Synthesis, Properties, Magnetism-Structure Relation of Lanthanide-based Metal-Organic Frameworks with (Ethylenedithio) Acetic Acid

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1. Bond length and bond angel of compounds 1-5

Table S1 Bond lengths and angle of compounds 1-5

Compound 1			
La(1)-O(4)	2.520(2)	La(1)-O(2W)	2.535(3)
La(1)-O(1)#1	2.530(2)	La(1)-O(1W)	2.553(3)
La(1)-O(3)#2	2.564(2)	La(1)-O(6)	2.616(2)
La(1)-O(2)#3	2.596(2)	La(1)-O(5)	2.639(3)
La(1)-O(1)#3	2.747(2)	La(1)-O(4)#2	2.912(3)
O(4)-La(1)-O(1)#1	148.91(8)	O(1)#1-La(1)-O(2W)	139.14(8)
O(4)-La(1)-O(2W)	71.82(9)	O(4)-La(1)-O(1W)	138.53(9)
O(1)#1-La(1)-O(1W)	72.54(9)	O(4)-La(1)-O(3)#2	108.67(8)
O(2W)-La(1)-O(1W)	67.00(8)	O(1)#1-La(1)-O(3)#2	76.27(8)
O(2W)-La(1)-O(3)#2	87.94(9)	O(4)-La(1)-O(2)#3	79.35(8)
O(1W)-La(1)-O(3)#2	75.02(10)	O(1)#1-La(1)-O(2)#3	108.74(7)
O(2W)-La(1)-O(2)#3	73.20(9)	O(3)#2-La(1)-O(2)#3	156.36(10)
O(1W)-La(1)-O(2)#3	84.37(10)	O(4)-La(1)-O(6)	74.40(8)
O(1)#1-La(1)-O(6)	77.30(8)	O(1W)-La(1)-O(6)	142.20(9)
O(2W)-La(1)-O(6)	135.38(8)	O(3)#2-La(1)-O(6)	76.12(8)
Symmetry code: #1 x,y,z-1; #2 -x+3,-y+1,-z-1; #3 -x+2,-y+1,-z;			
Compound 2			
Nd(1)-O(3)	2.439(3)	Nd(1)-O(1W)	2.496(3)
Nd(1)-O(2)#1	2.468(3)	Nd(1)-O(1)#3	2.531(3)
Nd(1)-O(2W)	2.486(3)	Nd(1)-O(5)	2.563(3)
Nd(1)-O(4)#2	2.491(3)	Nd(1)-O(6)	2.586(3)
Nd(1)-O(2)#3	2.735(3)		
O(3)-Nd(1)-O(2)#1	148.59(10)	O(2W)-Nd(1)-O(1W)	67.68(10)
O(3)-Nd(1)-O(2W)	71.60(10)	O(4)#2-Nd(1)-O(1W)	76.15(11)
O(2)#1-Nd(1)-O(2W)	139.51(9)	O(3)-Nd(1)-O(1)#3	79.37(10)
O(3)-Nd(1)-O(4)#2	106.39(9)	O(2)#1-Nd(1)-O(1)#3	109.43(8)
O(2)#1-Nd(1)-O(4)#2	78.20(9)	O(2W)-Nd(1)-O(1)#3	74.14(10)

Symmetry code: #1 x,y,z-1; #2 -x+2,-y,-z; #3 -x+1,-y,-z+1;

Compound 3

Eu(1)-O(2)#1	2.356(4)	Eu(1)-O(4)	2.472(4)
Eu(1)-O(3)#2	2.406(4)	Eu(1)-O(6)	2.512(4)
Eu(1)-O(1W)	2.422(5)	Eu(1)-O(5)	2.530(4)
Eu(1)-O(2W)	2.422(5)	Eu(1)-O(3)	2.735(4)
Eu(1)-O(1)#3	2.433(4)		
O(2)#1-Eu(1)-O(3)#2	148.14(15)	O(2)#1-Eu(1)-O(4)	79.53(14)
O(2)#1-Eu(1)-O(1W)	139.05(18)	O(3)#2-Eu(1)-O(4)	110.07(13)
O(3)#2-Eu(1)-O(1W)	72.81(16)	O(1W)-Eu(1)-O(4)	83.98(17)
O(2)#1-Eu(1)-O(2W)	71.37(18)	O(2W)-Eu(1)-O(4)	75.08(16)
O(3)#2-Eu(1)-O(2W)	139.93(17)	O(1)#3-Eu(1)-O(4)	154.97(15)
O(1W)-Eu(1)-O(2W)	68.22(19)	O(2)#1-Eu(1)-O(6)	73.15(14)
O(2)#1-Eu(1)-O(1)#3	103.91(14)	O(3)#2-Eu(1)-O(6)	78.10(13)
O(3)#2-Eu(1)-O(1)#3	80.33(13)	O(1W)-Eu(1)-O(6)	141.67(16)

Symmetry code: #1 -x+1,-y+1,-z+2; #2 -x+1,-y+1,-z+1; #3 x+1,y,z-1

Compound 4

Gd(1)-O(2)#1	2.329(3)	Gd(1)-O(4)	2.457(3)
Gd(1)-O(3)#2	2.392(3)	Gd(1)-O(6)	2.505(3)
Gd(1)-O(1W)	2.403(3)	Gd(1)-O(5)	2.530(3)
Gd(1)-O(1)#3	2.414(3)	Gd(1)-O(3)	2.732(3)
Gd(1)-O(2W)	2.422(3)		
O(2)#1-Gd(1)-O(3)#2	147.79(10)	O(2)#1-Gd(1)-O(4)	79.63(11)
O(2)#1-Gd(1)-O(1W)	139.46(12)	O(3)#2-Gd(1)-O(4)	110.28(9)
O(3)#2-Gd(1)-O(1W)	72.75(11)	O(1W)-Gd(1)-O(4)	83.84(12)
O(2)#1-Gd(1)-O(1)#3	102.99(11)	O(1)#3-Gd(1)-O(4)	154.60(11)
O(3)#2-Gd(1)-O(1)#3	81.18(9)	O(2W)-Gd(1)-O(4)	75.01(11)
O(1W)-Gd(1)-O(1)#3	77.99(12)	O(2)#1-Gd(1)-O(6)	72.67(10)
O(2)#1-Gd(1)-O(2W)	71.52(12)	O(3)#2-Gd(1)-O(6)	78.20(9)

Symmetry code: #1 -x+1,-y+1,-z+1; #2 -x+1,-y+1,-z; #3 x+1,y,z-1

Compound 5

La(1)-O(5)	2.479(3)	La(1)-O(1W)	2.606(3)
La(1)-O(4)#1	2.500(3)	La(1)-O(6)#3	2.607(3)
La(1)-O(1)	2.545(3)	La(1)-O(5)#3	2.681(3)
La(1)-O(2W)	2.563(4)	La(1)-O(2)#3	2.700(3)
La(1)-O(3)#2	2.566(3)	La(1)-O(4)#2	2.836(3)
O(5)-La(1)-O(4)#1	144.92(9)	O(2W)-La(1)-O(3)#2	72.81(11)
O(5)-La(1)-O(1)	69.83(9)	O(5)-La(1)-O(1W)	136.06(10)
O(4)#1-La(1)-O(1)	145.24(9)	O(4)#1-La(1)-O(1W)	77.11(10)
O(5)-La(1)-O(2W)	69.98(11)	O(1)-La(1)-O(1W)	69.88(10)
O(4)#1-La(1)-O(2W)	78.65(11)	O(2W)-La(1)-O(1W)	130.88(12)
O(1)-La(1)-O(2W)	132.07(11)	O(3)#2-La(1)-O(1W)	76.70(11)
O(5)-La(1)-O(3)#2	75.27(9)	O(5)-La(1)-O(6)#3	119.36(9)
O(4)#1-La(1)-O(3)#2	110.43(9)	O(4)#1-La(1)-O(6)#3	78.35(9)

O(1)-La(1)-O(3)#2	72.65(10)	O(1)-La(1)-O(6)#3	80.03(10)

Symmetry code: #1 x,y,z-1 #2 -x-2,-y-2,-z #3 -x-2,-y-3,-z-1

2. Our QMC fitting program

Algorithm of our fitting Program: The quantum Monte Carlo calculations were performed by our fitting program, where we call LOOP module of ALPS and utilize four parameters (*J*, *g*, *zJ* and *TIP*) to further fit the experimental result by use of exhaustive-iterative method. The corresponding formulas were shown in Eq(1)-Eq(4). The running procedure of our program is presented in the Figure S1. In the beginning, the range of four parameters (*J*, *g*, *TIP*, *zJ*) were respectively set to that: [J_1: J_2], [g_1: g_2], [TIP_1: TIP_2], [zJ_1: zJ_2], and the step sizes were set to Δ*J*, Δ*g*, Δ*TIP* and Δ*zJ*. Until the running is over, the best parameters (R_b, J_b, g_b, TIP_b, zJ_b) were obtained.

$$\chi_u = \frac{1}{NT}\left\langle \left(\sum_{i=1}^{N} S_i^z \right)^2 \right\rangle \qquad \text{Eq(1)}$$

where uniform magnetic susceptibility χ_u is dimensionless.

Convert uniform magnetic susceptibility to χ_T (cm^3mol^{-1})

$$\chi_T(cm^3mol^{-1}) = Ag^2\chi_u \qquad \text{Eq(2)}$$

where *g* is Landég-factor and *A* is unit constant.

Import *zJ* and *TIP* factor to χ_T , as bellows:

$$\chi_T'(cm^3mol^{-1}) = \frac{(\chi_T + TIP)}{1 + \frac{zJ(\chi_T + TIP)}{0.1303g^2}} \qquad \text{Eq(3)}$$

The reliability factor *R* is obtained by

$$R = \frac{\sum(\chi_T'T - \chi_{obs}T)^2}{\sum(\chi_{obs}T)^2} \qquad \text{Eq(4)}$$

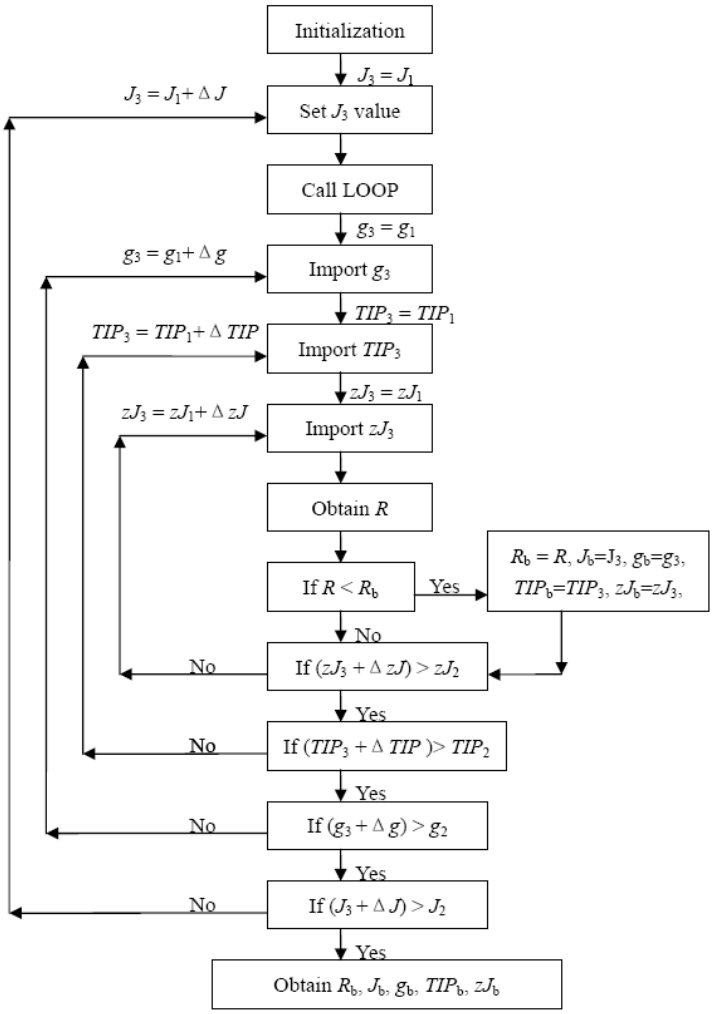

Figure S1 the procedure of our program.

3. XRD curves of compounds 1-5

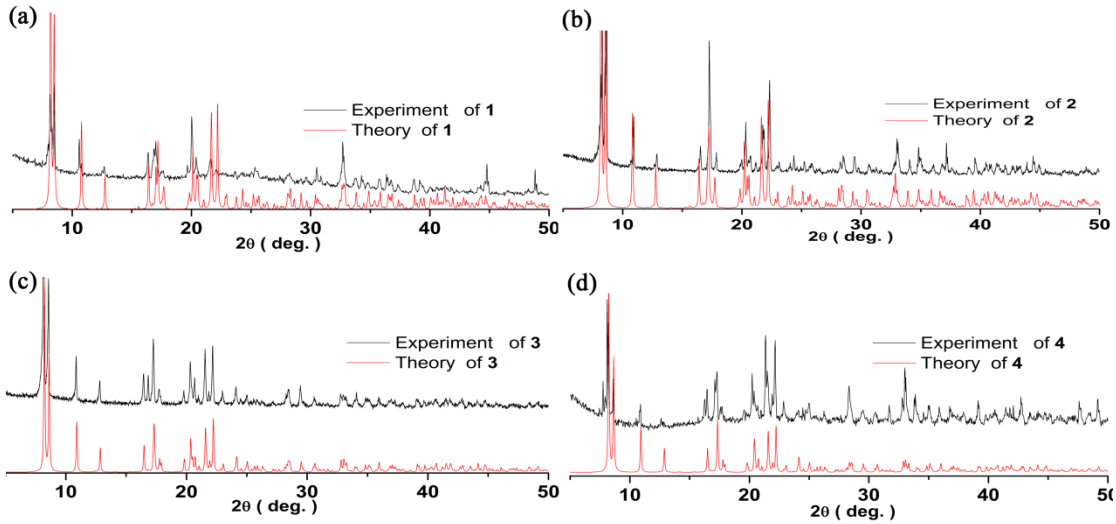

Fig.S2 XRD curves of compounds 1-4.

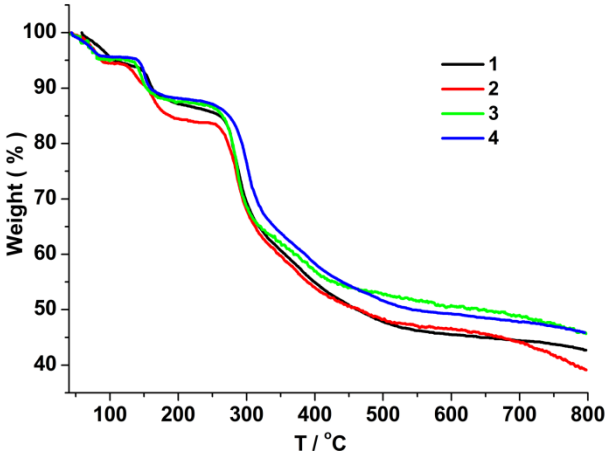

Fig. S3 TGA curve of compounds **1-4** in the range from room temperature to 800 °C.

4. Magnetic properties of 2 and 3.

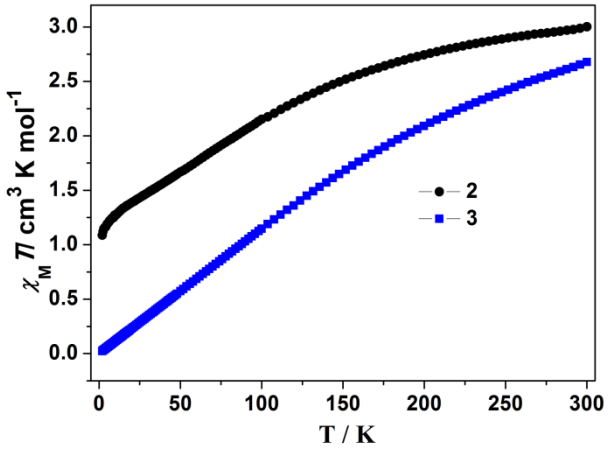

Figure S4 χ_mT vs T of **2** (black) and **3** (blue).

5. Lattice constant *a vs* U plot

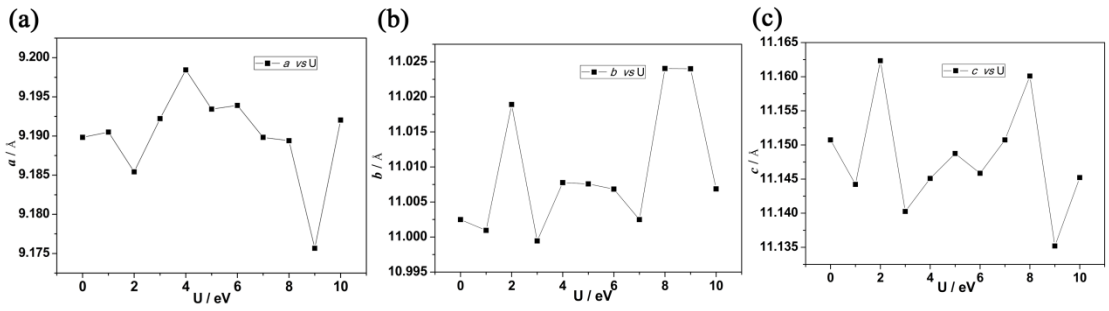

Figure S5 the plot of lattice parameter against U of optimized geometries.

5. PDOS obtained by normal PBE functional

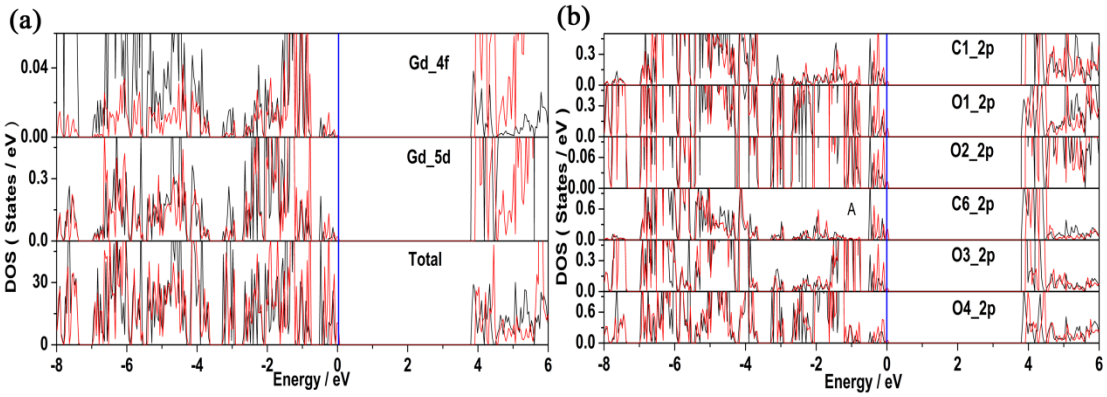

Figure S6 DOS of 4 at the level of PBE functional without VDW-df correction in spin-up (black) and spin-down (red) with Fermi level (blue line).