High field EPR investigations of quantum and environmental effects in single molecule nanomagnets

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We report high frequency electron paramagnetic resonance (EPR) investigations of a series of high spin (total spin up to S = 10) manganese, iron, cobalt and nickel complexes which have been shown to exhibit single molecule magnetism, including low temperature (below ~ 1K) hysteresis loops and resonant magnetic quantum tunneling. A cavity perturbation technique enables high sensitivity oriented single crystal EPR measurements spanning a very wide frequency range (16 to 200+ GHz). Fitting of the frequency and field orientation dependence of EPR spectra allows direct determination of the effective spin Hamiltonian parameters. Studies for a range of materials with varying (approximately axial) site symmetries facilitates an assessment of the role of transverse anisotropy (terms in the Hamiltonian that do not commute with S_z) in the magnetic quantum tunneling phenomenon. We also examine quantitatively the temperature dependence of the EPR linewidths and line shifts, for fixed frequency measurements with an applied magnetic field along the easy axis. Simulations of the obtained experimental data take into account intermolecular spin-spin interactions (dipolar and exchange), as well as distributions in both the uniaxial anisotropy parameter D and the Lande g-factor. These findings could have important implications for the mechanism of quantum tunneling of magnetization in nanomagnets, as well as providing deeper insights into the interactions which give rise to quantum decoherence.

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