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Pushing the Limits of Magnetic Anisotropy in a Mononuclear Ni(II) Single-Molecule Magnet: a High-Field EPR Study LAKSHMI BHASKARAN, Department of Physics, Florida State University (NHMFL), Tallahassee, Florida, USA, KATIE MARRIOTT, MARK MURRIE, Department of Chemistry, University of Glasgow, Scotland, UK, STEPHEN HILL, Department of Physics, Florida State University (NHMFL), Tallahassee, Florida, USA — Single-Molecule Magnets (SMMs) are potential candidates for nanoscale magnetic information storage, where slow magnetization dynamics (bistability) is realized at low temperatures due to a magnetic anisotropy barrier separating the "spin-up" and "spin-down" states of the SMMs. Here, we report spectroscopic evidence for a huge easy-axis anisotropy in a trigonal bipyramidal (TBP) $[Ni^{II}Cl_3(Me\text{-}dabco)_2]$ complex with an orbitally degenerate ground state. Single-crystal EPR studies were carried out in a 35T resistive magnet at the NHMFL. A very strong angle-dependence of the spectrum was observed within a few degrees of the hard plane, suggesting a huge zero-field-splitting (zfs) parameter, $|D|>300$ cm^{-1}, associated with first order spin-orbit coupling. This value is considerably larger than previously reported for a Ni^{II} TBP complex [1], and is thought to be due to the rigidity of the ligand that prevents Jahn-Teller type effects that can reduce D [2]. This is confirmed by the small value of the rhombic parameter, $|E|= 0.66$ cm^{-1}.

[1] R. Ruamps et al., JACS, 135, 3017-3026 (2013).
[2] M. Gruden-Pavlovic et al., Chem. Sci., 5, 1453-1462 (2014)

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