## Aging, rejuvenation, and memory effects in Ising and Heisenberg spin glasses

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We have compared aging phenomena in the  $Fe_{0.5}Mn_{0.5}TiO_3$  Ising spin glass and in the  $CdCr_{1.7}In_{0.3}S_4$ Heisenberg-like spin glass by means of low-frequency ac susceptibility measurements. At constant temperature, aging obeys the same " $\omega t$  scaling" in both samples as in other systems. Investigating the effect of temperature variations, we find that the Ising sample exhibits *rejuvenation and memory* effects which are *qualitatively* similar to those found in other spin glasses, indicating that the existence of these phenomena does not depend on the dimensionality of the spins. However, systematic temperature cycling experiments on both samples show important *quantitative* differences. In the Ising sample, the contribution of aging at low temperature to aging at a slightly higher temperature is much *larger* than expected from thermal slowing down. This is at variance with the behavior observed until now in other spin glasses, which show the opposite trend of a free-energy barrier growth as the temperature is decreased. We discuss these results in terms of a strongly renormalized microscopic attempt time for thermal activation and estimate the corresponding values of the barrier exponent  $\psi$  introduced in the scaling theories.

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#### I. INTRODUCTION

Spin glasses, an intriguing class of disordered and frustrated magnetic systems, have been the focus of many efforts for some years. Below their glass transition temperature, they are out of equilibrium on all experimental time scales: their dynamics is slow and *nonstationary*: it is the *aging* phenomenon, widely studied experimentally (see various references in Refs. 1 and 2), theoretically<sup>3–6</sup> and numerically.<sup>7</sup> Still, the nontrivial dependence of aging phenomena on temperature changes ("rejuvenation and memory effects" <sup>8</sup>) is yet poorly understood,<sup>9</sup> and their interpretation in terms of the slow growth of "spin-glass-ordered domains" remains unclear.<sup>5,8,10,11</sup>

Until now, rejuvenation and memory effects have only been studied in samples of weak anisotropy, with Heisenberg-like spins.<sup>1,2,8,12,13</sup> Comparison of these experiments with numerical simulations of the Edwards-Anderson model (Ising spins), which is presently the subject of intensive efforts,<sup>7</sup> may thus not be fully relevant. Therefore, it is of particular interest to perform experiments on *Ising* compounds along the same procedures as in previous investigations.

In this paper, we present ac susceptibility measurements on the  $Fe_{0.5}Mn_{0.5}TiO_3$  spin glass, which is a wellcharacterized Ising system.<sup>14</sup> Previous studies<sup>15</sup> have shown that some rejuvenation is visible upon decreasing the temperature. But memory effects, and their interplay with rejuvenation, have not yet been investigated. Here, we want to address the questions of a possible universality of rejuvenation and memory phenomena in Ising and Heisenberg spin glasses and of their quantitative comparison in various systems.

In the Ising sample, we could check that aging at constant temperature follows the usual  $\omega t$  scaling (in agreement with previous dc experiments<sup>15</sup>). We find that *rejuvenation and* 

*memory* effects are also present, although more "spread out" in temperature than in other spin glasses.<sup>8,16</sup> For a quantitative comparison, we have taken similar data on the  $CdCr_{1.7}In_{0.3}S_4$  Heisenberg-like<sup>17</sup> spin glass,<sup>20,1</sup> which allow us to detect and characterize marked differences.

In the spin glasses explored so far, aging experiments in which a small temperature cycle is applied from T to  $T - \Delta T$  and back to T have shown that the contribution on the aging of the  $T - \Delta T$  part of the cycle decreases extremely rapidly for increasing  $\Delta T$ .<sup>12,1</sup> This drastic slowing down can be seen, in terms of thermal activation, as an *increase* of the free-energy barrier heights as the temperature is decreased.<sup>13</sup> On the contrary, in the present Ising sample, we find that this separation of time scales between T and  $T - \Delta T$  is *weaker* than expected from thermal slowing down; in other words, this would imply the unlikely scenario of a barrier height *decrease* upon cooling.

We propose to interpret this result as a sign that the elementary attempt time involved in thermal fluctuations is *much longer* than the paramagnetic time of  $\sim 10^{-12}$  s, pointing out the relevance of critical fluctuations below  $T_g$ . This same picture can also be applied to the results from the other (Heisenberg-like) sample, and in Sec. V we discuss all results in terms of the barrier exponent  $\psi$  introduced in scaling theories.<sup>5</sup>

## **II. BASIC FACTS: AGING AT CONSTANT TEMPERATURE**

The Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> compound has been characterized as a typical Ising spin glass system.<sup>14</sup> The thermal variation of the susceptibility, when measured in a field *parallel* to the hexagonal *c* axis of the ilmenite-type crystalline structure, shows a sharp cusp at  $T_g = 20.7$  K which is absent in the *perpendicular* direction. The Ising character is well evidenced by the very small ratio of perpendicular to parallel magnetization (of about 7%).<sup>14</sup> The sample used in this study



FIG. 1. Out-of-phase susceptibility  $\chi''(\omega,t)$  vs  $\omega t$  for the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> sample. The six curves have been vertically shifted (see text). *t* is the total time elapsed since the quench (age). The inset shows the same scaling for the in phase susceptibility  $\chi'$ .

is a single crystal with the shape of a rectangular parallelepiped  $(2 \times 2 \times 5 \text{ mm}^3)$ , having its long axis parallel to the *c* axis. The measurements were made along this axis, using a commercial superconducting quantum interference device (SQUID) magnetometer (Cryogenic Ltd, U.K.) with a 1.7-Oe peak value of the ac field.

Previous dc experiments on Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> (Ref. 15) have shown that the relaxation rate of the zero-field-cooled magnetization shows a broad maximum in the region  $\ln t \sim \ln t_w$ , compatible with a  $t/t_w$  scaling. Here we study the frequency ( $\omega$ ) dependence of aging in the ac susceptibility, in order to test the validity of  $\omega t$  scaling (equivalent to  $t/t_w$ scaling in dc experiments). We quench the sample from above  $T_g$  to a given temperature  $T < T_g$  (here 19 and 15 K) and measure the subsequent relaxation of the ac susceptibility at this constant temperature T as a function of time t. Each point consists in the successive measurements of eight frequencies in the range 0.04-8 Hz. As in the rest of the paper, we use the data in the paramagnetic regime (assuming  $\chi''=0$ ) for checking and correcting slight frequencydependent phase shifts in the detection setup.

We present in Fig. 1 a scaling plot for the imaginary part  $\chi''$  of the ac susceptibility  $\chi''(\omega,t)$  at  $T_0 = 19$  K. The curves have been shifted on the vertical scale in such a way that they superimpose as a function of  $\omega t$ . This (usual) procedure accounts for the stationary part  $\chi''_0(\omega)$  of the susceptibility (i.e., its limit as  $t \rightarrow \infty$ ), which can on rather general grounds<sup>1</sup> be subtracted in order to separate the aging part. The aging part can be seen to satisfactorily obey an  $\omega t$  scaling. This also holds for the real part  $\chi'$  at 19 K (inset of Fig. 1) and as well for both  $\chi'$  and  $\chi''$  at 15 K. We have fitted the curves of Fig. 1 (and the corresponding ones at 15 K) to a power law

$$\chi''(\omega,t) - \chi''_0(\omega) = A(\omega t)^{-b}.$$
 (1)

The best fit yields  $b=0.14\pm0.03$  for both temperatures. For a direct comparison, we have taken data on the



FIG. 2. Out-of-phase susceptibility  $\chi''(\omega,t)$  of the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> sample during a negative temperature cycle. The inset shows that, despite the strong relaxation at 16 K (rejuvenation effect), both parts at 18 K are in continuation of each other (memory effect).

CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> thiospinel compound ( $T_g = 16.7$  K) at the same reduced temperatures. They yield, by the same analysis,  $b = 0.18 \pm 0.03$ . Let us note that, since the value of *b* is not strongly constrained (as indicated by the error bars), we cannot conclude that a significant difference in *b* exists between both systems. However, the aging magnitude *A* is better determined and shows a clear temperature dependence in the Ising system (20% decrease from 19 to 15 K, while it is constant in the thiospinel in this same range of temperature).

We have also analyzed the stationary susceptibility  $\chi_0(\omega)$ , as determined from both the superposition of the relaxation curves at different frequencies (Fig. 1) and the nonlinear fit of Eq. (1). The stationary susceptibility of the Ising sample fits well to a power law  $\chi''_0 \propto \omega^{\alpha}$  with a small exponent  $\alpha \sim 0.1$ , in the same range (0.02–0.10: see Ref. 21) as commonly observed in other (Heisenberg-like) spin glasses. We have checked that the real part  $\chi'_0$  of the susceptibility obeys the corresponding law  $\chi''_0 = \chi'_{\text{stat}} - \mathcal{A}\omega^{\alpha}$  with the same exponent values.

Thus, from our ac measurements, the low-frequency behavior of this Ising sample at *constant* temperature is similar to that of Heisenberg-like systems.

### III. REJUVENATION AND MEMORY EFFECTS IN THE ISING SAMPLE

Temperature cycling protocols have revealed the "rejuvenation and memory" effects in several (Heisenberg-like) spin glasses.<sup>12,8,1,16</sup> We have applied the same procedures to the Ising sample. In a first experiment (Fig. 2), the sample is quenched from above  $T_g$  to  $T_1=18$  K and the relaxation of the ac susceptibility at frequency 0.4 Hz is recorded as a function of time. After a time  $t_1=4000$  s, the sample is subjected to a negative temperature cycle of duration  $t_2$ = 4000 s and amplitude 2 K. Cooling from  $T_1=18$  K to



FIG. 3. Out-of-phase susceptibility  $\chi''(\omega,t)$  vs temperature at  $\omega = 0.04$  Hz for the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> sample. The solid lines show a reference behavior for continuous cooling and reheating (the reheating curve is slightly lower than the cooling curve). Open diamonds: when cooling is stopped at 19, 15, and 10 K (respectively,  $0.9T_g$ ,  $0.7T_g$ , and  $0.5T_g$ ),  $\chi''$  relaxes due to aging (very weakly at 10 K), but when cooling resumes  $\chi''$  merges with the reference curve. Solid circles: when reheating after cooling with stops for aging, the memory of aging is retrieved. The inset shows the same results for  $\omega = 0.04$  Hz.

 $T_2$ =16 K induces a strong restart of aging (see Fig. 2): it is the so-called *rejuvenation* effect. The renewed relaxation seems to take place independently of the previous equilibration stage at  $T_1$ . But when the temperature is turned back to  $T_1$  after the cycle, we see that the two relaxations at 18 K are in continuation of each other (inset of Fig. 2). The system has actually kept a *memory* of the state reached before the cycle, and the strong relaxation at 16 K had no effect on the relaxation at 18 K.

In a second experiment, we apply a procedure in which rejuvenation and memory effects can be visualized as "dips" in the thermal variation of  $\chi'$ .<sup>8,16</sup> Starting from the paramagnetic phase, the ac susceptibility is measured (at 0.04, 0.4, and 4 Hz in one run) while continuously cooling down to 3 K at a rate of 0.001 K/s, except for three stops of 7 h at 19, 15, and 10 K, during which we let the sample age at constant temperature. Then the temperature is continuously raised back to 30 K at the same rate. In a separate run, we determine *reference curves* (solid lines in Fig. 3) corresponding to the same protocol *without stops*.

At each aging temperature,  $\chi''$  slowly relaxes downwards with time, as seen in Fig. 3. The lower the frequency, the stronger the relaxation, in agreement with the  $\omega t$  scaling (previous section). Rejuvenation effects are clearly observed at the two higher aging temperatures of 19 and 15 K; when cooling is resumed after aging,  $\chi''$  rises up and merges with the reference curve (although this reference curve itself is out of equilibrium). The other important feature is seen on the  $\chi''$ curve measured upon reheating continuously from 3 K. When approaching 15 and 19 K,  $\chi''$  departs from the reference curve and traces back a dip which displays the memory of the past relaxation at those temperatures. The rejuvenation and memory effects obtained in this Ising sample are *qualitatively* similar to those observed in other spin glasses.<sup>8,16</sup>

However, in the experiment of Fig. 3 with successive agings stops at  $0.9T_g$ ,  $0.7T_g$ , and  $0.5T_g$ , the observed relaxation decreases significantly from one stop to another (by a factor of 2.5 between  $0.9T_g$  and  $0.7T_g$ ) and becomes undistinguishable at  $0.5T_g$ . This feature makes a difference with the CdCr<sub>1.7</sub>In<sub>0.3</sub>S spin glass<sup>8</sup> and may indicate some "accumulation" of aging, which would in consequence progressively die out during a long cooling procedure with multiple stops.

In fact, the comparison of the  $\chi''$  relaxations at  $0.7T_g$  after a slow cooling (0.001 K/s as above) and a direct quench does not show any difference between both samples: in the slow cooling case, the relaxation is for both samples ~70% of that obtained after the direct quench (of course, rejuvenation effects do not exclude that the very last part of the cooling procedure induces a weak but nonzero cooling rate effect<sup>8</sup>). But in a multiple-stop procedure like the one in Fig. 3, we could check that in this Ising sample the relaxation at the *lower* temperature is *more* influenced by aging at the higher temperature than in the thiospinel. The relaxation at  $0.7T_g$ (second stop) only amounts to ~25% of that obtained after a direct quench, whereas this proportion is of ~ 50% for the thiospinel in the same conditions.

In other words, aging processes in this Ising sample are more sensitive to the thermal history (cooling rate effect) than in other spin glasses, as already suggested by the spreading in temperature of the dips in Fig. 3.

# IV. QUANTITATIVE EXPERIMENTS: THERMAL SLOWING DOWN AND BARRIER GROWTH

The procedure of negative temperature cycling (like presented in Fig. 2) is "simpler" than the continuous cooling and reheating procedure of Fig. 3 and can yield more direct information on the influence of time and temperature on the rejuvenation and memory effects. We now discuss a series of such measurements on both samples, with *small* temperature intervals. After quenching from above  $T_g$  to  $T < T_g$  at time zero, the temperature T is kept constant for a time  $t_1$ = 7700 s, after which it is lowered to  $T - \Delta T$  for a time  $t_2$ = 23 650 s, and finally raised back to T for a time  $t_3$ = 6000 s.

The relaxation of the ac susceptibility at frequency  $\omega = 0.1$  Hz is recorded during the whole procedure. As working temperatures, we chose equivalent values in units of  $T_g$  for both samples:

(i) 
$$T = 18 \text{ K} = 0.87 T_g$$
 and  $T = 15 \text{ K} = 0.72 T_g$   
for Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub>,

(ii) 
$$T = 14 \text{ K} = 0.84 T_g$$
 and  $T = 12 \text{ K} = 0.72 T_g$   
for  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$ .



FIG. 4. Effect of a negative temperature cycling ( $\Delta T = -1$  K) on the time dependence of  $\chi''(\omega = 0.1$  Hz) in the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> sample. The inset shows a plot of the data points recorded at T = 15 K during time  $t_1$  before the cycle and  $t_3$  after the cycle. The points after the cycle have been superposed on a standard isothermal relaxation curve by shifting the time scale. The decay during  $t_3$  is a continuation of the initial decay with an effective time interval  $t_{\text{eff}}=4500$  s.

On the other hand, the temperature intervals have been adjusted in order to produce about the same effect in both samples: therefore, they are different, even in units of  $T_g$ :  $\Delta T = 0.25$ , 0.5, 0.75, and 1 K for Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> and  $\Delta T = 0.1$ , 0.2, 0.3, and 0.4 K for CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub>.

An example experiment is presented in Fig. 4 for the Ising sample at T=15 K. The effect of a  $\Delta T=1$  K negative cycling is compared with an isothermal reference curve at T= 15 K. For this small temperature interval, *no jump* (such as the one seen in Fig. 2) in the relaxation signs up any important rejuvenation effect as the temperature is decreased, whereas a *small jump* of  $\chi''$  is visible for the thiospinel sample in the same conditions. When the temperature is increased back to T,  $\chi''$  rises up to a value which is higher than the isothermal reference at T. This final relaxation can be superposed with the isothermal reference by a shift backwards of a time  $t_2 - t_{\text{eff}}$  with  $t_{\text{eff}} = 4500$  s (see inset of Fig. 4). The effective time  $t_{\rm eff}$  accounts for the effect at T of the processes which occurred at  $T - \Delta T$  during  $t_2$ . After  $t_1$  $+t_2$ , the state of aging can be considered as an additive combination of aging at T before the cycle, which has hence been memorized, with a contribution  $t_{\rm eff}$  from aging at T  $-\Delta T$ . It is interesting to compare this result with a simple estimate of the effect of thermal slowing down. Considering simply that aging involves thermally activated jumps over *fixed* free-energy barriers, the effective time  $t_{\text{eff}}^{TA}$  at  $\hat{T}$  which corresponds to  $t_2$  at  $T - \Delta T$  reads

$$(T - \Delta T) \log\left(\frac{t_2}{\tau_0}\right) = T \log\left(\frac{t_{\text{eff}}^{TA}}{\tau_0}\right), \qquad (2)$$

where  $\tau_0$  is an attempt time, which we choose for now as a microscopic spin-flip time, say,  $\tau_0 = 10^{-12}$  s. Surprisingly,



FIG. 5. Effective waiting times extracted from negative temperature cycling experiments on the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> and CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> samples, normalized to the duration of the cycle. For a comparison, the lines (*TA*) show the estimate for a simple thermal slowing down scenario [Eq. (2) with  $\tau_0 = 10^{-12}$  s].

one gets  $t_{\text{eff}}^{TA}$ =1915 s, a value smaller than  $t_{\text{eff}}$ =4500 s. That is, aging at the lower temperature has a *larger* contribution than expected from thermal activation over *fixed height* barriers.

This result is in sharp contrast with those obtained for the thiospinel sample and other spin glasses, in which the effective contribution of aging at a lower temperature is always smaller than expected from simple thermal slowing down, a general result that has been discussed in terms of a growth of free-energy barriers as the temperature is lowered.<sup>12,13</sup>

The whole set of results on both samples is summarized in Fig. 5, which displays the measured values of the effective times  $t_{\rm eff}(T,\Delta T)$ , in comparison with the calculated estimates for thermal activation over *fixed height* barriers [Arrhenius law, Eq. (2), with  $\tau_0 = 10^{-12}$  s]. Clearly, two different behaviors are obtained: for the thiospinel (Heisenberg-like) sample, we always have  $t_{\rm eff} < t_{\rm eff}^{TA}$ , whereas  $t_{\rm eff} > t_{\rm eff}^{TA}$  for the Ising one.

### V. PHYSICAL INTERPRETATION: A TEMPERATURE-DEPENDENT LENGTH SCALE

Our result that, in the Ising sample, the contribution of aging slows down less than expected when the temperature is lowered is, at first sight, puzzling. However, this conclusion is based on the assumption that the relevant elementary time scale is the paramagnetic spin-flip time  $\tau_0 \sim 10^{-12}$  s. In Fig. 5, the position of the curves calculated for thermal activation depends on the choice of the attempt time  $\tau_0$ ; they would move upwards for larger values of  $\tau_0$ . Another way to formulate this is proposed in Fig. 6, where we have plotted  $\log[t_{eff}/t_2^{T-\Delta T/T}]$  as a function of  $\Delta T/T$ . If barriers of the same height are crossed during the experiments at T and  $T-\Delta T$  with an attempt time  $\tau_0$ , one should observe a straight line of slope  $\log \tau_0$  [Eq. (2)]. One can see very clearly that the



FIG. 6. Effective waiting times extracted from the experiments, plotted in such a way that thermal activation over fixed height barriers [Eq. (2)] corresponds to a straight line of slope  $\log \tau_0$  ( $\tau_0$  in *s*). Thus, the average slope of each set of data shows an estimate of  $\log \tau_0^{\text{eff}}$ , where  $\tau_0^{\text{eff}}$  would be an effective attempt time for fixed height barriers. In addition to the present data from the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> and CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> samples (same as in Fig. 5), we have plotted comparable results obtained earlier with the Ag:Mn 2.7% sample (Ref. 13).

Ising data correspond to an effective attempt time  $\tau_0^{\text{eff}}$  much larger than  $10^{-12}$  s, whereas the CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> Heisenberglike spin glass favors  $\tau_0^{\text{eff}} \ll 10^{-12}$  s. A comparison can also be made with a third example, the Ag:Mn 2.7% spin glass, in which the anisotropy should still be weaker. In this sample, very detailed studies on the effect of tiny temperature variations have been performed in *dc* experiments, <sup>13</sup> yielding a precise characterization of the free-energy barrier growth as the temperature is lowered. Figure 6 shows that the corresponding effective attempt times  $\tau_0^{\text{eff}}$  are still shorter.

A natural possibility to explain the very large increase of the attempt time in the Ising sample is the proximity of the spin-glass transition which leads to critical slowing down. Dynamic critical scaling above  $T_g$  tells us that characteristic times  $\tau$  are diverging like the spin-glass correlation length  $\xi$ as a power law of temperature:

$$\tau \approx \xi^{z} \sim \left(\frac{T - T_{g}}{T_{g}}\right)^{-z\nu}.$$
(3)

By analogy with the ferromagnetic case, in which the correlation length gives the size of the typical (critical) fluctuations above as well as below the transition with the same exponent, we shall assume that the attempt time in the spinglass phase is governed by critical fluctuations below  $T_g$ . During aging, the characteristic length *l* over which spins are correlated grows progressively with time, as identified in experiments on the effect of field variations on the dynamics<sup>22</sup> (with good quantitative agreement with numerical simulations<sup>7</sup>). We propose that the attempt time corresponding to the length *l* is proportional to  $l^z$  and that the time  $\tau(l)$  which is needed to flip the *l*-sized cluster of spins is governed by thermal activation over a barrier  $B_T(l)$ , in such a way that<sup>23</sup>

$$\tau(l) = \tau_0 l^z \exp\left(\frac{B_T(l)}{k_B T}\right),\tag{4}$$

where *l* is in units of the lattice constant. The temperature and length dependence of  $B_T(l)$  can also be expressed by analogy with the ferromagnetic case and in the spirit of the scaling theories.<sup>5</sup> One can write that

$$B_T(l) = \Upsilon(T) l^{\psi}, \tag{5}$$

where  $\Upsilon(T)$  is a wall stiffness which should vanish at  $T_g$  like in ferromagnets:

$$\Upsilon(T) = \Upsilon_0 T_{\varrho} [(T_{\varrho} - T)/T_{\varrho}]^{\psi \nu}.$$
 (6)

This corresponds to the assumption that barriers on the scale of the correlation length  $\xi$  are of order  $k_B T_g$ .

Replacing the attempt time  $\tau_0$  by a longer time  $\tau_0 l^z$  in Eq. (2) will indeed qualitatively account for our experimental results on the Ising sample (weak slope  $\log \tau_0^{\text{eff}}$  of the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> data in Fig. 6). The  $\tau_0 l^z$  term is dominant in Eq. (4) provided one is in a regime where  $B_T(l) \leq k_B T$ , i.e., for temperatures sufficiently close to  $T_g$ . For lower temperatures, one may expect the growth of the barriers when the temperature is decreased [i.e., the  $\Upsilon(T)$  variation, Eq. (6)] to be the dominant effect, therefore leading to a much stronger slope  $\log \tau_0^{\text{eff}}$ . Our results indicate that the Ising sample is dominated by the critical regime, whereas the Heisenberg-like samples are rapidly dominated by the barriers (see below the values z,  $\nu$ , and  $\psi$ ).

We have adjusted our experimental data to Eqs. (4)–(6) in the following way. What comes out from each negative cycling experiment is the equivalence between an aging time  $t_2$ at  $T_2 = T_1 - \Delta T$  and an effective time  $t_{eff}$  at  $T_1$  (to be fully rigorous, the equivalence should also involve aging during  $t_1$ at  $T_1$ , but we checked that this does not affect the results). In terms of the characteristic length scales involved, this equivalence corresponds to  $l(t_{\text{eff}}, T_1) = l(t_2, T_2)$ . Using Eq. (4), we can numerically extract  $\Upsilon_0$  and  $\psi$  from the experiments by a best fit procedure,<sup>23</sup> which allows one in most cases to obtain  $l(t_{\text{eff}}, T_1) = l(t_2, T_2)$  within about 0.5%. In practice, both parameters are strongly correlated in the fit. We therefore present the results obtained with a reasonable value  $\Upsilon_0 = 1$ . Choosing higher (lower) values of  $\Upsilon_0$  yields lower (higher) values of  $\psi$ , but all qualitative trends remain the same. On the other hand, we have taken zv from results of dynamic critical scaling and estimated  $\nu$  from the static scaling exponents  $\beta$  and  $\gamma$  using  $\nu = (2\beta + \gamma)/d$ .<sup>14,20,24</sup> Thus we have  $\nu = 1.7$  and  $z\nu = 10.5$  for  $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$  and  $\nu = 1.3$  and  $z\nu = 7$  for CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub>, and in addition we have used  $\nu = 1.4$  and  $z\nu = 5$  (Refs. 24 and 25) for a comparison with older results on Ag:Mn 2.7%.<sup>13</sup> Varying slightly zv and vdoes not affect the following conclusions ( $\psi$  may vary by  $\pm 0.1$ ).

For a given temperature T, no systematic variation of  $\psi$  with  $\Delta T$  can be seen; therefore, we have fitted together all

TABLE I. Barrier exponent  $\psi$ , extracted from the negative cycling experiments using Eq. (4) with  $\nu$  and z as listed. The first two lines correspond to the ac measurements presented in this paper; the Ag:Mn 2.7% result has been deduced from previous dc data (Ref. 13).

	ν	zν	ψ
Fe <sub>0.5</sub> Mn <sub>0.5</sub> TiO <sub>3</sub>	1.7	10.5	0.3-0.7
CdCr <sub>1.7</sub> In <sub>0.3</sub> S <sub>4</sub>	1.3	7	1.1
Ag:Mn 2.7%	1.4	5	1.3

 $\Delta T$  results at each *T*. The results are as follows (see a summary in Table I). In the Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> Ising sample, we find  $\psi \sim 0.3$  at  $0.87T_g$  and  $\psi \sim 0.7$  at  $0.72T_g$ . This rather small value of  $\psi$  is associated to very large values of the renormalized attempt time: we find that  $\tau_0 l^z \sim 2500$  s at  $0.87T_g$  and 5 s at  $0.72T_g$ . Hence, the thermal activation hardly plays any role, in particular at  $0.87T_g$ : the dynamics is primarily probing the critical regime, in qualitative agreement with the "anomalous" effect of temperature shifts reported above. This might also explain why our estimate for  $\psi$  differs between the two temperatures, since the scaling behavior of  $B_T(l) \sim l^{\psi}$  implicitly assumes that  $l \geq \xi$ .

On the other hand, the CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> thiospinel sample (Heisenberg like) shows a markedly different behavior. Nevertheless, our theoretical analysis in terms of temperature-dependent barriers and a renormalized attempt time can be applied to this sample. About the same value of  $\psi \approx 1.1$  is favored for both temperatures. The higher value of  $\psi$  confirms the stronger separation of the different aging contributions for different temperatures, as already emphasized in the previous section.

We have also reanalyzed the former dc data of the Ag:Mn 2.7% spin glass.<sup>13</sup> In this third case, the above analysis leads to  $\psi \sim 1.3$ , indicating a still stronger effect of temperature (see Ref. 23 for a more detailed discussion), in agreement with the trend observable in Fig. 6.

### VI. CONCLUSION

We have applied temperature variation protocols in experiments on the Ising  $Fe_{0.5}Mn_{0.5}TiO_3$  spin glass. We find that the aging phenomena in the Ising sample present rejuvenation and memory effects which are qualitatively similar to those already evidenced in other (Heisenberg-like) spin glasses.<sup>26</sup>

The interest in the question of the "universality" of rejuvenation and memory phenomena in spin glasses has been recently revived by the numerous numerical simulations<sup>7</sup> of the Edwards-Anderson model (Ising spins), in which these effects could not yet clearly be seen. Our results indicate that the dimensionality of the spins should not be at the origin of this discrepancy, which we rather ascribe to the difference in time scales. The magnetization and susceptibility experiments are performed at macroscopic times which are up to  $\sim 10^{17}$  times the microscopic spin flip time ( $\sim 10^{-12}$  s), whereas numerics have been until now limited to  $\sim 10^{5-6}$  steps, exploring a much shorter range of correlation lengths.

It is plausible that, in simulations, these aging lengths are not enough different at different temperatures to allow the "separation of length scales"<sup>23</sup> which is necessary to observe rejuvenation and memory.

When studied in more details, aging in the Ising sample shows a new behavior. First, it dies out rather rapidly towards low temperatures and even seems completely frozen below  $0.5T_{g}$ . Second, negative temperature cycling experiments show that the separation of time scales in a negative cycle is here much weaker than in other spin glasses and, surprisingly, even weaker than expected from thermally activated dynamics with a microscopic attempt time. We interpret this effect as the signature of the mesoscopic or even macroscopic value of the attempt time for thermal activation, which we associate with a characteristic time  $\tau \propto l^z$  for critical fluctuations. In this framework, we have analyzed the temperature variation of the free-energy barriers within scaling hypotheses inspired from the droplet models of the spinglass phase<sup>5</sup> and found that the analysis could be applied equally well to both the Ising and the Heisenberg samples. The values of the barrier exponent  $\psi$  that we extract from the experiments on the Ising sample along this line are of the order of  $\sim 0.5$ . In comparison, the same experiments on the CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> Heisenberg-like spin glass yield  $\psi \approx 1.1$ . Using former data (from dc experiments<sup>13</sup>) on Ag:Mn, we found  $\psi \cong 1.3$ .

Of course, the data on both Heisenberg-like samples do not by themselves imply considering that the attempt time  $\tau_0$ for thermal activation [Eq. (2)] should be mesoscopic. Using a microscopic  $\tau_0 \sim 10^{-12}$  s, a rapid growth of the barriers with decreasing temperature was reported in Ref. 13, a result that was interpreted there as a sign that all barriers eventually diverge in the spin-glass phase. Here, we rather propose that barriers vanish continuously at  $T_g$  and are always finite (for finite l) for  $T < T_g$  (see also Ref. 23). The experimental data, over a limited time range, cannot really discriminate these two scenarios, although the latter has a rather natural physical motivation.

In any case, the central point of the present paper is that, for the Ising sample, a microscopic  $\tau_0$  would correspond to an unphysical decrease of the barriers at low temperatures  $(\psi < 0)$ . Allowing for a renormalized attempt time, we find that  $\psi$  is positive in the Ising sample, but significantly smaller than in the other systems. This small value implies a weaker separation of the time and length scales<sup>27</sup> as the temperature is lowered. This conclusion does not depend on the details of the theoretical analysis and is clearly visible in several qualitative aspects of the data (e.g., Figs. 5 and 6).

It is worth noticing that, within the three samples compared here, the estimated  $\psi$  values are found to increase with decreasing spin anisotropy, at variance with the intuitive expectation that walls made of anisotropic spins might be softer (i.e., with lower  $\psi$ ). This may be related to results from mesoscopic electrical noise measurements,<sup>28</sup> which show that the temperature dependence of stationary dynamics is stronger in CuMn spin glasses (equivalent to barrier growth for decreasing temperature) than in AuFe (Arrhenius behavior) compounds of much stronger anisotropy. On the other hand, a systematic dependence of the static critical exponents at the spin-glass transition as a function of anisotropy has been recently noted.<sup>29</sup> The question of a different nature of the spin-glass phase for Ising and Heisenberg spins is a long-standing issue and must be considered in the light of the present and forthcoming experimental and numerical results.

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