



Three-axis anisotropic exchange coupling in the single-molecule magnets $\text{NEt}_4[\text{Mn}^{\text{III}}_2(5\text{-Brsalen})_2(\text{MeOH})_2 \text{M}^{\text{III}}(\text{CN})_6]$ ($\text{M}=\text{Ru}, \text{Os}$)

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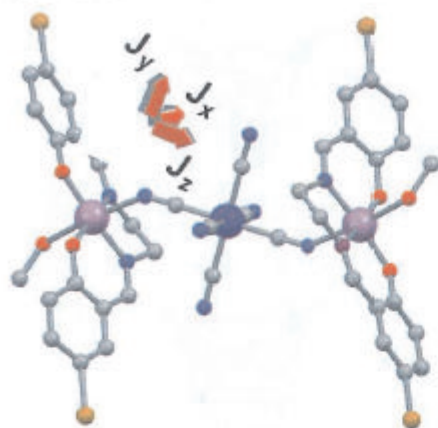
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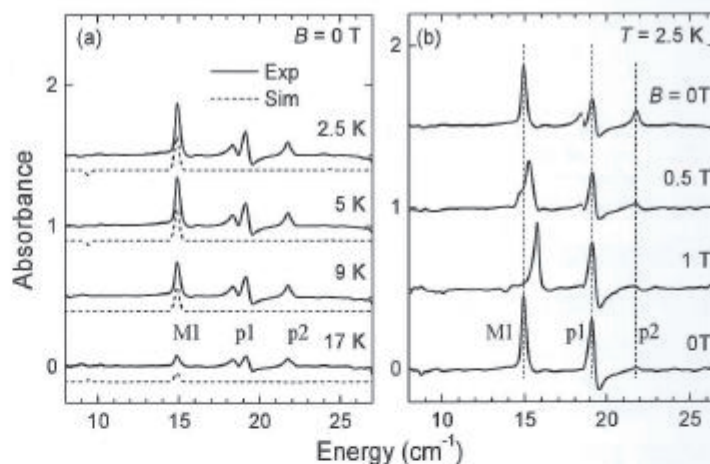
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The introduction of 4d and 5d ions into single-molecule magnets (SMMs) represents an intriguing path to enhance their properties [1]. Strong spin-orbit coupling and the diffuse 4d and 5d orbitals as compared to the 3d case enable large magnetic anisotropies and strong exchange coupling. Here we study isostructural Mn-M-Mn trinuclear SMMs, with $\text{M}=\text{Ru}$ and Os , by frequency-domain Fourier-transform THz electron paramagnetic resonance (FDFT-EPR), inelastic neutron scattering (INS) and SQUID magnetometry [2,3]. We find that the exchange coupling between Mn^{III} and Os^{III} ions is anisotropic and of three-axis type, i.e., it is antiferromagnetic along x and z axes while it is ferromagnetic along y . A further analysis yields that this is a consequence of orbitally dependent exchange [4,5] providing a profound understanding of the magnetic behavior of the orbitally degenerate Ru^{III} and Os^{III} ions.



(a)



(b)

Figure: (a) Structure of the Mn-Os-Mn SMM from X-ray diffraction. The NEt_4^+ counterion and H atoms have been omitted. Color code: Mn, purple; Os, dark blue; O, red; N, blue; C, grey; Br, brown. (b) FDFT THz-EPR spectra obtained on Mn-Os-Mn.

Literature: [1] X.-Y. Wang, C. Avendano, K. R. Dunbar, *Chem. Soc. Rev.* 2011, **40**, 3213; [2] K. S. Pedersen, J. Dreiser, J. Nehr Korn, M. Gysler, M. Schau-Magnussen, A. Schnegg, K. Holldack, R. Bittl, S. Piligkos, H. Weihe, P. Tregenna-Piggott, O. Waldmann, J. Bendix, *Chem. Commun.* 2011, **47**, 6918; [3] J. Dreiser, K. S. Pedersen, A. Schnegg, K. Holldack, J. Nehr Korn, M. Sigrist, P. Tregenna-Piggott, H. Mutka, H. Weihe, V. S. Mironov, J. Bendix, O. Waldmann, *Chem. Eur. J.* 2013, **19**, 3693; [4] J. J. Borrás-Almenar, J. M. Clemente-Juan, E. Coronado, A. V. Pali, B. S. Tsukerblat, *J. Phys. Chem. A* 1998, **102**, 200; [5] V. S. Mironov, L. F. Chibotaru, A. Ceulemans, *J. Am. Chem. Soc.* 2003, **125**, 9750.