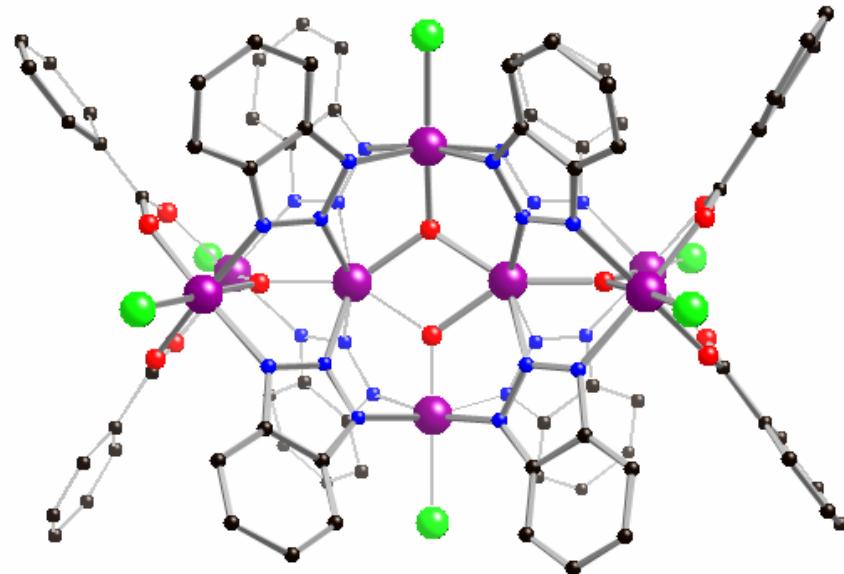
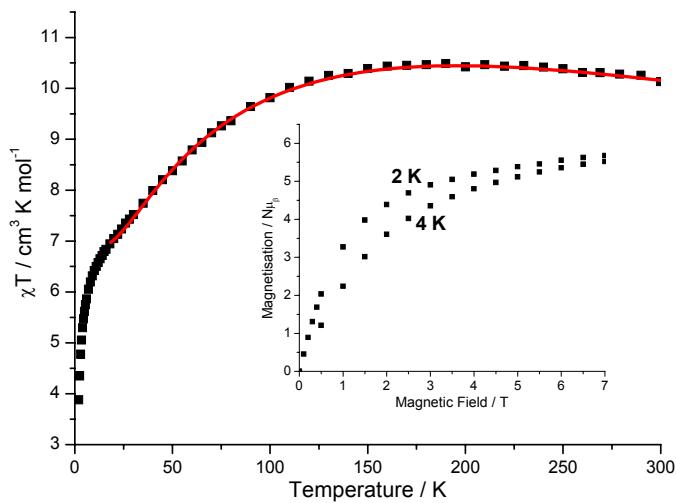


**Large spin, magnetically anisotropic octametallic vanadium(III) clusters with strong ferromagnetic coupling: Supplementary Information**

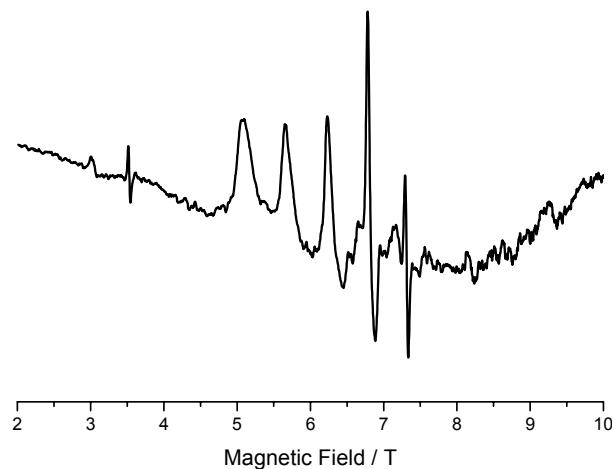
Rachel Shaw,<sup>a</sup> Floriana Tuna,<sup>a</sup> Wolfgang Wernsdorfer,<sup>b</sup> Anne-Laure Barra,<sup>c</sup> David Collison,\*<sup>a</sup> and Eric J. L. McInnes\*<sup>a</sup>



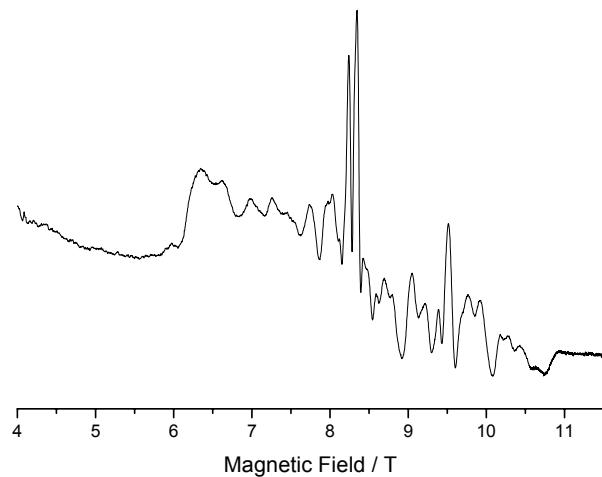
**Figure S1.** View of the structure of the dianion of **2**. Scheme: V (purple), O (red), N (blue), Cl (green), C (black), H omitted for clarity.



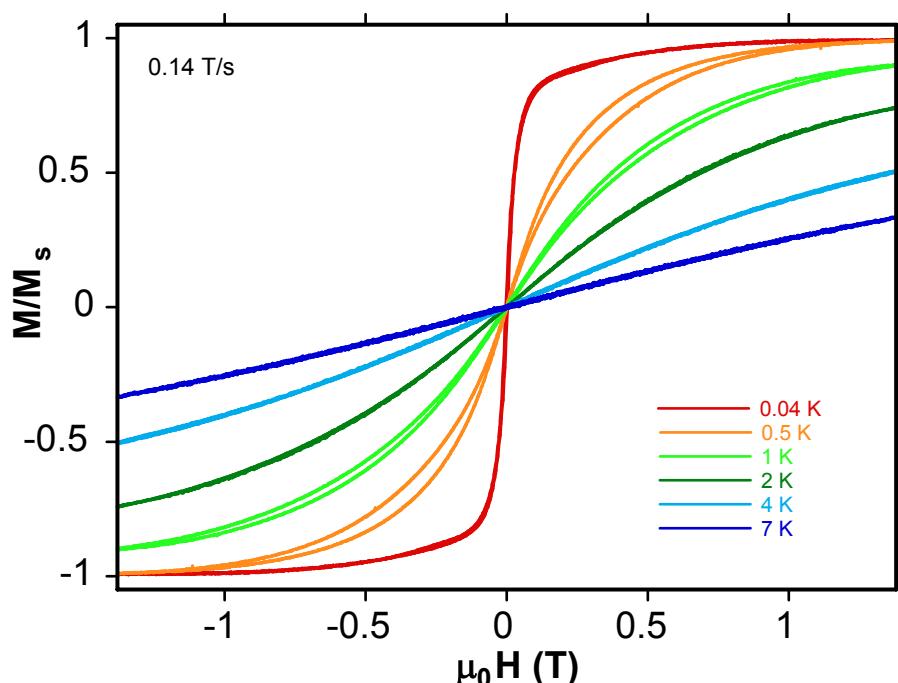
**Figure S2.**  $\chi T$  vs.  $T$  for **2** measured in 1 T applied magnetic field (black squares) and best fit down to 18 K (red line) using the coupling scheme in the insert of Fig. 2 and spin Hamiltonian (1) in the main text. Best fit parameters:  $J_1 = +118$ ,  $J_2 = -49$ ,  $J_3 = -6$  and  $J_4 = -11$  cm<sup>-1</sup> with  $g = 1.9$ . Inset: low temperature  $M$  vs.  $H$  for **2**.



**Figure S3.** 190 GHz EPR spectrum at 5 K of a loose polycrystalline sample of **1**, confirming the W-band observations in the main text.



**Figure S4.** 230 GHz EPR spectrum at 5 K of a polycrystalline sample of **1** restrained in eicosane wax. Note the very different form of the spectrum to that in Fig. S3, confirming that the crystallites orient in the field when loose.



**Figure S5.** Low temperature  $M$  vs.  $H$  ( $M$  normalised to saturation value  $M_s$ ) loops for a single crystal of **1** measured on a micro-SQUID array.

**Table S1.** Bond valence sum calculations for the vanadium ions in **1** and **2**, using parameters from N. E. Brese and M. O'Keefe, *Acta Cryst.* 1991, **B47**, 192-197.

	Complex 1				Complex 2			
	V(III)	V(IV)	V(V)	Assignment	V(III)	V(IV)	V(V)	Assignment
V1	3.21	3.35	3.46	V(III)	3.22	3.36	3.47	V(III)
V2	3.22	3.33	3.45	V(III)	3.29	3.40	3.53	V(III)
V3	3.32	3.44	3.57	V(III)	3.25	3.36	3.49	V(III)
V4	3.43	3.47	3.53	V(III)	3.42	3.46	3.52	V(III)