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Observation of Highly Forbidden Single-Photon Transitions in a Ni_4 Single-Molecule Magnet[1] YIMING CHEN, Amherst College and University of Massachusetts at Amherst, MOHAMMAD D. ASHKEZARI, Amherst College, RAFAEL CASSARO, University of Massachusetts at Amherst, JONATHAN FRIEDMAN, Amherst College — We report electron-spin resonance experiments on a crystal of the single-molecule magnet (SMM) $[Ni(hmp)(dmb)Cl]_4$ (hereafter Ni_4), which is an $S = 4$ system with large uniaxial anisotropy. At 115.54 GHz and low magnetic fields (below the anisotropy field for the SMM), we observe two weak resonances that correspond to highly forbidden transitions between magnetic sublevels, one corresponding to $\Delta m \approx 6$ and another corresponding to $\Delta m \approx -7$. The interpretation of the observed transitions is confirmed by following how the peak positions change with the angle between the sample's easy axis and the applied field. The selection rules forbidding these transitions are lifted by tunneling between m states. The observed forbidden transitions can be viewed as tunneling-assisted direct single-photon transitions between spin states. Equivalently, the forbidden transitions can be interpreted as resonant tunneling between one spin state of the molecule and the dressed state of another spin state. The forbidden transitions have much narrower line widths than the allowed transitions, which suggests that the lines are not inhomogeneously broadened by local fields. Assuming homogeneous broadening, we infer a decoherence time T_2 of $\sim$0.5 ns.

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