Supporting Information

A Layered Hybrid Rare-Earth Double Perovskite with Two Continuous Reversible Phase Transitions Induced By Unusual Two Driving Gears of Fan-Like Rotation Movements

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Table S1 Hydrogen Bonds for **1** at 233 K

D-H-A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
N5-H5-O2[1]	0.98	2.32	3.101(5)	135.6
N5-H5-O3[1]	0.98	2.01	2.963(6)	164.7
N6-H6-O1[2]	0.98	2.00	2.941(5)	159.2

N6-H6-O2[2]	0.98	2.30	3.043(5)	132.2

Symmetric code: [1]-1/2+X,1-Y,+Z; [2]+X,1+Y,+Z

Table S2 Hydrogen Bonds for **1** at 273 K

D-H-A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
N5-H5-O1	0.91	2.14	3.002(8)	157.6
N5-H5-O2	0.91	2.25	3.024(8)	142.6
N6-H6-O2	0.91	2.35	3.070(7)	135.6
N6-H6-O3	0.91	2.10	2.986(8)	165.0

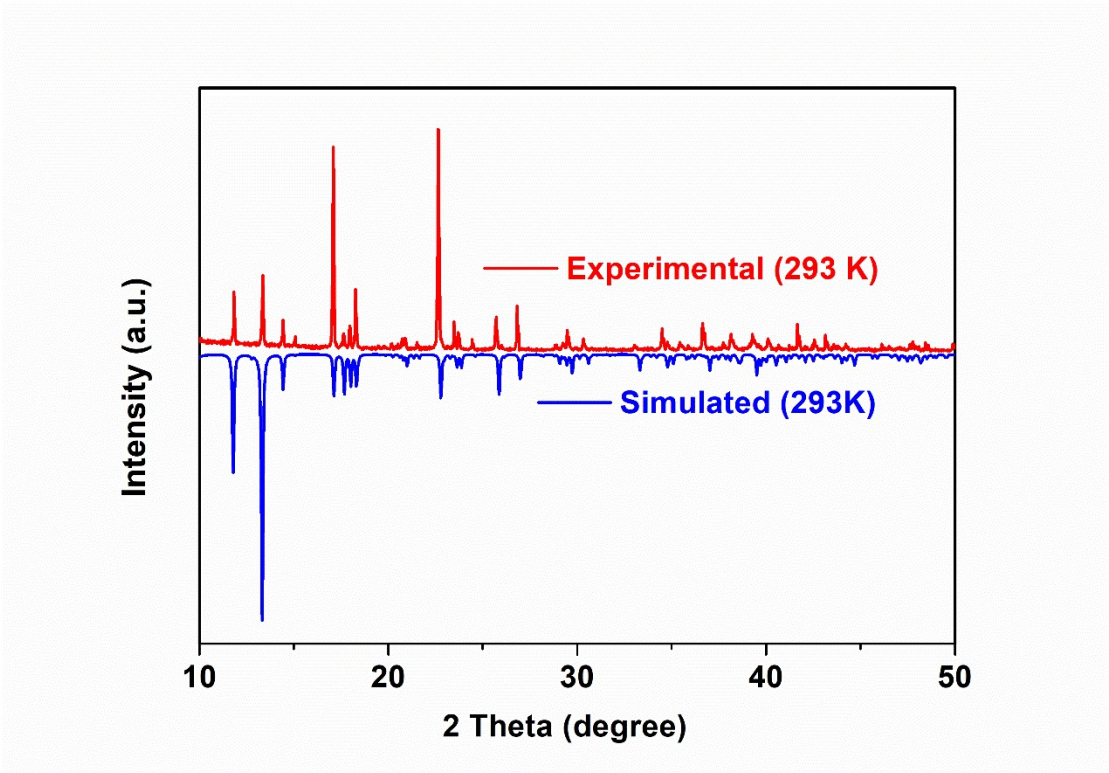

Figure S1 The simulated and experimental PXRD of **1** at 293 K

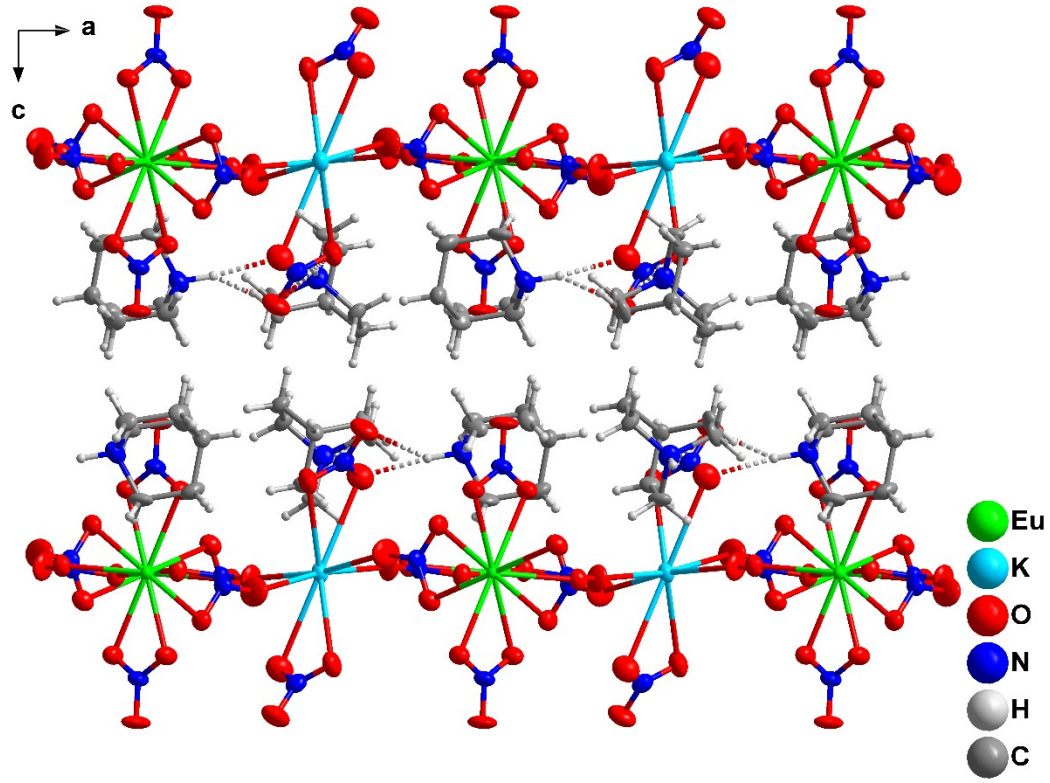

Figure S2 The crystal packing diagram of **1** along *b* axis at 233 K.

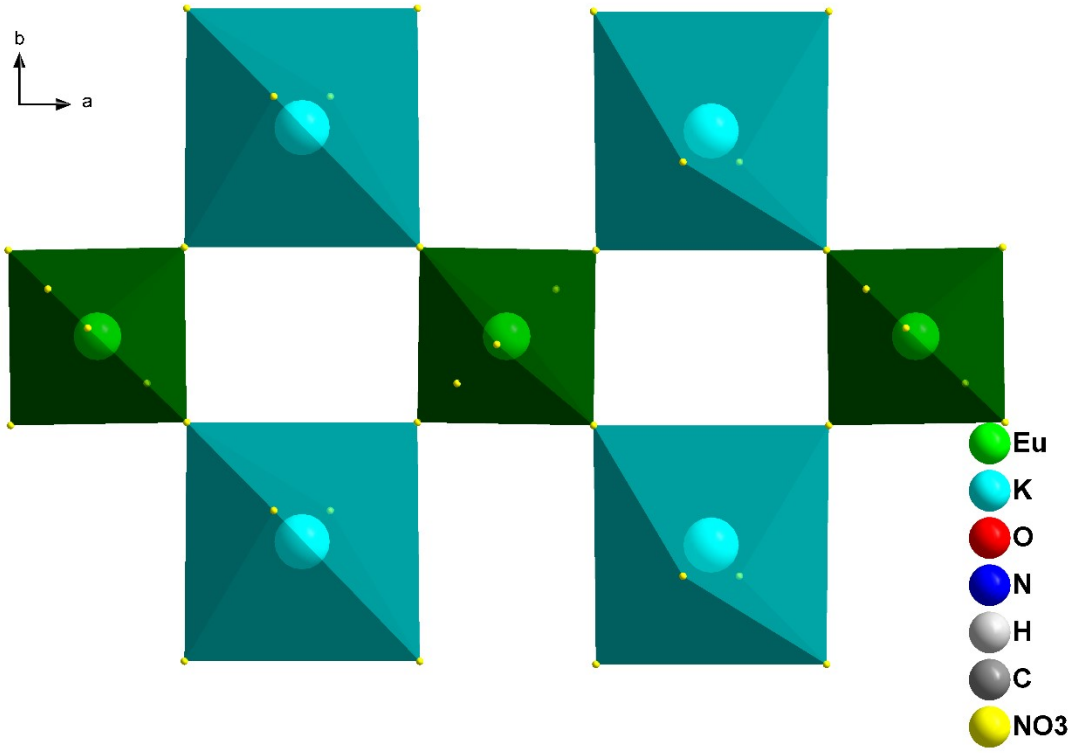

Figure S3 The inorganic framework of **1** at 233 K

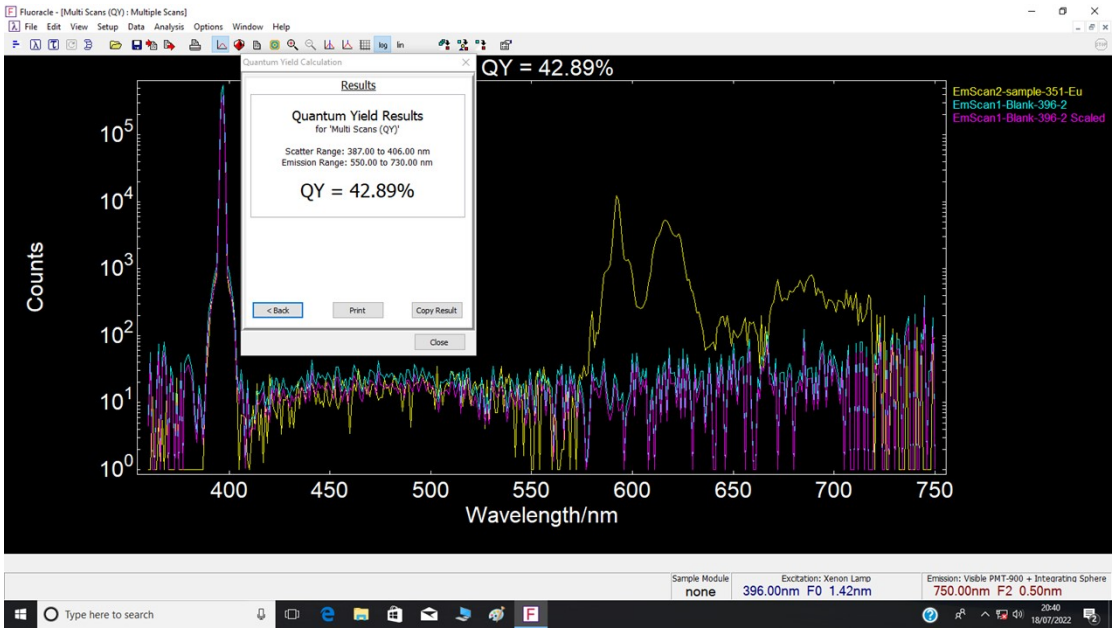

Figure S4 The absolute quantum yield (QY) of **1** in the solid state using an integrating sphere at room temperature.