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Coherent manipulation of mononuclear lanthanide-based single-molecule magnets SAITI DATTA, SANHITA GHOSH, JUREK KRZYSTEK, STEPHEN HILL, National High Magnetic Field Laboratory and Department of Physics, Florida State University, Tallahassee, FL 32310, ENRIQUE DEL BARCO, Department of Physics, University of Central Florida, Orlando, FL 32816, SALVADOR CARDONA-SERRA, EUGENIO CORONADO, Instituto de Ciencia Molecular, UniVersidad de Valencia, Poli'gono la Coma s/n, 46980 Paterna, Spain —
Using electron spin echo (ESE) spectroscopy, we report measurements of the longitudinal (T_1) and transverse (T_2) relaxation times of diluted single-crystals containing recently discovered mononuclear lanthanide-based single-molecule magnets (SMMs) encapsulated in polyoxometallate cages [AlDamen *et al. J. Am. Chem. Soc.* 130, 8874 – 8875 (2008)]. This encapsulation offers the potential for preserving bulk SMM properties outside of a crystal, e.g. in molecular spintronic devices. The magnetic anisotropy in these complexes arises from the spin-orbit splitting of the ground state J multiplet of the lanthanide ion in the presence of a ligand field. At low frequencies only hyperfine-split transitions within the lowest ground state $\pm m_J$ doublet are observed. Spin relaxation times were measured for a holmium complex, and the results were compared for different hyperfine transitions and crystal dilutions. Clear Rabi oscillations were also observed, indicating that one can manipulate the spin coherently in these complexes.

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