The pyrochlore lattice is a model for geometrical magnetic frustration. In rare earth pyrochlore titanates $R_2\text{Ti}_2\text{O}_7$, frustration arises from the subtle interplay of single ion anisotropy, exchange, and dipolar interactions. Depending on the balance between these factors, one may observe spin ice ($R=$Ho, Dy) [1] or spin liquid ($R=$Tb) behaviors, or complex magnetic orders stabilized by first order transitions ($R=$Er, Yb, Gd). A major role in the selection of the ground state is played by the single ion anisotropy that arises from the trigonal crystal electric field (CEF) at the rare earth site.

In the pyrochlore lattice, selection between Ising, Heisenberg, or XY models cannot be based, as usual, on the analysis of the macroscopic properties because of the presence of four anisotropy axes, namely, the <111> axes. Then only an average over the four local axes can be measured by classical methods, and no direct information can be obtained about the anisotropy of the local magnetic susceptibility of the $R$ ion, or of the local exchange or dipolar tensor. There is thus a clear motivation to get a direct access to these quantities and to quantify the axial or planar anisotropy in the rare earth pyrochlores.

We have shown that the site susceptibility approach [2] developed for polarized neutron diffraction is an accurate tool for obtaining the local susceptibility tensor $\chi$. The measured thermal variations of the components of $\chi$ are quantitatively compared with calculations using CEF [3]. To obtain good agreement with the data, one needs to introduce an anisotropic molecular field tensor, which takes into account both the first neighbor exchange and the dipolar coupling [4].

Single crystals of $\text{Ho}_2\text{Ti}_2\text{O}_7$, $\text{Tb}_2\text{Ti}_2\text{O}_7$, $\text{Er}_2\text{Ti}_2\text{O}_7$, and $\text{Yb}_2\text{Ti}_2\text{O}_7$ were grown by the floating-zone technique, using a mirror furnace. Their crystal structures were determined by zero-field neutron diffraction at 100 K and 5 K, within the space group $Fd-3m$.

Neutron diffraction measurements were performed at the ORPHEE reactor of the Laboratoire Léon Brillouin. Polarized neutron flipping ratios were measured on the Super-6T2 spectrometer using neutrons of incident wavelength $\lambda_n=1.4$ Å, and on the 5C1 spectrometer ($\lambda_n=0.84$ Å). For each compound, 100–200 flipping ratios were measured, at selected temperatures in the range 2–270 K, in a magnetic field of 1 T applied parallel to the [110] direction.

The thermal evolutions of $\chi_{||}$ and $\chi_{\perp}$ are shown in Fig.1–4 for the four compounds. In the insets are represented the susceptibility ellipsoids at 5 K (lower) and 250 K (upper), whose axes have been scaled by temperature to compensate for the Curie-Weiss decrease, together with the magnetic moments induced by the field applied along [110]. The data were interpreted using a single ion approach, where the trigonal symmetry CEF and Zeeman Hamiltonian is written as

$$H = \sum_{n,m} B_n C_n^m + g_J \mu_B J \cdot H$$
In order to take into account exchange and dipole-dipole interactions, we introduced an anisotropic molecular field tensor \( \{ \lambda_{||}, \lambda_{\perp} \} \) and assumed a field component of 1T either parallel or perpendicular to the ternary axis.

**Ho2Ti2O7 (Fig. 1):** At low temperature, the ellipsoid elongations \( \chi_{||} \) increase so markedly that they degenerate into needles, while \( \chi_{\perp} \) is very small, lying at the limit of experimental precision. These features are explained by the CEF level scheme of Ho

\[ \text{Ho}^{3+} \quad (J=8, g_J=5/4) \]

The ground state is very close to the extremely anisotropic non-Kramers doublet \( |J=8; J_z=\pm8> \), which has a moment of 10 \( \mu_B \) along the ternary axis and \( \chi_{\perp} = 0 \), and the first excited state lies at about 250 K above the ground state. For \( \chi_{||} \), introduction of a small positive (ferromagnetic) value \( \lambda_{||} = 0.05(1) \ T/\mu_B \) improves the agreement with the data.

**Tb2Ti2O7 (Fig. 2):** The CEF anisotropy of Tb

\[ \text{Tb}^{3+} \quad (J=6, g_J=3/2) \]

is considerably smaller than that of Ho

\[ \text{Ho}^{2+} \]

At room temperature, it is practically isotropic due to the relatively large number of populated CEF levels. Decreasing the temperature, it evolves progressively into an Ising type with a ratio of ellipsoid axes of the order of 10. Introducing an anisotropic \( \lambda \) tensor improves the agreement, especially for \( \chi_{\perp} \), with the AFM values: \( \lambda_{||} = -0.05(2) \ T/\mu_B \) and \( \lambda_{\perp} = -1.0 (2) \ T/\mu_B \). Both components of the \( \lambda \) tensor, which corresponds to an effective (exchange+dipole) low temperature interaction, are found to be of the AFM type.

**Er2Ti2O7 (Fig. 3):** The anisotropy of Er

\[ \text{Er}^{3+} \quad (J=15/2, g_J=6/5) \]

is rather weak, and a spherical shape for the magnetic ellipsoids is recovered above 100 K. At low temperature, \( \chi_{\perp} > \chi_{||} \), is in agreement with the planar magnetic ordering. We find the \( \lambda \) tensor is also anisotropic and of the AFM type: \( \lambda_{||} = -0.45(5) \ T/\mu_B \) and \( \lambda_{\perp} = -0.15(1) \ T/\mu_B \). The in-plane effective exchange is thus stronger than its component along the ternary axis, which reinforces the XY character determined by the CEF anisotropy.

**Yb2Ti2O7 (Fig. 4):** Yb

\[ \text{Yb}^{3+} \quad (J=7/2, g_J=8/7) \]

shows a planar anisotropy. Its magnetic ellipsoids are disk-shaped, like in Er2Ti2O7, but a strong anisotropy persists up to much higher temperature because the excited Kramers doublets lie much higher in energy (700–1000 K). A ferromagnetic value \( \lambda_{||} = 2.5(5) \ T/\mu_B \) is needed to reproduce \( \chi_{||}(T) \). The \( \lambda_{||} \) is much stronger than \( \lambda_{\perp} = 0 \), which results in a moment, at the temperature below 0.25 K, lying along the <111> axis, out of the rule of the strong CEF easy plane anisotropy.

In conclusion, polarized neutron diffraction allowed us to determine the local susceptibility tensor in the \( R_2Ti_2O_7 \) series, inaccessible by macroscopic measurements in single crystals. Its temperature dependence in the paramagnetic phase cannot be entirely accounted for by the sole crystal field anisotropy: a molecular field tensor \( \lambda \) must be introduced, which could be important for theory.

**References:**


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