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Structural and magnetic disorder in defect spin ice $Ho_2Ti_{1.5}Sc_{0.5}O_{6.75}$

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Geometrically frustrated systems have been a topic of discussion for a number of years with particular interest in quantum spin liquids, states of quantum magnets in which electronic spins reside in macroscopic superpositions of infinitely many microstates, and spin-ice, with an important example being the pyrochlore $Ho_2 Ti_2O_7$ [1]. The Ho ions of this sample are arranged in corner-sharing tetrahedra with an O ion, commonly labelled as O(1), in the centre of the tetrahedra, and every pair of Ho ions having a nearest neighbour O, commonly labelled as O(2), outside of the tetrahedra. The ground state configuration of this material is with two spins pointing in (towards the O) and two out (away from the O) of the tetrahedra. This two-in-two-out configuration is obeyed over a very long range at low temperatures, resulting in the appearance of bow tie like structures in reciprocal space called pinch points. As the temperature is increased, these pinch points broaden due to the emergence of magnetic monopoles [2], tetrahedra violating the two-in-two-out rule. Recently it has been predicted that for spin-ice materials with non-Kramers ions, such as $Ho_2Ti_2O_7$, it is possible to tune between classical and quantum spin liquid behaviour with long-range entanglement via the controlled introduction of structural disorder [3].

Measurements on highly defective single-crystal $Ho_2Ti_{1.5}Sc_{0.5}O_{6.75}$ at a temperature of 50 mK were performed using the diffractometer D7 at the Institut Laue-Langevin. The capability of this instrument to use polarized neutrons was used to properly separate the magnetic and nuclear contributions. Density functional theory calculations were performed and compared with the nuclear diffuse scattering data. We find that the oxygen vacancies are on the O(2) sites next to pairs of rare-earth ions. Crystal electric field measurements and point charge model calculations show that for this structural defect the nearest neighbouring H0 ions have non-magnetic singlet ground states [4]. The magnetic diffuse scattering data showed well-defined pinch points in agreement with Monte Carlo modelling and as predicted for the removal of spins from spin ice [5].

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