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 TICuCl$_3$ under pressure.

Rüegg et al., PRL 100, 205701 (2008)
In many cases, underlying field theory is known.

Magnetic order-disorder transition (e.g. TlCuCl$_3$):

$\varphi^4$ (or Landau-Ginzburg-Wilson) theory for antiferromagnetic order parameter $\varphi_\alpha(x, t)$

$$S = \int d^d r dt \frac{1}{2} \left[ c^2 (\nabla \varphi_\alpha)^2 + (\partial_t \varphi_\alpha)^2 + m_0 \varphi_\alpha^2 \right] + \frac{u_0}{24} (\varphi_\alpha^2)^2$$

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:

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

LiHoF$_4$: Magnetism dominated by dipolar interactions;
Exchange coupling small
(\(\rightarrow\) transition is of mean-field character)

\(B_c = 5.1\) T (at \(T \rightarrow 0\))

\(T_c = 1.53\) K (at \(B = 0\))
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

![Diagram of LiHoF$_4$ structure]

CEF levels as fct of transverse field

![Graph of CEF levels vs transverse field strength]
**Hyperfine coupling in LiHoF$_4$**

**Ho:** Nuclear spin $I=7/2$, hyperfine coupling $A \approx 0.04$ K

$\rightarrow$ Energy of hyperfine coupling of order $0.5$ K

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

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**Hyperfine-induced stabilization of ordered phase**

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**Incomplete softening of electronic mode at $B_c$**

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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

\[ T \]

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

→ QPT smeared by hyperfine coupling

\[ T \]

Eisenlohr / Vojta, PRB 103, 064405 (2021)
Apply field tilted by angle $\phi$ from hard axis ($\rightarrow$ couples to order parameter)

Transition smeared into crossover for finite $\phi$

Crossover field $B^*$ may be defined from susceptibility maximum.
Leonau theory: $B^* - B_c \sim \phi^{2/3}$
Magnetic ac susceptibility along easy axis as function of field for different angles $\phi$
Magnetic ac susceptibility along easy axis as function of field for different angles $\phi$

**Ordered FM** ($\chi$ limited by demagnetization effects)

**Transition**

Sharp phase transition at any finite angle $\phi$!
First order?
Why not smeared?

T = 1.2 K

Wendl / Eisenlohr et al., Nature 609, 65 (2022)
LiHoF₄: More susceptibility in tilted fields

Magnetic ac susceptibility along easy axis

As $T \to 0$:

Transition field very sensitive to $\phi$

Height of jump in $\chi$

Wendl / Eisenlohr et al., Nature 609, 65 (2022)
LiHoF₄: 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?
Modelling including domains

Microscopic ingredients

\[ H_{\text{mic}} = -K \sum_{i<j} J_{ij} \cdot J_{ij} + \sum_i \left[ V_{\text{CEF}}(J_i) + A J_i \cdot L_i \right] - \mu_B B \cdot \sum_i (gJ_i + g_N L_i) \]

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

Domains

Sheet-like domains (assumed)

Stray-field energy computed exactly

\[ \mu \int d^2 \mathbf{r} d^2 \mathbf{r}' \frac{\sigma(\mathbf{r})\sigma(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \]

Domain sizes as variational parameters

Stray fields induce effective antiferromagnetic coupling between domains!

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

Biltmo et al., EPL 87, 27007 (2009)
Coupled mean-field theory

Microscopic part, separately for each domain:

$$H_{\text{mic}}^\text{MF} = -nK(J \cdot \mathbf{J} - \frac{\mathbf{J}^2}{2}) + V_{\text{CF}}(J) + AJ \cdot I - \mu_B B \cdot (gJ + g_N I)$$

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

$$E_{\text{dom}} = M \sum_\alpha \left( c_{1}^{\alpha} J_{1}^{\alpha} J_{1}^{\alpha} + c_{2}^{\alpha} J_{2}^{\alpha} J_{2}^{\alpha} + c_{12}^{\alpha} J_{1}^{\alpha} J_{2}^{\alpha} \right)$$

Combined m-f theory:

$$H_{1}^\text{MF} = \left( -\frac{n}{2}K + \frac{c_{i}^{x}}{1 - v} \right) \left( 2J_{1}^{x} J_{1}^{x} - (J_{1}^{x})^2 \right)$$

$$+ \frac{c_{12}^{x}}{1 - v} \left( J_{2}^{x} J_{1}^{x} - \frac{1}{2} J_{1}^{x} J_{2}^{x} \right)$$

$$+ \left( -\frac{n}{2}K + \frac{c_{i}^{z}}{1 - v} \right) \left( 2J_{1}^{z} J_{1}^{z} - (J_{1}^{z})^2 \right)$$

$$+ \frac{c_{12}^{z}}{1 - v} \left( J_{2}^{z} J_{1}^{z} - \frac{1}{2} J_{1}^{z} J_{2}^{z} \right)$$

$$- \frac{n}{2}K \left( 2J_{1}^{y} J_{1}^{y} - (J_{1}^{y})^2 \right) + H_{\text{ion}}(\mathbf{J}_{1}),$$

$$H_{2}^\text{MF} = \left( -\frac{n}{2}K + \frac{c_{i}^{x}}{v} \right) \left( 2J_{2}^{x} J_{2}^{x} - (J_{2}^{x})^2 \right)$$

$$+ \frac{c_{12}^{x}}{v} \left( J_{1}^{x} J_{2}^{x} - \frac{1}{2} J_{1}^{x} J_{2}^{x} \right)$$

$$+ \left( -\frac{n}{2}K + \frac{c_{i}^{z}}{v} \right) \left( 2J_{2}^{z} J_{2}^{z} - (J_{2}^{z})^2 \right)$$

$$+ \frac{c_{12}^{z}}{v} \left( J_{1}^{z} J_{2}^{z} - \frac{1}{2} J_{1}^{z} J_{2}^{z} \right)$$

$$- \frac{n}{2}K \left( 2J_{2}^{y} J_{2}^{y} - (J_{2}^{y})^2 \right) + H_{\text{ion}}(\mathbf{J}_{2}),$$
Results: Phase transitions and domains

Key quantity: Domain volume ratio $\nu = \frac{D_2}{D_1+D_2}$

Transverse field

Domains vanish at microscopic transition

Tilted field

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)
Why is "hyperfine nose" suppressed for tilted fields?

Interplay of non-Kramers & hyperfine physics

\( \phi=0 \):
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 \(T\)

\( \phi>5^\circ \):
\(|J|\) variation weak
→ hyperfine coupling does no longer stabilize FM phase
Quantum phase transitions

Two types of (continuous) quantum phase transitions in LiHoF\(_4\):

1) Multi-domain ferromagnet $\leftrightarrow$ Single-domain paramagnet for $\phi=0$
   
   Domains disappear by vanishing magnetization
   
   Exponents $\alpha=0$, $\beta=1/2$, $\gamma=1$, $\delta=3$ (mean-field) due to dipolar interactions

2) Multi-domain $\leftrightarrow$ Single-domain for $\phi\neq0$
   
   Domains disappear by vanishing minority domain volume
   
   Exponents $\alpha=0$, $\beta=1$, $\gamma=0$, $\delta=1$

Mesoscale antiferromagnetism

Wendl / Eisenlohr et al., Nature 609, 65 (2022)
Heat capacity of LiHoF$_4$

(A)

$C/R$

$B = 0 \, T, \Phi = 0^\circ$

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
LiHoF₄ 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
Susceptibility diverges according to $\chi \sim (B - B_c)^\gamma$ with $\gamma = 1$ after removing demagnetization effects.
Domain-wall freezing

(A) $f$ (Hz)
- 5011
- 1011
- 511
- 120
- 60
- 10

$T = 139$ mK
$B_{HC} = 0.16$ mT

(B) $f$ (Hz)
- 5011
- 1011
- 511
- 120
- 60
- 10
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)
- Stray field energy (exact): calculate from magnetostatic potential of surface charges

\[ E_s = \frac{1}{2} \frac{\mu_0}{4\pi} \int d^2r \Phi_s(\vec{r}) \sigma(\vec{r}) \quad \Phi_s(\vec{r}) = \frac{\mu_0}{4\pi} \int d^2r' \frac{\sigma(\vec{r}')}{|\vec{r}' - \vec{r}|} \quad \sigma(\vec{r}) = \vec{m}(\vec{r}) \cdot \vec{n}(\vec{r}) \]

- Domain wall energy (estimate)

\[ E_{dw} = \sigma_{dw} A_{dw} N_{dw} \frac{\left| \vec{m}_1 - \vec{m}_2 \right|^2}{f^2} \]