Supplementary material for

Magnetocaloric properties of the magnetic refrigerant

For our investigations, we use gadolinium as the refrigerant and its magnetocaloric properties were measured by the physical property measurement system (PPMS). However, limited by the magnetic field intensity of the PPMS, only the properties under 1-7 T were measured. Thus, the molecular field theory (MFT) [1,2] is used to calculate the properties under the other magnetic fields. The equations required for the calculation are as follows. The total entropy of the MCM at constant pressure can be expressed as

$$S(H,T) = S_m(H,T) + S_l(H,T) + S_e(H,T) \tag{1}$$

where S_m is the magnetic entropy, S_l is the lattice and S_e is the electron contribution to the total entropy. Generally, we can assume that the lattice and electron parts of the entropy depend only on temperature. In that case, the isothermal magnetic entropy change ΔS_m can be defined as

$$\Delta S_m(\Delta H,T) = S(H_2,T) - S(H_1,T) = S_m(H_2,T) - S_m(H_1,T) \tag{2}$$

where $\Delta H = H_2 - H_1$ denotes the change of the magnetic field.

The magnetic entropy, lattice entropy and electronic entropy are given by the following relations [3].

Magnetic entropy:

$$S_m(H,T) = Nk_B\left[\ln\frac{\sinh\left(\frac{2J+1}{2J}y\right)}{\sinh\left(\frac{1}{2J}y\right)} - yB_J(y)\right] \tag{3}$$

where $B_J(y)$ is the Brillouin function and given by

$$B_J(y) = \frac{2J+1}{2J}\coth\left(\frac{2J+1}{2J}y\right) - \frac{1}{2J}\coth\left(\frac{1}{2J}y\right) \tag{4}$$

where y is defined as:

$$y = \frac{gJ\mu_B H}{k_B T} + \frac{3T_C J B_J(y)}{T(J+1)} \tag{5}$$

Here, N is the number of magnetic atoms. J and T_C respectively represent the total angular quantum number and curie temperature of the magnetocaloric material. k_B, μ_B and g are the Boltzmann constant, Bohr magneton and Lander factor, respectively.

Lattice entropy:

$$S_l(T) = \frac{R}{M_m}\left[12\left(\frac{T}{T_D}\right)^3 \int_0^{T/T_D} \frac{y^3}{e^y - 1}dy - 3\ln\left(1 - e^{-T/T_D}\right)\right] \tag{6}$$

where R is the molar gas constant, M_m is the molar mass and T_D is the Debye temperature of the material.

Electronic entropy:

$$S_e(T)=\gamma_e T \tag{7}$$

where γ_e is the electron specific heat coefficient, which is generally 10^{-3}~10^{-4} J mol^{-1} K^{-1}. Correspondingly, the total specific heat capacity consists of the magnetic heat specific capacity, lattice heat specific capacity and electronic heat specific capacity, expressed as [4]:

$$c(H,T)=c_m(H,T)+c_l(T)+c_e(T)=T\left(\frac{\partial S_m(H,T)}{\partial T}\right)+T\left(\frac{\partial S_l(T)}{\partial T}\right)+T\left(\frac{\partial S_e(T)}{\partial T}\right) \tag{8}$$

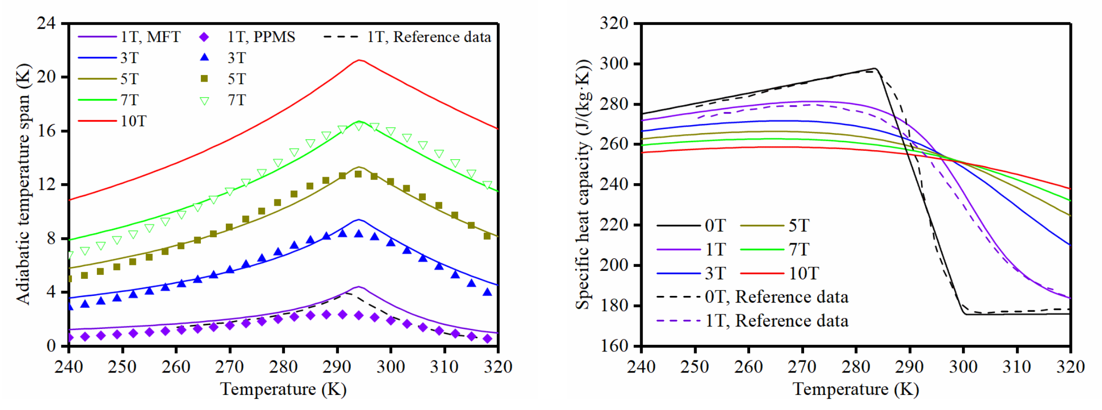
where c_m, c_l and c_e are the magnetic heat specific capacity, lattice heat specific capacity and electronic heat specific capacity, respectively. With the Maxwell relations, the adiabatic temperature change ΔT_{ad} of gadolinium can be readily calculated as:

$$\Delta T_{ad}(\Delta H,T)=-\frac{T\Delta S_m(\Delta H,T)}{c(H,T)} \tag{9}$$

The relevant parameters of gadolinium are shown in Supplementary Table S1. Supplementary Fig. S1 shows the calculated magnetocaloric properties, and the results are in good agreement with both measured and reference data [5].

Supplementary Table S1 The related parameter values of gadolinium [3].

g	J	T_D (K)	T_C (K)	M_m (kg mol^{-1})	ρ (kg m^{-3})
2	3.5	173	294	0.157	7901



(a) Adiabatic temperature change (b) Specific heat capacity

Supplementary Fig. S1. Magnetocaloric Properties of the gadolinium.

Supplementary References

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[5] Kitanovski, A.; Tušek, J.; Tomc, U.; Plaznik, U.; Ožbolt, M.; Poredoš, A. *Magnetocaloric Energy Conversion*; Springer International Publishing: Berlin/Heidelberg, Germany, 2015.