Experimental study of the temperature-field phase diagram of spin-glasses

Y. Yeshurun,* L. J. P. Ketelsen, and M. B. Salamon
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801
(Received 20 January 1982; revised manuscript received 30 April 1982)

Studies of magnetic viscosity are presented for the amorphous $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$ as a function of temperature ($4.2 \leq T \leq 36$ K) and field (50 Oe $\leq H \leq$ 10 kOe). The slow changes of magnetization with time are described by $M(t) = M_1 + S \ln t$. The coefficient S is found to be field and temperature dependent. For the remanent magnetization, $|S(H)|$ increases with field and then maintains a roughly constant value. For the in-field relaxation, $S(H)$ exhibits a maximum and for each temperature we find a critical field for which $S = 0$. The line of critical fields is identified as the de Almeida–Thouless line.

Spin-glasses are random magnetic systems with the magnetic spins frozen in random directions at low temperature.[1,2] One of the characteristic properties of spin-glass systems is the slow response to a change in magnetic field. This phenomena, known as magnetic viscosity, has recently attracted new theoretical[3–5] and experimental[6–11] efforts. We report here an experimental study of the field and temperature dependence of the magnetic viscosity in amorphous $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$ spin-glass. This study was stimulated by recent theoretical results[5,12–14] which predict a line of phase transitions in the field-temperature (H-T) phase diagram of spin-glasses. In the present study we find experimentally a line of critical fields $H_c(T)$ defined by the highest field for which viscosity phenomena is still observed at temperature T. This line, which gives the boundaries for vanishing viscosity, separates the spin-glass phase from the paramagnetic phase and acquires the features predicted theoretically.

The ac susceptibility of spin-glasses shows a sharp cusp at a well-defined temperature[1] T_f, an observation which has prompted considerations of the possibility of a thermodynamic phase transition. Models of the Edwards and Anderson (EA) type[15] such as the Sherrington-Kirkpatrick[16] (SK) mean-field solution, indeed produces cusp in the susceptibility, and have been very useful in explaining experimental data. de Almeida and Thouless[12] (AT), however, showed instabilities in the SK solution. For a spin-glass system they calculated a line in the H-T plane above which the SK solution is correct. The equation of this line is known and its asymptotic behavior for $(T_f - T)/T_f \equiv \tilde{t} \rightarrow 0$ is given by

$$H = A\tilde{t}^{3/2} \quad . \tag{1}$$

Several recent theories[5,13,14] have the AT line as a line of paramagnetic (PM)–spin-glass (SG) transitions. Sompolinsky,[5] using a dynamic approach, shows explicitly that this line marks the disappearance of irreversible phenomena in the high-temperature regime. Thus, from a theoretical point of view, the existence of a transition line in the H-T plane is well established and its physical meaning is well understood.

Magnetic viscosity, being one of the most basic features of spin-glasses, has been studied intensively. Most of the experimental findings (logarithmic time dependence, field dependence of the relaxation rates in a constant temperature, etc.) have been reproduced by Monte Carlo calculations[17–19] based on the EA spin-glass model. Almost no effort has been made, however, to interpret the viscosity results in terms of phase transitions. References 9–11 are the few exceptions. Knitter and Kouvel[9] found a line of critical fields defined at each temperature by vanishing of viscosity effects. Their method involves stirring the magnetic spins by turning the external field on and off. The method here is based on a different procedure, to be described below. The present study is the first one to use viscous phenomena for identification and characterization of the AT line. It is also interesting to note that most of the new experimental $S(H,T)$ data here are in striking similarity with the Monte Carlo results.

The system chosen for this study, $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$, has been previously characterized[20,21] as a spin-glass, with an ac susceptibility cusp at $T_f \cong 41$ K. Ribbons ($5 \times 1 \times 0.05$ mm^3) of total weight of 19.8 mg were prepared by centrifugal spin quenching[22] and mounted in a vibrating sample magnetometer with the longest axis parallel with the applied magnetic field to minimize demagnetization effects. The temperature of the sample was measured with a 25-μm thermocouple mounted $\sim$2 mm from the sample and having a good thermal contact with it. We find that the sample temperature can be maintained constant to within 0.05 K during times much greater than those required ($\sim$12 min) to take relaxation data. Direct measurement of the sample temperature is crucial when the relaxation rate is slow since small temperature drifts can cause comparable magnetiza-

tion changes.

Three different procedures were used. (i) The sample was cooled in zero field (ZFC) from $T \cong 60\ \text{K} > T_f$ to the desired temperature T. The field was increased abruptly to a value H and changes in the magnetization recorded for approximately 12 min. (ii) The field of step (i) was kept constant until quasiequilibrium (defined by a constant reading of magnetization over a period of several minutes) was achieved. Then the field was turned off and the relaxation recorded for ~12 min. (iii) After cooling in a field (FC) from $T \cong 60$ K to $T < T_f$ the field H was turned off and the relaxation of the thermoremanent magnetization (TRM) from equilibrium state[23] was recorded for approximately 12 min. In all cases we find that the changes in the magnetization can be fitted to

$$M(t) = M_1 + S \ln t \qquad (2)$$

in the range $t_1 = 1$ min to $t_2 = 12$ min. This fit can usually be extended to lower t_1 values. At low temperature and high fields, however, deviations from $\ln(t)$ are found. These might be attributed to the changes in time evolution predicted theoretically[4] but need more accurate determination. To avoid this complication we restrict ourselves to the above time interval and focus on the field and temperature dependence of the coefficient S in Eq. (2).

The results obtained upon following the first procedure (ZFC and application of a constant field) can be summarized as follows: (a) For a constant temperature, S increases with field, peaks at $H_m(T)$, then decreases and vanishes at $H_c(T)$. (See Fig. 1—open circles—for typical results.) The amplitude of the peak S_m and its position $H_m(T)$ decrease with increasing temperature. We observe a parabolic dependence of S on H for most of the field interval (see Fig. 2). This enables extrapolation to $S = 0$ with quite reasonable accuracy. Note, however, that the parabolic decrease fails at high fields, above $H = H_t$, where the coefficient S levels off to a certain fraction of $S_{\max}$ (20% and 7% of $S_{\max}$ at 4.2 and 19 K, respectively; undetectable, with the present sensitivity, for $T > 30$ K) and then decreases slowly to zero.

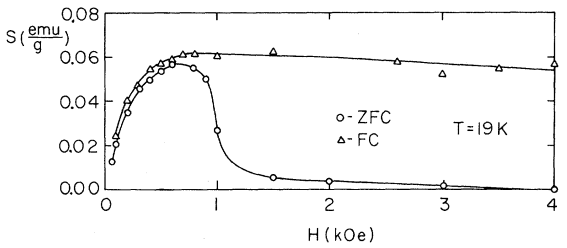

FIG. 1. Relaxation rates at $T = 19$ K as a function of field after zero-field-cooling process (circles) and after a field-cooling process (triangles).

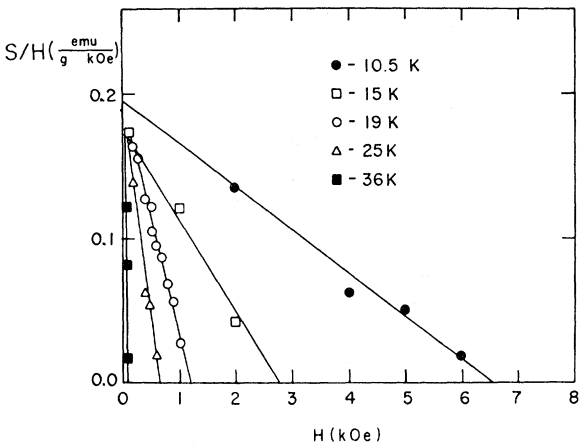

FIG. 2. Field dependence of S/H at several isotherms. The parabolic dependence of $S(H)$ is observed for most of the fields interval. Points which are part of the tail are not shown here.

Hereafter we refer to the high-field part of the $S(H)$ curve as a "tail." The tail is extrapolated to $S = 0$ by fitting the data to a power law, $S \propto (1 - H/H_c)^{\nu}$. The experimental errors[24] in the small-S values limit the accuracy in determining the parameters for the best fit. We find the exponent ν to vary between 1 and 1.5, whereas $H_c(T)$ is determined to within an accuracy of, typically, 20%. (b) For a constant field, S increases with temperature, peaks at $T_m(H)$ and then decreases and vanishes at $T_c(H)$. The magnitude of the peak S_m increases and its location T_m decreases with increasing field (see Fig. 3 for typical results). The tailing phenomena is not observed in $S(T)$ curves. The lines described by $T_m(H)$ and $T_c(H)$ coincide, within experimental error,[24] with the $H_m(T)$ and $H_c(T)$ lines defined above.

The validity of logarithmic decay law, such as Eq. (2), has been much debated. Of course, a power law $M \sim t^{-a}$ with $a \ll 1$ cannot be distinguished from a logarithmic relaxation. Monte Carlo studies strongly support logarithmic behavior and, in fact, give results for $S(H)$ strikingly similar to those reported here (see, e.g., Fig. 16 in Ref. 18, and the discussion associated with it). This point has been discussed recently by Bray and Moore[25] who point out the close association between a logarithmic decay and a nonzero density of zero-energy eigenstates.

In Fig. 4, we summarize the information derived from $S(H)$ curves by plotting two lines, labeled p and t, which define vanishing of viscosity phenomena. The lower branch (p) is found via parabolic extrapolation to a zero value of $S(H)$. The upper branch (t) describes the vanishing of the tail. Line p is the borderline between two viscous regimes in the H-T diagram, Fig. 4, whereas line t separates viscous and nonviscous regimes. It is quite tempting to identify these lines as the two transition lines predicted by

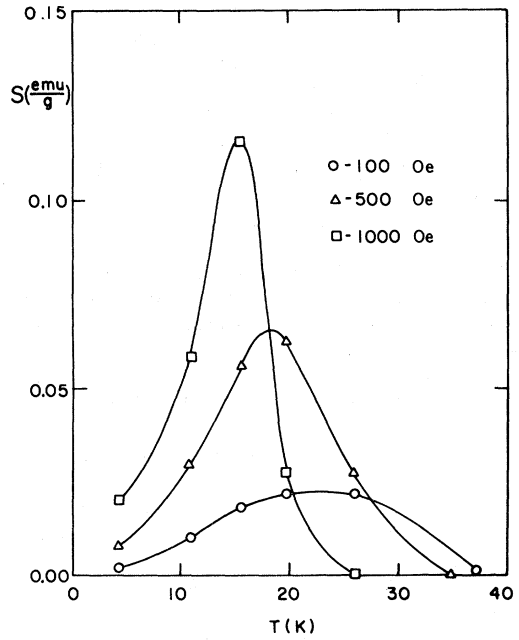

FIG. 3. Temperature dependence of the relaxation rate S for 100 Oe (circles), 500 Oe (triangles), and 1 kOe (squares).

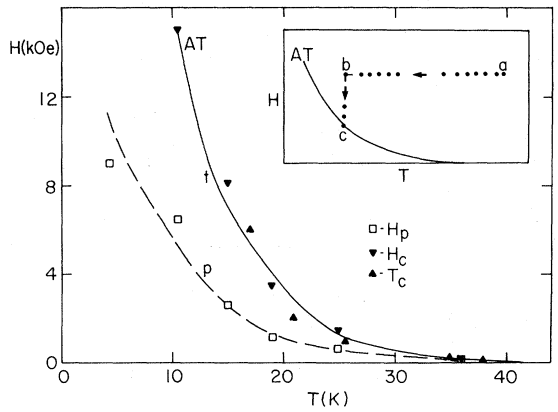

FIG. 4. Temperature/field phase diagram for $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$. Line p is the loci of $S=0$ found via parabolic extrapolation of $S(H)$. The solid line (t) is the de Almeida–Thouless line. Inset describes schematically a field-cooled process. At a point b, above the AT line, the field is turned off. A rapid relaxation to c is followed by a logarithmic decay in the spin-glass phase. The rate of the slow decay is independent of H (see text).

Gabay and Toulouse[14] for the non-Ising case (see Fig. 1 of Ref. 14). At present, however, there are no theoretical reasons for a change in the viscous characteristics between the two viscous regimes. It thus seems speculative at this stage to identify both lines as transition lines on the basis of the present data alone. Furthermore, some anisotropy is induced in the amorphous ribbons in the process of preparation.[22] This suggests more Ising-type spin-glass behavior and thus only one line of transitions is expected.[14] We therefore identify line t as the AT line. This line exhibits the main characteristics of the AT line. First, it marks the boundary between reversible and irreversible behavior. Second, for this line $H=H_0\tilde{t}^{1.6\pm0.2}$ in good agreement with the predicted behavior [Eq. (1)]. The field $H_0\simeq 40$ kOe, almost an order of magnitude smaller than predicted, reflecting, probably, spin-clustering effects.[26]

To complete our discussion, we mention briefly results obtained via procedures (ii) and (iii). Figure 1 shows results for TRM decay at 19 K in comparison with the in-field relaxation, procedure (i), at the same temperature. The most interesting phenomenon here is the roughly constant value of $S(H)$ at high fields [similar results are obtained via procedure (ii) and are also reproduced in Monte Carlo experiments[18]]. Figure 1 shows clearly asymmetry in the response to a symmetric change in the external field (turning the field on or off). This asymmetry and the plateau in $S(H)$ can be explained in terms of the AT line. In the inset of Fig. 4 we describe schematically a FC process. Starting at point a ($T>T_f$) the system is field cooled to point b in the PM regime (above the AT line). Then the field is turned off. The paramagnetic system responds quickly and relaxes at a point c on the borderline between PM and SG phases. From this point the TRM decays logarithmically. It is clear that the rate of relaxation of the TRM is independent of the field provided that point b is in the paramagnetic regime.

We have found, in summary, that the sudden application of a field to a spin-glass, cooled in zero field, leads to a relaxation where amplitude depends on the field step. We take the tendency for this relaxation to vanish above a critical field as evidence of a field-induced transition between SG and PM phases, as first suggested by de Almeida and Thouless. Certainly, much experimental and theoretical work remains to be done to elucidate these novel phase changes.

ACKNOWLEDGMENTS

We are grateful to Dr. H. Sompolinsky for discussing the experimental implications of Ref. 5. One of us (Y.Y.) has benefited greatly, during a stay at the Polytechnic Institute of New York, from discussions with Professor R. D. Parks. Thanks are due to Professor T. Egami and Dr. K. V. Rao for discussion of their results, Ref. 11. The helpful comments of Dr. J. Souletie are acknowledged. The sample used for this study was kindly provided by Dr. H. S. Chen. This work has been supported in part by National Science Foundation and Materials Research Laboratory Grant No. DMR79-23999.

*Permanent and present address: Department of Physics, Bar-Ilan University, Ramat-Gan, Israel.

[1]For a recent review of experimental work, see J. A. Mydosh, J. Magn. Magn. Mater. 7, 237 (1978).

[2]For a recent review of theoretical work, see K. Binder in *Proceedings of the Enschede Summer School of Fundamental Problems in Statistical Mechanics,* edited by E. G. D. Cohen (North-Holland, Amsterdam, 1981).

[3]S. K. Ma and J. Rudnick, Phys. Rev. Lett. 40, 589 (1978).

[4]H. Sompolinsky and A. Zippelius, Phys. Rev. Lett. 47, 359 (1981).

[5]H. Sompolinsky, Phys. Rev. Lett. 47, 935 (1981).

[6]C. N. Guy, J. Phys. F 5, L242 (1975); 7, 1505 (1978); 8, 1309 (1979).

[7]J. Ferré, J. Pommier, J. P. Renard, and K. Knorr, J. Phys. C 13, 3697 (1980).

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

[9]R. W. Knitter and J. S. Kouvel, J. Magn. Magn. Mater. 21, L316 (1980).

[10]Y. Yeshurun and M. B. Salamon, J. Phys. C 14, L575 (1981).

[11]T. Kudo, T. Egami, and K. V. Rao, J. Appl. Phys. 53, 2214 (1982).

[12]J. R. L. de Almeida and D. J. Thouless, J. Phys. A 11, 983 (1978).

[13]G. Parisi, Phys. Rev. Lett. 23, 1754 (1979); J. Phys. A 13, L115 (1980); 13, 1887 (1980).

[14]M. Gabay and G. Toulouse, Phys. Rev. Lett. 47, 201 (1981).

[15]S. F. Edwards and P. W. Anderson, J. Phys. F 5, 965 (1975).

[16]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. 35, 1792 (1975); S. Kirkpatrick and D. Sherrington, Phys. Rev. B 17, 4384 (1978).

[17]K. Binder and K. Schröder, Phys. Rev. B 14, 2142 (1976).

[18]W. Kinzel, Phys. Rev. B 19, 4595 (1979).

[19]C. Dasgupta, S. K. Ma, and C. K. Hu, Phys. Rev. B 20, 3837 (1979).

[20]M. B. Salamon, K. V. Rao, and H. S. Chen, Phys. Rev. Lett. 44, 596 (1980).

[21]Y. Yeshurun, M. B. Salamon, K. V. Rao, and H. S. Chen, Phys. Rev. B 24, 1536 (1981). Note: Fe concentration ($x = 0.36$) is found here via electron microprobe measurements. Nominal concentration ($x = 0.40$) appears in Ref. 20 for the same composition.

[22]H. S. Chen and C. E. Miller, Mater. Res. Bull. 11, 49 (1976).

[23]A. P. Malozemoff and Y. Imry, Phys. Rev. B 24, 489 (1981).

[24]Experimental errors defined here by the range of reproducibility. Error bars as a result of fitting data to Eq. (2) are much smaller.

[25]A. J. Bray and M. A. Moore, Phys. Rev. Lett. 47, 120 (1981).

[26]The break in $S(H)$ suggests that line p might be the AT line and, if this is the case, the tail reflects viscous phenomena, the origin of which is not a spin-glass phase. The exponent of line p is similar to the one found for line t but H_0 is smaller (see Fig. 4) pushing line p further away from the predicted line.