Experimental Report

Proposal: **5-41-762** **Council:** 4/2014

Title: Determination of the magnetic structure of the Shastry-Sutherland Magnet Neodymium Tetraboride

This proposal is a new proposal

Researh Area: Physics

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Samples: NdB4

Instrument	Req. Days	All. Days	From	To
D10	6	10	02/09/2014	08/09/2014
			17/09/2014	21/09/2014
D1B	2	1	03/09/2014	04/09/2014

Abstract:

NdB4 crystallises into a tetragonal structure, where the Nd ions for layers in the ab-plane that are topologically equivalent to the Shastry-Sutherland lattice. Other members show a range of novel and unusual results believed to be due to the competition between the magnetic and quadrupolar interactions in this systems. There are very few reports investigating NdB4, thus we propose to investigate the magnetic structure of NdB4 using the D10 spectrometer in zero applied field for 4 temperature, 20 K, 8 K, 6 K and at the base temperature of the cryostat. Boules of NdB4 have been produced and we are confident that single crystals will be grown soon.

The rare earth borides shows a diverse range of interesting and unusual behaviour. The rare earth tetraborides (RB$_4$) are particularly interesting, as they are a rare experimental realisation of the Shastry-Sutherland lattice (SSL). RB$_4$ crystallises into a tetragonal structure, where the R ions form layers of squares and equilateral triangles in the ab-plane which map to the SSL. Here the nearest neighbour ions form an orthogonal dimer lattice with in-built geometrical frustration [1]. It is believed the competition between the magnetic dipole and electric quadrupole interactions is crucial to establishing a ground state, giving rise to the variety of properties observed. However, the role geometric frustration plays is not yet fully understood [2]. Some of the properties observed in this family include fractional magnetisation plateaux [3, 4], unexpected by theory, complex phase diagrams [5, 6] and magnetic structures that can differ significantly between R ions [7, 8].

NdB$_4$ shows successive phase transitions at T_{N1} = 7 K and T_{N2} = 4.6 K for $H \parallel$ [001], with an additional transition at T_Q = 17.2 K for $H \perp$ [001]. It has been suggested that $T_{\mathrm{N1}} < T < T_Q$ is an ordered quadrupolar phase, $T_{\mathrm{N2}} < T < T_{\mathrm{N1}}$ is an incommensurate antiferromagnetic state, while $T < T_{\mathrm{N2}}$ is an ordered antiferromagnetic state [9]. Large single crystals of NdB$_4$ have been grown at Warwick using the floating zone method. Isotopically enriched boron, ^{11}B was used to reduce the absorption of neutrons. We have characterised the crystals using both temperature and field dependent magnetisation measurements. Interestingly our field dependent magnetisation shows a magnetisation plateaux corresponding to M/M_s = $1/5$ at 1.7 T. All of our results were consistent with previously published results [9].

We have carried out single crystal and powder neutron experiments on D10 and D1B respectively in order to determine the zero field magnetic structure. We were also given time on CYCLOPS in order to index the Laue patterns and find the hkl values of the incommensurate peaks. We observed incommensurate peaks of type $\mathbf{q}$ = (δ, δ, δ'), where δ = 0.15 and δ' = 0.4 in the intermediate range $T_{\mathrm{N2}} < T < T_{\mathrm{N1}}$. In the low temperature phase ($T < T_{\mathrm{N2}}$) we observed commensurate peaks of type $\mathbf{q}$ = 0 with additional incommensurate peaks of type $\mathbf{q}$=(0, δ'', δ'), where δ'' = 0.18. The temperature evolution of the intensity of these incommensurate peaks is shown in Fig. 1.

Refinement of the high temperatures phase ($T > T_Q$), has confirmed that NdB$_4$ crystallises into tetragonal structure, space group $P4/mbm$ and the fractional coordinates in agreement with previous measurements [10]. The comparison between the calculated ($F2calc$) and observed ($F2obs$) squared structure factor is shown in Fig. 2(a). The neutron Laue photographs for each phase using CYCLOPS are shown in Fig. 3. There is a slight difference between the high temperature photo and the phase between $T_{\mathrm{N1}} < T < T_Q$. However, we were unable to index the peaks so we cannot make any firm determinations of the structure or whether it is solely an ordered quadrupolar phase. Initial refinement of the intermediate temperature phase, $T_{\mathrm{N2}} < T < T_{\mathrm{N1}}$, has shown, due to a reduction of the symmetry, three inequivalent magnetic sites form, giving rise to a complicated helical structure. This result is not very convincing and further refinement is definitely needed.

The low temperature phase, $T < T_{\mathrm{N2}}$ showed the commensurate component orders into a simple antiferromagnetic structure with the spins aligned parallel to the c-axis. The incommensurate component shows there are two inequivalent magnetic sites for the Nd ions. These moments form an elliptical helix structure with moments rotating around the a or b axis. A comparison between $F2calc$ and $F2obs$ (Fig. 2(b)) suggests the structure is promising. These are our initial results and a great deal of work still needs to be done.

It would be interesting to investigate the phase between $T_{\mathrm{N1}} < T < T_Q$ in greater detail, so that we can determine the nature of the phase. Additionally the lab based measurements in field have shown some promising results, so extending the measurement to look at the field induced structure associated with the fractional magnetisation plateaux would be interesting.

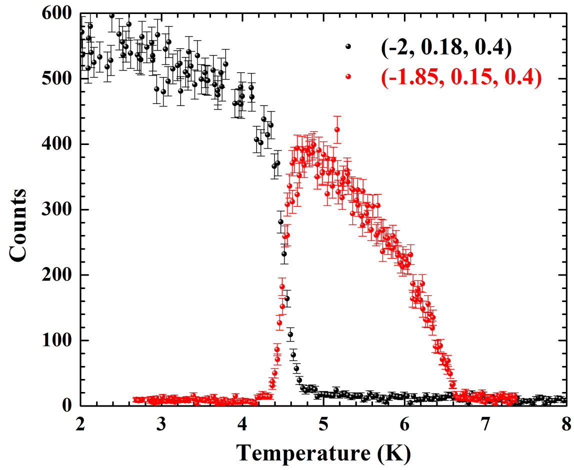

Figure 1: Temperature dependence of the intenesity of the two incommensurate peaks in NdB_4. Characterisation has shown the purely incommensurate phase occurs between T_{N1} =7.1 and T_{N2} =4.6, while the mixed commensurate and incommensurate phase occurs below T_{N2}, consistent with our results.

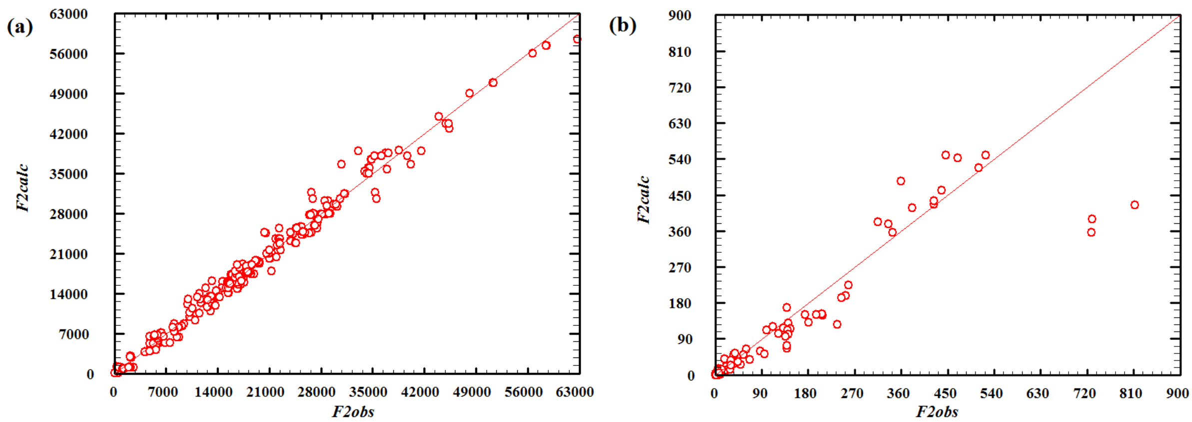

Figure 2: Comparison of the calculated and observed structure factors for (a) the high temperature (300 K) structure refinement and (b) the low temperature incommensurate component refinement.

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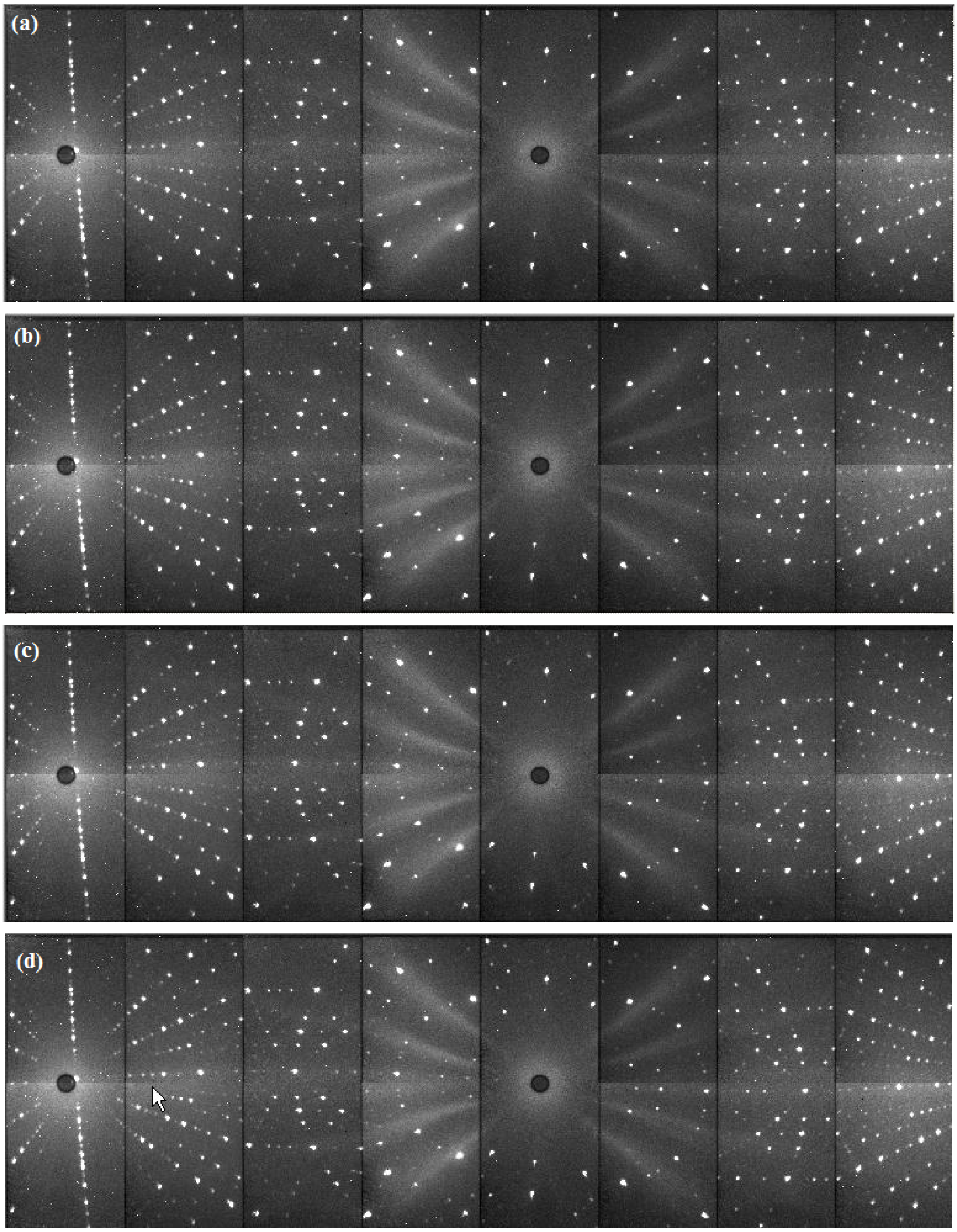

Figure 3: Neutron Laue photographs taken using CYCLOPS. The photos were taken at (a) T = 20 K, (b) T = 13 K, (c) T = 6 K and (d) T = 1.8 K.