

Acoustomagnetic pulse experiments in $\text{LiNbO}_3/\text{Mn}_{12}$ hybrids

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We report here on the influence of surface acoustic waves (SAWs) on the magnetization of a Mn_{12} -acetate single crystal. The crystal was mounted on the surface of a piezoelectric LiNbO_3 substrate containing an interdigital transducer for the excitation of SAWs. The magnetization of the crystal was measured using a rf superconducting quantum interference device with a time resolution of 1 μs . The piezoelectric material was excited by SAW pulses of different frequencies produced by applying microwave pulses to the transducer. Our data show that molecular magnets onto the LiNbO_3 surface can be used as very sensitive detectors of the SAW frequency and intensity. © 2006 American Institute of Physics. [DOI: 10.1063/1.2158705]

There has been a growing interest in recent years in using high-frequency surface acoustic waves (SAWs) to control the optical, electronic, and magnetic properties of advanced materials. These waves have been used to produce efficient light modulators and switches, as well as to transport single carriers,¹ excitons,² and spins.³ Except for the studies of SAW generation through magnetostriction,⁴ the interaction between high-frequency SAWs and ferromagnetic materials has received little attention. The SAW fields, however, offer unique possibilities to modulate the magnetic properties, in particular the ones of magnetic molecules. Single molecule magnets provide a very promising avenue for the study of quantum phenomena^{5,6} on the nanometric scale, and indeed a remarkable number of very important experimental findings has been reported in the last years (for reviews, see Refs. 7–9). In this letter we show results of the combination of piezoelectric materials and single molecule magnets with the aim of establishing a correlation between acoustic phenomena and quantum spin dynamics.

Hybrid piezoelectric interdigital transducers (IDTs) deposited on the 128 YX cut of LiNbO_3 substrates were used in the experiments. We employed the special transducer design described in Ref. 10 which yields devices capable of generating multiple harmonics with a fundamental frequency of 111 MHz, up to a maximum frequency of approximately 0.9 GHz. Most of the experiments were performed by exciting the second harmonic at 222 MHz. A Mn_{12} single crystal (cylindrical shape, 1 mm height, 0.2 mm diameter) was glued along its largest dimension directly on the IDTs, using commercial silicon grease, and the hybrid sample was inserted in the cryostat of a commercial magnetometer. We recorded simultaneously the reflection coefficient S_{11} and the magnetization of the Mn_{12} as a function of temperature (5–300 K) and magnetic field ($|H_{\text{max}}|=5$ T). The temperature of the IDT and of the helium gas, which acts as a heat ex-

changer, were monitored using two thermometers. The measurement of the reflection coefficient S_{11} was performed by using an Agilent network analyzer. The magnetization dynamics was accessed by exciting the IDTs with short microwave pulses and by detecting the magnetization using a rf-superconducting quantum interference device (SQUID). The microwave pulses were generated using an Agilent signal generator, which allows one to select the shape, duration and energy of the pulses in the frequency range between 250 kHz and 4 GHz. Most of the experiments were performed using rectangular microwave pulses of constant energy and different duration. The microwaves were transported to the transducer placed in the cryostat using coaxial cables, which introduce an attenuation of 10 dBm. The fast magnetization measurements (time resolution of 1 μs) were carried out at constant temperature and magnetic field by continuously reading the voltage variation detected by the rf-SQUID. To perform these measurements, we first cooled the sample down to the desired temperature under an applied magnetic field, and then we applied microwave pulses with widths in the range between 10 μs and 100 ms, separated by 100 ms.

The magnetic properties of the Mn_{12} crystal glued on the surface of the LiNbO_3 transducer were found to be similar to those previously published.^{5–9,11} Above the magnetic blocking temperature, the material exhibits superparamagnetic behavior, while below that temperature, the magnetization as a function of the magnetic field shows resonant spin tunneling transitions at multiple values of 0.45 T. Figure 1 shows the time evolution of the magnetization induced by SAW pulses of different durations (10 μs , 100 μs , 1 ms) at $T=8.5$ K and $H=4$ kOe. A magnification of the curve for the pulse time of 100 μs in the time window between 0 and 0.3 ms is shown in the inset of the figure. In all cases, the magnetization starts changing at about 30 μs after the application of the pulse, and the maximum departure from its initial value, ΔM , occurs well after the end of the pulse. The duration of this postpulse delay is always on the order of 30 μs , even for the

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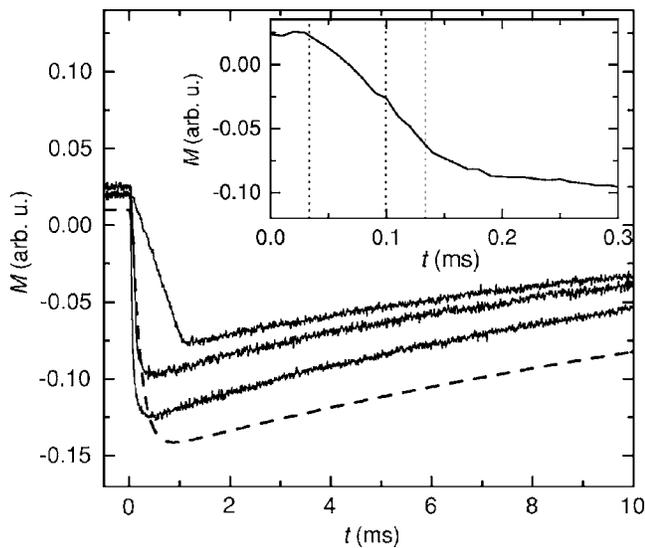


FIG. 1. Experimental time evolution of the magnetization of the Mn_{12} crystal for SAW pulses of different durations (10 μs , 100 μs , and 1 ms, respectively, from bottom to top curve) at $T=8.5$ K and $H=4$ kOe. The dashed line corresponds to a curve calculated using the theoretical model, for a pulse of 100 μs , under the same conditions of magnetic field and temperature. The inset shows a magnification of the time evolution curve for the pulse of 100 μs in the time range between 0 and 0.3 ms.

shortest pulse (10 μs). The time for the magnetization to recover its initial value after the microwave pulse finishes is, in contrast, always of the order of 50 ms (partially shown in Fig. 1).

In Fig. 2 we show the frequency dependence of S_{11} (lower curve) and ΔM (upper curve) when microwave pulses of 20 μs with a nominal power of 10 dB m and frequencies from 0.1 to 1 GHz are applied to the transducer, at $T=10$ K and $H=5$ kOe. Even though the Mn_{12} crystals are subject to the electric field produced by the IDT fingers during the whole frequency scan, changes in magnetization are only observed at the acoustic resonant frequencies. Similar results were obtained at different temperature and magnetic field conditions. This result unambiguously shows that fast magnetic measurements are able to detect the frequency of the different SAWs generated. The full width at half maximum of ΔM at the different resonant frequencies detected with this novel method gives frequency resolution and quality factor values similar to those determined by measuring

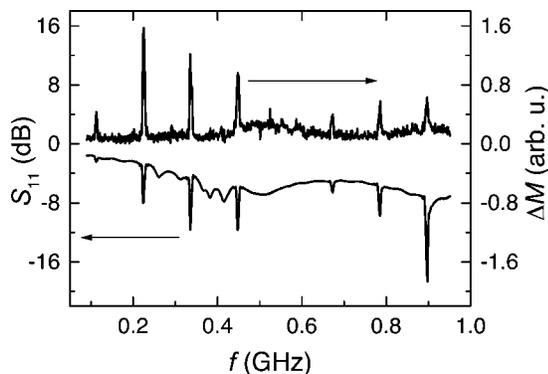


FIG. 2. Frequency dependence of the reflection coefficient S_{11} (lower curve, left axis) and the variation of magnetization of the Mn_{12} crystal, ΔM (upper curve, right axis), measured at $T=10$ K and $H=5$ kOe for a SAW pulse of 20 μs with a nominal power of 10 dB m.

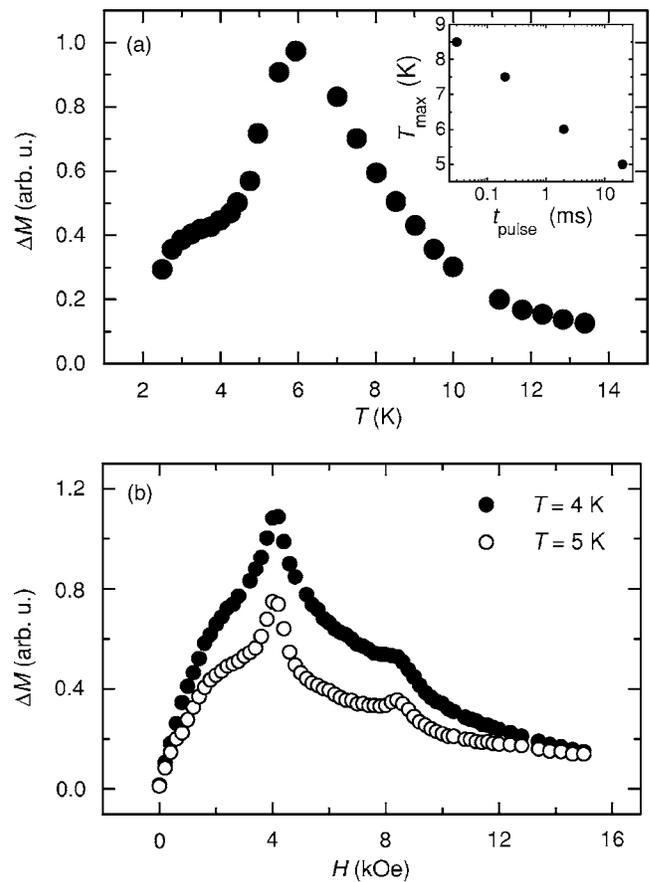


FIG. 3. Temperature dependence of the variation of magnetization of the Mn_{12} crystal measured at $H=4$ kOe for a SAW pulse of 2 ms (a), and magnetic field dependence of the variation of magnetization measured at $T=4$ K (solid circles) and 5 K (open circles) for the same pulse duration (b). The inset in the upper panel shows the pulse time dependence of the temperature at which the variation of magnetization presents the maximum.

the reflected energy using the network analyzer.

The temperature dependence of ΔM for pulses of 2 ms when $H=4$ kOe is shown in Fig. 3(a). A maximum for ΔM is observed at $T \sim 6$ K, while a small shoulder can also be distinguished between 3 and 4 K. The temperature T_{max} of that maximum is plotted in a semilog scale as a function of the pulse duration in the inset of the upper panel of that figure. The linear dependence, $T_{\text{max}} \propto 1/\ln(t_{\text{pulse}})$, clearly indicates that T_{max} corresponds to the blocking temperature of Mn_{12} , which is determined in each measuring process by the duration of the pulse. Figure 3(b) presents the magnetic field dependence of ΔM in the range between 0 and 15 kOe, at two different temperatures, $T=4$ and 5 K, for a pulse of 2 ms. For both curves, ΔM shows a strong peak at $H \sim 4.5$ kOe and a shallow maximum at $H \sim 9.0$ kOe, as it corresponds to the spin tunneling resonant fields of the Mn_{12} crystal.⁵⁻⁹

To explain these observations we have computed the time evolution of the population of the 21 discrete levels of the spin $S=10$ of the Mn_{12} molecules during the application of the acoustic pulse and afterwards. The energies of the spin levels are determined by the Hamiltonian

$$H = -DS_z^2 - aS_z^4 + g\mu_B H_z S_z, \quad (1)$$

where $D \sim 0.6$ K, $a \sim 8 \times 10^{-4}$ K,⁷⁻⁹ and the magnetic field is applied along the z direction to be aligned with the magnetic easy axis of the Mn_{12} crystal. We note that the SAW quanta in the present experiments, of the order of 10 mK, are con-

siderably smaller than the spin levels spacing (0.6–12 K). In addition, the time delay between the application of the acoustic pulse and the changes in magnetization, of the order of 30 μs , is considerably larger than the time required by the SAW to transit between the IDT and the Mn_{12} crystal. This delay indicates that the magnetization changes are not induced by the coherent acoustic vibrations generated by the transducers, but by the incoherent phonon bath resulting from the thermalization of such vibrations. The process can be therefore treated by considering the spin levels in thermal contact with the phonons of the lattice and assuming different temperatures for the spins, T_S , and for the lattice, T_l , that satisfy the differential equations

$$dT_l/dt = W(t) + \beta(T_l - T_0) - \alpha(T_l - T_S), \quad (2)$$

and

$$dT_S/dt = \alpha(T_l - T_S), \quad (3)$$

where α and β are time constants of heat transport, T_0 is the temperature of the helium bath, and $W(t)$ is the pulse function that introduces the heat into the system. A curve calculated for $T=8.5$ K, $H=4$ kOe, and a pulse time of 100 μs is shown as a dashed line in Fig. 1. The curve reproduces to great accuracy the behavior of the experimental data.

According to this model, the four distinct regions in the curves presented in Fig. 1 can be understood as follows: (i) the magnetization does not change noticeably during the first 30 μs , because the thermal phonon distribution is not generated immediately, so this elapsed time may be attributed to the lifetime of the SAWs; (ii) the quasilinear variation of the magnetization between $t=30$ μs and the end of the pulse is associated to the variation of the population of the different spin levels in the two energy wells, as a consequence of the continuous thermal flow between the lattice and the spins, generated by the decay of the energy of the SAWs; (iii) the variation of magnetization during the next 30 μs once the pulse has finished is due to the thermalization of the energy released by the last living SAWs; (iv) after this variation, the magnetization recovers its initial value in a time of the order of $\tau \sim 50$ ms. This value can be used to estimate an upper bound for the heat diffusivity as $\kappa = l^2/\tau \cong 5 \times 10^{-5}$ m^2/s ,

where l is a characteristic length for heat propagation typically on the order of 1 mm. The model reproduces also with great accuracy the temperature and magnetic field dependences of ΔM shown in Fig. 3.

In conclusion, we have shown the use of molecular magnets to determine the frequencies of SAWs generated by pulse microwaves in piezoelectrics. We have seen that the magnetic blocking temperature of the Mn_{12} crystal is determined by the duration of the microwave pulse and the lifetime of the SAWs in the LiNbO_3 is on the order of 30 μs . The results have been interpreted in the framework of a theoretical model based on the absorption of thermalized acoustic vibrations by the spin levels. This is a new way to study very fast spin tunneling processes in molecular magnets.

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