SUPPLEMENTARY INFORMATION

for

# Magnetoelastic coupling in the stretched diamond lattice of $TbTaO_4$

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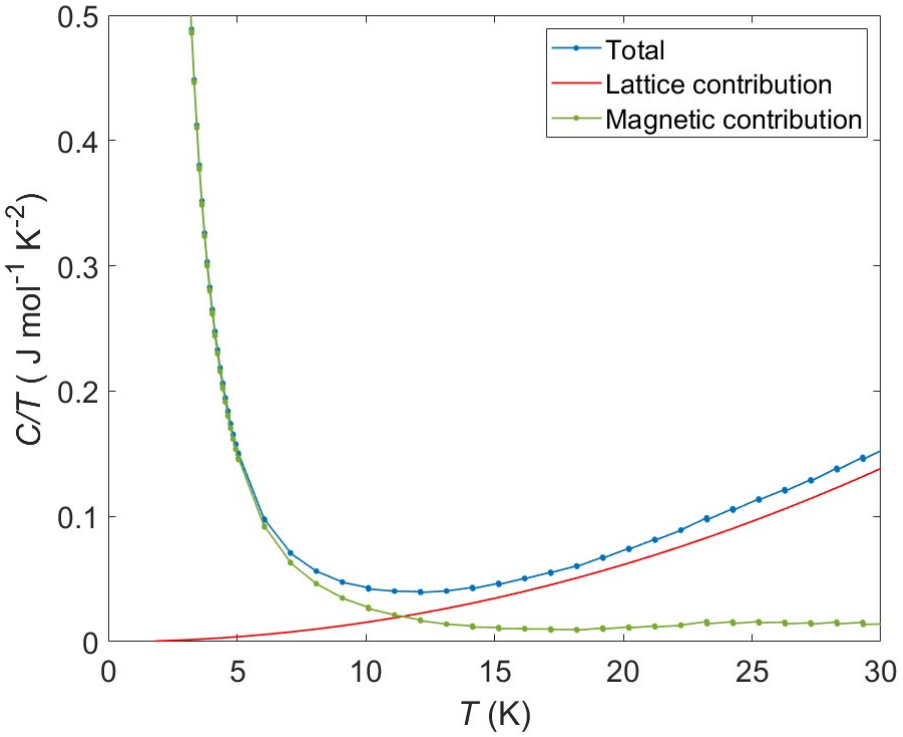



Fig. S1 Heat capacity data for *M*-$TbTaO_4$. Blue dots: total heat capacity (after subtraction of the Ag contribution from the raw data). Red line: Estimated lattice contribution using the Debye model with $\Theta_d$=370 K[1]. Green dots: magnetic contribution, $C_{mag}=C_{total}- C_{lattice}$.

Table S1: Refined Lattice parameters and atomic positions of *M*-$TbTaO_4$ from powder X-ray diffraction at room temperature. The O atomic positions were fixed at value from previous neutron diffraction data[2, 3]. Both $Tb^{3+}$ and $Ta^{5+}$ ions lie on (0.25, y , 0) sites.

| Unit cell | |
|---|---|
| Space group | *I2/a* |
| *a*/ Å | 5.38264(15) |
| *b*/ Å | 11.0182(3) |
| *c*/ Å | 5.06692(13) |
| *β*/° | 95.6844(14) |
| Volume/ Å$^3$ | 299.025(14) |
| $R_{wp}$/% | 7.21 |
| $\chi^2$ | 2.10 |
| **Atomic positions** | |
| *yTb* | 0.6180 (3) |
| *yTa* | 0.1502 (2) |
| *xO1* | 0.090 |
| *yO1* | 0.461 |
| *zO1* | 0.252 |
| *xO2* | -0.003 |
| *yO2* | 0.717 |
| *zO2* | 0.290 |

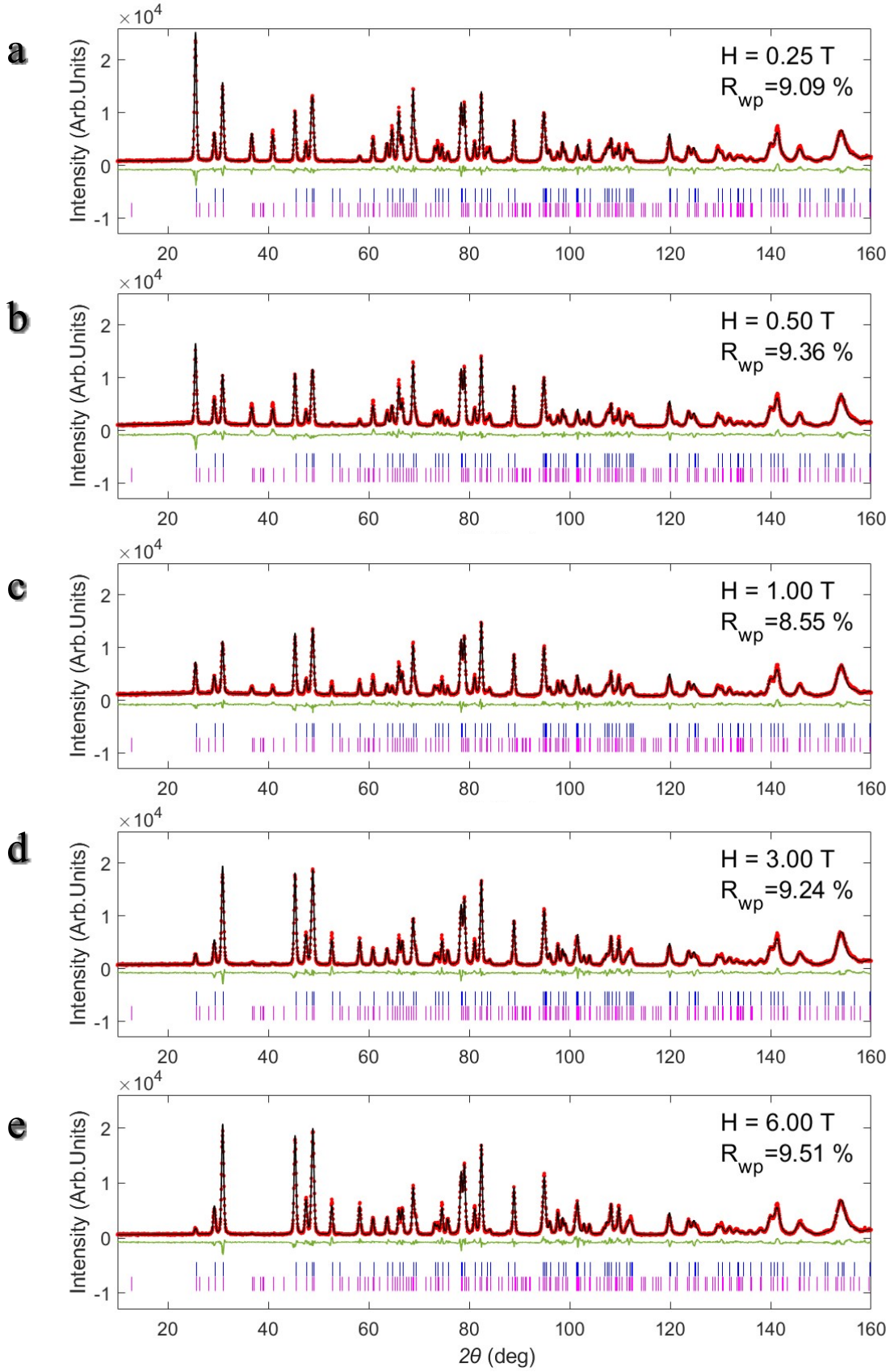



Fig. S2 (a-e) Refined PND data of *M*-TbTaO4 collected at 1.6 K with 0.25 T, 0.50 T, 1 T, 3 T and 6 T. Red dots, experimental data; black line, calculated intensities; green line, difference pattern; tick marks, nuclear (blue), magnetic (pink) Bragg reflection positions.

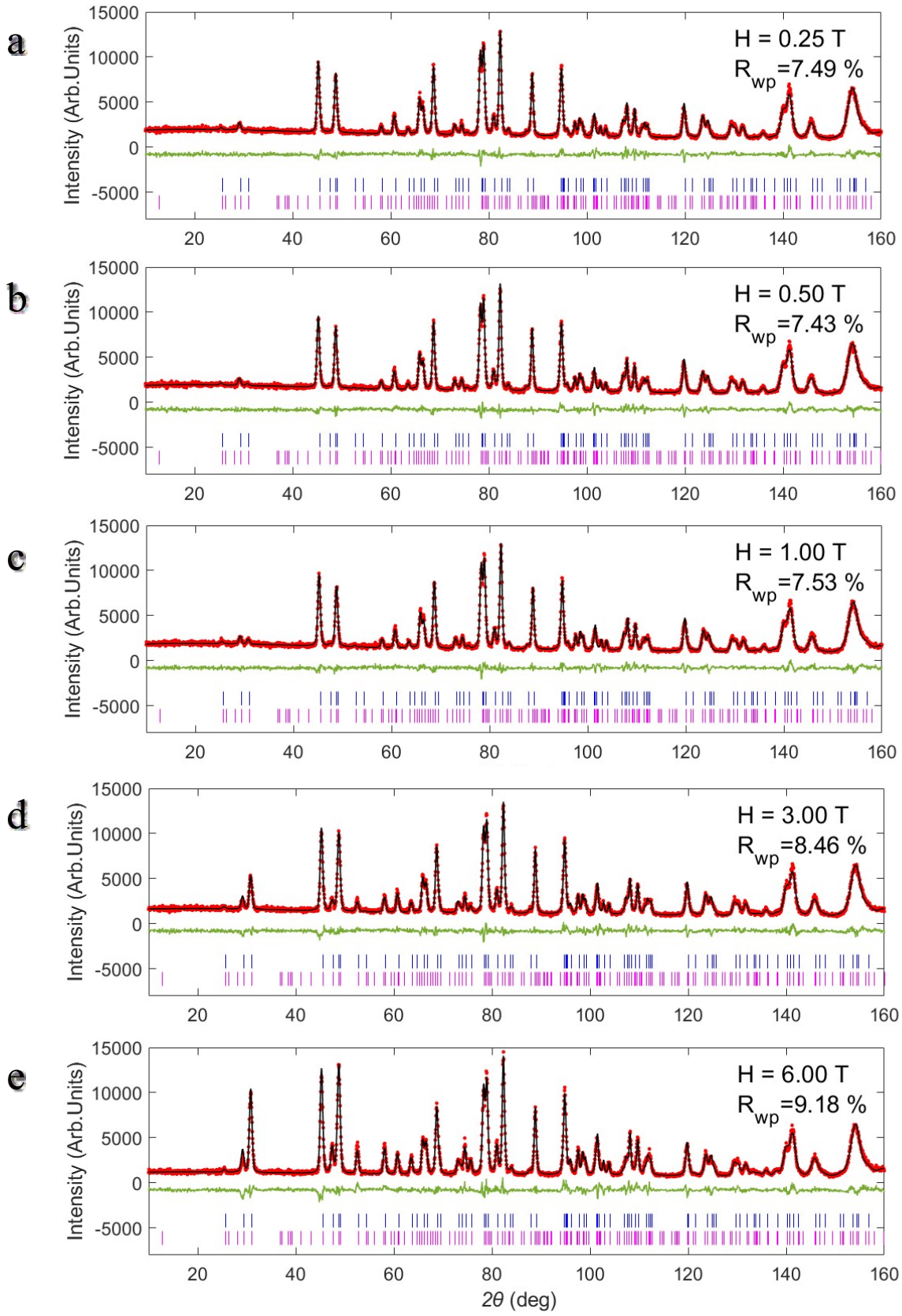



Fig. S3 (a-e) Refined PND data of *M*-$TbTaO_4$ collected at 20.0 K with 0.25 T, 0.50 T, 1 T, 3 T and 6 T. Red dots, experimental data; black line, calculated intensities; green line, difference pattern; tick marks, nuclear (blue), magnetic (pink) Bragg reflection positions.

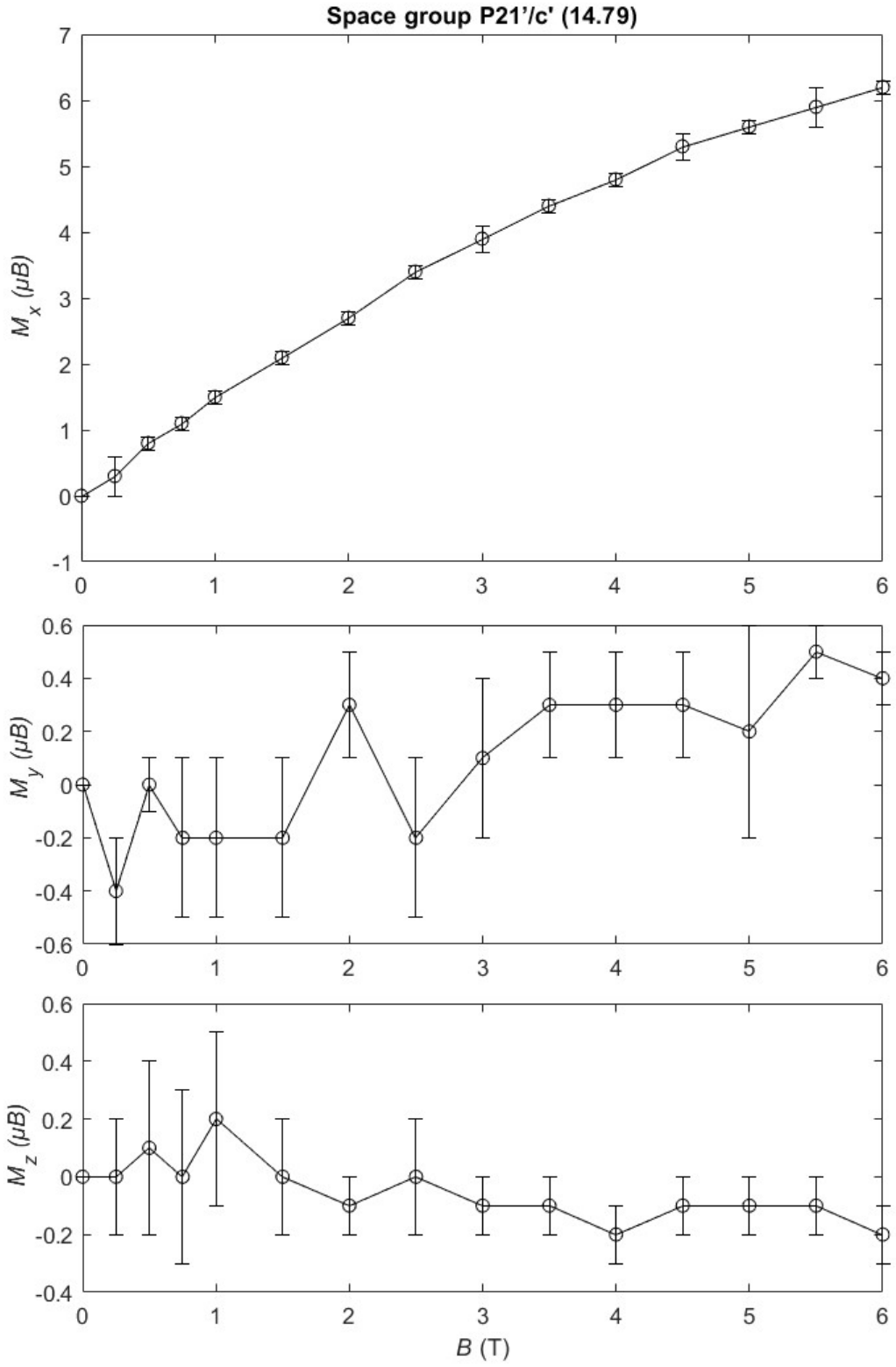



Fig. S4 Evolution of the $Tb^{3+}$ lattice-coordinate magnetic moments along x, y and z axes as a function of magnetic field at 20.0 K from powder neutron diffraction (PND) data.

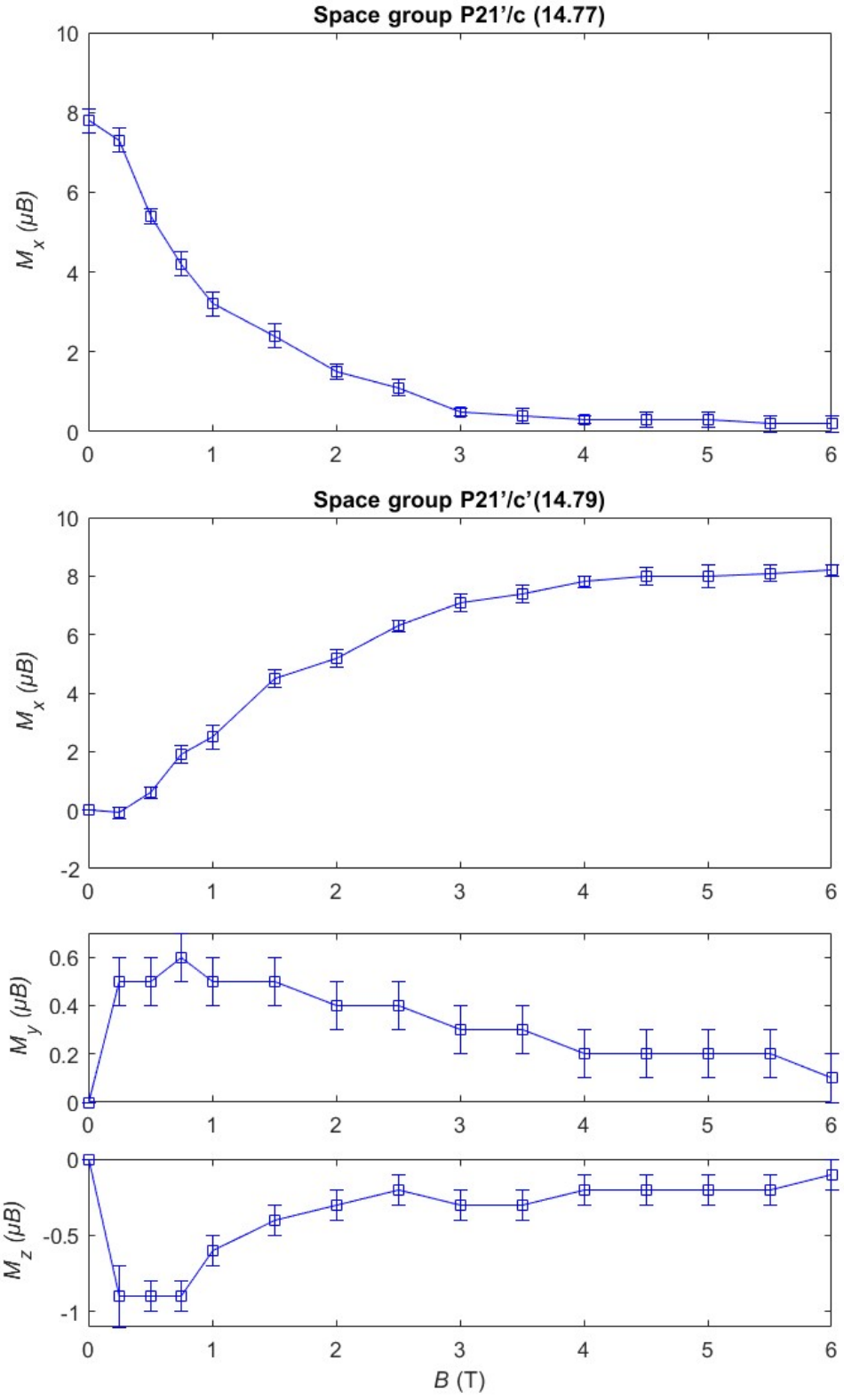
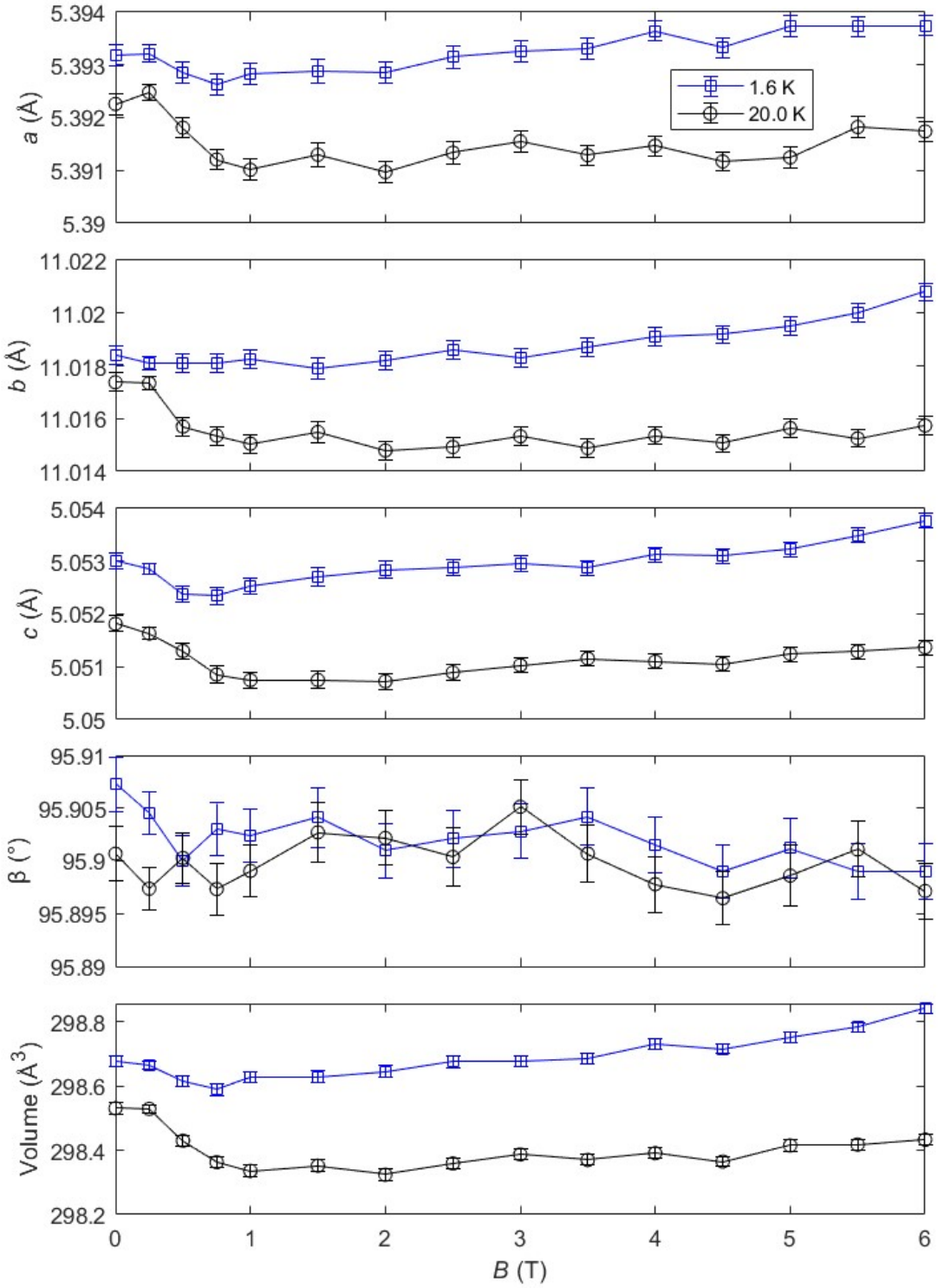



Fig. S5 Evolution of the $Tb^{3+}$ lattice-coordinate magnetic moments along x, y and z axes as a function of magnetic field at 1.6 K from powder neutron diffraction (PND) data. The magnetic structure is modelled with a mixing of two magnetic space group. The phase fractions are fixed and identical to each other, while the magnetic moments are refined independently.


5.394
5.393
5.392
5.391
5.39
a (Å)
1.6 K
20.0 K
11.022
11.02
11.018
11.016
11.014
b (Å)
5.054
5.053
5.052
5.051
5.05
c (Å)
95.91
95.905
95.9
95.895
95.89
β (°)
298.8
298.6
298.4
298.2
Volume (Å3)
0
1
2
3
4
5
6
B (T)

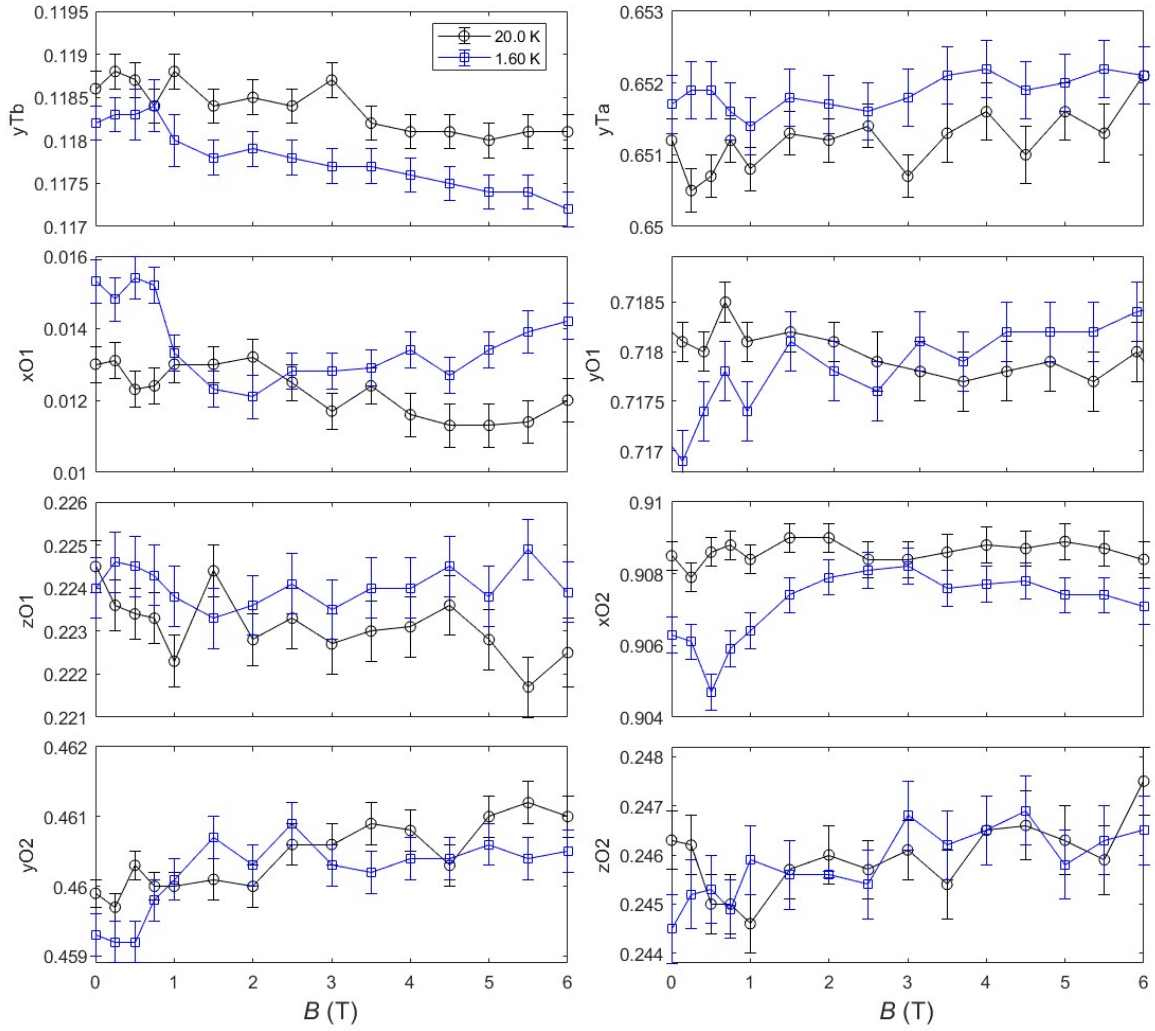



Fig. S7 Refined atomic positions of *M*-$TbTaO_4$ at 1.6 K and 20K as a function of magnetic field. Obtained from Rietveld refinement of powder neutron diffraction (PND) data collected on HRPT, PSI. λ = 2.45 Å.

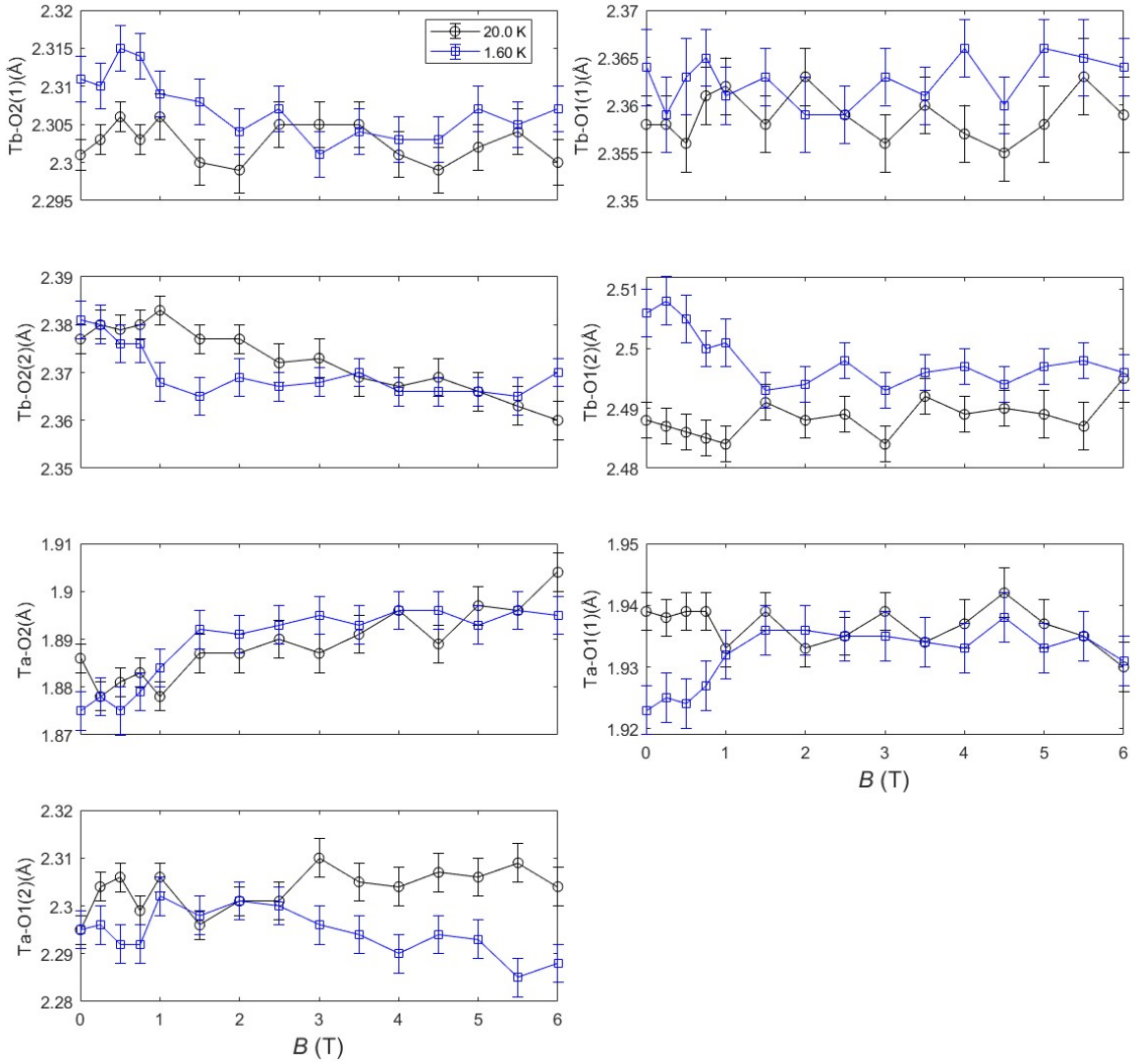



Fig. S8 Refined interatomic distances of *M*-$TbTaO_4$ at 1.6 K and 20K as a function of magnetic field. Obtained from Rietveld refinement of powder neutron diffraction (PND) data collected on HRPT, PSI. λ = 2.45 Å.

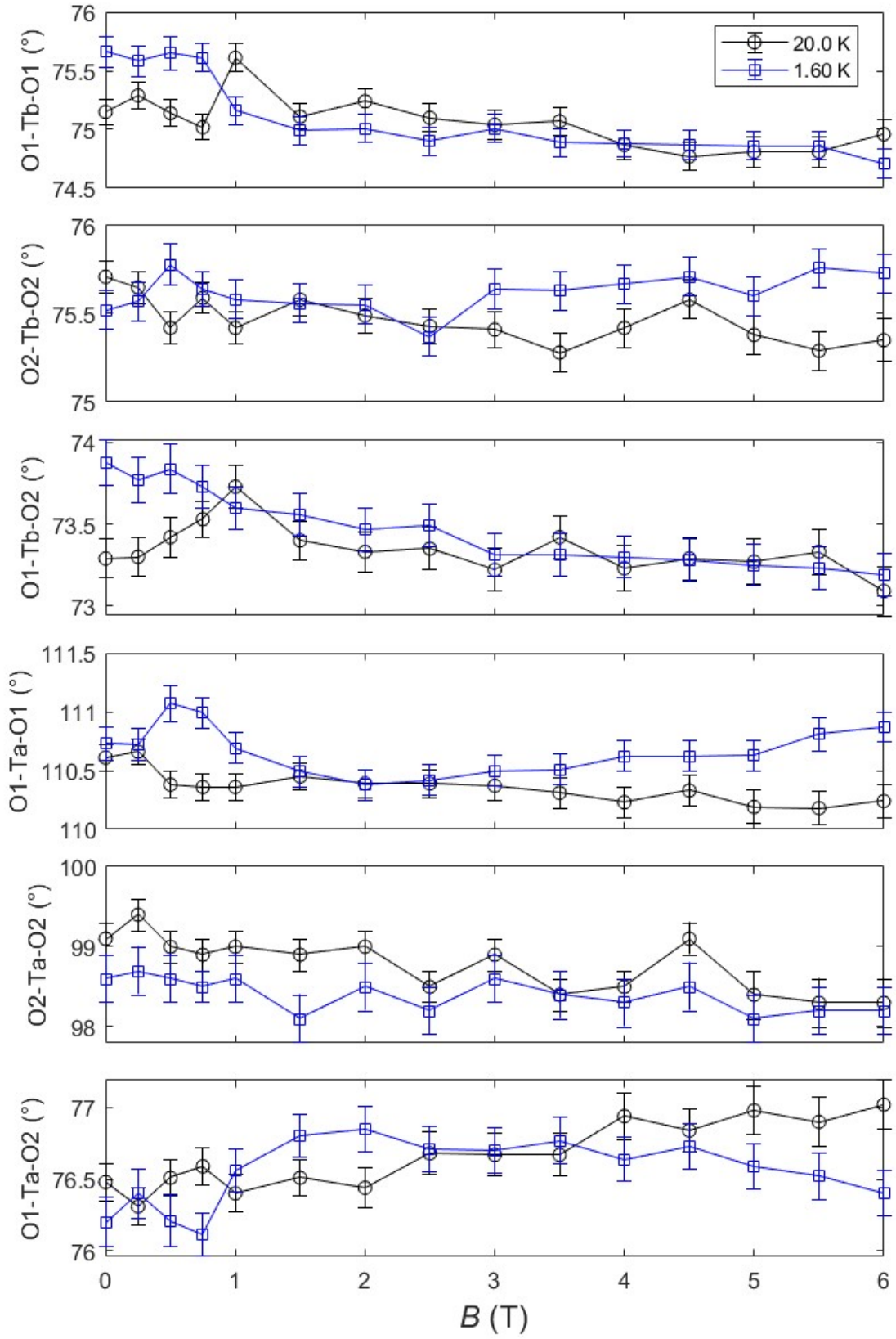



Fig. S9 Refined bond angles of *M*-$TbTaO_4$ at 1.6 K and 20K as a function of magnetic field. Obtained from Rietveld refinement of powder neutron diffraction (PND) data collected on HRPT, PSI. λ = 2.45 Å.

# Reference


1. E. Gopal, *Specific heats at low temperatures*, Springer Science & Business Media, 2012.

2. N. D. Kelly, L. Yuan, R. L. Pearson, E. Suard, I. P. Orench and S. E. Dutton, *Phys Rev Mater*, 2022, **6**, 044410.

3. N. D. Kelly, C. V. Colin, S. E. Dutton, V. Nassif, I. P. Orench and E. Suard, *Institut Laue-Langevin*, 2021, DOI: 10.5291/ILL-DATA.5-31-2854.