# The magnetism of $Yb_2Ti_2O_7$

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**Novel Directions in Frustration and Critical Magnetism**
**Nordita, Stockholm**

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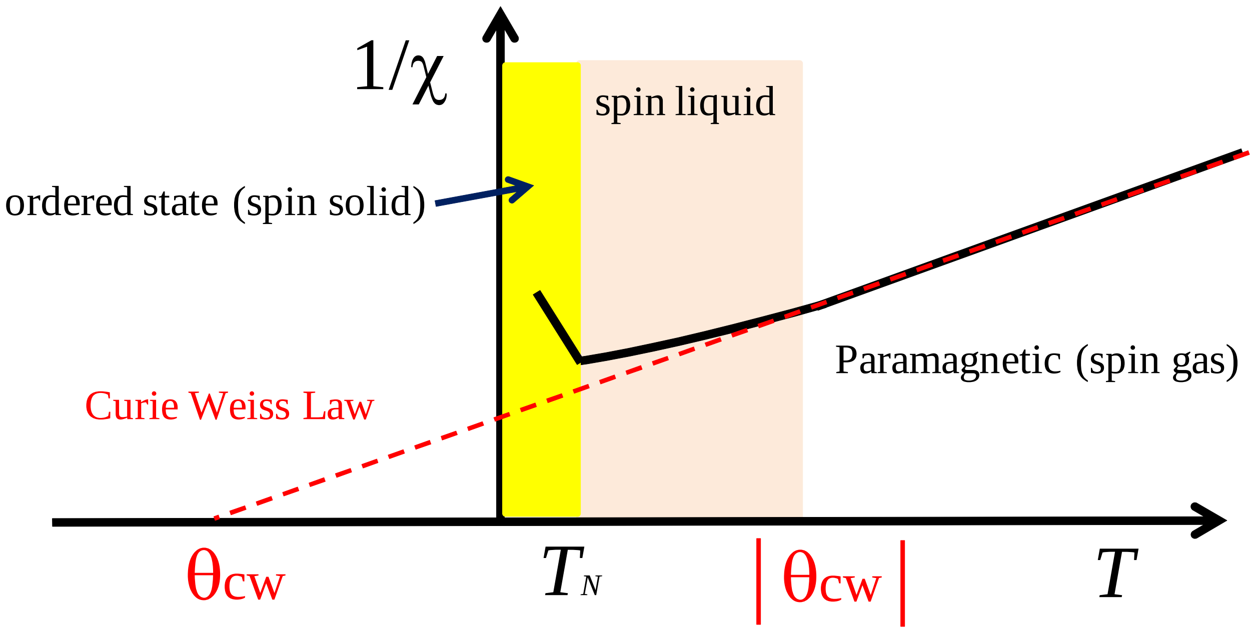
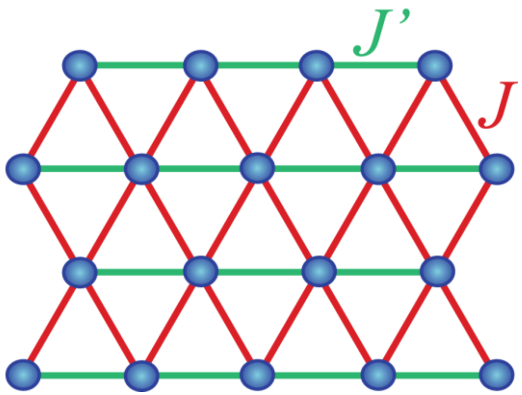
# Frustration parameter



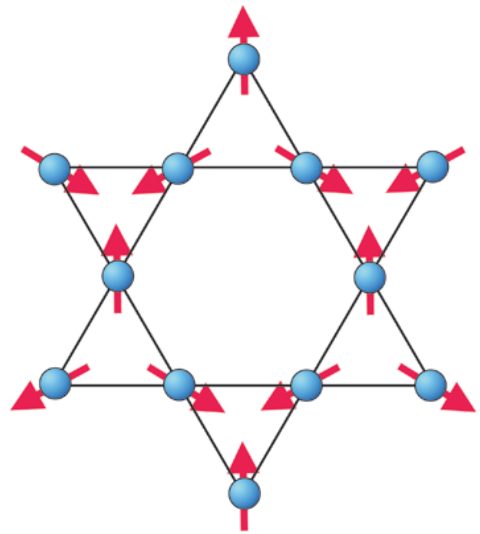

Frustration parameter, $f = |\theta_{cw}| / T_N$

When f >> 1, strongly frustrated magnets

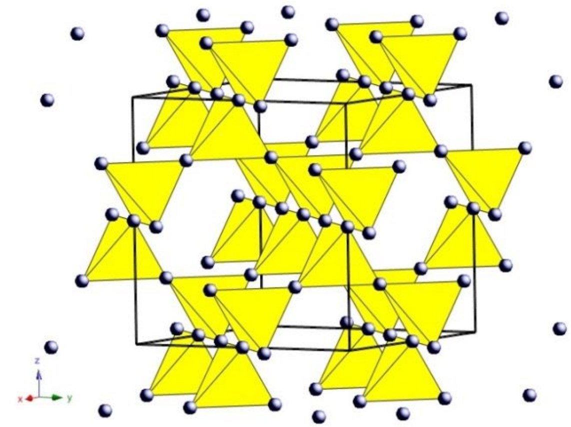
# Some candidates




A. Sachdev, Physics **2**, 90 (2009)



K. Matan et al., PRB **83**, 214406 (2011)



## Triangular lattice:

$Ba_3CuSb_2O_9$
κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$
$EtMe_3Sb[Pd(dmit)_2]_2$

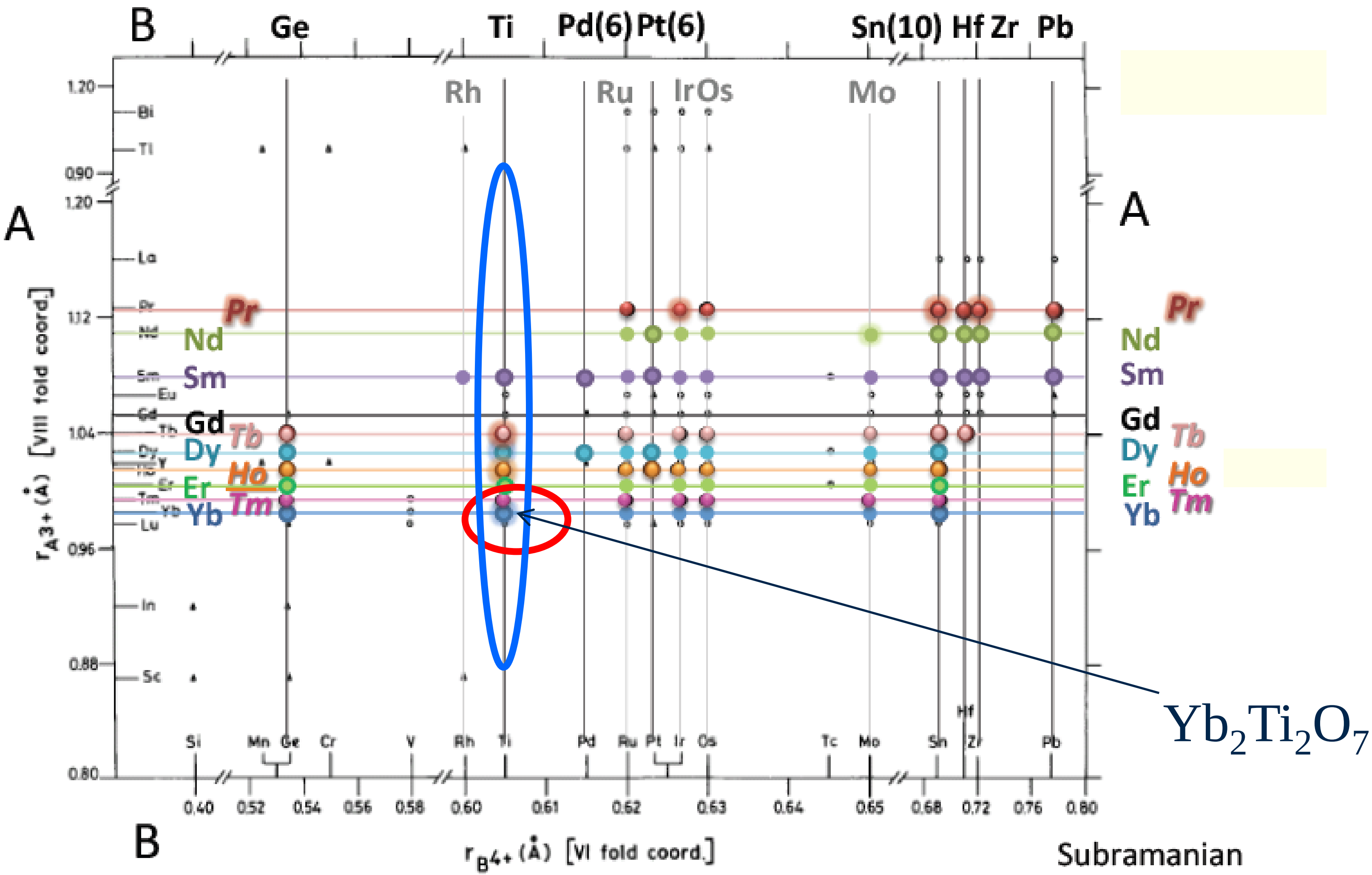
## Kagome lattice:

$ZnCu_3(OH)_6Cl_2$
$Cu_3V_2O_7(OH)_6 \cdot 2H_2O$
$BaCu_3V_2O_3(OH)_2$

## Pyrochlore lattice:

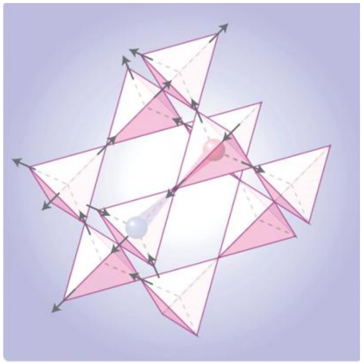
$Na_2IrO_3$
$Na_4Ir_3O_8$
$Pr_2Zr_2O_7$
$Yb_2Ti_2O_7$

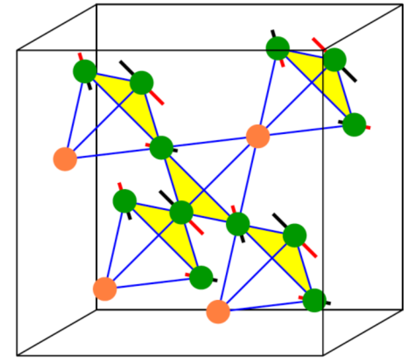
# Magnetic pyrochlore oxides $A_2B_2O_7$




Easy to grow large titanate crystals

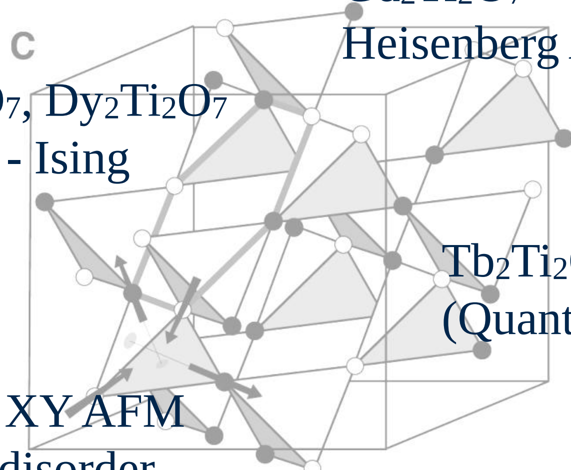
Modified by: S. Onoda



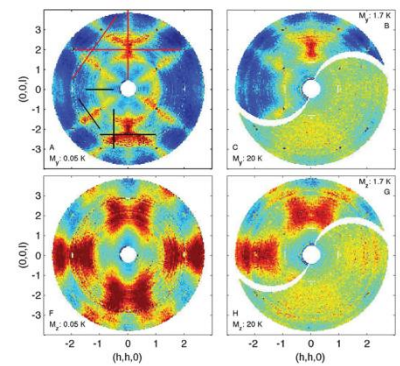


$Gd_2Ti_2O_7$
Heisenberg AFM

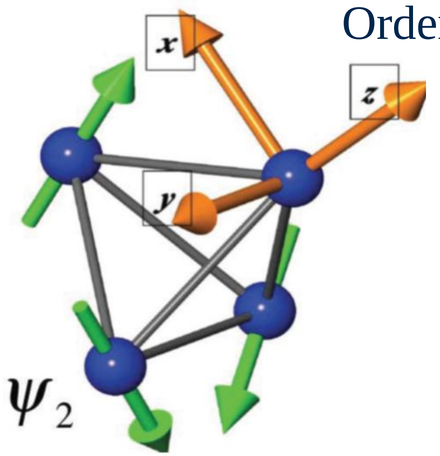
$Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$
Spin ice - Ising



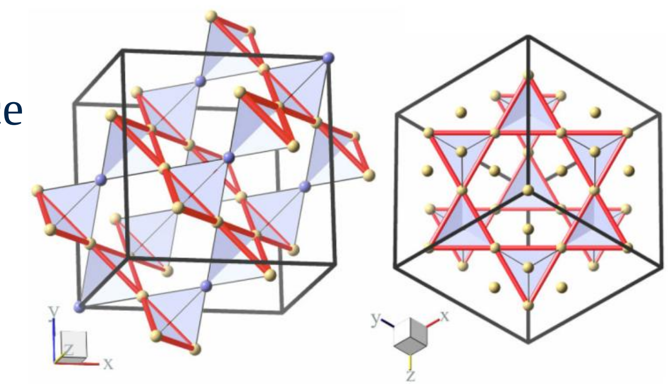

$Tb_2Ti_2O_7$
(Quantum) spin liquid




$Er_2Ti_2O_7$
Quantum XY AFM
Order by disorder



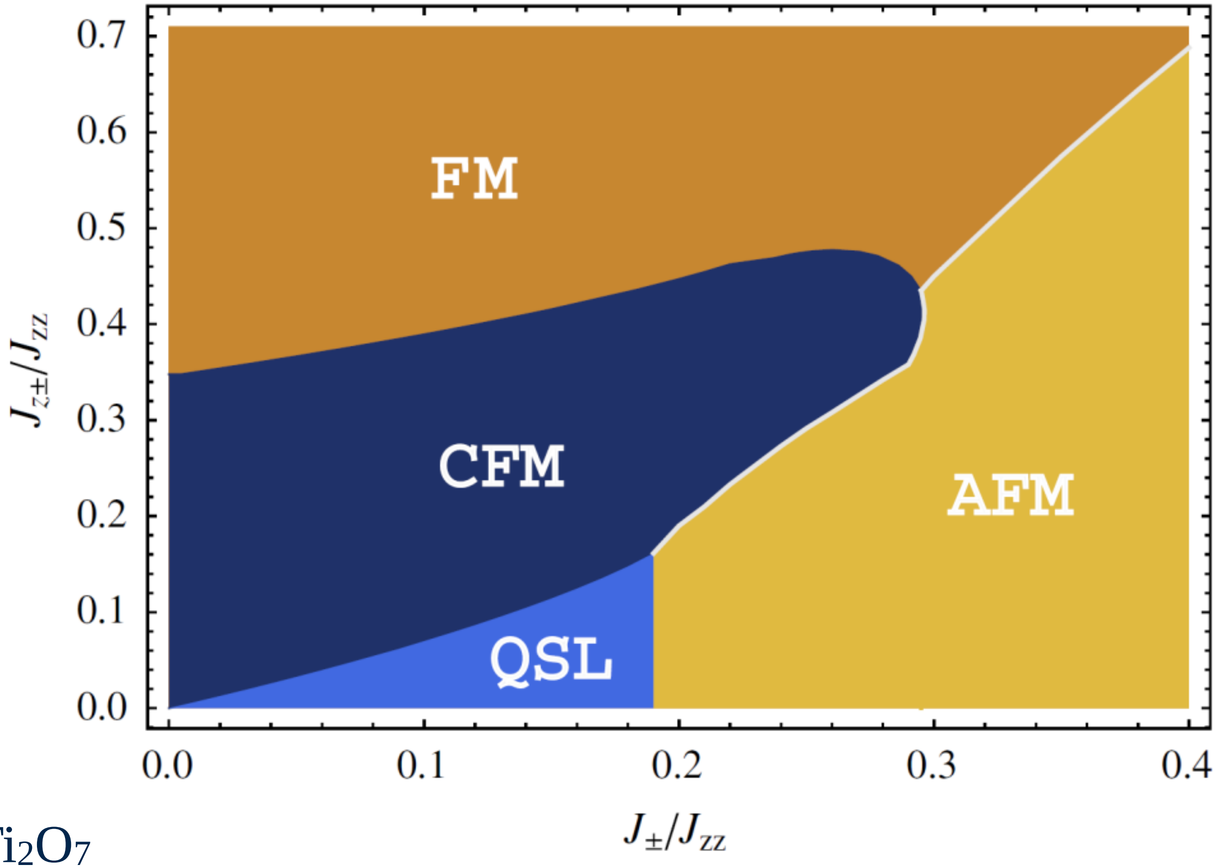

$Yb_2Ti_2O_7$
Quantum spin ice




J. S. Gardner, M. J. P. Gingras, and J. E. Greedan, Rev. Mod. Phys. **82**, 53 (2010).

$Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$
Spin ice - Ising

$Tb_2Ti_2O_7$
(Quantum) spin liquid




$Er_2Ti_2O_7$
Quantum XY AFM
Order by disorder

$Yb_2Ti_2O_7$
Quantum spin ice?

L. Savary and L Balents, Phys. Rev. Lett **108**, 037202 (2012).

# $Yb_2Ti_2O_7$ - a pyrochlore with long-range FM order

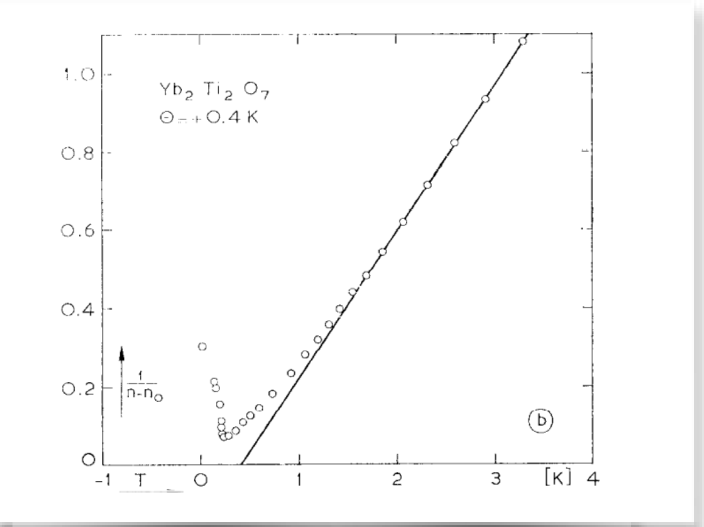
$Yb^{3+}$ ions with an effective spin- ½ .

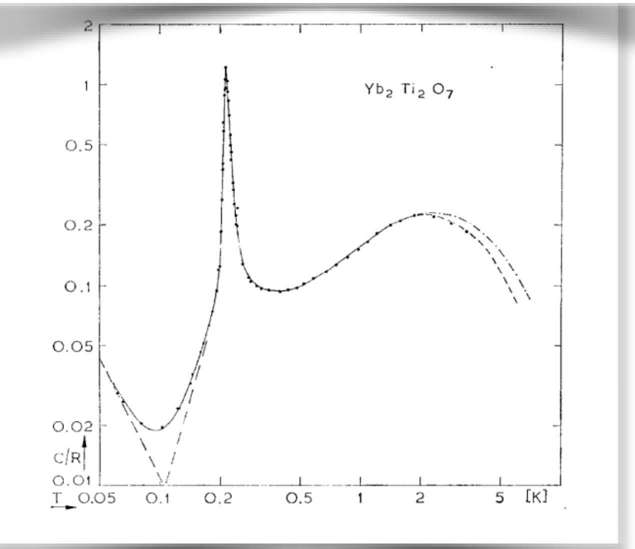
Magnetic susceptibility follows Curie-Weiss law with $\theta = +0.4$ K.

Heat capacity suggests $Yb_2Ti_2O_7$ orders (ferromagnetically) at 0.214 K.

Entropy of 0.671*R* recovered by 6 K.

cf. Full *R*ln2 entropy associated for effective spin-½  of 0.693*R* – simple!




H. W. J. Blöte, R. F. Wielinga and W. J. Huiskamp, Physica ***43***, *549 (1969).*

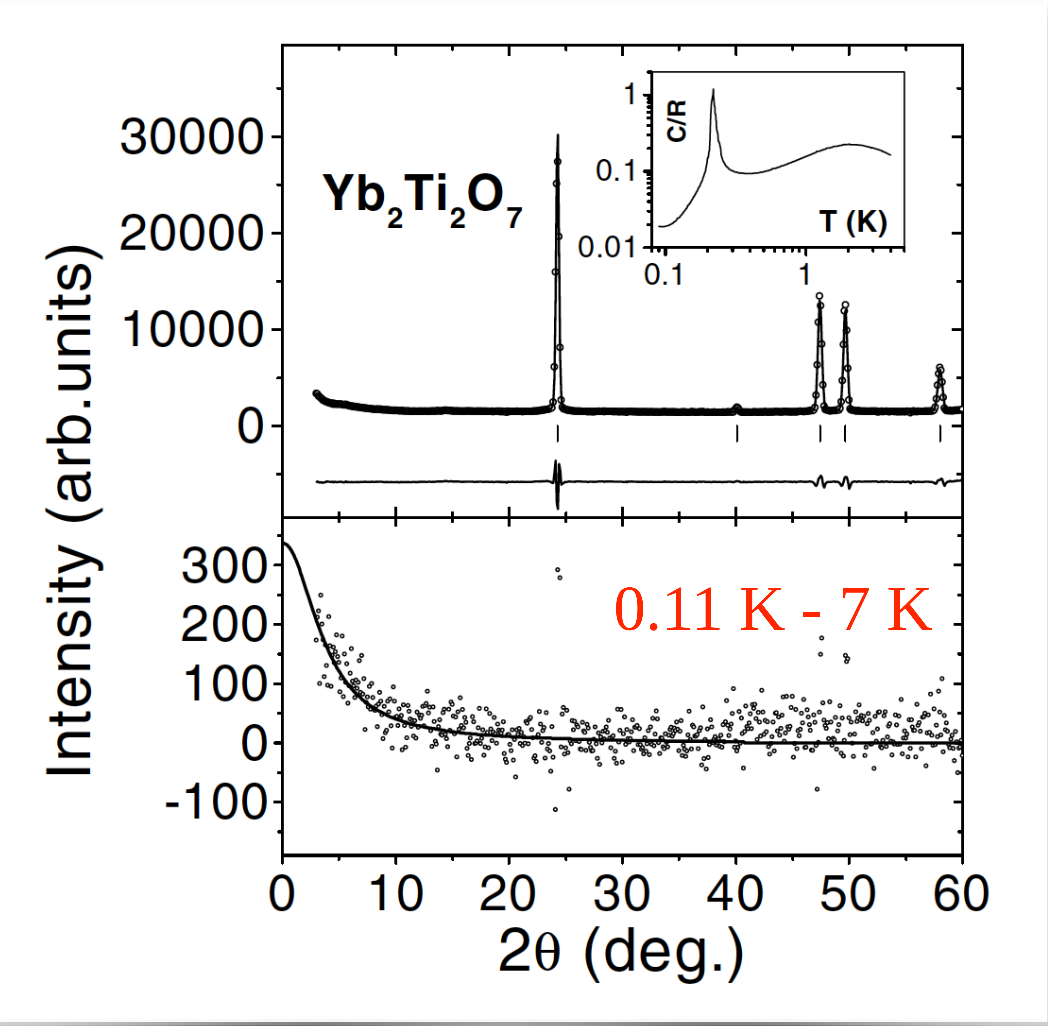
# No long-range order in polycrystalline $Yb_2Ti_2O_7$

Neutron diffraction, muon spin relaxation, and $^{170}$Yb Mössbauer.

First-order transition in spin-dynamics at 0.24 K.

**BUT**

No long-range order.




J. A. Hodges *et al*. Phys. Rev. Lett. **88** 077204 (2002).

# $Yb_2Ti_2O_7$

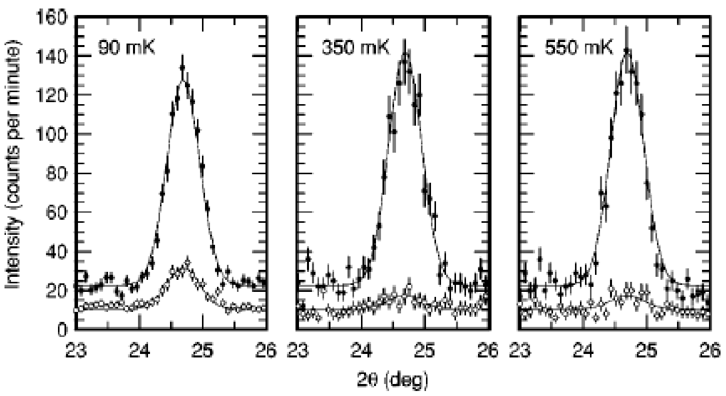



FIG. 3. Spin-flip (open circles) and non-spin-flip (closed circles) scattering at the (111) Bragg position. Above 240 mK there is only a small amount feed through due to incomplete polarization. At 90 mK, a peak is clearly seen in the spin-flip data.

J. S. Gardner *et al.*, PRB 70 180404(R) (2004)

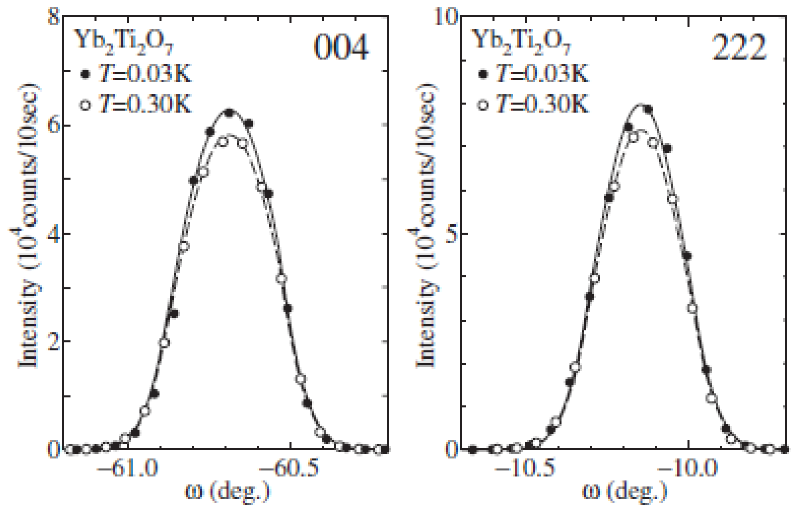



Fig. 3. Profiles of the $\omega$-scans for 004 and 222 reflections taken at 0.03 K and 0.30 K. Solid and Broken lines are guides for the eye.

Y. Yasui *et al.*, JPSJ **72** 3014 (2003)

No neutron depolarization

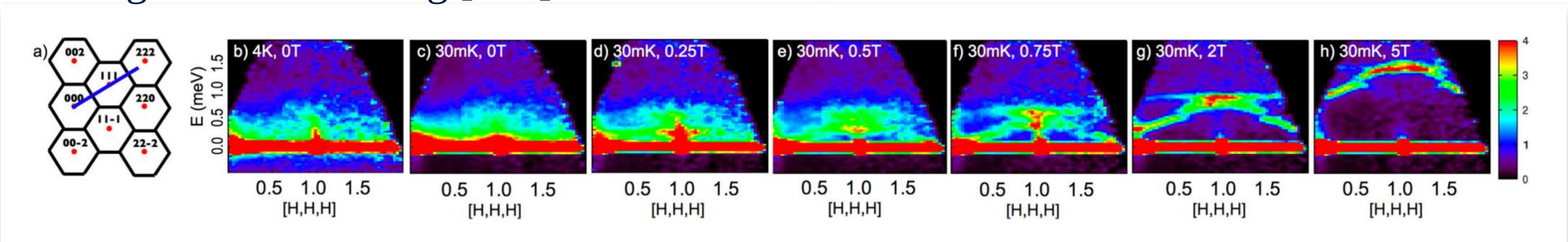
ferromagnetic

No $Yb_2Ti_2O_7$ powder shows LRO.
Only Yasui’s crystal shows LRO

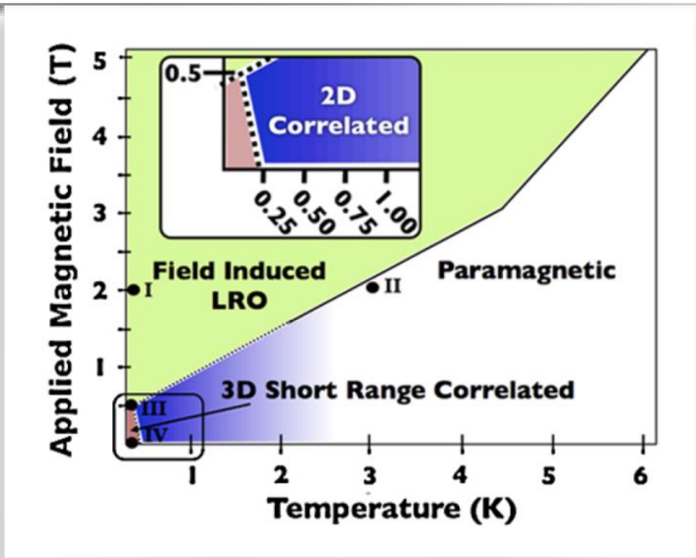
# No long-range order in single-crystal $Yb_2Ti_2O_7$

Neutron scattering suggests strong quasi-two-dimensional (2D) spin correlations at low T.

Correlations give way to long-range order under the application of modest (0.5 T) magnetic fields along [$1\bar{1}0$].




Transition at 210 mK involves a crossover from 2D correlated state to a short-ranged 3D correlated state.




K. A. Ross, J. P. C. Ruff, C. P. Adams, J. S. Gardner, H. A. Dabkowska, Y. Qiu, J. R. D. Copley, and B. D. Gaulin, Phys. Rev. Lett. **103**, 227202 (2009).

# Quantum spin ice approach

The $Yb^{3+}$ 4f magnetic moment at a site *r is described with the pseudospin-1/2 operator* $\hat{\boldsymbol{S}}_{\boldsymbol{r}} = (\hat{S}^x_{\boldsymbol{r}}, \hat{S}^y_{\boldsymbol{r}}, \hat{S}^z_{\boldsymbol{r}})$

$$\boldsymbol{m}_{\boldsymbol{r}} = \mu_B \left( g_\perp (\hat{S}^x_{\boldsymbol{r}} \boldsymbol{x}_{\boldsymbol{r}} + \hat{S}^y_{\boldsymbol{r}} \boldsymbol{y}_{\boldsymbol{r}}) + g^\parallel \hat{S}^z_{\boldsymbol{r}} \boldsymbol{z}_{\boldsymbol{r}} \right)$$

the *g-tensor components* $g^\perp = 4.18$ and $g^\parallel = 1.77$

where the *z* direction is taken along the <111> direction

$$H_D = \frac{\mu_0}{4\pi} \sum_{\langle \boldsymbol{r}, \boldsymbol{r} \rangle} \left[ \frac{\boldsymbol{m}_{\boldsymbol{r}} \cdot \boldsymbol{m}_{\boldsymbol{r}}}{(\Delta r)} - 3 \frac{(\boldsymbol{m}_{\boldsymbol{r}} \cdot \Delta \boldsymbol{r})(\Delta \boldsymbol{r} \cdot \boldsymbol{m}_{\boldsymbol{r}})}{(\Delta r)^5} \right],$$

$\boxed{\text{Spin ice}}$ + $\boxed{\text{Quantum fluctuations}}$ = $\boxed{\text{Quantum spin Ice}}$

$$H_{se} = J \sum_{\langle \boldsymbol{r}, \boldsymbol{r} \rangle}^{n.n.} \left[ \hat{S}^z_{\boldsymbol{r}} \hat{S}^z_{\boldsymbol{r}} + \frac{\delta}{2} \left( \hat{S}^+_{\boldsymbol{r}} \hat{S}^-_{\boldsymbol{r}} + h.c. \right) + \frac{q}{2} \left( e^{2i\phi_{\boldsymbol{r},\boldsymbol{r}}} \hat{S}^+_{\boldsymbol{r}} \hat{S}^+_{\boldsymbol{r}} + h.c. \right) + \frac{K}{2} \left( e^{i\phi_{\boldsymbol{r},\boldsymbol{r}}} (\hat{S}^z_{\boldsymbol{r}} \hat{S}^+_{\boldsymbol{r}} + \hat{S}^+_{\boldsymbol{r}} \hat{S}^z_{\boldsymbol{r}}) + h.c. \right) \right].$$

S. Onoda

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  - *MuSR*
  - *magnetization*
- Sample dependent
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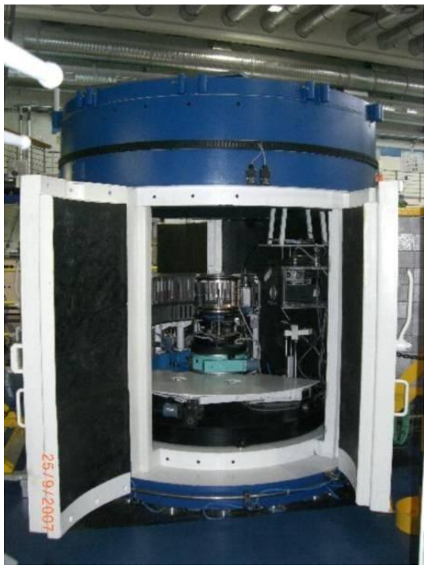
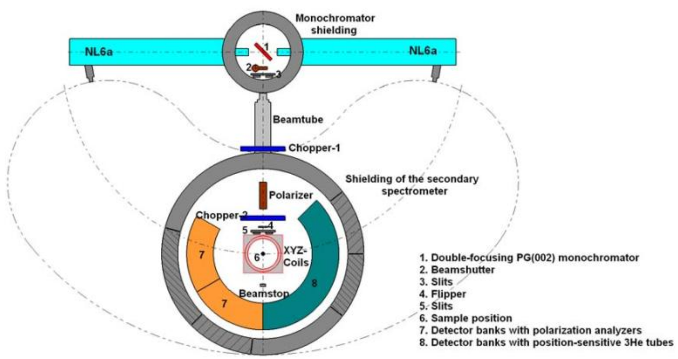
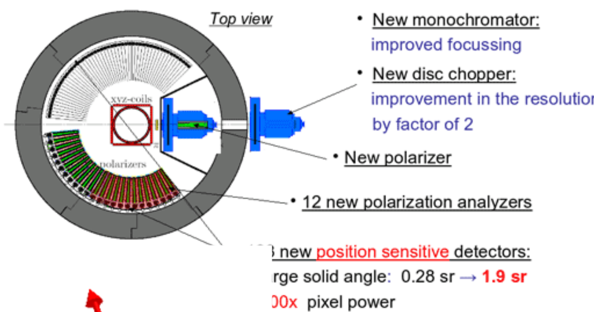
# $Yb_2Ti_2O_7$

**Single crystals were grown from IR furnace (Y. Yasui, Meiji U.)**

# DNS@FRM-II, Germany
# Diffuse Neutron Scattering

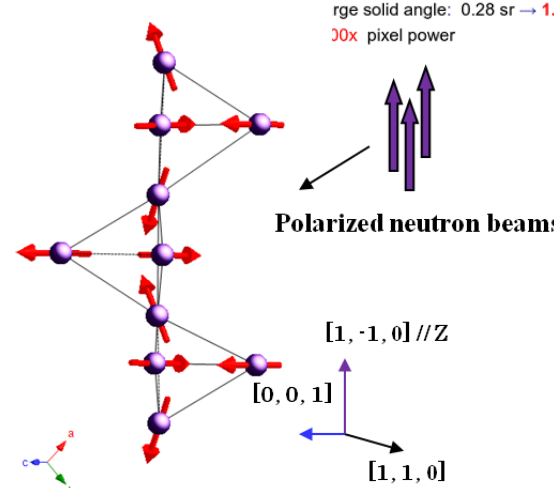




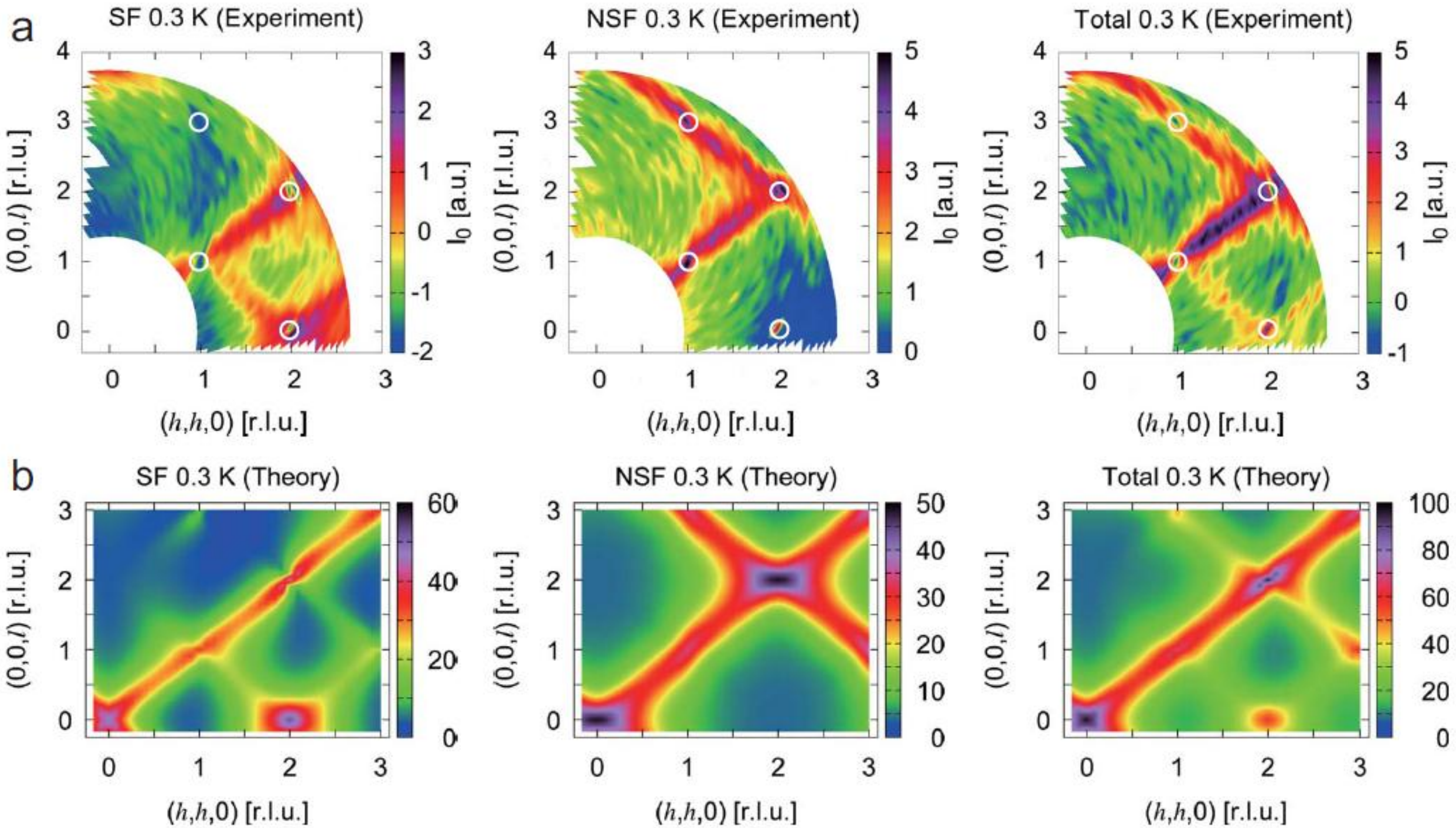

Spin-flip channel

Non-Spin-flip channel



# Diffuse neutron scattering with polarization analysis

Calculated by RPA $$I(Q) = \sum_n \sum_{i,j=1}^{4} f^{\mu\nu}_{\boldsymbol{Q},i,j} \frac{\phi^{\mu}_{-\boldsymbol{Q},n,i}\phi^{\nu}_{\boldsymbol{Q},n,j}}{3+\epsilon_{\boldsymbol{Q},n}/T}.$$



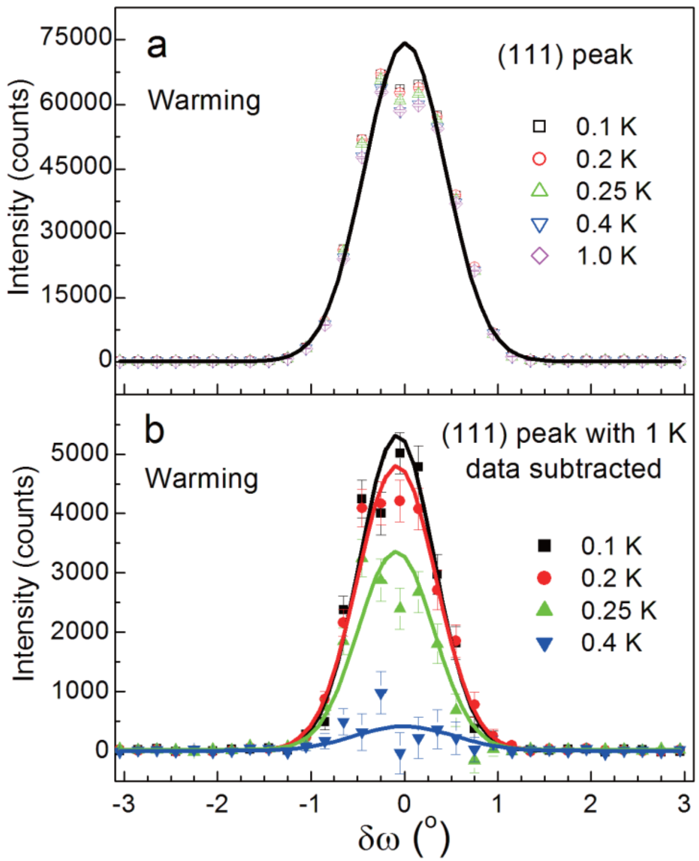

$$f^{\mu\nu}_{\boldsymbol{Q},i,j} = \frac{1}{4|\boldsymbol{Q}|^4}\left[\boldsymbol{Z}\cdot(\boldsymbol{Q}\times(g_\mu \boldsymbol{e}^\mu_i \times \boldsymbol{Q}))\right]\left[\boldsymbol{Z}\cdot(\boldsymbol{Q}\times(g_\nu \boldsymbol{e}^\nu_j \times \boldsymbol{Q}))\right],$$

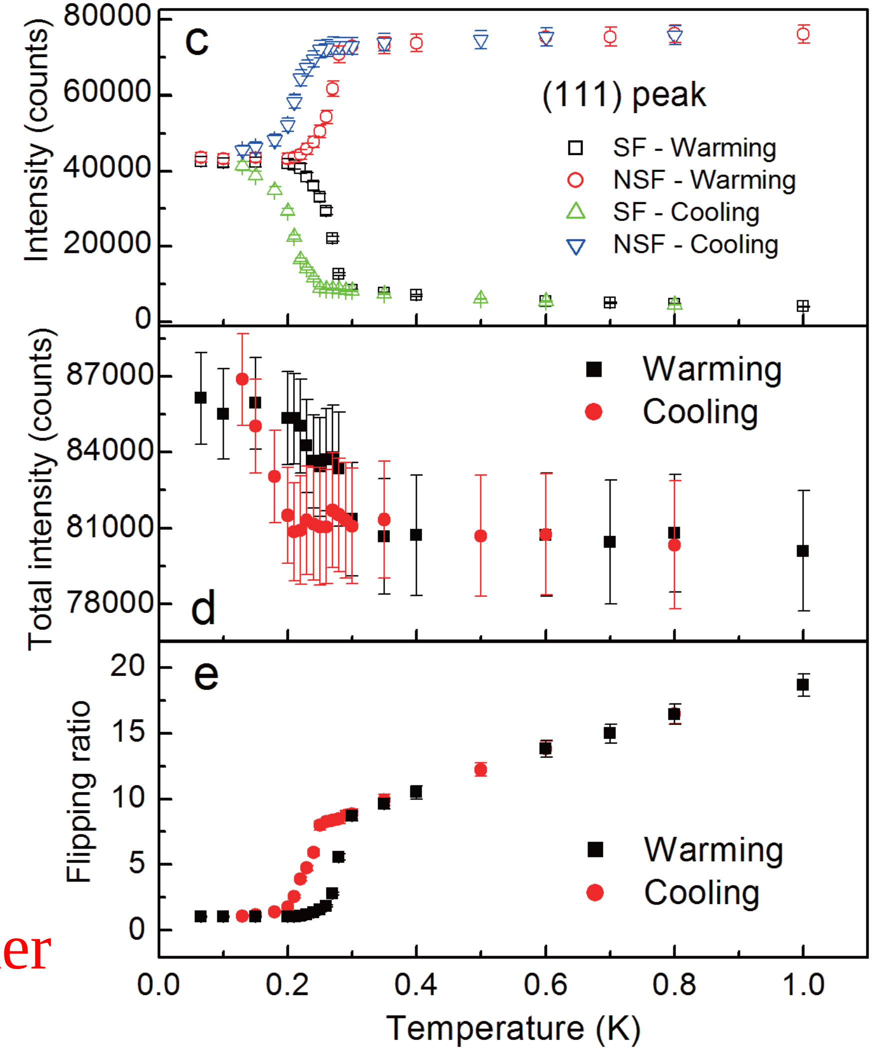
with $\boldsymbol{Z} = (1,-1,0)/\sqrt{2}$ for the NSF channel and $$f^{\mu\nu}_{\boldsymbol{Q},i,j} = \frac{1}{4|\boldsymbol{Q}|^4}\left[\boldsymbol{Q}\times(g_\mu \boldsymbol{e}^\mu_i \times \boldsymbol{Q})\right]\cdot\left[\boldsymbol{Q}\times(g_\nu \boldsymbol{e}^\nu_j \times \boldsymbol{Q})\right]$$

for the total. The SF channel is obtained as the difference between the above two.

L. J. Chang/ S. Onoda *et al.*, Nat. Commun. 3, 992 (2012)

# Peak (111) intensities



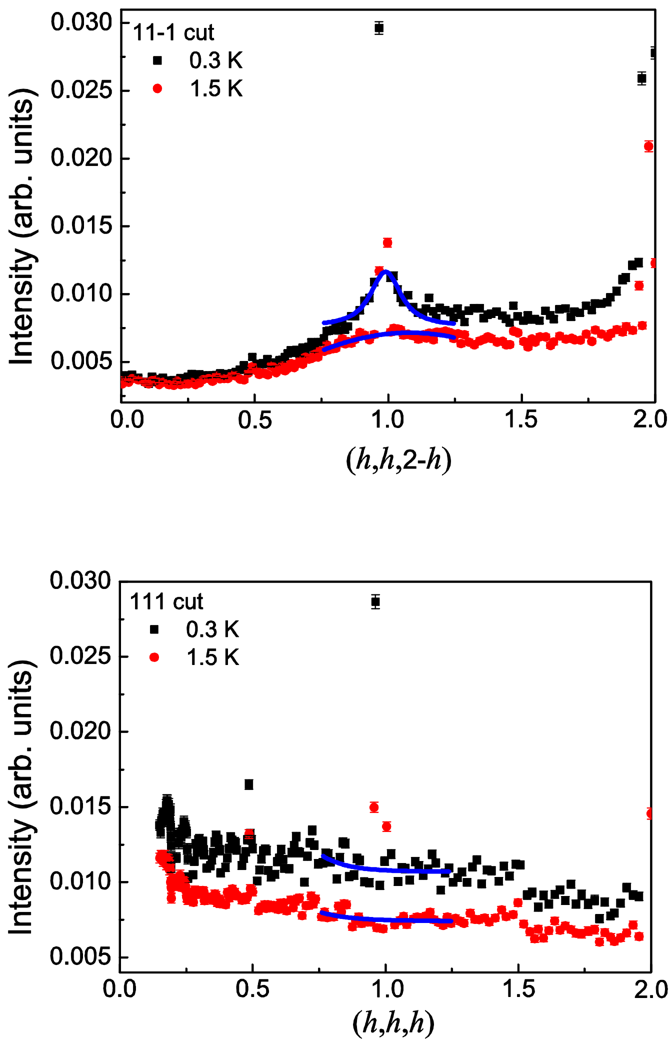

Depolarization: ferromagnetic order

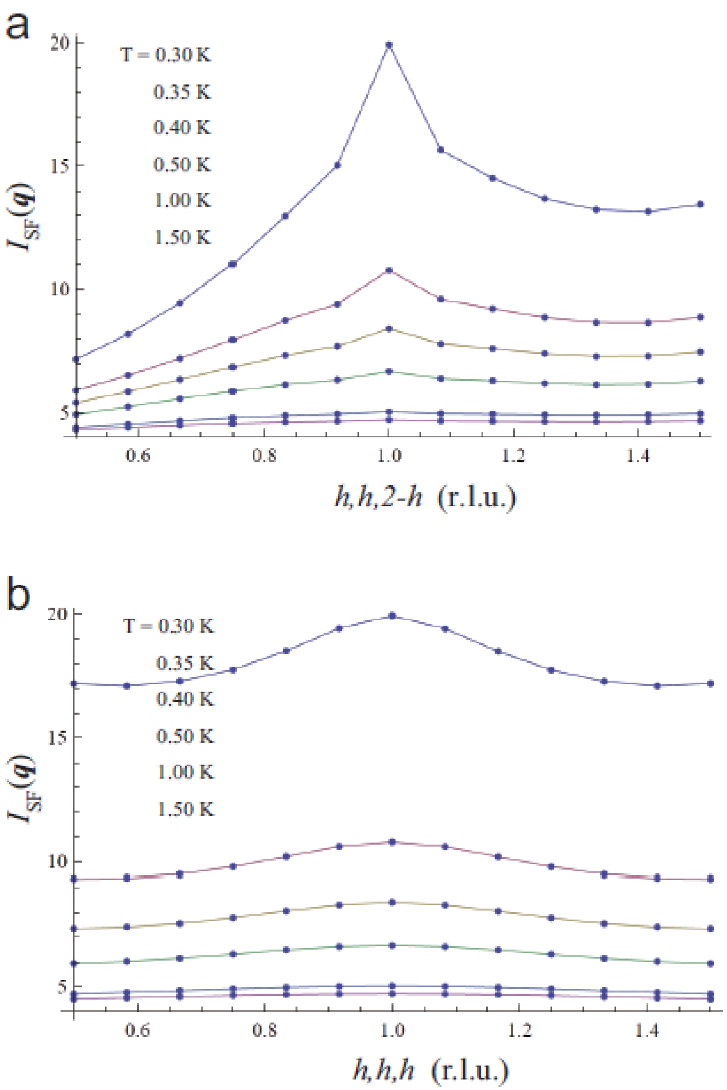
First-order transition

The widths of 111 peaks are identical

L. J. Chang/ S. Onoda *et al.*, Nat. Commun. 3, 992 (2012)

# Pinch point cut

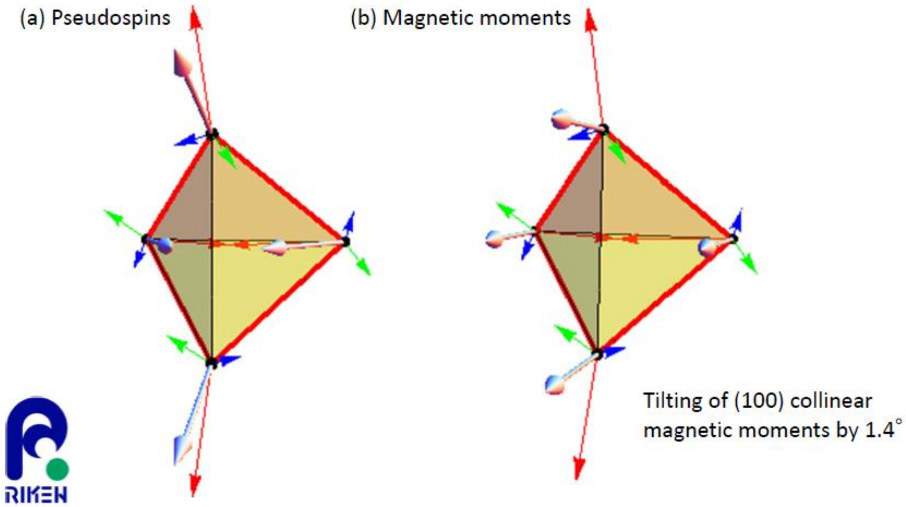



L. J. Chang/ S. Onoda *et al.*, Nat. Commun. 3, 992 (2012)

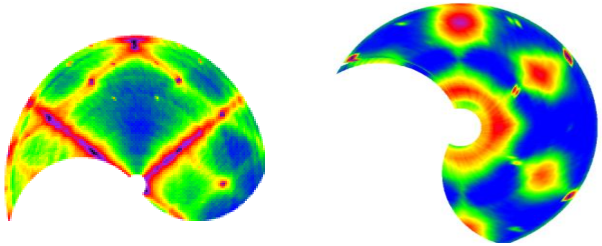
# Phase diagram

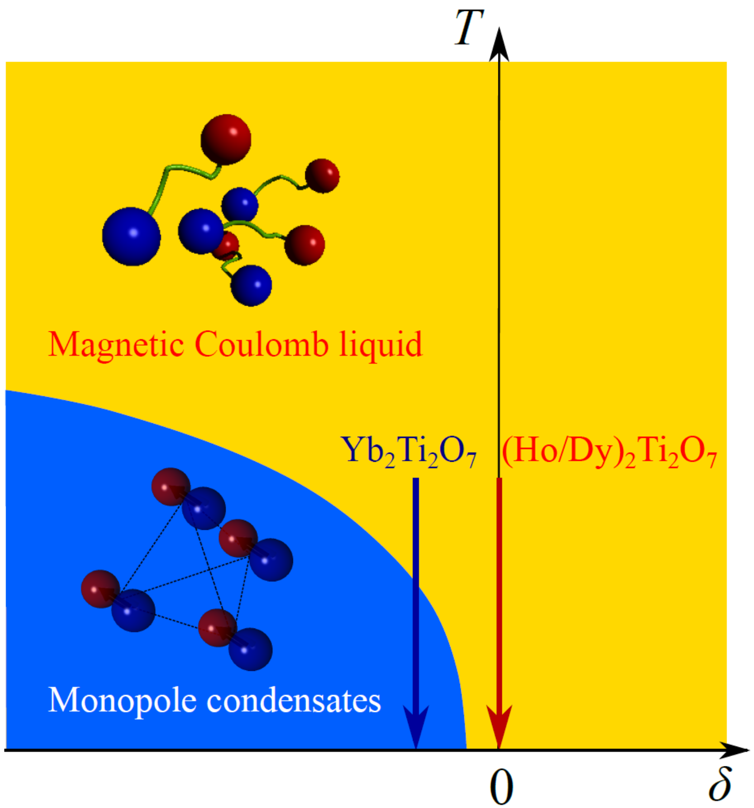
## Ground-state pseudospins and magnetic moments




moment amplitude $1.3\mu_B$

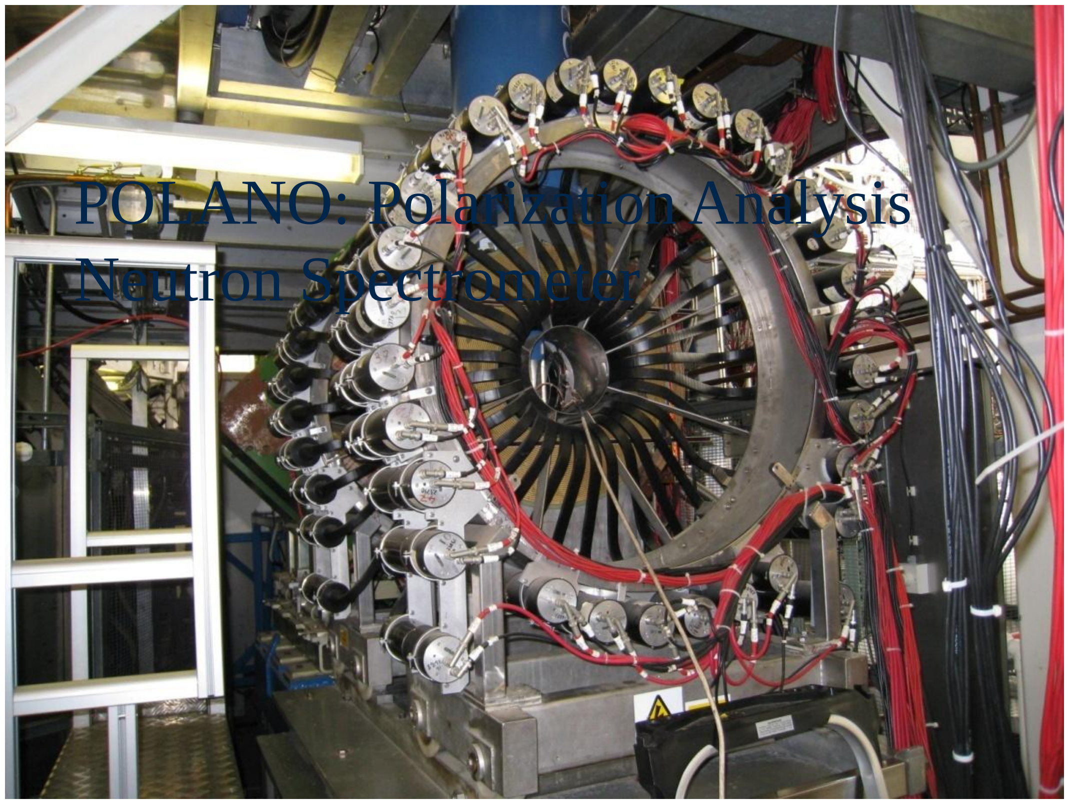
Nearly collinear ferromagnet
(~1 degree canting) , M//[100]






L. J. Chang/ S. Onoda *et al.*, Nat. Commun. 3, 992 (2012)

# MuSR experiments @ μSR, ISIS

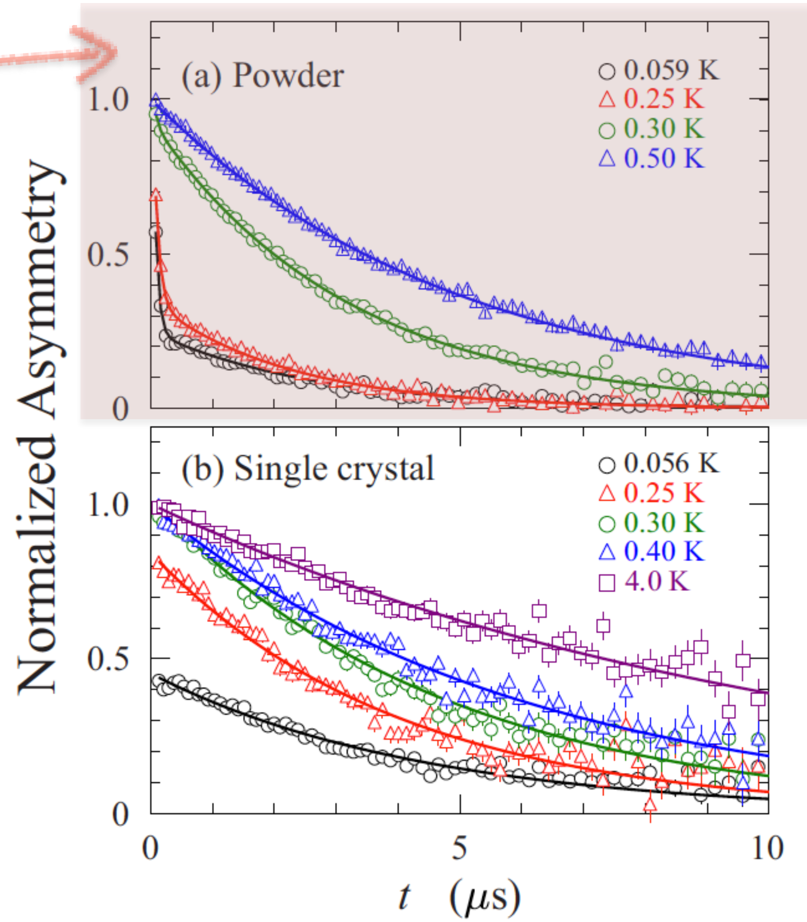


POLANO: Polarization Analysis Neutron Spectrometer

# Zero-field μSR – powder sample

$A(t)$ normalized by $A(t = 0, T = 0.6\ \mathrm{K})$.

**Powder**




At 0.5 K, exponential decay of $A(t)$ - indicates Yb spins are fluctuating, yielding a slow relaxation of the muon spins.

At 0.25 K, [slightly below $T_C$ determined from the $C(T)$], steep drop in $A(t < 0.5\ \mu s)$, followed by a slow relaxation.

Initial drop indicates muon spins are depolarized more quickly in the pulse duration of 70 ns.

Strong evidence Yb moments are static or quasi-static within $\mu$SR time window (10 ps to 1 $\mu$s).

L.-J. Chang *et al*., Phys. Rev. B **89**, 184416 (2014).

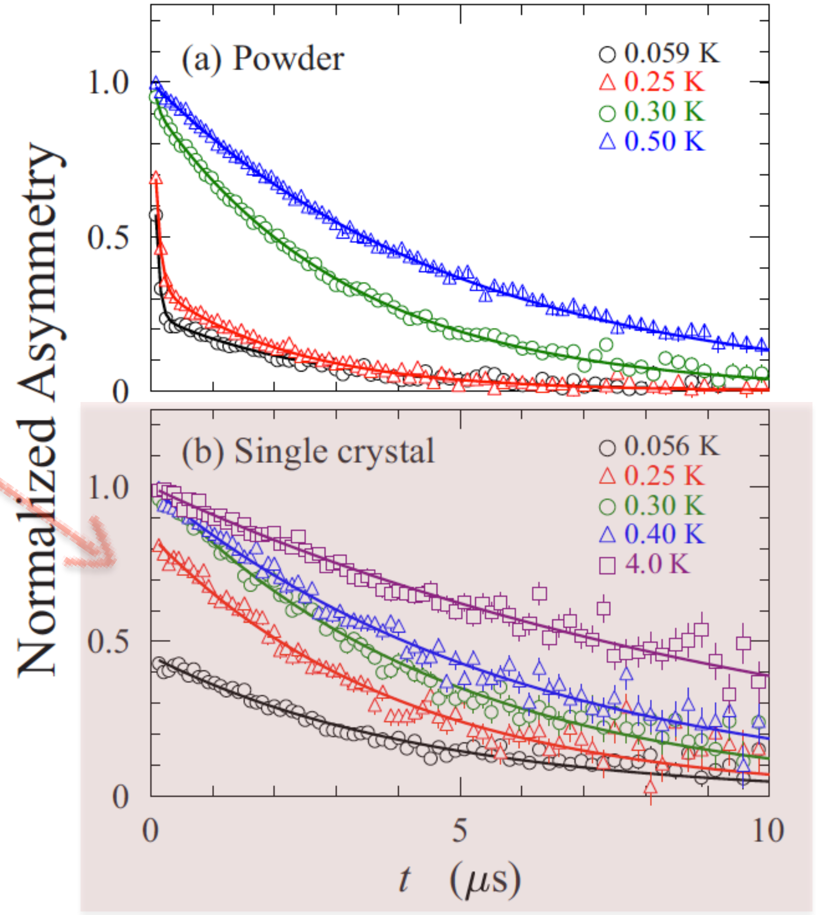
# Zero-field μSR – single crystal sample

**Single crystal**

At high $T$ (4.0, 0.4, and 0.3 K) relaxation of muon spins is slightly slower than in powder.

At $T$ = 0.25 K, we again observe a rapid initial drop in the asymmetry followed by a slow relaxation.

Size of the initial decrease in asymmetry is reduced from that of the powder.




L.-J. Chang *et al.*, Phys. Rev. B **89**, 184416 (2014).

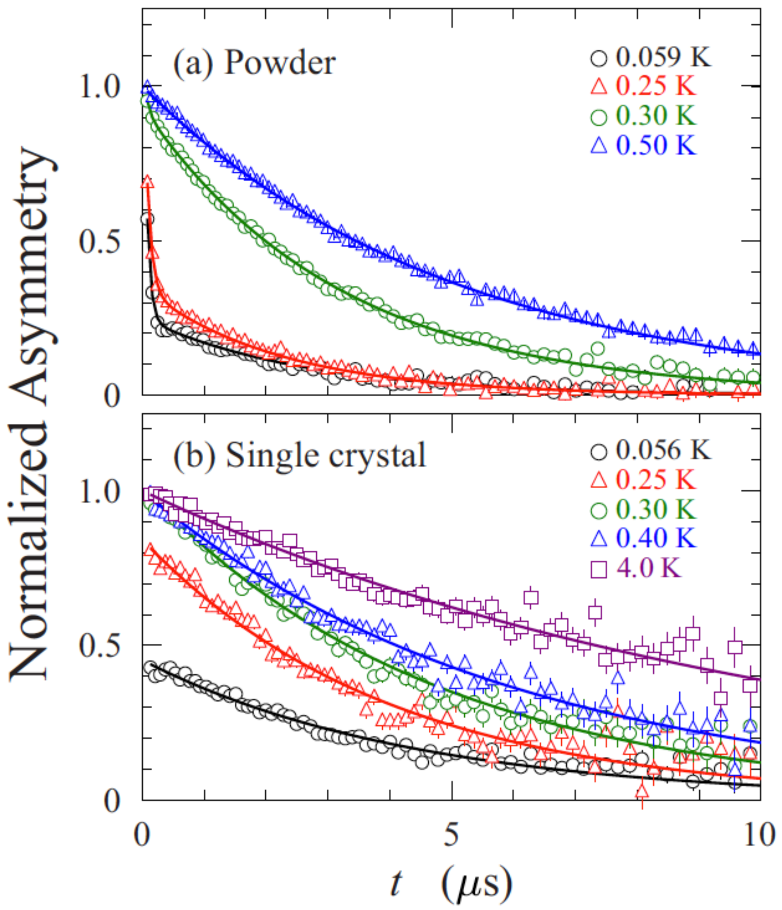
# Zero-field μSR - *A*(*t*)

$\mu$SR time spectra can be analysed using:

$$A(t) = A_1\exp(-\lambda_1 t) + A_2\exp(-\lambda_2 t)\ .$$

fast

slow




L.-J. Chang *et al*., Phys. Rev. B **89**, 184416 (2014).

# Zero-field μSR - $A(t)$

$\mu$SR time spectra can be analysed using:

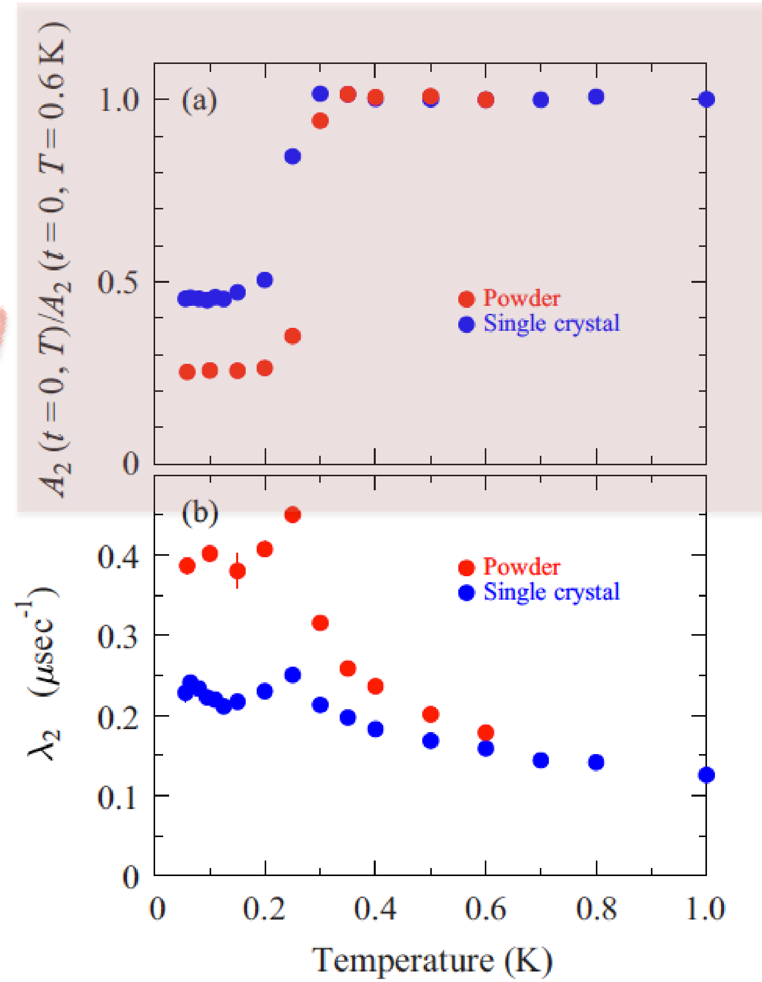
$$A(t) = A_1 \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t) .$$

fast          slow

$A_2(t = 0)$ is nearly $T$ independent at high $T$.

Below ~0.3 K, $A_2(t = 0)$ falls to an almost constant value of ¼ and ½ its high-$T$ value for powder and single crystal respectively.

=> volume fraction of static or quasi-static moments is ~80 - 100%.



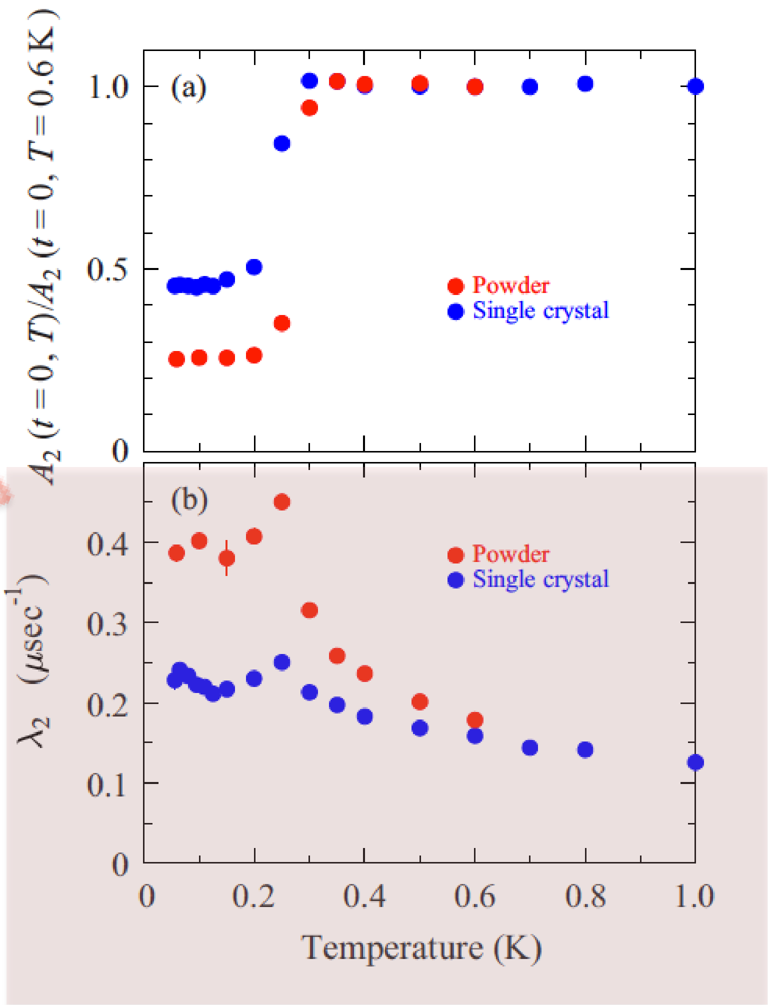

L.-J. Chang *et al.*, Phys. Rev. B **89**, 184416 (2014).

# Zero-field μSR - λ(*t*)

Relaxation rate $\lambda_2$ same at ~1 K, reaching a maximum at ~0.25 K.

$\lambda_2$ for powder enhanced by a factor of 2, compared with single-crystal.

Peak in $\lambda_2$ indicates that the time scale of magnetic fluctuations increases and crosses $\mu$SR time window, (10 ps to 1 $\mu$s), around this $T$.



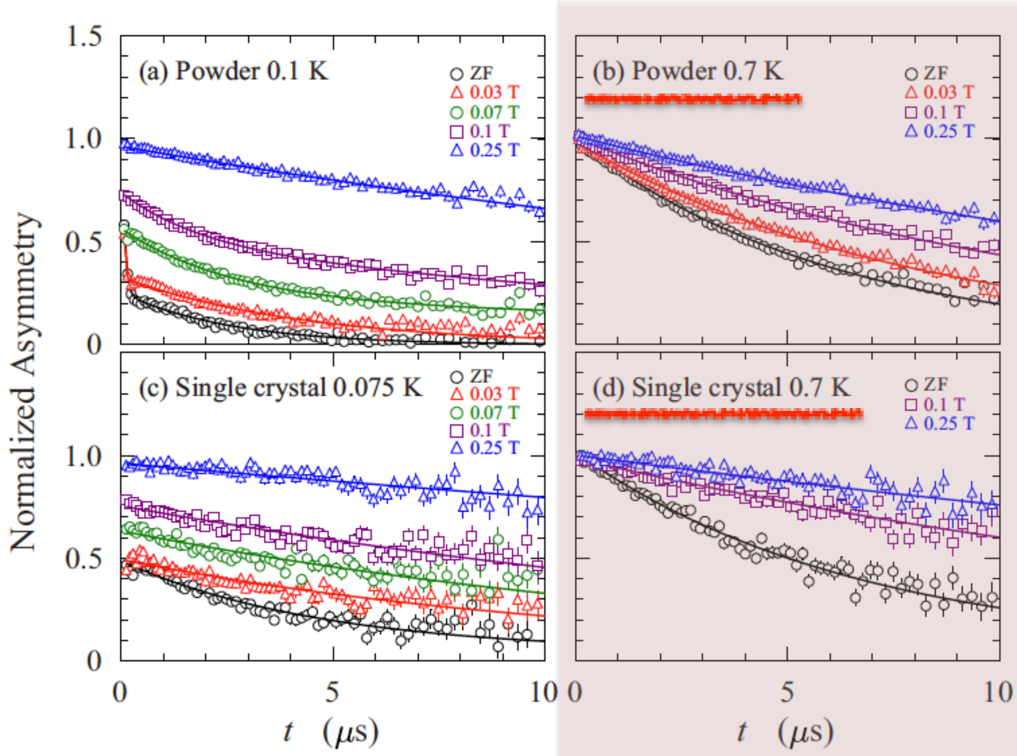

L.-J. Chang *et al.*, Phys. Rev. B **89**, 184416 (2014).

# LF-field μSR

$\lambda_2$ decreases monotonically with increasing longitudinal field both well below and well above $T_C$.

At 0.7 K, $A(t)$ still shows a single slow exponential time decay even in a magnetic field of 0.25 T.

Some of the magnetic excitations lie in the $\mu$SR time window in this $T$ and field range.



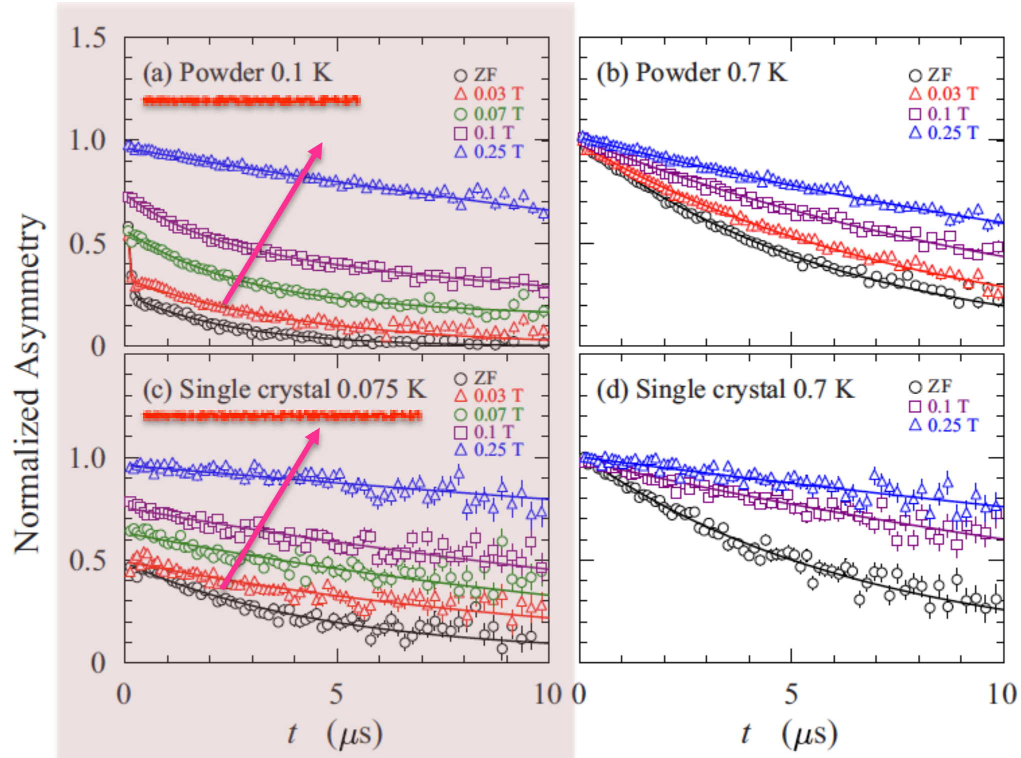

L.-J. Chang *et al*., Phys. Rev. B **89**, 184416 (2014).

# LF-field μSR

Below $T_C$ (~0.1 K) $A(t)$ shifts upward with increasing field, with no crossing.

Shift accompanied by recovery in asymmetry.

Initial drop in asymmetry disappears at 0.25 T - approx. order of $T_C$.




L.-J. Chang *et al.*, Phys. Rev. B **89**, 184416 (2014).

# LF-field μSR

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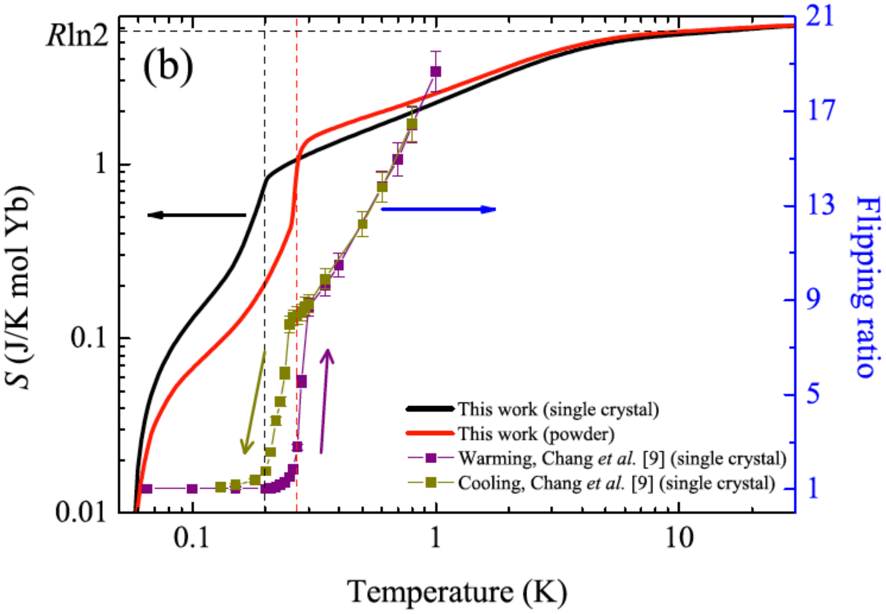
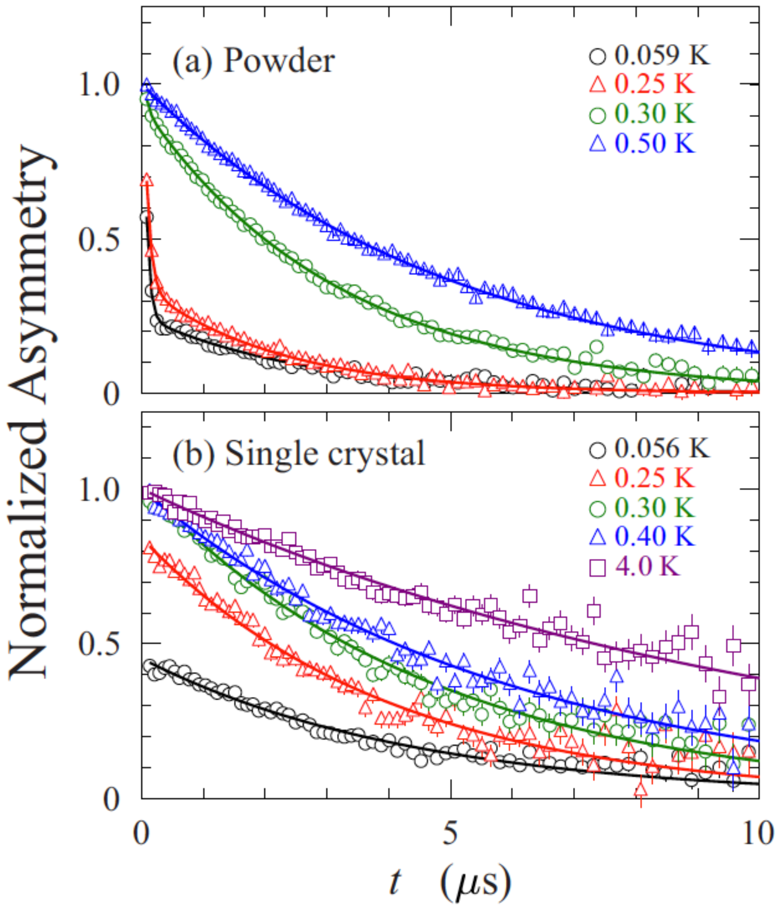
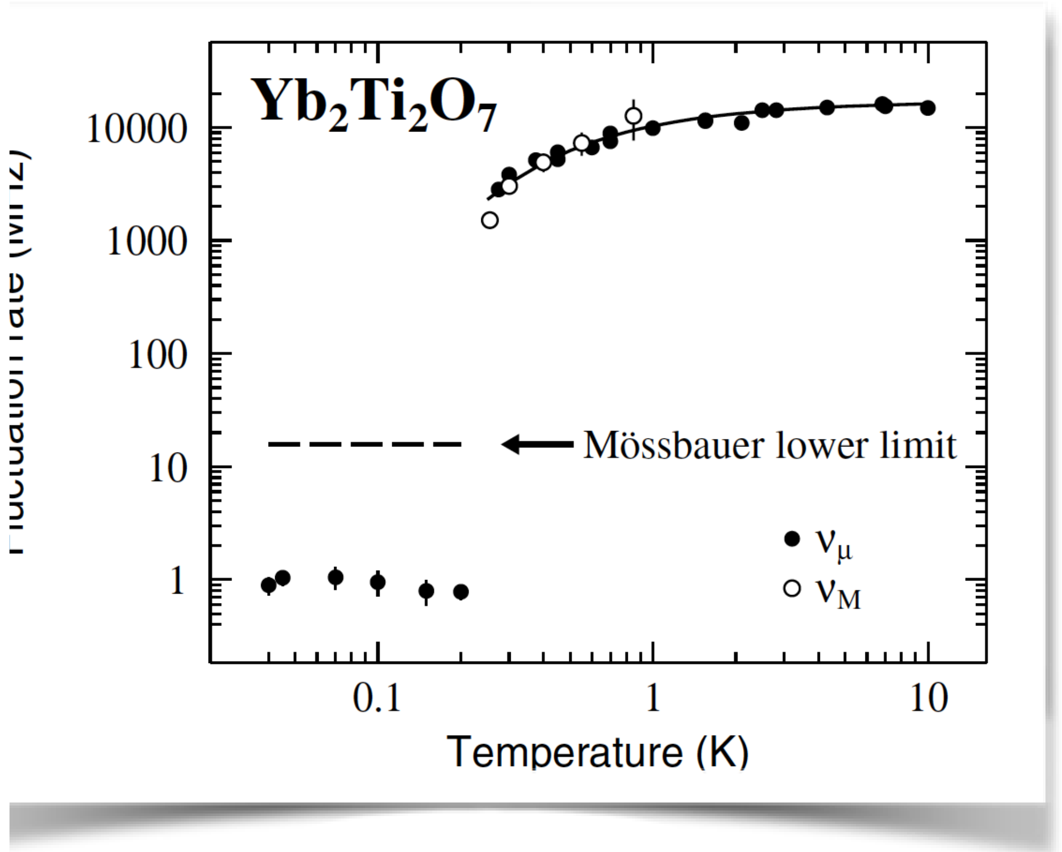



Muon spectroscopy ⇒ static or quasi-static momen s.
Full entropy from heat capacity ⇒ long-range orde

L.-J. Chang *et al.*, Phys. Rev. B **89**, 184416 (2014).

# Other μSR studies




First-order transition in spin fluctuation rate.

J. A. Hodges *et al*. Phys. Rev. Lett. **88**, 077204 (2002).
P. Dalmas de Réotier *et al*. Physica B **374-375**, 145 (2006).

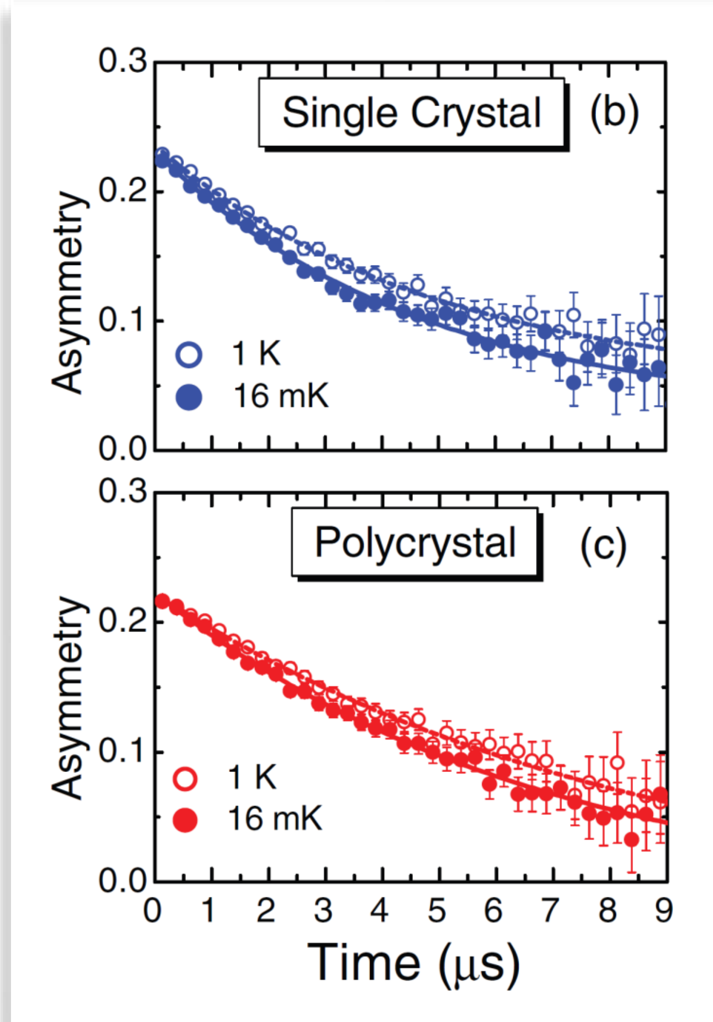
# Other μSR studies

No difference in A($t$) data between high $T$ (1 K) and base $T$ (16 mK) for single crystal or powder.

No long-range magnetic order.

BUT

Also no change in fluctuation rate.




R. M. D. D' Ortenzio *et al*. Phys Rev B **88**, 134428 (2013).

# Discussions on the MuSR results

We had carried out MuSR experiments on powder sample, “long-ranged order” crystal, and a crystal without HC peak.

- Emergence of static (<1 MHz) internal magnetic moments observed for powder and “long-ranged order” crystal.
- Temperature vs relaxation rate of the crystal without HC peak is close to that reported in the D. M. D’Ortenzio paper. → magnetic interaction is weaker in this sample (small fraction of the crystal contributed to magnetic order)
- The initial drop in the asymmetry on our single-crystal is similar to that on the powder worked by Hodges et al. (Volume fraction of the static moments is ~80% in our single crystal.)

# Magnetisation - *M*(*T*)

Powder and single crystal samples.

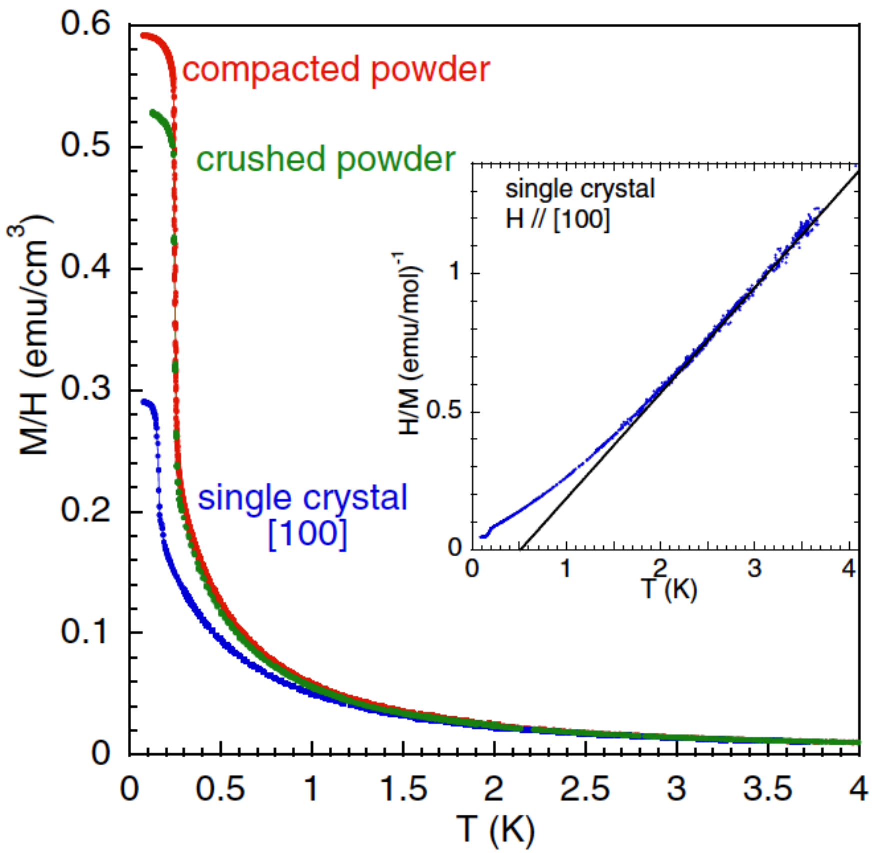
Low-*T*, SQUID magnetometers.

Crushed powder with Cu paste.

*M*/*H* vs *T* for *H*//[100] and [110];
*H* = 5 Oe.

At high *T*, Curie - Weiss law.

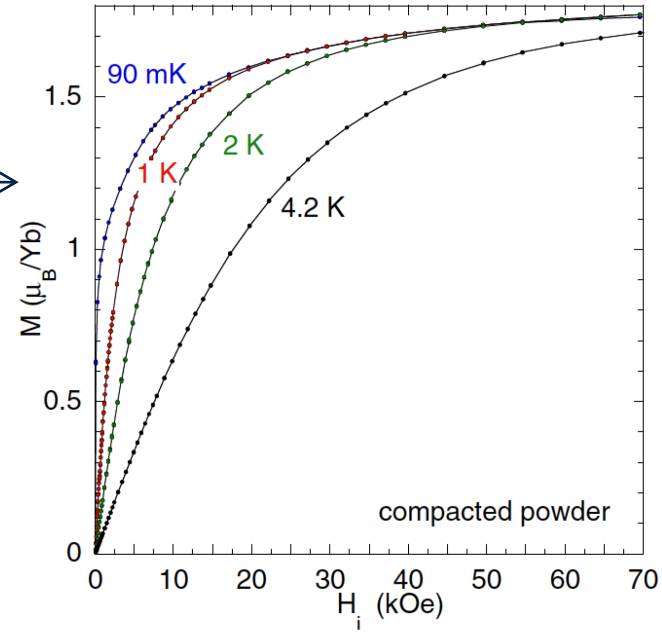
At low *T*, abrupt rise limited by the demagnetisation factor of the samples.




E. Lhotel *et al.*, Phys. Rev. B **89**, 224419 (2014).

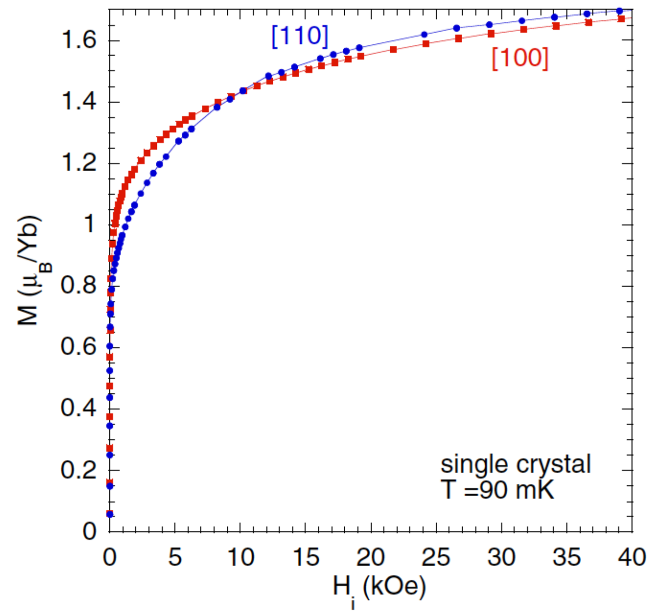
# Magnetisation - *M*(*H*)

Magnetisation saturates at 1.75 $\mu_B$/Yb.



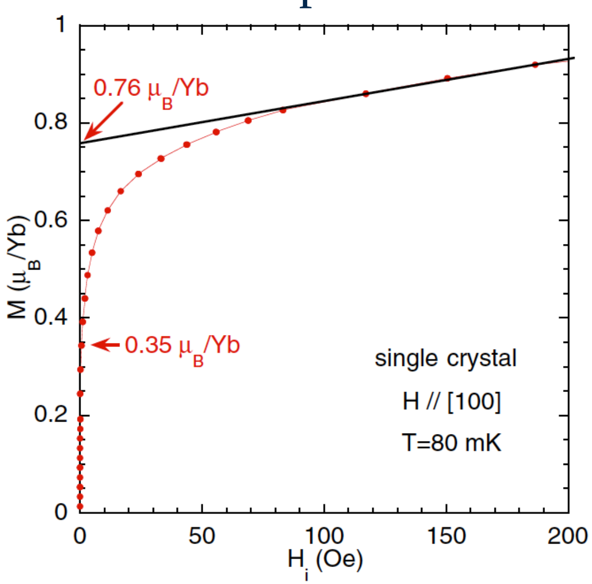

Above 2 K, no clear anisotropy.

Below 1 K, *M*(*H*) curves start to separate.



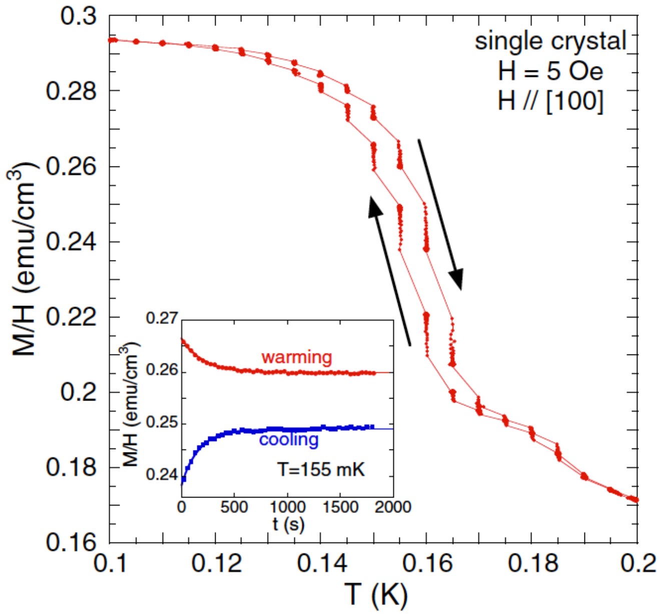

**Ferromagnetic Order**

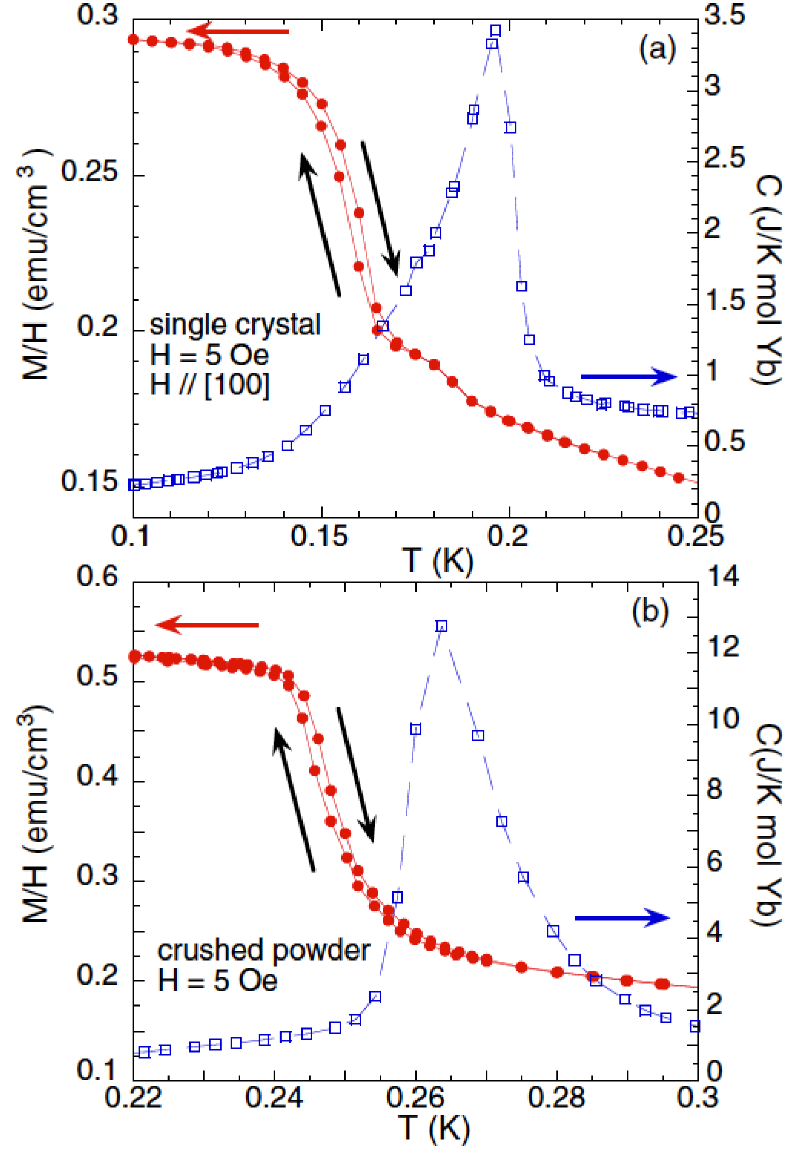
Ordered moment (along [100]) estimated from extrapolation to $H \to 0$.




E. Lhotel *et al.*, Phys. Rev. B **89**, 224419 (2014).

# Magnetisation - first order transition




Strong relaxation.

Equilibrium is only reached after ~600 seconds.

Small hysteresis indicates 1st-order transition.

Increase in $M(T)$ appears below maximum in $C(T)$.

# Magnetisation - first order transition

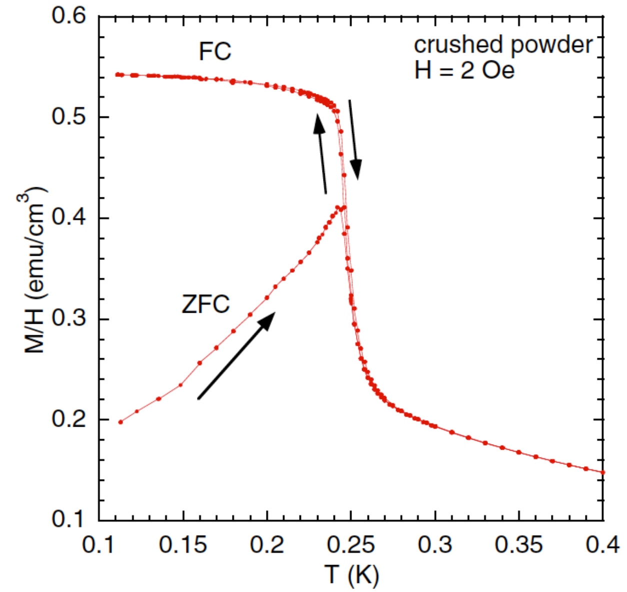
Zero-field cooled, field-cooled magnetisation show irreversibility.

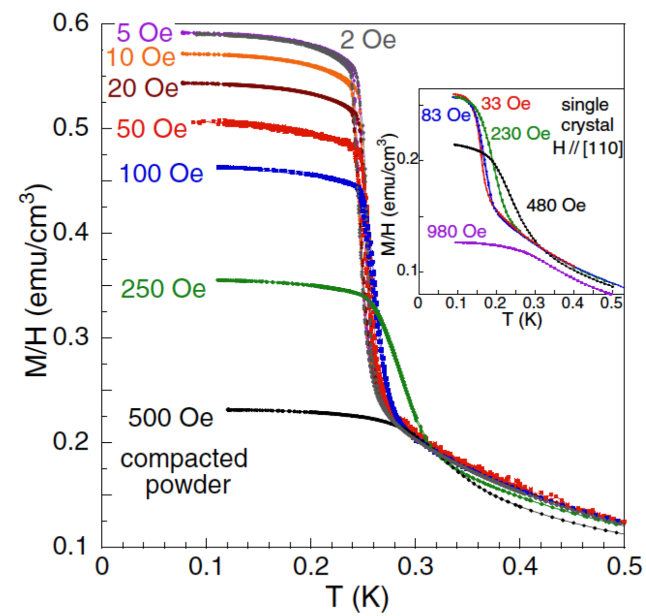
Domains in FM ordered state.

ZFC-FC suppressed and transition broadened and shifted to higher $T$ in fields up to 500 Oe.

Hysteresis in $M(H)$ for powder;
$H_c \approx 10$ Oe @ 80 mK.
$H_c \approx 5$ Oe @ 200 mK.

Results qualitatively similar in single crystal.




E. Lhotel *et al.*, Phys. Rev. B **89**, 224419 (2014).

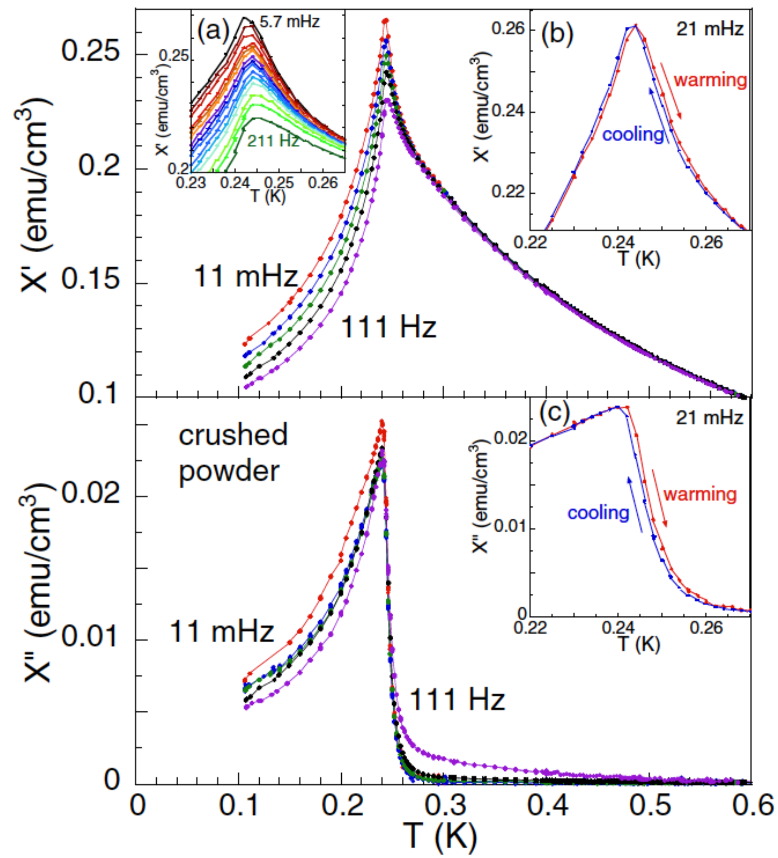
# AC susceptibility

$\chi'(T)$ sharp peak at transition (243 mK).

Peak in $\chi'(T)$ coincides with onset of signal in $\chi''(T)$.

No frequency dependence.

Small $T$ hysteresis in $\chi'$ and $\chi''$.




E. Lhotel *et al.*, Phys. Rev. B **89**, 224419 (2014).

# Contents

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  - *polarized neutron studies*
  - *MuSR*
  - *magnetization*
- **Sample dependent**
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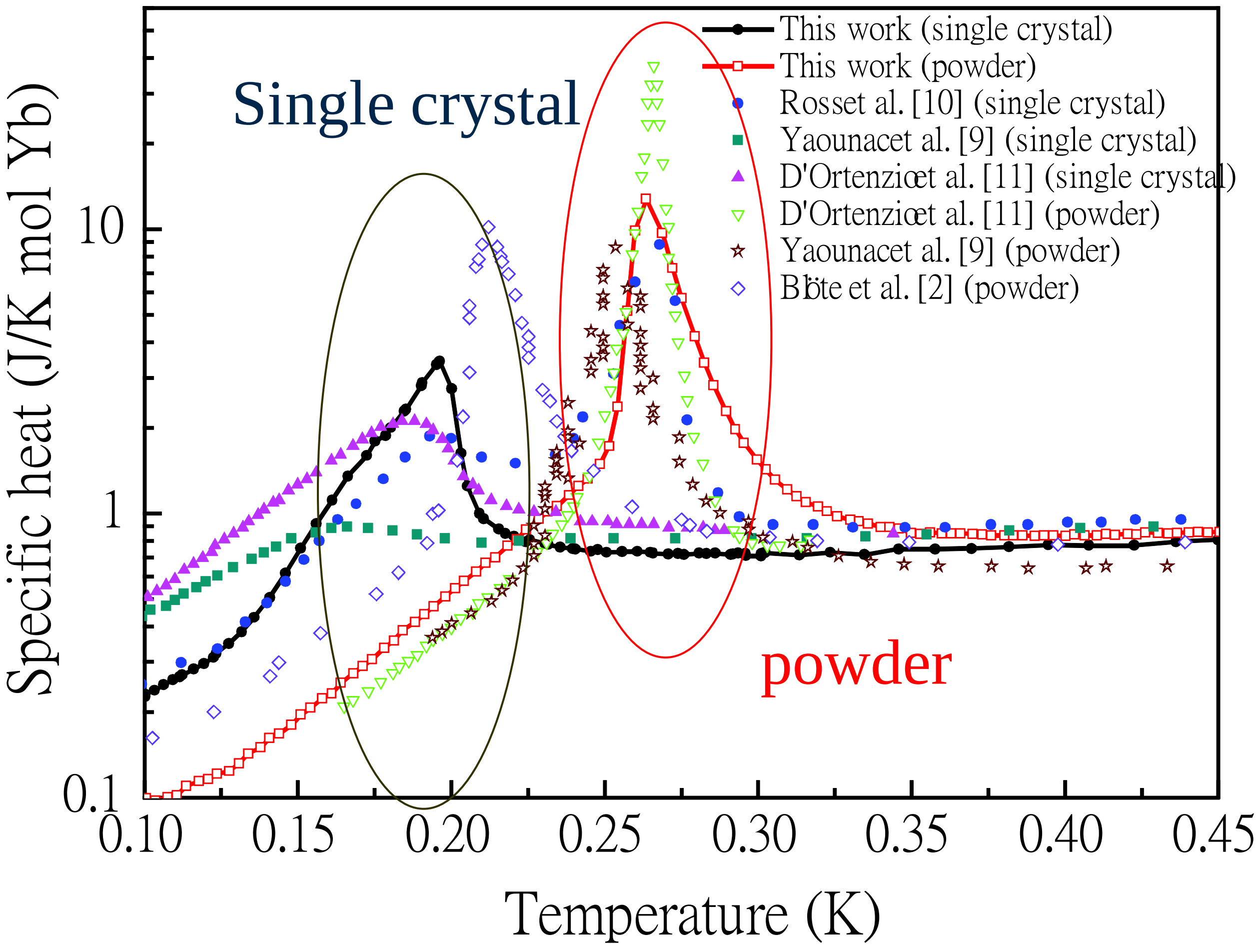
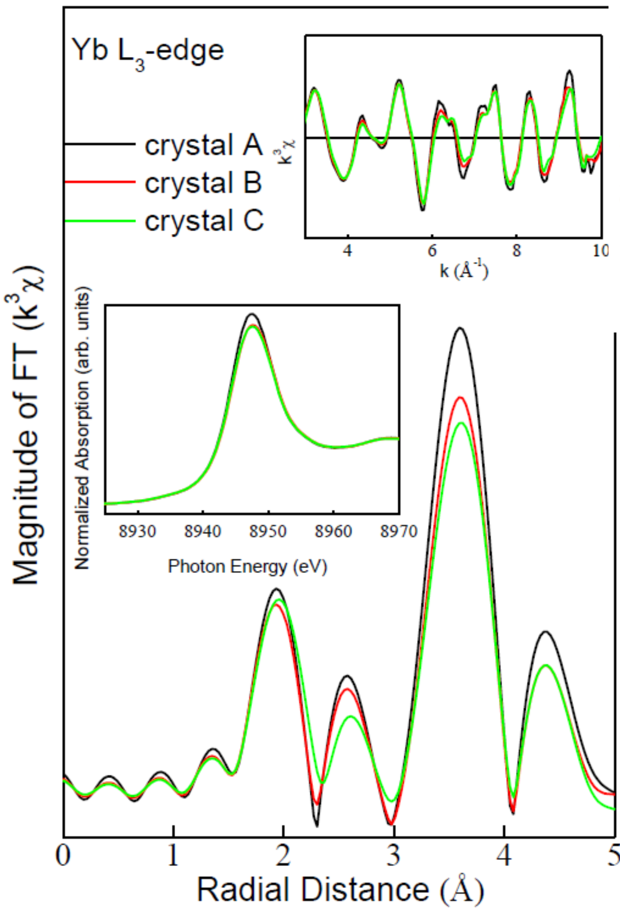
# Sample dependent

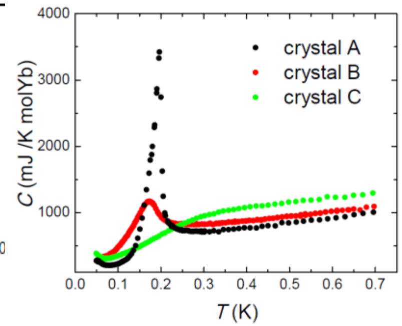


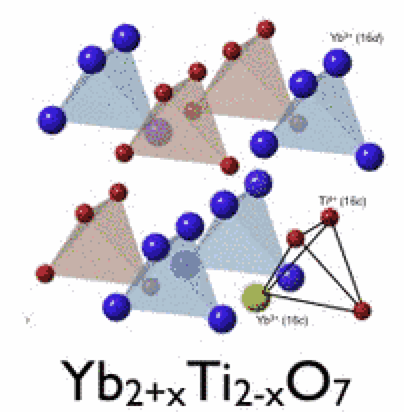

L. J. Chang *et al.*, PRB **89**, 184416 (2014)

# Sample dependent

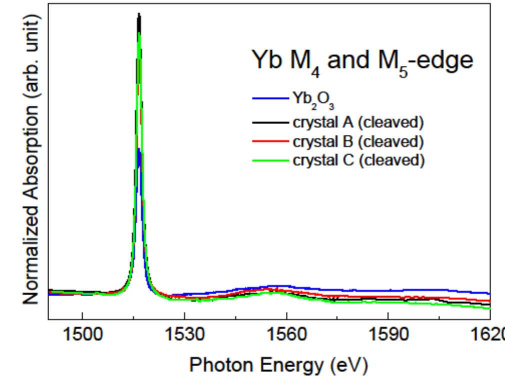
## EXAFS

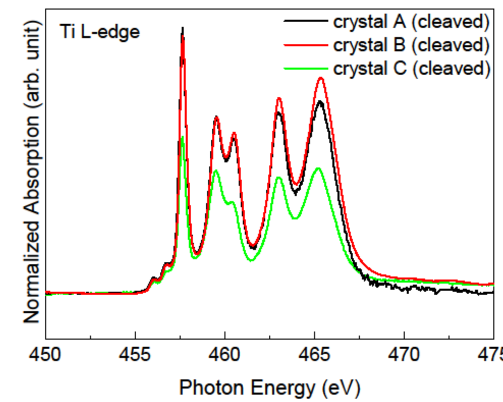


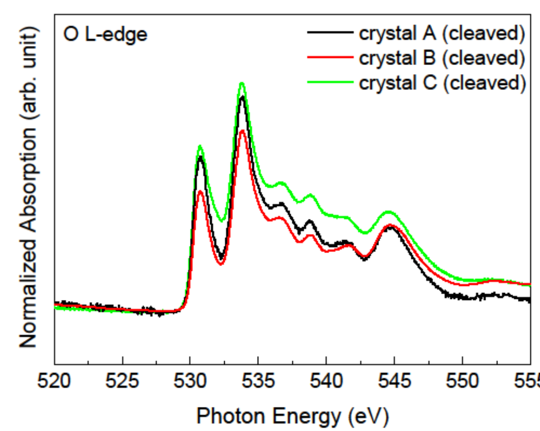




L. J. Chang/ S. Onoda *et al.*, Nat. Commun 3, 992 (2012).




K.A. Ross *et al.*, proposed "stuffed" $Yb_2(Ti_{2-x}Yb_x)O_{7-x/2}$ with x = 0.046
PRB 86, 174424 (2012)

No charge state variations

# Summary

- $Yb_2Ti_2O_7$, a quantum spin ice, first CEF ~ 620 K

- T > ~0.24 K: Rod-structure, quantum spin ice (Pinch points (magnetic Coulomb phase))

- T < ~0.24 K:

  Confirmed ferromagnetic order in crystals and powder:

  Neutron scattering, MuSR, DC magnetization (*L. J. Chang et al.*, *E. Lhotel et al.*)