Spin-Freezing Process in a Spin-Glass: $Cd_{0.6}Mn_{0.4}Te$

M. Ayadi[a] and J. Ferré
Laboratoire de Physique des Solides, Université de Paris–Sud, 91405 Orsay, France

A. Mauger
Groupe de Physique des Solides de l'Ecole Normale Supérieure, Université Paris 7, 75251 Paris, France

and

R. Triboulet
Laboratoire de Physique des Solides, Centre National de la Recherche Scientifique, 92190 Meudon, France
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The time dependence of the thermoremanent magnetization has been investigated for times 10^{-2} s $\leq t \leq 10^2$ s in $Cd_{0.6}Mn_{0.4}Te$. The sudden appearance of a slow relaxation regime at temperature $T_{f0} = 13.10 \pm 0.03$ K, superimposed on a much faster regime varying continuously through T_{f0}, supports the phase-transition hypothesis.

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A full description of spin-glass freezing must take time effects into account.[1] Below the static freezing temperature T_{f0} macroscopic relaxation times appear and are related to the breakdown of the ergodicity. The usual way to describe the slow relaxation of the remanent magnetization $M_R(t)$ is to postulate a statistical distribution $g(\tau)$ of the relaxation times τ and to assume additive contributions:

$$M_R(t) = \int_0^\infty g(\tau) e^{-t/\tau} d\tau. \quad (1)$$

This approach has been widely used[2-5] to describe the relaxation of M_R in the vicinity of the spin-glass freezing temperature. Within this model, the distribution $g(\tau)$ spreads suddenly to long times at T_{f0} on lowering of the temperature. However, this distribution does not reduce to a Dirac function just above T_{f0} because of the slow relaxation of the ramified spin structure[6] or the finite probability of having clusters with unfrustrated spins.[7,8] For the last case, the corresponding "Griffiths" intermediate phase is less extended above T_{f0} in systems having long-range interactions—as in metallic alloys (CuMn, AgMn, AuFe, etc.), for which the frustration is more evenly distributed on all spins—than in most insulating spin-glasses ($Eu_xSr_{1-x}S$, amorphous manganese aluminosilicates, etc.).

Interesting questions arise naturally: Do all spins couple together cooperatively at T_{f0}? How is $g(\tau)$ modified at this temperature? Lundgren, Svedlindh, and Beckman[4] proposed that the onset of ordering at T_{f0} may be observed only for measuring times longer than a critical time τ_{crit} and suggested a simplified picture for the spin-glass state consisting of two coexisting phases, one ordered and the other characterized by a broad spectral distribution of relaxation times with a finite maximum value. In this picture, also implicitly used by Nordblad and Lundgren,[9] the evolution of $g(\tau)$ with temperature does not show any singularity at T_{f0}. However, these proposals have never been checked experimentally on real systems.

In this Letter we report time-dependent properties of the remanent magnetization in the diluted semiconductor spin-glass $Cd_{0.6}Mn_{0.4}Te$,[10,11] in the vicinity of its freezing temperature $T_{f0} = 13.10$ K, to verify these proposals directly. At low temperature it behaves like an insulator with short-range antiferromagnetic interactions between Mn^{2+} ions. Following De Sèze,[12] the spin-glass state is due to frustration in such a diluted fcc antiferromagnet. We evidence two distinct relaxation regimes: Above T_{f0} the relaxation is thermally activated, while this process coexists with a much slower relaxation regime just below. This last process is well described by a Kohlrausch law at low enough temperature. This new result proves unambiguously the coexistence of two phases just below the freezing temperature defined by the appearance of a slow relaxation regime, related to the spin-glass ordering.

The crystal, prepared by the Bridgman technique, has been cut and polished as a thin plate ($4 \times 4 \times 1.65$ mm^3). The high quality of our sample has been checked previously.[13] The time relaxation of the thermoremanent magnetization (TRM) has been measured by Faraday rotation (FR) at 6328 Å. The light beam propagated along the initially applied magnetic field direction, normally to the plate. We checked that the FR in the sample was proportional to the magnetization from direct measurements with a Foner-type magnetometer. The FR is a very convenient method to measure small variations of the magnetization of our sample ($\delta M \sim 10^{-5}$ emu) even at short time.

The magnetic field was generated by a small copper coil of seventy turns, which allowed us to switch off

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the field used, $H = 56$ Oe, in less than 1 ms. This value of H represents the lowest field for which the signals are large enough to allow a quantitative analysis with our present experimental setup. The same qualitative time dependence of the TRM is observed at lower fields. The field used is small enough to stay in the spin-glass phase, since it is 2 orders of magnitude lower than necessary to reach the saturated magnetization,[10] and because of the very weak variation of T_{f0} with H.[14] To avoid any artifact associated with a possible weak overshoot of the coil, we have only reported the data for times exceeding 10 ms. The temperature was detected by a calibrated carbon thermometer located against the sample, and was stabilized within ± 0.01 K during the experimental time ($t < 1000$ s). The FR was measured by a sensitive modulation technique of the state of polarization of the light.[15] For times 1 s $< t < 10^3$ s, the relaxation of the TRM following the switching off of the magnetic field at $t = 0$ was continuously recorded. For intermediate and shorter time measurements (10 ms $< t <$ 20 s), the magnetic field was generated by a square-wave current in the coil, with a period (1 s $< T <$ 200 s) much larger than the time required by the sample to reach the magnetized or demagnetized state at equilibrium. The FR corresponding to the relaxation of the TRM was detected and averaged on a digital storage oscilloscope over up to 256 times, depending on the time range investigated. In both cases, the residual signal for the demagnetized state was deduced from a zero-field–cooled scan.

As for other spin-glasses, the TRM of $Cd_{0.6}Mn_{0.4}Te$ is dependent on aging effects.[16] Below 12.75 K, we chose the waiting time $t_w = 180$ s spent during field cooling of the sample from T_{f0}. However, in the close vicinity of the freezing temperature ($0.95 < T/T_{f0} < 1.05$), we checked that the FR related to the TRM is not affected by the waiting time within the experimental uncertainty.

The static freezing temperature $T_{f0} = 13.10 \pm 0.03$ K is defined from the break in the slope of the slow–field-cooled magnetization curve. Moreover, it corresponds to the temperature of the cusp of the zero-field–cooled magnetic dc susceptibility obtained during a slow heating process.[13] It is consistent with that found recently from step temperature changes in magnetization.[17]

We found that irreversible effects still exist above T_{f0} [Fig. 1(a)], in agreement with the frequency dependence of the ac susceptibility.[13] As the temperature approaches T_{f0} from above, a thermally activated relaxation process appears in the millisecond range. In a crude approximation, the fast regime may be fitted by a $\log t$ law, but a precise functional form cannot be deduced, given the weakness of the Faraday rotation. The characteristic time τ_m for which the TRM vanishes increases rapidly when the temperature is lowered.

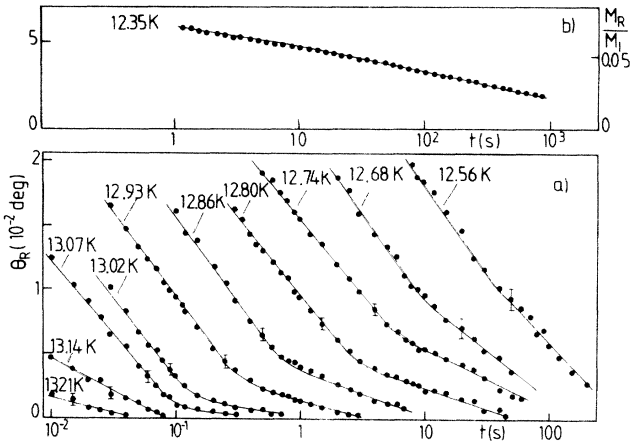

FIG. 1. Relaxation of the TRM of $Cd_{0.6}Mn_{0.4}Te$ measured by Faraday rotation, corresponding to an initially applied field of 56 Oe: (a) in the vicinity of $T_{f0} \simeq 13.10$ K, and (b) at $T = 0.94 T_{f0}$. In the ordinate, we also give the value M_R/M_i of the TRM normalized by the initial value of the magnetization under field. The experimental data may be well fitted by the Kohlrausch decay form with $n = 0.80$ and $\tau = 300$ s. Each dot on the curves corresponds to a local time average of the continuously recorded signal.

A new slower relaxation regime, involving a distribution at long times, appears suddenly at T_{f0} [Fig. 1(a)] and grows at the expense of the faster component when the temperature decreases down to 12.5 K. It is important to point out that both regimes coexist between 13.1 and 12.5 K, as evidenced by the significant break of the slope of the $\theta_R(\log t)$ relaxation curves at τ_m, still corresponding to the disappearance of the faster regime. Within the experimental errors, the temperature dependence of τ_m on both sides of T_{f0} may be described by the same Vogel-Fulcher law:

$$\tau_m = \tau_0 \exp[E_a/k_B(T_m - T_0)]. \quad (2)$$

Above T_{f0} the value of $\tau_m(T)$ is deduced from the vanishing of the out-of-phase component of the magnetic susceptibility $\chi''(\nu_m)$ for the corresponding frequency $\nu_m = (2\pi\tau_m)^{-1}$. As found experimentally[18] for other spin-glasses, τ_m diverges at a temperature T_0 which is smaller than the freezing temperature T_{f0}.

Assuming $\nu_0 = (2\pi\tau_0)^{-1} = 10^{12}$ Hz, we get reasonable fitting parameters $T_0/T_{f0} = 0.77$, $E_a/k_B = 190$ K (Fig. 2) as compared to other data.[18] For comparison, we have also shown in the same figure the variation of $T_f(\nu)$, defined as the cusp temperature of the ac susceptibility measured at frequency ν. We have also reported the point $T = T_{f0}$ corresponding to $\tau_m = \infty$, assuming that slow (1 mK/s) zero-field or field-cooled scans may be considered as quasistatic measurements. Below 12.5 K, only the slower regime is observed [Fig.

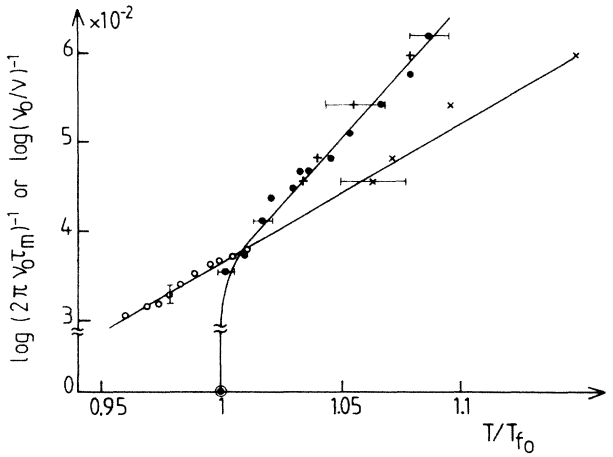

FIG. 2. Temperature dependence of (closed circles) the cusp of the real part of the ac susceptibility $\chi'_{ac}(\nu)$ measured at the ν frequency; (plusses) the inflection point of the imaginary part of the ac susceptibility $\chi''_{ac}(\nu)$; (crosses) the appearance of irreversibility, i.e., $\chi''_{ac}(\nu) \to 0$; and (open circles) τ_m corresponding to the vanishing of the faster relaxation regime. We have also reported the quasistatic point (circled dot) at $T = T_{f0}$ (see text).

1(b)], and it is fitted by the Kohlrausch stretched-exponential decay

$$M_R(t) = M_0 \exp[(-t/\tau)^{1-n}], \quad (3)$$

with long characteristic times τ ($\tau = 300$ s at $T/T_{f0} = 0.96$) and n not far from 1, as already found in several spin-glasses.[19,20]

The slower regime, appearing at T_{f0}, is associated with a cooperative behavior in the ordered state. Since the faster regime coexists with this slower one, just below T_{f0}, all the spins do not participate in the freezing. We expect that correlated magnetic regions are still present in the sample and involve long lifetimes.[21] It is hard to estimate the proportion of frozen spins at T_{f0} from our data, since the order parameter $q(t) = \overline{\langle S_i(0) S_i(t) \rangle}$ increases slowly with decrease of temperature just below T_{f0}. Our results demonstrate that the ordered state grows rapidly, with lowering temperature, at the expense of "clusters" responsible for the faster regime. This is consistent with a divergence of the magnetic correlation length at T_{f0} corresponding to a magnetic percolation process,[7] but as the temperature is decreased, more and more fluctuating spins are bound to the infinite cluster. Our results are inconsistent with a progressive freezing of the spins, and the abrupt appearance of long characteristic times at T_{f0} rather point out a phase transition. However, the faster regime, still present above T_{f0}, cannot be interpreted as a critical slowing down, since τ_m continues to increase even below T_{f0}. This makes it difficult to demonstrate a critical behavior in the vicinity of the freezing temperature. The increase of τ_m is inconsistent with the development of one ordered state at the expense only of clusters with the largest relaxation times. It is important to note that a chemical clustering cannot explain our experimental data.

Consequently, our experimental results are in qualitative agreement with models based on hierarchically constrained dynamics,[22] and we are led to to the conclusion that the faster regime, present above T_{f0}, is related to weakly coupled magnetic unfrustrated clusters,[7,8] which can survive below T_{f0}, in its vicinity. From the above data, it is clear that the short- and long-time behaviors of the relaxation of the TRM have a different origin, and that the slow dynamics of $Cd_{0.6}Mn_{0.4}Te$ cannot be explained within a simple model[9] assuming a continuous change of the relaxation at T_{f0}.

While the temperature dependence of τ_m may be fitted by a Vogel-Fulcher law above and below T_{f0}, the variation of $T_f(\nu)$, defined as the temperature of the cusp of the ac susceptibility, may also be described by another Vogel-Fulcher law above T_{f0}, but tends suddenly towards T_{f0} in the static limit[13] (Fig. 2 and as already found[15] for $Eu_{0.4}Sr_{0.6}S$). This behavior has been also reported for AuFe from zero-field–cooled experiments.[4] The breakdown of the Vogel-Fulcher law for $T_f(\nu)$ at low frequency is directly related to the appearance of very long characteristic times for the dynamical process at T_{f0} and does not assume *a priori* the existence of an upper limit for the static susceptibility of spin-glasses as invoked previously for CuMn.[9] As mentioned earlier,[22,23] the cusp in susceptibility has no universal meaning concerning the dynamics, and irreversible effects appear at temperatures higher than $T_f(\nu)$ at the corresponding frequency.

In spite of its insulating character, $Cd_{0.6}Mn_{0.4}Te$ shows long-time dynamical properties similar to those met in archetypal spin-glasses like CuMn, in the sense that just below T_{f0} the involved relaxation times become very long, pointing out a sudden freezing of the spin system.

In conclusion, we have shown the existence of two relaxation processes in $Cd_{0.6}Mn_{0.4}Te$, which coexist below the spin-glass quasistatic freezing temperature T_{f0}. The fast relaxation regime is associated with the existence of spin-correlated regions resulting from intrinsic local magnetic inhomogeneities.[21] Such a clustering is inherent to any spin-diluted insulating material at a microscopic scale, and results from the short-range nature of the magnetic interactions.[6] It follows that our conclusions should be generalized to other insulating spin-glasses. Similar results should be observed in other members of the family of the so-called semimagnetic semiconductors.

In another insulating spin-glass, $Eu_{1-x}Sr_xS$, the frustration is different in nature and results from the competition between ferromagnetic and antiferromag-

netic interactions, and not from the particular topology of the lattice. This material and related compounds then form a distinct class of spin-glasses where the time scale of the slower regime is reduced. In such a case, it may be more difficult to separate experimentally the two regimes which have closer relaxation rates near T_{f0}.[20]

In spite of this difficulty, very refined susceptibility measurements on a wide frequency scale have permitted us to reach the critical slowing down just above the freezing temperature of $Eu_{0.4}Sr_{0.6}S$.[24] Similar experiments could be planned on $Cd_{0.6}Mn_{0.4}Te$.

In metallic spin-glass alloys, the local magnetic inhomogeneities have a less significant effect on the slow dynamics above the freezing temperature, because of the long-range nature of the magnetic interactions. In such a case, the typical magnetic correlation length will be of the order of the magnitude of the range of interactions. In the vicinity of T_{f0}, the dynamical behavior is consequently closer to a classical critical slowing down. The Griffiths phase[8] is expected to be less extended than in insulating spin-glasses, since the frustration is more equally distributed over spins. Nevertheless, the investigation of spin dynamics led Lundgren, Svedlindh, and Beckman[4] to propose the coexistence of two distinct phases in the spin-glass state of AuFe. This is an indication that our conclusions may even be generalized to metallic spin-glasses. Unfortunately, short time measurements cannot be performed as easily in metallic alloys as in insulators, especially by means of magneto-optical techniques.

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[a]Present address: Département de Physique de la Faculté des Sciences de Tunis, Campus Universitaire, Le Belvédère, Tunisie.

[1]G. Toulouse, in *Congrès de la Société Française de Physique* (Les Editions de Physique, Les Ulis, 1981), p. 3.

[2]J. J. Préjean and J. Souletie, J. Phys. (Paris) **41**, 1335 (1980).

[3]C. A. M. Mulder, A. J. van Duyneveldt, and J. A. Mydosh, Phys. Rev. B **23**, 1384 (1981).

[4]L. Lundgren, P. Svedlindh, and O. Beckman, Phys. Rev. B **26**, 3990 (1982).

[5]L. E. Wenger, in *Heidelberg Colloquium on Spin Glasses*, edited by J. Ehlers, K. Hepp, R. Kippenhahn, H. A. Weidenmüller, and J. Z. Harz, Lecture Notes in Physics Vol. 192 (Springer-Verlag, Berlin, 1983), p. 60.

[6]A. T. Ogielski, Phys. Rev. B **32**, 7384 (1985).

[7]M. Cyrot, Solid State Commun. **39**, 1009 (1981).

[8]M. Randeira, J. P. Sethna, and R. G. Palmer, Phys. Rev. Lett. **54**, 1321 (1985).

[9]P. Nordblad and L. Lundgren, J. Phys. Soc. Jpn. **54**, 1681 (1985).

[10]S. Oseroff and F. G. Gandra, J. Appl. Phys. **57**, 3421 (1985).

[11]J. K. Furdyna, J. Appl. Phys. **53**, 7637 (1982).

[12]L. De Sèze, J. Phys. C **10**, L353 (1977).

[13]M. Ayadi, P. Nordblad, J. Ferré, A. Mauger, and R. Triboulet, J. Magn. Magn. Mater. **54–57**, 1223 (1986).

[14]H. Kett, W. Gebhardt, U. Krey, and J. K. Furdyna, J. Magn. Magn. Mater. **25**, 215 (1981).

[15]J. Ferré, J. Rajchenbach, and H. Maletta, J. Appl. Phys. **52**, 1697 (1981).

[16]R. V. Chamberlin, Phys. Rev. B **30**, 5393 (1984).

[17]P. Nordblad, P. Svedlindh, J. Ferré, and M. Ayadi, J. Magn. Magn. Mater. (to be published).

[18]J. L. Tholence, Solid State Commun. **35**, 113 (1980).

[19]R. V. Chamberlin, G. Morzukewich, and R. Orbach, Phys. Rev. Lett. **52**, 867 (1984).

[20]J. Ferré, M. Ayadi, R. V. Chamberlin, R. Orbach, and N. Bontemps, J. Magn. Magn. Mater. **54–57**, 211 (1986).

[21]A. E. Jacobs, Phys. Rev. B **32**, 7430 (1985).

[22]R. G. Palmer, D. L. Stein, E. Abrahams, and P. W. Anderson, Phys. Rev. Lett. **53**, 958 (1984).

[23]A. P. Malozemoff, Y. Imry, and B. Barbara, J. Appl. Phys. **53**, 7672 (1982).

[24]N. Bontemps, J. Rajchenbach, R. V. Chamberlin, and R. Orbach, J. Magn. Magn. Mater. **54–57**, 1 (1986).