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

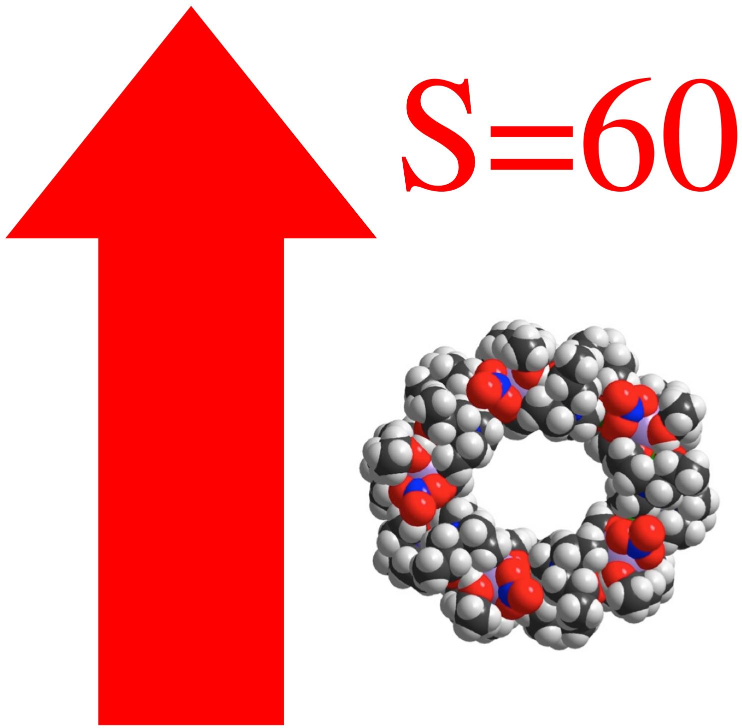
Jürgen Schnack

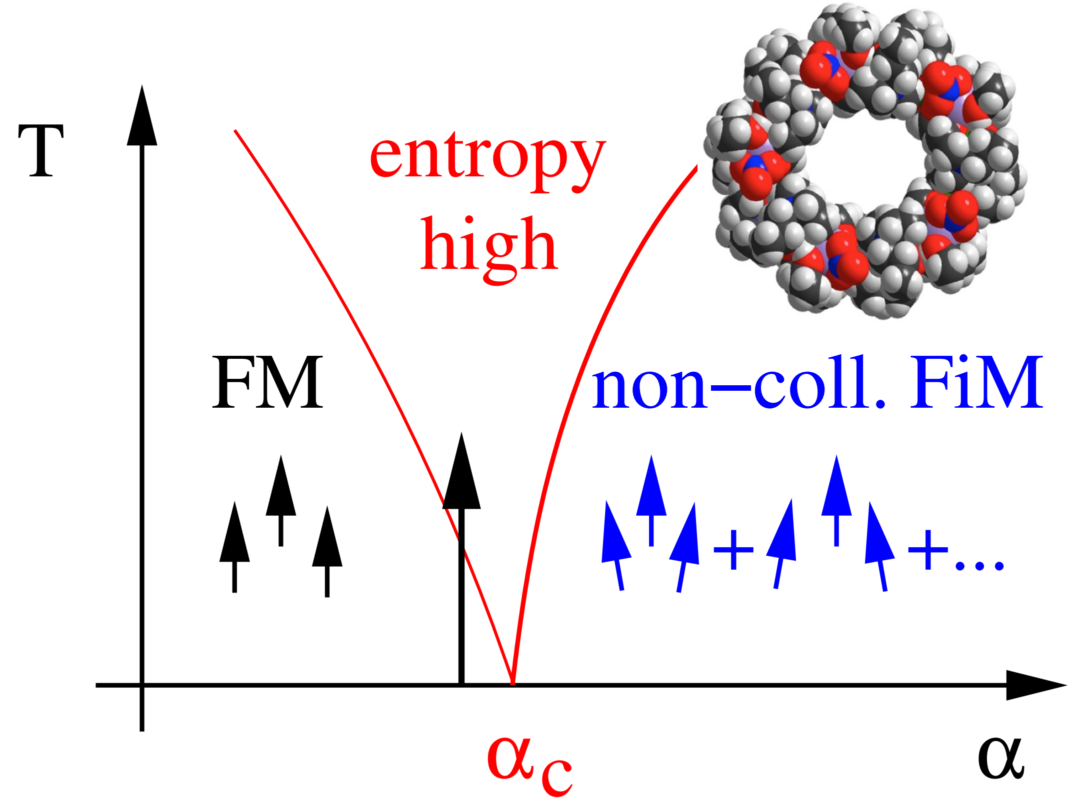
Department of Physics – University of Bielefeld – Germany

http://obelix.physik.uni-bielefeld.de/∼schnack/

4th International Conference on Bimetallic Complexes, Karlsruhe, 11 October 2018

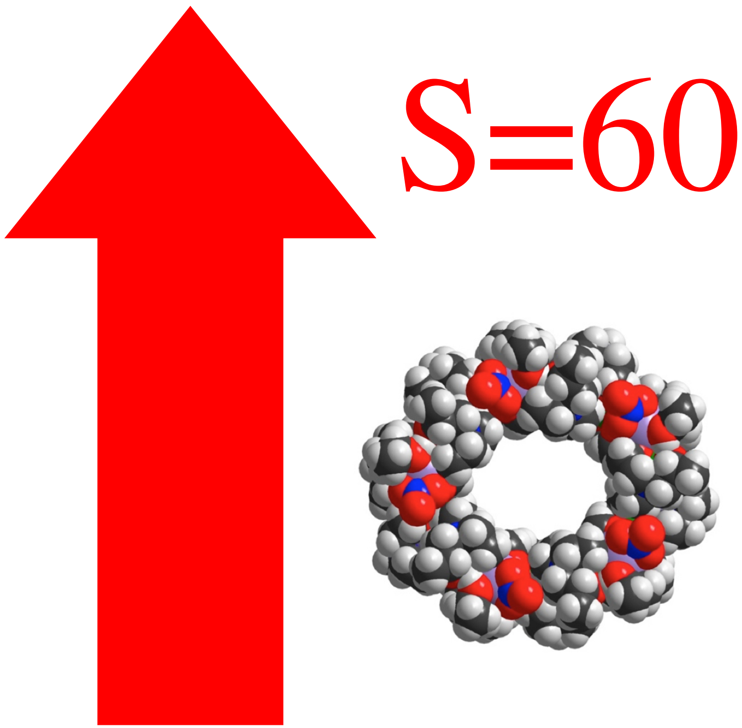
$Gd_{10}Fe_{10}$ – summary

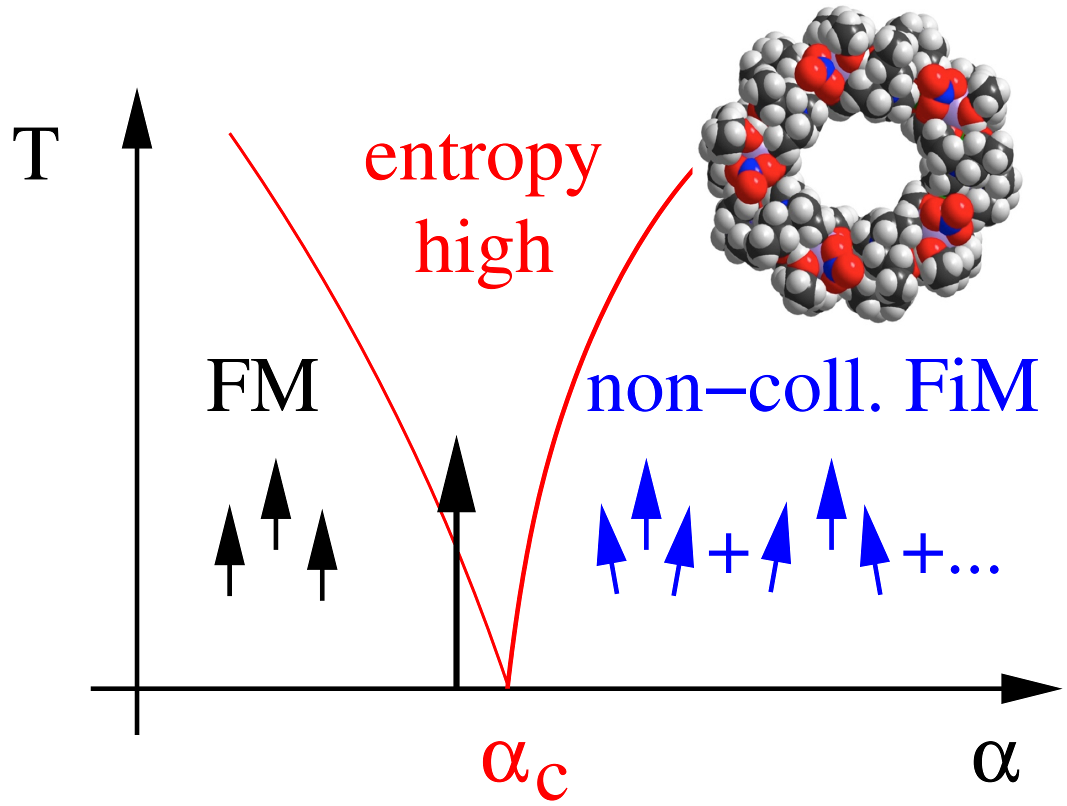




A. Baniodeh, N. Magnani, Y. Lan, G. Buth, C.E. Anson, J. Richter, M. Affronte, J. Schnack, A.K. Powell,
High Spin Cycles: Topping the Spin Record for a Single Molecule verging on Quantum Criticality,
npj Quantum Materials **3**, 10 (2018)

$Gd_{10}Fe_{10}$ – summary

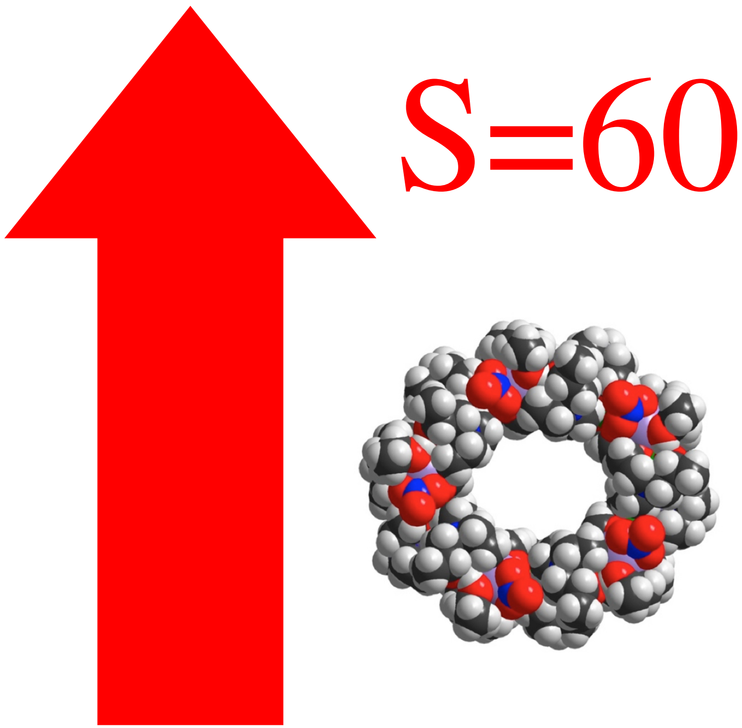


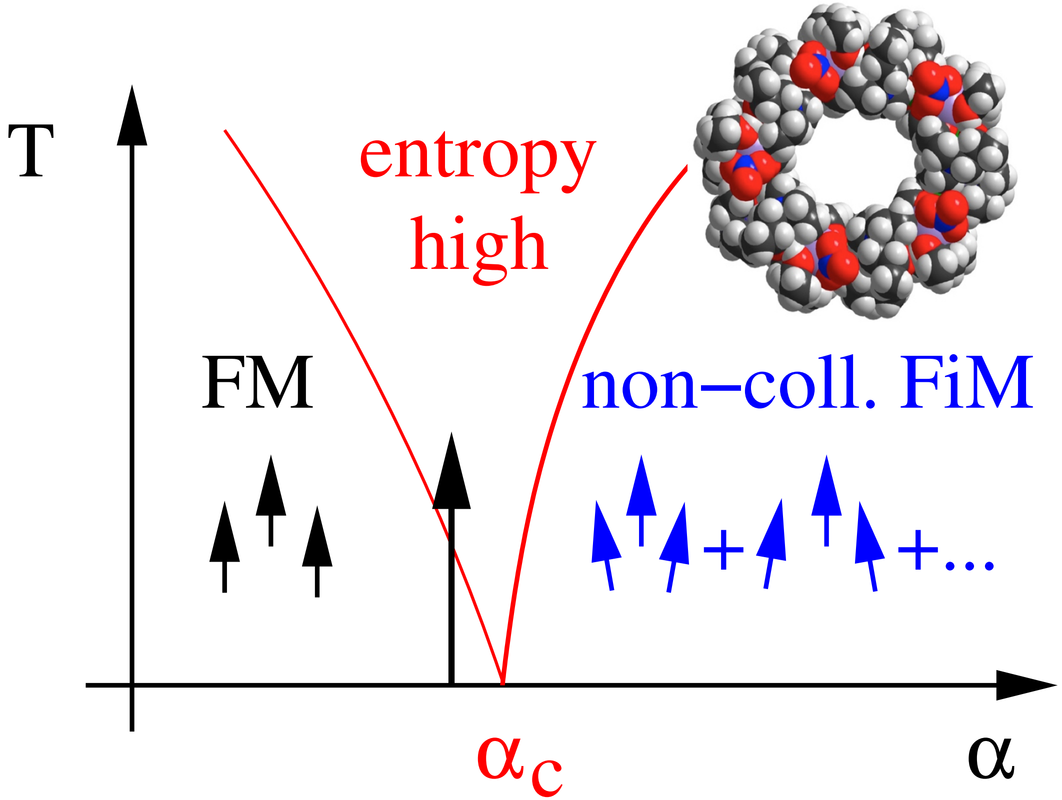


How do we know?

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

$Gd_{10}Fe_{10}$ – summary



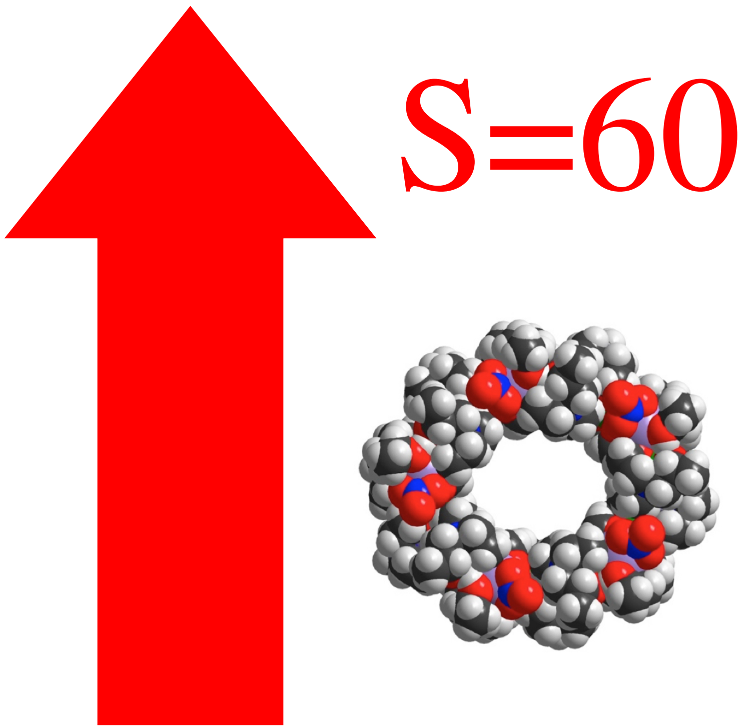


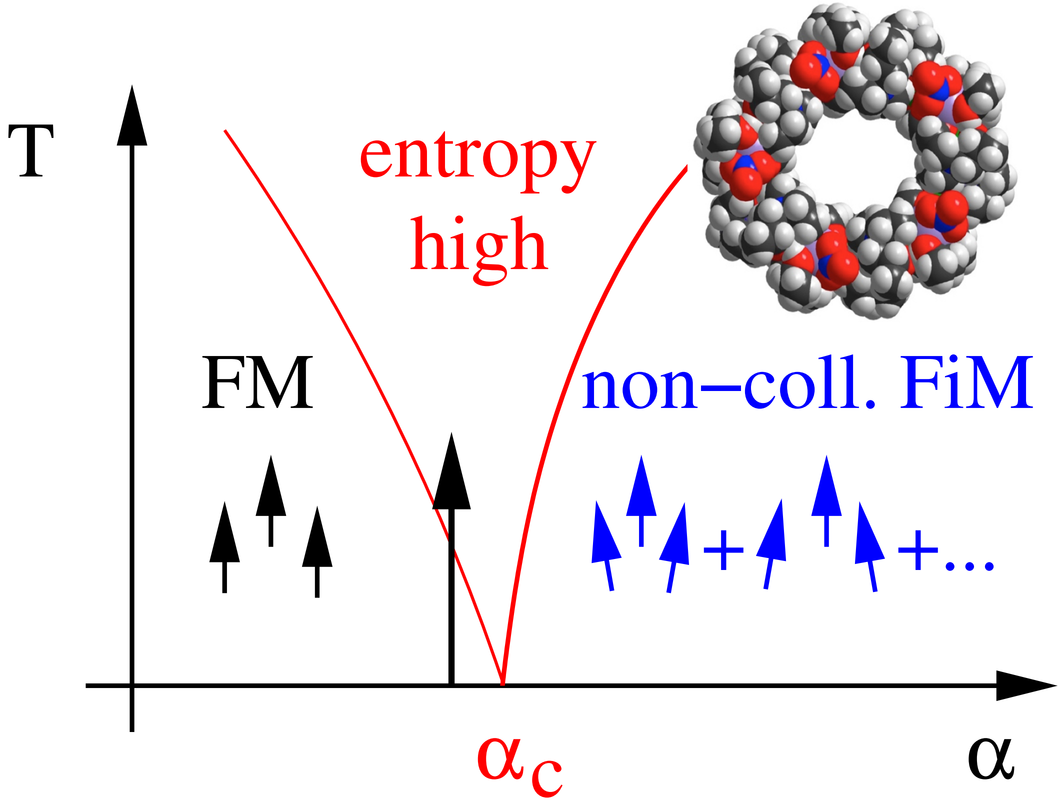
How do we know?

What is a QPT?

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

$Gd_{10}Fe_{10}$ – summary





How do we know?

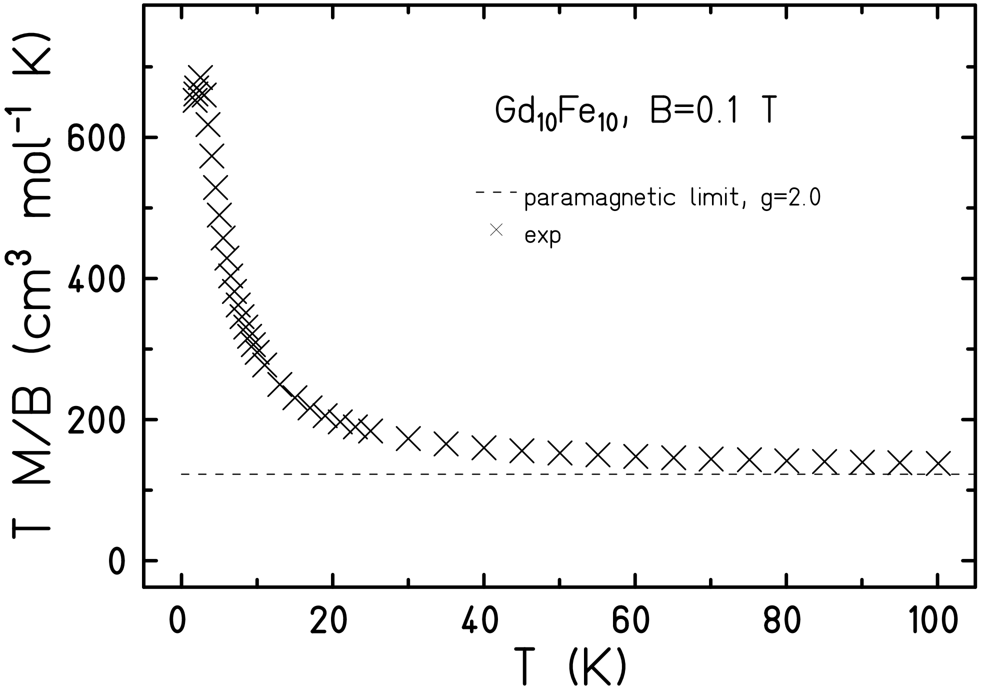
What is a QPT?

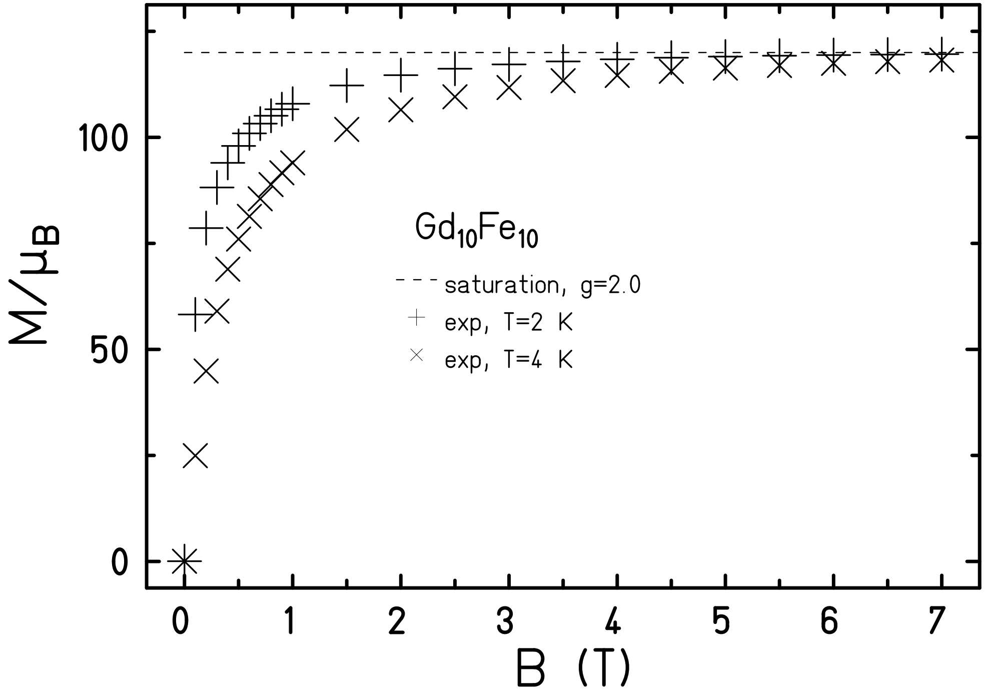
In $Gd_{10}Fe_{10}$?

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

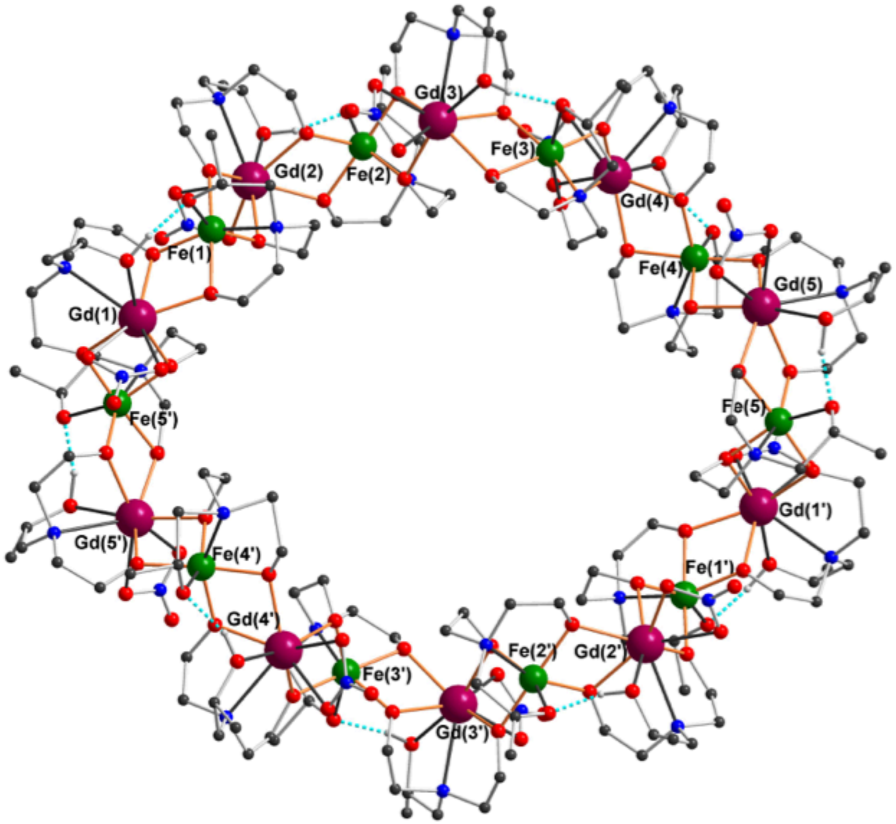
Start: experimental data

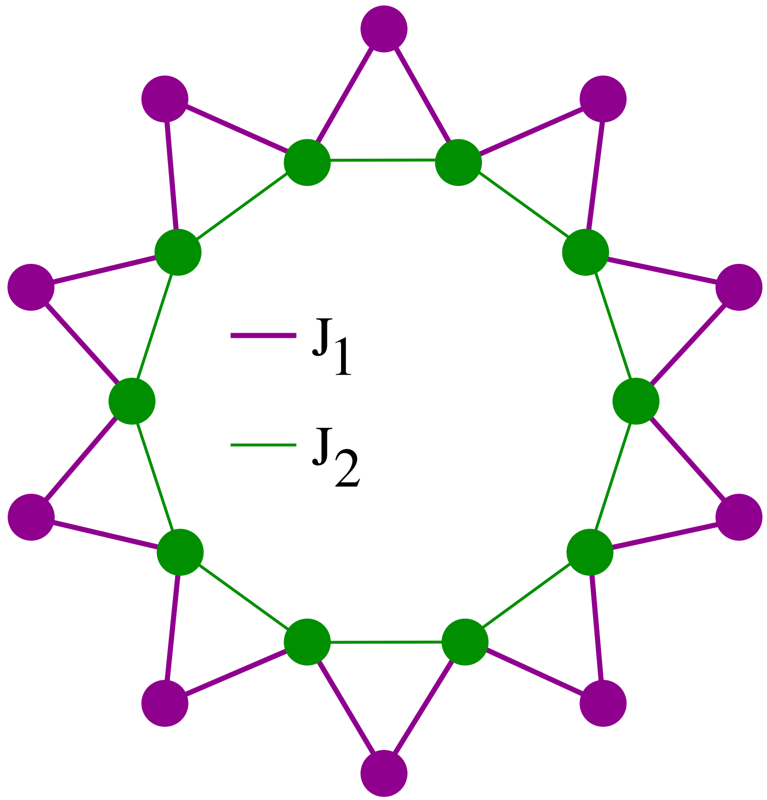
$Gd_{10}Fe_{10}$ – How to rationalize the experimental data?





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





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

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

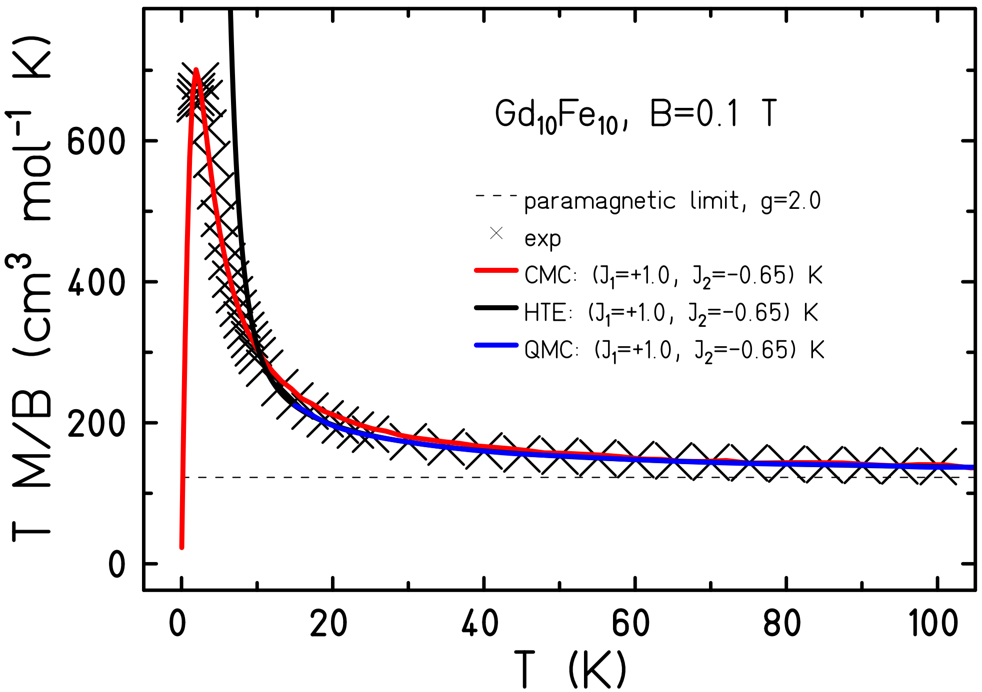
Model Hamiltonian

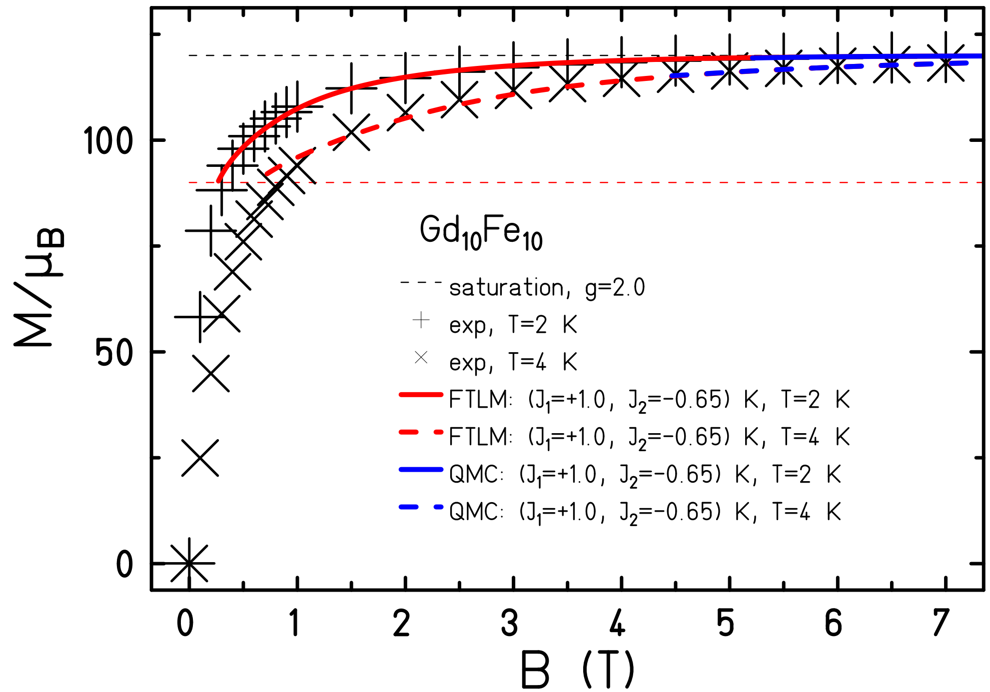
$$\underset{\sim}{H} = -2J_1 \sum_i \underset{\sim}{\vec{s}}_{\text{Gd},i} \cdot \left(\underset{\sim}{\vec{s}}_{\text{Fe},i} + \underset{\sim}{\vec{s}}_{\text{Fe},i+1} \right) -2J_2 \sum_i \underset{\sim}{\vec{s}}_{\text{Fe},i} \cdot \underset{\sim}{\vec{s}}_{\text{Fe},i+1} + g\,\mu_B\,B \sum_i \left(\underset{\sim}{s}^z_{\text{Gd},i} + \underset{\sim}{s}^z_{\text{Fe},i} \right)$$

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; Dimension of largest Hilbert space $< 10^5$.

- High-temperature series expansion: $\mathcal{O} \approx \sum_{\mu=0}^{\mu_{\text{max}}} o_\mu \, T^{-\mu}$,
  o_μ known up to $\mu_{\text{max}} = 6$ for mixed spin systems; $\mu_{\text{max}} = 11$ otherwise [1].

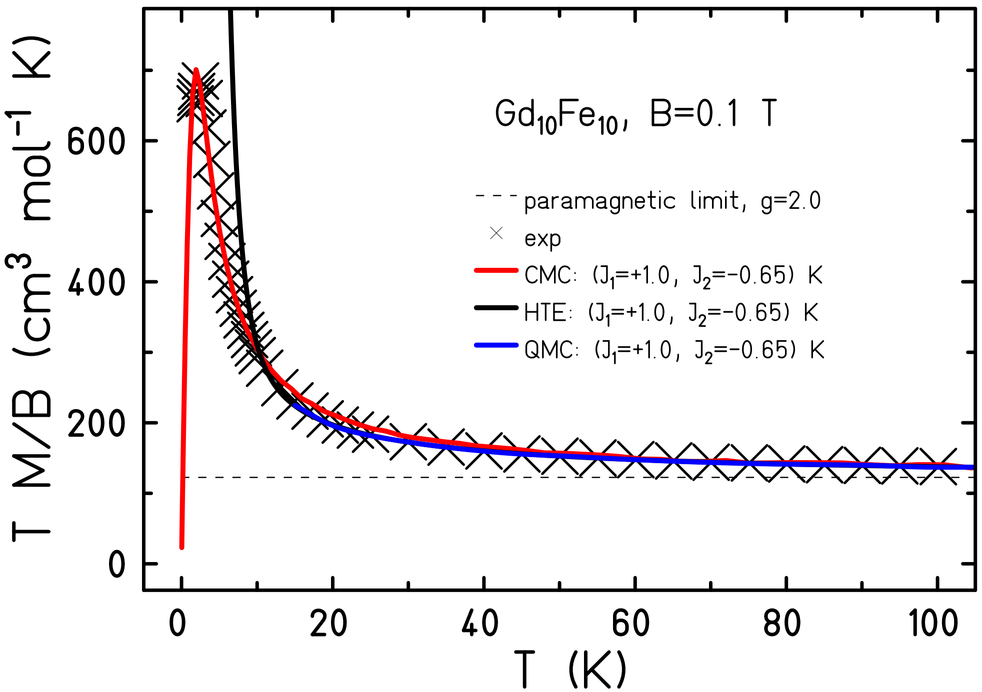
- Finite Temperature Lanczos Method (FTLM): pseudo-spectrum, low-lying levels good, approximation of partition function, time-evolution; DoH $< 10^{10}$ [2].

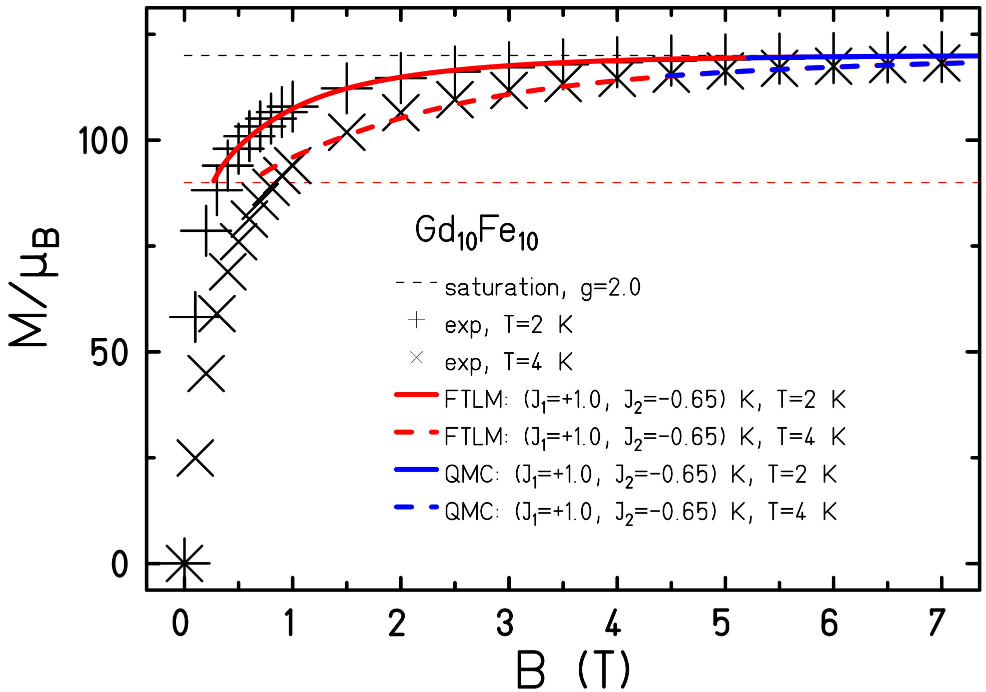
- Quantum Monte Carlo (QMC): approximation of partition function, observables; bad/no convergence for competing interactions (frustration) due to negative sign problem; otherwise HUGE systems possible [ALPS].

- 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.

$Gd_{10}Fe_{10}$ – Methods

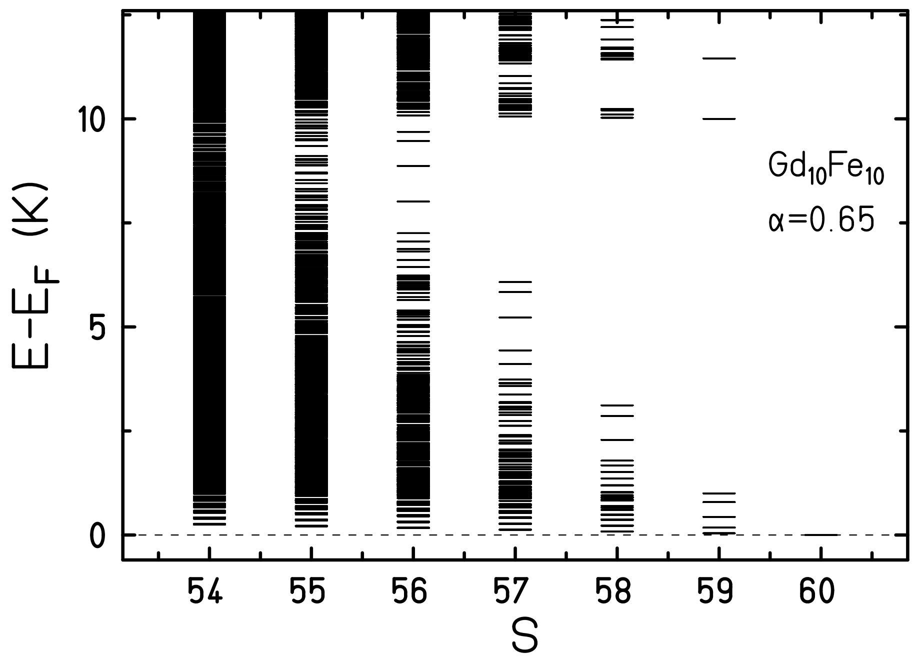


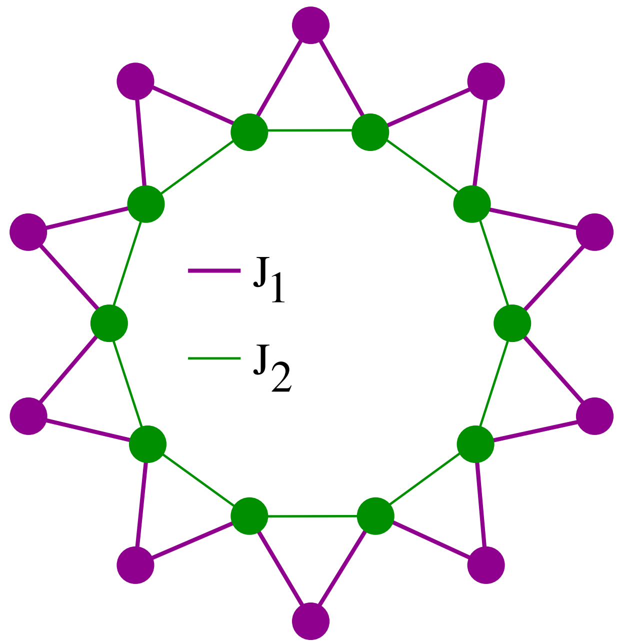


Methods: HTE, QMC, CMC, FTLM $\Rightarrow J_1 = 1.0$ K, $J_2 = -0.65$ K

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

$Gd_{10}Fe_{10}$ – $S = 60$



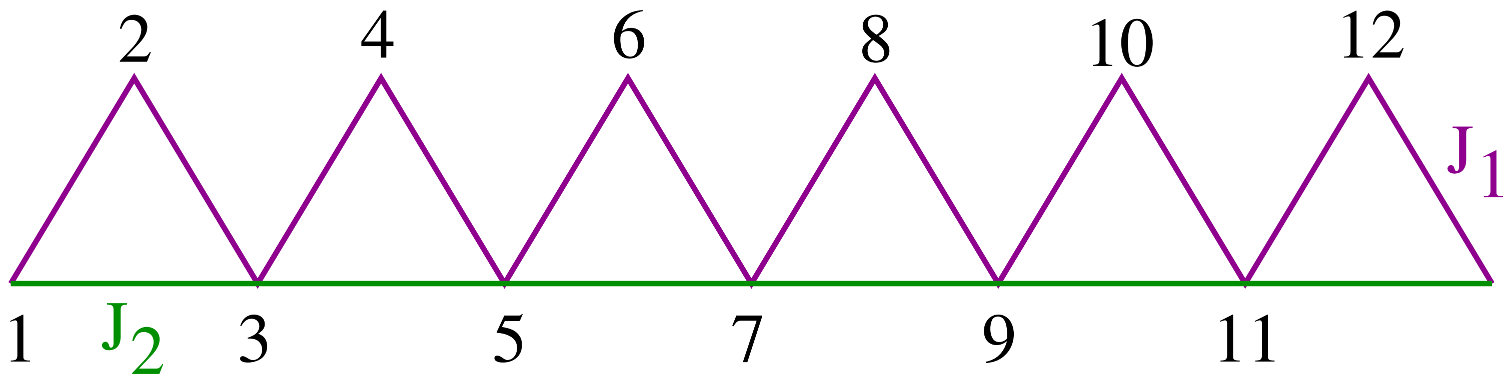


⇒ $S = 60$, largest ground state spin of a molecule to date

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

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



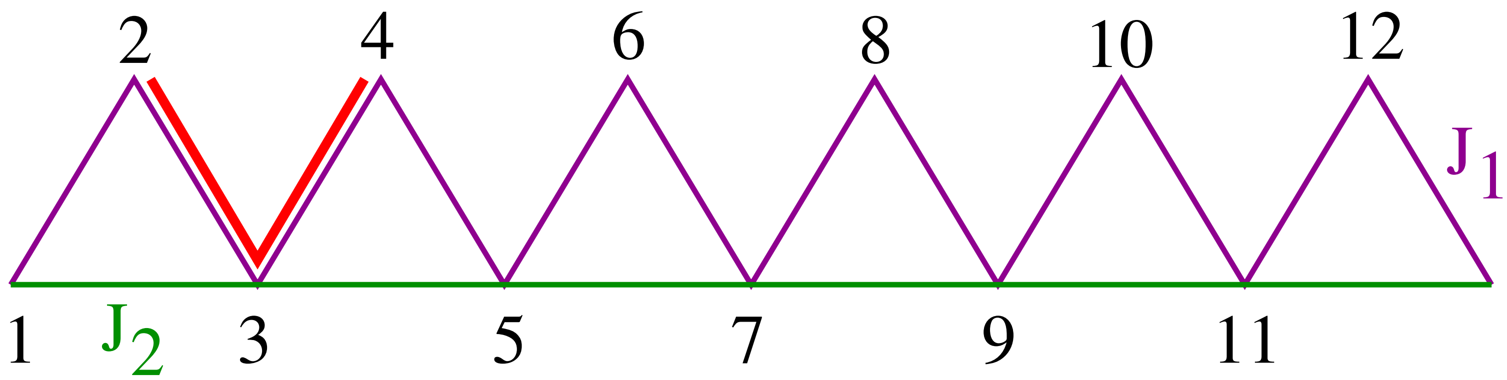
⇒ special properties for $J_1 > 0$ (ferro) and $J_2 < 0$ (af) at certain α_c

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



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

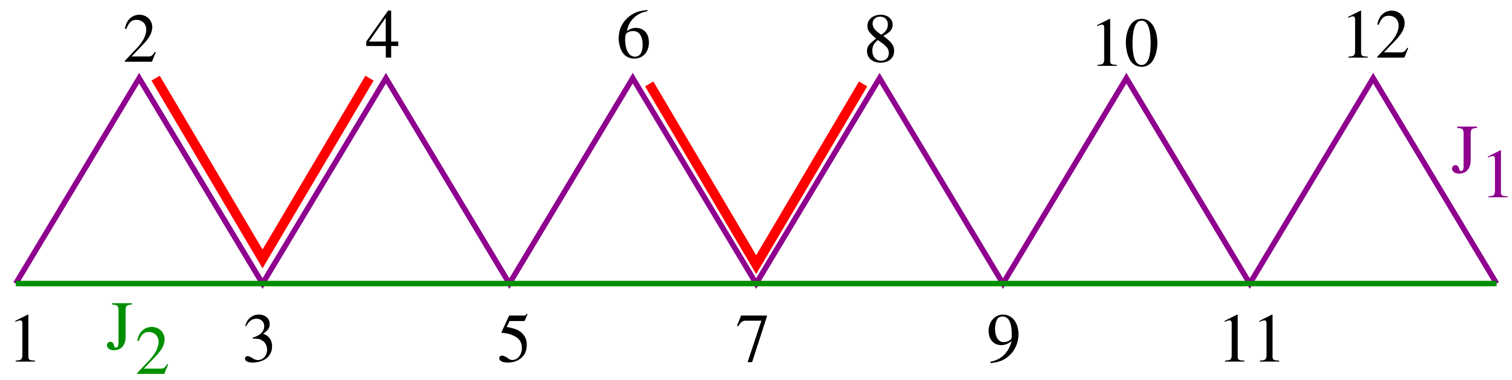
$\Rightarrow$ $\mid$ 1 localized magnon at (2,3,4) $\rangle = (\underset{\sim}{s}_2^- + \underset{\sim}{s}_4^- + 2\underset{\sim}{s}_3^-) \mid F\,\rangle$;

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

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

(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

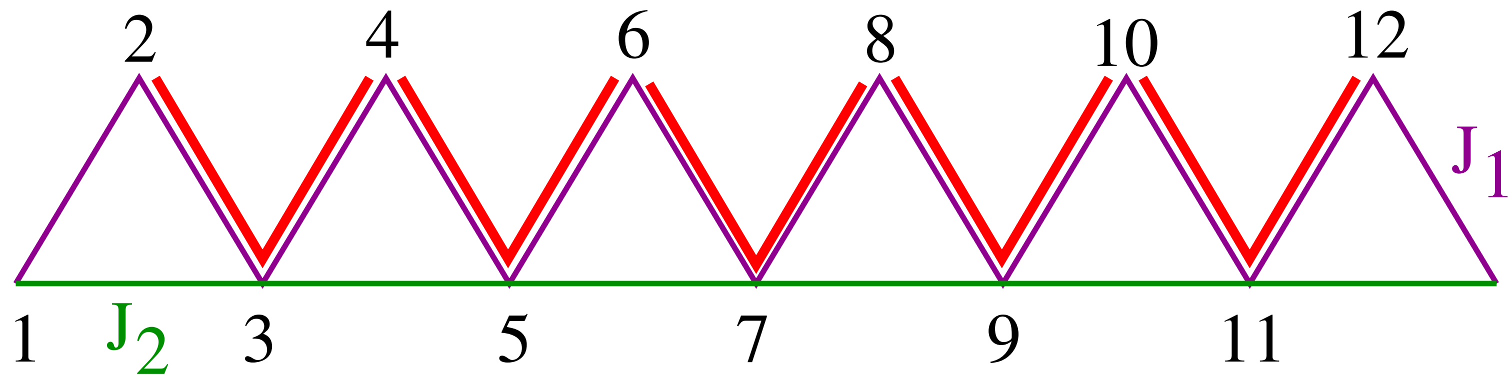


$\Rightarrow$ $|\,$2 localized magnons$\,\rangle$; $E = E_F, M = S_{\text{max}} - 2$

$\Rightarrow$ 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



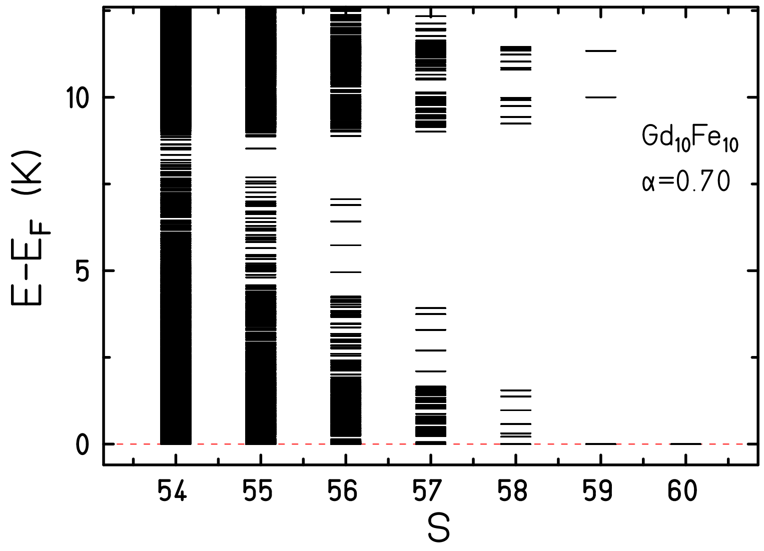
⇒ $|$ max. number of localized magnons $\rangle$; $E = E_F, M = S_{\text{max}} - N/2$

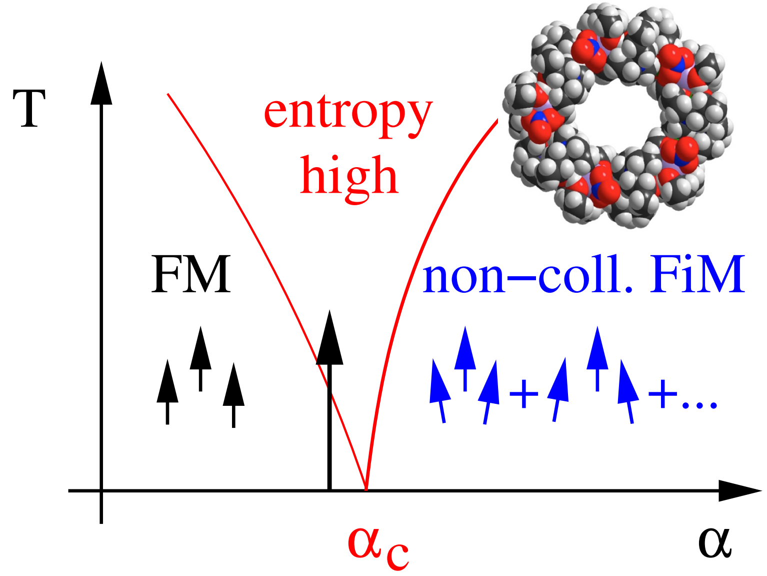
⇒ Macroscopic number of localized magnons. Degenerate with $|\, F \,\rangle$.

⇒ 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 $S = 54$.

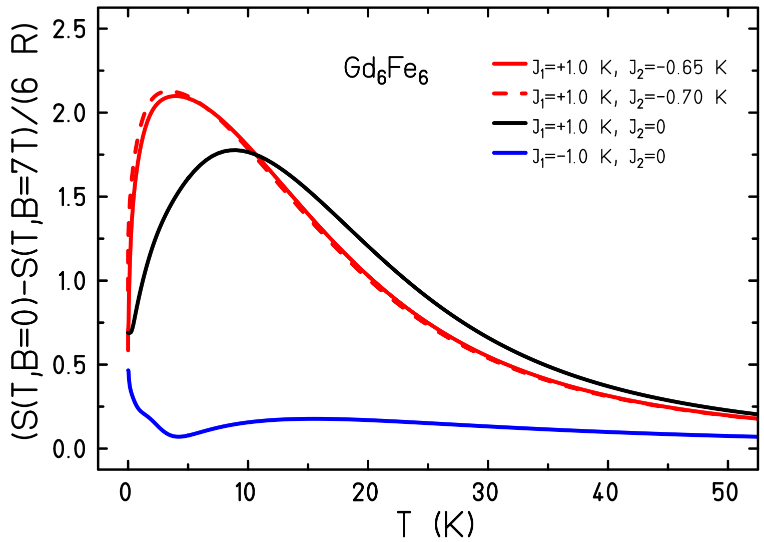
($\Delta S = N/4 + 1$ in general)

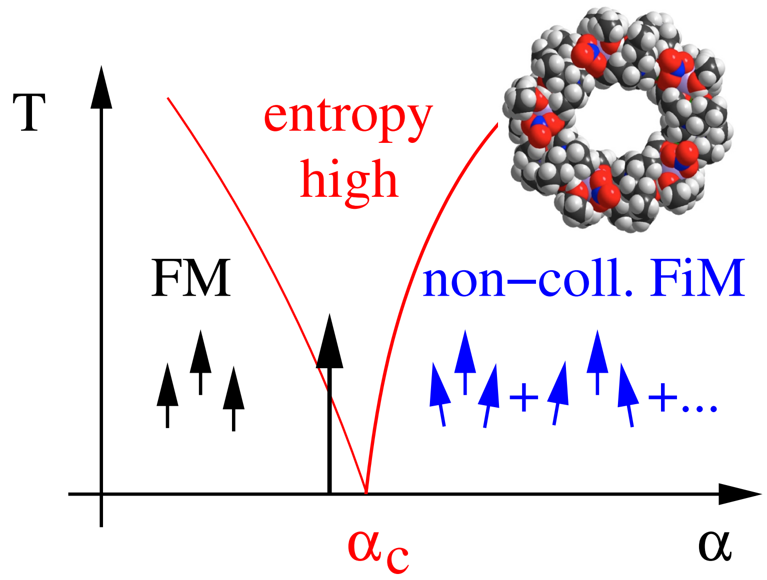
A. Baniodeh *et al.*, *npj Quantum Materials* ***3****, 10 (2018)*

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.

$Gd_{10}Fe_{10}$ – $T > 0$





$\Rightarrow$ although QPT and QCP at $T = 0$,

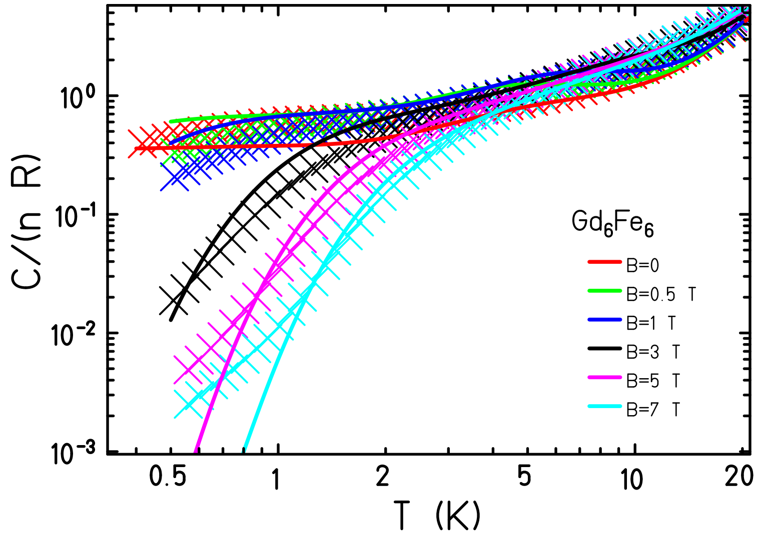
noticeable at elevated temperatures (arrow);

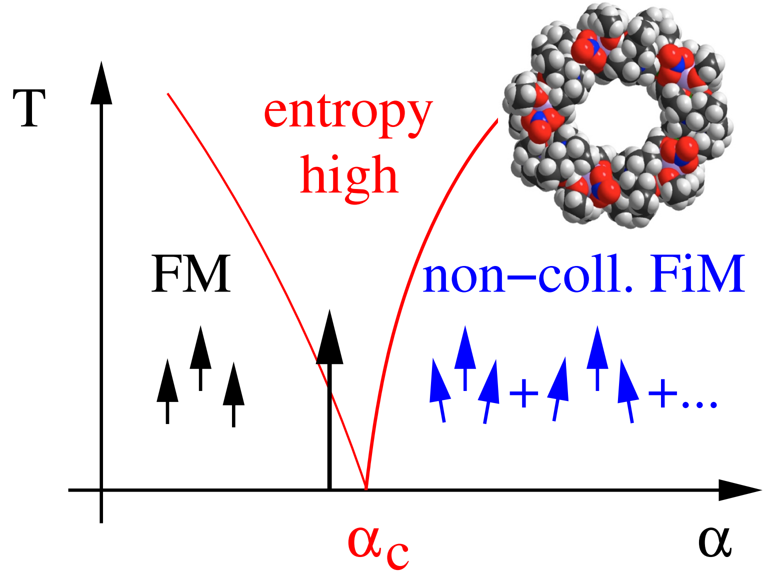
$\Rightarrow$ example isothermal entropy change:

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

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

$Gd_{10}Fe_{10}$ – heat capacity





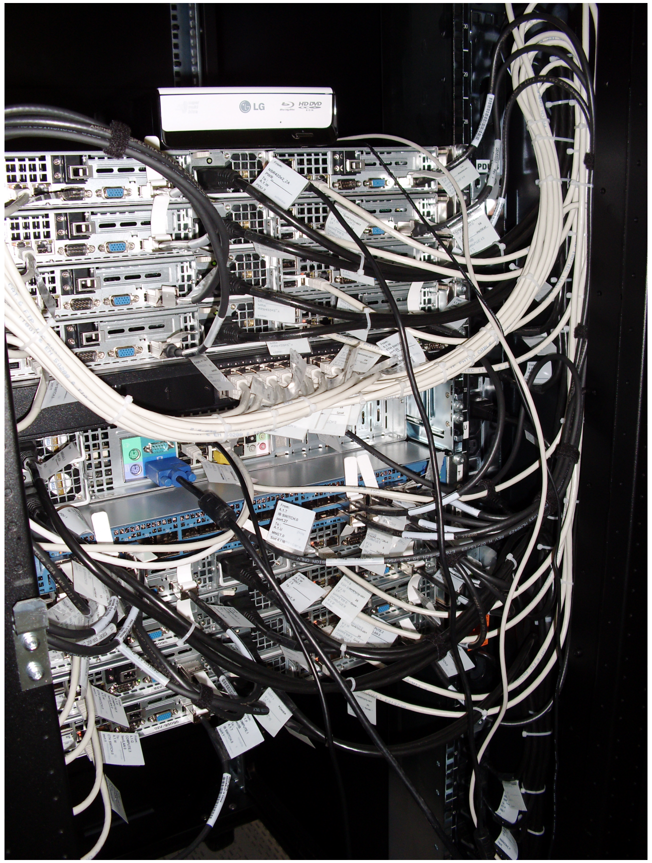
⇒ heat capacity assumes very large values even down to lowest temperatures;

⇒ 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)
- 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)

60

. . . is not the only number.

There is also . . .

42

PHYSICAL REVIEW B **98**, 094423 (2018)

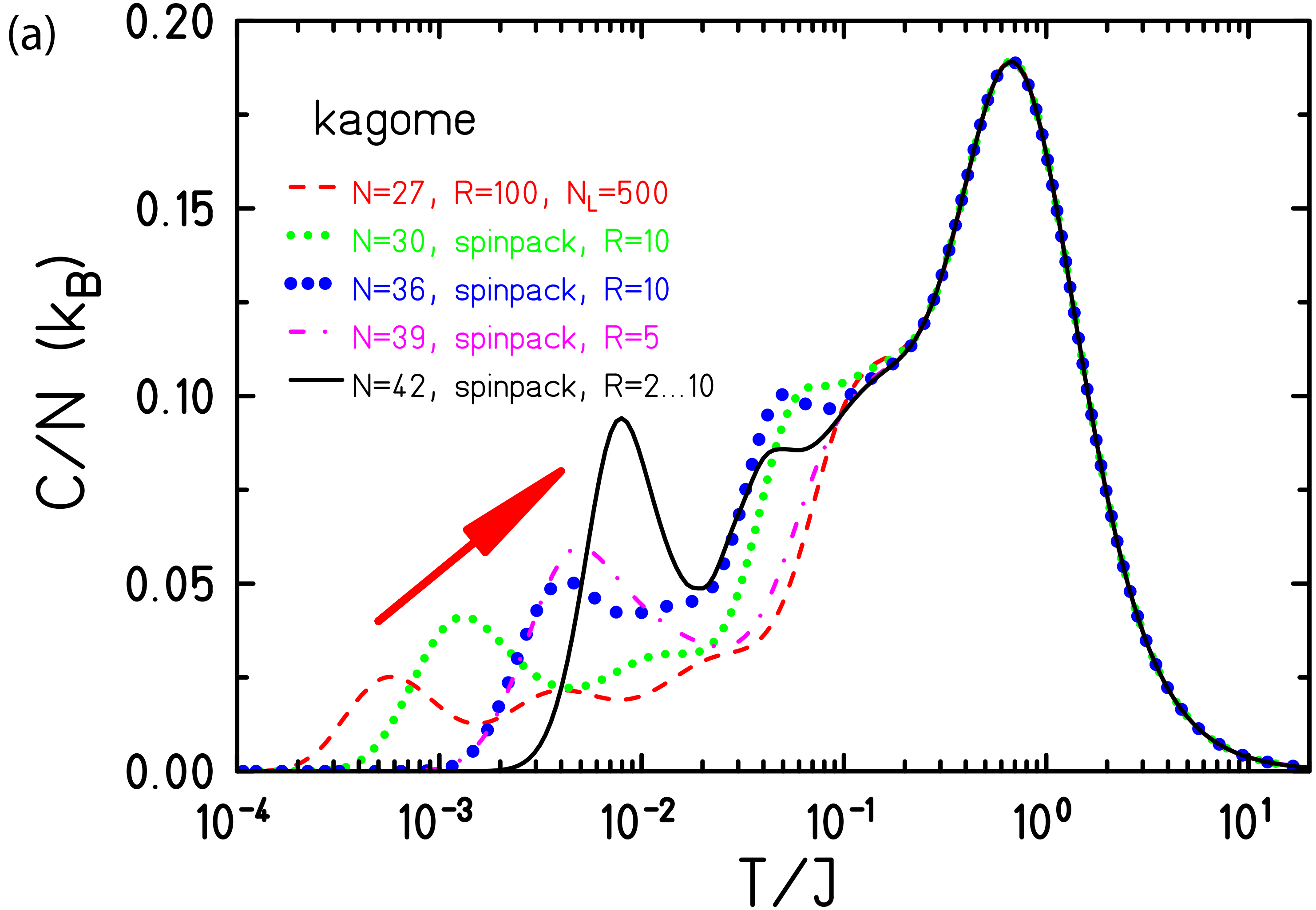
Magnetism of the $N = 42$ kagome lattice antiferromagnet

Jürgen Schnack,[1,*] Jörg Schulenburg,[2] and Johannes Richter[3,4,†]
[1]*Fakultät für Physik, Universität Bielefeld, Postfach 100131, D-33501 Bielefeld, Germany*
[2]*Universitätsrechenzentrum, Universität Magdeburg, D-39016 Magdeburg, Germany*
[3]*Institut für Physik, Universität Magdeburg, P.O. Box 4120, D-39016 Magdeburg, Germany*
[4]*Max-Planck-Institut für Physik Komplexer Systeme, Nöthnitzer Straße 38, 01187 Dresden, Germany*

(Received 14 June 2018; revised manuscript received 21 August 2018; published 25 September 2018)

For the paradigmatic frustrated spin-half Heisenberg antiferromagnet on the kagome lattice we performed large-scale numerical investigations of thermodynamic functions by means of the finite-temperature Lanczos method for system sizes of up to $N = 42$. We present the dependence of magnetization as well as specific heat on temperature and external field and show in particular that a finite-size scaling of specific heat supports the appearance of a low-temperature shoulder below the major maximum. This seems to be the result of a counterintuitive motion of the density of singlet states towards higher energies. Other interesting features that we discuss are the asymmetric melting of the $1/3$ magnetization plateau as well the field dependence of the specific heat that exhibits characteristic features caused by the existence of a flat one-magnon band. By comparison with the unfrustrated square-lattice antiferromagnet the tremendous role of frustration in a wide temperature range is illustrated.

DOI: 10.1103/PhysRevB.98.094423



(a)
kagome
N=27, R=100, N_L=500
N=30, spinpack, R=10
N=36, spinpack, R=10
N=39, spinpack, R=5
N=42, spinpack, R=2...10
C/N (k_B)
T/J
0.20
0.15
0.10
0.05
0.00
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}

42 means:
no open questions anymore.

Otherwise, please ask.

Thank you very much for your attention.

The end.

Molecular Magnetism Web

www.molmag.de

Highlights. Tutorials. Who is who. Conferences.