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The relative influences of disorder and of frustration on the glassy dynamics in magnetic systems

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Abstract

The magnetization relaxations of three different types of geometrically frustrated magnetic systems have been studied with the same experimental procedures as previously used in spin glasses. The materials investigated are $Y_2Mo_2O_7$ (pyrochlore system), $SrCr_{8.6}Ga_{3.4}O_{19}$ (piled pairs of Kagomé layers) and $(H_3O)Fe_3(SO_4)_2(OH)_6$ (jarosite compound). Despite a very small amount of disorder, all the samples exhibit many characteristic features of spin glass dynamics below a freezing temperature T_g, much smaller than their Curie–Weiss temperature θ. The ageing properties of their thermoremanent magnetization can be well accounted for by the same scaling law as in spin glasses, and the values of the scaling exponents are very close. The effects of temperature variations during ageing have been specifically investigated. In the pyrochlore and the bi-Kagomé compounds, a decrease of temperature after some waiting period at a certain temperature T_p reinitializes ageing, and the evolution at the new temperature is the same as if the system were just quenched from above T_g. However, as the temperature is raised back to T_p, the sample recovers the state it had previously reached at that temperature. These features are known in spin glasses as rejuvenation and memory effects. They are clear signatures of the spin glass dynamics. In the Kagomé compound, there is also some rejuvenation and memory, but much larger temperature changes are needed to observe the effects. In that sense, the behaviour of this compound is quantitatively different from that of spin glasses.

(Some figures in this article are in colour only in the electronic version)

It was usually thought that both disorder and frustration were necessary to obtain spin glass dynamics (see, for example, [1–3]). However, several frustrated but non-disordered antiferromagnets, rather than condensing into a spin liquid state as theoretically expected,

were found to enter a glassy state below a well defined freezing temperature. As in usual spin glasses, below this temperature T_g their dynamic response to a small magnetic field becomes very slow and depends on the time t_w spent below T_g ('ageing'; see, for example, [1–3]). The question thus arises whether these glassy materials are really similar to standard spin glasses. We have addressed this question by investigating three examples of such antiferromagnets using the same experimental procedures as previously used in the detailed study of the dynamical properties of spin glasses. The three examples correspond each to a different topology of the frustrated spin network. The first one is $Y_2Mo_2O_7$ ('YMO'), where the spins ($S = 3/2$) are on a three-dimensional (3D) pyrochlore lattice [4, 5]. The second one is $SrCr_{8.6}Ga_{3.4}O_{19}$, ('SCGO'), where the spins are located on bi-layers of 2D Kagomé lattices, each bi-layer being well separated from the others [6, 7]. The third one is a jarosite compound $(H_3O)Fe_3(SO_4)_2(OH)_6$, where the spins ($S = 5/2$) are located on genuine 2D Kagomé layers, with small interlayer couplings [8].

At high temperature, all three compounds exhibit a Curie–Weiss behaviour with a large negative value for the Curie–Weiss temperature θ, indicating strong antiferromagnetic interactions between spins. The values of θ are much larger than the observed freezing temperatures T_g. The ratio $|\theta|/T_g$ is of order 10 for the (3D) YMO sample [4], about 140 for SCGO [6], and about 60 for the (2D) Kagomé sample [9]. This reveals that frustration plays a key role in the low-T physics.

Below T_g a clear separation between zero field cooled (ZFC) and field cooled (FC) magnetizations is found. This is a standard indication suggesting that the low-temperature phase is a spin glass phase. The spin glass transition is further confirmed by other measurements:

(i) in the Kagomé sample an analysis of the critical dynamics was carried out over 10 decades in frequency (including Mössbauer experiments) and it yielded the critical exponents usually found in standard spin glasses [10];
(ii) in YMO [4] it was shown that the nonlinear susceptibility diverges at T_g, as expected for spin glasses.

In addition, both in YMO [5] and in the Kagomé sample [11], neutron diffraction experiments do not detect any long range ordering.

The amount of disorder is very small in all these materials. In YMO the main source of disorder might be a small percentage of Mo–Mo bonds whose lengths are slightly different from the standard one [5]. In SCGO an obvious source of disorder could arise from the non-stoichiometry of the compound which leads to a Cr coverage of the magnetic sublattice of $x = 8.6/9 = 95\%$, i.e. to an amount of disorder of 5%. However, since it was shown that T_g is an increasing function of x [6], one expects a spin glass phase even in the stoichiometric compound. Thus, the imperfect coverage of the magnetic sublattice does not seem to trigger the spin glass phase. Last, in the Kagomé sample [9], the coverage of the Fe network is higher than 97%, leading, as in the two other compounds, to an upper bound of a small percentage of disorder.

To further investigate the spin glass phase in these samples, we first performed thermoremanent magnetization (TRM) experiments. In the measurements described here, the sample is quenched from above T_g to $T_m = 0.7 \times T_g$ in a magnetic field of $H = 50$ Oe. After a certain waiting time t_w at T_m, the field is switched off and the magnetization M is recorded as a function of time t. The results are shown in figure 1. They look very similar to those of standard spin glasses, displaying a very slow t_w dependent relaxation. The larger t_w, the slower the relaxation, and the larger the remanent magnetization at a given time t. This indicates that more and more spin correlations develop as time t_w becomes larger [1–3, 12].

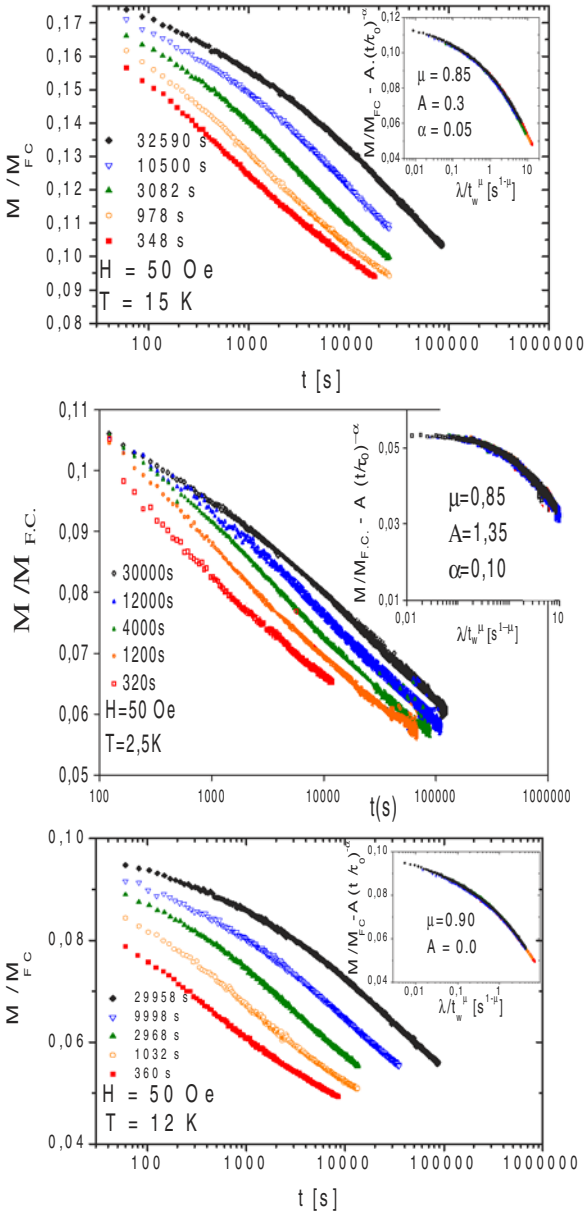

Figure 1. Thermoremanent magnetization experiments carried out at $T = 0.7 \times T_g$ (see the text). From top to bottom: YMO, SCGO, Kagomé samples. The longer the waiting time t_w, the slower the relaxation when the magnetic field is switched off. Just as in standard spin glasses, the ageing magnetization $M(t, t_w) - A(\tau_0/t)^\alpha$ can be scaled (see the insets) as a function of λ/t_w^μ, where $\lambda = t_w[(1 + t/t_w)^{1-\mu} - 1]/[1 - \mu]$, i.e. $\lambda/t_w^\mu \simeq t/t_w^\mu$ for $t \lesssim t_w$. The best fit gives parameters very close to those found in standard spin glasses. For YMO $\mu = 0.85$, $\alpha = 0.05$, $A = 0.3$; for SCGO $\mu = 0.85$, $\alpha = 0.10$, $A = 1.35$; for the Kagomé sample $\mu = 0.90$ and $A \simeq 0.0$.

As can be seen in figure 1, the whole set of TRM experiments yields results very similar to those of spin glasses. Moreover, applying the standard scaling procedure used in spin glasses, (see, for example, [2]) allows a perfect scaling of all the $M(t, t_w)$ data on a unique curve. This is

obtained by fitting the relaxation curves to an expression with two additive terms. The first one is a stationary (t_w independent) power law relaxation $A(\tau_0/t)^\alpha$ which is predominant at very small times. The second term is a function $F(t, t_w)$ of t and t_w. This function is found to scale as t/t_w^μ at short t with μ being an exponent close to 1. In this short t region, t_w is approximately the age of the system, i.e. the time spent at the measuring temperature. At larger t, the real age must be taken as $t + t_w$; in other words, the system continues to age as one measures its relaxation. It can then be shown that the correct scaling variable should be rewritten as λ/t_w^μ, where $\lambda = t_w[(1 + t/t_w)^{1-\mu} - 1]/[1 - \mu]$. Note that one recovers $\lambda/t_w^\mu \simeq t/t_w^\mu$ for $t \lesssim t_w$, as expected. The insets of figure 1 show the plots of the function F versus this reduced variable for the present samples. The scaling is shown to apply quite well. Setting the microscopic attempt time to $\tau_0 = 10^{-12}$ s, one finds fitting parameters very close to those of the standard spin glasses (see the caption of figure 1), especially for the ageing exponent μ which is slightly below 1, indicating the usual 'sub-ageing' phenomenon.

Beyond these standard TRM measurements which are performed at constant temperature (after the initial quench), experiments involving temperature variations during ageing below T_g usually reveal very striking characteristic features in standard spin glasses [13, 14]. For example, it is now well established that ageing at any $T_p < T_g$ has no apparent effect on the state at all other temperatures sufficiently different from T_p. This was found, for instance, in ZFC experiments in which modified cooling protocols were applied [13, 14]. While in the usual ZFC protocol the system is cooled (at a constant cooling rate) in zero field from above T_g down to the lowest temperature, in the modified protocols, the cooling is interrupted at some temperature(s) T_p and resumed after some waiting time t_w. In both cases, a small dc field is applied at the lowest temperature reached, and the magnetization is recorded while the system is heated up at a constant rate. The ZFC magnetization, as measured in the modified protocol, clearly showed marked singularities ('dips') centred around the temperature(s) T_p. Far enough from T_p, it recovered the values of the usual (reference) ZFC magnetization (see [13, 14]). This showed that ageing at T_p during t_w did not affect the system at a temperature T different from T_p. It also implied that the system kept the memory of ageing at T_p while it was at lower temperatures and was able to retrieve the aged state previously reached at T_p. These results proved that in spin glasses different spin correlations are building up at different temperatures, and that the correlations built up at any temperature remain imprinted at lower temperatures. *In these experiments, the width of the dips gives an idea of the 'temperature selectivity' of the spin correlations*. The results of such ZFC experiments are displayed in figure 2 for the three considered compounds. In this figure, the differences between the modified ZFC and the reference ZFC data are plotted as a function of temperature. Clear singularities are observed around the various chosen values of T_p exactly as in spin glasses. The (3D) YMO sample as well as the SCGO material present narrow dips characterized by a value $\delta T/T_g = 0.17 \pm 0.03$ very close to the values found in standard spin glasses: for instance, $\delta T/T_g = 0.16 \pm 0.03$ in the $CdCr_{1.7}In_{0.3}S_4$ Heisenberg spin glass for comparable t_w, while $\delta T/T_g = 0.21 \pm 0.03$ in the $Fe_{0.5}Mn_{0.5}TiO_3$ Ising spin glass. This is in contrast with the results on the Kagomé (2D) compound, where $\delta T/T_g = 0.39 \pm 0.05$ is remarkably larger.

The 'temperature selectivity' has been studied more quantitatively by measuring the effect at some temperature T of ageing at a slightly smaller temperature $T - \Delta T$. This experiment yields a TRM measurement with the following procedure:

(i) after field cooling from above T_g to T, the sample is kept for a short time $t_1 = 500$ s at constant T,

(ii) it is then further cooled to $T - \Delta T$ and aged during $t_2 = 9000$ s before being reheated to T where it stays for another short time $t_3 = t_1$,

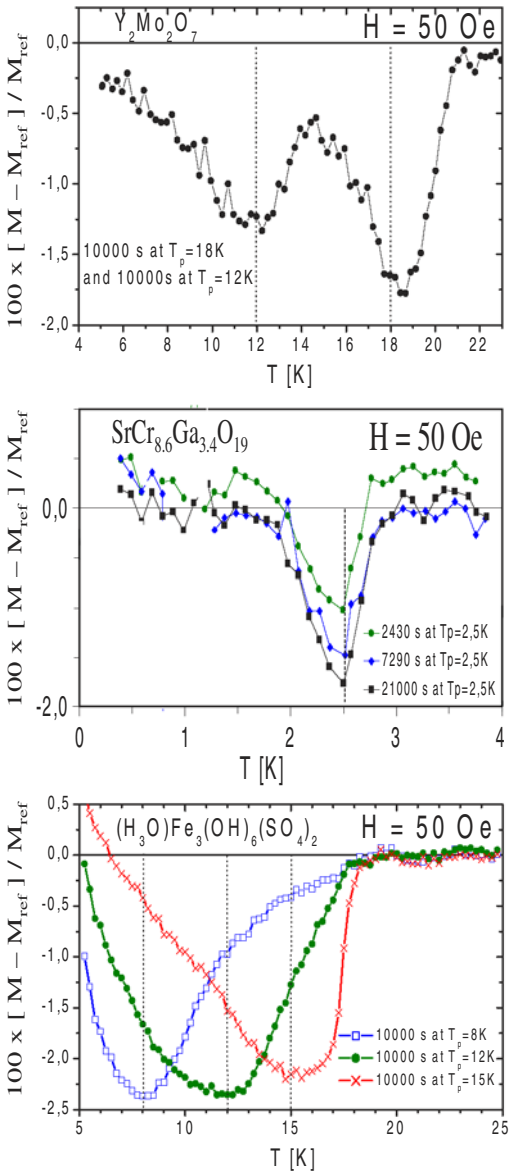

Figure 2. Memory dip experiment (see text): $M_{\rm ref}$ is measured in a standard ZFC experiment, while $M(T)$ corresponds to the experiment where the cooling was interrupted for a time $t_{\rm w}$ at a temperature $T_{\rm p}$ (marked by the dotted lines). From top to bottom: YMO, SCGO, Kagomé samples. The width of the memory dip is quite small in YMO and in SCGO (just as in standard spin glasses). It turns out to be much larger in the Kagomé sample.

(iii) last, the field is switched off and the TRM curve is recorded.

From these experiments, an effective time $t_2^{\rm eff}$ can be defined such as to superimpose the measured TRM with a purely isothermal TRM recorded at temperature T with waiting time $t_1 + t_2^{\rm eff} + t_3$. In other words, $t_2^{\rm eff}$ is a measure of the effect at T of ageing at $T - \Delta T$ for a time t_2. The actual quantitative results for the presently studied materials are reported in

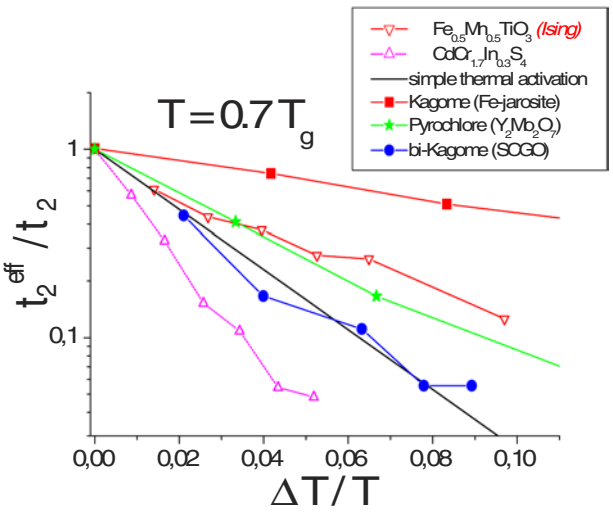

Figure 3. $t_2^{\rm eff}/t_2$ plotted versus $\Delta T/T$ (see text): the data for $Fe_{0.5}Mn_{0.5}TiO_3$ and $CdCr_{1.7}In_{0.3}S_4$ are plotted as examples of, respectively, an Ising spin glass and an Heisenberg spin glass. The steeper the slope in this plot, the more 'temperature-selective' the ageing (solid line: prediction assuming that ageing occurs via thermal activation with an activation energy independent on T). In contrast to SCGO and YMO data, which fall within the standard spin glass area, the Kagomé data are well above this region, with a very small slope.

figure 3 and compared to those obtained in [15] for the $CdCr_{1.7}In_{0.3}S_4$ Heisenberg spin glass and for the $Fe_{0.5}Mn_{0.5}TiO_3$ Ising spin glass. The results for YMO and SCGO are clearly in agreement with those of spin glasses: the slopes of $t_2^{\rm eff}/t_2$ versus ΔT for these two systems are indeed within the values delimited by the Heisenberg and Ising cases. As for the Fe-jarosite, the effective time $t_2^{\rm eff}$ does not vary much with the change of temperature. The system ages almost as quickly at $T - \Delta T$ as at T, and builds up the same pattern of spin correlations at both temperatures.

The conclusion of this investigation is that, in spite of their very small amount of disorder, the two frustrated antiferromagnets, the YMO pyrochlore and the bi-Kagomé SCGO, behave exactly as standard spin glasses. The Fe-jarosite Kagomé compound, though presenting many similarities in terms of the existence of a freezing temperature and the presence of ageing effects, has quite different dynamical properties as a function of temperature. It appears, indeed, in figure 3 that $t_2^{\rm eff}$ remains very close to t_2 even for larger ΔT, meaning that ageing at $T - \Delta T$ contributes significantly to the age of the system at T. The behaviour is not very temperature selective, implying that the pattern of the spin correlations is not very temperature dependent. The two-dimensional character of the jarosite compound may be at the origin of the observed differences with the two other materials [16].

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