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The Role of Antisymmetric Exchange on the Quantum Interference between States of Different Spin Length in a dimeric Molecular Nanomagnet.

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We report direct evidence of quantum oscillations of the *total spin length* of a dimeric molecular nanomagnet through the observation of quantum interference associated with tunneling trajectories between states having different spin quantum numbers. As we outline, this is a consequence of the unique characteristics of a molecular Mn₁₂ wheel which behaves as a (weak) ferromagnetic exchange-coupled molecular dimer: each half of the molecule acts as a single-molecule magnet (SMM), while the weak coupling between the two halves gives rise to an additional internal spin degree of freedom within the molecule, namely that its total spin may fluctuate. This extra degree of freedom accounts for several magnetization tunneling resonances that cannot be explained within the usual giant spin approximation. More importantly, the observation of quantum interference provides unambiguous evidence for the quantum mechanical superposition involving entangled states of both halves of the wheel. Magnetization results obtained in two other versions of this compound, in which the ligands have been modified, show that slight variations of the relative distance between the Mn ions determine whether the molecule behaves as a rigid magnetic unit of spin $S = 7$ or as two exchange-coupled halves of spin $S = 7/2$. We analyze the effect of the Dzyaloshinskii-Moriya antisymmetric exchange interaction in a molecule with a centre of inversion symmetry and propose a formal model to account for the observed broken degeneracy that preserves the molecular inversion symmetry.