(Quasi) 2D Quantum Magnets

when theory (tries to) meet experiments

Nicolas Laflorencie

Laboratoire de Physique Théorique
CNRS
FERMI
Université Paul Sabatier (Toulouse)

(Quasi) 2D Quantum Magnets

when theory (tries to) meet experiments

Nicolas Laflorencie

Laboratoire de Physique Théorique
CNRS
FERMI
Université Paul Sabatier (Toulouse)

- Does dimension really matter ?
- The case of Han purple (quasi-2D)
- Monolayer $CrCl_3$ (2D)
- Conclusions

Does dimension really matter?

Does dimension really matter?

The MWH theorem

VOLUME 17, NUMBER 22 PHYSICAL REVIEW LETTERS 28 NOVEMBER 1966

ABSENCE OF FERROMAGNETISM OR ANTIFERROMAGNETISM IN ONE- OR TWO-DIMENSIONAL ISOTROPIC HEISENBERG MODELS*

N. D. Mermin† and H. Wagner‡
Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York
(Received 17 October 1966)

It is rigorously proved that at any nonzero temperature, a one- or two-dimensional isotropic spin-S Heisenberg model with finite-range exchange interaction can be neither ferromagnetic nor antiferromagnetic. The method of proof is capable of excluding a variety of types of ordering in one and two dimensions.

David Mermin (1935 -)

Herbert Wagner (1935 -)

- Continuous symmetry + Short-Range interaction

No Finite-Temperature Ordering for $\mathrm{D} \leq 2$

- 1D (spin chains, ladders…)
- 2D planes, bilayers, finite number of layers

PHYSICAL REVIEW VOLUME 158, NUMBER 2 10 JUNE 1967

Existence of Long-Range Order in One and Two Dimensions

P. C. HOHENBERG
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received 24 October 1966)

It is pointed out that a rigorous inequality first proved by Bogoliubov may be used to rule out the existence of quasi-averages (or long-range order) in Bose and Fermi systems for one and two dimensions and $T \neq 0$.

Pierre Hoenberg
(1934 - 2017)

Does dimension really matter?

The MWH theorem

VOLUME 17, NUMBER 22 PHYSICAL REVIEW LETTERS 28 NOVEMBER 1966

ABSENCE OF FERROMAGNETISM OR ANTIFERROMAGNETISM IN ONE- OR TWO-DIMENSIONAL ISOTROPIC HEISENBERG MODELS*

N. D. Mermin† and H. Wagner‡
Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York
(Received 17 October 1966)

It is rigorously proved that at any nonzero temperature, a one- or two-dimensional isotropic spin-S Heisenberg model with finite-range exchange interaction can be neither ferromagnetic nor antiferromagnetic. The method of proof is capable of excluding a variety of types of ordering in one and two dimensions.


David Mermin (1935 -)


Herbert Wagner (1935 -)

- Continuous symmetry + Short-Range interaction

No Finite-Temperature Ordering for $\mathrm{D} \leq 2$

- 1D (spin chains, ladders…)
- 2D planes, bilayers, finite number of layers

PHYSICAL REVIEW VOLUME 158, NUMBER 2 10 JUNE 1967

Existence of Long-Range Order in One and Two Dimensions

P. C. HOHENBERG
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received 24 October 1966)

It is pointed out that a rigorous inequality first proved by Bogoliubov may be used to rule out the existence of quasi-averages (or long-range order) in Bose and Fermi systems for one and two dimensions and $T \neq 0$.


Pierre Hoenberg
(1934 - 2017)

The special case of 2D

Long-Range Order only at T=0

The special case of 2D

Berezinskii
(1935-1980)

Kosterlitz
(1943 -)

Thouless
(1934-2019)

VL Berezinskii
Sov. Phys. JETP **34** 610 (1971)

J M Kosterlitz and D J Thouless
J. Phys. C **6** 1181 (1973)

The special case of 2D

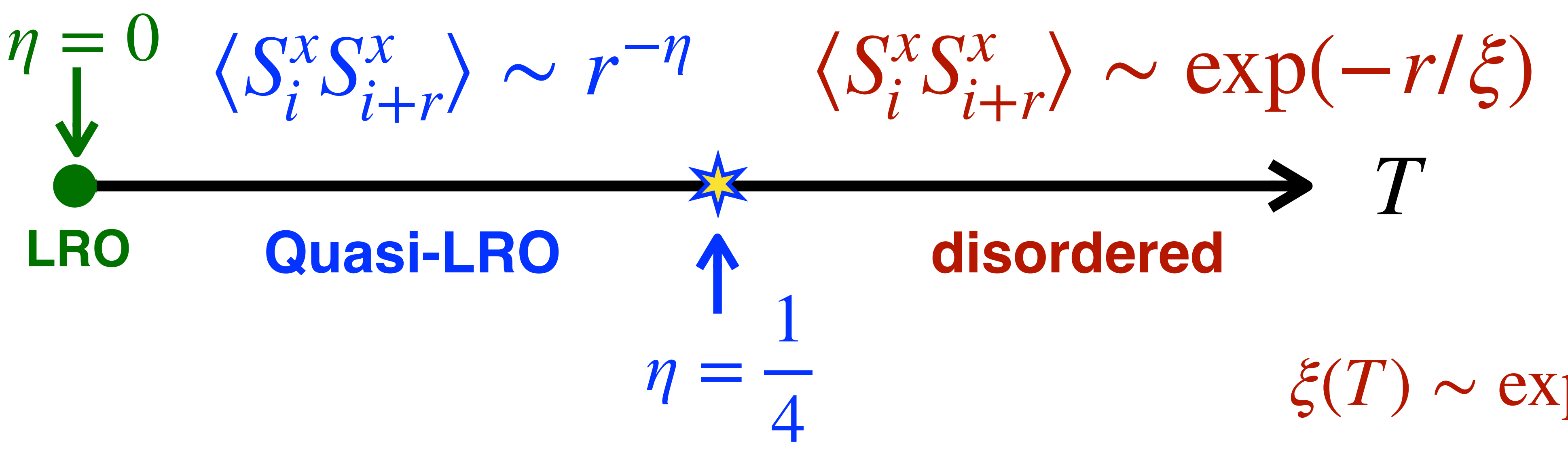
Berezinskii
(1935-1980)

Kosterlitz
(1943 -)

Thouless
(1934-2019)

VL Berezinskii
Sov. Phys. JETP **34** 610 (1971)

J M Kosterlitz and D J Thouless
J. Phys. C **6** 1181 (1973)



$$\xi(T) \sim \exp\left(b\sqrt{\frac{T_{\rm BKT}}{T - T_{\rm BKT}}} \right)$$

Example: the 2D XY model

$$\mathcal{H}_{\rm XY} = -J \sum_{\langle ij \rangle} \left(S_i^x S_j^x + S_i^y S_j^y \right)$$

Berezinskii
(1935-1980)

Kosterlitz
(1943 -)

Thouless
(1934-2019)

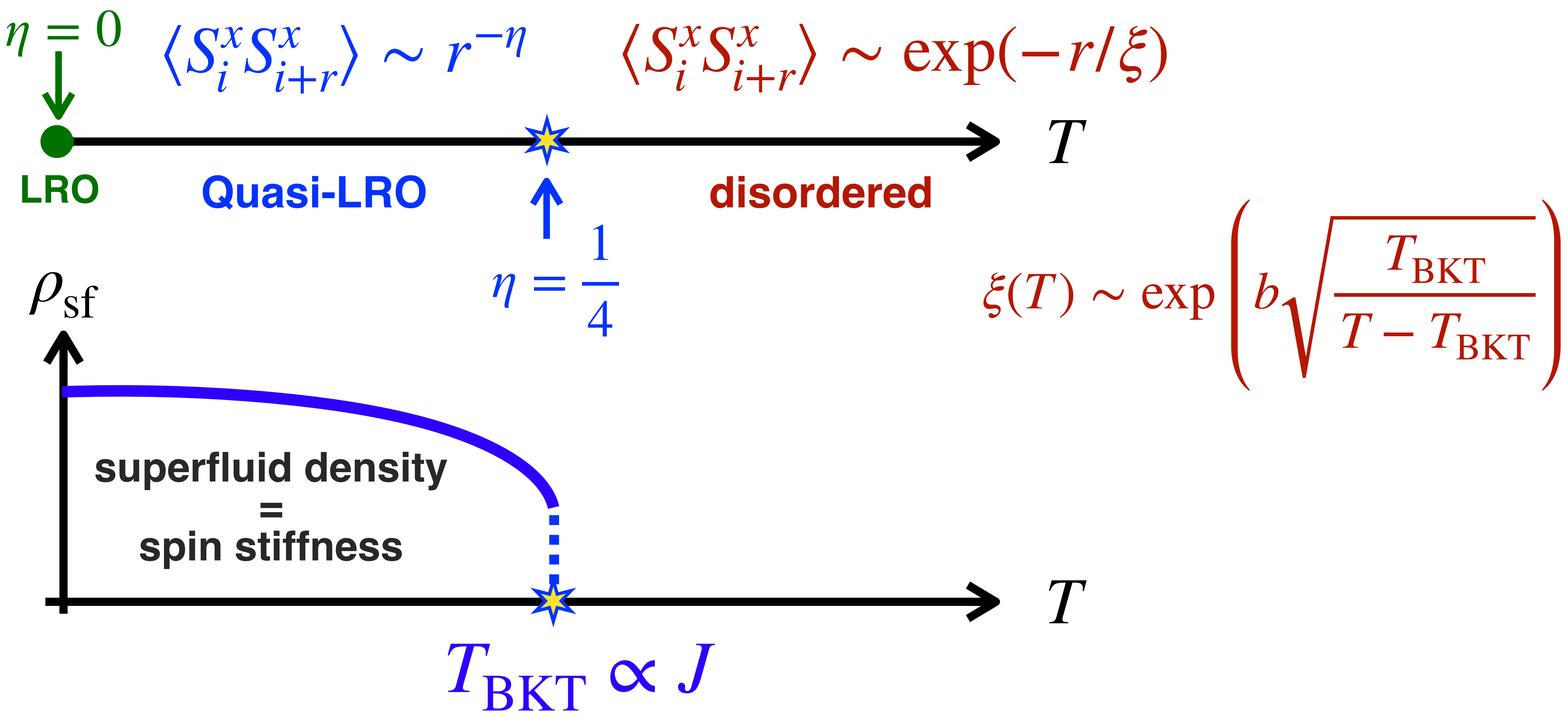
The special case of 2D

VL Berezinskii
Sov. Phys. JETP **34** 610 (1971)

J M Kosterlitz and D J Thouless
J. Phys. C **6** 1181 (1973)

Example: the 2D XY model

$$\mathscr{H}_{\mathrm{XY}} = -J \sum_{\langle ij \rangle} \left(S_i^x S_j^x + S_i^y S_j^y \right)$$



The special case of 2D

Berezinskii (1935-1980)

Kosterlitz (1943 -)

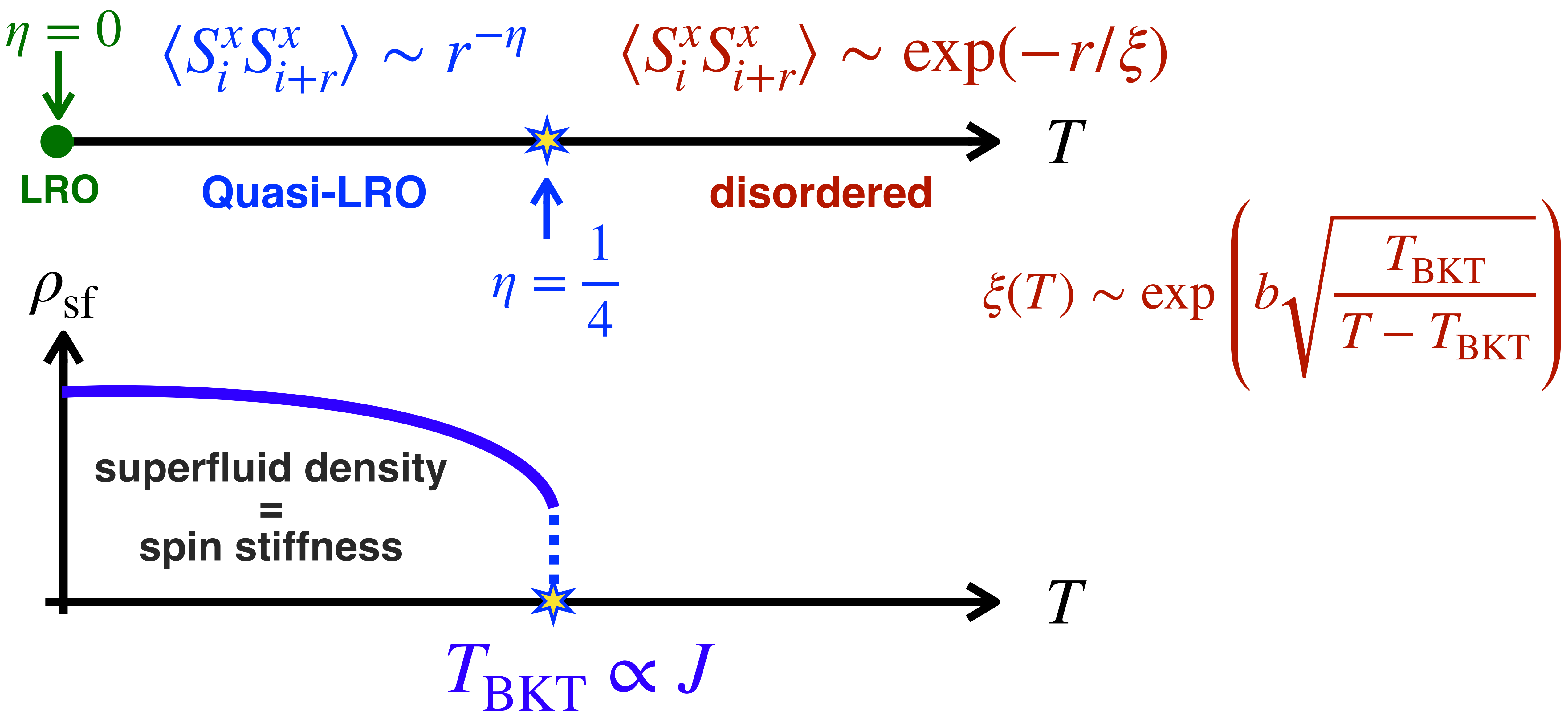
Thouless (1934-2019)

VL Berezinskii
Sov. Phys. JETP **34** 610 (1971)

J M Kosterlitz and D J Thouless
J. Phys. C **6** 1181 (1973)

Example: the 2D XY model

$$\mathcal{H}_{\mathrm{XY}} = -J\sum_{\langle ij\rangle}\left(S_i^x S_j^x + S_i^y S_j^y\right)$$



BKT Physics may be relevant in many situations

- **Layered spin systems**

Magnetization and universal sub-critical behaviour in two-dimensional *XY* magnets

S T Bramwell† and P C W Holdsworth‡

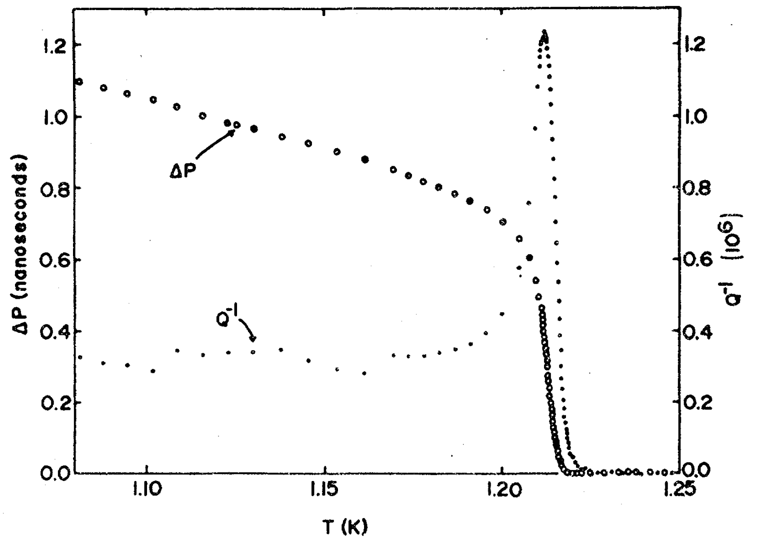
Signatures for Berezinskii-Kosterlitz-Thouless critical behavior in the planar antiferromagnet $BaNi_2V_2O_8$

E. S. Klyushina, J. Reuther, L. Weber, A. T. M. N. Islam, J. S. Lord, B. Klemke, M. Månsson, S. Wessel, and B. Lake
Phys. Rev. B **104**, 064402 – Published 2 August 2021

- **Superfluid helium films**

Study of the superfluid transition in two-dimensional ^{4}He films

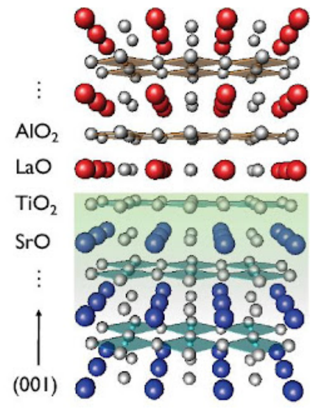
D. J. Bishop* and J. D. Reppy



- **Superconductors**

Superconducting Interfaces Between Insulating Oxides

N. Reyren,[1] S. Thiel,[2] A. D. Caviglia,[1] L. Fitting Kourkoutis,[3] G. Hammerl,[2] C. Richter,[2] C. W. Schneider,[2] T. Kopp,[2] A.-S. Rüetschi,[1] D. Jaccard,[1] M. Gabay,[4] D. A. Muller,[3] J.-M. Triscone,[1] J. Mannhart[2]*



- **Cold atoms**

Nature **594**, 191 (2021)

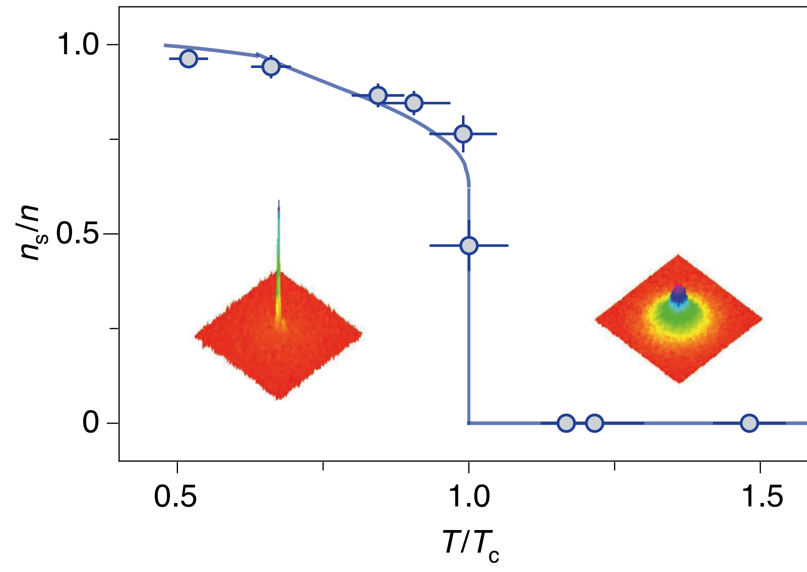
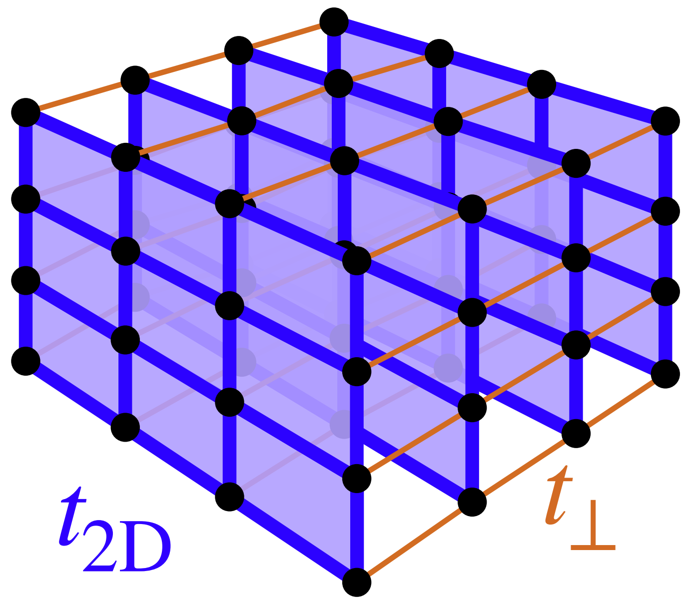


Fig. 1 | Universal jump in superfluid density.

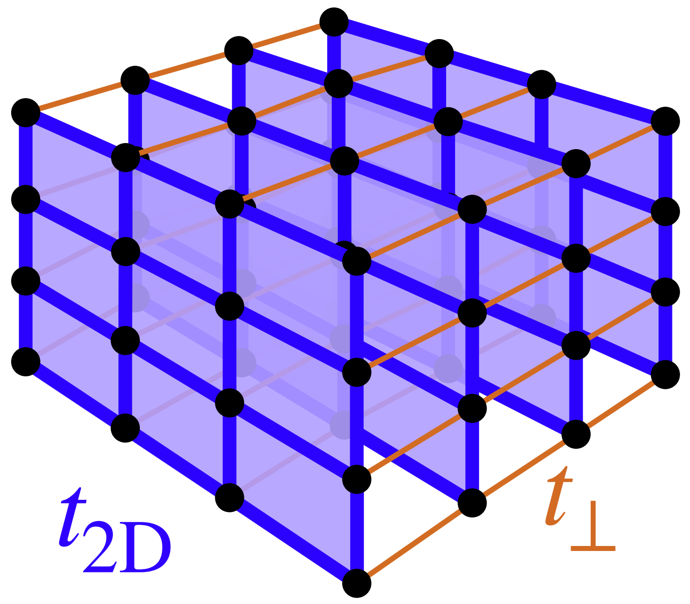
Quasi-2D: significance of energy scales

Weakly (3d) coupled superfluid/superconducting planes

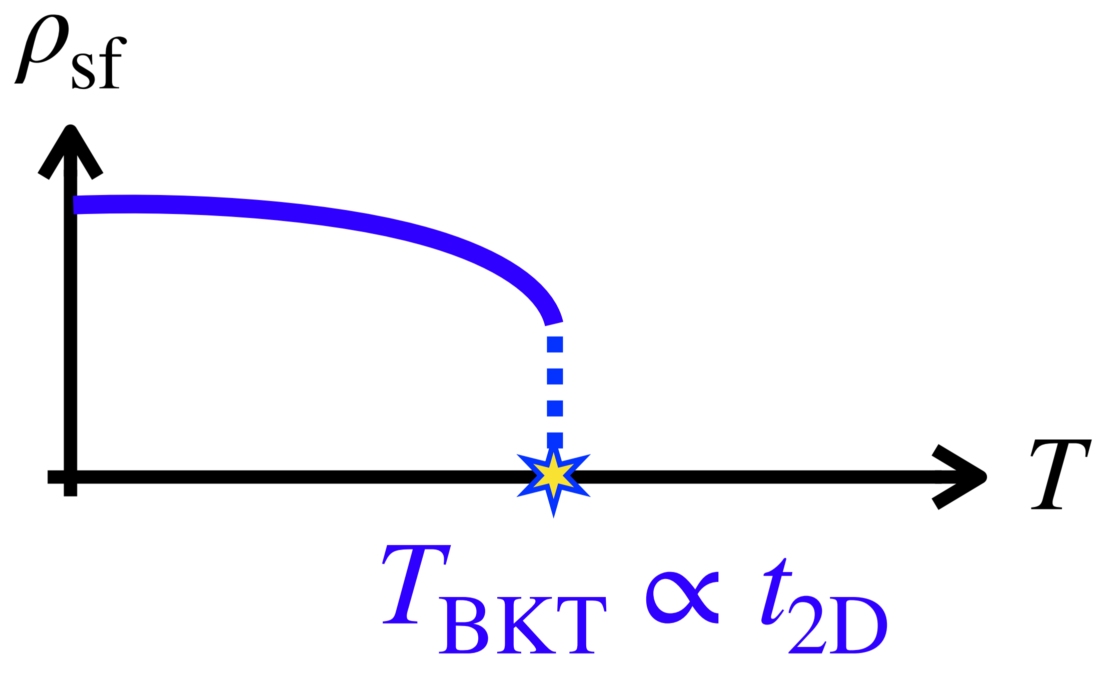


Quasi-2D: significance of energy scales

Weakly (3d) coupled superfluid/superconducting planes

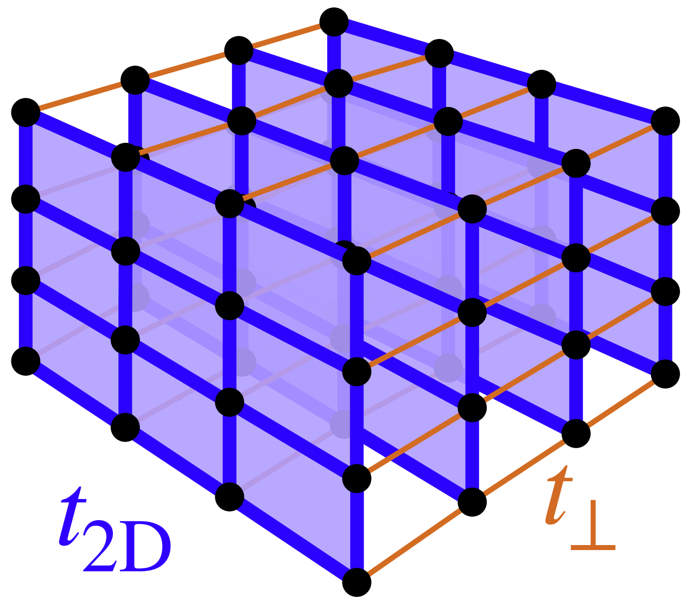


- When decoupled $(t_\perp = 0)$ each individual layer displays BKT physics

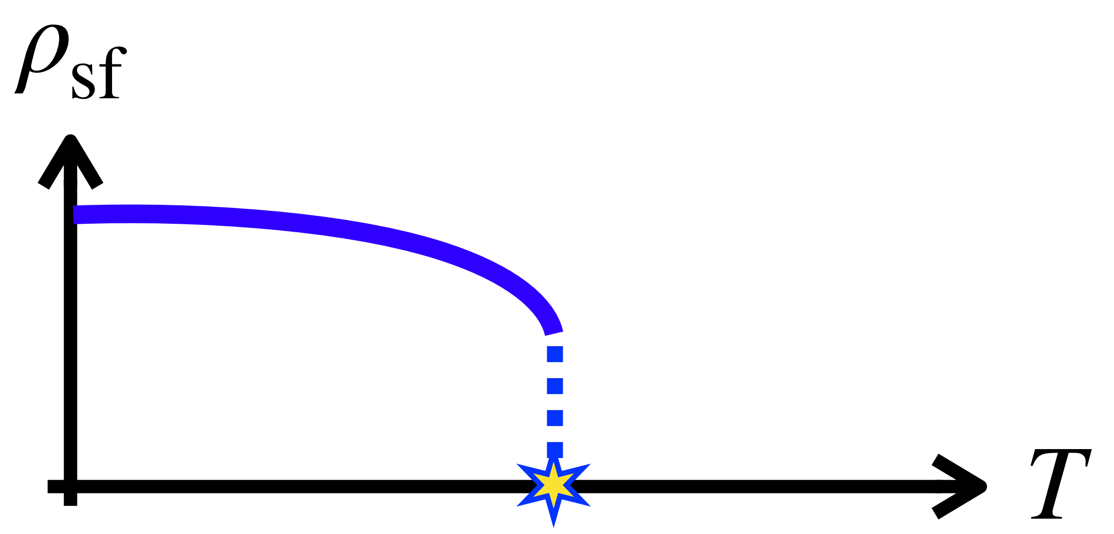


Quasi-2D: significance of energy scales

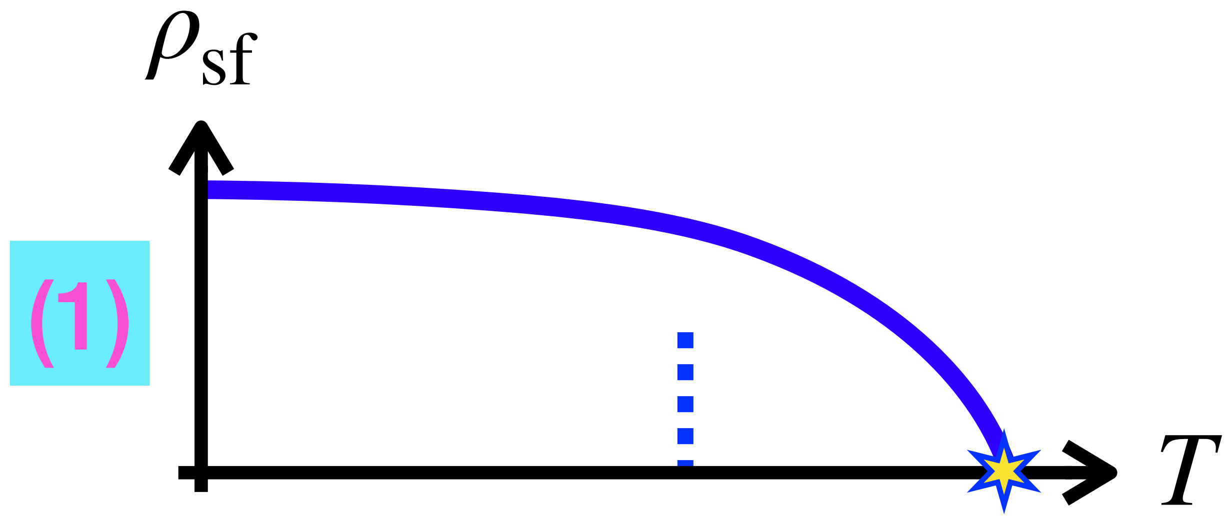
Weakly (3d) coupled superfluid/superconducting planes

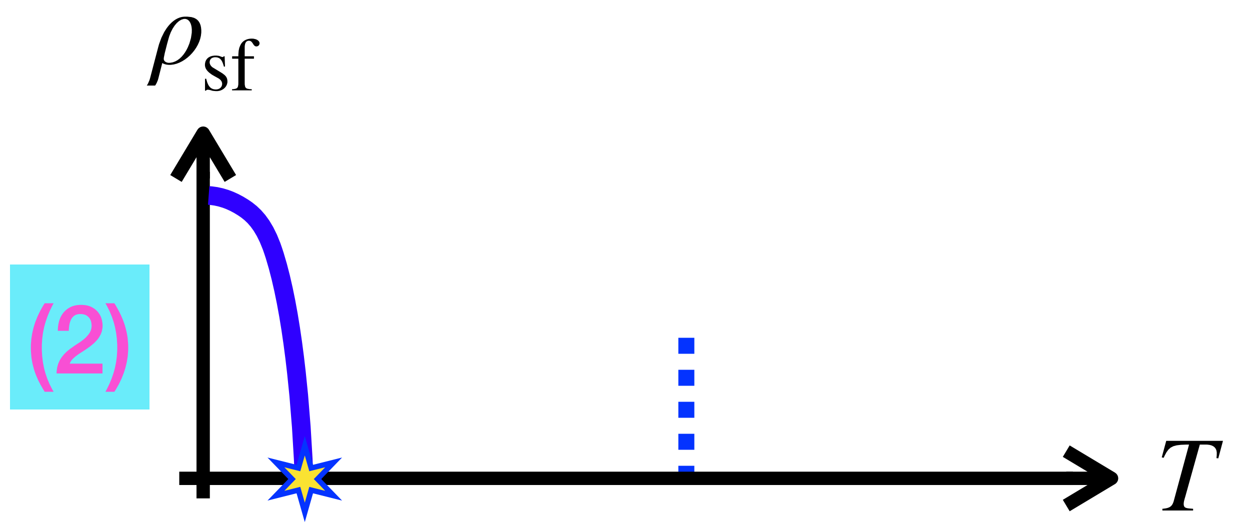


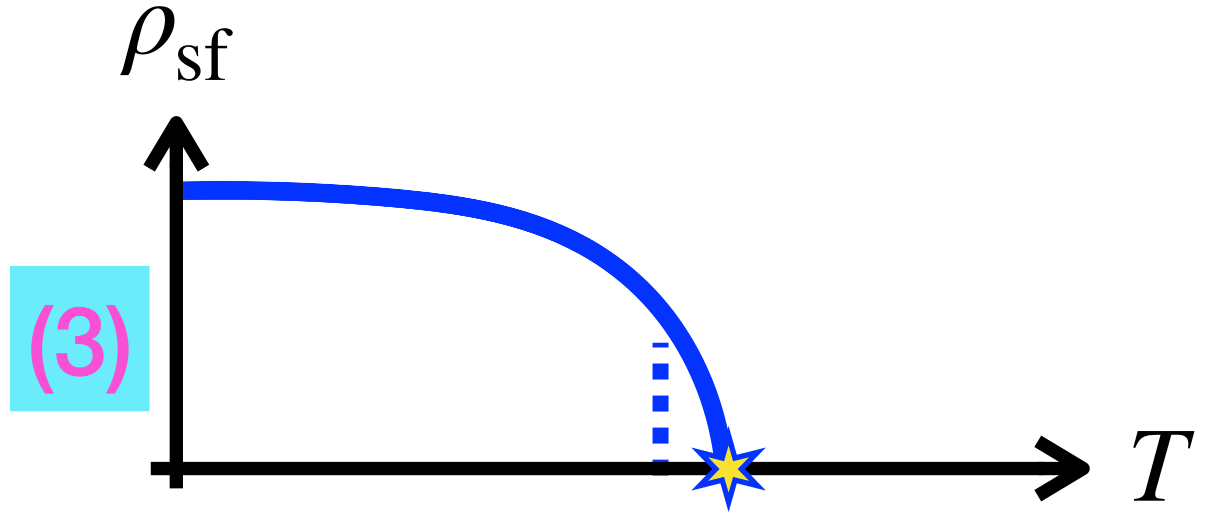
- When decoupled $(t_\perp = 0)$ each individual layer displays BKT physics
- Weakly 3D coupled layers $(t_\perp / t_{2D} \ll 1) \Rightarrow$ 3D ordering at a finite T_c



$T_{BKT} \propto t_{2D}$







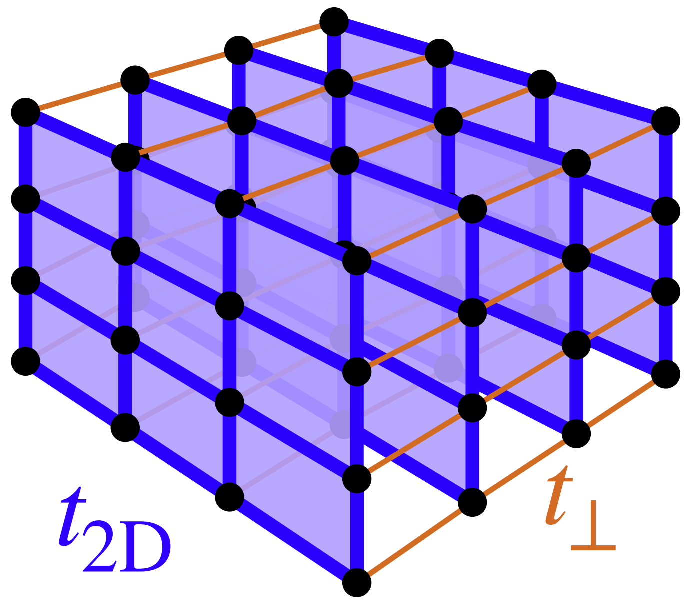
$$T_c^{3D} = T_{BKT}\left(1 + \frac{a}{\ln^2\left(t_\perp / t_{2D}\right) + c}\right)$$

$T_c^{3D} \propto t_\perp$

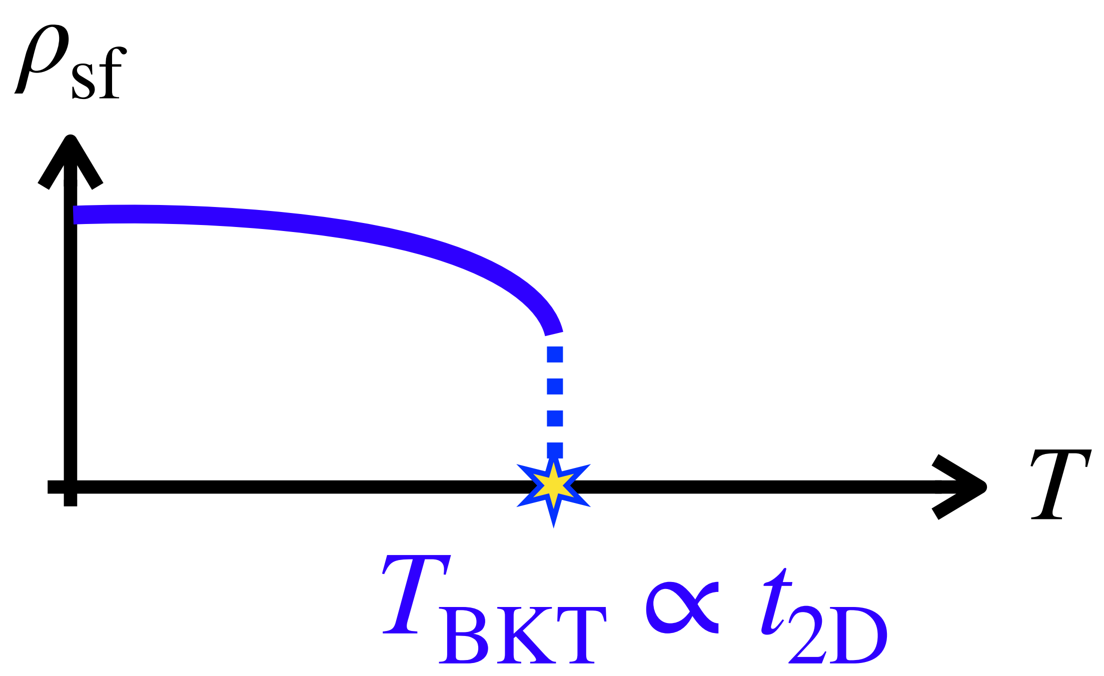
$T_c^{3D} = T_{BKT} + a t_\perp$

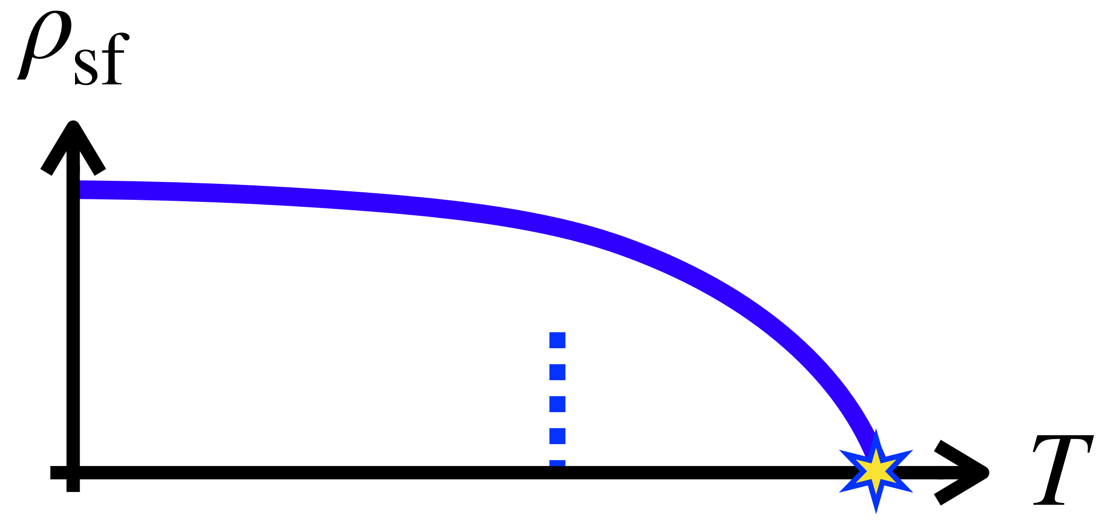
Quasi-2D: significance of energy scales

Weakly (3d) coupled superfluid/superconducting planes



- When decoupled $(t_\perp = 0)$ each individual layer displays BKT physics
- Weakly 3D coupled layers $(t_\perp / t_{2D} \ll 1) \Rightarrow$ 3D ordering at a finite T_c

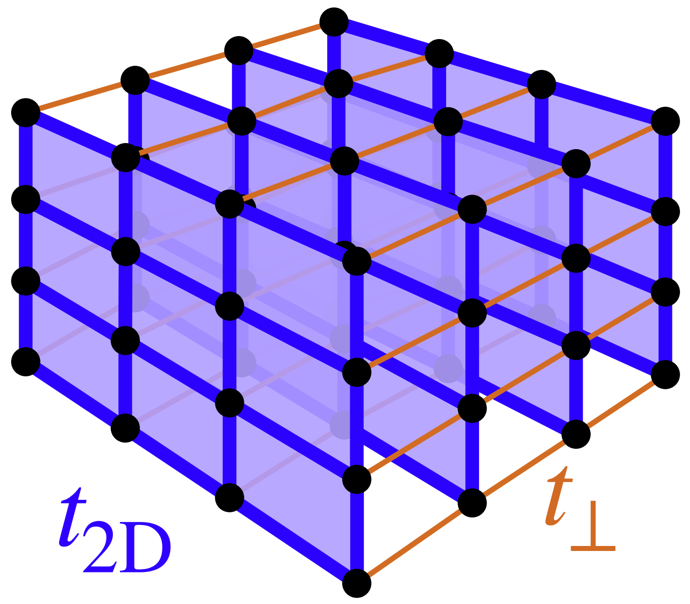




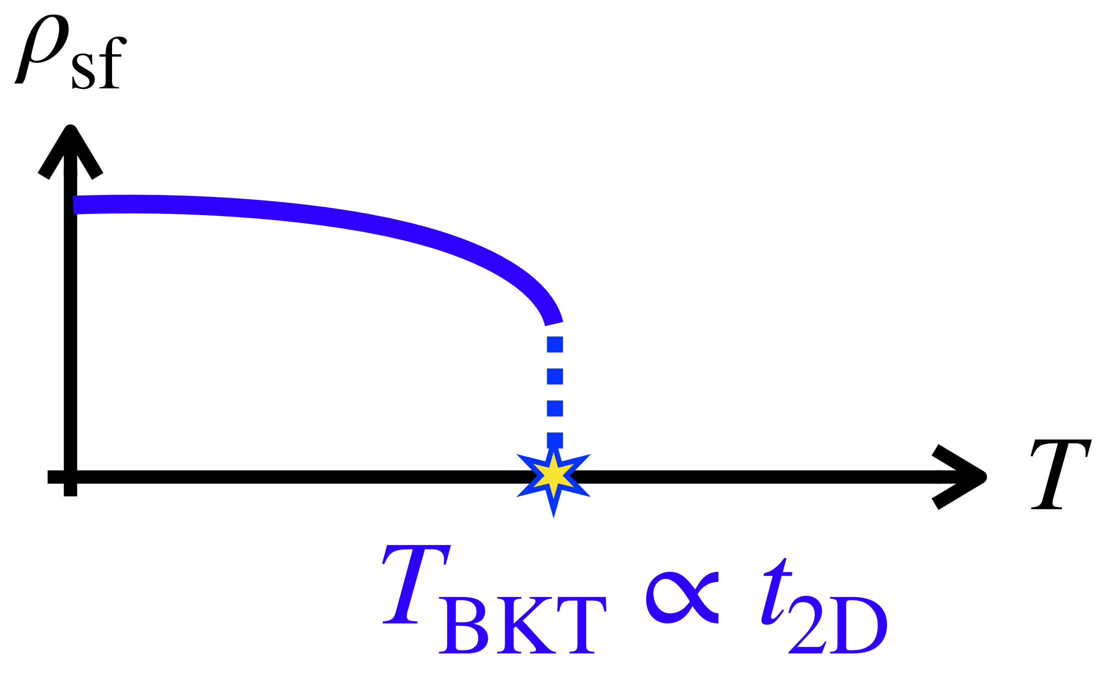
$$T_c^{3D} = T_{BKT}\left(1 + \frac{a}{\ln^2\left(t_\perp / t_{2D}\right) + c}\right)$$

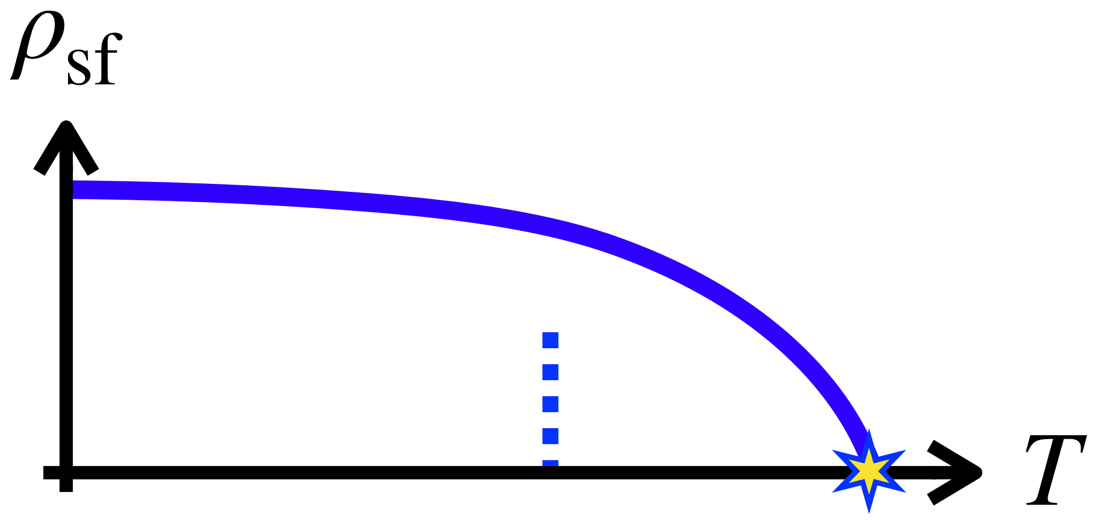
Quasi-2D: significance of energy scales

Weakly (3d) coupled superfluid/superconducting planes



- When decoupled $(t_\perp = 0)$ each individual layer displays BKT physics
- Weakly 3D coupled layers $(t_\perp/t_{2D} \ll 1) \Rightarrow$ 3D ordering at a finite T_c





$$T_c^{3D} = T_{BKT}\left(1 + \frac{a}{\ln^2\left(t_\perp/t_{2D}\right) + c}\right)$$

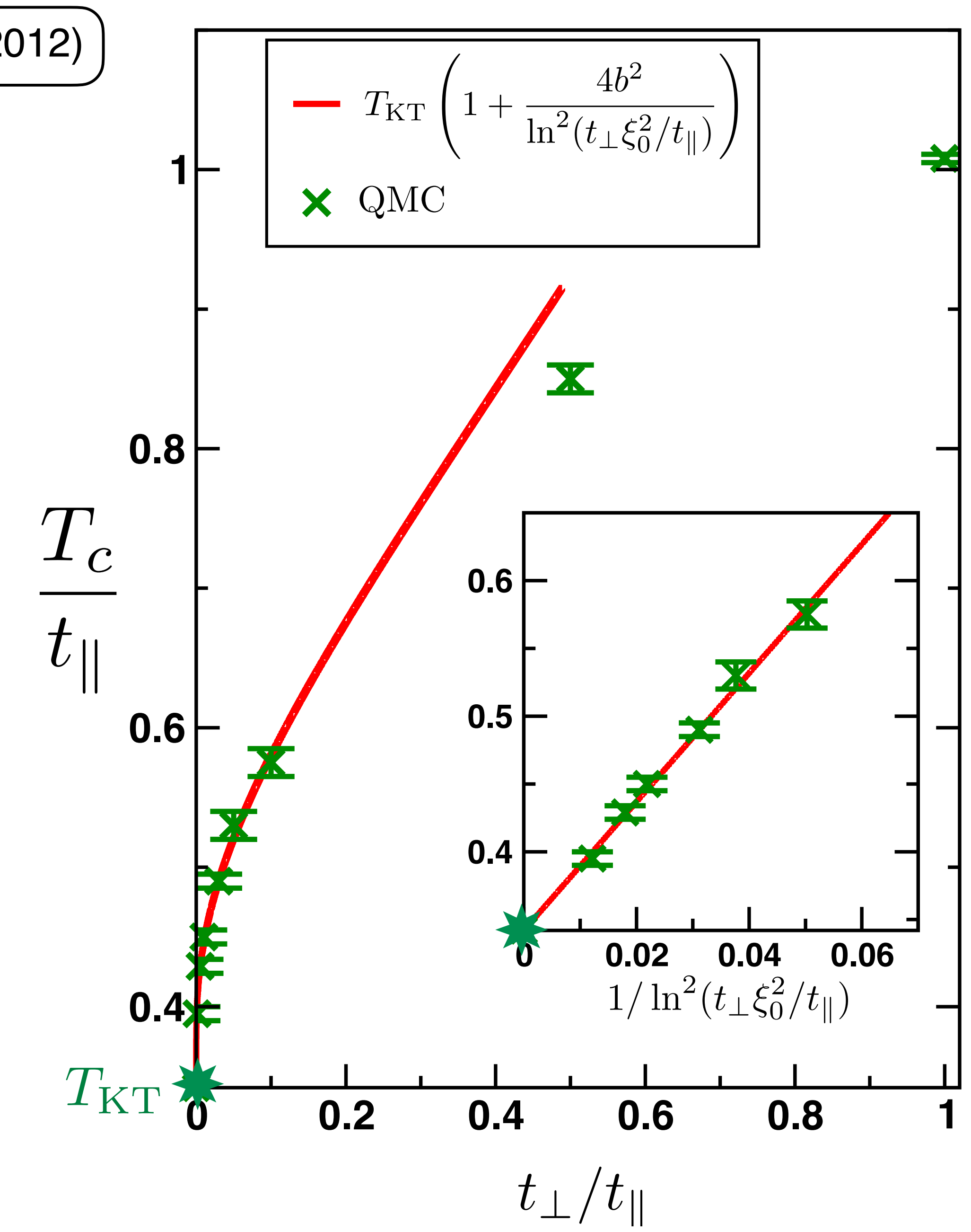
- **Mean-Field argument**

$$\xi_{2D}(T) \approx \xi_0 \exp\left(b\sqrt{\frac{T_{BKT}}{T - T_{BKT}}}\right)$$

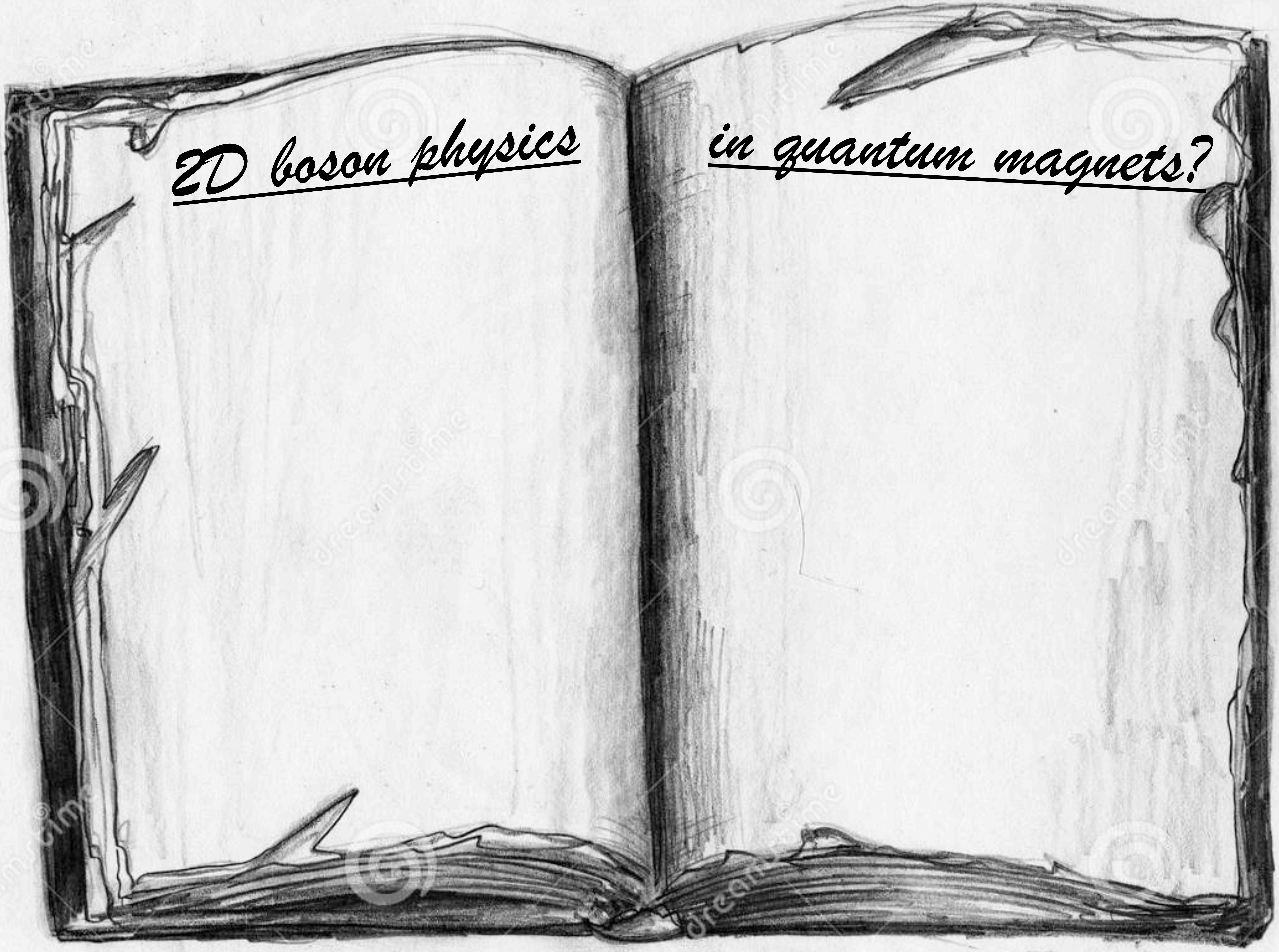
3D effects when $\xi_{2D}^2(T) \times t_\perp \sim t_{2D}$

Quantum Monte Carlo simulations of the 2D XY model

NL Europhysics Letters **99**, 66001 (2012)



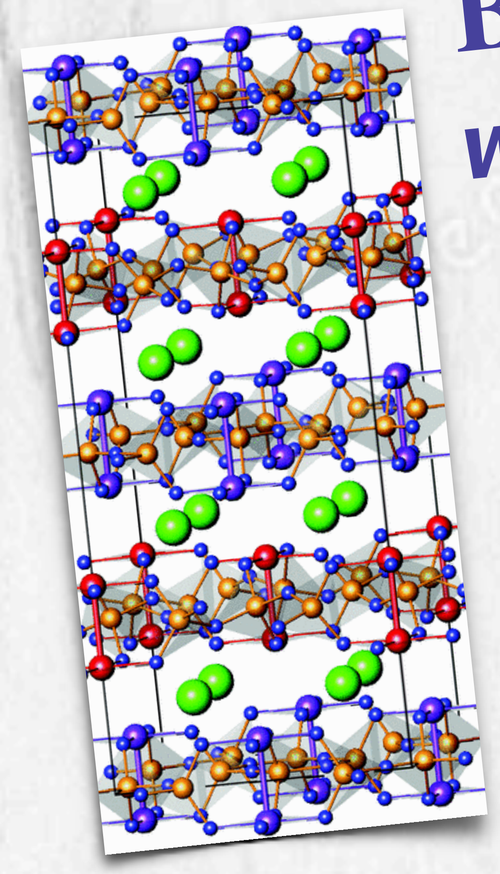
Strong T_c enhancement by very weak $t_\perp$


2D boson physics
in quantum magnets?

2D boson physics in quantum magnets?

The (long story of) the Han purple

[-200 … 2022]



$BaCuSi_2O_6$

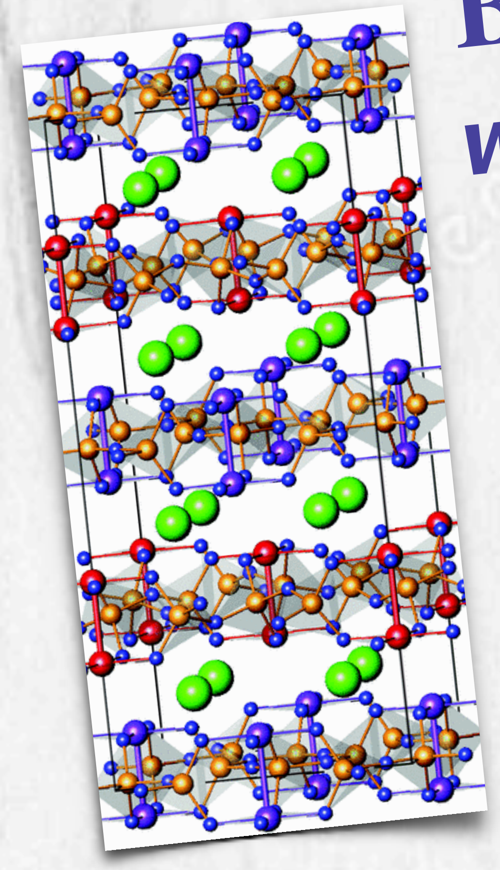
weakly coupled $S=1/2$ bilayers

2D boson physics in quantum magnets?

The (long story of) the Han purple [-200 … 2022]

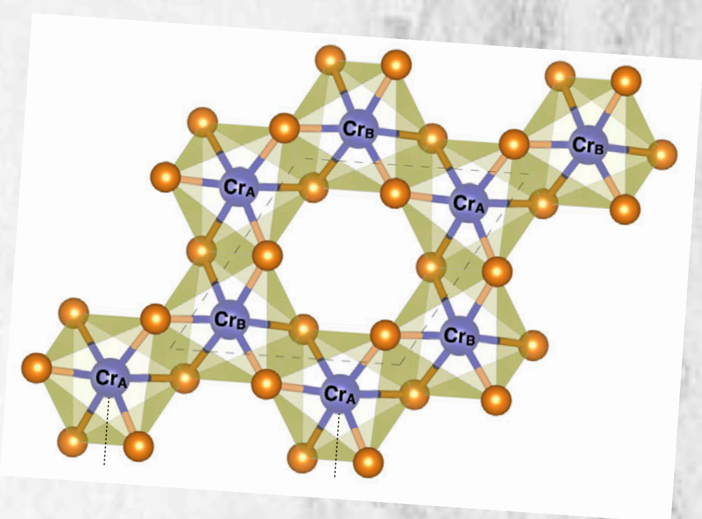
$BaCuSi_2O_6$

weakly coupled $S=1/2$ bilayers



The monolayer halide $CrCl_3$

$S=3/2$ Honeycomb Magnet



Season One

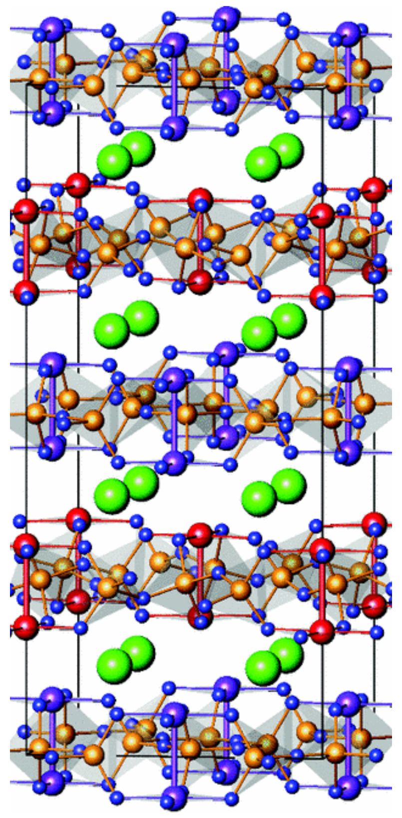
[-200 … 2004]

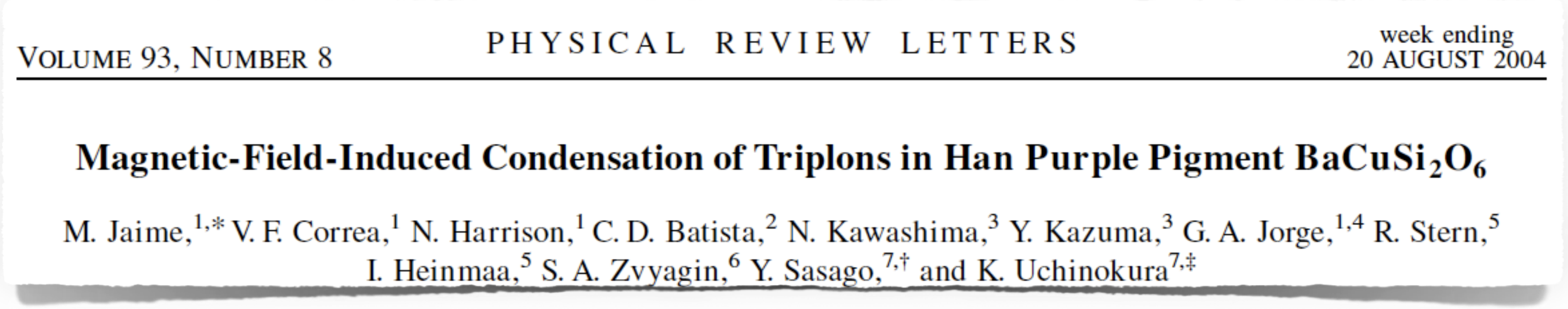
The Han purple

BaCuSi$_2$O$_6$

weakly coupled S=1/2 bilayers

From the Han Dynasty to modern quantum magnetism

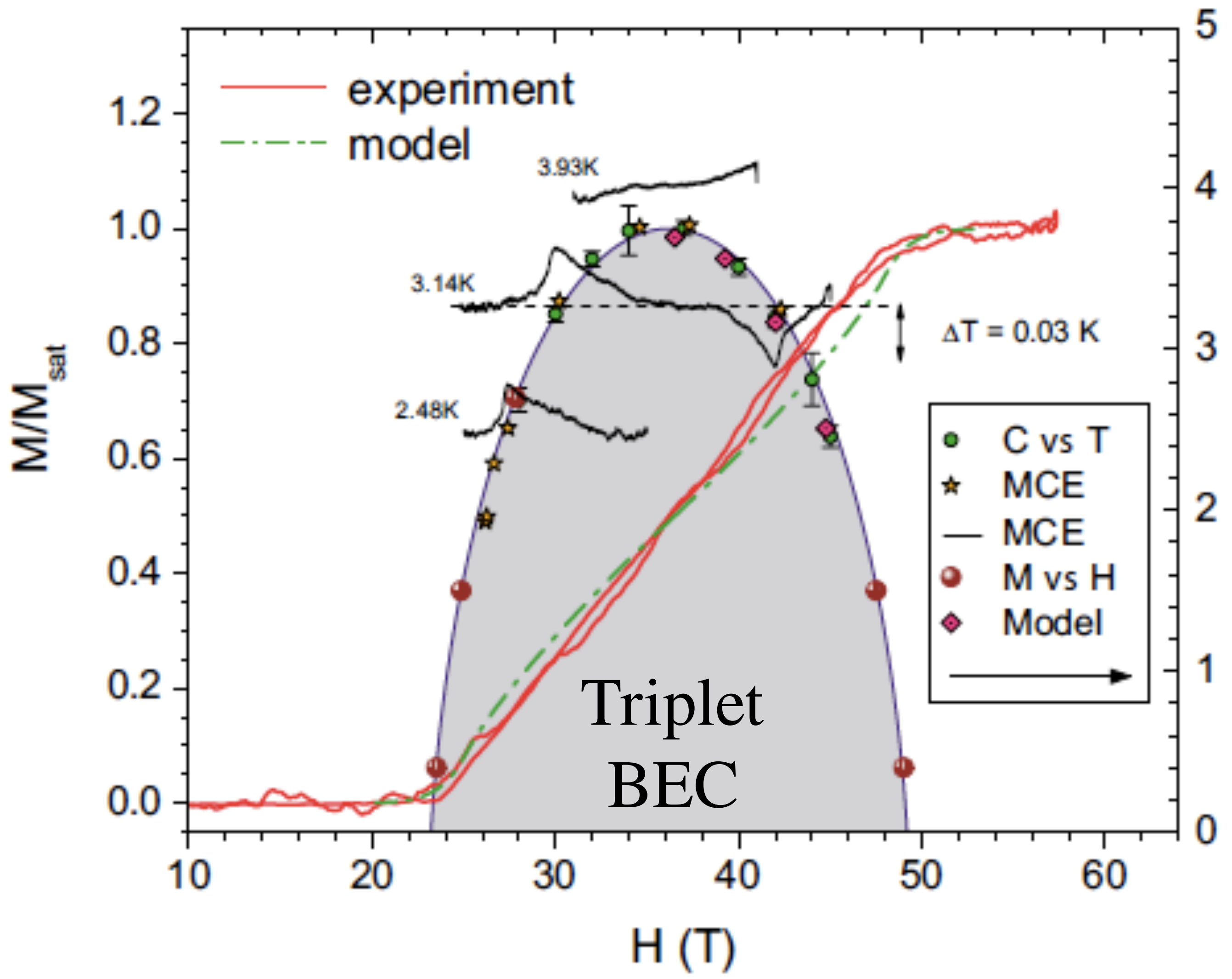




VOLUME 93, NUMBER 8 PHYSICAL REVIEW LETTERS week ending 20 AUGUST 2004

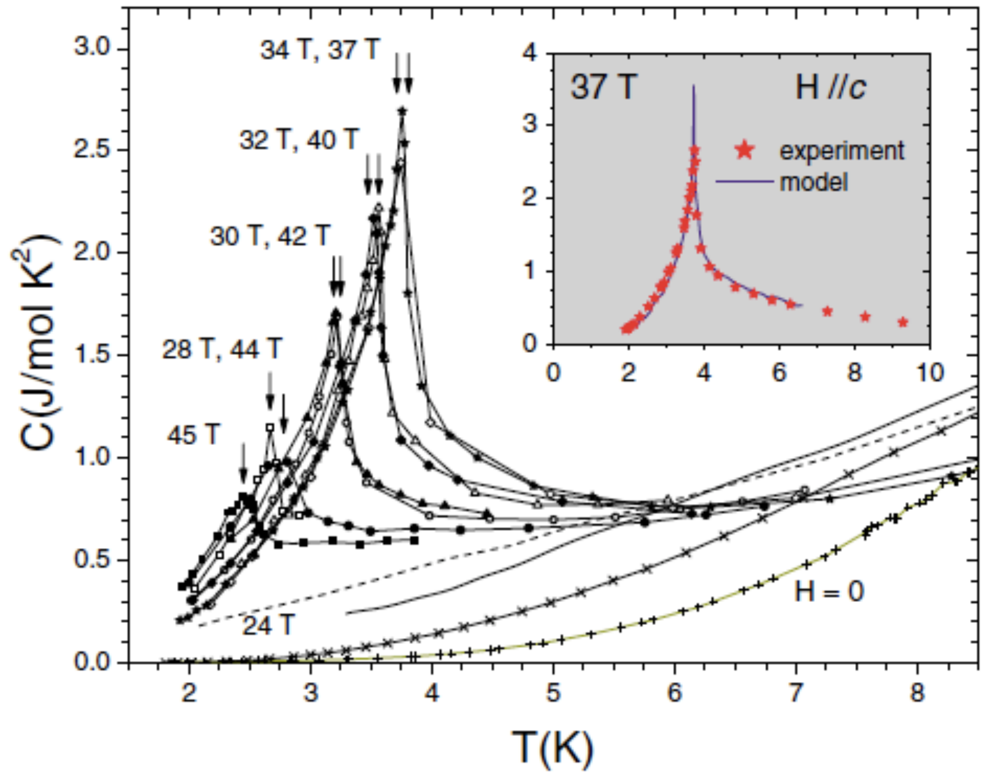
Magnetic-Field-Induced Condensation of Triplons in Han Purple Pigment BaCuSi$_2$O$_6$

M. Jaime,[1,*] V. F. Correa,[1] N. Harrison,[1] C. D. Batista,[2] N. Kawashima,[3] Y. Kazuma,[3] G. A. Jorge,[1,4] R. Stern,[5] I. Heinmaa,[5] S. A. Zvyagin,[6] Y. Sasago,[7,†] and K. Uchinokura[7,‡]



λ – anomaly

BEC (3D XY) universality class



Season One

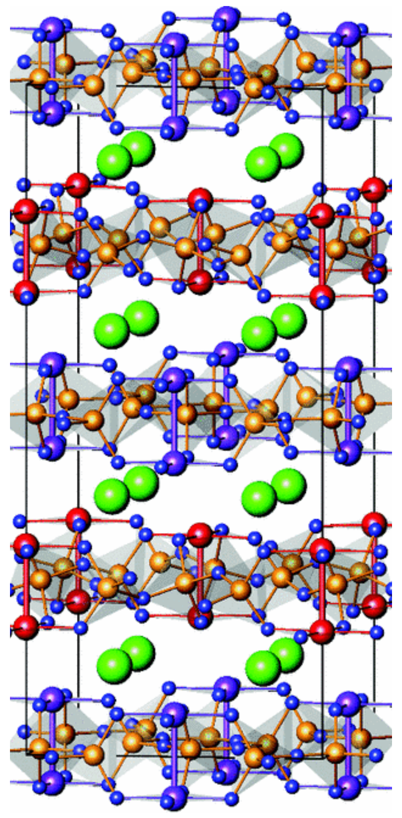
[-200 … 2004]

The Han purple

From the Han Dynasty to modern quantum magnetism

$BaCuSi_2O_6$

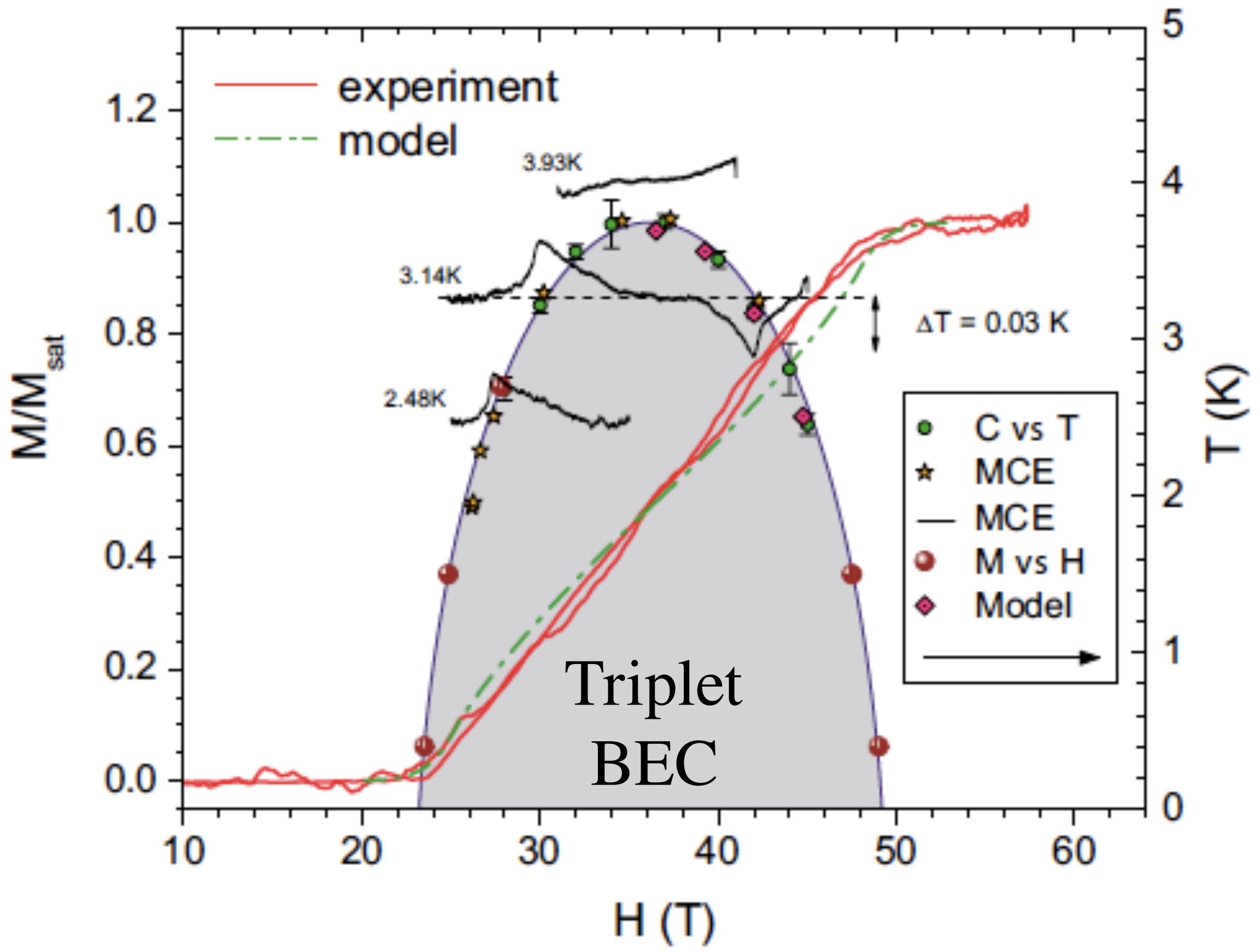
weakly coupled S=1/2 bilayers



VOLUME 93, NUMBER 8 PHYSICAL REVIEW LETTERS week ending 20 AUGUST 2004

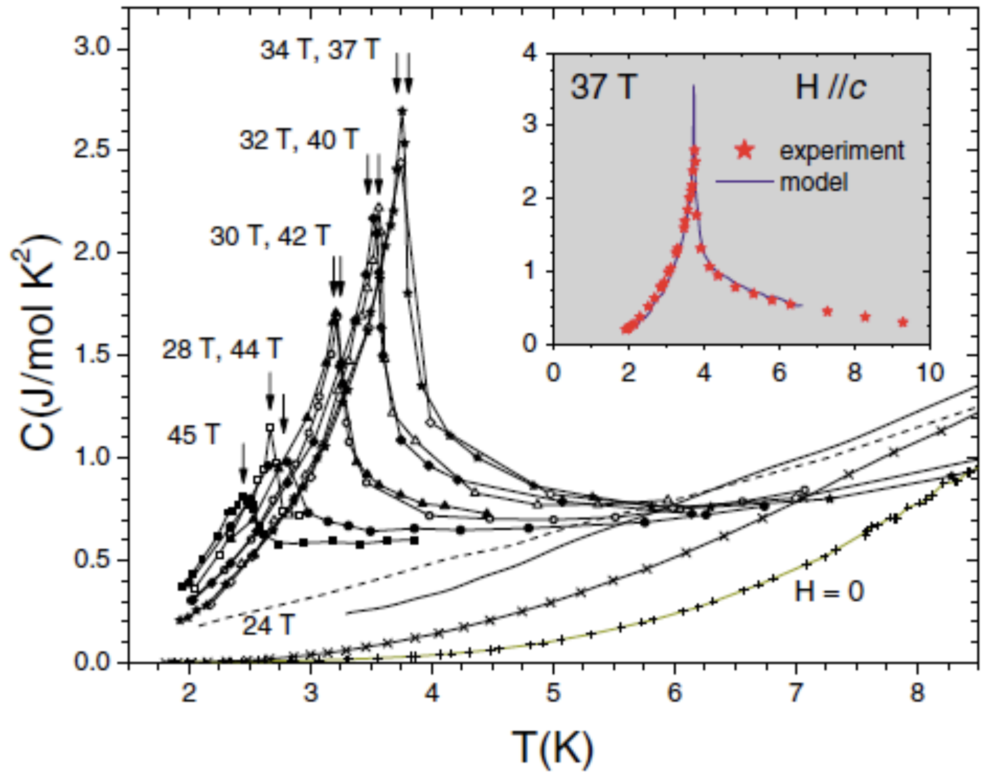
Magnetic-Field-Induced Condensation of Triplons in Han Purple Pigment $BaCuSi_2O_6$

M. Jaime,[1,*] V. F. Correa,[1] N. Harrison,[1] C. D. Batista,[2] N. Kawashima,[3] Y. Kazuma,[3] G. A. Jorge,[1,4] R. Stern,[5] I. Heinmaa,[5] S. A. Zvyagin,[6] Y. Sasago,[7,†] and K. Uchinokura[7,‡]



λ – anomaly

BEC (3D XY) universality class

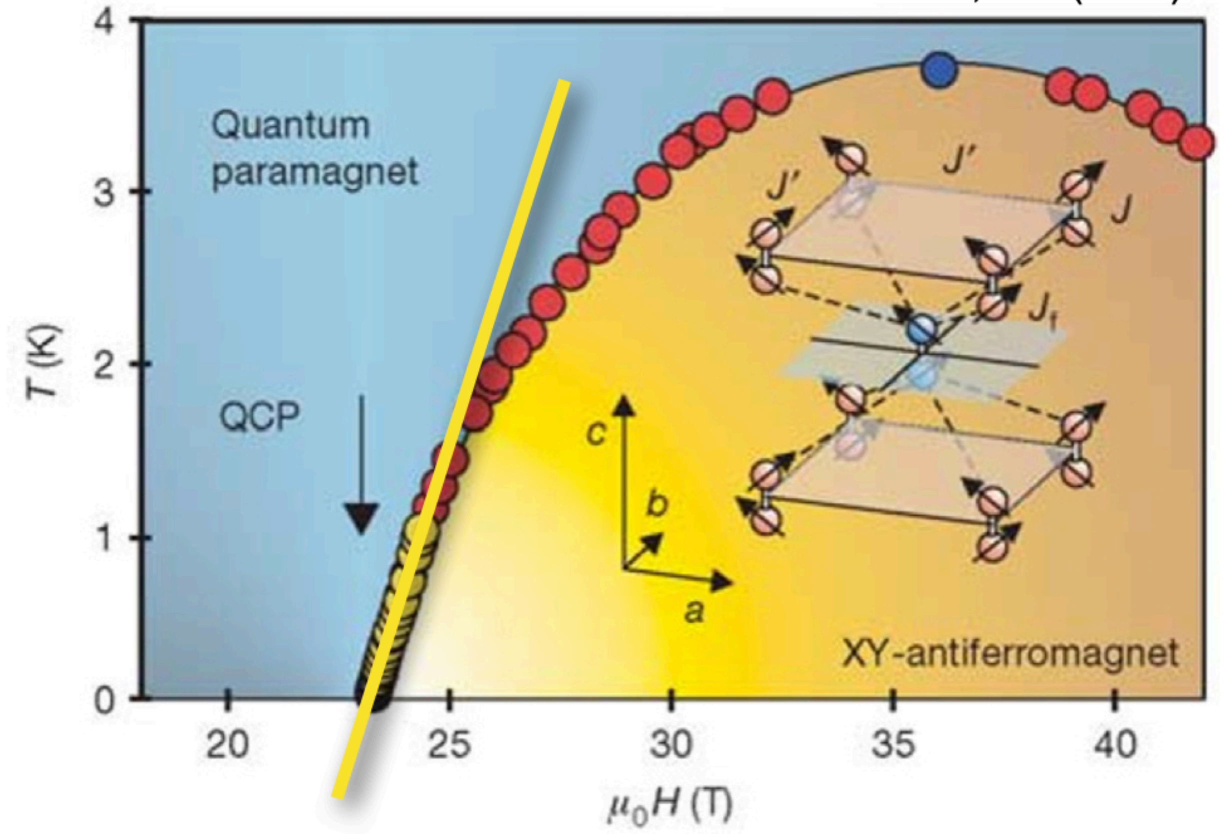


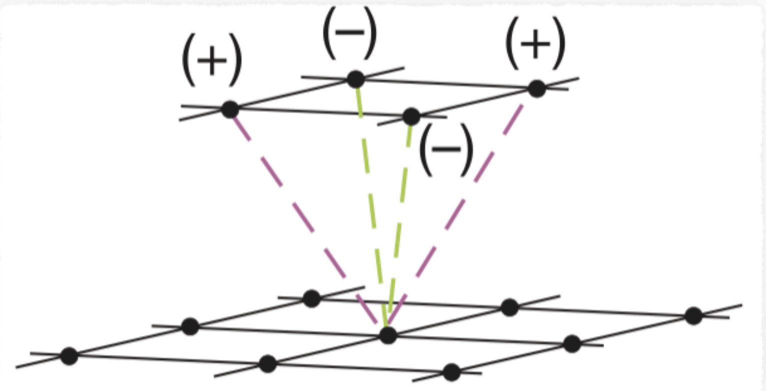
Season two

[2006]

Frustration induced dimensional reduction

Sebastian *et al.* Nature **441**, 617 (2006)



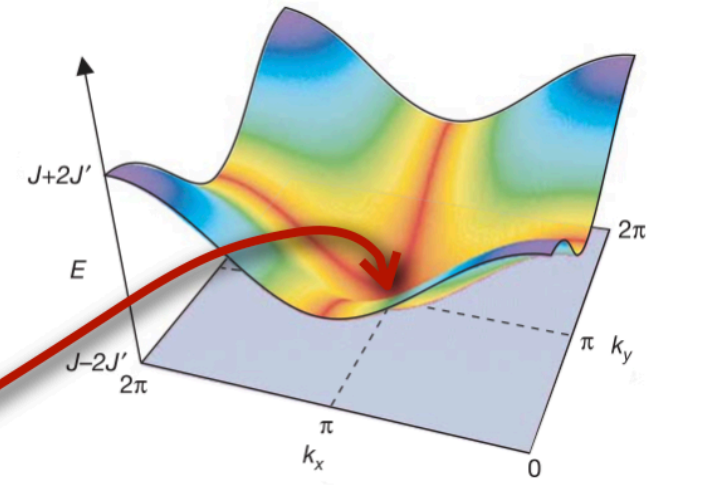


$$T_{\rm BEC} \propto (H - H_c)^{\phi=1}$$

$\phi = 2/D$ (shift exponent for BEC)

Triplet dispersion

$$E(\vec{k}) = J_{\parallel}(\cos k_x + \cos k_y) + 2J_{\perp}\left[\cos\left(\frac{k_x}{2}\right)\cos\left(\frac{k_y}{2}\right)\right]\cos k_z$$



Interlayer hopping vanishes at the BEC point $(k_x, k_y) = (\pi, \pi)$

Interlayer frustration prevents Bose-condensed triplets to 3D disperse
⇒ Dimensional reduction

Season 3

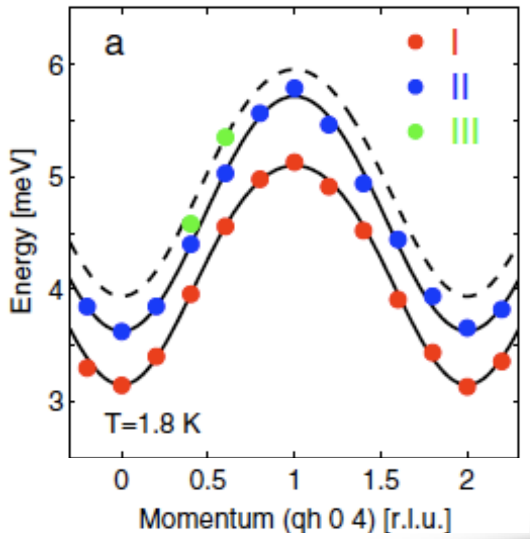
[2007]

Neutrons and NMR: A new hope?

Multiple Magnon Modes and Consequences for the Bose-Einstein Condensed Phase in $BaCuSi_2O_6$

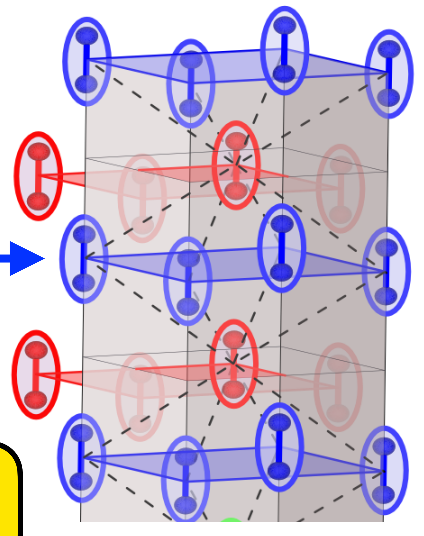
Ch. Rüegg,[1] D. F. McMorrow,[1,2] B. Normand,[3] H. M. Rønnow,[4] S. E. Sebastian,[5] I. R. Fisher,[5] C. D. Batista,[6] S. N. Gvasaliya,[4] Ch. Niedermayer,[4] and J. Stahn[4]

PRL **98**, 017202 (2007)



$J_B \simeq 4.7$ meV (54.5 K)

$J_A \simeq 4.27$ meV (49.5 K)

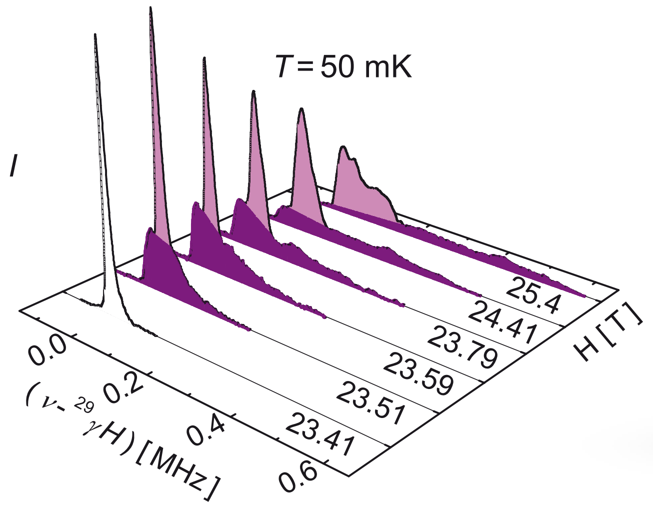


Two types of bilayers

Nuclear magnetic resonance evidence for a strong modulation of the Bose-Einstein condensate in $BaCuSi_2O_6$

S. Krämer,[1] R. Stern,[2] M. Horvatić,[1] C. Berthier,[1] T. Kimura,[3] and I. R. Fisher[4]

PRB **76**, 100406(R) (2007)



Triplet population strongly modulated along the c-axis

Season 3

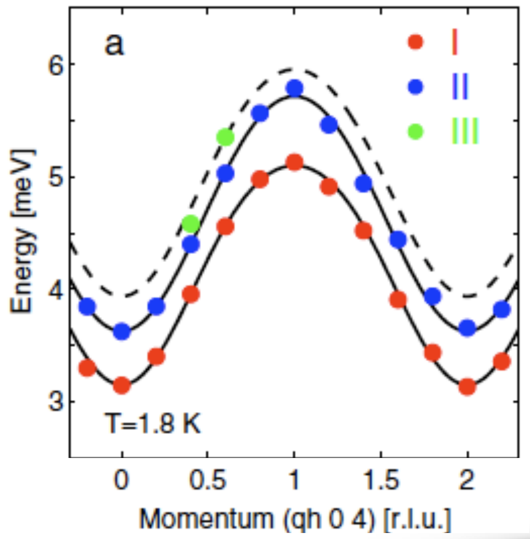
[2007]

Neutrons and NMR: A new hope?

Multiple Magnon Modes and Consequences for the Bose-Einstein Condensed Phase in $BaCuSi_2O_6$

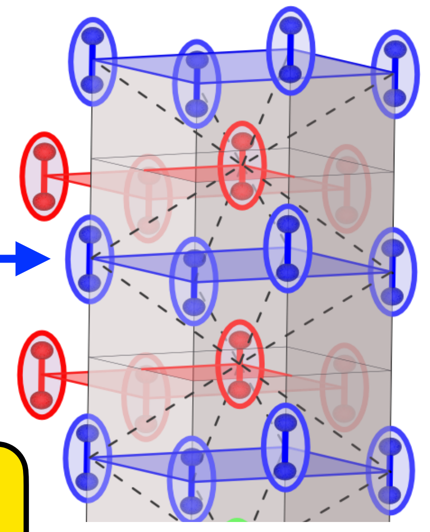
Ch. Rüegg,[1] D. F. McMorrow,[1,2] B. Normand,[3] H. M. Rønnow,[4] S. E. Sebastian,[5] I. R. Fisher,[5] C. D. Batista,[6] S. N. Gvasaliya,[4] Ch. Niedermayer,[4] and J. Stahn[4]

PRL **98**, 017202 (2007)



$J_B \simeq 4.7$ meV (54.5 K)

$J_A \simeq 4.27$ meV (49.5 K)

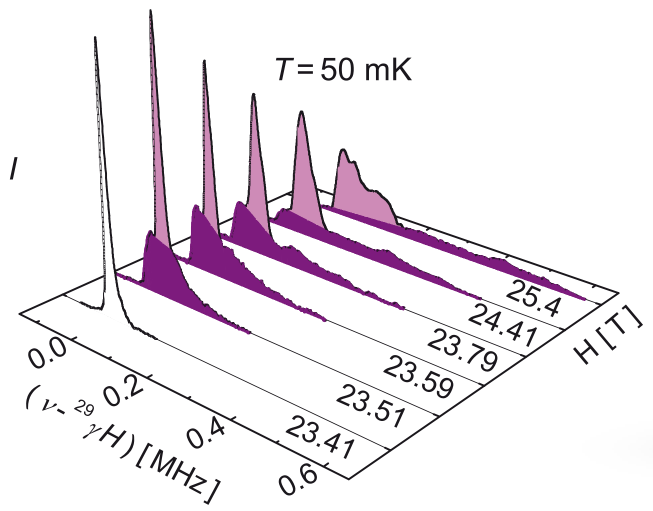


Two types of bilayers

Nuclear magnetic resonance evidence for a strong modulation of the Bose-Einstein condensate in $BaCuSi_2O_6$

S. Krämer,[1] R. Stern,[2] M. Horvatić,[1] C. Berthier,[1] T. Kimura,[3] and I. R. Fisher[4]

PRB **76**, 100406(R) (2007)



Triplet population strongly modulated along the c-axis

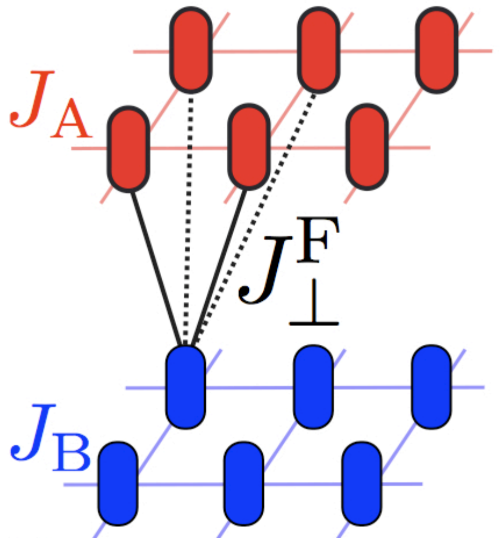
SeasoN Four

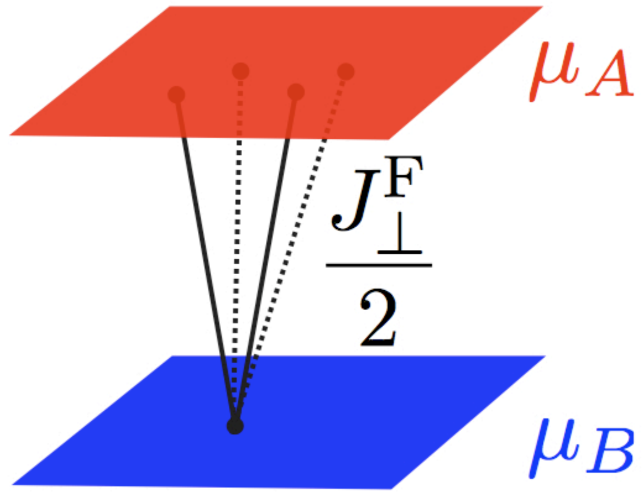
[2009 - 2011]

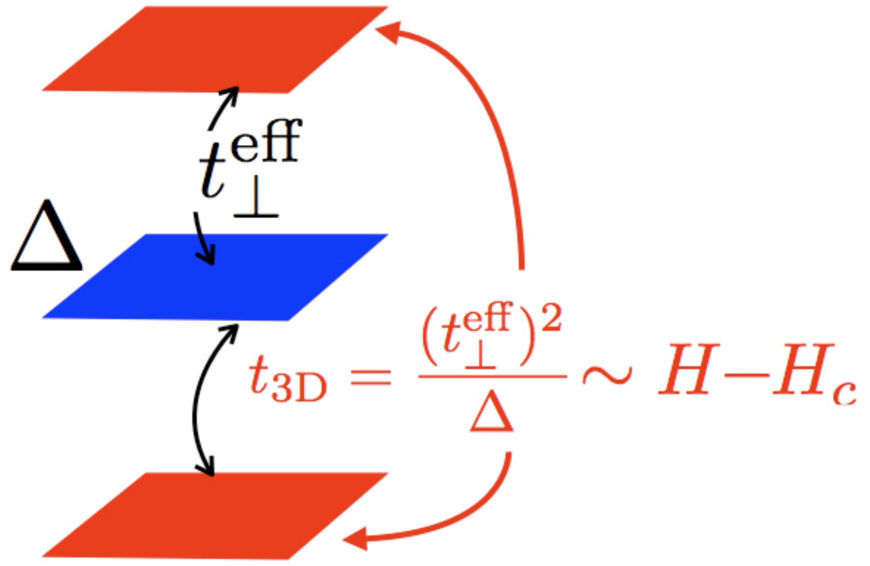
Theory

NL and F. Mila PRL (2009)
NL and F. Mila PRL (2011)

Frustrated inequivalent bilayers → dimer-boson mapping → **Frustrated bosonic layers** → semi-classical expansion → **Non-frustrated effective bosonic model**



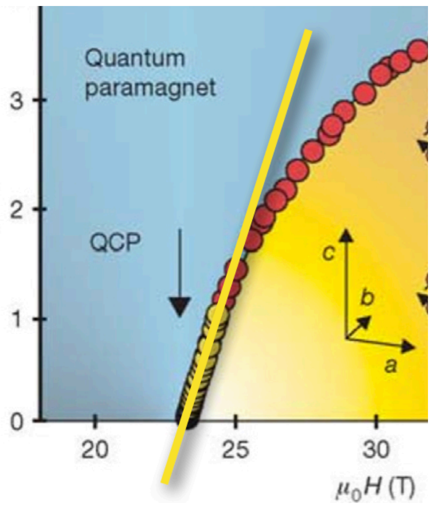




Consequences for the critical temperature

$$T_{\rm BEC} \approx \frac{\hbar^2}{2k_B(m_x m_y m_z)^{1/3}}\left[\kappa(H-H_c)^{2/3}\right]$$

$$\frac{\hbar^2}{2(m_x m_y m_z)^{1/3}} = J_\parallel{}^{2/3} t_{3D}{}^{1/3} \Rightarrow T_{\rm BEC} \sim (H-H_c)^{\frac{2}{3}+\frac{1}{3}}$$



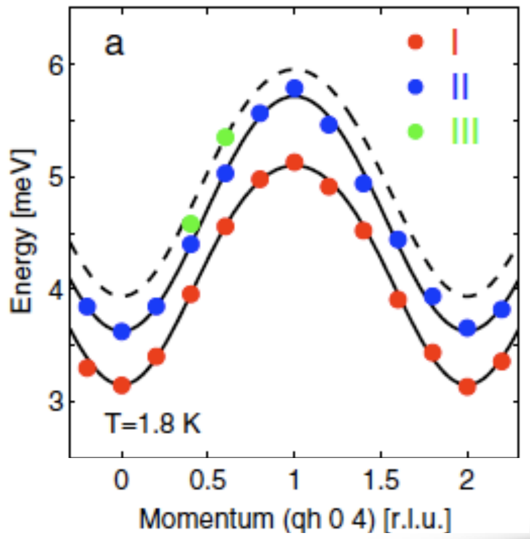
Neutrons and NMR: A new hope?

Season 3 [2007]

Multiple Magnon Modes and Consequences for the Bose-Einstein Condensed Phase in $BaCuSi_2O_6$

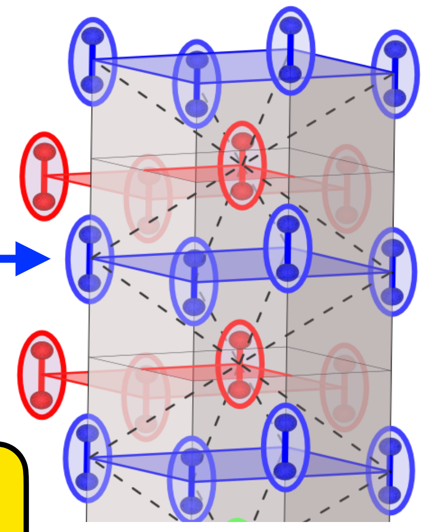
Ch. Rüegg,[1] D. F. McMorrow,[1,2] B. Normand,[3] H. M. Rønnow,[4] S. E. Sebastian,[5] I. R. Fisher,[5] C. D. Batista,[6] S. N. Gvasaliya,[4] Ch. Niedermayer,[4] and J. Stahn[4]

PRL **98**, 017202 (2007)



$J_B \simeq 4.7$ meV (54.5 K)

$J_A \simeq 4.27$ meV (49.5 K)

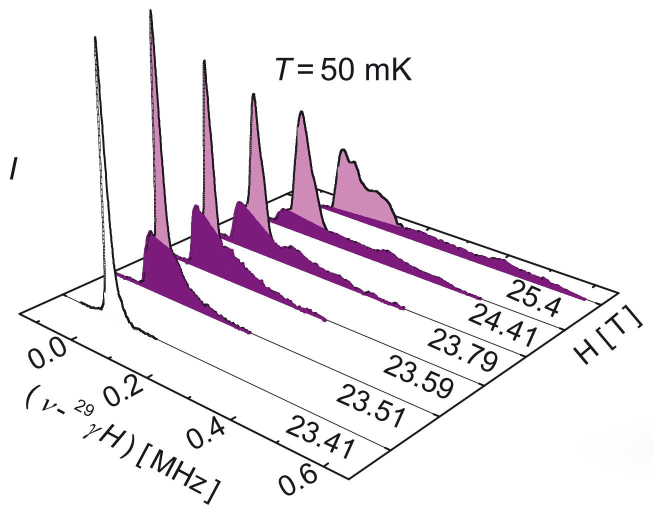


Two types of bilayers

Nuclear magnetic resonance evidence for a strong modulation of the Bose-Einstein condensate in $BaCuSi_2O_6$

S. Krämer,[1] R. Stern,[2] M. Horvatić,[1] C. Berthier,[1] T. Kimura,[3] and I. R. Fisher[4]

PRB **76**, 100406(R) (2007)



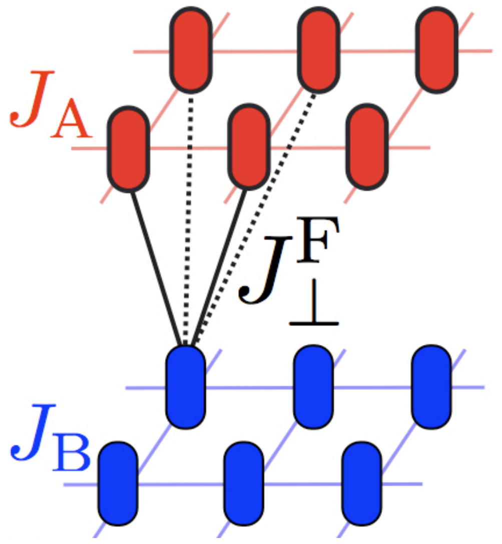
Triplet population strongly modulated along the c-axis

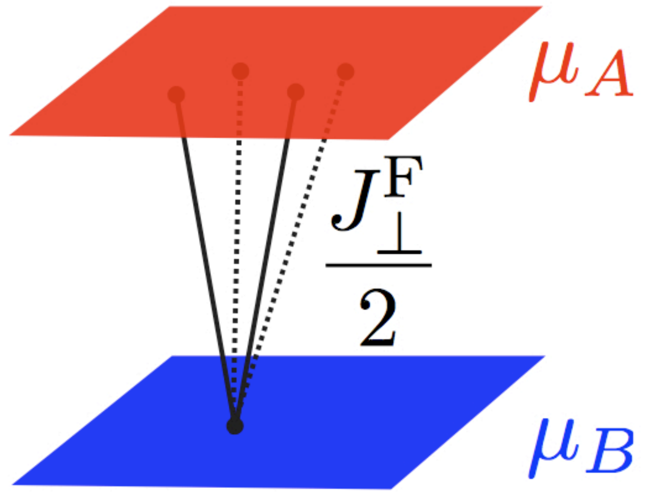
SeasoN Four [2009 - 2011]

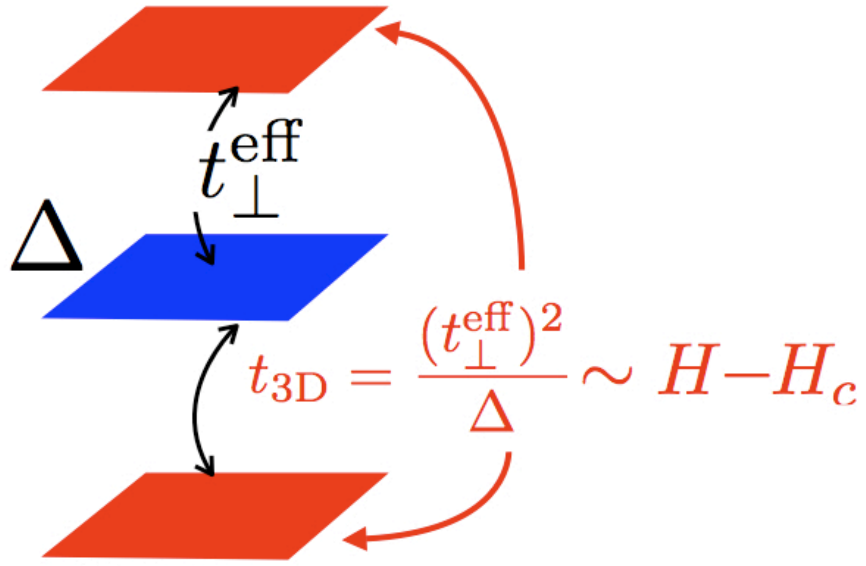
Theory

NL and F. Mila PRL (2009)
NL and F. Mila PRL (2011)

Frustrated inequivalent bilayers → (dimer-boson mapping) → **Frustrated bosonic layers** → (semi-classical expansion) → **Non-frustrated effective bosonic model**



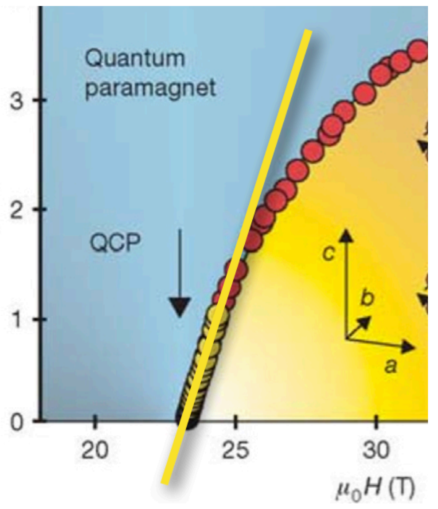




Consequences for the critical temperature

$$T_{BEC} \approx \frac{\hbar^2}{2k_B(m_x m_y m_z)^{1/3}}\left[\kappa(H-H_c)^{2/3}\right]$$

$$\frac{\hbar^2}{2(m_x m_y m_z)^{1/3}} = J_\parallel{}^{2/3} t_{3D}{}^{1/3} \Rightarrow T_{BEC} \sim (H-H_c)^{\frac{2}{3}+\frac{1}{3}}$$



SeasoN Five [2013]

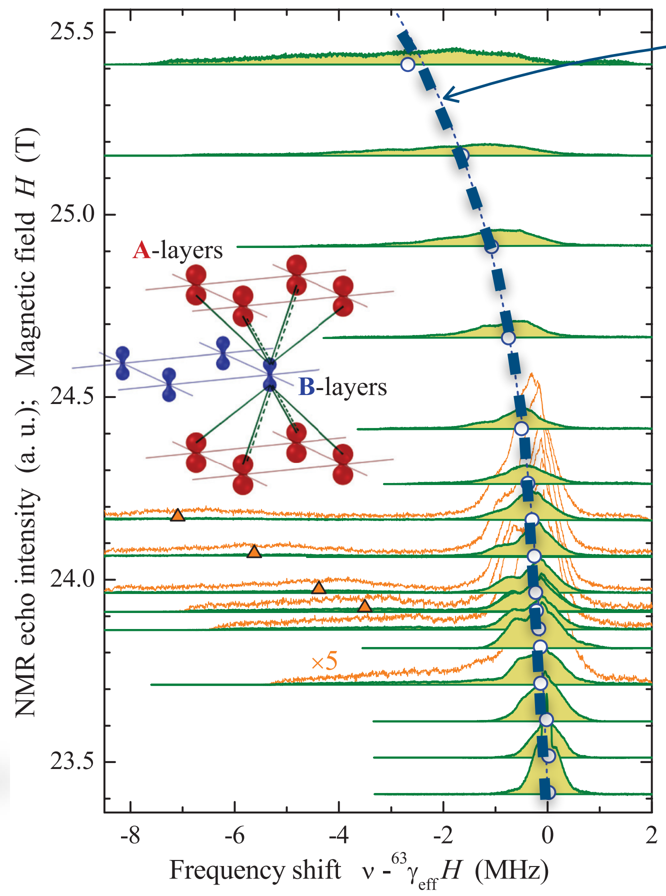
NMR strikes back
The fall of frustration?

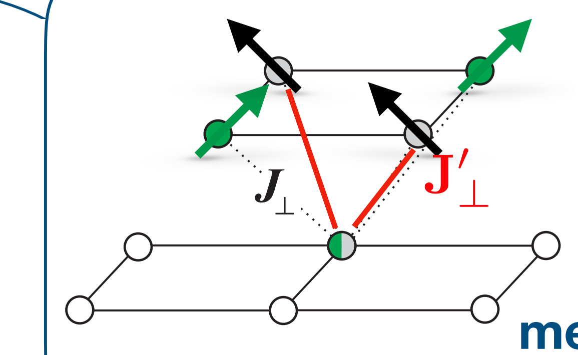
Spatially resolved magnetization in the Bose-Einstein condensed state of $BaCuSi_2O_6$: Evidence for imperfect frustration

S. Krämer,[1,*] N. Laflorencie,[2,†] R. Stern,[3] M. Horvatić,[1] C. Berthier,[1] H. Nakamura,[4] T. Kimura,[4] and F. Mila[5]

PRB **87**, 180405(R) (2013)

Precise determination of B-layer population by NMR





Perfect frustration has to be broken to explain the measured triplet populations!

No perfect frustration in $BaCuSi_2O_6$

See also DFT calculations by Mazurenko *et al*, PRL **112**, 107202 (2014)

Season Six

[2020]

The return of the neutrons

PRL **124**, 177205 (2020)

Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in $BaCuSi_2O_6$

S. Allenspach,[1,2] A. Biffin,[3] U. Stuhr,[3] G. S. Tucker,[3,4] S. Ohira-Kawamura,[5] M. Kofu,[5] D. J. Voneshen,[6] M. Boehm,[7] B. Normand,[1] N. Laflorencie,[8] F. Mila,[4] and Ch. Rüegg[1,2]

Season Six

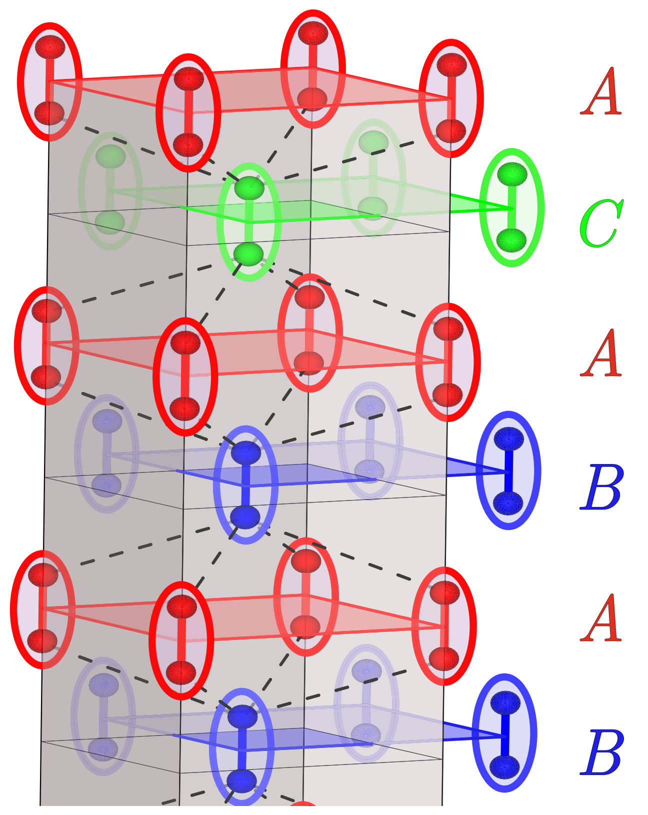
[2020]

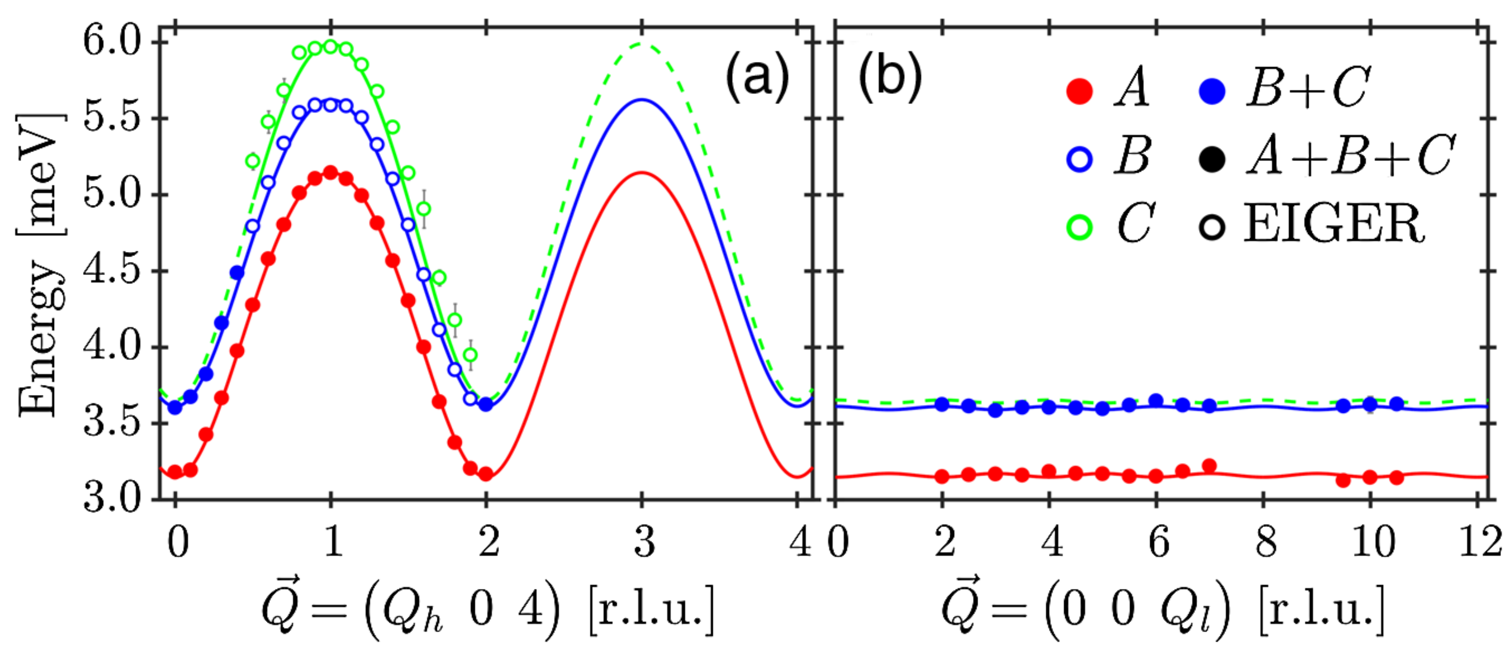
The return of the neutrons

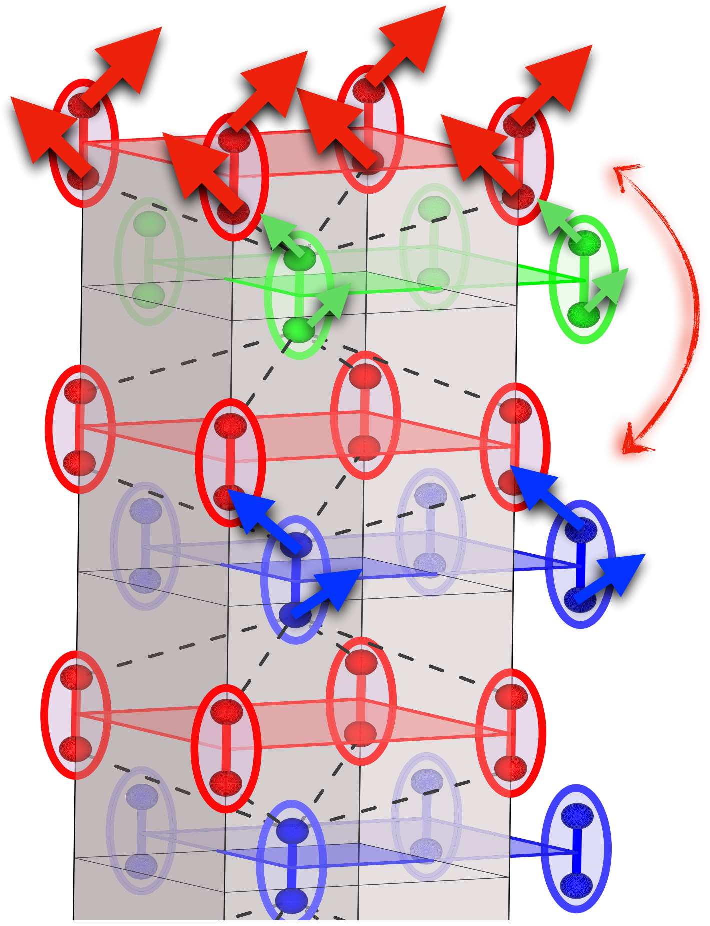
PRL **124**, 177205 (2020)

Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in $BaCuSi_2O_6$

S. Allenspach,[1,2] A. Biffin,[3] U. Stuhr,[3] G. S. Tucker,[3,4] S. Ohira-Kawamura,[5] M. Kofu,[5] D. J. Voneshen,[6] M. Boehm,[7] B. Normand,[1] N. Laflorencie,[8] F. Mila,[4] and Ch. Rüegg[1,2]







In plane $t_{2D} \sim 3\,K$

Effective transverse tunneling

$$t_{3D}^{eff}(h) = t_{3D}^{0} + a_1 h + a_2 h^2 + \cdots$$

At the quantum critical point H_{c1}

$$t_{3D}^{0} \approx \frac{4J_\perp^2}{J_B - J_A} \sim 100\ \mathrm{mK}$$

3 types of bilayers

A – B – A – B – A – C …

$J_A = 4.275(5)$ meV

$J_B = 4.72(1)$ meV

$J_C = 4.95(2)$ meV

Ferromagnetic (effective) intralayer couplings

$J_\parallel^A = -0.480(3)$ meV

$J_\parallel^B = -0.497(8)$ meV

$J_\parallel^C = -0.57(1)$ meV

Small FM interlayer coupling

$J_\perp = -0.04(1)$ meV

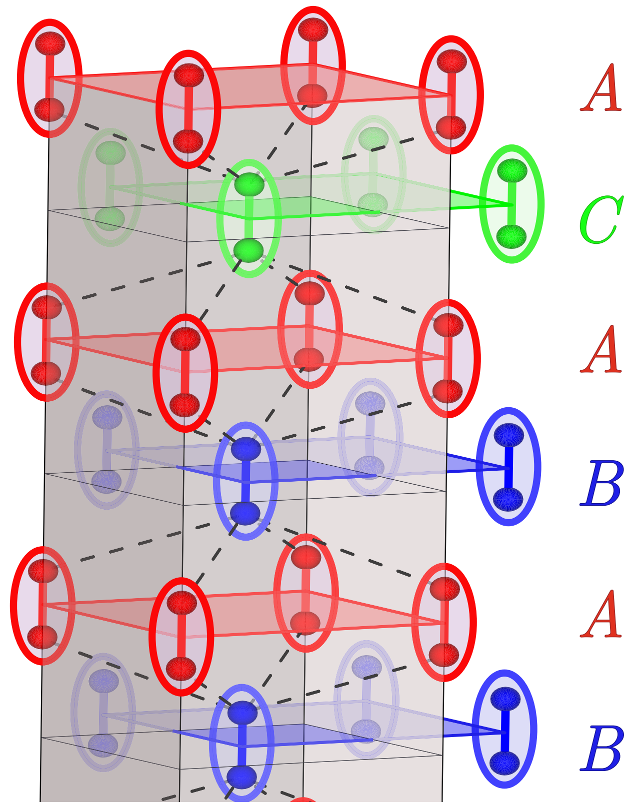
Season Six
[2020]

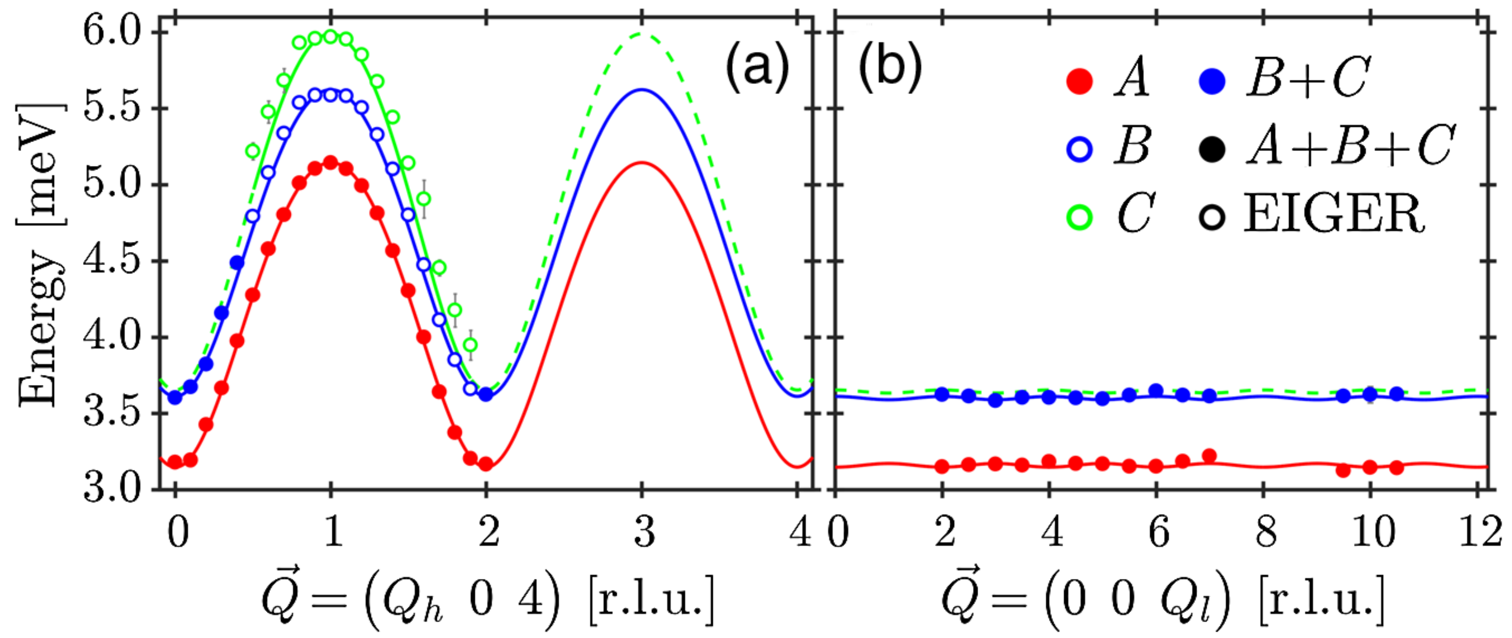
The return of the neutrons

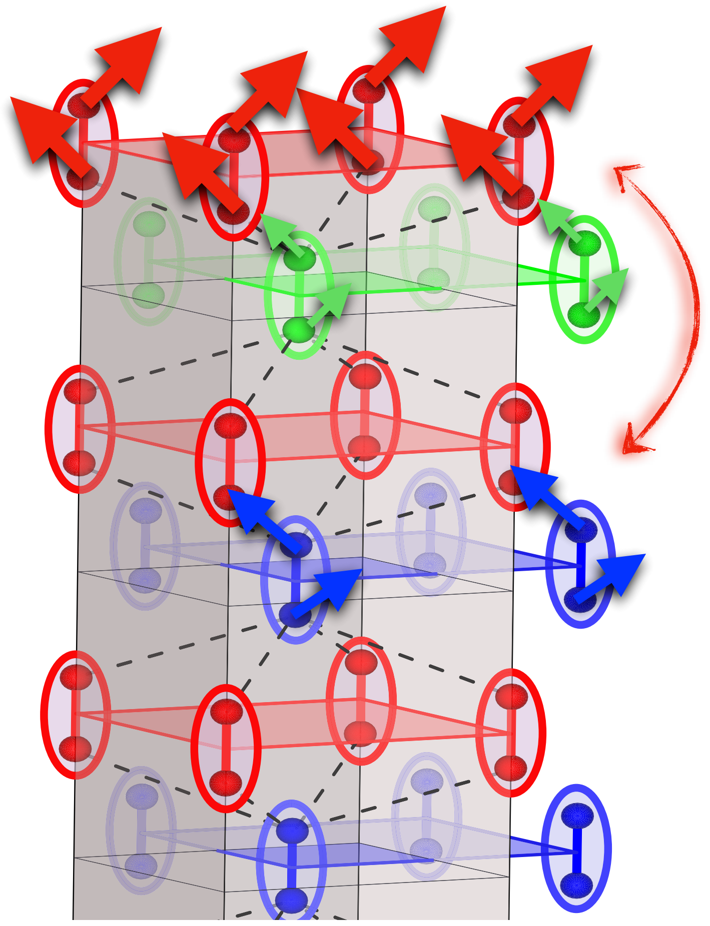
PRL **124**, 177205 (2020)

Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in $BaCuSi_2O_6$

S. Allenspach,[1,2] A. Biffin,[3] U. Stuhr,[3] G. S. Tucker,[3,4] S. Ohira-Kawamura,[5] M. Kofu,[5] D. J. Voneshen,[6] M. Boehm,[7] B. Normand,[1] N. Laflorencie,[8] F. Mila,[4] and Ch. Rüegg[1,2]







In plane $t_{2D} \sim 3\,\mathrm{K}$

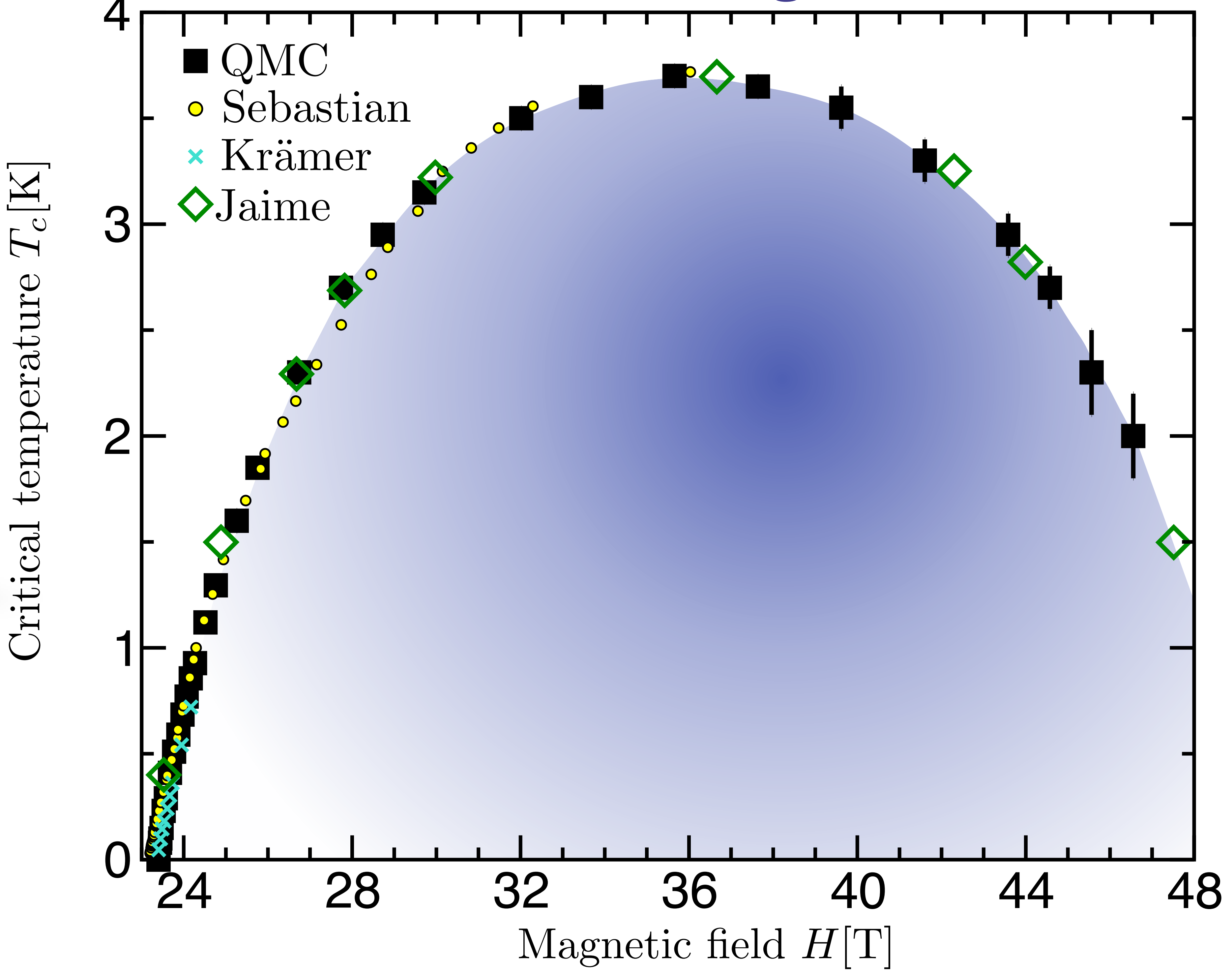
Effective transverse tunneling

$$t_{3D}^{\mathrm{eff}}(h) = t_{3D}^{0} + a_1 h + a_2 h^2 + \cdots$$

At the quantum critical point H_{c1}

$$t_{3D}^{0} \approx \frac{4J_\perp^2}{J_B - J_A} \sim 100\ \mathrm{mK}$$

Phase diagram



3 types of bilayers

A – B – A – B – A – C …

$J_A = 4.275(5)$ meV

$J_B = 4.72(1)$ meV

$J_C = 4.95(2)$ meV

Ferromagnetic (effective) intralayer couplings

$J_\parallel^A = -0.480(3)$ meV

$J_\parallel^B = -0.497(8)$ meV

$J_\parallel^C = -0.57(1)$ meV

Small FM interlayer coupling

$J_\perp = -0.04(1)$ meV

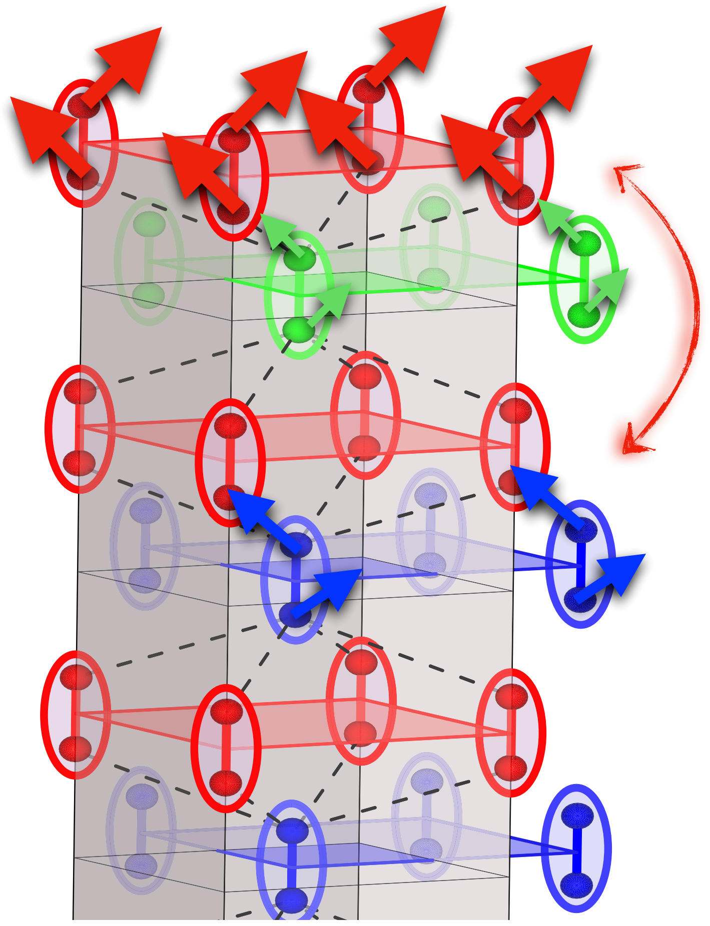
Season Six
[2020]

The return of the neutrons

PRL **124**, 177205 (2020)

Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in $BaCuSi_2O_6$

S. Allenspach,[1,2] A. Biffin,[3] U. Stuhr,[3] G. S. Tucker,[3,4] S. Ohira-Kawamura,[5] M. Kofu,[5] D. J. Voneshen,[6] M. Boehm,[7] B. Normand,[1] N. Laflorencie,[8] F. Mila,[4] and Ch. Rüegg[1,2]



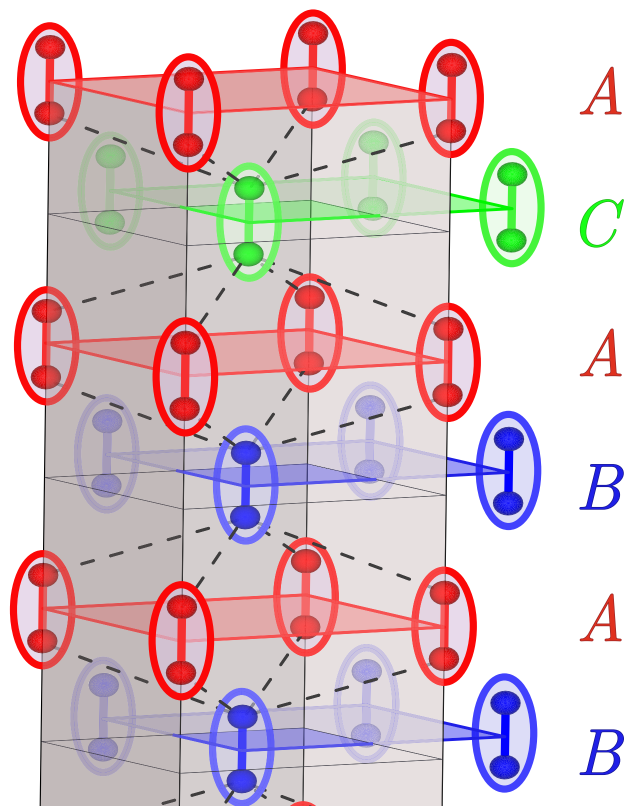
In plane $t_{2D} \sim 3\,\mathrm{K}$

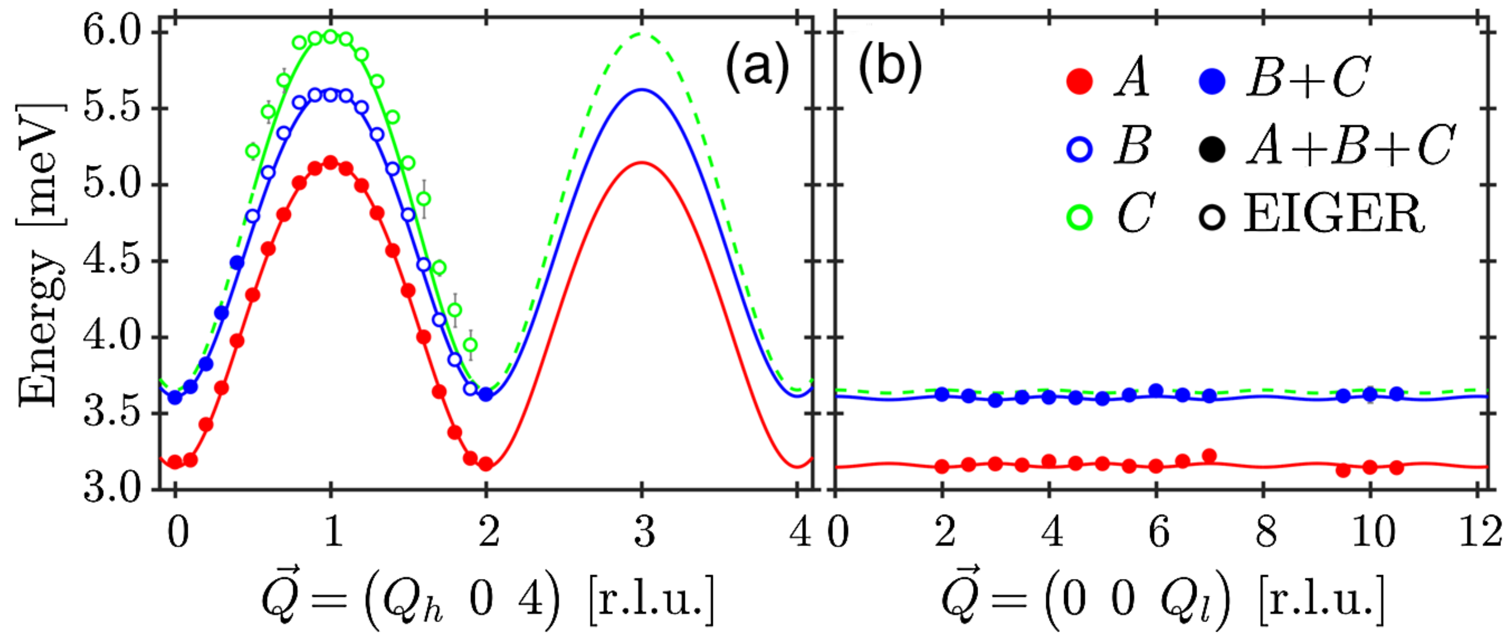
Effective transverse tunneling

$$t_{3D}^{\mathrm{eff}}(h) = t_{3D}^{0} + a_1 h + a_2 h^2 + \cdots$$

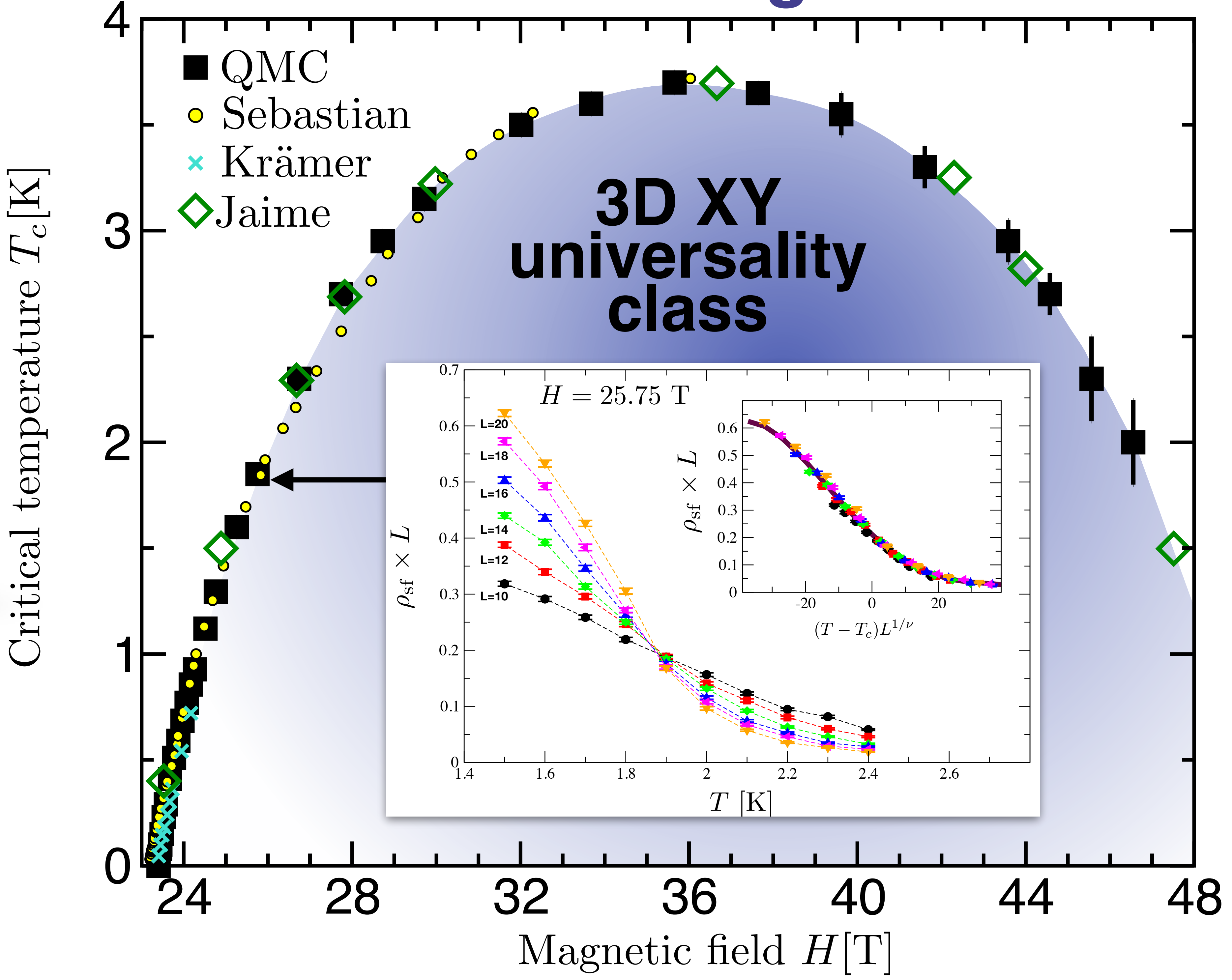
At the quantum critical point H_{c1}

$$t_{3D}^{0} \approx \frac{4J_\perp^2}{J_B - J_A} \sim 100\ \mathrm{mK}$$





Phase diagram



3 types of bilayers

A – B – A – B – A – C …

Ferromagnetic (effective) intralayer couplings

Small FM interlayer coupling

$J_A = 4.275(5)$ meV

$J_B = 4.72(1)$ meV

$J_C = 4.95(2)$ meV

$J_\parallel^A = -0.480(3)$ meV

$J_\parallel^B = -0.497(8)$ meV

$J_\parallel^C = -0.57(1)$ meV

$J_\perp = -0.04(1)$ meV

Season Six

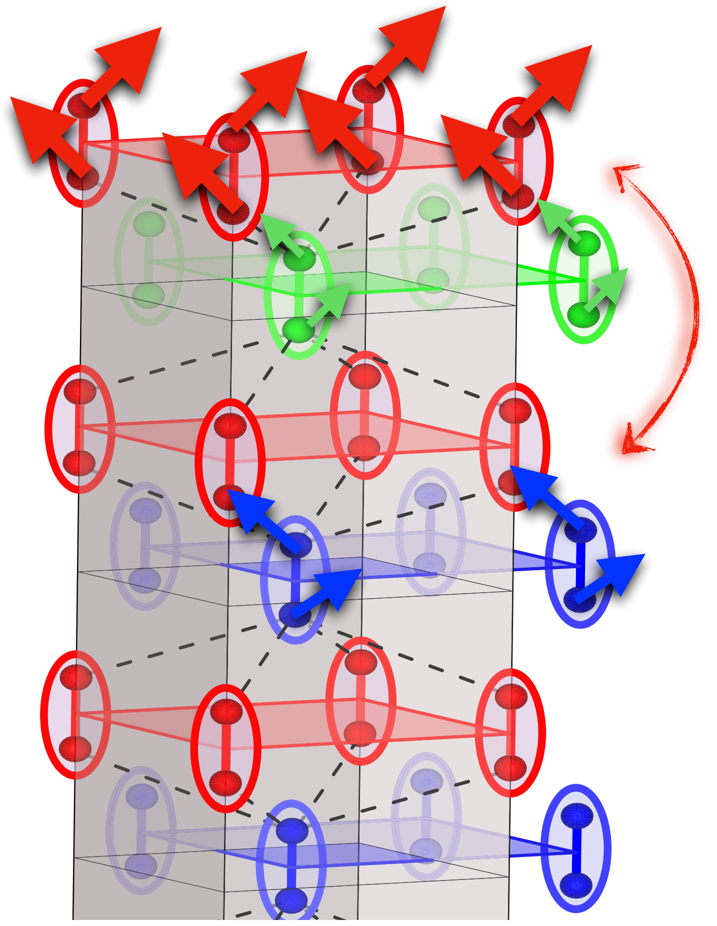
[2020]

The return of the neutrons

PRL **124**, 177205 (2020)

Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in $BaCuSi_2O_6$

S. Allenspach,[1,2] A. Biffin,[3] U. Stuhr,[3] G. S. Tucker,[3,4] S. Ohira-Kawamura,[5] M. Kofu,[5] D. J. Voneshen,[6] M. Boehm,[7] B. Normand,[1] N. Laflorencie,[8] F. Mila,[4] and Ch. Rüegg[1,2]



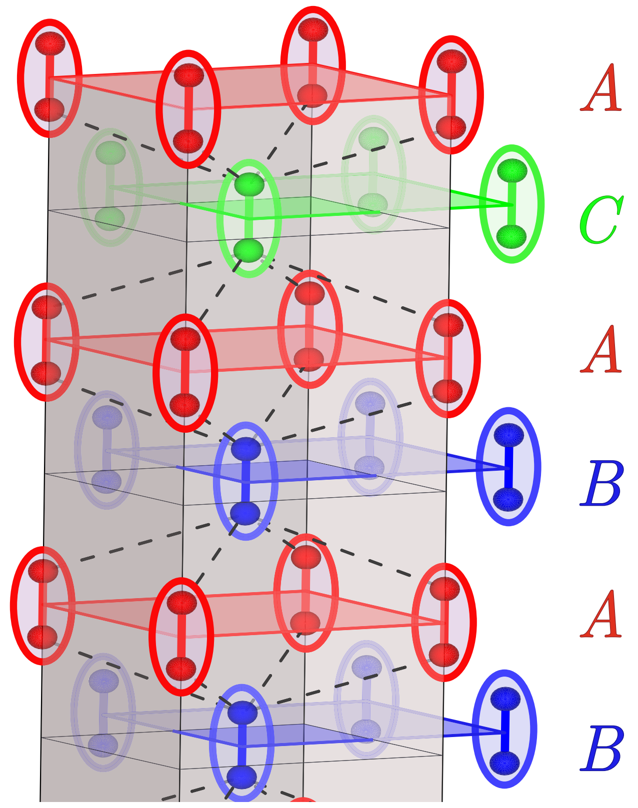
In plane $t_{2D} \sim 3\,\mathrm{K}$

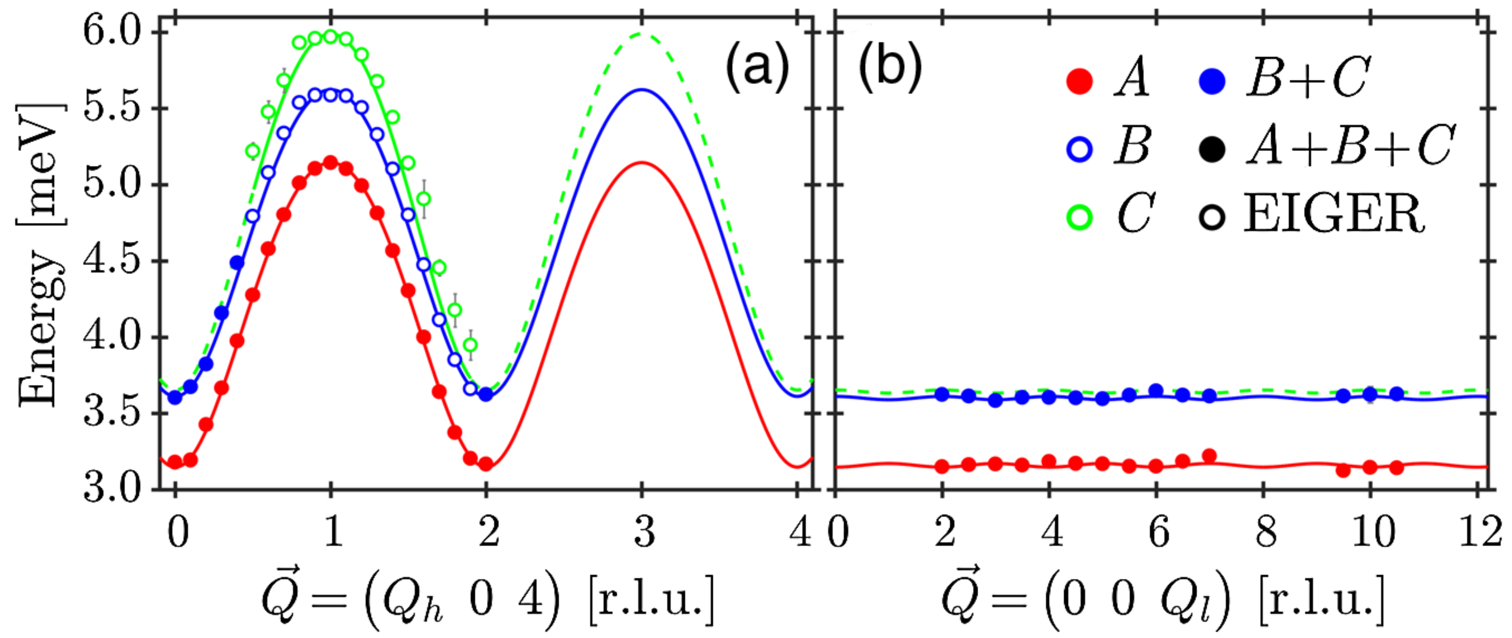
Effective transverse tunneling

$$t_{3D}^{\mathrm{eff}}(h) = t_{3D}^{0} + a_1 h + a_2 h^2 + \cdots$$

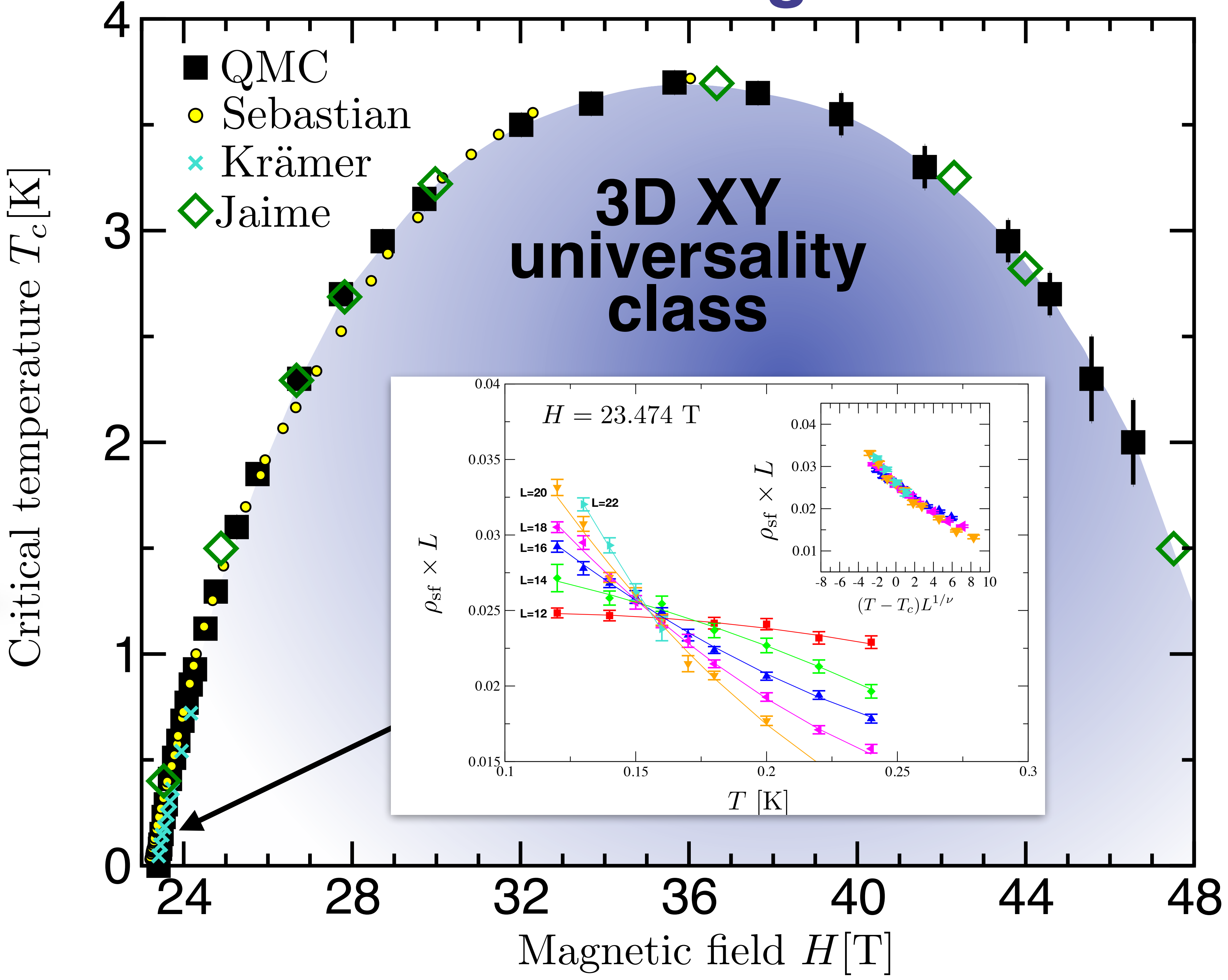
At the quantum critical point H_{c1}

$$t_{3D}^{0} \approx \frac{4J_\perp^2}{J_B - J_A} \sim 100\ \mathrm{mK}$$





Phase diagram



3 types of bilayers

A – B – A – B – A – C …

$J_A = 4.275(5)$ meV

$J_B = 4.72(1)$ meV

$J_C = 4.95(2)$ meV

Ferromagnetic (effective) intralayer couplings

$J_\parallel^A = -0.480(3)$ meV

$J_\parallel^B = -0.497(8)$ meV

$J_\parallel^C = -0.57(1)$ meV

Small FM interlayer coupling

$J_\perp = -0.04(1)$ meV

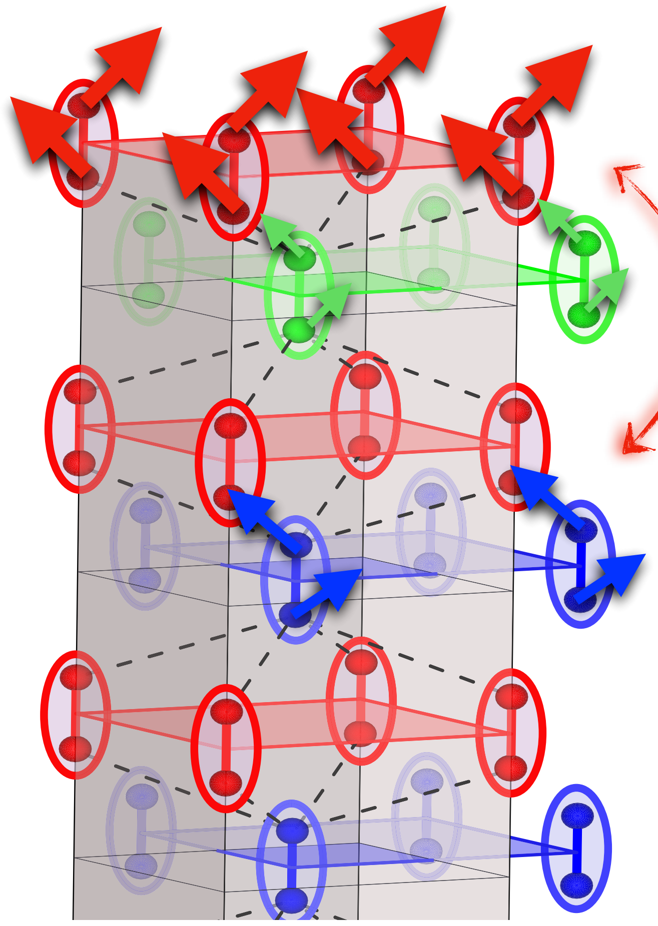
Season Six
[2020]

The return of the neutrons

PRL **124**, 177205 (2020)

Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in $BaCuSi_2O_6$

S. Allenspach,[1,2] A. Biffin,[3] U. Stuhr,[3] G. S. Tucker,[3,4] S. Ohira-Kawamura,[5] M. Kofu,[5] D. J. Voneshen,[6] M. Boehm,[7] B. Normand,[1] N. Laflorencie,[8] F. Mila,[4] and Ch. Rüegg[1,2]



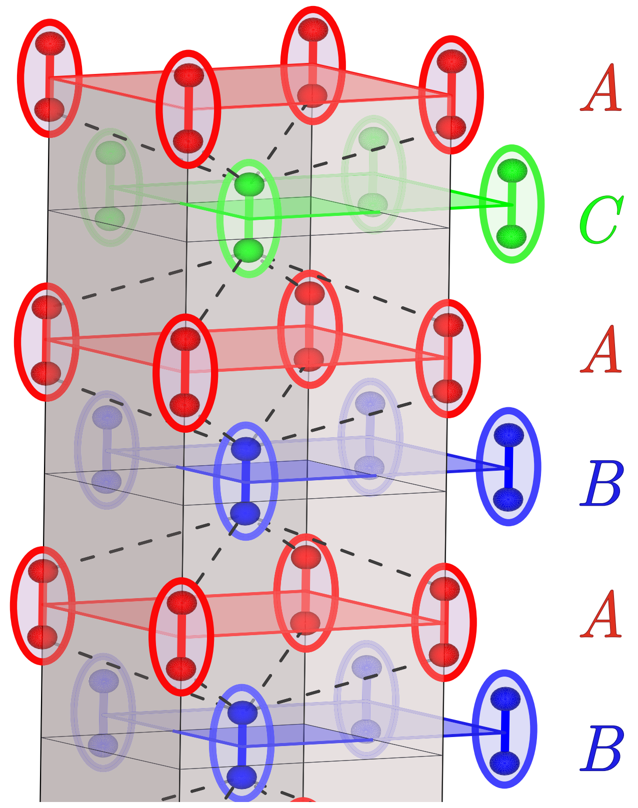
In plane $t_{2D} \sim 3\,\mathrm{K}$

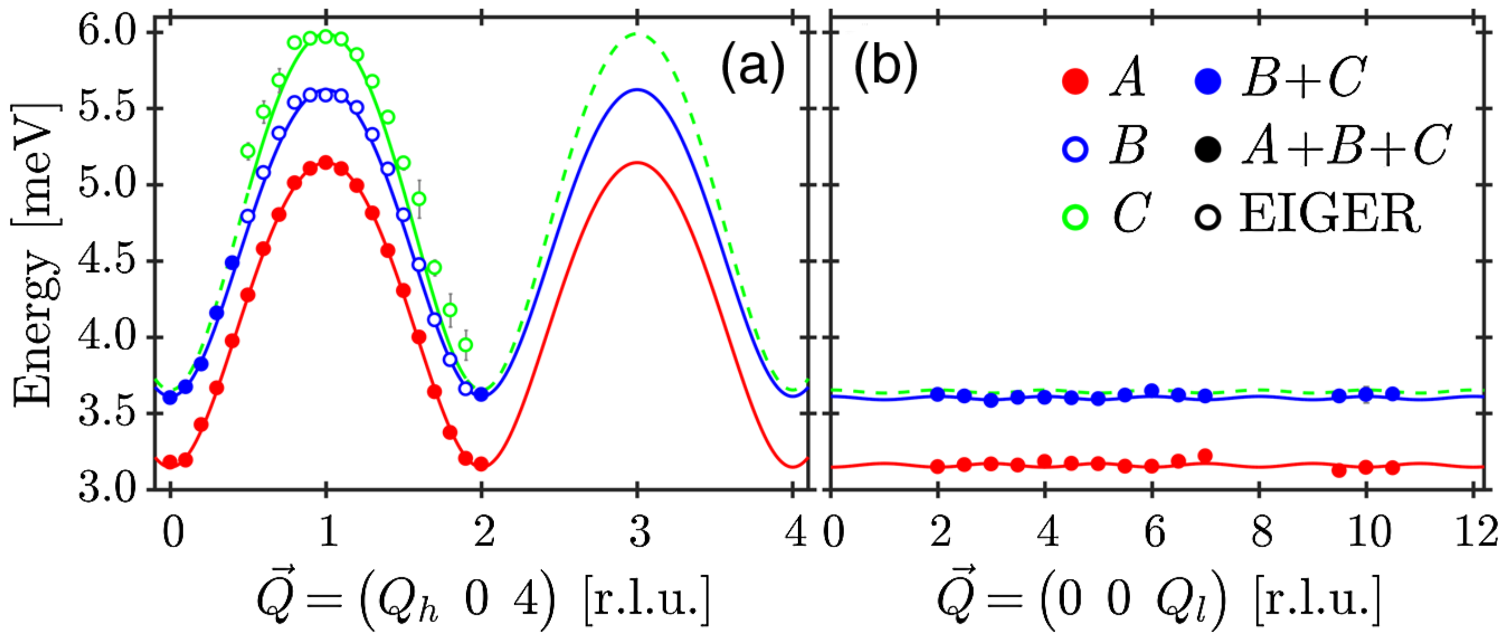
Effective transverse tunneling

$$t_{3D}^{\mathrm{eff}}(h) = t_{3D}^{0} + a_1 h + a_2 h^2 + \cdots$$

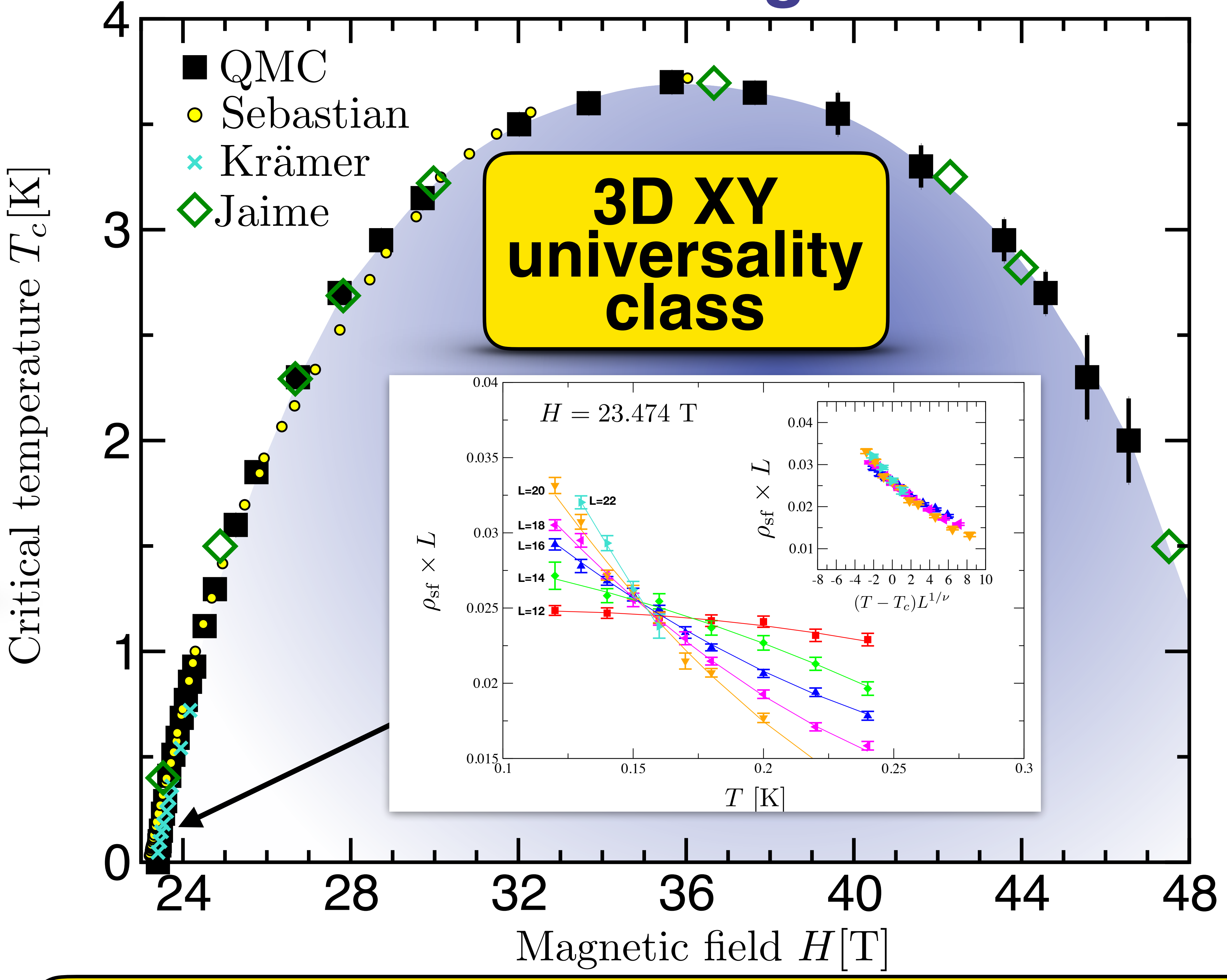
At the quantum critical point H_{c1}

$$t_{3D}^{0} \approx \frac{4J_\perp^2}{J_B - J_A} \sim 100\ \mathrm{mK}$$





Phase diagram



3 types of bilayers

A – B – A – B – A – C …

$J_A = 4.275(5)$ meV

$J_B = 4.72(1)$ meV

$J_C = 4.95(2)$ meV

Ferromagnetic (effective) intralayer couplings

$J_\parallel^A = -0.480(3)$ meV

$J_\parallel^B = -0.497(8)$ meV

$J_\parallel^C = -0.57(1)$ meV

Small FM interlayer coupling

$J_\perp = -0.04(1)$ meV

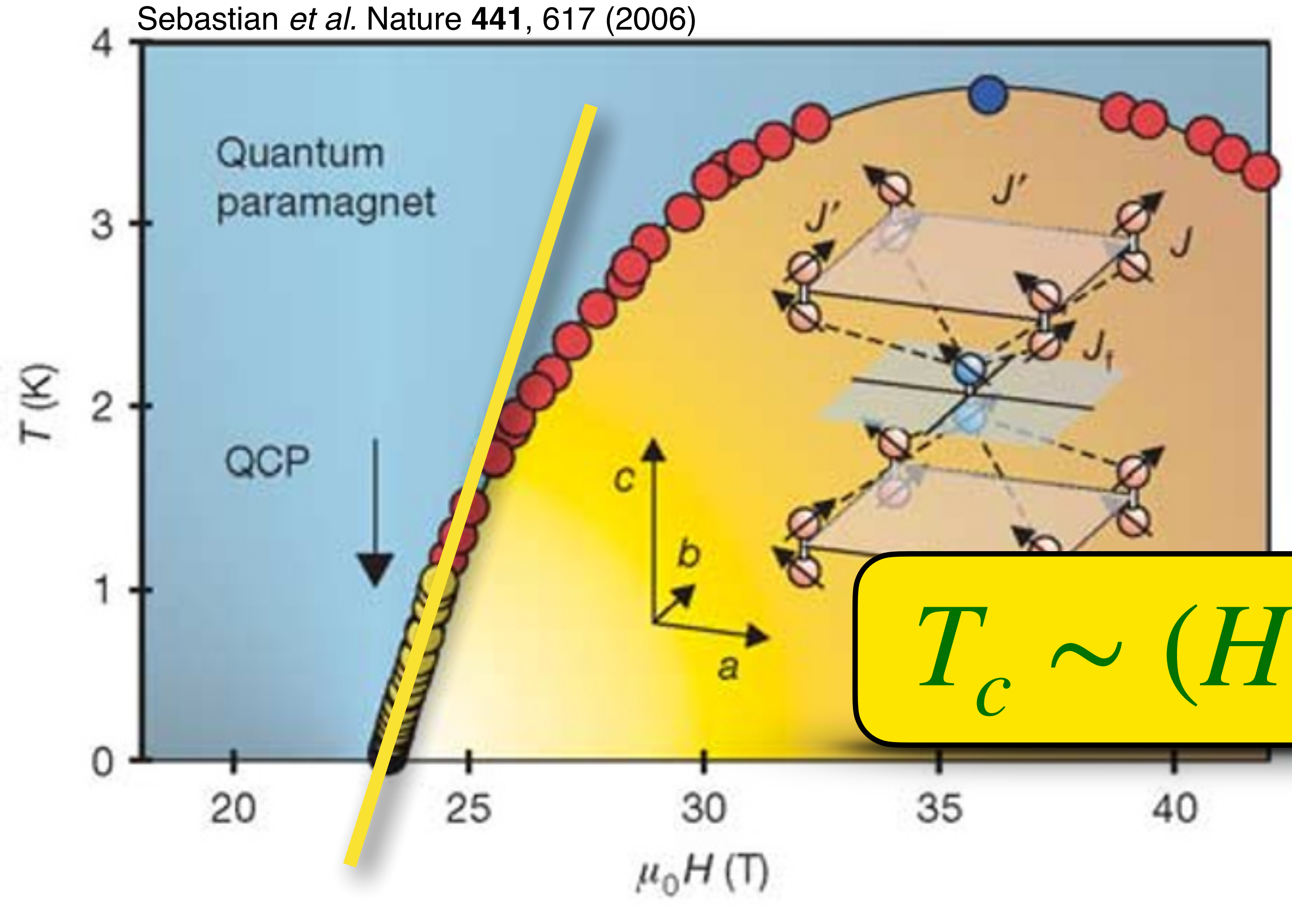
Overall excellent agreement QMC *vs.* Experiments

BonuS

BonuS

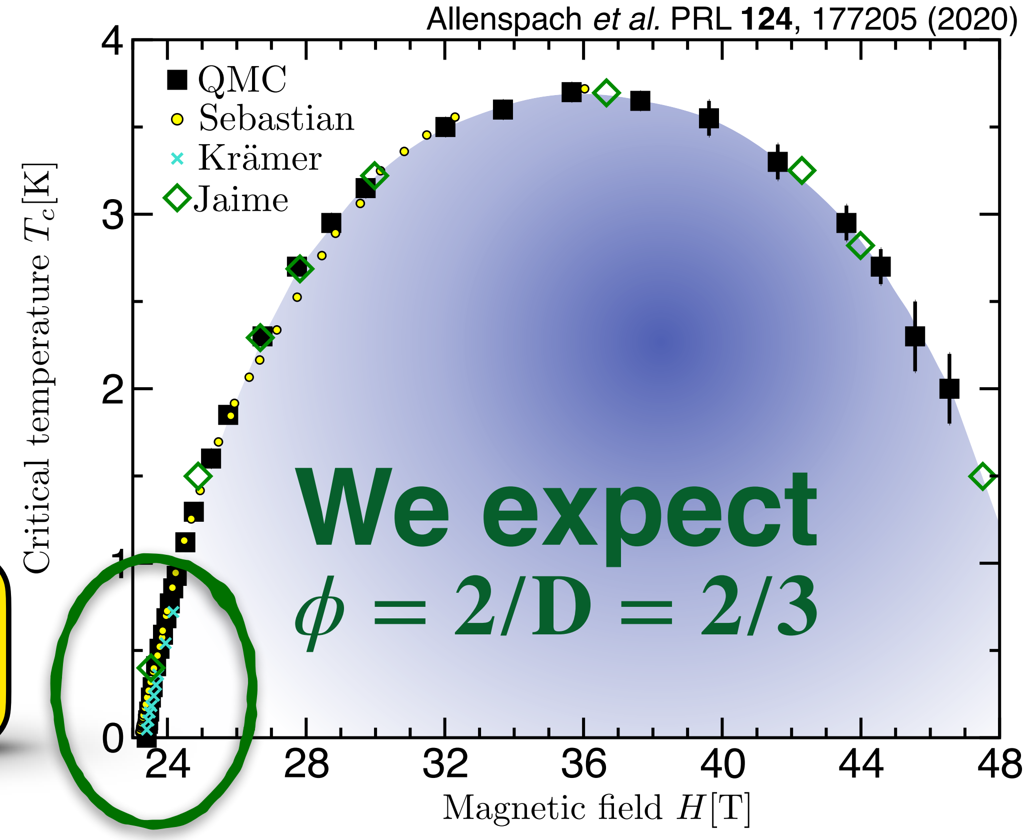
End of the ϕ crisis?

Sebastian *et al.* Nature **441**, 617 (2006)



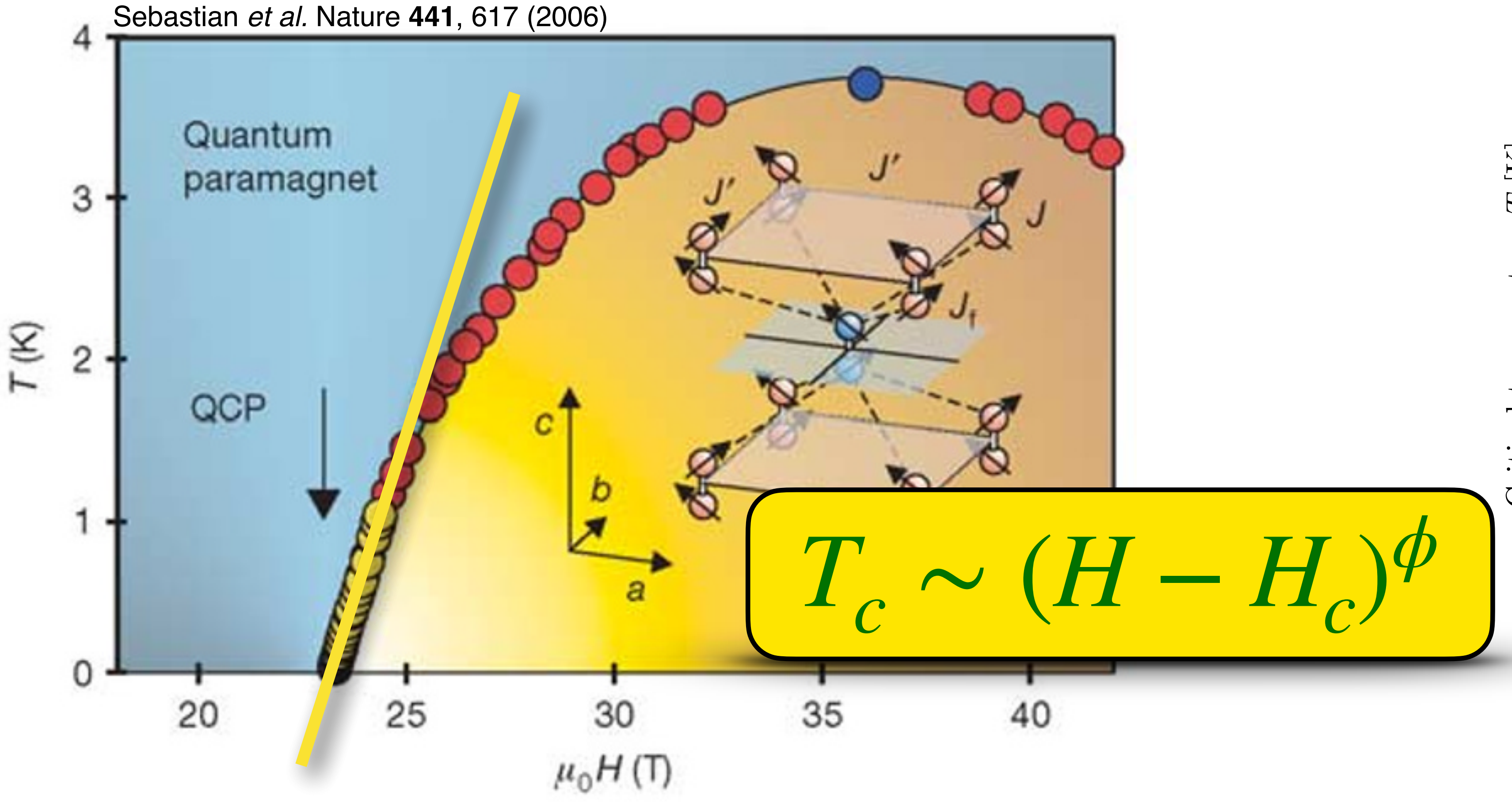
$$T_c \sim (H - H_c)^{\phi}$$

Allenspach *et al.* PRL **124**, 177205 (2020)

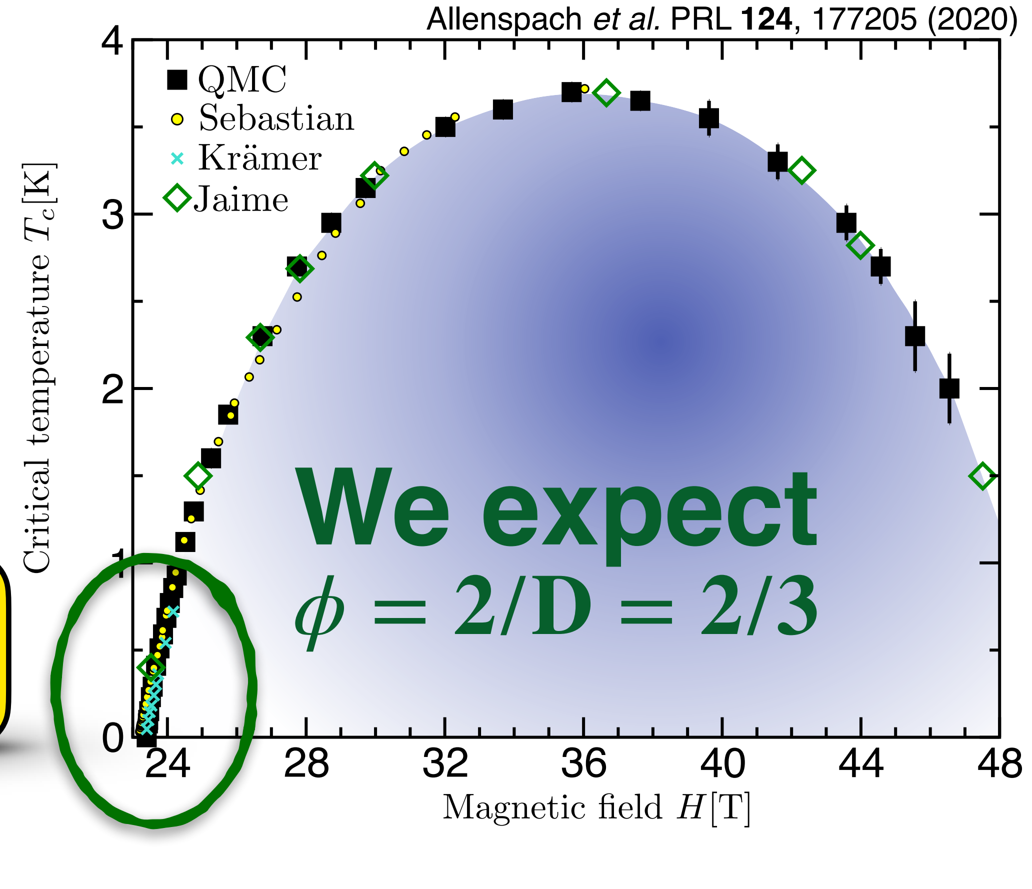


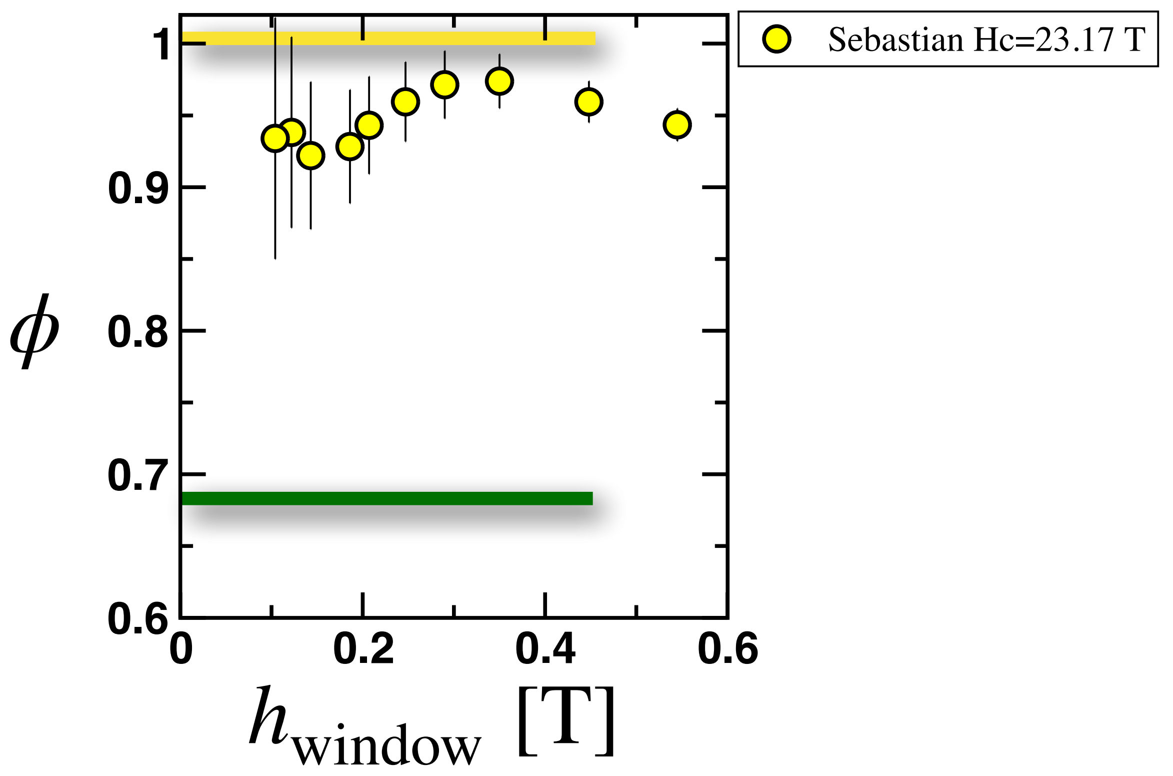
End of the ϕ crisis?

Sebastian *et al.* Nature **441**, 617 (2006)



Allenspach *et al.* PRL **124**, 177205 (2020)

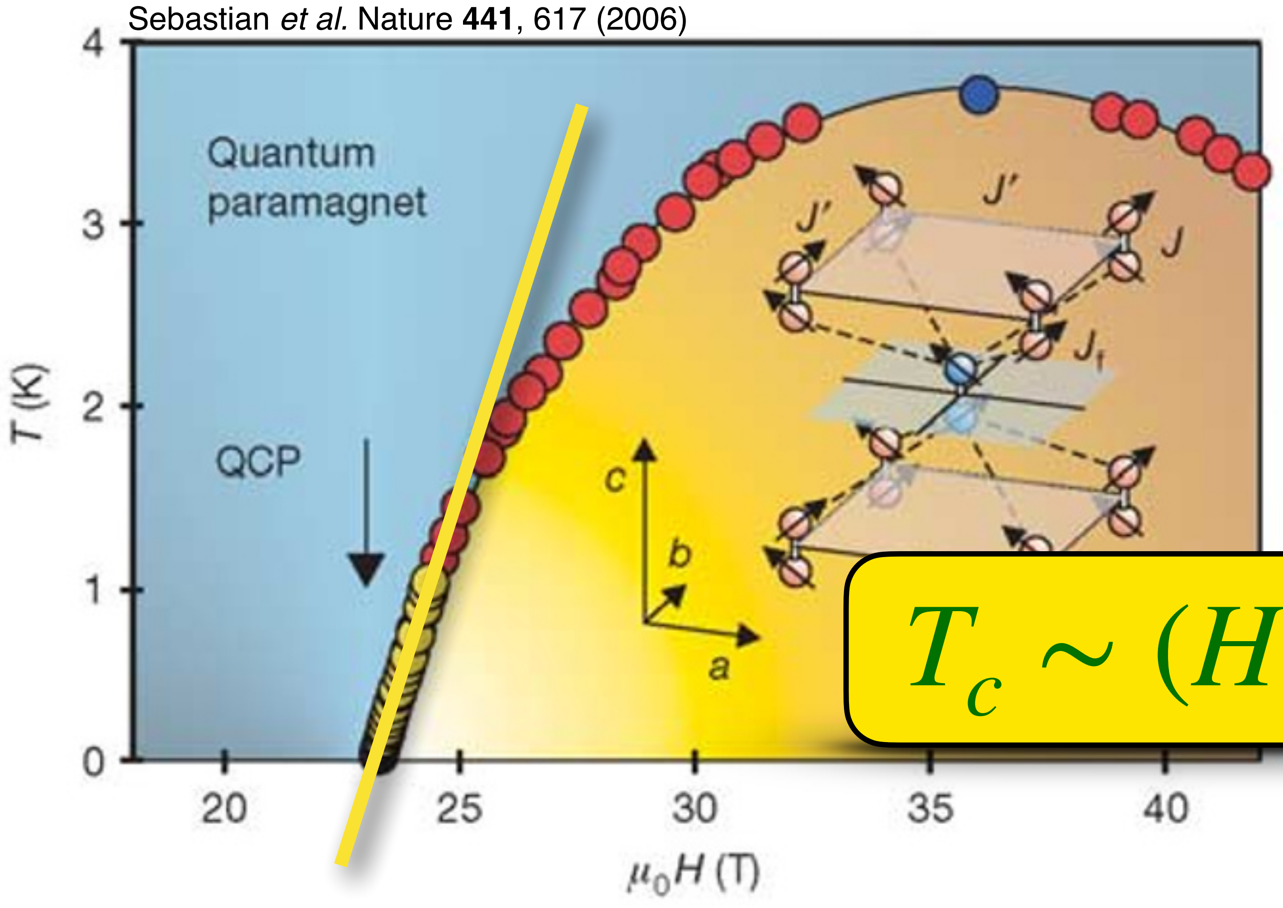




BonuS

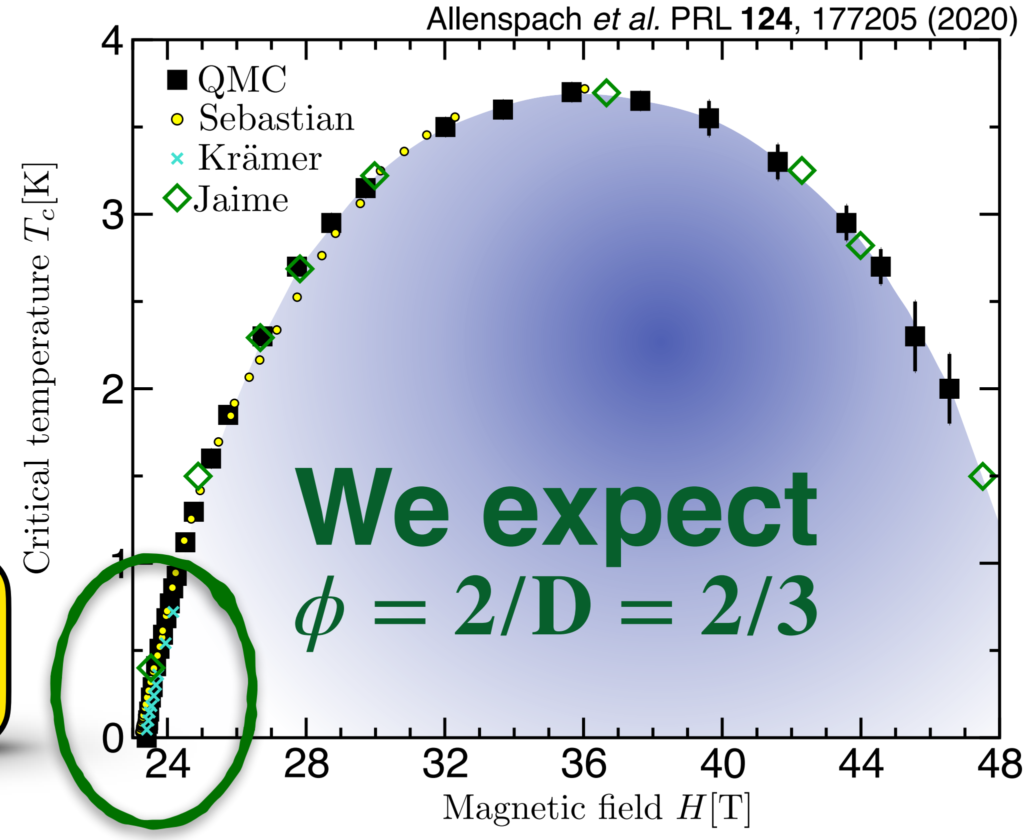
End of the ϕ crisis?

Sebastian *et al.* Nature **441**, 617 (2006)

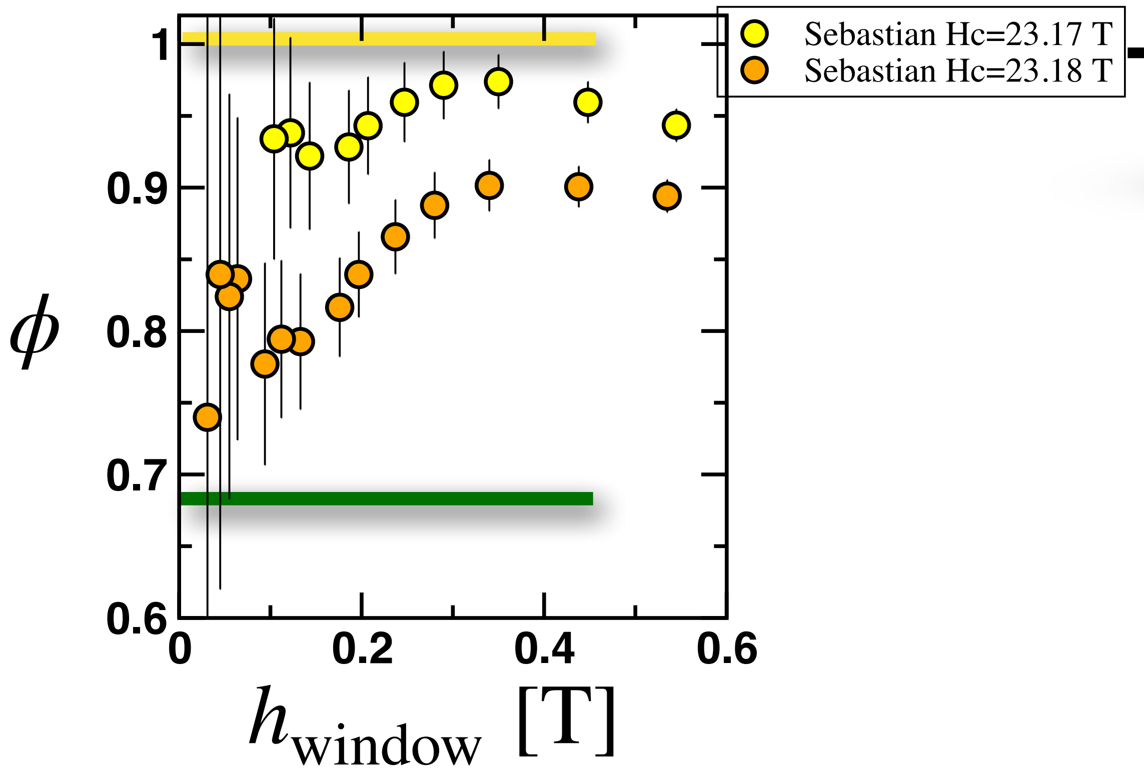


$T_c \sim (H - H_c)^{\phi}$

Allenspach *et al.* PRL **124**, 177205 (2020)



We expect
$\phi = 2/D = 2/3$

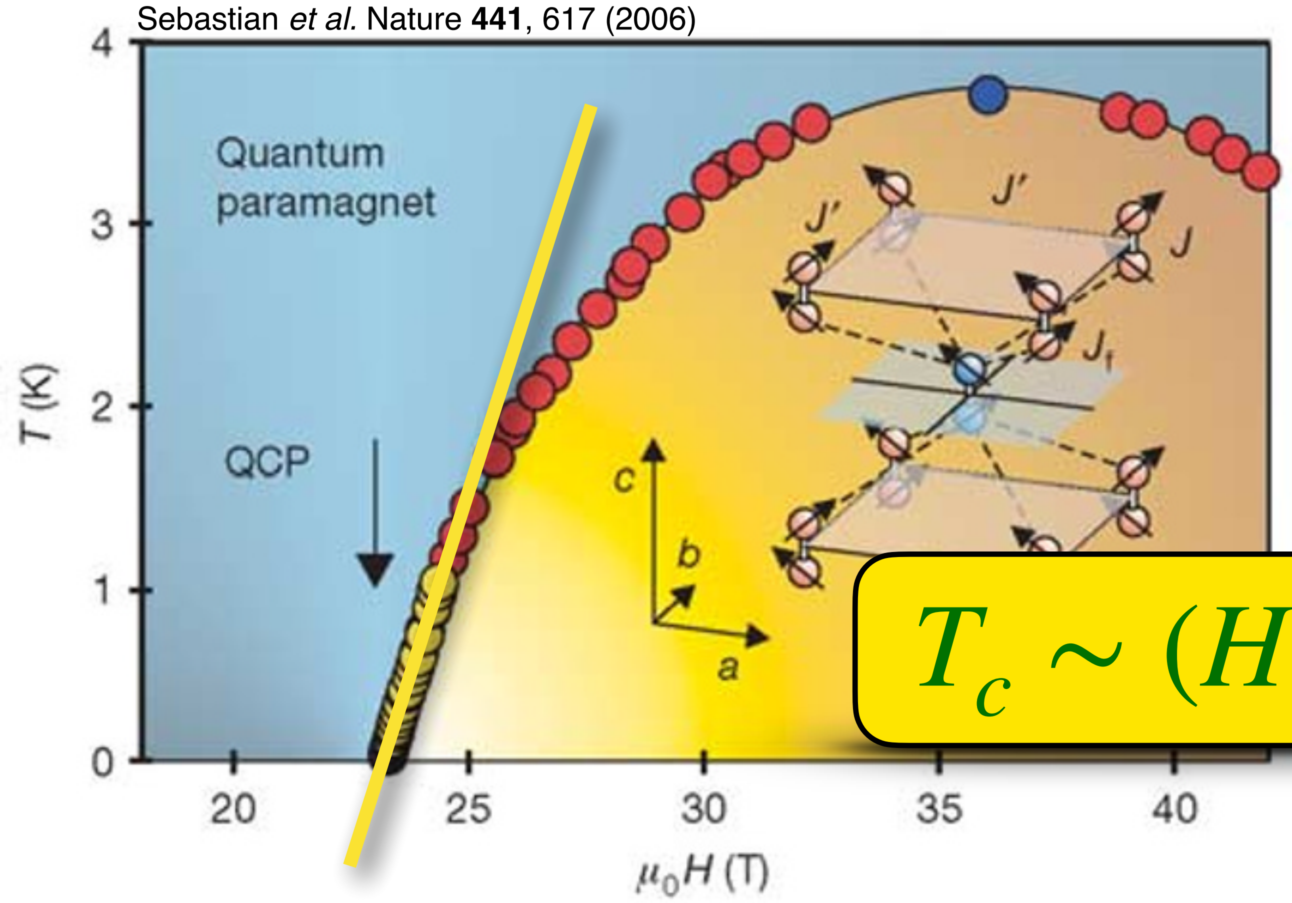


extreme sensitivity to the critical field!

BonuS

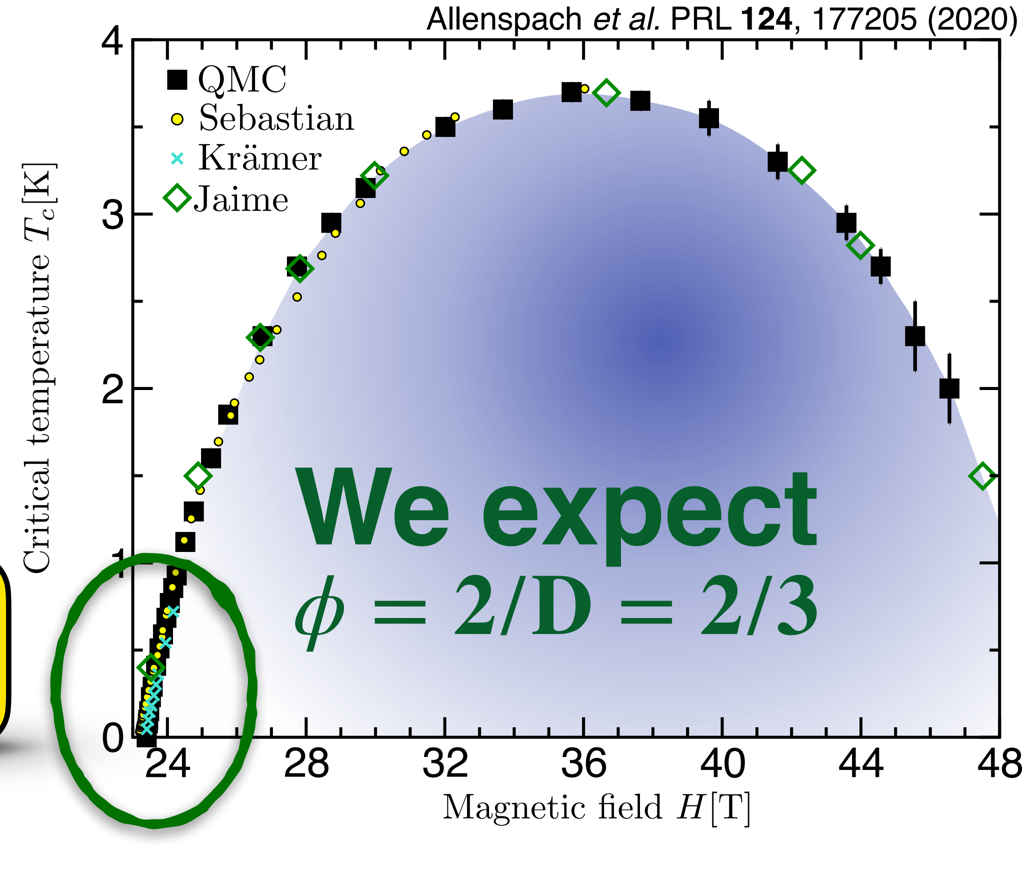
End of the ϕ crisis?

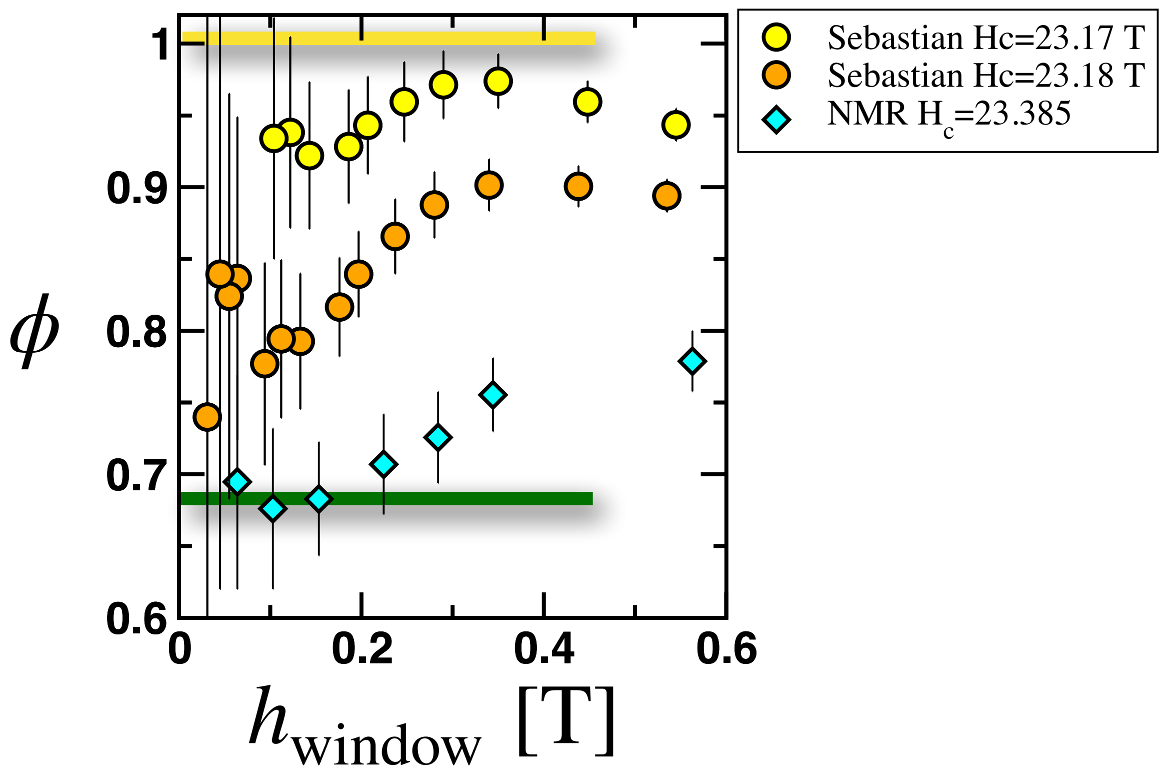
Sebastian *et al.* Nature **441**, 617 (2006)



$T_c \sim (H - H_c)^{\phi}$

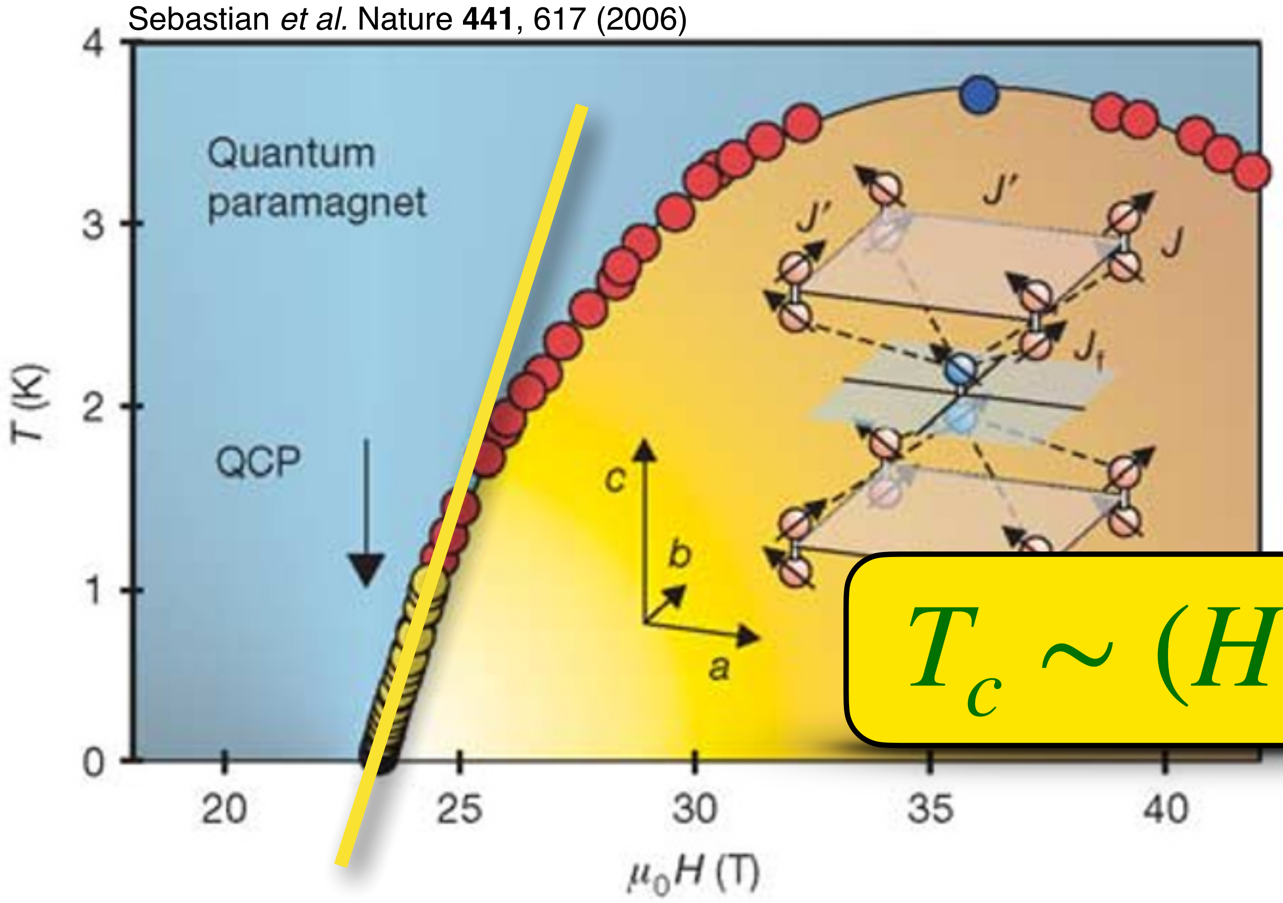
Allenspach *et al.* PRL **124**, 177205 (2020)





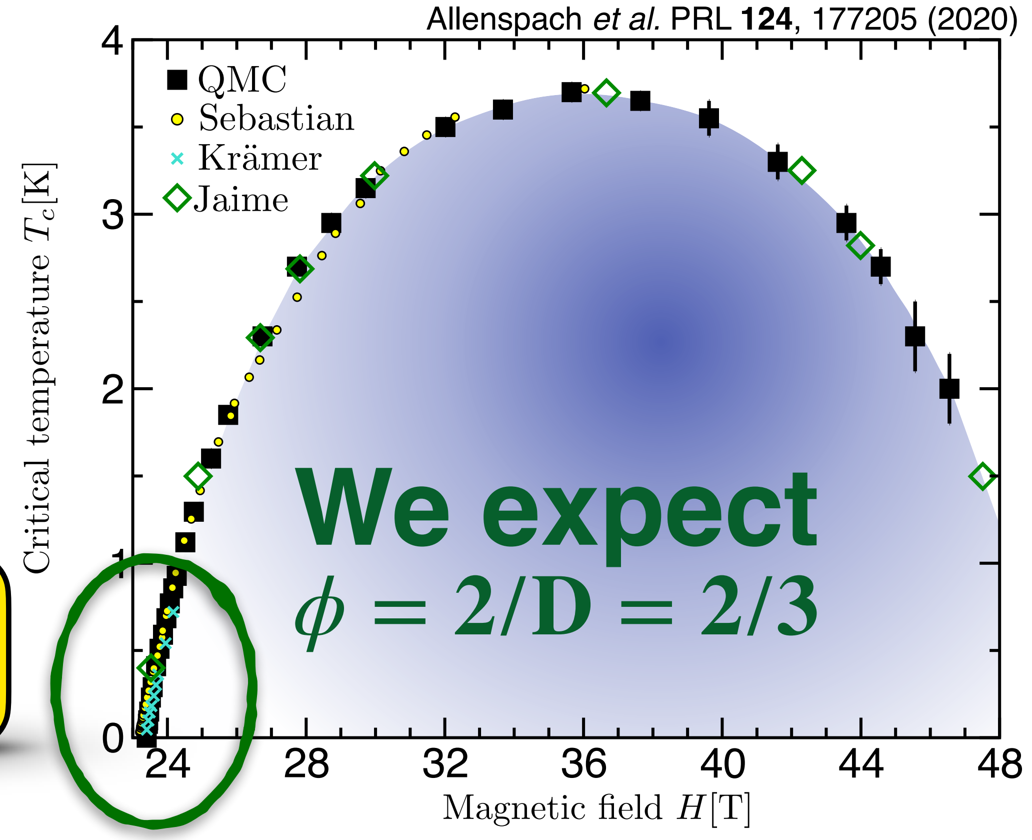
End of the ϕ crisis?

Sebastian *et al.* Nature **441**, 617 (2006)

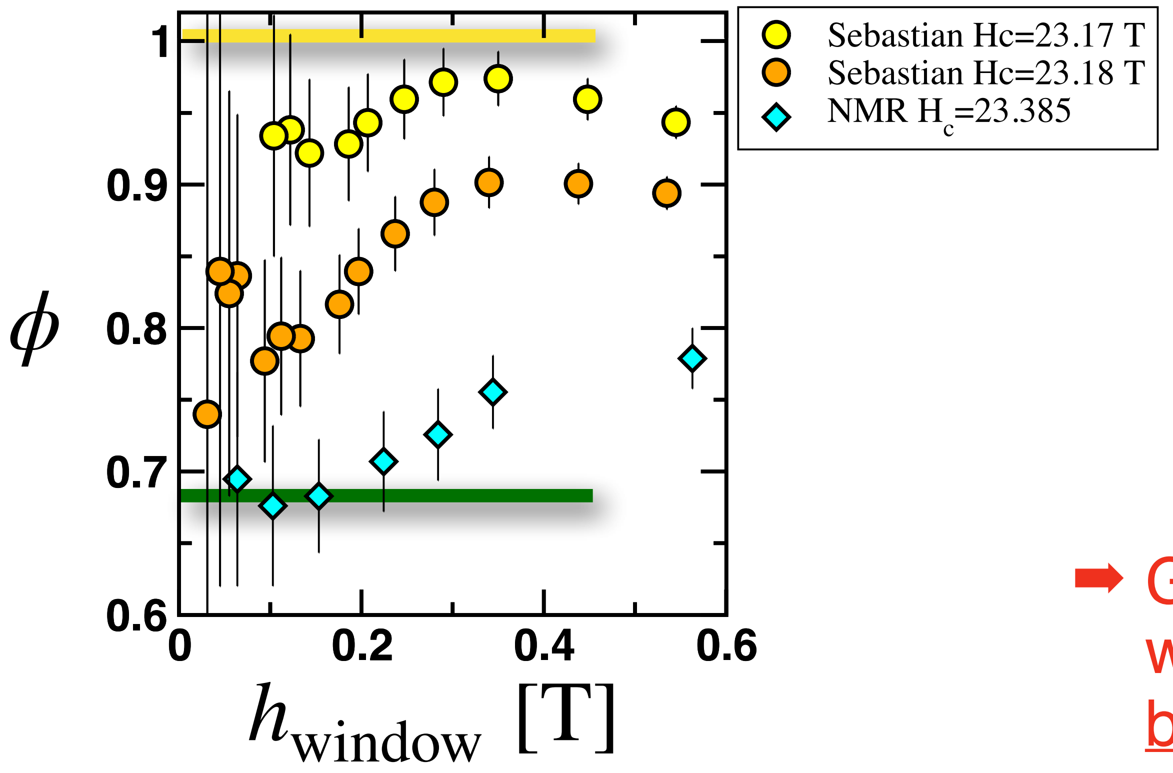


$T_c \sim (H - H_c)^{\phi}$

Allenspach *et al.* PRL **124**, 177205 (2020)



We expect
$\phi = 2/D = 2/3$



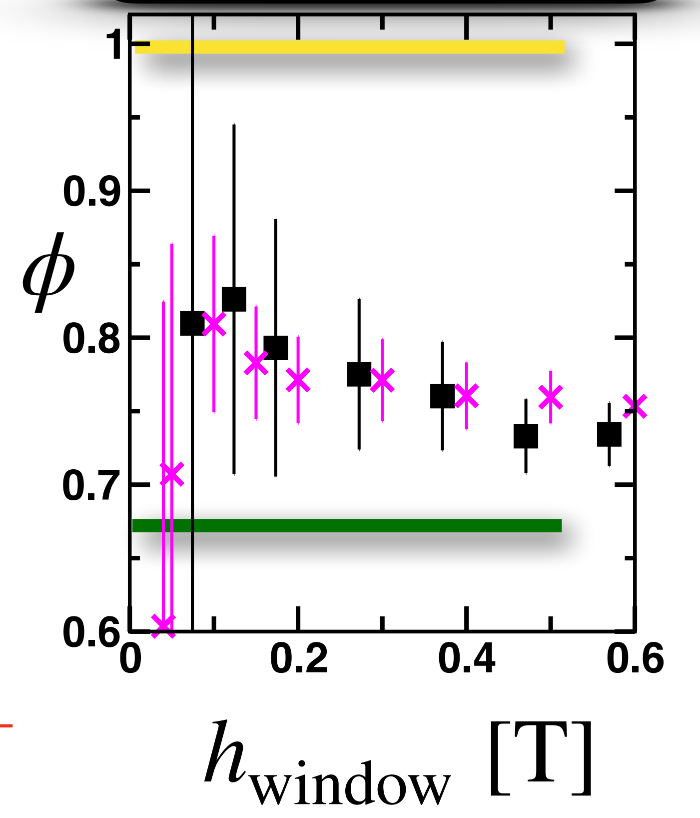
$$t^0_{3D} \approx \frac{4J_\perp^2}{J_B - J_A} \sim 100\ \text{mK}$$

$$t_{2D}/t^0_{3D} \approx 30$$

➡ Genuine 3D BEC criticality with $\phi = 2/3$ observed only below 0.05 T ($\sim$ 100 mK)

➡ Above, ϕ is non-universal

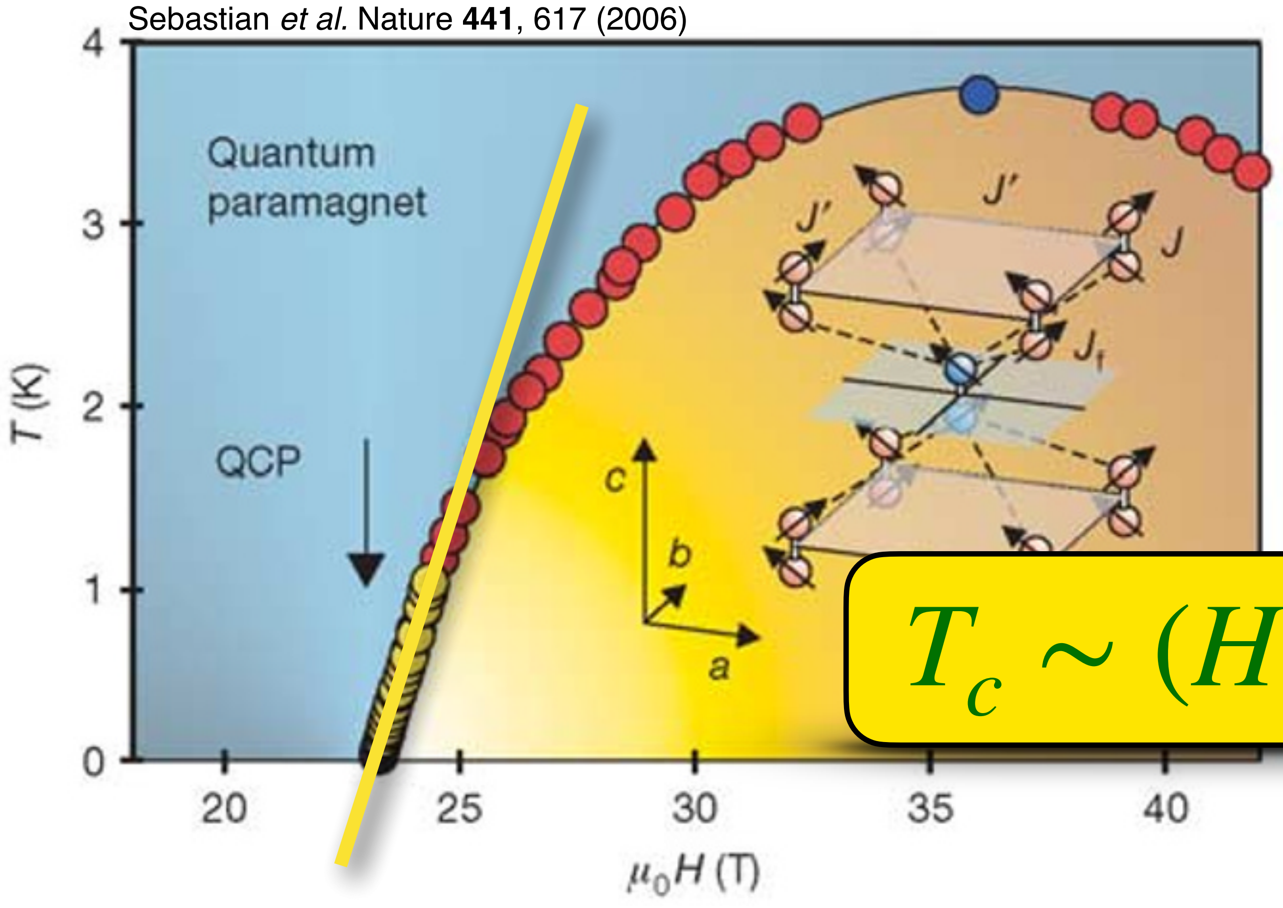
QMC estimate



Bonus

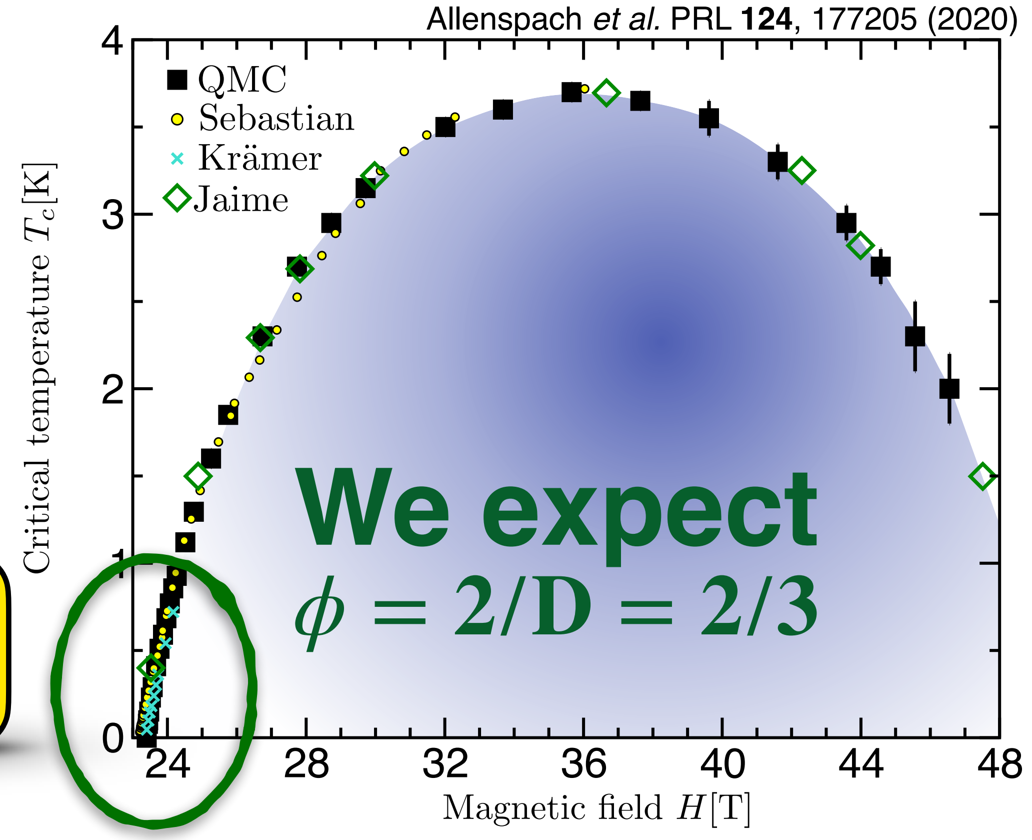
End of the ϕ crisis?

Sebastian *et al.* Nature **441**, 617 (2006)

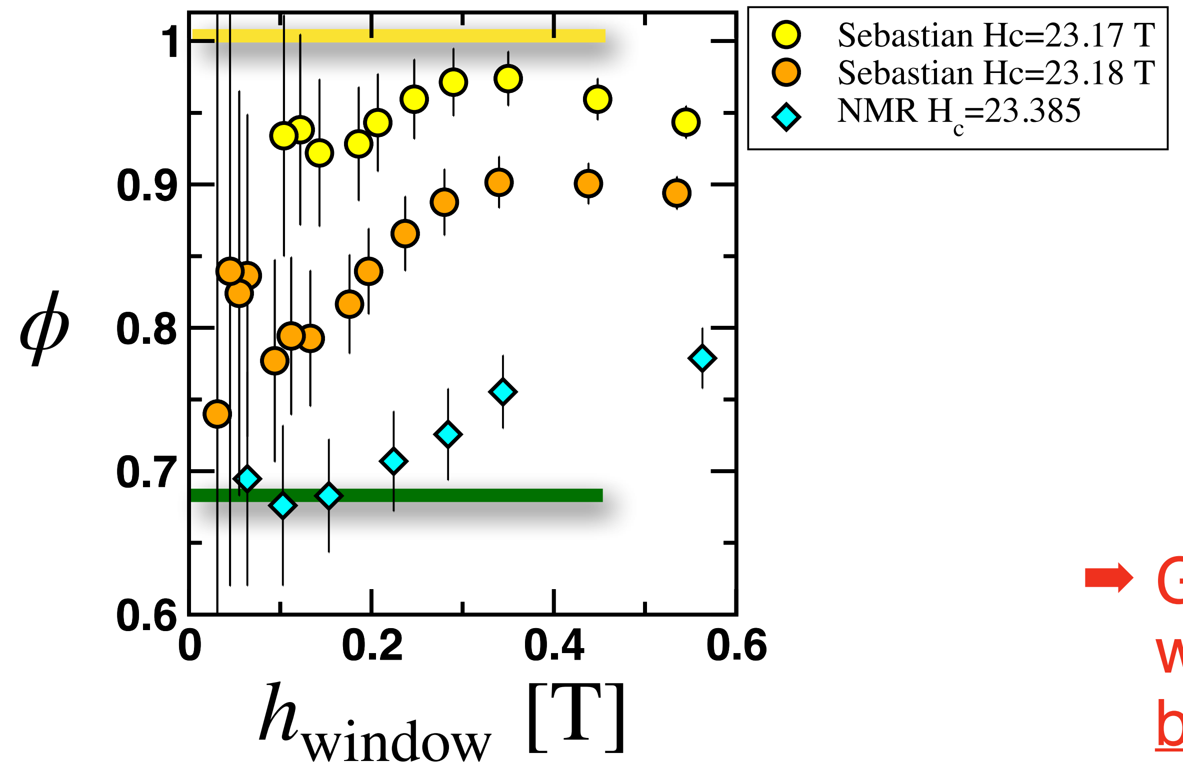


$T_c \sim (H - H_c)^{\phi}$

Allenspach *et al.* PRL **124**, 177205 (2020)



We expect
$\phi = 2/D = 2/3$



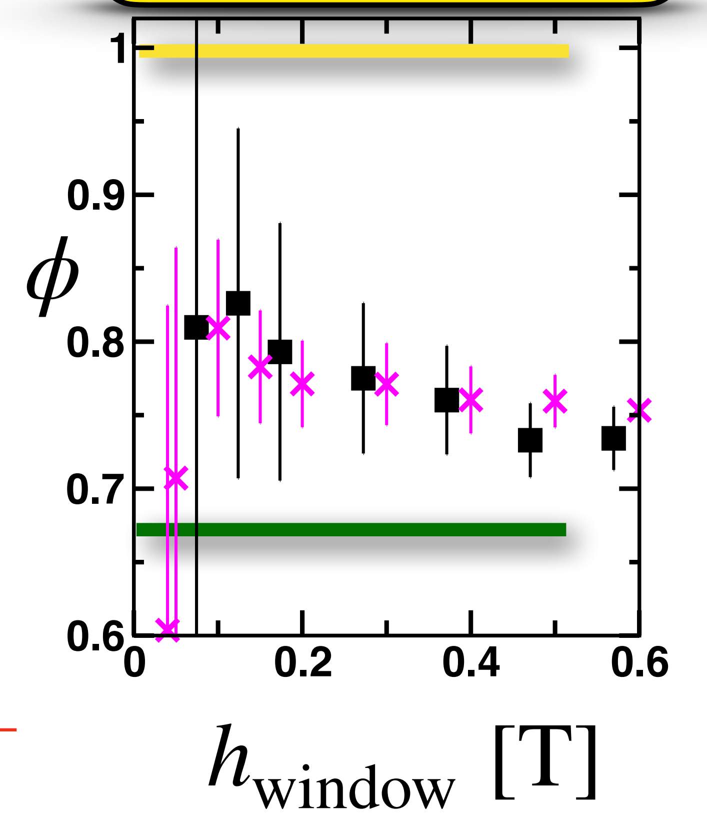
$$t^0_{3D} \approx \frac{4J_\perp^2}{J_B - J_A}$$
~ 100 mK

$$t_{2D}/t^0_{3D} \approx 30$$

➡ Genuine 3D BEC criticality with $\phi = 2/3$ observed only below 0.05 T (~ 100 mK)

➡ Above, ϕ is non-universal

QMC estimate



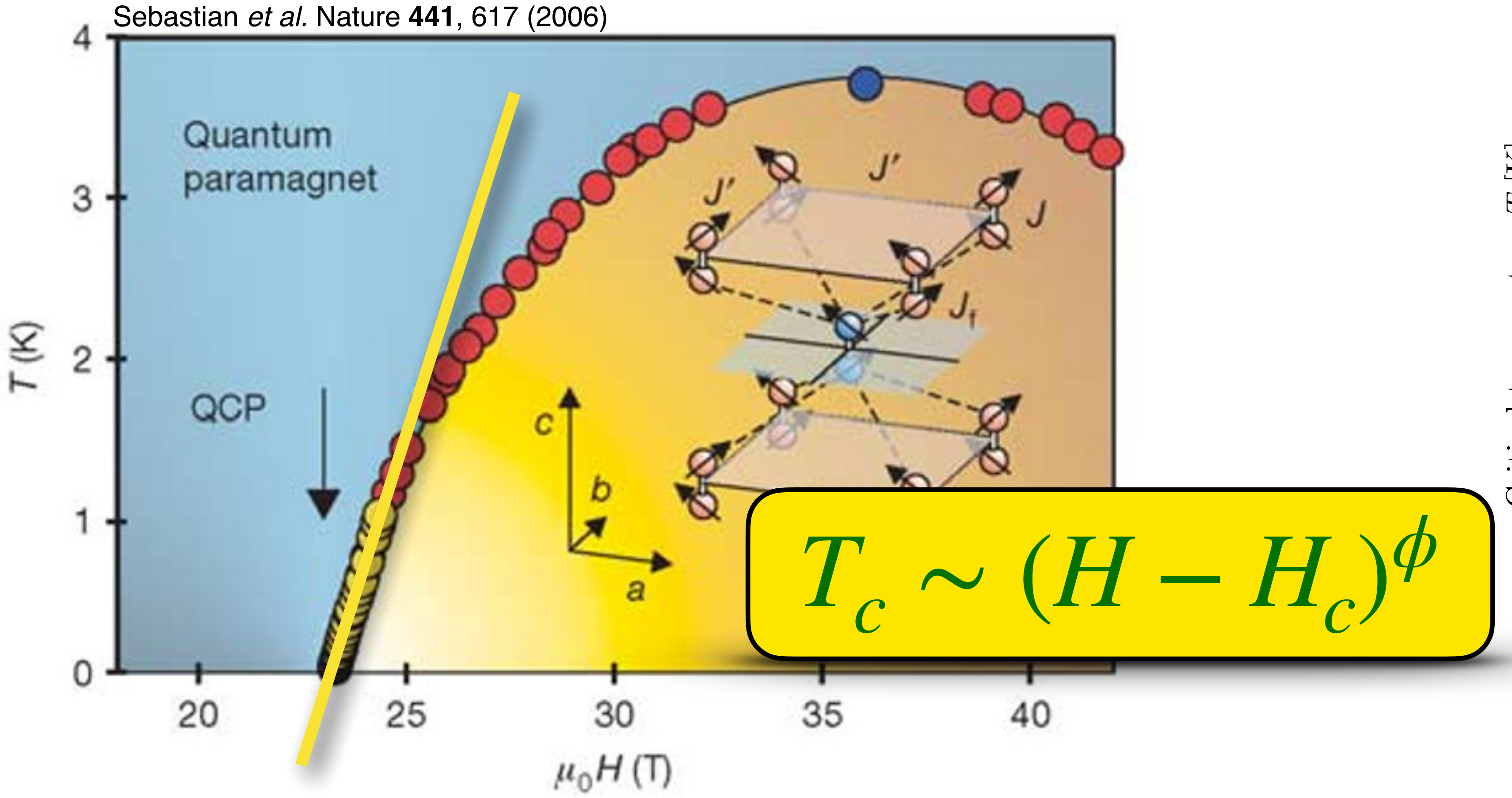
2D physics very hard to see, even when 3D couplings are very weak

End of the ϕ crisis?

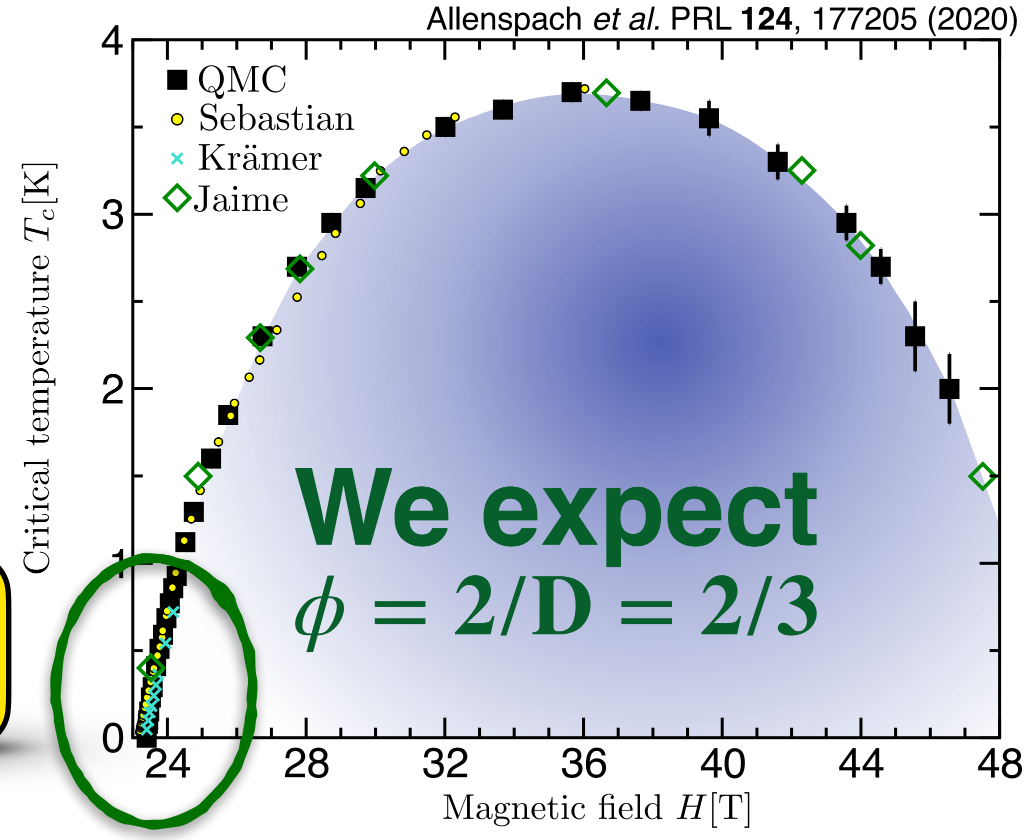
Bonus

Epilogue

Sebastian *et al.* Nature **441**, 617 (2006)



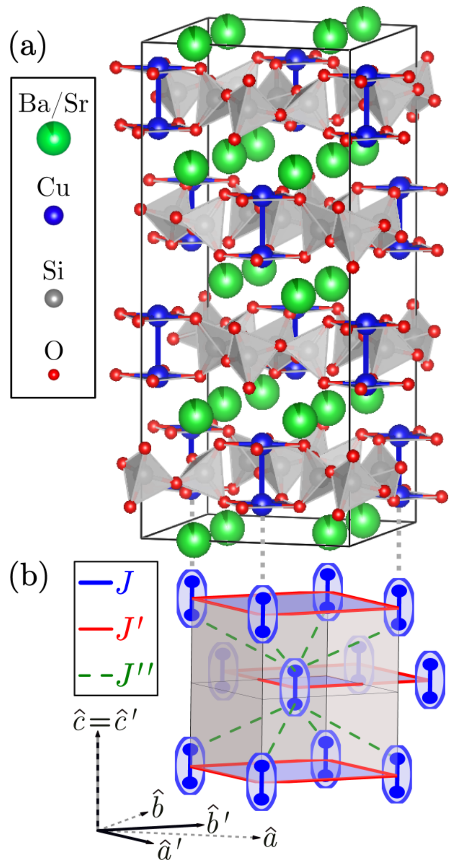
Allenspach *et al.* PRL **124**, 177205 (2020)

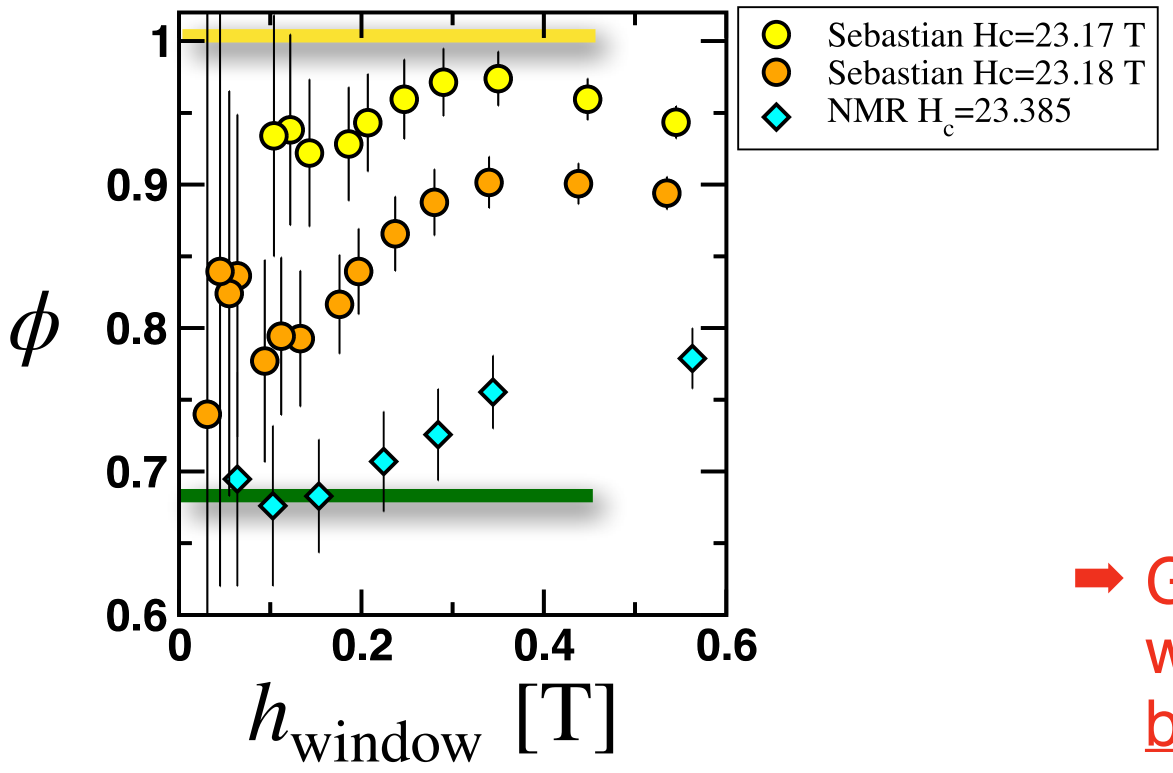


Phys. Rev. Research **3**, 023177 (2021)

Revealing three-dimensional quantum criticality by Sr substitution in Han purple

Stephan Allenspach,[1,2] Pascal Puphal,[3,4,5] Joosep Link,[6] Ivo Heinmaa,[6] Ekaterina Pomjakushina,[3] Cornelius Krellner,[4] Jakob Lass,[7,8] Gregory S. Tucker,[7,9] Christof Niedermayer,[7] Shusaku Imajo,[10] Yoshimitsu Kohama,[10] Koichi Kindo,[10] Steffen Krämer,[11] Mladen Horvatić,[11] Marcelo Jaime,[12] Alexander Madsen, Antonietta Mira,[13,14] Nicolas Laflorencie,[15] Frédéric Mila,[9] Bruce Normand,[1,9,16] Christian Rüegg,[1,2,9,17] Raivo Stern,[6] and Franziska Weickert[18]





$$t^0_{3D} \approx \frac{4J_\perp^2}{J_B - J_A}$$

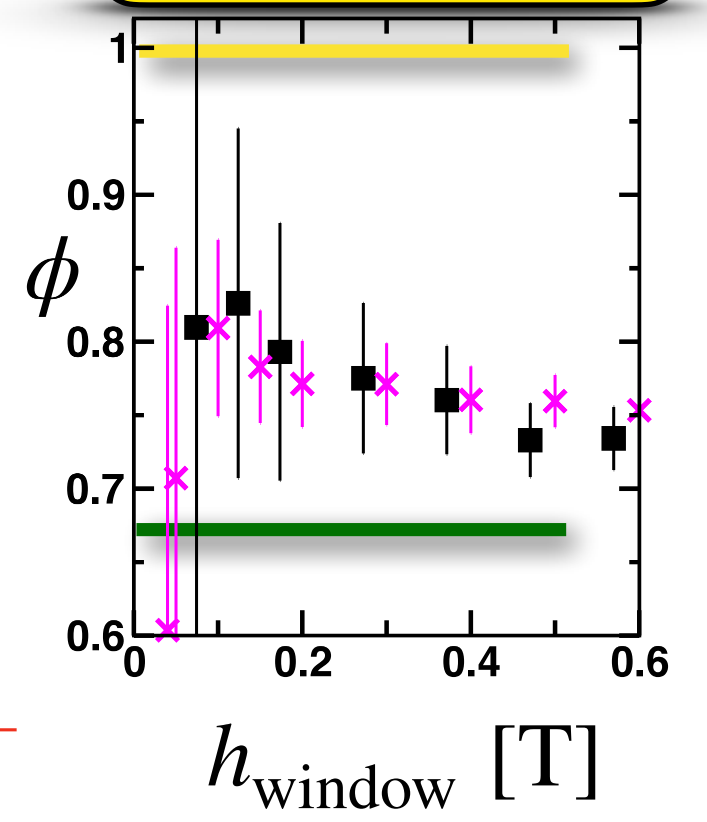
~ 100 mK

$t_{2D}/t^0_{3D} \approx 30$

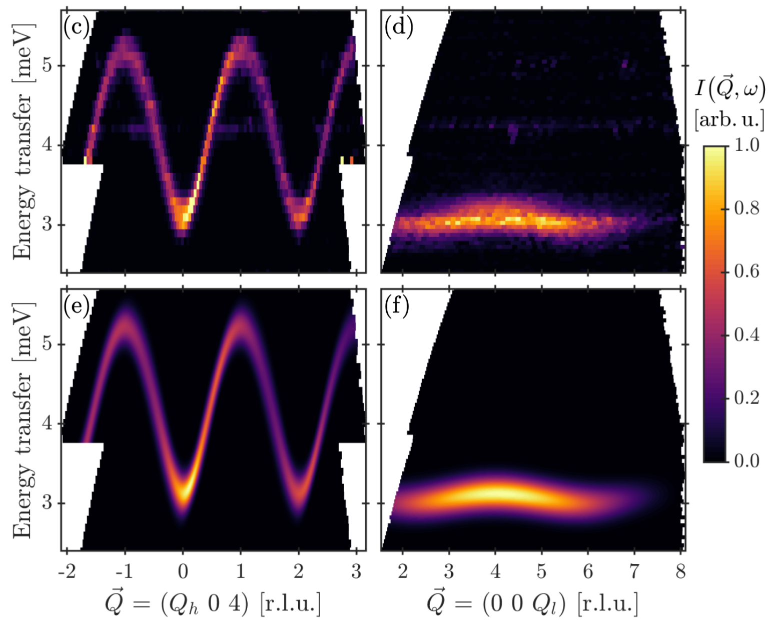
➡ Genuine 3D BEC criticality with $\phi = 2/3$ observed only below 0.05 T (~ 100 mK)

➡ Above, ϕ is non-universal

QMC estimate



$t_{2D}/t_{3D} \approx 6$



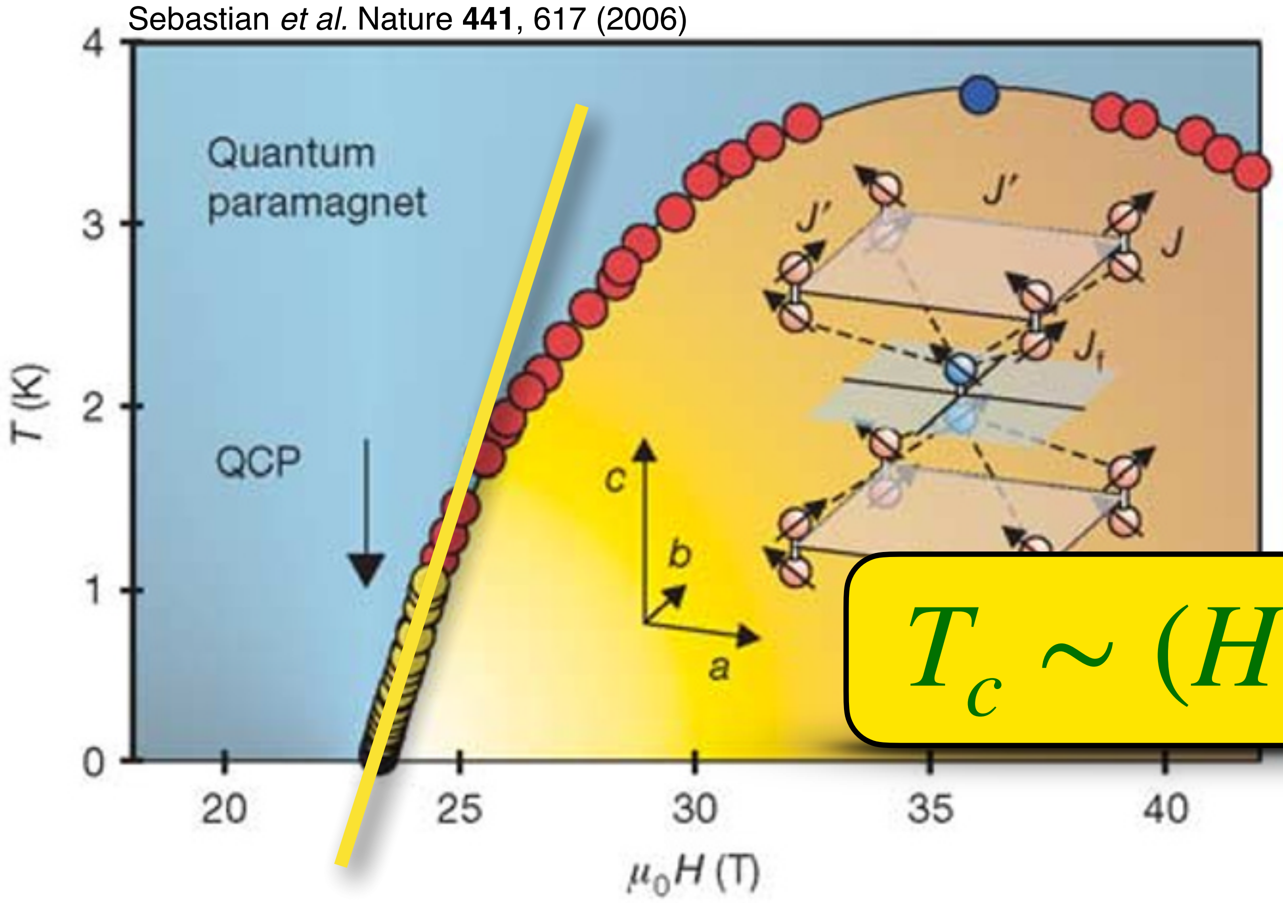
2D physics very hard to see, even when 3D couplings are very weak

Bonus

Epilogue

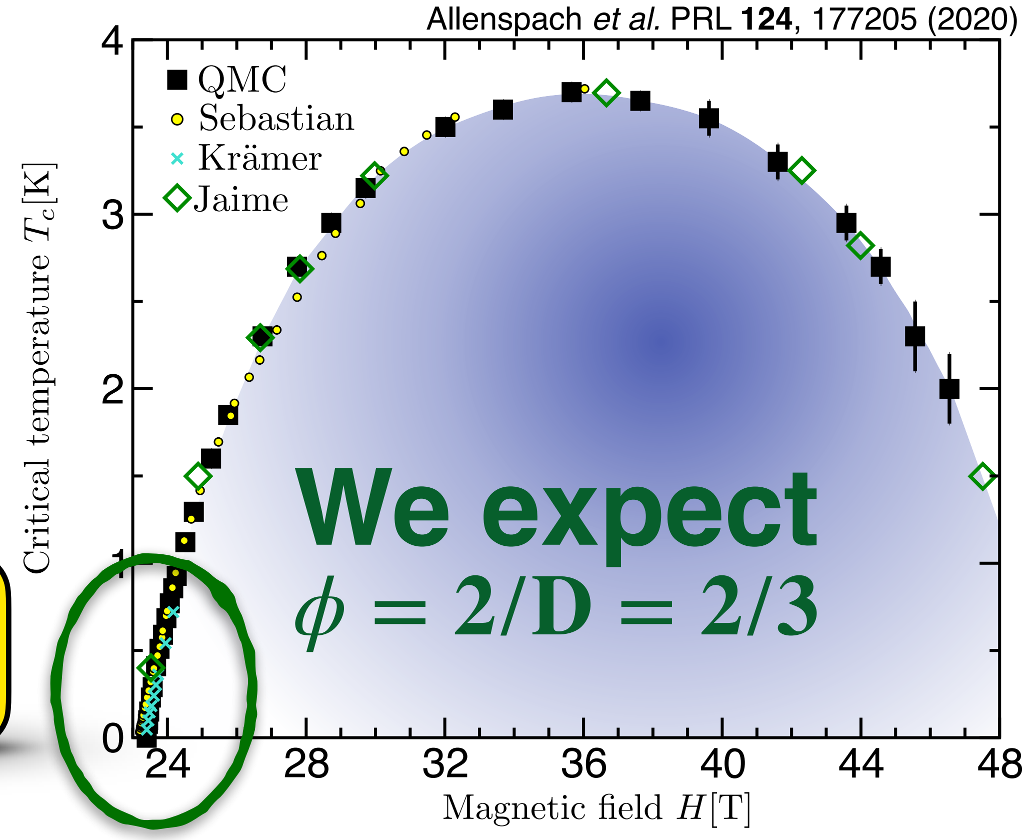
End of the ϕ crisis?

Sebastian *et al.* Nature **441**, 617 (2006)

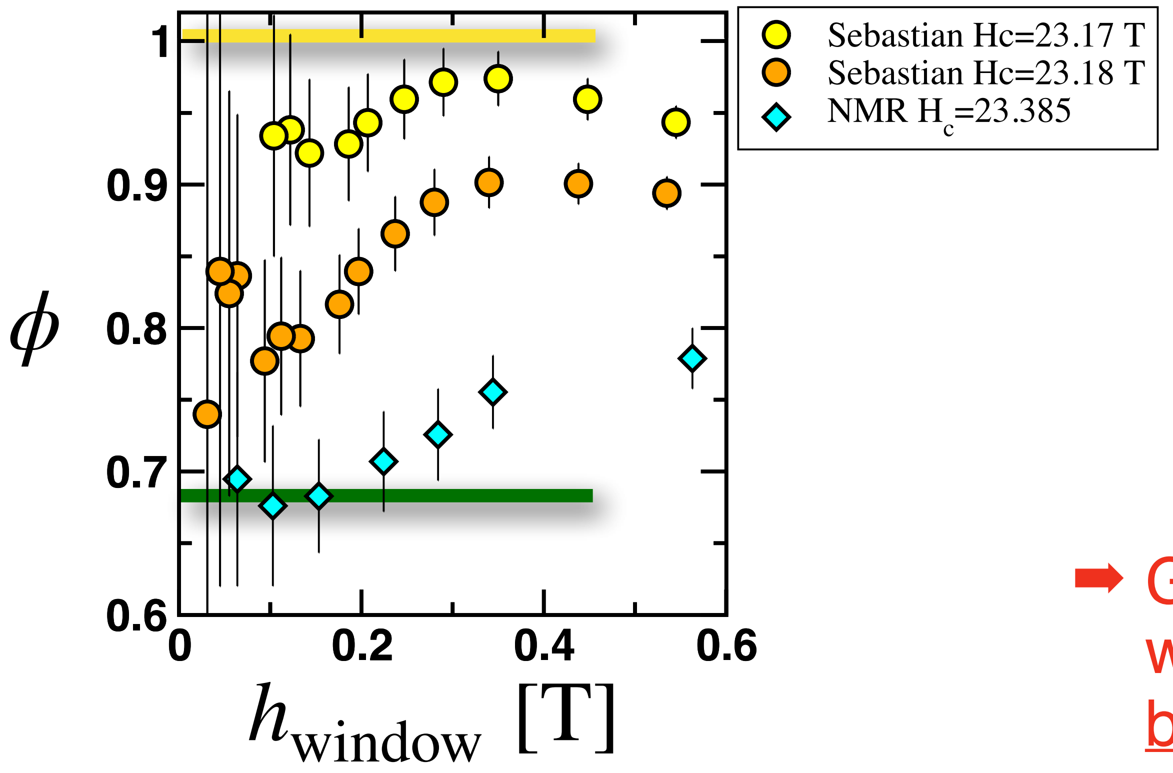


$T_c \sim (H - H_c)^{\phi}$

Allenspach *et al.* PRL **124**, 177205 (2020)



We expect $\phi = 2/D = 2/3$



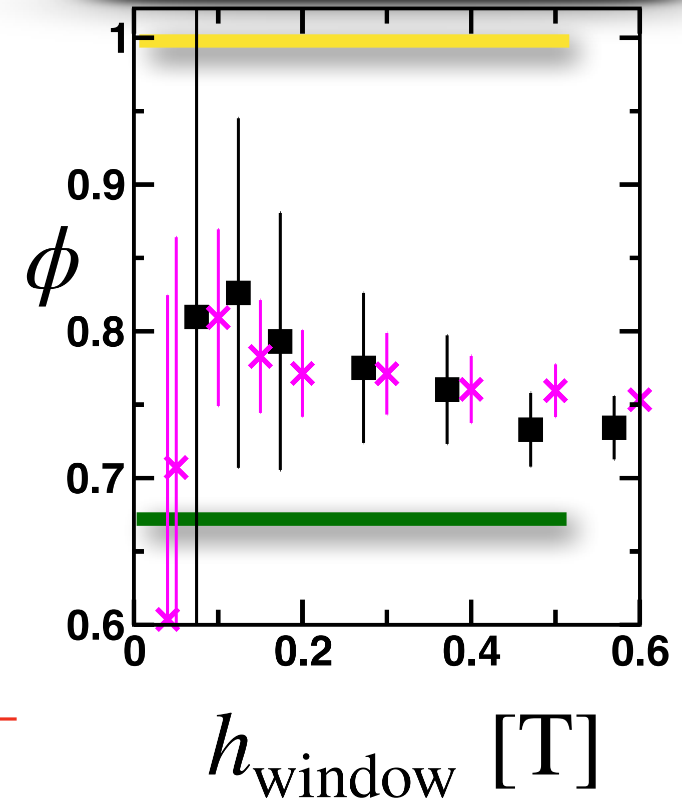
$$t^0_{3D} \approx \frac{4J_\perp^2}{J_B - J_A} \sim 100 \text{ mK}$$

$$t_{2D}/t^0_{3D} \approx 30$$

➡ Genuine 3D BEC criticality with $\phi = 2/3$ observed only below 0.05 T (~ 100 mK)

➡ Above, ϕ is non-universal

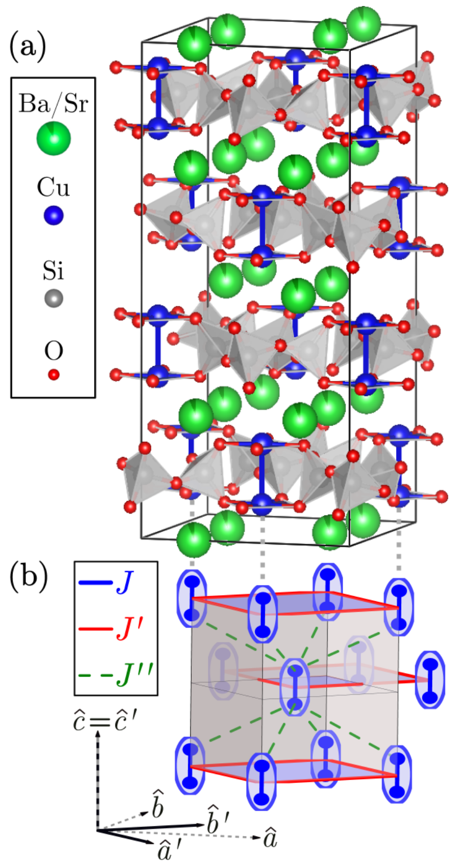
QMC estimate



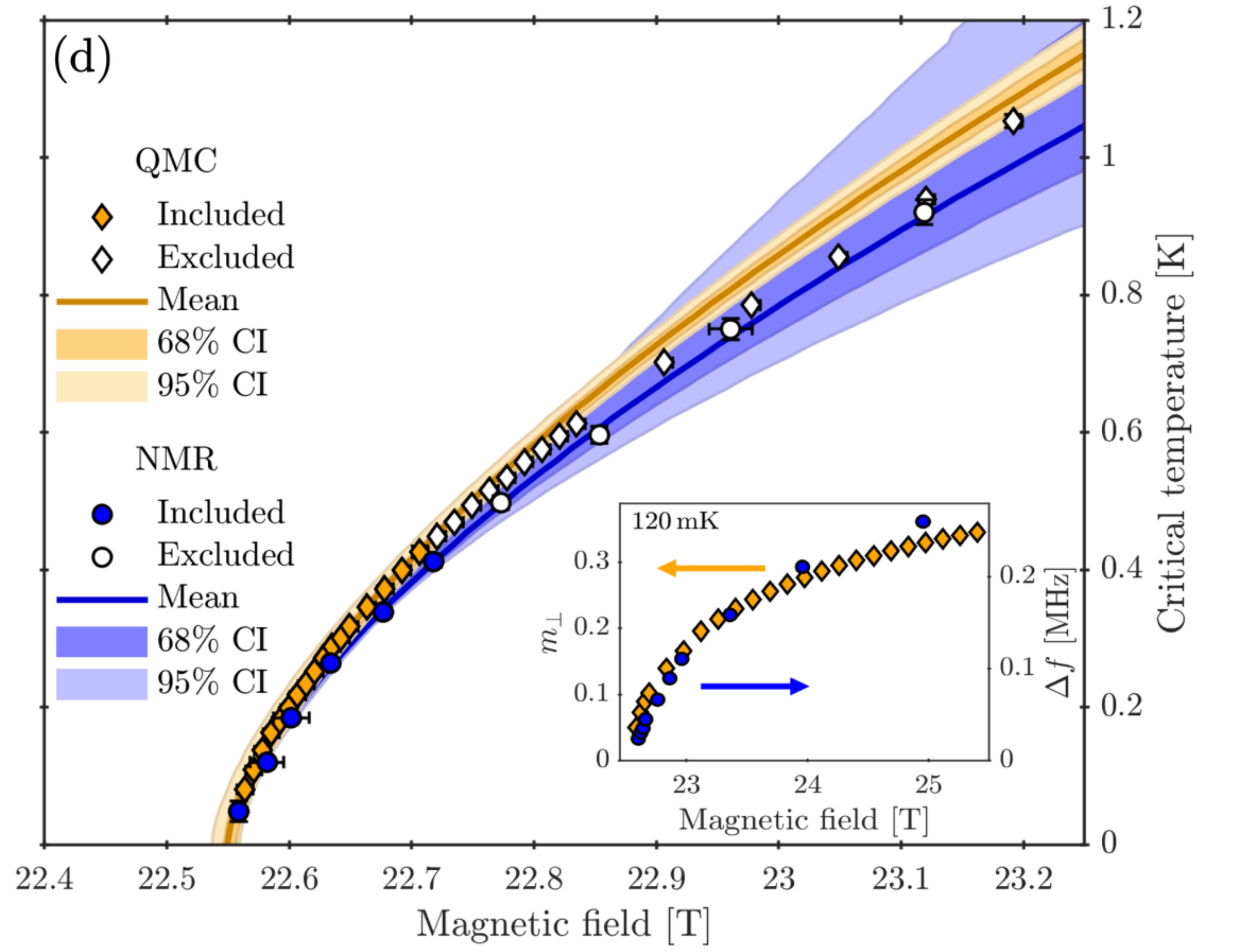
Phys. Rev. Research **3**, 023177 (2021)

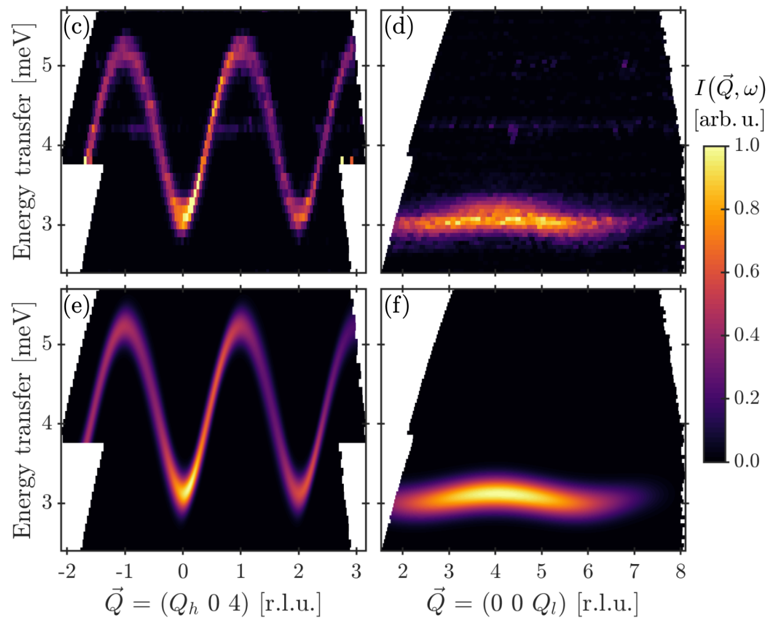
Revealing three-dimensional quantum criticality by Sr substitution in Han purple

Stephan Allenspach,[1,2] Pascal Puphal,[3,4,5] Joosep Link,[6] Ivo Heinmaa,[6] Ekaterina Pomjakushina,[3] Cornelius Krellner,[4] Jakob Lass,[7,8] Gregory S. Tucker,[7,9] Christof Niedermayer,[7] Shusaku Imajo,[10] Yoshimitsu Kohama,[10] Koichi Kindo,[10] Steffen Krämer,[11] Mladen Horvatić,[11] Marcelo Jaime,[12] Alexander Madsen Antonietta Mira,[13,14] Nicolas Laflorencie,[15] Frédéric Mila,[9] Bruce Normand,[1,9,16] Christian Rüegg,[1,2,9,17] Raivo Stern,[6] and Franziska Weickert[18]

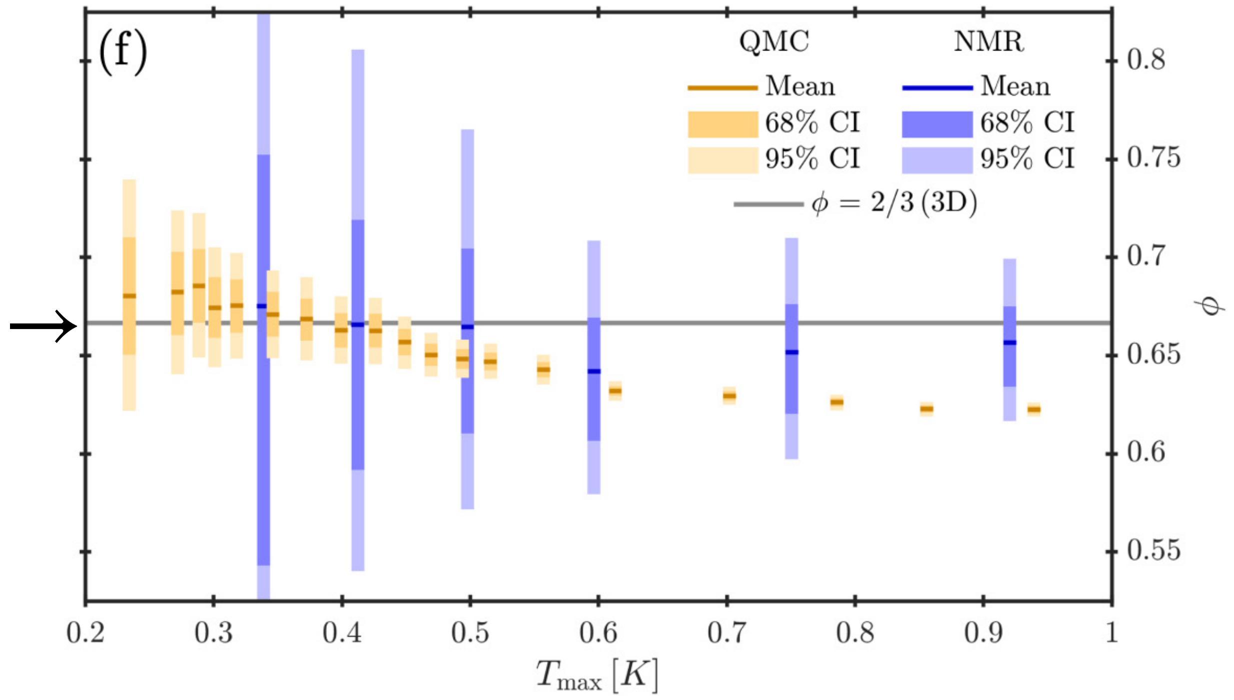


$$t_{2D}/t_{3D} \approx 6$$





$\phi = \frac{2}{3} \rightarrow$



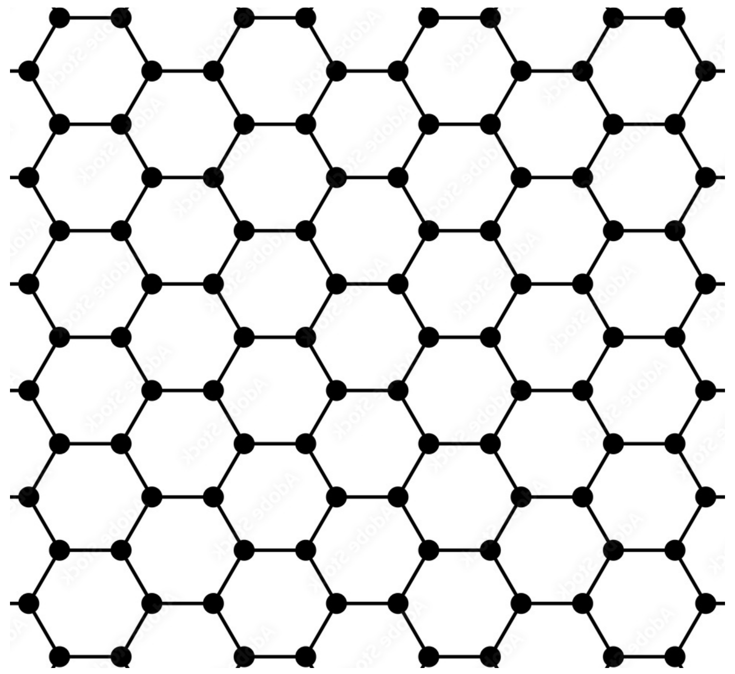
2D physics very hard to see, even when 3D couplings are very weak

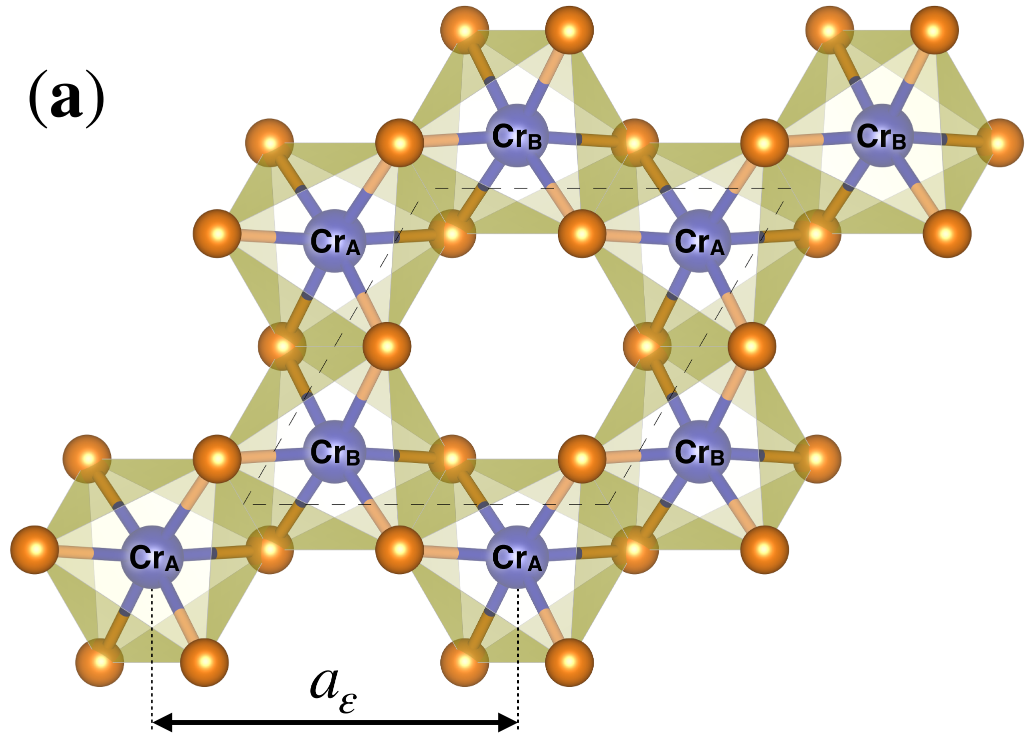
Monolayer Halides $CrCl_3$: welcome to flatland

$CrCl_3$

- Honeycomb lattice $S = 3/2$ Heisenberg model + single-ion anisotropy

$$\hat{H} = J_{\varepsilon} \sum_{\langle \boldsymbol{r}, \boldsymbol{r}' \rangle} \hat{\boldsymbol{S}}_{\boldsymbol{r}} \cdot \hat{\boldsymbol{S}}_{\boldsymbol{r}'} + K_{\varepsilon} \sum_{\boldsymbol{r}} \left(\hat{S}^z_{\boldsymbol{r}} \right)^2$$





Monolayer Halides $CrCl_3$: welcome to flatland

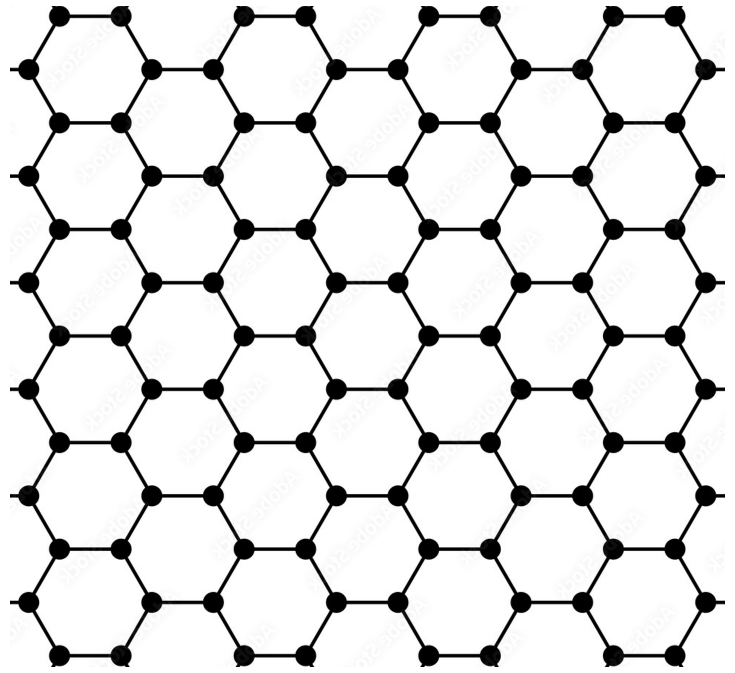
$CrCl_3$

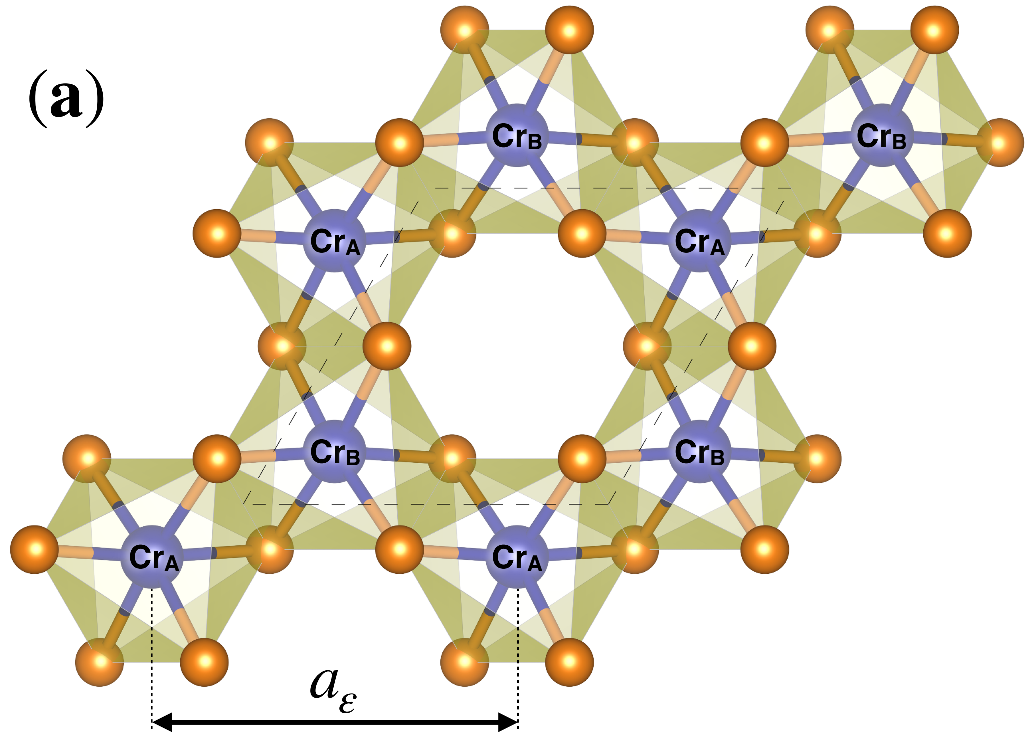
- **Honeycomb lattice $S = 3/2$ Heisenberg model + single-ion anisotropy**

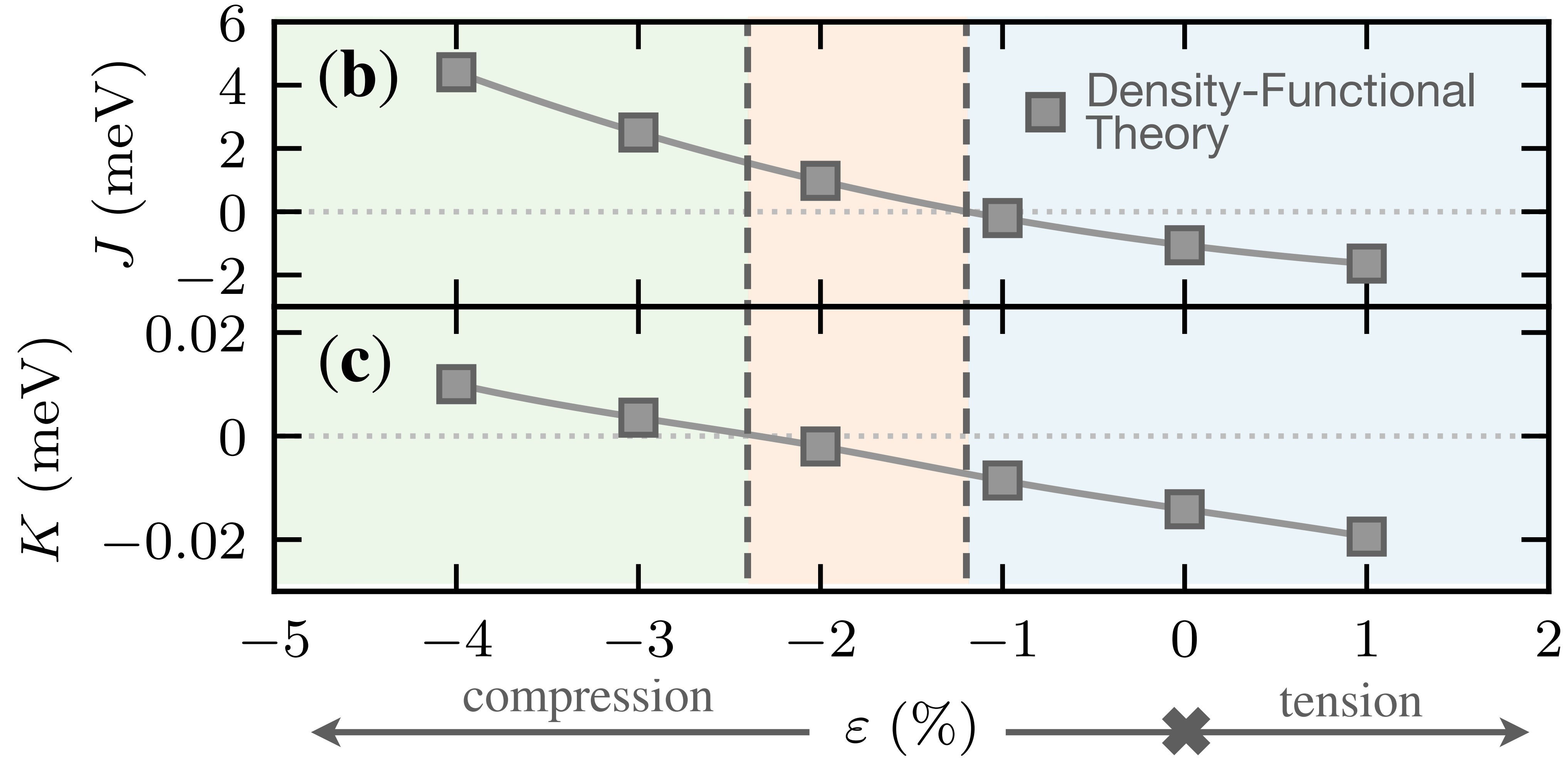
$$\hat{H} = J_\varepsilon \sum_{\langle \boldsymbol{r},\boldsymbol{r}'\rangle} \hat{\boldsymbol{S}}_{\boldsymbol{r}} \cdot \hat{\boldsymbol{S}}_{\boldsymbol{r}'} + K_\varepsilon \sum_{\boldsymbol{r}} \left(\hat{S}^z_{\boldsymbol{r}}\right)^2$$

■ Density functional theory calculations

Yaroslav Kvashnin (Uppsala)





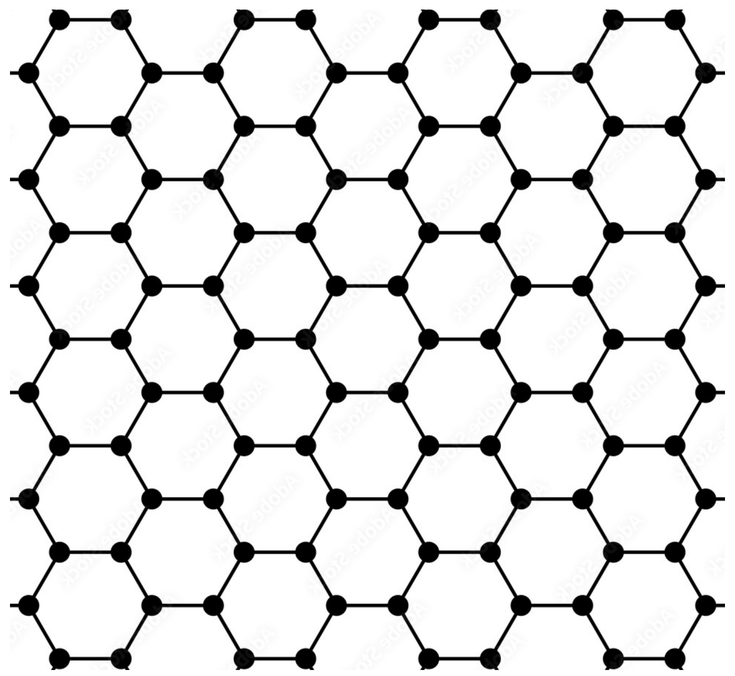


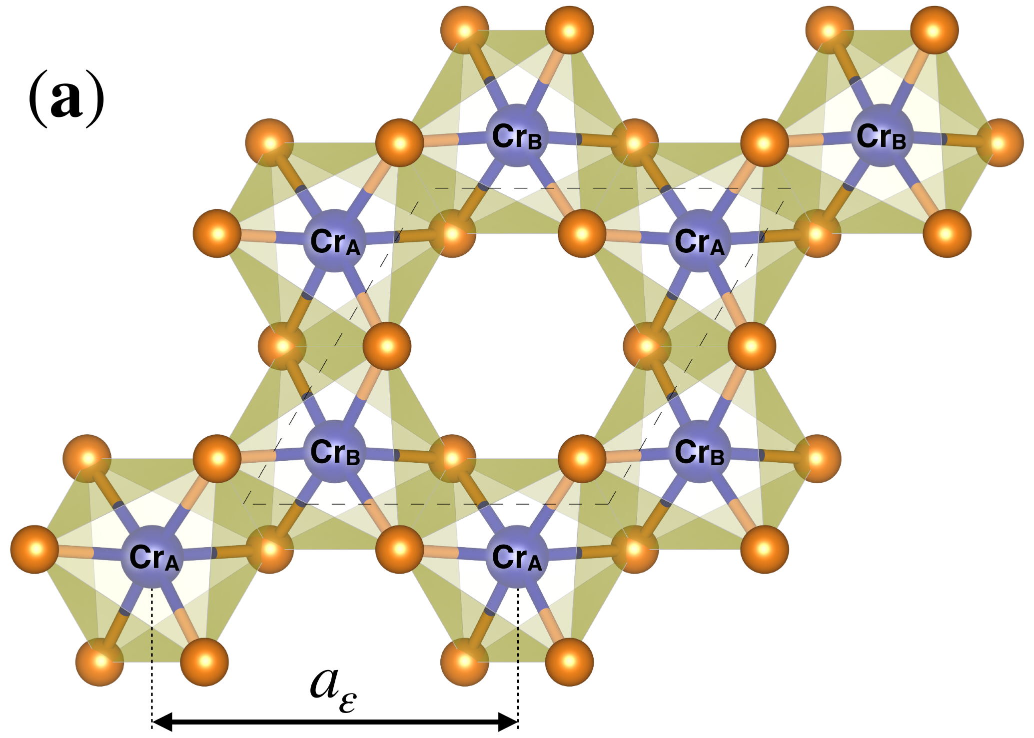
Monolayer Halides $CrCl_3$: welcome to flatland

$CrCl_3$

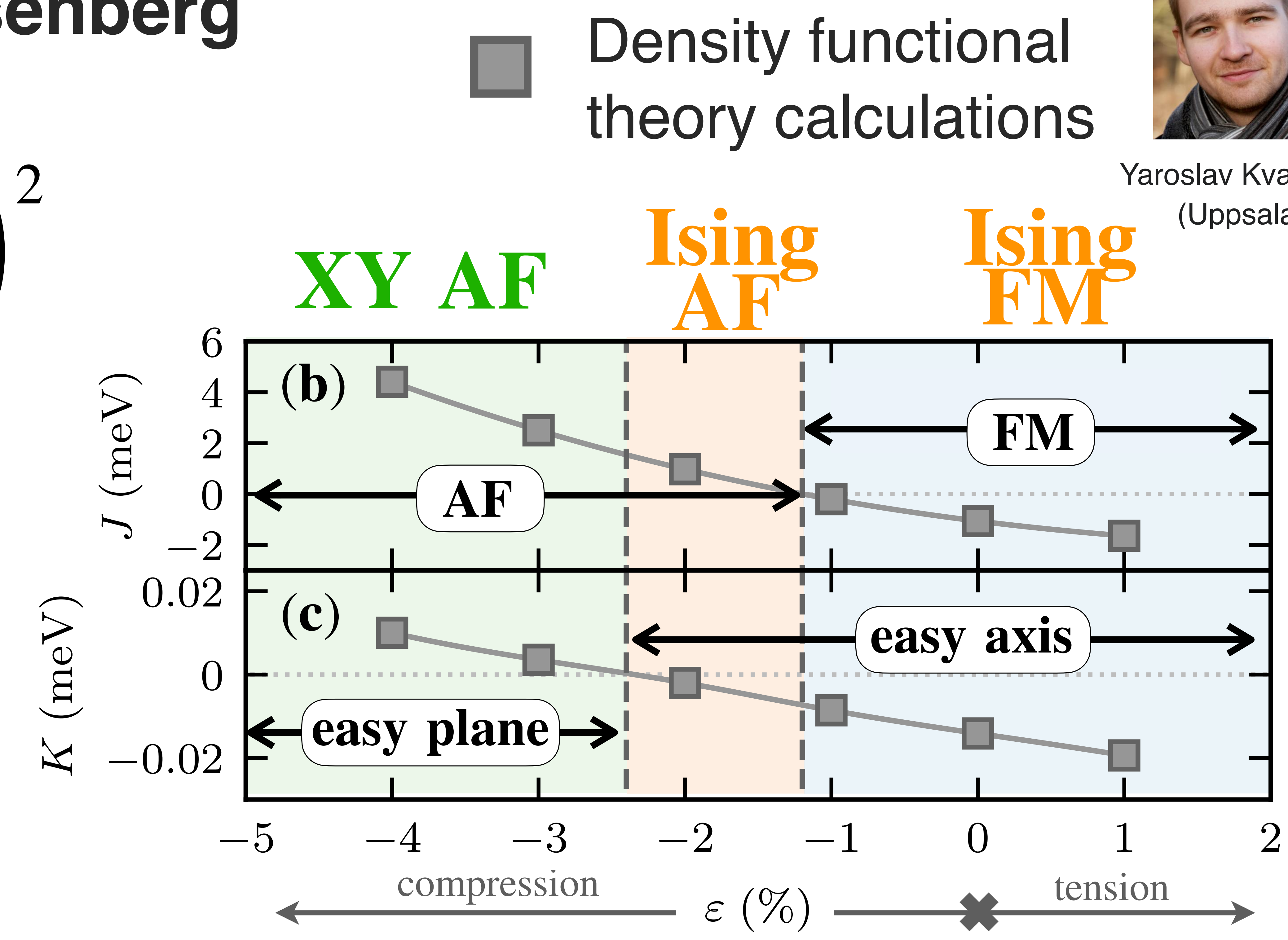
- **Honeycomb lattice $S = 3/2$ Heisenberg model + single-ion anisotropy**

$$\hat{H} = J_\varepsilon \sum_{\langle \boldsymbol{r},\boldsymbol{r}' \rangle} \hat{\boldsymbol{S}}_{\boldsymbol{r}} \cdot \hat{\boldsymbol{S}}_{\boldsymbol{r}'} + K_\varepsilon \sum_{\boldsymbol{r}} \left(\hat{S}^z_{\boldsymbol{r}} \right)^2$$





Yaroslav Kvashnin (Uppsala)

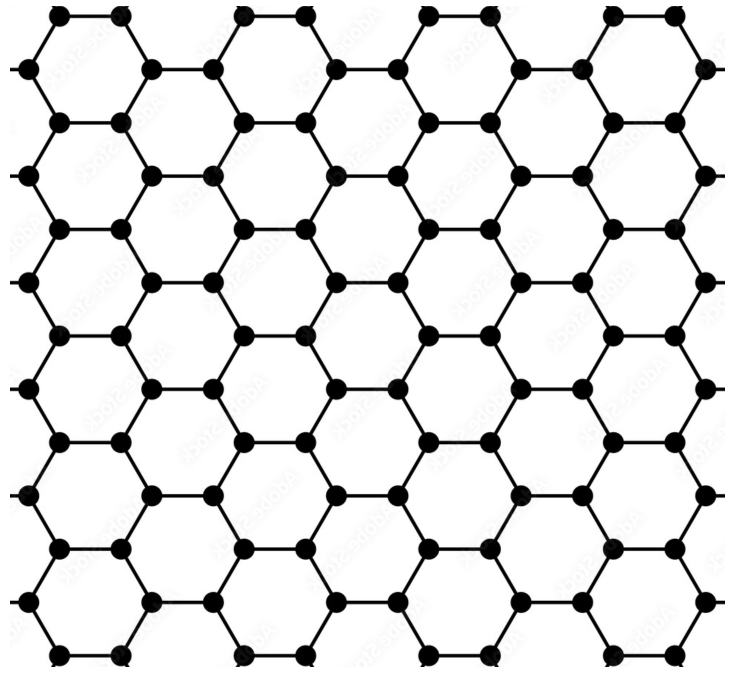


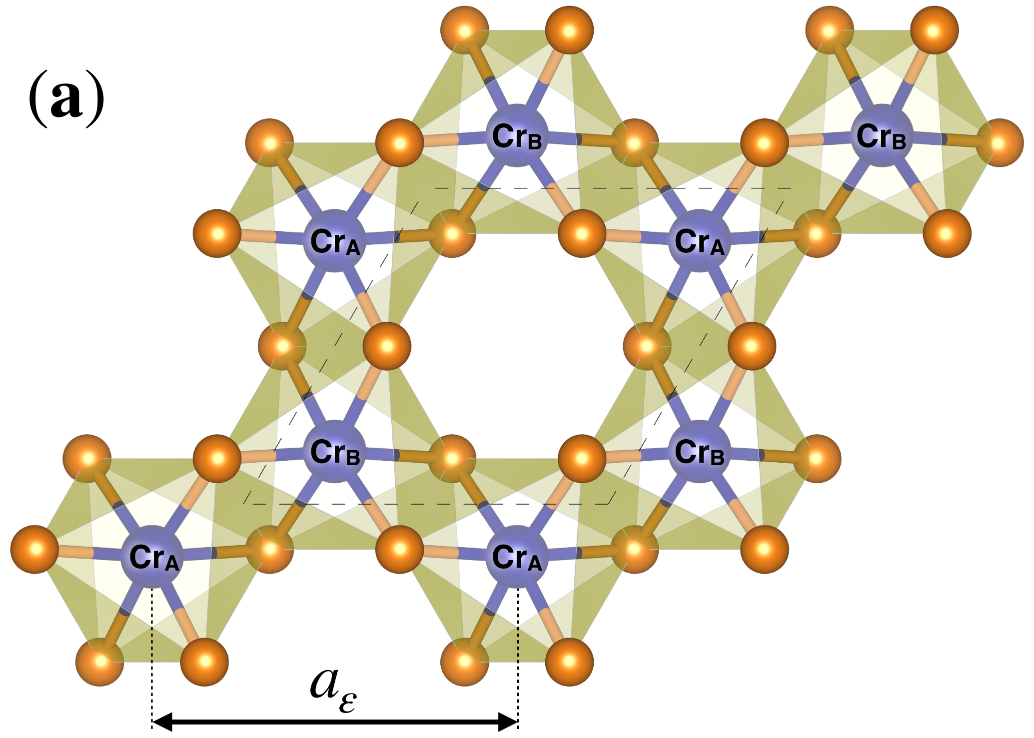
Monolayer Halides $CrCl_3$: welcome to flatland

$CrCl_3$

- **Honeycomb lattice $S = 3/2$ Heisenberg model + single-ion anisotropy**

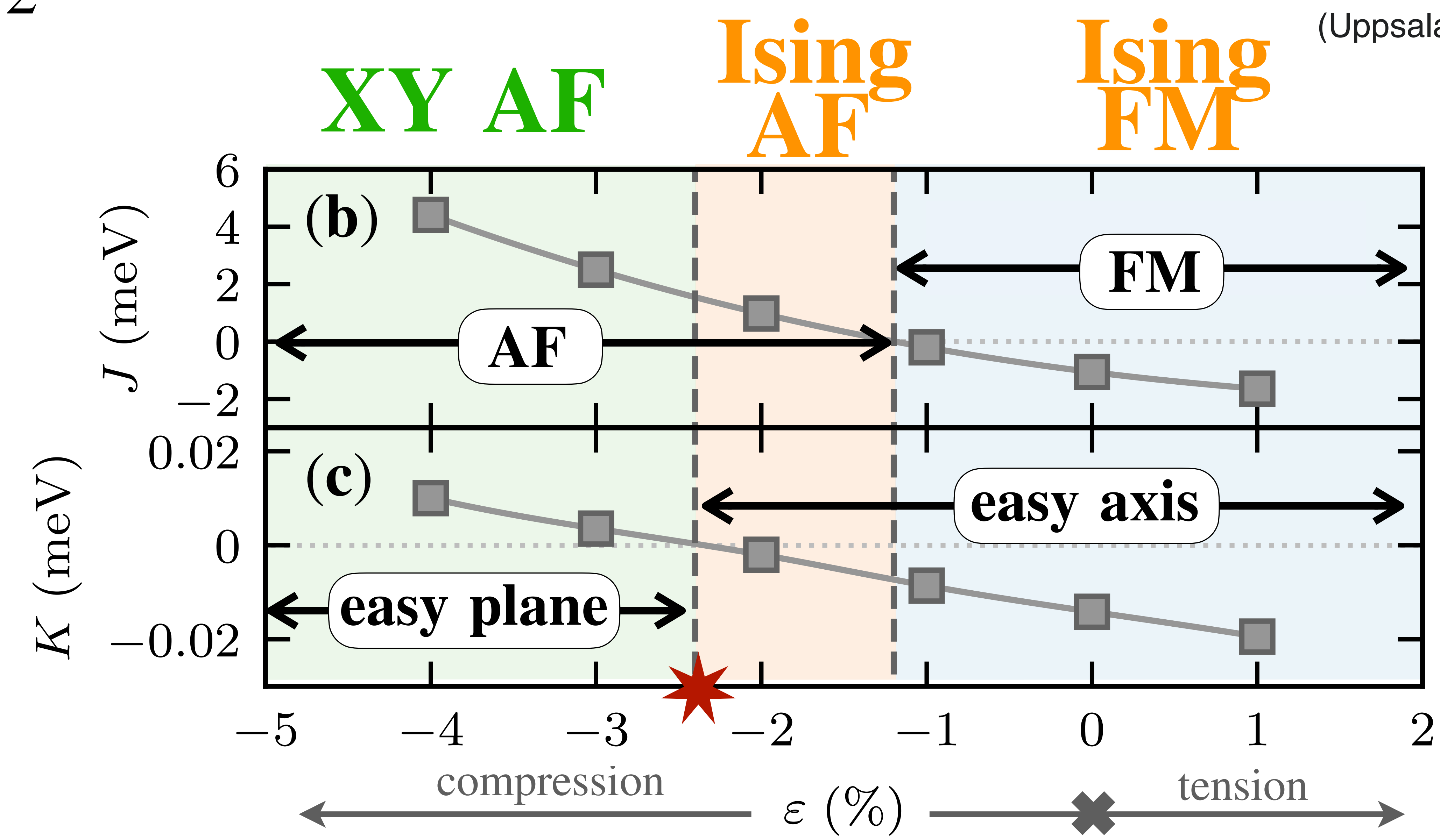
$$\hat{H} = J_\varepsilon \sum_{\langle r, r' \rangle} \hat{S}_r \cdot \hat{S}_{r'} + K_\varepsilon \sum_r \left(\hat{S}^z_r\right)^2$$





■ Density functional theory calculations

Yaroslav Kvashnin (Uppsala)

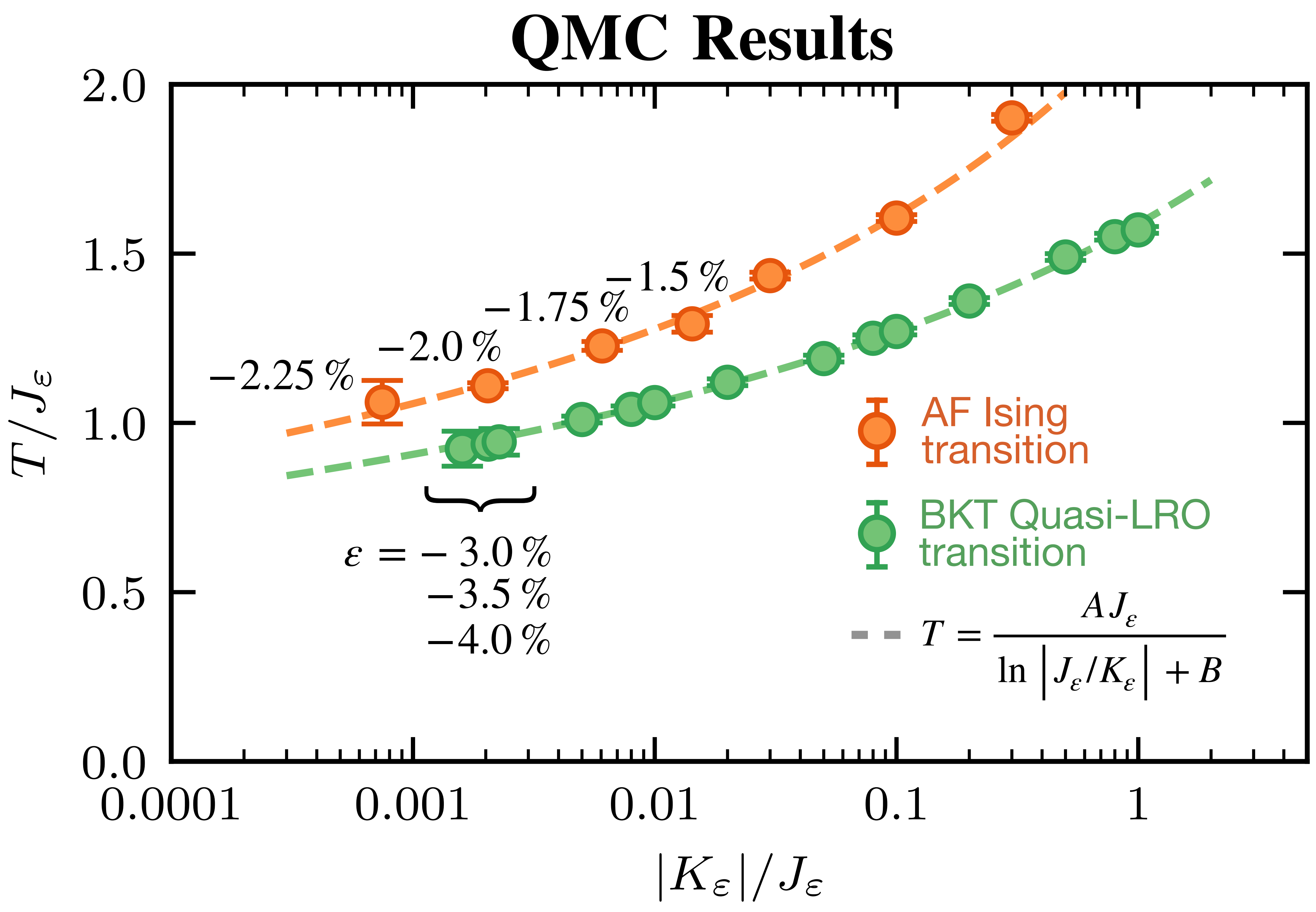


- **Can we reach "large" BKT and Ising critical temperatures ?**

★ $K = 0$ Heisenberg AF (ordered only at $T = 0$) $\xi(T) = \xi_0 \exp(\frac{2\pi J \rho_s}{T})$ (Chakravarty, Halperin, Nelson, PRL 60, 1057 (1988))

- Small $K > 0$ XY AF: Start to see easy-plane correlations when $KS^2\xi^2(T^*) \approx J \Rightarrow T_{\mathrm{BKT}} = \frac{4\pi J \rho_s}{\ln(J/K) + B}$
- Small $K < 0$ Ising AF: easy-axis correlations $\Rightarrow T_c = \frac{4\pi J \rho_s}{\ln(J/|K|) + B}$

Very strong logarithmic growth of the critical temperatures



PHYSICAL REVIEW LETTERS **127,** 037204 (2021)

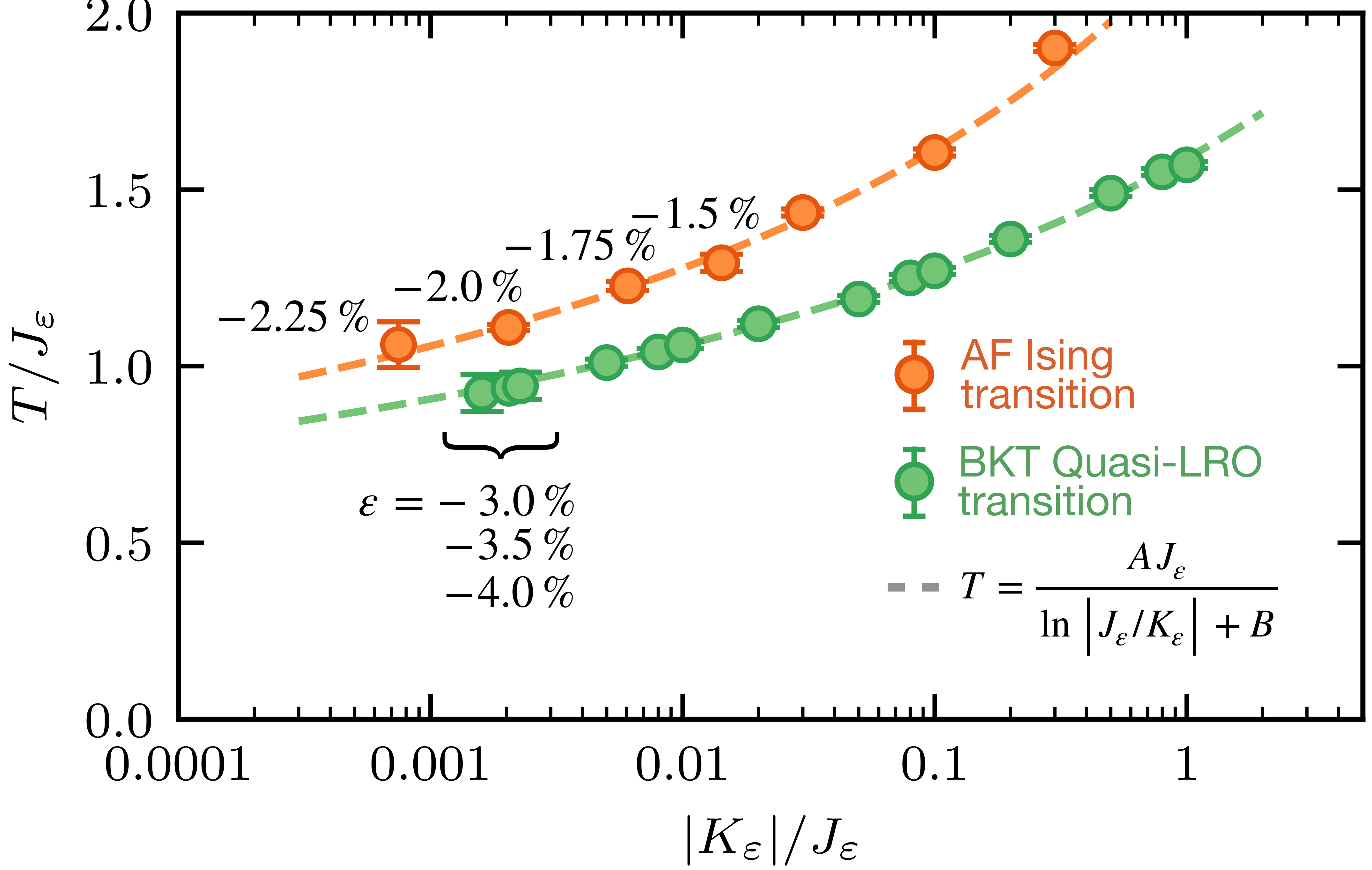
Monolayer $CrCl_3$ as an Ideal Test Bed for the Universality Classes of 2D Magnetism

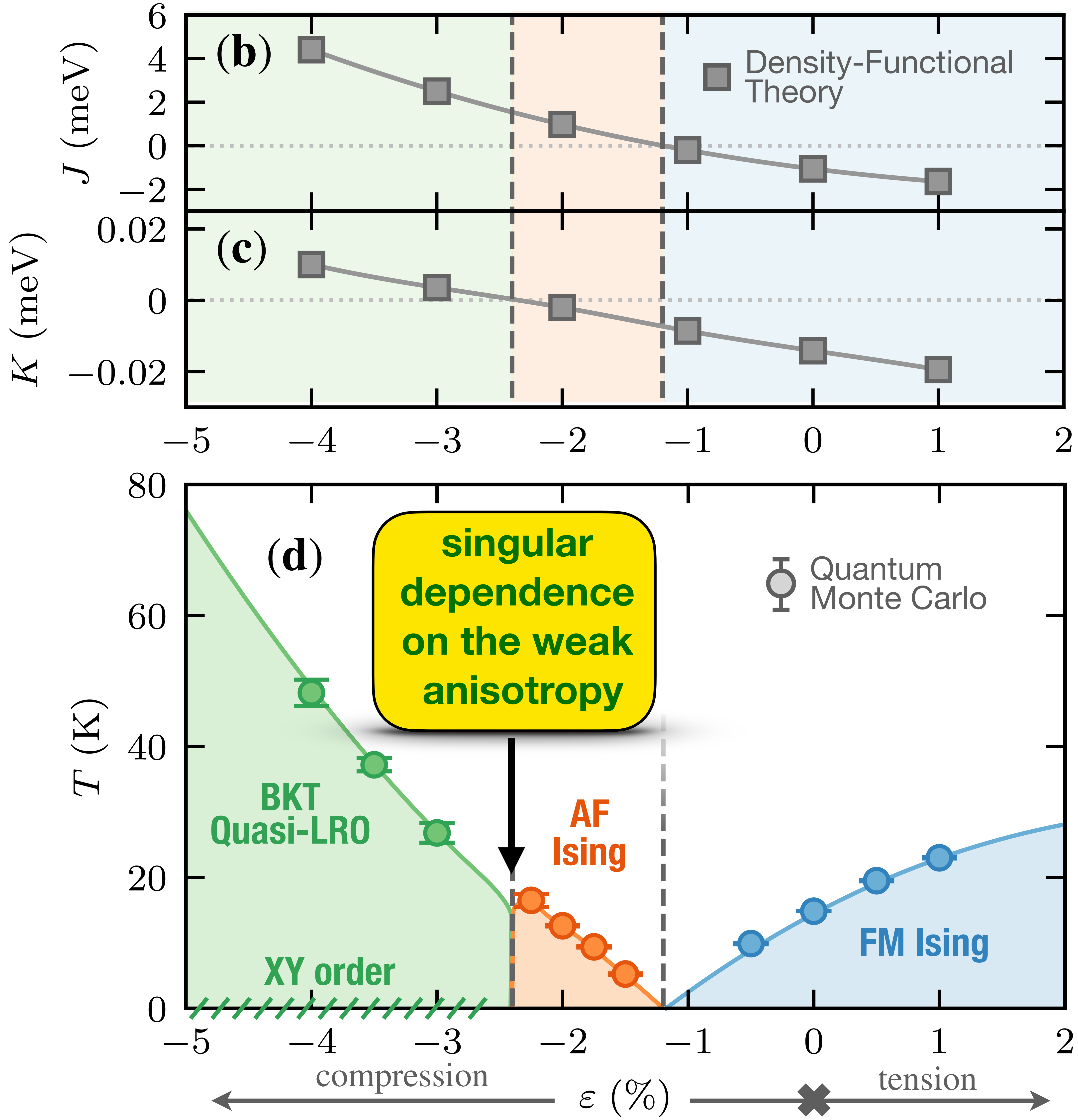
M. Dupont,[1,2] Y. O. Kvashnin,[3] M. Shiranzaei,[3] J. Fransson,[3] N. Laflorencie,[4] and A. Kantian[5,3]
[1]*Department of Physics, University of California, Berkeley, California 94720, USA*
[2]*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*
[3]*Department of Physics and Astronomy, Uppsala University, Box 516, S-751 20 Uppsala, Sweden*
[4]*Laboratoire de Physique Théorique, IRSAMC, Université de Toulouse, CNRS, UPS, 31062 Toulouse, France*
[5]*SUPA, Institute of Photonics and Quantum Sciences, Heriot-Watt University, Edinburgh EH14 4AS, United Kingdom*

Very strong logarithmic growth of the critical temperatures

Theoretical phase diagram

QMC Results





PHYSICAL REVIEW LETTERS **127,** 037204 (2021)

Monolayer $CrCl_3$ as an Ideal Test Bed for the Universality Classes of 2D Magnetism

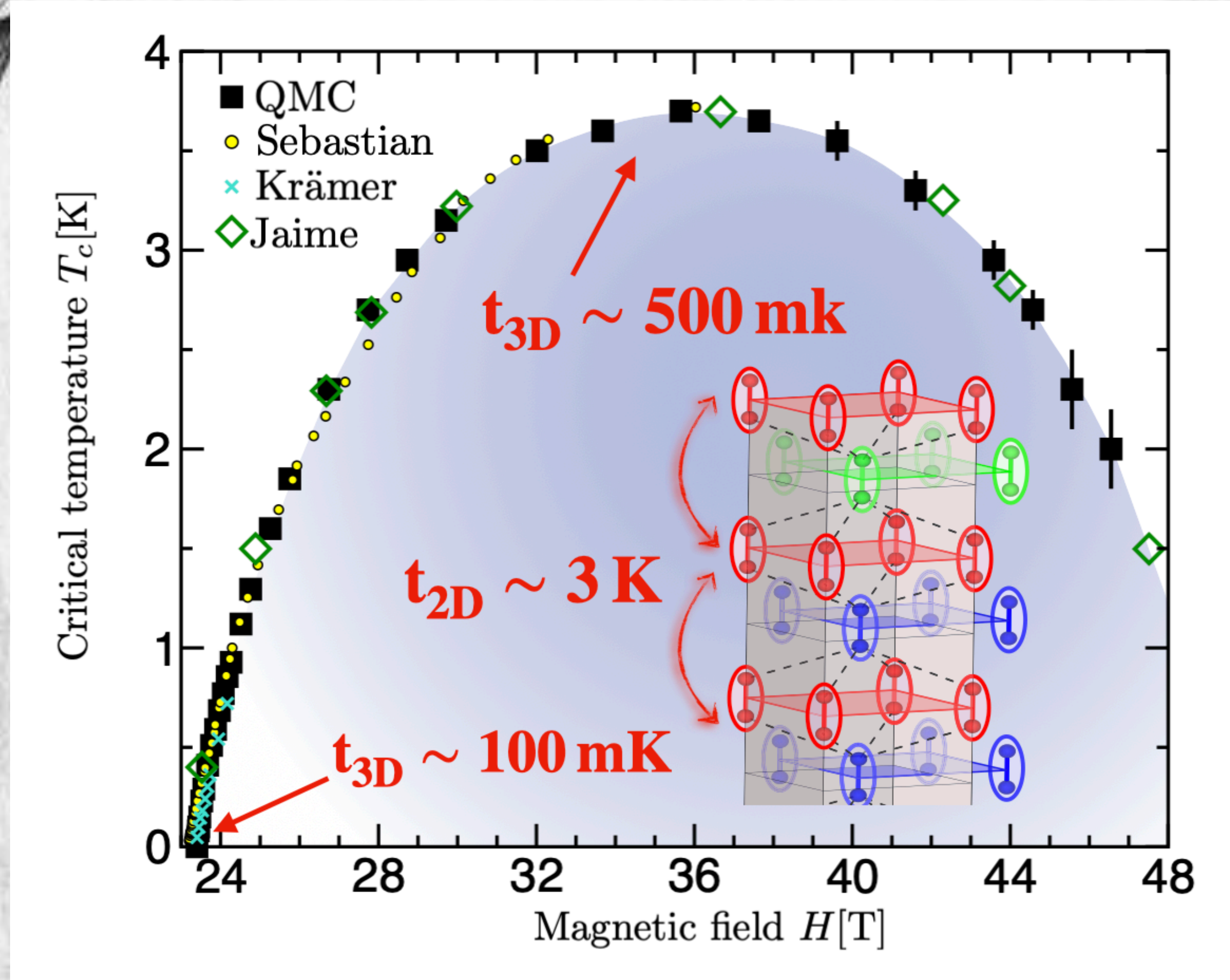
M. Dupont,[1,2] Y. O. Kvashnin,[3] M. Shiranzaei,[3] J. Fransson,[3] N. Laflorencie,[4] and A. Kantian[5,3]

[1]*Department of Physics, University of California, Berkeley, California 94720, USA*
[2]*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*
[3]*Department of Physics and Astronomy, Uppsala University, Box 516, S-751 20 Uppsala, Sweden*
[4]*Laboratoire de Physique Théorique, IRSAMC, Université de Toulouse, CNRS, UPS, 31062 Toulouse, France*
[5]*SUPA, Institute of Photonics and Quantum Sciences, Heriot-Watt University, Edinburgh EH14 4AS, United Kingdom*

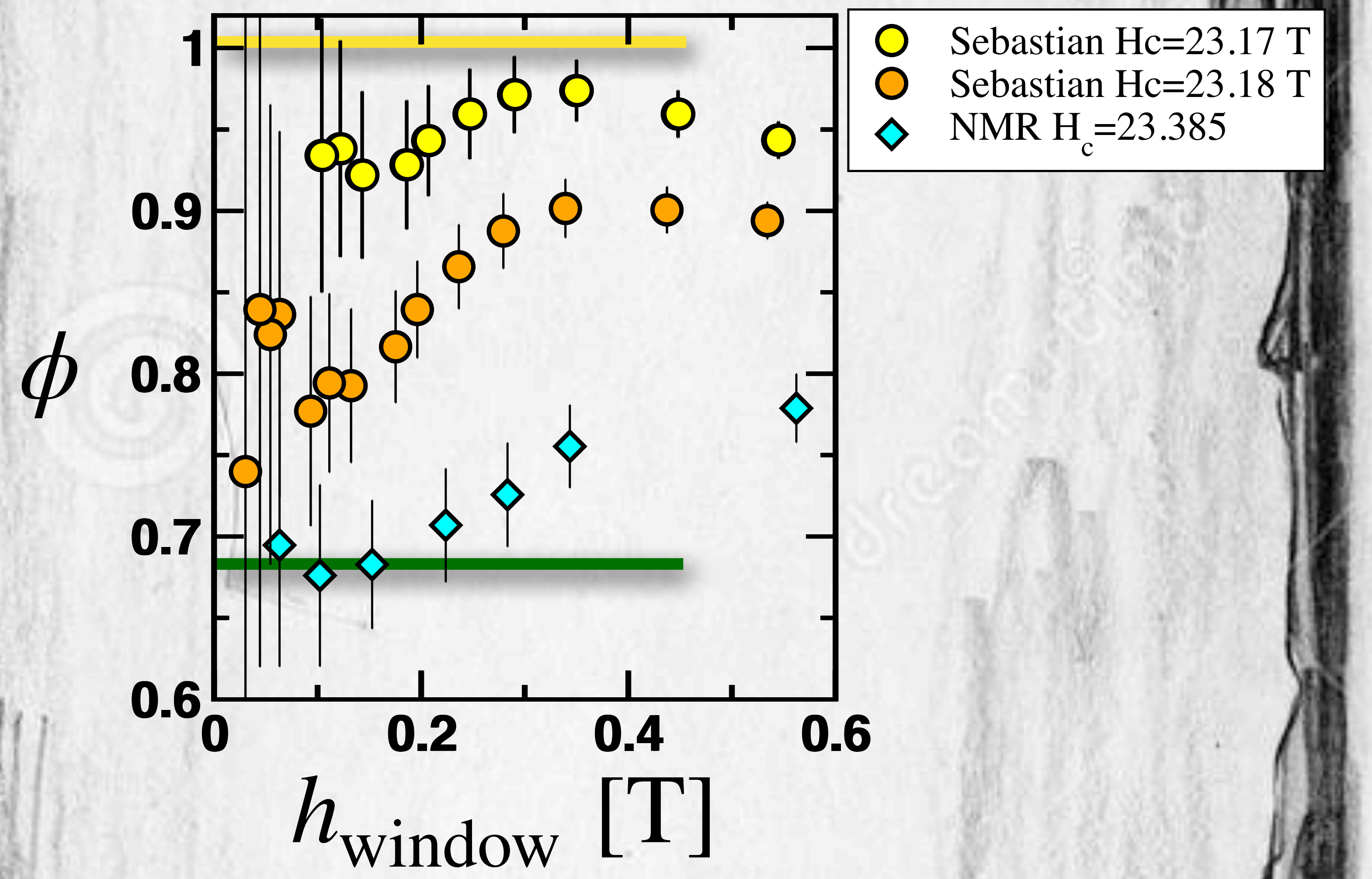
Conclusions

Tiny anisotropies (real space or spin space) can have dramatic effects on critical temperatures

$$T_c^{3D} = T_{BKT}\left(1 + \frac{a}{\ln^2\left(t_{3D}/t_{2D}\right) + c}\right)$$



Critical exponents are very sensitive numbers



Starring

$BaCuSi_2O_6$

$CrCl_3$

Bruce

Stephan

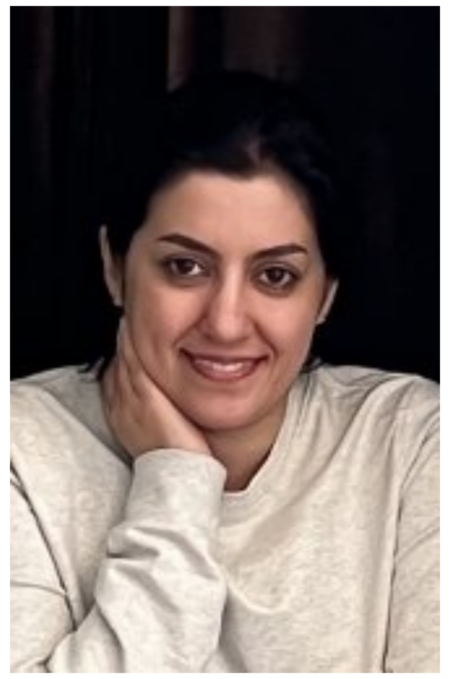
Christian

Mladen

Franziska

Maxime

Yaroslav



Marhoo

Frédéric

Steffen

Raivo

Claude

Jonas

Adrian