Table S1 Selected interatomic distances d /pm for

$$(CH_3NH_3)_2[Gd_2(CCl_3COO)_6(H_2O)_6](CCl_3COO)_2 \cdot 2CCl_3COOH \ (\mathbf{1}) \text{ and}$$

$$(NH_3CH_3)_2[Gd_4(OH)_4(CCl_3COO)_{10}(H_2O)_6] \cdot 2H_2O \ (\mathbf{2})$$

(1)		d /pm	(2)		d /pm
Gd1	–O11	238.4(4)	Gd1	–O1	239.3(8)
	–O12[a)]	237.1(4)		–O2	238.8(9)
	–O21	239.5(5)		–O4	237.1(8)
	–O21[a)]	290.9(4)		–O01	238.9(8)
	–O22[a)]	247.5(4)		–O12	240.7(12)
	–O31	233.2(4)		–O41	244.8(11)
	–O1W	243.3(4)		–O71	234.7(9)
	–O2W	239.4(4)		–O1W	246.9(10)
	–O3W	248.2(5)	Gd2	–O2	244.2(8)
	–Gd1[a)]	420.2(3)		–O3	236.2(9)
N1	–O22[b)]	294.4(3)		–O4	237.6(9)
	–O41[c)]	295.3(3)		–O42	234.9(11)
	–O42[d)]	280.6(1)		–O81	232.9(13)
	–O51[e)]	307.8(3)		–O3W	244.8(10)
	–O2W[c)]	311.2(2)		–O4W	241.7(11)
C11	–O11	124.1(7)		–O6W	242.5(12)
	–O12	123.6(7)	Gd3	–O1	239.2(9)
C21	–O21	123.9(8)		–O2	241.8(8)
	–O22	124.0(7)		–O3	242.9(8)
C31	–O31	123.8(7)		–O02	250.3(10)
	–O32	122.3(7)		–O21	243.4(11)
C41	–O41	122.0(9)		–O62	251.4(10)
	–O42	124.9(9)		–O82	241.6(13)
C51	–O51	118.9(8)		–O91	239.7(10)
	–O52	129.3(9)		–O2W	249.7(8)
			Gd4	–O1	240.1(8)
				–O3	238.7(9)
				–O4	239.4(9)
				–O11	236.0(11)
				–O31	233.3(10)
				–O51	232.3(9)
				–O61	240.9(9)
				–O5W	249.5(13)
			Gd1	–Gd2	382.6(1)
				–Gd3	387.3(1)
				–Gd4	382.2(1)
			Gd2	–Gd3	385.3(1)
				–Gd4	380.3(1)
			Gd3	–Gd4	388.0(1)
			N1	–O22[c)]	280.3(2)
				–O72	279.4(3)
				–O7W	284.0(3)
			N2	–O02	289.2(3)
				–O62	282.3(3)
				–O2W	284.2(3)

Symmetry transformations used to generate atoms:

[a)]–x,–y,–z+1; [b)]–x,–y+1,–z+1; [c)]x,y+1,z; [d)]–x+1,–y+1,-z; [e)]–x+1,–y+2,–z

Fig. S1 Comparison of measured ($\bigcirc$,$H^{(\mathrm{ir})}$:500 Oe; Δ, $H^{(\mathrm{ir})}$:1000 Oe) and calculated (—) effective Bohr magneton numbers for **1**.
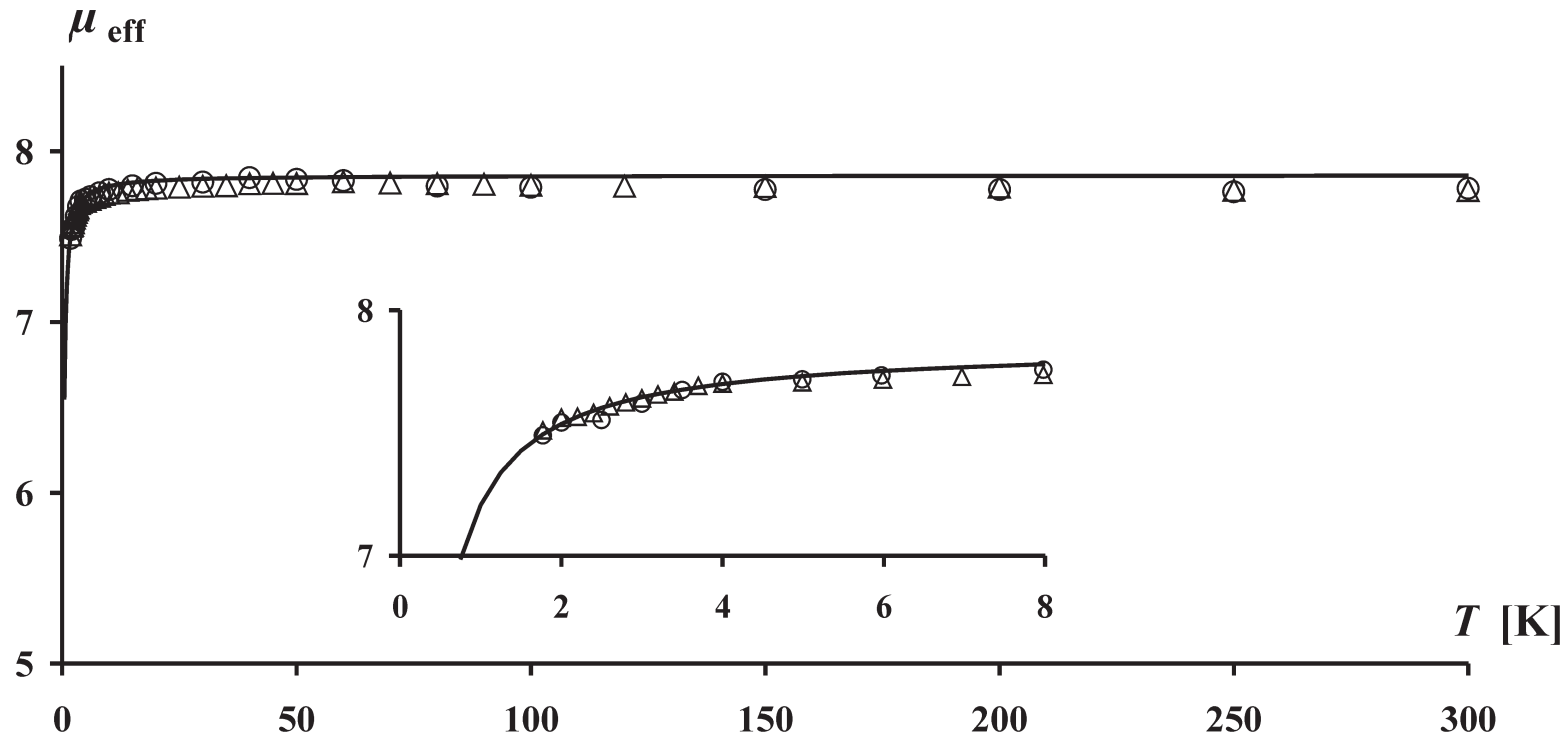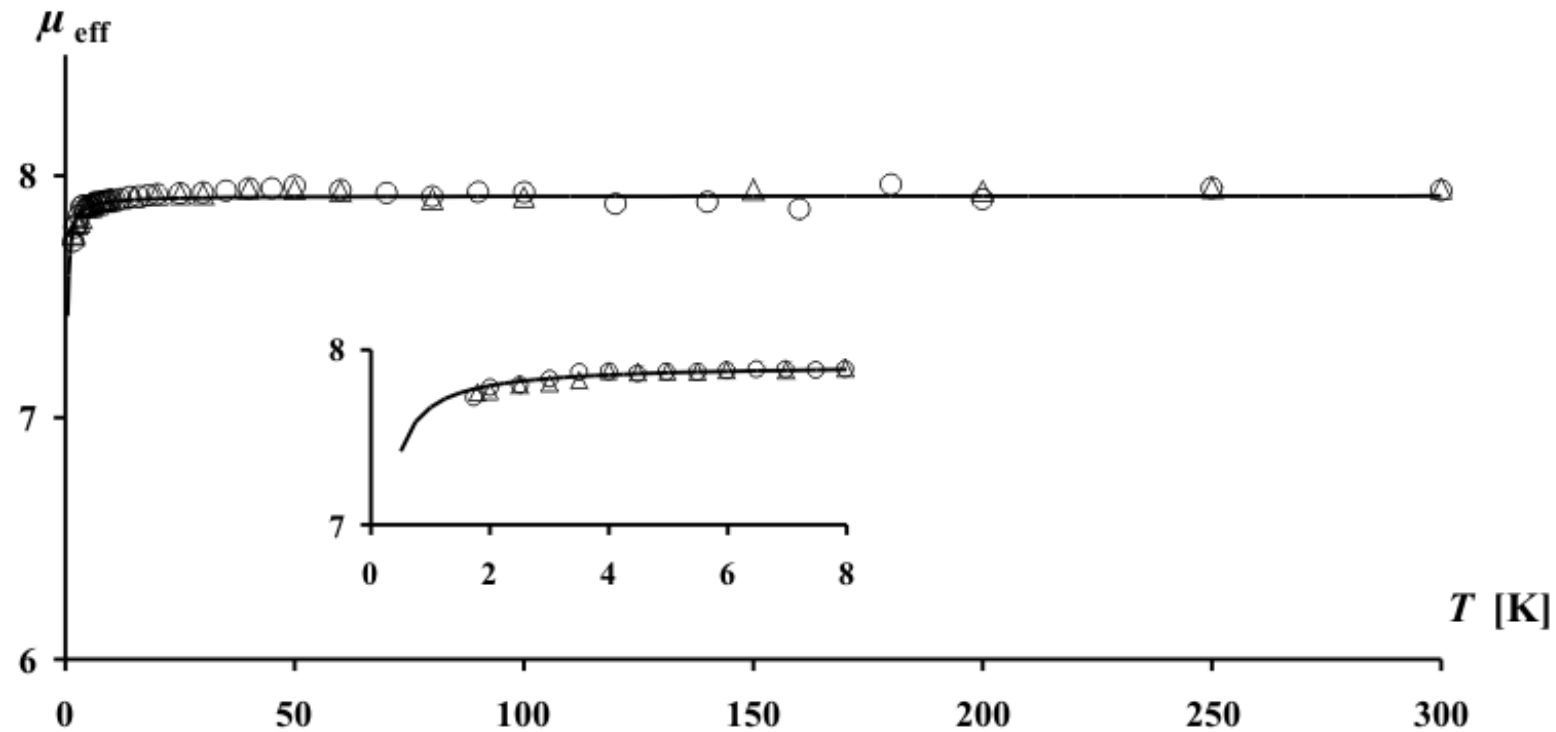
Fig. S2 Comparison of measured ($\bigcirc$, $H^{(ir)}$:500 Oe; $\triangle$, $H^{(ir)}$:1000 Oe) and calculated (—) effective Bohr magneton numbers for **3**.