Effect of disorder on the magnetization of a spin glass

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The magnetization of spin glasses cooled in zero field show a characteristic logarithmic increase with time. If the sample is held at a measuring temperature just below the transition temperature for a waiting time t_w the dependence on $\ln t$ is faintly sigmoidal; so, the derivative with respect to $\ln t$ has a maximum, which is at about $\ln t_w$. We present results that show that this logarithmic time dependence also occurs in the nondisordered, fully frustrated pyrochlore $Y_2Mo_2O_7$. However, it differs from that in conventional spin glasses in that the maximum in the derivative is very broad and is not centered on t_w. The introduction of disorder by replacing some of the Mo with nonmagnetic Ti has the effect of markedly increasing the time-dependent moment, and restores the usual spin-glass behavior. [S0163-1829(99)02701-0]

INTRODUCTION

$Y_2Mo_2O_7$ is a member of a class of geometrically frustrated antiferromagnets. The source of the frustration is evident from Fig. 1: the structure of this material corresponds to a network of corner sharing tetrahedra. The Mo^{4+} are located on the corners of the tetrahedra, and are antiferromagnetically coupled. Frustration results from the impossibility of satisfying this requirement for all four spins.

A characteristic feature of spin glasses is that the magnetization of a sample cooled in zero field to a temperature just below the glass transition temperature T_g depends on how long it is held in zero field before the magnetic field is applied.[1,2] Upon the application of a small magnetic field, the magnetization evolves logarithmically in time. Waiting in zero field for a time t_w before turning on the magnetic field, has an important effect: the longer t_w the lower the resultant magnetization, and the dependence of the magnetization on $\ln t$ becomes slightly sigmoidal. Therefore, if the curve is differentiated with respect to $\ln t$ the derivative $S = \partial M/\partial \ln t$, displays a maximum, and if the temperature is about $0.9T_g$ the maximum occurs at about $\ln t_w$.

It is often stated that spin glasses are required to be both frustrated and disordered. Spin-glass behavior has been shown to exist in the pyrochlore $Y_2Mo_2O_7$ (Refs. 3,4) which is not disordered, although every spin is frustrated. What is particularly puzzling is that theory[5,6] and numerical results[7] appear to show that, at least classically, such systems do not order even at zero temperature. However, it is difficult to see how a permanent moment can be produced by cooling in a field if no static moments exist in the system, and it should be noted that recent neutron results[8] show a very weak, diffuse peak, indicating that permanent moments exist. It is clear, however, that if the magnetic order is associated with a domain structure the domains must be small and may have a very broad size distribution. In addition a wide spectrum of relaxation times would make correlations difficult to detect. The purpose of the work to be reported here was to investigate whether characteristic spin-glass aging phenomena exist in this material, and what effect the introduction of disorder, resulting from the replacement of some of the Mo with nonmagnetic Ti might have.

EXPERIMENT

The moment was measured as a function of time in a Quantum Design superconducting quantum interference device magnetometer. The samples were powders, and were identical to those used in obtaining the experimental data reported in Ref. 4. As emphasized in those publications neutron and x-ray diffraction studies of the nondisordered $Y_2Mo_2O_7$ show that there is no measurable mixing between the Y^{3+} and Mo^{4+} sublattices. Nor can any oxygen vacancies be detected, thus any random disorder due to oxygen vacancies is below the 1% detectability level. The samples were first cooled in zero field from a temperature well above Tg. A field of 5 Oe was then applied after either no waiting or waiting for 3000 sec.

RESULTS AND DISCUSSION

The magnetization of the three samples showed the usual $\ln t$ dependence which decreased if t_w was increased. Differences between the nondisordered sample and the others only became evident in S. The magnetizations as a function of $\ln t$

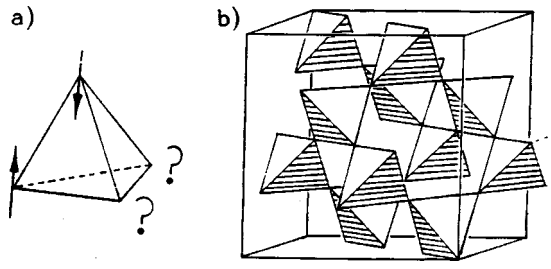

FIG. 1. (a) Illustrates the frustration resulting from four antiferromagnetically coupled spins on a single tetrahedron. (b) The three-dimensional network of corner sharing tetrahedra.

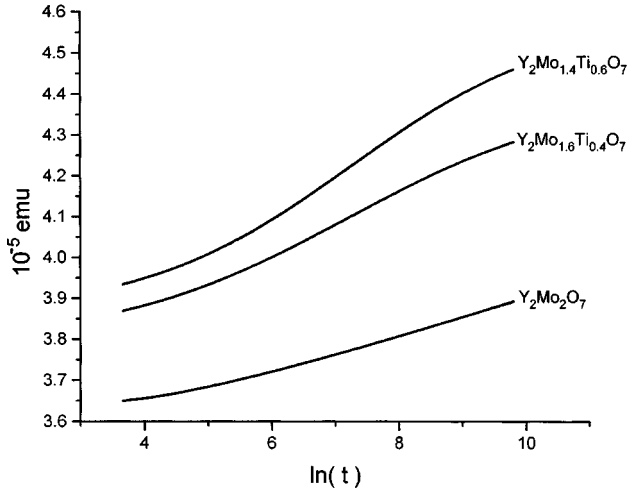

FIG. 2. Moment as a function of time for samples cooled in 0 magnetic field from above T_g. A field of 0.005 T was applied after waiting in zero field for 3000 sec. The data are for nondisordered $Y_2Mo_2O_7$ at 15 K (T_g for this material is 23 K), $Y_2Mo_{1.6}Ti_{0.4}O_7$ at 10 K (Tg=15 K), and $Y_2Mo_{1.4}Ti_{0.6}O_7$ at 8 K (T_g=12 K).

are shown in Fig. 2 for a waiting time of 3000 sec at temperatures equal to $0.6T_g$. The derivative is plotted in Fig. 3. The reason for this choice of measuring temperature is that recently Eftimova[9] has shown that the magnitude of S is strongly temperature dependent, with a maximum at about $0.6T_g$. Recent measurements have confirmed this in other spin glasses.[10]

The results for the nondisordered material are unlike those published[1,2] for other spin glasses. On the introduction of disorder by the substitution of nonmagnetic Ti for Mo, conventional spin-glass behavior is obtained. This difference can be explained by the presence of percolation clusters in the disordered material, and their absence in the nondisordered sample.

A domain model has been shown to completely account for the peak in $S=\partial M/\partial \ln t$.[11] Those arguments will now be summarized briefly: A number of different theoretical approaches[12–17] yield the result that the domain size R is

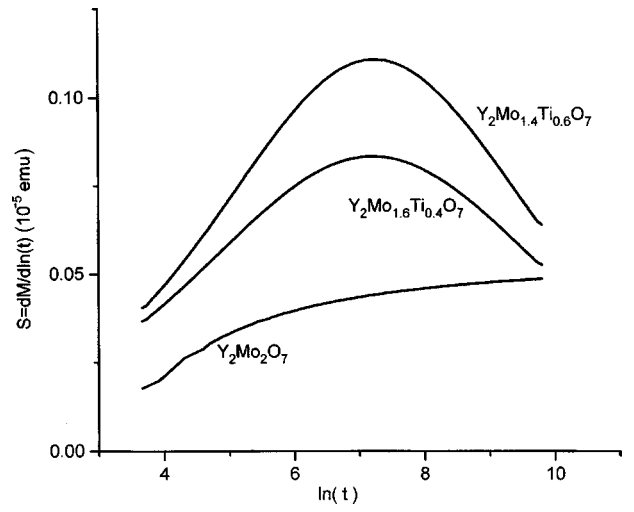

FIG. 3. The derivatives of the curves in Fig. 2 with respect to $\ln t$.

proportional to $\ln t$; so, in general we may write

$$R=A[\ln(t/\tau)]^a=A\ \ln^a(t/\tau), \tag{1}$$

where A can depend on temperature and the value of a depends on the model, varying between 0.88 (Refs. 11,17) and 4.[16]

It is clear that if t is measured from the end of a waiting time t_w

$$R=A\ \ln^a\left(\frac{t_w+t}{\tau}\right)\equiv A\ \ln^a(g+k). \tag{2}$$

The number of spins in a domain will be $R^D=A^D\ \ln^{aD}(g+k)$, where D is the fractal dimension, $D=2.5$ on a fractal structure.[18]

The relaxation time may be approximated by $\tau=\omega_0^{-1}e^{(Q+E)/T}$ where ω_0 is an attempt frequency, Q is an intrinsic crystal field barrier to spin reversal, E is an effective medium[19] barrier due to the interaction of the spins in the sequence with their neighbors. In a magnetic field H this expression becomes

$$\tau^{\pm}=\omega_0^{-1}e^{(Q+E\pm\mu H)/T}=\tau e^{\pm h}. \tag{3}$$

When a magnetic field is applied the domains continue to grow, but at different rates. The new volumes must be in thermal equilibrium, and the sample develops a moment proportional to the field and the new volume. With $b=aD$, the average moment per domain for vanishingly small fields is

$$m_d=A^Dh[\ln^b(g+k)-\ln^b g]. \tag{4}$$

In a disordered system, the spins can be expected to lie on a percolation structure. At concentrations below the percolation threshold the system will consist of a number of separated clusters. Above the threshold an infinite cluster appears which can be viewed as a collection of clusters[20] some of which are connected, this is the ''blobs and links'' model.[21]

The cluster size distribution can be approximated by $s^{\theta}e^{-Cs^{\nu}}$ where s is the number of spins in the cluster[20] and C is a constant on the order of the reciprocal of the number of spins in the average cluster. In three dimensions, above the percolation threshold $\theta\sim\frac{1}{9}$, and $\nu=\frac{2}{3}$, below $\theta\sim-\frac{3}{2}$ and $\nu=1$.

Clusters smaller than $\ln^a g$ will be single domain. After the field is applied domain growth will take place in the larger clusters at different rates for the two spin orientations. As a consequence clusters with $\ln^a g<R^{\pm}\leqslant\ln^a(g+ke^{\pm h})$ will become single domain. The difference between their volumes will contribute to the moment. The number of spins in the domain is $\sim R^D$ and this contribution to the moment is

$$\delta M_{\text{cluster}}\propto\int_{\ln^b g}^{\ln^b(g+ke^h)}s^{1+\theta}e^{-Cs^{\nu}}-\int_{\ln^b g}^{\ln^b(g+ke^{-h})}s^{1+\theta}e^{-Cs^{\nu}} \tag{5}$$

$$\approx h\frac{k}{(g+k)}[\ln^{(2+\theta)b-1}(g+k)e^{-C\ \ln^{\nu b}(g+k)}]. \tag{6}$$

Differentiation of Eq. (6) with respect to $\ln t$ leads to a term $k/(g+k)-[k/(g+k)]^2=gk/(g+k)^2$, which has a maximum at $k=g$. In Ref. 11 it was demonstrated that Eqs. (6)

and (4) can account for the dependence of S on t_w and $\ln t$, including the strong dependence of the initial value of S on waiting time.

If this explanation is correct, and the peak in S is a result of a percolation structure, it should not be present in a nondisordered material. Due to eventual saturation a maximum will appear, but it should be quite unlike that in the disordered material. Reference to Fig. 3 reveals that this is indeed the case.

Furthermore, the magnitude of the peak value of S depends on the number of clusters whose size is on the order of the domain size. This number must increase as the concentration decreases for concentrations above the percolation limit, which explains the larger value of S for $Y_2Mo_{1.4}Ti_{0.6}O_7$.

CONCLUSIONS

From these results it may be concluded that although $Y_2Mo_2O_7$ is a frustrated system with no ordered ground state, and the magnetization increases with $\ln t$, the absence of a peak in $S = \partial M/H\partial \ln t$ distinguishes it from the classical spin glasses. The substitution of nonmagnetic Ti for some of the Mo, thereby introducing structural disorder, restores conventional spin-glass behavior and strongly enhances the magnetization.

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