Propagation of Magnetic Avalanches in $Mn_{12}Ac$ at High Field Sweep Rates

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Time-resolved measurements of the magnetization reversal in single crystals of $Mn_{12}Ac$ in pulsed magnetic fields, at magnetic field sweep rates from 1.5 kT/s up to 7 kT/s, suggest a new process that cannot be scaled onto a deflagrationlike propagation driven by heat diffusion. The sweep rate dependence of the propagation velocity, increasing from a few 100 m/s up to the speed of sound in $Mn_{12}Ac$, indicates the existence of two new regimes at the highest sweep rates, with a transition around 4 kT/s that can be understood as a magnetic deflagration-to-detonation transition.

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Manganese-12 Acetate ($Mn_{12}Ac$) is a magnetically bistable molecule showing a combination of high spin properties ($S = 10$) and strong magnetic anisotropy [1], resulting in a prototype model system for studies of quantum effects in the magnetization reversal such as resonant spin tunneling [2–5], and quantum selection rules for the absorption of electromagnetic radiation and the lifetime of the spin states [6–9]. Spin states in $Mn_{12}Ac$ were expected to be blocked at temperatures below 3.5 K, with an expected relaxation time of already more than 1 h at 2.5 K. However a much faster relaxation, at a rate increasing resonantly for a discrete number of applied magnetic fields along the magnetic anisotropy axis, was observed experimentally and was attributed to quantum tunneling between degenerate spin states. This low temperature behavior is now well understood and can be observed macroscopically as steps in the hysteresis curves of the magnetization, known as thermally assisted quantum tunneling of the magnetization (QTM) [2–10].

In experiments using large crystals or higher magnetic field sweep rates, an even faster relaxation mode of the magnetization, attributed to the occurrence of spin-phonon avalanches, was observed and it was shown that the reversal occurred inhomogeneously throughout the crystals [11,12]. These magnetic avalanches are now known to propagate throughout the crystal as a front moving at a constant speed, similar to deflagration in a flammable chemical substance [13]. More detailed studies of the process of magnetic deflagration, involving a novel method of controlled ignition of avalanches by the use of surface acoustic waves, show an enhancement of the propagation speed at the same discrete number of applied magnetic fields where resonant spin tunneling is seen and a detailed theory of this process has been proposed [14,15].

Current understanding of deflagration assumes a propagation driven by heat diffusion, even though the possibility of a superradiant coupling between clusters of $Mn_{12}Ac$ in a single crystal was shown theoretically [16]. Attempts to detect coherent electromagnetic radiation produced during such superradiant transitions have led to promising results [17]. Furthermore, previous studies of avalanches in $Mn_{12}Ac$, performed in pulsed magnetic fields, indicate a close relation to a collective electromagnetic spin relaxation [18].

In this Letter we present studies of the propagation of magnetic avalanches in $Mn_{12}Ac$ as observed in pulsed magnetic fields. An unexpected fast propagation is observed at the highest magnetic field sweep rates up to 7 kT/s, thus showing the existence of a new regime for the magnetization reversal in $Mn_{12}Ac$ that possibly involves an enhanced radiative coupling [19].

The experiments were performed at the Pulsed Fields facility of the Catholic University of Leuven. Our setup allows the application of magnetic field pulses, with maxima up to 70 T and a typical pulse duration of about 20 ms, by discharging a capacitor bank through a specially designed magnet coil [20].

During such a field pulse the magnetization reversal was detected using a set of compensated measurement coils. These sensors are coils with their inner windings wound in the opposite direction as their outer windings as shown in Fig. 1(a). If the number of inner and outer windings of such coil is tuned correctly, the sensor can be shown to be insensitive to fields that are homogenous over the area of the sensor. Because of the small size of these sensors compared to the magnet coil, the applied field pulses are effectively homogenous over the area of the sensors. The result is that our measurements are sensitive only to the changing magnetization of the sample and insensitive to the applied field pulses. Tuning of the number of windings is done outside of the cryostat, before mounting the sensors. If the number of windings is not perfectly tuned, the induced voltage will see both the changing magnetization and a small background component proportional to the

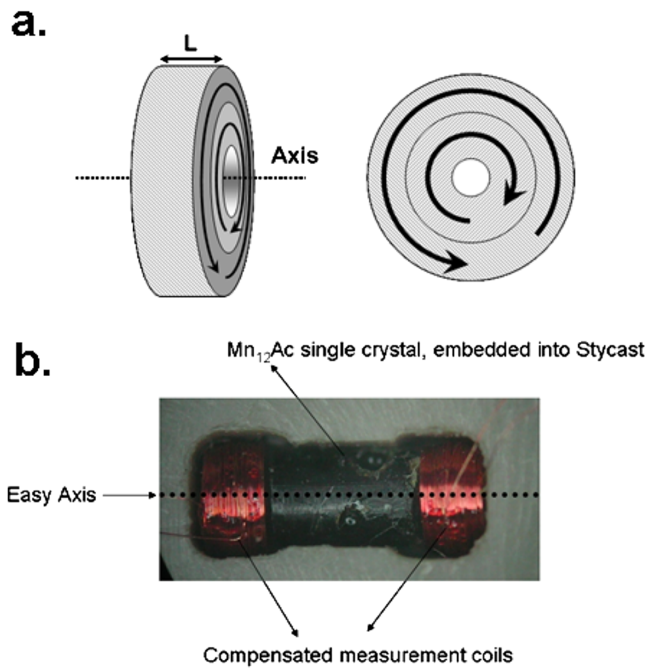

FIG. 1 (color online). (a) Typical sensor used to perform magnetization measurements in pulsed fields. Typical dimensions are $L \sim 1$ mm, an inner diameter of less than 1 mm and an outer diameter of less than 4 mm. (b) Cylindrical Mn_{12}Ac single-crystal sample, embedded into Stycast, and mounted between two compensated measurement coils. The easy axis of the crystal and the axis of each measurement coil are aligned.

applied field pulse. A set of amplifiers and a coil measuring only the applied field pulse are typically present to subtract this background component during our experiments. If the sample size is small enough to fit entirely inside the sensor, the voltage induced in such a sensor can be shown to be proportional to $\frac{dM}{dt}(t)$. For larger samples, as are typically used in our experiments, the induced voltage in such sensors will also depend on the distance from the propagation front to the sensor and scales approximately with the inverse cube of this distance.

The first stage of the applied field pulses (from 0 to a few Tesla) is nearly linear with time. As will be clear from our results presented below, the observed magnetization reversals of a Mn_{12}Ac single crystal occurs well within this time frame where the field variation is still linear, therefore allowing us to study propagation effects as a function of very high effective field sweep rates from 1.6 kT/s up to 7 kT/s. During our experiments the mm-sized single crystal sample was always immersed in liquid ^{3}He, to maintain a constant temperature of about 600 mK.

To study propagation effects, the reversal of the magnetization was detected on opposite ends of the easy axis of a cylindrical Mn_{12}Ac single crystal using two sensors as shown in Fig. 1(b). It must be noted that at any time t the induced voltage in our sensors will be approximately proportional to $\frac{dM}{dt}(t, d)$, with d the distance between the sensor and the position where the magnetization reversal occurs. For a propagating avalanche front the shape of the measured response will therefore depend on both the local magnetization reversal time (relaxation time) and the velocity of propagation. It can be shown, however, that a maximal response is consistently obtained when the reversal occurs closest to the center of such a sensor, therefore allowing the measurement of an effective time delay in between the reversal of the magnetization on opposite ends of the easy axis of the crystal.

As shown in Fig. 2, a time shift between the reversal of the magnetization on opposite ends of the easy axis of our Mn_{12}Ac crystal is observed clearly at the lowest effective field sweep rate of about 1.6 kT/s, but seems to disappear gradually towards the highest magnetic field sweep rates we used. For an effective field sweep rate of about 7 kT/s the magnetization reversal becomes practically indistinguishable from an instantaneous reversal throughout the crystal. This observation is made more quantitative in Fig. 3 by plotting the sweep rate dependence of the time shift, as obtained by looking for a maximum in the correlation of the two peaks in the dynamic susceptibility registered by the two sensors. Again this time shift decreases gradually as a function of rising magnetic field sweep rate, but drops to nearly zero at the highest field sweep rates. As explained earlier, the shape of the peeks measured by our sensors, as shown in Fig. 2, will depend both on the distance between the sensors and the propagation front, and therefore the velocity of propagation, and the typical microscopic relaxation time of the magnetization. Fitting the observed peeks using the measured propagation velocities, results in a microscopic relaxation time in the order of

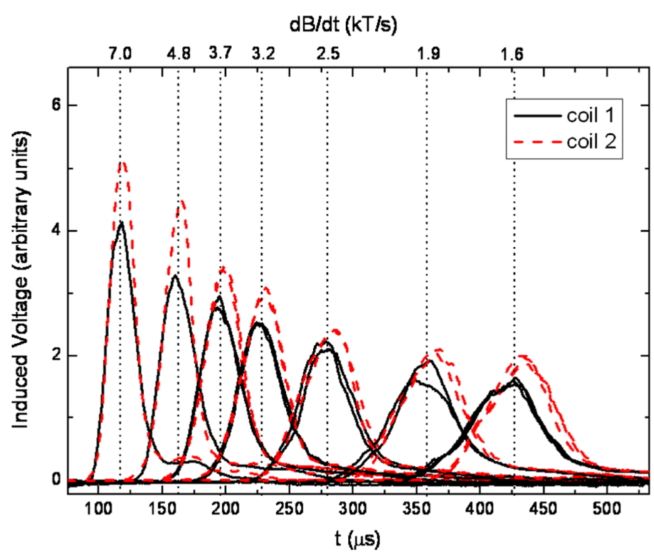

FIG. 2 (color online). Peaks in the dynamic susceptibility that correspond to a fast reversal of the magnetization as measured on opposite ends of the easy axis of a single crystal of Mn_{12}Ac at high magnetic field sweep rates.

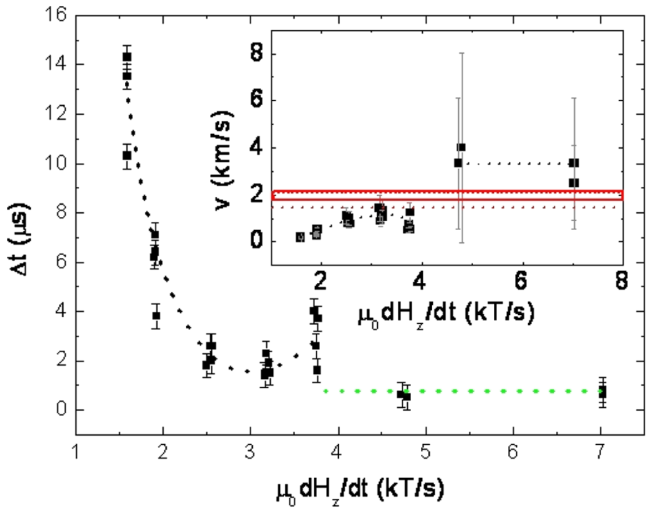

FIG. 3 (color online). Time shifts between the reversal of the magnetization on opposite ends of a single crystal of $Mn_{12}Ac$ at different high magnetic field sweep rates. The inset shows the resulting speeds, if deflagrationlike propagation of the magnetization reversal is assumed. The horizontal red solid lines and marked area correspond to a theoretical value and a range of experimental values, respectively, for the speed of sound in $Mn_{12}Ac$, as taken from [23]. Data points on the black dotted line correspond to an observable time delay, whereas those on the green dotted line correspond to a time delay indistinguishable from zero.

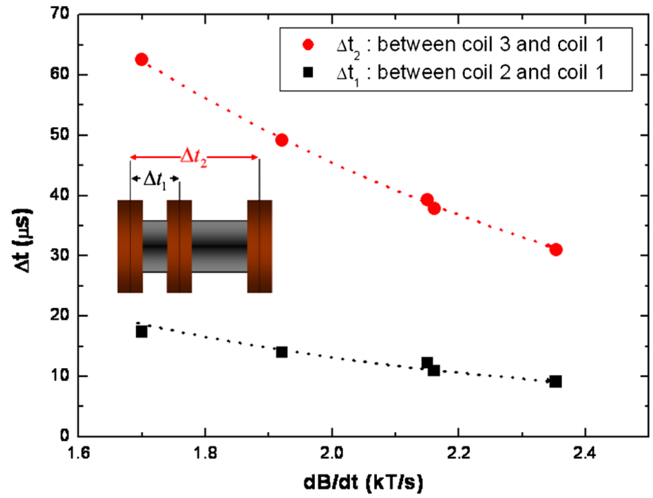

FIG. 4 (color online). Time shifts between the reversal of the magnetization of another single crystal of $Mn_{12}Ac$, now using a set of 3 sensors mounted along the easy axis of the crystal.

10 μs. The magnetization reversal is always seen to occur around the second resonant field at 0.9 T.

The most important point to emphasize about these results is that the observed time difference is too small to correspond to a deflagrationlike propagation of an avalanche front from one side of the crystal to the other as observed in [12,13]. Such deflagrationlike propagation is expected to occur at a constant speed of a few meters per second and thus should correspond to time differences of the order of 1 ms, whereas the largest time difference observed in our experiments is about 14 μs.

To provide evidence that propagation effects are involved in the process and to show that the observed effects are not an artifact of the particular $Mn_{12}Ac$ single crystal we used, an extra set of measurements were done, using a new $Mn_{12}Ac$ single crystal and using a set of three measurement coils, rather than just two, with the first and third sensor mounted on opposite sides of the easy axis of a cylindrical single crystal and the second sensor mounted in between, closest to sensor 1. The observed time differences are shown in Fig. 4. Though slightly larger time differences than for the previous sample are observed, the delays are still much smaller than expected for a deflagrationlike process driven by heat diffusion. The differences in time delays for both crystals can be ascribed to the different dimensions of both crystals as the total volume of the crystal is expected to play an important role in the energy-transfer mechanism [21]. More important is to note the fact that the reversal is consistently observed to occur first in the first sensor, then in the second and finally in the third, providing extra evidence that propagation effects are indeed involved.

Such propagation from one side of the easy axis to the other is furthermore consistent with what was observed in Refs. [13,14,22]. We should also note that the total size of the single crystal and sensors is small compared to the magnet coil used to apply field pulses, therefore resulting in an external field that is very homogenous in the area of the crystal and sensors. Care was also taken to ensure alignment of the crystal's easy axis with the axis of the magnet coils, and it is therefore unlikely that the initial ignition point of the propagation, on one side of the crystal, is determined in any way by the geometry of our setup, but it should rather be seen as an intrinsic property of the crystal.

If then such a propagation of one side to the other on the easy axis is assumed, the small time differences, as seen in our experiments, cannot correspond to an observation of magnetic deflagration as previously proposed, but should instead be seen as part of a new regime for the low temperature magnetic behavior of $Mn_{12}Ac$. More specifically, a transition from magnetic deflagration into this new regime at high sweep rates might be observed, as the time difference drops gradually towards high sweep rates for sweep rates up to about 4 kT/s, but drops very suddenly to nearly zero at sweep rates above 4 kT/s.

The main difference with magnetic deflagration is thus the unexpected high velocity of propagation. As the sample is about 2 mm long, the observed time differences would correspond to velocities ranging from 150 m/s up to 3500 m/s, whereas the maximum velocity expected for magnetic deflagration is only about 30 m/s. These high propagation speeds can therefore not be understood if heat diffusion is the only energy-transfer mechanism driving the propagation, as suggested recently [15]. Instead we would propose that an enhanced radiative coupling between the $Mn_{12}Ac$ clusters is required. Furthermore we would like to note the remarkable fact that the speed of sound in $Mn_{12}Ac$, as taken from [23], is somewhere around 2000 m/s and this value almost exactly separates our data for sweep rates below the observed transition at 4 kT/s from our data for sweep rates above 4 kT/s. The high sweep rate regime would then correspond to a regime where the front propagates at speeds higher then the speed of sound and therefore our data might correspond to the first observation of a magnetic deflagration-to-detonation transition.

In conclusion, we would like to state that a new regime for the low temperature magnetic behavior of $Mn_{12}Ac$ was observed at high magnetic field sweep rates up to 7 kT/s, with a transition into this regime at around 4 kT/s. As an increase in the propagation velocity towards higher sweep rates is found, starting from a few 100 m/s up to the speed of sound in $Mn_{12}Ac$, the observed transition could be related to a magnetic deflagration-to-detonation transition. These high propagation velocities require a faster energy-transfer mechanism than solely heat diffusion, most likely related to an enhanced radiative coupling between the molecular clusters. This new regime, observed in our measurements, is therefore a suitable candidate for further research of radiative and possibly superradiant phenomena in molecular magnets.

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