PROBING GIANT MAGNETIC ANISOTROPIES IN MONONUCLEAR SINGLE-MOLECULE MAGNETS USING VERY HIGH-FIELD EPR

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The development and study of molecular nanomagnets has witnessed tremendous progress in recent years, with potential applications on the horizon. Of particular interest are so-called single-molecule magnets (SMMs) that display slow magnetic relaxation below a characteristic blocking temperature, $\mathbf{T}_B$, due to a combination of a large magnetic moment and appreciable spin-orbit (SO) anisotropy. Early efforts aimed at increasing $\mathbf{T}_B$ focused on polynuclear clusters and maximization of the molecular spin state, $\boldsymbol{S}$. However, this becomes challenging for large clusters whilst simultaneously maintaining the molecular anisotropy. Thus, a more direct route to increasing $\mathbf{T}_B$ involves optimization of the magnetic anisotropy, albeit for simpler molecules in which one can exert synthetic control over the ligand field (LF). In particular, certain transition metals residing in high-symmetry coordination environments can experience orbital degeneracies and very strong first-order contributions to their SO anisotropy. This talk will highlight recent work involving $\boldsymbol{S} = 1$ Ni^{II} [1] and V^{III} [2] complexes subjected to rigid trigonal coordination environments in the solid state that are relatively stable against symmetry lowering Jahn-Teller distortions [3]. The resulting giant anisotropies associated with these species have been measured using very high-field (up to 35 T) EPR techniques.

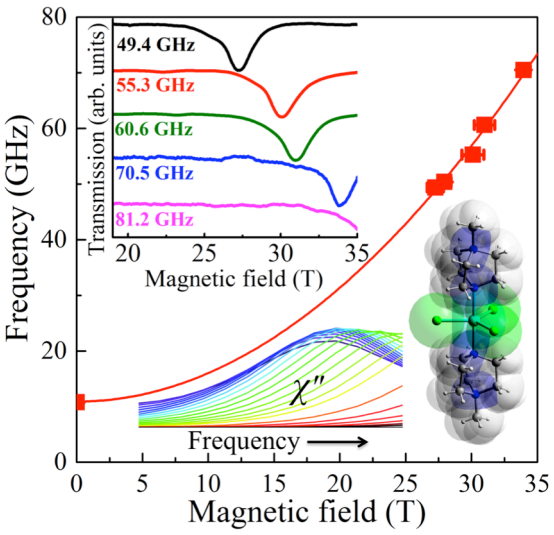

Fig. 1. Frequency-dependent high-field EPR spectra (upper inset) and peak positions (main panel) obtained with the magnetic field applied in the hard plane of $[Ni(MDABCO)_2Cl_3]ClO_4$ (right inset); frequency-dependent out-of-phase AC susceptibility peaks (lower inset) indicate slow magnetic relaxation - see [1] for details.

[1] K. E. R. Marriott, L. Bhaskaran, C. Wilson, M. Medarde, S. T. Ochsenbein, S. Hill and M. Murrie, Chem. Sci., 2015; DOI: 10.1039/C5SC02854J.
[2] M. R. Saber, M. Atanasov, L. Bhaskaran, K. Thirunavukkuarasu, S. Hill, F. Neese and K. R. Dunbar (submitted).
[3] R. Ruamps, R. Maurice, L. Batchelor, M. Boggio-Pasqual, R. Guillot, A.-L. Barra, J. Liu, E.-E. Bendief, S. Pillet, S. Hill, T. Mallah and N. Guihery, J. Am. Chem. Soc. **135**, 3017-3026 (2013).

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