

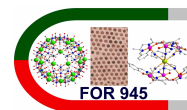
High Spin Cycles: Topping the Spin Record for a Single Molecule verging on Quantum Criticality

Jürgen Schnack

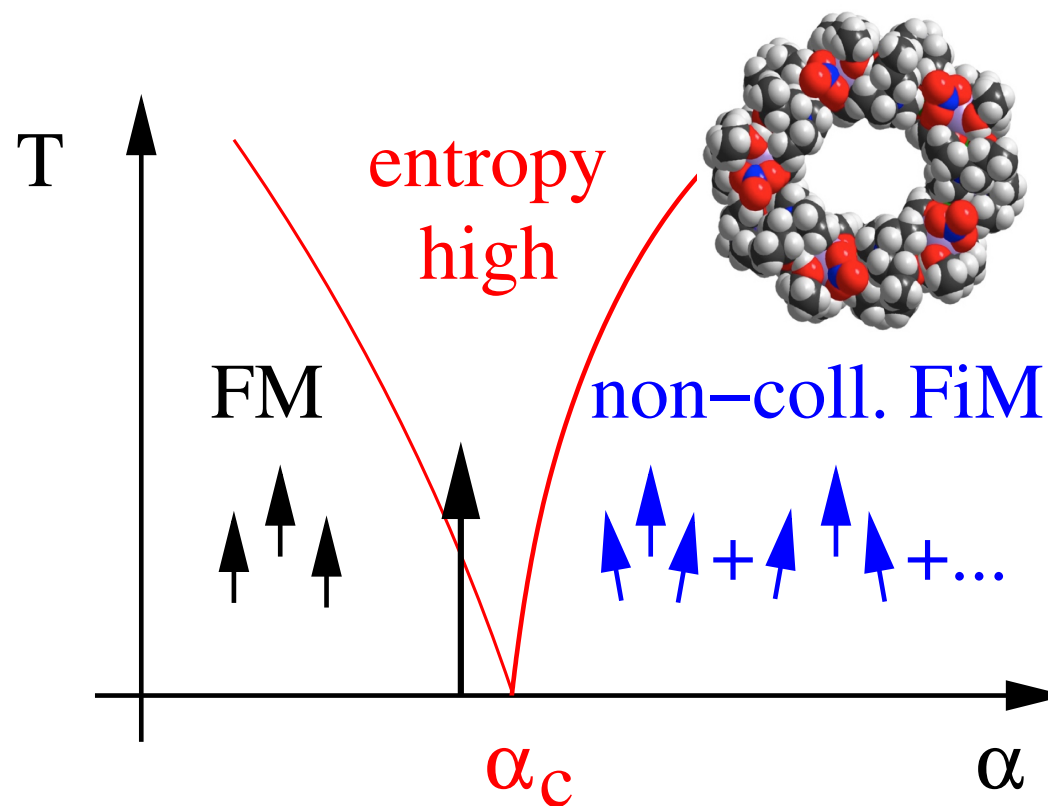
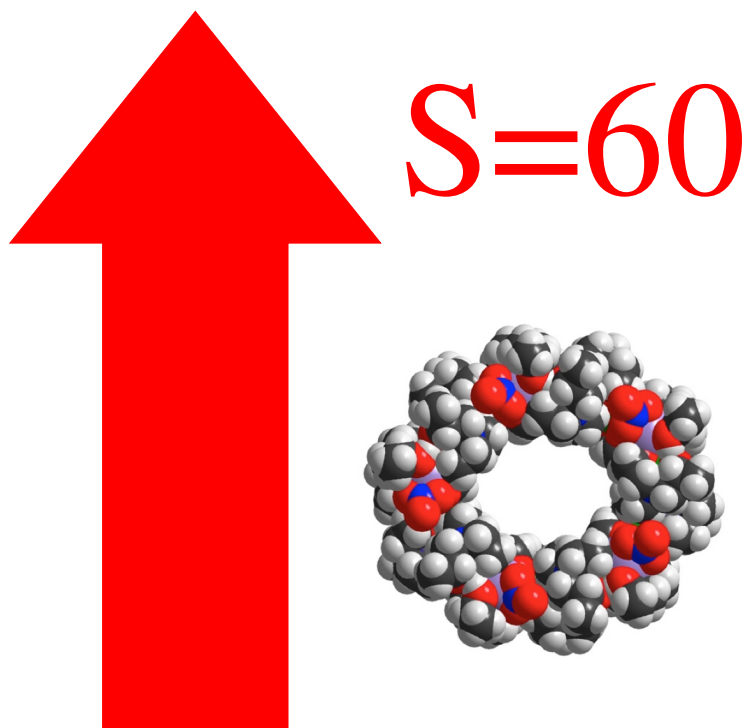
Department of Physics – University of Bielefeld – Germany

<http://obelix.physik.uni-bielefeld.de/~schnack/>

The Joint European Magnetic Symposia, Mainz, 4 September 2018



Gd₁₀Fe₁₀ – summary

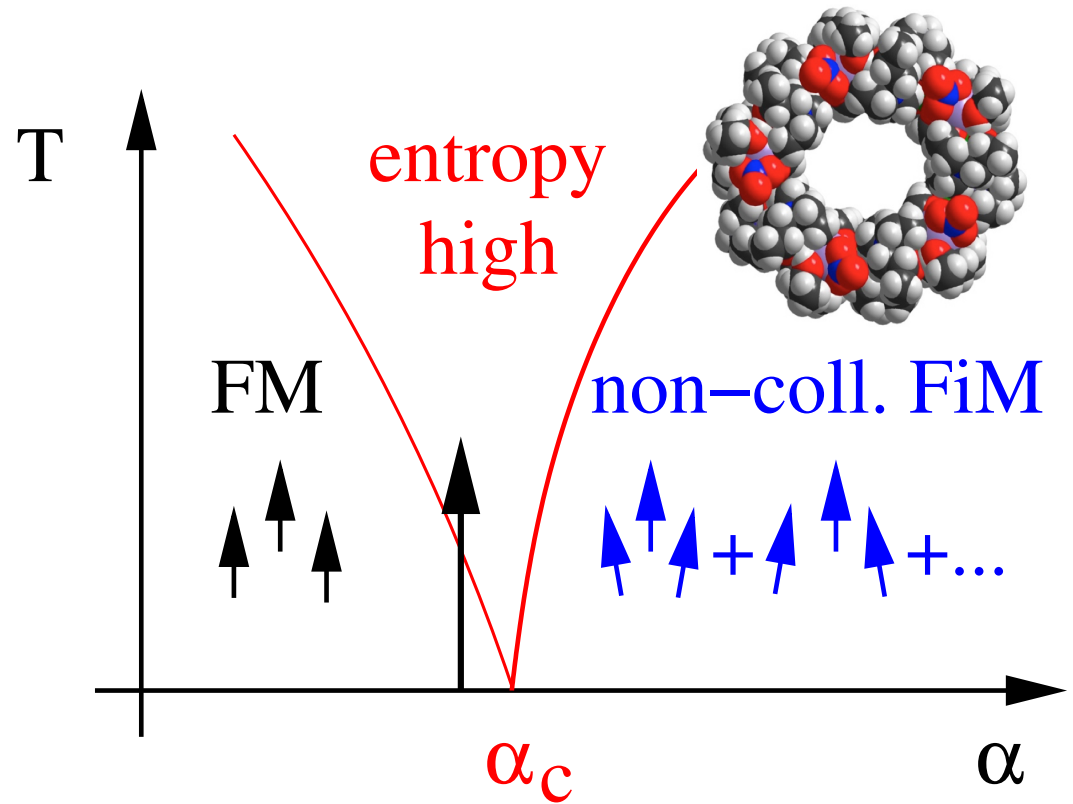
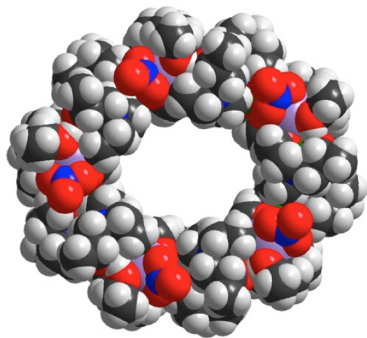


A. Baniodeh, N. Magnani, Y. Lan, G. Buth, C.E. Anson, J. Richter, M. Affronte, J. Schnack, A.K. Powell, *High Spin Cycles: Topping the Spin Record for a Single Molecule verging on Quantum Criticality*, npj Quantum Materials **3**, 10 (2018)

Gd₁₀Fe₁₀ – summary



$S=60$

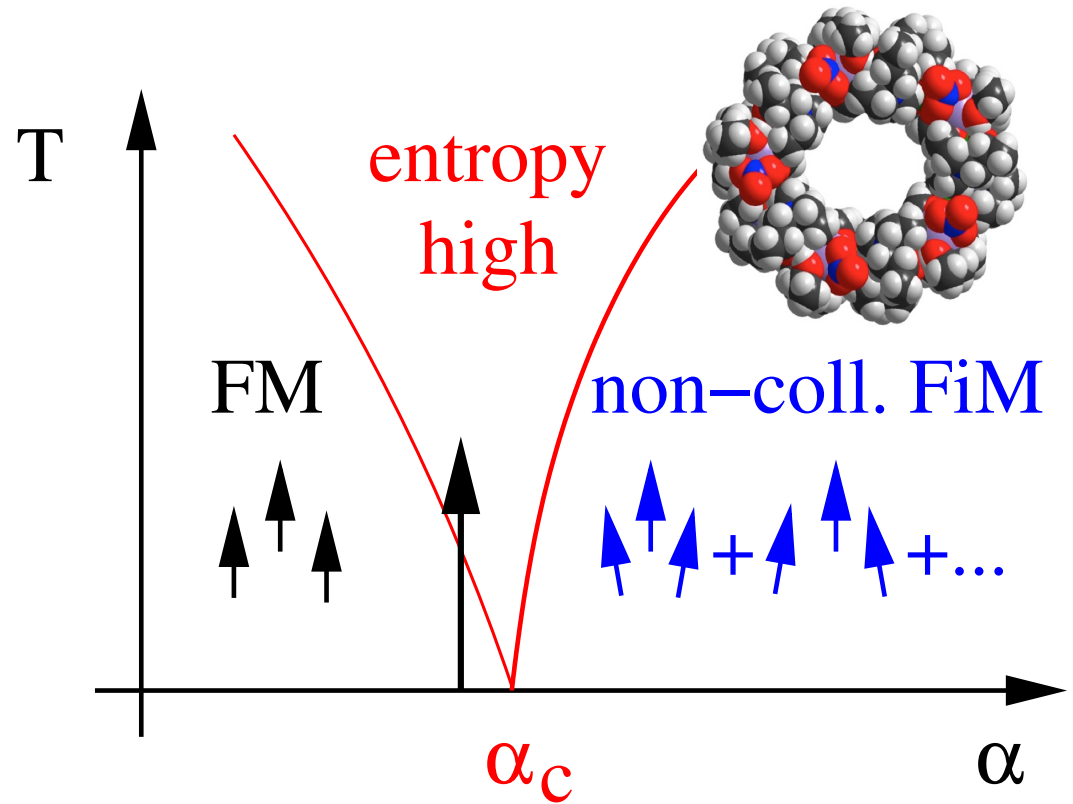
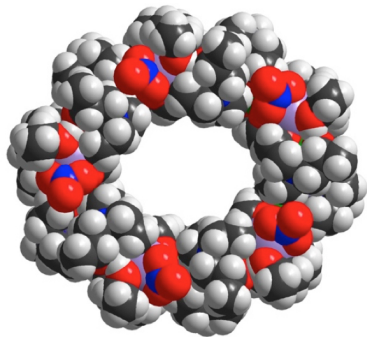


How do we know?

Gd₁₀Fe₁₀ – summary



S=60



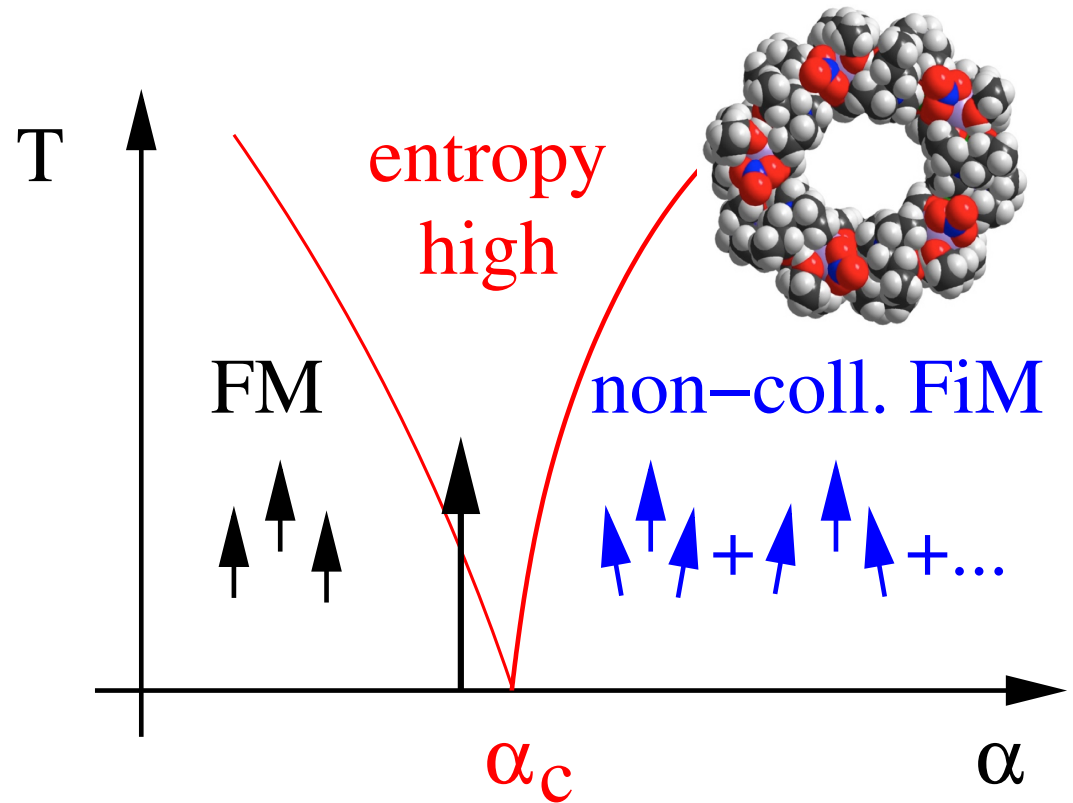
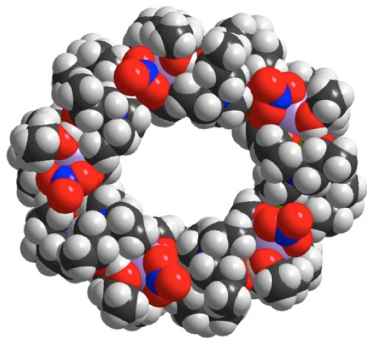
How do we know?

What is a QPT?

Gd₁₀Fe₁₀ – summary



$S=60$

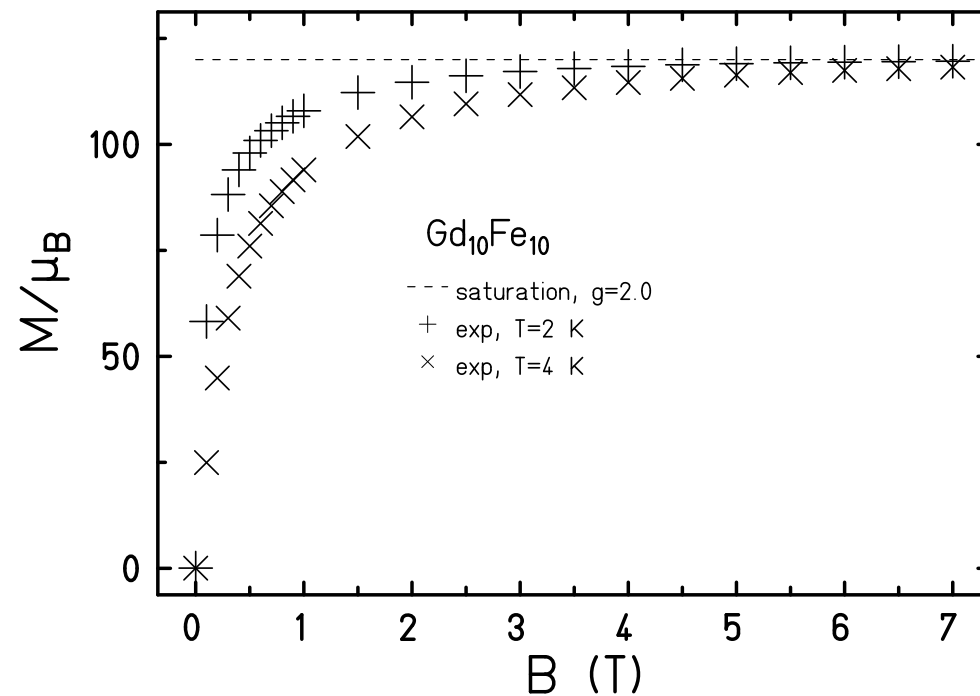
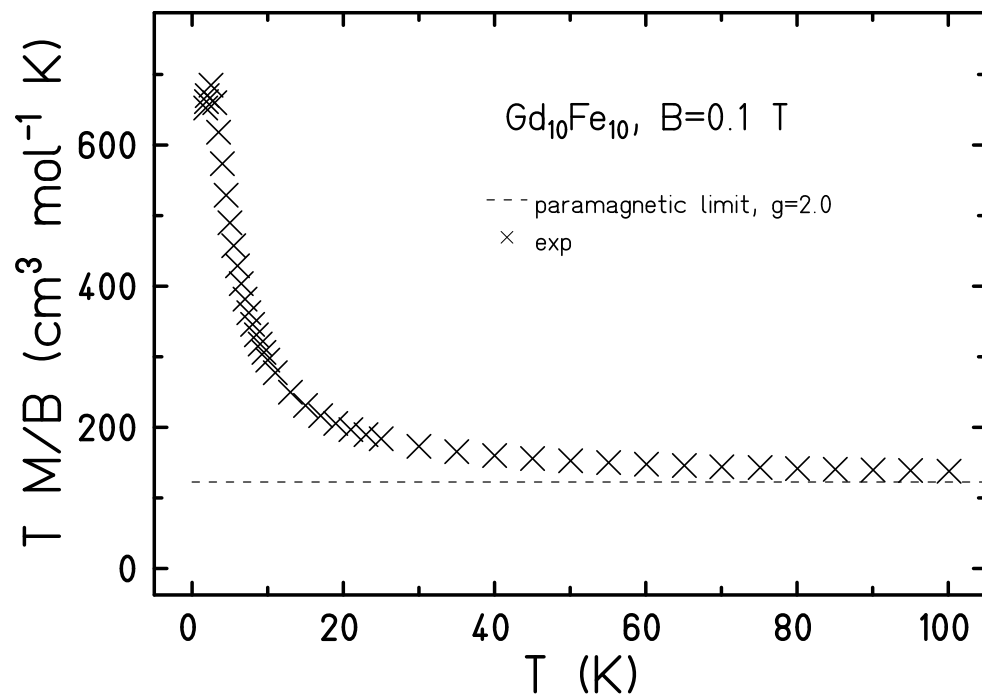


How do we know?

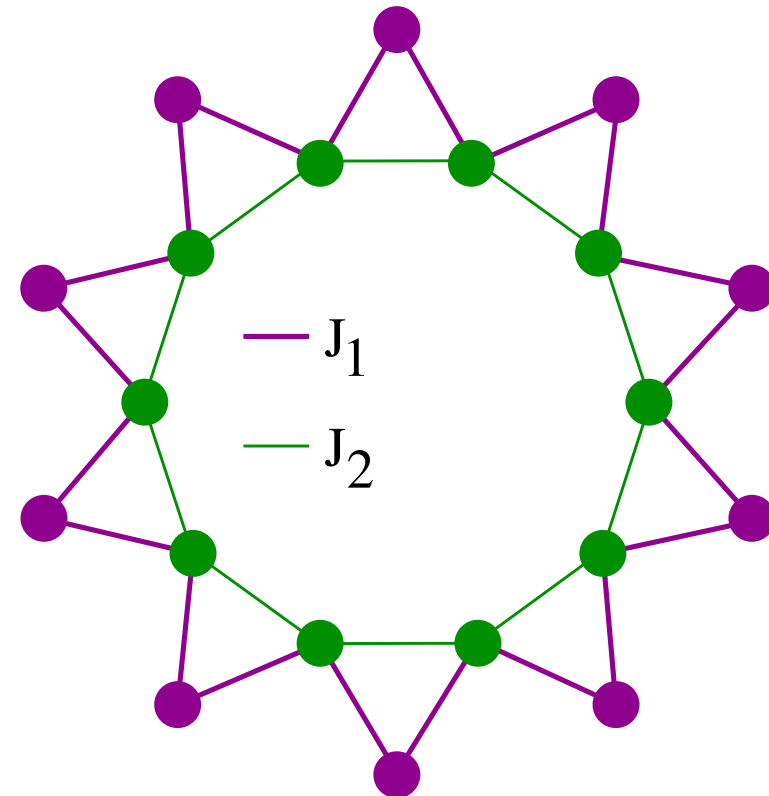
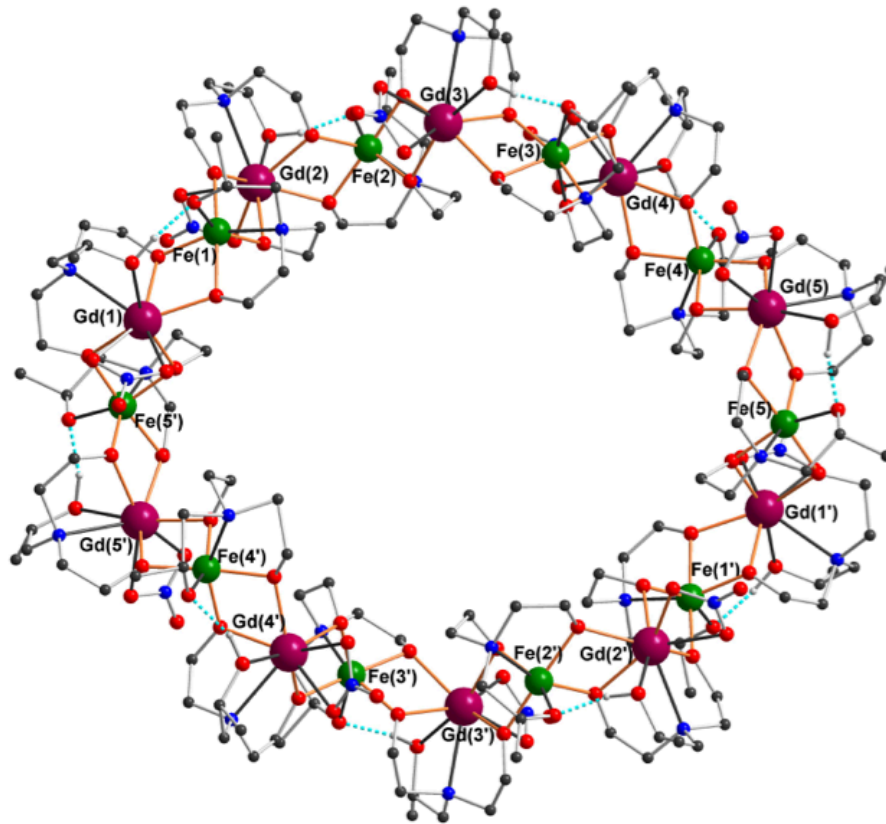
What is a QPT?
In Gd₁₀Fe₁₀?

Start: experimental data

Gd₁₀Fe₁₀ – How to rationalize the experimental data?



Gd₁₀Fe₁₀ – structure = delta chain



green: Fe ($s = 5/2$), purple: Gd ($s = 7/2$)

A. Baniodeh *et al.*, *npj Quantum Materials* **3**, 10 (2018)

Model Hamiltonian

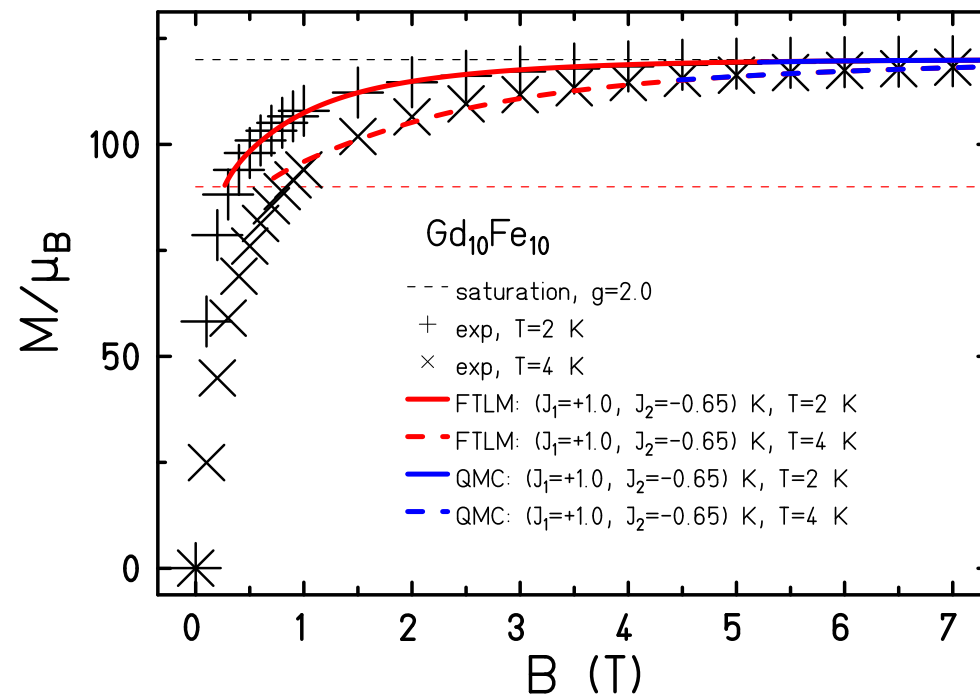
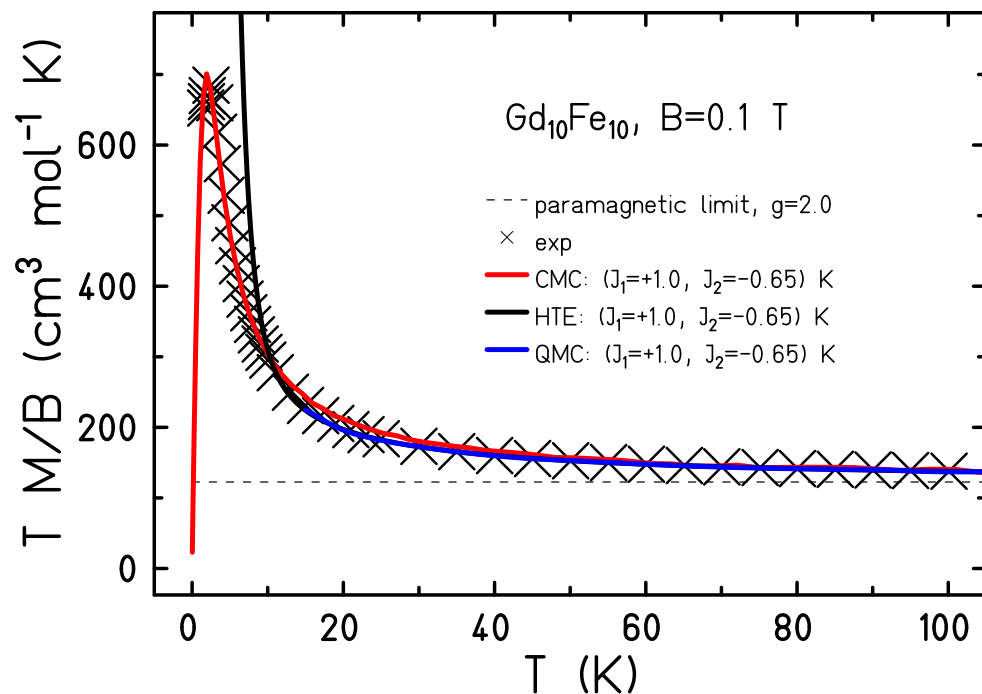
$$\begin{aligned} \underline{H} = & -2J_1 \sum_i \underline{\vec{S}}_{\text{Gd},i} \cdot \left(\underline{\vec{S}}_{\text{Fe},i} + \underline{\vec{S}}_{\text{Fe},i+1} \right) \\ & -2J_2 \sum_i \underline{\vec{S}}_{\text{Fe},i} \cdot \underline{\vec{S}}_{\text{Fe},i+1} + g \mu_B B \sum_i \left(\underline{S}_{\text{Gd},i}^z + \underline{S}_{\text{Fe},i}^z \right) \end{aligned}$$

Dimension of Hilbert space

$$(2s_{\text{Gd}} + 1)^{10} (2s_{\text{Fe}} + 1)^{10} \approx 6.5 \cdot 10^{16}$$

What would you do?

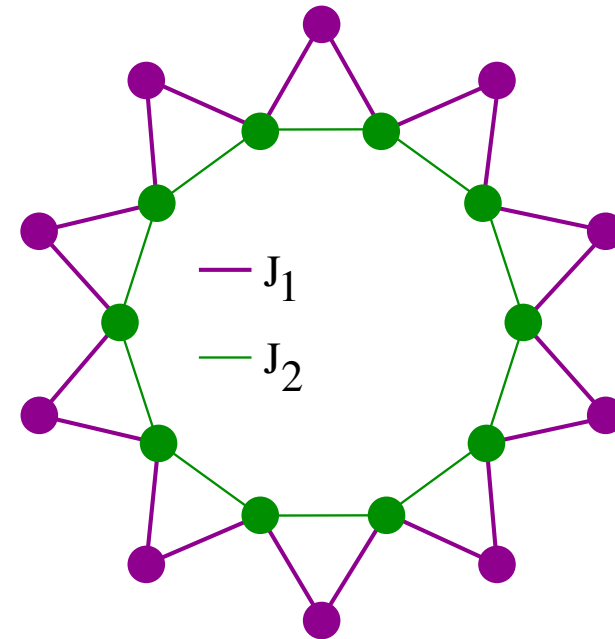
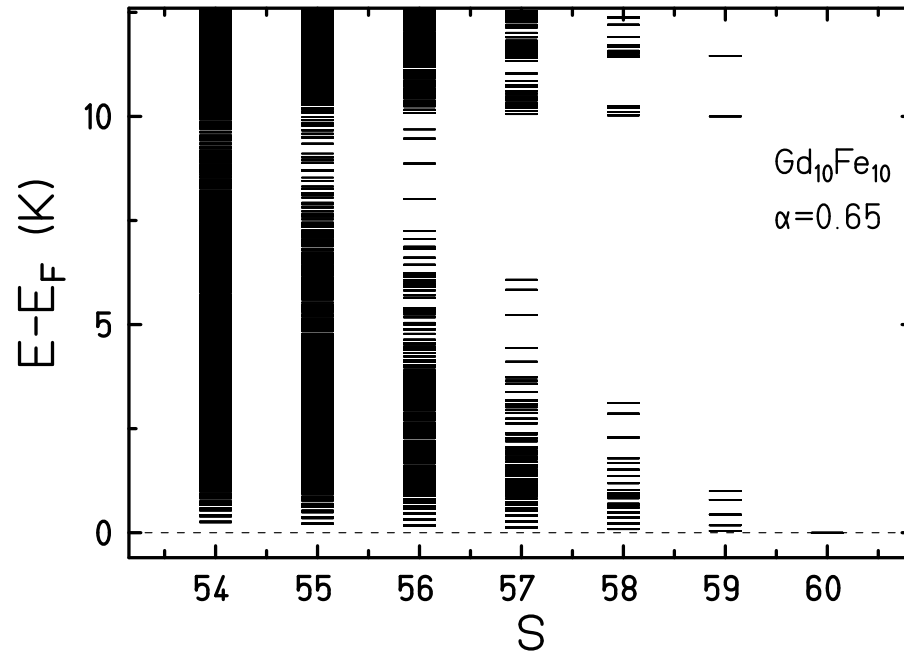
Gd₁₀Fe₁₀ – Methods



Methods: HTE, QMC, CMC, FTLM $\Rightarrow J_1 = 1.0$ K, $J_2 = -0.65$ K

A. Baniodeh *et al.*, *npj Quantum Materials* **3**, 10 (2018)

Gd₁₀Fe₁₀ – $S = 60$

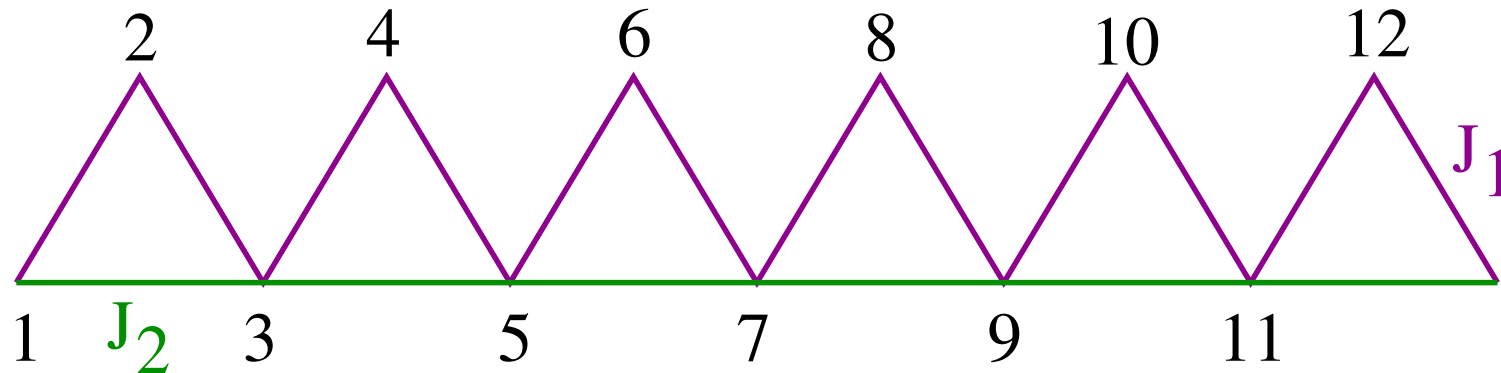


$\Rightarrow S = 60$, largest ground state spin of a molecule to date

$\Rightarrow \alpha_{\text{Gd}_{10}\text{Fe}_{10}} = |J_2|/J_1 = 0.65$ What if J_2 stronger?

A. Baniodeh *et al.*, *npj Quantum Materials* **3**, 10 (2018)

Excursus: sawtooth (delta) chain



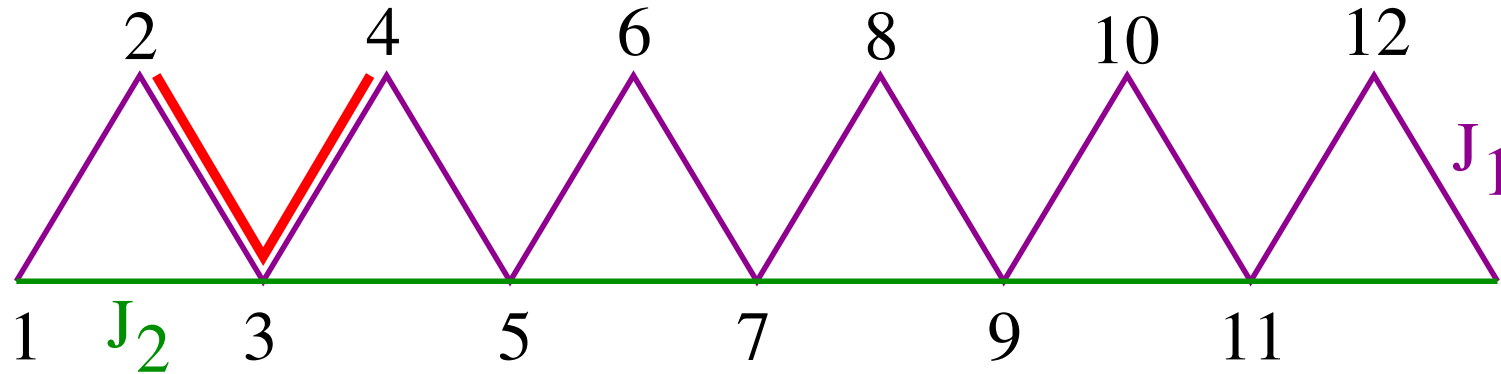
⇒ special properties for $J_1 > 0$ (ferro) and $J_2 < 0$ (af) at certain α_c

e.g. $\alpha_c = |J_2|/J_1 = 0.5$ if $s_i = 1/2 \forall i$

⇒ flat band of (multi-) magnon states; huge ground state degeneracy (1,2)

- (1) V. Y. Krivnov, D. V. Dmitriev, S. Nishimoto, S.-L. Drechsler, and J. Richter, Phys. Rev. B **90**, 014441 (2014).
 (2) D. V. Dmitriev and V. Y. Krivnov, Phys. Rev. B **92**, 184422 (2015).

Excursus: sawtooth (delta) chain



$\Rightarrow |F\rangle = |S = S_{\max}, M = S_{\max}\rangle$ fully polarized ferromagnetic state

$\Rightarrow |1 \text{ localized magnon at } (2,3,4)\rangle = (\tilde{s}_2^- + \tilde{s}_4^- + 2\tilde{s}_3^-) |F\rangle;$

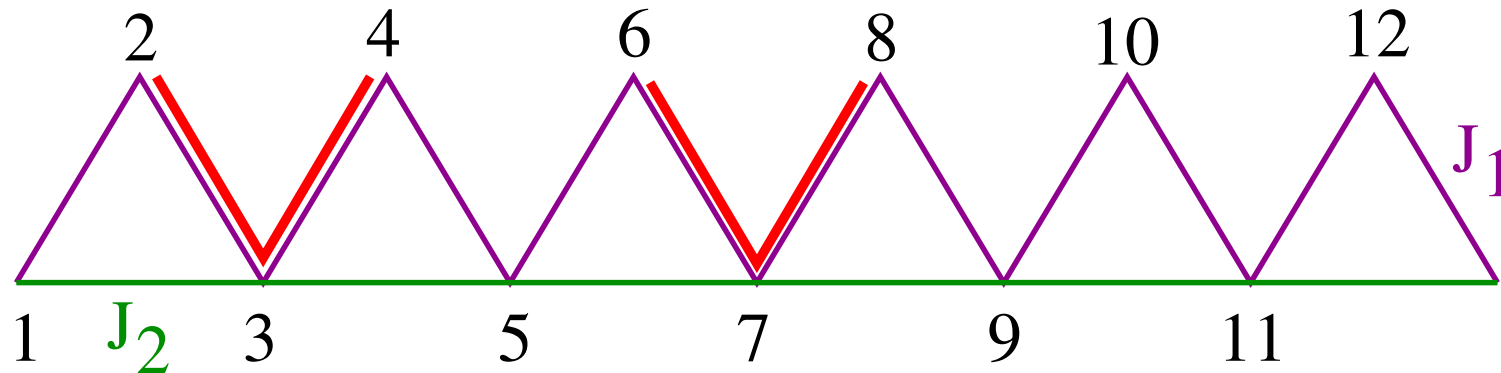
$$E = E_F, M = S_{\max} - 1$$

\Rightarrow Can be everywhere. Flat band in one-magnon space. Degenerate with $|F\rangle$.

(1) V. Y. Krivnov, D. V. Dmitriev, S. Nishimoto, S.-L. Drechsler, and J. Richter, Phys. Rev. B **90**, 014441 (2014).

(2) D. V. Dmitriev and V. Y. Krivnov, Phys. Rev. B **92**, 184422 (2015).

Excursus: sawtooth (delta) chain

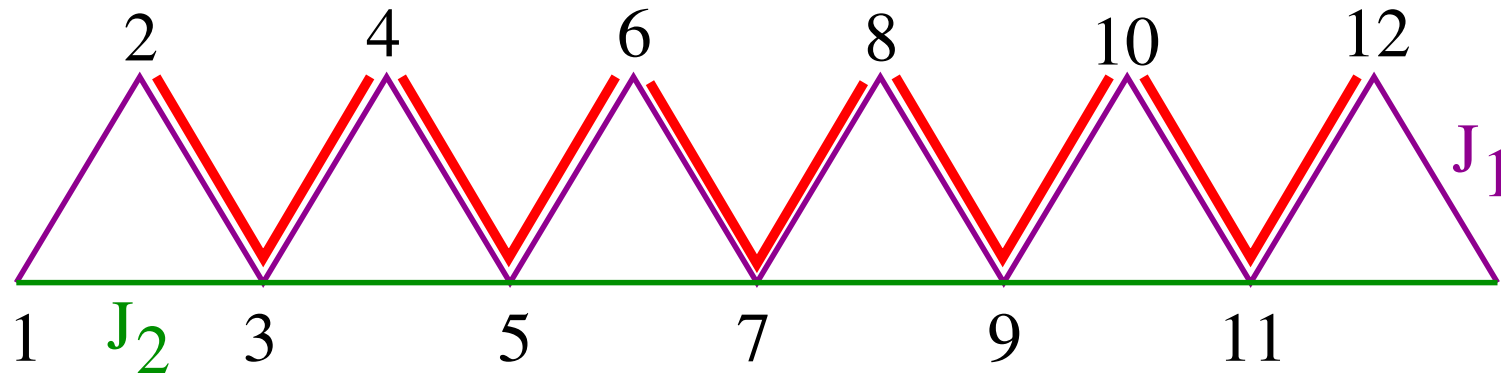


⇒ $|2 \text{ localized magnons}\rangle; E = E_F, M = S_{\max} - 2$

⇒ Can be everywhere. Flat band in two-magnon space. Degenerate with $|F\rangle$.

- (1) V. Y. Krivnov, D. V. Dmitriev, S. Nishimoto, S.-L. Drechsler, and J. Richter, Phys. Rev. B **90**, 014441 (2014).
- (2) D. V. Dmitriev and V. Y. Krivnov, Phys. Rev. B **92**, 184422 (2015).

Excursus: sawtooth (delta) chain



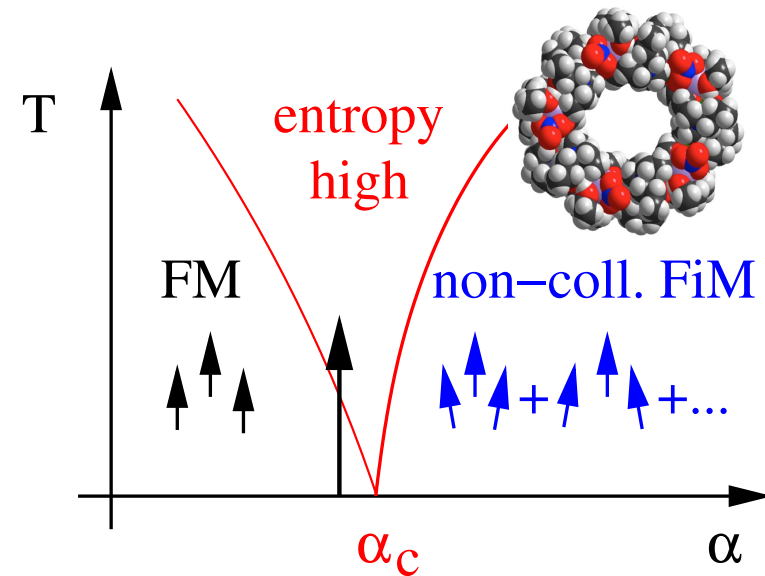
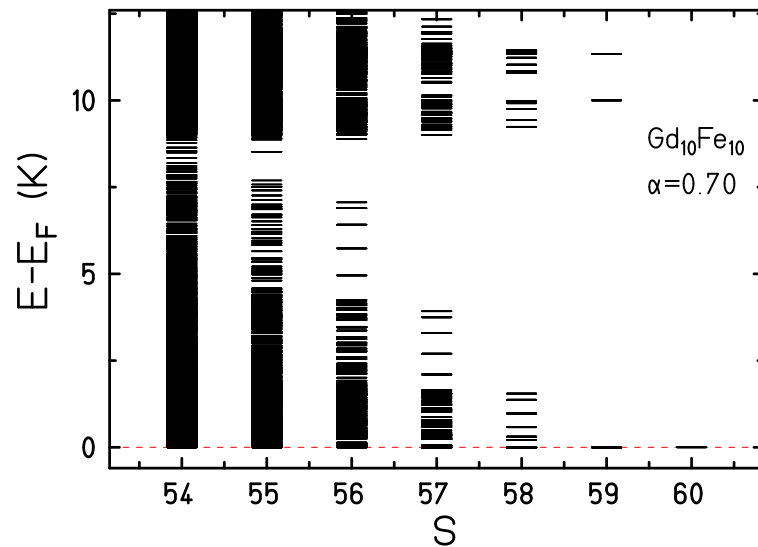
⇒ | max. number of localized magnons \rangle ; $E = E_F, M = S_{\max} - N/2$

⇒ Macroscopic number of localized magnons. Degenerate with $| F \rangle$.

⇒ Extensive entropy.

(1) V. Y. Krivnov, D. V. Dmitriev, S. Nishimoto, S.-L. Drechsler, and J. Richter, Phys. Rev. B **90**, 014441 (2014).

(2) D. V. Dmitriev and V. Y. Krivnov, Phys. Rev. B **92**, 184422 (2015).

Gd₁₀Fe₁₀ – QCP

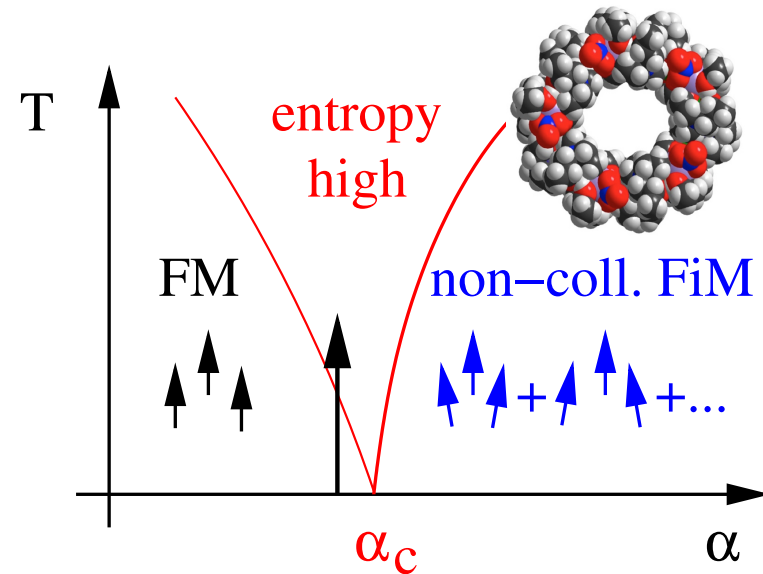
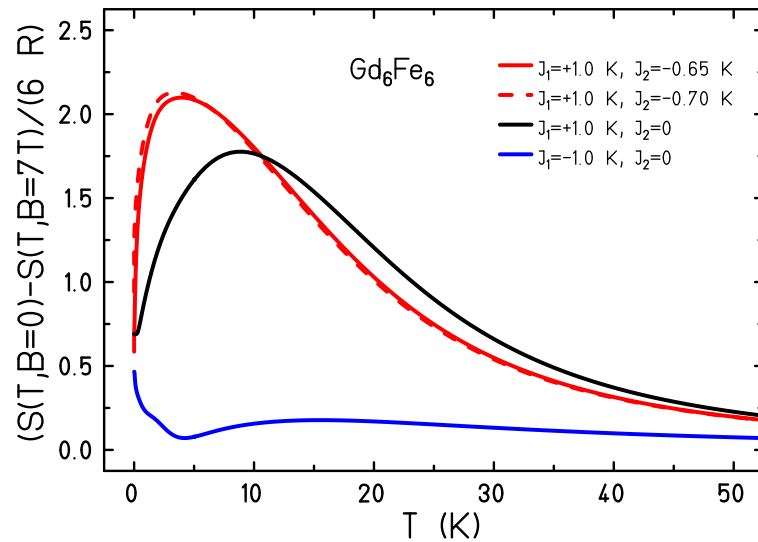
⇒ for $s_1 = 5/2$ and $s_2 = 7/2$: $\alpha_c = 0.70$

⇒ as function of α Quantum Phase Transition at α_c
 from $S = 60$ ground state to ground state with $S = 54$.
 ($\Delta S = N/4 + 1$ in general)

A. Baniodeh *et al.*, *npj Quantum Materials* **3**, 10 (2018)

Quantum Phase Transition

Non-analytic behavior of thermodynamic functions at $T = 0$ for variation of another external parameter, e.g. field, pressure; here α – maybe varied by pressure.

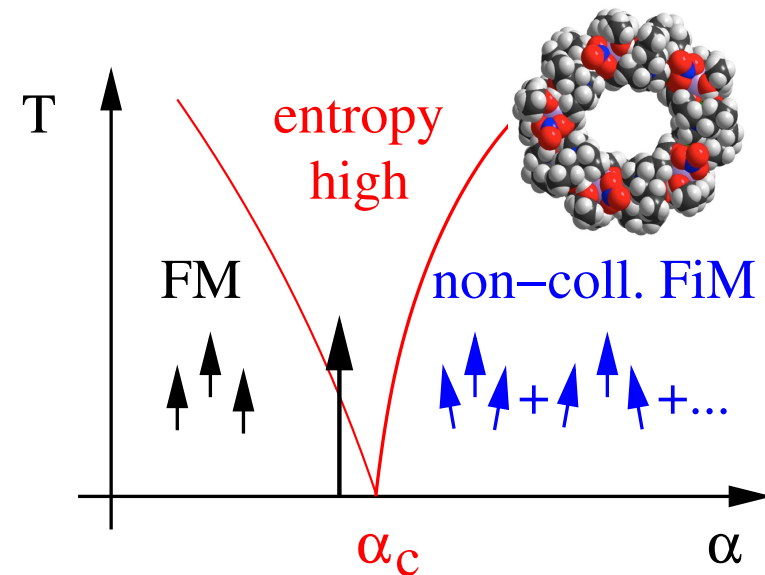
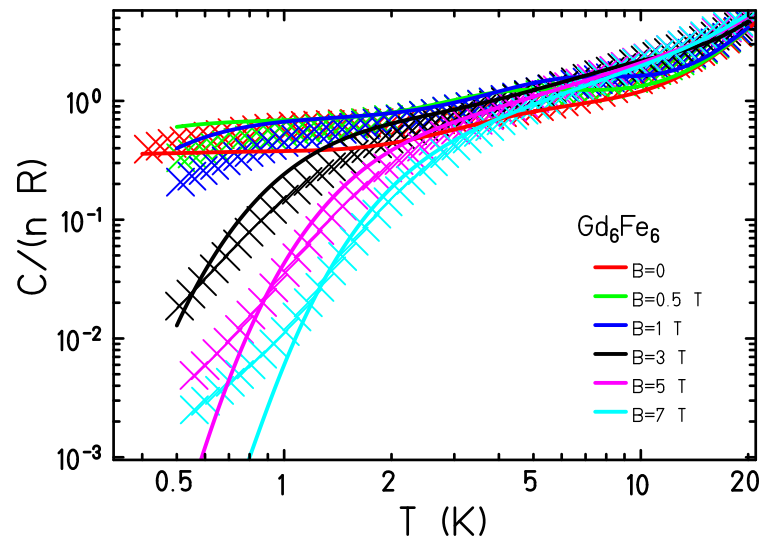
Gd₁₀Fe₁₀ – $T > 0$ 

\Rightarrow although QPT and QCP at $T = 0$,
noticeable at elevated temperatures (arrow);

\Rightarrow **example isothermal entropy change:**
little difference between $\alpha = 0.70$ and $\alpha = 0.65$.

A. Baniodeh *et al.*, *npj Quantum Materials* **3**, 10 (2018)

Gd₁₀Fe₁₀ – heat capacity



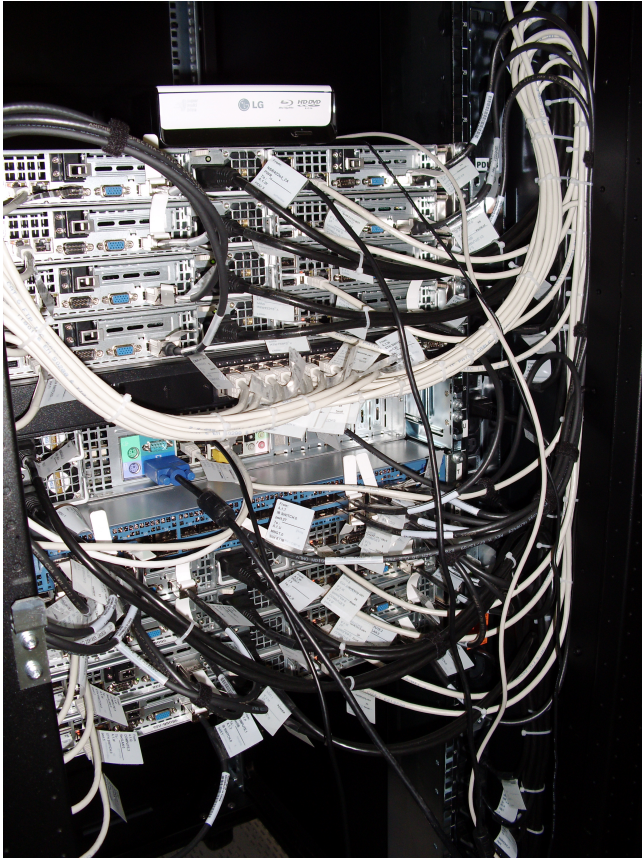
⇒ heat capacity assumes very large values even down to lowest temperatures;

⇒ **evaluated by means of FTLM for a smaller (hypothetical) system Gd₆Fe₆;**

⇒ **magnetic field separates $S = 60$ ground state, C drops.**

A. Baniodeh *et al.*, *npj Quantum Materials* **3**, 10 (2018)

Gd₁₀Fe₁₀ – Summary



- Sawtooth chain has a rich phase diagram: magnetization plateaux, magnetization jumps, flat bands, quantum phase transitions.
 - Gd₁₀Fe₁₀ is a lucky punch.
 - Largest ground state spin of a single molecule to date: $S = 60$.
 - Quantum Phase Transition observable in a molecule with structure of a sawtooth chain.
- ⇐ And yes, we use big computers.

A. Baniodeh, N. Magnani, Y. Lan, G. Buth, C.E. Anson, J. Richter, M. Affronte, J. Schnack, A.K. Powell, *High Spin Cycles: Topping the Spin Record for a Single Molecule verging on Quantum Criticality*, npj Quantum Materials **3**, 10 (2018)

Many thanks to my collaborators



- C. Beckmann, M. Czopnik, T. Glaser, O. Hanebaum, Chr. Heesing, M. Höck, N.B. Ivanov, H.-T. Langwald, A. Müller, R. Schnalle, Chr. Schröder, J. Ummethum (Bielefeld)
- **K. Bärwinkel, H.-J. Schmidt, M. Neumann (Osnabrück)**
- M. Luban (Ames Lab); P. Kögerler (Aachen, Jülich, Ames); D. Collison, R.E.P. Winpenny, E.J.L. McInnes, F. Tuna (Man U); L. Cronin, M. Murrie (Glasgow); E. Brechin (Edinburgh); H. Nojiri (Sendai, Japan); A. Postnikov (Metz); M. Evangelisti (Zaragosa); A. Honecker (U de Cergy-Pontoise); E. Garlatti, S. Carretta, G. Amoretti, P. Santini (Parma); A. Tennant (ORNL); Gopalan Rajaraman (Mumbai); **M. Affronte (Modena)**
- **J. Richter**, J. Schulenburg (Magdeburg); B. Lake (HMI Berlin); B. Büchner, V. Kataev, H.-H. Klauß (Dresden); **A. Powell**, W. Wernsdorfer (Karlsruhe); E. Rentschler (Mainz); J. Wosnitza (Dresden-Rossendorf); J. van Slageren (Stuttgart); R. Klingeler (Heidelberg); O. Waldmann (Freiburg)

Thank you very much for your
attention.

The end.

Molecular Magnetism Web

www.molmag.de

Highlights. Tutorials. Who is who. Conferences.