Activated dynamics in a two-dimensional Ising spin glass: $Rb_2Cu_{1-x}Co_xF_4$

C. Dekker, A. F. M. Arts, and H. W. de Wijn
Faculty of Physics and Astronomy, University of Utrecht, P.O. Box 80.000, 3508 TA Utrecht, The Netherlands

A. J. van Duyneveldt and J. A. Mydosh
Kamerlingh Onnes Laboratorium, Rijksuniversiteit Leiden, P.O. Box 9506, 2300 RA Leiden, The Netherlands
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The complex ac susceptibility of the two-dimensional Ising spin glass $Rb_2Cu_{0.782}Co_{0.218}F_4$ is measured as a function of the temperature and frequency. The results are first analyzed in terms of the Cole-Cole formalism. It is found that the median relaxation time τ_c diverges towards $T_c=0$ K over as many as 16 decades according to activated dynamics, i.e., $\tau_c \propto \exp[(b/T)^{1+\psi\nu}]$. The distribution of relaxation times appears to be nearly symmetric in $\ln\tau$, covering at least 10 decades in width. These findings are corroborated with analyses based on exponential-logarithmic and stretched-exponential forms of the time decay of the spin-spin correlation function. The out-of-phase susceptibility is further found to be in conformity with activated dynamic scaling over a wide range of temperatures and frequencies. For the critical exponents the various methods of analysis yield, on the average, $\gamma=4.4\pm0.2$, $\nu=2.4\pm0.3$, $\theta=-0.41\pm0.05$, and $\psi=0.8\pm0.2$.

I. INTRODUCTION

The subject of spin glasses (SG) has challenged both experimentalists and theoreticians for over 15 years. A primary and long-standing controversy concerned the very existence of the SG equilibrium state. The consensus has now been reached that the lower critical dimensionality of short-range Ising SG is between two and three,[1] i.e., in the case of dimensionality $d=3$ an equilibrium ordered phase is achieved at a finite critical temperature T_c, whereas $d=2$ systems only order at $T_c=0$.[2] With this central question being settled, the attention has focused on the intriguing issue of the dynamics. In the critical dynamics of SG systems, the lattice dimensionality again plays a crucial role. For $d=3$, the critical behavior of the short-range Ising SG is accepted to be of the conventional type. Both numerical simulations[3] of the $d=3$ $\pm J$ Ising model and susceptibility studies of $d=3$ Ising SG, notably $Fe_{0.5}Mn_{0.5}TiO_3$,[4] have shown that above T_c the characteristic relaxation time τ_c diverges according to

$$\tau_c \propto \xi^z \propto (T-T_c)^{-z\nu} ,$$

where z is the dynamic critical exponent and ν the exponent of the correlation length. In the case of systems with a zero-temperature critical point, however, recent theoretical arguments[5-11] have made one realize that static fluctuations associated with the randomness prevail over dynamic thermal fluctuations. Accordingly, the critical dynamics in a $d=2$ SG is anticipated to be governed by thermally activated relaxation over energy barriers. This study aims at providing a comprehensive experimental verification of activated dynamics in an actual $d=2$ system.[12] This is accomplished in $Rb_2Cu_{1-x}Co_xF_4$, a nearly ideal realization of the $d=2$ Ising SG with random nearest-neighbor bonds, by investigating the frequency dependence of the complex susceptibility.

The fundamental concept of activated dynamics in random systems is that the free energy of low-lying excitations of length scale L is given by $F\sim L^{\theta}$, with $\theta<0$ for $d=2$. As a result, excitations of arbitrary low energy destroy the SG order at a finite temperature. The low-temperature divergence of the correlation length, $\xi \propto T^{-\nu}$, is determined by $\nu=1/|\theta|$ from the condition that F is of order T. At shorter length scales, relaxation over energy barriers of height $B\sim L^{\psi}$, with $\psi\geq 0$, controls the fluctuations. For the characteristic relaxation time τ_c this leads to[9]

$$\ln\left[\frac{\tau_c}{\tau_0}\right]=\frac{B}{T}\propto\frac{\xi^{\psi}}{T}\propto\frac{1}{T^{1+\psi\nu}} , \quad (1)$$

with τ_0 the microscopic single-spin relaxation time. Activated dynamics thus involves a much faster divergence of τ_c than according to a conventional power law. Note that the randomness further induces a distribution of barrier heights with a typical spread ΔB of again $\sim L^{\psi}$, implying an extremely broad distribution of relaxation times.

Below, the complex susceptibility of $Rb_2Cu_{1-x}Co_xF_4$ is analyzed in several ways. First, we extract characteristic relaxation times to be identified with the time τ_c in Eq. (1). These analyses are based on the Cole-Cole description (Sec. III A) and, alternatively, on the time decay of the dynamic correlation function (Sec. III B). Irrespective of the particular analysis, however, evidence for activated dynamics is found from a fast divergence of τ_c towards $T_c=0$ extending over a great many decades in time. The divergence appears to be in excellent accord with Eq. (1) with, on the average, $\psi\nu=1.9\pm0.3$. Further, an extremely broad distribution of the relaxation times, with $\Delta B/B$ of order unity, is established (Sec. III C). Another independent analysis of the data relies on dynamic scaling of the out-of-phase susceptibility, an approach not requiring specific assumptions to be made

about the distribution of relaxation times. Dynamic scaling appears feasible over a wide range of temperatures and frequencies (Sec. IV), and is shown to yield essentially the same result for $\psi\nu$ ($\psi\nu=2.2\pm0.3$). Moreover, dynamic scaling provides the critical exponent γ, which, in turn, allows estimates for ν, θ, and ψ.

II. EXPERIMENTAL RESULTS

$Rb_2Cu_{1-x}Co_xF_4$ is a random mixture of the archetypal square-lattice antiferromagnet Rb_2CoF_4 ($S_{\text{eff}}=\frac{1}{2}$, $T_N=103.0$ K), having the layered K_2NiF_4 structure, and the isostructural ferromagnet Rb_2CuF_4 ($S=\frac{1}{2}$, $T_c=6.05$ K). The mixed system exhibits SG behavior for $0.18<x<0.40$.[13] The competition between strongly Ising-type ferromagnetic and antiferromagnetic nearest-neighbor interactions make $Rb_2Cu_{1-x}Co_xF_4$ closely resemble the short-range Ising Edwards-Anderson model for $d=2$.[2] The present single crystal has $x=0.218$, and is part of the same Czochralski-grown single crystal of which the static nonlinear susceptibility has been studied previously.[2]

The differential-susceptibility data were collected at a set of discrete temperatures and frequencies ranging from 0.3 Hz to 50 kHz by use of conventional mutual-inductance methods. The in-phase and out-of-phase parts were recorded simultaneously with phase-sensitive detection techniques. The susceptibility was checked to be linear at the driving-field amplitudes used, which were typically 1 G. The measured susceptibility $X=X'-iX''$ was converted to the intrinsic susceptibility $\chi=\chi'-i\chi''$ by correcting for demagnetization, i.e., $\chi^{-1}=X^{-1}-4\pi N$,[14] or

$$\chi'=\frac{X'-4\pi N(X'^2+X''^2)}{(1-4\pi NX')^2+(4\pi NX'')^2}\ , \tag{2a}$$

$$\chi''=\frac{X''}{(1-4\pi NX')^2+(4\pi NX'')^2}\ , \tag{2b}$$

with N the relevant demagnetization factor of the roughly ellipsoidal sample. The sample was oriented such that the c (Ising) axis was along the driving field. No external dc fields were applied. The temperature was servostabilized to within a few mK, and measured with two calibrated carbon-glass resistors.

The in-phase and out-of-phase susceptibilities $\chi'(\omega,T)$ and $\chi''(\omega,T)$ are, for a selection of frequencies, presented versus the temperature in Fig. 1. At first glance, χ' resembles the results typical for a $d=3$ SG, in line with what recently has been argued.[9] A clear maximum at a frequency-dependent temperature T_f is observed, which is indicative of freezing into a nonequilibrium state. There are, however, a number of important distinctions from the short-range $d=3$ Ising case:[4] (i) the apparent freezing at $T_f\approx3.4$ K occurs much below the temperature associated with the typical spread in exchange energies, viz., $\Delta J\approx35$ K;[15] (ii) the maximum of $\chi'(\omega,T)$ at T_f is very strongly dependent on the frequency, varying in height by a factor of approximately 3 in our frequency range; (iii) the susceptibility deviates from being isothermal already at temperatures as high as $2T_f$; (iv) above T_f the frequency dependence of χ'' is extremely weak, even on a logarithmic scale; and (v), although at higher temperatures χ'' shows the usual increase with increasing frequency, χ'' decreases with ω below approximately 4 K. Finally, χ'' exhibits an inflection point, although a faint one, at a few tenths of a K above T_f, goes through a maximum at typically 0.2 K below T_f, and decreases strongly towards lower temperatures.

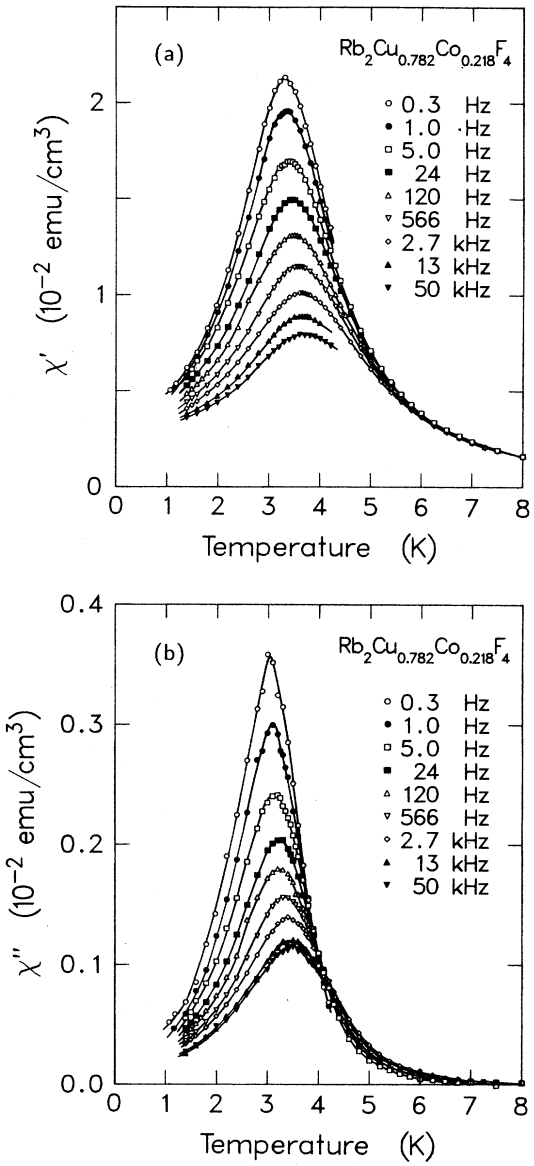

FIG. 1. (a) In-phase linear susceptibility $\chi'(\omega,T)$ for $Rb_2Cu_{0.782}Co_{0.218}F_4$ vs the temperature. Data were taken at frequencies $\omega/2\pi$ ranging from 0.3 Hz to 50 kHz, and with the driving field along the c axis. Solid lines are guides to the eye. (b) Same as (a), but out-of-phase linear susceptibility $\chi''(\omega,T)$.

III. RELAXATION TIMES

In this section, we analyze the data of Fig. 1 with the primary aim to deduce the relaxation times and to com-

pare their temperature dependence with Eq. (1). This is independently accomplished in terms of the phenomenological description of Cole and Cole (Sec. III A) and, alternatively, by use of the Fourier transform of the time decay of the spin-correlation function (Sec. III B). For the latter, two forms will be considered, viz., the exponential-logarithmic decay and the stretched-exponential decay.

A. Cole-Cole analysis

The Cole-Cole formalism[16] involves a modeling of the dynamics at a given temperature onto a distribution of relaxation times that is symmetric on a logarithmic time scale. Substantial justification for this assumption is found in Eq. (1) as well as the experiments themselves. According to Eq. (1), applicable to the case of activated dynamics, a distribution in the barrier height B reflects itself in a distribution of $\ln\tau$ rather than τ. Experimental evidence for the distribution is found from the frequency dependence of χ'', which, for broad distributions and correspondingly weak dependences of χ'' on the frequency, quite directly mirrors the distribution of relaxation times. As it appears (cf. Fig. 4), χ'' indeed is a broad and nearly symmetric function of $\ln\omega$.

The Cole-Cole equation may be written

$$\chi(\omega)=\chi_S+\frac{\chi_0-\chi_S}{1+(i\omega\tau_c)^{1-\alpha}}\ , \tag{3}$$

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. (3) returns to the Debye equation appropriate for relaxation with a single time constant. Equation (3) may be decomposed into

$$\chi'(\omega)=\chi_S+\frac{\chi_0-\chi_S}{2}\left[1-\frac{\sinh[(1-\alpha)\ln(\omega\tau_c)]}{\cosh[(1-\alpha)\ln(\omega\tau_c)]+\sin(\frac{1}{2}\alpha\pi)}\right], \tag{4a}$$

$$\chi''(\omega)=\frac{\chi_0-\chi_S}{2}\left[\frac{\cos(\frac{1}{2}\alpha\pi)}{\cosh[(1-\alpha)\ln(\omega\tau_c)]+\sin(\frac{1}{2}\alpha\pi)}\right]. \tag{4b}$$

Although Eqs. (4) may be adjusted directly to the experimental data with χ_0, χ_S, α, and τ_c as adjustable parameters, it is advantageous to first perform a fit to the locus of χ in the complex plane (Cole-Cole diagram),

$$\chi''(\chi')=-\frac{\chi_0-\chi_S}{2\tan[\frac{1}{2}\pi(1-\alpha)]}\left[\left[\frac{\chi_0-\chi_S}{2}\right]^2-\left[\frac{\chi_0-\chi_S}{2\tan[\frac{1}{2}\pi(1-\alpha)]}\right]^2-\left[\chi'-\frac{\chi_0+\chi_S}{2}\right]^2\right]^{1/2}. \tag{5}$$

This yields χ_S, χ_0, and α, and permits τ_c to be extracted from a fit of Eqs. (4) with χ_0, χ_S, and α set at the values determined in the fitting of Eq. (5). Note that in the relevant part of the diagram Eq. (5) represents a circular arc of size $(1-\alpha)\pi$ cutting the χ' axis at $\chi'=\chi_S$ and $\chi'=\chi_0$. At the maximum of χ'', $\omega\tau_c=1$.

Figure 2 shows some representative Cole-Cole diagrams, i.e., Eq. (5) with the fitted values of the parameters inserted, together with the data points. Note that χ'/χ_0 and χ''/χ_0 are the more natural variables for a comparison of χ at different temperatures. The most noteworthy feature of Fig. 2 is the shift of the data points from a nearly isothermal susceptibility at 6.75 K, via a maximum in χ'' around 4 K, to an almost vanishing susceptibility at 3.40 K. This, in fact, corresponds with a τ_c evolving from $\tau_c \ll 10^{-6}$ s to $\tau_c \gg 1$ s. The relaxation time τ_c thus moves rapidly through our time window, which covers slightly over 5 decades, within only a fraction of a K. Below 3.4 K the fits do not give reliable values for χ_0 and τ_c. As to the resultant fitting parameters, χ_S was found equal to zero within errors in all cases. The temperature dependence of χ_0 will be discussed below in Sec. III D. The parameter α, shown in Fig. 3(a), amounts to 0.902 ± 0.002 at 3.40 K. It decreases linearly with increasing temperature, but remains above 0.80. At temperatures so low that $\omega\tau_c \gg 1$, Eq. (4b) simplifies to $\chi''(\omega)\propto\omega^{-(1-\alpha)}$, which still allows a determination of α. Good fits are found, yielding, for example, $\alpha=0.92\pm0.01$ at 2.00 K. These truly high values of α evidence that an

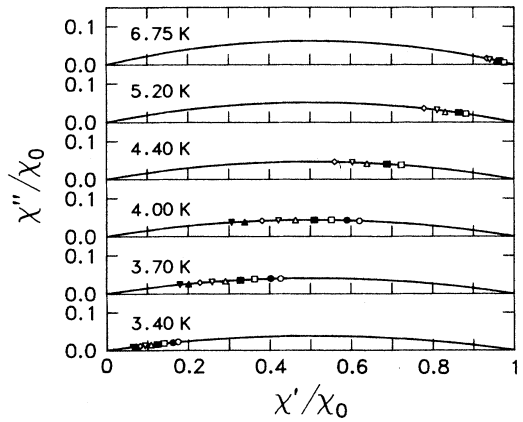

FIG. 2. Cole-Cole diagram for a selection of temperatures. Susceptibilities are normalized to the isothermal susceptibility χ_0. Symbols correspond to the frequencies in Fig. 1. Solid lines are fits of Eq. (5).

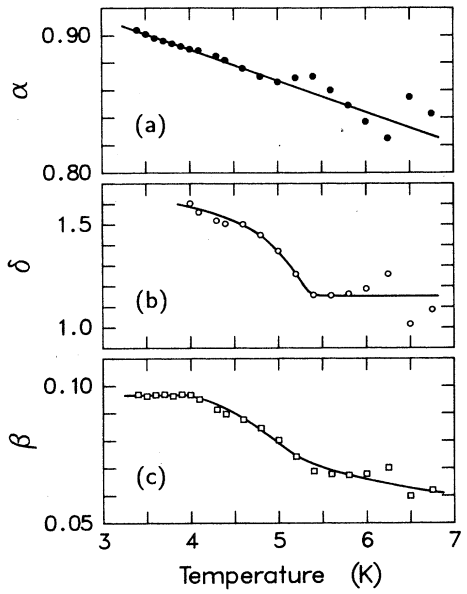

FIG. 3. Temperature dependences of the exponents α, δ, and β occurring in Eqs. (3), (7), and (8), respectively. Solid lines are guides to the eye.

extremely broad distribution of relaxation times persists throughout the entire temperature range studied. Note that for constant $\Delta B/B$ the parameter α should decrease with increasing temperature. Given the output values of χ_S, χ_0, and α, we subsequently extract, for each temperature, τ_c by simultaneous adjustment of Eqs. (4a) and (4b) to the data for, respectively, χ' and χ''. For the temperatures selected in Fig. 2, the result of these fits is shown in Fig. 4 as a function of the frequency. It is evident that the Cole-Cole approach indeed provides an adequate description of the data. The results for τ_c are presented in Fig. 5.

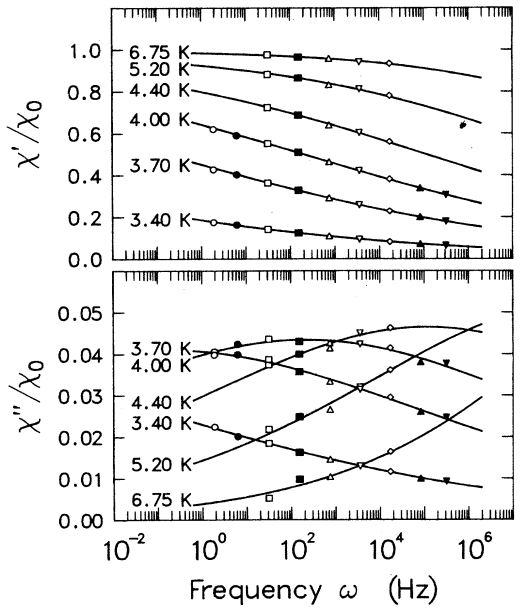

FIG. 4. Frequency dependences of $\chi'(\omega,T)$ and $\chi''(\omega,T)$ for a selection of temperatures. Susceptibilities are normalized to the isothermal susceptibility χ_0. Solid lines are fits of Eqs. (4).

We now turn to an examination of the temperature dependence of the relaxation time τ_c, which constitutes the principal result of the present Cole-Cole analysis of the complex susceptibility. In Fig. 5, it is manifest that the dynamic response of our system slows down by as many as 16 decades in time in the range of temperatures considered. The most likely source for such a dramatic divergence clearly is activated dynamics, and we examine this first. The solid line in Fig. 5, then, represents a fit of $\tau_c=\tau_0\exp[(b/T)^{1+\psi\nu}]$, appropriate to a system with a zero critical temperature [cf. Eq. (1)]. Indeed, the divergence is found to be excellently described ($\chi^2\approx 1$). The fit given in Fig. 5 extends over the temperature range of 3.7–6.8 K, and yields $\psi\nu=2.2\pm0.2$, $\tau_0=(2\pm1)\times10^{-13}$ s, and $b=10.8\pm0.6$ K. Note that the finite value for $\psi\nu$ excludes a simple Arrhenius description, which would correspond to $\psi\nu=0$. The result for $\psi\nu$ will be further discussed below. The result for the microscopic single-spin time τ_0, which corresponds to an exchange energy of order 40 K, is of quite reasonable magnitude, and so is the value for b, which is a measure for the temperature-dependent barrier height B through $B=b^{1+\psi\nu}/T^{\psi\nu}$. Below 3.5 K, the expression for τ_c is apparently inadequate, but for plausible reasons. Here, the relaxation times have become many orders of magnitude longer than the upper bound of the experimental time window ($\sim$1 s). In fact, the range of the agreement may be extended to lower temperatures, albeit by only 0.2 K, by adding "dc" points[2,13] (typical time scale of 30 s) to our set of data. These were deduced from the time-dependent

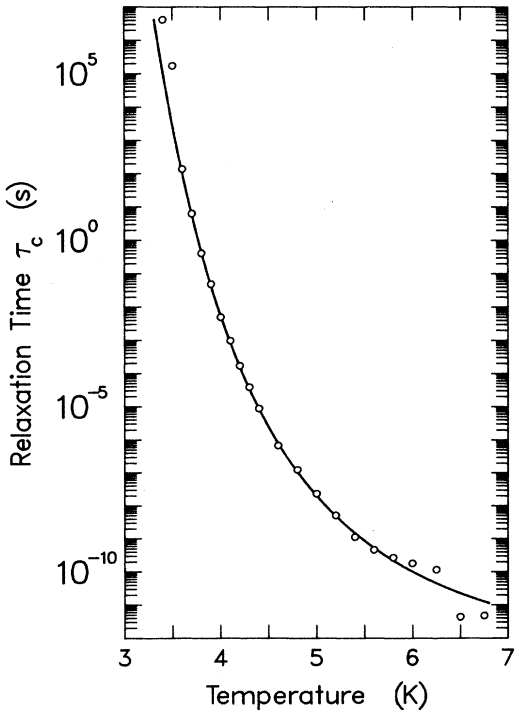

FIG. 5. Temperature dependence of the relaxation time τ_c, derived from the Cole-Cole analysis. Solid line is a fit of Eq. (1).

zero-field-cooled magnetization M in a small field $H=10$ G via the well-known "$\pi/2$ rule," i.e., $\chi'=M/H$ and

$$\chi''=-(\pi/2)d(M/H)/d\ln t \ .$$

Note that τ_c rises so steeply that even extension to time scales of a week would not bring us lower in temperature than, say, another 0.2 K.

To establish the occurrence of activated dynamics in the present SG more firmly, we point out that the data in Fig. 5 definitely are at variance with power-law dynamics as well as a finite T_c. A fit of a power law of the form $\tau_c \propto T^{-z\nu}$, diverging at $T_c=0$, entirely fails ($\chi^2\approx 40$), 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 conceivably be caused by residual coupling among the magnetic layers. The dynamics would then be governed by ordinary critical slowing down, i.e., $\tau_c \propto (T-T_c)^{-z\nu}$. Such a power law appears to yield fits of reasonable quality ($\chi^2=1$–4), but the resultant $z\nu=15\pm1$ is unacceptably large. More importantly, the value of 3.26 ± 0.07 K found for T_c is incompatible with $T_f=2.97\pm0.02$ K measured at a time of 90 s.[2,13] Forcing T_c to lower values drives $z\nu$ to values of over 20, and deteriorates the fit. A finite T_c also markedly worsens the fit in the case of activated dynamics. For instance, when setting τ_0 at the reasonable value of 2×10^{-13} s, activated dynamics with a finite T_c results in $\chi^2=5$ for $T_c=1.0$ K, gradually increasing to $\chi^2=90$ for $T_c=3.0$ K. Note that the inadequacy of the latter fit also rules out the Vogel-Fulcher law,[1] which in our notation corresponds to $\psi\nu=0$ and finite T_c. In summary, therefore, the temperature dependence of the relaxation time is excellently, and uniquely, accounted for in terms of activated dynamics with a vanishing critical temperature.

B. Correlation-function analysis

At this point, it is important to ascertain to what extent this conclusion and the result for $\psi\nu$ depend on the assumptions underlying the Cole-Cole description, more specifically, identifying the median value of a distribution on a logarithmic time scale with the characteristic relaxation time τ_c in Eq. (1). An alternative method to derive the relevant dynamic quantities from the data is to assume a certain form for the spin-correlation function

$$q(t)=\langle S_z(0)S_z(t)\rangle \ ,$$

and next to calculate, by use of the fluctuation-dissipation theorem, the susceptibility from the relation[17]

$$\chi(\omega)=-\chi_0\int_0^\infty \left[e^{-i\omega t}\frac{d}{dt}q(t)\right]dt \ . \tag{6}$$

First, we consider the exponential-logarithmic decay

$$q(t)=\exp\left[-\left[\frac{\ln(t/\tau_0)}{\ln(\tau_c/\tau_0)}\right]^\delta\right] , \tag{7}$$

which is expected to be suitable for cases where thermally activated processes dominate the relaxation, and $\ln t$ rather than t is the appropriate time parameter.[6] From Monte Carlo simulations of the $d=3$ random-field Ising system, for which the dynamics is expected to be closely similar to the $d=2$ Ising SG, this form for $q(t)$ was found to describe the long-time tail of the decay with an exponent $\delta=3$.[18]

The real and imaginary parts of $\chi(\omega)$ obtained from Eqs. (6) and (7) by numerical integration have been fitted, at fixed temperatures, to the data with χ_0, δ, and τ_c as adjustable parameters, while adopting $\tau_0=10^{-13}$ s. The results for $\ln(\tau_c/\tau_0)$ versus the temperature are plotted double-logarithmically in Fig. 6. The resultant temperature dependences of δ and χ_0 are presented in Figs. 3(b) and 7, respectively. The fits appeared to be of a quality comparable to the Cole-Cole fits for temperatures ranging from 5.0 to 7.0 K. In this range, the exponent δ is approximately constant and equal to $\delta=1.15\pm0.05$, indicating that the decay of $q(t)$ is very close to algebraic ($\delta=1$). Accordingly, the decay is not dominated by the relaxation of isolated locally ordered clusters, for which, at long times, Eq. (7) has been derived to hold with $\delta=d/(d-1)=2$.[19] Below 4.8 K, the fits gradually deteriorate, and the value of δ increases to $\delta=1.6$ at 4.0 K. As seen is Fig. 6, in this temperature regime a sharp increase in $\ln(\tau_c/\tau_0)$ is apparent. Upon lowering the temperature further, fits with realistic values for the parameters could not be achieved. For a comparison of the relaxation times derived from the exponential logarithm with those from the Cole-Cole analysis, we have replotted the latter in Fig. 6. Clearly, the exponential-logarithmic decay, as much as the Cole-Cole analysis, establishes activated dynamic behavior [cf. Eq. (1)], although with a slightly different value for the exponent $\psi\nu$. In the tem-

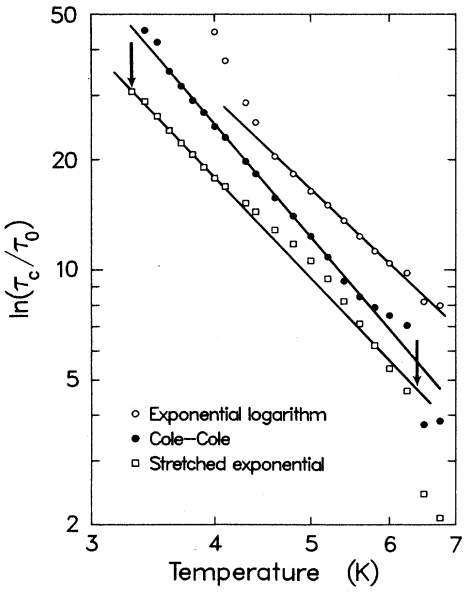

FIG. 6. Double-logarithmic plot of $\ln(\tau_c/\tau_0)$ vs the temperature. Here, τ_c is derived from Eqs. (3), (7), and (8) (open circles, solid circles, and squares, respectively). For clarity, the latter data are shifted downwards by dividing $\ln(\tau_c/\tau_0)$ by 1.35 (cf. arrows). Solid lines represent fits of Eq. (1).

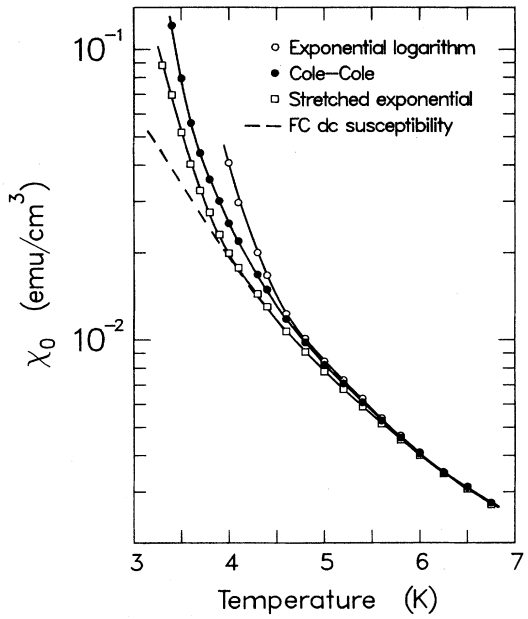

FIG. 7. Temperature dependence of the isothermal susceptibility χ_0, as derived from the various analyses. Solid lines are guides to the eye. The dashed line represents the field-cooled (FC) "dc" susceptibility (Ref. 2).

perature range where δ is approximately constant, a straight line according to Eq. (1) has been fitted to $\ln \ln(\tau_c/\tau_0)$ vs $\ln T$ (Fig. 6) as derived from the exponential-logarithmic decay, to yield $\psi\nu = 1.6 \pm 0.2$.

The second form for $q(t)$ we consider is the stretched exponential, or Kohlrausch, decay

$$q(t) = \exp[-(t/\tau_c)^{\beta}] . \tag{8}$$

This dependence is widely used to describe relaxation processes in SG and disordered media in general. As a case in point, extensive numerical simulations of the $d = 3$ $\pm J$ Ising SG (Ref. 3) have shown that this form excellently represents the behavior of $q(t)$ at long times. Similarly to Eq. (7), Eq. (8) substituted in Eq. (6) has, at fixed temperatures, been adjusted to the measured $\chi(\omega)$ with χ_0, β, and τ_c as parameters. The fits turn out to be of good quality for a substantial range of temperatures. In fact, from 7 down to 3.4 K the fits are essentially of the same quality as the fits of the Cole-Cole expression. In comparison with the exponential-logarithmic decay, the fits have somewhat improved below ≈ 4.6 K ($\chi^2 \approx 1$ compared with $\chi^2 \approx 2$), but they deteriorated above this temperature ($\chi^2 \approx 0.6$ compared with $\chi^2 \approx 0.3$). The temperature dependences of the fitted χ_0 and β are presented in Figs. 7 and 3(c), respectively. The results for $\ln(\tau_c/\tau_0)$ are given in Fig. 6, for the sake of clarity of presentation downshifted as indicated. The relaxation times nearly equal the Cole-Cole results, and thus once more evidence activated dynamics. From the slope of the pertinent solid line in Fig. 6, which represents a least-squares adjustment of Eq. (1), we find $\psi\nu = 1.9 \pm 0.2$.

As to the exponent β, its value appears to increase from $\beta = 0.06$ at 6.75 K to a constant value of $\beta = 0.09$ below 4 K. These very small values again evidence an extremely slow decay of $q(t)$, and sharply contrast both numerical[3] and experimental[4] results in $d = 3$ SG. In the latter, β typically varies from 0.3 just above the SG transition temperature to 1 at the Curie temperature of the nonrandom Ising system. For the present system, nonexponential decay of the correlation function may be expected below temperatures as high as $T \approx 100$ K, the transition temperature of Rb_2CoF_4.[20] To understand the *decrease* of β with temperature it is necessary to realize that in the event of extremely slow decays only a very small fraction of the total decay of $q(t)$ is actually probed. Although this fraction, which is determined by the experimental time window (10^{-5}–1 s), can be accurately described by Eg. (8), $q(t)$ viewed over a longer time interval may be functionally different. An increase of the temperature, in fact, results in shifting the part of the decay of $q(t)$ probed to times long compared with τ_c. The concommittant decrease of the fitted exponent β then shows that the functional dependence of $q(t)$ drops slower than according to Eq. (8) with fixed β. Apparently, as was shown above, the long-time tail of $q(t)$ is better represented by the exponential-logarithmic decay, Eq. (7). According to the same reasoning, the near constancy of β below 4 K suggests that $q(t)$ is quite well described by the stretched-exponential form for $\ln t \lesssim \ln \tau_c$. The two time decays of $q(t)$ considered thus seem to apply best to different temperature regimes, but in a reversed order compared to $d = 3$ SG.[3] These considerations, however, do not detract from the conclusion that the associated temperature dependences of τ_c establish activated dynamic behavior, and support the identification of τ_c in the Cole-Cole equation as the characteristic relaxation time of the $d = 2$ SG. Further, all deduced values for $\psi\nu$ are mutually consistent with an average of $\psi\nu = 1.9 \pm 0.3$.

C. Relaxation-time distribution

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 the Cole-Cole equation, Eq. (3). We have

$$g(\tau) = \frac{1}{2\pi} \frac{\sin(\alpha\pi)}{\cosh[(1-\alpha)\ln(\tau/\tau_c)] - \cos(\alpha\pi)} , \tag{9}$$

which is normalized according to $\int_{-\infty}^{+\infty} g(\tau) d \ln\tau = 1$. In Fig. 8 some representative distributions are shown. The salient point here is that the profiles $g(\tau)$, which on a logarithmic scale are symmetric about τ_c, turn out to be extremely broad, even at temperatures substantially above T_f. In fact, at 5.8 K the distribution spans as many as 10 decades in time (full width at half maximum), spreading by another 5 decades when going to 3.4 K. Meanwhile, the distribution is seen to shift as a whole towards larger times over a distance significantly beyond its width.

It is of interest to compare the Cole-Cole distribution with the distribution of relaxation times $g'(\tau)$ associated with the correlation function $q(t)$. This distribution may be defined by the relation

$$q(t) = \int_0^{\infty} g'(\tau) \exp(-t/\tau) d \ln\tau . \tag{10}$$

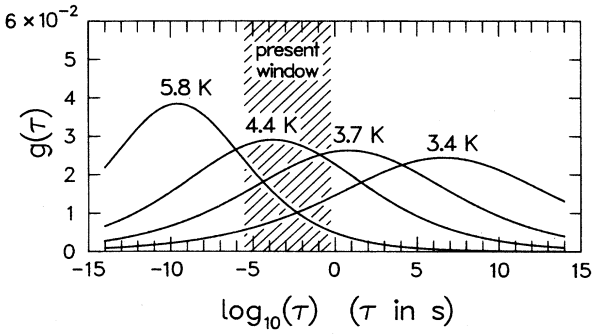

FIG. 8. Distribution of relaxation times $g(\tau)$ according to Eq. (9) for a selection of temperatures.

For a stretched-exponential decay [Eq. (8)], with small values of β, i.e., $\beta|\ln(t/\tau_c)| \ll 1$, the inverse Laplace transform of $q(t)$ can be written as the series expansion[17]

$$g'(\tau)=\beta\exp[-\tfrac{1}{2}\beta^2\ln^2(\tau/\tau_c)-1][1+O(\beta^2)] , \quad (11)$$

which, to leading order, is a Gaussian distribution on a $\ln\tau$ scale. Apparently, in the present case the Cole-Cole and stretched-exponential descriptions are closely related and differ only with respect to the precise shape of their otherwise symmetric logarithmic distributions. The Cole-Cole distribution carries more weight in its wings, which affects the decay of the associated correlation function at long times. In fact, in this limit the Cole-Cole $q(t)$ reduces to the simple algebraic form $q(t)\approx t^{\alpha-1}$, which is close to the exponential-logarithmic decay [Eq. (7)] for $\delta\approx 1$.

D. Isothermal susceptibility

The temperature dependences of the isothermal susceptibility χ_0 as derived with the above three fitting procedures are presented in Fig. 7. The dependences appear to be quite similar, except for a gradual disparity below ≈ 5 K. Above this temperature, the χ_0 found further coincide with the results from the dc susceptibility (dashed line in Fig. 7).[2] A monotonic increase with decreasing temperature is observed which is much faster than the simple Curie law $\chi \propto 1/T$ anticipated for a $d=2$ SG with a symmetric distribution of random exchange couplings. Apparently the susceptibility is enhanced by the presence of short-range ferromagnetic correlations. Predominantly ferromagnetic, or for that matter antiferromagnetic, correlations are indeed expected to develop in any SG system exhibiting an asymmetric distribution of exchange interactions, even if the interactions average out to zero. In $Rb_2Cu_{0.782}Co_{0.218}F_4$, the antiferromagnetic J's are -37 and -90 K, while the ferromagnetic J's are typically 20 K.[15] Nevertheless, this system forms a SG because the concentrations of Cu and Co are such that the weaker ferromagnetic bonds are more abundant. When cooling, at first short-range antiferromagnetic order develops, followed by short-range ferromagnetic order upon further cooling. Accordingly, the susceptibility initially drops below the Curie law, but rises above it at lower temperatures. Ultimately, the growth of the correlations is, of course, limited by the SG correlation length, so that the susceptibility approaches the Curie law. In the present system, χ_0 is antiferromagnetically suppressed down to ≈ 15 K, below which temperature it starts catching up, crossing the Curie law at ≈ 7.5 K. More precisely, from 7 down to 3.5 K χ_0 from the Cole-Cole analysis follows the phenomenological quasicritical power-law divergence $\chi_0 \propto (T-T^*)^{-\gamma}$ with $\gamma=1.67$ and $T^*=2.99$ K. Similarly, χ_0 from the stretched exponential yields $\gamma=1.68$ and $T^*=2.80$ K. These results for γ conform essentially with the $d=2$ Ising value $\gamma=1.75$. The ultimate crossover to a Curie law is not observed, presumably because below 3.5 K χ_0 escapes determination with the present experimental time window.

IV. DYNAMIC SCALING

An alternative analysis of the data, which extends to lower temperatures and yields additional information on the critical exponents, is based on scaling the susceptibility in the frequency domain according to activated dynamics. 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 $\ln(\omega\tau_0)/\xi^{\psi}$,[8] and an activated dynamic scaling form for $\chi''(\omega,T)$ then follows by use of the fluctuation-dissipation theorem, which connects the correlation function to χ''. We thus have

$$\chi'' T^{-p}=\mathcal{F}[-\ln(\omega\tau_0)T^q] , \quad (12)$$

in which $\mathcal{F}$ is a scaling function, and

$$p\equiv -1-\nu(2-\eta-\psi)=-1-\gamma+\psi\nu ,$$

$q\equiv\psi\nu$, and τ_0 are scaling parameters; γ and η are critical exponents in the usual notation. In the derivation of Eq. (12) it is assumed that the relaxation spectrum is a slowly varying function of ω, a condition that is amply met in $Rb_2Cu_{0.782}Co_{0.218}F_4$.

In Fig. 9, 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 data of Fig. 1(b) have been included. The frequency $\omega/2\pi$ thus runs from 0.3 to 50 000 Hz, while T varies from 1 to approximately 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. The maximum of the scaling function corresponds to $\omega\tau_c\approx 1$, with τ_c at the pertinent temperature taken from the Cole-Cole analysis. The right-hand flank is associated with $\omega\tau_c<1$, or higher temperatures, and conversely the left-hand side with $\omega\tau_c>1$. The result for $q=\psi\nu$ and τ_0 are in good agreement with the corresponding results of the relaxation time analyses of the previous section, which is gratifying in that it demonstrates consistency of the two approaches. The results for p and $q=\psi\nu$, when substituted into the relation $p=-1-\gamma+\psi\nu$, yield for the critical exponent of the order-parameter susceptibilty $\gamma=4.2\pm0.6$, which excellently compares with the result $\gamma=4.5\pm0.2$ deduced from the static nonlinear susceptibility.[2]

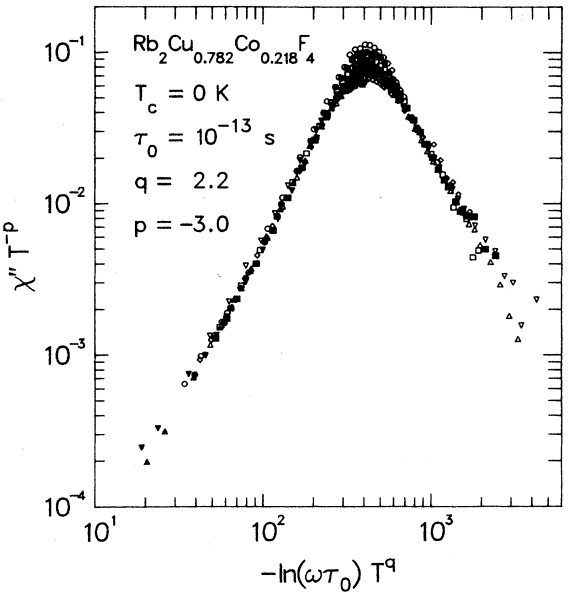

FIG. 9. Activated-dynamic-scaling plot of the $\chi''(\omega,T)$ data for temperatures from 1 to 7 K and frequencies from 0.3 Hz to 50 kHz. Symbols correspond to the frequencies in Fig. 1.

A somewhat surprising conclusion deduced from Fig. 9 is that, apart from deviations near $\omega\tau_c \approx 1$, 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. In this context we recall that the system would only attain equilibrium if it were left to relax for a time exceeding the longest relaxation time present in the spectrum. Although this condition is evidently violated in the low-temperature regime, the time to take a data point, of order 10^2 s in the present experiment, is much larger than the experimental time ω^{-1}, and consequently the susceptibility will, at the relevant ω, probe fluctuations that closely resemble those of true equilibrium.

V. CONCLUDING REMARKS

The results from the Cole-Cole analysis, the frequency transforms of the dynamic correlation functions, and dynamic scaling all confirm the physical concepts underlying the theory of activated dynamics in SG: The dynamics is governed by thermally activated relaxation over barriers B that are distributed in height with a spread $\Delta B \sim B$, and diverge with decreasing temperature as some power of the correlation length. That $\Delta B/B$ indeed is of order unity at all temperatures may be verified from the relaxation-time spectra obtained with the Cole-Cole analysis by converting them to distributions of barrier heights by use of Eq. (1). The temperature dependence of $g(\tau)$ observed in $Rb_2Cu_{0.782}Co_{0.218}F_4$ is probably exemplary for the spectrum of relaxation times in the $d=2$ Ising SG. Indeed, it sharply contrasts, at least in a qualitative way, with the results available for $d=3$ SG above T_c.[21,22] In the latter case, upon approaching T_c from above, $g(\tau)$ suddenly expands toward dc time scales by developing a long-time tail rather than a shift of the entire distribution, while the time corresponding to the maximum in $g(\tau)$, although increasing, remains short ($<10^{-5}$ s).

The reason for the success of the Cole-Cole description for this $d=2$ SG has become apparent from the analyses based on the decay of the dynamic correlations. At short times ($\ln t \lesssim \ln \tau_c$) the decay is well described by a stretched exponential with a small exponent ($\beta \approx 0.09$), while at long times ($\ln t \gtrsim \ln \tau_c$) the decay is closer to algebraic. Both features are contained in the Cole-Cole equation with α close to unity. Below 3.4 K, the relaxation-time spectrum has, as a whole, shifted to such long times that analyses based on the determination of a characteristic relaxation time are rendered unfeasible. But here, dynamic scaling, directly probing fluctuations in the "short"-time tail of the distribution, still allows a determination of $\psi\nu$. The value of $\psi\nu$ depends slightly on the way the data are analyzed. From the various methods of analysis an averaged value of $\psi\nu = 2.0 \pm 0.3$ is found.

We finally derive a complete set of critical exponents for $Rb_2Cu_{0.782}Co_{0.218}F_4$. Combining the present result $\gamma = 4.2 \pm 0.6$ with $\gamma = 4.5 \pm 0.2$ from the static susceptibility,[2] we arrive at $\gamma = 4.4 \pm 0.2$. This value favors a comparison of the present system with the $\pm J$ model for $d=2$ Ising SG, for which estimates of γ range from 4 to 5.3, rather than the Gaussian model for $d=2$ Ising SG, which leads to $\gamma \approx 7$.[23] 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 finite.[15] From the scaling relation $\gamma = \nu(2-\eta)$ we then deduce $\nu = 2.4 \pm 0.3$ upon adopting for η a small positive value ($\eta = 0.2 \pm 0.2$), as appropriate to the $d=2$ $\pm J$ Ising SG.[7] This implies $\theta = -1/\nu = -0.41 \pm 0.05$. Finally, by use of our result $\psi\nu = 2.0 \pm 0.3$ it follows that $\psi = 0.8 \pm 0.2$, which agrees with the upper limit $\psi \leq d-1$ set by theory.[6,8,11] A recent rigorous numerical calculation of the energy barriers in $d=2$ systems infers $\psi = 1$.[24]

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