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Origin of the fast magnetization tunneling in [Ni(hmp)(tBuEtOH)Cl]4 JON LAWRENCE, CEM KIRMAN, STEPHEN HILL, University of Florida, EN-CHE YANG, DAVID HENDRICKSON, University of California at San Diego, UNIVERSITY OF FLORIDA, PHYSICS DEPT. COLLABORATION, UNIVERSITY OF CALIFORNIA AT SAN DIEGO, CHEMISTRY DEPT. COLLABORATION — High-frequency (40-360 GHz), angle-dependent EPR data have been collected for single-crystals of $[\mathrm{Ni(hmp)(dmb)Cl}]_4$, and $[\mathrm{Ni_{0.02}Zn_{0.98}(hmp)(dmb)Cl}]_4$. The all-nickel complex behaves as a single-molecule magnet (SMM) at low temperatures, displaying hysteresis and magnetic quantum tunneling. However, in spite of its high symmetry (S_4), the relaxation is found to be very fast. We show that the origin of this behavior is related to a 4^{th}-order transverse crystal-field interaction, $B_4^4(S_+^4 + S_-^4)$, which produces a significant tunnel-splitting (~10 MHz) of the $m_s = \pm 4$ ground state of this $S = 4$ SMM. The fourth-order (B_4^4) and uniaxial (D) crystal-field strengths can be related to the directionality and magnitude of the single-ion interactions (D_i and E_i) at the individual Ni^{II} sites, as determined for the doped crystals. Variable-temperature EPR measurements also reveal the locations of excited spin states ($S = 3$, 2, etc..), enabling estimates of intra-molecular exchange coupling strengths.

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