Investigations into the Magnetic and Crystal Field Excitations of the Orthorhombically Distorted Perovskites $TbVO_3$ and $CeVO_3$

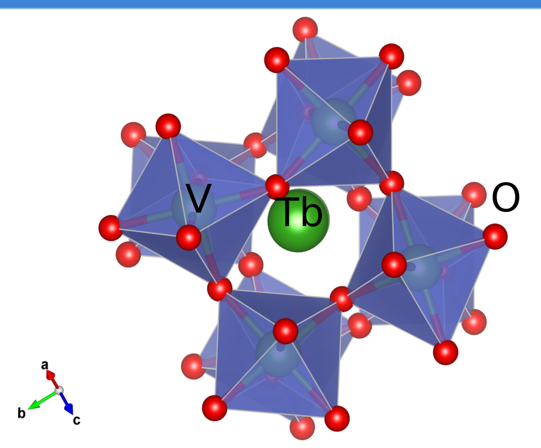
Joel O'Brien[a], N. Reynolds[a], P. Rovillain[a,b], S.A. Danilkin[b], K. Schmalzl[c], M. Reehuis[d], R. Mole[b], D. Yu[b], S. Miyasaka[e], F. Fujioka[e], Y. Tokura[e], B. Keimer[f], G. McIntyre[b], and C. Ulrich[a,b]

[a]School of Physics, The University of New South Wales, Sydney, NSW 2052, Australia.
[b]ACNS, ANSTO, Lucas Heights, NSW 2234, Australia.
[c]Institut Laue Langevin, BP 156, 38042 Grenoble Cedex 9, France.
[d]Helmholtz-Zentrum for Materials and Energy, Berlin, Germany.
[e]Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan.
[f]Max-Planck-Institute for Solid State Research, Stuttgart, Germany.

Introduction

- Inelastic neutron scattering performed on series of vanadates to categorise and examine the interplay between crystal field and magnetic excitations.
- Unexpected splitting of the magnon peaks was observed in YVO_3, which can be explained by an 'Orbital Peirels State' [1].
- For RVO_3 (*R*=Dy, Tb, Pr, Ce) additional crystal field excitations are observed in inelastic neutron scaterring experiments.
- Unexpected shift of the crystal field energies at the magnetic phase transition.
- In this work: - Inelastic neutron scattering experiments
  - Crystal field calculations to explain the observed shifts for C-type antiferromagnetic $CeVO_3$

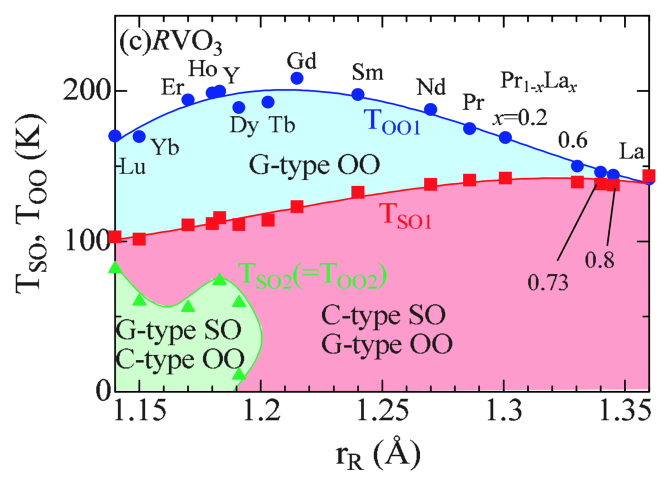
Crystal Structure



The crystal structure of RVO_3 (space group Pbmn) consists of corner-sharing VO_6 octahedra.

The magnetic structure is C-type antiferromagnetic (antiferromagnetic in the ab-plane and ferromagnetic along the c-axis) below the Neel transition temperature of approximately 110 K [2-5].

The rare earth element (Dy, Tb, Pr, Ce) is located in an open cage surrounded by the VO_6 octahedra.

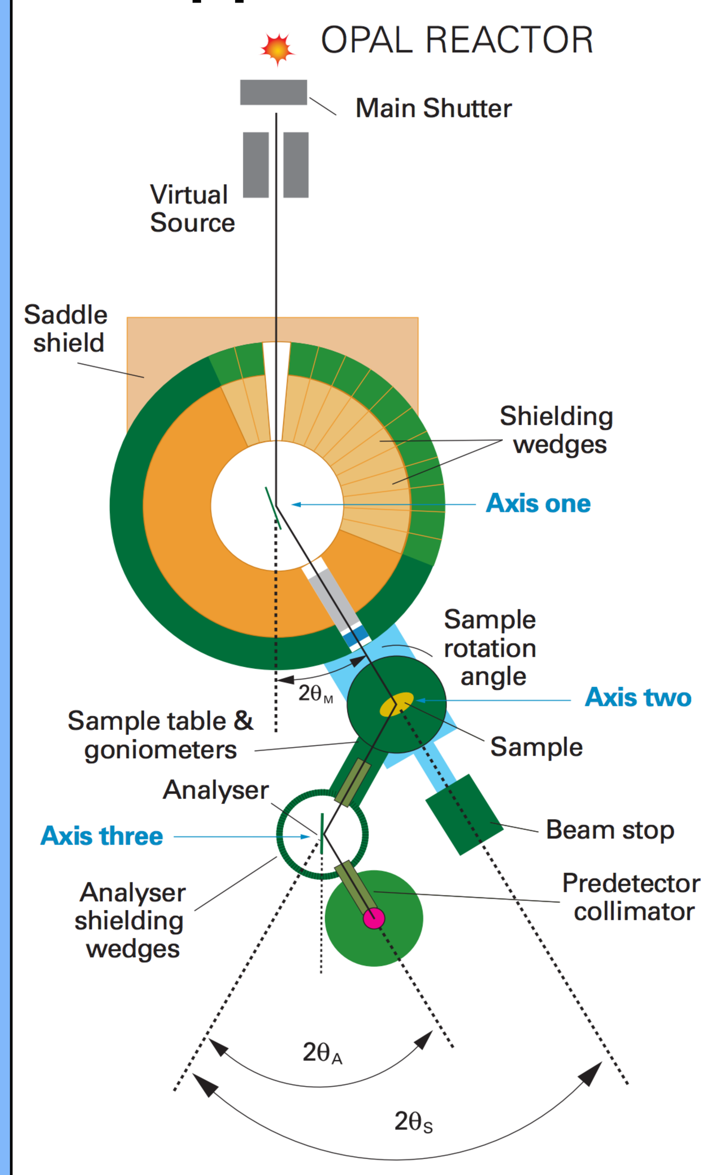


Phase diagram of rare earth vanadate series [3,4].

Methods

Elastic neutron scattering was used for determination of crystallographic and magnetic structure. Experiment performed at the HZB in Berlin, Germany [2,5].

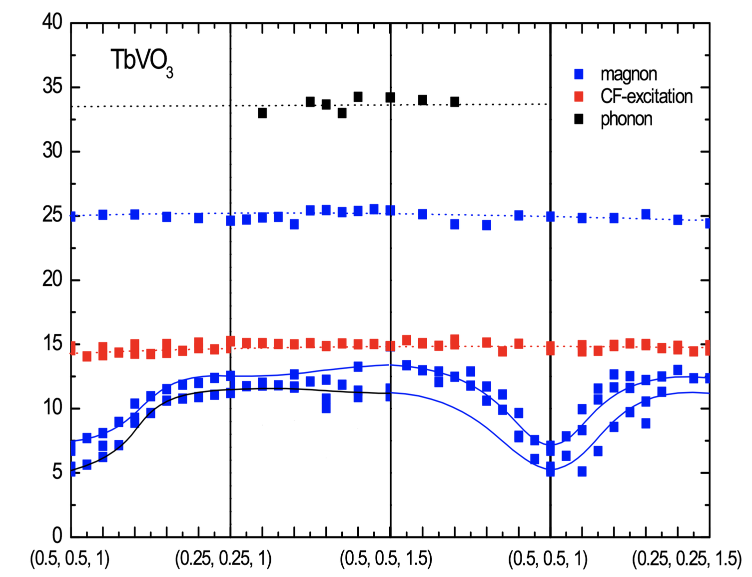
Inelastic neutron scattering data taken at TAIPAN, ANSTO and IN22 at the ILL, Grenoble, France [1].



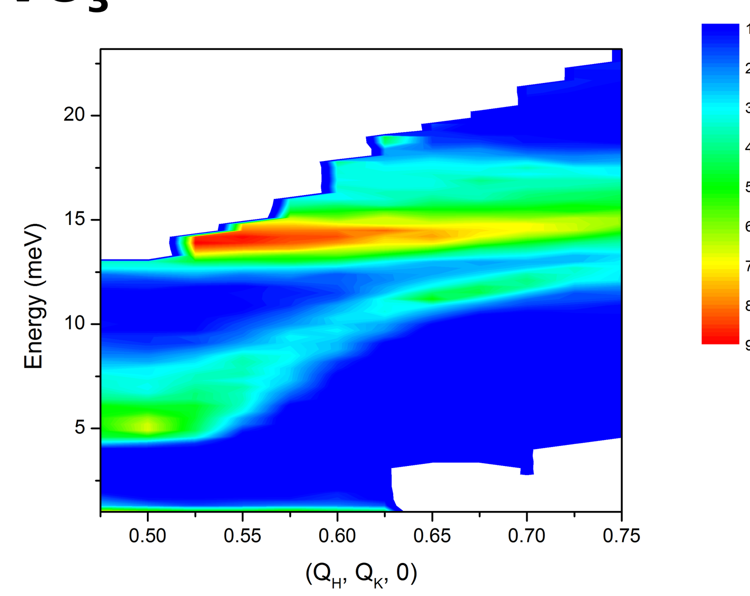
Triple Axis Spectrometer TAIPAN at ANSTO.

Inelastic Neutron Scattering

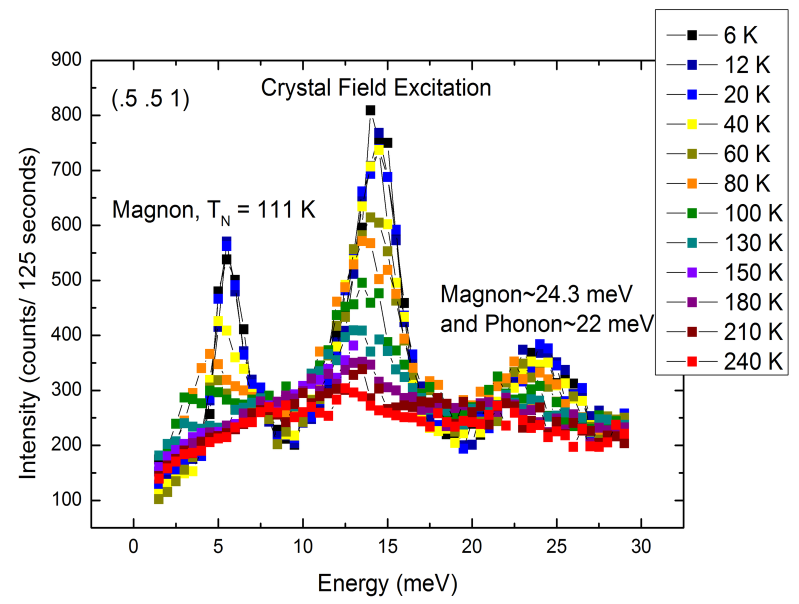
$TbVO_3$



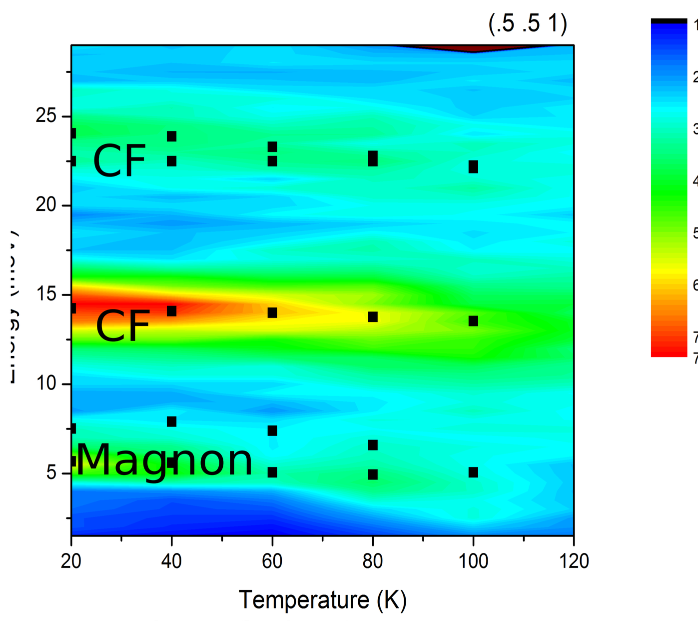
Dispersion relation of the magnon and crystal field (CF) excitations of $TbVO_3$.
Data taken at ILL, Grenoble, France.



Dispersion scan of $TbVO_3$.
Phonon excitation at 17meV, crystal field excitation at 14.5meV and magnon excitation between 5meV and 12meV. Data taken at ILL, Grenoble, France.

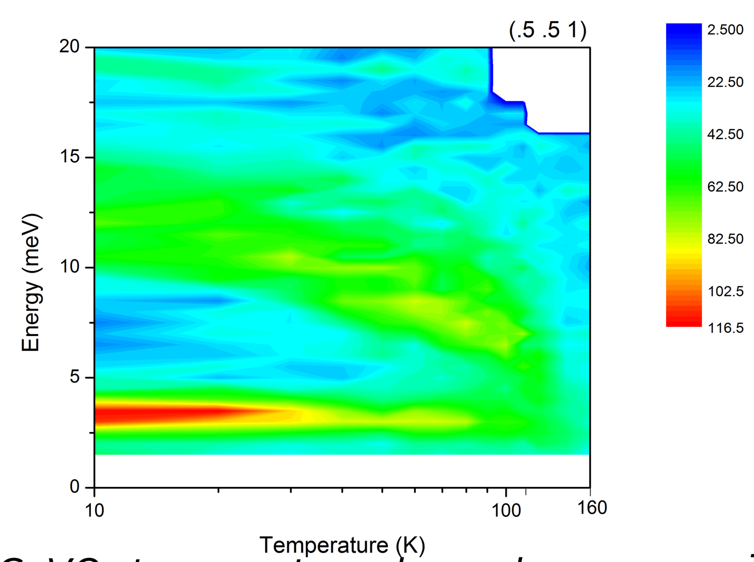


Temperature dependence from triple axis scan of $TbVO_3$.
Data taken at Taipan, ANSTO.

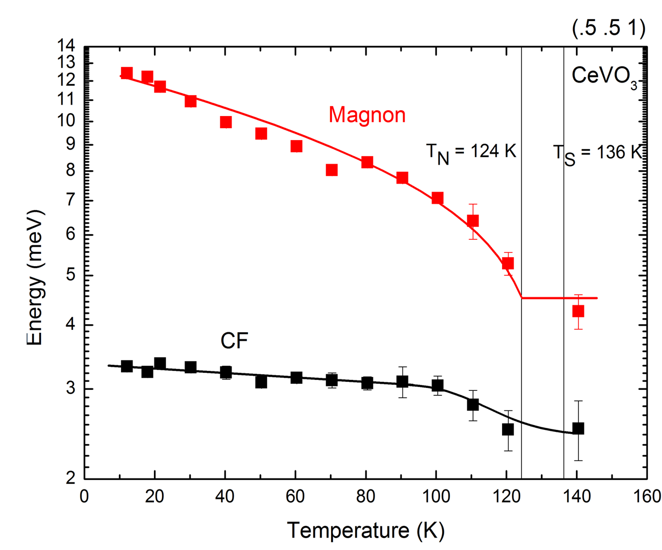


Contour plot of $TbVO_3$ temperature dependence. Black dots show fitted peak positions of excitations of the CF and magnon.

$CeVO_3$



$CeVO_3$ temperature dependence scan.
Crystal field excitation at 3meV and magnon excitation between 5-15meV.
Data from Taipan, ANSTO.



Temperature dependence of $CeVO_3$.
The crystal field excitation energy decreases by half at the magnetic phase transition.

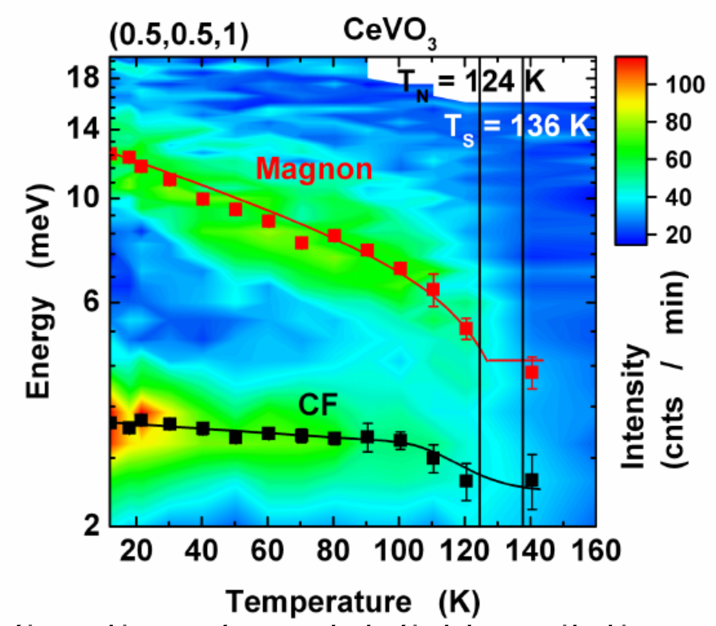
Unexpected shift of crystal field excitations at the magnetic phase transition.

Point-charge Crystal Field Calculations

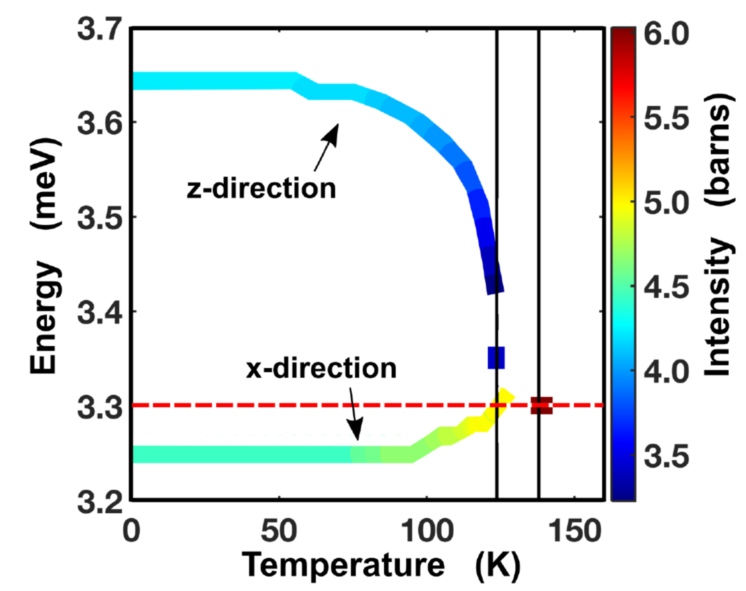
The program McPhase was used for crystal field calculations:

- Point charge model used to generate Steven's Parameters
- Steven's Parameters describe the 4f orbital configuration inside the crystal
- Steven's Parameters were then refined to fit to the experimental data
- Magnetic field generated by the spin order of vanadium ions was estimated at the atomic radius of Ce^{3+} ion inside unit cell to be 6.9 T, agreeing with μSR measurements of similar materials
- The temperature dependency of the corresponding internal magnetic field was then simulated by McPhase

Experimental Data



Results of Simulation



Confirmation of crystal field excitation energies with program McPhase. Simulating magnetic field in the z-direction (along ferromagnetic crystallographic c-axis) leads to a positive shift in crystal field excitation energy, reproducing the correct shape and order of magnitude of the internal magnetic field from the vanadium spins.

Magnetic field applied in the x-direction (in the antiferromagnetic crystallographic ab-plane) leads to a slight negative shift in crystal field excitation energy, which further confirms that the mechanism of the crystal field energy shift is due to the vanadium spins ferromagnetically ordered parallel to the c-axis.

These results suggest an internal magnetic exchange field is responsible for the shift of the crystal field energies at the magnetic phase transition, that is a Zeeman Effect

References

[1] C. Ulrich, et al., Phys Rev. Lett. 91, 257202 (2003).
[2] M. Reehuis, et al., Eur. Phys. J. B 64, 27–34 (2008).
[3] J. Fujioka et al., Phys. Rev. B 82, 144425 (2010).
[4] S. Miyasaka et al., Phys. Rev. B 68, 100406 (2003).
[5] M. Reehuis, et al., Phys. Rev. B 83, 064404 (2011).
[6] N. Reynolds, Masters Thesis, School of Physics, UNSW, (2013).

Contact

Joel O'Brien, email: j.obrien@student.unsw.edu.au