RAPID COMMUNICATION

High pressure synthesis and characterization of the pyrochlore Dy$_2$Pt$_2$O$_7$: A new spin ice material

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The cubic pyrochlore Dy$_2$Pt$_2$O$_7$ was synthesized under 4 GPa and 1000 °C and its magnetic and thermodynamic properties were characterized by DC and AC magnetic susceptibility and specific heat down to 0.1 K. We found that Dy$_2$Pt$_2$O$_7$ does not form long-range magnetic order, but displays characteristics of canonical spin ice such as Dy$_2$Ti$_2$O$_7$, including (1) a large effective moment 9.64 $\mu_B$ close to the theoretical value and a small positive Curie–Weiss temperature $\theta_{CW} = +0.77$ K signaling a dominant ferromagnetic interaction among the Ising spins; (2) a saturation moment $\sim 4.5 \mu_B$ being half of the total moment due to the local (111) Ising anisotropy; (3) thermally activated spin relaxation behaviors in the low ($\sim$ 1 K) and high ($\sim$ 20 K) temperature regions with different energy barriers and characteristic relaxation time; and most importantly, (4) the presence of a residual entropy close to Pauling’s estimation for water ice.

Keywords: Dy$_2$Pt$_2$O$_7$, pyrochlore oxide, spin ice, high-pressure synthesis

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1. Introduction

Geometrically frustrated magnets can display rich and diverse phenomena, providing an excellent platform for experimental realizations of collective magnetism that was predicted theoretically.\cite{1,2} The recognition of a spin ice state in the cubic pyrochlore compounds Ho$_2$Ti$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$ represents such an example that successfully maps to the well-known water–ice problem.\cite{3-6} In these pyrochlores, the magnetic rare-earth ions, Ho$^{3+}$ or Dy$^{3+}$, with a large magnetic moment of $\sim 10 \mu_B$ reside on the vertices of the corner-sharing tetrahedra, forming a geometrically frustrated lattice. A uniaxial local crystalline electric field (CEF) around Ho$^{3+}$ or Dy$^{3+}$ results in a nearly perfect easy-axis anisotropy, forcing the rare-earth’s spin to point along the local (111) axis that joins the centers of two neighboring tetrahedra.\cite{7,8} The combination of dipolar and magnetic exchange interactions favors ‘two-in, two-out’ spin configurations on each tetrahedron, which has been termed ‘spin ice’ in direct analogy to the ‘two-short, two-long’ proton bond disorder in water ice. The Pauling’s zero-point entropy $S_p = (R/2) \ln(3/2)$, where $R$ is the ideal gas constant, for water ice has also been confirmed in the spin ices having a macroscopically degenerate ground state.\cite{5}

The low-temperature magnetic properties of a pyrochlore spin ice are mainly controlled by the magnetic exchange ($J_{nn}$) and the dipole–dipole interaction ($D_{nn}$) of the nearest-neighbor spins. For these pyrochlore spin ice materials, $J_{nn}$ is usually antiferromagnetic, while $D_{nn}$ is ferromagnetic and can be calculated as $D_{nn} = 5/3(\mu_0/4\pi)\mu^2/2r_{nn}$, where $r_{nn}$ is the nearest-neighbor rare-earth distance. $D_{nn}$ is estimated to be $\sim 2.35$ K for Ho$_2$Ti$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$. The physics of pyrochlore spin ices can be largely captured by the dipolar spin ice model (DSIM) proposed by den Hertog and Gingras.\cite{9} In the theoretical phase diagram based on DSIM,\cite{9} the spin ice state is stable for $J_{nn}/D_{nn} > -0.91$, and a $Q = 0$ antiferromagnetically ordered state is favored for $J_{nn}/D_{nn} < -0.91$. Since $J_{nn}$ is more sensitive to $r_{nn}$ than $D_{nn}$, by synthesizing Ho$_2$Ge$_2$O$_7$ and Dy$_2$Ge$_2$O$_7$,\cite{10,11} which have smaller lattice parameter in comparison to the corresponding titanates and stannates, our previous studies have successfully modified the ratio of $J_{nn}/D_{nn}$. The lowest value of $-0.73$ is achieved in Dy$_2$Ge$_2$O$_7$,\cite{11} which

\[ J_{nn}/D_{nn} = -0.73 \]

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is still located inside the spin ice regime. A further enhancement of $J_{nn}$ is needed in order to achieve a transition from spin ice to a $Q = 0$ state.

Besides the chemical pressure effects caused by the size of $B$-cation in $Ln_2B_2O_7$ ($Ln$ = rare earth), our recent studies on the platinum-based pyrochlores $Ln_2Pt_2O_7$ have revealed an additional effect of the nonmagnetic Pt$^{4+}$ ions,\cite{12,13} i.e., the spatially more extended Pt-5d orbitals and thus the enhanced Pt 5d–O 2p hybridizations can modify the CEF and influence the exchange interactions. In comparison with Gd$_2B_2O_7$ ($B$ = Ge, Ti, Sn), the antiferromagnetic transition temperature of Gd$_2Pt_2O_7$ is substantially enhanced because the empty Pt-$e_g$ orbitals provide extra superexchange pathways.\cite{13} Thus, we expect an alternative routine to modify $J_{nn}/D_{nn}$ in the Pt-based Ising pyrochlores.

In this work, we have prepared the pyrochlore compound Dy$_2Pt_2O_7$ under high pressure, and characterized its magnetic and thermodynamic properties via magnetic susceptibility and specific heat measurements down to 0.1 K. We find that Dy$_2Pt_2O_7$ does not exhibit any long-rang magnetic order at low temperature, but displays canonical spin ice characteristics, including the thermally activated spin dynamics and the presence of Pauling’s zero-point entropy. By comparing the low-temperature magnetic specific heat to DSIM, we obtain a ratio of $J_{nn}/D_{nn} = -0.56$ for Dy$_2Pt_2O_7$. Our results demonstrate that Dy$_2Pt_2O_7$ is a new spin ice compound with enhanced superexchange interaction $J_{nn}$.

2. Experimental details

The sample preparations and characterizations in the present study are similar to those performed in our previous work.\cite{12} The cubic pyrochlore Dy$_2Pt_2O_7$ sample was prepared under 4 GPa and 1000 °C by using a Kawai-type multi-anvil module (Max Voggenreiter GmbH). The resultant high-pressure products were first washed with warm aqua regia to remove a small amount of platinum metal and unreacted Dy$_2O_3$; the obtained powders were then pressed into pellets and subjected to heat treatment at 900 °C for 10 h to facilitate the measurements of the bulk physical properties.

Phase purity of the obtained powder and pellet samples was examined by powder x-ray diffraction (XRD) at room temperature. Structural parameters were extracted from the XRD patterns via Rietveld refinement with the FullProf program. DC magnetic susceptibility was measured with a commercial magnetic property measurements system (MPMS-III, Quantum Design) in the temperature range from 1.8 K to 300 K under an external magnetic field of $\mu_0H = 0.1$ T. AC magnetic susceptibility measurements in the temperature range $2 K < T < 50 K$ were performed in a physical property measurement system (PPMS, Quantum Design). AC susceptibility measurements from 70 mK to 2 K were carried out in an Oxford dilution refrigerator with the mutual induction method; an excitation current of ~ 1 mA with frequencies ranging from 117 Hz to 1517 Hz was applied to the primary coil during the measurements. Specific-heat data down to 0.1 K were collected by the PPMS with a dilution refrigerator insert.

3. Results and discussion

3.1. Structure characterizations

Figure 1 (a) shows the powder XRD pattern of Dy$_2Pt_2O_7$, which is confirmed to be single phase with the pyrochlore structure. The XRD pattern can be refined well with a cubic pyrochlore structure defined by the Fd-3m (No. 227) space group with the Dy atom at 16d (1/2,1/2,1/2), the Pt atom at 16c (0,0,0), the O1 atom at 48f (x,1/8,1/8), and the O2 atom at 8b (3/8,3/8,3/8) site, respectively. The obtained lattice constant $a = 10.1913(1)$ Å for Dy$_2Pt_2O_7$ is in excellent agreement with the previously reported value of 10.202 Å.\cite{14} In addition, the lattice constant $a$ scales linearly with the ionic radius (IR) of the $B^{4+}$ ions for the series of Dy$_2B_2O_7$ ($B$ = Sn, Pt, Ti, Ge), as depicted in Fig. 1(b).

![Figure 1](image_url)

**Fig. 1.** (a) Room-temperature powder XRD pattern of Dy$_2Pt_2O_7$ and the results of the Rietveld refinement. (b) Lattice parameter $a$ (left) and the Curie–Weiss temperature $\theta_{CW}$ (right) as a function of the ionic radius of the $B^{4+}$ ions in the series of Dy$_2B_2O_7$ ($B$ = Sn, Pt, Ti, Ge).

3.2. DC magnetic susceptibility

The magnetic properties of Dy$_2Pt_2O_7$ were first characterized by DC magnetization measurements in the temperature range from 1.8 K to 300 K. Figure 2(a) displays the DC magnetic susceptibility $\chi(T)$ and its inverse $\chi^{-1}(T)$ measured under $\mu_0H = 0.1$ T after zero-field-cooled (ZFC) and field-cooled (FC) processes from room temperature. No sign of long-range magnetic ordering was observed down to 1.8 K.
A Curie–Weiss (CW) fitting has been applied to $\chi^{-1}(T)$ in the temperature range 10–50 K, i.e., $\chi^{-1}(T) = (T - \theta_{CW})/C$, which yields an effective moment $\mu_{eff} = 9.62 \mu_B$ per Dy$^{3+}$ and a CW temperature $\theta_{CW} = +0.77$ K for Dy$_2$Pt$_2$O$_7$. The obtained $\mu_{eff}$ is very close to the theoretical value of 9.55 $\mu_B$ for free Dy$^{3+}$ ion with $J = 15/2$, and the small positive $\theta_{CW}$ signals a net ferromagnetic interaction between the Ising spins located on the vertices of the corner-shared tetrahedra.\(^{[15]}\) As seen in Fig. 1(b), the positive $\theta_{CW}$ decreases roughly linearly with decreasing IR of $B^{4+}$ and approaches to zero for the Ge-based pyrochlore. Such a monotonic variation of $\theta_{CW}$ versus IR($B^{4+}$) suggests that the nearest-neighbor distance, $r_{nn}$, remains the dominant factor that governs the balance between the antiferromagnetic $J_{nn}$ and the ferromagnetic $D_{nn}$ between the nearest-neighbor Ising spins.

The isothermal magnetization $M(H)$ curves measured in fields up to 5 T at 2 K and 5 K are shown in Fig. 2(b) for Dy$_2$Pt$_2$O$_7$. As seen in the classical spin ices, a typical ferromagnetic behavior is observed and the saturation moment reaches $\sim 4.5 \mu_B$ per Dy$^{3+}$, which is about half of the total moment available at each site due to the local (111) Ising magnetic anisotropy and the powder averaging.\(^{[16]}\) From these DC magnetic measurements on Dy$_2$Pt$_2$O$_7$, we find nearly identical behaviors as those well-characterized classical spin ices Dy$_2$B$_2$O$_7$ ($B = $ Sn, Ti, Ge). Below we further provide the AC magnetic susceptibility and thermodynamics evidences in support of the spin-ice behavior for Dy$_2$Pt$_2$O$_7$.

### 3.3. AC magnetic susceptibility

Figure 3 shows the AC magnetic susceptibility of Dy$_2$Pt$_2$O$_7$ in the temperature range (a)–(c) below 2 K and (d)–(f) above 2 K. For $T < 2$ K, the real $\chi'(T)$ and the imaginary part $\chi''(T)$ are normalized by their maximum values.

Fig. 2. (a) Temperature dependence of the DC magnetic susceptibility $\chi(T)$ and its inverse $\chi^{-1}(T)$ for Dy$_2$Pt$_2$O$_7$. The solid line represents the Curie–Weiss fitting curve and the fitting parameters are given in the figure. (b) The isothermal magnetization curves $M(H)$ measured at $T = 2$ K and 5 K for Dy$_2$Pt$_2$O$_7$.

![Figure 2](https://example.com/figure2.png)

![Figure 3](https://example.com/figure3.png)

Fig. 3. The real part $\chi'(T)$ and the imaginary part $\chi''(T)$ of the AC magnetic susceptibility for Dy$_2$Pt$_2$O$_7$ in the temperature range (a), (b) below 2 K and (d), (e) above 2 K. (c) and (f) The linear fit to the Arrhenius plot of the characteristic temperatures $T_1$ and $T_2$, respectively.
As can be seen in Figs. 3(a) and 3(b), $\chi'(T)$ in the frequency range from 117 Hz to 1317 Hz drops quickly below $\sim 1.6$ K, and $\chi''(T)$ displays a broad shoulder at a slightly lower temperature. Both $\chi'(T)$ and $\chi''(T)$ are frequency dependent and the shoulder shifts to higher temperatures upon increasing frequency. Such a slow spin dynamics is typical for classical spin ices and has been attributed to the formation of spin-ice configurations.$^{[17,18]}$ Due to the ambiguity of the maximum in $\chi''(T)$, here we define the maximum of $d\chi'/dT$ as a characteristic temperature $T_1$ whose frequency dependence can be fitted with an Arrhenius formula, $f = j_0 \exp(-E_{\text{act}}/k_B T_1)$. Fig. 3(c), yielding an activation energy $E_{\text{act}} = 267.4$ K and a characteristic relaxation time $\tau_{\text{dy}} = 1/f_{\text{dy}} = 3.18 \times 10^{-10}$ s.

Another distinct feature in the AC susceptibility of Dy-pyrochlore spin ices is the presence of a high-temperature peak around 15 K.$^{[17-20]}$ Similar behaviors are also observed in Dy$_2$Pt$_2$O$_7$, Figs. 3(d) and 3(e). For $f \geq 333$ Hz, we observe a clear drop in $\chi'(T)$ and a corresponding maximum in $\chi''(T)$ at temperature $T_2$, which starts at $\sim 18$ K and moves to higher temperatures with increasing frequency. Similarly, a linear fitting to the Arrhenius plot of $\ln f$ versus $1/T_2$, Fig. 3(f), gives the activation energy $E_{\text{act}} = 267.4$ K and the characteristic relaxation time $\tau_{\text{dy}} = 1/f_{\text{dy}} = 1.66 \times 10^{-9}$ s, which are also consistent with the reported values of $E_{\text{a}} = 210$ K and $\tau_0 = 1.39 \times 10^{-9}$ s for the polycrystalline Dy$_2$Ti$_2$O$_7$. Since the energy barrier $E_{\text{dy}} \sim 300$ K is close to the energy scale set by the first excited CEF level from the ground state doublet, the spin relaxation processes are thermally activated for $T \geq T_2$.

### 3.4. Specific heat

Figure 4(a) shows the low-temperature specific heat $C(T)$ of Dy$_2$Pt$_2$O$_7$, which displays a Schottky-like peak centered at about 1.1 K. Both the peak position $\sim 1$ K and the magnitude $\sim 3$ J/(mol-Dy)$^{-1}$K$^{-1}$ are very similar to those of the classical Dy-pyrochlore spin ices, Dy$_2$B$_2$O$_7$ ($B = \text{Sn, Ti, Ge}$).$^{[11]}$ The magnetic contribution $C_{\text{m}}$ was obtained by subtracting from the measured total specific heat $C_{\text{total}}$ the lattice contribution $C_{\text{lat}}$. which was taken from the specific heat of isostructural, nonmagnetic Lu$_2$Pt$_2$O$_7$. As shown in Fig. 4(a), the magnetic entropy obtained via integrating $C_{\text{m}}/T$ over the investigated temperature range approaches a constant value of $\sim 4.2$ J/(mol-Dy)$^{-1}$K$^{-1}$, which falls considerably short of the expected value of $R\ln 2 \approx 5.76$ J/(mol-Dy)$^{-1}$K$^{-1}$ for Dy$^{3+}$ with a ground Karmers doublet. The resultant residual entropy $1.56$ J/(mol-Dy)$^{-1}$K$^{-1}$ is close to the Pauling’s zero-point entropy $S_{\text{p}} = (R/2)\ln(3/2) = 1.68$ J/(mol-Dy)$^{-1}$K$^{-1}$, providing an important evidence for the spin-ice state in Dy$_2$Pt$_2$O$_7$.$^{[5]}$

![Fig. 4.](image)

(b) Comparison of the magnetic specific heat $C_{\text{m}}$ of Dy$_2$B$_2$O$_7$ ($B = \text{Sn, Pt, Ti, Ge}$) pyrochlores.

### 3.5. Discussion

Based on these above characterizations, we can conclude that Dy$_2$Pt$_2$O$_7$ is a new spin ice characterized by: (i) a large effective moment 9.64 $\mu_B$ close to the theoretical value, and a small positive Curie-Weiss temperature $\theta_{\text{CW}} = 0.77$ K signaling a dominant ferromagnetic interaction among the Ising spins; (ii) a saturation moment $\sim 4.5$ $\mu_B$ being half of the total moment due to the local $\{111\}$ Ising anisotropy; (iii) thermally activated spin relaxation behaviors in the low $(\sim 1$ K) and high $(\sim 20$ K) temperature regions with different energy barriers and characteristic relaxation time; and most importantly, (iv) the presence of a residual entropy close to Pauling’s estimation for water ice.

Although it is not unexpected that Dy$_2$Pt$_2$O$_7$ displays typical behaviors of canonical spin ice, the effects of nonmagnetic Pt$^{4+}$ are noteworthy. For this purpose, we have compared the $C_{\text{m}}$ of Dy$_2$Pt$_2$O$_7$ with those of Dy$_2$B$_2$O$_7$ ($B = \text{Sn, Ti, Ge}$) in Fig. 4(b). For those latter spin ices, it has been shown that the $C_{\text{m}}$ peak shifts to lower temperatures and its height also increases upon reducing the ionic radius of B$^{4+}$ ions or $r_{\text{nn}}$.$^{[11]}$ This is consistent with the prediction of DSIM due to the enhancement of $J_{\text{nn}}$ with respect to $D_{\text{nn}}$.$^{[9]}$ If considering only the lattice parameter or $r_{\text{nn}}$, the $C_{\text{m}}$ peak of Dy$_2$Pt$_2$O$_7$ should locate in between those of Dy$_2$Sn$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$. However, as shown in Fig. 4(b), the $C_{\text{m}}$ peak of Dy$_2$Pt$_2$O$_7$ is higher than that of Dy$_2$Sn$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$, and the $C_{\text{m}}$
Based on the prediction of DSIM\cite{9} and $D_{mn} = 2.29$ K for Dy$_2$Ti$_2$O$_7$, the $C_m$ peak temperature $T_{peak}$ corresponds to a $J_m/D_{mn} = -0.56$, which predicts $J_m = -1.28$ K and $J_{eff} = J_m + D_{mn} = 1.01$ K. These values are also listed in Table 1. For comparison, Dy$_2$Ti$_2$O$_7$ has $J_m/D_{mn} = -0.49$, $J_m = -1.15$ K, and $J_{eff} = 1.20$ K\cite{11}. Because the ionic radius of Pt$^{4+}$ (0.625 Å) is larger than that of Ti$^{4+}$ (0.605 Å), the observed larger $|J_m|$ and $|J_m/D_{mn}|$ in Dy$_2$Pt$_2$O$_7$ than those of Dy$_2$Ti$_2$O$_7$ suggest that other factors beyond the chemical pressure effect should play a role in determining the low-temperature magnetic properties of Dy$_2$Pt$_2$O$_7$. As in the case of Gd$_2$Pt$_2$O$_7$\cite{13} we attribute the enhanced $|J_m|$ in Dy$_2$Pt$_2$O$_7$ to the extra superexchange pathways through the empty $e_g$ orbitals of Pt$^{4+}$ via Dy$-$O$-$Pt$-$O$-$Dy. This contribution becomes prominent in Dy$_2$Pt$_2$O$_7$ having the spatially more extended Pt 5d orbitals.

Table 1. Lattice parameter and selected magnetic parameters for the Dy-pyrochlore spin ice Dy$_2$B$_2$O$_7$ ($B = \text{Ge, Ti, Pt, Sn}$).

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4. Conclusion

In summary, we have synthesized the cubic pyrochlore Dy$_2$Pt$_2$O$_7$ under 4 GPa and 1000 °C, and confirmed it to be a new classical spin ice as Dy$_2$Ti$_2$O$_7$. The magnetic specific heat of Dy$_2$Pt$_2$O$_7$ signals a moderate enhancement of $|J_m|$, but the ratio $J_{mn}/D_{mn} = -0.56$ remains located in the spin ice regime as predicted by DSIM. Our work demonstrates that the $J_{mn}/D_{mn}$ can be effectively tuned by replacing the B-site cation of Ising pyrochlores. But further explorations are needed to realize a transition from spin ice to an antiferromagnetically ordered state by varying $J_{mn}/D_{mn}$ in a larger range as predicted by the DSIM.

References