HoTbTi$_2$O$_7$, the mixtures of spin ice and spin liquid

L.J. Chang$^{a,b,*}$, H. Terashita$^b$, W. Schweika$^b$, Y.Y. Chen$^c$, J.S. Gardener$^{d,e}$

$^a$Nuclear Science and Technology Development Center, National Tsing Hua University, Hsinchu 300, Taiwan
$^b$IFF Scattering Methods, Research Center Jülich, 52425 Jülich, Germany
$^c$Institute of Physics, Academia Sinica, Nankang Taipei 115, Taiwan
$^d$Department of Physics, Brookhaven National Laboratory, Upton, NY, 11973-5000, USA
$^e$NIST Center for Neutron Research, Gaithersburg, MD, 20899-8562, USA

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Abstract

Polycrystalline samples of Ho$_{2-x}$Tb$_x$Ti$_2$O$_7$ ($x = 0.5, 1, and 1.5$) have been prepared and characterized. No long-range order is observed for HoTbTi$_2$O$_7$ in magnetization and specific heat measurements down to 2 K. The low-energy magnetic excitation measurements suggests that HoTbTi$_2$O$_7$ possesses both characteristics of spin ice and spin liquid in the ground state. © 2006 Elsevier B.V. All rights reserved.

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Pyrochlore oxides with a general formula A$_2$B$_2$O$_7$ (A: rare earth, B: transition metal) have attracted much attention due to the geometrical frustrations and interesting low temperature properties. Ho$_2$Ti$_2$O$_7$ had been reported to have a spin-ice configuration with a net ferromagnetic interaction explained by an Ising-type anisotropy model [1]. Tb$_2$Ti$_2$O$_7$ 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$_2$O$_7$, together with its structural data.

Polycrystalline samples Ho$_{2-x}$Tb$_x$Ti$_2$O$_7$ ($x = 0.5, 1.0,$ 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.

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 $\theta_{CW}$ 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 $10.29 \mu_B$, 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$_2$Ti$_2$O$_7$. 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...
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 \( \sim 30 \) 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 HoTbTi2O7 shows the characteristics of both spin ice and spin liquid below 30 K.

In addition, the low-energy excitations of HoTbTi2O7 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 \( \sim 1.3 \) mV (not shown). This may resemble the intrinsic transitions of Ho2Ti2O7 [5].

For summary, we prepared the polycrystalline pyrochlore samples of Ho2–\( x \)Tb\( x \)Ti2O7 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 HoTbTi2O7, which shows both characteristics of spin-ice state (Ho2Ti2O7) and spin-liquid state (Tb2Ti2O7).

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References