Electronic Supplementary Material (ESI) for RSC Advances.

ELECTRONIC SUPPLEMENTARY INFORMATION

to

Two unique star-like [$Mn^{IV}Mn^{III}_2Ln^{III}$] clusters: magnetic relaxation phenomena†

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SYNTHETIC DETAILS

All manipulations were performed under aerobic conditions using materials (reagent grade) and solvents as received.

General synthetic strategy applicable to **1**-**2**:

$Mn(ClO_4)_2{\cdot}6H_2O$ (120 mg, 0.33 mmol), $Gd(NO_3)_3{\cdot}6H_2O$ (0.33 mmol, 149 mg), H_3L^2 (90 mg, 0.33 mmol), NH_4SCN (76 mg, 1 mmol), and NEt_3 (~ 1 mmol) were dissolved in MeOH (~ 30 mL) forming a yellow suspension that was left upon stirring for ~45' to yield a brown solution. The solution was then filtered and left undisturbed to evaporate at room temperature. Dark-brown single-crystals suitable for X-ray crystallography were formed (for the Gd analogue, **1**) after ~ 4 days in ~35% yield, and they were washed with Et_2O (2 x 5 ml) and dried in air. Elemental Anal. calcd (found) for **1**: C 52.21 (52.32), H 4.49 (4.32), N 4.29 (4.18).

For **2**, the same exactly procedure was followed as in the case of **1**, with the use of $Dy(NO_3)_6{\cdot}6H_2O$ (0.33 mmol, 150 mg) instead of $Gd(NO_3)_3{\cdot}6H_2O$. Dark-brown crystals were formed after ~ 4 days in ~30% yield, and they were washed with Et_2O (2 x 5ml) and dried in air. Elemental Anal. calcd (found) for **2**: C 52.05 (52.12), H 4.48 (4.26), N 4.28 (4.16) %.

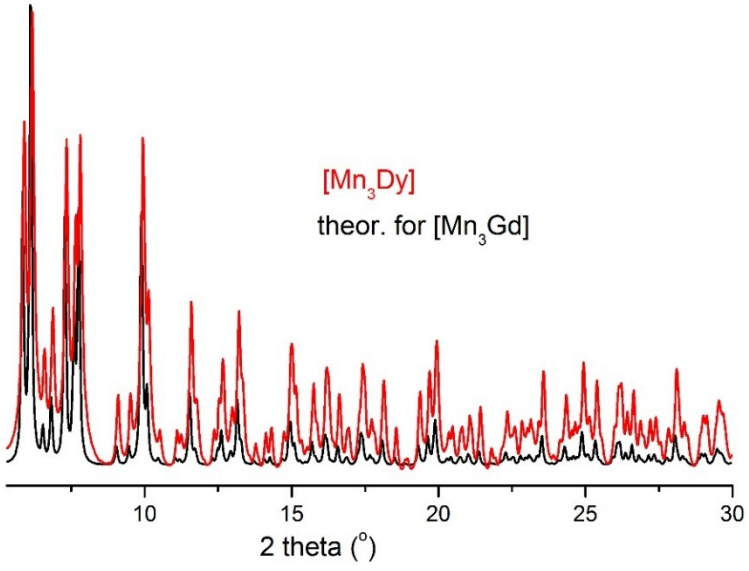

Fig. S1 Experimental PXRD pattern for the Dy analogue (red) compared with the theoretical pXRD pattern of the Gd analogue (black).

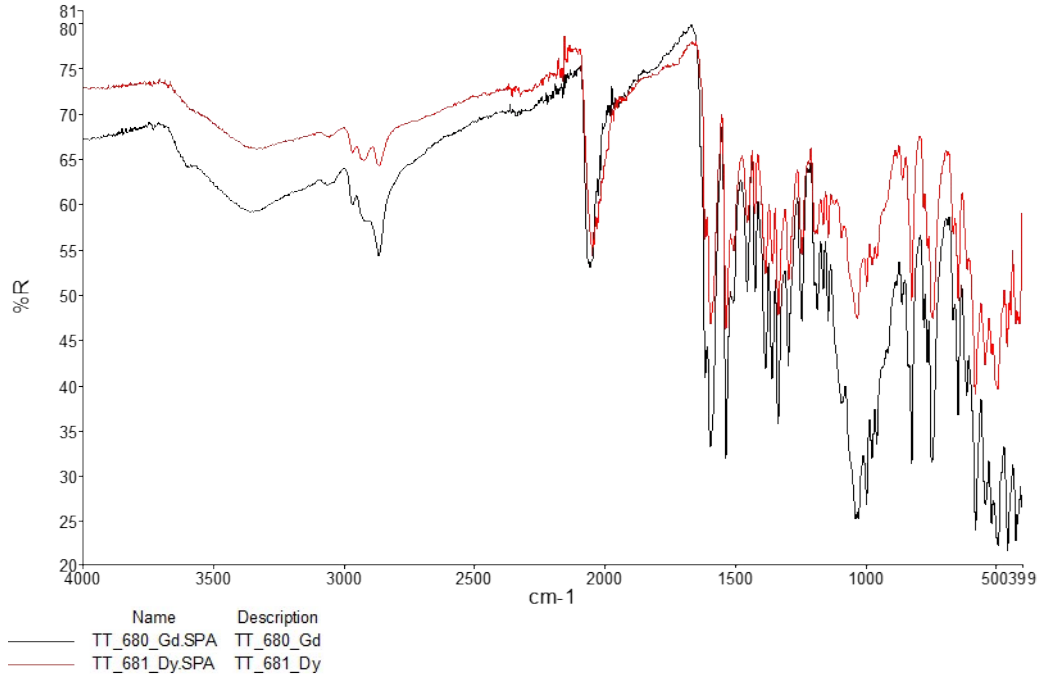

Fig. S2 IR spectra comparison for the Dy (black line) and Gd (red line) analogues.

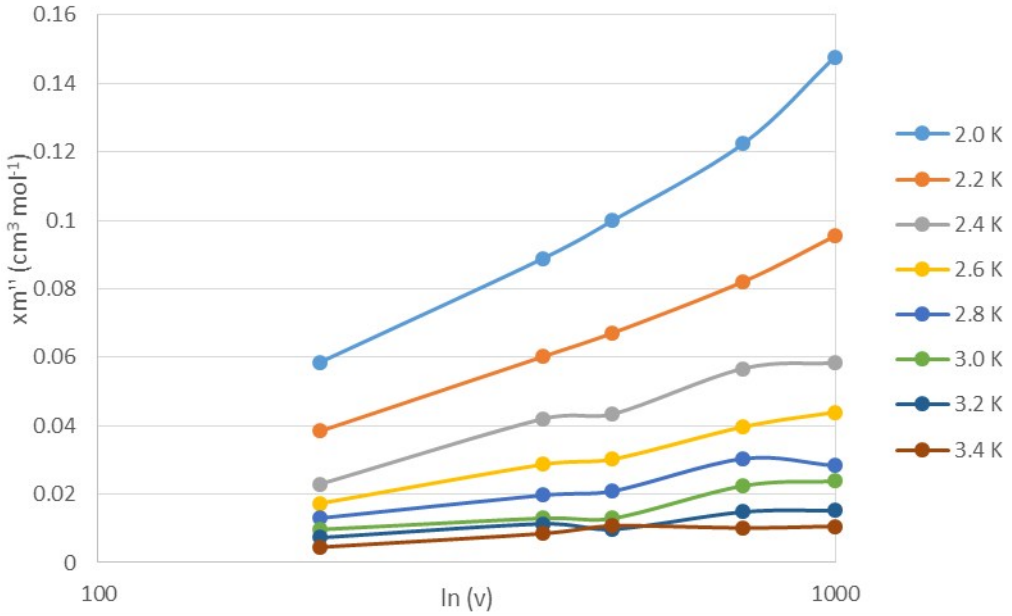

Fig. S3 Plot of the out-of-phase χ_M'' signals *vs.* ln(ν) for complex **2** at the indicated temperatures

OP-8	D8h Octagon	28.898
HPY-8	C7v Heptagonal pyramid	21.922
HPY-8	D6h Hexagonal bipyramid	14.454
CU-8	Oh Cube	8.926
SAPR-8	**D4d Square antiprism**	**1.510**
TDD-8	D2d Triangular dodecahedron	1.744
JGBF-8	D2d Triangular dodecahedron	14.422
J14	-	24.322
JBTPR-8	C2v Biaugmented trigonal prism J50	2.577

Table S1. SHAPE analysis of the lanthanide center in complex **1**.