Emergence of mesoscale quantum phase transitions in a ferromagnet

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1. Recap quantum criticality
2. Transverse-field Ising model & $LiHoF_4$
3. Experimental results in **tilted** fields
4. Modelling: Stray fields and domains
5. Mesoscale quantum criticality

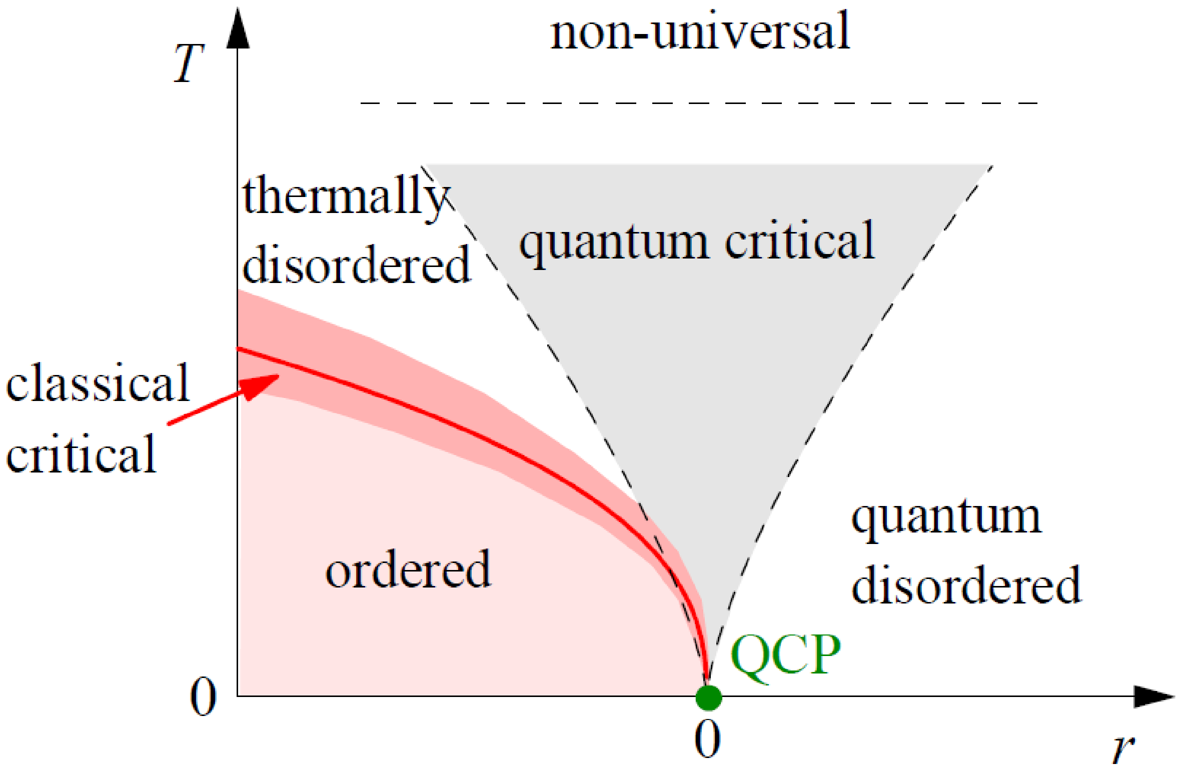
What is a phase transition?

A change in the collective properties of a macroscopic number of atoms.

What is a quantum phase transition?

A phase transition at $T = 0$,
driven by “quantum fluctuations”.

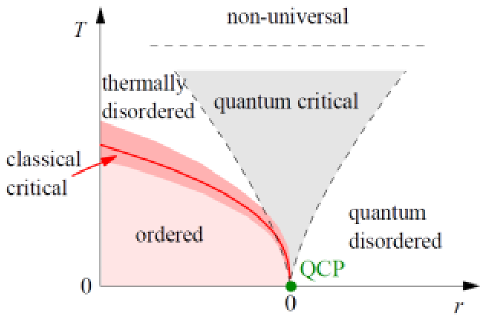
What is a quantum phase transition?



Experimentally observed in many compounds,
e.g. in $TlCuCl_3$ under pressure.

Rüegg *et al.*, PRL **100**, 205701 (2008)

Landau-Ginzburg-Wilson description



In many cases, underlying field theory is known.

Magnetic order-disorder transition (e.g. $TlCuCl_3$):

φ^4 (or Landau-Ginzburg-Wilson) theory for
antiferromagnetic order parameter $\varphi_\alpha(\vec{x},\tau)$.

$$\boxed{\mathcal{S} = \int d^d r d\tau \frac{1}{2}\left[c^2(\vec{\nabla}\varphi_\alpha)^2 + (\partial_\tau\varphi_\alpha)^2 + m_0\varphi_\alpha^2\right] + \frac{u_0}{24}(\varphi_\alpha^2)^2}$$

**Coarse-grained description of
microscopic (physical or emergent) degrees of freedom**

$LiHoF_4$: An Ising ferromagnet

Textbook example for quantum criticality

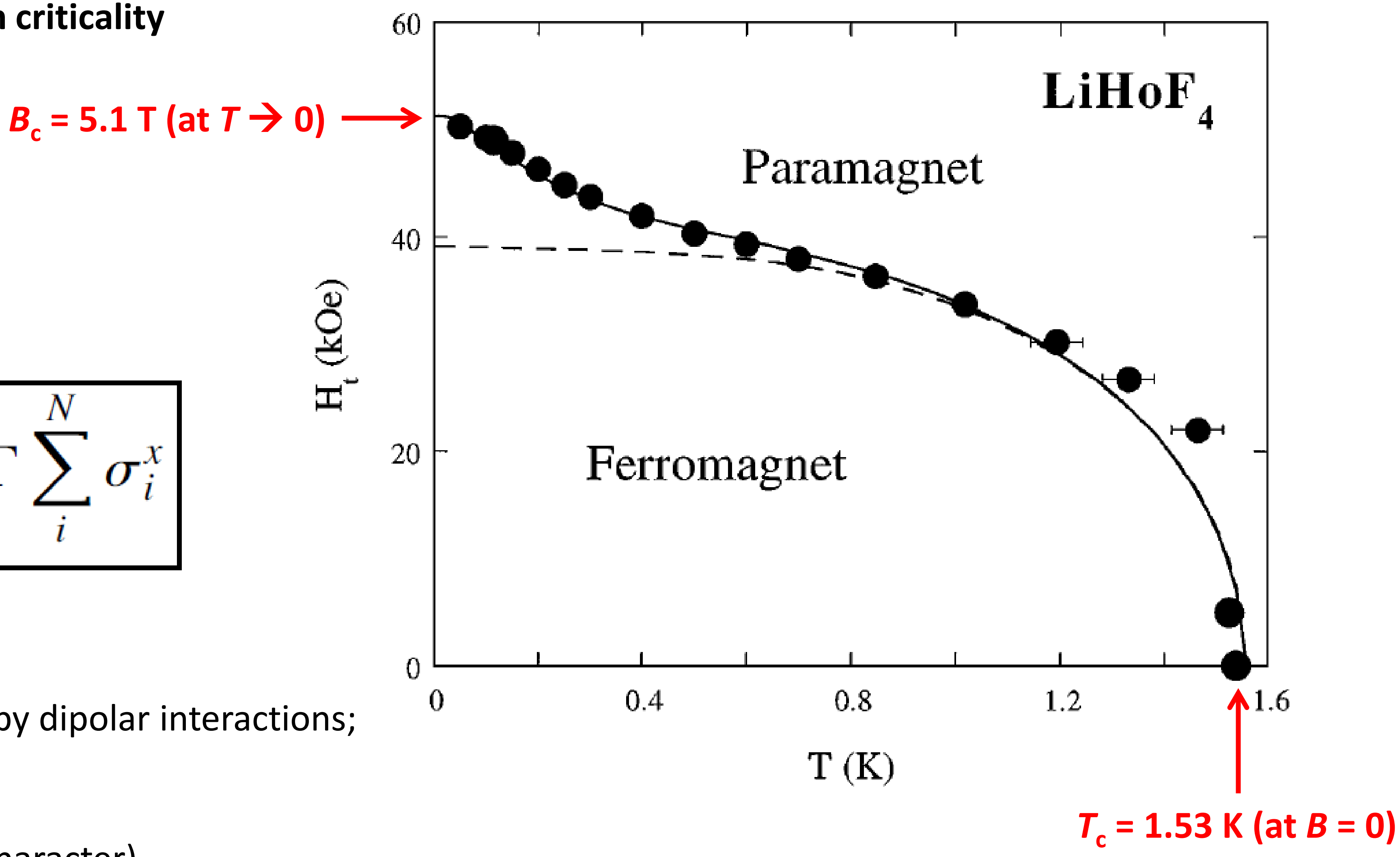
Ising model in transverse field:

$$\boxed{H = \sum_{i,j}^{N} J_{ij}\sigma_i^z\sigma_j^z - \Gamma\sum_{i}^{N}\sigma_i^x}$$

$LiHoF_4$: Magnetism dominated by dipolar interactions;

Exchange coupling small

(→ transition is of mean-field character)



Bitko / Rosenbaum / Aeppli, PRL **77**, 940 (1997)

$LiHoF_4$: Non-Kramers moments

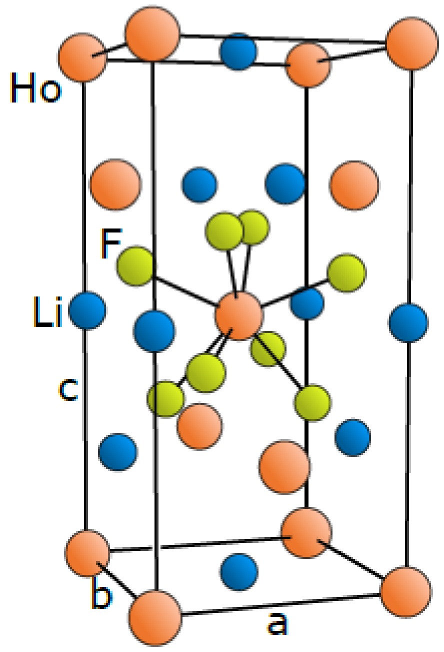
Ho^{4+}:

J=8 electronic moments

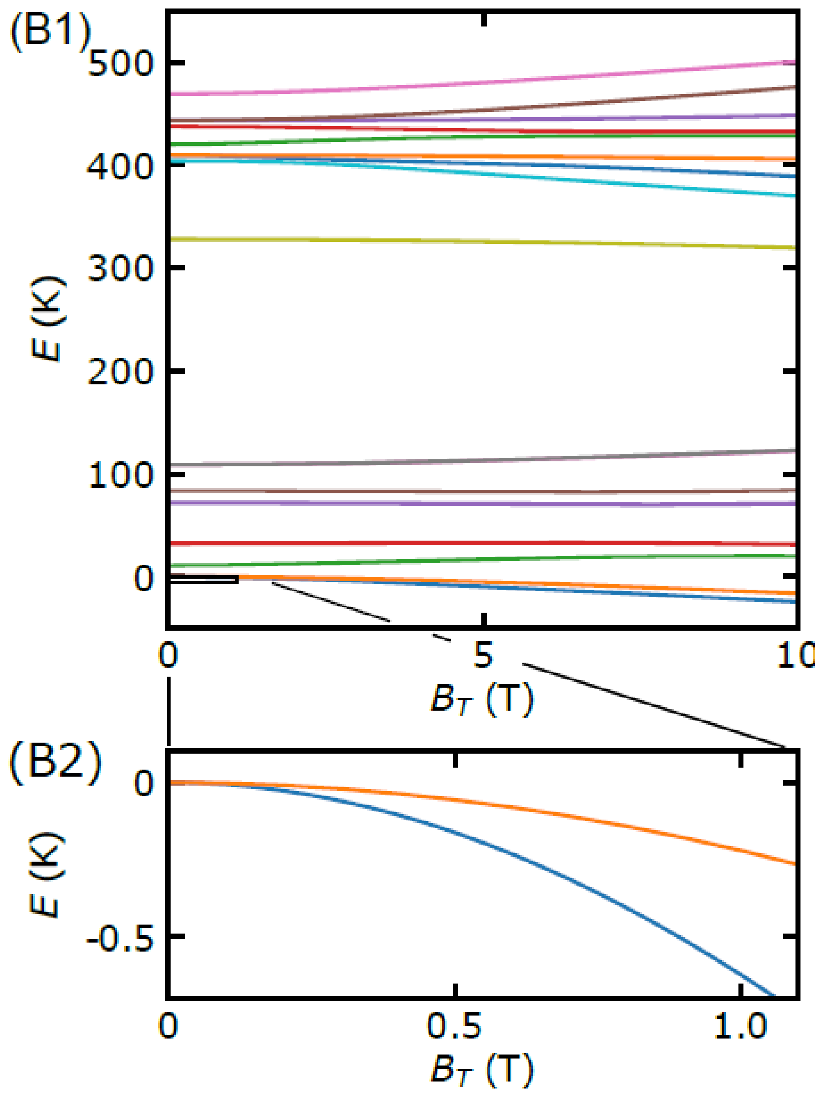
Lowest CEF state of Ho in $LiHoF_4$:

non-Kramers doublet (11 K gap to next level)

with large moment along z axis, but zero moment along x & y



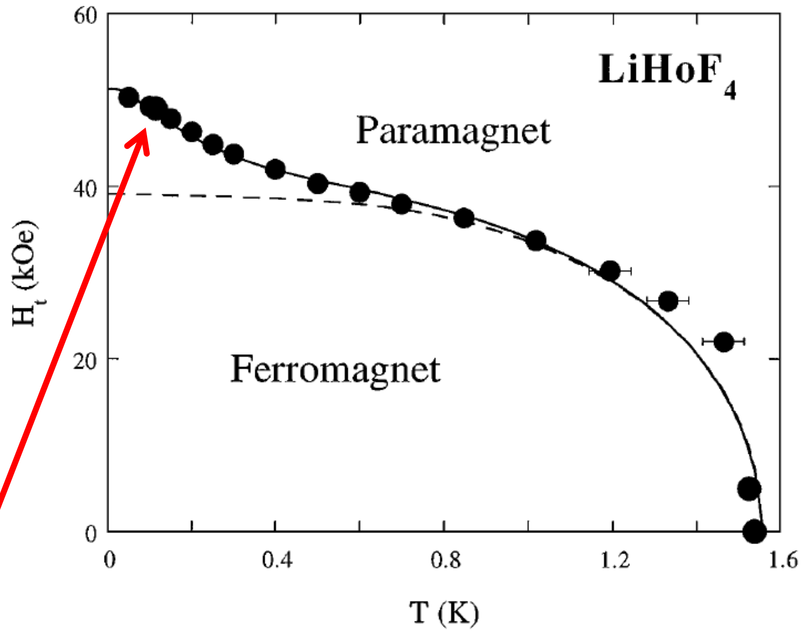
CEF levels as fct of transverse field



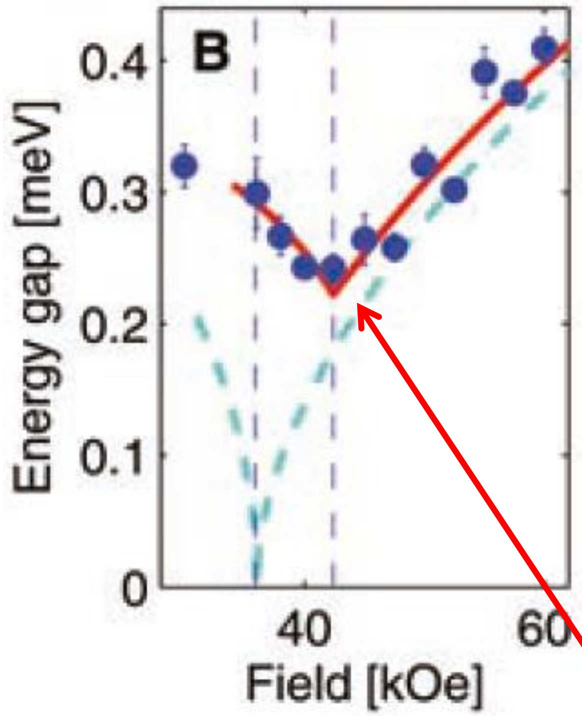
Hyperfine coupling in $LiHoF_4$

Ho: Nuclear spin I=7/2, hyperfine coupling A ≈ 0.04 K
→ Energy of hyperfine coupling of order 0.5 K

Excitations are mixed electro-nuclear modes, observable e.g. in neutron scattering



Hyperfine-induced stabilization of ordered phase



Incomplete softening of electronic mode at B_c

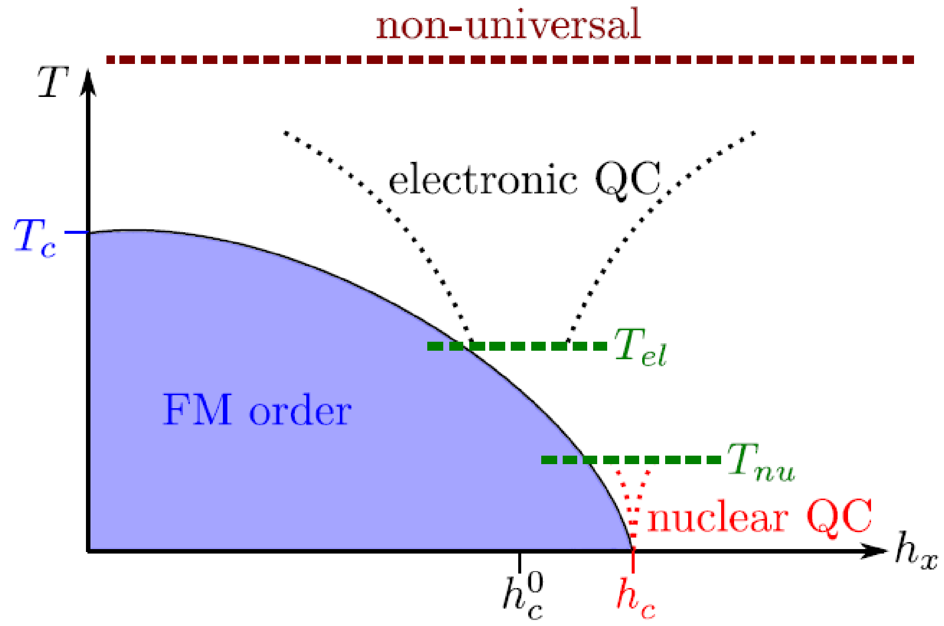
Bitko / Rosenbaum / Aeppli, PRL **77**, 940 (1997)

Ronnow *et al.*, Science **308**, 389 (2005)

QPT + nuclear spins: Two scenarios

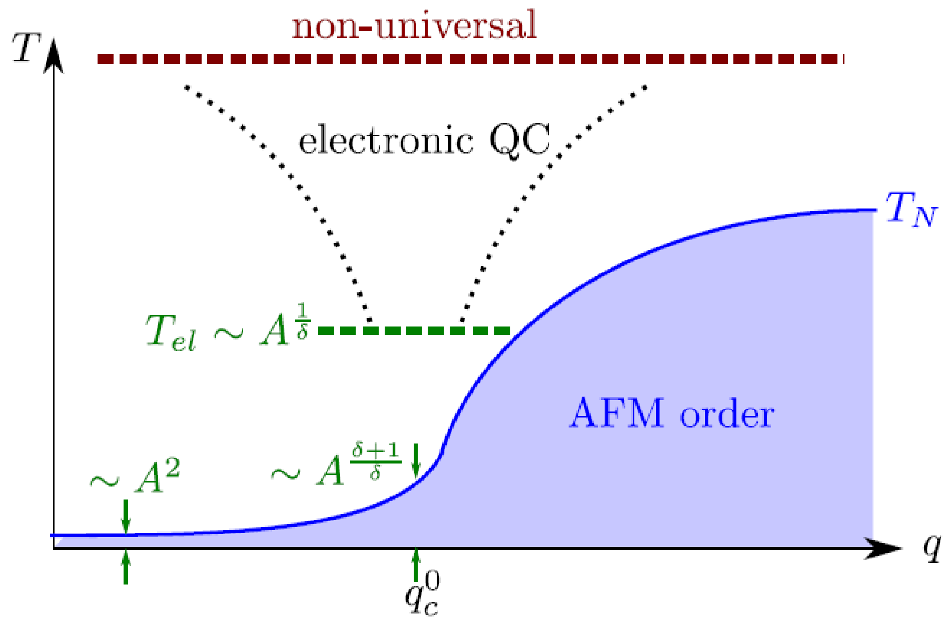
Time reversal broken near electronic QPT
(e.g. transverse-field Ising)

→ QPT shifted by hyperfine coupling



Time reversal unbroken near electronic QPT
(e.g. coupled dimers)

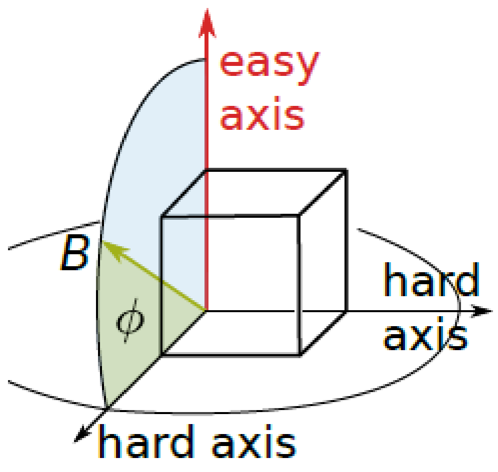
→ QPT smeared by hyperfine coupling



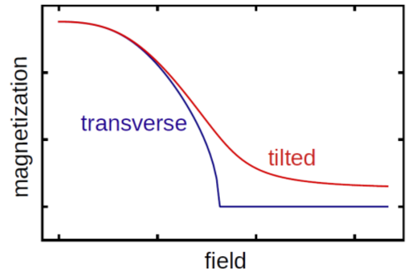
Eisenlohr / Vojta, PRB **103**, 064405 (2021)

Ising ferromagnet in tilted field (expectation)

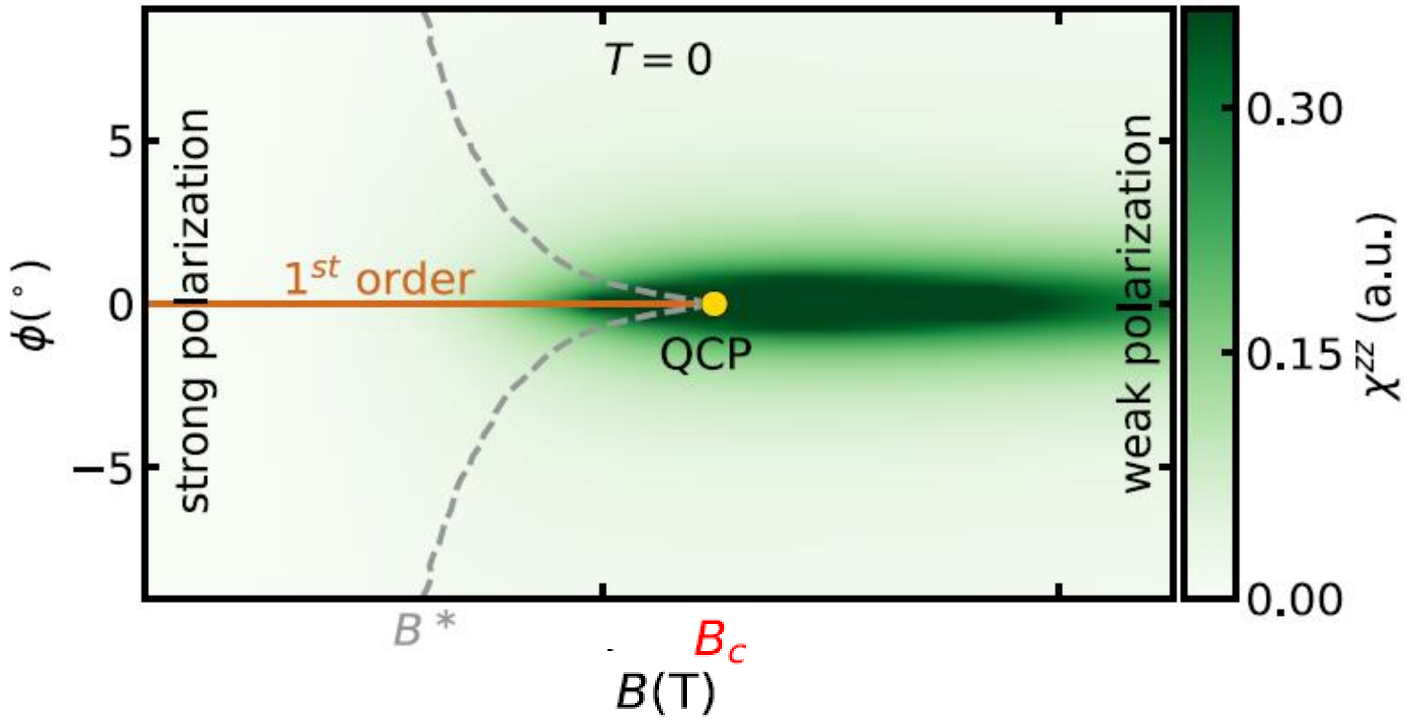
Apply field
tilted by angle ϕ from hard axis
(→ couples to order parameter)



Transition smeared into crossover for finite ϕ

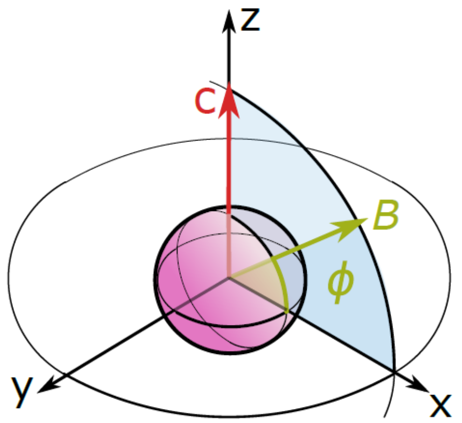


Crossover field B^* may be defined from susceptibility maximum.
Landau theory: $B^* - B_c \sim \phi^{2/3}$



$LiHoF_4$: Susceptibility in tilted fields

Magnetic ac susceptibility
along easy axis as function of field
for different angles ϕ



LiHoF$_4$: Susceptibility in tilted fields

Magnetic ac susceptibility
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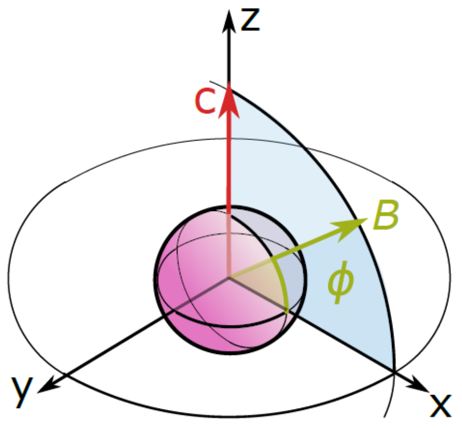
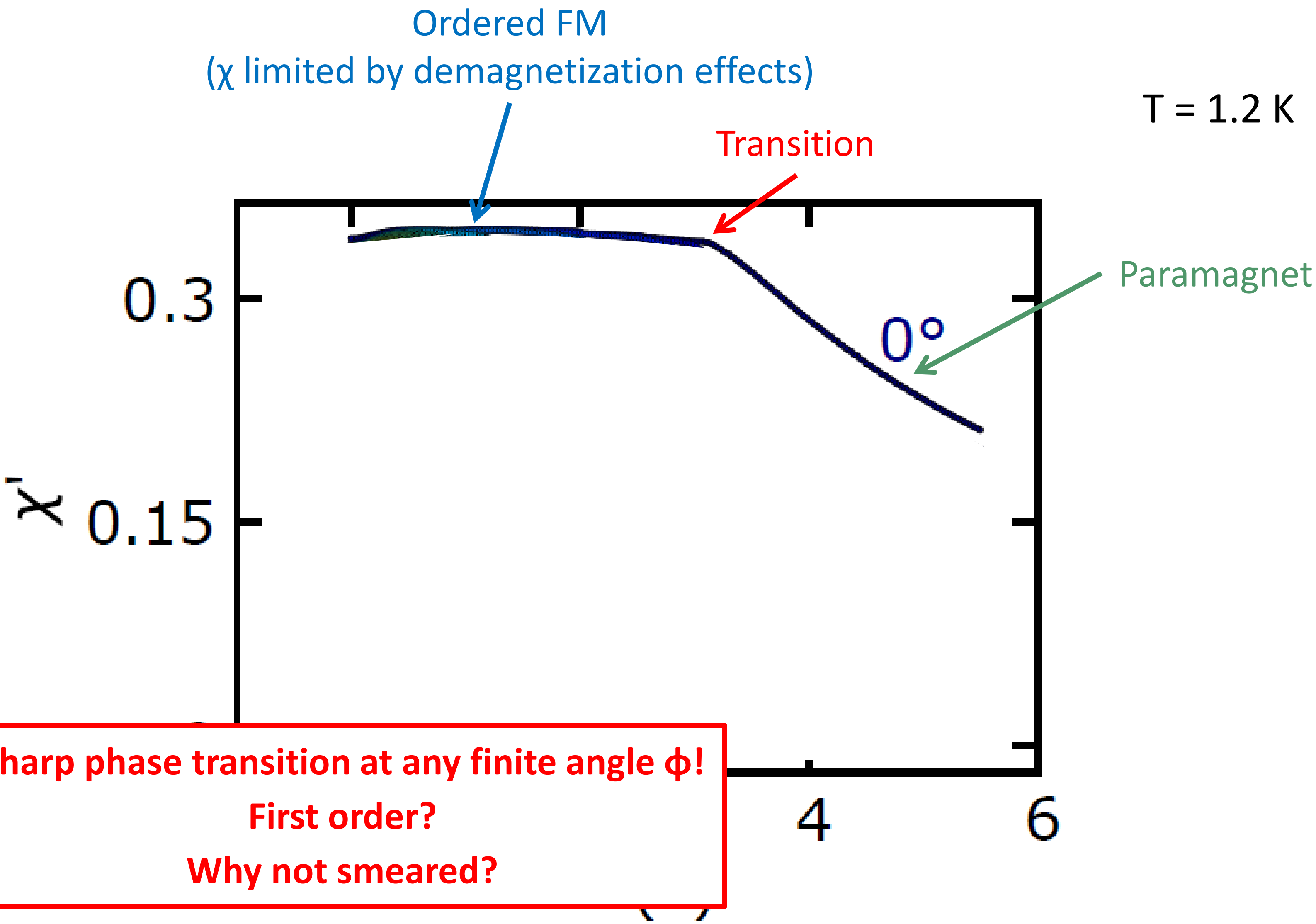


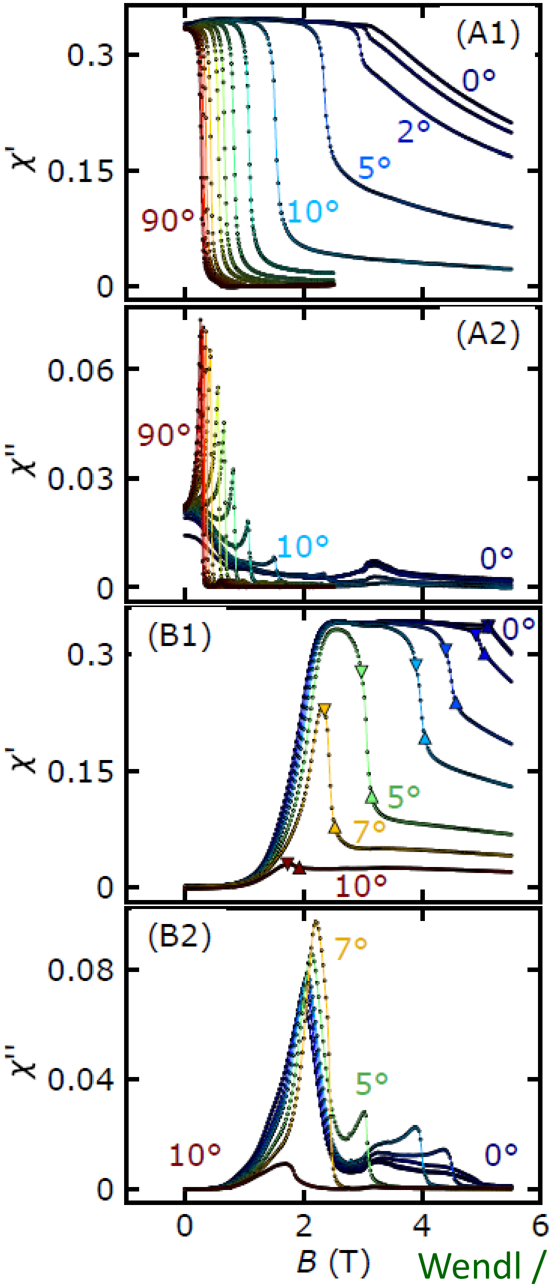


Sharp phase transition at any finite angle ϕ!
First order?
Why not smeared?

Wendl / Eisenlohr *et al.*, Nature **609**, 65 (2022)

$LiHoF_4$: More susceptibility in tilted fields

Magnetic ac susceptibility along easy axis



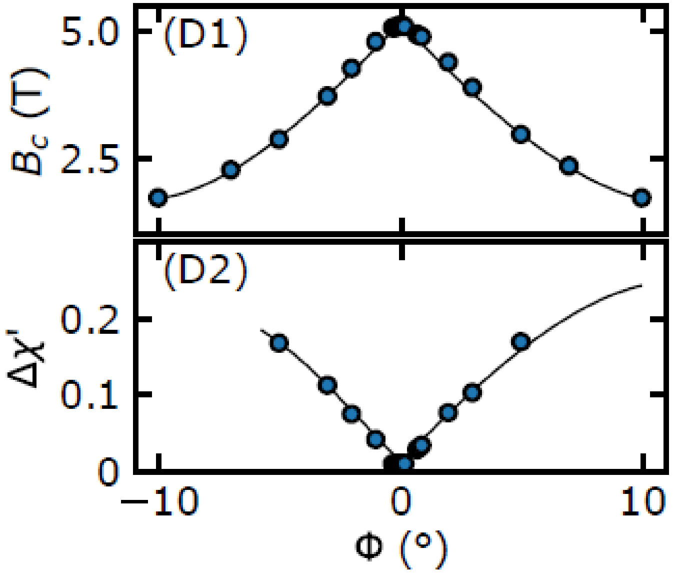
T = 1.2 K

T = 60 mK

As T→0:

Transition field very sensitive to ϕ

Height of jump in χ

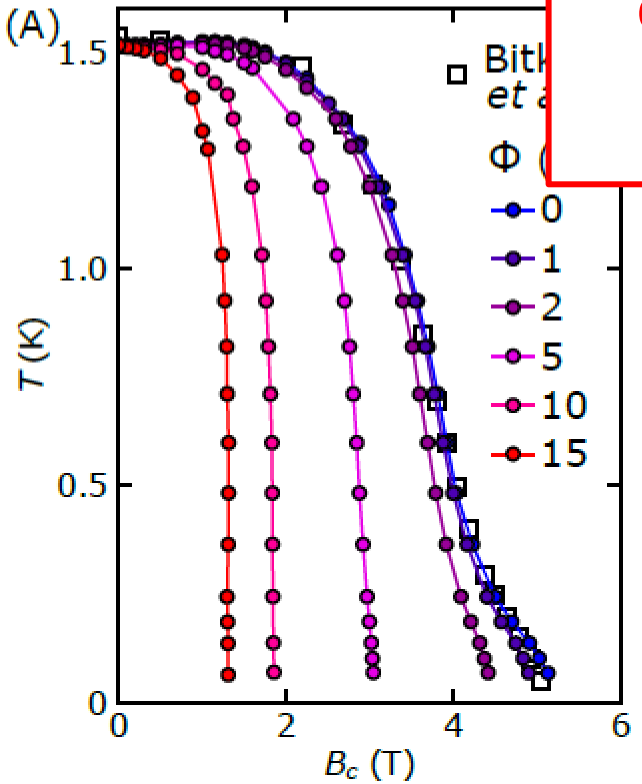


Wendl / Eisenlohr *et al.*, Nature **609**, 65 (2022)

$LiHoF_4$: Phase diagram in tilted fields

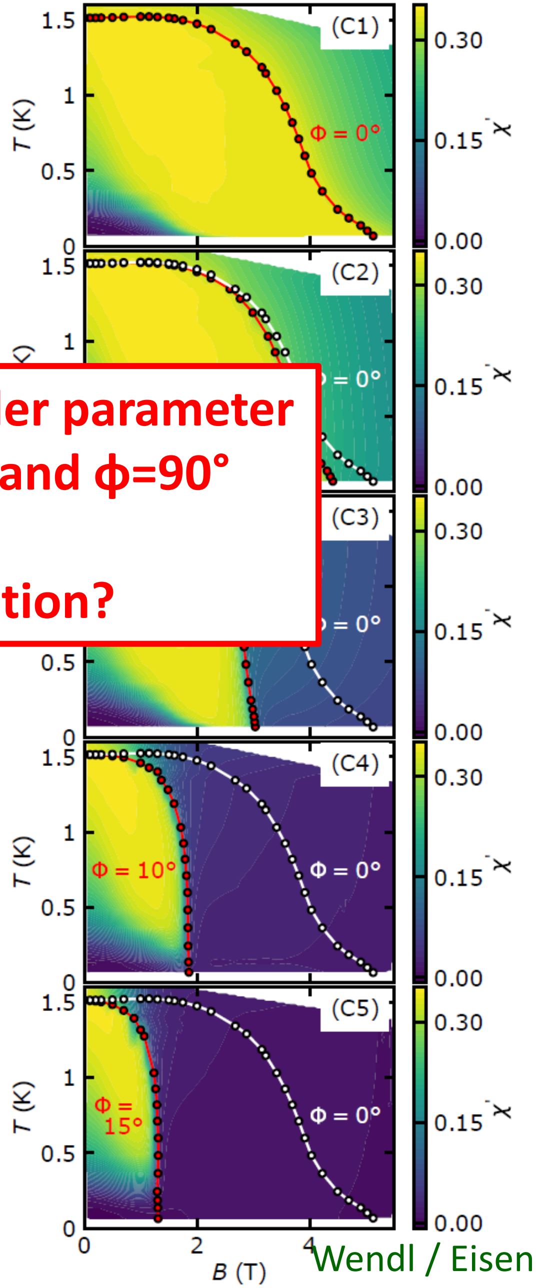
Transition field strongly suppressed with increasing ϕ

Hyperfine-induced ``nose´´ disappears beyond ϕ=5°



Physics beyond microscopic order parameter
Continuity between small ϕ and ϕ=90°

→ domain-driven transition?



Wendl / Eisenlohr *et al.*, Nature **609**, 65 (2022)

Modelling including domains

Microscopic ingredients

$$H_{\mathrm{mic}} = -K \sum_{\langle ij \rangle} \mathbf{J}_i \cdot \mathbf{J}_j + \sum_i [V_{\mathrm{CF}}(\mathbf{J}_i) + A\mathbf{J}_i \cdot \mathbf{I}_i] - \mu_B \mathbf{B} \cdot \sum_i (g\mathbf{J}_i + g_N \mathbf{I}_i)$$

All CEF levels (required for tilted fields!)
All nuclear levels + hyperfine coupling

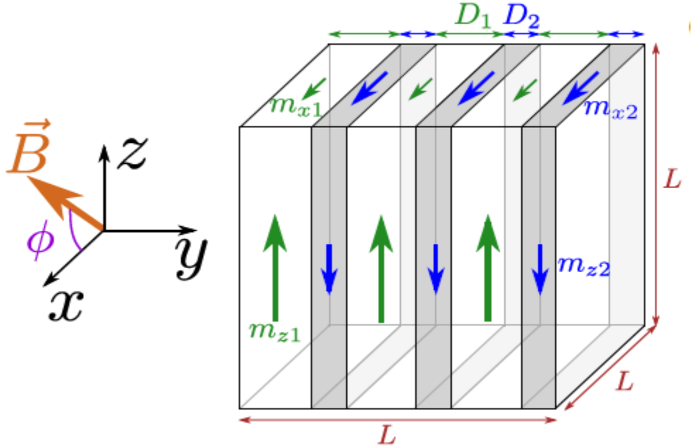
Interactions approximated as Heisenberg,
treated at mean-field level

→ m-f theory with 17x8-dim Hilbert space

Chakraborty *et al.*, PRB **70**, 144411 (2004)
Tabei *et al.*, PRB **78**, 184408 (2008)
McKenzie *et al.*, PRB **97**, 214430 (2018)

Domains

Sheet-like domains (assumed)



Stray-field energy computed exactly

$$\mu_0 \int d^2r d^2r' \frac{\sigma(\mathbf{r})\sigma(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$$

taken from Monte Carlo

Biltmo *et al.*, EPL **87**, 27007 (2009)

ational parameters

Combine into consistent mean-field theory for transverse-field Ising ferromagnet w/ domains

Stray fields induce effective antiferromagnetic coupling between domains!

Coupled mean-field theory

Microscopic part, separately for each domain:

$$H_{\mathrm{mic}}^{\mathrm{MF}} = -nK(\mathbf{J}\cdot\bar{\mathbf{J}} - \frac{\bar{\mathbf{J}}^2}{2}) + V_{\mathrm{CF}}(\mathbf{J}) + A\mathbf{J}\cdot\mathbf{I} - \mu_B\mathbf{B}\cdot(g\mathbf{J} + g_N\mathbf{I})$$

Domain energies, written as effective interaction for domains 1 and 2:

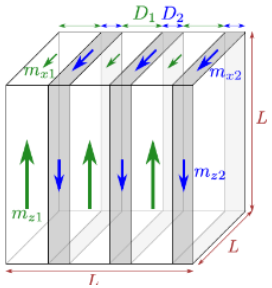
$$E_{\mathrm{dom}} = M\sum_{\alpha}\left(c_1^{\alpha}\bar{J}_1^{\alpha}\bar{J}_1^{\alpha} + c_2^{\alpha}\bar{J}_2^{\alpha}\bar{J}_2^{\alpha} + c_{12}^{\alpha}\bar{J}_1^{\alpha}\bar{J}_2^{\alpha}\right)$$

Combined m-f theory:

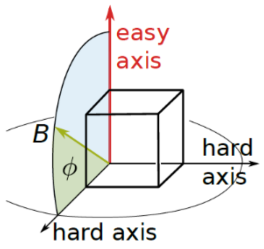
$$\begin{aligned}
H_1^{\mathrm{MF}} &= \left(-\frac{n}{2}K + \frac{c_1^x}{1-v}\right)\left(2\bar{J}_1^x J_1^x - (\bar{J}_1^x)^2\right) \\
&+ \frac{c_{12}^x}{1-v}\left(\bar{J}_2^x J_1^x - \frac{1}{2}\bar{J}_1^x\bar{J}_2^x\right) \\
&+ \left(-\frac{n}{2}K + \frac{c_1^z}{1-v}\right)\left(2\bar{J}_1^z J_1^z - (\bar{J}_1^z)^2\right) \\
&+ \frac{c_{12}^z}{1-v}\left(\bar{J}_2^z J_1^z - \frac{1}{2}\bar{J}_1^z\bar{J}_2^z\right) \\
&- \frac{n}{2}K\left(2\bar{J}_1^y J_1^y - (\bar{J}_1^y)^2\right) + H_{\mathrm{ion}}(\hat{\mathbf{J}}_1),
\end{aligned}$$

$$\begin{aligned}
H_2^{\mathrm{MF}} &= \left(-\frac{n}{2}K + \frac{c_2^x}{v}\right)\left(2\bar{J}_2^x J_2^x - (\bar{J}_2^x)^2\right) \\
&+ \frac{c_3^x}{v}\left(\bar{J}_1^x J_2^x - \frac{1}{2}\bar{J}_1^x\bar{J}_2^x\right) \\
&+ \left(-\frac{n}{2}K + \frac{c_2^z}{v}\right)\left(2\bar{J}_2^z J_2^z - (\bar{J}_2^z)^2\right) \\
&+ \frac{c_{12}^z}{v}\left(\bar{J}_1^z J_2^z - \frac{1}{2}\bar{J}_1^z\bar{J}_2^z\right) \\
&- \frac{n}{2}K\left(2\bar{J}_2^y J_2^y - (\bar{J}_2^y)^2\right) + H_{\mathrm{ion}}(\hat{\mathbf{J}}_2),
\end{aligned}$$

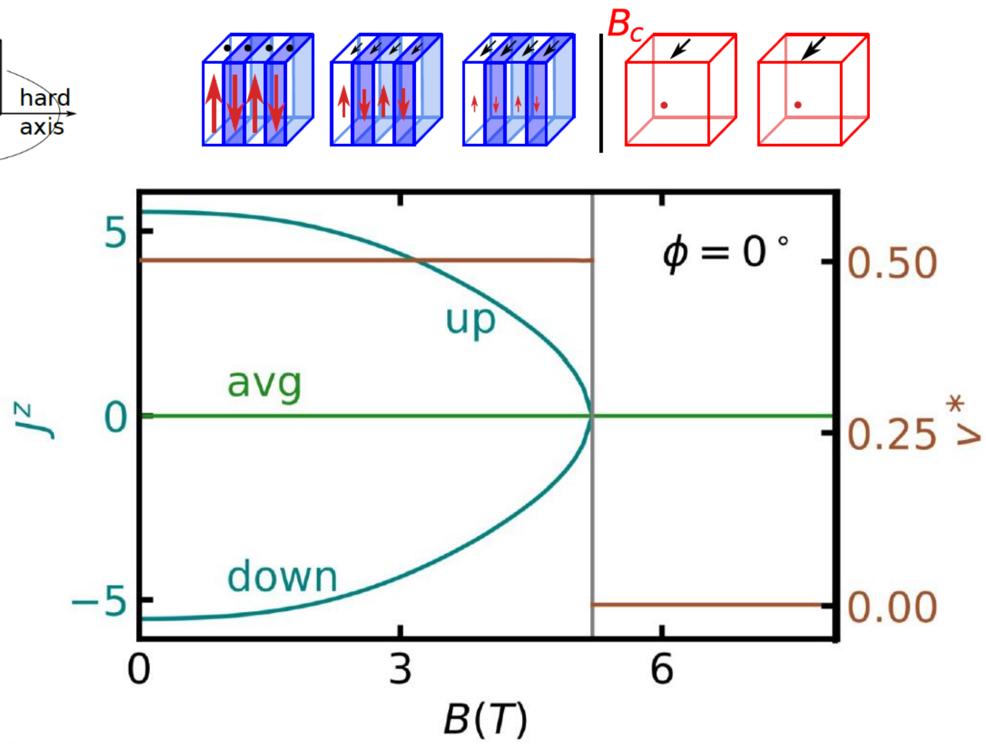
Results: Phase transitions and domains



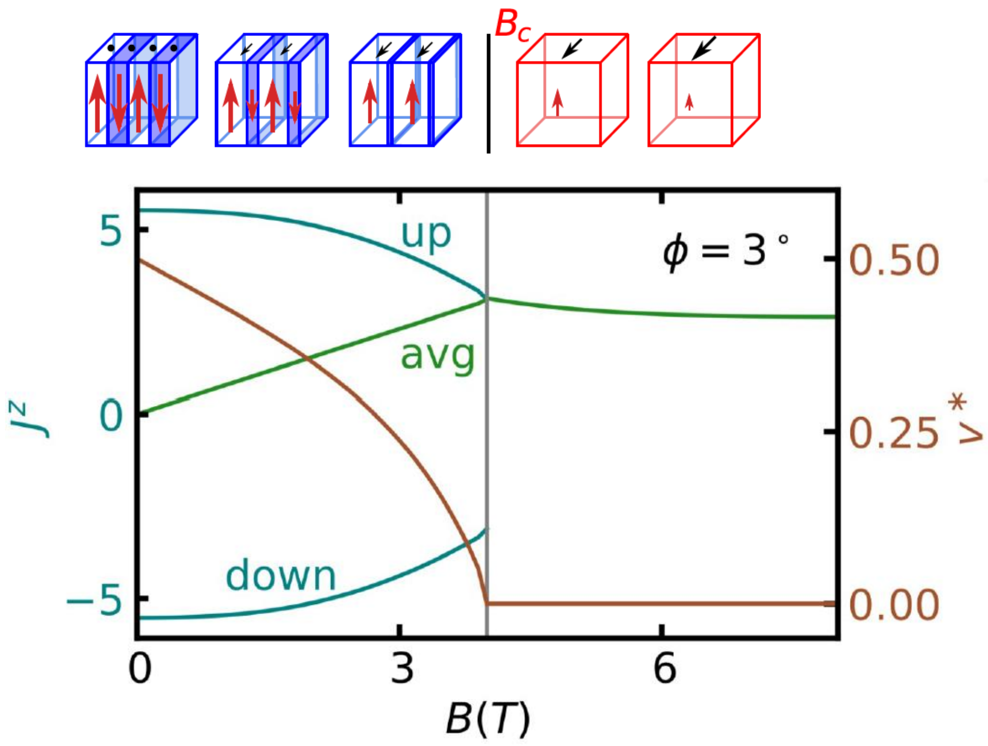
Key quantity: Domain volume ratio $v = D_2 / (D_1+D_2)$



Transverse field



Tilted field

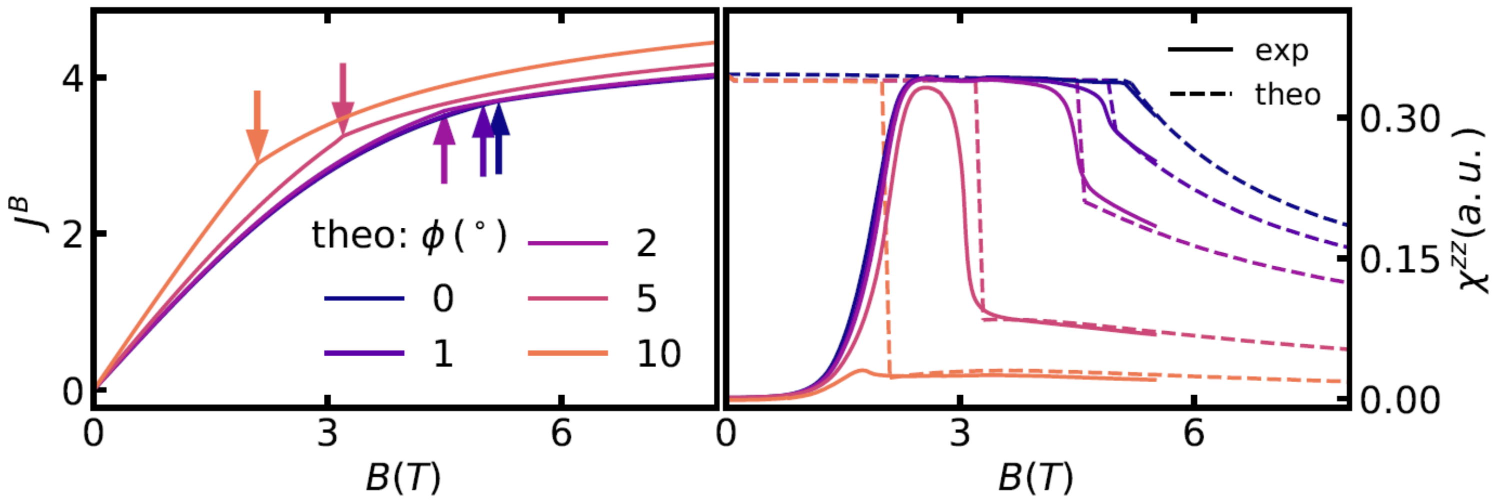


Domains vanish at microscopic transition

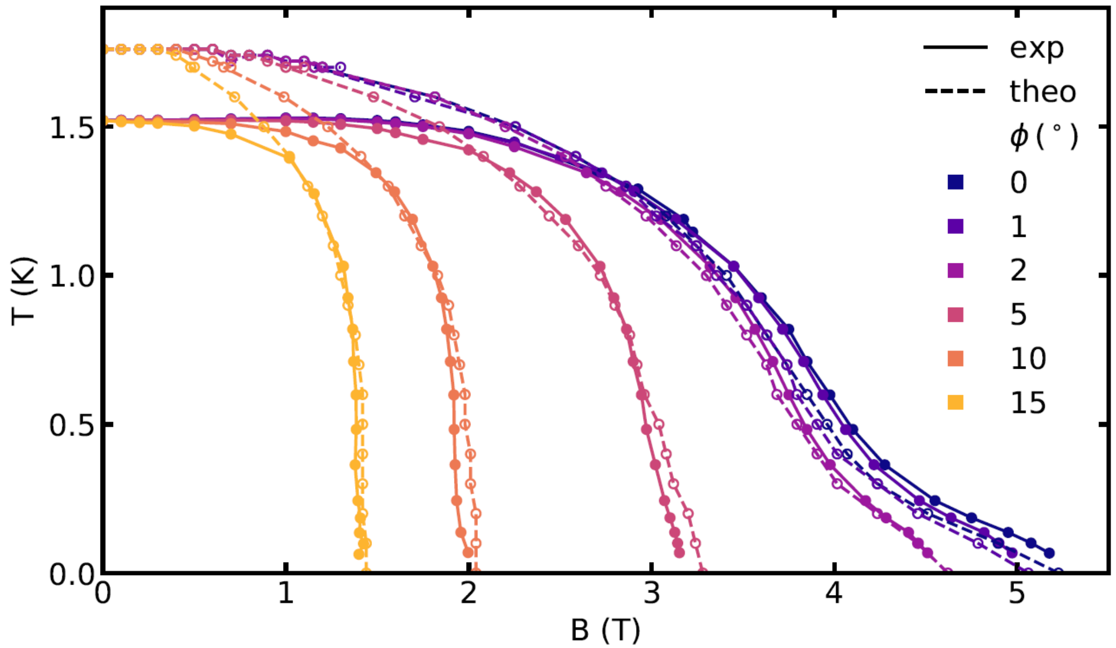
Minority domains are expelled

Theory vs. experiment

Magnetization along field | Susceptibility along easy axis



Phase diagram



Excellent quantitative agreement (*J* is only fit parameter)

Wendl / Eisenlohr *et al.*, Nature **609**, 65 (2022)

Non-Kramers moments & nuclear spins

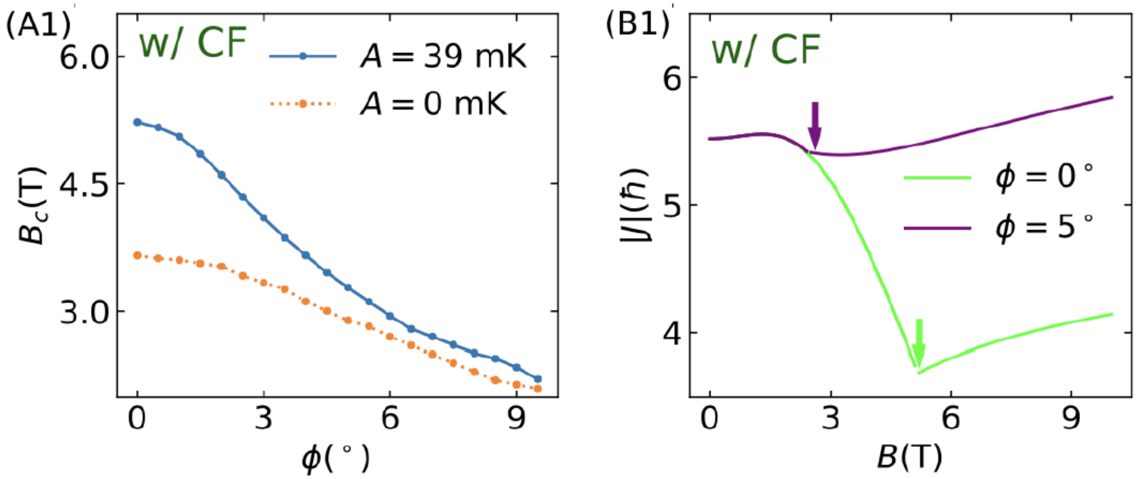
Why is ``hyperfine nose“ suppressed
for tilted fields?

Interplay of non-Kramers & hyperfine physics

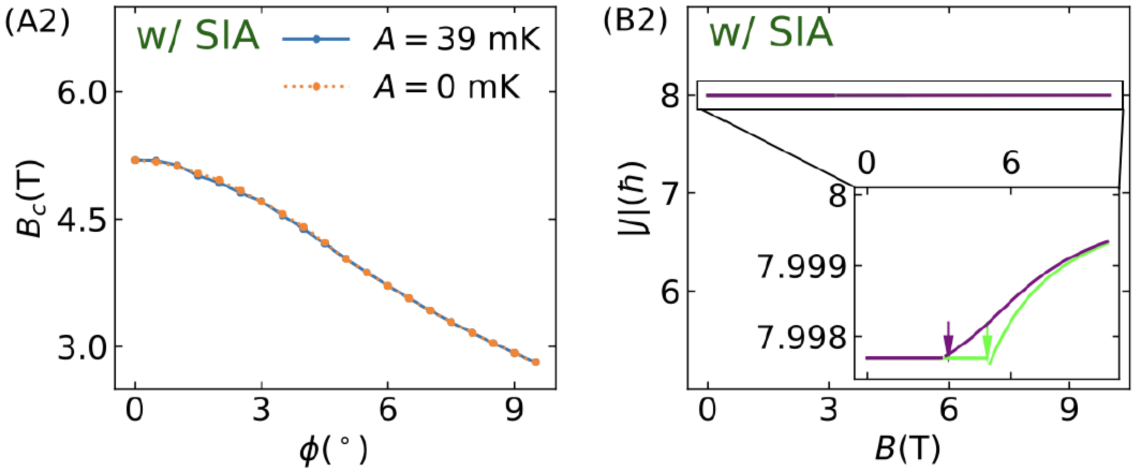
ϕ=0:
Electronic moment |*J*| strongly varies as fct of B
(because *x* component is small)
→ hyperfine coupling gains more energy in FM phase
→ FM phase stabilized at low T

ϕ>5°:
|*J*| variation weak
→ hyperfine coupling does no longer stabilize FM phase

Calculation w/ full CEF Hamiltonian



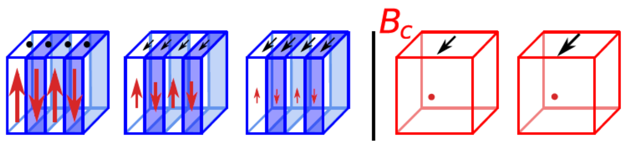
Calculation w/ Kramers + single-ion anisotropy



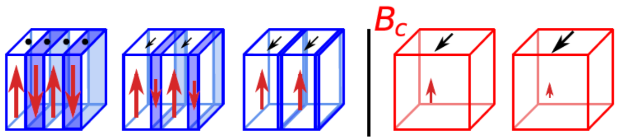
Quantum phase transitions

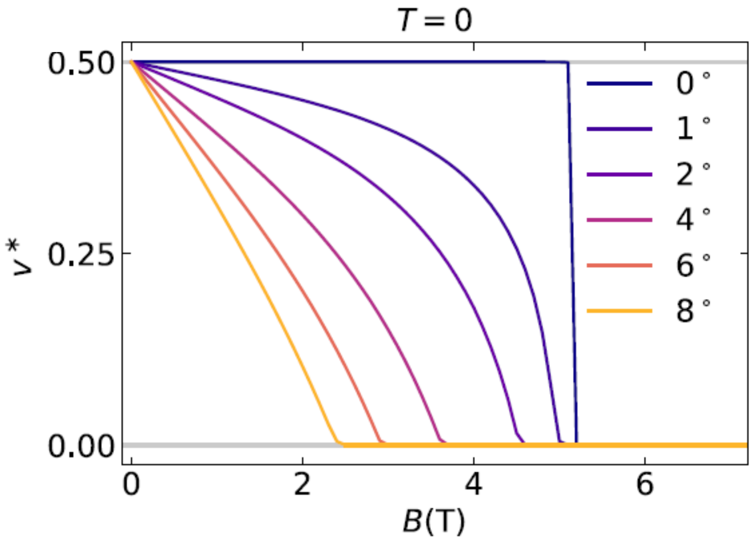
Two types of (continuous) quantum phase transitions in $LiHoF_4$:

1) Multi-domain ferromagnet ←→ Single-domain paramagnet for ϕ=0
   Domains disappear by vanishing magnetization
   Exponents α=0, β=1/2, γ=1, δ=3 (mean-field) due to dipolar interactions



2) Multi-domain ←→ Single-domain for ϕ≠0
   Domains disappear by vanishing minority domain volume
   Exponents α=0, β=1, γ=0, δ=1

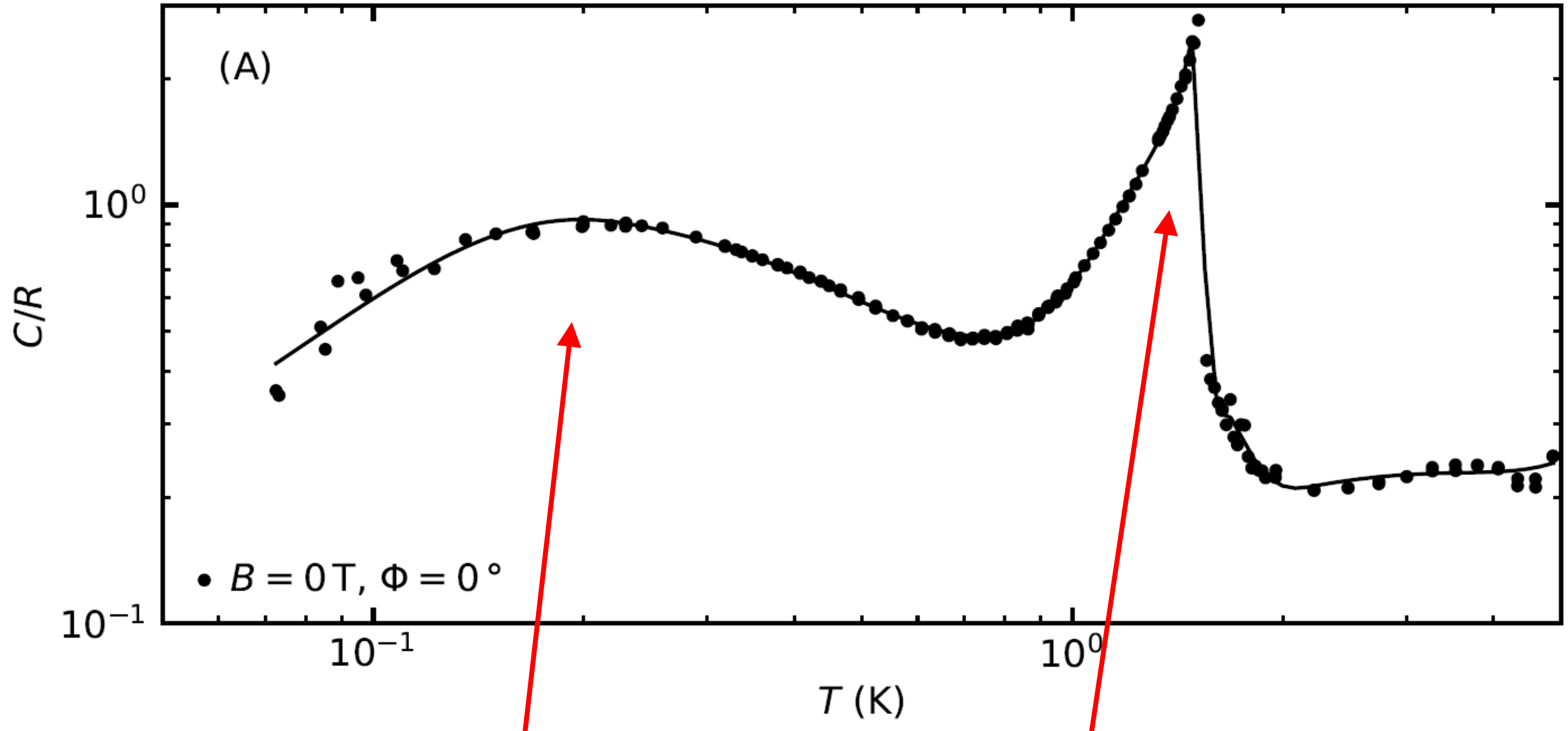




Mesoscale antiferromagnetism

Wendl / Eisenlohr *et al.*, Nature **609**, 65 (2022)

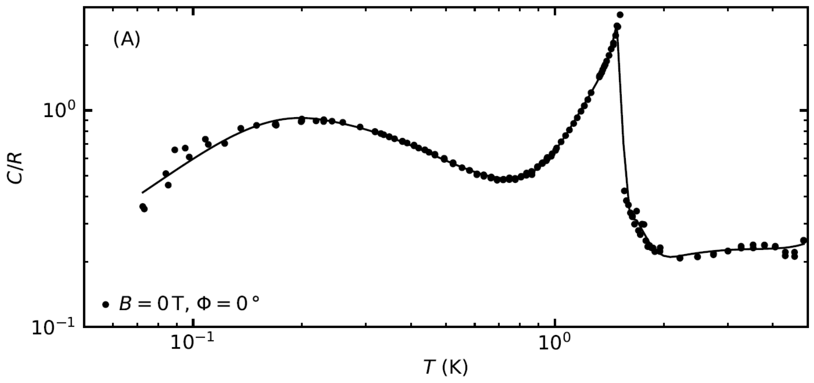
Heat capacity of $LiHoF_4$

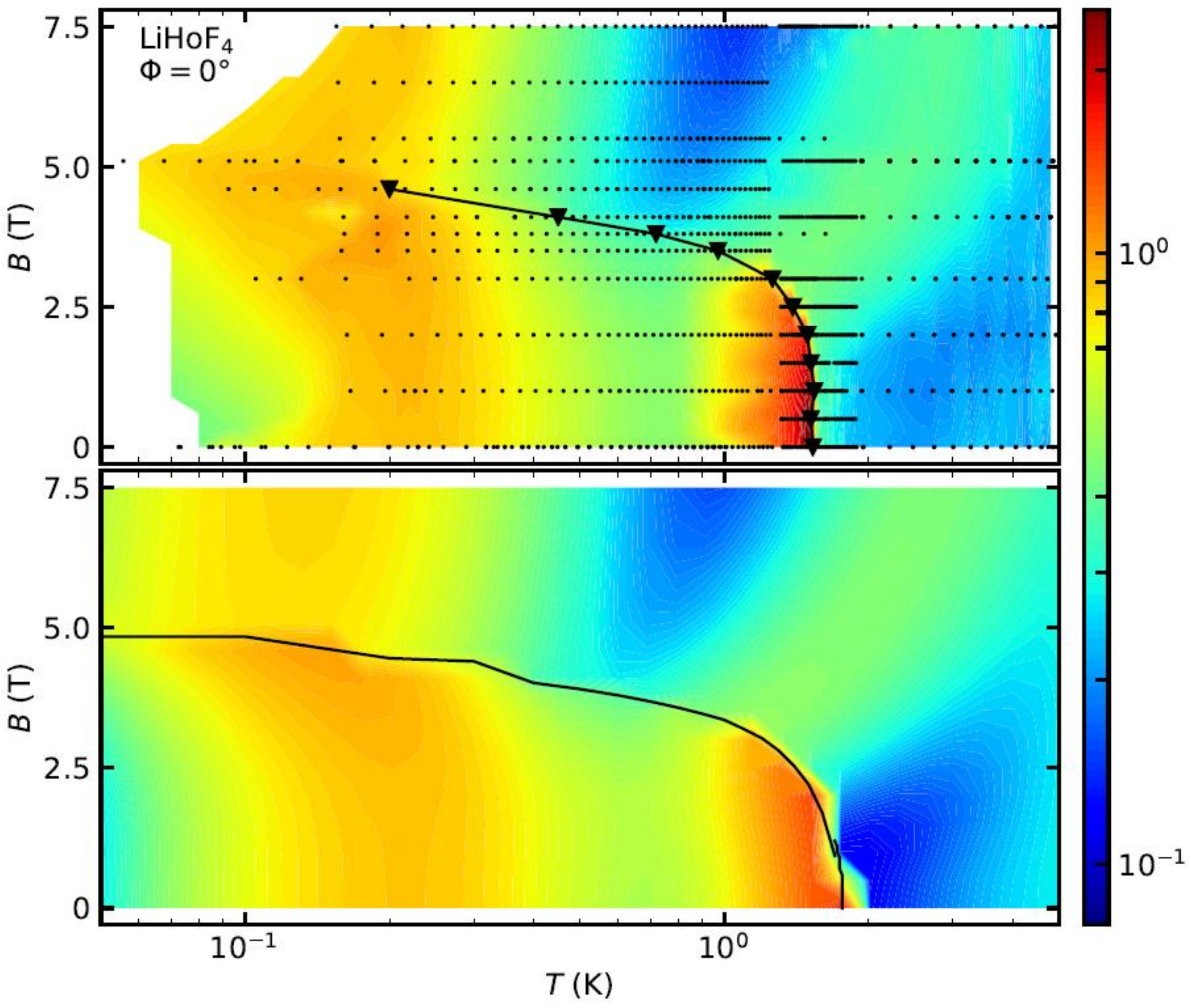


Nuclear spins

Transition

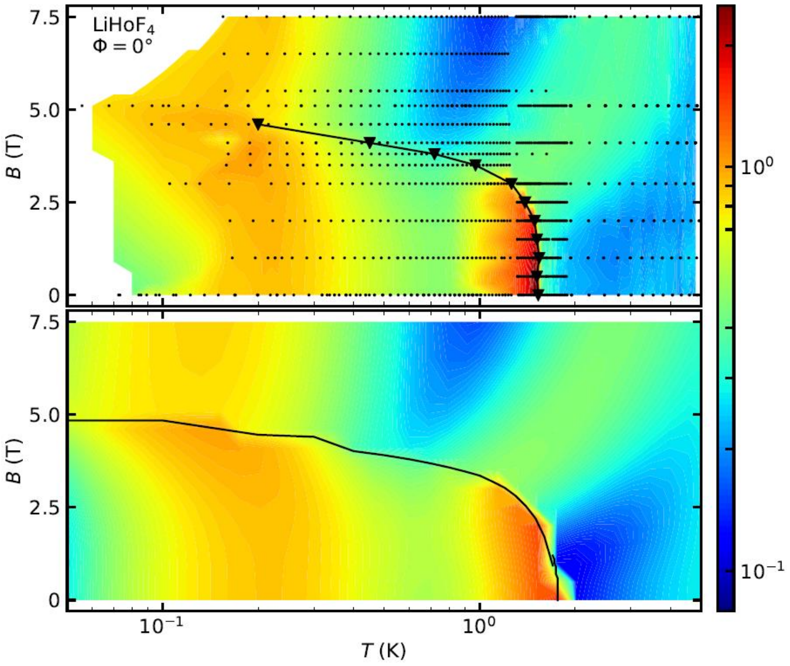
Heat capacity of $LiHoF_4$: transverse field

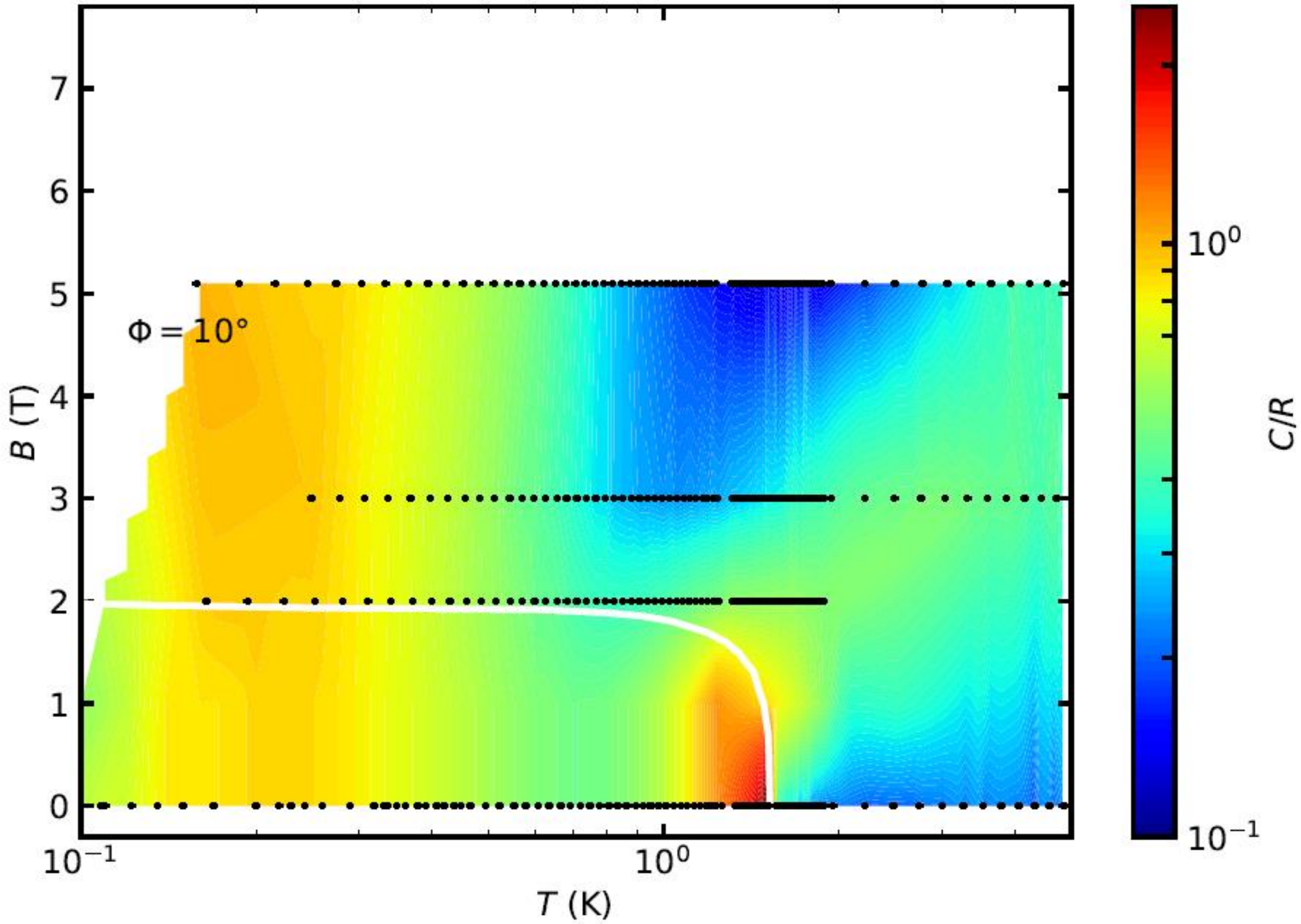




Wendl *et al.*, unpublished

Heat capacity of $LiHoF_4$: transverse & tilted field





Wendl *et al.*, unpublished

Summary & Outlook

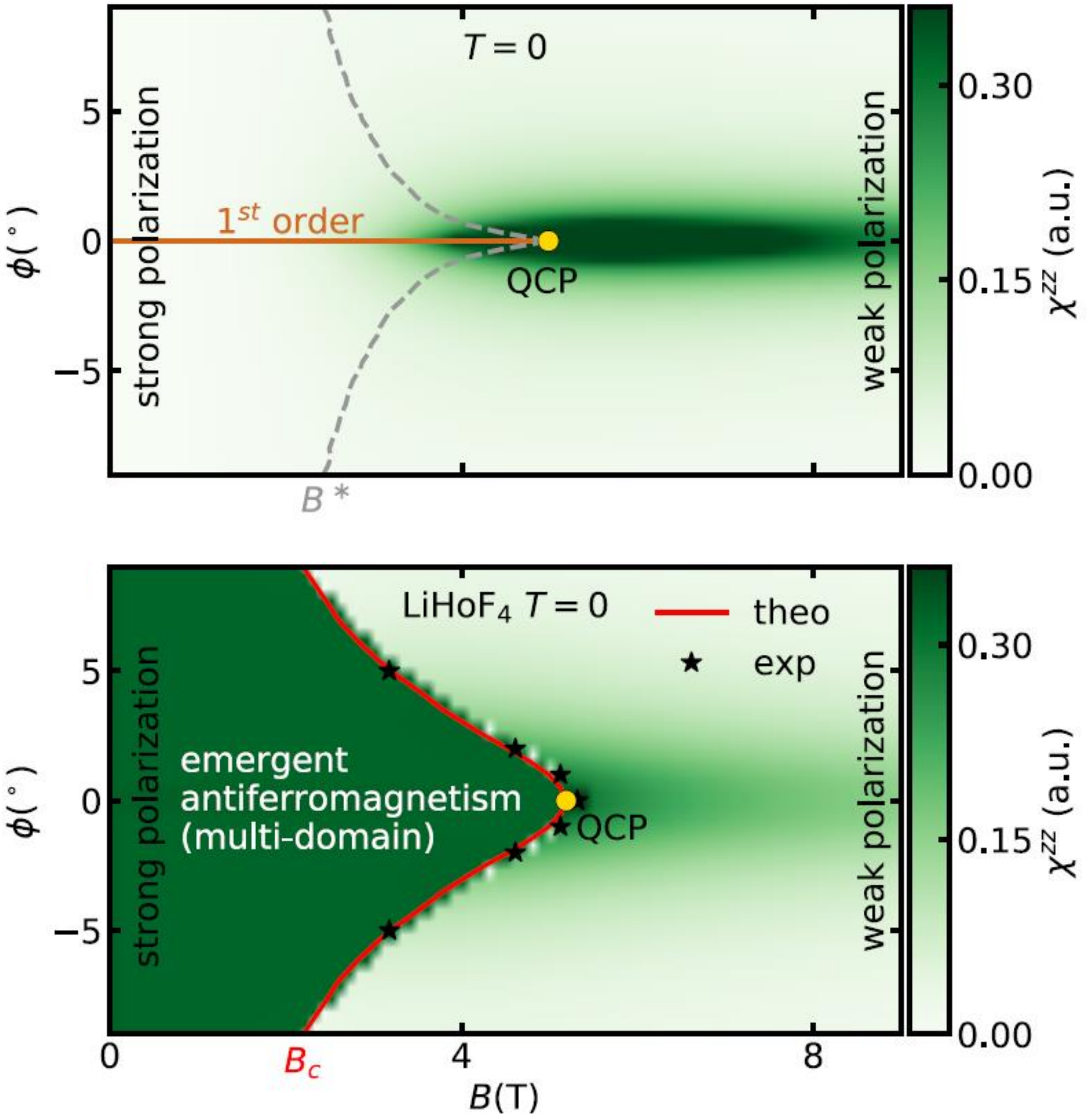
$LiHoF_4$ displays sharp phase transition even under tilted fields.

Theory including domains is in excellent agreement w/ experiment.

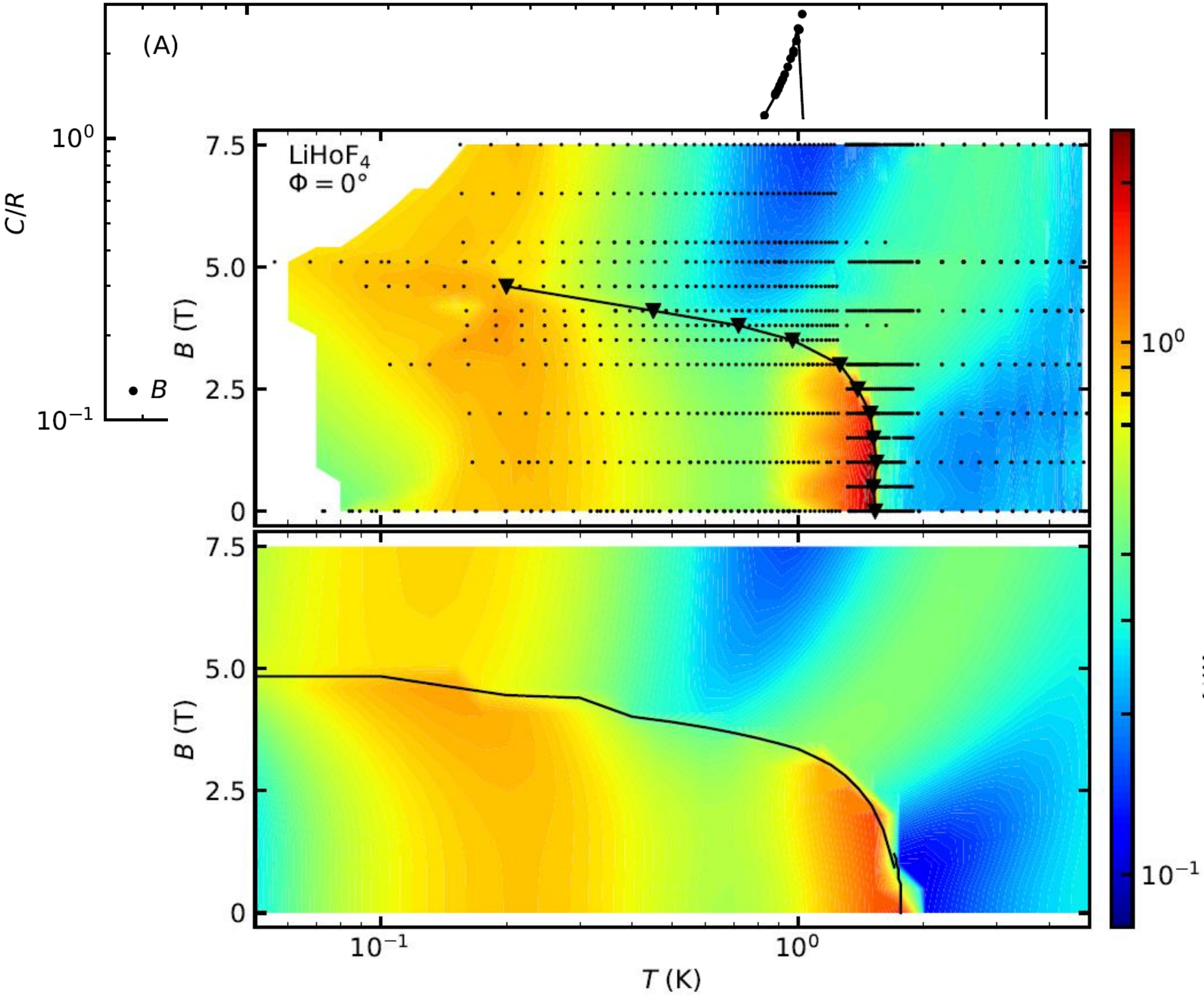
Open questions:

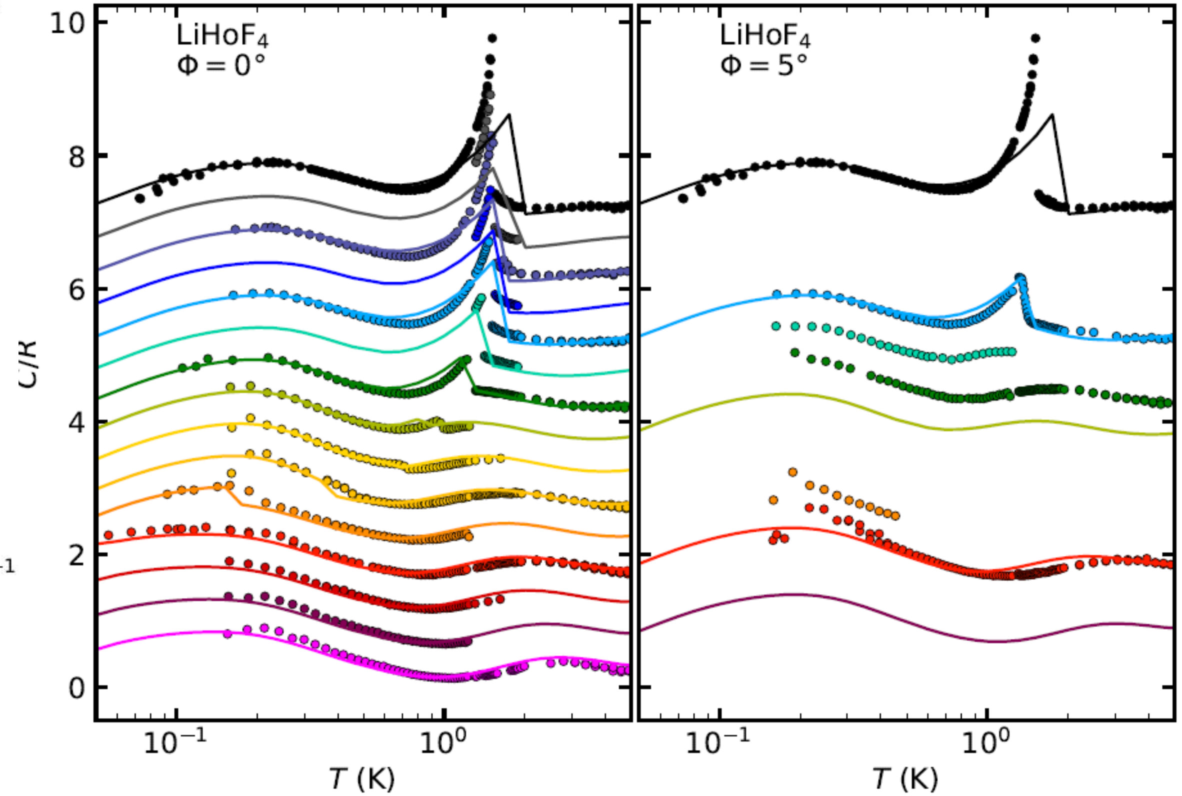
Are domain-driven transitions always of m-f type?
Role of (quantum) fluctuations?

Related transitions in
other domain-forming systems (ferroelectrics)?

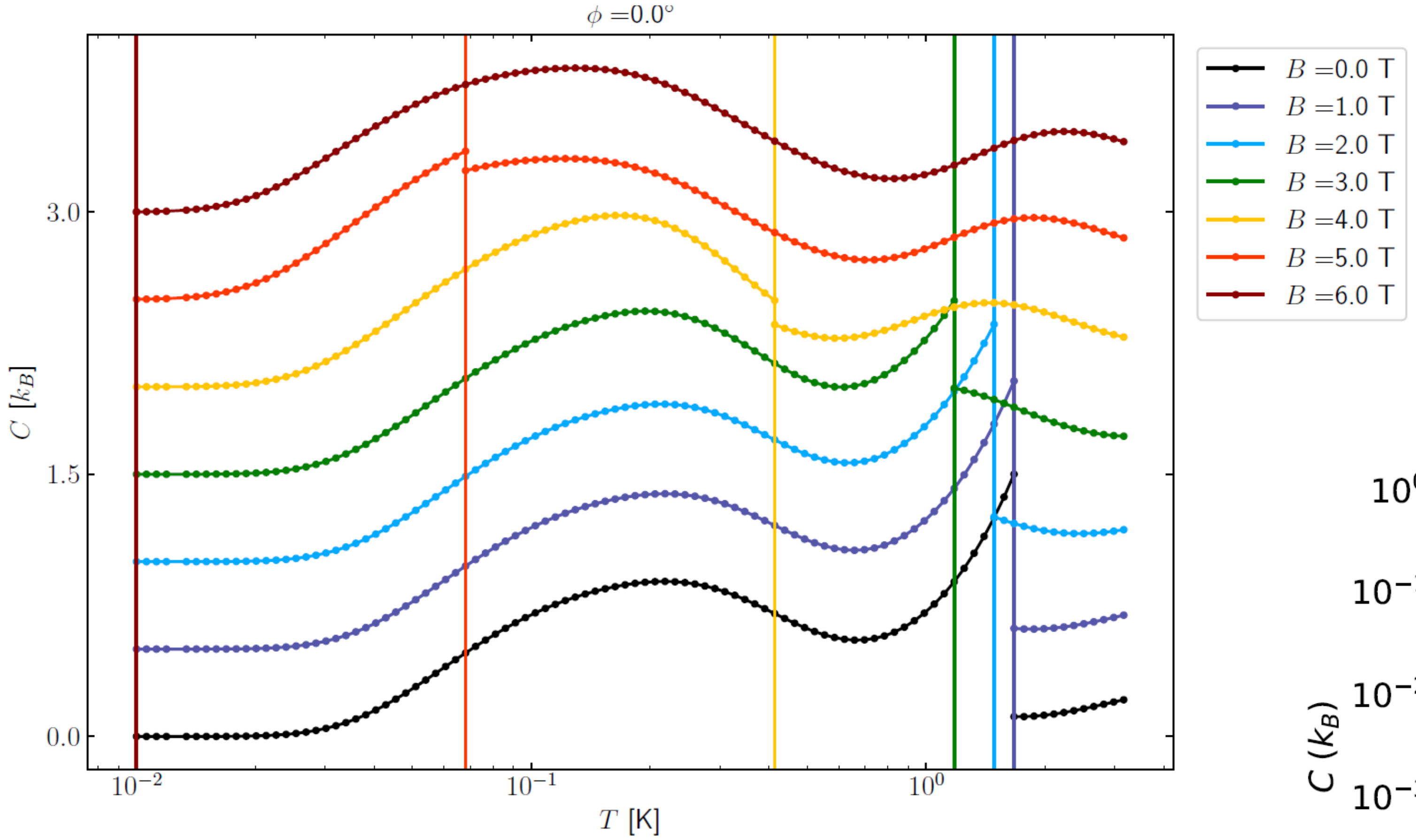


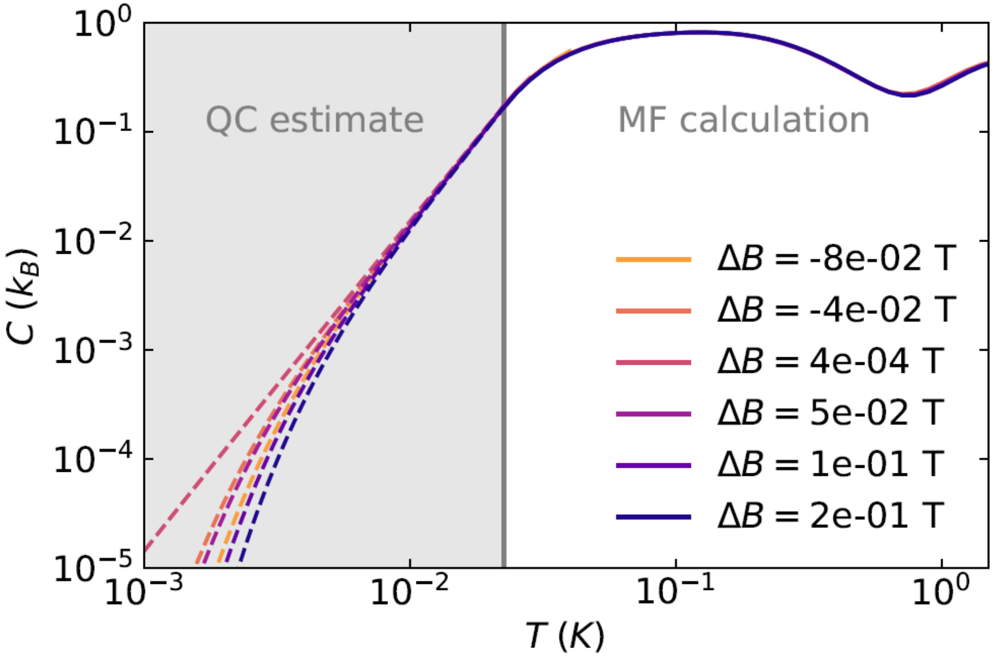
Heat capacity - experiment





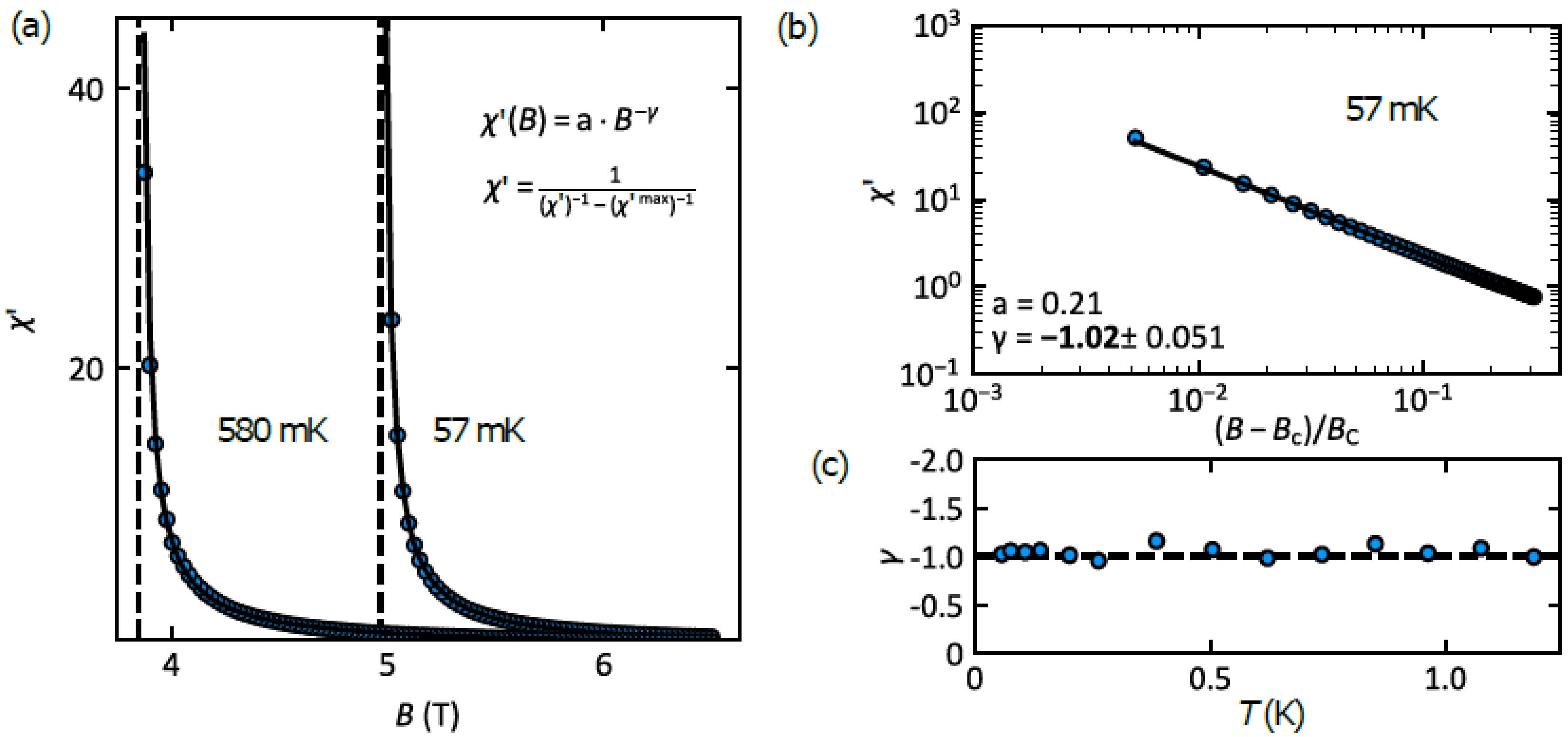
Heat capacity - theory



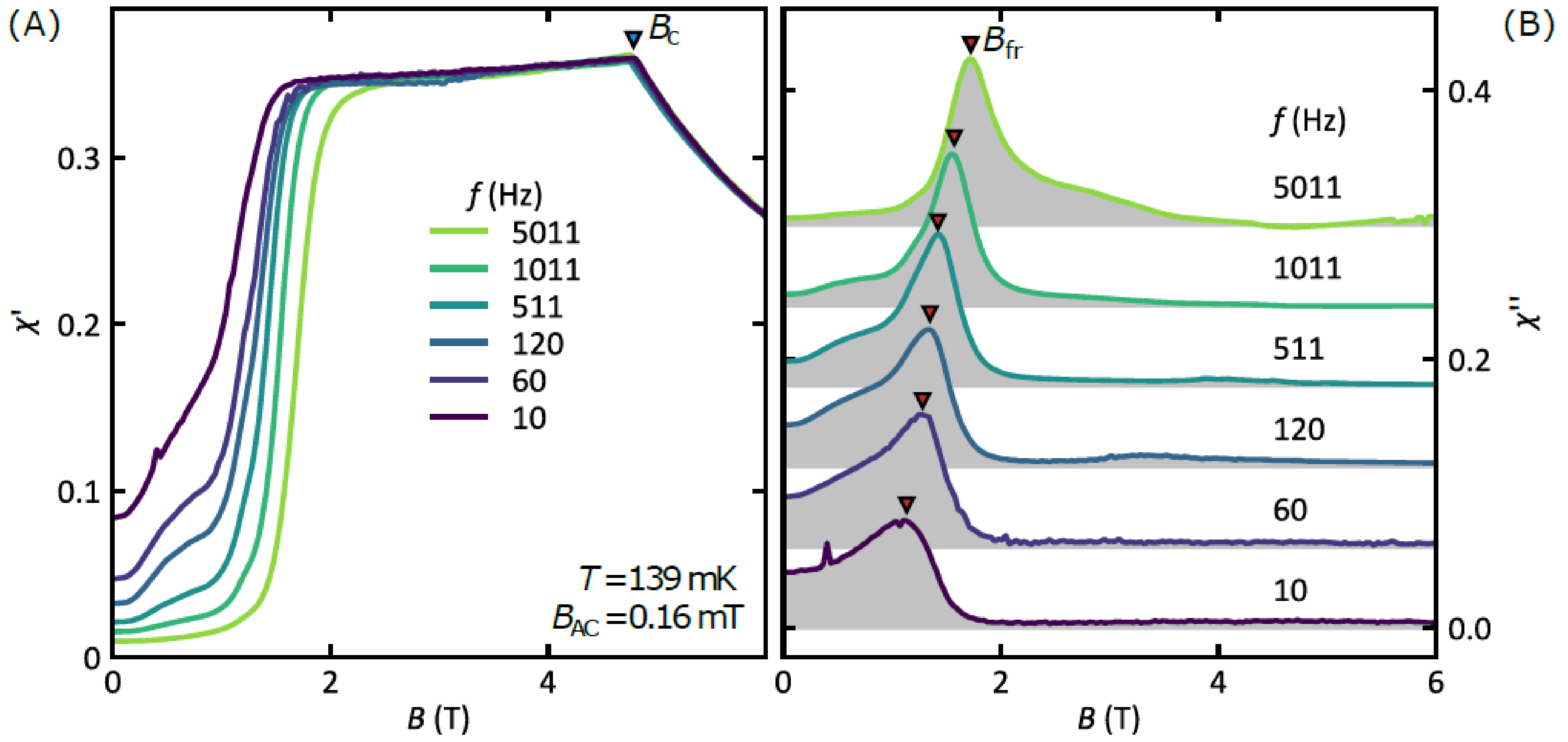


Criticality for transverse field

Susceptibility diverges according to $\chi \sim (B\text{-}B_c)^{-\gamma}$ with γ=1 after removing demagnetization effects



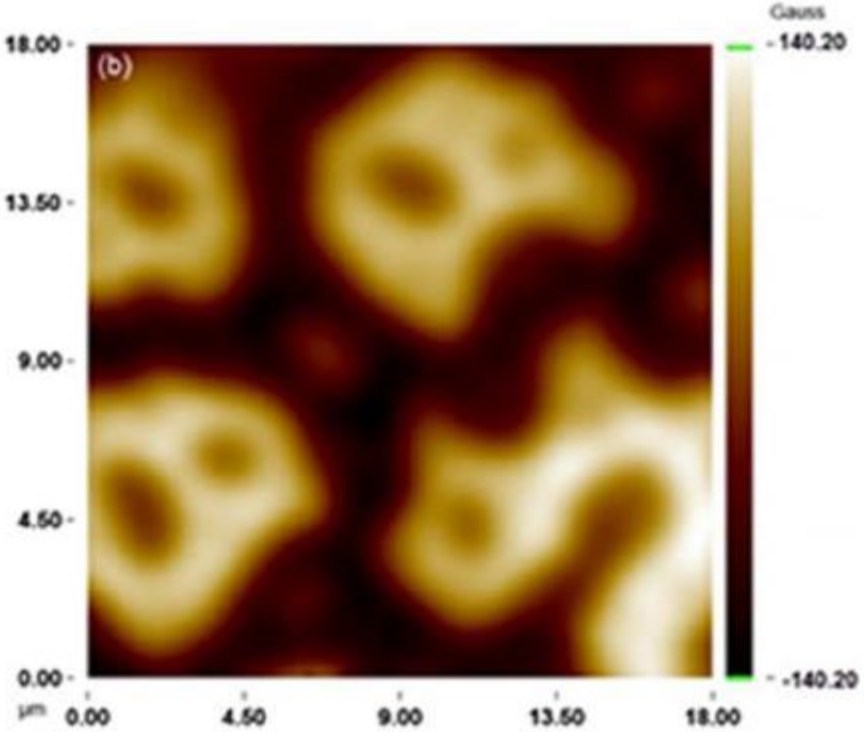
Domain-wall freezing



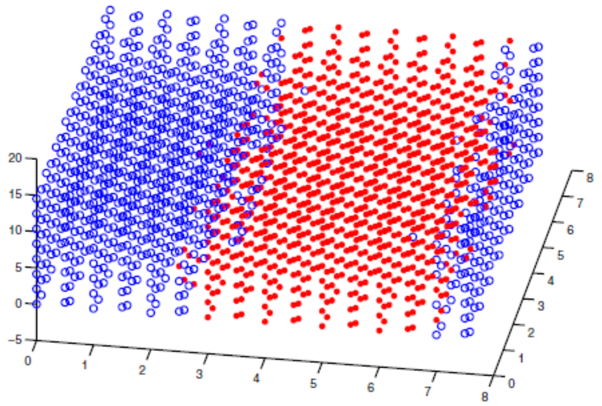
Domains in $LiHoF_4$

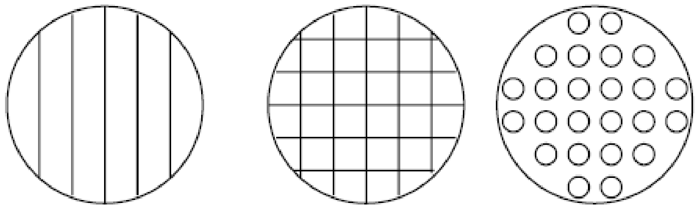
Needle-like domains along easy axis

Scanning-Hall probe image



MC simulation





Karci *et al.*, Rev. Sci. Inst. **85**, 103703 (2014)

Biltmo *et al.*, EPL **87**, 27007 (2009)

Domain energetics

- Stray field energy (exact): calculate from magnetostatic potential of surface charges

$$E_s = \frac{1}{2}\frac{\mu_0}{4\pi}\int \mathrm{d}^2 r\,\Phi_s(\vec{r})\sigma(\vec{r}) \qquad \Phi_s(\vec{r}) = \frac{\mu_0}{4\pi}\int \mathrm{d}^2 r'\,\frac{\sigma(\vec{r}')}{|\vec{r}' - \vec{r}|} \qquad \sigma(\vec{r}) = \vec{m}(\vec{r})\cdot\vec{n}(\vec{r})$$

- Domain wall energy (estimate)

$$E_{\mathrm{dw}} = \sigma_{\mathrm{dw}} A_{\mathrm{dw}} N_{\mathrm{dw}} \frac{|\vec{m}_1 - \vec{m}_2|^2}{f^2}$$