

**Table S1.** Single-ion principal components of the  $g$  matrix of the Mn(III) centres in **1** and **2** and orientations with respect to the crystallographic frame of reference  $XYZ$  by means of Euler angles.

	$g_{xx}$	$g_{yy}$	$g_{zz}$	$\alpha/^\circ$	$\beta/^\circ$	$\gamma/^\circ$
Mn1 <b>(1)</b>	1.978	1.992	1.999	60.10	100.64	345.98
Mn1 <b>(2)</b>	1.977	1.990	1.998	83.97	89.70	315.67
Mn2 <b>(1)</b>	1.982	1.994	1.998	38.01	141.29	144.04
Mn2 <b>(2)</b>	1.978	1.992	1.998	144.19	117.42	340.11
Mn3 <b>(1)</b>	1.980	1.992	1.998	281.22	132.66	31.42
Mn3 <b>(2)</b>	1.978	1.993	1.998	210.52	125.14	6.02

**Table S2.** Angles, in degrees, between the Jahn-Teller orientation and the AOM calculated  $D_{ZZ}$  component of the anisotropy tensor for the Mn(III) sites Mn1, Mn2, and Mn3, for compounds **1** and **2**.

	Mn1	Mn2	Mn3
<b>1</b>	4°	7°	4°
<b>2</b>	15°	14°	13°

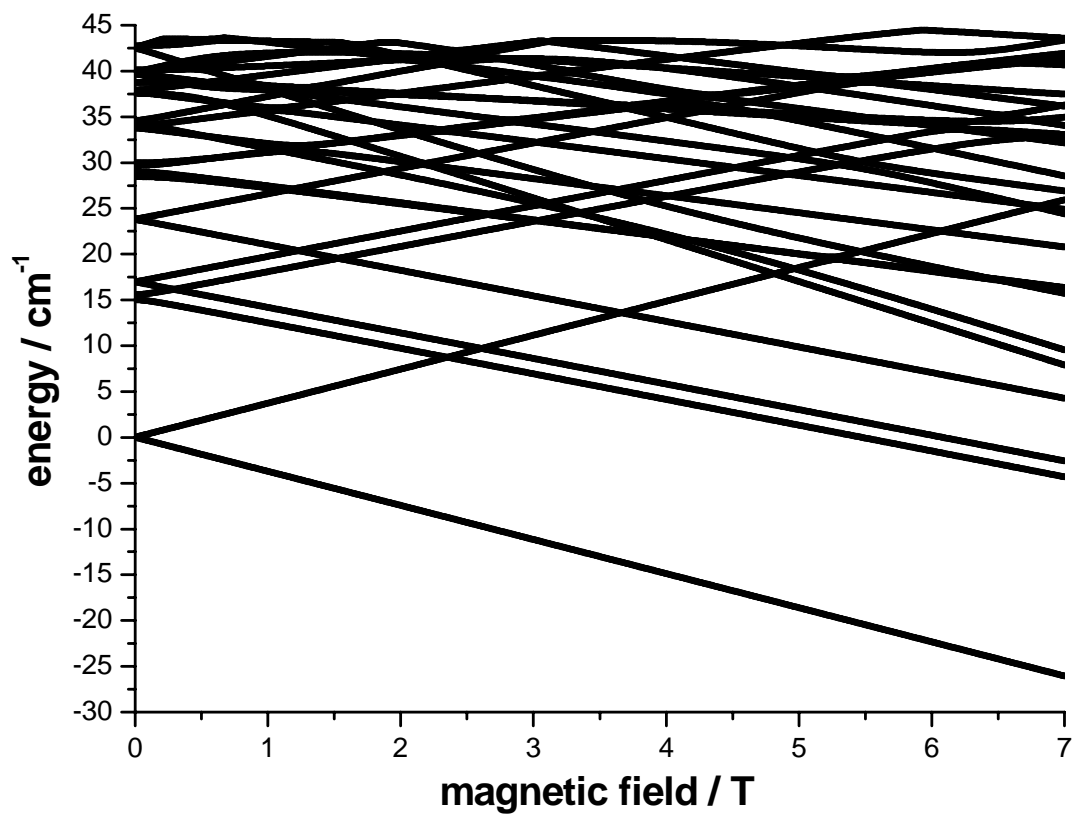


Figure S1. Field dependence of the eigenvalues of the twenty eight lowest lying eigenstates of **1** calculated as described in the text. The magnetic field is applied along the quantization axis (Z-axis).

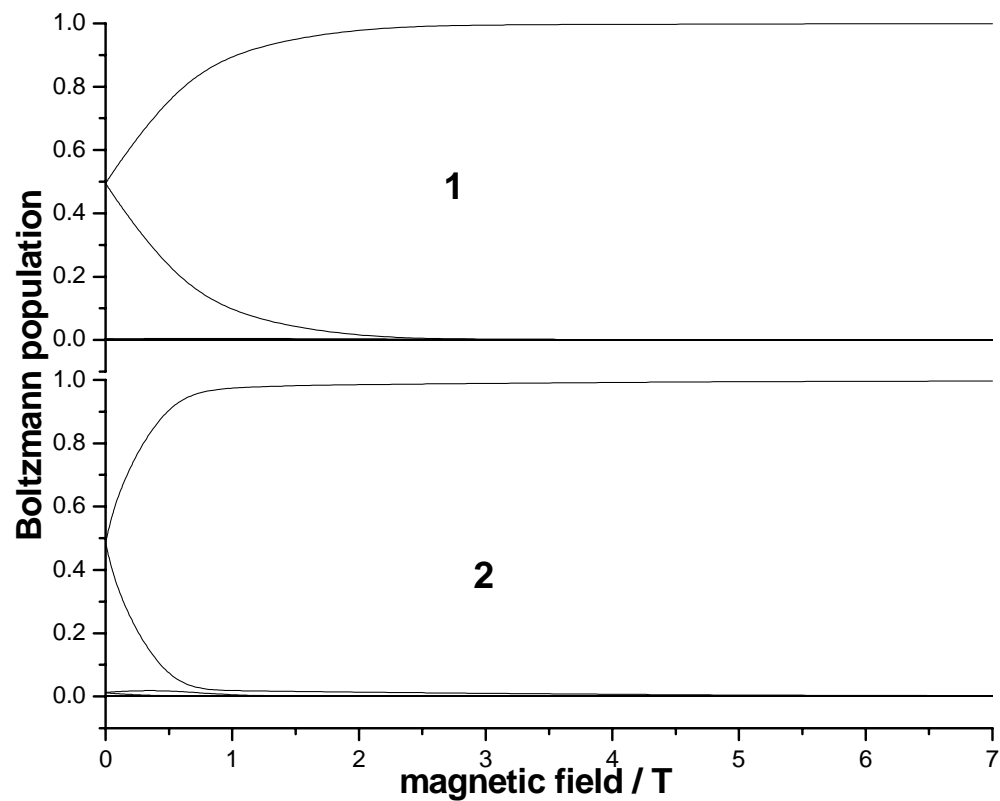


Figure S2. Field dependence of the Boltzmann population of the twenty eight lowest lying eigenstates of **1** (top) and of the forty eight eigenstates of **2** (bottom) determined by diagonalisation of the anisotropic spin-Hamiltonian (5) at 4.2 K.

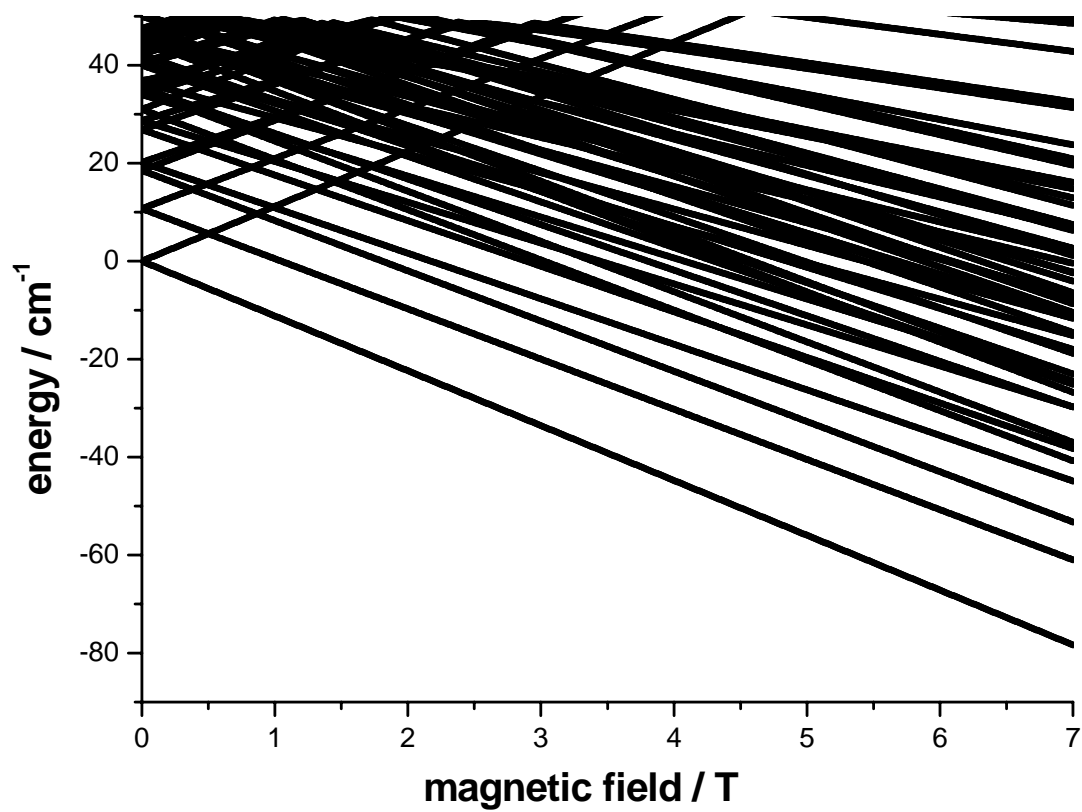


Figure S3. Field dependence of the eigenvalues of the forty eight lowest lying eigenstates of **2** calculated as described in the text. The magnetic field is applied along the quantization axis (Z-axis)