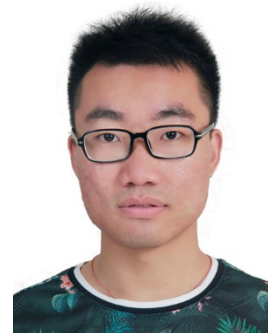


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2. "Uranocenium: Synthesis, Structure and Chemical Bonding", F.-S. Guo et al., *Angew. Chem. Int. Ed.*, **2019**, 58, 10163.
3. "Main Group Chemistry at the Interface with Molecular Magnetism", F.-S. Guo et al., *Chem. Rev.*, **2019**, 119, 8479
4. "Magnetic hysteresis up to 80 kelvin in a dysprosium metallocene single-molecule magnet", F.-S. Guo et al., *Science*, **2018**, 362, 1400.
5. "A dysprosium metallocene single-molecule magnet functioning at the axial limit", F.-S. Guo et al., *Angew. Chem. Int. Ed.*, **2017**, 56, 11445.
6. "Strong direct exchange coupling and single-molecule magnetism in indigo-bridged lanthanide dimers", F.-S. Guo et al., *Chem. Comm.*, **2017**, 53, 3130.
7. "Anion-Templated Assembly and Magnetocaloric Properties of a Nanoscale {Gd₃₈} Cage versus a {Gd₄₈} Barrel", R. Ohtani, M. Ohba, S. Kitagawa et al., *Chem. Eur. J.*, **2013**, 19, 14876.
8. "A large cryogenic magnetocaloric effect exhibited at low field by a 3D ferromagnetically coupled Mn(II)–Gd(III) framework material", F.-S. Guo et al., *Chem. Comm.*, **2012**, 48, 12219.
9. "Polynuclear and Polymeric Gadolinium Acetate Derivates with Large Magnetocaloric Effect", F.-S. Guo et al., *Inorg. Chem.*, **2012**, 51, 405.

Magnetic Hysteresis above 77 K in a Dysprosium Metallocene Single-Molecule Magnet

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Single-molecule magnets (SMMs) containing only one metal center are a type of nanomagnet that may represent the lower size limit for molecule-based magnetic information storage materials.^[1-2] The current drawback is that all SMMs require liquid-helium cooling to show magnetic memory effects. We now show that a strategy in which two key structural parameters within the metallocene framework – i.e. the Dy-Cp_{cent} distances and the Cp-Dy-Cp bending angle – are rendered short and wide, respectively, through a careful choice of ligand substituent produces an axial crystal field of sufficient strength to furnish the first SMM with a blocking temperature above 77 K.

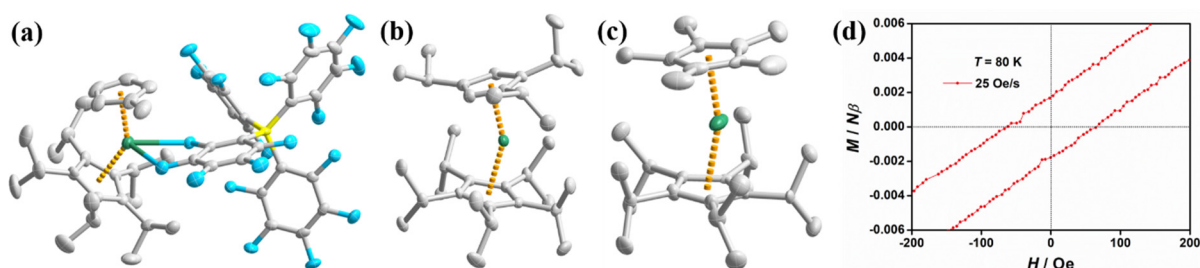


Figure 1. (a-c) Structures of the dysprosium metallocene complexes. (d) Magnetic hysteresis of the “5*” cation at 80 K with a sweep rate of 25 Oe/s.

A dysprosium metallocene cation, $[(Cp^{iPr5})Dy(Cp^*)]^+$ (“5*” cation, see Figure 1c), was targeted with cyclopentadienyl substituents of sufficient bulk to produce a wide Cp-Dy-Cp angle, but not too bulky such that close approach of the ligands became hindered. A new record energy barrier of $U_{eff} = 1,541 \text{ cm}^{-1}$ is described. The blocking temperature of $T_B = 80 \text{ K}$ for the 5* cation ushers in an era of high-temperature SMMs, thus overcoming an essential barrier towards the development of nanomagnet devices that function at practical temperatures.^[3]

Acknowledgments: The author thanks the MSCA Fellowship (grant 653784) and ERC (CoG 646740) for financial support; and Prof. Richard A. Layfield, Prof. Ming-Liang Tong and Dr. Akseli Mansikkamäki for the kind cooperation.

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