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The occurrence of avalanches in a single crystal of Mn_{12}-acetate

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Recent studies of Mn_{12}-acetate have focused on the possible observation of superradiance triggered by avalanches of the magnetic relaxation [J. Tejada, E. M. Chudnovsky, J. M. Hernandez, and R. Amigo, Appl. Phys. Lett. **84**, 2373 (2004); M. Bal *et al.*, Phys. Rev. B, **70**, 140403 (2004)]. It is therefore of interest to determine the conditions for such avalanches. For several single crystals of Mn_{12}-acetate of different dimensions, the distribution of magnetic fields at which magnetic avalanches occur at different temperatures is reported for field sweep rates of 5 and 10 mT/s. No avalanches were observed above 0.8 K for these conditions. For temperatures below 0.8 K, avalanches were found between 2.0 and 4.2 T, with about half occurring below the field (3.2 T) above which appreciable relaxation proceeds by ground state tunneling. At the level of statistics accumulated in these experiments, there appears to be no clear preference for an avalanche to occur at or near a resonant magnetic field, and a surprising number occur at fields below the expected onset of appreciable ground state tunneling in the absence of avalanches. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1854419]

I. INTRODUCTION

Molecular nanomagnets, or single molecule magnets, exhibit interesting quantum mechanical phenomena at low temperatures, such as quantum tunneling of magnetization[1,2] and Berry phase oscillations,[3] and have possible applications as memory storage and qubits for quantum computers.[4] One of the several prototypical systems, Mn_{12}-acetate was synthesized by Lis in 1980.[5] It contains a regular array of magnetic clusters, each composed of 12 Mn atomic spins strongly coupled together to give a net spin $S=10$. Hysteretic behavior is observed in the M versus H curve below a blocking temperature of 3 K, due to slow magnetic relaxation in the presence of the sizable anisotropy barrier of ≈ 54 K. Prior to the discovery by Friedman *et al.*[1] of regularly spaced multiple steps in the hysteresis due to resonant spin tunneling, early experiments by Paulsen *et al.*[6] found sharp single-step reversal of the magnetization. In order to study the behavior of these materials under controlled conditions, a great deal of attention has been paid to avoiding uncontrolled magnetic avalanches which tend to occur at low temperatures in larger samples. As a result, avalanche effects have not been fully investigated yet. Stimulated by a suggestion of Chudnovsky and Garanin,[7] recent studies of Mn_{12}-acetate have focused on the possible observation of superradiance triggered by avalanches of the magnetic relaxation.[8–10] It is therefore of interest to determine the conditions for such avalanches.

Paulsen *et al.*[6] measured hysteresis curves for a cluster of several crystals of Mn_{12}-acetate using a magnetic field sweep rate of 0.3 mT/s at various temperatures and magnetic field directions. Avalanches always occurred at temperatures below 1.5 K. The cause of avalanche effects was inferred as “a kind of chain reaction or thermal runaway” caused by the energy which was released through quantum tunneling of magnetization.[6] No comparison was possible of the conditions for magnetic relaxation with and without avalanches since avalanches always occurred. Fominaya *et al.*[11] observed heat emissions caused by magnetic relaxation; del Barco *et al.*[12] numerically confirmed the enhancement of magnetic relaxation due to heating caused by magnetic relaxation for a zero-field-cooled oriented powder sample of Mn_{12}-acetate at temperatures above 1.9 K in a pulsed magnetic field with a sweep rate of 140 T/s. In an experimental search for superradiance during avalanches in a cluster of Mn_{12}-acetate crystals, Tejada *et al.*[8] have reported that the magnetization reversal occurs in a time on the order of 0.1 sec at temperatures above 1.7 K. More recent measurements made by Bal *et al.*[9] indicate faster switching times on the order of milliseconds. In both cases avalanches occurred above 1.7 K for a large cluster of crystals at magnetic fields corresponding to tunneling resonances, and they occurred at the same, reproducible, magnetic field for a given fixed temperature.

II. EXPERIMENTAL RESULTS

The magnetization of single crystals of Mn_{12}-acetate was measured using a 10×10 μm^2 Hall sensor composed of a

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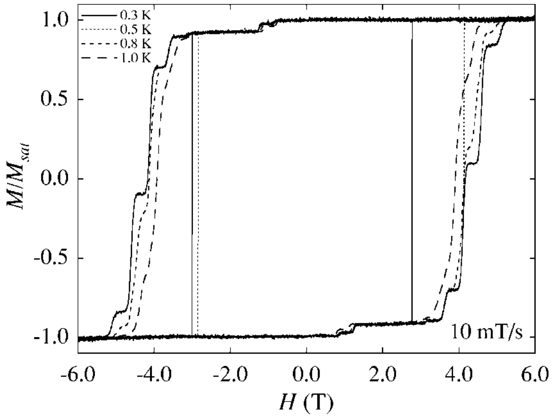

FIG. 1. Magnetization vs magnetic field applied along the easy axis of Mn_{12}-acetate at four different temperatures, as labeled. Data is shown for sample *A* using a magnetic field sweep rate of 10 mT/s. Avalanches, which occur at random magnetic fields, are shown at 0.3 and 0.5 K.

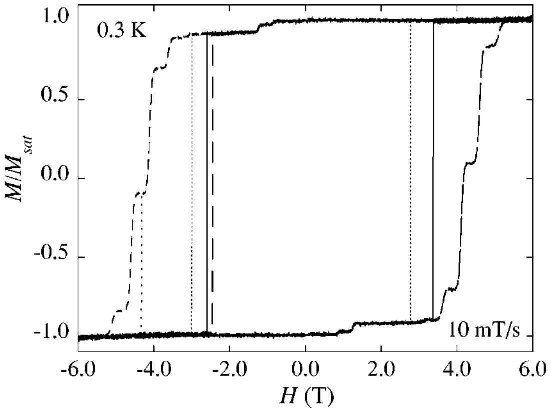

FIG. 2. Magnetization vs magnetic field applied along the easy axis of Mn_{12}-acetate sample at ≈0.3 K using a magnetic field sweep rate of 10 mT/s for sample *A*, showing several avalanches. The fields at which avalanches occurred were distributed as shown in Fig. 3.

two-dimensional electron gas in a GaAs/AlGaAs heterostructure using a small excitation current of 1 μA at temperatures down to 0.3 K. A longitudinal magnetic field, swept at a constant rate, was applied along the easy axis. Good thermal contact was provided by direct immersion in ^{3}He liquid. Data were obtained for three single crystal samples of Mn_{12}-acetate of dimensions $0.3\times0.3\times1.5$ mm^3 (sample *A*), $0.4\times0.4\times1.8$ mm^3 (sample *B*), and $0.15\times0.15\times0.2$ mm^3 (sample *C*). A ruthenium oxide thermometer mounted directly on the sample was used to monitor the temperature during magnetic field sweeps. Temperature increases of up to 200 mK were recorded during the resonances for controlled quantum tunneling of magnetization in the absence of avalanches. During avalanches, the temperature increased from 0.3 K to between 1.4 and more than 5 K.[13]

The magnetization is shown as a function of magnetic field in Fig. 1 at four different temperatures between 0.3 and 1.0 K for a magnetic field sweep rate of 10 mT/s. In agreement with earlier measurements, the hysteresis curves at 0.3 and 0.5 K coincide, indicating that the magnetic relaxation is completely dominated by spin tunneling from the ground state of the metastable potential well. Small changes are observed at the two higher temperatures, indicating that thermally assisted tunneling is playing a measurable role. Also shown in Fig. 1 are four avalanche events, at 0.3 and 0.5 K. Note that a second species of Mn_{12}-acetate that is known to be present in all samples has a lower potential barrier and a faster relaxation that is expected to bring it to saturation at 1.5 T for the magnetic sweep rates used in our experiments.[14]

Figure 2 shows five curves obtained at ≈0.3 K using a field sweep rate of 10 mT/s. Six magnetic avalanches are shown that occurred at six different magnetic fields. The probability of an avalanche was different for different samples. In the temperature range between 0.3 and 0.6 K, avalanches occurred roughly 50% of the time for sample *A* and 100% of the time for sample *B*. This difference may be associated with different sample history, sample quality, sample size, or some other factor.

For three single crystals of Mn_{12}-acetate Fig. 3 shows the distribution of magnetic fields at which magnetic avalanches occur at different temperatures above 0.3 K for field sweep rates of 5 and 10 mT/s. No avalanches were recorded at temperatures above 0.8 K. For temperatures below 0.8 K, magnetic avalanches were distributed randomly between 2.0 and 4.2 T. Approximately half the avalanches occur below the field (3.2 T) above which appreciable relaxation proceeds under controlled conditions. At the level of statistics accumulated in these experiments, there appears to be no clear preference for an avalanche to occur at or near a resonant magnetic field, and a surprising number occur at fields below the expected onset of appreciable tunneling in the absence of avalanches.

III. CONCLUSION AND DISCUSSION

To summarize, we have shown that for our samples avalanches were found between 2.0 and 4.2 T at temperatures below 0.8 K, with about half occurring below the field (3.2 T) at which appreciable relaxation proceeds by ground

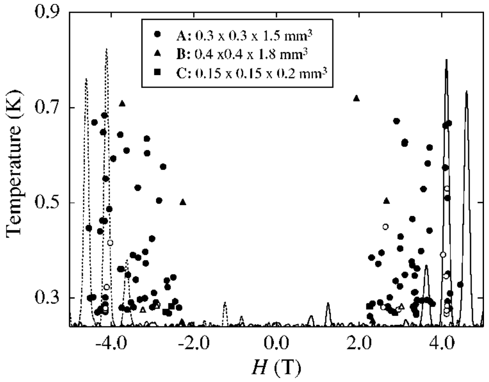

FIG. 3. Magnetic fields at which avalanches occurred at different temperatures above 0.3 K. The absence of data points above 0.8 K reflects the fact that no avalanches were observed above this temperature (Ref. 15). Note that points at positive and negative fields represent different data, and are not simply reflections about $H=0$. Closed and open symbols represent sweep rates of 10 and 5 mT/s, respectively. To indicate the resonant magnetic fields for ground state tunneling, the first derivative of the *M* vs *H* curves at 0.3 K without avalanches is shown in arbitrary units by the solid and dashed curves, respectively, for magnetic field sweeping upward and downward at 10 mT/s.

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state tunneling. Unexpectedly, there is no clear preference for an avalanche to occur at or near a resonant magnetic field, and a surprising number were found at fields below the expected onset of appreciable tunneling in the absence of avalanches.

We have observed avalanches in single crystals only at temperatures below 0.8 K which occur at randomly distributed magnetic fields. These differ from the avalanches that have been observed above 1.7 K in large clusters of crystals glued together,[9,10] which occur at a specific, reproducible magnetic field. We suggest that avalanches are triggered by global heating in the large clusters. In the case of smaller single crystals, local nucleation of magnetic relaxation with or without defects, or perhaps some other mechanism, may be involved. Further work is needed to clarify the origin and the occurrence of avalanches in molecular nanomagnets.

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[13] We note that the temperature also increased during field sweeps when no avalanches occurred. For example, for sample A at a sweep rate of 10 mT/s, the sample thermometer readings were 0.5, 0.65, and 0.86 K when the bath temperature was, respectively, 0.3, 0.6, and 0.84 K.
[14] See W. Wernsdorfer, cond-mat/0405501.
[15] More data were obtained in some temperature intervals than others, and the density of points does not therefore reflect the probability of obtaining an avalanche. Although preliminary results do indicate that the probability of avalanches decreases as the temperature is raised from 0.3 to 0.8 K, additional measurements are needed to establish this beyond doubt.

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