High-spin cycles showing quantum criticality

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The Bielefeld conspiracy

The story goes that the city of **BIELEFELD** in the German state of North Rhine-Westphalia **DOES NOT** actually **EXIST**. Rather, its existence is merely propagated by an entity known only as THEM, which has conspired with the authorities to create the illusion of the city's existence.

The origins of and reasons for this conspiracy are not a part of the original theory. Speculated originators jokingly include the CIA, Mossad, or aliens who use Bielefeld University as a disguise for their spaceship.

Do you know anybody from Bielefeld?

https://en.wikipedia.org/wiki/Bielefeld_Conspiracy

The Bielefeld conspiracy

Around 2,000 People Vie to Prove German City Doesn't Exist

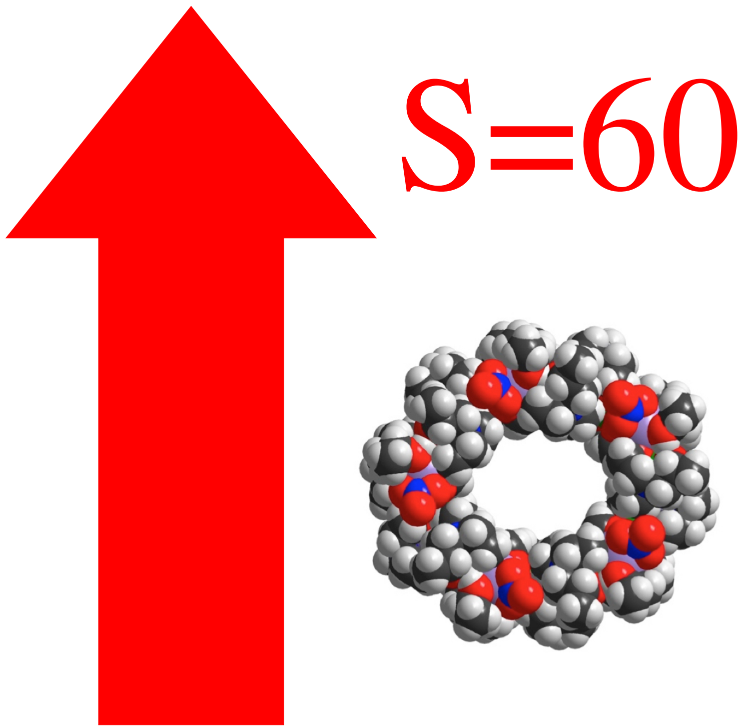
By The Associated Press

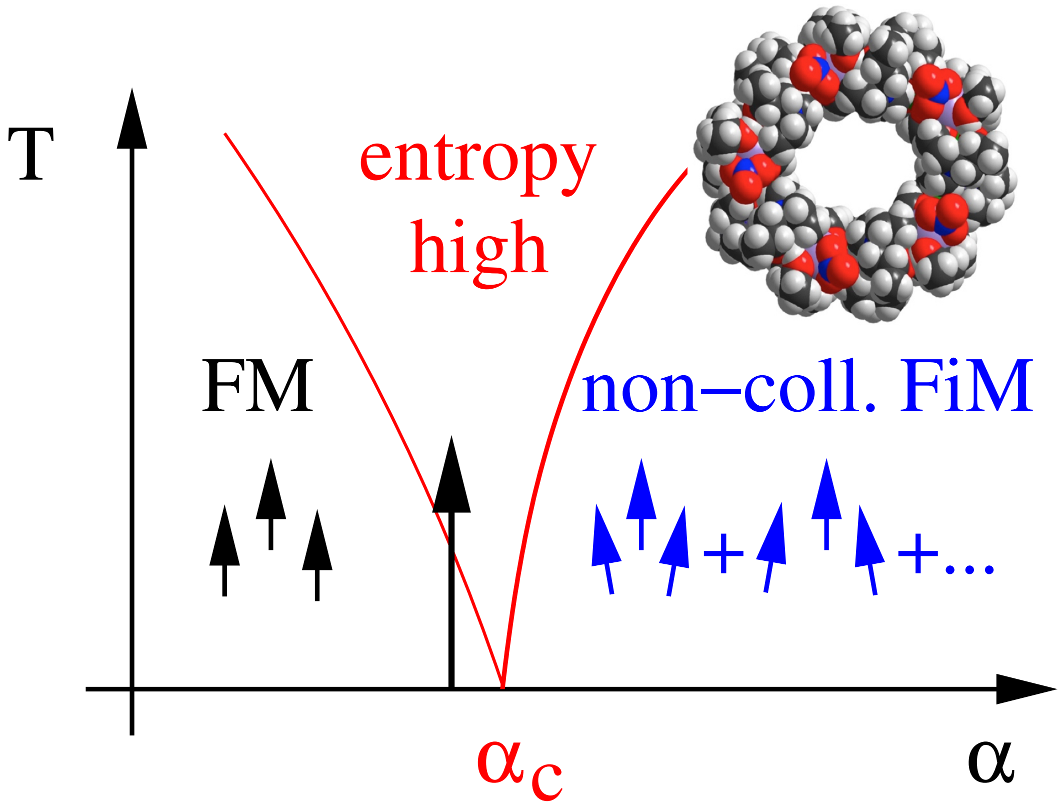
Sept. 5, 2019

BERLIN — Around 2,000 people are vying to win 1 million euros ($1.1 million) offered by a German city to anyone who can deliver solid proof it doesn't exist.

Officials in Bielefeld announced the competition last month, saying that there are "no limits to creativity" but only incontrovertible evidence will be rewarded. The idea that the western city doesn't

$Gd_{10}Fe_{10}$ – 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)

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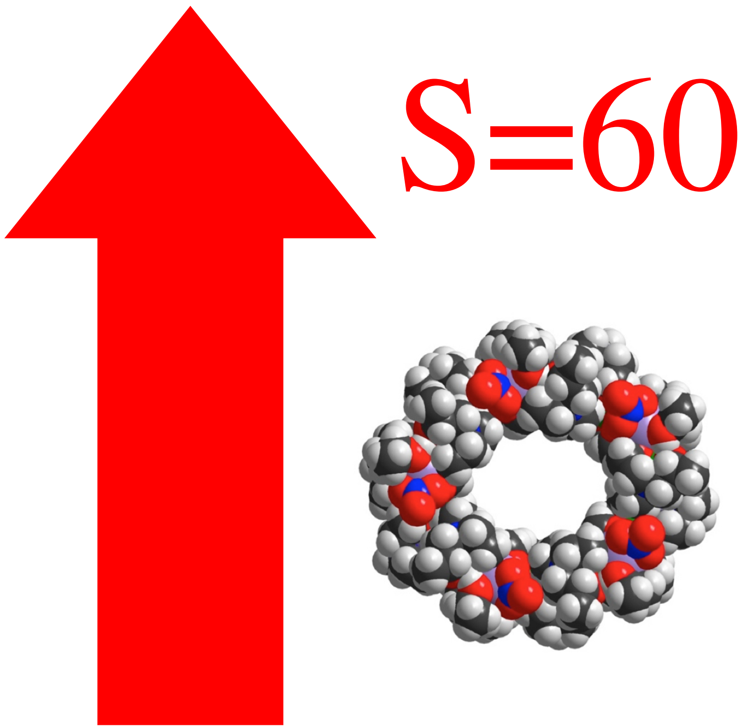
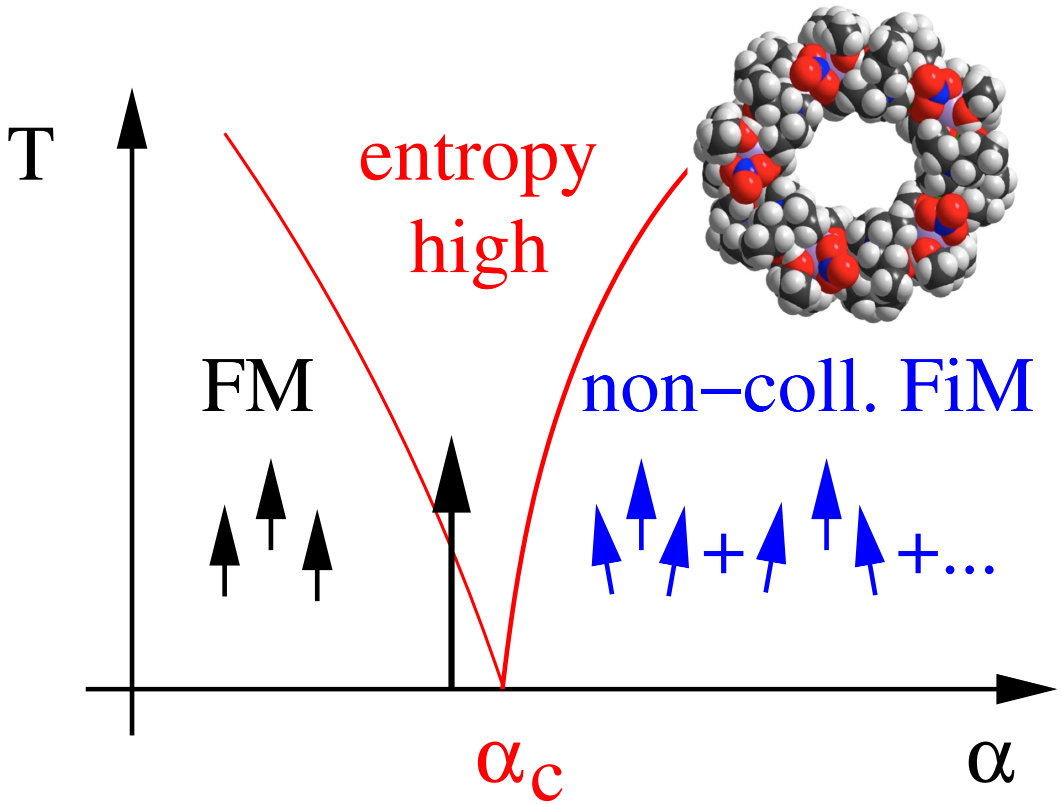
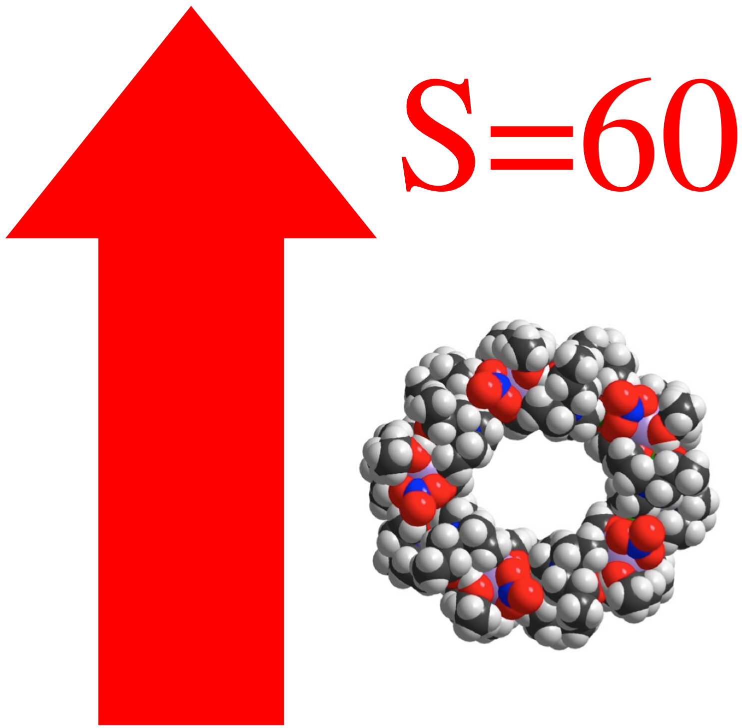


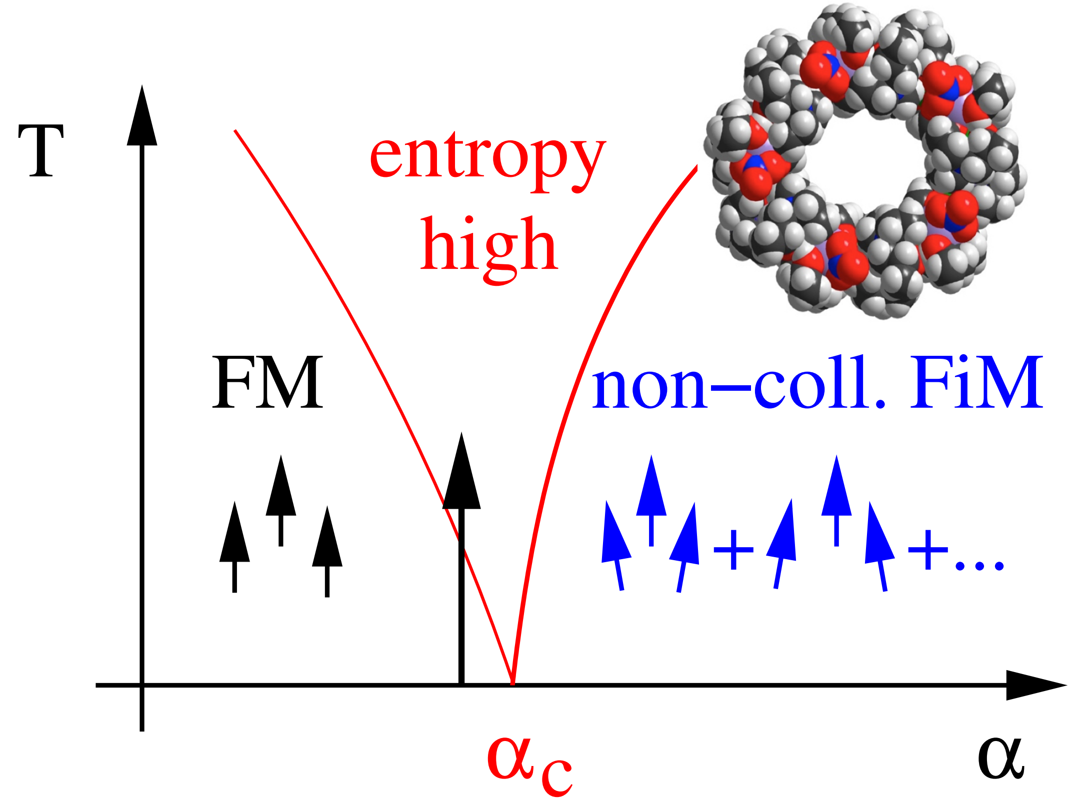


How do we know?

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

$Gd_{10}Fe_{10}$ – summary





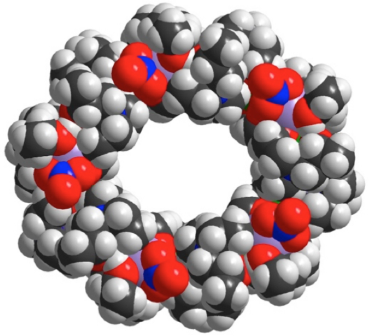
How do we know?

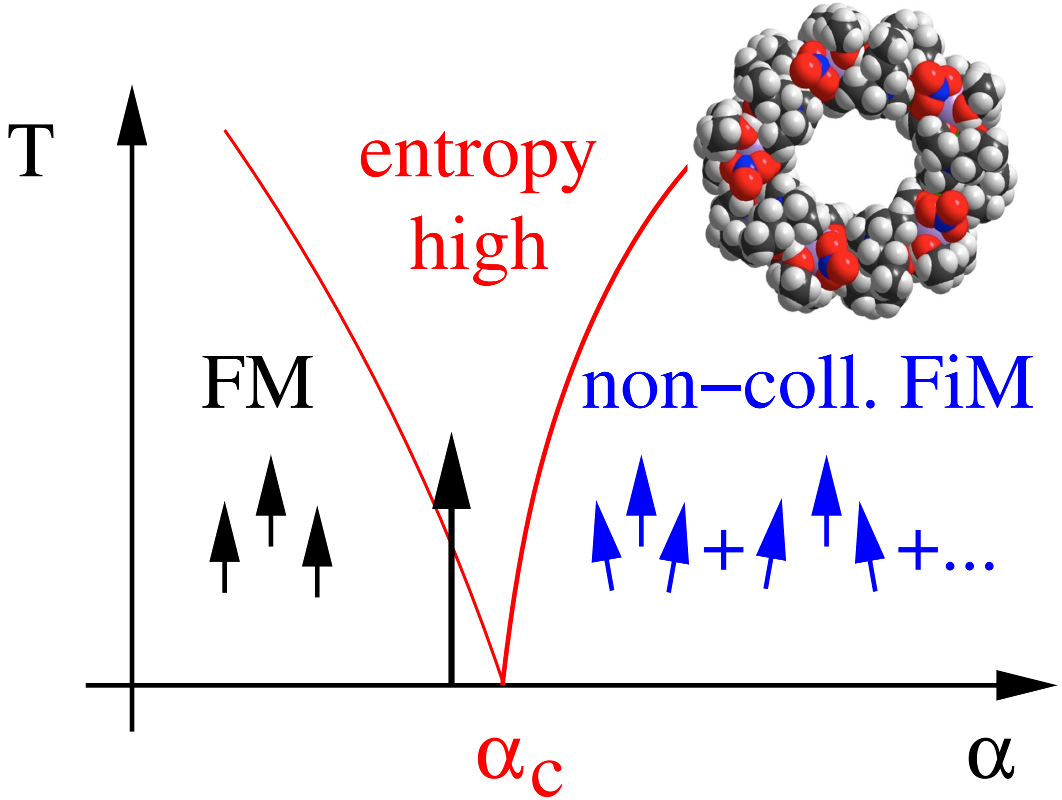
What is a QPT?

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

$Gd_{10}Fe_{10}$ – summary

S=60





How do we know?

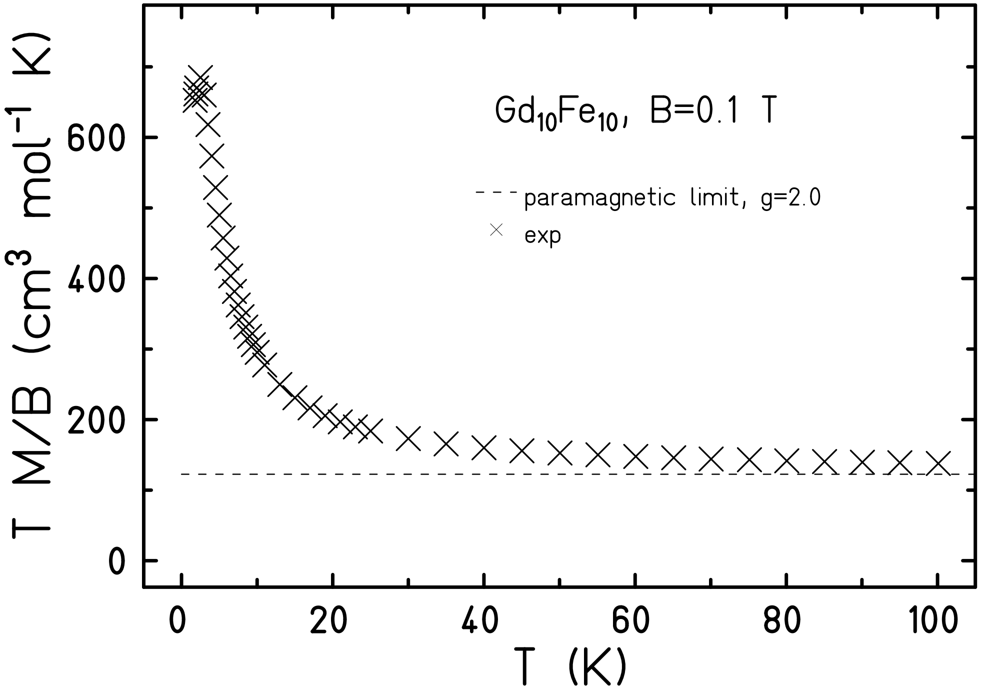
What is a QPT?

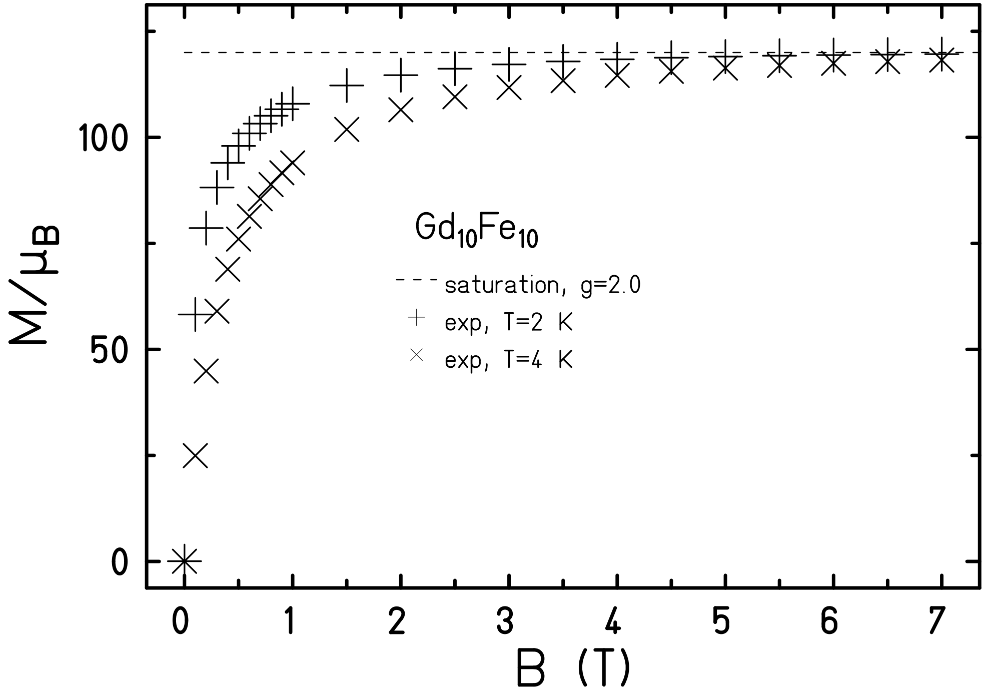
In $Gd_{10}Fe_{10}$?

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

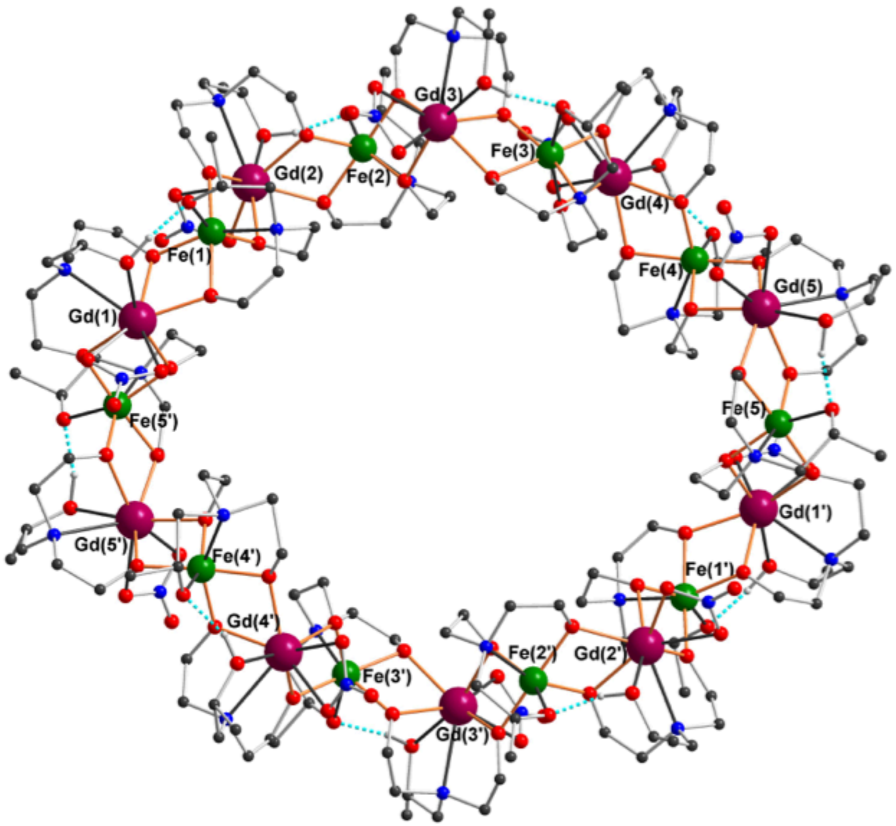
Start: experimental data

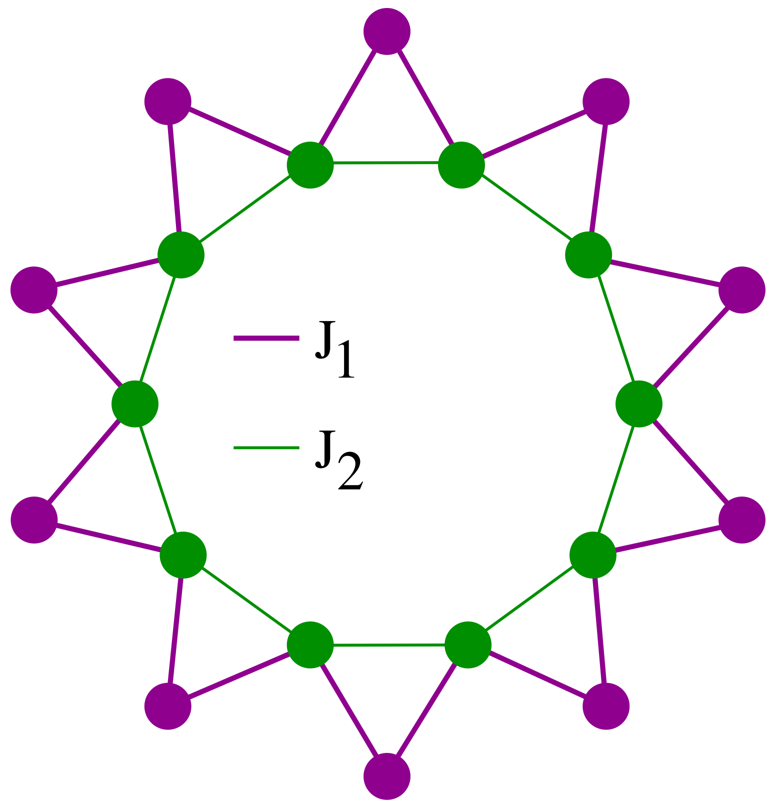
$Gd_{10}Fe_{10}$ – How to rationalize the experimental data?





$Gd_{10}Fe_{10}$ – structure = delta chain





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

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Model Hamiltonian

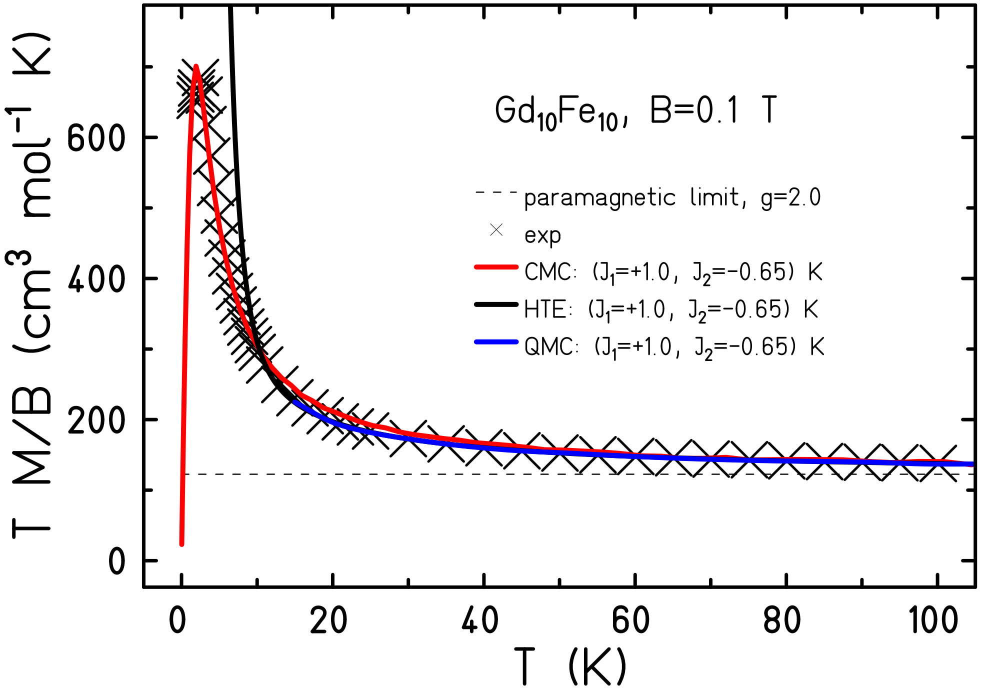
$$\underset{\sim}{H} = -2J_1 \sum_i \underset{\sim}{\vec{s}}_{\text{Gd},i} \cdot \left(\underset{\sim}{\vec{s}}_{\text{Fe},i} + \underset{\sim}{\vec{s}}_{\text{Fe},i+1} \right)$$

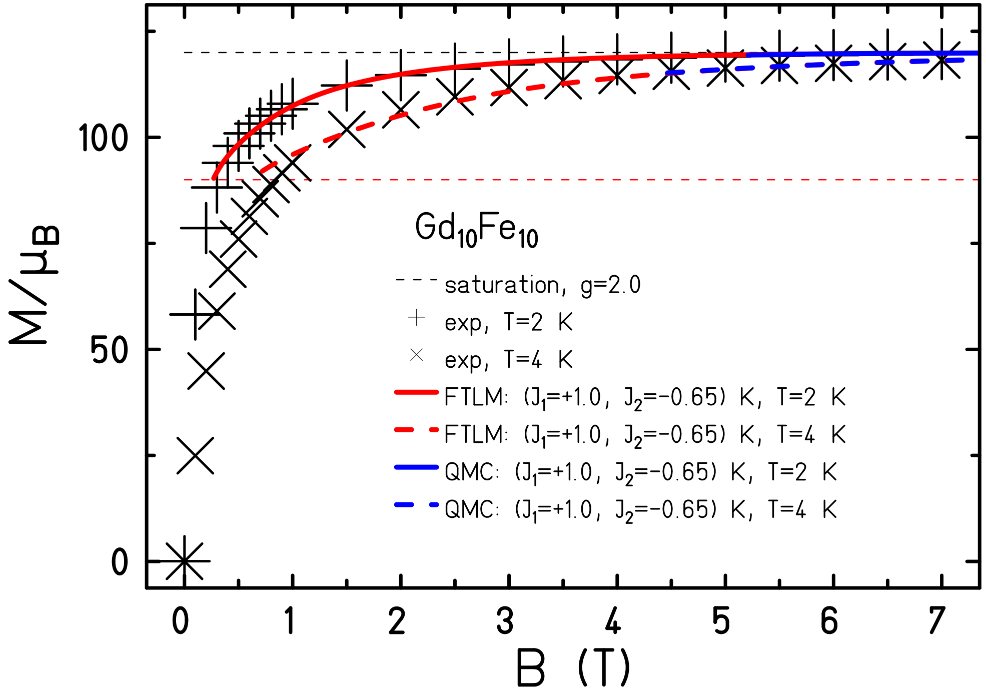
$$-2J_2 \sum_i \underset{\sim}{\vec{s}}_{\text{Fe},i} \cdot \underset{\sim}{\vec{s}}_{\text{Fe},i+1} + g\,\mu_B\,B \sum_i \left(\underset{\sim}{s}^z_{\text{Gd},i} + \underset{\sim}{s}^z_{\text{Fe},i} \right)$$

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

What would you do?

$Gd_{10}Fe_{10}$ – Methods





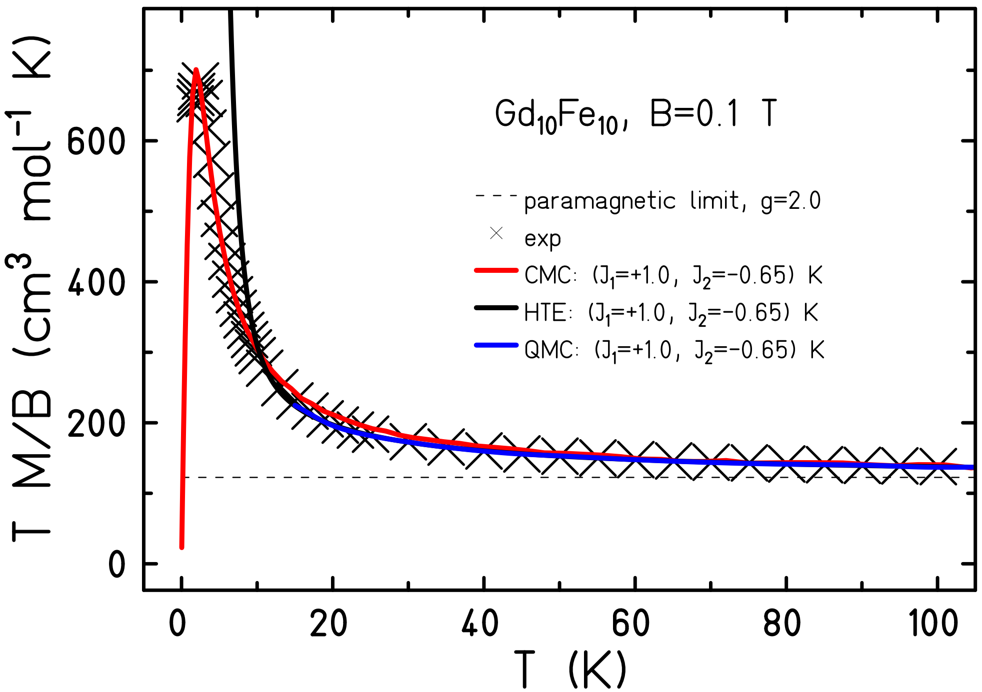
Methods: HTE, QMC, CMC, FTLM $\Rightarrow J_1 = 1.0$ K, $J_2 = -0.65$ K

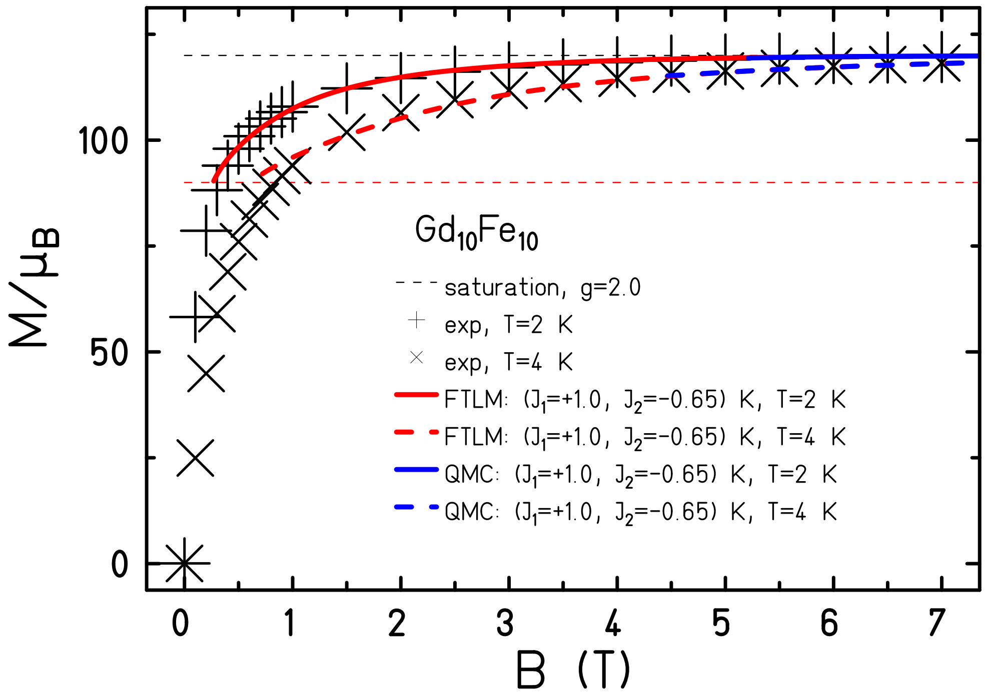
A. Baniodeh *et al., npj Quantum Materials* ***3****, 10 (2018)*

Summary: theory methods

- Complete diagonalization: exact; spectra, transitions, observables, time-evolution; Dimension of largest Hilbert space $< 10^5$.
- High Temperature Series Expansion (HTE): write thermodynamic functions as a Taylor series in β, evaluate coefficients according to your model; powers of order 10 possible.
- Finite Temperature Lanczos Method (FTLM): pseudo-spectrum, low-lying levels good, transitions, observables, time-evolution; DoH $< 10^{10}$.
- Quantum Monte Carlo (QMC): observables; bad/no convergence for competing interactions (frustration) due to negative sign problem; otherwise HUGE systems possible.
- Classical Monte Carlo (CMC): treat spins as classical vectors; good approximation for large spins; no quantum effects; really HUGE systems possible.

Gd$_{10}$Fe$_{10}$ – Methods





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

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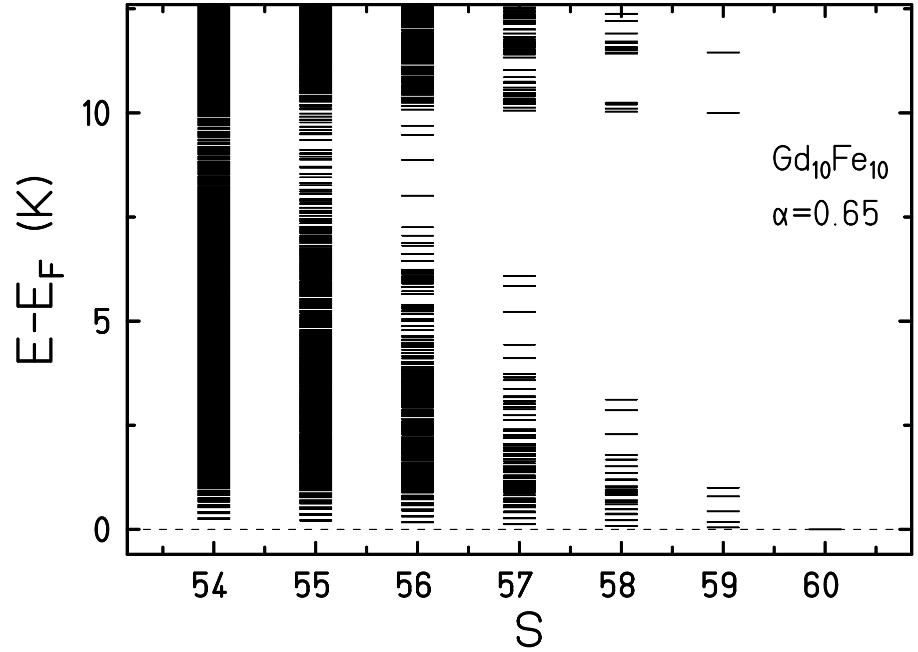
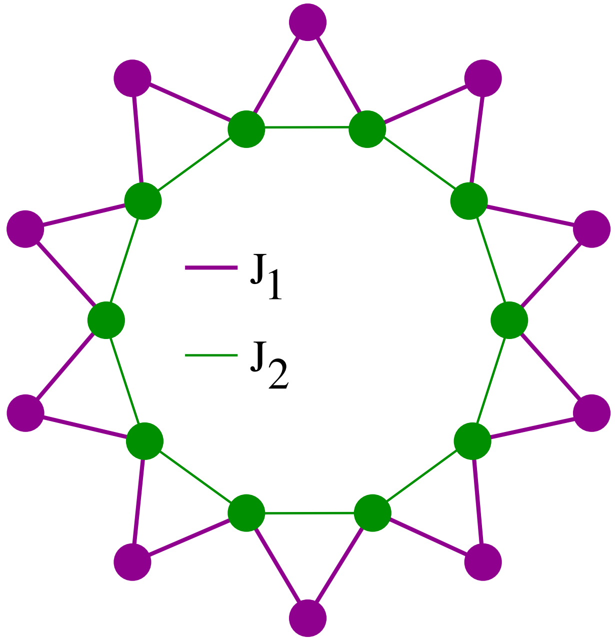
$Gd_{10}Fe_{10}$ – $S = 60$





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

⇒ $\alpha_{Gd_{10}Fe_{10}} = |J_2|/J_1 = 0.65$ What if J_2 stronger?

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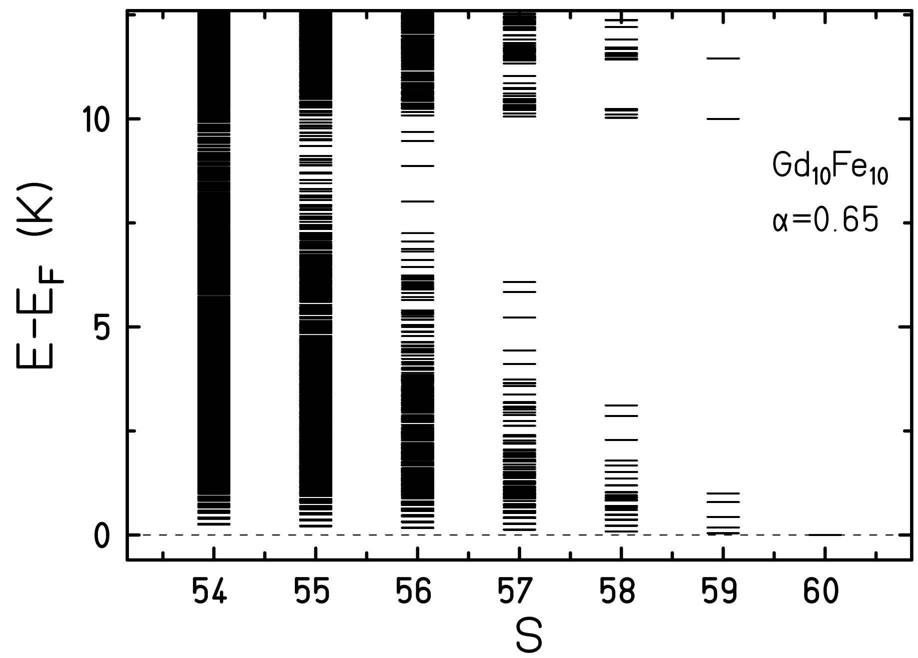
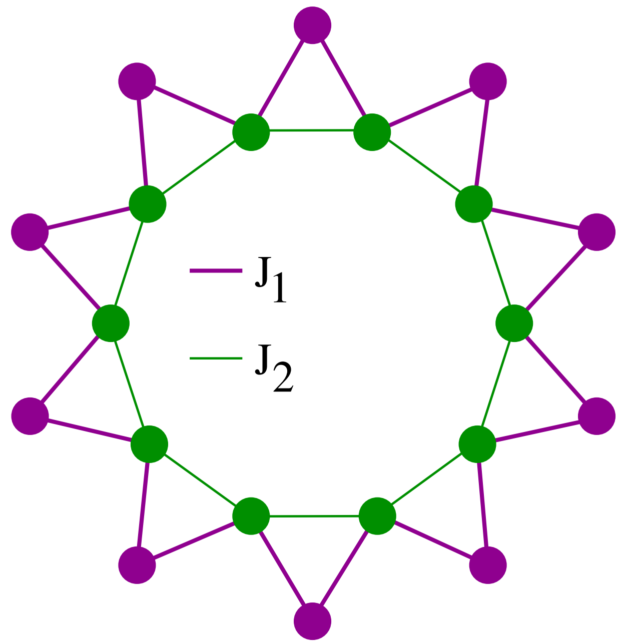
$Gd_{10}Fe_{10}$ – $S = 60$



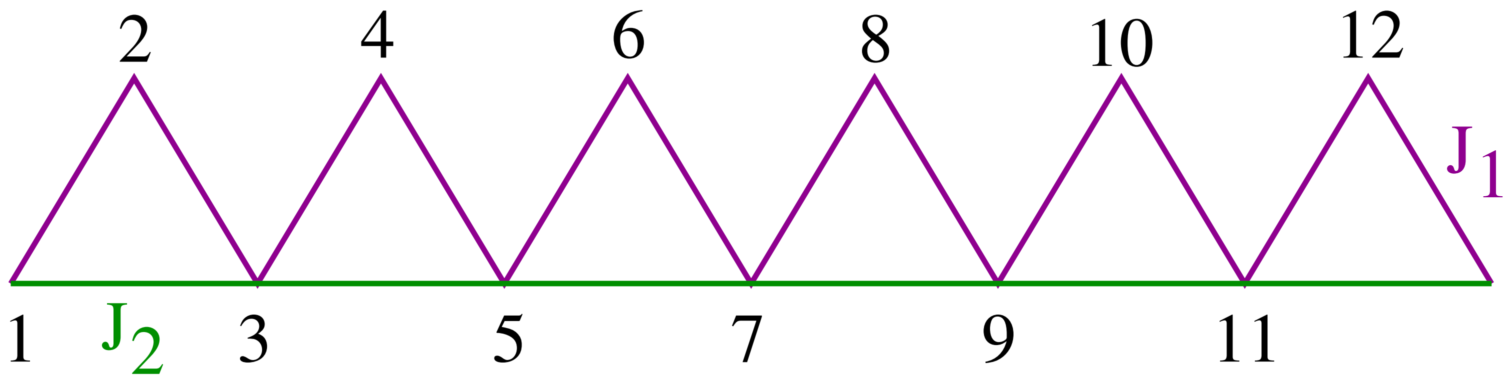


$\Rightarrow$ $S = 60$, largest ground state spin of a molecule ~~to date~~ for about one month

$\Rightarrow$ $\alpha_{Gd_{10}Fe_{10}} = |J_2|/J_1 = 0.65$ What if J_2 stronger?

Wei-Peng Chen, Jared Singleton, Lei Qin, Agustin Camon, Larry Engelhardt, Fernando Luis, Richard E. P. Winpenny, Yan-Zhen Zheng, Quantum Monte Carlo simulations of a giant $\{Ni_{21}Gd_{20}\}$ cage with a $S = 91$ spin ground state, Nature Communications **9**, 2107 (2018)

Excursus: sawtooth (delta) chain



$\Rightarrow$ 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 $s_i = 1/2\ \forall i$

$\Rightarrow$ 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).

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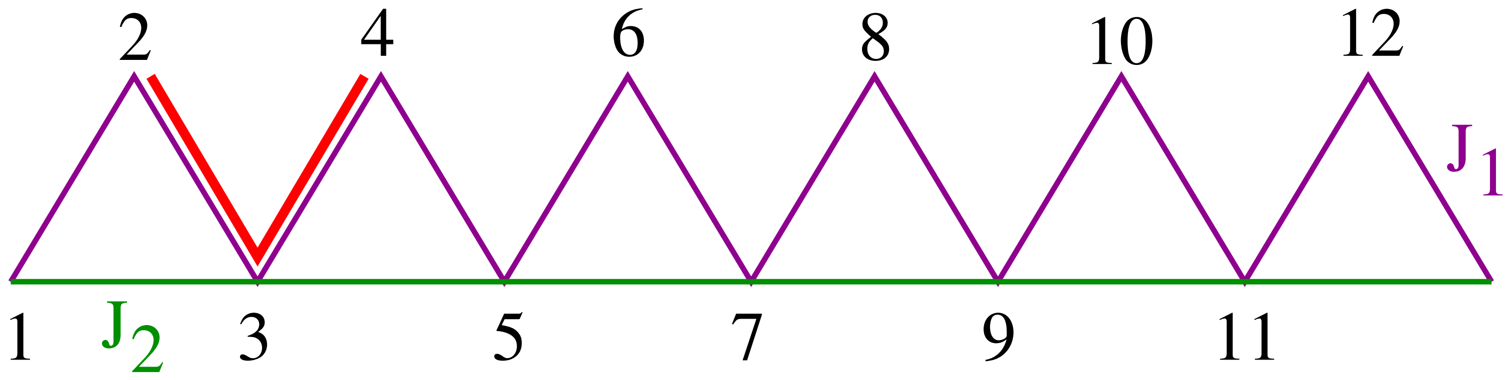

$\Rightarrow$ $|\, F\,\rangle = |\, S = S_{\text{max}}, M = S_{\text{max}}\,\rangle$ fully polarized ferromagnetic state

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

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

$\Rightarrow$ 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).
(3) J. Schnack, Contemporary Physics (2019), doi:10.1080/00107514.2019.1615716

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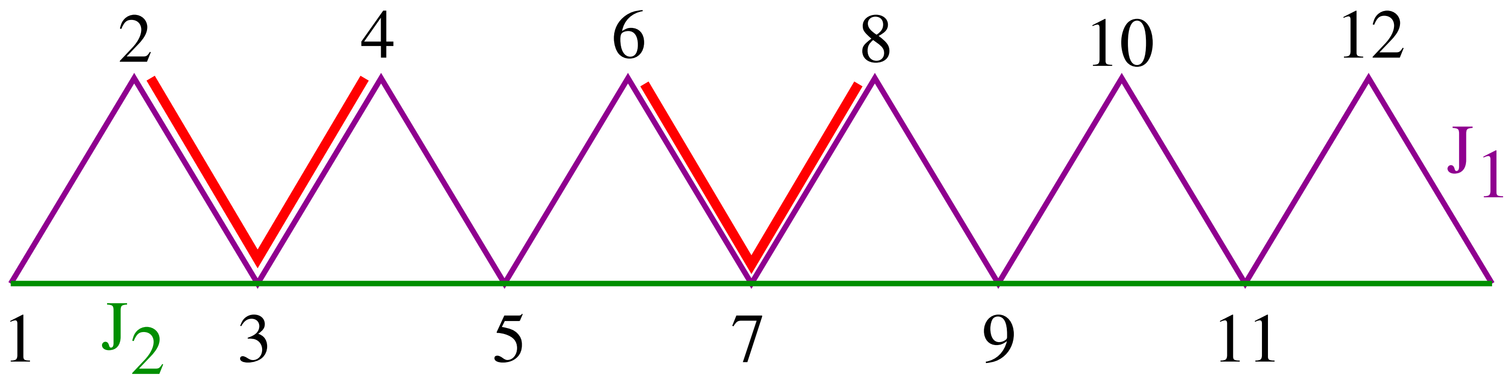
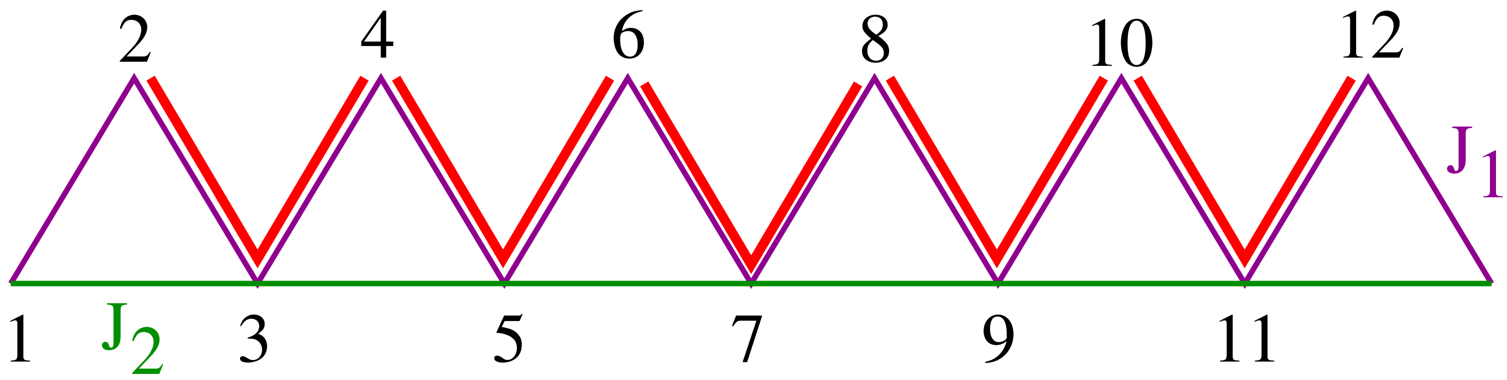


$\Rightarrow$ $|$ 2 localized magnons $\rangle$; $E = E_F, M = S_{\text{max}} - 2$

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

Excursus: sawtooth (delta) chain



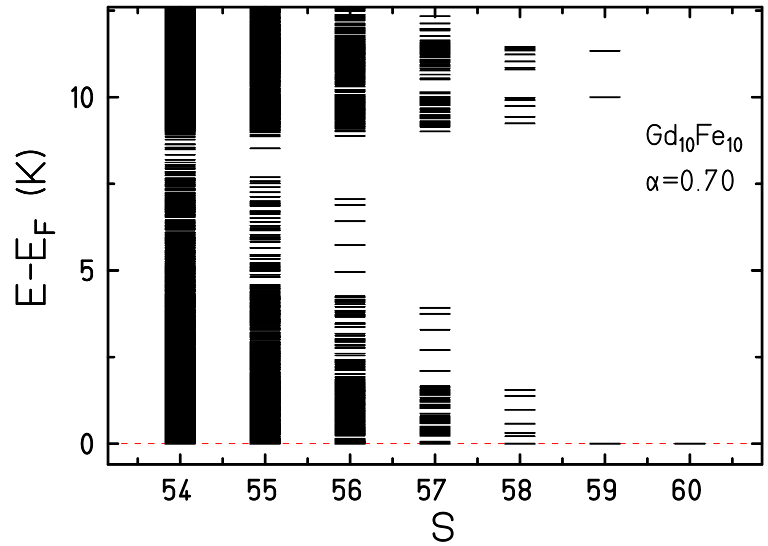
⇒ | max. number of localized magnons ⟩; $E = E_F, M = S_{\text{max}} - N/2$

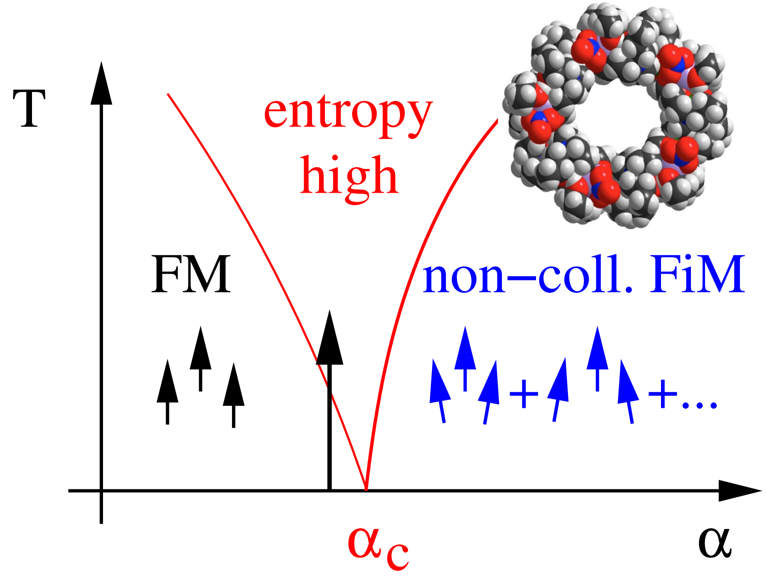
⇒ Macroscopic number of localized magnons. Degenerate with $|\, F\,\rangle$.

⇒ 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).

$Gd_{10}Fe_{10}$ – 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 $S = 54$.

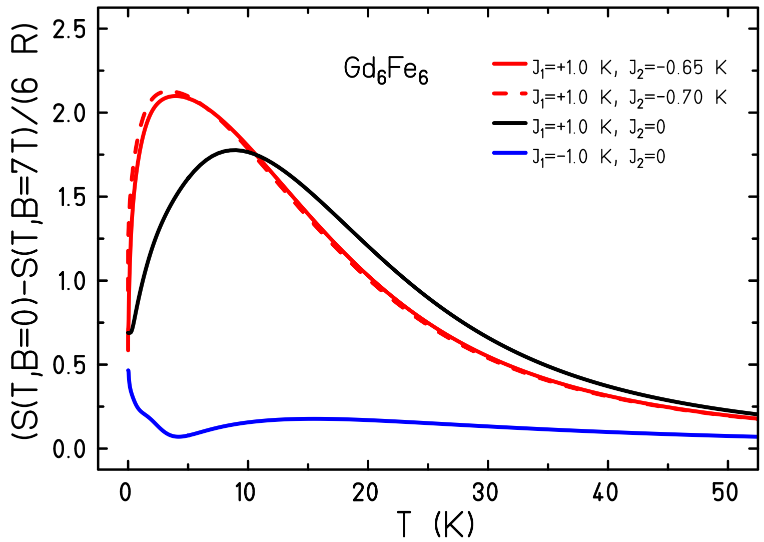
($\Delta S = N/4 + 1$ in general)

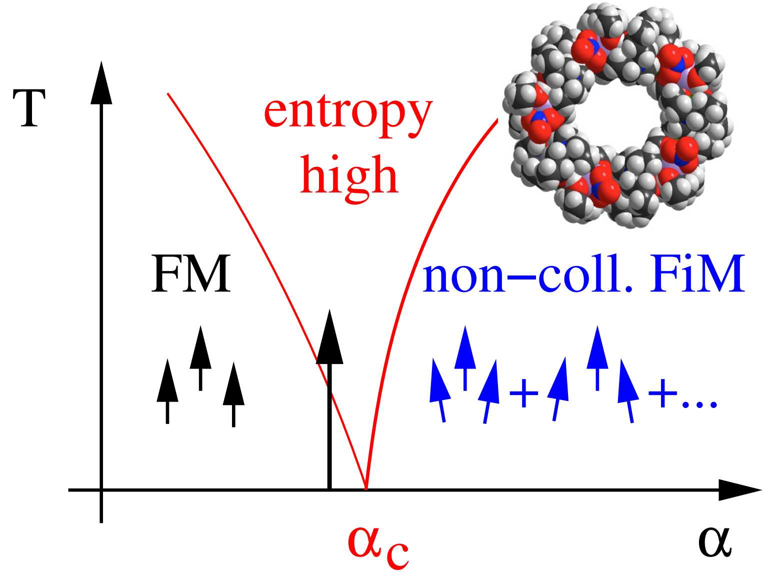
A. Baniodeh *et al.*, *npj Quantum Materials* ***3****, 10 (2018)*

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.

$Gd_{10}Fe_{10}$ – $T > 0$





⇒ although QPT and QCP at $T = 0$,

noticeable at elevated temperatures (arrow);

⇒ example isothermal entropy change:

little difference between $\alpha = 0.70$ and $\alpha = 0.65$.

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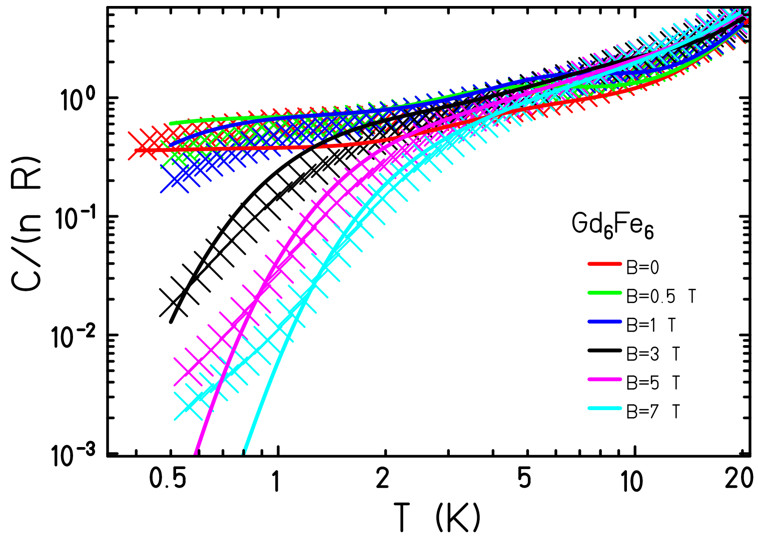
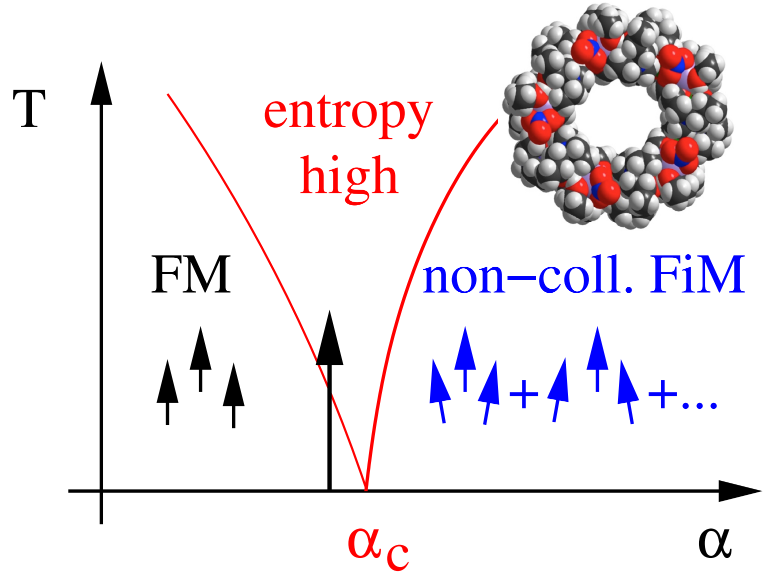
$Gd_{10}Fe_{10}$ – heat capacity





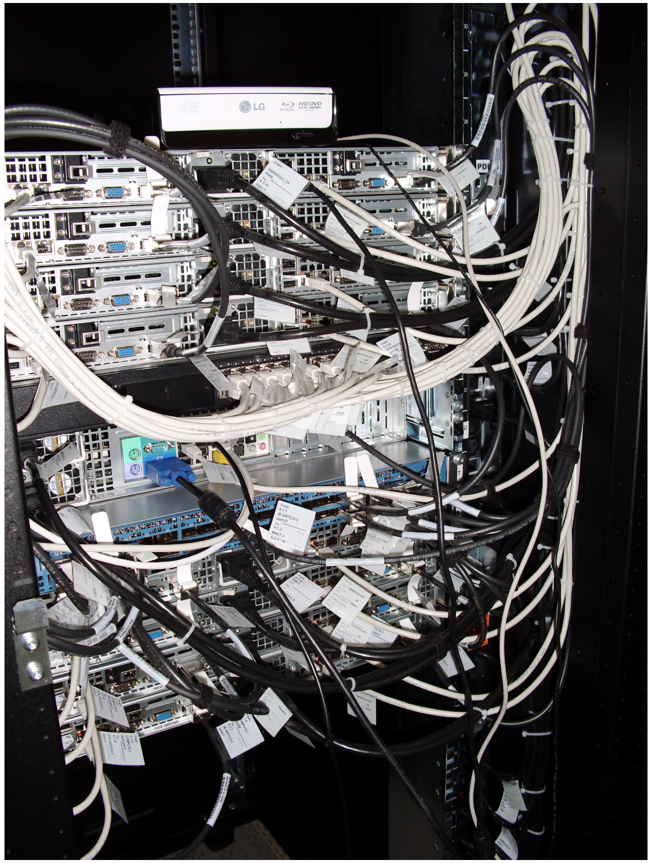
⇒ heat capacity assumes very large values even down to lowest temperatures;

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

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

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

$Gd_{10}Fe_{10}$ – Summary



- Sawtooth chain has a rich phase diagram: magnetization plateaux, magnetization jumps, flat bands, quantum phase transitions.
- $Gd_{10}Fe_{10}$ 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.

$\Leftarrow$ 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)
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- **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.

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