Low Temperature Nuclear Orientation Studies of the Magnetic Structures of $RNiAl_4$ in Applied Magnetic Fields

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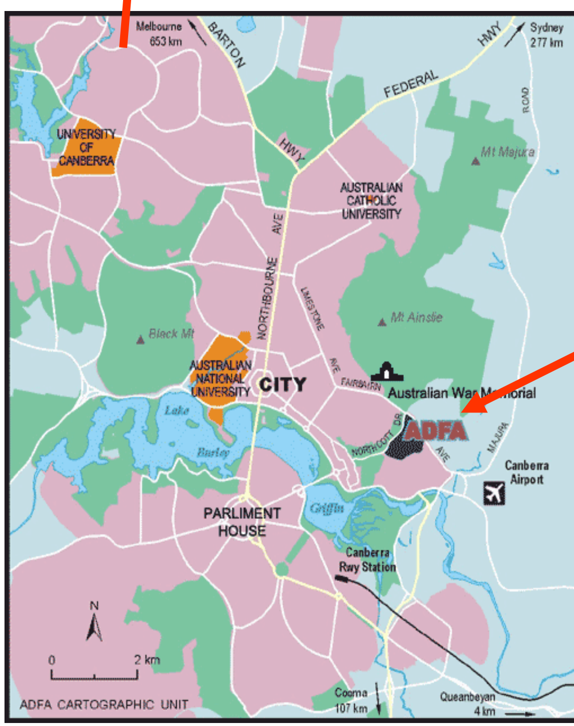
The University of New South Wales
at the
Australian Defence Force Academy

Location:
Canberra
Australian
Federal
Capital



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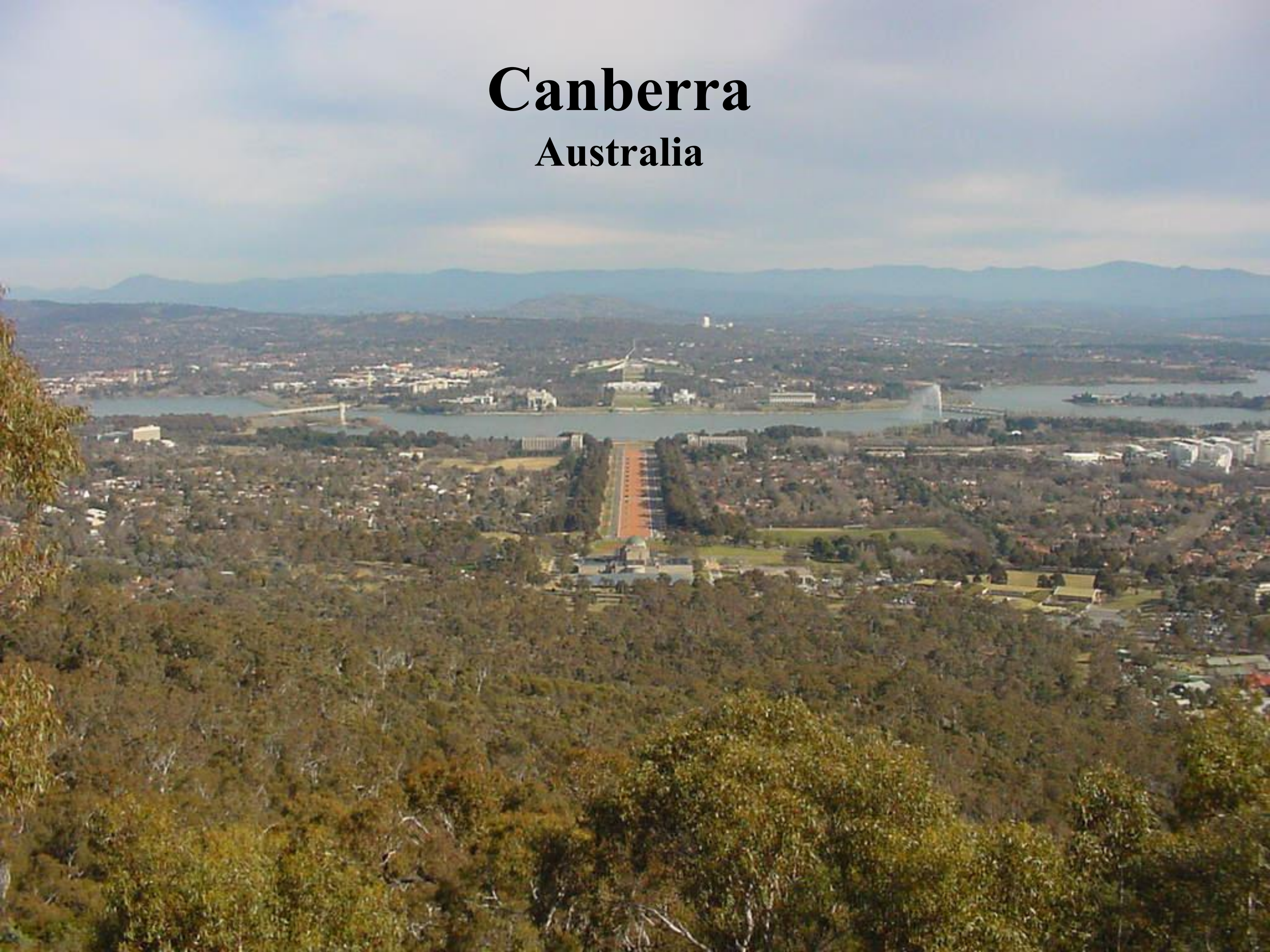
Canberra
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$RNiAl_4$ Systems

R = rare earth (or Y)

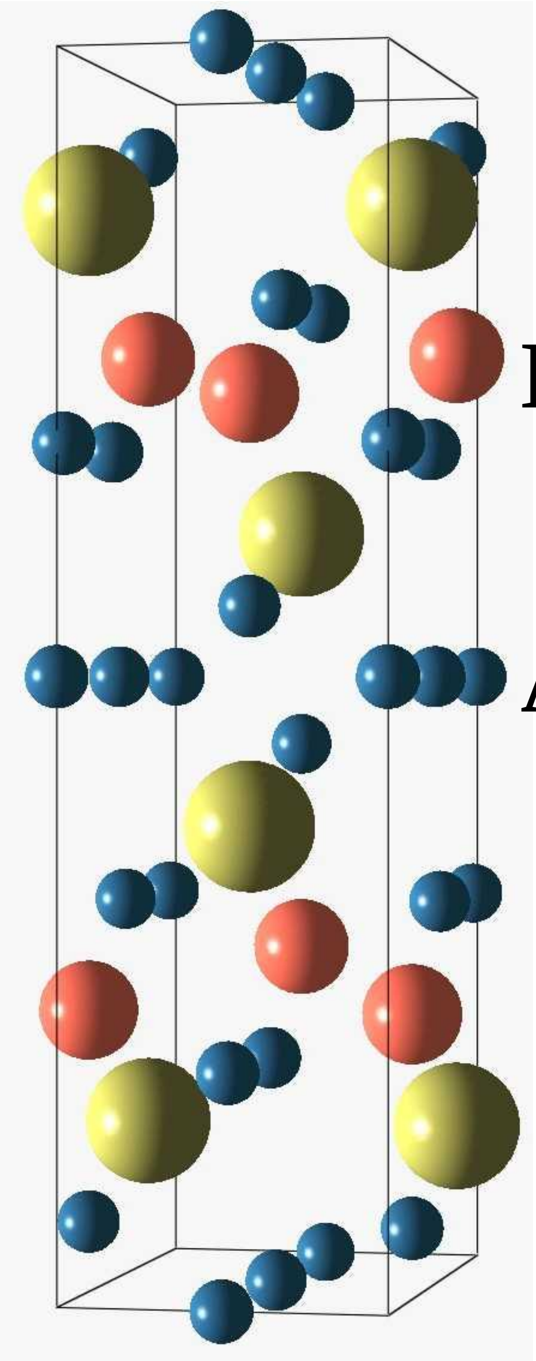
- Orthorhombic structure (Cmcm) with R in single sites and nonmagnetic Ni

- exhibit antiferromagnetic order and metamagnetism (B field driven transitions)

- CEF driven magnetic anisotropies dependent on choice of R

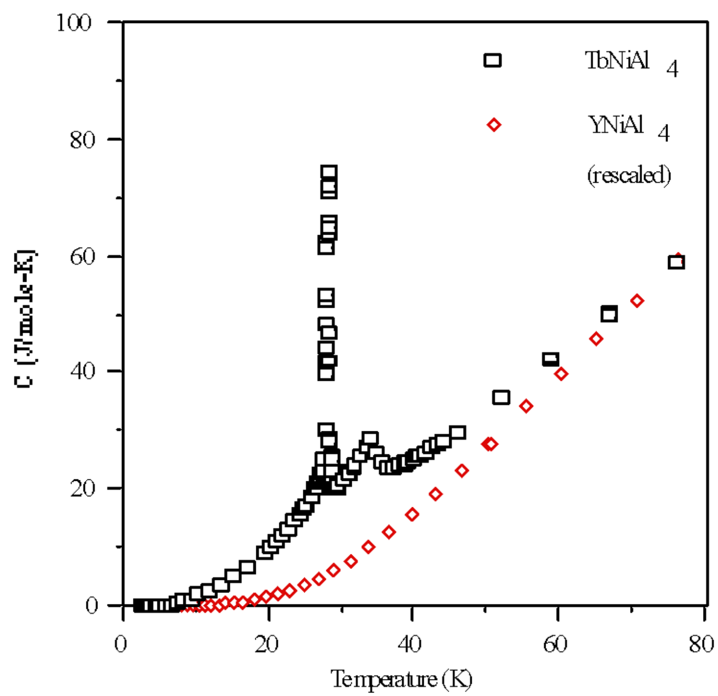
- R = Pr, Gd, Tb & Dy known to have 3 resolved phases v *T*

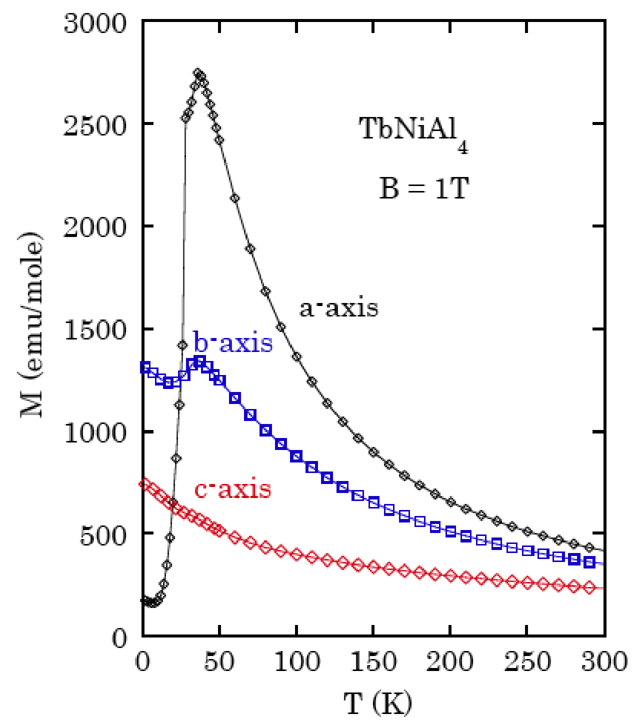
- started LTNO studies to look at anisotropy in mixed R compounds *but other things came up*



Examples: $TbNiAl_4$, $NdNiAl_4$

- $TbNiAl_4$ is one of the most interesting compounds in the series
- In zero B field - T_N ~34.0 K $T_{N'}$ ~28.0 K, *a* axis linear antiferromagnet (AF) alignment in low temp phase, incommensurate AF in intermediate phase

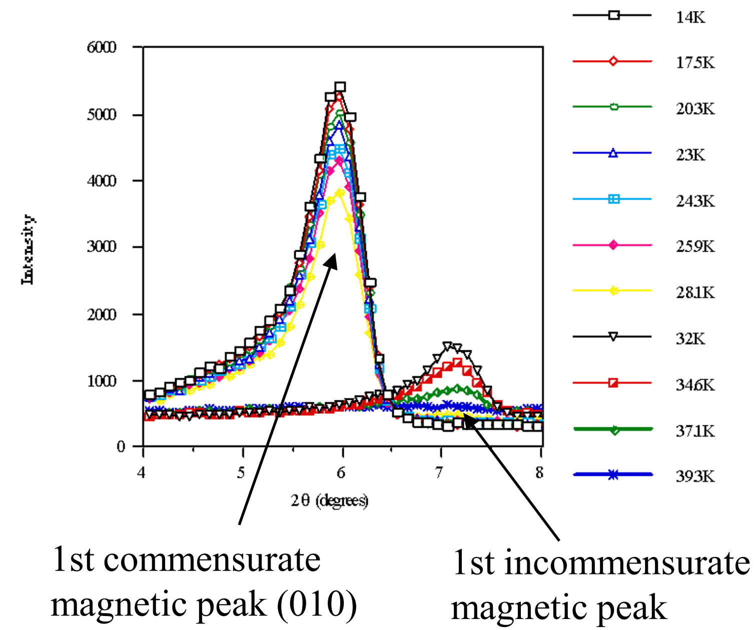




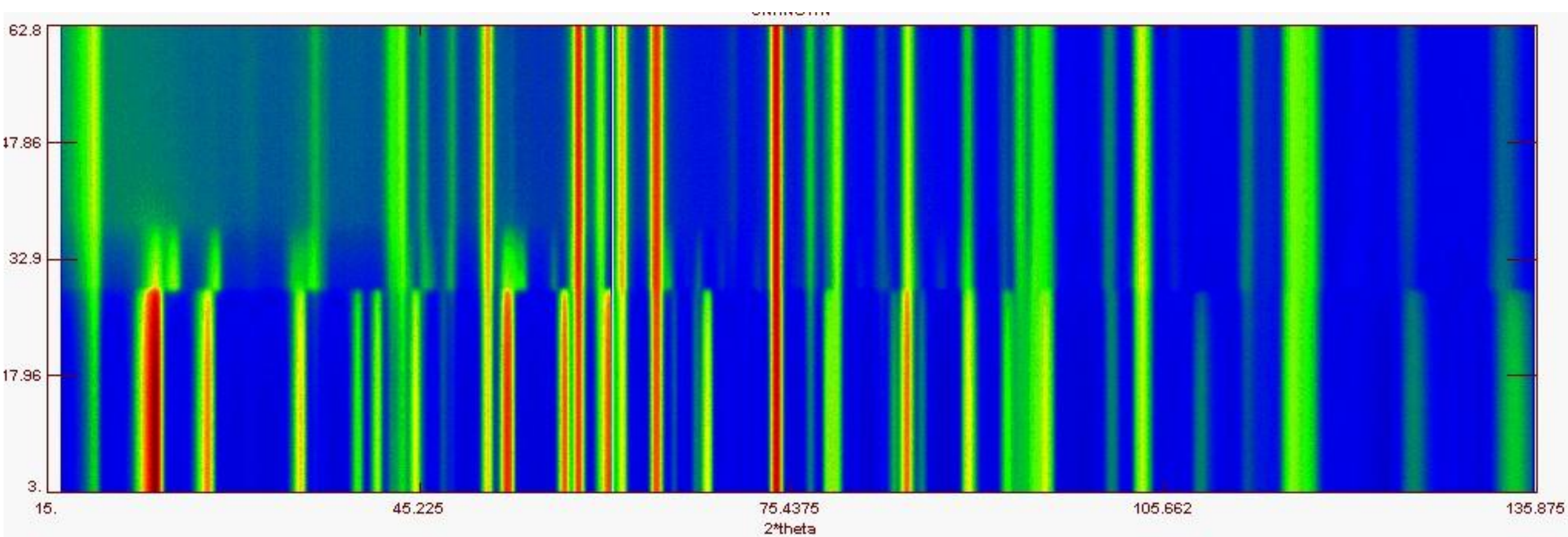
$TbNiAl_4$ – powder neutron diffraction v T (zero applied B)

- Confirms low *T* phase linear AF with ***a*** axis alignment & (010) propagation

- Intermediate phase incommensurate AF, (0.172 1 0.038) propagation



*(W.D. Hutchison et al., JMMM **301** (2006) 352.)*



TbNiAl$_4$ - v B at low T

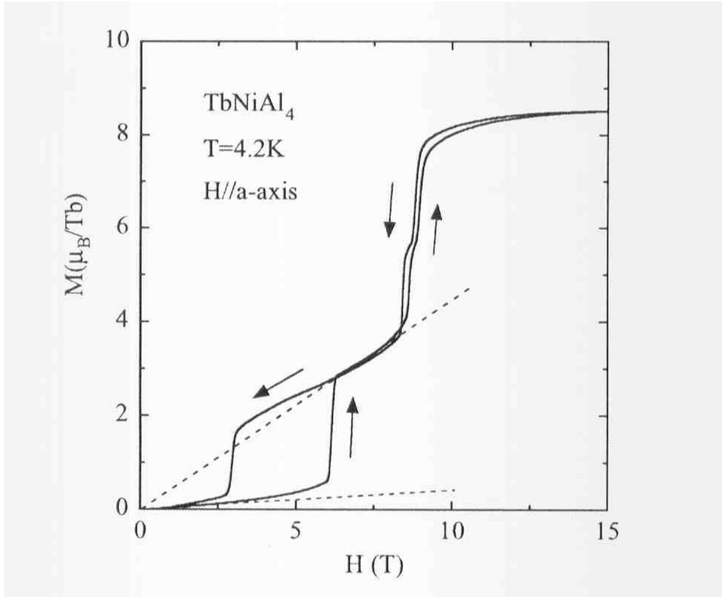
• TbNiAl$_4$ has 3(or 4) phases in the B field manifold at low temp.

• Low field is AF, high field is Ferro. What is the nature of the intermediate phase?

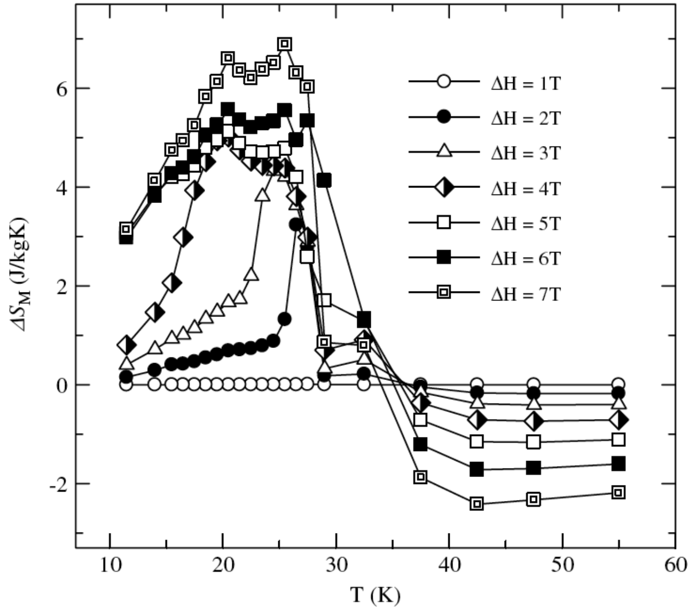
• There is a large negative MCE (magnetocaloric effect) associated with 1st transition - intermediate phase is higher entropy than low field phase

• Studied with neutron diffraction

• Studied via LTNO

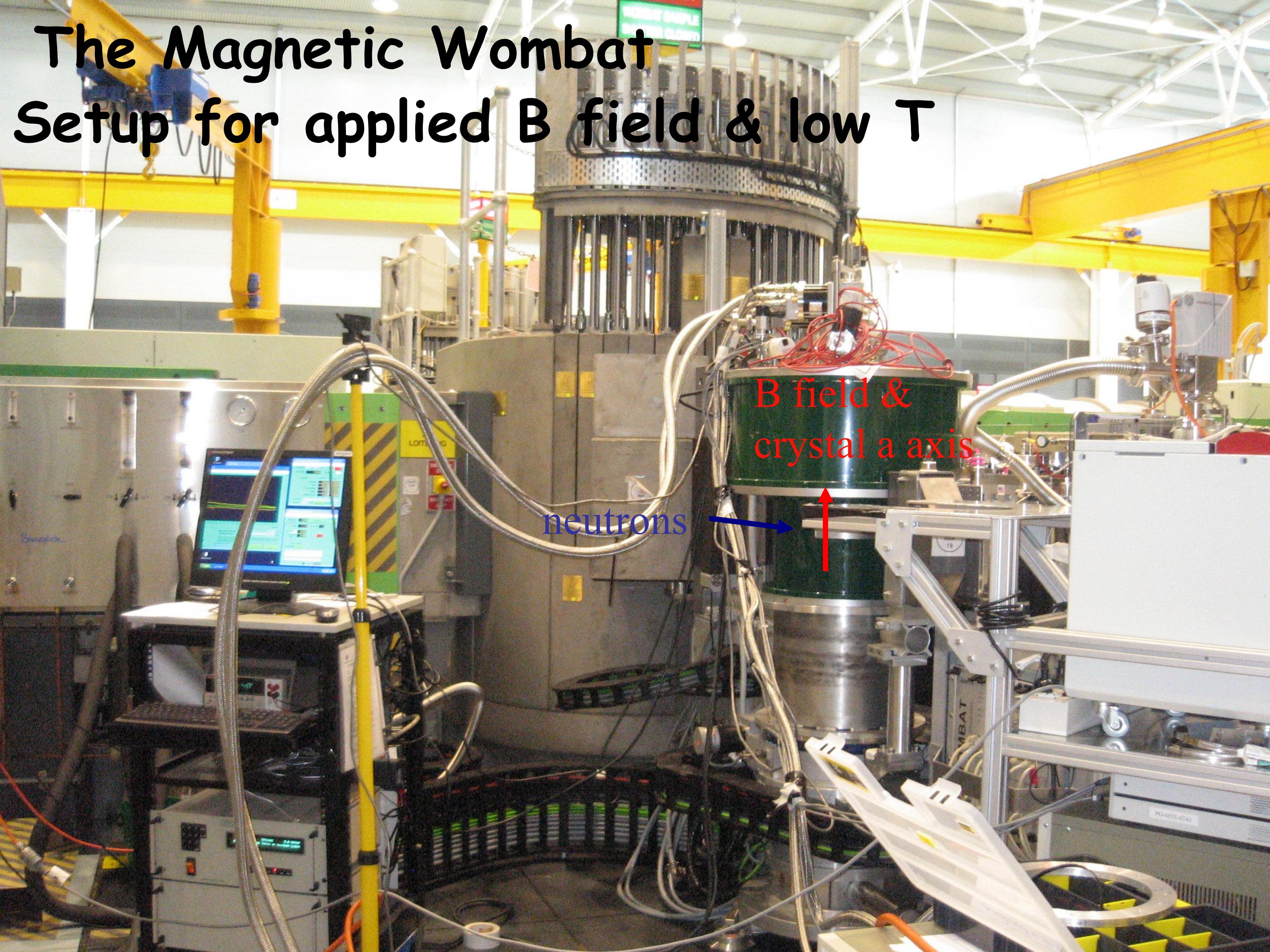


(*T. Mizushima et al., JMMM* ***272*** *(2004) e475.)*

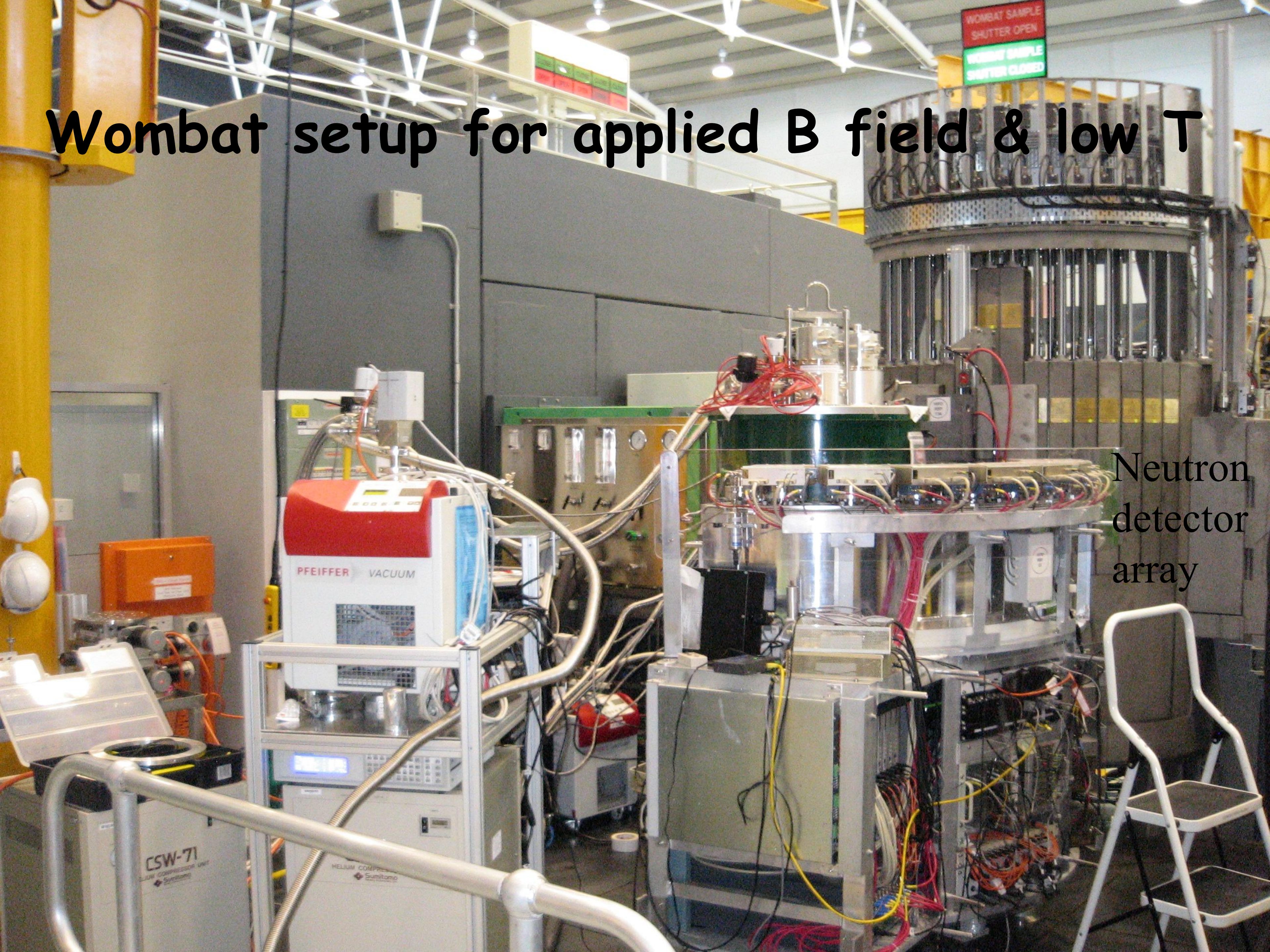


(*L. Li et al., Solid State Comm.* ***149*** *(2009) 932.)*

The Magnetic Wombat
Setup for applied B field & low T

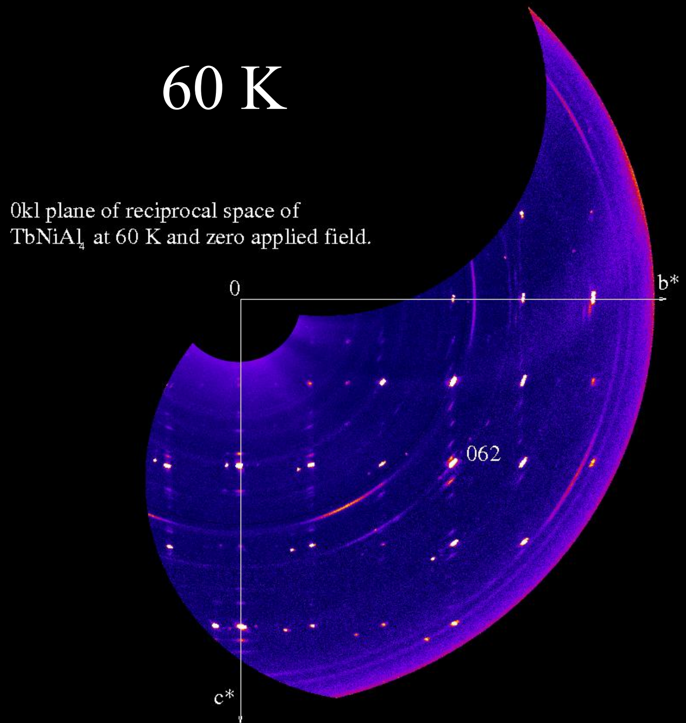


Wombat setup for applied B field & low T

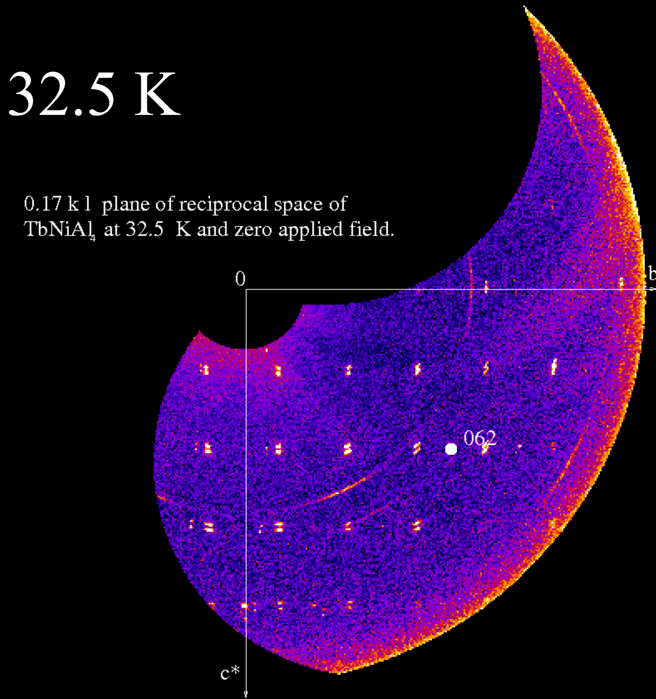


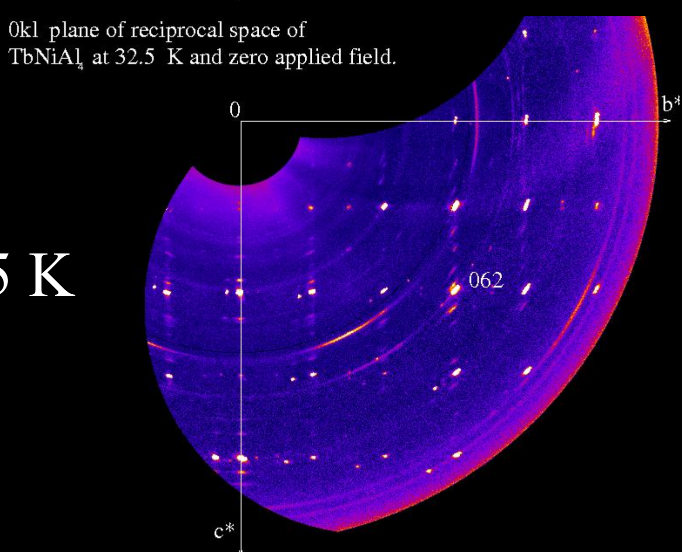
$TbNiAl_4$ – single crystal diffraction (*a* plane) zero field

60 K

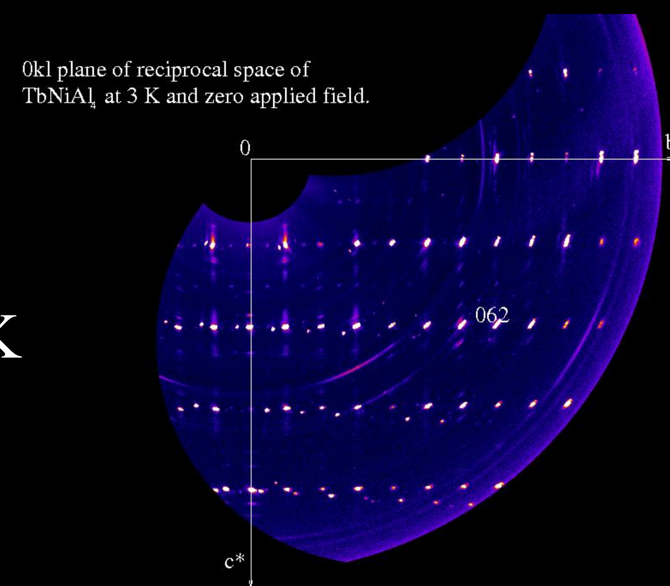


32.5 K





32.5 K



3 K

$TbNiAl_4$ - single crystal diffraction (*a* plane) zero field hk-1 slices

QuickTime™ and a
TIFF (LZW) decompressor
are needed to see this picture.

60 K

QuickTime™ and a
TIFF (LZW) decompressor
are needed to see this picture.

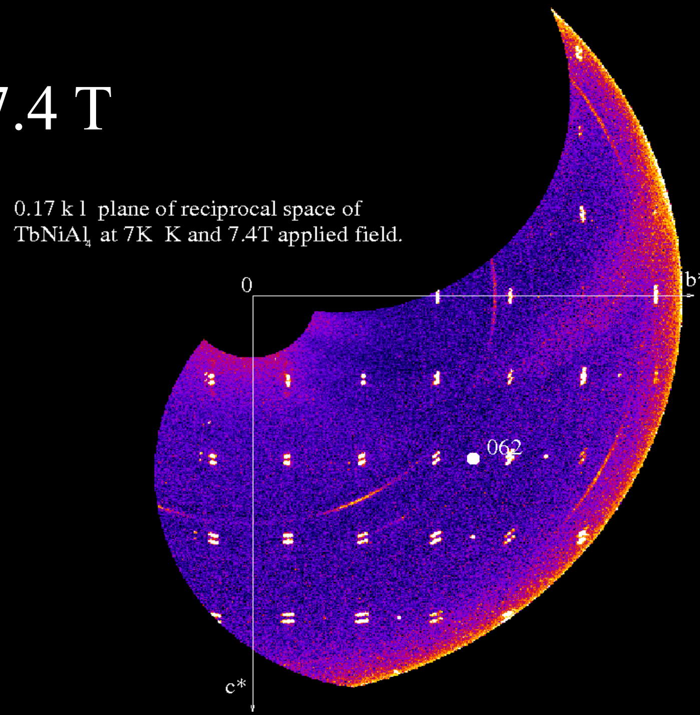
32.5 K

QuickTime™ and a
TIFF (LZW) decompressor
are needed to see this picture.

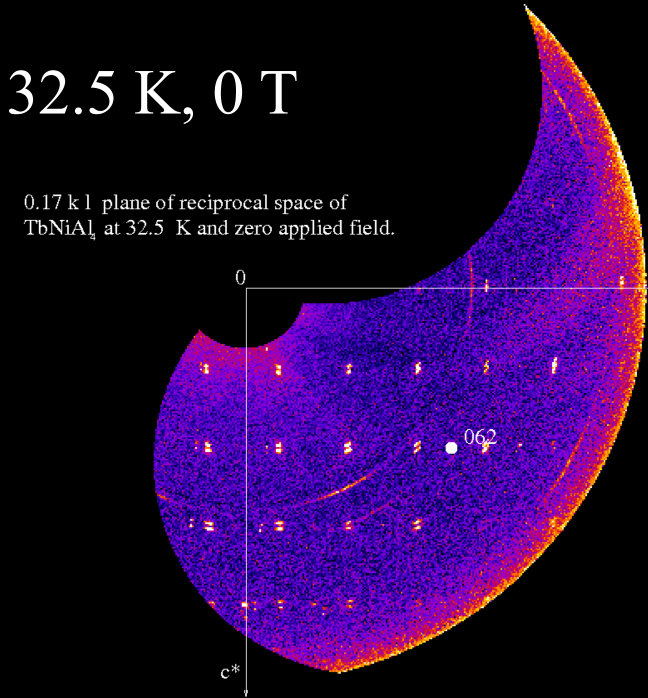
3 K

$TbNiAl_4$ – single crystal diffraction (*a* plane) v B field

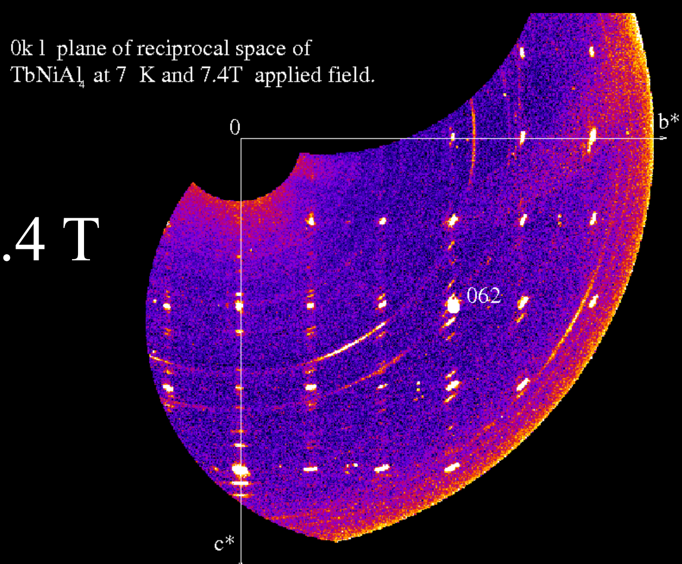
7 K, 7.4 T



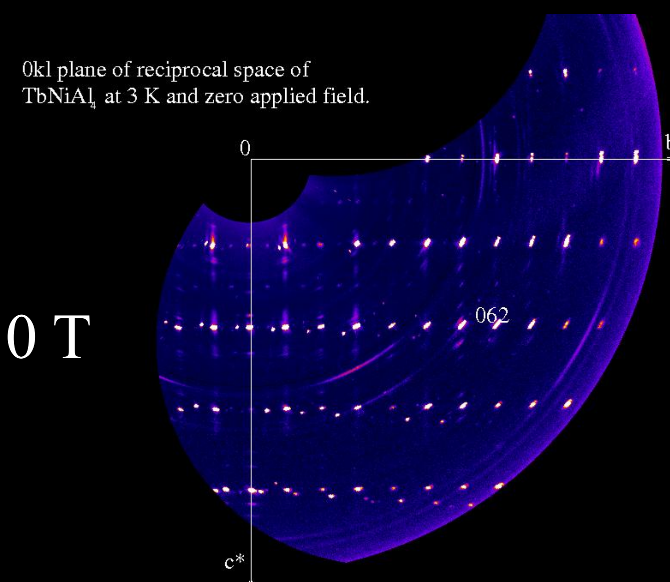
32.5 K, 0 T



7 K, 7.4 T



3 K, 0 T



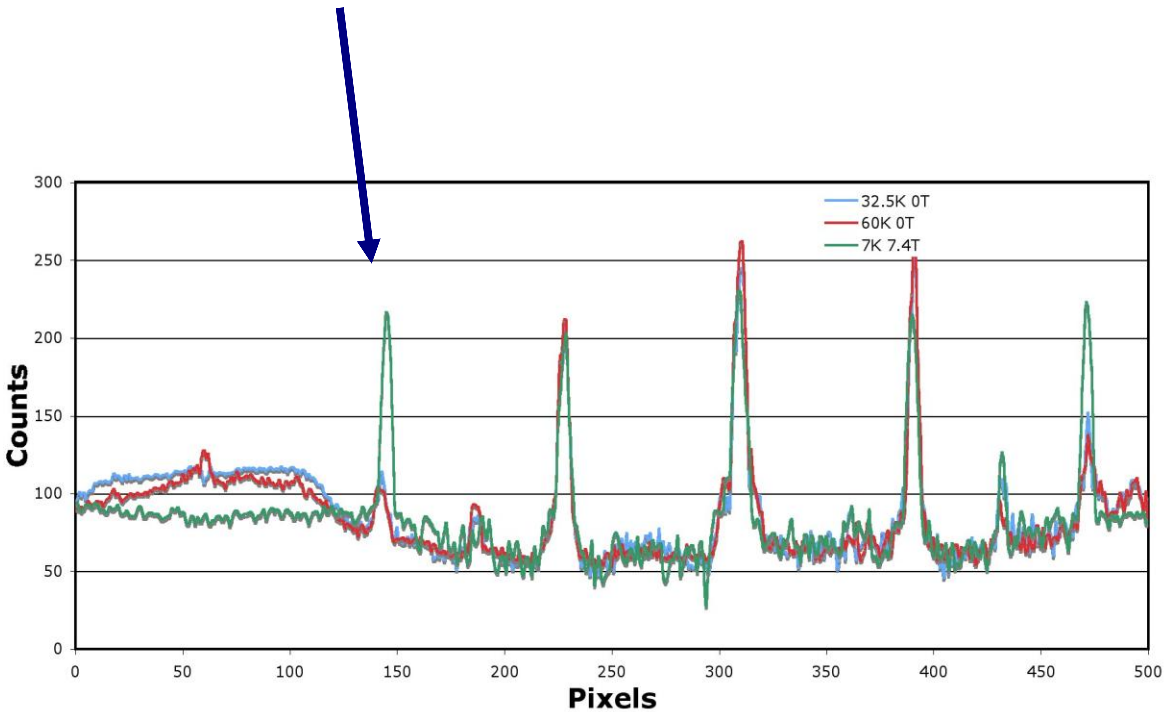
$TbNiAl_4$ - single crystal diffraction (*a* plane) v B field

QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

7 K, 7.4 T

QuickTime™ and a
TIFF (LZW) decompressor
are needed to see this picture.

32.5 K, 0 T



Matching positions of reflections for these two conditions. But also increased intensity at low angle nuclear peaks suggests presence of ferro moments.

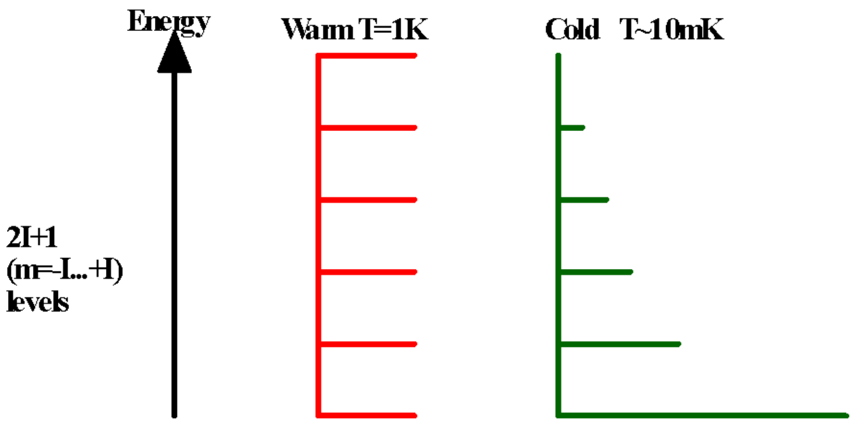
Low Temperature Nuclear Orientation (LTNO)

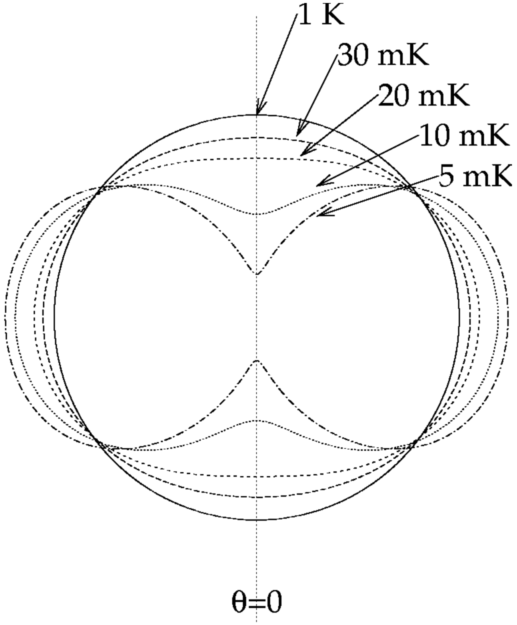
Nuclei with non-zero spin I, magnetic moment μ, in a magnetic field B

2I+1 nuclear Zeeman levels populated according to Boltzmann distribution

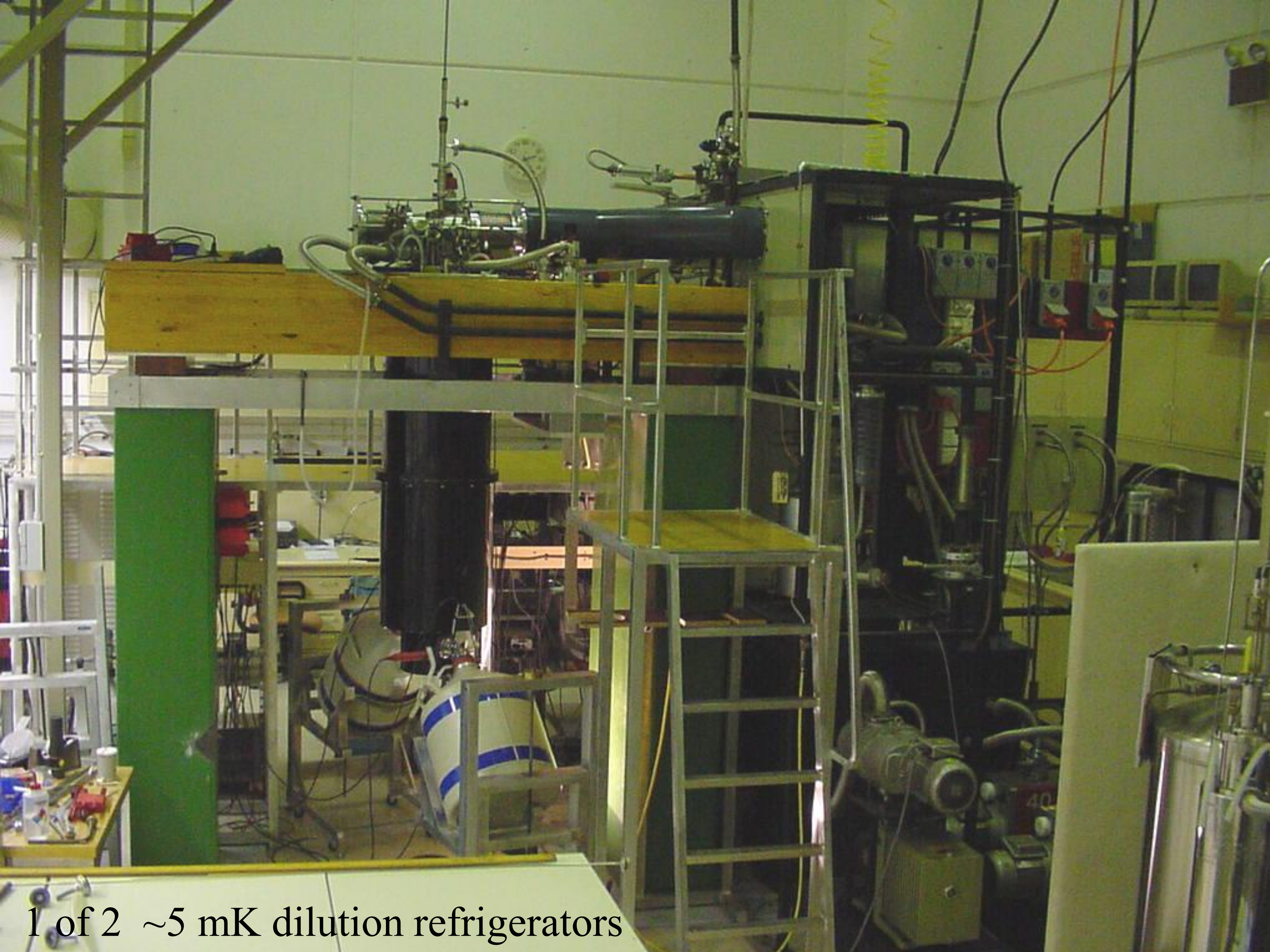
$$P(m) = \exp\left(\frac{E(m)}{kT}\right) / \sum_m \exp\left(\frac{E(m)}{kT}\right) \quad \text{where} \quad E(m) = -m\mu B / I$$

Radiative detection - If spatially oriented nuclei decay the emitted radiation (e.g. γ-ray) can also show spatial anisotropy in emission





NB - Orientation of nuclei will match that of (atomic) magnetic moments via the magnetic hyperfine interaction



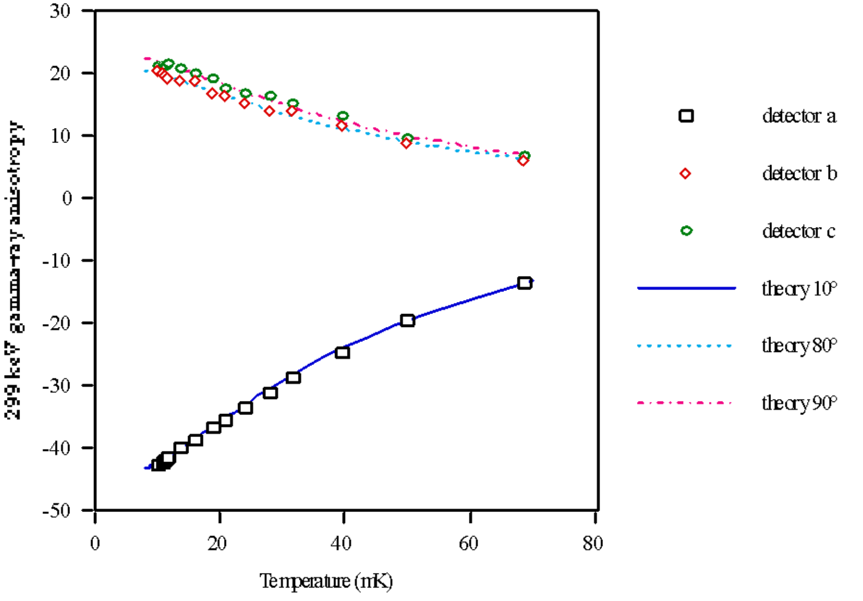
1 of 2 ~5 mK dilution refrigerators

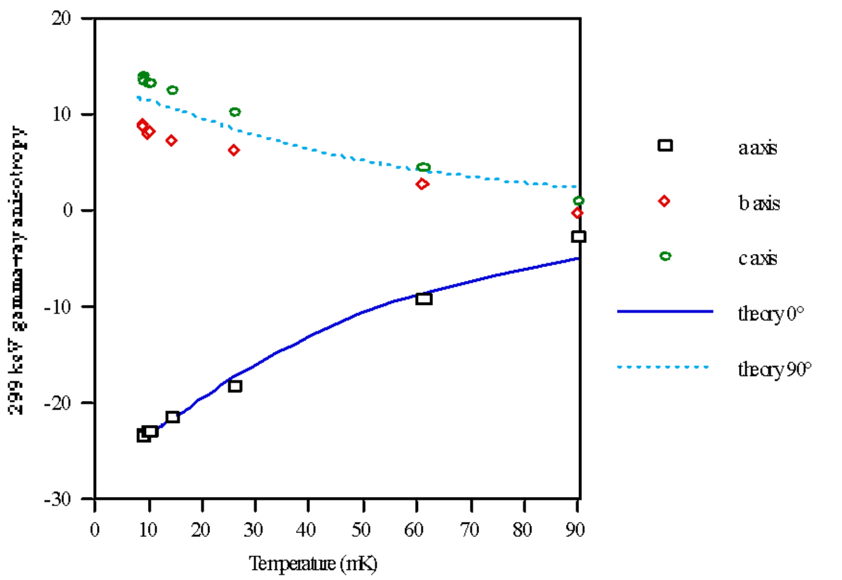
$TbNiAl_4$ - LTNO
Annealing - large effect

- LTNO data: 299 keV γ-ray of ^{160}Tb (^{160}Dy daughter) neutron activated in single crystal (^{54}Mn)Ni thermometer

- Top: annealed post activation. Lines are theoretical fits based on B_{hyp} = 290 T, P/h = 130 MHz and 90% of the nuclei contribute to orientation.

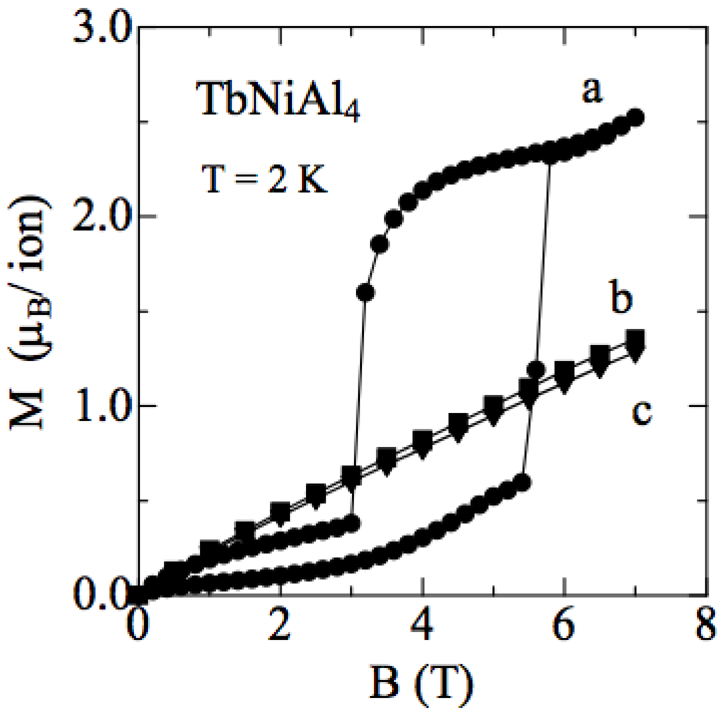
- Bottom: unannealed shows only ~50% of the anisotropy. Lines have B_{hyp} = 290 T, P/h = 130 MHz but only 47% of the nuclei contribute to orientation.

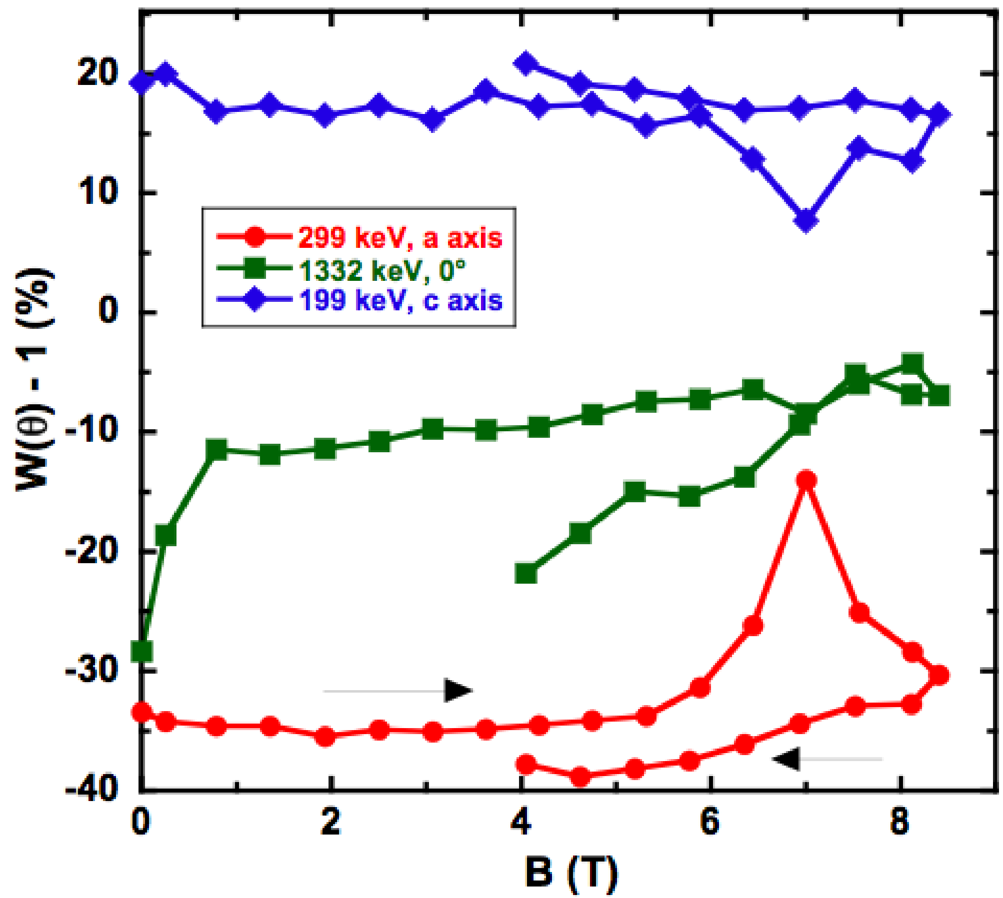




(W.D. Hutchison et al., *Hyperfine Interact.* **177** (2007) 38.)

$TbNiAl_4$ - LTNO
B field sweep





• LTNO data: 299 keV γ-ray of ^{160}Tb (^{160}Dy daughter) in single crystal $TbNiAl_4$ (annealed post activation), (^{60}Co)Co thermometer.

• Sweep B up: Non-thermal loss of anisotropy at phase transition, recovers to near full above transition (No spin flop). Sweep B down: Effect not present at same field on sweep down (hysteresis).

$NdNiAl_4$ - LTNO

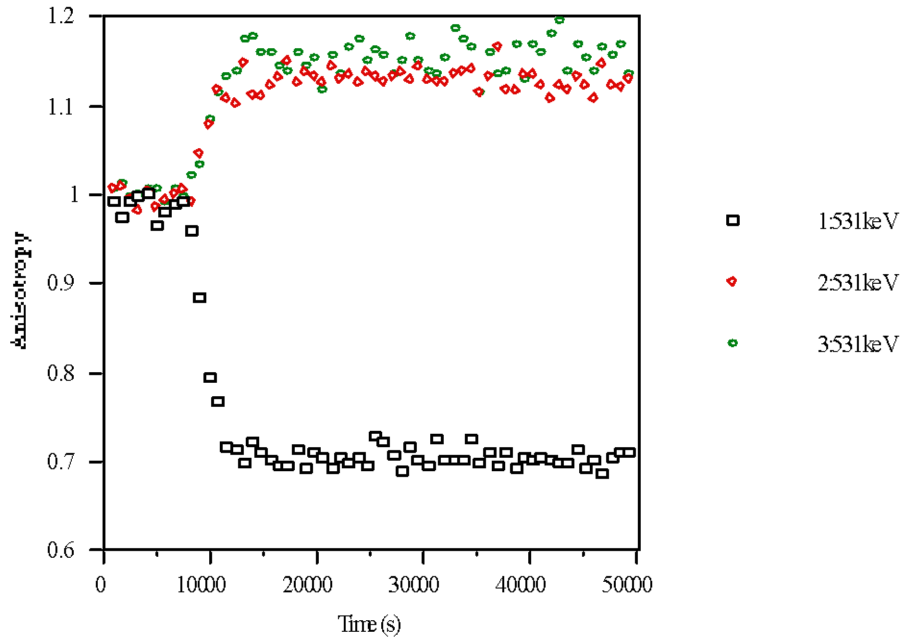
annealing - small effect

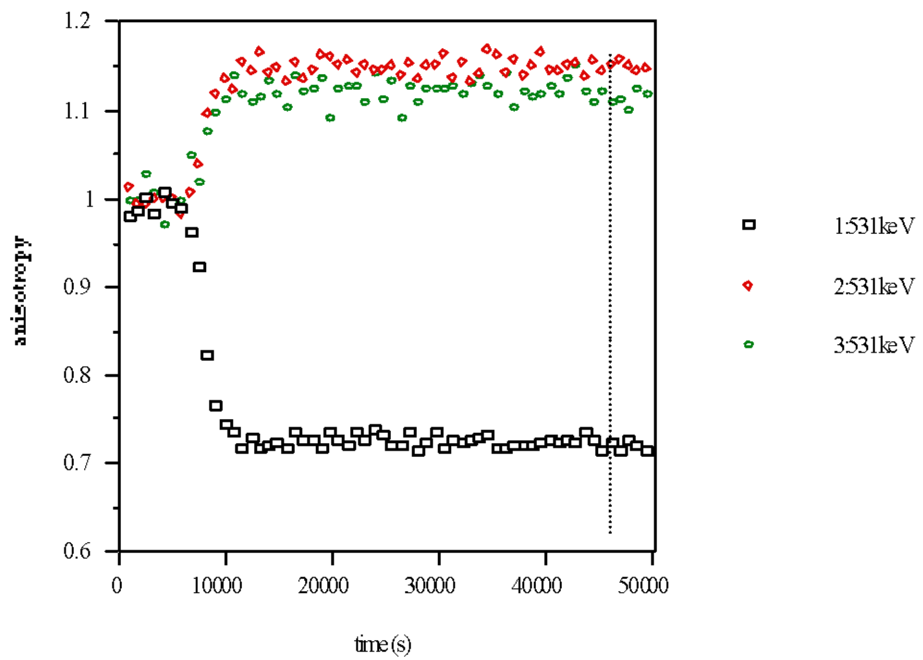
• LTNO data B_{ext} = 0.5 T (^{60}Co)Co thermometer

: 531 keV γ-ray of ^{147}Nd (^{147}Pm daughter) neutron activated in single crystal.

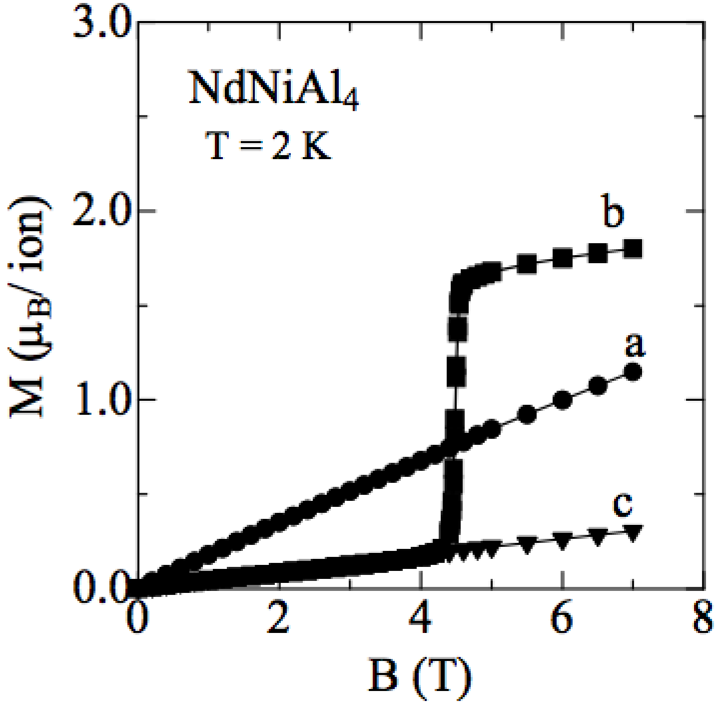
• Top: annealed post activation.

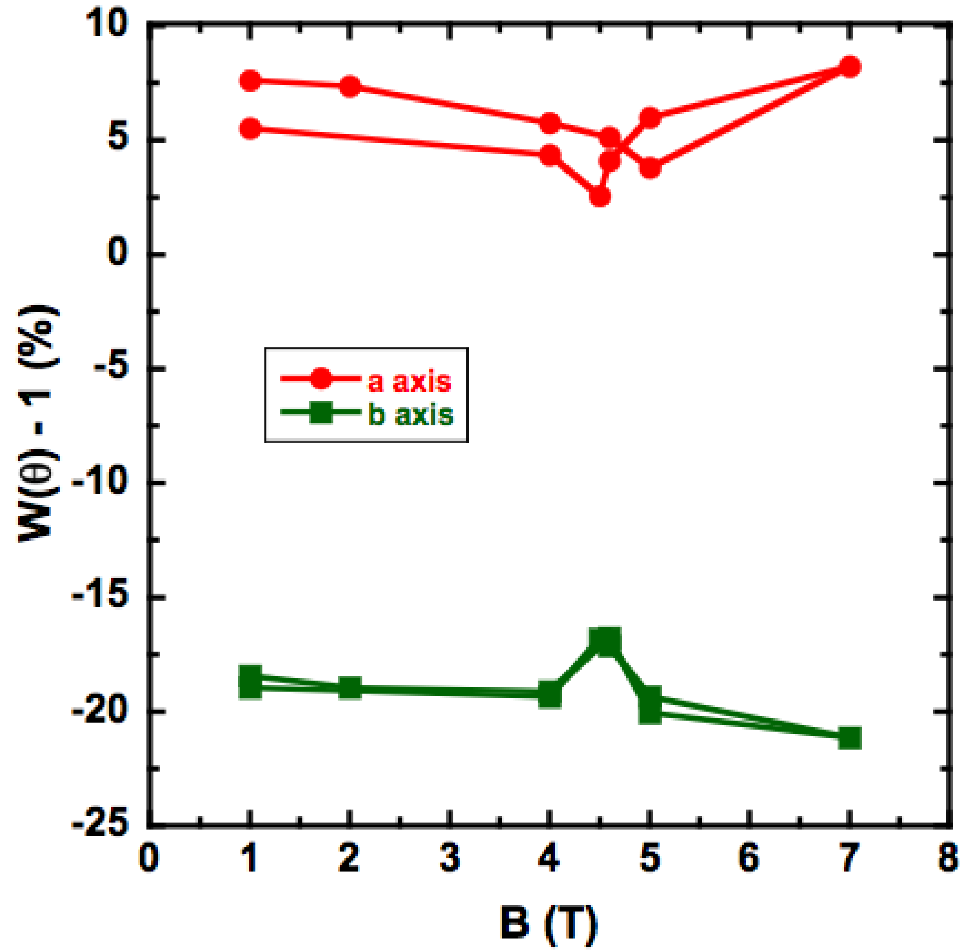
• Bottom: unannealed shows ~92% of the anisotropy of top.





$NdNiAl_4$ - LTNO
B field sweep





- LTNO data : 531 keV γ-ray of ^{147}Nd (^{147}Pm daughter) neutron activated in single crystal (^{54}Mn)Ni thermometer held at 7 mK.
- Small deviation in anistropy at phase transition field, no hysteresis.

Discussion

• $TbNiAl_4$ and $NdNiAl_4$, no spin flop, rather just a temporary fanning out of the moments during the first field induced transition.

• For $TbNiAl_4$ - LTNO consistent with the findings of neutron diffraction study whereby the magnetic phase beyond the first transition is interpreted as a mixture of ferromagnetic and incommensurate AF components. But implies continued *a* axis alignment.

• The large inverse MCE found in $TbNiAl_4$ is most significant at much higher temperatures (20 to 30 K), where the incommensurate AF component is no doubt larger. However, the fanning out of the moments observed during the transition may also play a role in entropy considerations.