Coherent Manipulation and Decoherence of S=10 Single-Molecule Magnets

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Introduction

Single-molecule magnets (SMMs) are nanoscale quantum magnets which exhibit quantum tunneling of magnetization (QTM) and quantum phase interference (Berry phase). As solid-state based spin systems, SMMs form a unique class of materials that have a high-spin, and whose spin state and interaction can be easily tuned by changing peripheral organic ligands and solvate molecules. Because the molecules within the crystal lattice of SMMs interact very weakly with each other, properties of a single SMM can be deduced from measurements of a macroscopic ensemble. Although quantum phenomena observed in SMMs have been investigated extensively, couplings between SMMs and their environment are still poorly understood for proposed applications to dense and efficient quantum memory, computing, and molecular spintronics devices.

Experimental

Spin relaxation times (T_1 and T_2) of S=10 Fe_8 SMM single crystals were investigated using 240 GHz continuous wave (cw) and pulsed EPR spectrometers at the NHMFL, EMR program.

Results and Discussion

We observed spin echoes of the m_S=–10↔–9 transition in S=10 Fe_8 SMM single crystals. This is the first observation of spin echoes from high-spin SMM single crystals.

The spin-lattice relaxation time T_1 was directly measured at 1.3 K using a stimulated echo sequence. We found that stimulated echoes are detectable for more than a few milliseconds and that the echo decay curve shows two relaxation rates with a short time of T_{short} = 1.0 ± 0.1 μs and a long time of T_{long} = 948 ± 108 μs. Because of the small excitation bandwidth, a strong spectral diffusion is expected. Therefore we currently speculate that the T_{short} is due to spectral diffusion and the T_{long} is the spin-lattice relaxation time T_1. Surprisingly, this value of T_1 is more than two orders of magnitude longer than previous findings.

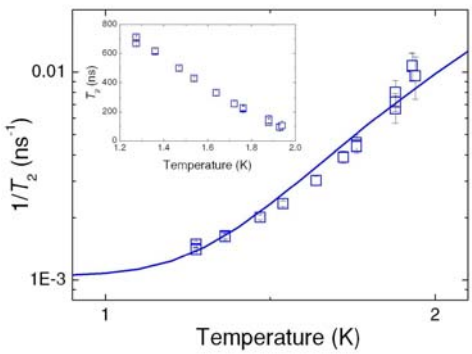

Fig.1. Temperature dependence of T_2. Solid line shows the fit by the spin flip-flop model.

Fig.1 shows the temperature dependence of T_2. Observation of a strong temperature dependence of T_2 suggests that the main decoherence mechanism at higher temperatures is due to coupling to fluctuating neighboring electron spins which is often pictured as a spin bath [1]. Since the cw EPR spectra show that Fe_8 SMMs dominate the population of electron spins in the sample crystal, the source of the Fe_8 spin decoherence is fluctuations of the Fe_8 spin bath itself. T_2 below 1 K is limited by other temperature-independent decoherence sources which are likely nuclear moments and phonon.

Conclusions

We demonstrated the first direct determination of T_1 and T_2 for Fe_8 SMMs single crystals using spin echoes. Spin decoherence is significantly suppressed by polarizing the Fe_8 spin bath. See Ref. [2] for details.

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References

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[2] Takahashi, S., *et al*., submitted to Phys. Rev. Lett., arXiv:0810.1254.