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Geometric-Phase Interference in a Mn_{12} Single-Molecule Magnet with Truly Fourfold Symmetry[1]

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A single-molecule magnet (SMM) is a large-spin system with an anisotropy barrier separating preferred "up" and "down" orientations. The spin can tunnel between these directions when an external longitudinal magnetic field brings levels in opposite wells into resonance. When there exist more than one energetically equivalent paths for tunneling, those paths can interfere, a geometric-phase effect that modulates the rate at which spins flip direction. The interference can be controlled by a magnetic field applied perpendicular to the spin's easy magnetization axis. In a ground-breaking experiment, Wernsdorfer and Sessoli [1] found oscillations in the probability of spin tunneling as a function of the field applied along the hard axis of the Fe_8 SMM. This observation confirmed a theoretical prediction by Garg [2]. Similar geometric-phase interference has been observed in other SMMs that have effective two-fold symmetry, where tunneling involves the interference between two equal-amplitude paths. Such interference effects have not previously been seen in systems with four-fold rotational symmetry. In recent work [3], my group has seen evidence of the observation of a geometric-phase interference effect in the Mn_{12}-^{t}BuAc SMM, a variant of the bellwether Mn_{12}-Ac SMM that has true four-fold rotational symmetry (being free of the solvent disorder that breaks the four-fold symmetry in the latter). The spin relaxation rate as a function of the applied transverse magnetic field shows a modulated behavior, with retarded relaxation near where one expects destructive interference between tunneling paths associated with excited states. Tuning the direction of the transverse field away from the hard axis washes out the observed interference effect by favoring one tunneling path over others. Detailed master-equation calculations are used to fit the observed behavior and yield anisotropy parameters consistent with values determined by other groups. Unlike previous observations of geometric-phase interference, which involved ground-state tunneling, the interference effect we observe in Mn_{12}-^{t}BuAc takes place in the thermally assisted tunneling regime where tunneling occurs near the top of the barrier. The interference effect enables us to clearly identify which levels participate in the thermally assisted process. Some preliminary results on geometric-phase interference in a version of Mn_{12}-Ac that is crystalized without solvent disorder will also be presented.

[1] W. Wernsdorfer and R. Sessoli, Science **284**, 133 (1999).
[2] A. Garg, Europhys. Lett. **22**, 205 (1993).
[3] S. T. Adams *et al.*, Phys. Rev. Lett., **110**, 087205 (2013).

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