Supplementary information

Recent Advances in Computational Modelling of Mononuclear Actinide Single Molecule Magnets

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Computational details:

The ab initio calculations on the crystal structures of complexes **3**, **5**, **8** and **39** have been performed on the MOLCAS8.2 programme package.[1] The scaler relativistic effect in our calculations has been considered the Douglas-Kroll-Hess (DKH) Hamiltonian.[2] We have employed the Cholesky decomposition technique to reduce the size of the disk space.[3] The basis set in our calculations was taken from the ANO-RCC library implemented in the MOLCAS8.2 programme package. We have used the VTZP basis set for U, the VDZP basis set for the atoms in the first coordination sphere, and the VDZ basis set for the atoms from the secondary coordination sphere onwards. We have considered CAS(3,7) (three electrons in seven 5f orbitals) active space for our CASSCF calculations. Within this active space, we have computed the energies of 35 quartets and 112 doublets. All the quartets and doublets have been mixed by the RASSI-SO module to compute the energies of spin-orbit coupled states. Finally, the g tensors, QTM, TA-QTM, etc., have been calculated with the SINGLE_ANISO module of MOLCAS.

Table S1: The CASSCF/RASSI-SO/SINGLE_ANISO computed energies and g factors five ground KDs of **7**.

Energy (cm^{-1})	g_x	g_y	g_z
0.0	0.434	1.178	4.736
100.8	0.133	0.345	4.548
242.3	0.460	0.994	3.601
351.1	3.440	2.205	0.077
555.4	0.363	0.528	4.510

Table S2: The CASSCF/RASSI-SO/SINGLE_ANISO computed energies and g factors five ground KDs of **9**.

Energy (cm^{-1})	g_x	g_y	g_z
0.0	0.687	1.357	4.655
229.3	3.562	1.833	0.039
325.3	2.967	2.031	0.183
624.2	3.369	2.421	1.090
762.8	0.627	1.054	5.014

Table S3: The CASSCF/RASSI-SO/SINGLE_ANISO computed energies and g factors five ground KDs of **12**.

Energy (cm^{-1})	g_x	g_y	g_z
0.0	0.268	1.462	3.718
260.7	2.337	1.609	0.453
399.0	0.337	1.302	3.025

642.3	0.951	1.753	2.709
1135.6	2.933	2.567	1.771

Table S4: The CASSCF/RASSI-SO/SINGLE_ANISO computed energies and g factors five ground KDs of **39**.

Energy (cm^{-1})	g_x	g_y	g_z
0.0	0.400	0.424	4.826
180.2	0.513	0.748	4.073
199.6	1.455	2.030	3.277
432.6	2.886	2.833	0.359
693.3	2.949	2.934	1.381

1. F. Aquilante, J. Autschbach, R. K. Carlson, L. F. Chibotaru, M. G. Delcey, L. De Vico, I. Fdez. Galván, N. Ferré, L. M. Frutos and L. Gagliardi, Molcas 8: New capabilities for multiconfigurational quantum chemical calculations across the periodic table, *J. Comput. Chem.*, 2016, **37**, 506-541.
2. M. Reiher and A. Wolf, *Relativistic quantum chemistry: the fundamental theory of molecular science*, John Wiley & Sons, 2014.
3. F. Aquilante, P.-Å. Malmqvist, T. B. Pedersen, A. Ghosh and B. O. Roos, Cholesky decomposition-based multiconfiguration second-order perturbation theory (CD-CASPT2): application to the spin-state energetics of Co^{III} (diiminato)(NPh), *J. Chem. Theory Comput.*, 2008, **4**, 694-702.