Exponential equilibration by slow cooling in the planar random-anisotropy magnet: Experiments and simulations

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Neutron measurements of the equilibration of the staggered magnetization in $DyAs_{0.35}V_{0.65}O_4$ are compared with Monte Carlo simulations of spin dynamics in a planar random-anisotropy magnet. The simulation results are in agreement with striking observed relaxation phenomena: when cooled rapidly to a low temperature, no magnetic ordering is observed; however when cooled in small steps an ordered magnetic moment appears, which is found to equilibrate exponentially with time at temperatures through and below the transition temperature. In contrast to the freezing of spins in other random systems, the time scale of the relaxation in this system does not increase significantly even at the lowest temperatures.

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I. INTRODUCTION

The presence of disorder in magnetic systems generally leads to a characteristically slow (glassy) relaxation of the magnetization at low temperatures. In typical spin glasses there is no sharp magnetic ordering transition and no spin equilibration on laboratory time scales.[1] Models for analysis and simulations of disordered magnets generally include random exchange and random uniaxial anisotropy, which are believed to be the most relevant mechanisms for spin-glass properties.[2] There is an extensive literature on the effects of both random exchange and random uniaxial anisotropy, with most emphasis on random exchange interactions, which are certainly important in the commonly studied metal alloy systems. However some recent experiments[3] and simulations[4] have demonstrated the important role of random anisotropy on the spin freezing and relaxation processes in these systems. It should be possible to study the effects of random anisotropy separately from random exchange by choosing systems where the latter should not be significant. This can be achieved in crystalline samples by not diluting or mixing the magnetic ions but instead by partial substitution of neighboring ions. Some experiments in such a system, $DyAs_xV_{1-x}O_4$, were reported previously[5,6] and are further discussed here. Measurements of the magnetization relaxation in this material, which is believed to be a random planar anisotropy magnet (RPAM) showed surprising behavior:[6] for example, in contrast to other random magnetic systems the magnetization relaxation times did not become very long at lower temperatures but remained roughly constant or even decreased. To address the question as to whether this property was a fundamental characteristic of RPAM systems or whether it had some other origin, we have carried out Monte Carlo simulations on the RPAM model and compared the results with experiments. A good agreement was found between the simulations and the observed relaxation behavior, confirming that it is a characteristic of the RPAM system.

II. EXPERIMENTS

First the sample and the experiments will be briefly described: further information can be found in Refs. 5 and 6. In $DyAs_{0.35}V_{0.65}O_4$ the Dy-Dy interactions are expected to drive an antiferromagnetic ordering transition at a temperature of a few Kelvin. The As/V substitutions generate uniaxial random anisotropy at each Dy site without significantly altering the Dy-Dy interactions. The Dy spins lie in the basal plane of the tetragonal structure and order parallel to one or the other basal plane axes depending on the local anisotropy. In neutron experiments carried out at the Canadian Neutron Beam Centre, Chalk River, the growth of the antiferromagnetic (100) peak was studied as the sample was cooled below 1.6 K. No ordering field is applied in these experiments: the peak intensity grows in response to temperature reductions giving a direct measure of the staggered magnetization. It should be noted that the study of magnetic peaks by neutron scattering has rarely been an effective technique in investigations of spin glasses and other disordered magnets. In many samples the severe chemical disorder destroys the lattice periodicity; in others where there may be a well defined lattice the random magnetic interactions lead to a disordered short-range spin freezing structure that gives only a very broad magnetic diffraction peak. The present system is very favorable for diffraction studies since the lattice is only slightly disordered, and it is known that planar interactions are able to drive a magnetic transition with quasi-long-range order even in the presence of strong random anisotropy.[7] As shown previously[5,6] the neutron experiments do show a clear but slightly rounded magnetic transition at $T_c = 1.6$ K with a magnetic peak that is quite narrow but not resolution limited, implying correlation lengths of about 40 nm.

Our experiments showed that when the sample was cooled rapidly to a temperature well below 1.6 K there was no detectable (100) peak and hence there is no magnetic ordering, even after waiting 24 h. However, when the tem-
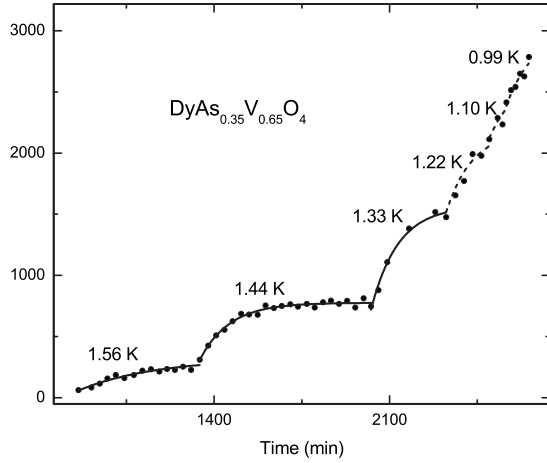
FIG. 1. Growth of (100) magnetic peak intensity while cooling in small steps. Solid lines are fits to exponential equilibration and dashed lines are guides to the eye.

perature was reduced in small steps (typically 0.1 K) and held constant for a few hours at each temperature while the (100) peak was scanned repeatedly, the magnetic peak grew and reached a stable intensity. To achieve temperatures below 1 K a ^{3}He cryostat was used, but since its cycle time was limited to ~30 h the time available at each temperature was significantly restricted. As a result the experimental statistics are relatively poor but the main results are nevertheless clear. The growth of the magnetic peak intensity as a function of time is shown in Fig. 1 along with exponential fits. The long set of data at 1.44 K shows exponential equilibration most convincingly. The equilibration times obtained for the fits were as follows: 307 ± 135 min at 1.56 K; 118 ± 10 min at 1.44 K; and 107 ± 23 min at 1.33 K. At the three lowest temperatures where the data were too sparse to allow unrestricted fitting, the dashed lines are exponential fits with the relaxation time set equal to 107 min. They are guides to the eye but provide evidence that the relaxation times are not lengthening rapidly at these low temperatures, in contrast to the pronounced freezing of the spin dynamics found in most disordered magnets.

The above results can be put into meaningful context by comparison with results for identical experiments carried out on a sample with reduced As substitution, $DyAs_{0.17}V_{0.83}O_4$. In this sample the structural disorder was not large enough to inhibit the tetragonal to orthorhombic transition that occurs in pure $DyVO_4$ at 14 K but instead it only suppressed the transition temperature to about 8 K.[8] As a result the random crystal fields are overwhelmed by the uniform orthorhombic distortion and we do not expect a random-anisotropy effect on the magnetic transition although some disorder still exists. In this sample a strong (100) magnetic peak appeared, signaling magnetic ordering at 2.3 K. In contrast to the behavior of the sample with 35% As concentration, which remains tetragonal on average with random anisotropy axes, the (100) peak was resolution limited indicating long-range order and its growth showed no observable time dependence. Although these neutron experiments cannot access the actual relaxation times, we expect that they are a characteristic of a pure system, which is of order a spin-flip time h/J extended by critical slowing down.
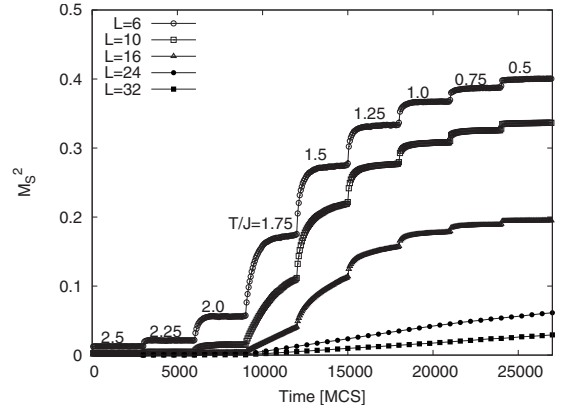

FIG. 2. Relaxation of the staggered magnetization for different system sizes (from top to bottom $L=6$, 10, 16, 24, and 32). The anisotropy to exchange ratio is $D/J=7$.

III. RPAM SIMULATIONS

The system is described by the following classical Heisenberg Hamiltonian:

$$H = -J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j - D \sum_i (\vec{n}_i \cdot \vec{S}_i)^2, \qquad (1)$$

where D and J are the anisotropy and the exchange couplings, respectively. The spin variable $\vec{S}_i$ is a three component unit vector associated with the ith node of a cubic lattice and the first sum runs over all nearest-neighbor pairs of spins. $\vec{n}_i$ is a unit random vector that defines the local easy axis direction of the anisotropy at site i. These easy axes are quenched variables chosen from a given distribution: the RPAM model is defined by choosing an isotropic distribution on a circle in the x-y plane.

The simulations were performed in a system of $N=L^3$ spins using a Monte Carlo Metropolis algorithm with periodic boundary conditions. In the case of the RPAM we set $J>0$ when the neighboring spins are in the x-y plane and $J<0$ in the other case. Thus when $D=0$ we have a planar antiferromagnet.

In order to follow the cooling protocol outlined in Ref. 5, we started every run at a temperature above the ordering temperature (around $T/J=1.8$) and then reduced it in uniform steps. Immediately after each step the relaxation of the root-mean-square staggered magnetization is measured during a time period larger than the longest observed relaxation time. Times are expressed in Monte Carlo steps (MCS) where a MCS is defined as a complete cycle of N-spin update trials. The results are shown in Fig. 2 for $D/J=7$ and different system sizes; each curve was averaged over 10^2-10^3 samples. For system sizes $L=24$ and 32 only temperatures larger than $T/J=1.75$ are shown. It can be seen that the relaxation times show qualitatively the same behavior as in Fig. 1, in particular the relaxation times do not increase significantly at low temperatures. Figure 2 also shows that the staggered magnetization takes lower values in the whole temperature range when the system size increases, suggesting, as expected,[7] that the ordered state has zero staggered magnetization in the thermodynamic limit. In contrast, the
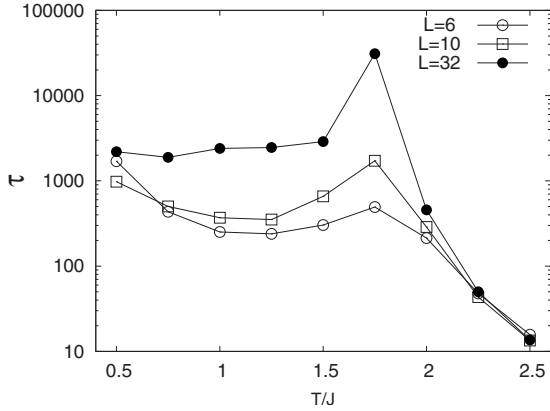
FIG. 3. Relaxation time vs temperature for the RPAM model (obtained from fittings of curves shown in Fig. 2).

FIG. 4. Relaxation of M_S^2 after quenching to $T/J=0.5$ ($L=10$) together with logarithmic fit. For comparison the relaxation after slow cooling down to the same temperature for $L=10$ as shown in Fig. 2 is also included.

magnetic peak observed experimentally is not much smaller than that for the conventional antiferromagnet $DyAs_{0.17}V_{0.83}O_4$. This disagreement could arise from a small difference between the interactions in the real system from those assumed in the model, for example the distribution of anisotropy directions may not be uniform in the basal plane.

The relaxation curves shown in Fig. 2 are well fitted by exponential functions and we show the relaxation times τ in Fig. 3 as a function of temperature. A distinct peak in the relaxation times can be observed at $T/J=1.75$ for every system size; the height of the peak increases with the system size. This behavior strongly resembles the critical slowing down properties for a system undergoing a conventional second-order phase transition. Below T_c the relaxation times drop to a plateau where they remain relatively unchanged down to low temperatures (especially for large L). Our experiments can only provide data below T_c for comparison but they show the same qualitative behavior as the simulations below T_c, with τ initially decreasing and then remaining roughly constant.

In contrast, when we quenched the system to $T < T_c$ in our simulations the staggered magnetization was found to grow on a time scale much greater than those observed for slow cooling. Figure 4 illustrates this behavior for a quench to $T/J=0.5$. The slow relaxation can be fitted by a logarithmic function as shown, although a stretched exponential also gives an adequate fit.

The value of D/J appropriate to our sample is unknown but we expect it to be large because in $DyVO_4$ the ion-lattice interactions are strong, and they induce a crystallographic phase transition at a temperature much higher than the magnetic transition. We determined relaxation times in simulations with D/J values of seven and ten and show the results in Fig. 5. While the critical temperature and temperature dependence seem to be independent of the anisotropy, the most remarkable result is the rapid, perhaps exponential, increase in the low-temperature relaxation time with D/J. This suggests an Arrhenius mechanism and can be understood by the increase in the local barriers due to the anisotropy. A comparison of MCS and experimental time scales confirms that a large D/J value is appropriate, but we have not attempted to match the time scales by repeating the simulations with
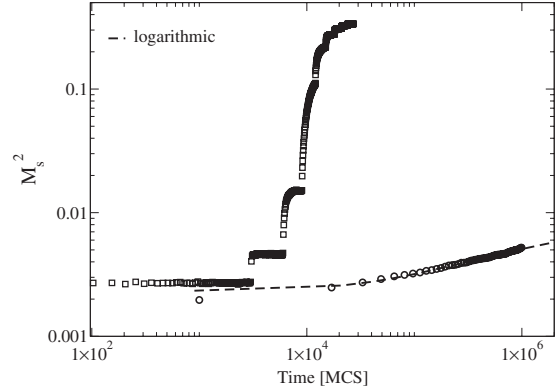
larger D/J values because the model is too simple in some respects, for example, the magnetic interactions are assumed nearest neighbor only and only a single value of D is used.

IV. DISCUSSION

The relaxation properties observed here are quite different from those of other spin systems. For example, if non-interacting spins are cooled in an applied field, exponential equilibration occurs with a characteristic time constant, and if the system is quenched and observed on a much shorter time scale there would indeed be no observed relaxation. The key difference is that in that system, unlike ours, the relaxation time does not depend on the cooling rate. Moreover, since relaxation depends on thermal processes, it slows dramatically at lower temperatures in contrast to the temperature-independent behavior that we observed under slow cooling. If we compare our system with typical spin glasses, in both cases the spins freeze and are unable to order after rapid cooling. However our system differs in being able to equilibrate fully even at the lowest temperatures when cooled slowly. In some magnetic systems the observation of temperature-independent relaxation has been explained by a
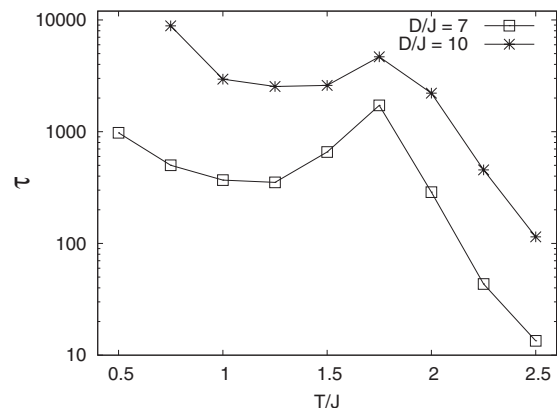

FIG. 5. Relaxation times in the RPAM model for $L=10$ and different anisotropy strengths.

quantum tunneling mechanism.[9] However those observations are concerned with the response of the system to a magnetic field, whereas in our case we are dealing with a purely thermal response, namely, the growth of the staggered magnetization following a reduction in temperature. This requires energy exchange between the spin system and a thermal reservoir, a scenario quite different from that in which quantum tunneling operates.

In the hope of clarifying the relaxation mechanism in our system and determining if it is unique to the RPAM system, we have carried out further simulations using the same cooling protocol on the $3d$ RAM system and the pure Heisenberg model. Full results will be reported later[10] but preliminary results show some significant similarities. Similar to the RPAM system, the $3d$ RAM system shows glassy dynamics when cooled rapidly but equilibrates exponentially when cooled slowly. The relaxation times do not show a well defined peak, consistent with the expected lack of long-range order for the $3d$ RAM[2,11] but reach a plateau with little variation with temperature at lower temperatures. In the pure Heisenberg system the magnetization does not show glassy dynamics but the relaxation times show a temperature dependence that is qualitatively similar to the RPAM system, with a well defined peak corresponding to critical slowing down and a plateau at lower temperatures. The actual relaxation times are, of course, many orders of magnitude shorter. The surprising inference is that random anisotropy does not qualitatively alter the relaxation dynamics if the system is slowly cooled, although the relaxation times are increased by orders of magnitude. Thus it appears that the free-energy barriers due to random anisotropy in this system are not too high, and under slow cooling the system is able to reach states that are close to the equilibrium state. It should be noted that a distribution of energy barrier heights is expected in the experimental system because of the random atomic substitutions. Although in the simulations a single D value is assumed, the anisotropy energy barriers encountered by coupled spins should also have a random height distribution because each block of spins will have a different combination of favorable and unfavorable axes. Thus it is unexpected that the relaxation on slow cooling is found to follow simple exponential time dependence in both the experiments and the simulations. In the case of rapid quenching the system evidently gets stuck in a local minimum of the rough free-energy landscape and relaxation is thereby inhibited.

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