the resultant $\psi\nu = 3.4 \pm 0.2$ obviously is in error.

[17]See compilation in Table II of R. N. Bhatt and A. P. Young, in Ref. 2, p. 215.