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Probing magnetic quantum tunneling in Fe_8 with muons

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Abstract

We present a μSR study of the Fe_8 molecules from which an evidence of quantum tunneling of the magnetization (QTM) can be seen, in the form of drastic changes in the local field experienced by the muon.

Keywords: High spin molecules; Quantum tunneling of magnetization

Technology today has reached a point where a single atom and a single molecule can be manipulated. This, essentially, allows reduction of the size of a magnetic memory unit from a billion atoms per bit [1], used today, to a single molecule. The difficulty lies in the stability of the stored information. The spin polarization, and hence the memory stored in a single molecule, can be lost via magnetic quantum tunneling. As a result considerable effort is being exerted to understand the mechanism of magnetic quantum tunneling in the hope of one day preventing it. The research in this field is concentrated on two special molecules Mn_{12} [2,3] and Fe_8 [4]. The latter molecule is particularly interesting since it exhibits 'pure' quantum tunneling of the magnetization, namely, the tunneling rate is temperature independent below 360 mK [4–7]. Therefore, there is an ongoing effort to make Fe_8 films. Once this goal is achieved there will be a need to confirm that the molecules in the films behave as the molecules in the bulk. This is not a simple task since measurements that have been used up to date such as susceptibility, specific heat, NMR, etc. need a large amount of sample and cannot be applied on thin films. Here we show that the muon spin rotation (μSR) technique, which is applicable to films [8], is sensitive to the quantum nature of the Fe_8 molecule.

The full formula of Fe_8 is $[Fe_8O_2(OH)_{12}(C_6H_{15}N_3)_6]Br_7(H_2O)Br \cdot 8H_2O$ [9]. It has a ground spin state of $S = 10$ and the most important terms in the Hamiltonian are given by $H = -DS_z^2 - g\mu_B \mathbf{S} \cdot \mathbf{H}$ where $D = 0.275$ K [5] and $\mathbf{H}$ is the external field. When the field is applied along the easy axis ($\mathbf{z}$) direction, the energy levels of this Hamiltonian are given by $E_m = -Dm^2 - g\mu_B mH$, where m is the quantum number of the spin in the $\mathbf{z}$ direction running from -10 to 10. In zero field the ground state is given by $m = \pm 10$. At

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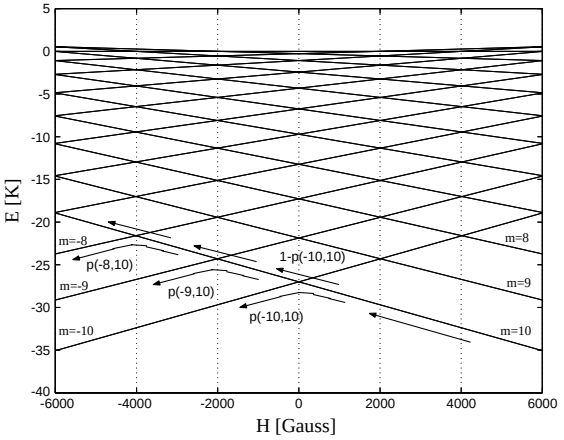

Fig. 1. Energy levels of the different spin states as a function of applied field $\mathbf{H}\|\mathbf{z}$.

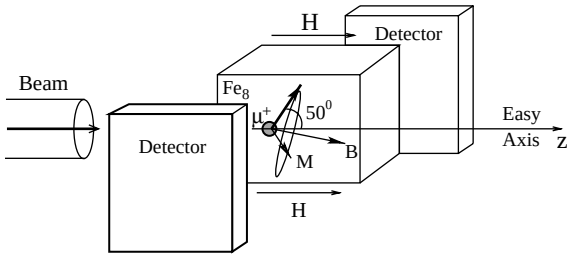

Fig. 2. The arrangement of the Fe_8 single crystal in the experimental apparatus. μ^+ is the muon, **M** is the magnetization, **B** and **H** are the internal and external magnetic fields, and **z** is the easy axis of the Fe_8 crystals.

certain 'matching' field values $H_n = 0, \pm 2.1, \pm 4.2$ kG etc. states with different m's (e.g. $m = +10$ and $m = -9$) can have identical energies, and spin transitions between these states may occur while maintaining energy conservation. This is demonstrated in Fig. 1 where the energy levels as a function of applied magnetic field are depicted for each quantum number m. The fields at which different lines cross each other are matching fields. However, the Hamiltonian H cannot induce tunneling between different spin states since it commutes with S_z, and the tunneling is induced by additional small terms in the Hamiltonian such as spin-phonon interaction [10,11], dipolar interactions, and nuclear fluctuations [12].

When applying a high positive field at low temperature, all molecules are polarized in the $m = 10$ spin state. As the field is decreased nearly to zero, the spins remain in this state. However, when the applied field becomes negative the ground state switches to the $m = -10$ state. As a result some of the spins tunnel from the $m = 10$ to the $m = -10$ state with probability $p(-10, 10)$. This tunneling takes place when the field is close to the first matching field $H_n = 0$. However, if the field sweep is fast, not all spins will have a chance to make the transition, and some will be 'stuck' in the $m = 10$ state. Transitions out of the $m = 10$ state can be ignored when $H \neq H_n$, since it takes years for the magnetization to reach equilibrium at such low temperatures [4]. Therefore, the second opportunity for the spins to escape the $m = 10$ and reach the $m = -10$ state is when the field is negative so that the $m = -9$ state has the same energy as the $m = 10$ state. Similarly, the third chance is when the field is further reduced so that the $m = -8$ state has the same energy as the $m = 10$ state, etc. This is demonstrated in Fig. 1 by the arrows. Each time a matching field H_n is crossed, we expect a change in the sample's magnetization.

Our μSR measurements were performed on a few aligned Fe_8 single crystals which were glued on a small silver plate. In these measurements we apply a magnetic field parallel to the easy axis **z** of the Fe_8 single crystals. The initial polarization of the muon is 50° relative to **z**, and the counters are perpendicular to **z**. This setup is demonstrated in Fig. 2. Also shown in this figure are a possible direction of the magnetization and the internal local magnetic field $\mathbf{B} = \mathbf{H} + \mathbf{M}$ around which the muons rotate. All measurements were performed at a temperature of 40 mK to minimize thermal activation effects.

These measurements were performed using a three step field cycle [13]: (I) a field of $H = +2$ T was applied for 15 min to polarize the Fe_8 molecules[1] (II) the field was swept to an intermediate negative magnetic field H_i with a sweep rate v, (III) eventually the field was swept back to $H = -50$ G where all measurements were performed. The only variable that changes between

[1] Experiments show that applying a field 2 T for longer times does not change the muon asymmetry.

the different runs is H_i. It allows us to sweep through different matching fields H_n's and to control the sample magnetization as explained above. A procedure similar to steps I and II is used in all other experiments on high spin molecules. The uniqueness of the μSR experiment is in step III, namely, all measurements are performed in the same field.

In Fig. 3 we present the asymmetry as a function of time for different intermediate fields H_i. The field sweep rate is $v = 0.24$ T/min. The particular field cycle for each measurement is indicated in the figure. One can clearly see that depending on H_i the asymmetry is different (especially between 0.5 and 2 μs), and that all asymmetries differ except for (b) and (c). The reason for this is that no other matching field is crossed and therefore there is no difference in the magnetization between these two cases. However, the runs (b) and (c) differ strongly from the other runs, for which new matching fields were crossed.

We analyze the data by fitting the asymmetry of all runs to a function of the form

$$A(t) = A_{10}\sin(\omega_{10}t)e^{-\lambda_{10}t} + A_{\pm 10}\sin(\omega_{\pm 10}t)e^{-\lambda_{\pm 10}t}, \quad (1)$$

where A_{10}, ω_{10} and λ_{10} represents the fraction of muons which experience a magnetic field mainly from molecules with $m = 10$ spin state. Similarly $A_{\pm 10}$, $\omega_{\pm 10}$ and $\lambda_{\pm 10}$ represent the fraction of muons which interact also with negative spin states. The precession frequencies ω_{10} and $\omega_{\pm 10}$ represent the different local fields experienced by the muons inside the Fe_8 single crystal. The fitting curves are the solid lines in Fig. 3, ω_{10} was found to be 13.4(3) MHz, common to all curves, as expected for muons which do not experience a change in their local field. The values of A_{10} and $A_{\pm 10}$, and $\omega_{\pm 10}$ are presented in Fig. 4(a) and (b) respectively. The changes in $\omega_{\pm 10}$ when matching fields are crossed indicate a change in the internal field, or in the spin direction of those Fe_8 molecules close to the muon and affecting its polarization. Similarly the observed decrease in A_{10} accompanied by an increase in $A_{\pm 10}$ indicate quantum transitions to the unpopulated spin state.

Thus we have been able to reveal the quantum nature of the molecules using muons. While this is an expensive method to measure magnetization

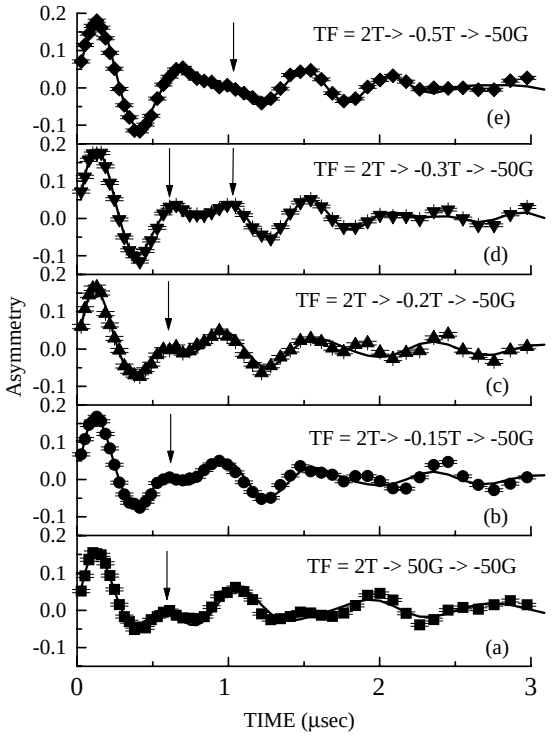

Fig. 3. The asymmetry as a function of time for different field cycles as indicated in the figure. These field cycles vary only in the intermediate field H_i. The arrow shows the region in time where asymmetry varies most.

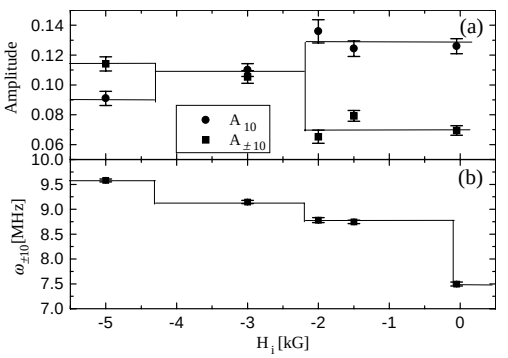

Fig. 4. (a) The amplitudes associated with the different fractions of muons, and (b) $\omega_{\pm 10}$ as a function of H_i. The amplitudes and frequency change only when different matching fields are crossed.

changes it might be the best or the only method to probe films of magnetic molecules.

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