Electronic Supplementary Information (ESI) for

Study of a Magnetic-Cooling Material $Gd(OH)CO_3$

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Table S1. Selected Bond Lengths (Å) for $Gd(OH)CO_3$.[a]

Gd(1)-O(1)	2.288(8)
Gd(1)-O(1)#1	2.307(7)
Gd(1)-O(3)#2	2.4955(17)
Gd(1)-O(3)	2.4955(17)
Gd(1)-O(2)#3	2.515(5)
Gd(1)-O(2)#4	2.515(5)
Gd(1)-O(2)#5	2.555(5)
Gd(1)-O(2)#6	2.555(5)
Gd(1)-O(2)#7	2.752(5)
Gd(1)-O(2)	2.752(5)

[a]Symmetry transformations used to generate equivalent atoms:
#1 x-1/2,y,-z+1/2 #2 x,y-1,z
#3 x+1/2,y,-z+1/2 #4 x+1/2,-y-1/2,-z+1/2
#5 -x+1/2,-y,z+1/2 #6 -x+1/2,y-1/2,z+1/2
#7 x,-y-1/2,z

Table S2. Selected Bond Angles (°) for $Gd(OH)CO_3$.[a]

O(1)-Gd(1)-O(1)#1	135.5(2)	O(3)#2-Gd(1)-O(2)#6	71.4(2)
O(1)-Gd(1)-O(3)#2	89.91(19)	O(3)-Gd(1)-O(2)#6	121.7(2)
O(1)#1-Gd(1)-O(3)#2	81.35(19)	O(2)#3-Gd(1)-O(2)#6	95.09(12)
O(1)-Gd(1)-O(3)	89.91(19)	O(2)#4-Gd(1)-O(2)#6	68.33(19)
O(1)#1-Gd(1)-O(3)	81.35(19)	O(2)#5-Gd(1)-O(2)#6	51.2(2)
O(3)#2-Gd(1)-O(3)	155.0(3)	O(1)-Gd(1)-O(2)#7	73.69(19)
O(1)-Gd(1)-O(2)#3	72.4(2)	O(1)#1-Gd(1)-O(2)#7	67.66(19)
O(1)#1-Gd(1)-O(2)#3	140.47(16)	O(3)#2-Gd(1)-O(2)#7	49.3(2)
O(3)#2-Gd(1)-O(2)#3	133.5(2)	O(3)-Gd(1)-O(2)#7	107.0(2)
O(3)-Gd(1)-O(2)#3	69.7(2)	O(2)#3-Gd(1)-O(2)#7	145.87(12)
O(1)-Gd(1)-O(2)#4	72.4(2)	O(2)#4-Gd(1)-O(2)#7	108.37(18)
O(1)#1-Gd(1)-O(2)#4	140.47(16)	O(2)#5-Gd(1)-O(2)#7	144.66(8)
O(3)#2-Gd(1)-O(2)#4	69.7(2)	O(2)#6-Gd(1)-O(2)#7	113.41(5)
O(3)-Gd(1)-O(2)#4	133.5(2)	O(1)-Gd(1)-O(2)	73.69(19)
O(2)#3-Gd(1)-O(2)#4	64.0(2)	O(1)#1-Gd(1)-O(2)	67.66(19)
O(1)-Gd(1)-O(2)#5	140.24(18)	O(3)#2-Gd(1)-O(2)	107.0(2)
O(1)#1-Gd(1)-O(2)#5	77.4(2)	O(3)-Gd(1)-O(2)	49.3(2)
O(3)#2-Gd(1)-O(2)#5	121.7(2)	O(2)#3-Gd(1)-O(2)	108.37(18)
O(3)-Gd(1)-O(2)#5	71.4(2)	O(2)#4-Gd(1)-O(2)	145.87(12)
O(2)#3-Gd(1)-O(2)#5	68.33(19)	O(2)#5-Gd(1)-O(2)	113.41(5)
O(2)#4-Gd(1)-O(2)#5	95.09(12)	O(2)#6-Gd(1)-O(2)	144.66(8)
O(1)-Gd(1)-O(2)#6	140.24(18)	O(2)#7-Gd(1)-O(2)	57.9(2)
O(1)#1-Gd(1)-O(2)#6	77.4(2)		

[a]Symmetry codes:

#1 x-1/2,y,-z+1/2 #2 x,y-1,z
#3 x+1/2,y,-z+1/2 #4 x+1/2,-y-1/2,-z+1/2
#5 -x+1/2,-y,z+1/2 #6 -x+1/2,y-1/2,z+1/2
#7 x,-y-1/2,z

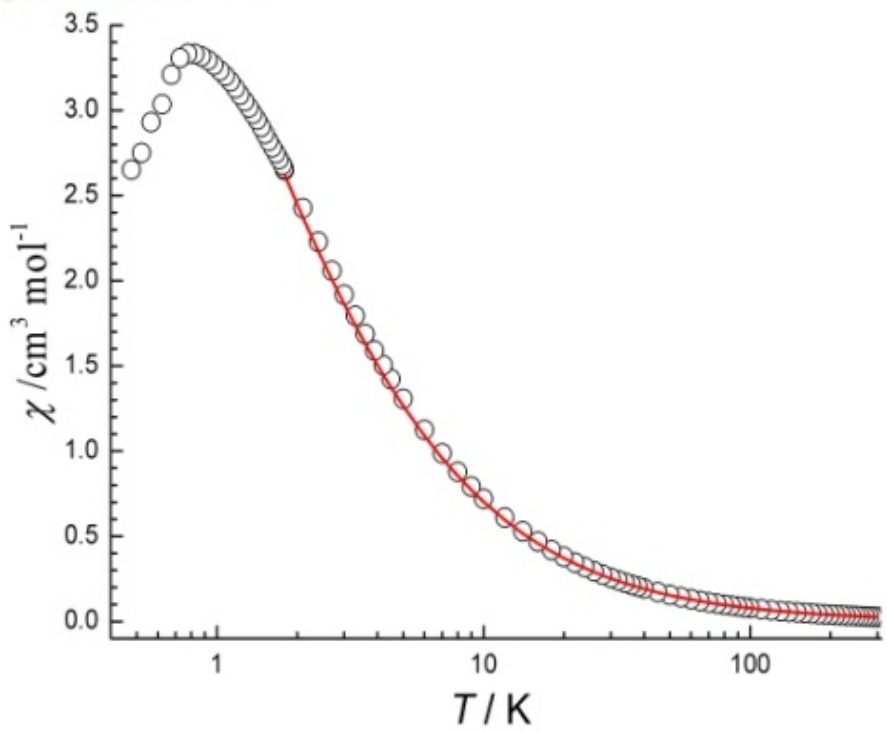

Figure S1 Temperature-dependencies of the magnetic susceptibility (χ_m). The temperature range is 0.46 K ~ 300 K and the applied field is 0.2 T. The red solid line represents the least-square fit for Curie-Weiss law.

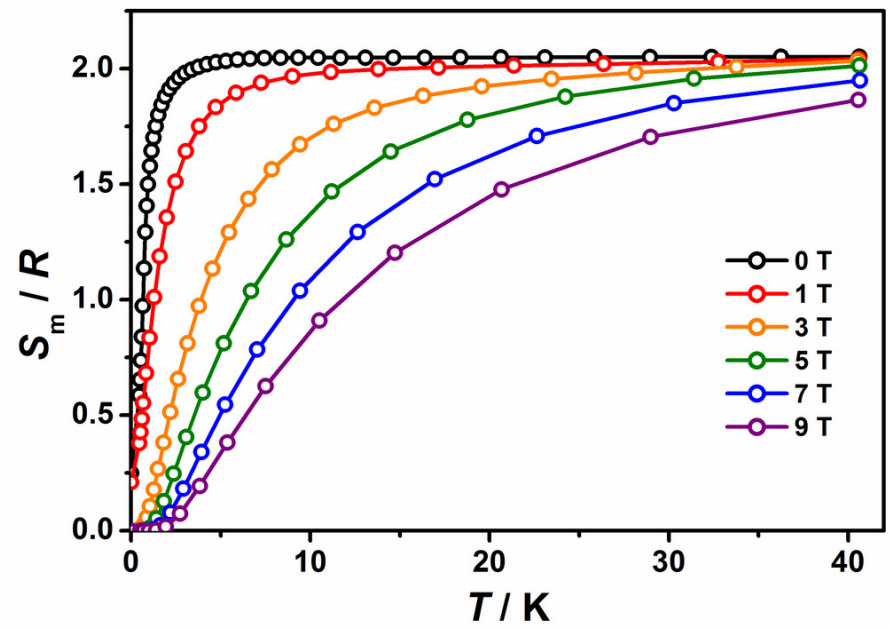

Figure S2 Temperature-dependencies of the magnetic entropy normalized to the gas constant in selected applied fields. Lines are guides to the eyes.

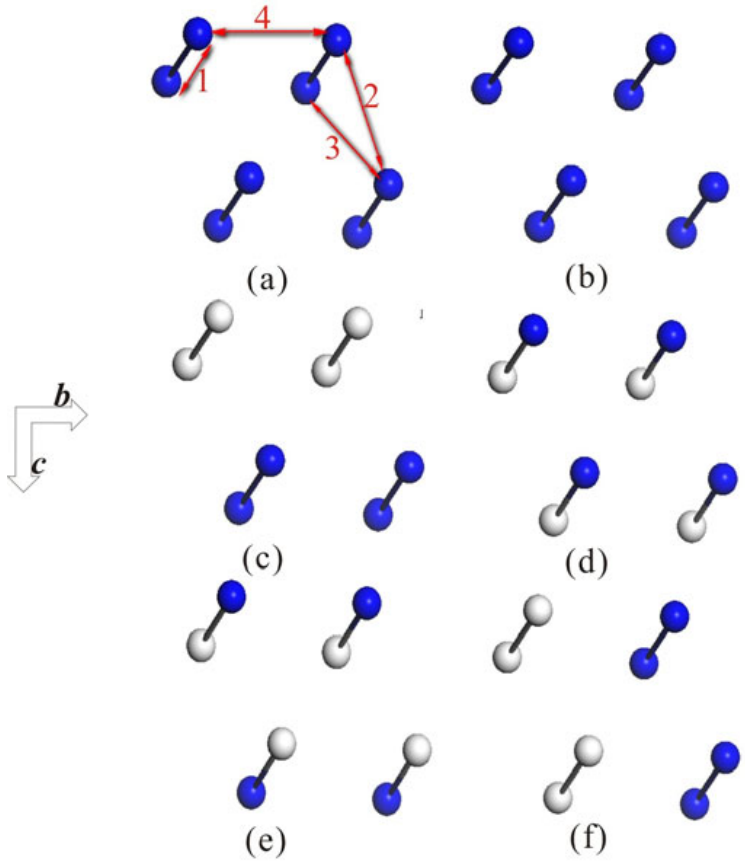

Figure S3 (a) Schematic representation of the spin exchange paths J_1-J_4 of a $Gd(OH)CO_3$ supercell (a 2b c) projected in *bc* plane. (b)-(f) are the five ordered spin states FM, AFM1, AFM2, AFM3, AFM4 of $Gd(OH)CO_3$. The blue ball and white ball represent the Gd^{3+} ions with up-spin and down-spin, respectively.

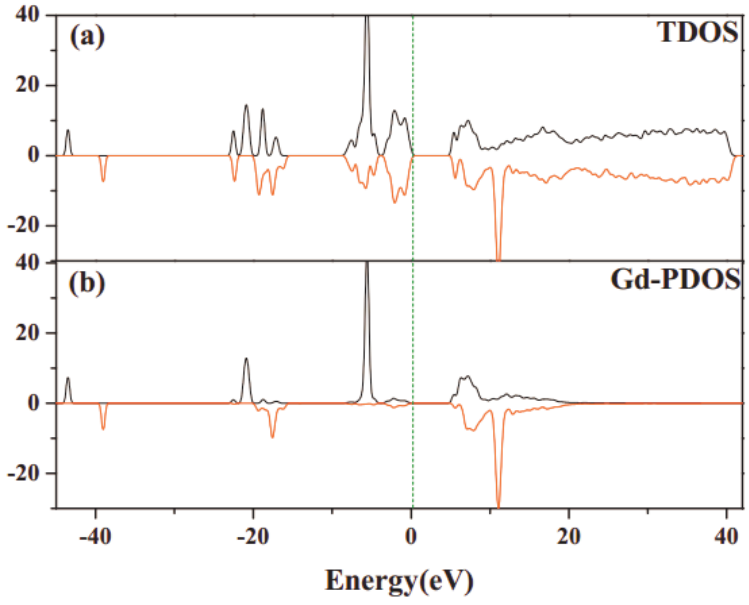

Figure S4 Spin polarized DOS of $Gd(OH)CO_3$ in the FM configuration. (a) Total density of states (b) projected density of states of the Gd atom.