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Tunnel splitting distributions and dipolar shuffling in Mn_{12}-acetate ☆

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Abstract

Analysis of magnetization data for the single molecule magnet, Mn_{12}-acetate, recorded for a magnetic field that is swept back and forth across each resonance, indicates that the transverse anisotropy is distributed and that the full transverse Hamiltonian, $\hat{V}_T = g_x\mu_B\hat{S}_x + E(\hat{S}_x^2 - \hat{S}_y^2) + (C/2)(\hat{S}_+^4 + \hat{S}_-^4)$ is needed to explain the data.

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Keywords: Single molecule magnet; Quantum tunneling; Distribution

The magnetization of Mn_{12}-acetate has been measured using micro-Hall bars in response to a magnetic field parallel to the anisotropy axis that is swept back and forth across each ground state energy resonance corresponding to steps $N = 4, \ldots, 9$. The data shown in Fig. 1 were taken at fixed sweep rate of 7.15 mT/s and at a temperature of $T = 0.25$ K.[1]

The fraction of molecules remaining in the metastable well $f_{N,j}$ after sweeping past the Nth resonance j times is related to the normalized magnetization M_j of the Mn_{12}-acetate crystal by the expression, $f_{N,j} = (1 - M_j)/2$. The inset to Fig. 1 details how $f_{N,j}$ was determined for the first few oscillations for resonance, $N = 7$. The probability of remaining in the metastable well for the Nth resonance is given by the Landau–Zener–Stueckelberg formula, $P_N = \exp(-\pi\Delta_N^2/2v_N)$, where Δ_N is the tunnel splitting and $v_N = g_z\mu_B(2S - N)\,dH_z/dt$ is the energy sweep rate for the Nth resonance.

The fraction of molecules remaining in the metastable well, $f_{N,j}$, is shown as a function of $j/(dH_z/dt)$ in Fig. 2 for each resonance, N, for three different field sweep rates, dH_z/dt. The data scale for all resonances except at the slowest sweep rate, $dH_z/dt = 0.45$ mT/s for $N = 7, \ldots, 9$. This suggests that dipolar shuffling, an effect proposed by Liu et al. [1] that results in a redistribution of internal magnetic fields whenever an individual molecule tunnels, does not play a significant role for the data taken at $dH_z/dt = 7.15$ mT/s and $dH_z/dt = 12.7$ mT/s.

The fraction $f_{N,j}$ is expected to be an exponential function of j for an ensemble of identical molecules, for in this case, $f_{N,j} = f_{N,0}(P_N)^j$. In contrast, the measured dependence is markedly non-exponential and has been explained in terms of a distribution of tunnel splittings [2]. Distributed tunnel splittings may be due to crystal defects such as edge dislocations, as suggested by Chudnovsky and Garanin [3], or to the different

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[1] Data (not shown) were also taken for sweep rates, 0.45 and 12.7 mT/s at $T = 0.25$ K.

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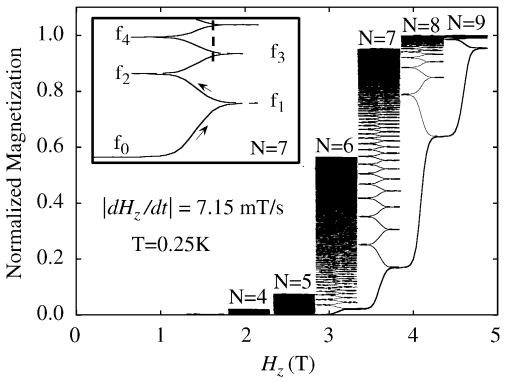

Fig. 1. Normalized magnetization versus magnetic field for a field that is swept back and forth across each resonance. The inset shows the first few oscillations for step $N = 7$.

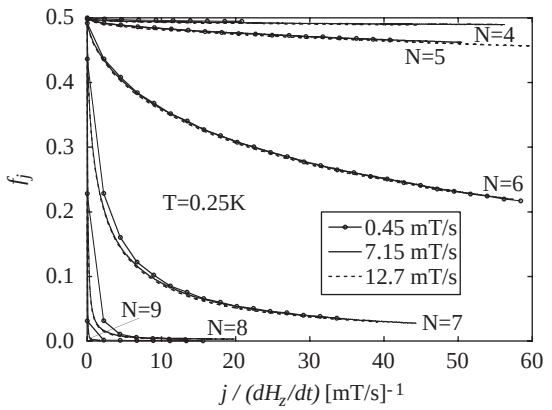

Fig. 2. The fraction of molecules remaining in the metastable well as a function of the oscillation number, $j/(\mathrm{d}H_z/\mathrm{d}t)$ for three different sweep rates.

coordination geometries of the manganese(III) ions, as pointed out by Cornia et al. [4]. In either case, $f_{N,j}$ is determined by integrating over the distribution, $\tilde{f}_{(X)}$: $f_{N,j} = \int_{-\infty}^{+\infty} \prod_{k=0}^{N-1} P_k(X)(P_N(X))^j \tilde{f}(X)\,\mathrm{d}X$; this expression reflects the fact that the field is swept once through each of $N-1$ previous resonance in the process of reaching the Nth resonance under study.

In order to determine the distribution from our experimental data, we have attempted to choose the form of $\tilde{f}(X)$ (as shown in Fig. 3) to fit all the curves

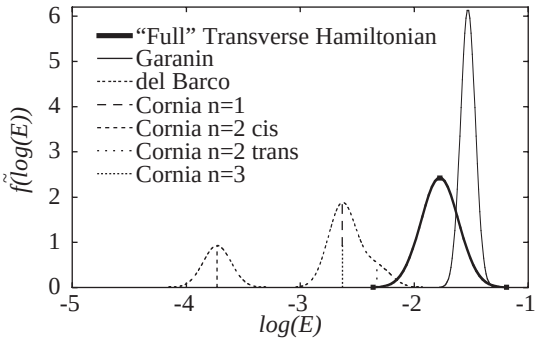

Fig. 3. The log-Gaussian distribution of transverse anisotropy, $\tilde{f}(\log(E))$, determined by fitting the data depicted in Fig. 2 for four different models of transverse anisotropy.

shown in Fig. 2 *simultaneously*. We found that the discrete distribution (vertical lines) of transverse anisotropy, $E(\hat{S}_x^2 - \hat{S}_y^2)$, suggested by Cornia et al. does not yield the shape of any of the curves. By adding width to each of these values (dashed curves), as suggested by del Barco et al. [5], the curves can all be fit individually. However, it is impossible to fit *all* the curves simultaneously. A simultaneous fit for all curves can be obtained by including a 1° misalignment of the crystal, incorporating fourth-order transverse anisotropy, $(C/2)(\hat{S}_+^4 + \hat{S}_-^4)$ with $C = 10$ μK, as well as a distribution of transverse anisotropy (thick solid curve). The values of E in this case are consistent with those reported by Hill et al. [6]. Also shown (thin solid curve) are previously reported data [2] that give a partial fit to the data but neglect fourth-order transverse anisotropy. This indicates that it is important to use the full transverse Hamiltonian, $\hat{V}_\mathrm{T} = g_x\mu_\mathrm{B}\hat{S}_x + E(\hat{S}_x^2 - \hat{S}_y^2) + (C/2)(\hat{S}_+^4 + \hat{S}_-^4)$.

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