

# Slow and static spin correlations in $\text{Dy}_{2+x}\text{Ti}_{2-x}\text{O}_{7-\delta}$

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**Abstract.** The static and dynamic spin correlations in the spin-ices  $\text{Dy}_{2.3}\text{Ti}_{1.7}\text{O}_{6.85}$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$  have been studied in polarised neutron diffraction and neutron spin echo experiments. The measurements reveal that, below 100 mK, the magnetic scattering broadens and shifts to higher  $|\mathbf{Q}|$  upon stuffing the pyrochlore lattice with additional  $\text{Dy}^{3+}$  ions. These observations can be related, by means of reverse Monte Carlo simulation, to the modified distribution of near neighbour distances and an overall more antiferromagnetic character of the near neighbour couplings. The dynamic measurements show that the spin correlations are slower in the stuffed system. These results will be discussed and compared to the holmium analogues.

## 1. Introduction

Geometrically frustrated magnetism has been studied by physicists for several decades now because it allows for the possibility of exotic ground states. [1] This usually refers to the inability to form a ground state with long range order at sufficiently low temperature, because the spatial arrangement of the magnetic ions on the underlying lattice is incompatible with near neighbour couplings. [2]

Face centered cubic pyrochlore oxides display a particularly rich variety of unusual phenomena at low temperature which are related to geometrically induced frustration. Earlier studies have found partially ordered states in  $\text{Gd}_2\text{Ti}_2\text{O}_7$  and  $\text{Tb}_2\text{Sn}_2\text{O}_7$ , [3, 4] a order by disorder transition in  $\text{Er}_2\text{Ti}_2\text{O}_7$ , [5] and spin ice behavior in  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$ . [6, 7] Spin glass states with very little, or no, positional disorder have been found in  $\text{Y}_2\text{Mo}_2\text{O}_7$  [8],  $\text{Tb}_2\text{Mo}_2\text{O}_7$ , [9, 10] and  $\text{Sr}_2\text{CaReO}_6$ , an ordered perovskite with face centered cubic symmetry. [11]

Frustrated magnets also possess unusual dynamical properties, [12] for example the  $\text{Tb}^{3+}$  spins in  $\text{Tb}_2\text{Ti}_2\text{O}_7$ , [13, 14] are still very dynamic at 17 mK,  $\text{Yb}_2\text{Ti}_2\text{O}_7$ , [15] has a first order transition which prematurely ends before the sample freezes and spin ice compounds have been shown to host magnetic monopoles which propagate through the sample at low temperatures. [16, 17, 18, 19, 20]

The spin ices stand out as unique examples for frustrated systems with effective *ferromagnetic* interactions and have therefore been studied extensively. [21, 22, 23, 24, 25, 26, 27, 28] Besides thermodynamic variables such as temperature and magnetic field, the chemical composition of the systems may be altered to study the relationship of structural and magnetic properties. To this end, ‘stuffed’ spin ice (SSI) materials with general formula  $\text{R}_{2+x}\text{Ti}_{2-x}\text{O}_{7-\delta}$  ( $\text{R} = \text{Ho}$  or  $\text{Dy}$ ) have been synthesized, in which additional magnetic ions replace the non-magnetic  $\text{Ti}^{4+}$  ions and creating new magnetic pathways ( $\delta > 0$ , as the oxygen content needs to be adjusted for charge balance). [29, 30, 31] Due to the way the R and Ti sublattices penetrate each other, one creates additional neighbours for a fraction of the magnetic R ions as one *stuffs* the lattice. These new neighbours are at a distance of  $\sim 3.6 \text{ \AA}$ , equal to the length of the A-site tetrahedron. The additional magnetic exchange pathways represent a major disturbance of the magnetic system and introduce positional disorder. Surprisingly however, it has been found that the holmium ‘stuffed’ spin ice systems have the same entropy per spin at low temperature as the ‘unstuffed’ spin ice. [29, 30, 31]. Ehlers *et al.* [31] also showed that although the magnitude of the relaxation times changed with stuffing, the general characteristics associated with spin ice, and monopole formation, were qualitatively the same up to  $x = 0.3$ . [31, 32]

Ueland *et al.* [33] studied the low temperature thermodynamic properties of stuffed dysprosium titanate,  $\text{Dy}_{2+x}\text{Ti}_{2-x}\text{O}_{7-\delta}$ . They found that the additional Dy - Dy pathways released the zero point entropy completely for  $x \geq 0.3$ , which is significantly different to the results from  $\text{Ho}_{2+x}\text{Ti}_{2-x}\text{O}_{7-\delta}$ . [29] They also measured the magnetic susceptibility at millikelvin temperatures of both  $\text{Ho}_{2.67}\text{Ti}_{1.33}\text{O}_{6.67}$  and  $\text{Dy}_{2.67}\text{Ti}_{1.33}\text{O}_{6.67}$ , finding evidence of persistent spin dynamics, where it was noted that the Ho based compound was less susceptible at 100 mK. [29]

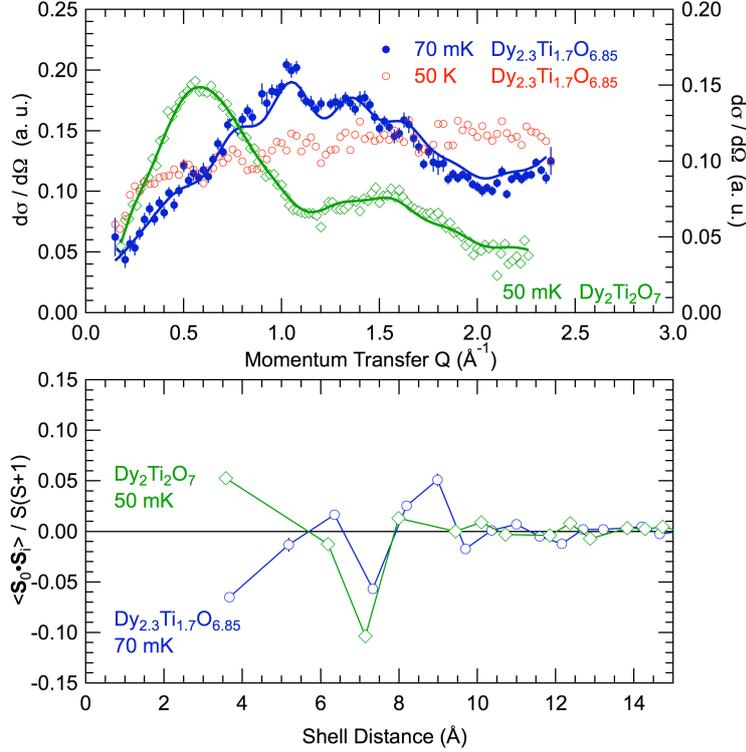
## 2. Experimental

In this paper we present results of neutron scattering experiments from Dy based pyrochlores. Due to the high absorption cross section of naturally occurring Dy, we studied small ( $\approx 500 \text{ mg}$ ), polycrystalline samples with  $x = 0$  and  $x = 0.3$ , enriched in the less absorbing  $^{162}\text{Dy}$  isotope. These were prepared and characterized using procedures described elsewhere. [30] Neutron scattering experiments were performed at several instruments including the D7 polarised time-of-flight spectrometer at the ILL using a wavelength of  $4.8 \text{ \AA}$  and the spin echo spectrometers IN11 and IN15 also at the ILL using  $5.5 \text{ \AA}$  and  $6.5 \text{ \AA}$  neutrons respectively. Both spectrometers had a 15 %  $\Delta\lambda/\lambda$ . To investigate the slower spin dynamics, we extended the dynamical range studied in  $\text{Dy}_2\text{Ti}_2\text{O}_7$  using NG5NSE at the NCNR using  $6.0 \text{ \AA}$  neutrons and a 17 %  $\Delta\lambda/\lambda$ . In each experiment the sample was mounted in a dilution refrigerator and cooled to base temperature,  $T > 50 \text{ mK}$ . NSE has been used successfully for studies of frustrated magnets, [12, 34] where a significant amount of spectral weight from low energy excitations exists.

## 3. Results

High quality, polycrystalline samples were shown to be single phase – no evidence of a fluorite phase was found in X-ray diffraction – and to have the high temperature bulk properties similar to those already published. [29]

The top panel of Fig. 1 shows the magnetic scattering of both samples at  $T < 70 \text{ mK}$  and for  $\text{Dy}_{2.3}\text{Ti}_{1.7}\text{O}_{6.85}$  at 50 K as obtained at D7 by full  $xyz$ -polarisation analysis. At 50 K the magnetic scattering in stuffed Dy-spin ice is almost featureless, consistent with a weakly coupled spin system. The decrease in magnetic scattering as  $S|\mathbf{Q}|$  approaches  $0 \text{ \AA}^{-1}$  is consistent with



**Figure 1.** Upper panel: Quasielastic magnetic scattering and fit to  $d\sigma/d\Omega$  resulting from reverse Monte Carlo simulation. The broad diffuse scattering from both pure and stuffed Dy spin ice, taken below 70 mK and a 50 K data set from  $\text{Dy}_{2.3}\text{Ti}_{1.7}\text{O}_{6.85}$ . Lower panel: time averaged spin correlations for near neighbour shells. Error bars represent  $\pm 1\sigma$  from counting statistics.

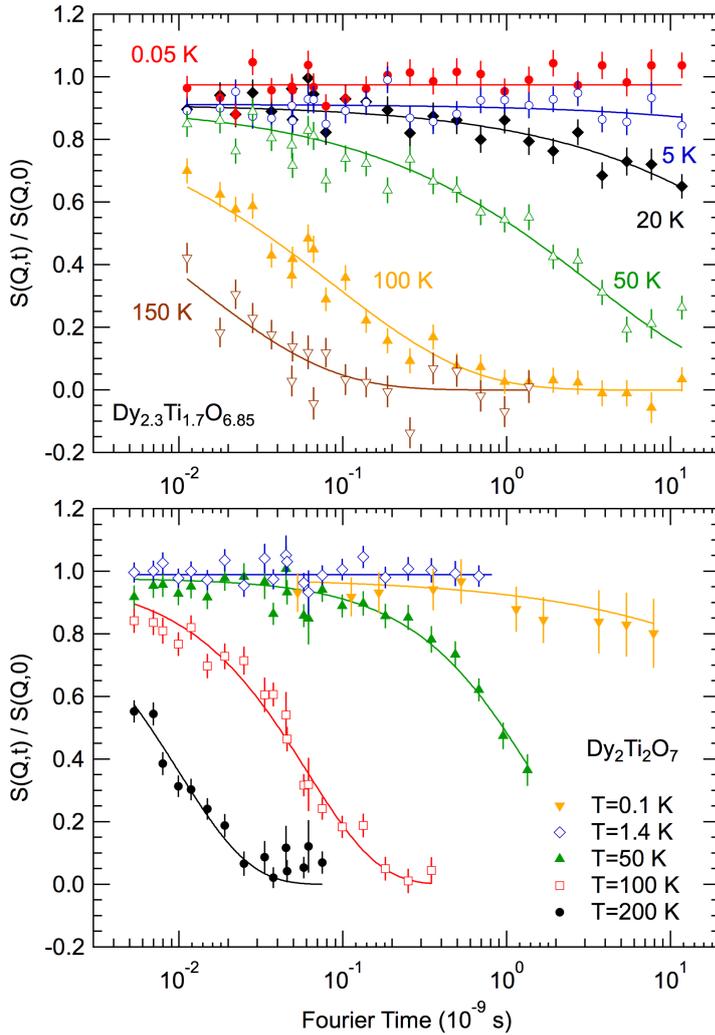
some weak antiferromagnetic correlations still persistent at these elevated temperatures. Similar results have been seen in  $\text{Ho}_2\text{Ti}_2\text{O}_7$  where magnetic correlations, albeit weak, were seen up to 800 K. [27] As the temperature is lowered the magnetic structure factor monotonically increases from  $S|\mathbf{Q}| = 0 \text{ \AA}^{-1}$  to a maximum at  $\approx 1.05 \text{ \AA}^{-1}$ . This is significantly different to the pure spin ice, where  $S|\mathbf{Q}|$  peaks at  $0.55 \text{ \AA}^{-1}$ , and is consistent with the more antiferromagnetic nature of the coupling first found by Ueland *et al.* [33] The scattering from  $\text{Dy}_2\text{Ti}_2\text{O}_7$  is also considerably sharper than that in the stuffed compound, indicative of a longer correlation length, but neither samples shows magnetic Bragg peaks, indicating the absence of true long range magnetic order.

These data were analysed using a reverse Monte-Carlo (rmc) technique. For powder-averaged data, the scattering arising from the correlation of a given magnetic moment  $J_0$  with near neighbours arranged in shells at distances  $R_n$  can be written as

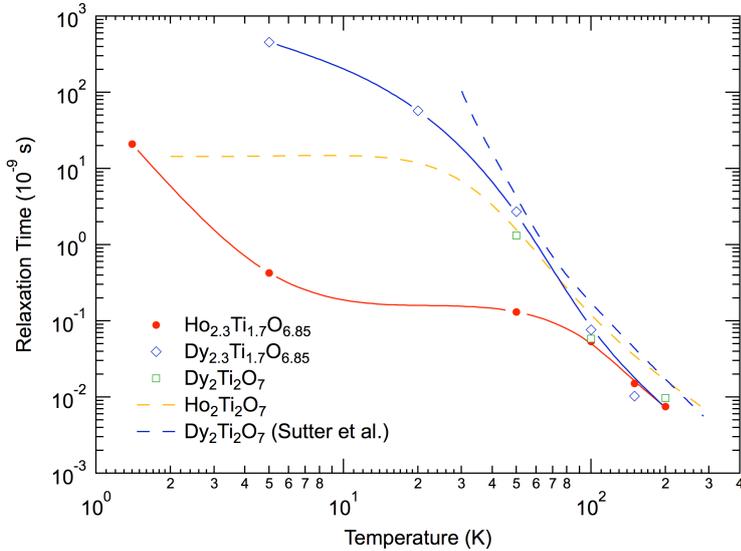
$$S(Q) = \text{const} \cdot f^2(Q) \cdot g_J^2 \cdot J(J+1) \times \left\{ 1 + \sum_n Z_n \frac{\langle \mathbf{J}_0 \cdot \mathbf{J}_n \rangle \sin(QR_n)}{J(J+1) QR_n} \right\} \quad (1)$$

where  $f(Q)$  is the magnetic form factor,  $J$  is the angular momentum quantum number,  $g_J$  is the Landé-factor and  $Z_n$  is the occupation in the  $n$ -th shell. The rmc simulation was performed on a cube of  $6 \times 6 \times 6$  unit cells with magnetic moments situated at all the  $16d$  (Dy) sites and an appropriate amount on the  $16c$  (Ti) site when stuffed. No preferred moment direction in the lattice was assumed. Starting with an arbitrary spin configuration, a small number

of spins was randomly chosen which were turned in one step of the simulation to a random new direction. The step was then accepted if the fit to the measured data was improved, and repeated if not. Many individual rmc runs (with typically  $\sim 10^5$  steps) were performed until convergence, and then linearly combined to produce the final best fit. The resulting fits, describe the data extremely well and are also shown in the upper panel in Fig. 1. The lower panel of Fig. 1 summarizes the rmc results for the shell-averaged couplings (note the additional shells in the  $\text{Dy}_{2.3}\text{Ti}_{1.7}\text{O}_{6.85}$  sample, due to the extra magnetic ions). The most striking result is that



**Figure 2.** NSE spectra of the  $x = 0.30$  (upper panel) and pure (lower panel) Dy-based spin ice samples. The data was measured near the maximum in  $S|\mathbf{Q}|$  for each sample ( $|\mathbf{Q}| = 1.0 \text{ \AA}^{-1}$  for  $x = 0.30$  and between  $0.3 \text{ \AA}^{-1}$  and  $1.1 \text{ \AA}^{-1}$  for the pure sample) to improve statistics. Like earlier spin ice measurements, it was found that the NSE spectra for the pure compound was  $S|\mathbf{Q}|$  independent. The lines are fits to exponential relaxation functions (see text), but clearly the stuffed sample is *stretched* ( $\beta = 0.52$ ), indicative of multiple relaxation rates, even at 150 K, where one would expect only single ion processes. At 5 K, the stuffed sample is  $\approx 90\%$  static at 10 ns and static at 50 mK. The pure sample looks static at 1.4 K, but low temperature measurements on NG5NSE showed that there are extremely slow dynamics even at 100 mK.



**Figure 3.** Temperature dependence of observed relaxation times as measured by NSE on several spin ice compounds. Note the regime with a small if not zero energy barrier (plateau in this figure) is not seen in the Dy based materials in this frequency and temperature window. Nuclear forward scattering results reproduced from Sutter *et al.* [39]

the nearest neighbour shell is antiferromagnetically coupled in the stuffed sample, with a small ferromagnetic component on the  $3^{rd}$  shell, whilst it is ferromagnetically coupled in the pure ice case.

The slow spin dynamics in both compounds are shown in Fig. 2. On the (0.01 to 10) ns timescale,  $\text{Dy}_{2.3}\text{Ti}_{1.7}\text{O}_{6.85}$  looks static with a normalised intermediate scattering function of 0.98(4) at 50 mK. By 500 mK (not shown), this falls to an average of 0.96(4) and 0.90(4) by 5 K. This loss of signal corresponds to very fast dynamics with little spectral weight, and may be attributed to paramagnetic (uncorrelated) Dy spins. Over this temperature range however, the  $\text{Dy}_{2.3}\text{Ti}_{1.7}\text{O}_{6.85}$  spectra show very little *relaxation*, indicative of very slow spin fluctuations, since there is at best a very slight downturn observable at long times. At higher temperatures the spectra relax fully within the NSE time window.

The lower panel shows the neutron spin echo spectra from pure  $\text{Dy}_2\text{Ti}_2\text{O}_7$  for comparison. At times shorter than 1 ns this sample looks static at 1.4 K. This is very different to pure  $\text{Ho}_2\text{Ti}_2\text{O}_7$  where the normalised intermediate scattering function is 0.95(1), even at 0.3 K. [26, 27] Data collected on NG5NSE, out to 10 ns, at 100 mK revealed a slow relaxation processes within this spin ice, comparable with  $\text{Dy}_{2.3}\text{Ti}_{1.7}\text{O}_{6.85}$  at 5 K.

On further examination of the data from the two samples, one also notes that the relaxation process within the stuffed compound is very drawn out. The pure pyrochlore compounds  $\text{Dy}_2\text{Ti}_2\text{O}_7$  and  $\text{Ho}_2\text{Ti}_2\text{O}_7$  [26, 27] relax with a single relaxation rate at all temperatures studied. Intuitively, creating local disorder may result in multiple relaxation processes and a stretched exponential relaxation, however diluting the magnetic A-site with non-magnetic Y or La, does not alter this relaxation process significantly. [36, 37, 38] Stuffing however does not only create local distortions but also extra exchange pathways at the nearest neighbour distance. Since the additional magnetic ions are inserted into the lattice randomly, that is around any given A-site, Dy can have between zero and six magnetic B-site neighbours. These extra pathways must be crucial for the creation of the multiple relaxation processes elucidated by the stretched exponential relaxation required to describe the stuffed spin ice data. Similar conclusions can be

made from  $\chi''(T)$ . [40] It should be noted here however that these data are very different from the spin liquid,  $\text{Tb}_2\text{Ti}_2\text{O}_7$  where a stretched exponential was also used to fit the low temperature data [41] but most of the magnetic signal relaxed within 5 ps and dilution broadens the relaxation functions further. [42]

Figure 3 summarizes the spin relaxation times observed in the spin ices in the nanosecond window. At high temperatures, where the relaxation process is dominated by a spin flip process by way of the first excited crystalline electric field state, all compounds have a relaxation time of  $\approx 10^{-2}$  ns. As the system cools to 2 K, it is observed that the Ho-based compounds are faster than the isostructural Dy-based counterparts but qualitatively their properties are the same. One possible explanation for this involves the *nuclear* spin system in the Ho-based materials. [43] Holmium has only one natural isotope with a large nuclear spin ( $I = 7/2$ ) which couples strongly through a hyperfine interaction and can speed up the relaxation of the electronic spin system. In naturally abundant dysprosium compounds, 40 % of the isotopes have a 5/2 spin. However, in our studies, using samples enriched in the  $^{162}\text{Dy}$  isotope, no more than 1% of the compound has a 5/2 spin. These differences should be studied further and kept in mind when comparing susceptibility and Dy-based neutron results. From this study, it has been argued that stuffing the lattice makes the time scales of the spin correlations become slower, thus one could imagine now looking at Ho/Dy solid solutions to secure the desired relaxation times to study the system with other methods, especially optical spectroscopies.

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