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

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A single crystal of high-spin single-molecule magnets (SMMs) is an attractive testbed for quantum science and technologies. High-spin SMMs are suitable for applications to dense quantum memory and computing devices. Because SMM clusters are identical and interact weakly, the ensemble properties of single crystals of SMMs reflect the properties of a single cluster. However coherent manipulation of high-spin SMM crystals has never been demonstrated due to strong spin decoherence. For spins in the solid state, an interaction with fluctuations of surrounding spin bath is a major source of spin decoherence. One approach to reduce spin bath fluctuations is to bring the spin bath into a well-known quantum state that exhibits little or no fluctuations. A prime example is the case of a fully polarized spin bath. In diamond, spin decoherence has been quenched using high-frequency pulsed electron paramagnetic resonance (EPR) [1]. We present coherent manipulation and decoherence of a single-crystal of S=10 Fe8 SMMs. Through polarizing a spin bath in Fe8 single-molecule magnets at 4.6 T and 1.3 K, we demonstrate that spin decoherence is significantly suppressed to extend the spin decoherence time (T_2) up to 700 ns [2]. Investigation of temperature dependence of spin relaxation times reveals the nature of spin decoherence. This work is collaboration with J. van Tol, C. C. Beedle, D. N. Hendrickson, L.-C. Brunel, and M. S. Sherwin.

[1] S. Takahashi, R. Hanson, J. van Tol, M. S. Sherwin, and D. D. Awschalom, *Phys. Rev. Lett.* **101**, 047601 (2008).
[2] S. Takahashi, J. van Tol, C. C. Beedle, D. N. Hendrickson, L.-C. Brunel, and M. S. Sherwin, arXiv: 0810.1254.