Electronic Supplementary Information (ESI) for

Magnetocaloric Effect and Thermal Conductivity of $Gd(OH)_3$ and $Gd_2O(OH)_4(H_2O)_2$

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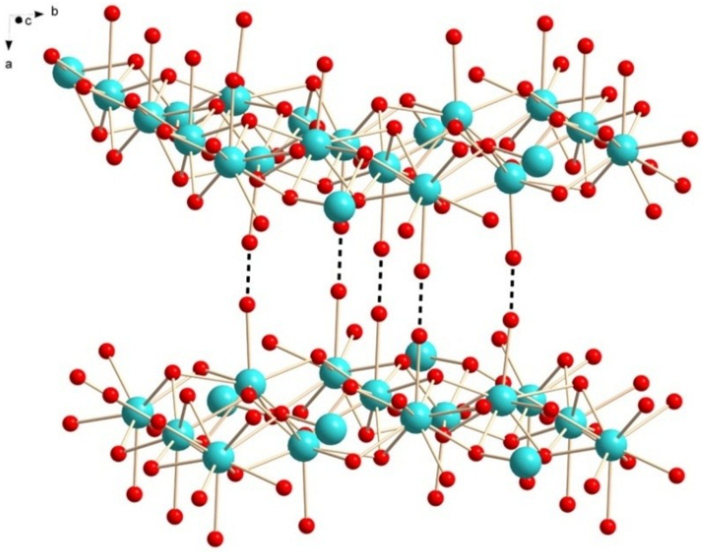

Fig. S1 The 3D structure of compound **2**. Gd blue, O red, H atoms are omitted for clarity.

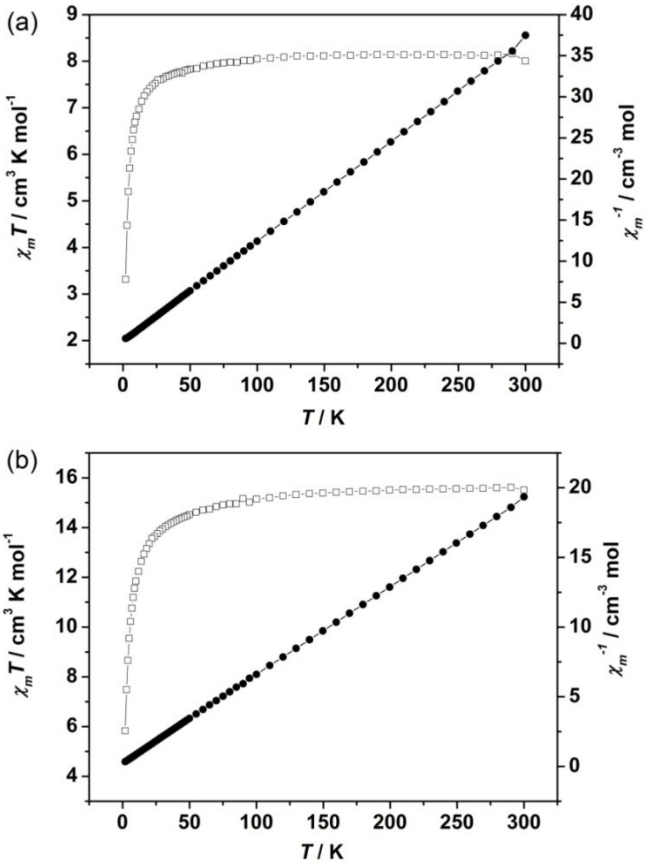

Fig. S2 Plots of temperature dependence of $\chi_M T$ (□) and χ_M^{-1}(●) for **1** and **2** under 1000 Oe dc field between 2 and 300 K.

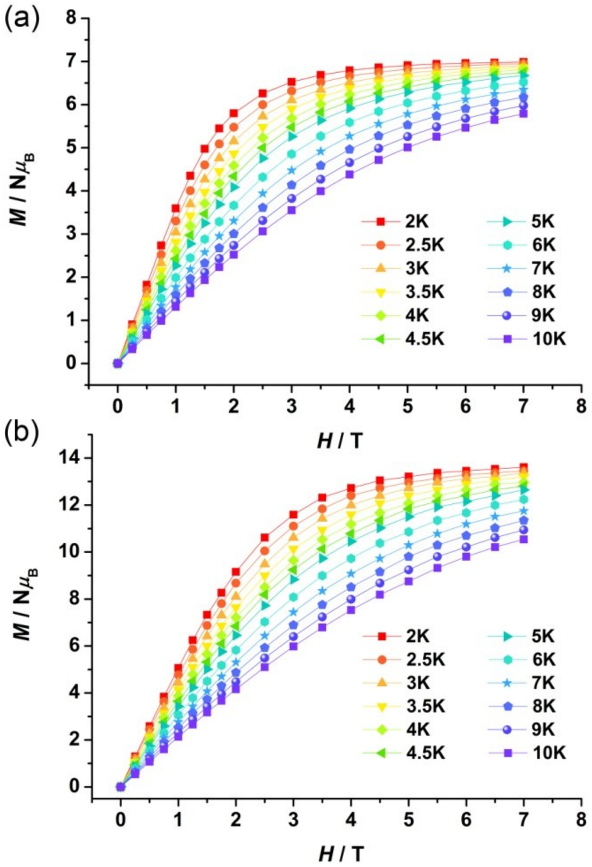

Fig. S3 Field-dependent magnetization plots at indicated temperatures for compounds **1** and **2**.

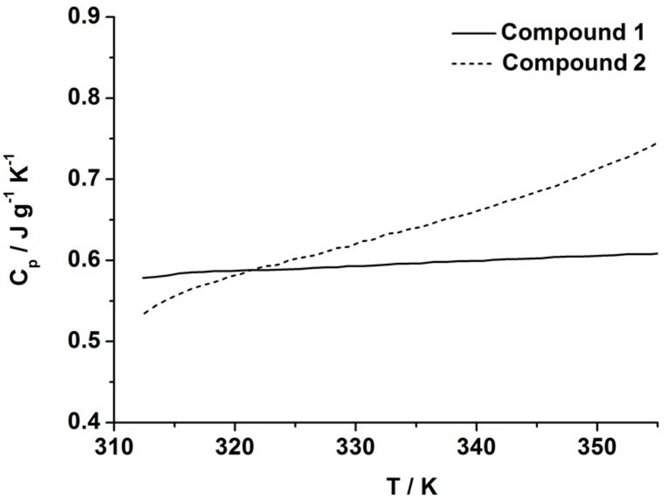

Fig. S4 Specific heat capacity at indicated temperatures for compounds **1** and **2**.

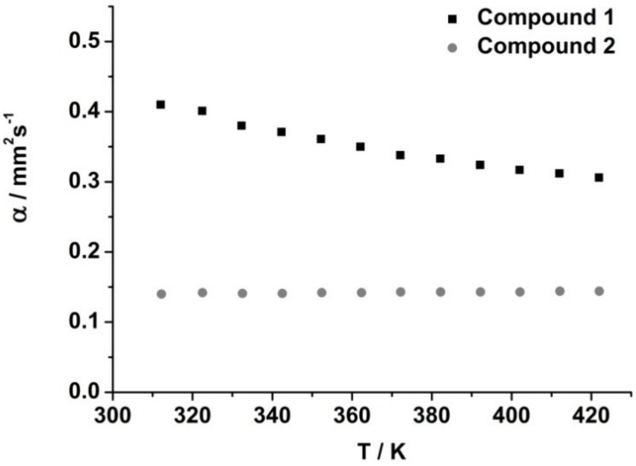

Fig. S5 The thermal diffusion coefficient of compounds **1** and **2** between 312 and 352K.

The thermal conductivity of the compounds was performed on the basis of the powder samples. The detailed condition is: the grain size of the compounds is less than 2μm, the pressure is 12 MPa, and the diameter of pellet of the sample is 13 mm.

The estimating of thermal conductivity (κ) was conducted according to the reported method.[1]

$$\kappa = \frac{1}{T} \cdot \frac{A}{\gamma^2 \varepsilon^3} T_{mp}{}^{3/2} \rho^{2/3} M^{1/3} n^{-1/3} \mu^{-3/2} \quad Eq.\ (1)$$

Where T_{mp} is the melting point, ρ is the density, M and n are the molecular mass and the number of atoms per formula unit respectively. μ is the harmonic average of all the atom masses in a cell. All the aforementioned parameters could be calculated or measured if the crystal cell is determined. Owing to the negligible change with temperature, all these parameters can be considered as constant coefficient P. We could rewrite *Eq. (1)* as follow:

$$\kappa = \frac{1}{T} \cdot \frac{A}{\gamma^2 \varepsilon^3} P \quad Eq.\ (2)$$

Here A is also a independent constant, while γ is the Gruneisen anharmonicity parameter and ε is the factor that corresponding to the atomic distance at melting point (a) and the average atomic distance (a_0) ($a = \varepsilon a_0$). Further more, γ is a function about the specific heat C_v[2]:

$$\gamma = \frac{3\beta B V m}{C_v} \quad Eq.\ (3)$$

Where β is the thermal expansion coefficient, B is the isorthermal bulk modulus and V_m is the molar volume. Thus, we could eventually simplify the thermal conduction expression as follow:

$$\kappa = A \cdot \frac{C_v{}^2}{T} \quad Eq.\ (4)$$

Where all the temperature-independent parameters are integrated into a constant of A.

To estimate the isochoric specific heat C_v, we employed the expression of Debye Model at the full-temperature range:

$$\frac{C_v}{Nk} = 9\left(\frac{T}{T_D}\right)^3 \int_0^{\frac{T_D}{T}} \frac{x^4 e^x}{(e^x - 1)^2} dx \quad Eq.\ (5)$$

Here N is the Avogadro constant, k is the Boltzmann constant and T_D is the Debye temperature. As the isobaric specific heat C_p (the difference between C_v and C_p can be ignored for solid state sample[3]) of compound **1** had been measured via DSC instrument, the T_D can be obtained through fitting the measurement result perfectly. (See Fig. S6)

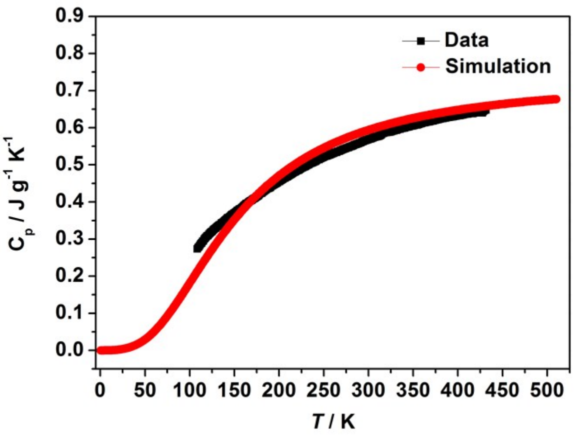

Fig. S6 The calculated curve (Red) using $T_D = 620$ K matches the measurement result perfectly.

To obtain the constant A, we firstly transform *Eq. (4)* slightly into *Eq. (6)*.

$$\frac{\kappa}{C_v^2} = \frac{A}{T} \quad \text{Eq. (6)}$$

Then, the plot of κ measured by Thermal Conduction Instrument (NETZSCH LFA457/2/G) divided by C_v^2 *vs.* temperature was fitted to give the constant $A = 1096.82$ g^2 K^2 J^{-1} s^{-1}m^{-1}. (See Fig. S7)

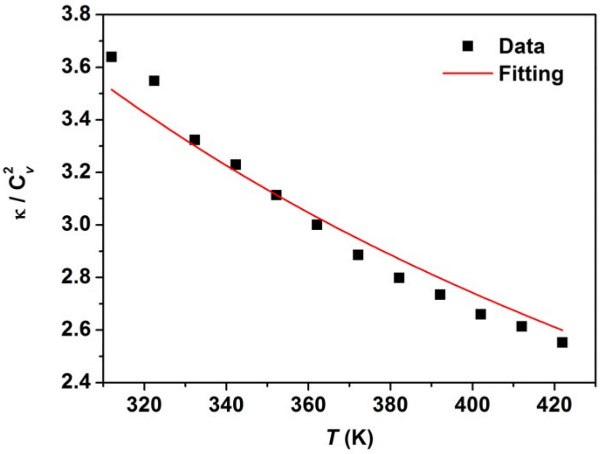

Fig. S7 The polt of κ divided by C_v^2 *vs.* temperature. The non-linear fitting using the function of $y = ax^b$ give the constant $A = 1096.82$ g^2 K^2 J^{-1} s^{-1}m^{-1}.

Measurements of the low-temperature specific heat between 0.43-5.1K for compound **1** has been reported in the early research.[4] Substituting low-temperature specific heat and the constant A into *Eq. (6)*, we obtained temperature-dependent thermal conductivity for compound **1** as shown in Fig. S8.

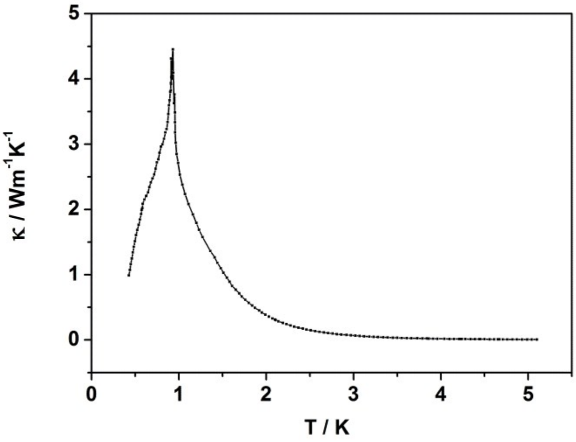

Fig. S8 temperature-dependent thermal conductivity for compound **1** between 0.43-5.1K

The calculated curve shows a sharp peak of 4.45 W m^{-1} K^{-1} at 0.93K, which is larger than the value measured near the room temperature. Though some errors may exists during the estimating of thermal conductivity (κ), The result still reflect the trend of thermal conductivity at low temperature.

Reference

1 R. Gaumé, B. Viana, D. Vivien, J. Roger, D. Fournier, *Appl. Phys. Lett.*, 2003, **83**, 1355.
2 J. G. Yin, Y. Hang, *J. Synth. Cryst.*, 2012, **41**, 247.
3 R. X. Li, *J. B. Inst. Pet. T.*, 1994, **2**, 1.
4 A. T. Skjeltorp, C. A. Catanese, H. E. Meissner, W. P. Wolf, *Phys. Rev. B.*, 1972, **7**, 2062.

Single-crystal X-ray structure determination: Data collections were performed on an Agilent technologies Super Nova Micro Focus single Crystal Diffractometer using Mo-Kα radiation (λ = 0.71073 Å) at 100K. Absorption corrections were applied by using the multi-scan program. The structures were solved by direct methods, and non-hydrogen atoms were refined anisotropically by least-squares on F^2 using the SHELXTL-97 program (*SHELXTL 6.10*, *Bruker Analytical Instrumentation*, Madison, WI, **2000**). The hydrogen atoms were generated geometrically. CCDC 1043508-1043509 for **1** and **2** contain the supplementary crystallographic data for this paper. The data can be obtained free of charge from The Cambridge Crystallorgaphic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.