SYNEMAG 2012

October 17-19th, 2012, Grenoble

Field-induced magnetic behavior in quasi-1D Ising-like antiferromagnet $BaCo_2V_2O_8$: a detailed single crystal neutron diffraction study

Béatrice GRENIER

UJF & CEA, INAC/SPSMS/MDN
Grenoble, France

grenier@ill.fr

Outline

- Introduction on Ising-like quasi-1D systems

- The $BaCo_2V_2O_8$ compound
  - crystallographic structure
  - previous results

- Single-crystal neutron diffraction results *
  - experimental
  - magnetic structure in the Néel phase
  - Néel-LSDW phase transition and (H, T) phase diagram
  - magnetic structure in the LSDW phase

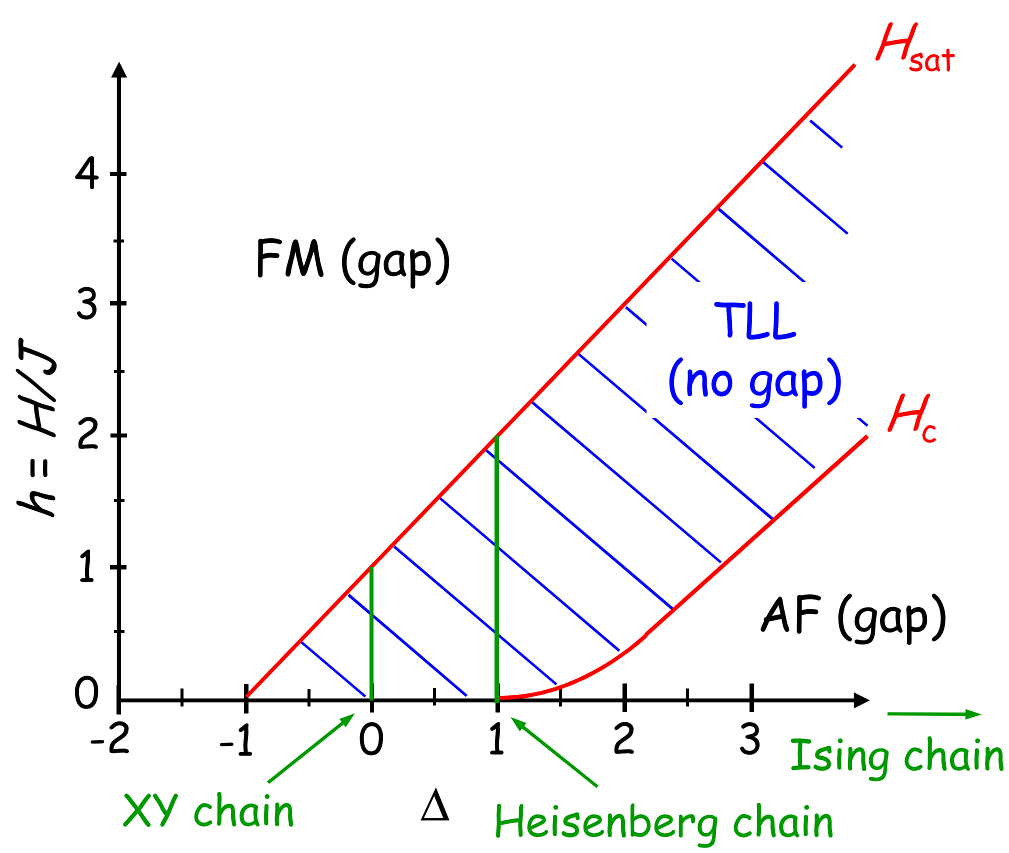
- Conclusion

* E. Canévet, B. Grenier, M. Klanjšek, C. Berthier, M. Horvatić, V. Simonet, and P. Lejay, *arXiv: 1210.3253 (2012); submitted to Phys. Rev. B*

Introduction on Ising-like quasi-1D systems

Hamiltonian of a $S = \frac{1}{2}$ 1D XXZ AF chain system in a magnetic field:

$$\mathcal{H} = J \sum_i \{\Delta S_i^z S_{i+1}^z + (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y)\} - g\mu_B \sum_i S_i^z H$$



Spin-spin correlation functions of the Tomonaga-Luttinger liquid (TLL):

Staggered transverse correlation:

$$\langle S_0^x S_r^x \rangle \simeq (-1)^r \, |r|^{-\eta}$$

Incommensurate longitudinal correlation:

$$\langle S_0^z S_r^z \rangle - M_z^2 \simeq \cos(2k_F r) \, |r|^{-1/\eta}$$

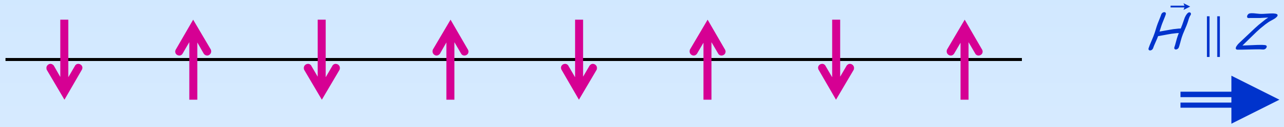
with $k_F = \pi(1/2 - M_z)$

Introduction on Ising-like quasi-1D systems

XXZ chain with weak 3D interaction

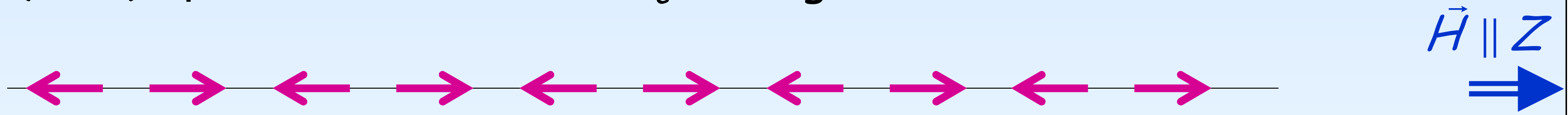
- Heisenberg chain ($\Delta = 1$): $\eta < 1$ as soon as $H \neq 0$ $\rightarrow$ transverse fluctuations dominate

transverse staggered (XY Néel)

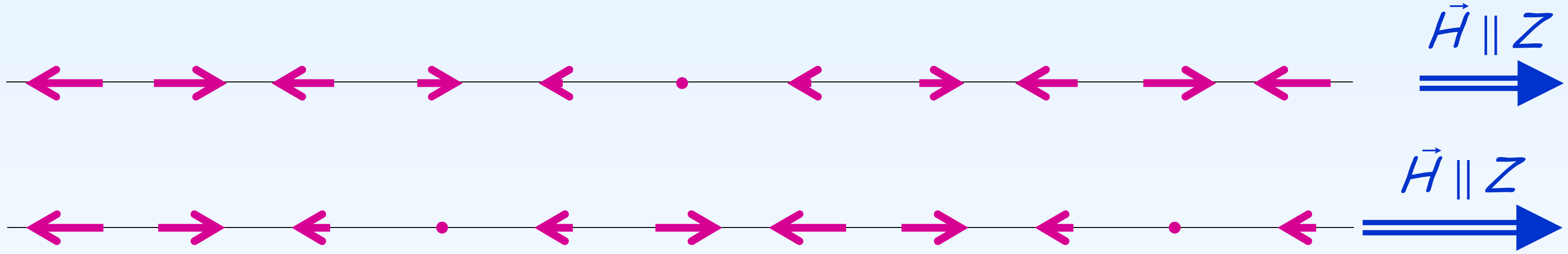


- Ising-like chain ($\Delta > 1$): $\eta > 1$ at low H above H_c $\rightarrow$ longitudinal fluctuations dominate

$H_c > H \geq 0$: gapped Néel

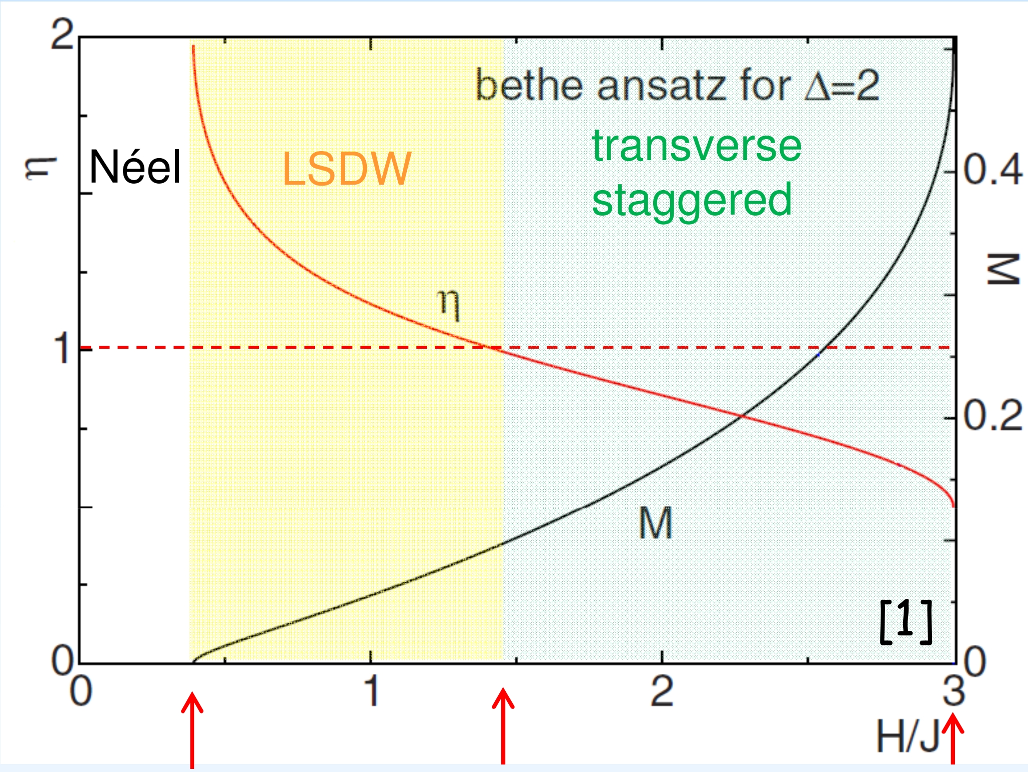


$H^* > H > H_c$: longitudinal spin density wave (LSDW)



$H > H^*$: $\eta < 1$ $\rightarrow$ transverse staggered (XY Néel)

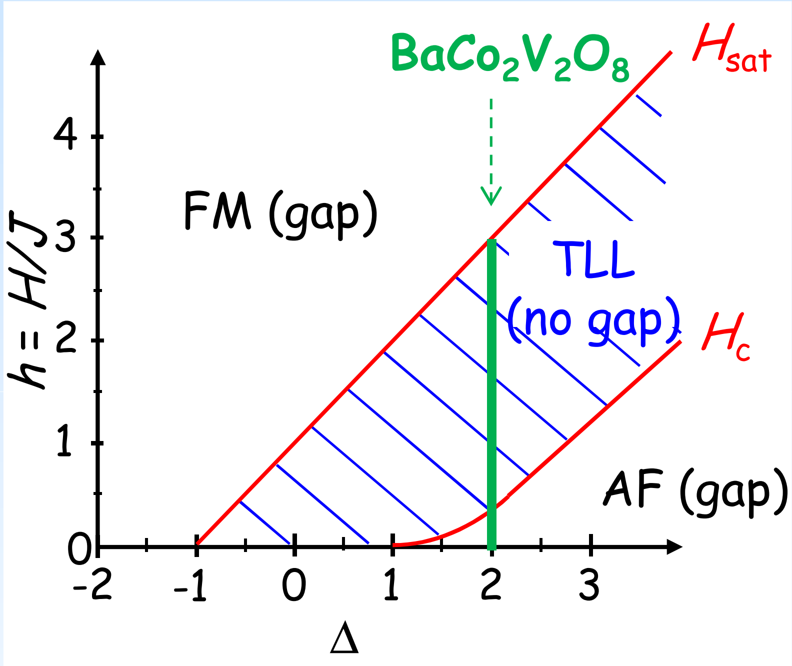
Introduction on Ising-like quasi-1D systems



H_c 3.9 T

H^* ~15 T

H_{sat} 22.7 T



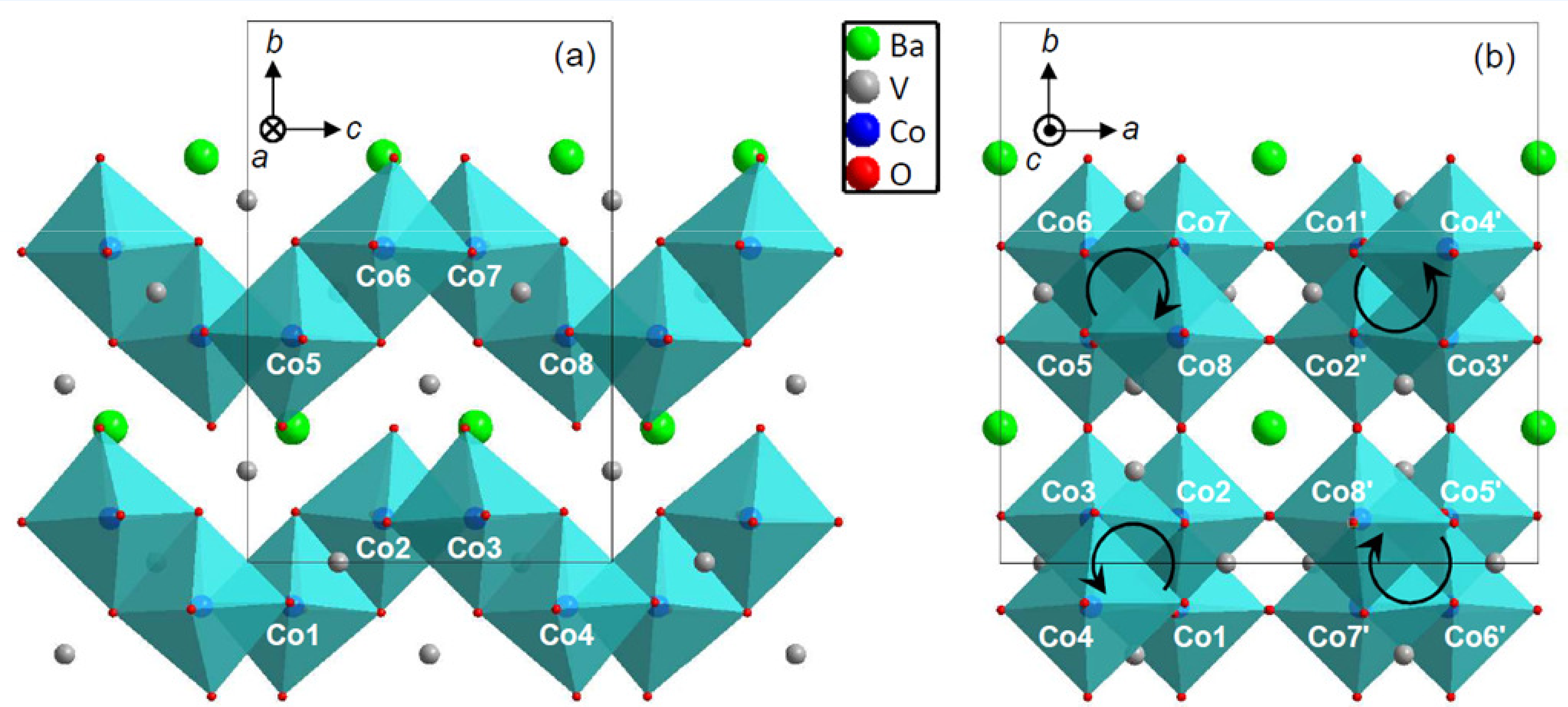
[1] Okunishi and Suzuki, PRB **76**, 224411 (2007)

The $BaCo_2V_2O_8$ compound: *crystallographic structure*

$I4_1/acd$ space group with $a = 12.444$ Å and $c = 8.415$ Å at $T = 300$ K

R. Wichmann and Hk. Müller-Buschbaum, Z. Anorg. Allg. Chem. **532**, 153 (1986)

4 screw chains ||c-axis per unit cell → 16 Co^{2+} with spin 3/2 (effective spin $S = \frac{1}{2}$)



The $BaCo_2V_2O_8$ compound: *previous results*

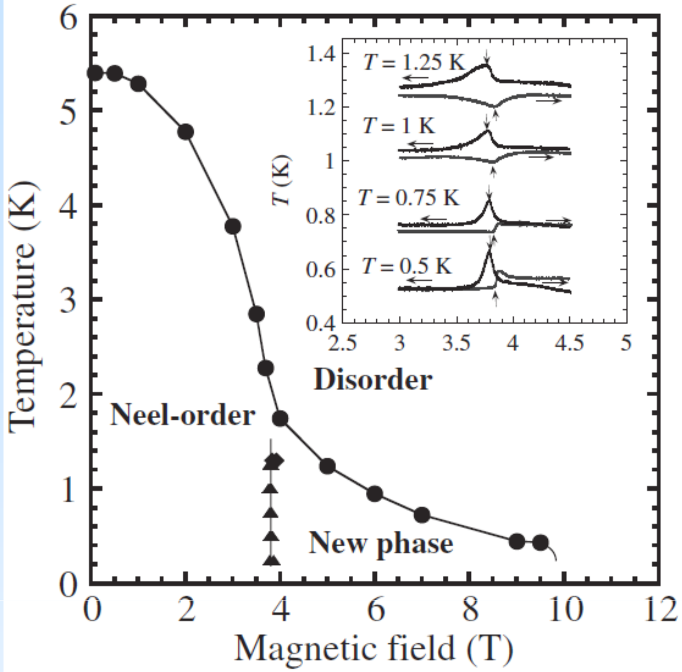
- "Discovery" of $BaCo_2V_2O_8$:
Crystal growth + Magnetic measurements
He *et al.*, Chem. Mater. **17**, 2924 (2005)
Kimura *et al.*, J. Phys.: Conf. Series **51**, 99–102 (2006)

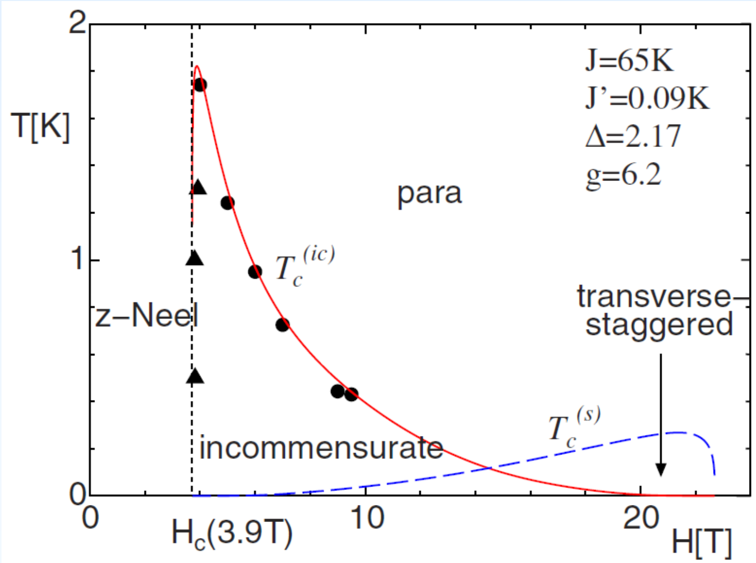
- Theoretical predictions applied to $BaCo_2V_2O_8$
Okunishi and Suzuki, PRB **76**, 224411 (2007)

- *H* - *T* diagram from specific heat
Kimura *et al.*, PRL **100**, 057202 (2008)

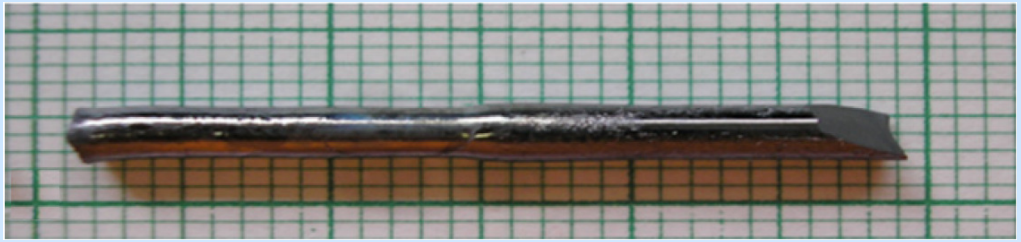
- First observation of IC satellites up to *H* = 5 T by single-crystal neutron diffraction
Kimura *et al.*, PRL **101**, 207201 (2008)

- Determination of the magnetic structure at *H* = 0 by powder neutron diffraction
Kawasaki *et al.*, PRB **83**, 064421 (2011)





Results: *Experimental*



- Crystal synthesis @ Institut Néel

- Specific heat measurements @ CEA

- Magnetic structure in zero field
(Néel phase) on **D15 & D23** @ ILL

- Magnetic structure in the LSDW phase
& incommensurability vs field on **D23** @ ILL
H || easy axis (chain *c*-axis)
6 T / 12 T + dilution
Vertical collimation 20'

Results: *Magnetic structure in the Néel phase*

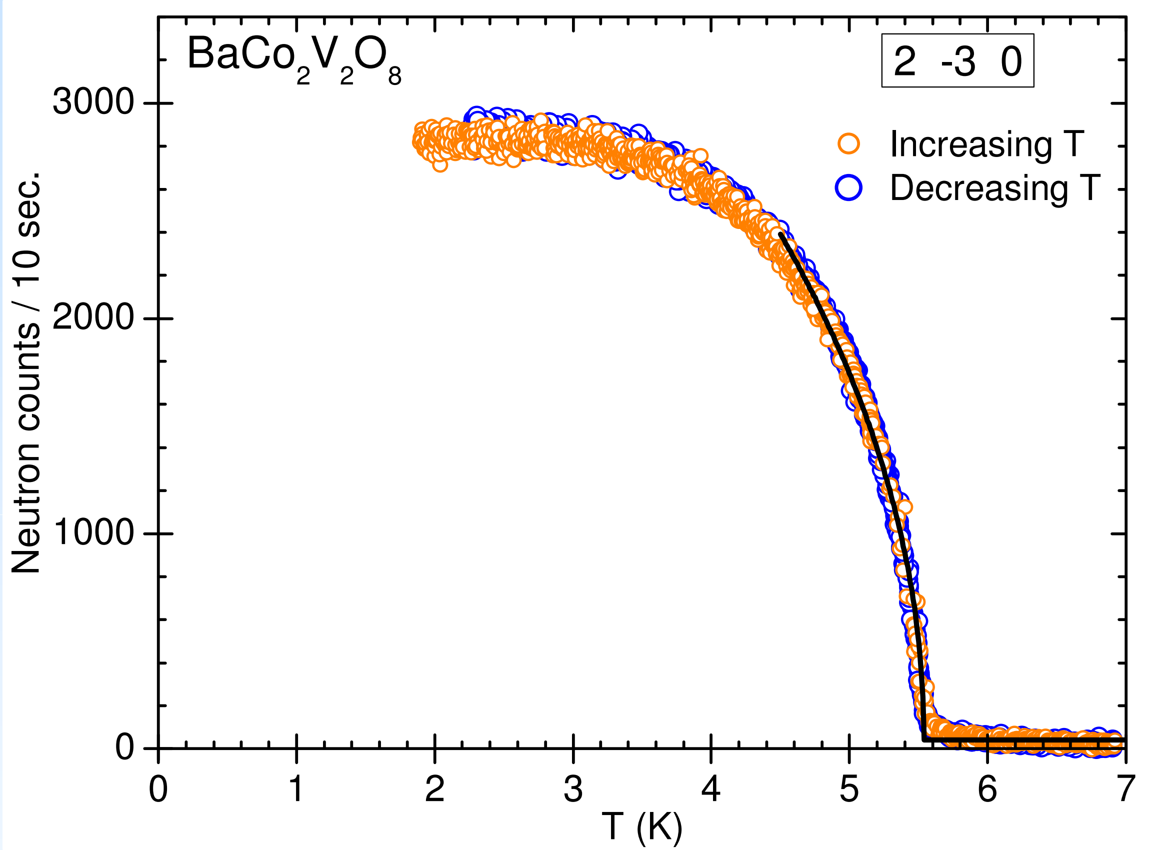
Propagation vector:

$\vec{k}_{AF} = (1,0,0)$

$\vec{Q}_{AF} = \vec{H} + \vec{k}_{AF}$

where $\vec{H} = (h,k,l)$

with $h + k + l = 2n$



Critical exponent $\beta = 0.32 \Rightarrow$ 3D Ising universality class

Results: *Magnetic structure in the Néel phase*

Nuclear structure:

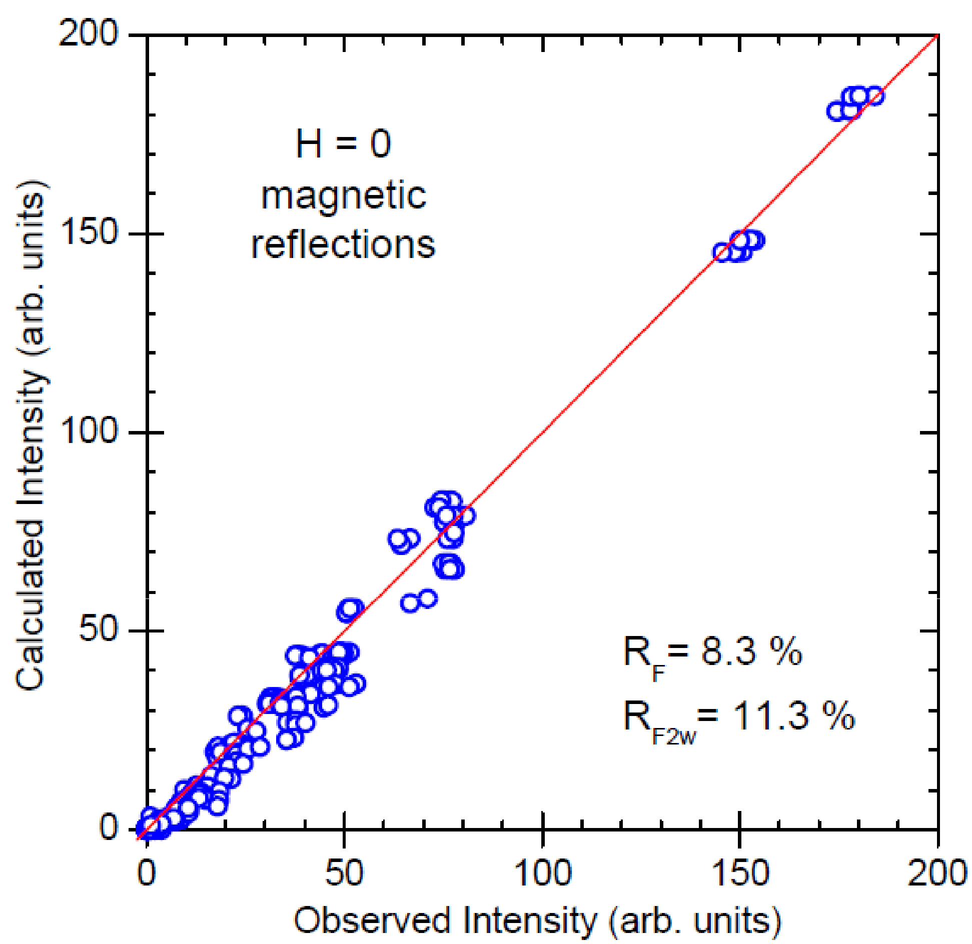
Same structure below T_N as above

Magnetic structure:

313 reflections collected (113 independent)
up to $\sin\theta/\lambda$ = 0.55

Refinement with Fullprof

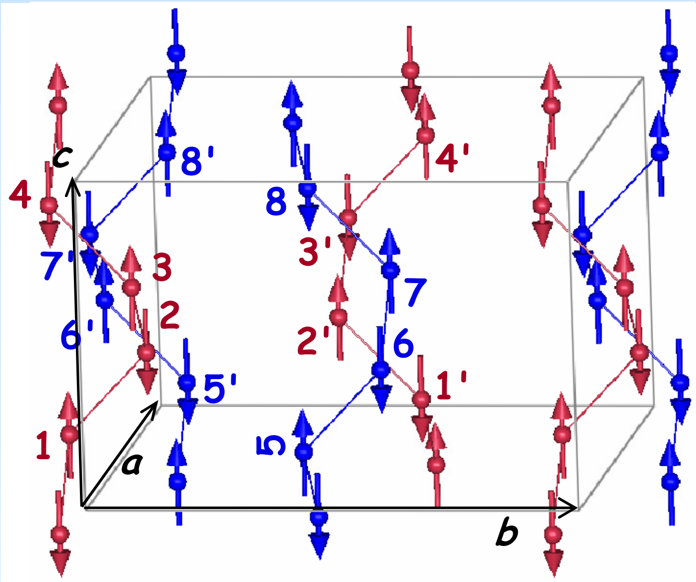
→ one of the four magnetic structures predicted by group theory with two magnetic domains of populations 49.5(4)% and 50.5(4)%

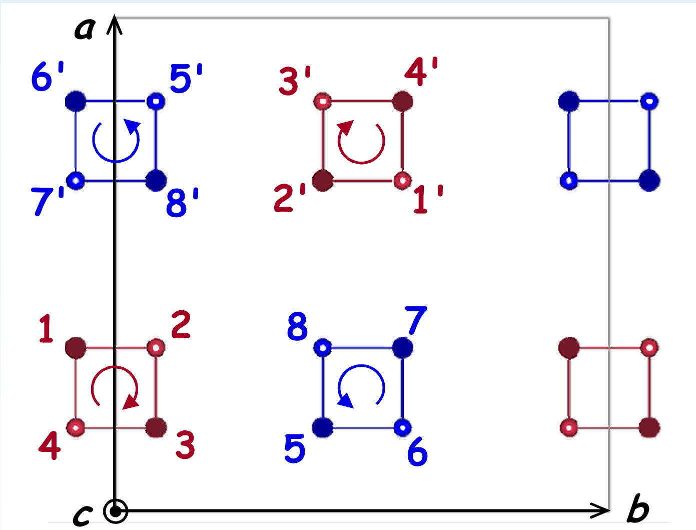


h	k	l	$\sin\theta/\lambda$	I_{obs}	I^{tot}_{calc}	I^{1}_{calc}	I^{2}_{calc}
1	0	0	0.0403	0.00(0.23)	0.00	0.00	0.00
1	1	1	0.0823	24.39(0.37)	28.70	14.20	14.50
$\bar{2}$	$\bar{3}$	0	0.1452	178.43(1.57)	181.15	181.15	0.00
$\bar{3}$	$\bar{2}$	0	0.1452	184.13(1.24)	184.73	0.00	184.73
$\bar{2}$	$\bar{4}$	1	0.1896	32.27(0.64)	33.00	19.87	13.13
$\bar{4}$	$\bar{2}$	1	0.1896	32.72(0.64)	33.15	12.87	20.28

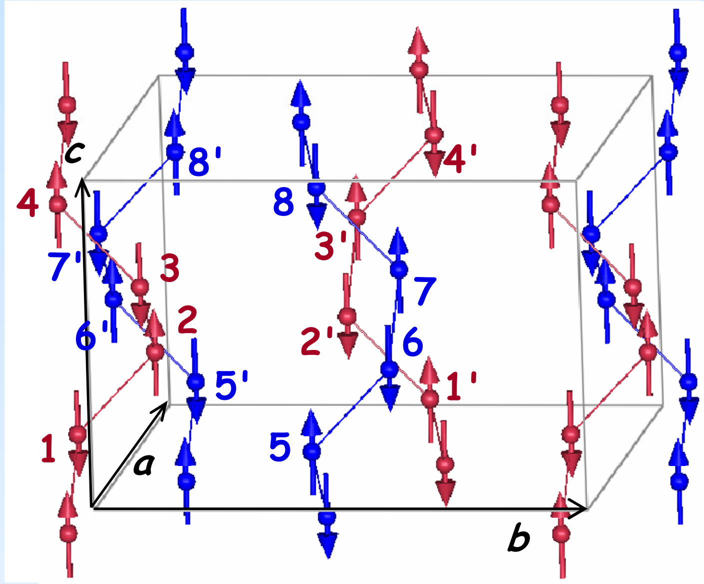
Results: *Magnetic structure in the Néel phase*

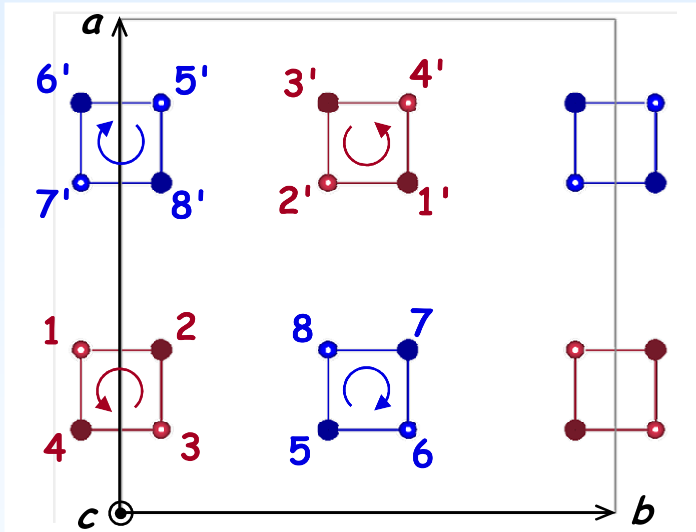
Magnetic domain #1





Magnetic domain #2





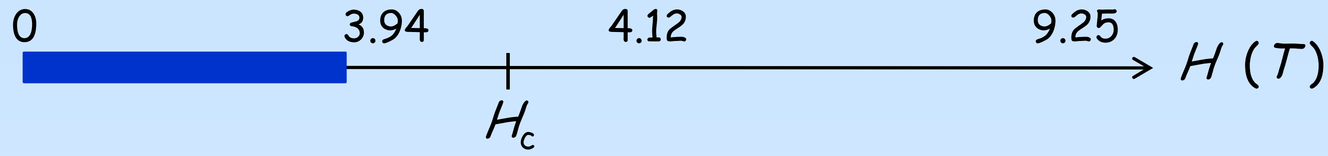
Screw axis 4_1

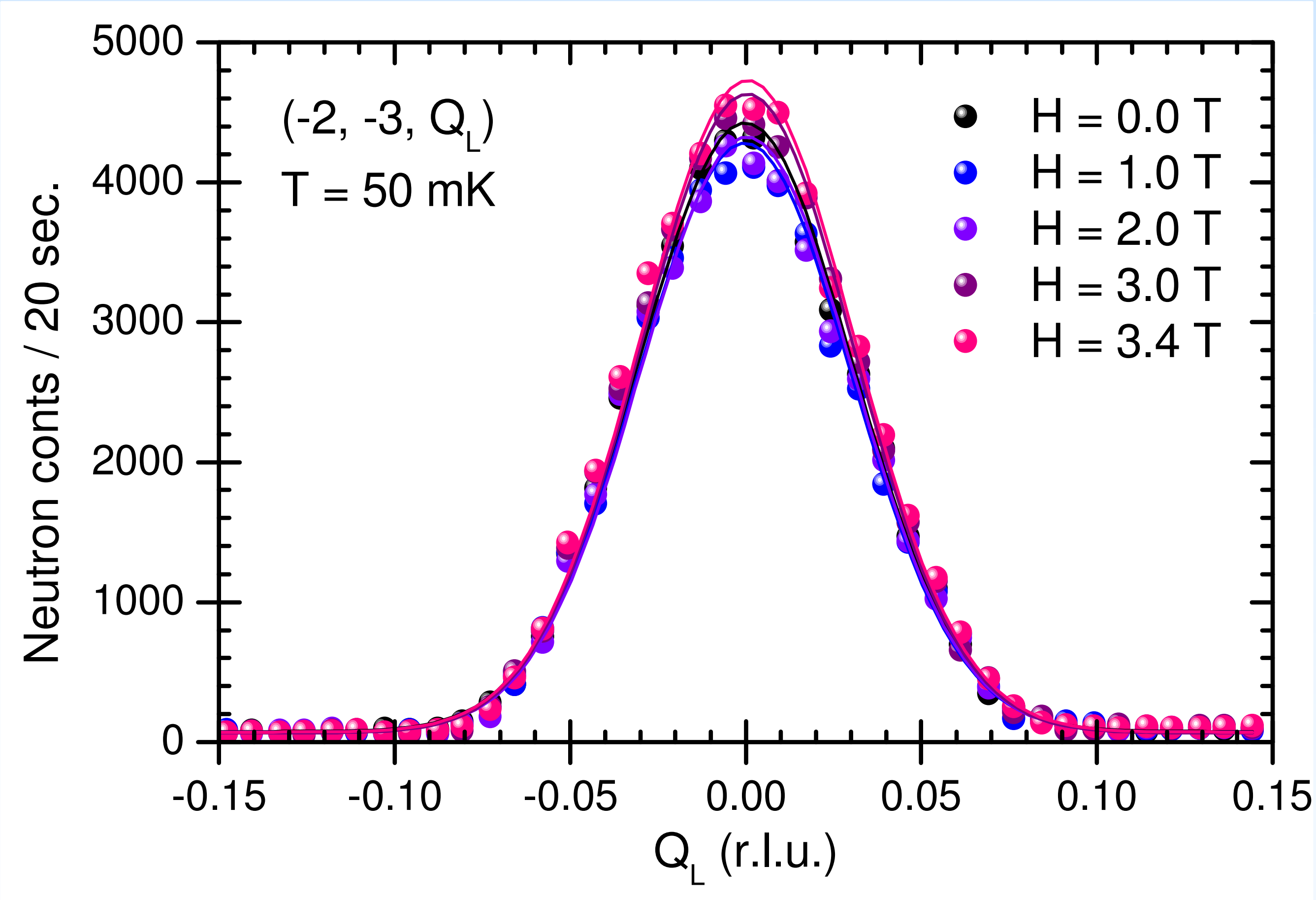
⇔

$m_x = m_y = 0$

$m_z = 2.267(3)\mu_B / Co^{2+}$

Results: *Néel-LSDW phase transition*



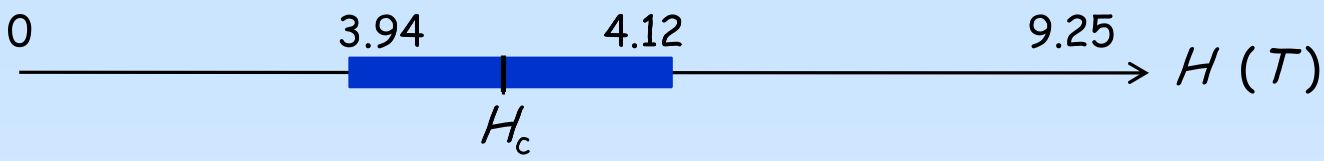


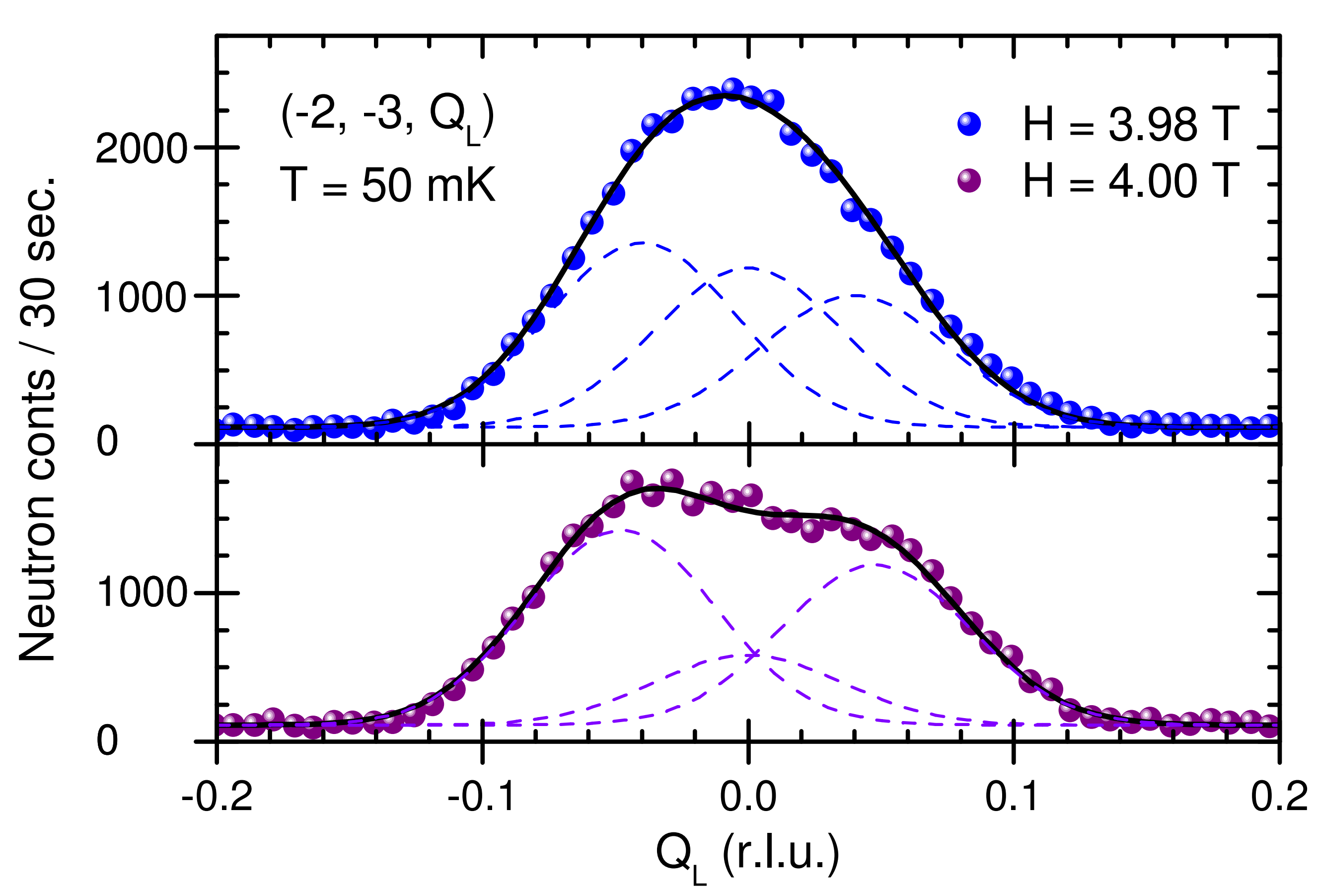
$0 \leq H \leq 3.92$ T:

Néel phase

$\vec{k}_{AF} = (1,0,0)$

Results: *Néel-LSDW phase transition*





$3.94 \leq H \leq 4.12$ T:

Coexistence between Néel & LSDW phases

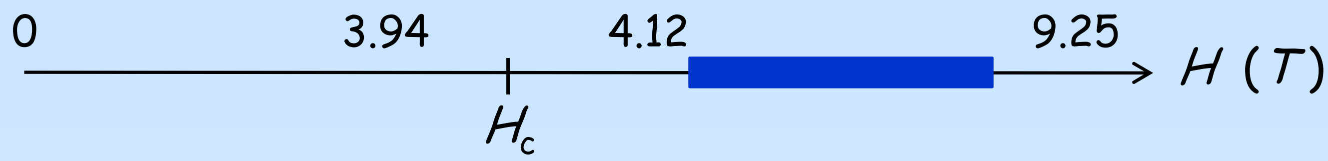
$\vec{k}_{AF} = (1,0,0)$

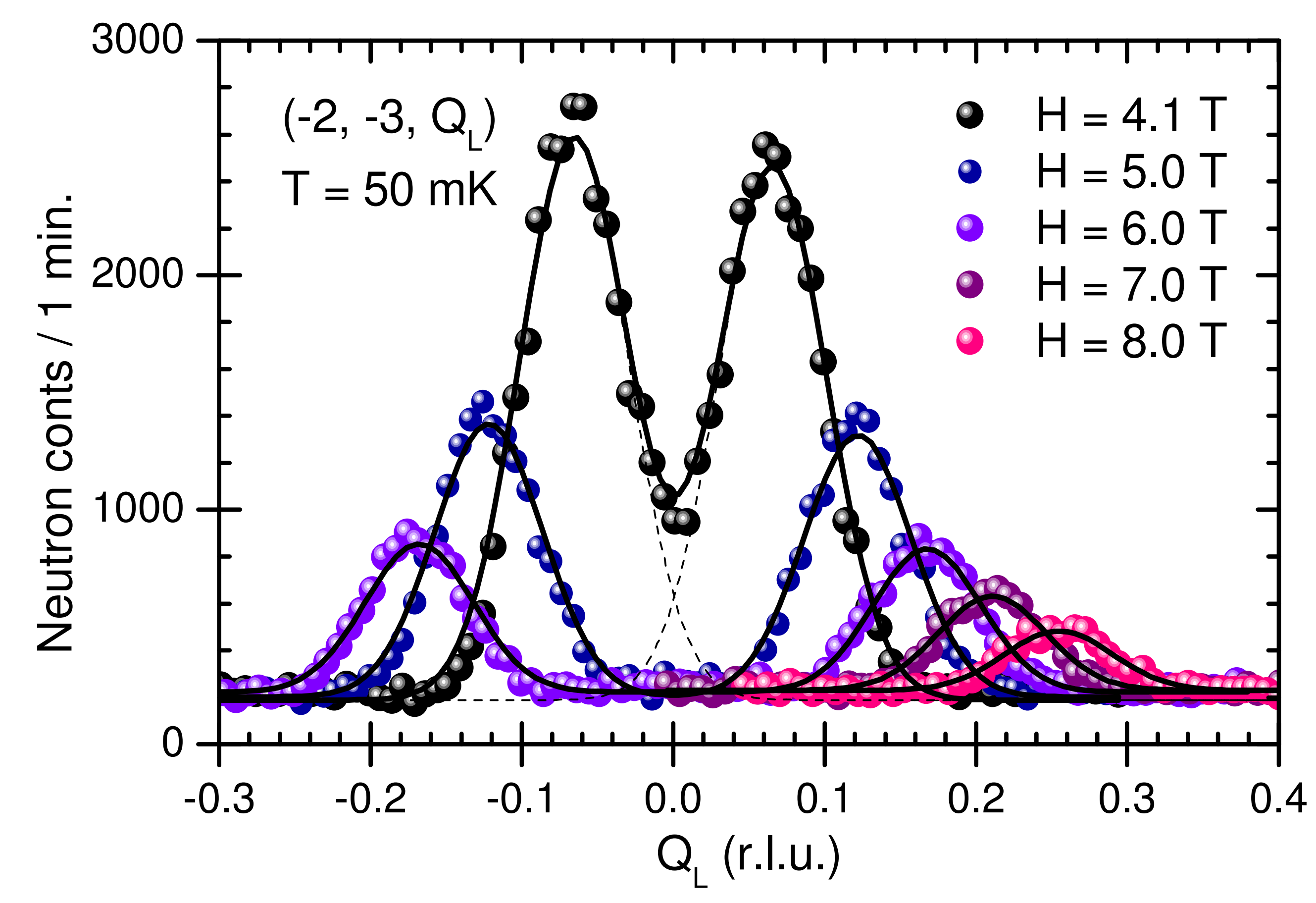
$\vec{k}_{LSDW} = (1,0,\delta)$

$0.03 < \delta < 0.07$

$\vec{Q}_{LSDW} = \vec{H} \pm \vec{k}_{LSDW}$

Results: *Néel-LSDW phase transition*





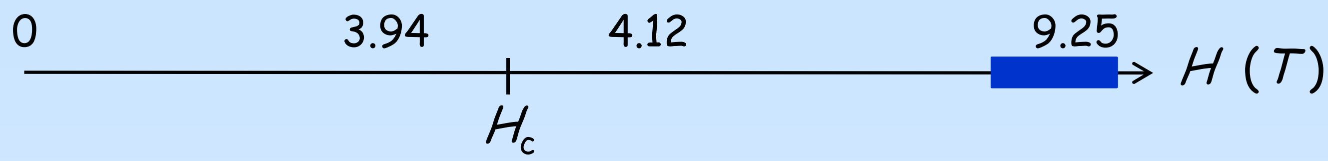
$4.14 \leq H \leq 9.25$ T:

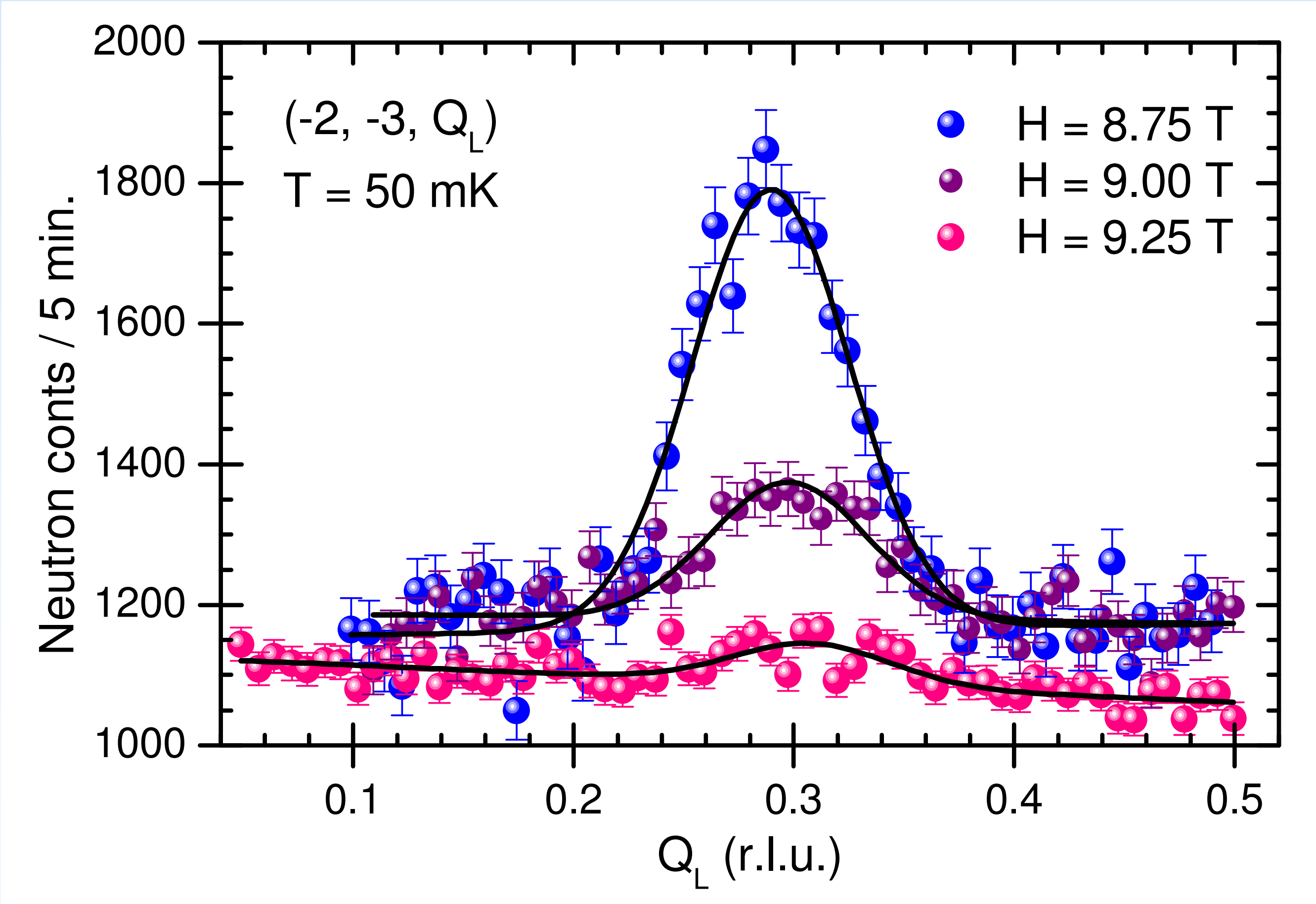
LSDW phase

$\vec{k}_{LSDW} = (1, 0, \delta)$

$0.07 \leq \delta \leq 0.31$

Results: *Néel-LSDW phase transition*



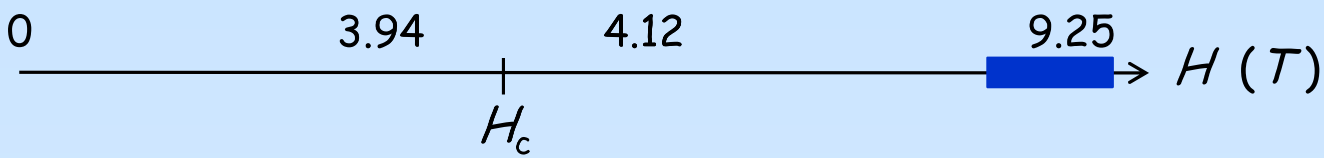


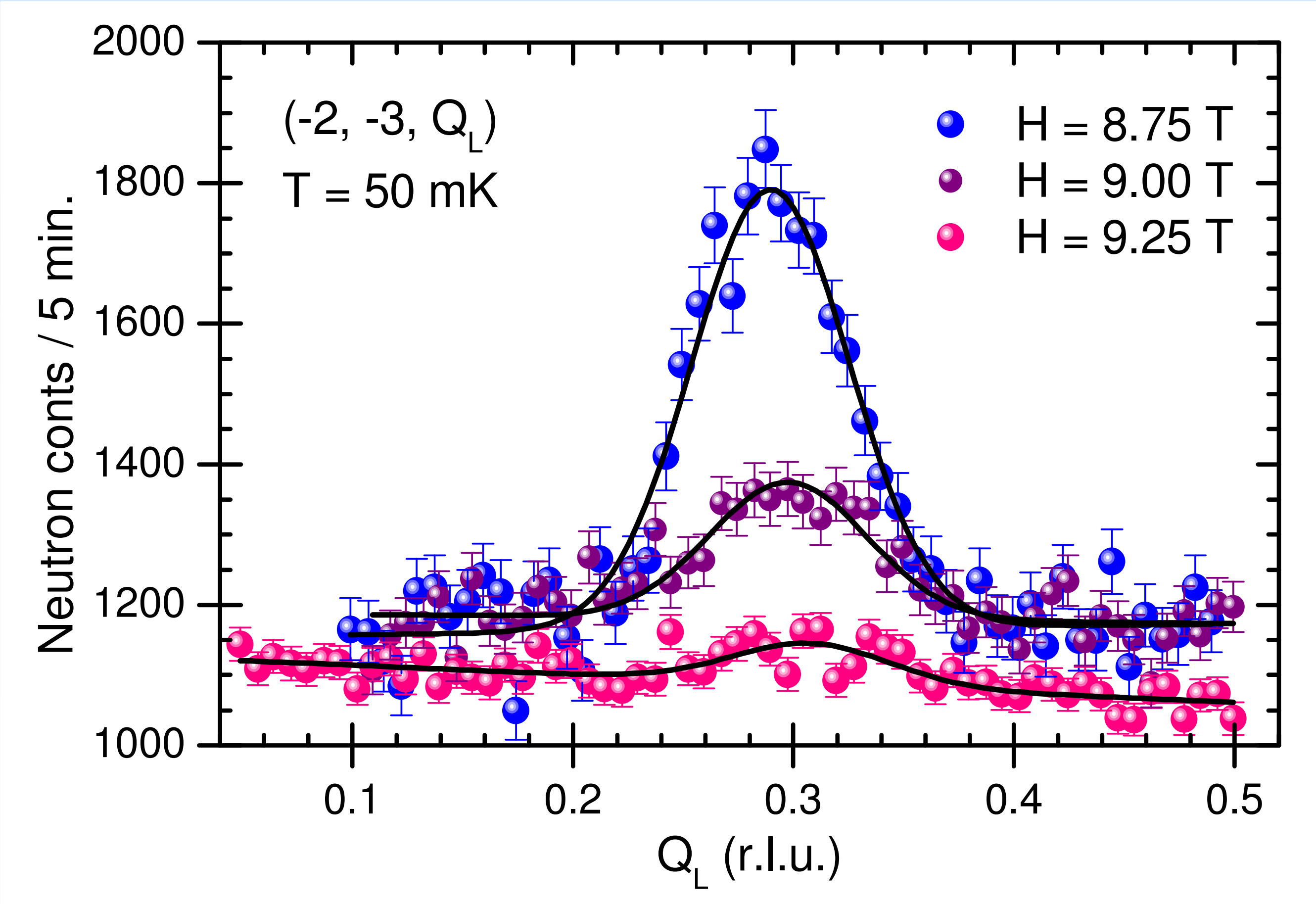
$4.14 \leq H \leq 9.25$ T:

LSDW phase

$$\vec{k}_{LSDW} = (1, 0, \delta)$$
$$0.07 \leq \delta \leq 0.31$$

Results: *Néel-LSDW phase transition*





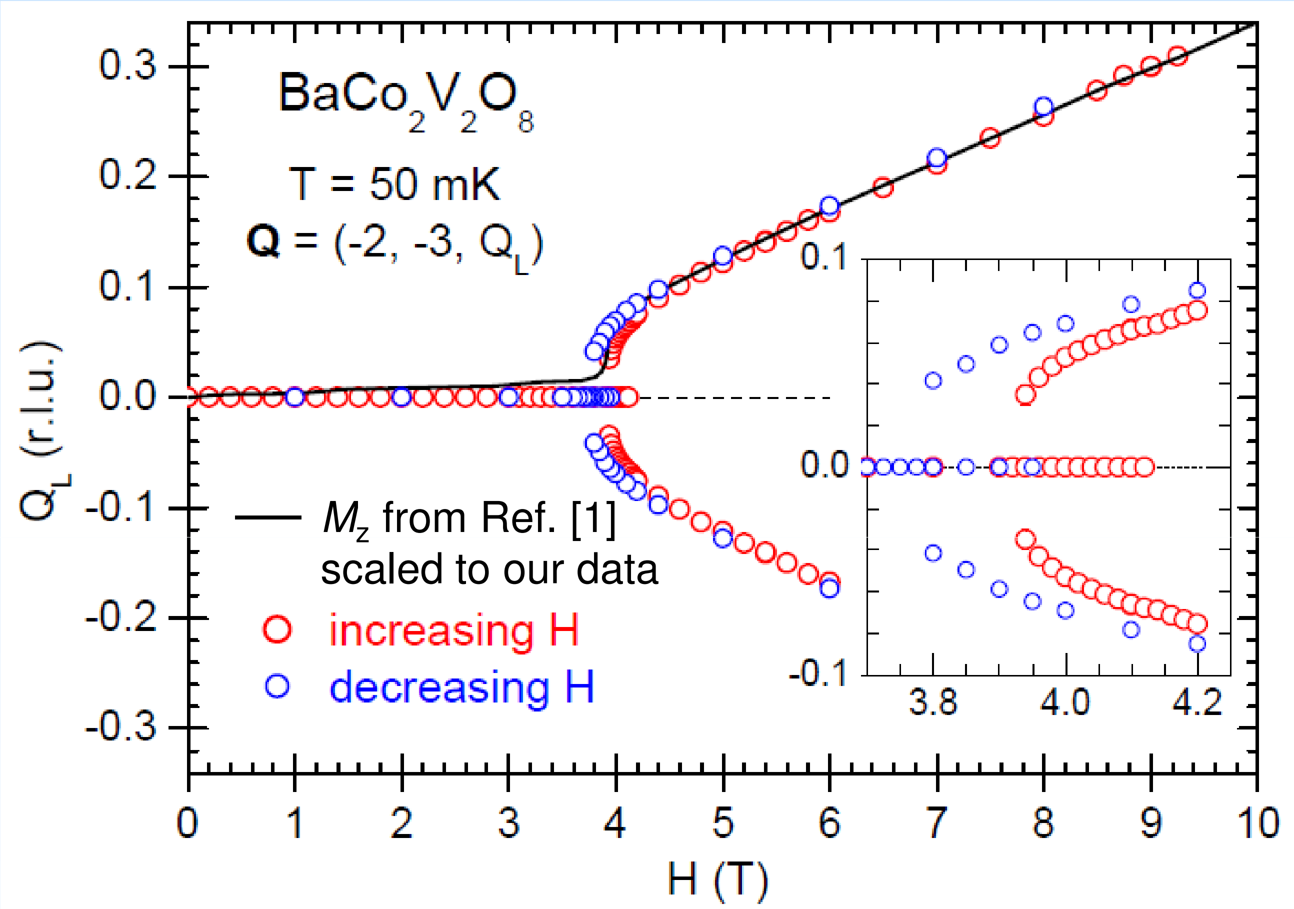
$H \geq 9.5$ T:

$I = 0$

from NMR [1]: new phase

[1] M. Klanjšek *et al.*, arXiv: 1202.6376 (2012)

Results: *Néel-LSDW phase transition*



- Field hysteresis ~ 0.2 T

⇓

1st order transition

- Theory applies: $M_z = 1/2\ g\mu_B S$ [2]

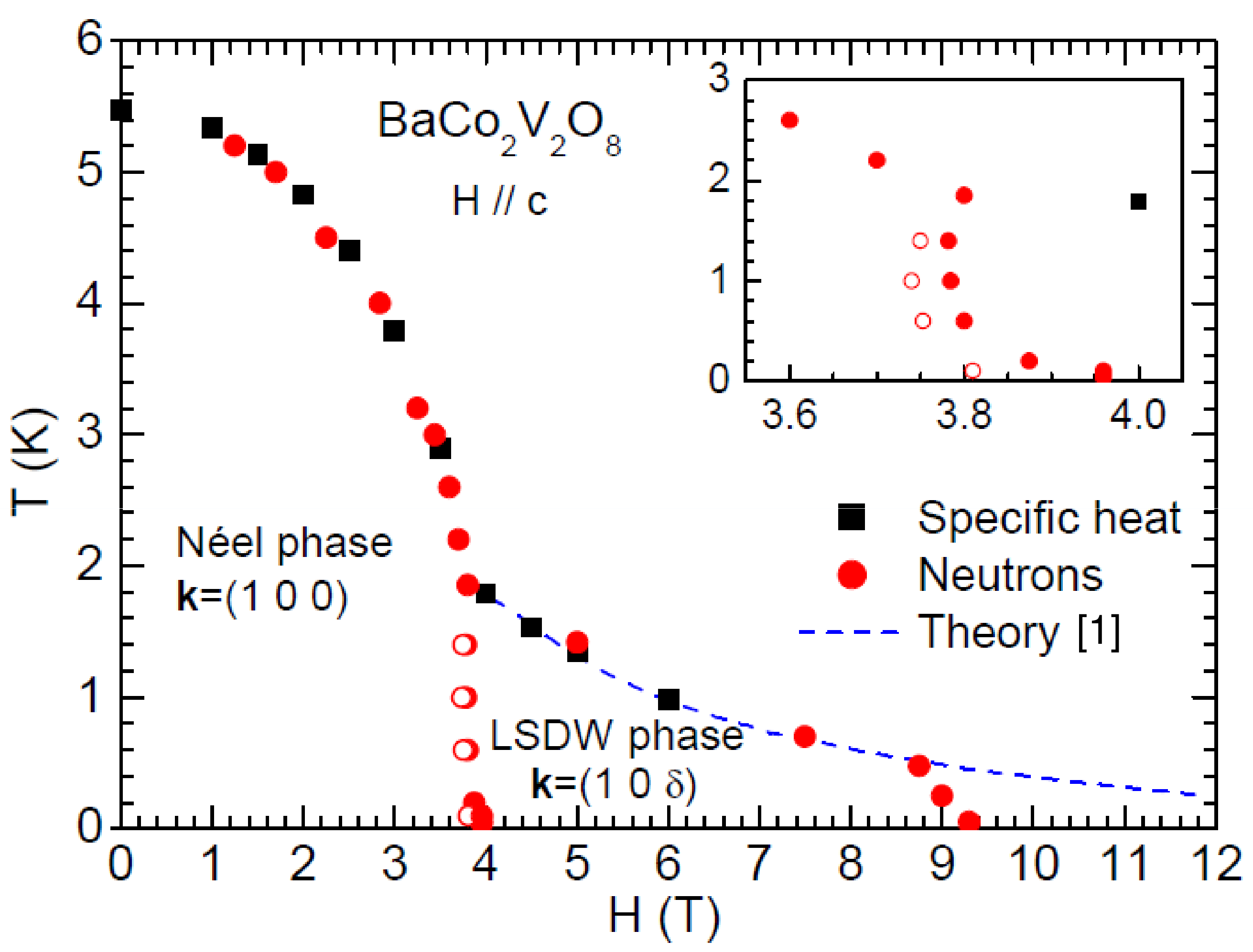
⇓

$g = 7.2$

(to be compared to $g = 6.2$ [3])

[1] Kimura *et al.*, PRL 100, 057202 (2008)
[2] Haldane, PRL 45, 1358 (1980)
[3] Kimura *et al.*, J. Phys.: Conf. Ser. **51**, 9910 (2006)

Results: *H -T phase diagram*



[1] Okunishi and Suzuki, PRB 76, 224411 (2007)

Results: *Magnetic structure in the LSDW phase*

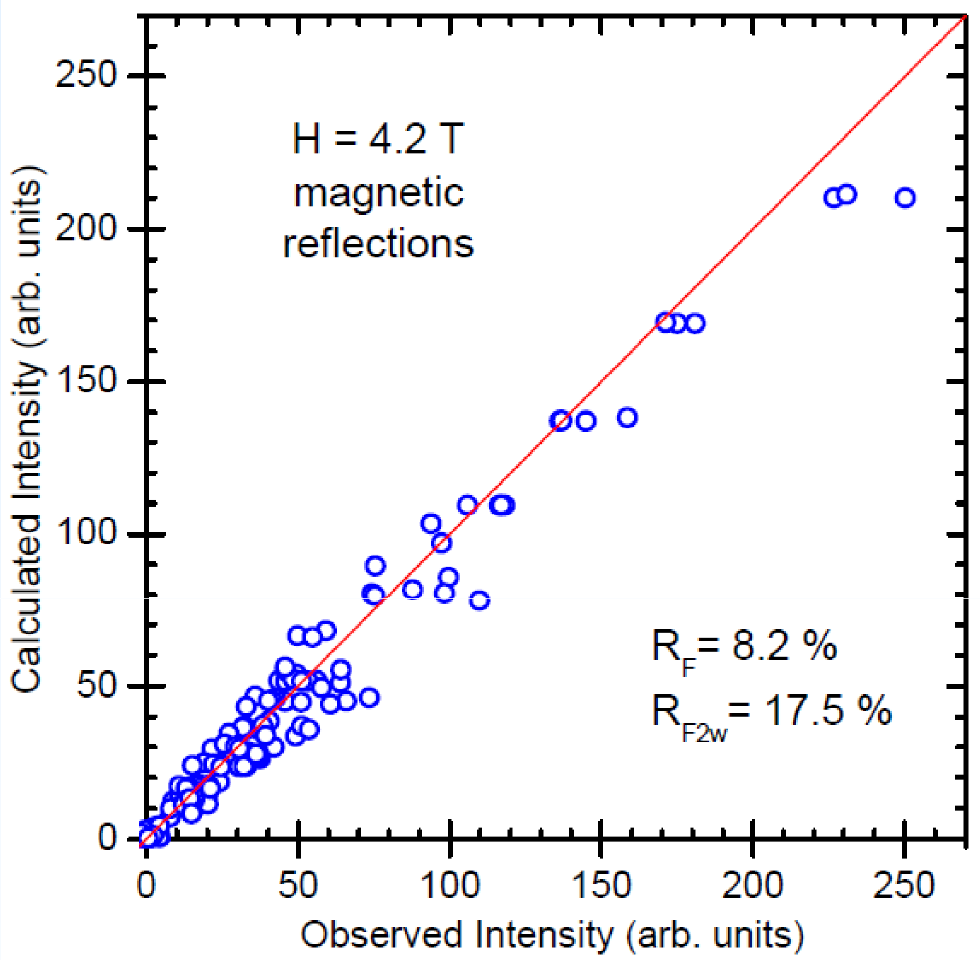
***H* = 4.2 T coming down from 6 T**

Nuclear structure:
Same as in zero magnetic field

Magnetic structure:
129 reflections collected (49 independent)

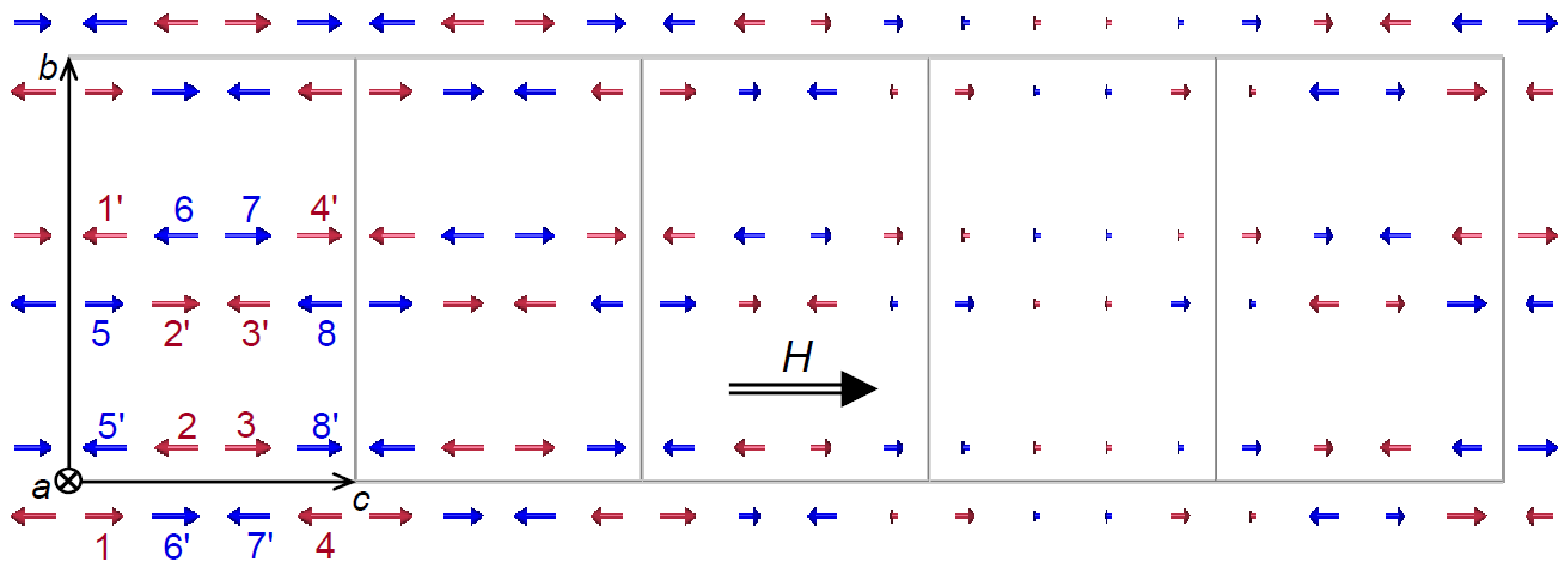
Continuity with that found in zero field

Two magnetic domains of populations
38.3(9)% and 61.7(9)%
⇑
small misalignment of the field
H_a = 0.11 T, H_b = 0.04 T



Results: *Magnetic structure in the LSDW phase*

Magnetic domain #1



Amplitude of the sine wave modulation = 1.398(6)μ_B/Co^{2+}

Conclusion & perspectives

CONCLUSION

- **Magnetic structures**:

- At $H = 0$: confirmation of the structure determined on powder
  opportunity for a visualization of the magnetic domains
- At $H = 4.2$ T: refined for the first time → continuity with zero field
  first direct evidence of the LSDW phase in $BaCo_2V_2O_8$

- **Critical exponent** β: $BaCo_2V_2O_8$ belongs to the 3D Ising universality class

- δ **(*H*)** in perfect agreement with theory assuming $g = 7.2$

- ***H* – *T* phase diagram** in perfect agreement with theory up to ~9 T then not any more
→ new phase seen by NMR [M. Klanjšek *et al.*, arXiv: 1202.6376 (2012)]: ferromagnetic SDW

PERSPECTIVES

- Neutron diffraction: study of this new phase
- Inelastic neutron scattering: study of the spin dynamics

Collaborations:

E. Canévet (ILL & UJF-Grenoble)

M. Klanjšek (Jozef Stefan Institute, Ljubljana, Slovenia)

C. Berthier and M. Horvatić (LNCMI-Grenoble)

V. Simonet and P. Lejay (Institut Néel, Grenoble)

Thank you