

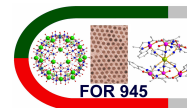
High Spin Cycles: Topping the Spin Record for a Single Molecule verging on Quantum Criticality

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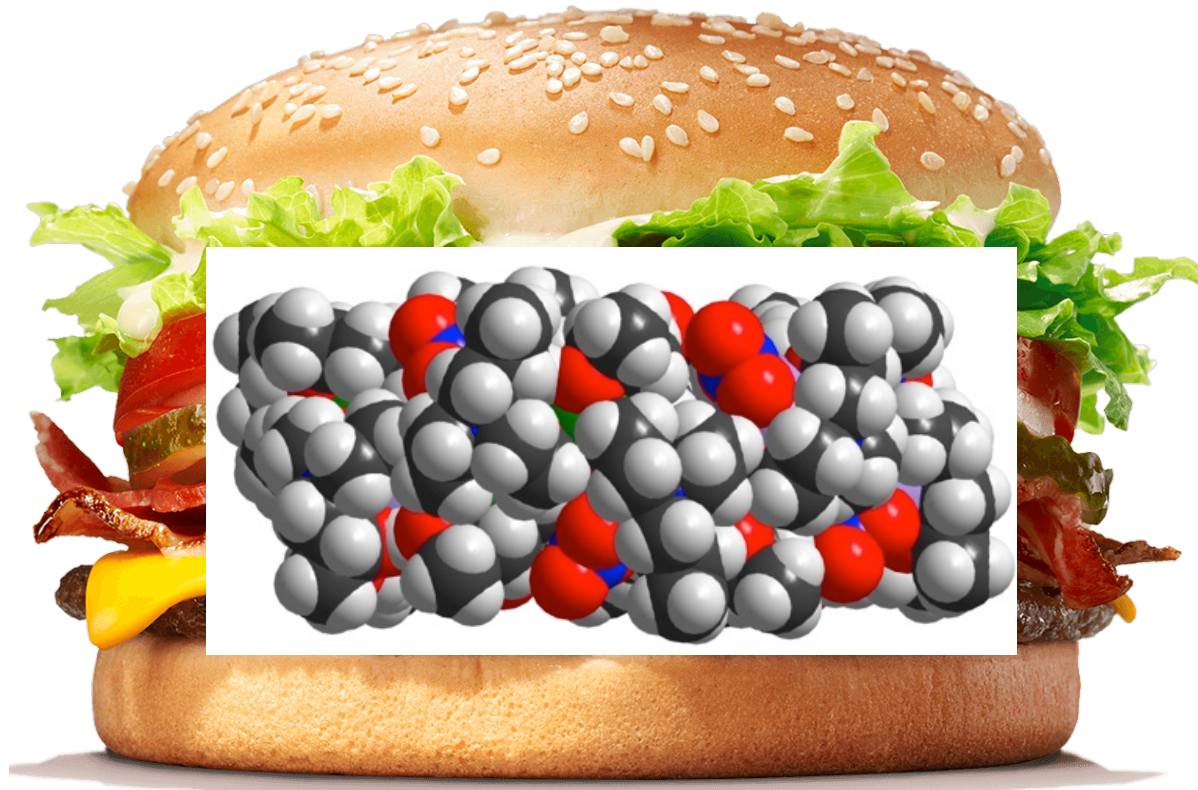
Modern didactics teaches us

All information goes to the Crocodile Brain first.

The CrocBrain is able to ask only two questions:

Is it dangerous?

Can one eat it?

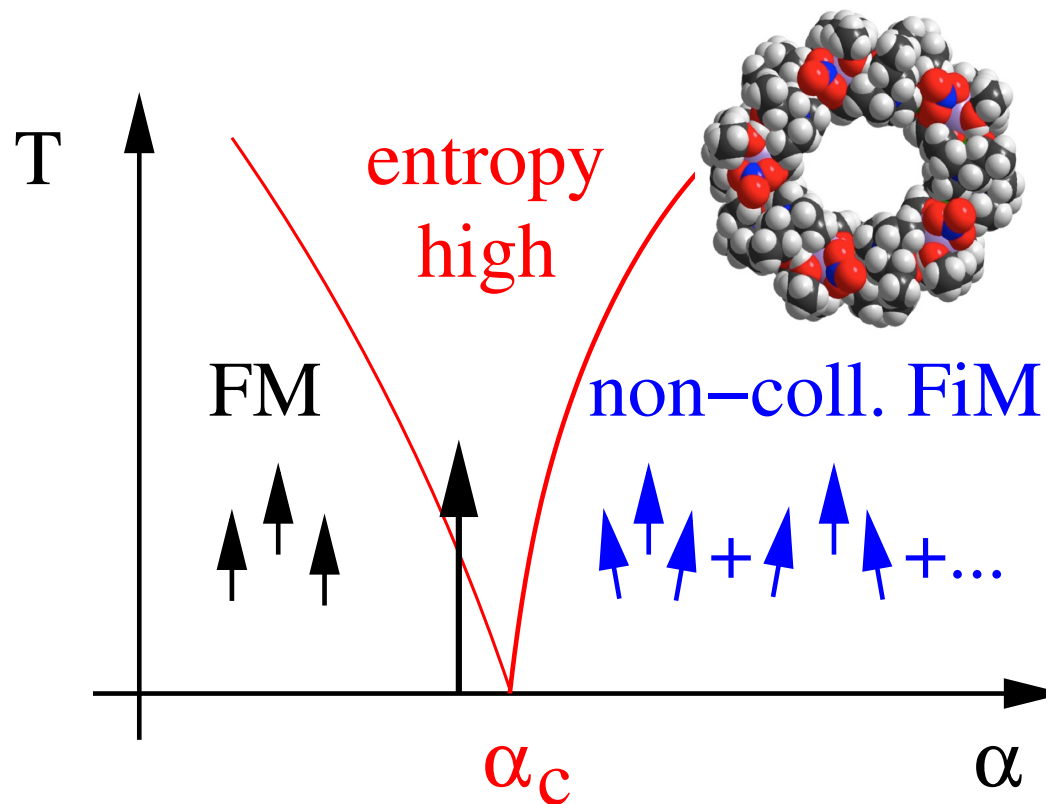
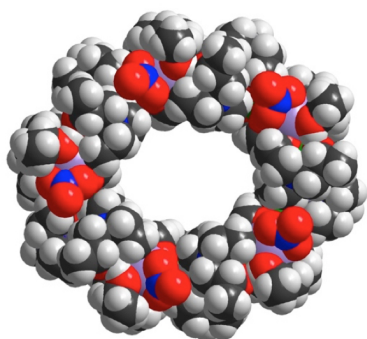


The mouth-watering $Gd_{10}Fe_{10}$!
(It's not dangerous.)

Gd₁₀Fe₁₀ is yummy



S=60

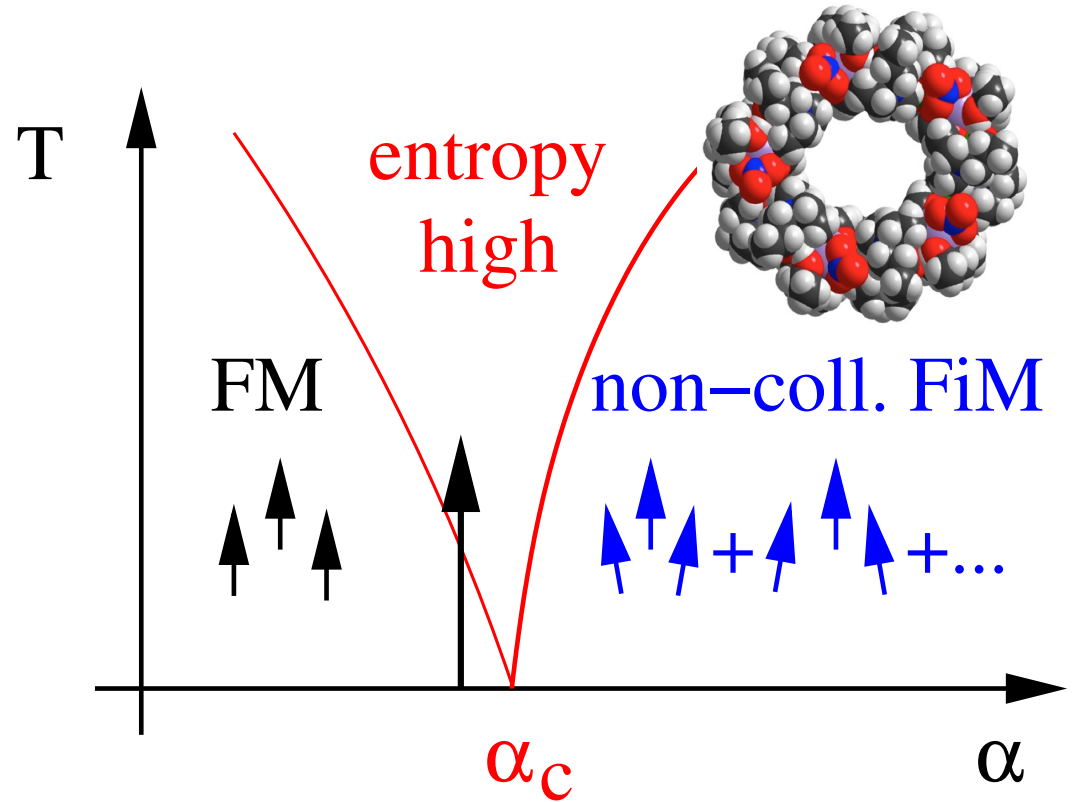
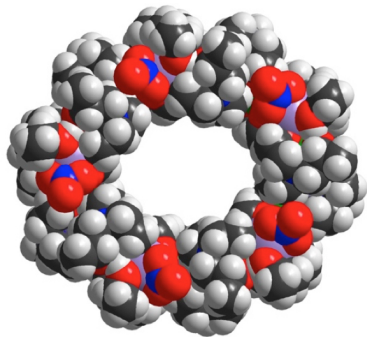


A. Baniodeh, N. Magnani, Y. Lan, G. Buth, C.E. Anson, J. Richter, M. Affronte, J. Schnack, A.K. Powell, *High Spin Cycles: Topping the Spin Record for a Single Molecule verging on Quantum Criticality*, npj Quantum Materials **3**, 10 (2018)

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$S=60$

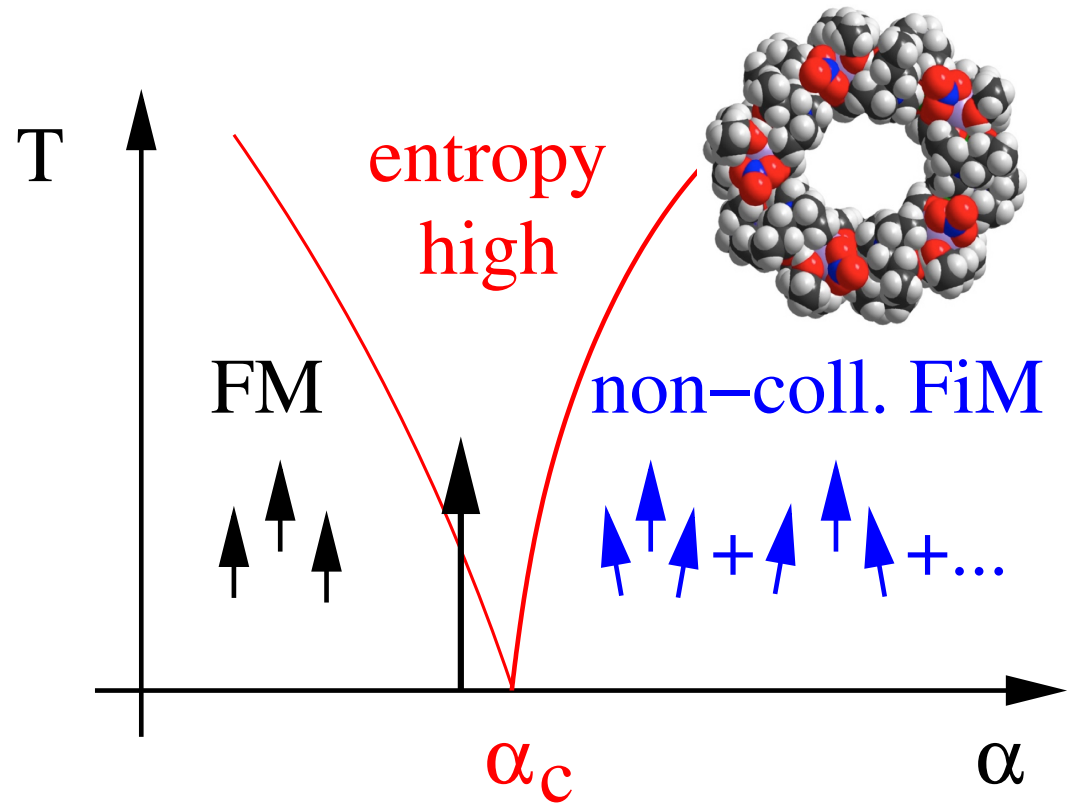
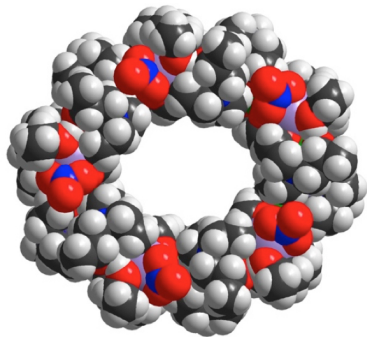


How do we know?

Gd₁₀Fe₁₀ is yummy



S=60



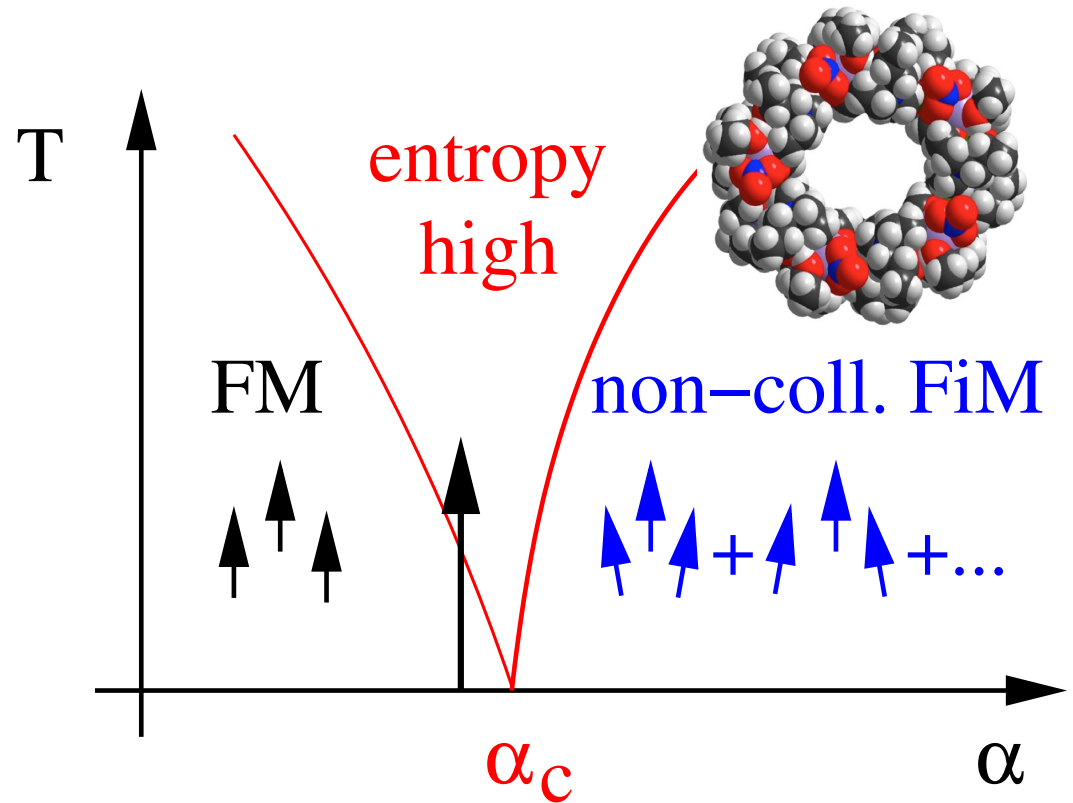
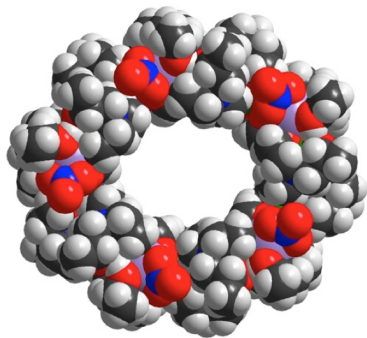
How do we know?

What is a QPT?

Gd₁₀Fe₁₀ is yummy



S=60

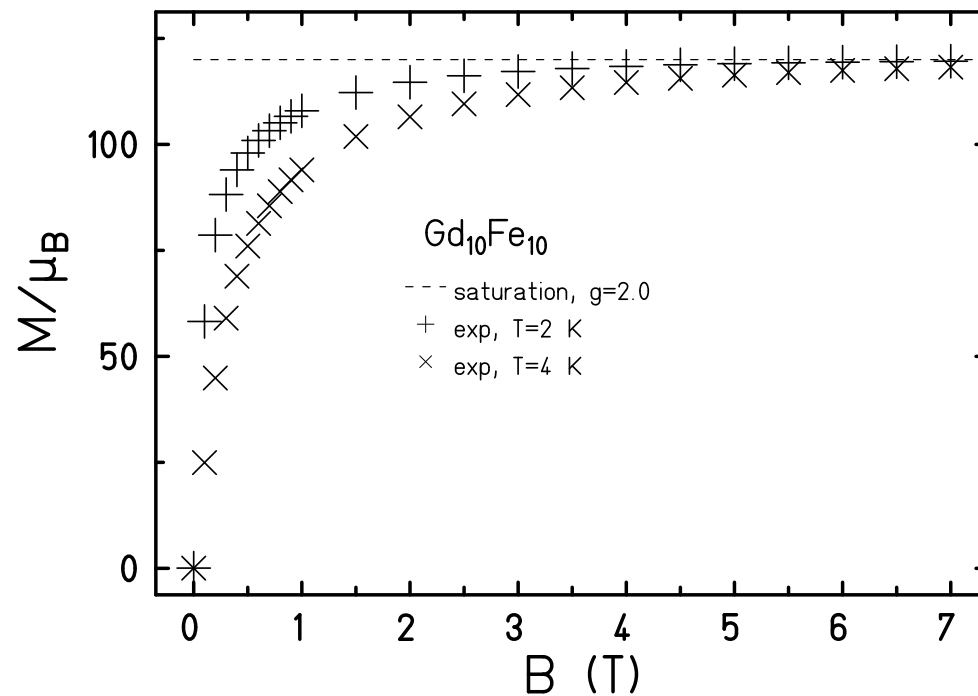
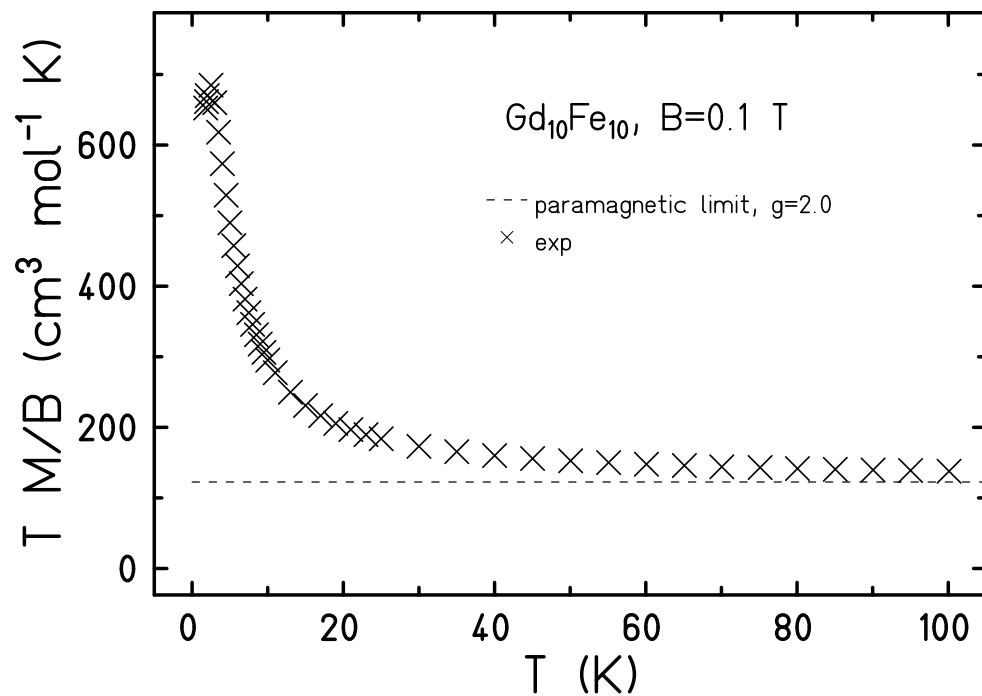


How do we know?

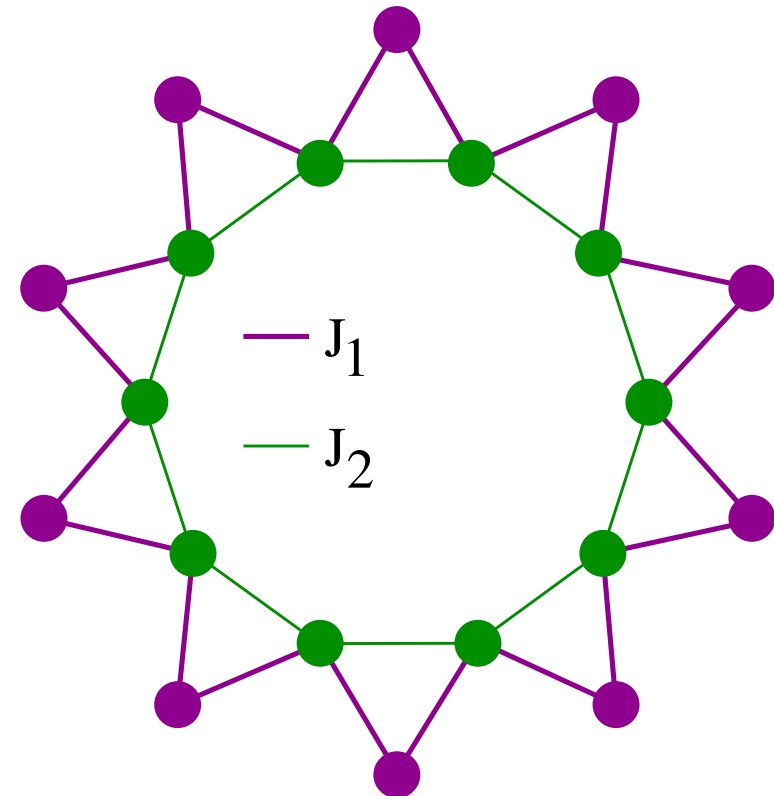
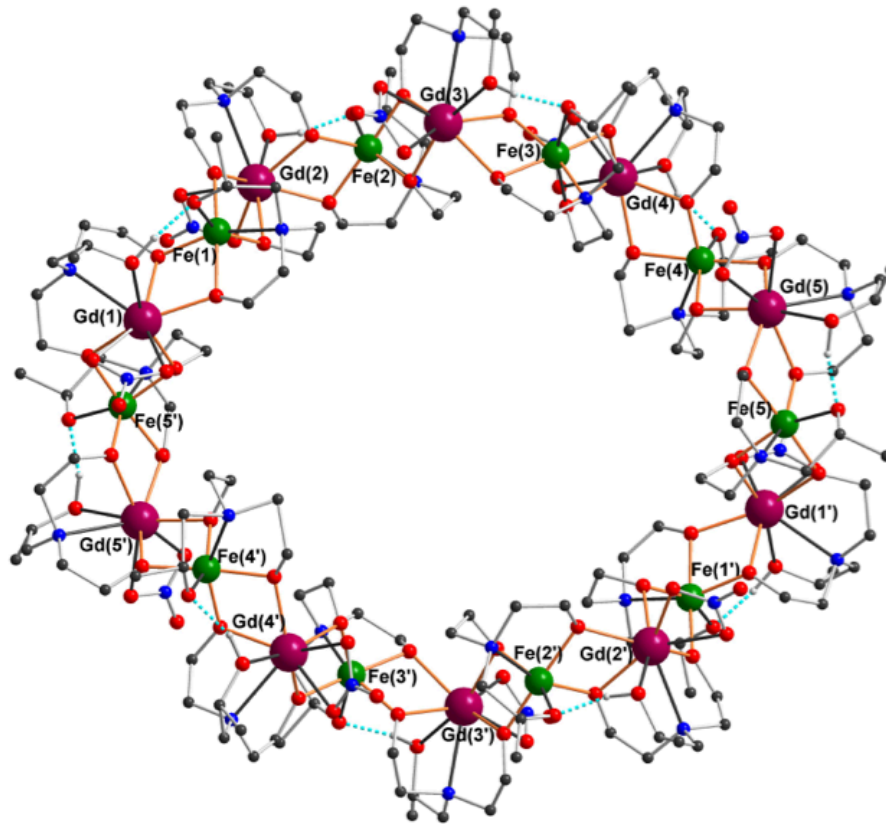
What is a QPT?
In Gd₁₀Fe₁₀?

Start: experimental data

Gd₁₀Fe₁₀ – How to rationalize the experimental data?



Gd₁₀Fe₁₀ – structure = delta chain



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

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Model Hamiltonian

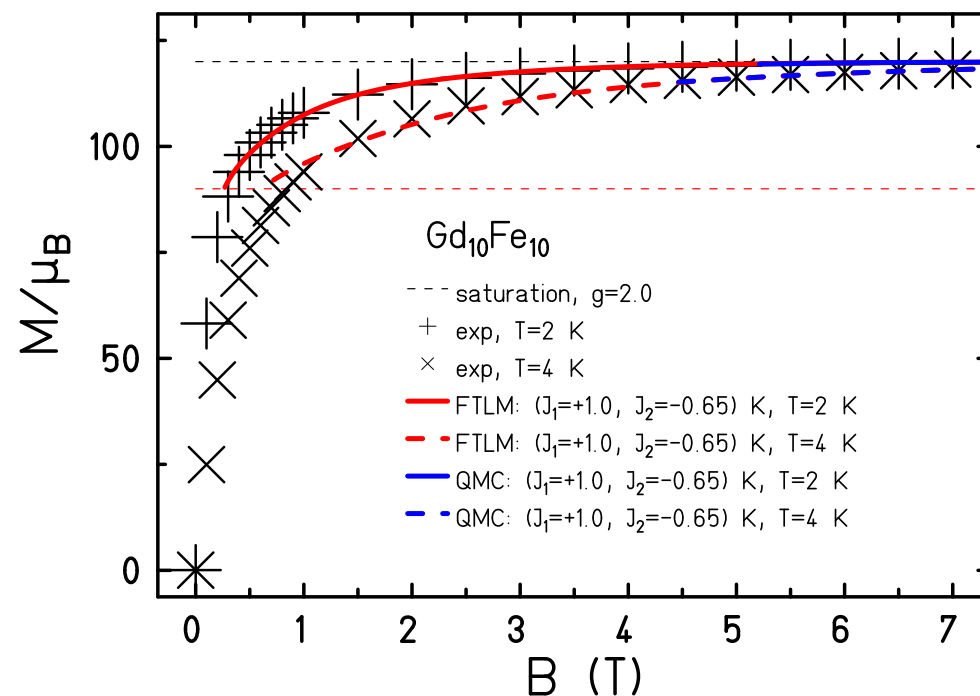
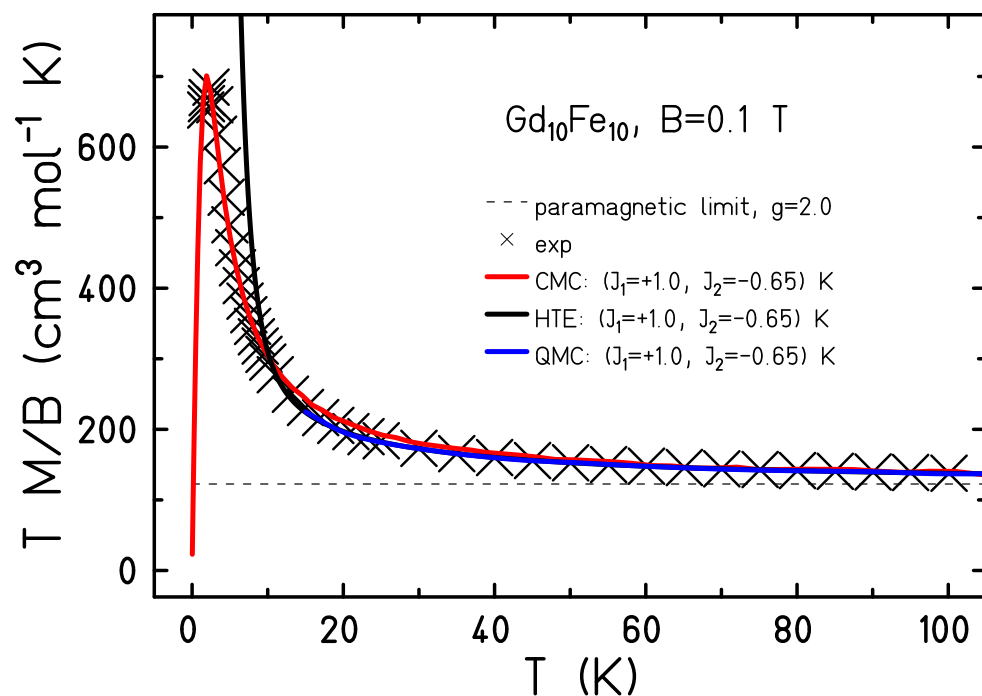
$$\begin{aligned} \underline{H} = & -2J_1 \sum_i \underline{\vec{S}}_{\text{Gd},i} \cdot \left(\underline{\vec{S}}_{\text{Fe},i} + \underline{\vec{S}}_{\text{Fe},i+1} \right) \\ & -2J_2 \sum_i \underline{\vec{S}}_{\text{Fe},i} \cdot \underline{\vec{S}}_{\text{Fe},i+1} + g \mu_B B \sum_i \left(\underline{S}_{\text{Gd},i}^z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

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

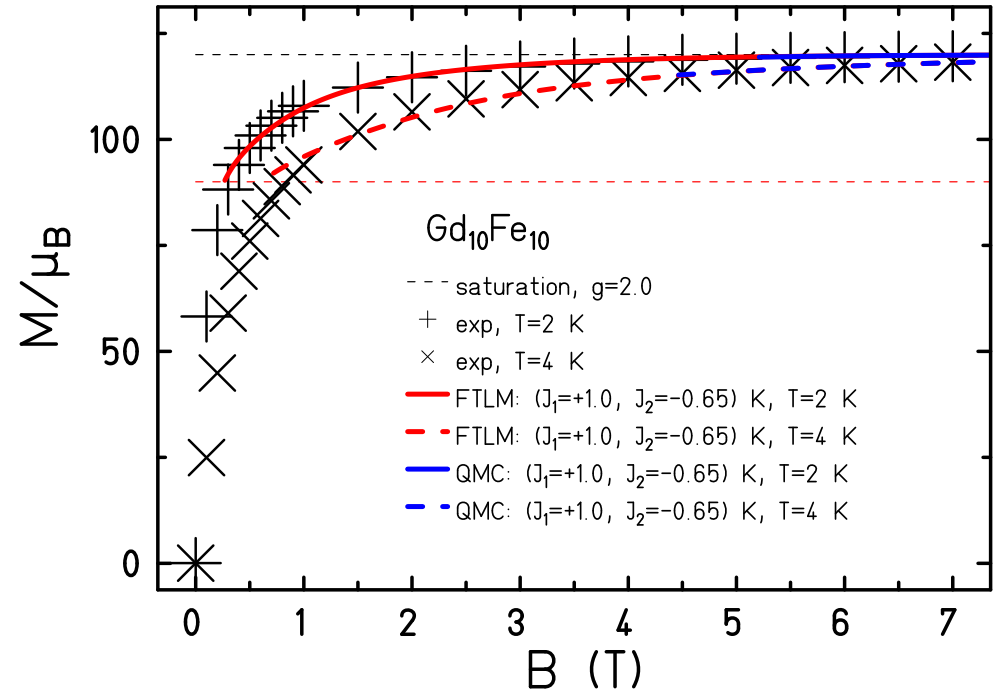
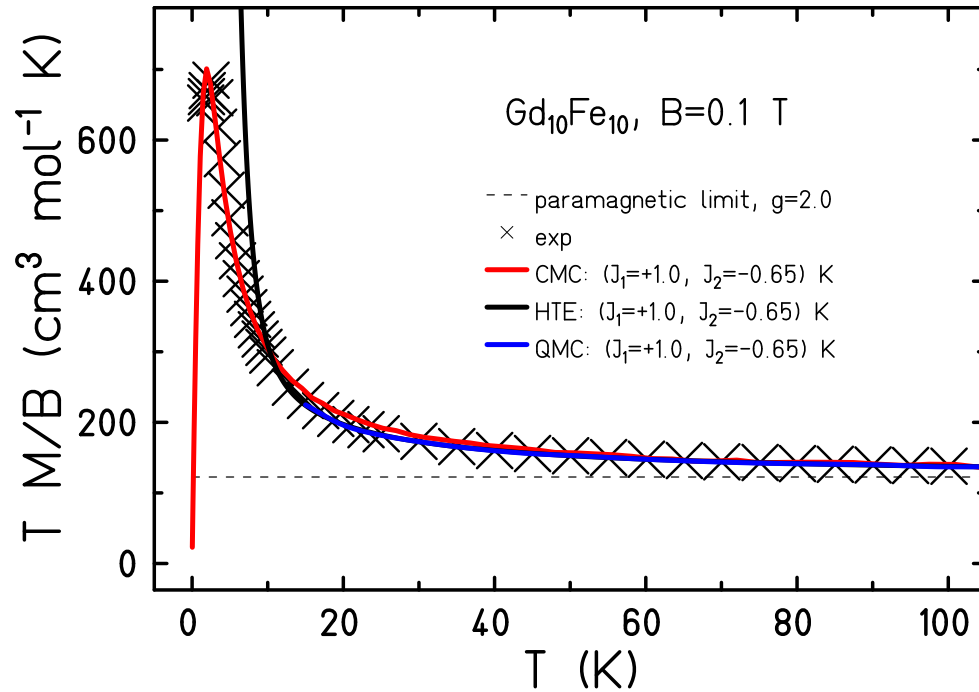
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Summary: theory methods

- **Complete diagonalization:** exact; Dimension of largest Hilbert space $< 10^5$.
- **High-temperature series expansion:** $\mathcal{O} \approx \sum_{\mu=0}^{\mu_{\max}} o_{\mu} T^{-\mu}$,
 o_{μ} known up to $\mu_{\max} = 6$ for mixed spin systems; $\mu_{\max} = 11$ otherwise [1].
- **Finite Temperature Lanczos Method (FTLM):** pseudo-spectrum, low-lying levels good, approximation of partition function, time-evolution; DoH $< 10^{10}$ [2].
- **Quantum Monte Carlo (QMC):** approximation of partition function, observables; bad/no convergence for competing interactions (frustration) due to negative sign problem; otherwise HUGE systems possible [ALPS].
- **Classical Monte Carlo (CMC):** spins are classical vectors; reasonable approximation for large spins such as $s = 5/2$ and $s = 7/2$.

[1] H.-J. Schmidt, A. Lohmann, J. Richter, Phys. Rev. B 84, 104443 (2011); Phys. Rev. B 89, 014415 (2014). [2] J. Jaklic and P. Prelovsek, Phys. Rev. B **49**, 5065 (1994); J. Schnack and O. Wendland, Eur. Phys. J. B **78** (2010) 535-541.

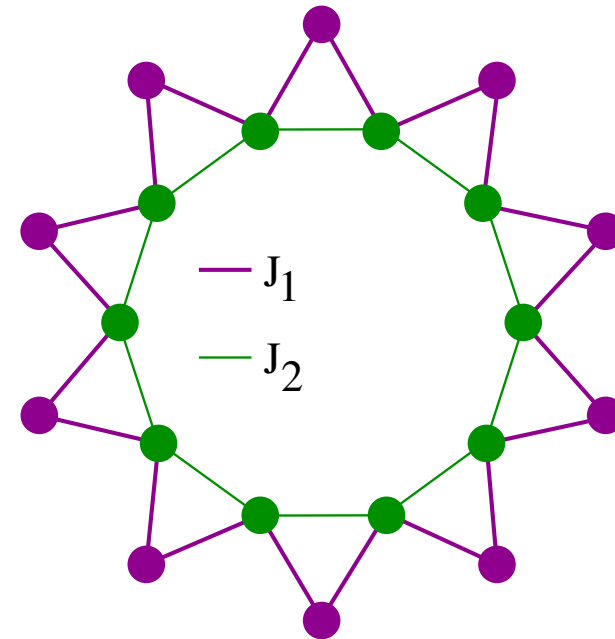
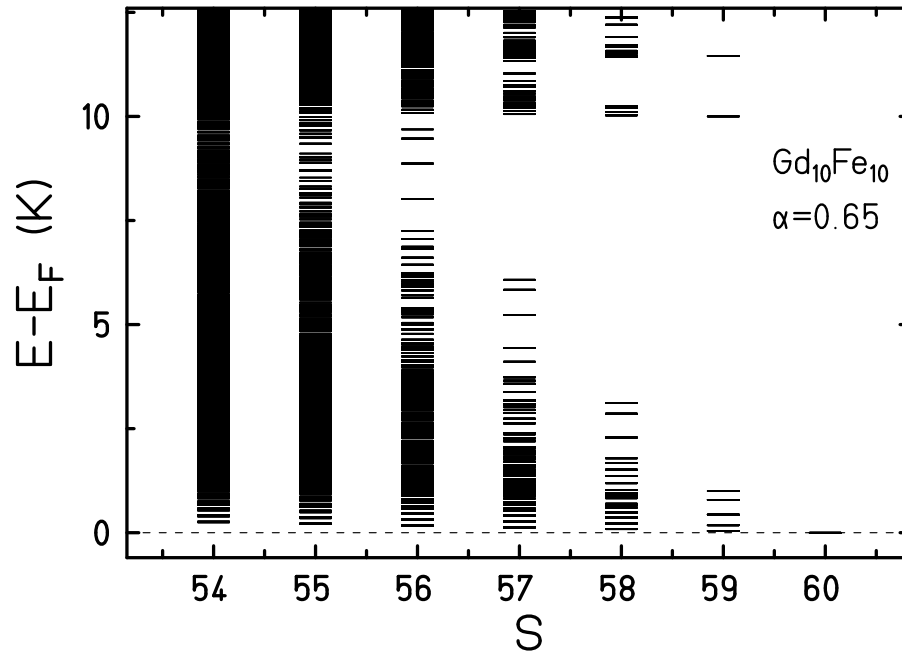
Gd₁₀Fe₁₀ – Methods



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

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Gd₁₀Fe₁₀ – $S = 60$

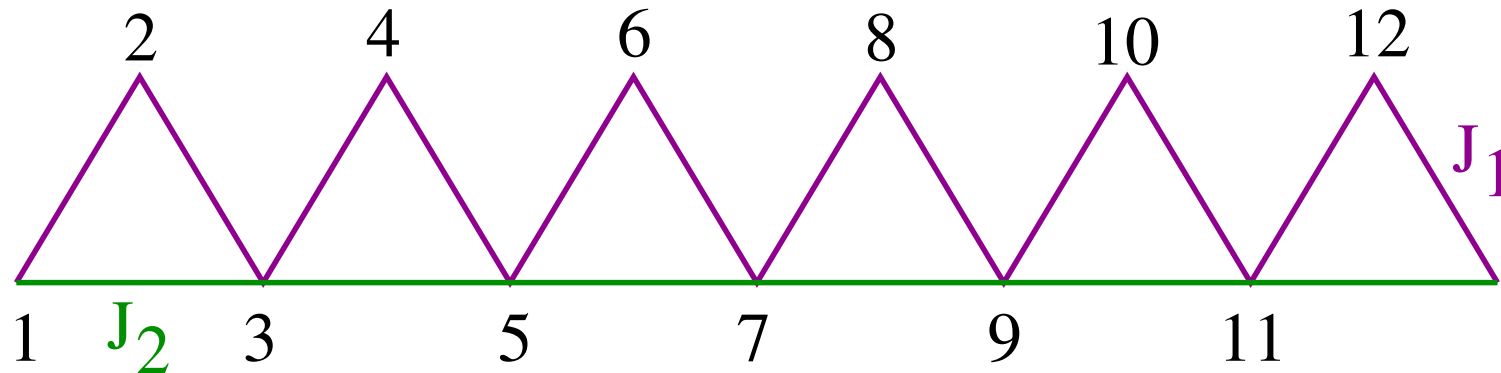


⇒ $S = 60$, largest ground state spin of a molecule to date

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

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Excursus: sawtooth (delta) chain



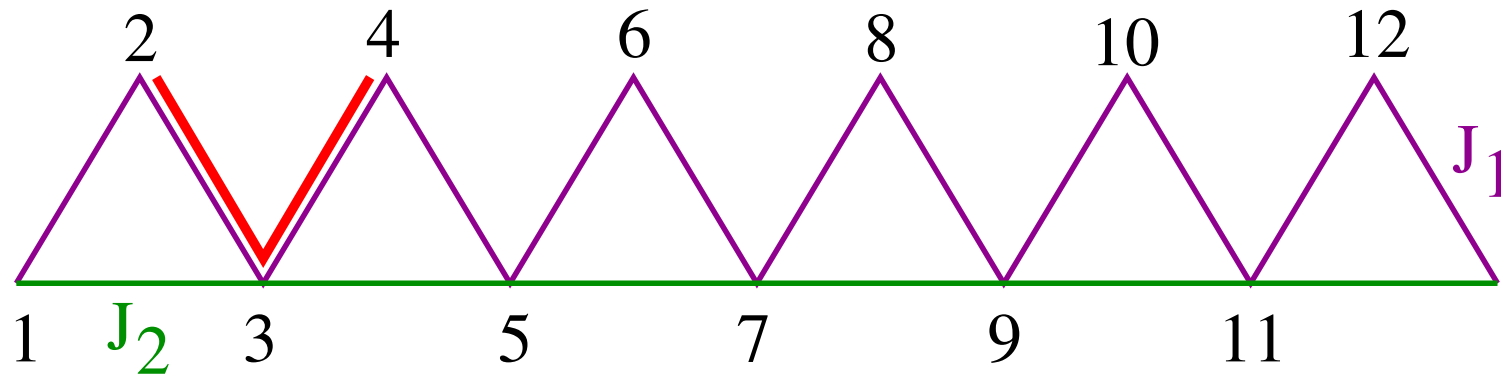
⇒ special properties for $J_1 > 0$ (ferro) and $J_2 < 0$ (af) at certain α_c

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

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

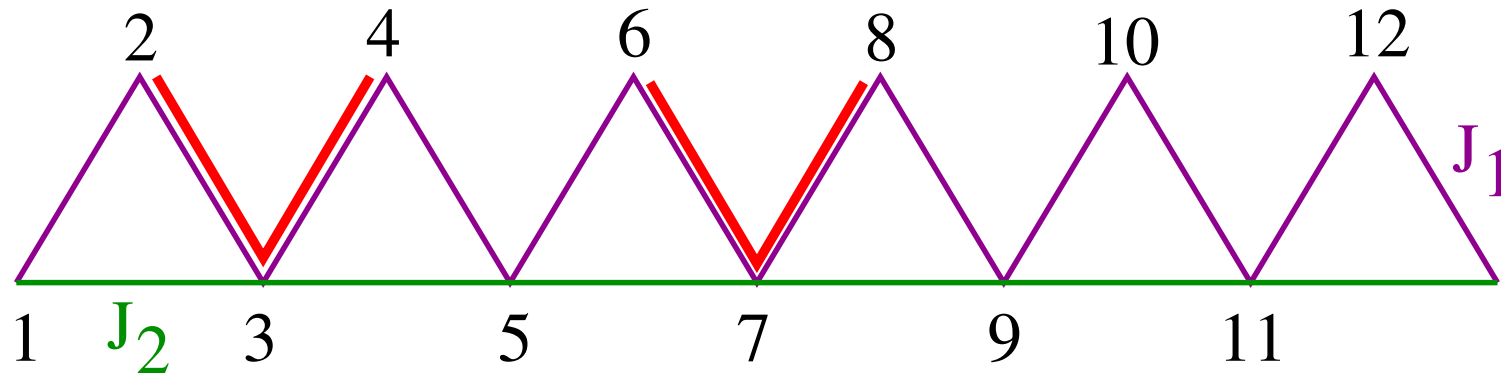
$$E = E_F, M = S_{\max} - 1$$

\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).

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Excursus: sawtooth (delta) chain

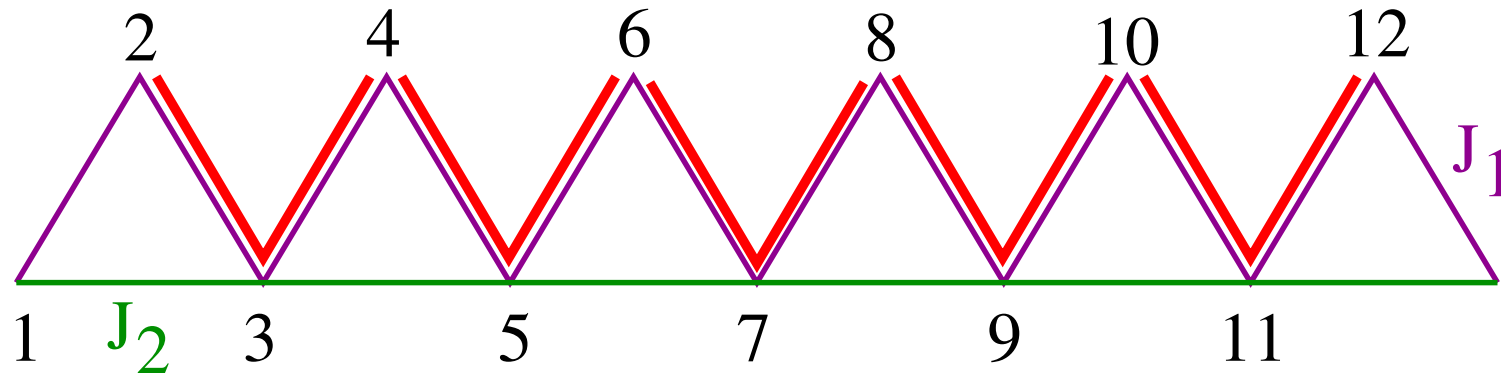


\Rightarrow $|2 \text{ localized magnons}\rangle; E = E_F, M = S_{\max} - 2$

\Rightarrow 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).
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Excursus: sawtooth (delta) chain



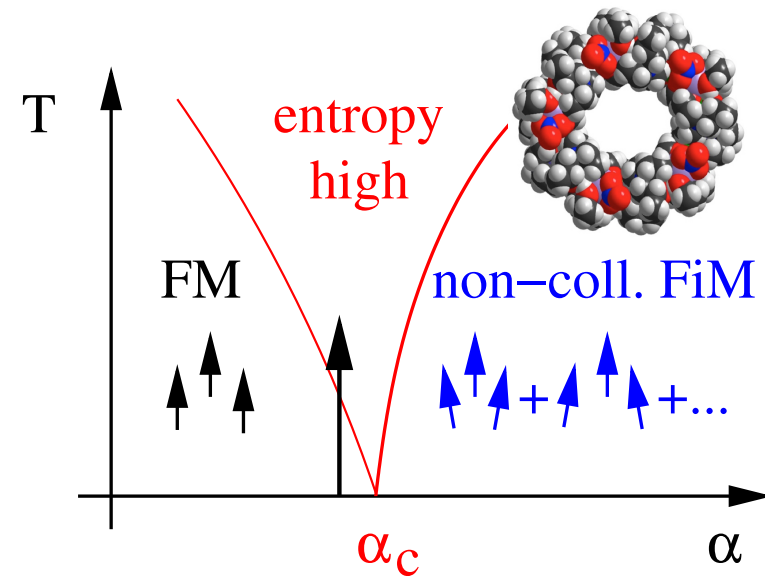
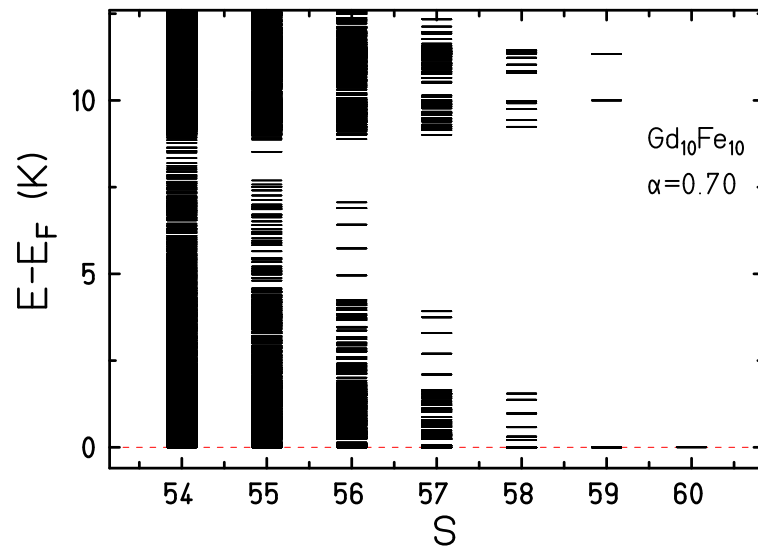
⇒ | max. number of localized magnons \rangle ; $E = E_F, M = S_{\max} - N/2$

⇒ 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

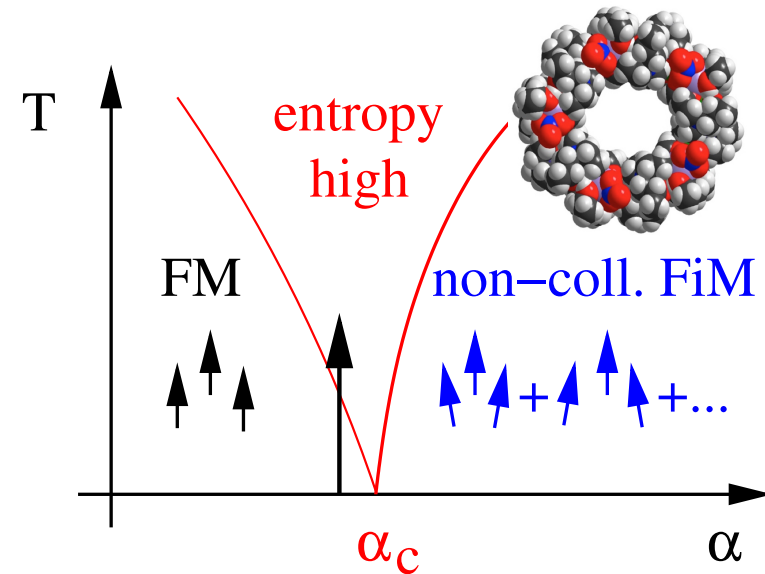
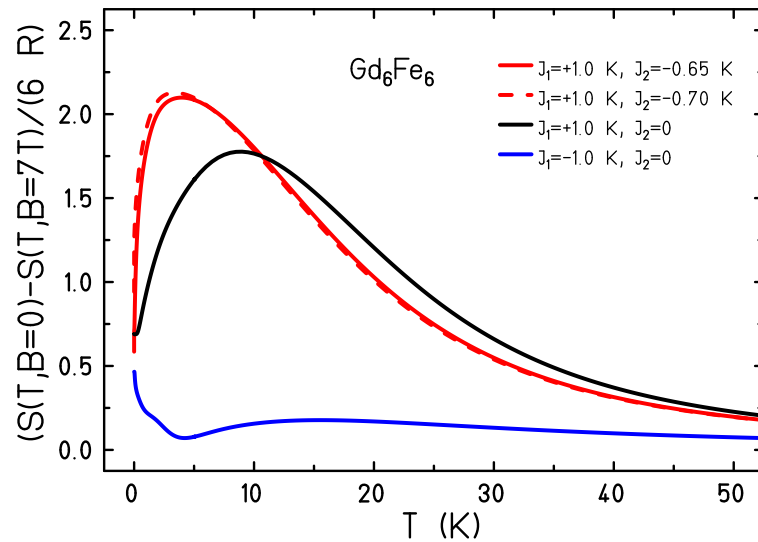
\Rightarrow for $s_1 = 5/2$ and $s_2 = 7/2$: $\alpha_c = 0.70$

\Rightarrow 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)

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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.

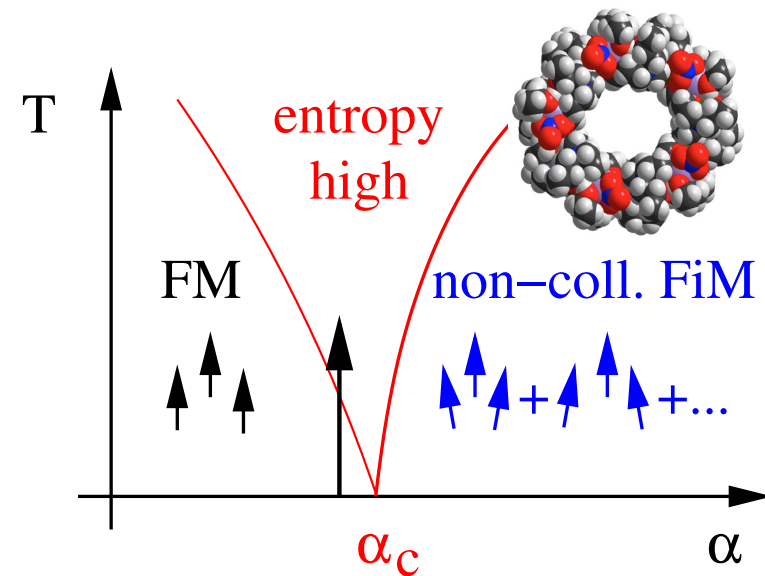
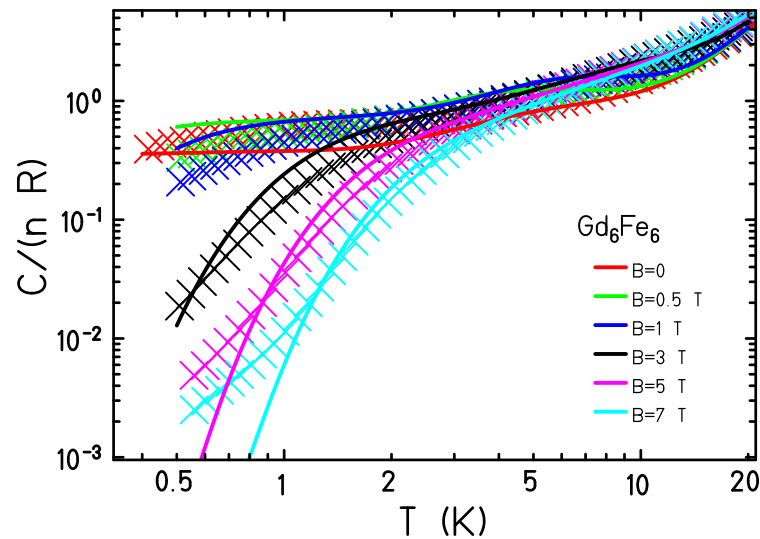
Gd₁₀Fe₁₀ – $T > 0$ 

\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 $\alpha = 0.65$.

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Gd₁₀Fe₁₀ – heat capacity



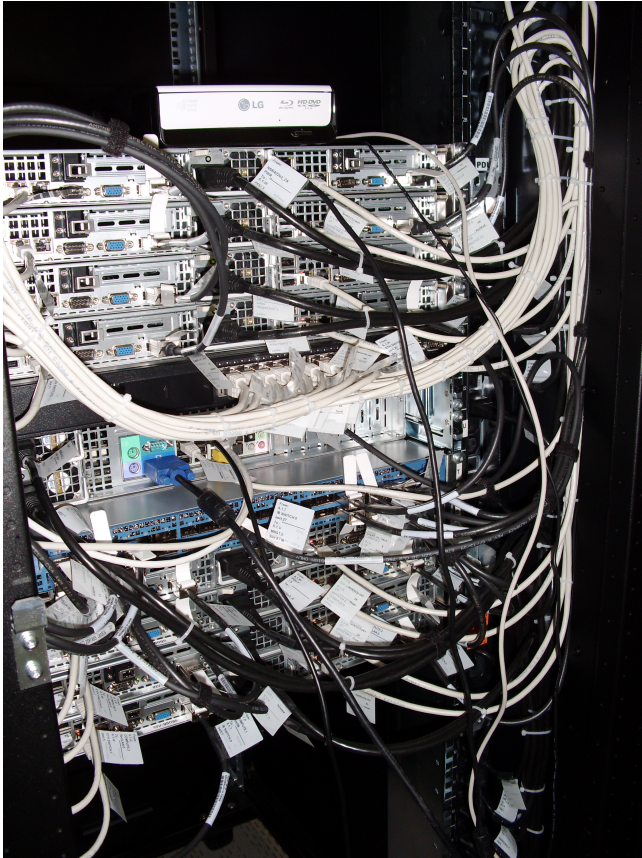
⇒ heat capacity assumes very large values even down to lowest temperatures;

⇒ evaluated by means of FTLM for a smaller (hypothetical) system Gd_6Fe_6 ;

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

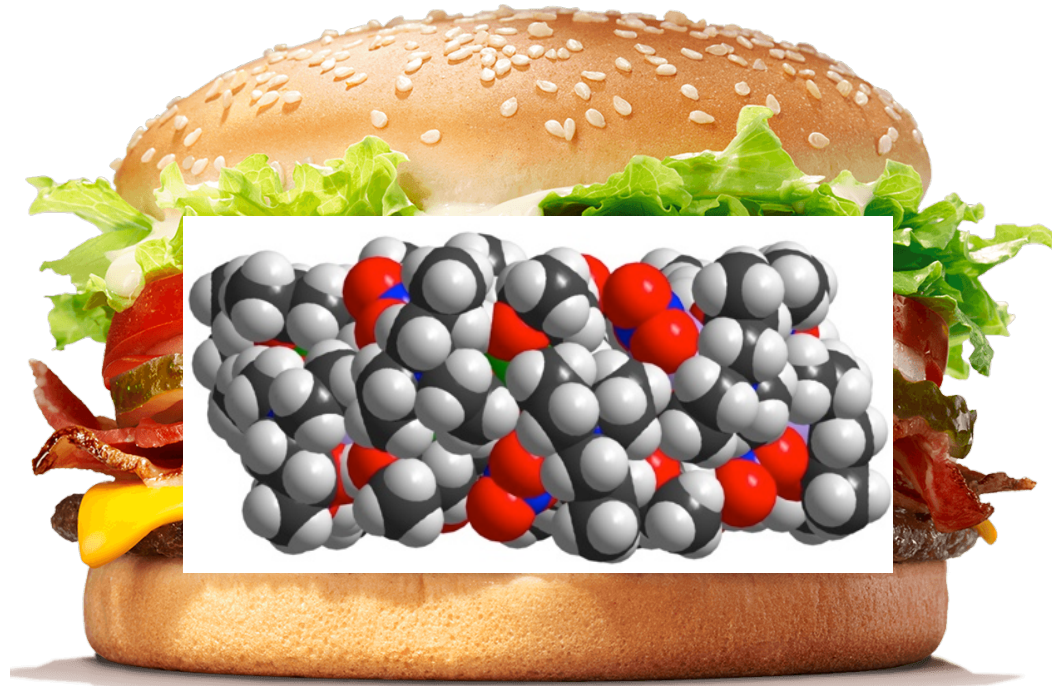
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Gd₁₀Fe₁₀ – Summary



- 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.

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Man does not live by bread alone!



Many thanks to my collaborators



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Thank you very much for your
attention.

The end.

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