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Journal of Magnetism and Magnetic Materials 272–276 (2004) e739–e740
journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Thermally assisted tunneling for a distribution of tunnel splittings in Mn_{12}-acetate ☆

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Abstract

Scaling of magnetic relaxation data for Mn_{12}-acetate due to ground-state tunneling at 0.24 K is extended to thermally assisted tunneling at higher temperature. Calculated magnetic relaxation using a distribution of tunnel splittings due to second-order transverse anisotropy is compared with measured magnetization curves for temperatures between 0.24 and 1.9 K.

PACS: 75.45.+j; 75.60.Ej

Keywords: Single molecule magnet; Quantum tunneling; Distribution

Mn_{12}-acetate is a well-studied single-molecule magnet which exhibits quantum tunneling of magnetization manifested as steps in the magnetic relaxation below the blocking temperature for specific values of magnetic field applied parallel to the anisotropy axis. For ground-state tunneling (GST) at 0.24 K, the step heights have been analyzed using the Landau–Zener–Stueckelberg (LZS) formula; the primary symmetry breaking term (which drives tunneling) has been shown to be a distributed second-order transverse anisotropy, $E(\hat{S}_x^2 - \hat{S}_y^2)$, which is not allowed by four-fold crystal symmetry [1]. Possible sources of E are crystal dislocations, as suggested by Chudnovsky and Garanin [2], and solvent disorder within a molecule, as suggested by Cornia et al. [3]. In this paper the analysis of [1] is extended to include thermally-assisted tunneling (TAT) for higher temperatures.

For a distribution of tunnel splittings, the fraction of molecules that tunnels depends on what part of the distribution has not tunneled at the previous resonances. Instead of the step-function approximation for the LZS formula used in Ref. [1], numerical calculations were performed by integrating over a distribution of E for each of 11 GST resonances, 69 TAT resonances and five over-the-barrier hopping processes[1] for steps number $k = 1$–11.[2] The formulae in Ref. [4] were used to calculate the tunnel splittings and the tunneling probability for the incoherent LZS transition. The same formula was used for over-the-barrier hopping processes by taking the limit $\delta\varepsilon_{n,k} \ll \Delta_{n,k}$, where $\Delta_{n,k}$ is the tunnel splitting and $\delta\varepsilon_{n,k}$ is the energy difference to the lower adjacent energy level. In Fig. 1, calculated magnetization curves and the experimental data are plotted for

☆ This work was supported by NSF grants DMR-0116808, CHE-0095031, DMR-0103290, and CHE-0123603, and by ISF Grant 8003/02.

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[1] This process occurs when one of the energy level is at the top of the energy barrier.

[2] Some of TAT resonances occur before the GST resonance of the following step number.

0304-8853/$ - see front matter
doi:10.1016/j.jmmm.2003.12.054

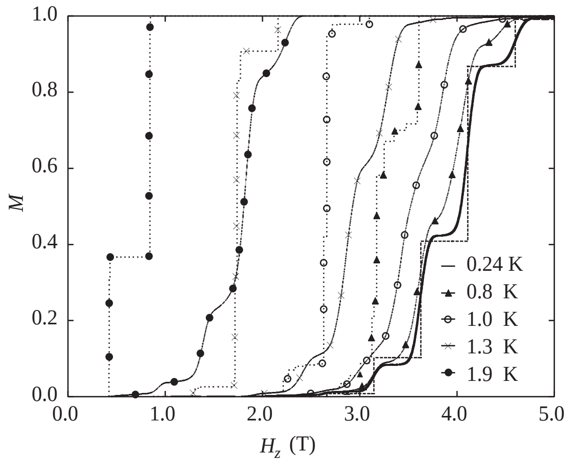

Fig. 1. Calculated magnetization curves (dotted lines) are plotted with experimental data (solid lines) for different temperatures.

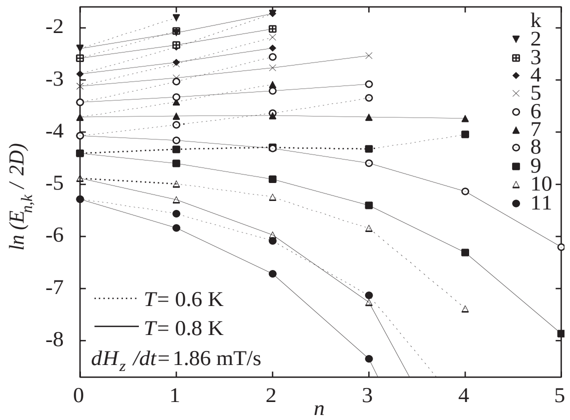

Fig. 2. $E_{n,k}$ is the calculated value of E which would give $1 - 1/e$ of tunneling probability (larger if $E > E_{n,k}$) at the kth step for the nth excited state [4].

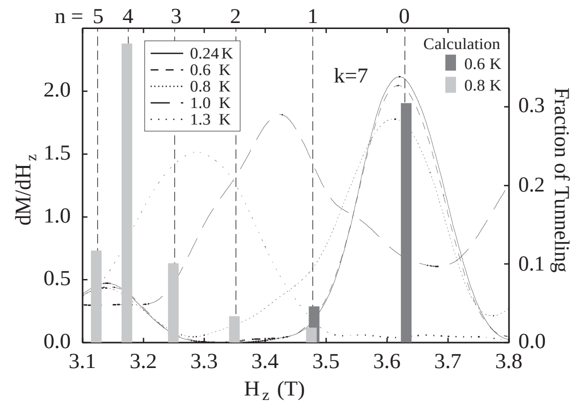

Fig. 3. Measured dM/dH_z (solid and dotted lines) and calculated fraction of tunneling (bars) are plotted for $k = 7$.

different temperatures. The distribution of E was used in our calculations that provides the best fit to the plateau heights for GST at 0.24 K. The data show widths of $\approx$0.15 T at the steps due to internal fields and a small distribution of longitudinal anisotropy. This has a negligible effect on the step heights.

There are significant discrepancies between the calculation and the data. Relatively large values of E are required to fit the GST at 0.24 K while for higher temperatures, tunneling is too large, allowing its saturation to come at much lower H_z than the experimental data. Even with smaller E, no distribution of E was found that fit the magnetization curves for temperatures above 0.5 K. As seen in Fig. 2, the presence of E produces a very abrupt transition from GST to TAT at the top of the barrier as the temperature increases [4]. In Fig. 3, calculated fractions of tunneling (bars) show an abrupt transition with the presence of a distribution of tunnel splittings. Note that higher excited states come into resonance at lower H_z than the lower energy states. A relatively more gradual transition has been found in experimental data [5] as seen in Fig. 3.

Neglecting the fourth-order longitudinal anisotropy may cause a small deviation. Including fourth-order transverse anisotropy and transverse magnetic field could improve the fit to some degree. We note that the possible role of the $S = 9$ manifold [6] and spin-spin cross-relaxation [7] may need to be considered.

We thank Nurit Avraham for help with the manuscript.

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