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# Effect of quantum tunneling on the ignition and propagation of magnetic avalanches in $\mathrm{Mn}_{12}$ acetate

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Using a wire heater to ignite magnetic avalanches in fixed magnetic field applied along the easy axis of single crystals of the molecular magnet $\mathrm{Mn}_{12}$ acetate, we report fast local measurements of the temperature and time-resolved measurements of the local magnetization as a function of magnetic field. In addition to confirming maxima in the velocity of propagation, we find that avalanches trigger at a threshold temperature which exhibits pronounced minima at resonant magnetic fields, demonstrating that thermally assisted quantum tunneling plays an important role in the ignition as well as the propagation of magnetic avalanches in molecular magnets.

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First synthesized in 1980,[1] $\mathrm{Mn}_{12}$ acetate ($[\mathrm{Mn}_{12}\mathrm{O}_{12}(\mathrm{CH}_3\mathrm{COO})_{16}(\mathrm{H}_2\mathrm{O})_4]\cdot 2\mathrm{CH}_3\mathrm{COOH}\cdot 4\mathrm{H}_2\mathrm{O}$, hereafter referred to as $\mathrm{Mn}_{12}$-ac) is a prototypical molecular magnet that is particularly interesting for its high spin and high bistable anisotropy.[2] The magnetic core of each $\mathrm{Mn}_{12}$-ac molecule is composed of 12 Mn atoms strongly coupled to form a rigid spin $S=10$ cluster at low temperatures; strong uniaxial magnetic anisotropy along the tetragonal symmetry axis provides an $\approx 60$ K barrier against spin reversal and robust bistability at temperatures below the blocking temperature of $\approx 3$ K. Composed of $\approx 10^{18}$ nominally identical magnetic molecules regularly arranged on a tetragonal lattice, $\mathrm{Mn}_{12}$-ac samples have served as a platform for the study of a wide variety of interesting magnetic phenomena. In particular, macroscopic quantum tunneling of magnetization was first observed in this material[3,4] below the blocking temperature whenever a magnetic field applied parallel to the anisotropy axis brought into alignment a pair of energy levels on opposite sides of the anisotropy barrier corresponding to different spin projections, as illustrated in Fig. 1(a) (see Refs. 5–8 for reviews).

Although abrupt reversals of the magnetization, referred to as magnetic avalanches, have been regularly observed in molecular magnets,[9] they received little attention until relatively recently. Avalanches were thought to entail a thermal runaway process in which the reversing spins release heat, causing the relaxation of the remaining spins in the crystal.[9] Indeed, both direct and indirect measurements of the heat released during an avalanche have confirmed their thermal nature.[10] Recent experiments of Suzuki *et al.*[11] have revealed that the magnetization reversal does not occur homogeneously throughout the sample but travel instead with constant velocity as a narrow interface between regions of opposing magnetization. In light of the thermal nature of the process and the relatively slow velocity of propagation of the interface ($\approx 10$ m/s), Suzuki *et al.*[11] have suggested this is "magnetic deflagration," in analogy with the very similar process of chemical combustion referred to as chemical deflagration.[12]

Based on measurements of the time evolution of the total magnetization of $\mathrm{Mn}_{12}$-ac crystals during avalanches triggered by surface acoustic waves at fixed magnetic fields, Hernandez-Minguez *et al.*[13,14] have reported maxima in the velocity of propagation of avalanches in $\mathrm{Mn}_{12}$-ac at "resonant" magnetic fields where the anisotropy barrier is effectively lowered by quantum tunneling of the spins. The velocity maxima were attributed to thermally assisted quantum deflagration.

In the present Brief Report, we report the results of experiments designed to elucidate the role of quantum mechanics (i.e., spin tunneling) in the ignition and propagation of magnetic avalanches in $\mathrm{Mn}_{12}$-ac. Using a wire heater to ignite magnetic avalanches in fixed magnetic field applied along the easy axis of single crystals of the molecular magnet $\mathrm{Mn}_{12}$ acetate, we report fast local measurements of the temperature and time-resolved measurements of the local magnetization as a function of magnetic field. We find that avalanches ignite at a reproducible threshold temperature, and this temperature exhibits pronounced minima at magnetic fields corresponding to thermally assisted tunneling across the anisotropy barrier. Additionally, we find maxima for the velocity of propagation of the avalanches, albeit in a higher range of magnetic field than those reported by Hernandez-Minguez *et al.*[13,14]

All measurements reported here were performed on single crystals of $\mathrm{Mn}_{12}$-ac with typical dimensions of $1.5\times 0.3\times 0.3\ \mathrm{mm}^3$ immersed in liquid $^3$He at approximately 300 mK. Germanium thin film resistance thermometers of $40\times 100\ \mu\mathrm{m}^2$ dimensions were deposited by e-gun evaporation on heated GaAs substrates in vacuum. The crystal was mounted using a thin layer of thermally conductive Apiezon M grease [see Fig. 1(b)]. In order to make good thermal contact with the heater, the entire assembly, including thermometer, sample, and heater, was encased in Apiezon M grease, as shown in Fig. 1. To minimize thermal gradients between the crystal and the thermometer, care was taken to place the heater as close as possible to the sample (roughly 1 mm above the crystal) and the minimum heater power was used that still triggered avalanches.

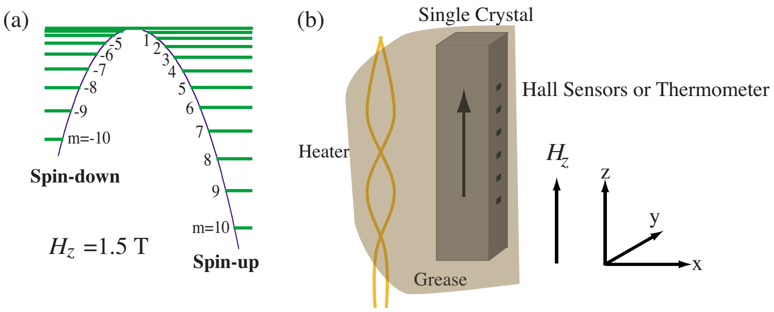

FIG. 1. (Color online)(a) Energy level diagram for a longitudinal magnetic field of 1.5 T. (b) Schematic diagram of a crystal mounted on (i) an array of the Hall sensors used to measure the magnetization or (ii) a germanium thermometer used to measure the temperature. The heater, crystal, and sensors are all encased in Apiezon M grease.

It is well known that all crystals of $\mathrm{Mn}_{12}$-ac contain a small amount of a second species of spin $S=10$ molecules that have a lower anisotropy barrier of roughly 45 K. This minor species is homogeneously distributed in the crystal at typical levels between 5% and 8%.[2,15] The presence of the minor species was found to have a significant influence on both the temperature for ignition and the propagation velocity of the avalanches. Consequently, we used the following protocol to "quench" the effect of the minor species' spin relaxation:[16] after fully magnetizing the crystal in the "up" direction, the field was swept to a value in the opposite (downward) direction that is large enough to flip the minor species downward but small enough that it leaves the major species intact ($\sim$2 T). Bringing the magnetic field back to zero then yields a crystal with the major (spin "up") and minor (spin "down") species fully magnetized in opposite directions. This allows the magnetic relaxation of minor and major species of $\mathrm{Mn}_{12}$-ac to be studied independently. We will report a detailed study of the interplay between the two species in another publication. For samples prepared as described above, we report the behavior of avalanches of the major species where the minor species plays no role, having already relaxed along the direction of the applied field.

Our studies of avalanches of the major species were carried out using the following experimental protocol. After preparing the sample as described in the preceding paragraph, the magnetic field was ramped at 10 mT/s to a preassigned value in a direction opposite to the polarization of the major species, with the sample immersed in liquid $^3$He at 300 mK. It is important to note that this temperature is well below the blocking temperature of $\approx$3 K so that below about 3 T, there was negligible reduction of the magnetization by relaxation via tunneling as the field was swept through the resonant fields; the sample thus remained fully magnetized. The wire heater was then turned on (and left on) at fixed magnetic field, and the temperature of the sample was monitored by measuring the resistance of the Ge thermometer using standard four-terminal techniques.

A typical curve showing the temperature as a function of time is shown in Fig. 2. A spike occurs at $t \approx 13$ ms when the heater is turned on. The heater remains on and the subsequent slow rise in temperature between $t \approx 13$ and $t \approx 30$ ms reflects the gradual heating of the entire sample in response to the power provided by the heater. The sharp rise in temperature at $t \approx 30$ ms signals the sudden release of heat associated with the ignition of an avalanche at a threshold temperature. The inset shows the data on an expanded time scale. Measurements were repeated several times at a given field and were reproducible within a given run. Similar data were taken at many different (fixed) magnetic fields.

Figure 3 show the threshold temperature required to ignite avalanches plotted as a function of the magnetic field for fixed fields between 0.4 and 2.0 T. Sharp dips in the ignition temperature occur at magnetic fields denoted by vertical lines. These magnetic fields correspond to thermally assisted spin tunneling across the anisotropy barrier in $\mathrm{Mn}_{12}$-ac,[3,17] effectively reducing the anisotropy barrier.

As mentioned earlier, the absolute value of the ignition temperature was reproducible within a given experimental run but varied by as much as 0.25 K from one run to another. This is undoubtedly due to uncontrolled thermal gradients that were different depending on the thermal connection between the thermometer and the sample. For example, the thickness of the layer of Apiezon M grease was perforce different for different runs. It is important to note that strong minima were observed in *all* runs at the same magnetic fields. This behavior is robust and reproducible.

Garanin and Chudnovsky have recently provided a de-

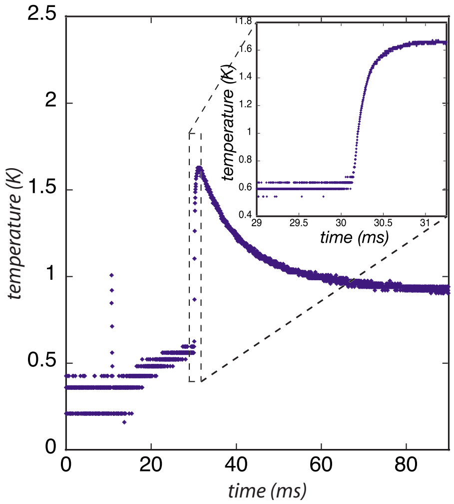

FIG. 2. (Color online) Temperature recorded by the thermometer in contact with the crystal for an avalanche triggered at 1.85 T. The narrow peak at 13 ms is the electrical noise when the heater is turned on. The abrupt rise at 30 ms is due to heat released by the avalanche. The inset shows data taken near the ignition point with higher resolution for the same avalanche. The noise at low temperatures derives from two factors: a nonlinearity in the thermometer that limits the resolution for temperatures below 0.4 K and the noise associated with digitizing (continuous) data acquired by the oscilloscopes.

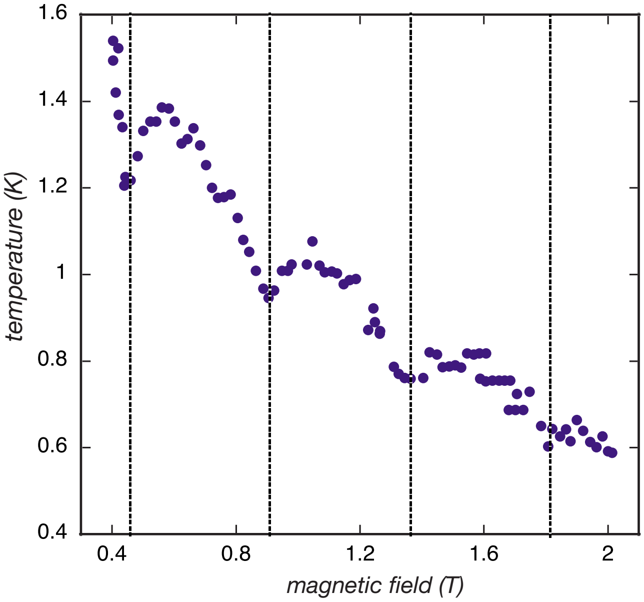

FIG. 3. (Color online) Temperature required to ignite avalanches plotted as a function of magnetic field. All data were taken for field-cooled samples. The vertical lines denote the magnetic fields where sharp minima occur in the ignition temperature corresponding to thermally assisted tunneling near the top of the anisotropy barrier (Ref. 17). The overall decrease in ignition temperature is due to the reduction of the anisotropy barrier as the field is increased.

tailed theoretical foundation for the newly discovered process of magnetic deflagration by extending many of the results from the classical theory of combustion to the process of spin reversal in molecular magnets.[18] Consistent with the data shown in Fig. 3, their theory predicts a significant drop in the threshold temperature required to trigger avalanches at the resonant values of magnetic field where the barrier against spin reversal is effectively reduced due to resonant quantum spin tunneling.

We now present measurements of the magnetization obtained in separate experimental runs for similar $\mathrm{Mn}_{12}$-ac crystals. Time-resolved measurements of the local magnetization were obtained from measurements of the transverse component of the magnetic field $B_x$ during an avalanche using six $30 \times 30\ \mu\mathrm{m}^2$ two-dimensional electron gas GaAs Hall sensors placed along the crystal to probe a significant fraction of its length. Figure 4 shows the time of arrival at each sensor of the narrow interface between regions of the sample with antiparallel magnetizations. The velocity of propagation of the avalanche is then deduced from the known spacing between the sensors, as shown in the inset (see Refs. 11 and 19 for experimental details).

The velocity of avalanches is shown in Fig. 5, where the filled circles denote results for avalanches that entail the reversal of the full magnetization from one direction to the other along the $c$ axis, and the open circles are for zero-field-cooled samples where the magnetization changes by half the amount, from zero to full magnetization. The ignition temperatures shown in Fig. 3 are also plotted as triangles for comparison. The vertical lines drawn in Fig. 5 denote the magnetic fields at which minima occur in the ignition temperature and maxima occur for the velocity. It is interesting to note that the minimum ignition temperature occurs at higher field than the corresponding velocity maximum (see

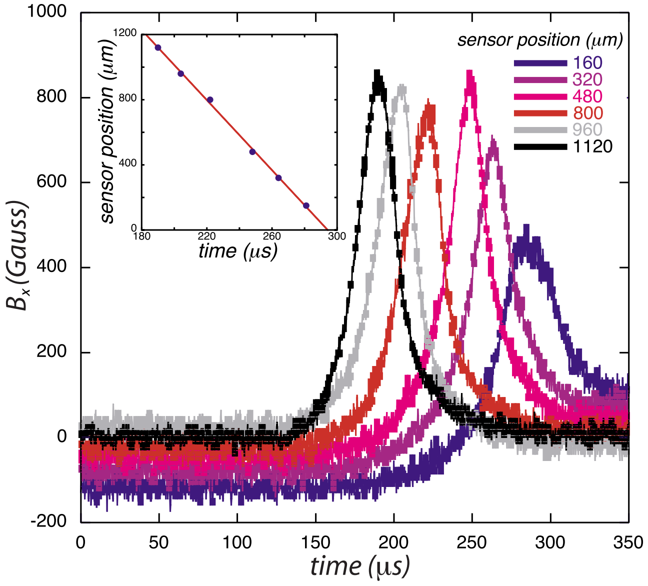

FIG. 4. (Color online) Signals recorded by six Hall sensors in contact with the crystal for an avalanche triggered at 2 T. The inset shows sensor position versus the time at which the sensor recorded peak amplitude. A straight line fit yields a velocity of 10.6 m/s.

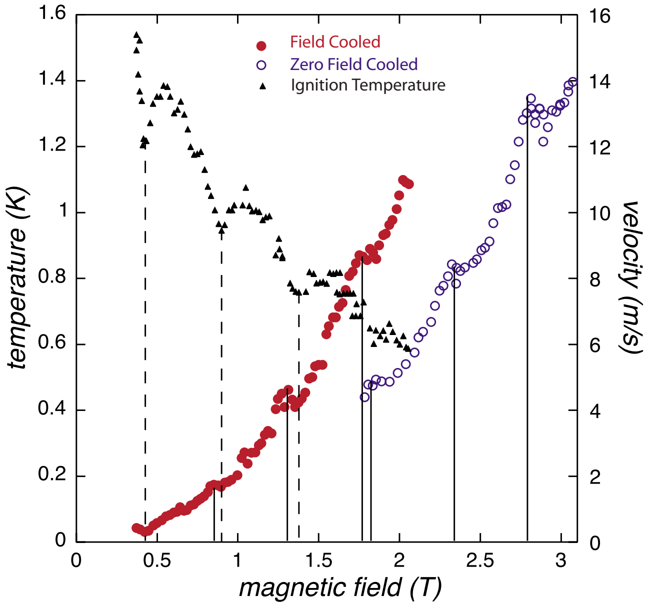

FIG. 5. (Color online) Velocity of propagation of avalanches (right-hand $y$ axis) for field-cooled (filled circles) and zero-field-cooled (open circles) samples versus magnetic field at which the avalanche was triggered. The triangles show the ignition temperature for field-cooled samples (left-hand $y$ axis). The solid vertical lines drawn from the bottom denote velocity maxima; the dashed vertical lines drawn from the top denote ignition temperature minima. The overall increase of the velocity with increasing magnetic field is due to the decrease of the anisotropy barrier.

data at $H \approx 0.9$ T and $H \approx 1.35$ T). In a similar manner, the velocity maximum for the zero-field-cooled sample at $H \approx 1.8$ T is at a slightly higher magnetic field than that for the field-cooled case. In both cases, the higher resonant field indicates that the tunneling takes place for energy level crossings that are deeper in the potential well.[17] A detailed study of this effect will be published elsewhere.

The velocity of propagation increases as the field is raised and the barrier to spin reversal is reduced. From measurements at ≈2.1 K, Hernandez-Minguez *et al.*[13,14] have reported maxima at 0.9 T and 1.35 T, in agreement with predictions of the theory of Garanin and Chudnovsky.[18] Although maxima are barely discernible at low magnetic fields at the lower (initial) temperatures of our experiments, they become evident at higher magnetic fields. Further study is required to determine the conditions (e.g., temperature, size, and direction of magnetic field) for observing the effect of quantum tunneling on the velocity of propagation of avalanches.

To summarize, using fast local measurements of the temperature and time-resolved measurements of the local magnetization, we have shown that quantum tunneling of the magnetization plays a significant role in determining the threshold temperature for the ignition as well as the propagation of avalanches, as predicted by Garanin and Chudnovsky.[18]

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