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$HoTbTi_2O_7$, the mixtures of spin ice and spin liquid

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Abstract

Polycrystalline samples of $Ho_{2-x}Tb_xTi_2O_7$ ($x = 0.5$, 1, and 1.5) have been prepared and characterized. No long-range order is observed for $HoTbTi_2O_7$ in magnetization and specific heat measurements down to 2 K. The low-energy magnetic excitation measurements suggests that $HoTbTi_2O_7$ possesses both characteristics of spin ice and spin liquid in the ground state.

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Pyrochlore oxides with a general formula $A_2B_2O_7$ (A: rare earth, B: transition metal) have attracted much attention due to the geometrical frustrations and interesting low temperature properties. $Ho_2Ti_2O_7$ had been reported to have a spin-ice configuration with a net ferromagnetic interaction explained by an ising-type anisotropy model [1]. $Tb_2Ti_2O_7$ shows a fluctuating spin-liquid state at low temperatures as explained by an antiferromagnetic Heisenberg model [2]. In this report, we describe the physical properties of the mixed compound, $HoTbTi_2O_7$, together with its structural data.

Polycrystalline samples $Ho_{2-x}Tb_xTi_2O_7$ ($x = 0.5$, 1.0, and 1.5) were synthesized using a standard solid-state reaction [3]. Room temperature X-ray powder diffraction data showed no secondary impurity phases within our instrumental resolution of 0.03°. Lattice parameters are $a = 10.134$, 10.117, and 10.103 Å for $x = 0.5$, 1.0, and 1.5, respectively with space group Fd3m.

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Magnetic susceptibility $\chi(T)$ are shown in Fig. 1 for $x = 0.5$, 1.0, and 1.5. No magnetic transition is observed down to 2 K; however, small change is observed below 5 K for all samples. The Curie–Weiss law, $\chi = C/(T–\theta_{CW})$, was fitted to the data, where C is the Curie constant and θ_{CW} the Curie–Weiss temperature. The effective moments p_{eff} were determined for the temperature range from 200 to 400 K.

It is found that $\theta_{CW} = -14.2$, -11.7, and -10.3 K, and $p_{eff} = 9.93$, 10.08, and 10.29 μ_B, for $x = 0.5$, 1.0, and 1.5, respectively. p_{eff} increase as x increase. These observations suggest that antiferromagnetic state is preferred to ferromagnetic state in these compounds.

Specific heat measurements were carried out using a thermal relaxation method down to 0.5 K. No magnetic transition is observed. These results are consistent with those from neutron powder diffraction measurements [3]. Specific heat C_p for $x = 1$ is shown in Fig. 2.

Lattice specific heat C_l is estimated from the nonmagnetic iso-structural compound $Y_2Ti_2O_7$. Magnetic specific heat C_m is estimated below 30 K by subtracting C_l. No anomaly is observed. Although no nuclear contribution is considered here [4], a broad peak, which is possibly from spin ice state, can be seen around 1.9 K. The broad peak

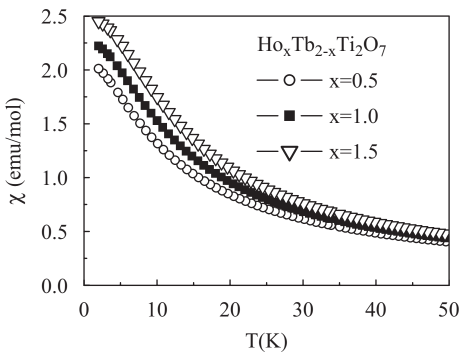

Fig. 1. Temperature dependence of inverse magnetic susceptibility $1/\chi$ for $Ho_{2-x}Tb_xTi_2O_7$ with $x = 0.5$, 1.0, and 1.5.

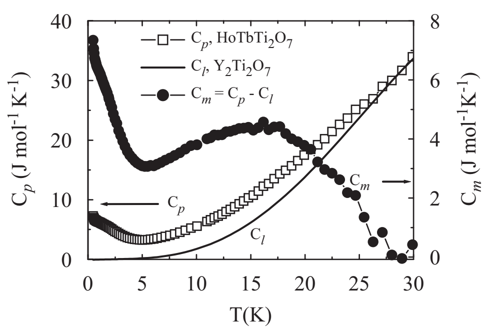

Fig. 2. Specific heat C_p for $HoTbTi_2O_7$ as a function of temperature. Lattice specific heat C_l is estimated from $Y_2Ti_2O_7$. Magnetic specific heat C_m is calculated from C_p by subtracting C_l.

around 6 K in the spin-liquid state for $x = 0$ [2] is shifted to 15 K. This observation is consistent with the results from $\chi(T)$, where the $\chi(T)$ deviates from the Curie–Weiss law below ~30 K. The peak at 1.5 K for $x = 0$ may merge into the peak at 1.9 K. Detailed analysis of magnetic specific heat and magnetic entropy will be discussed elsewhere [3]. From the discussion above, it can be concluded that $HoTbTi_2O_7$ shows the characteristics of both spin ice and spin liquid below 30 K.

In addition, the low-energy excitations of $HoTbTi_2O_7$ powder samples were investigated at the time-of-flight (TOF) spectrometer diffuse neutron scattering (DNS) at FRJ-2, Germany. The wavelength of 4.75 Å was chosen to achieve a good energy resolution. The inelastic neutron scattering data at 10 K are shown in Fig. 3. The first excited mode is observed at ~2 meV, which is reminiscent of the crystal-field level previously reported for $Tb_2Ti_2O_7$ [2]. Above 100 K, this mode disappears, however, a new mode

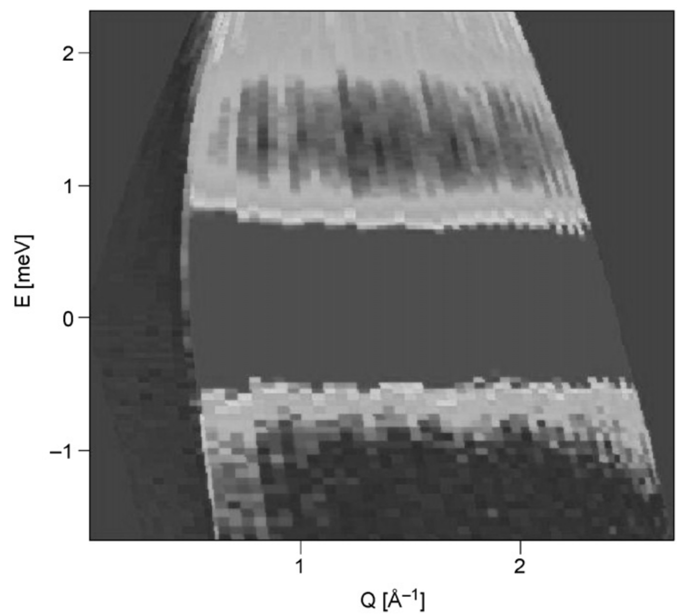

Fig. 3. Time-of-flight neutron scattering performed at DNS for $HoTbTi_2O_7$ at 10 K with a wavelength of 4.75 Å.

is observed at ~1.3 mV (not shown). This may resemble the intrinsic transitions of $Ho_2Ti_2O_7$ [5].

For summary, we prepared the polycrystalline pyrochlore samples of $Ho_{2-x}Tb_xTi_2O_7$ and studied magnetic and structural properties. No long-range order was observed in magnetic susceptibility, specific heat and the DNS measurements down to the lowest temperature. Specific heat and the DNS measurements, however, suggest a possible ground state of $HoTbTi_2O_7$, which shows both characteristics of spin-ice state ($Ho_2Ti_2O_7$) and spin-liquid state ($Tb_2Ti_2O_7$).

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