Supporting Information File

Observation of Large Magnetocaloric Effect in a 2D Gd(III) based Coordination Polymer

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Synthesis of $[Gd(C_4O_4)(OH)(H_2O)_4]_n$: 3ml 0.1(M) methanolic solution of $Gd(NO_3).6H_2O$ was carefully layered above the 3ml 0.1(M) aqueous solution of lithium squarate in a layering tube for slow solvent diffusion. The tube was gently kept at room temperature for few weeks. Colourless shiny diffraction quality needle like crystals were appeared at the junction. Selected IR data(KBr pellet): 3191(w), 2971(s) (NH), 1719 (m), 1468(m), 1259(m), 1098(m), 998(m), 972(m), 840(m),738 (s)cm-1; Selected Raman data: 343(vs), 434(s), 657(w), 961(m), 1042(m), 1426(m), 1851(m).(Fig. S7)

X-ray Crystallography. X-ray crystallographic data were collected at 140K on a Bruker Apex II CCD diffractometer using graphite monochromated MoKα (λ =0.71073) radiation. Data collection was performed using φ and ω scan. The data reduction and cell refinement was performed with the SAINT program and adsorption correction was done using SADABS. The structure was solved using direct methods followed by full matrix least square refinements against F^2(all data HKLF 4 format) using SHELXTL. In anisotropic refinement few of the atoms become NPD. So, they were refined isotropically. This could be due to unsuitable adsorption correction of the needle like crystal. We are unable to locate the position of hydrogen atoms of the coordinated water molecules.

Physiscal Measurements. Elemental analysis was performed on an Elementar vario Micro Cube instrument. IR spectrum was recorded on a Perkin-Elmer spectrometer from KBr pellets. Raman spectra was recorded on a Jovin-Yvon Labram Raman spectrometer.

Magnetic Measurements. Magnetic measurements were performed using a Quantum Design PPMS set-up. The measured values were corrected for the experimentally measured contribution of the sample holder, while the derived susceptibilities were corrected for the diamagnetism of the samples, estimated from Pascal's tables[(12)].

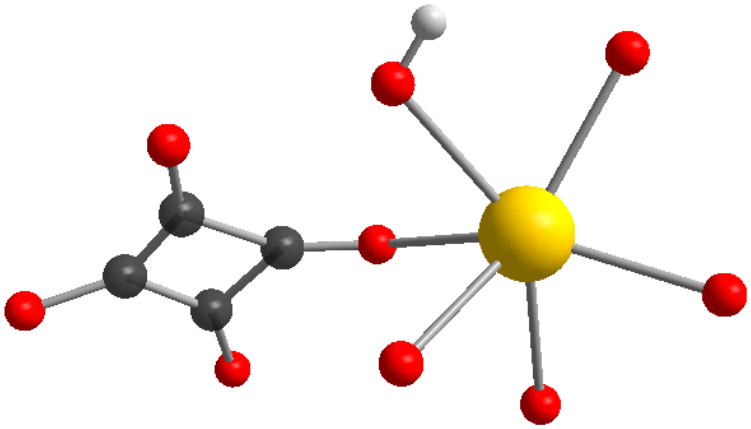

Fig. S1. Asymmetric unit of the compound. Colour code, yellow: Gadolinium, red: Oxygen, grey: Carbon, light grey: Hydrogen

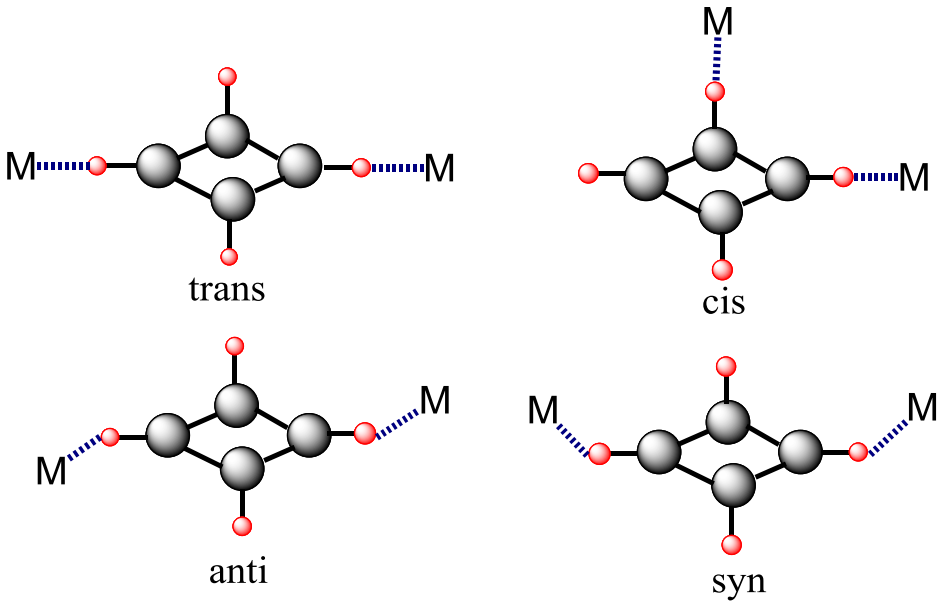

Fig. S2 Different bridging modes of the squarate ligand.

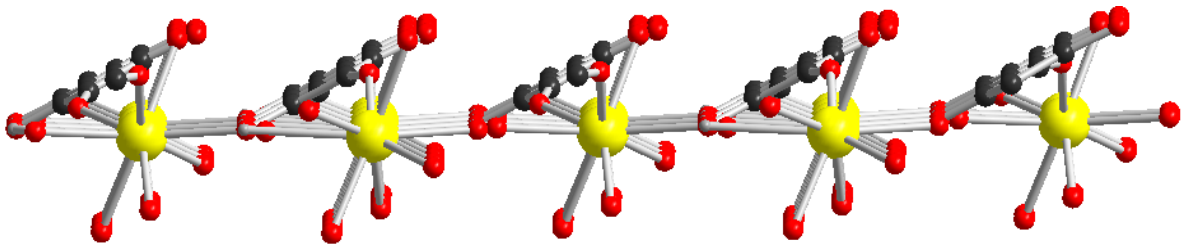

Fig. S3. View along *b*-axis of the compound **1**.

Table S1. Hydrogen bonding distances (Å) and angles (deg) for the compound **1.**

D-H….A <D-H-A(deg)	Symmetry operation	D-H(Å)	A…H(Å)	D….A(Å)
O6-H6-O8 153.89	x,-y+1/2+1,+z-1/2	0.820	2.504	3.261
O6-H6-O3 107.89	x,+y+1,+z	0.820	2.384	2.749
O6-H6-O2 123.33	x,-y+1/2,+z-1/2	0.820	2.343	2.846

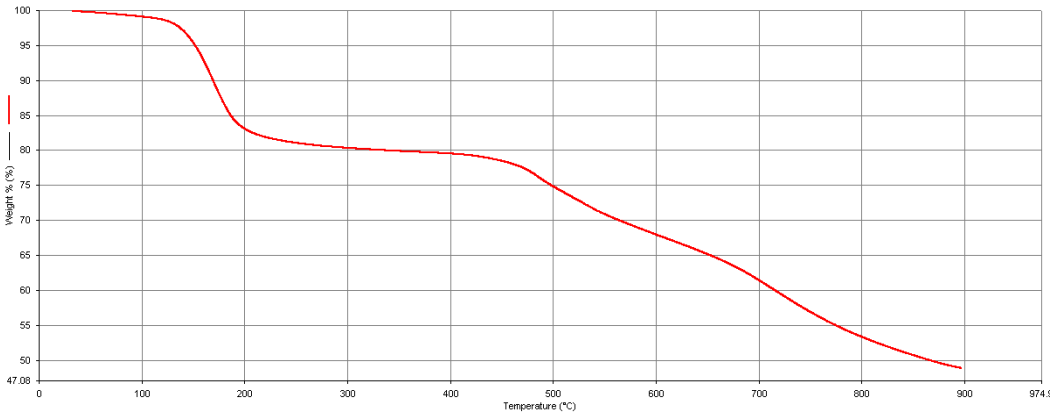

Fig. S4. Thermogravimetric data for $[Gd(C_4O_4)(OH)(H_2O)_4]_n$

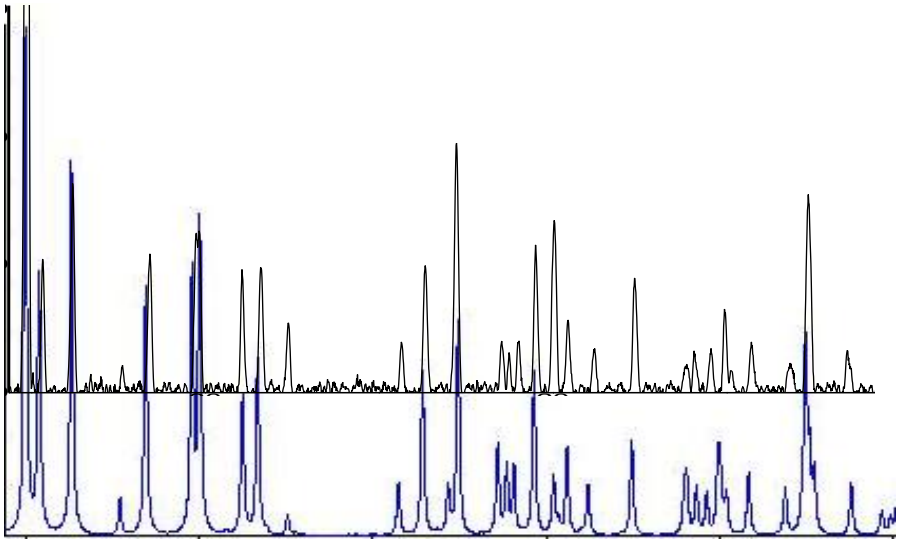

Fig. S5. Experimental (black) and simulated (blue) powder XRD data for $[Gd(C_4O_4)(OH)(H_2O)_4]_n$.

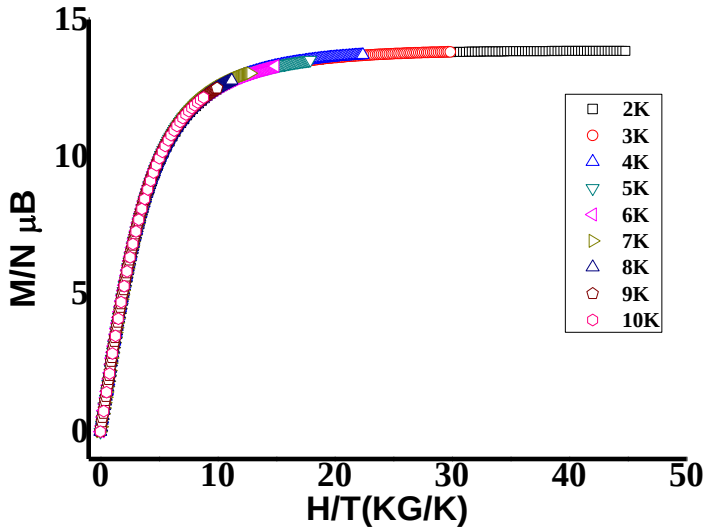

Fig. S6. Isothermal normalized magnetizations vs. field/temperature plot for two Gd^{3+} ions, collected for temperatures ranging from 2 to 10 K.

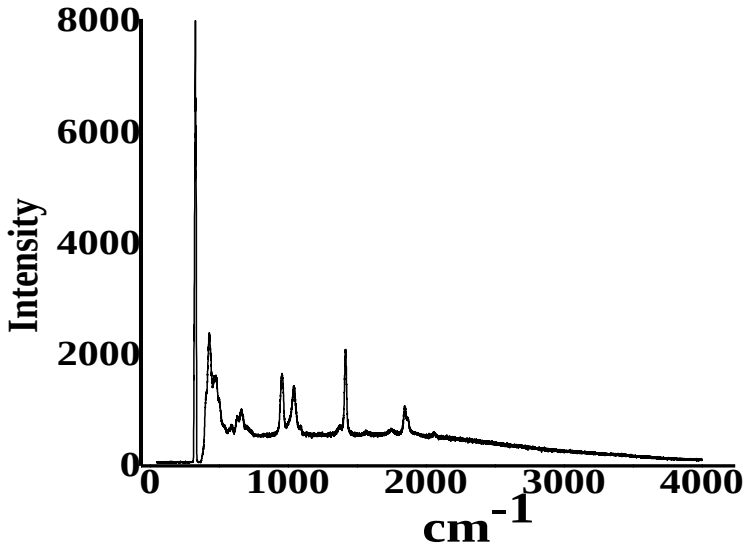

Fig. S7. Raman spectrum of $[Gd(C_4O_4)(OH)(H_2O)_4]_n$.