# Aging in spin glasses as a random walk: Effect of a magnetic field

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We investigate the role of a magnetic field on the aging properties of spin glasses, focusing both on ac susceptibility measurements with an additional field variation and on thermoremanent magnetization (TRM) relaxation (to which the field variation is inherent). We propose a scaling of the TRM decay curves for various amplitudes of the magnetic field. Regarding aging as a random walk in a set of traps, we analyze the transient modification of the dynamics after a field change as twofold: an average reduction of relaxation times, well described by a Zeeman reduction of the energy barriers (hard traps), together with an "emptying out" of some other relaxation modes (fragile traps) corresponding to a partial quench effect. The fragile traps are associated with the longest time scales, that is to processes involving a large number of spins. A quantitative analysis of the results allows us to estimate a typical number of spins involved in both types of traps, as well as to discuss the growth of the energy barriers as a function of the number of spins.

### I. INTRODUCTION

Aging occurs when the physical properties of a system evolve with time, giving rise to nonstationary dynamics and to the breakdown of time translational invariance for the response of the system to an external perturbation. Aging phenomena have been known in some branches of the material science,<sup>1</sup> but their analysis has remained limited to a phenomenological description, and until recently<sup>2</sup> they had no place in any *ab initio* theoretical construction. Aging effects were initially seen as an experimental difficulty, since the measured properties vary during the measurement. For example, in spin glasses, the decay of the thermoremanent magnetization (TRM) and the relaxation of the zero-field-cooled magnetization have been found to depend on the waiting time before the field variation, the maximum decay rate being obtained after a time of the order of the waiting time itself.<sup>3-5</sup> Careful analyses of these aging phenomena, and especially of the effect of slight temperature changes, have later shown that aging can teach us a lot about the hidden properties of the spin-glass phase.<sup>6</sup> The hierarchical organization of the metastable states as a function of temperature, inferred from the temperature variation experiments,<sup>6</sup> brings the experimental spin glass closer to its rather abstract description in the Parisi solution of the meanfield problem.<sup>7</sup> Conversely, since the real spin glass is always found out of equilibrium, the theory is now focused on out-of-equilibrium properties, and its most recent analytical and numerical developments<sup>8</sup> show that aging phenomena indeed occur within the original Sherrington-Kirkpatrick (SK) model.9

The dynamics of the spin glass is commonly investigated by recording the response to a small field variation (except in the case of measurements of the spontaneous magnetic noise<sup>10</sup>). In the present paper, we want to address the question of how far aging is affected by the field variations which are used in the diverse experimental scenarios. We discuss TRM measurements for various field amplitudes; but the most quantitative information is obtained from low-frequency ac susceptibility measurements, during which a small dc field variation (comparable to those used for TRM studies) is applied.

In a standard experiment, the spin glass is quenched from the paramagnetic phase, above the freezing point  $T_g$ , down to some temperature  $T_0$ . We call age the time elapsed at the constant temperature  $T_0$ ; this time governs the observed properties. For example, in a TRM decay measurement, the sample is cooled in a field, which is removed after a waiting time  $t_w$ . Aging is visible in the  $t_w$ dependence of the decay curves, or more precisely in their age dependence, the age being in that case the sum of the waiting time plus the duration of the relaxation measurement. Phenomenological scaling relations have been established to account in detail for these aging effects;<sup>5,6</sup> they can be summarized in the following way. We describe the spin-glass dynamics by a distribution  $g(\tau, t_a)$  of relaxation times  $\tau$  at an age  $t_a = t + t_w$ ; the age dependence of the distribution is found to be

$$g(\tau, t_a) = G(\tau/t_a^{\mu}) , \qquad (1)$$

where  $\mu$  is an exponent ~0.9 for  $0.4 < T/T_g < 0.9$ . Thus, over most of the temperature range, aging is equivalent to a "logarithmic shift" of relaxation times, with a constant of proportionality slightly lower than 1. This point can be given a physical meaning, which is briefly discussed at the end of Sec. IV. For our present purpose, it will be sufficient to retain (as an approximation) that the TRM relaxation essentially depends on the ratio  $t/t_w$ . Correspondingly, the ac susceptibility at frequency  $\omega$  relaxes as the age  $t_a$  increases, and mainly depends on the product  $\omega \cdot t_a$ .

The occurrence of aging, together with an approximate  $t/t_w$  or  $\omega \cdot t$  scaling, can be intuitively understood within the scenario of *weak ergodicity breaking*,<sup>11</sup> which we briefly recall. Due to frustration, the spin glass cannot find equilibrium in the low-temperature phase; within a

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configuration space description, it wanders in a selfsimilar mountainous landscape of metastable states, surrounded by energy barriers of all possible heights. This general picture is implied by the evidence, in the experiments, of dynamic processes at all time scales. In Ref. 11, this slow evolution is sketched by a random walk among a collection of "traps" with random trapping times  $\tau$ , all equally accessible. To each trap is associated a certain magnetization M and ac susceptibility  $\chi_{\tau}(\omega)$ . In order to obtain the properties of the real sample, one should average over a collection of magnetically decorrelated subsystems with the appropriate probability distribution. The distribution of trap depths is taken as an exponential, as suggested by the mean-field results.<sup>7</sup> For thermally activated processes, this then leads to the following distribution of trapping times:

$$\psi(\tau) = \frac{\tau_0^4}{\tau^{1+x}} \quad (\text{for } \tau \gg \tau_0) , \qquad (2)$$

where x (from the distribution of barrier heights) is a temperature dependent parameter describing the structure of the phase space. The crucial point is that x < 1 in the spin-glass phase, and that in consequence the mean value of  $\psi(\tau)$  is divergent; the mean time needed to explore the whole set of traps (and thus to reach ergodicity) is infinite. In this sense, ergodicity is *weakly* broken: although there are always larger and larger barriers to be crossed, the system never gets trapped in a finite region of phase space. This is at variance with usual ergodicity breaking, where systems reach rather quickly an equilibrium configuration, albeit in a restricted sector of phase space.

After a time  $t_w$ , the system has explored a very large number of short-lifetime traps, which actually contribute (as far as x < 1) to a very modest part to the total elapsed time. As usual with extremely broad distributions, the significant contributions arise from the largestalthough infrequent—events. Thus, after a random walk during  $t_w$ , the system has the largest probability to be found in a trap of characteristic time of order  $t_w$  itself. At that point,  $t_w$  fixes the time scale of the dynamics, and the microscopic time  $\tau_0$  becomes irrelevant. For TRM experiments, it has been shown<sup>11</sup> that the magnetization decay depends on the ratio  $t/t_w$ . A similar formulation can be given in the case of ac experiments;<sup>11</sup> assuming that the out-of-phase susceptibility  $\chi_{\tau}''(\omega)$  of a  $\tau$  trap is a function of  $\omega \cdot \tau$  only, peaked around  $\omega \cdot \tau = 1$  (e.g., a simple Debye form), one finds by averaging over the subsystems that the out-of-phase part  $\chi''(\omega, t)$  of the ac susceptibility reads

$$\chi''(\omega,t) = \chi_0 (\omega \cdot t)^{x-1} + \chi_{\infty} \quad (\text{for } t \gg 1/\omega) , \qquad (3)$$

where  $\chi_0$  is a constant. We have added in Eq. (3) the equilibrium value  $\chi_{\infty}$ , which naturally appears when some stationary "bottom of the traps" dynamics is introduced, as recently discussed in Ref. 12 (see also Sec. V). The  $\omega \cdot t$  dependence of  $\chi''(\omega, t)$  is equivalent to the  $t/t_w$ scaling of the TRM. We mainly concentrate here on the *dissipative* component of the ac susceptibility because it is the most sensitive in relative value to aging effects. The results presented here have been obtained with the  $CdCr_{1.7}In_{0.3}S_4$  insulating compound, elaborated by M. Noguès at the Meudon-Bellevue CNRS laboratory. This sample has been previously characterized as a spin glass, exhibiting a *static* and *dynamic* critical behavior<sup>13</sup> around  $T_g = 16.7$  K. It is worth noticing that this system presents in all respects the same behavior as the Ag:Mn<sub>2.6%</sub> metallic compound.<sup>5,6</sup>

The paper is organized as follows. In Sec. II, we describe the behavior of the aging out-of-phase susceptibility when small field variations are applied. We analyze the results with the help of the trap model,<sup>11</sup> and examine the manner in which the traps are affected by a field variation. This prompts us to go one step beyond the "phase-space" description of the trap model, by associating to a trap a typical number of spins to be flipped to escape. However, the general features of the effect on aging of field variations that we deduce from both  $\chi''$  and TRM measurements are experimental facts, and therefore independent of any model. In Sec. III, we examine TRM measurements for various fields amplitudes along the same line, and in Sec. IV we use both TRM and  $\chi''$  results to develop a global quantitative description. Finally, in Sec. V, we use our analysis to relate the number of spins to the energy barrier associated with metastable states.

## II. AGING OF THE ac SUSCEPTIBILITY AND dc FIELD VARIATIONS

#### **A. Experiments**

The low-frequency (0.1 and 1 Hz) ac susceptibility in presence of a superposed dc field has been measured with an rf superconducting quantum interference device magnetometer, the lock-in detection being performed by a digital method. Details on this setup and the experimental procedure can be found in Ref. 14, where other results of the same series of  $\chi''$  measurements are also presented. Let us only recall that, upon applying or removing a dc field of 5-30 Oe, a huge varying magnetization adds up to the response of the spin glass to the small (0.3 Oe) ac probe field. For each point, this magnetization drift has been measured; during the time needed for the lock-in detection (two ac periods), it has been approximated linearly, and accounted for in the digital analysis.

At the beginning of the measurement, the sample is in the paramagnetic phase at  $T=1.3T_g$ ; it is cooled down to  $0.7T_g$  (12 K, temperature of all the present  $\chi''$  measurements) in a few minutes. At this point, the spin glass is out of equilibrium and starts aging; we continuously record the ac susceptibility, which slowly decreases. Aging is particularly visible (in relative value) in the out-ofphase component  $\chi''$ , on which we focus here. Figure 1 presents the  $\chi''$  relaxation as a function of the age (time elapsed after quench); in the first part of the experiment (age < 350 min), the relaxation shows the aging evolution started at the quench. After 350 min, we apply a dc field of 5-30 Oe. This range of values remains small compared to the field of order 1 kOe which, at this temperature, would bring the spin glass back to the paramagnetic 1052

phase.<sup>14</sup> The field is kept applied during a second period of 350 min; it is then removed, and the  $\chi''$  measurement is pursued during another 350 min.

In Fig. 1, the first obvious consequence of applying the dc field is that the  $\chi''$  relaxation is restarted. In terms of metastable states or traps, we can say that the field variation  $\Delta H$  has produced an increased population rate in the short-time traps (of order  $\tau = 1/\omega$ ), therefore increasing  $\chi''(\omega)$ . We associate a typical Zeeman energy  $E(\Delta H)$  to the field variation  $\Delta H$ ; the lifetime  $\tau$  of the traps will be reduced to  $\tau'$  by a multiplicative factor  $\alpha$  such that

$$\tau' = \alpha \cdot \tau$$
 and  $\alpha = \exp(-E(\Delta H)/k_B T)$ , (4)

where  $k_B$  is the Boltzmann constant and T the temperature. At the time  $t_1$  of the perturbation, the system has the largest probability<sup>11</sup> to be found in  $\tau$  traps such that  $\tau \sim t_1$ ; if the perturbation reduces this value to  $\alpha \cdot \tau_1$ , the system will now behave as if its age was also reduced to  $\alpha \cdot \tau_1$ . This is the trend observed in Fig. 1, but a careful examination shows that this does not work quantitatively. The  $\chi''$  relaxation after the field variation cannot be exactly superimposed onto some part of the one following the quench; a horizontal translation, corresponding to a decrease of the effective age, yields too smooth an effect. The measured relaxation restarts more abruptly, as if for part of the subsystems the effect was more like a *new quench* rather than a reduction of the age.

In addition, a decrease of the effective age is not sufficient on its own to explain the frequency dependence of the results. Figure 2 shows similar experiments, performed for two different values  $\omega = 0.1$  and 1 Hz of the frequency. At 0.1 Hz, the field is applied after 350 min as in Fig. 1, whereas at 1 Hz, it is applied after only 35 min, in order to keep the product  $\omega \cdot t_1$  constant. The curves, plotted as a function of  $\omega \cdot t$ , have been vertically translated in order to make them coincide in their first part. Part of this vertical translation accounts for the difference in the equilibrium values of  $\chi''$  at 0.1 and 1 Hz, which cannot be disentangled from a slight uncertainty on the



FIG. 1. Relaxation of the out-of-phase susceptibility  $\chi''$  (in arb. units) at frequency  $\omega = 1$  Hz and constant temperature 12 K, as a function of the time (age) following a quench from above  $T_g = 16.7$  K. The sample is (as in all figures) the CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> insulating spin glass. After 350 min, a static field  $\Delta H$  (=5, 9, 15, or 30 Oe) is applied, producing a renewed  $\chi''$  relaxation. After another 350 min, the field is removed. The inset sketches the procedure.



FIG. 2. Comparison of the effect on  $\chi''$  of a dc field variation in two experiments at frequencies  $\omega = 0.1$  and 1 Hz. The field variation is applied at a time  $t_1$  (=350 and 35 min, respectively) such that the product  $\omega \cdot t_1$  is kept constant. The curves are plotted as a function of  $\omega \cdot t$ , and have been vertically shifted in order to superpose both relaxations before the field variation. At constant  $\omega \cdot t$ , the effect of the perturbation is seen to be stronger for the lowest frequency (longest  $t_1$ ).

phase setting of the lock-in detection at each frequency. The coincidence of both relaxations after the quench shows that the  $\omega \cdot t$  scaling, suggested by the trap model, indeed works fairly well.

In contrast, Fig. 2 clearly shows that the effect of the field perturbation is weaker at higher frequency and constant  $\omega \cdot t_1$ , thus shorter  $t_1$ . This feature has been systematically observed for all the values of  $\Delta H$  that we have explored. We interpret this result in the following way: the relevant traps (those which are significantly occupied) are more affected by the field variation in the (0.1 Hz, 350 min) experiment than in the (1 Hz, 35 min) experiment. At the time of the perturbation, these relevant traps have a lifetime of the order of the age, i.e., larger in the 0.1 Hz case; hence, the deeper the trap, the stronger its coupling to the field. We are thus led to go beyond the purely phase-space description that we have used up to now, by specifying how a  $\tau$  trap "couples" to the magnetic field. One can think of a typical number N of spins, which must be flipped to escape from the bottom of the  $\tau$ trap of depth B(N):

$$\tau = \tau_0 \exp(B(N)/k_B T) , \qquad (5)$$

 $\tau_0$  being some attempt time, to be specified later. B(N) is expected to increase with N. We now explicit the energy variation  $E(\Delta H)$  in Eq. (4) as a Zeeman coupling of the field with the group of N spins, where "spins" denote the elementary magnetic objects to which the field couples; they can be single spins, but also renormalized groups of spins. Let us say that they have a magnetic moment Mequal to  $m\mu_B$ . Due to the random nature of the interactions and the frustration they cause, the net uncompensated moment for a group of N spins is of the order of  $\sqrt{N}$ , and thus we write

$$E(\Delta H) \equiv E(\Delta H, N) = \sqrt{N} m \mu_B \Delta H .$$
(6)

The reduction factor  $\alpha$  is thus smaller (stronger perturba-

tion) for the deeper traps which are explored in a longer experiment; this is the correct qualitative behavior observed in Fig. 2.

We give in Fig. 3 a tentative sketch for qualitatively summarizing our picture. Figure 3 depicts the freeenergy landscape after a field variation. Two sets of traps are shown, corresponding to two different values of the total magnetization. The perturbation has tilted the landscape; the new value of the field now favors the magnetization corresponding to the set of traps located downhill. In our analysis, the deeper the traps, the larger the number of spins involved, and the larger the extension of the traps in the configuration space. In Fig. 3, we have pictured the deeper traps as craters with a very enlarged opening. In such a situation, the overall slope of the landscape clearly has a more drastic effect on the deeper traps.

#### B. Quantitative analysis

The exact analytical calculation of the  $\chi''$  relaxation after a field variation within the trap model is beyond the scope of this paper. We have instead worked out a simple approximation which allows us to reproduce the experimental results with two physically meaningful parameters. From Eqs. (4) and (5), the effect of the field is to reduce the lifetime  $\tau$  of a trap to  $\tau'$  such that

$$\tau'(N) = \alpha \cdot \tau(N) = \tau_0 \exp\left\{\frac{B(N) - E(\Delta H, N)}{k_B T}\right\}.$$
 (7)

From Eq. (3), the  $\chi''$  relaxation after the quench follows a  $t^{x-1}$  power law, as can be checked in Fig. 4 (the dotted line corresponds to x = 0.76, which is the value obtained from the analysis of the TRM data<sup>15</sup>). Just after the field variation at  $t_1$ , the reduction of the age from  $t_1$  to  $\alpha \cdot t_1$  alone would produce a  $(t-t_1+\alpha \cdot t_1)^{x-1}$  shape, but such a shape (dashed lines in Fig. 4) does not account well for our measurement. The observed shape indeed shows a rather abrupt restart of the relaxation; it suggests that,



FIG. 3. Schematic picture of the free-energy landscape after a field variation. The magnetically unfavorable metastable states have been tilted, and their depth has diminished; the deeper the trap, the larger its extension, and the stronger the effect of the field perturbation (in agreement with the result in Fig. 2).



FIG. 4. Example data with fits to equations in the text. The dotted line is a power-law fit of the relaxation following the quench [Eq. (3), x = 0.76 from TRM data (Ref. 15)]. The solid lines correspond to our model for the effect of the field perturbation [Eqs. (11), (A1), and (A2)], with fitted values of  $\alpha$  and p; for the last stage of the experiment, the limiting cases [Eqs. (A1) and (A2)] of *full memory* (FM) and *no memory* (NM) are displayed (see Appendix A). The dashed lines are calculated with p=0 in Eq. (11), that is with the only effect of an *age reduction* (" $\alpha$  effect"). They show that the "*p* effect" of a *partial quench* is needed to reproduce the data (the observed departure has been found systematically).

for part of the subsystems, the effect of the field variation is similar to that of the initial quench. This is what would happen if these subsystems were sitting at  $t_1$  in traps which have coupled so strongly to the field that they have been immediately emptied once the field is switched on (the existence of such fragile traps was indeed recently established within the SK model by one of  $us^{16}$ ). A way to account for this effect is to assume that B(N) grows less rapidly than  $E(\Delta H, N) \sim \sqrt{N}$ , in order to obtain that  $\tau'(N)$  in fact decreases for large N (this will be checked for self-consistency in Sec. V). In that case, the traps with sufficiently large N will indeed be completely washed out by the field variation, which corresponds to the observed effect on  $\chi''$ . This occurs beyond a critical size  $N^*$ , which can be defined in the following way: a trap is emptied out if its escape time after the field perturbation is shorter or equal to the "probe time" of the experiment, which is the inverse frequency  $1/\omega$ . Thus,  $N^*$  is defined in such a way that:

$$\tau'(N^*) = \alpha \cdot \tau(N^*) = 1/\omega , \qquad (8)$$

which means in terms of energy depths of traps

$$\frac{1}{\omega} = \tau_0 \exp\left\{\frac{B(N^*) - E(\Delta H, N^*)}{k_B T}\right\}.$$
(9)

The critical size  $N^*$  is associated to a characteristic trapping time  $\tau^*$  which, in the absence of field perturbation, reads

$$\tau^* = \tau(N^*) = \tau_0 \exp\left\{\frac{B(N^*)}{k_B T}\right\}.$$
 (10)

Our approximation simply consists in dividing the set of traps in two families. The first one, corresponding to  $\tau \leq \tau^*$ , is that of the "hard" traps which are only partially affected by the field. They yield a behavior which is characteristic of a reduced age, which we approximate by a mean multiplicative factor  $\langle \alpha \rangle$ , averaged over the family. The second one, such that  $\tau > \tau^*$ , is that of the "fragile" traps, which are completely destabilized by the perturbation. The notion of a threshold size  $N^*$  is reminiscent of the treatment of a field perturbation  $\Delta H$  in the "domain theory" of Ref. 17, in which the authors state that two thermodynamic equilibrium states differing by  $\Delta H$  are indistinguishable up to a certain ( $\Delta H$  dependent) "overlap length" (see also Ref. 18). However, we insist that our definition of  $N^*$  is purely dynamical (and not thermodynamical), and thus explicitly depends on the probe frequency [see Eq. (8)].

We define p as the proportion of subsystems which are in this fragile region at the time  $t_1$  of the perturbation. At time t from the quench, the  $\chi''$  relaxation after a field variation at time  $t_1$  can thus be written as the sum of the two corresponding terms:

$$\chi^{\prime\prime}(\omega,t>t_1) = p \chi^{\prime\prime}(\omega,t-t_1) + (1-p)\chi^{\prime\prime}(\omega,\langle\alpha\rangle t_1 + t - t_1) .$$
(11)

The functional dependence of  $\chi''$  on  $\omega$  and t is determined from the power-law behavior following the quench [see Eq. (3)].

Thus, we summarize the effect of a field variation in two components: an average reduction of the trapping times (concerning what we call hard traps), together with a sudden emptying out of the deepest traps (fragile traps). After the field change, we describe the jump of  $\chi''$  as a redistribution of the population of the traps within an invariant distribution of trapping times. The system is suddenly attracted towards another set of traps, which has the same statistical properties as the previous one; the reduction or temporary extinction of some relaxation times reflects the fact that the system shifts away from the unfavorable region of the configuration space. Then, aging resumes; the new set of traps still has the initial distribution [Eq. (2)]. As time elapses, deeper and deeper traps will progressively be populated again, and the vestiges of the perturbation will slowly become insignificant.

Let us now turn to the third phase of the experiments: at a time  $t_2 (=2t_1$  in our case), the static field has been removed. Figures 1, 2, and 4 show that the  $\chi''$  relaxation is again restarted, and that the effect is slightly weaker than when the field was established at  $t_1$ . Part of the aging during  $t_1$  remains effective after the field cycle; there is a *memory effect*, for which we have estimated upper and lower bounds in Appendix A.

For each of our  $\chi''$  measurements, we have chosen the values  $\langle \alpha \rangle$  and p to fit the data. An example is shown in Fig. 4 (solid lines). The relaxation during the second part of the experiment, when the field is applied, can be satisfactorily reproduced by Eq. (11); concerning the third part, we have only constrained  $\langle \alpha \rangle$  and p by imposing that the experimental curve lies in between the two bounds given by Eqs. (A1) and (A2) of Appendix A. We will discuss later the results on  $\langle \alpha \rangle$ , in comparison with

TABLE I. Values of the proportion p of subsystems in the region of *fragile* traps  $[\tau > \tau^*, \text{ Eq. (10)}]$ , deduced from the three series of  $\chi''$  measurements.  $t_w$  denotes the duration of each phase of the experiment; starting from the quench, the field is applied at  $t_1 = t_w$ , and removed at  $t_2 = 2t_w$ .

ω (Hz)	$t_w$ (min)	H = 5 Oe	H=9 Oe	H = 15 Oe	H=30 Oe
0.1	350	0.01	0.10	0.35	
1	350	0.01	0.02	0.15	0.60
1	35	< 0.01	< 0.01	0.07	

the TRM experiments. The p values are summarized in Table I.

Comparing the two series of results at 1 Hz, one sees in Table I that p increases with the duration of the experiment. Indeed, at fixed field and frequency, the limit  $\tau^*$  [defined by Eq. (10)] above which the traps are fragile is fixed; the longer the waiting time, the larger the number of subsystems which are found in long-lifetime traps, hence the larger the proportion p of them beyond  $\tau^*$ . Also, the correct qualitative behavior as a function of the frequency  $\omega$  is found; at fixed  $t_{\omega}$  and field, a higher frequency yields larger values of  $N^*$  and  $\tau^*$  [see Eqs. (9) and (10)]; the proportion p of subsystems beyond  $\tau^*$  is therefore expected to be lower.

At this point, we have a quantitative estimate of the effect of the field on the aging evolution of  $\chi''$  in the spin glass. We now use the same guideline to analyze the effect of the field in another class of experiments: the relaxation of the thermoremanent magnetization.

## **III. FIELD SCALING OF THE TRM RELAXATION**

In a TRM experiment, the sample is cooled from above  $T_g$  in a given field, and kept at constant  $T < T_g$  under this field during a time  $t_w$ . Then the field is removed, and the decay of the remanent magnetization is measured. The relaxation function is sensitive to the field amplitude; it is faster for a larger field. In a previous paper,<sup>19</sup> we have parametrized the field dependence of the relaxation function. We now reanalyze an extended set of this previous series of data, obtained with the same CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub> sample as for the  $\chi''$  data.

Repeating identical experimental procedure, we have explored field amplitudes ranging from H = 10 to 100 Oe (previous data<sup>20</sup> with H=0.1 Oe, taken with a different setup, cannot be directly included in the present analysis). The measurements have been performed by extracting the sample from the pick-up coils, a method which yields the full value of the magnetization at each point. This enables us to compare not only the shapes but also the amplitudes of the decay curves obtained for various field values. We have normalized the remanent magnetization to its field-cooled (FC) value (indeed, a measurable relaxation of the FC magnetization in another sample has already been quoted,<sup>21</sup> but in our case we have checked<sup>6</sup> that no relaxation of the field-cooled magnetization is found within the accuracy of the present set of TRM measurements).

A set of TRM curves measured at T = 12 K for  $t_w = 30$ 

min and 10–100 Oe field amplitudes is presented in Fig. 5. For increasing field, it appears clearly that the remanent magnetization represents a decreasing fraction of the FC value (11% reduction from 10 to 50 Oe, 27% from 10 to 100 Oe). This effect is larger than the non-linearity of the magnetization,<sup>13</sup> which reduces the FC susceptibility by 3.5% from 10 to 50 Oe and 6.5% from 10 to 100 Oe. Also, the measured relaxation becomes steeper. On a  $t/t_w$  plot like the insert of Fig. 5, it can be seen that the overall qualitative effect of increasing the field is compatible with the age reduction deduced from the  $\chi''$  experiments. This can be checked by trying to adjust the different curves on the same master curve as a function of the modified variable  $t/(\langle \alpha(H) \rangle t_w)$ , where  $\langle \alpha(H) \rangle$  has the same meaning as for  $\chi''$ .

However, the  $\langle \alpha \rangle$  reduction does not give a complete scaling of the curves. One finds that the initial falloff of the amplitude, increasing with the field, is not well reproduced by the  $\langle \alpha \rangle$  reduction, and necessitates the introduction of a second parameter, as was the case for the  $\chi''$ data. We have defined p as the proportion of subsystems located in the region of fragile traps which are washed out by the field variation. At the field cutoff in the TRM experiment, some of these subsystems will indeed jump directly into the region of more favorable magnetization, thus contributing to the initial falloff of the magnetization. We have rescaled the magnetization amplitude by a factor  $\kappa(H)$ , which we qualitatively relate in Appendix B to the parameter p from the  $\chi''$  analysis.

Figure 5 shows this two-parameter scaling of the  $(T=12 \text{ K}, t_w=30 \text{ min})$  set of TRM curves. The continuity of the obtained "master relaxation function" is quite good. What is remarkable is that increasing fields give access to a time region of the master curve which is not easily accessible at lower fields; the reduction of the effective age of the system allows us to measure the relax-



FIG. 5. TRM relaxations at T=12 K for  $t_w=30$  min, with values of the applied field ranging from 10 to 100 Oe. The magnetization has been normalized to the field-cooled value. The insert shows the decay curves as a function of  $t/t_w$ . In the main part of the figure, the curves have been rescaled in order to place them on a unique master curve: the factor  $\alpha$  accounts for the *age-reduction* effect, and the parameter  $\kappa$  for the *partial quench* effect. These parameters are related to those deduced from the  $\chi''$  measurements (see text). In this rescaled plot, the higher-field curves are in continuation of the lower-field ones, giving access to an extended time region.

ation function for very large values of  $t/(\langle \alpha \rangle t_w)$ . The long-time power-law behavior, for instance, is only visible at the very end of the 10–20 Oe curves; but it is fully pictured by the 100 Oe relaxation, which is obviously a power law with the same exponent (slope in the log-log plot of Fig. 5) as that of the end of the low-field curves. This is in agreement with the prediction made in Refs. 11, 12, and 15 that the long-time part of the TRM relaxation is a power law.

We do not have an extensive set of comparable measurements at various  $t_w$  for each of the field values explored. Also, we do not aim to establish here a complete scaling of the TRM data as a function of both field and waiting time, since the  $t/t_w$  scaling is only approximate (Sec. I). We limit ourselves for now to field scaling at fixed  $t_w$ . Figure 5 shows our most complete data set; we have also obtained a similar scaling, although with less data, at 12 K for  $t_w = 350$  min (2 curves) and at 13 K for  $t_w = 30$  and 300 min (3 curves for each). Interestingly, the longer  $t_w$  data favor lower values of  $\langle \alpha \rangle$ ; this corresponds to the fact that the averaging of  $\alpha$  is performed over a set of traps among which the deepest ones are more and more populated for increasing  $t_w$ . Simple arguments within the trap model, or following Parisi,<sup>22</sup> suggest that  $\langle \alpha \rangle$  should be a decreasing function of the reduced variable  $Ht_w^{\lambda}$ . Our data is compatible with a power law of exponent  $\lambda \sim 0.2$  (this determination cannot be very accurate because of the insufficient amount of data).

A systematic study of the TRM decay for various field amplitudes has been recently performed on a Cu:Mn spin glass by Chu, Kenning, and Orbach.23 We could check that all qualitative trends which are found in their results are compatible with ours. They describe a landscape with barriers of all heights very close to the picture considered here. Nevertheless, we believe that the existence of a distribution of trapping times with infinite mean [Eq. (2)] is a crucial point for a quantitative description of aging since it is at the heart of the most visible evidence of aging, which is that after  $t_w$  the characteristic relaxation times of the system are of the order of  $t_w$  itself. The picture of the effect of the field as a "Zeeman tilting of the phase space" has indeed emerged from discussions with these authors; our present point, however, is that all traps are not equally affected by the field change.

The effect of the field amplitude on the relaxation of the zero-field-cooled magnetization has also been investigated by Djurberg, Mattson, and Nordblad.<sup>24</sup> The authors mainly study the logarithmic derivative of the relaxation, which is a good approximation to the distribution of response times.<sup>3</sup> This quantity is shown to peak at shorter times for sufficiently large fields, in qualitative agreement with our conclusion of an age-reduction by a factor  $\langle \alpha(H) \rangle$ .

## IV. COMPARISON OF THE RESULTS FROM TRM AND $\chi''$ MEASUREMENTS

We first consider the mean reduction factor  $\langle \alpha \rangle$ . The quantity has the same meaning in both experiments, and can be used to quantitatively specify the Zeeman cou-

pling  $E(\Delta H, N)$  of the field to the N spins associated with a  $\tau$  trap.

Figure 6 presents a set of  $\langle \alpha \rangle$  values deduced from both kinds of measurements. Within the error bars, it is clear that a behavior which is common to both experiments is found.  $\langle \alpha \rangle$  is an average value over the family of hard traps visited after  $t_w$ ; this average value yields a typical size  $\tilde{N}$ , characteristic of the order of the magnitude of the relevant hard traps:

$$\ln\langle\alpha\rangle = E(\Delta H, \tilde{N})/k_B T .$$
<sup>(12)</sup>

We have assumed in Eq. (6) that the energy depth variation  $E(\Delta H, \tilde{N})$  of the trap for a field change  $\Delta H$  is proportional to  $\Delta H$ . Figure 6 shows that the approximation works well up to around 50 Oe; in that range, the magnetic moment can be considered independent of the field. This is obviously a low-field approximation; for higher fields, the interactions locking the relative orientations of the spins in a random way should no longer be dominant, and an extensive polarization of the N spins is expected. The magnetization of a set of N spins will then grow faster than  $\sqrt{N}$ , presumably proportionally to  $N\Delta H$  (or slightly slower than  $\Delta H$  due to nonlinearities), thus leading to  $E(\Delta H) \propto \Delta H^2$ . This is indeed the general trend which can be seen in Fig. 6: above 50 Oe,  $\ln\langle \alpha \rangle$  varies faster than linearly with the field.

The slope of  $\ln\langle \alpha \rangle$  versus  $\Delta H$  is  $\sqrt{N}m\mu_B/k_BT$  [Eqs. (4) and (6)]. Let us now specify the meaning of the elementary magnetic moment  $m\mu_B$ . From the study of the susceptibility of spin glasses above  $T_g$ , it is known (see Ref. 14 and references therein) that the elementary magnetic objects which produce the paramagneticlike behavior are not individual spins, but renormalized groups of spins. This has been observed in different samples by different laboratories; in the case of our sample,<sup>13,20</sup> the measured Curie constant indicates a magnetic moment equivalent to  $\sim 60$  ferromagnetically coupled  $Cr^{3+}$  ions as basic renormalized entities. Using this value, we find  $\tilde{N} = 670$ , which means that escaping from the hard traps typically involves flipping of the order of 700 elementary groups of  $Cr^{3+}$  ions. We can now check afterwards the validity of the linear approximation in Eq. (6); the energy  $\sqrt{N}m\mu_B H$  becomes of the order of the mean interaction energy  $k_B T_g$  for  $H \approx 45$  Oe ( $T_g = 16.7$ 



FIG. 6. Field dependence of the parameter  $\alpha$  [Eq. (4)] from both  $\chi''$  (open circles) and TRM (full diamonds) experiments at 12 K. The solid line is a fit to a linear dependence of the Zeeman energy  $\ln \alpha$  with field [Eqs. (4) and (6)]. Departures from this approximation are expected above ~45 Oe (see text).

K). It is therefore natural that the linear approximation fails above this field range.

We give in Appendix B a brief discussion of the comparison between the values of the scaling factor  $\kappa$ , obtained from the initial falloff in the TRM decay, with the parameter p characterizing the "partial quench effect" in the  $\chi''$  experiments. We now analyze the set of  $p(\chi'')$  results, which allows us to specify the range of  $\tau^*$  and  $N^*$ values which are explored in the measurements.

p is the proportion of subsystems in traps greater than  $\tau^*$  at the time  $t_w$ . In the  $t_w < \tau^*$  limit, one finds, within the trap model, that p is given to first approximation by

$$p \approx \frac{\sin \pi x}{x \, \Gamma(x+1)} \left\{ \frac{t_W}{\tau^*} \right\}^x \,, \tag{13}$$

where  $\Gamma$  is the Euler gamma function. More accurately, we have estimated  $\tau^*$  from our fitted values of p by using an expansion up to terms of order  $(t_w/\tau^*)^{x+2}$ . Interestingly, at this stage, without any assumption concerning either  $\tau_0$  or the dependence of  $\tau$  (or B) upon N, we can estimate  $N^*$  from the values of  $\tau^*$ : from Eqs. (6), (9), and (10), we may write

$$N^* = \left[\frac{k_B T}{m\mu_B H} \ln \omega \tau^*\right]^2.$$
(14)

The  $\tau^*$  and  $N^*$  results are summarized in Table II. Note that the logarithmic dependence of  $N^*$  on  $\tau^*$  means that our assumption on the relation between p and  $\tau^*$  is not too crucial for our later conclusions. Table II shows that  $N^*$ , of the order of  $10^{5-6}$  groups of renormalized spins is much larger than the typical number of spins in hard traps  $\tilde{N}$ , as it should be, and remains smaller than the number of spins within a micron size grain  $\sim 10^{10}$ . Experiments on mesoscopic samples of about 107-8 spins should thus display interesting modifications of the aging behavior.  $\tau^*$  is found of the order of 100 times  $t_w$  for H=5 Oe, and rapidly approaches the experimental time window as H increases above a few tens of Oe. This is in agreement with the experimental fact that the aging phenomena in TRM relaxations are insensitive to the field amplitude in the 0.1-20 Oe range;<sup>19</sup> the observed relaxation itself does indeed depend on the field amplitude, mainly becoming faster for increasing field, but the effect of aging on the dynamic properties [as, e.g., parametrized by  $\mu$  in Eq. (1)] remains the same. Beyond this low-field range, departures have been previously quoted;<sup>19</sup> these may also be correlated with the appearance of nonlinear effects.13

One may wonder whether the influence of a field variation, which is shown here to produce a sudden depopulation of the traps beyond  $\tau^*$ , could also be responsible for the observed systematic deviations<sup>5,6,15</sup> from a pure  $t/t_w$ scaling. We show here that  $\tau^*$  presents a very rapid variation with the field; if this effect was to be at the origin of the fact that  $\mu$  [Eq. (1)] is lower than 1,  $\mu$  should go closer and closer to 1 as the field is reduced. In polymer glasses,<sup>1</sup> a gradual decrease of  $\mu$  as the stress increases is observed, but very low stress data are lacking and the question of the zero-stress limit of  $\mu$  is not settled. In our

TABLE II. Values of  $\tau^*$  and  $N^*$  [from Eqs. (13) and (14)], calculated from the values of p which have been fitted to the different  $\chi''$  measurements.

$\omega$ (Hz) $t_w$ (sec)		$\tau^*(H)$ (sec)			$N^*(H)$				
		5	9	15	30	5	9	15	30
0.1	$2.1 \times 10^{4}$	$1.9 \times 10^{6}$	$1.0 \times 10^{5}$	$2.4 \times 10^{4}$		$3.9 \times 10^{6}$	6.8×10 <sup>5</sup>	$1.8 \times 10^{5}$	
1	$2.1\!\times\!10^4$	$1.9 \times 10^{6}$	$7.8 \times 10^{5}$	$6.1 \times 10^{4}$	$1.5 \times 10^{4}$	$5.5 \times 10^{6}$	$1.5 \times 10^{6}$	$3.5 \times 10^{5}$	$6.7 \times 10^{4}$
1	$2.1\!\times\!10^3$	$> 1.9 \times 10^{5}$	$> 1.9 \times 10^{5}$	$1.6 \times 10^{4}$		$> 3.9 \times 10^{6}$	$> 1.2 \times 10^{6}$	$2.7  imes 10^5$	

case, we have recalled above numerous TRM results where no sign of a  $\mu$  approaching 1 at very low fields [as low as 0.1 Oe (Refs. 5 and 20)] was found. Thus, the systematic departure from a pure  $t/t_w$  scaling is likely to be related to another important feature of experimental spin glasses. We briefly recall how these deviations can be interpreted<sup>15</sup> in the context of the trap model.<sup>11</sup> If the number of traps per subsystem is large but finite, and distributed with some typical value  $N^{t}$ , then those subsystems smaller than  $N^t$  will eventually reach ergodicity within the time of the measurement: their dynamics will no more depend on  $t_w$  ("interrupted aging"). As shown in Ref. 15, this produces an effective scaling very close to the one given in Eq. (1), with  $\mu < 1$ , and a good collapse of the TRM curves for various  $t_w$  and two different samples. The result is a value of a typical time  $t_{erg}$  needed to reach ergodicity in a significant fraction of the system, of the order of a few  $10^6$  sec (tens of days) in the region  $0.6-0.9T_g$ . Of course, this does not mean that aging suddenly disappears after  $t_{erg}$ , which must be understood as a crossover time scale. Thus, since the experiments indicate that this latter effect ( $\mu < 1$ ) persists in the limit of vanishing fields, we conclude that both kinds of crossover time scales are found in spin glasses: the effect of the field amplitude on the dynamics only appears for sufficiently high fields, such that the threshold value  $\tau^*(H,\omega)$  becomes smaller than  $t_{\rm erg}$ .

## V. SIZE DEPENDENCE OF THE FREE-ENERGY BARRIERS

We have obtained that a typical number N of (possibly renormalized) spins should be associated to a trapping time  $\tau$ ; N is the typical number of spins which must be flipped for escaping from the trap. This allowed us to quantify the Zeeman coupling of the field to the traps. We have now determined a set of coupled  $\tau^*$ , N\* values (Table II); we can tentatively use them to determine—or at least constrain—the possible shape of  $\tau(N)$ , i.e., of the corresponding free-energy B(N), together with the acceptable range for the attempt time  $\tau_0$  [Eq. (5)]. [Note that Eq. (14) does *not* give the explicit dependence of  $\tau$  on N, due to the presence of H.]

A natural assumption for B(N), inspired from Fisher and Huse's *droplet* description,<sup>18</sup> is

$$B(N) = E_0 N^{\nu} , \qquad (15)$$

where  $E_0$  fixes the energy scale, and v is some exponent which, as stated in Sec. II B, is expected to be smaller than 1/2 so that  $\tau'(N)$  becomes a decreasing function of

N for large N. From the arguments of Fisher and Huse,<sup>18</sup> one should expect 0.06 < v < 0.66 (the lower bound being a reasonable guess. Note that v is equivalent to  $\psi/3$  in Ref. 18). We have fitted the shape Eq. (15) to the  $\tau^*, N^*$ values deduced from the  $\chi''$  measurements. Despite the low number of points entering the fit (3+4, correspond)ing to the first two lines in Table II), some general trends can be observed. First, we have fixed  $\tau_0$  to the microscopic value of  $10^{-12}$  sec. This yields  $E_0 = 26k_BT$ , a rather large value for the reversal of the elementary magnetic entities (N=1). However, the exponent is found to have the remarkably low value v=0.03; this allows the exploration of a large range of  $N(1 \rightarrow 75000)$  within the experimental time window. The tendency to low values for v persists if  $\tau_0$  is set to 1 sec instead of being microscopic, which yields v=0.10 and  $E_0=3k_BT$ . Thus in any case, we find that the time scale  $\tau(N=1)$  for elementary processes is macroscopic (0.1 to 10 sec) rather than microscopic.

The small value of v suggests that the power law [Eq. (15)] could as well be a logarithmic dependence, as suggested by Rieger:<sup>8</sup>

$$B(N) = E_1 \ln(N/N_0) . (16)$$

We cannot discriminate between Eqs. (15) and (16); indeed, we only know a set of  $N(\tau)$  values in the limited region of  $N \sim 10^{5-6}$ . The logarithmic fit gives  $E_1 = 1.25k_BT$ , independently of the choice of  $\tau_0$ , which in this formulation is only correlated to the value of  $N_0$ . Trying  $\tau_0 = 10^{-12}$  sec yields the completely unphysical value  $N_0 = 1.10^{-8}$ , whereas a more likely value  $N_0 = 1$  is obtained for  $\tau_0 \approx 0.01$  sec ( $\tau_0 = 1$  sec corresponds to  $N_0 = 43$ ). Again, we are led to consider that the characteristic time for the elementary processes relevant to our experimental time window is in the micro- rather than the microscopic time scale.

This is a very interesting point, which raises the question of what "trap" really means. All along this paper, we have sketched the spin-glass behavior (at a fixed temperature) as a random walk among a *single type* of trap. On the other hand, we know that another class of experiments,<sup>6</sup> in which slight temperature variations are applied during aging, show that the metastable states are hierarchically organized, and that the bottom nodes of the hierarchical tree develop into new branches as the temperature is lowered. The experiments analyzed here have been performed at constant temperature after the quenching procedure; in that case, only one level of the hierarchical tree is explored, whereas the temperature variation experiments give access to higher or lower levels in the tree. At a fixed temperature, the system wanders among the "bottom nodes" of the tree as described in the trap model; these "bottom nodes" are not elementary configurations, but rather free-energy valleys involving a larger number of configurations, which can subdivide into other valleys at lower temperatures—see the recent discussion given in Ref. 12. The attempt time  $\tau_0$  corresponds to elementary jump processes between the bottom nodes; we interpret the macroscopic value that we obtain as indicating that the bottom nodes at a given temperature proceed from the renormalization of the lower branches of the tree (which, otherwise, would only be revealed by decreasing the temperature).  $\tau_0$  can thus be understood as a renormalized value; it may therefore be of a macroscopic order of magnitude.

We have thus tentatively obtained the dependence of the trapping time  $\tau(N)$  upon a corresponding number of elementary magnetic objects N, and also the fashion in which a  $\tau$  trap is modified by a field variation. Figure 7 displays, using Eq. (16), the variation of the energy depth of the traps due to a field variation of a given amplitude, as a function of N.

### **VI. CONCLUSION**

In this paper, we have addressed the question of the effect of the magnetic field on the free-energy landscape of a spin glass. The aging phenomena contain much information about the complex geometry of this landscape, since the evolution of the macroscopic properties reveals the slow spreading of the system among valleys and mountains, towards the distant horizon of equilibrium. Most experimental investigations involve a field variation; however, the ac susceptibility is close to the limit of zero applied field, and we have used this highly sensitive probe to study the effect of an additional field variation, with an amplitude comparable to that used for TRM decay experiments. We have made a direct and quantitative comparison of both classes of experiments.

Aging, together with its main consequence of an ap-



FIG. 7. After a given field variation  $\Delta H$  (=5, 9, 15, or 30 Oe), dependence of the depths of the traps  $\Delta E = B(N) - E(\Delta H, N)$  [Eq. (7)], in units of  $k_B T$ , on the corresponding number of spins N (decimal logarithm scale). We have used the logarithmic dependence of B(N) [Eq. (16)] with  $\tau_0$ =0.01 sec. The dashed lines stand for the typical value  $\Delta E(N^*) = k_B T \ln(1/\omega \cdot \tau_0)$  [Eq. (9)]. In a  $\chi''$  experiment at frequency  $\omega$ , this typical value defines the cross over  $N^*$  between hard  $(N < N^*)$  and fragile  $(N > N^*)$  traps.

proximate  $t/t_w$  or  $\omega \cdot t$  dependence of the dynamics, can be usefully described by a random walk in a wide distribution of traps.<sup>11,15</sup> We have used this picture as a guideline to interpret our experimental results. From the relaxation of  $\chi''$  during aging, and from the modification of this relaxation when an additional dc field is changed, we find that the effect of the field is twofold. Firstly, all traps have their depth reduced by a Zeeman energy term E, which helps to escape towards a region of more favorable magnetization. Secondly, the depth reduction is more important for the deeper traps. Thinking of the perturbation as a general tilt of the landscape, this assertion suggests that deeper traps have a larger extension in the configuration space: the larger opening of the deeper traps will make them more sensitive than the smaller ones to a given tilt angle.

This selective coupling of the field to the traps has led us to associate to a given trap depth B a typical number N of spins (or basic renormalized entities). The Zeeman term E is found proportional to the field H, up to  $\sim 50$ Oe; it is likely to represent the coupling to the field of frustrated clusters of frozen magnetic moment, such that  $E \propto \sqrt{N}H$ . The shape of the restart of the  $\chi''$  relaxation after a field change shows a partial quench effect: from this observation, we infer that some of the deepest traps are immediately emptied by the field change, or in other words that the field-perturbed trap depth B(N)-E(N) becomes a decreasing function of N for large N. We thus describe the  $\chi''$  results with the help of two fitting parameters: an average  $\langle \alpha \rangle = \langle \exp(-E(N)/k_BT) \rangle$  of the Zeeman effect on the small-N traps which are only partially perturbed (hard traps). This corresponds to a mean reduction of the effective age at the time of the perturbation from  $t_1$  to  $\langle \alpha \rangle t_1$ . The second parameter is the proportion p which is the requenched population from the region of unfavorable large-N traps which are completely washed out (fragile traps). The limit between both families is found for trapping times of the order of  $10^{5-6}$  sec for  $H \sim 10$  Oe.

We have analyzed along the same line a set of TRM experiments performed with various field amplitudes (10-100 Oe). We find that the effect of increasing the field amplitude is actually to reduce the effective age by the same factor  $\langle \alpha \rangle$  as found in  $\chi''$ ; the  $t/t_w$  scaling variable is to be replaced by  $t/(\langle \alpha \rangle t_w)$ . The contribution of the fragile traps also appears; it is seen as a slight increase of the initial falloff of the magnetization for increasing fields. Let us remark that, in the case of the TRM experiments, the measurement is only sensitive to magnetization changes, and ignores further aging among deeper and deeper traps, which will however continue within an unchanged distribution of trapping times.

We have been able to rescale the set of TRM curves measured at various fields (but fixed  $t_w$ ) on the same master curve.<sup>14</sup> For different  $t_w$  values, more accurate assumptions than the simplified  $t/t_w$  scaling should be used; this is beyond our present purpose. The analysis of the parameters fitted to both  $\chi''$  and TRM allows us to specify the function B(N), which expresses the size dependence of the trapping times. The trap depths vary as a power law of N with a small exponent, or can be represented as a logarithmic growth:  $B(N)=1.25k_BT\ln(N/N_0)$ . In the latter case, the physical condition that  $N_0 > 1$  implies that the attempt time  $\tau_0$  associated with thermally activated escape from the traps is macroscopic, namely  $\tau_0=0.01$  sec for  $N_0=1$ .

Indeed, the present stage of the trap model<sup>11,15</sup> which we have used here is that of a "one-level tree," which we know to be insufficient to account for the effect on aging of temperature variations, or even the existence of stationary dynamics. The temperature variation experiments support a hierarchical organization of the metastable states.<sup>6</sup> A recent extension<sup>12</sup> of the trap model to a Parisi-tree-type organization of the states shows that, at a given temperature, the relevant metastable states for aging correspond to a given level of the tree; these states indeed consist in sets of hierarchically lower-level states, among which ergodicity can be reached. We see in the appearance of a macroscopic value for  $\tau_0$  an indication that the basic thermal jumps are actually occurring among renormalized sets of lower-level states.

Our data analysis allows for the rescaling of TRM relaxations for various fields onto the same graph. In our picture, the field appears capable of extending the experimental time window in which the relaxation is explored; as a function of the reduced scaling variable, the 100 Oe curve is clearly the continuation for longer observation times of the curves at lower fields. Meanwhile, increasing the field brings closer and closer to the experimental times the crossover time scale  $\tau^*$  due to the fragile traps. The limit of higher fields has been known for a long time from the pioneering TRM studies:<sup>25</sup> aging disappears. Thus, slight field variations teach us instructive subtleties of the spin-glass phase, but stronger perturbations erase all this information. Indeed, the mechanics of glassy polymers<sup>1</sup> have already displayed very similar phenomena: weak stresses reveal aging in the slow strain of the materials, whereas high stresses have been quoted<sup>1</sup> to erase the effect of previous aging.

#### ACKNOWLEDGMENTS

We would like to thank F. Lefloch and M. Ocio for their participation in the experiments, and also for numerous stimulating discussions all along this work. This work was initiated after discussions with R. Orbach; we are grateful to him for his interest in this work, and also to D. Chu, with whom fruitful exchanges have allowed a constructive comparison of the results on different samples.

### APPENDIX A: MEMORY EFFECT AT THE END OF THE FIELD CYCLING

When the field was applied at  $t_1$ , the hard traps had their lifetime reduced by  $\langle \alpha \rangle$ ; some of the subsystems in this region may have stayed within these magnetically unfavorable traps when, at  $t_2$ , the field has been removed. These traps now have a favorable magnetization, and aging will continue among them. For these memorykeeping subsystems, there is no perturbation at  $t_2$ . We do not intend to calculate this effect in all details, but we



FIG. 8. Schematic diagram demonstrating the full-memory (FM) and no-memory (NM) scenarios for field-cycling experiments.

state what can be expected in two limiting cases of

(i) full memory (FM): none of the (1-p) subsystems is perturbed at  $t_2$ . ( $\iff$  effect of aging during  $[t_1, t_2]$  negligible: subsystems in hard traps at  $t_1$  are still in the same trap at  $t_2$ .)

(ii) no memory (NM): all of the (1-p) subsystems are perturbed at  $t_2$ , as at  $t_1$ . ( $\Leftrightarrow$  the effect of the field perturbation at  $t_1$  has been forgotten: all subsystems in hard traps at  $t_1$  have decayed into new traps before  $t_2$ .)

These two limiting hypotheses can be sketched in the way shown in Fig. 8, yielding simple formulas for  $\chi''$ : (ii) FM case

$$\chi''(\omega, t > t_2) = p^2 \chi''(\omega, t - t_2) + p(1-p)\chi''[\omega, \langle \alpha \rangle (t_2 - t_1) + t - t_2] + (1-p)\chi''(\omega, t) ;$$
(A1)

(ii) NM case

$$\chi^{\prime\prime}(\omega,t>t_{2}) = p\chi^{\prime\prime}(t-t_{2})$$

$$+p(1-p)\chi^{\prime\prime}[\omega,\langle\alpha\rangle(t_{2}-t_{1})+t-t_{2}]$$

$$+(1-p)^{2}\chi^{\prime\prime}[\omega,\langle\alpha\rangle(\langle\alpha\rangle t_{1}+t_{2}-t_{1})$$

$$+t-t_{2}]. \qquad (A2)$$

### APPENDIX B: COMPARISON BETWEEN $\kappa$ (TRM) AND $p(\chi'')$

The scaling factor  $\kappa$ , obtained from the TRM results, accounts for the field dependence of the "almost instantaneous" falloff of the magnetization after the field cutoff. This initial falloff is the sum of all relaxation processes occurring before the first measurement, which in that case is performed after  $\sim 5-10$  sec. In terms of traps, these processes can indeed be associated with the p proportion of the subsystems which are in these fragile traps that are completely washed out by the field change. But the two experiments give in fact access to different quantities;  $\chi''$  essentially measures the number of subsystems which, after having been kicked out of the fragile traps, restart aging and are therefore found in short lifetime traps of order  $1/\omega$ . On the other hand, the TRM decay measures a magnetization decrease: only those of these restarting subsystems which go to a region of different magnetization are contributing, and also at time t all processes having occurred before t are integrated. Therefore, it is not straightforward to make a detailed comparison of both quantities  $\kappa$ (TRM) and  $p(\chi'')$ ; we can discuss them at least qualitatively. Using the previous parametrization of the TMR relaxation with the trap model,<sup>11</sup> we write that the TRM value for H=20 Oe is  $m_0=0.5$ (in units of  $m_{\rm FC}$ ) at time zero after the field cutoff, the  $\kappa$ scaling thus yields the values of the initial falloff  $\Delta m_I = 1 - m_0 / \kappa$ , which are listed in Table III. Table III shows that, as expected from the above remarks, the relative variation of  $\Delta m_I$  with the field is much weaker than that of  $p(\chi'')$  (see Table I). A crude comparison of their overall behavior shows that

$$\Delta m_I \cong (0.03 \text{ to } 0.10) p(\chi'') + 0.46$$
. (B1)

In this expression, 0.46 is thus the zero-field limit of  $\Delta m_I$ ;

TABLE III. Results of the  $\kappa$  scaling for TRM measurements at 12 K and  $t_w = 30$  min.  $\kappa = 1$  is arbitrarily fixed at the lowest field.  $\Delta m_I$  is the corresponding value of the initial falloff of the TRM, in units of the field-cooled magnetization  $m_{\rm FC}$ .

H (Oe)	10	20	50	100
κ	1	1.02	1.07	1.08
$\Delta m_I = 1 - m_0 / \kappa$	0.46	0.48	0.50	0.51

in addition, Eq. (B1) indicates that 3-10% of the subsystems which escaped from the fragile traps at the field change will change their magnetization within the first few seconds. The comparison cannot be pursued in more details without a complete analytical formulation of the model, which is not our present purpose.

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