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Magnetic correlations in pyrochlore spin ice as probed by polarized neutron scattering

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Abstract. Polarized neutron diffuse scattering experiments have been carried out on (1-10) and (111) oriented single crystals of the pyrochlore spin ice [1] compound $\text{Ho}_2\text{Ti}_2\text{O}_7$ in order to investigate the complex spin correlations in this material at low temperature. The incident neutron beam was polarized perpendicular to the horizontal scattering plane. Thus, the corresponding components of the spin correlations in and out of the scattering plane can be probed with spin-flip (SF) and non-spin-flip (NSF) scattering respectively via neutron polarization analysis. A strong anisotropy of the in-plane and out-of-plane spin scattering has been observed in both samples. The temperature dependence of magnetic diffuse scattering has also been measured. Here we concentrate on reporting the experimental results. In addition, a brief discussion of the possible nature of spin correlations in this pyrochlore spin ice will also be given.

1. Introduction

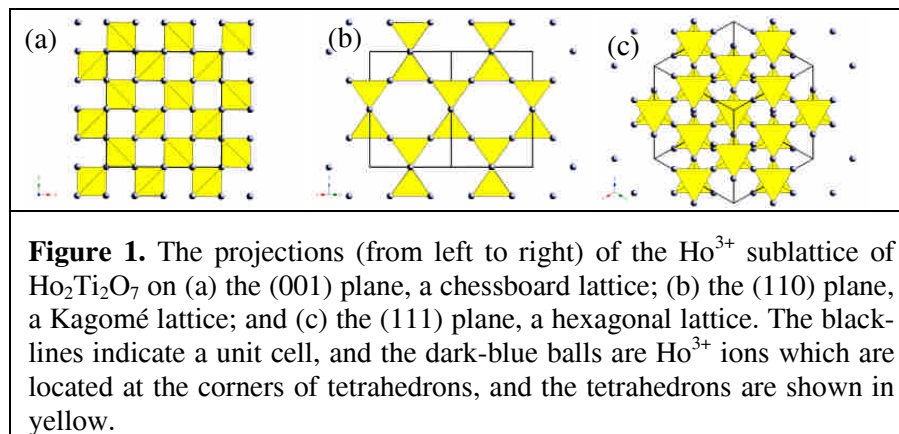
Geometrically frustrated magnets, in which the subtle balance between the energy terms of the exchange, dipolar interactions, single ion anisotropy, and the special local lattice geometries such as triangles or tetrahedrons can drive the systems into novel types of quantum-mechanical magnetic ground states with no classical equivalent, have attracted a great deal of attention from both experimental and theoretical physicists [1-13]. Pyrochlore oxides are regarded as ideal candidates for studying geometrically magnetic frustration in three dimensions because of their unusual structure of corner-sharing tetrahedrons which can lead to remarkable low-temperature magnetic properties [3]. These properties include a collective paramagnetism or spin liquid behavior [2,4-6], spin glass behavior but with very little structural disorder [7-9], and perhaps the most interesting and extensively studied the spin ice behavior [10-13].

$\text{Ho}_2\text{Ti}_2\text{O}_7$, $\text{Ho}_2\text{Sn}_2\text{O}_7$ and $\text{Dy}_2\text{Ti}_2\text{O}_7$ have been identified as spin ice compounds [13-16]. As in the case of water ice, the spin ice has a degeneracy of energetically preferred states which generates a residual finite entropy as the temperature approaches absolute zero [17]. The magnetic ground-states' organizing principles, or "ice rules", require that two spins should point in, and two out of each elementary tetrahedron in the lattice occupied by the rare earth atoms. The "two in, two out" ice rule was generally believed to be the result of a delicate balance between the interaction of the spins and the special geometry of the pyrochlore lattice. Theoretical simulations by Bramwell *et al.* suggested that $\text{Ho}_2\text{Ti}_2\text{O}_7$ can be described using a dipolar spin ice model [1], which indicates that the "ice-rule" has the configuration of two spins in and two spins out at the tetrahedral corners, and is from [111]

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anisotropy and ferromagnetic next neighbor coupling. In recent neutron diffuse scattering studies on the spin ice compounds, scattering intensities on the zone boundary have been observed for the spin ice $\text{Dy}_2\text{Ti}_2\text{O}_7$ [18]. A cluster-like correlation model, which was used to explain the zone boundary scattering intensities in the spinel ZnCr_2O_4 [19], was also used to understand the scattering intensities on the zone boundary in spin ice. However, Yavors'kii *et al.* showed that an equally satisfactory description is obtained via a microscopic theory in which the interaction terms in the Hamiltonian of the Monte Carlo simulations are fine-tuned [20] when includes the second and third nearest neighbor exchange couplings.

Pyrochlore oxides with the chemical formula $\text{A}_2\text{B}_2\text{O}_7$, where A is a rare-earth ion and B is a transition-metal ion, have a face-centered-cubic structure with space group $Fd-3m$ and eight molecules of its general formula in a unit cell. In the spin ice $\text{Ho}_2\text{Ti}_2\text{O}_7$, the Ho^{3+} ions occupy the $16d$ site with a local trigonal symmetry $3m$, and are the only magnetic ions in the specimen. Fig.1 shows the projections of the Ho^{3+} sublattice of $\text{Ho}_2\text{Ti}_2\text{O}_7$ on (a) the (001), (b) the (110), and (c) the (111) planes; the black-lines indicate a unit cell, and the dark-blue balls are Ho^{3+} which are located at the corners of tetrahedrons which are shown in yellow. The projection on the (001) plane displays a chessboard lattice; the (110) plane a Kagomé lattice; and the (111) a hexagonal lattice. These three orientations display the possible anisotropies in a cubic pyrochlore lattice.

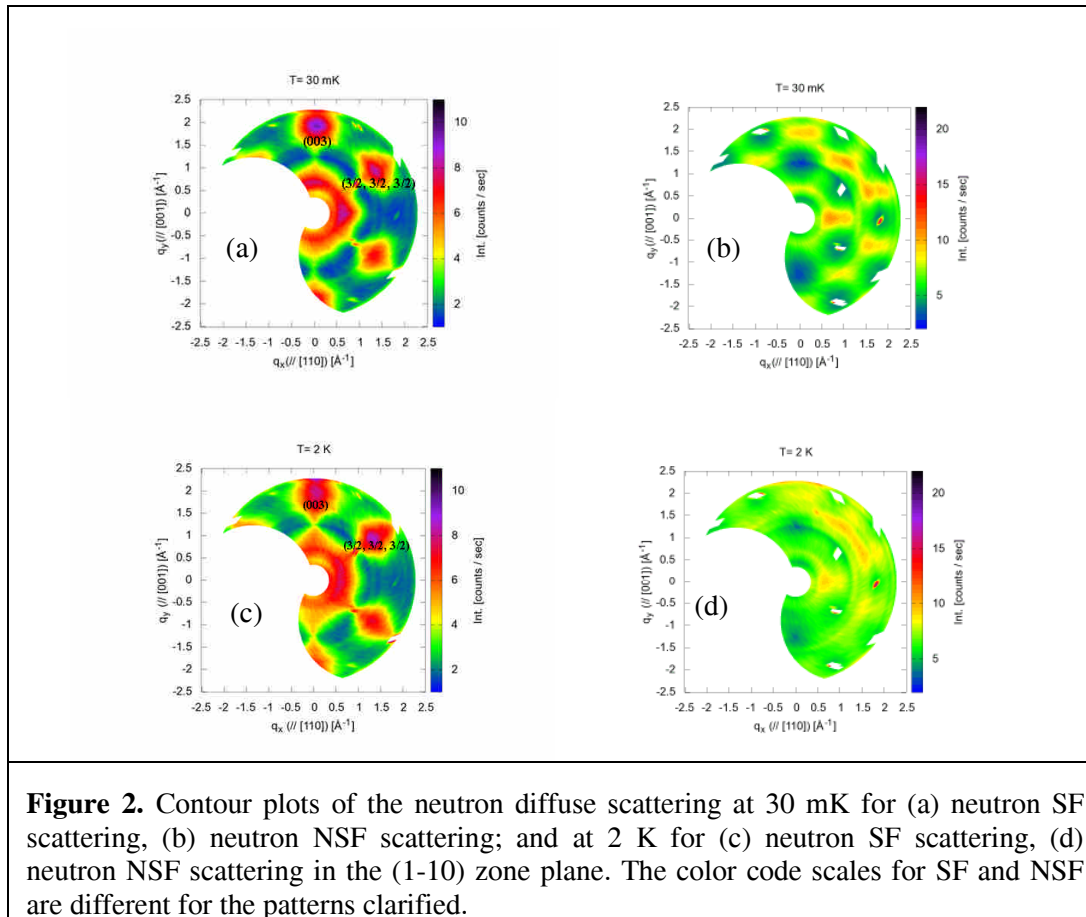


Neutron diffuse scattering is a powerful technique to probe short-range spin correlations in frustrated magnets, such as the pyrochlore spin ice. By combining this technique with longitudinal neutron polarization analysis, it is possible to separate the contributions of the components of spin correlations in and out of the scattering plane. Here we report the experimental results of neutron polarization analysis on the diffuse scattering for the single crystals $\text{Ho}_2\text{Ti}_2\text{O}_7$ aligned in the (1-10) and the (111) orientations.

2. Experimental

A $\text{Ho}_2\text{Ti}_2\text{O}_7$ single crystal ~ 0.6 cm in diameter, ~ 2 cm in length, and amber-red in color was grown in an infrared double mirror image furnace. The details of the crystal growth are given in reference [21]. Pieces of the crystals were cut, oriented by x-ray Laue diffraction, and mounted on an aluminum sample stick so that either the [1-10] or [111] zone-axis was perpendicular to the horizontal scattering plane. Neutron diffuse scattering was carried out at the newly commissioned polarized diffuse scattering spectrometer DNS at FRM-II. A $^3\text{He}/^4\text{He}$ dilution insert was used for the measurements in the mK range. The high-temperature data ($T > 1.7$ K) were obtained with a standard closed cycle cryostat. A neutron wavelength of 4.74 \AA was selected for all the experiments. Magnetic diffuse scattering with z-direction polarization analysis was used to separate the contributions from the in-plane (neutron SF scattering), and out-of-plane i.e. along the [1-10] or [111] zone-axis (neutron NSF

scattering) components of the spin correlations. The details for the neutron polarization analysis technique are described in reference [22].



3. Results and Discussion

Contour plots of neutron diffuse scattering at 30 mK are shown in Fig. 2(a) SF scattering, 2(b) NSF scattering in the (1-10) zone plane at 30 mK. The residual intensities at nuclear scattering positions for SF channel are due to the imperfect neutron polarization in the experiments. In Fig. 2(a), the results correspond to the magnetic correlations projected on the (1-10) plane, and are very similar to the simulated pattern for the dipolar spin ice model [1] calculated from three dimensional magnetic correlation. There is an unusual diffuse scattering related to long range spin correlation, along transverse Q at pinch points. The NSF scattering shown in Fig. 2(b) reveals the correlations among spin components perpendicular to the (hhl) plane. The zone boundary scattering and/or strong antiferromagnetic diffuse scattering was observed at the positions of $h = k = \text{odd}$ and $l = \text{odd}$. The strong intensity at (003) and by comparison the weaker features seen at (3/2, 3/2, 3/2) in Fig. 2(b) are consistent with the relative intensities predicted by the dipolar spin ice model for the region around (003) and (3/2, 3/2, 3/2), and the spread of the broad features along at (3/2, 3/2, 3/2) [1]. Yavors'kii *et al.* indicated that the zone boundary scattering pattern can be interpreted using either an independent small scale hexagonal spin cluster model [20], or an extensive generalized dipolar spin ice model (g-DSM), which has included the second and third nearest neighbor exchange couplings in the Monte Carlo simulations for another spin ice compound $\text{Dy}_2\text{Ti}_2\text{O}_7$. Pinch points were argued as the main differences between these two models. However, it is worth noting that the diffuse scattering patterns of g-DSM around (1/2, 1/2, 1/2) and (3/2, 3/2, 3/2) shown in reference [20] are made up of three

separate features. Our results shown in Fig. 2(b) also contain three separate features at these two positions, although the antiferromagnetic correlations for Ho^{3+} are not predicted in this g-DSM model. Fig. 2(c) shows the contour plot for the SF scattering in the (1-10) zone plane at 2 K. This pattern is reminiscent of the nearest neighbor spin ice model simulated by Bramwell *et al* [1]. The main differences between dipolar and nearest neighbor spin ice models are the four intense regions around (000) in the dipolar model, the relative intensities of the region around (003) and $(3/2, 3/2, 3/2)$, and the spread of the broad features along the diagonal [1]. With the exception of the variation of intensity around (000), which we cannot probe due to the limited q range in our experiments, Fig. 2(a) and Fig. 2(c) reflect these differences. The diffuse scattering pattern for the NSF scattering at 2 K is not clear (see Fig. 2(d)). This means that the projected magnetic correlations perpendicular to the (hhl) plane are very weak, or that the fluctuations out of the plane are faster than our measurements at this temperature. Fig. 2(c) and Fig. 2(d) seem to reveal the two-dimensional character of spin correlations in the (hhl) plane at 2 K at the first sight. However, the same magnitude of spin components in plane and out of plane, leading to the SF and NSF intensities respectively, is expected from the cubic symmetry of the compound. The diffuse scattering is clearly more pronounced in the SF scattering may be explained, for example, that in a projection along one edge of the tetrahedral, and within a tetrahedral, the vertical components in the nearest neighbor spin correlations is equal to zero in average for the ice-rule. A further study is necessary for clarifying.

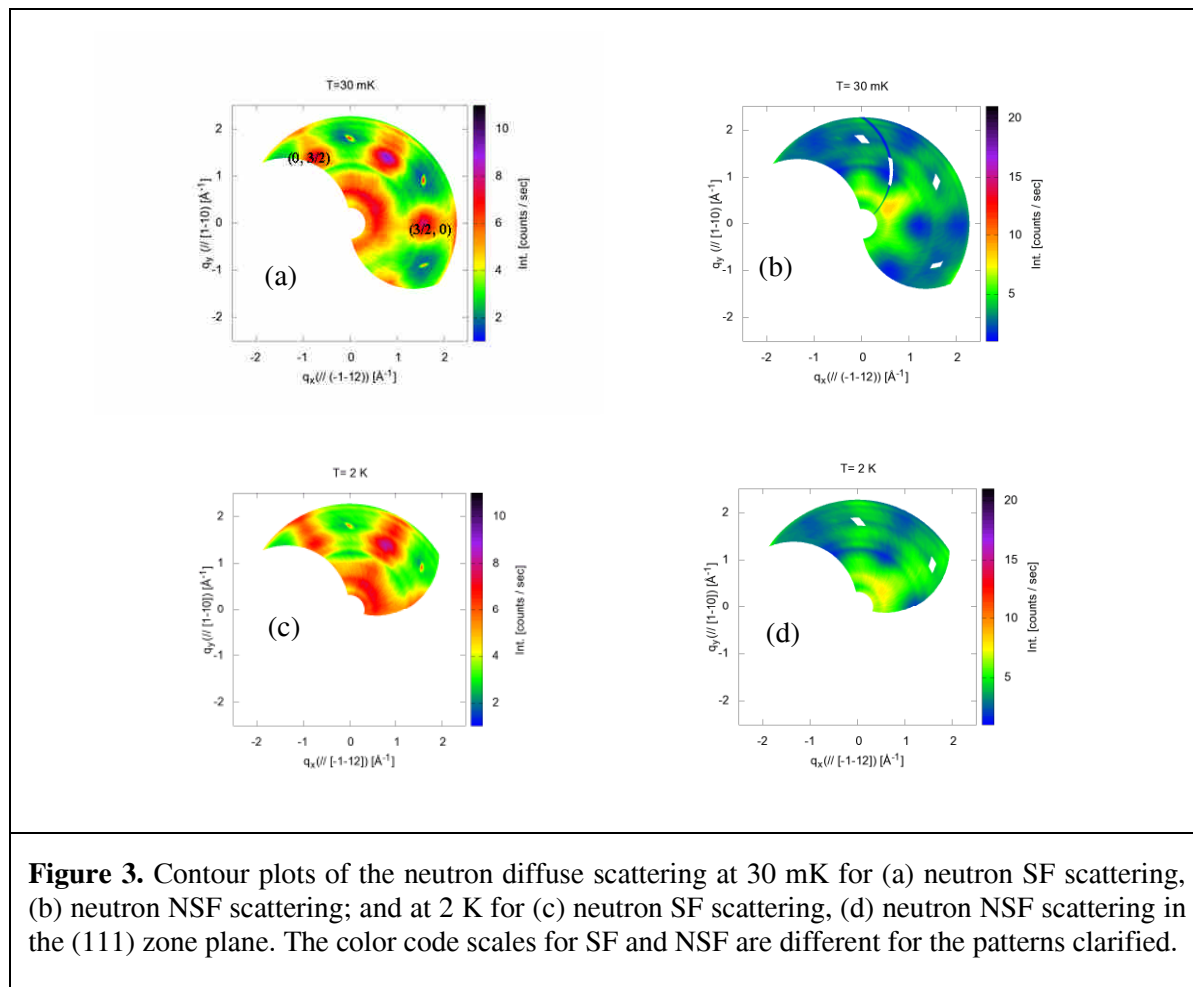


Figure 3 (a) – (d) manifest the SF and NSF scatterings in the (111) zone plane at different temperatures. In Fig. 3(a) and 3(b), a hexagonal and a stellate pattern were displayed around the center

(000) for SF and NSF respectively at 30 mK. Strong diffuse scattering intensities were also observed at (3/2,0), (3/2,3/2), and (0,3/2) in Fig. 3(a) when the diffraction plane is indexed according to a hexagonal lattice symmetry. Fig. 3(c) and 3(d) shows the contour plots of the SF and NSF scattering respectively at 2 K. These patterns are very similar to those shown in Fig. 3(a) and Fig. 3(b) for 30 mK except for the weaker overall intensities. The results are different to what we observed in the (1-10) scatterings, which display different patterns at 30 mK and 2 K in both SF and NSF channels. These results suggest that more detailed models are required to fully understand the spin ice freezing process and to completely explain the nature of the magnetic correlations in this material. Theoretical simulations and calculations are under way to investigate the magnetic correlations in the (111) orientation.

4. Conclusion

We have investigated the complex magnetic correlations in spin ice $\text{Ho}_2\text{Ti}_2\text{O}_7$ crystals aligned in the (1-10) and (111) orientations via polarized neutron diffuse scattering experiments. In the neutron SF scattering, the diffuse patterns suggest that there is a crossover from a regime above 2 K where nearest neighbor exchange interactions are most important to a dipolar-dipolar dominated regime below ~2 K. However, in the NSF scattering, a clear diffuse scattering at the Brillouin zone boundary was observed below 2 K. The NSF scattering patterns are also reminiscent of the zone boundary diffuse scattering pattern proposed in reference [20], in which the second and third nearest neighbor exchange interactions (the g-DSM model) [20] or the independent small scale hexagonal spin cluster model [19] need be taken into consideration. Further neutron diffuse scattering studies for different crystal orientations and non-magnetic doped specimens are processing to better understand the magnetic correlations appearing in the NSF scattering channel.

After submission of this paper the work of Fennel *et al.* appeared for scattering near pinch points which are indicative of separated dipoles and resemble the behavior of magnetic monopoles [23].

Acknowledgements

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