Aging in two-dimensional Ising spin glasses

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Aging of two-dimensional (2D) Ising spin glasses with short-range interactions $Rb_2Cu_{1-x}Co_xF_4$ ($T_c = 0$) is studied in the critical regime via the low-frequency ac susceptibility and via the dc magnetization for $x = 0.22$ and 0.33. After quenching to low temperatures, slow logarithmic relaxation is observed for both the in-phase and out-of-phase ac susceptibilities. Using the droplet scaling model, the spin-glass correlation length R is concluded to increase after a temperature quench as $R \propto (\ln t)^{1/\psi}$, with t in units of some microscopic time. The exponent $\psi = 1.0 \pm 0.1$ compares with recent theoretical results and numerical simulations for 2D short-range Ising spin glasses. The waiting-time dependence found for the field-cooled and zero-field-cooled dc magnetizations points to a markedly slower crossover from the quasiequilibrium to the nonequilibrium regime than in 3D. From experiments with different cooling histories, finally, the equilibrium spin-glass state is inferred to be strongly dependent on the temperature.

I. INTRODUCTION

Aging is an intriguing phenomenon intrinsic to random systems. After a system has been left in a nonequilibrium state, it manifests itself under stable external conditions in a reduction of the response to an external probe over long times. Since its discovery in spin glasses,[1] aging has become a major field of interest in random magnetic systems. In three-dimensional (3D) spin glasses aging is usually observed after a quench to a temperature below the transition temperature,[1–5] where the dynamics is slowed down as a consequence of the frustration of the magnetic interactions. In 2D, no spin-glass ordered phase exists at finite temperatures.[6–10] At low enough temperatures, however, the 2D spin-glass correlation length exceeds the typical length scale probed in the experiment. A 2D spin glass in the regime not too far above $T_c = 0$ is therefore anticipated to behave in a way similar to an ordered spin glass. Indeed, aging has been reported for the 2D spin glasses $Rb_2Cu_{0.78}Co_{0.22}F_4$ (Ref. 11) and $Rb_2Cu_{0.67}Co_{0.33}F_4$ (Ref. 12) as well as in thin films of the spin glass CuMn.[13] In the latter, aging was found to show qualitatively the same behavior as in 3D spin glasses. The results in $Rb_2Cu_{0.78}Co_{0.22}F_4$ and $Rb_2Cu_{0.67}Co_{0.33}F_4$, by contrast, indicate that aging in 2D Ising spin glasses markedly differs from the 3D case. In this paper, we present a more comprehensive study of aging in $Rb_2Cu_{1-x}Co_xF_4$, which for $0.18 < x < 0.40$ has proved to be a nearly perfect realization of a 2D Ising spin glass with random short-range Ising interactions.[10] A principal objective of the present study is to explore the aging mechanism of a system that has no ordered phase at finite temperatures.

Microscopic treatment of aging requires an understanding of the ground state, which in spin glasses self-evidently is of a very complex nature. No analytical solution is available for spin glasses with short-ranged interactions in 2D or 3D, and so one has to rely on more phenomenological models. These are based on spin-glass ordered domains[14,15] or on phase-space hierarchical relaxation.[16,17] Particularly suited to describe the domain growth in ordered Ising spin glasses is the treatment by Fisher and Huse[14] based on droplet scaling.[18–20] The basic idea is that the system progresses from one nonequilibrium state to the next, where each state is composed of "droplet" excitations, i.e., low-energy domains made up of spins that are collectively turned over with respect to the ground state. The droplets have free energies which scale with their linear size L as $E \sim \gamma L^{\theta}$, where γ is a measure of the magnetic stiffness, and θ is an exponent depending on the dimensionality. In 3D, $\theta \approx 0.2$. The fundamental long-time nonequilibrium process is assumed to be thermally activated growth of spin-glass ordered domains. This involves surmounting the activation barriers B associated with the creation and annihilation of the droplets. These barriers are taken to scale with the linear size L as $B \sim L^{\psi}$, where ψ is an exponent satisfying $\theta \leq \psi \leq d - 1$, with d the dimensionality.

For 2D, the exponent θ is negative [$\theta \approx -0.25$ (Ref. 21)], which correctly implies that excitations of arbitrary low energy destroy the spin-glass order at finite temperatures, i.e., $T_c = 0$. The equilibrium correlation length ξ diverges towards zero temperature according to $\xi \sim T^{-\nu}$, with $\nu = 1/|\theta|$, as determined from the fact that the free energy is of order k_BT. A necessary condition for the observation of aging phenomena in 2D is that the domain

dynamics is probed at length scales shorter than ξ, i.e., at sufficiently short-time scales. Since ξ is substantial in the 2D case if the temperature is sufficiently low, there is a range of temperatures in the critical regime for which the condition is fulfilled despite the failure to order. The droplet theory, which was originally developed for the spin-glass ordered phase, thus applies to the 2D case as well. In a recent paper,[22] it was shown that the lowest-lying excitations in the critical regime are a continuation of the lowest-lying excitations in the spin-glass ordered phase. This forms the theoretical basis for application of the droplet theory of Ref. 14 to the 2D spin glass.

The paper is organized as follows. We first present measurements on the relaxation to equilibrium of the ac susceptibility after a quench in temperature has left the spin glass in a nonequilibrium state. In interpreting the results, advantage is taken of the droplet scaling model,[14] and it is thereby shown that aging reflects the growth of the spin-glass correlation length by thermally activated dynamics. Second, we present the results of dc-susceptibility measurements taken with the field-cooled (FC) or zero-field-cooled (ZFC) procedure. The noteworthy conclusion here is that the crossover from equilibrium to nonequilibrium is slower than in the 3D case. Third, we consider the effect of aging at a certain temperature on the subsequent aging at a lower measuring temperature. The results are compared with those of experiments in 3D, where it was shown that the equilibrium correlations below T_c are strongly sensitive to temperature.[23,24]

II. EXPERIMENTAL DETAILS

The samples studied are single crystals of $Rb_2Cu_{1-x}Co_xF_4$, which is a random mixture of the archetypal square-lattice antiferromagnet Rb_2CoF_2 (layered K_2NiF_4 structure, effective spin $S = \frac{1}{2}$, $T_N = 103.0$ K), and the isostructural ferromagnet Rb_2CuF_2 ($S = \frac{1}{2}$, $T_c = 6.05$ K). Recently, the (x,T) diagram has been determined over the whole range of x.[10] For $0 < x < 0.18$, at the Cu-rich side, the system is ferromagnetic, while it is antiferromagnetic for $x > 0.40$. In between, for $0.18 < x < 0.40$, the system has been shown to be a nearly ideal realization of the $d = 2$ Ising spin glass with random nearest-neighbor bonds.[10] Samples with two different concentrations, $x = 0.22$ and 0.33, which together are representative of the whole spin-glass phase, have been used for the experiments. For $x = 0.22$, activated dynamics with a zero critical temperature was established by an analysis of the divergence of the median relaxation time above the freezing temperature,[25] which is defined as the temperature at which the in-phase susceptibility is maximum. This temperature depends on the frequency, and equals 3.5 K at 100 Hz. The sample with $x = 0.33$ is closer to the antiferromagnetic end of the (x,T) diagram, and the bond strengths nearly average out to zero. It was found to yield essentially the same freezing temperature and critical behavior.

Aging was, on the one hand, observed through the response of the system to a dc field applied from a variable waiting time t_w after a temperature quench in zero field (ZFC procedure). An equivalent dc method is cooling the sample in a field, removing the field after a waiting time, and recording the relaxing magnetization (FC procedure). Alternatively, an ac field was applied, and aging was observed through the relaxation of the ac susceptibility versus the time.

In the experiments on the $x = 0.22$ sample (performed in Saclay), the sample was positioned in one end of an astatic superconducting pickup coil connected to a rf superconducting quantum interference device (SQUID) (CEA LETI, Grenoble). The ac susceptibility was measured vs the time at frequencies down to 0.01 Hz. The ac probe field amounted to 6 mG peak to peak, and the dc background was less than 0.5 mG. The in-phase and out-of-phase components of the fundamental harmonic of the signal were simultaneously recorded by use of a spectrum analyzer. At the low frequencies used, the phase shift by the coils is negligible, and so the driving voltage could be used directly as a zero-phase reference. In the case of the dc measurements, the output voltage of the SQUID was sampled at regular time intervals by the use of a computer-controlled amplitude-to-digital converter. The sample was mounted in the cryostat in a vacuum chamber on a post made of polychlorotetrafluoroethylene, which at the top end was thermally connected to a heat sink held at 1.5 K. The temperature was servo stabilized to within 5 mK over long time intervals ($> 10^4$ s) by resistive heating of the post with reference to a calibrated carbon-glass resistance thermometer. The susceptibility measurements were calibrated with reference to a standard paramagnetic sample. At the end of each run, the zero-susceptibility signal, which was subtracted, was determined after lifting the sample out of the coils.

In the case of the data for $x = 0.33$ (taken in Utrecht), the sample was mounted in the cryostat within a double-walled glass tube filled with exchange gas. This assembly was positioned in one end of an astatic superconducting pickup coil connected to a dc SQUID (SHE). The measurement of the signal associated with the ac-susceptibility detection was similar to that with the rf SQUID above. The probe field was 0.1 G and the background less than 1 mG. For the dc ZFC measurements, the voltage output of the SQUID was measured with a Keithley 194A high-speed voltmeter. Some runs for the $x = 0.22$ sample were repeated with the dc SQUID to confirm the calibrations.

III. ac SUSCEPTIBILITY

The ac-susceptibility experiments were performed in the frequency range of 0.01–56 Hz after rapid cooling of the sample from 8 K or above, where the system is in equilibrium. The cooling times achieved were between 30 and 100 s, depending on the final temperature. Three regimes can be distinguished for the final temperature. The $x = 0.22$ spin glass exhibits only an in-phase susceptibility χ' for final temperatures above 7 K. For final temperatures between 7 and 3.5 K, the out-of-phase susceptibility χ'' rises gradually towards lower temperature, but χ' and χ'' both are independent of the time elapsed

since cooling. Below 3.5 K aging becomes manifest from a decrease of the ac susceptibility with time. In the case of the $x = 0.33$ spin glass, where $\Delta J/|J_{av}|$ is larger and the distribution of relaxation times is broader, aging effects are manifest at temperatures as high as 5 K, which is significantly larger than the freezing temperature of 3.5 K.

Typical sets of ac-susceptibility data for the $x = 0.22$ and 0.33 samples are presented semilogarithmically vs the time in Figs. 1 and 2, respectively. The data for $x = 0.22$ were taken after a quench in 45 s from 9 K to 3.2 K, which is close to the freezing temperature of 3.5 K. The measuring frequencies were selected between 0.01 and 10 Hz. The data for the $x = 0.33$ sample were taken at 3.7 and 4.2 K, again after a quench from 9 K, at frequencies between 0.14 and 56 Hz.[26] For both $x = 0.22$ and $x = 0.33$, χ' and χ'' are seen to decrease slowly with time in a logarithmic way. A slight curvature is however discernible in these decays, in particular at the lower frequencies, which indicates that at these frequencies equilibrium is not attained until after very long times. Anticipating the analysis below, we note that the values of χ'' extrapolated to infinite time, χ''_{eq}, are nearly independent of the frequency in the frequency range studied. Such a weak frequency dependence of χ''_{eq} is indicative of an extremely broad distribution of relaxation times.[27] As concerns the frequency dependences of the decay times, the latter increase towards lower frequencies for both χ' and χ'', again in logarithmic way. These frequency dependences are much weaker than in the 3D case, for which the decay time to equilibrium was found to scale roughly

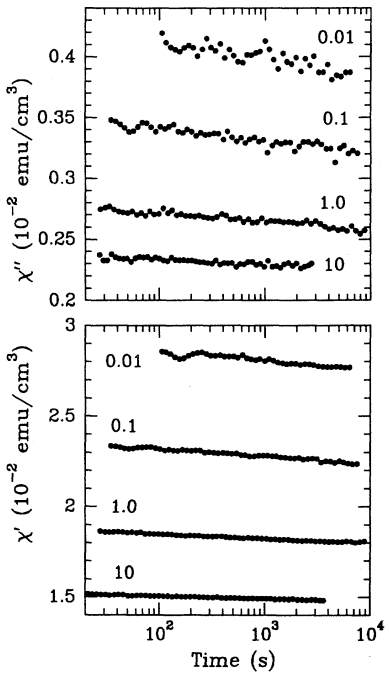

FIG. 1. Relaxation of the in-phase (χ') and out-of-phase (χ'') ac susceptibilities in $Rb_2Cu_{0.78}Co_{0.22}F_4$ after a quench in temperature from 9 to 3.2 K within 45 s. Labels denote the frequency in Hz.

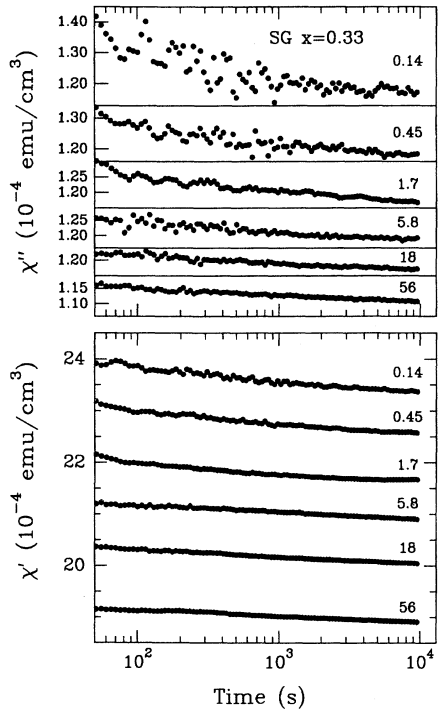

FIG. 2. Same as Fig. 1, but after a temperature quench to 3.7 K in $Rb_2Cu_{0.67}Co_{0.33}F_4$.

algebraically with the frequency, i.e., according to $\omega^{-\alpha}$ with $\alpha \approx 1$ (Refs. 1, 28) or $\alpha \approx 0.25$ (Ref. 5). Similar logarithmic dependences of χ' and χ'' have been observed at other temperatures. The slopes of χ' and χ'' vs $\log_{10}t$, however, diminish as the temperature increases. At the lowest temperatures, where the dynamics is very far from equilibrium, no flattening out of the curves is seen within the experimental time window.[11]

To relate the relaxation of the susceptibility to the domain size at time t, we rely on the results of the droplet scaling theory as developed in Ref. 14. After a quench in temperature, the system is thought to be left in a nonequilibrium state made up of ordered spin-glass domains of small size. Correlations subsequently start to develop, and the larger domains will grow at the expense of smaller ones, presumably by a collective turnover of the spins making up a smaller domain. At the end, the smaller domains thus have disappeared from the system. The driving force of this long-time relaxation towards equilibrium is assumed to be thermal activation. The relaxation time τ associated with surmounting an energy barrier of height B needed to turn over a droplet amounts to

$$\tau = \tau_0 \exp(B/k_B T) , \qquad (1)$$

in which T is the temperature and τ_0 denotes a microscopic attempt time of order 10^{-12} s. We recall at this point that $B \propto L^{\psi}$, where L is the linear size of the droplet. Because all droplets having $\tau \ll t$ are in equilibrium at a time t after the quench, the correlations extend over a distance

$$R(t) \propto [T \ln(t/\tau_0)]^{1/\psi} . \quad (2)$$

At a given temperature, the correlations are thus seen to grow as with power law of $\ln(t/\tau_0)$.

Quite similarly, when applying a probe field with an angular frequency ω, one measures the system at the length scale L_ω corresponding to times $t = 1/\omega$. With the definition $\omega_0 = 1/\tau_0$, L_ω is determined by

$$L_\omega \propto [T|\ln(\omega/\omega_0)|]^{1/\psi} . \quad (3)$$

Because the elapsed time $t \gg 1/\omega$, we have $L_\omega < R(t)$. The droplet excitations of length scale L_ω associated with the probe field have lower energies if their walls to some extent coincide with the domain walls of the larger domains of scale R. This may effectively be described by a reduction of the stiffness γ, which, of course, is proportional to the density of the walls of the $R(t)$ domains. On the basis of scaling arguments,[14] the reduction of the stiffness, $\Delta\gamma$, is expected to be a function of $L_\omega/R(t)$. In the event $L_\omega/R(t) \ll 1$, we have

$$\Delta\gamma \propto \left(\frac{L_\omega}{R(t)}\right)^{d-\theta} . \quad (4)$$

Here, d is the dimension of the magnetic lattice, and θ is the exponent relating the energy of the droplets to their size. It is noted that in general the magnitudes of the aging effect on the real and the dissipative parts of the susceptibility need not be equal.

The susceptibility is inversely proportional to the effective stiffness γ. A reduction in the stiffness, therefore, immediately affects the measured ac susceptibility χ' according to

$$\chi' = \chi'_{\rm eq}\left(1 - \frac{\Delta\gamma}{\gamma_{\rm eq}}\right)^{-1} , \quad (5)$$

in which $\Delta\gamma$ vanishes at large time scales. Equation (5) is equivalent to

$$\frac{\chi' - \chi'_{\rm eq}}{\chi'} = c'\left(\frac{|\ln(\omega/\omega_0)|}{\ln(t/\tau_0)}\right)^{(d-\theta)/\psi} , \quad (6)$$

where we have substituted Eqs. (2)–(4). A similar expression, of course, holds for χ''. The constants c' and c'' are independent of time and frequency. Equation (6) indeed represents a logarithmic dependence of the susceptibility on time and frequency in conformity with the present data for 2D spin glasses.

In order to investigate the validity of Eq. (6) for the 2D Ising spin glass in more detail, we have plotted in a single log-log graph the quantities $(\chi' - \chi'_{\rm eq})/\chi'$ and $(\chi'' - \chi''_{\rm eq})/\chi''$ for the various measuring ω against $|\ln(\omega/\omega_0)|/\ln(t/\tau_0)$. In Fig. 3 this is done for the $x = 0.22$ crystal at 3.2 K, and in Fig. 4 for the $x = 0.33$ crystal at 3.7 and 4.2 K. The equilibrium values $\chi'_{\rm eq}$ and $\chi''_{\rm eq}$ were adjusted for each ω so as to achieve the best scaling. After this procedure, all six data sets are seen to merge into straight lines, where the data for the various ω cover different parts of the line. The scaling plots Figs. 3 and 4 agree well with the the droplet scaling theory, which is a principal result of this investigation: (i) Both χ' and χ'' show the same functional dependence, which substantiates the idea that aging is associated with a slow increase in the stiffness towards its equilibrium value. (ii) The susceptibility data, which are taken over nearly three decades in time and frequency, fall on a single line. This confirms that measurements at time t, on the one hand, and at angular frequency $\omega = 1/t$, on the other, probe the same length scale, such as is expressed by Eqs. (2) and (3). (iii) In more detail, the functional dependence of the susceptibility on $|\ln(\omega/\omega_0)|/\ln(t/\tau_0)$ is recovered,

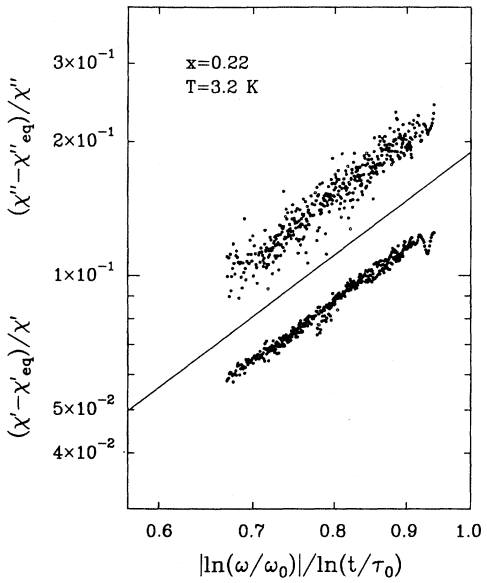

FIG. 3. Activated-dynamic scaling plot of the relaxation of the in-phase and out-of-phase susceptibilities in $Rb_2Cu_{0.78}Co_{0.22}F_4$ after a temperature quench to 3.2 K.

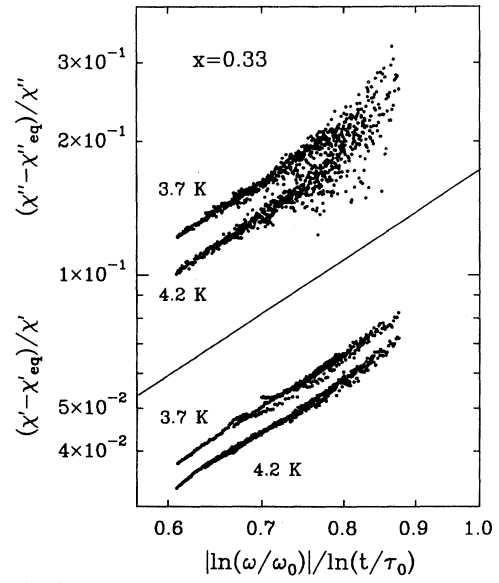

FIG. 4. Same as Fig. 3, but after temperature quenches to 3.7 and 4.2 K in $Rb_2Cu_{0.67}Co_{0.33}F_4$.

especially for the longer times and the higher frequencies. As for the fitted values of χ'_{eq} and χ''_{eq}, they depend monotonically on the frequency, as one would expect, and vary only weakly ($\lesssim 20\%$) within the limited frequency range. The quantity χ'_{eq} decreases with the frequency according to $\chi'_{eq} \propto \omega^{-0.03\pm0.01}$; χ''_{eq} increases with ω, and is equally well represented by $\chi''_{eq} \propto \omega^{0.04\pm0.01}$ and $\chi''_{eq} \propto |\ln(\omega/\omega_0)|^{-1.1\pm0.3}$. The latter form is in agreement with the droplet scaling theory applied to the 2D spin glass.[20]

We finally derive a value for ψ. According to Eq. (6) the slopes of the straight lines in Figs. 3 and 4 equal $(d-\theta)/\psi$. Adopting for θ the theoretical value -0.25, setting $d=2$ for the dimensionality, and taking the reasonable microscopic time of 2×10^{-13} for $\tau_0 = 1/\omega_0$, we obtain $\psi = 0.93 \pm 0.10$ for $x = 0.22$ and $\psi = 1.06 \pm 0.10$ for $x = 0.33$. On the average, we find $\psi = 1.0 \pm 0.1$ with inclusion of the uncertainty in τ_0. It is noted that scaling can be accomplished for a wide range of τ_0. The data can in fact be brought to coincidence for τ_0's as long as $\tau_0 = 10^{-9}$ s, which would have yielded the slightly higher value $\psi \approx 1.4$. Such a long τ_0 is, however, unphysical. The resulting ψ is in good accord with the previous result $\psi = 0.9 \pm 0.2$ from activated dynamics above the freezing temperature,[25] and with the theoretical upper limit $d-1$ suggested by computer simulations.[14,29]

IV. dc MAGNETIZATION

In the dc experiments, the sample was cooled down under the same conditions as in the ac-susceptibility measurements. For the $x = 0.22$ sample, the final temperatures ranged from 2.1 to 4 K, and the waiting times t_w ranged from 100 to 8100 s. Similarly to the ac measurements, three temperature regimes can be distinguished. Above $\sim$ 7 K, $\chi_{dc} = M/H$ is independent of the time. Below this temperature, χ_{dc} was found to relax, but a dependence on t_w, reflecting aging, is not observed until below 3.5 K. Figure 5 gives representative examples of the ZFC results at 2.5 and 3.0 K. Aging is apparent from the observed vertical displacement of the ZFC susceptibility curves as a function of t_w. It is emphasized that the form of these χ_{dc} vs $\log_{10}t$ curves in the regime of aging as well as their dependence on t_w are markedly different from what has been observed for 3D spin glasses.[4,30,31] In the latter, plots of χ_{dc} vs $\log_{10}t$ exhibit an inflection at times of order t_w, largely independent of temperature. In the present 2D system, by contrast, only a weak inflection is observed at temperatures above approximately 2.8 K. The point of inflection moves towards longer times for longer t_w and appears to be strongly dependent on the temperature.

In the quantitative analysis of the ZFC data, first the time t_{infl} at which the inflection occurs was extracted. More precisely, t_{infl} is defined as the time at which $d\chi_{dc}/d\log_{10}t$ is maximum. The results for $x = 0.22$ have been plotted vs t_w in Fig. 6. Within errors, linear dependences are found at temperatures above approximately 2.8 K, however with a slope that is strongly dependent on the temperature. Note that, in contrast

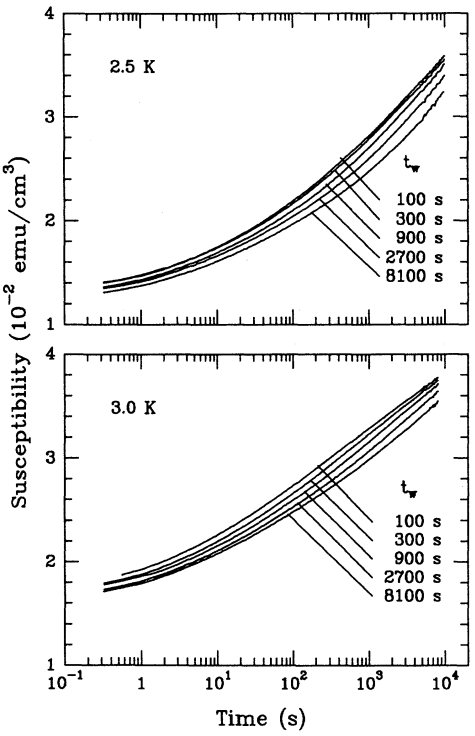

FIG. 5. ZFC susceptibility in $Rb_2Cu_{0.78}Co_{0.22}F_4$ vs the time for various waiting times following a temperature quench from 8 to 2.5 K (upper frame) and to 3.0 K (lower frame).

to 3D spin glasses, the slope equals unity only at about 3.0 K, and at temperatures above approximately 3.5 K has dropped to zero. In other terms, with increasing temperature the equilibrium correlation length becomes smaller than the length scale of the experiment, so that aging phenomena become indistinguishable. In Fig. 7, we

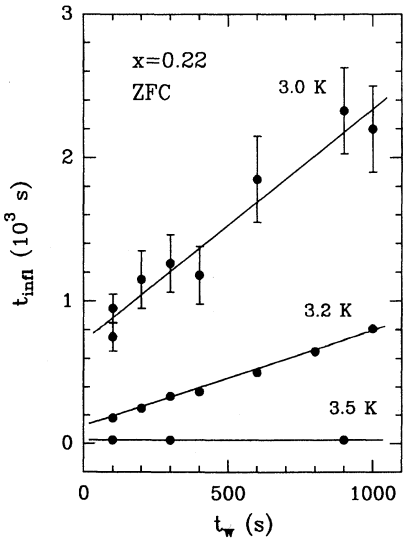

FIG. 6. Dependence of the inflection point of the ZFC χ_{dc}-vs-$\log_{10}t$ curves on the waiting time before applying the field in $Rb_2Cu_{0.78}Co_{0.22}F_4$ at various temperatures, as indicated. The solid lines are least-squares fitted linear dependences.

present similar results derived from the data for $x = 0.33$ at $T = 4.2$ K obtained with the FC technique. For the shorter t_w, the dependence of $t_{\rm infl}$ is again linear, with a slope smaller than unity. For t_w's approaching 10^4 s, however, $t_{\rm infl}$ is observed to saturate; i.e., aging ceases. This is exactly what is expected because the experimental length scale $R(t_w)$ becomes of the same order of magnitude as the finite equilibrium correlation length. We note that a saturation of aging has not yet been reported in spin glasses. The result is however reminiscent of the long-time dynamics in a network of charge-density-wave dislocations,[32] which can be interpreted as to exhibit a zero critical temperature.

In Fig. 5, the ZFC curves do not to converge for short observation times, which implies that even for the longest t_w the conditions of quasiequilibrium are not reached. This is again quite different from the 3D case,[4,30] for which the relaxation curves show only a weak dependence on t_w in the limit of short observation times. It is consistent, however, with the fact that the ac χ' decreases with the observation time up to the highest measuring frequencies (Fig. 1). This is borne out by inspection of the relation that exists between the dc susceptibility and the ac χ' in the case of a very broad spectrum of relaxation times,

$$\chi'(t,\omega)|_{t=t_w} = \chi_{\rm dc}(t,t_w)|_{t=1/\omega} \ . \tag{7}$$

It appears that the crossover from quasiequilibrium to nonequilibrium is much slower in the 2D short-range Ising spin glass than it is in most 3D spin glasses.

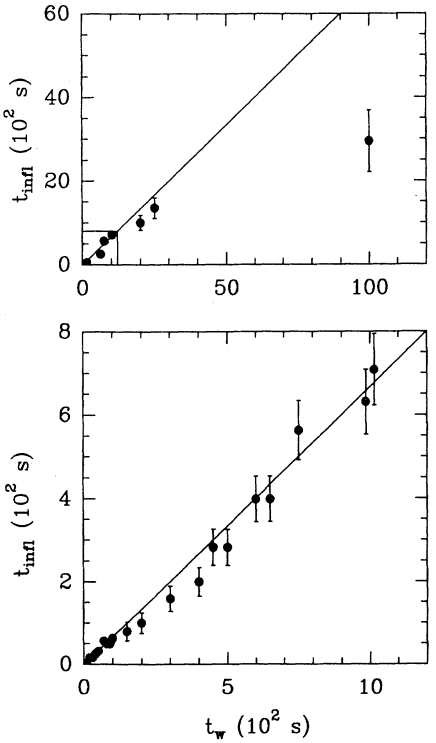

FIG. 7. Dependence of the inflection point of the FC $\chi_{\rm dc}$-vs-$\log_{10}t$ curves on the waiting time after removal of the field in $Rb_2Cu_{0.67}Co_{0.33}F_4$ at 4.2 K. The solid line in the lower frame is the least-squares fitted linear dependence.

The crossover spans logarithmic rather than linear time scales, as predicted by the droplet scaling theory.

In the present 2D spin glass above $T_c = 0$, aging is thus found to be less pronounced than in 3D spin glasses in the ordered phase. This can be made more quantitative by means of a phenomenological time scaling procedure. The procedure was pioneered in an analysis of the mechanical properties of glassy polymers,[33] and was previously applied to a number of 3D spin glasses, viz., $CsNiFeF_6$, AgMn, and $CdCr_{1.7}In_{0.3}S_4$.[4,31] Explicit account is given for the fact that the aging not only takes place during the waiting time, but persists during the subsequent observation time t. The total aging time thus is $t_a = t_w + t$. The starting point is that, to a good approximation, aging makes the whole distribution of relaxation times on a logarithmic scale, $g(\tau)$, shift towards longer times by an amount $\mu \log_{10} t_a$. It can then be shown that the process of aging is stationary with respect to the effective time λ defined by

$$\frac{\lambda}{t_w^\mu} = \frac{(t_w+t)^{1-\mu} - t_w^{1-\mu}}{1-\mu} \ , \tag{8}$$

rather than with respect to t_a. Consequently, the observed relaxation curves should fall on top of each other when plotted as a function of $\log_{10}[\lambda/t_w^\mu]$, because one is reconstructing the relaxation function the system would have at a constant given age. In the above 3D systems, collapse was indeed achieved for values of μ around 0.9 in a large range of temperatures.[4,31]

The ZFC data of the $x = 0.22$ spin glass have been put through the same scaling analysis. The best collapse of the curves in Fig. 5 was achieved for $\mu = 0.37 \pm 0.05$ at 2.5 K and $\mu = 0.33 \pm 0.05$ at 3.0 K. Figure 8 shows the

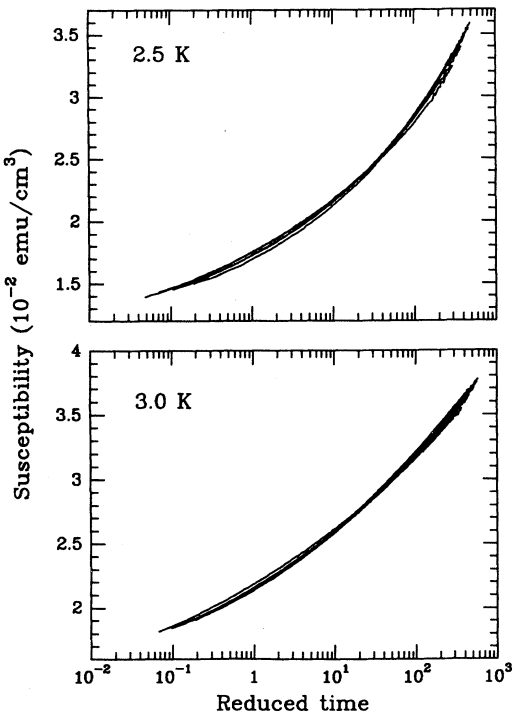

FIG. 8. ZFC susceptibility in $Rb_2Cu_{0.78}Co_{0.22}F_4$ at 2.5 and 3.0 K (Fig. 5) vs the reduced variable λ/t_w^μ [cf. Eq. (8)].

ZFC data of Fig. 5 as a function of λ/t_w^μ with these μ inserted. (In these fits, the finite cooling time has been counted as an additional waiting time of 100 s at 2.5 K, and of 45 s at 3.0 K.) It is emphasized that μ turns out to be smaller in 2D than in 3D, which confirms that aging processes have a slower time dependence in 2D. It should be appreciated, however, that the curves for the various t_w do not have exactly the same shape when plotted as a function of $\log_{10}(\lambda/t_w^\mu)$. As distinct from the 3D case, where the evolution of the dynamics during aging is correctly described by Eq. (8),[4,31] apparently no such transformation of the relaxation-time distribution exists in 2D.

V. TEMPERATURE STEPS

A characteristic in which the spin-glass ordered state differs from other ordered random-exchange magnets is the sensitivity of the equilibrium correlations to the temperature.[19] In random-exchange ferromagnets, for example, the equilibrium domains formed when passing through the transition from above will persist upon further lowering of the temperature. In spin glasses, by contrast, the sensitivity of the states to the temperature entails that the "large" domains formed near T_c at a lower temperature are small domains separated by networks of domain walls.

In 3D spin glasses, it has been demonstrated that minor temperature changes can substantially affect the process of aging.[23,31,34] Already a small decrease in temperature results in a another course of aging, which in a limited experimental time window imitates the effect of a quench all the way from above T_c. On the other hand, a very long time spent at a slightly lower temperature only minutely affects the relaxation dynamics at a higher temperature. Some of these phenomena have been interpreted[34] in the context of the droplet model. At different temperatures, aging aims at equilibrium states which are identical only up to a certain length scale, and this length scale varies strongly with the temperature.[19] In Refs. 23, 31, and 35, by contrast, this very asymmetric dependence of aging on heating and cooling has been interpreted in the framework of hierarchical organization of the metastable states as a function of the temperature.

In the case of 2D spin glasses, as opposed to 3D ones, the system is in the critical regime, and accordingly the equilibrium correlation length ξ depends on the temperature. As a result, raising the temperature to $T + \Delta T$ for some time will destroy all correlations beyond ξ at $T + \Delta T$. After return to the measuring temperature, the correlations will again grow towards ξ at T, which is larger than ξ at $T + \Delta T$. A condition for observation of this mechanism, of course, is that ξ is within reach in physically realistic times; i.e., the temperature must be sufficiently high.

In Fig. 9 the effect is presented which interim heating has on the ac susceptibility of $Rb_2Cu_{0.78}Co_{0.22}F_4$ at 2.5 K. Figure 9(a) shows the relaxation of χ' at a frequency of 0.01 Hz after a quench from 9 K. The relaxation was followed up to 3×10^4 s. At this point, the

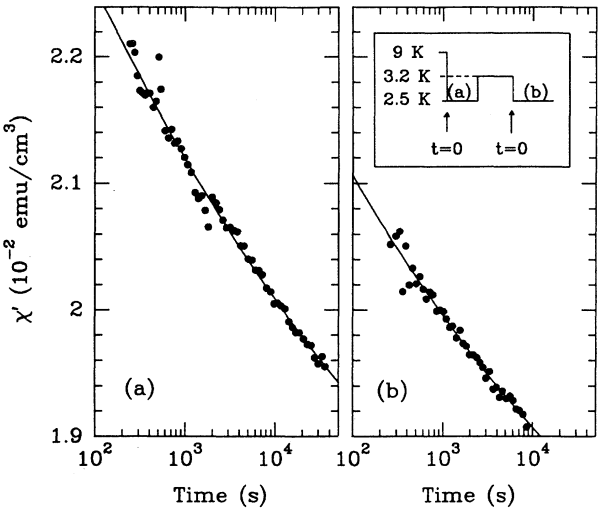

FIG. 9. In-phase ac susceptibility in $Rb_2Cu_{0.78}Co_{0.22}F_4$ at a frequency of 0.01 Hz vs the time after a quench from 9 to 2.5 K, waiting for 4×10^4 s, step heating to 3.2 K for 2×10^4 s, and a second quench back to 2.5 K.

sample was step heated to 3.2 K, where it was kept for another 2×10^4 s. The sample was subsequently cooled back to 2.5 K, and χ' was again recorded [Fig. 9(b)]. (The time origin of both traces is the moment the sample has reached 2.5 K.) Comparing Fig. 9(b) with Fig. 9(a), one sees that the aging at 2.5 K has in part been nullified by the temperature cycle to 3.2 K. The fits in Figs. 9(a) and 9(b) are based on Eq. (6), with $(d - \theta)/\psi = 2.25$ and $\omega = 2\pi \times 0.01$ rad/s inserted. They yield approximately the same χ_{eq}, but distinct constants c. As we have seen in connection with Fig. 1, aging also takes place at 3.2 K, but apparently with the emphasis in another part of phase space.

Figure 10 shows a set of ac-susceptibility relaxation

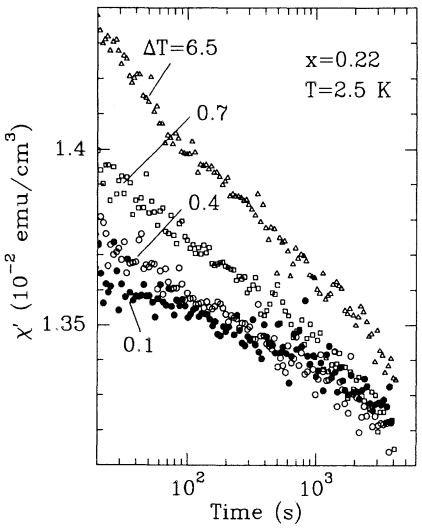

FIG. 10. In-phase ac susceptibility in $Rb_2Cu_{0.78}Co_{0.22}F_4$ at a frequency of 1 Hz vs the time after a quench from 9 to 2.5 K with an intermediate waiting time of 1000 s at $T = 2.5 + \Delta T$, with ΔT as indicated.

curves which all were taken at a measuring temperature of 2.5 K and a frequency of 1 Hz, but differ from one another in their prehistory. The time origin again is the moment at which the sample has reached the measuring temperature of 2.5 K. The samples were first quenched to a temperature of $2.5\,\mathrm{K}+\Delta T$, with $\Delta T = 0.7$, 0.4, and 0.1 K, and were subsequently held there for an intermediate waiting time of 10^3 s prior to the final quench to 2.5 K. For comparison, the temperature was also quenched directly from 9 to 2.5 K (i.e., $\Delta T = 6.5$ K). The traces are seen to be nearly linear on the semilogarithmic plot of χ' vs time, but have different slopes, the slope being smaller for smaller ΔT. More precisely, the relaxation curves satisfy Eq. (6), but with different values of c. Analysis also shows that the curves cannot be simply shifted by some effective time to coincidence with the data of the direct quench. This implies that the data for the various ΔT correspond to entirely different trajectories in phase space.

The conclusion drawn from these experiments is (i) that the trajectory followed in phase space drastically depends on the temperature history, and (ii) that the equilibrium states at different temperatures have widely different spatial spin configurations.

VI. CONCLUSIONS

In conclusion, aging effects have been observed in genuine 2D short-range Ising spin glasses. The relaxation of the susceptibility can be quantitatively accounted for within the framework of a droplet scaling model, in which the fundamental long-time nonequilibrium process is thermally activated growth of spin-glass ordered domains. These domains are a continuation in the critical regime above $T_c = 0$ of the droplet excitations in the ordered spin-glass phase. Equilibrium is established when the spin-glass correlations reach the equilibrium length $\xi \sim T^{-\nu}$. In comparison with the 3D spin glass below T_c, aging effects are less pronounced, and the crossover from equilibrium to nonequilibrium is observed on logarithmic, or epochlike, time scales. Only within a very narrow temperature range does this crossover behave in a way similar to the 3D case. In this respect, our results are at variance with the experiments on aging in thin CuMn films,[13] which yielded essentially the same behavior as in 3D. As in the 3D case, however, the equilibrium correlations are found to be quite sensitive to the temperature, and shifts in temperature by more than 1 K completely destroy the equilibrium correlations developed by the aging process.

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