Experimental report

Proposal:	DIR-275
Title:	Search for superstructure reflections in the magnetization plateau phases of TbB4
Research area:	Physics

This proposal is a continuation of 5-41-478

Council: 10/2022

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Samples: TbB4

Instrument	Requested days	Allocated days	From	To
IN22	8	8	11/05/2023	16/05/2023
			04/12/2023	10/12/2023

Abstract:

TbB4 has a tetragonal structure (P4/mbm) in which the network of Tb3+ ions lying in the c-plane is equivalent to the geometrically frustrated Shastry-Sutherland lattice (SSL). In zero-field, two successive antiferromagnetic transitions occur at $TN1=44$ K and $TN2=24$ K. Upon the application of a magnetic field along the c-axis, TbB4 is known for its devil-staircase-like magnetization process, stabilizing successive magnetization plateau phases for fields $16 ¿ mu0H ¿ 28$ T. Thanks to recent single crystal neutron diffraction studies performed in pulsed magnetic field up to 35 T, it has been possible to propose several magnetic models for the three magnetization plateau phases $M/MS = 2/9, 1/3$ and $½$ in agreement with the observed intensities of three integer Bragg peaks. In order to distinguish those models, we propose now to use the same experimental set-up at 2 K to search for their characteristic superstructure reflections at the positions (d d 0), (0 d 0) and (0 0 d), with d being 1/9, 1/3 or ½ (depending on M/MS).

Experimental report for DIR-275: Search for superstructure reflections in the magnetization plateau phases of TbB$_4$

IN22, May and Dec. 2023

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Scientific background:

TbB$_4$ belongs to the family of tetraborides which crystallize in the tetragonal space group *P4/mbm* and can be mapped to the Shastry-Sutherland lattice [1], a well-known example of a geometrically frustrated system with an exact ground state solution. The tetraborides have gathered a lot of interest due to their diverse magnetic properties ranging from simple antiferromagnetic structures with magnetic moments along the *c* axis for ErB$_4$ [2, 3] and DyB$_4$ [2] or within the *a-b* plane for GdB$_4$ [4, 5] and TbB$_4$ [3, 6, 7], but also more complex magnetic structures like in HoB$_4$ [8] and TmB$_4$ [9, 10]. The particularly exciting physics arise upon applying a magnetic field which induces a cascade of magnetic phase transitions related to fractional magnetization plateaus [5, 11-13].
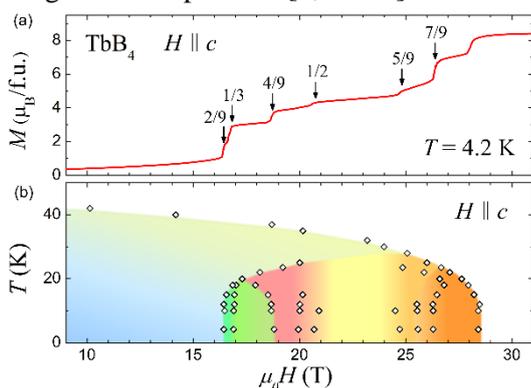
In the particular case of TbB$_4$, magnetization and magnetostriction measurements [12] performed upon the application of a high magnetic field perpendicular to the magnetic easy plane revealed a series of successive magnetization plateau phases and a complex phase diagram (Fig. 1).

Figure 1 from [15]: (a) Magnetization vs magnetic field at $T = 4.2$ K for an applied magnetic field along the *c* axis. (b) Temperature vs magnetic field phase diagram deduced from these magnetization measurements (both adapted from [12]).

Following neutron diffraction experiments [14] in pulsed magnetic fields up to 30 T, a model, consisting of *XY*-and Ising units, was proposed for the magnetic structure of the half-magnetization plateau $M/M_S = ½$ phase. Our recently published results [15] are in agreement with this model. In addition, the improved time resolution and better control of the sample temperature during the long-duration field pulses of our experiment allowed us to analyze not only the $M/M_S = ½$ state, but also the 1/3 and 2/9 phases. We were then able to propose an extension of the *XY* + Ising model to the different magnetization plateaus in agreement with the observed intensities of three integer Bragg peaks. This model consists of diagonal stripes of polarized spin plaquettes with an increased density of stripes proportionally to the macroscopic magnetization. However, even though this model explained the field-dependence of the scattered neutron intensities, a set of alternative models exists which differs concerning the modulation but yields exactly the same intensity distribution on the integer Bragg reflections.

To distinguish between these different models, we proposed to search for their characteristic superstructure reflections at Q = (δ δ 0), (0 0 δ), and (δ 0 0) with δ being 1/9, 1/3 or ½ (depending on M/M_S) by means of single-crystal neutron diffraction in pulsed magnetic fields up to 32 T. A single crystal of TbB$_4$ enriched with ^{11}B (99.52%) with a plaquette-like shape (with **c** normal to the platelet) and dimensions of roughly 2 x 2 x 1 mm^3 has been used.

The experiment was performed on the IN22 spectrometer (CRG-ILL) equipped with the pulsed field 2 K /40 T cryomagnet [16] and operated in a double-axis mode, without analyzer. A wavelength of 0.97 Å ($k_i = 6.1$ A^{-1}) provided by the (002) reflection of a pyrolytic graphite monochromator was used.

For this experiment, the sample was mounted with [1 1 0] and [0 0 1] in the scattering plane with the field parallel to **c**, giving us the possibility to investigate the positions (δ δ l) and (h h δ). We first checked the sample's orientation at room temperature on the strong nuclear peaks (1 1 0) and (2 2 0). We cooled down the sample to base temperature $T = 2$ K and measured the field dependence of their peak intensities, accumulating for each reflection a few tens of field pulses to get enough statistics.

Most of the time of the experiment was then dedicated to the search of field-induced diffracted signal at the positions (δ δ l) and (h h δ). Main results are summarized in fig. 2 (numerous other positions were investigated but are not reported here). Magnetic intensity was not found on (δ δ l) as shown on fig 2(a) and (b), but on the (h h δ) positions (fig 2(c)) which suggests a stacking model along the **c**-axis. The intensity was observed for δ = 1/3, 4/9 and ½ for fields covering the range of the 1/3, 4/9 and 5/9, and ½ magnetization plateau phases, respectively.
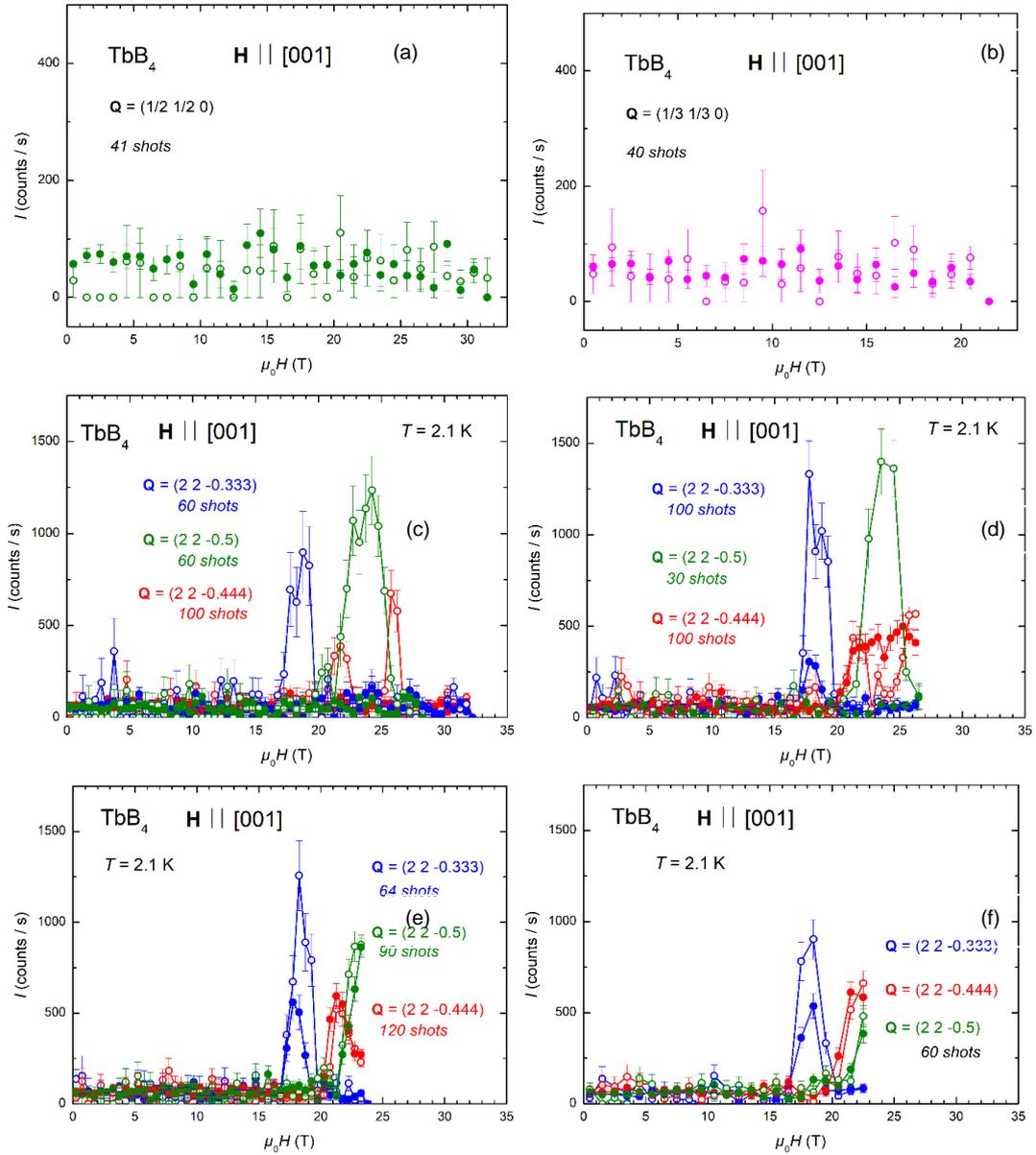
Figure 2: Magnetic field dependence of neutron diffracted intensities at: (a) $\mathbf{Q} = (½\ ½\ 0)$ in fields up to 31 T and (b) $\mathbf{Q} = (1/3\ 1/3\ 0)$ in fields up to 21 T. Magnetic field dependence of neutron diffracted intensities at $\mathbf{Q} = (2\ 2\ -1/3)$, $(2\ 2\ -½)$ and $(2\ 2\ -4/9)$ in fields up to (c) ~32 T, (d) 26 T, (e) ~23.5 T and (f) ~22.3 T. The magnetic field was applied along the *c* axis. Open and solid symbols correspond to rising and falling fields, respectively. All measurements were carried out at $T = 2.1$ K

More intriguingly, for fields up to 32 T, we only observed the magnetic satellites on increasing magnetic field (fig 2(c)). No signal was found neither on the (h h δ) nor on the (δ δ l) positions on decreasing magnetic field. Decreasing the field strengths to roughly the middle of each plateau states, the intensity of the corresponding magnetic satellites was only partially recovered (fig 2(d) to (f)). This strange microscopic behavior seems to be in contradiction with high field magnetization measurements and our previous neutron data [15] which show the existence of the plateau phases also on decreasing magnetic fields.

These data clearly call to further measurements to search for a field-induced magnetic signal on decreasing magnetic fields.

References

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