Quantum Magnetic Deflagration in Mn_{12} Acetate

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We report controlled ignition of magnetization reversal avalanches by surface acoustic waves in a single crystal of Mn_{12} acetate. Our data show that the speed of the avalanche exhibits maxima on the magnetic field at the tunneling resonances of Mn_{12}. Combined with the evidence of magnetic deflagration in Mn_{12} acetate [Y. Suzuki *et al.*, Phys. Rev. Lett. **95**, 147201 (2005)], this suggests a novel physical phenomenon: deflagration assisted by quantum tunneling.

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Magnetic properties of Mn_{12} acetate have been intensively studied after the magnetic bi-stability of this molecular cluster below 3.5 K was demonstrated [1]. The bistability is caused by a large spin of the cluster, $S = 10$, and by strong uniaxial magnetic anisotropy that provides a 65 K energy barrier between spin-up and spin-down states. At low temperature a magnetized Mn_{12} crystal exhibits two modes of magnetic relaxation. The first mode is a slow one. It manifests itself in a staircase hysteresis curve which is due to thermally assisted quantum tunneling of the magnetization [2]. The second relaxation mode, exhibited by sufficiently large crystals, is a much more rapid magnetization reversal that typically lasts less than 1 ms. It was initially studied by Paulsen and Park [3] and attributed to a thermal runaway or avalanche [see also Ref. [4]]. In the avalanche, the initial relaxation of the magnetization towards the direction of the field results in the release of heat that further accelerates the magnetic relaxation. Recent local magnetic measurements of Mn_{12} crystals [5] have demonstrated that during an avalanche the magnetization reversal occurs inside a narrow interface that propagates through a crystal at a constant speed of a few meters per second. It has been argued that this process is analogous to the propagation of a flame front (deflagration) through a flammable chemical substance. The conventional theory of deflagration, in the first approximation, yields the following expression for the velocity of the flame front [5–7]:

$$v = \sqrt{\frac{\kappa}{\tau_0}} \exp\left(-\frac{U}{2k_B T_f}\right). \quad (1)$$

Here U, τ_0, and T_f are the energy barrier, the attempt frequency, and the temperature of the "flame" in the expression $\tau = \tau_0 \exp(U/k_B T_f)$ for the "chemical reaction" time, and κ is thermal diffusivity. In the case of Mn_{12}, $\kappa \sim 10^{-5}$ m^2/s, $\tau_0 \sim 10^{-7}$ s, and the field dependence of the energy barrier, $U(H)$, is well known.

In a flammable chemical substance the potential barrier is a constant determined by the nature of the chemical reaction that transforms a metastable chemical into a stable chemical (e.g., a mixture of hydrogen and oxygen transforms into water). On the contrary, in molecular magnets the energy barrier, as well as the released energy, can be controlled by the magnetic field. At certain values of the magnetic field the spin levels on the two sides of the energy barrier come to resonance and thermally assisted quantum spin tunneling under the barrier takes place; see Fig. 1. Therefore, the effect of the tunneling is roughly equivalent to cutting out the top of the barrier. This well-understood effect is responsible for the staircase hysteresis curve in Mn_{12} and other molecular magnets. Because of this effect, one also should expect that the velocity of the avalanche, given by Eq. (1), increases at the resonant values of the magnetic field.

In the experiment of Suzuki *et al.* [5] avalanches were ignited in a stochastic way on sweeping magnetic field

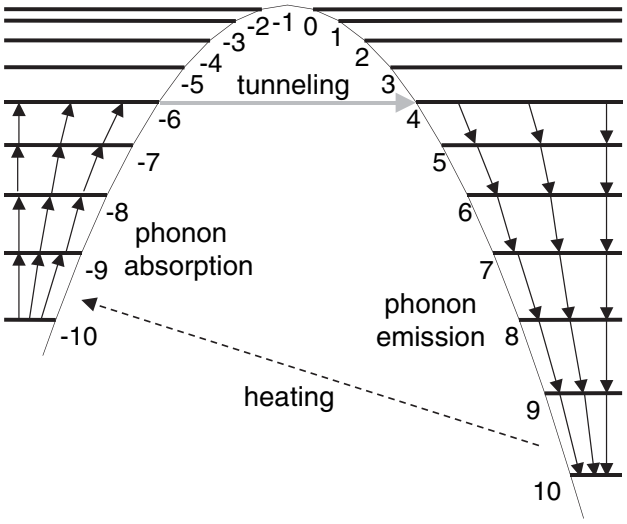

FIG. 1. Spin levels and magnetic avalanche in Mn_{12} acetate. At resonant fields, quantum tunneling effectively cuts out the top of the barrier, increasing the rate of relaxation between the left and right wells. Spin in the right well relax down the well releasing phonons that produce heat, which is absorbed by spins in the left well producing the avalanche.

between -5 T and 5 T. In such an experiment one cannot control the ignition process. Consequently, the probability that the avalanche occurs at a tunneling resonance is low, which may explain why no oscillation of v on H due to resonant spin tunneling has been observed. In this Letter we report a novel method of controlled ignition of avalanches in Mn_{12} acetate at constant magnetic field by means of surface acoustic waves (SAW). We demonstrate that the velocity of the deflagration front and the ignition rate oscillate on the magnetic field in accordance with the expectation that quantum spin tunneling lowers the barrier for the deflagration. Thus, in effect, Mn_{12} acetate exhibits a phenomenon which has not been seen in any other substance: slow burning (deflagration) assisted by quantum tunneling.

The acoustomagnetic experiments were performed by using hybrid piezoelectric interdigital transducers (IDT) deposited on the 128 YX cut of a $LiNbO_3$ substrate of dimensions $4 \times 12 \times 1$ mm^3, Fig. 2. The transducers [8] generate multiple harmonics of the fundamental frequency 111 MHz up to a maximum frequency of approximately 1.5 GHz. A single crystal of Mn_{12} acetate of dimensions $3 \times 1 \times 1$ mm^3 was glued directly onto the IDT, using commercial silicon grease.

Experiments have been carried out inside a commercial rf-SQUID Quantum Design magnetometer. The microwaves for the SAW generation were transported to the transducers with the help of coaxial cables which introduce attenuation smaller than 10 dB. The measurement of the reflection coefficient S_{11} was performed by using an Agilent network analyzer. We measured simultaneously the frequency dependence of S_{11} and the magnetization of the Mn_{12} crystals. Both the magnetization and S_{11} showed minima at the resonant frequencies of the IDT. The intensity of these minima depends on the energy of the SAW absorbed by the Mn_{12} single crystal. During the experiments, the temperature of the IDT attached to the sample and the temperature of the helium gas that provided heat exchange were independently monitored.

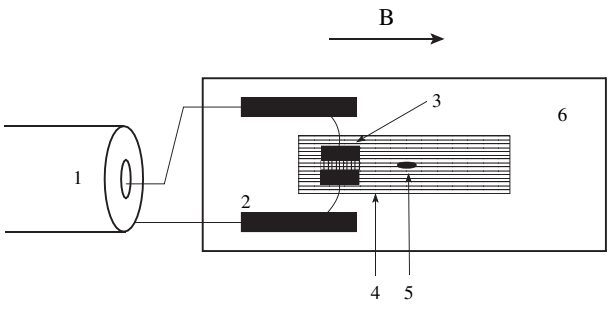

FIG. 2. Experimental setup. The microwave signal is taken by a coaxial cable (1) through a pair of conducting stripes (2) to the transducers (3), which generate the surface acoustic waves on the $LiNbO_3$ substrate (4). The Mn_{12} crystal (5) is glued directly on the substrate, with the easy anisotropy axis parallel to the magnetic field direction. The whole assembly is mounted on a plastic stand (6).

The magnetization dynamics was generated by exciting the IDTs with microwave pulses from a commercial Agilent signal generator. This generator permits selection of the shape, duration, and energy of the pulse in the frequency range 250 kHz–4 GHz. Most of the experiments were performed using rectangular microwave pulses of duration 10 μs–10 ms. Fast magnetization measurements (time resolution of 1 μs) were carried out at a constant temperature and constant magnetic field by continuously reading the voltage variation detected by the rf SQUID.

The magnetic properties of the Mn_{12} single crystals glued onto the surface of the transducer were found to be similar to those previously published [2,9–13]. Above the magnetic blocking temperature, T_B, the material exhibits superparamagnetic behavior with the magnetization following a $1/T$ law and a Brillouin dependence as a function of the ratio H/T. Below T_B, the magnetization as a function of the magnetic field shows resonant spin tunneling transitions at magnetic field values multiple of 0.45 T. In order to estimate the energy deposited by the SAW into the Mn_{12} crystal, we used microwave pulses of different duration in the superparamagnetic regime, $T > T_B$, where $M(H)$ is a known function determined by the thermal equilibrium among the spin states. The temperature variation of the Mn_{12} crystal, induced by the SAW for pulses of 1 ms duration, was 0.3 K.

To study magnetic avalanches, we first saturated the sample at -2 T and 2.1 K. The field was then varied at a constant rate of 300 Oe/s until a desired value of H was reached. Maintaining this field value, we delivered to the sample the SAW pulses of 1 ms to trigger the avalanche. These measurements have established a new method of igniting magnetization avalanches in molecular magnets. The advantage of using SAW is a total control over the field at which the avalanche takes place.

Figure 3 shows the time dependence of the magnetization change during avalanches triggered at different magnetic fields. The avalanche time, Δt, is defined as the time needed by the magnetization to change between the 2% and the 98% of the total variation. The speed, v, of the developed deflagration front can be obtained from the data as $v = l/\Delta t$, where l is the length of the sample. The dependence of v on H, extracted from the data, is shown in Fig. 4. The dotted line represents the fit of the data by Eq. (1) with the effective energy barrier cut by quantum tunneling near resonance fields. The field dependence of the effective barrier was taken from earlier experiments on magnetic relaxation [14,15]. The temperature of the flame, T_f, was obtained using measured specific heat of Mn_{12} acetate [16,17]. T_f is related to the specific heat, $C(T)$, through the relation

$$H\Delta M = \int_{T_i}^{T_f} dT C(T), \tag{2}$$

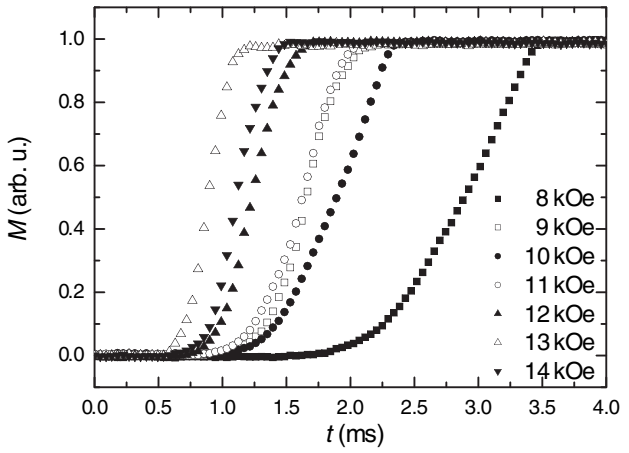

FIG. 3. Time dependence of the magnetization change during the avalanche at different magnetic fields for pulses of 1 ms duration starting at $t = 0$.

where H is the field at which the avalanche occurs, ΔM is the total change of magnetization, and T_i is the initial temperature. T_f was found to be in the range between 6 K and 9 K, depending on the magnetic field. The only fitting parameter was $\sqrt{\kappa\tau_0}$ for which the value of 2×10^{-6} m was obtained. This agrees with $\kappa \sim 10^{-5}$ m^2/s and $\tau_0 \sim 10^{-7}$ s, known from independent measurements.

Vertical lines in Fig. 4 represent positions of the tunneling resonances for Mn_{12} acetate. It is clear from the figure that the maxima in $v(H)$ correspond to the tunneling resonances. This observation is a clear evidence of the quantum aspect of the deflagration. To be precise, the magnetic deflagration corresponding to the avalanche is, of course, a thermal phenomenon, driven by thermal conductivity. However, the speed of the deflagration is also determined by the speed of ''chemical reaction'', which, in our case, is the rate of the transition between the two wells shown in Fig. 1. This rate is determined by both thermal activation to the excited spin levels and quantum tunneling between the levels. It is actually the thermally assisted quantum tunneling that accelerates deflagration at tunneling resonances. To our knowledge, slow burning (deflagration) assisted by quantum tunneling has never been observed in any chemical substance. In a crystal of molecular nanomagnet it becomes observable due to the possibility of controlling the speed of the deflagration by the magnetic field.

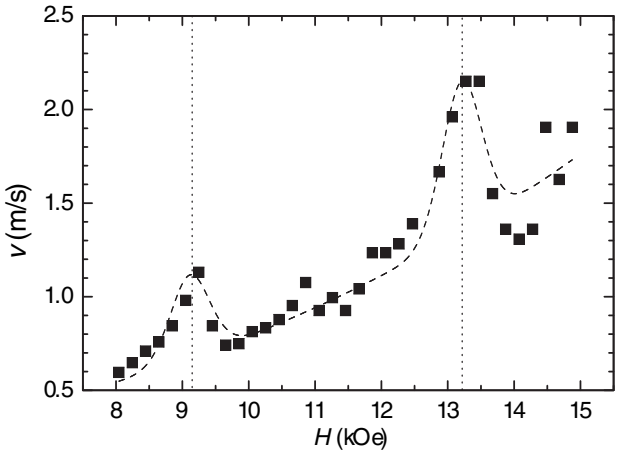

FIG. 4. Magnetic deflagration speed as a function of the magnetic field. The dashed line represents the fit by Eq. (1). Vertical lines show the positions of the tunneling resonances.

In conclusion, a new method of igniting magnetization reversal in molecular magnets by surface acoustic waves has been developed. We have observed a fundamentally new phenomenon: magnetic deflagration assisted by quantum tunneling. We have demonstrated that quantum tunneling can be seen not only in slow relaxation of molecular magnets (through staircase hysteresis loop) but also in the fast relaxation, that is, avalanche, which is a process equivalent to slow burning. This observation opens a new way for the study of quantum phenomena in molecular magnets, as well as for the experimental study of the complex deflagration physics.

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