# Dynamics of the spin-glass freezing in Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te

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We report on dynamic magnetic properties at low temperatures (4.2 < T < 30 K) of the disordered Heisenberg frustrated antiferromagnet Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te over an extended range of frequencies or of corresponding observation times  $(10^{-6} < t_{obs} < 3 \times 10^3 \text{ s})$ . At high frequencies, the real  $(\chi'_{\nu})$  and imaginary  $(\chi''_{\nu})$  parts of the susceptibility are determined from accurate Faraday-rotation experiments while the dynamics for longer characteristic times are investigated in a weak field, through superconducting quantum-interference device (SQUID) measurements of the magnetization. Starting from two different dynamic criteria, (i) the significant change of the relaxation of the in-field magnetization to step variations of the temperature and (ii) the appearance of long-time thermoremanent magnetization relaxation, we deduce a value  $T_{f0} = 12.9 \pm 0.1$  K for the freezing temperature. Fortunately, it also corresponds to the coalescence between the temperature of appearance of irreversibilities and that corresponding to the maximum of the ac susceptibility at very low frequencies. The Vogel-Fulcher law is unable to describe the dynamics over so large a frequency range. Thus, the spin freezing has been analyzed in terms of a critical slowing down above the static freezing temperature  $T_{f0}$  using the power law  $\tau/\tau_0 = A [(T - T_{f0})/T]^{-zv}$  with reasonable values of  $\tau_0 = 3.8 \times 10^{-14}$  s and of the dynamic critical exponent zv' = 9.7. This exponent is consistent with simulations obtained recently for three-dimensional Ising spin-glasses. We get independently a similar estimation of  $T_{f0}$  by other approaches. We also determined the (H, T) magnetic phase diagrams of  $Cd_{0.6}Mn_{0.4}$ Te for different values of  $t_{obs}$  up to 100 s. The field dependence of the temperature corresponding to the onset of irreversibilities, even for long observation times, looks like a Gabay-Toulouse line, associated with a transverse spin freezing. This behavior, uncommon for classical spin-glasses, is assumed to be related to the particular frustrated structure in this Heisenberg disordered antiferromagnet.

### I. INTRODUCTION

Among insulator spin-glasses, with spins coupled antiferromagnetically and localized in a frustrated lattice,  $Cd_{1-x}Mn_xTe$  has become the archetypical material.<sup>1</sup> The magnetic properties of  $Cd_{1-x}Mn_xTe$  are reviewed in Ref. 2. For x > 0.3, the magnetic interactions responsible for the spin-glass freezing are short range.<sup>3</sup> In this case, it is necessary to avoid any nonstatistical chemical inhomogeneity in the Mn spin distribution over the lattice. Such extrinsic chemical fluctuations, when they exist, are responsible for magnetic clustering effects which play a more important role than in metallic spin-glasses where the frustration is more spread out over the whole lattice, as a result of the longer range of the magnetic interactions.

Improvements in the crystal-growing procedure now make possible the preparation of samples for which the magnetic ions may be considered as randomly distributed over the lattice, the magnetic inhomogeneities being those intrinsic in nature. In such samples, the spin-glass freezing is clearly observed.<sup>4-7</sup> In particular, we have already reported data on the static and ac magnetic susceptibility, remanent magnetization, and nonlinear magnetization of  $Cd_{0.6}Mn_{0.4}Te^{.6,8,9}$ 

In this paper, we complete the dynamic study of the same homogeneous sample. The in-phase and out-ofphase components of the ac magnetic susceptibility  $\chi_{\nu}$ have been simultaneously measured in the frequency range 1.5 Hz  $< v < 8 \times 10^4$  Hz. Since the spin dynamics become slower as one approaches the static spin-glass freezing temperature from above, it is desirable to explore the spin relaxation at lower frequencies, or equivalently at longer observation times tobs corresponding to  $t_{\rm obs} \equiv (2\pi v)^{-1}$ . For this purpose, dynamic magnetization measurements have been performed in the time range  $0.3 < t_{obs} < 3 \times 10^3$  s. All the experimental results, put together, allow us to investigate the spin dynamics of the system over nine decades of observation times  $10^{-6} < t_{obs} < 3 \times 10^3$  s. This study is finally extended to the determination of the time-dependent magnetic phase diagram in the (H, T) plane under weak magnetic fields Η.

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# **II. EXPERIMENTAL TECHNIQUES**

Faraday rotation is a powerful technique to study dynamical properties of insulating spin-glasses over an extended time or frequency range.<sup>10</sup> In the present case we checked that, under weak magnetic field (H < 100 Oe), the Faraday rotation is proportional to the magnetization.

We have performed low-temperature measurements with an exchange-gas cryostat in the 4.2 < T < 30 K range. The Faraday rotation has been measured by using an attenuated He-Ne laser light beam (~1  $\mu$ W/mm<sup>2</sup> at  $\lambda = 6328$  Å), for which the sample is transparent. The sensitivity of the polarimeter is rather high since one uses a linear detection method associated with a modulation technique. The sample is cut as a nicely polished plate of 1.86 mm in thickness. The state of polarization of the light is changed by a photoelastic modulator working at a frequency f = 50 kHz before crossing the sample. Between polarizers at 45° from each other one can show that the 2f component of the light intensity detected by the photomultiplier is proportional to the Faraday rotation. A lock-in detection at 2f is used to measure a signal proportional to  $\sin(2\theta)$ , where  $\theta$  is the Faraday rotation angle. The dc part of the current on the photomultiplier is maintained automatically constant in order to be insensitive to light-beam fluctuations.

In magnetooptical measurements of the susceptibility at low frequencies (v < 1 kHz) two successive lock-in detections are used, the first at twice the photoelastic modulator frequency (2f) and the second on the ac magnetic field modulation ( $h \sim 1$  Oe) applied to the sample at a slower frequency v. The output signal is then proportional to the Fourier component of  $\theta$  at frequency v. Inphase and out-of-phase signals are directly proportional to the real ( $\chi'_v$ ) and imaginary ( $\chi''_v$ ) parts of the ac magnetic susceptibility, respectively. At higher frequencies (v > 1 kHz), the photoelastic modulator is removed, and the component of the photomultiplier signal at frequency v is directly measured by a single lock-in detection.

Very accurate long-time magnetization measurements have been performed at Uppsala with a sensitive SQUID magnetometer utilizing the SHE model No. 30 electronic control system.

### **III. SPIN DYNAMICS**

### A. Experimental results

We have already reported ac susceptibility data in the frequency range  $0.6 < v < 8 \times 10^4$  Hz.<sup>6</sup> In order to study dynamics at longer characteristic times, the zero-field-cooled (ZFC) magnetization curves  $M_{ZFC}(T)$  versus temperature have been measured at different observation times  $(0.3 < t_{obs} < 3 \times 10^3 \text{ s})$  following the application of a weak magnetic field (10 Oe). The results are reported in Fig. 1. As found in other spin-glasses, the temperature  $T_f$  corresponding to the cusp of  $\chi'_v(T)$  or  $M_{ZFC}(T)$  curves increases slightly with v (or  $t_{obs}^{-1}$ ).

The out-of-phase component of the ac susceptibility  $\chi_{\nu}''(T)$  has been also investigated in the whole range



FIG. 1. Zero-field-cooled magnetization as a function of temperature (dashed curves) for different observation times:  $t_{obs} = 0.5 \text{ s} (+), 3 \text{ s} (\odot), 30 \text{ s} (\times), 300 \text{ s} (\Box), and 3000 \text{ s} (\triangle)$ measured with the SQUID magnetometer (H = 10 Oe). The solid curve corresponds to the field-cooled magnetization  $M_{FC}$ (H = 10 Oe) scan at a cooling rate of 0.5 K/min.

 $1.5 < v < 8 \times 10^4$  Hz. We report in Fig. 2 typical data for  $\chi'_{\nu}(T)$  and  $\chi''_{\nu}(T)$  measured at a mean typical frequency. From our data, and as usually found in other spin-glasses, we may assume that the inflection point on the  $\chi''_{\nu}(T)$  curve stands at  $T_f(v)$ . In order to perform a more quantitative analysis of our data we have determined the quantity  $\varepsilon_{\nu} = \arctan(|\chi''_{\nu}|/\chi'_{\nu})$ , which measures the magnetic losses.<sup>11</sup> Typical  $\varepsilon_{\nu}(T)$  curves obtained at several typical frequencies are reported in Fig. 3. The maximum of  $\varepsilon_{\nu}(T)$  is located at a temperature close to that of  $\chi''_{\nu}(T)$  and slightly smaller than  $T_f(v)$  at all frequencies. These results are in a close agreement with those reported recently for  $Cd_{1-x}Mn_xTe$  (x = 0.45, 0.55) (Ref. 7) and for  $Hg_{1-x}Mn_xTe$ .<sup>12</sup>



FIG. 2. Experimental data of the temperature dependence of the in-phase  $(\chi'_{\nu})$  and out-of-phase  $(\chi''_{\nu})$  components of the ac magnetic susceptibility at 2.9 kHz.  $\chi'_{\nu}$  and  $\chi''_{\nu}$  are plotted in the same units.



FIG. 3. Temperature dependence of  $\varepsilon_{\nu \simeq} |\chi_{\nu}''| / \chi_{\nu}'$  deduced from ac susceptibility data for different values of the frequency.

For longer observation times, we have extended our study by magnetization measurements, starting from the general relation:<sup>13,14</sup>

$$\chi_{\nu}^{\prime\prime} = -\frac{\pi}{2} \frac{\partial \chi_{\nu}^{\prime}}{\partial \ln(2\pi\nu)} . \tag{1}$$

Assuming the absence of nonlinear field effects up to the field H = 10 G used in SQUID experiments, we can write  $\chi'_{\nu} = M_{ZFC}/H$ . Then this relation can be written under the following form:

$$\chi_{\nu}^{\prime\prime} = \frac{\pi}{2} \frac{1}{H} \frac{\partial M_{\rm ZFC}}{\partial \ln t_{\rm obs}} \,. \tag{2}$$

The quantity  $S_v = H^{-1} \partial M_{ZFC} / \partial \ln t_{obs}$  has been deduced from ZFC measurements for  $0.5 < t_{obs} < 3 \times 10^3$  s (Fig. 4).



FIG. 4. Plot of  $S = H^{-1} \partial M_{ZFC} / \partial \ln t_{obs}$  as a function of temperature for different values of the observation time  $t_{obs}$ , deduced from zero-field-cooled magnetization  $M_{ZFC}$  curves reported in Fig. 1 (same symbols are used for each  $t_{obs}$ ).

As expected, the value of  $T_f(v)$  deduced either from the maximum of  $\chi'_v(T)$  at high frequencies or from the inflection point of  $S_v(T)$  at low frequencies (or long observation times) have a common behavior as shown in Fig. 5.

At a given frequency, irreversible effects appear at a temperature  $T_i(v)$ , defined from the onset of  $\chi''_v$ , which is a few degrees higher than  $T_f(v)$ . However, the frequency dependencies of these two characteristic temperatures are related together. As shown by Bontemps *et al.*,<sup>11</sup> the critical dynamics are better related to  $T_i(v)$ . For each value of v (or  $t_{obs}$ ) we define the temperature  $T_i(v)$  at which  $|\chi''_v| / \chi'_v$  is equal to a small constant value; it is of course limited by the signal-to-noise ratio. In the present case this constant has been chosen equal to  $10^{-3}$  and the corresponding results are shown in Fig. 5.

An important question arises: is there a limit for  $T_f(v)$ when v tends to zero? From the response of the fieldcooled magnetization to step variations of the temperature it is possible to estimate the static freezing temperature  $T_{f0}=12.9\pm0.1$  K.<sup>9</sup> This value is consistent with the apparition of very long times in the relaxation of the thermoremanent magnetization (TRM).<sup>8</sup> Field-cooled (FC) measurements, even realized at a slow cooling rate, do not allow us to determine the equilibrium magnetization state. This is well illustrated in Fig. 1 where the ZFC magnetization measured at long observation times may be larger than FC magnetization obtained at a higher cooling rate. This result is due to the drastic change in the time-dependent behavior of the FC magnetization in the vicinity of  $T_{f0}$ .

# B. Analysis of the results and discussion

There are basically two different possible interpretations of the spin-glass freezing. First, in the cluster mod-



FIG. 5. Variation of the temperatures  $T_i$  and  $T_f$  with frequency in a Vogel Fulcher plot.  $T_i$  corresponds to the appearance of irreversibilities. It is defined either from  $|\chi''_{\nu}|/\chi'_{\nu}=10^{-3}$  at high frequencies ( $\odot$ ) or from the relation  $S_{\nu}=(\pi/2)(\partial M_{\rm ZFC}/\partial \ln t_{\rm obs})/M_{\rm ZFC}=10^{-3}$  at long observation times ( $\times$ ).  $T_f(\nu)$  corresponds to the cusp of the  $\chi_{\nu}(T)$  curve (+) or to the inflection point of the  $S_{\nu}(T)$  curve ( $\Box$ ).

el,<sup>15</sup> the spin-glass is considered as a collection of superparamagnetic clusters. For each cluster, the probability to overcome the anisotropy energy barrier E is expressed in terms of a relaxation time  $\tau$  related to E by the Arrhenius law  $\tau \propto \exp(-E/kT)$ . In this context, the spin-glass freezing takes place when  $t_{obs}$  is equal to the longest relaxation time of the distribution of magnetic clusters. Due to the unphysical experimental values for the shorter relaxation time, Tholence has interpreted data with a Vogel-Fulcher law:<sup>16</sup>

$$\tau = \tau_0 \exp[E/k(T - T_0)], \qquad (3)$$

with reasonable values of the atomic relaxation time  $\tau_0 = (2\pi v_0)^{-1}$ .  $T_0$  can be viewed as a phenomenologic parameter which describes the intercluster interaction. As shown on Fig. 5, the frequency dependence of  $T_f$  may be fit by such a Vogel-Fulcher law in the whole range  $2 \times 10^{-3} < v < 8 \times 10^4$  Hz, by taking E = 29 cm<sup>-1</sup>,  $T_0 = 11.77$  K, and  $\tau_0 = 10^{-12}$  s. Note that the numerical values of the fitting parameters E and  $T_0$  are not very sensitive to the choice of  $\tau_0$ . The relative variation of  $T_f$  per frequency decade  $\Delta T_f / T_f \Delta \ln v$  is  $2 \times 10^{-2}$  for Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te, intermediate between values reported in metallic spin-glasses  $(0.7 \times 10^{-2}$  in Cu:Mn) (Ref. 17) and for another insulator Eu<sub>1-x</sub>Sr<sub>x</sub>S (5 × 10<sup>-2</sup> for x = 0.4).<sup>10</sup>

As discussed above, two procedures may be used to analyze the dynamic behavior in a spin-glass, namely the measurements of the  $\nu$  (or  $t_{obs}$ ) dependence of either  $T_i$ corresponding to the appearance of irreversibilities, or  $T_f$ (Fig. 5). The static freezing temperature  $T_{f0}$  is then defined as the common limit of  $T_f(\nu)$  and  $T_i(\nu)$  at low frequency. Extrapolation of these plots in Fig. 5 leads to  $T_{f0}=12.8\pm0.1$  K, a value which is consistent with our previous determinations of  $T_{f0}$ .

The frequency dependencies of  $T_f$  and  $T_i$  are expected to be different since  $T_i$  is only related to the average correlation time  $\overline{\tau}$  which characterizes the distribution  $\rho(\tau)$ , whereas  $T_f$  depends on several moments of  $\rho(\tau)$ .<sup>18</sup>

 $\rho(\tau)$ , whereas  $T_f$  depends on several moments of  $\rho(\tau)$ .<sup>18</sup> In the restricted range  $2 \times 10^{-3} < \nu < 350$  Hz, it is still possible to fit the  $T_i(v)$  dependence by a Vogel-Fulcher law with the fitting parameters E = 37.8 cm<sup>-1</sup> and  $T_0 = 11.3$  K. According to expression (3),  $T_0$  should be the glass temperature. The value of  $T_0$  is inconsistent with the previous determination of  $T_{f0}$  and the crossing of the  $T_i(v)$  and  $T_f(v)$  curves on Fig. 5, pointing out the inability to fit our data with a Vogel-Fulcher law on a very extended frequency range, as already mentioned for other spin-glasses.<sup>19</sup> This is consistent with the fact that the Vogel-Fulcher law can always be viewed as an asymptotic form of a power law, expected for a phase transition. This is supported by the observation of a significant curvature of the  $T_i(v)$  curve at high frequencies up to  $8 \times 10^4$  Hz (Fig. 5). It is well known that the slow dynamics in spin-glasses depends on the wait time  $t_w$  spent below the freezing temperature.<sup>9</sup> We show in Fig. 6 that this effect can already be unambiguously observed on the TRM at a temperature 12 K, larger than  $T_0$ , ruling out definitively the validity of a Vogel-Fulcher law.

Let us try now to assume the existence of a phase tran-



FIG. 6. Relaxation of the thermoremanent magnetization in the spin-glass phase at 12 K for various wait times  $t_w = 10^2$  s (---),  $t_w = 10^3$  s  $(\cdot \cdot \cdot \cdot)$ ,  $t_w = 10^4$  s (---). All curves have been obtained after cooling the sample from 13.6 down to 12 K, at a rate of 1 K/min after switching off a field H = 10 Oe. The inflection point of these relaxation curves, revealed in the insert by the maximum of  $S_{\text{TRM}} = (H^{-1}) \partial M_{\text{TRM}} / \partial \ln t$ , stands at  $t \simeq t_w$ .

sition at  $T_{f0}$ .<sup>18,19</sup> The spin freezing may then be analyzed in terms of a critical slowing down above  $T_{f0}$  by using the power law

$$\tau/\tau_0 = A \left[ \frac{T - T_{f0}}{T} \right]^{-zv'}, \qquad (4)$$

where  $\nu'$  is the critical exponent for the correlation length  $\xi$ , and z the dynamic exponent relating  $\tau$  to  $\xi$  by  $\tau \simeq \tau_0 \xi^z$ . The  $T_i(\nu)$  variation for  $Cd_{0.6}Mn_{0.4}$ Te is consistent with the law in Eq. (4). The relative uncertainty on the determination of the fitting parameters  $z\nu'$  and  $\tau_0$ originates from that of  $T_{f0}$ . Assuming  $T_{f0}=12.80$  K, we obtain  $z\nu'=9.7$  and  $\tau_0=3.8\times10^{-14}$  s. Another set of parameters,  $z\nu'=7.9$  and  $\tau_0=1.5\times10^{-12}$  s, can be determined if one sets  $T_{f0}=12.90$  K. The corresponding critical plots are reported on Fig. 7. The shortest relaxation time  $\tau_0$  compares well with  $\hbar/(kT_{f0})=5.5\times10^{-13}$  s, as other critical fits reported for  $Cd_{1-x}Mn_xTe$  for other manganese concentrations<sup>7</sup> and in  $Eu_{0.4}Sr_{0.6}S.^{11}$  The obtained value for  $z\nu' \sim 8-10$  agrees with simulation data of Ogielski<sup>18</sup> in the case of 3d spin-glasses with shortrange magnetic interactions.

### IV. H-T PHASE DIAGRAM

# A. Generalities

The phase diagram of spin-glasses in the presence of an applied magnetic field H was first derived by de Almeida and Thouless (AT).<sup>20</sup> In Ising spin-glasses, treated in the Sherrington-Kirkpatrick (SK) molecular field approximation, for purely random interactions of infinite range, they evidenced an H-T instability AT line, corresponding to the onset of irreversibilities with the equation



FIG. 7. Dynamic scaling of the critical slowing down above the static freezing temperature  $T_{f0}$ .  $T_i$  is the temperature corresponding to the appearance of irreversibilities (see Fig. 5). The plots are given for two reasonable values of  $T_{f0}$ , namely  $T_{f0} = 12.80 \text{ K} (\circ)$  and 12.90 K ( $\times$ ).

$$H_C = \frac{2}{\sqrt{3}} (\tau^l)^{3/2} , \qquad (5)$$

where  $H_C = g\mu_B SH/(kT_{f0})$  is the reduced magnetic field, and  $\tau^l = [1 - T_i^l(H)/T_{f0}]$  is the reduced temperature. This line has been first interpreted by Toulouse<sup>21</sup> as due to a phase transition.

These calculations have been extended to the case of an isotropic Heisenberg spin system in the SK approximation.<sup>22</sup> Upon cooling, they predict first a freezing of the transverse components of the spins along the so-called Gabay-Toulouse (GT) line:

$$H_C = \frac{10}{\sqrt{23}} (\tau^t)^{1/2} , \qquad (6)$$

where  $\tau^t = [1 - T_i^t(H)/T_{f0}]$  corresponds to the transverse case. As the temperature further decreases, the longitudinal spin component, parallel to *H*, also freezes along a line similar to that derived by AT [expression (5)] but with a new coefficient  $(\frac{2}{5})^{1/2}$ . For Heisenberg systems, this AT line only describes a crossover behavior instead of a true phase transition.<sup>23</sup>

All these predictions are based on the hypothesis of a thermodynamic equilibrium which is far from reachable in usual experiments, so that the comparison between these calculations and experimental data may be somewhat hazardous. Some works even suggest that apparent transitions at finite temperature are artifacts coming from the finite measuring time.<sup>24,25</sup> It follows that, so far, the experimental observation of AT or GT lines does not support unambiguously the existence of an in-field phase transition in 3d spin-glasses, which is still a subject of controversy.<sup>26</sup> The determination of  $T_i^{t(l)}(H,t)$  lines gives, however, important information on spin-glasses because the  $H^2$  or  $H^{2/3}$  variations reflect different dynamics of the spin freezing.<sup>27</sup>

Herein, we report (H, T) phase diagram studies on  $Cd_{0.6}Mn_{0.4}Te$  in weak magnetic fields. We extend to low fields previous data obtained on the  $Cd_{0.45}Mn_{0.55}Te$  sam-

ple<sup>28</sup> and stress the effect of the time of measurement  $(10^{-3} < t < 100 \text{ s})$  on the results.

### **B.** Experimental procedures

In the millisecond range  $T_i(H, t_{obs})$  is deduced, like in Ref. 11, from the onset of irreversibilities in the temperature variation of  $\chi''_{\nu}$ , measured under a weak ac magnetic field ( $\simeq 0.1$  Oe), superposed on the static field H.  $\chi''_{\nu}$  is measured by Faraday rotation, following the procedure reported in Sec. II. At long observation times ( $0.5 < t_{obs} < 10^2$  s) the (H, T) diagrams were more accurately determined from SQUID measurements.

The time corresponding to the onset of irreversibilities, for several values of H and T, is determined by extrapolations of the ZFC and TRM magnetization relaxation curves to the equilibrium levels. It is then possible to plot a set of  $T_i(H)$  lines for different values of  $t_{obs}$ . The response of the system is assumed to be linear in the determination of these transition lines.

### C. Results and discussion

At temperatures significantly larger than  $T_{f0}$ , namely T > 14.2 K, we have shown<sup>8</sup> that the fast relaxation  $(t < 10^{-3} \text{ s})$  can be attributed to the dynamics of weakly coupled superparamagnetic entities. It is reasonable to assume that  $T_i(H,t)$  does not depend strongly on the value of H at short times, as already observed in  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}^{-11}$ 

The investigation of results at larger observation times is more interesting since the  $T_i(H)$  behavior is connected with a cooperative behavior.<sup>8</sup> The (H, T) phase diagrams for different values of  $t_{obs}$  are shown in Fig. 8. At first glance the variation of  $T_i$  with H at long times  $(1 < t_{obs} < 10^2 \text{ s})$  is consistent with that predicted for GT lines. This result can be extended to at least  $t_{obs} \simeq 10^{-3}$  s since  $\chi_{\nu}^{\prime\prime}(T)$  curves  $(5 < \nu < 10^3 \text{ Hz})$  obtained for different values of H in the range 0 < H < 120 Oe look identical within experimental errors, i.e., the difference  $T_i(H=120$ Oe) –  $T_i(H=0)$  does not exceed  $2 \times 10^{-2}$  K. This longtime GT behavior has been previously observed for Cd<sub>0.45</sub>Mn<sub>0.55</sub>Te (Ref. 28) at higher fields, up to 6.5 kOe, in agreement with Monte Carlo simulations showing that the  $T_i(H)$  line is related to the transverse spin freezing in the thermodynamic limit.<sup>29</sup> A similar result has also been reported in CsNiFeF<sub>6</sub>,<sup>30</sup> for which the field dependence of the temperature, corresponding to the appearance of weak irreversibilities, is GT-like [see figure (8) in Ref. 30]. These results, however, differ from many other ones in metallic spin-glasses,<sup>31</sup> or in the insulating spinglass Eu<sub>0.4</sub>Sr<sub>0.6</sub>S (Ref. 11) for which a strong dependence of  $T_i$  on H, characteristic of an AT-like line, has been observed at long times. We believe this difference is related to the particular frustrated random antiferromagnetic structure of such Heisenberg spin-glasses as  $Cd_{1-x}Mn_x$  Te or CsNiFeF<sub>6</sub>.

In insulators with negligible crystal-field anisotropy like  $Cd_{1-x}Mn_xTe$  or  $Eu_{1-x}Sr_xS$ , the anisotropy comes mainly from the dipole-dipole interaction  $E_{d-d}$ . A typical value is  $E_{d-d} \simeq 0.4$  K for these compounds. So why does



FIG. 8. (H, T) phase diagram determined from the vanishing of the thermoremanent magnetization at observation times  $t_{obs} = 1$ , 10, and 100 s by (a) a SQUID magnetometer or (b) by Faraday rotation at t = 0.50 s. The two methods give consistent results.

 $Eu_{0.4}Sr_{0.6}S$  behave like an Ising spin-glass while the (H-T) phase diagram of  $Cd_{0.6}Mn_{0.4}Te$  is more consistent with the Heisenberg case? A reasonable answer is that  $E_{d-d}$  has a value comparable to the exchange interaction between europium ions in  $Eu_{0.4}Sr_{0.6}S$ , while in  $Cd_{0.6}Mn_{0.4}Te$  the dipolar interaction is at least 1 order of magnitude smaller.

In addition to the GT line related to the onset of weak irreversibilities at long time (and defined from  $T_i$ ), an AT-type line corresponding to larger irreversibilities and more connected with  $T_f$  has already been observed at lower temperature in frustrated disordered antiferromagnets such as  $Cd_{0.55}Mn_{0.45}Te$  and  $CsNiFeF_6$ . This AT line, however, has not been determined in this work for  $Cd_{0.6}Mn_{0.4}Te$  since our in-field experimental procedures were restricted to too short times for the range of low magnetic fields we have explored.

The GT line corresponds to the freezing of the transverse components of the magnetization, i.e., perpendicular to the field-cooled direction.<sup>22</sup> Therefore it is desirable to explain why the magnetization or the Faraday rotation, measured along the field-cooled direction, can probe such transverse spin freezing. This feature can be attributed to the particular spin structure in disordered Heisenberg antiferromagnets where spins point towards all directions. The magnetic field applied during the cooling process is weak enough so that the spin structure is not altered significantly, hence a contribution of both longitudinal and transverse local spin components. The relaxation of the thermoremanent magnetization can be significantly different in various types of spin glasses, especially in diluted frustrated antiferromagnets.

# **V. CONCLUSION**

The dynamic magnetic properties of  $Cd_{0.6}Mn_{0.4}Te$  the archetypical pure Heisenberg spin-glass generated by randomness on a frustrated antiferromagnetic lattice have been extensively investigated for a wide range of observation times. The appearance of a slow relaxation regime below T = 14.2 K has been described in terms of a critical slowing down. Our results are better interpreted within a dynamic scaling approach in the temperature range 13.0 < T < 14.2 K, supporting previous suggestions<sup>1,5,7</sup> that there exists a phase transition at the static spin-glass freezing temperature  $T_{f0}=12.85\pm0.05$  K. This result has also been inferred from the study of the relaxation of the thermoremanent magnetization in the vicinity of  $T_{f0}$ .<sup>8</sup>

For different values of the observation times, up to 100 s, the (H,T) magnetic phase diagrams of  $Cd_{0.6}Mn_{0.4}Te$ have been determined in weak fields. The temperature dependence of the onset of irreversibilities is always well described by a Gabay-Toulouse (GT-like) line, characterizing the appearance of a transverse spin freezing. In this work, we have not proved the divergence of the relaxation time along the GT line as already done for  $Eu_{0.4}Sr_{0.6}S$  on the AT line supported by the in-field dynamic scaling. It will be interesting to investigate dynamic properties of other disordered frustrated antiferromagnets in the future, especially in two dimensions.

Theories must be developed in disordered frustrated antiferromagnets to test quantitatively their original dynamic properties.

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- <sup>1</sup>R. R. Galazka, S. Nagata, and P. H. Keesom, Phys. Rev. B 22, 3344 (1980); S. Oseroff and P. H. Keesom, in *Semiconductors* and *Semimetals*, edited by R. K. Williamson and A. C. Beer (Academic, New York, 1986).
- <sup>2</sup>N. B. Brandt and V. V. Moshchalkov, Adv. Phys. **33**, 193 (1984).
- <sup>3</sup>M. Escorne, A. Mauger, R. Triboulet, and J. L. Tholence, Physica B + C 107B, 209 (1981).
- <sup>4</sup>M. Escorne and A. Mauger, Phys. Rev. B 25, 4674 (1982).
- <sup>5</sup>S. Oseroff and F. G. Gandra, J. Appl. Phys. 57, 3421 (1985).
- <sup>6</sup>M. Ayadi, P. Nordblad, J. Ferré, A. Mauger, and R. Triboulet,

J. Magn. Magn. Mater. 54-57, 1223 (1986).

- <sup>7</sup>M. Saint-Paul, J. L. Tholence, and W. Giriat, Solid State Commun. **60**, 621 (1986).
- <sup>8</sup>M. Ayadi, J. Ferré, A. Mauger, and R. Triboulet, Phys. Rev. Lett. **57**, 1165 (1986).
- <sup>9</sup>P. Nordblad, P. Svedlindh, J. Ferré, and M. Ayadi, J. Magn. Magn. Mater. **59**, 250 (1986).
- <sup>10</sup>J. Ferré, J. Rajchenbach, and H. Maletta, J. Appl. Phys. 52, 1697 (1981).
- <sup>11</sup>N. Bontemps, J. Rajchenbach, R. V. Chamberlin, and R. Orbach, Phys. Rev. B 30, 6514 (1984).
- <sup>12</sup>C. Rigaux, A. Mycielski, G. Barilero, and M. Menant, Phys. Rev. B 34, 3313 (1986).
- <sup>13</sup>L. Lundgren, P. Svendlindh, and O. Beckman, J. Magn. Magn. Mater. 25, 33 (1981).
- <sup>14</sup>A. P. Malozemoff and E. Pytte, Phys. Rev. B 34, 6579 (1986).
- <sup>15</sup>L. Néel, Ann. Geophys. 5, 99 (1949).
- <sup>16</sup>J. L. Tholence, Solid State Commun. 35, 113 (1980).
- $^{17}$ J. L. Tholence, Physica B + C **126B**, 157 (1984).
- <sup>18</sup>A. T. Ogielski, Phys. Rev. B 32, 7384 (1985).
- <sup>19</sup>J. Souletie and J. L. Tholence, Phys. Rev. B 32, 516 (1985).

- <sup>20</sup>J. R. L. de Almeida and D. J. Thouless, J. Phys. A **11**, 983 (1978).
- <sup>21</sup>G. Toulouse, J. Phys. Lett. 41, L447 (1980).
- <sup>22</sup>G. Toulouse and M. Gabay, J. Phys. Lett. **42**, L103 (1981); M. Gabay and G. Toulouse, Phys. Rev. Lett. **47**, 201 (1981).
- <sup>23</sup>D. S. Sherrington, D. M. Cragg, D. Elderfield, and M. Gabay, J. Phys. Soc. Jpn. (Suppl.) 52, 229 (1983).
- <sup>24</sup>A. P. Young, Phys. Rev. Lett. 50, 917 (1983).
- <sup>25</sup>W. Kinzel and K. Binder, Phys. Rev. Lett. 50, 1509 (1983).
- <sup>26</sup>A. P. Young and R. N. Blatt, J. Magn. Magn. Mater. 54-57, 6 (1986).
- <sup>27</sup>J. Rajchenbach and N. Bontemps, J. Phys. Lett. **44**, L799 (1983).
- <sup>28</sup>H. Kett, W. Gebhardt, U. Krey, and J. K. Furdyna, J. Magn. Magn. Mater. 25, 215 (1981).
- <sup>29</sup>H. Kett, W. Gebhardt, and U. Krey, J. Magn. Magn. Mater. 46, 5 (1984).
- <sup>30</sup>C. Pappa, J. Hammann, and C. Jacoboni, J. Phys. C 17, 1303 (1984).
- <sup>31</sup>For a review, see C. Y. Huang, J. Magn. Magn. Mater. **25**, 215 (1986).