From chemistry to physics and back: sloppy calculations do the magic

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Annie, get your fun!

KIT, Karlsruhe, 18. 12. 2019

Magic Numbers

42 60 64 66 72

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Magic Numbers only increase.
Sorry, but that's physics:
2nd law of thermodynamics, or alike.

42 60 64 66 72

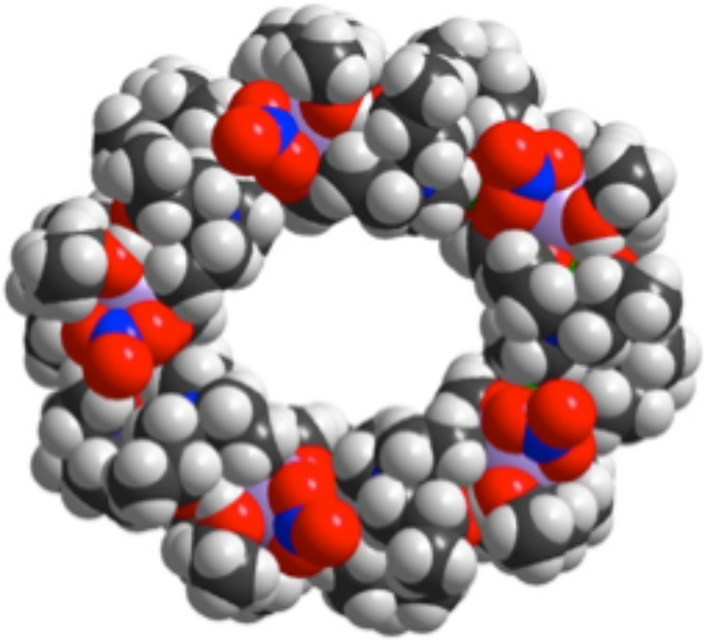
Random Numbers

The first 100 natural random numbers

0	1	2	3	4	5	6	7	8	9
10	11	12	13	14	15	16	17	18	19
20	21	22	23	24	25	26	27	28	29
30	31	32	33	34	35	36	37	38	39
40	41	42	43	44	45	46	47	48	49
50	51	52	53	54	55	56	57	58	59
60	61	62	63	64	65	66	67	68	69
70	71	72	73	74	75	76	77	78	79
80	81	82	83	84	85	86	87	88	89
90	91	92	93	94	95	96	97	98	99

How can we use Random Numbers to solve some problems that involve Magic Numbers?

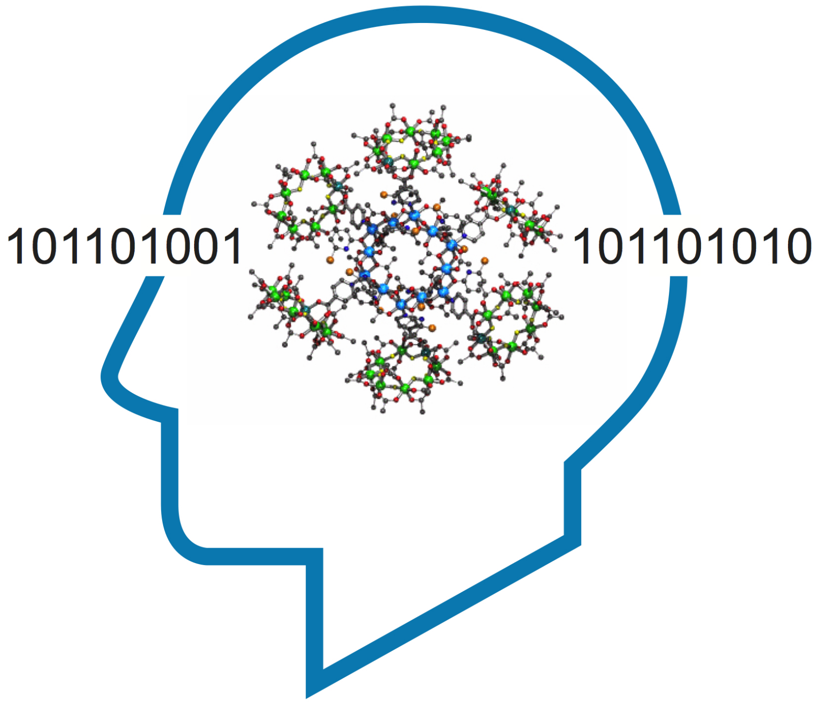
You have got a molecule!



$$S = 60!$$

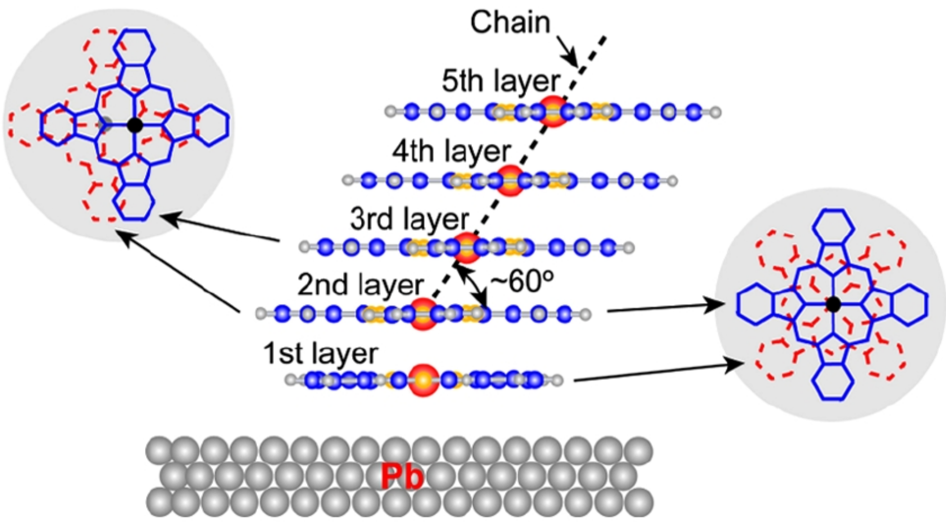
Congratulations!

You want to build a quantum computer!



Very smart!

You want to deposit your molecule!

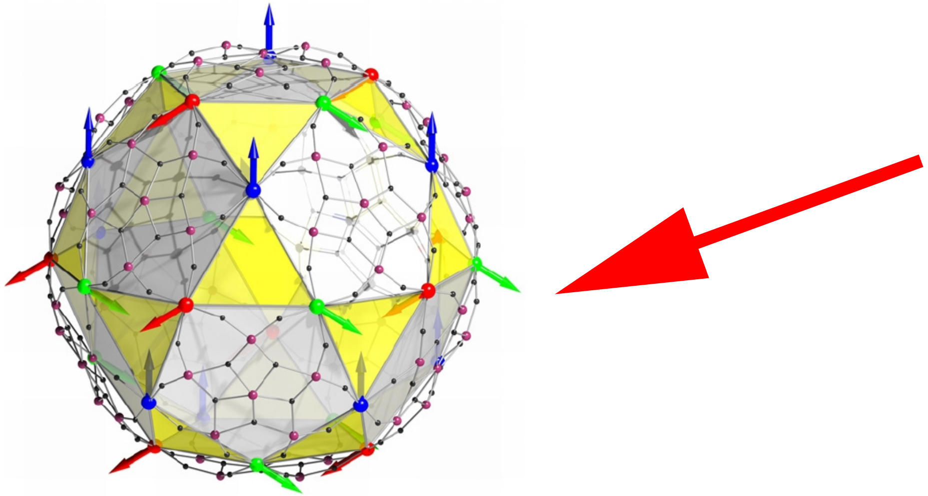


Next generation magnetic storage!

You have got an idea about the modeling!

$$\underset{\sim}{H} = -2\sum_{i<j} J_{ij}\, \underset{\sim}{\vec{s}}(i) \cdot \underset{\sim}{\vec{s}}(j) \quad + \quad g\,\mu_B\, B \sum_{i}^{N} \underset{\sim}{s}_z(i)$$

Heisenberg　　　　Zeeman



60 edges!

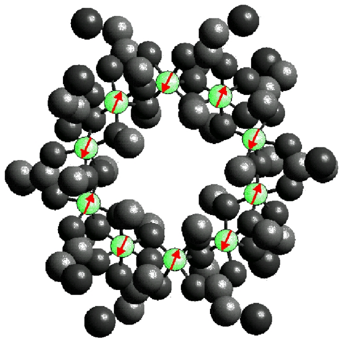
You have to solve the Schrödinger equation!

$$\underset{\sim}{H} \mid \phi_n \,\rangle = E_n \mid \phi_n \,\rangle$$

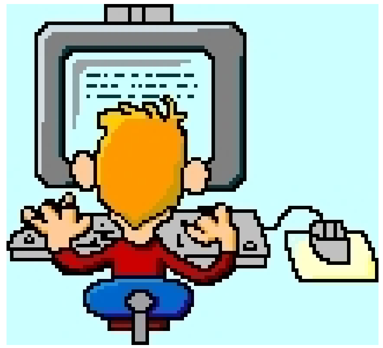
Eigenvalues E_n and eigenvectors $\mid \phi_n \,\rangle$

- needed for spectroscopy (EPR, INS, NMR);
- needed for thermodynamic functions (magnetization, susceptibility, heat capacity);
- needed for time evolution (pulsed EPR, simulate quantum computing, thermalization).

In the end it's always a big matrix!



$$\Rightarrow \begin{pmatrix} -27.8 & 3.46 & 0.18 & \cdots \\ 3.46 & -2.35 & -1.7 & \cdots \\ 0.18 & -1.7 & 5.64 & \cdots \\ \vdots & \vdots & \vdots & \cdots \end{pmatrix} \Rightarrow$$



$\mathrm{Fe}^{\mathrm{III}}_{10}$: $N = 10, s = 5/2$

Dimension=**60**,466,176. Maybe too big?

Can we evaluate the partition function

$$Z(T,B) \;=\; \text{tr}\left(\exp\left[-\beta \underset{\sim}{H}\right]\right)$$

without diagonalizing the Hamiltonian?

Yes, we can,

with magic built on randomness!

Solution I: trace estimators

$$\text{tr}\left(\underset{\sim}{Q}\right) \approx \langle\, r \,|\, \underset{\sim}{Q} \,|\, r \,\rangle$$

$$|\, r \,\rangle = \sum_{\nu} r_\nu \,|\, \nu \,\rangle \;, \quad r_\nu = \pm 1$$

- $|\, \nu \,\rangle$ some orthonormal basis of your choice;
  not the eigenbasis of $\underset{\sim}{Q}$, since we don't know it.
- $r_\nu = \pm 1$ random, equally distributed. Rademacher vectors.
- Amazingly accurate, bigger (Hilbert space dimension) is better.

M. Hutchinson, Communications in Statistics - Simulation and Computation **18**, 1059 (1989).

Solution II: Krylov space representation

$$\exp\left[-\beta \underset{\sim}{H}\right] \;\approx\; \underset{\sim}{\mathbb{1}} - \beta \underset{\sim}{H} + \frac{\beta^2}{2!}\underset{\sim}{H}^2 - \cdots \frac{\beta^{N_L-1}}{(N_L-1)!}\underset{\sim}{H}^{N_L-1}$$

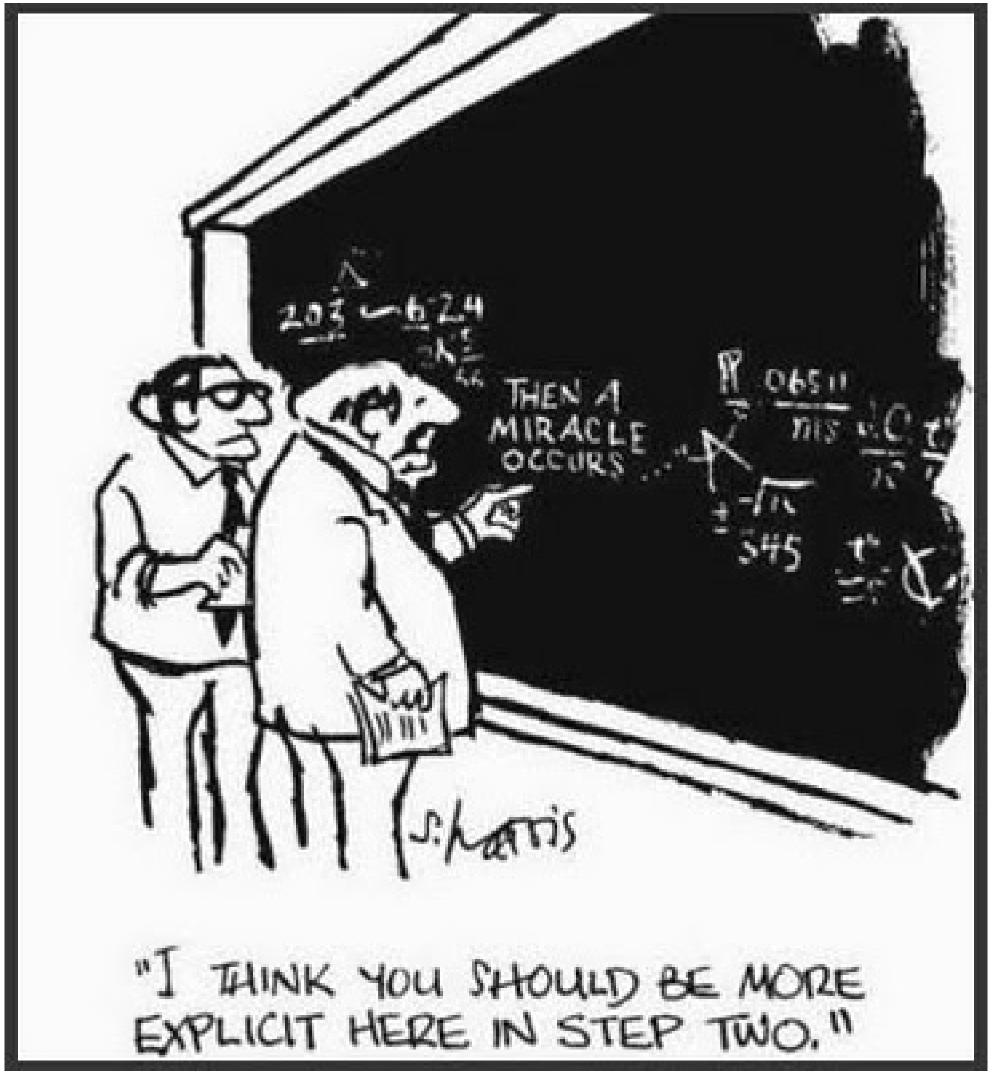
applied to a state $|\,r\,\rangle$ yields a superposition of

$$\underset{\sim}{\mathbb{1}}\,|\,r\,\rangle, \underset{\sim}{H}\,|\,r\,\rangle, \underset{\sim}{H}^2\,|\,r\,\rangle, \ldots \underset{\sim}{H}^{N_L-1}\,|\,r\,\rangle\ .$$

These (linearly independent) vectors span a small space of dimension N_L;
it is called Krylov space.

Let's diagonalize $\underset{\sim}{H}$ in this space!

Solution III: put everything together, then a miracle occurs



Partition function I: simple approximation

$$Z(T,B) \approx \langle\, r \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle \approx \sum_{n=1}^{N_L} e^{-\beta \epsilon_n^{(r)}} |\langle\, n(r) \,|\, r \,\rangle|^2$$

$$O^{\mathrm{r}}(T,B) \approx \frac{\langle\, r \,|\, \underset{\sim}{O} e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle}{\langle\, r \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle}$$

- Wow!!!
- One can replace a trace involving an intractable operator by an expectation value with respect to just ONE random vector evaluated by means of a Krylov space representation???

J. Jaklic and P. Prelovsek, Phys. Rev. B **49**, 5065 (1994).

Well, yes,

Well, yes,

but 60 is better!

Partition function II: Finite-temperature Lanczos Method

$$Z^{\text{FTLM}}(T,B) \approx \frac{1}{R}\sum_{r=1}^{R=\mathbf{60}}\sum_{n=1}^{N_L} e^{-\beta\epsilon_n^{(r)}} |\langle\, n(r) \,|\, r \,\rangle|^2$$

- Averaging over random vectors is better.
- $|\, n(r) \,\rangle$ n-th Lanczos eigenvector starting from $|\, r \,\rangle$ (Rademacher vectors).
- Partition function replaced by a small sum: $R = 1 \dots 60, N_L \approx 100$.

J. Jaklic and P. Prelovsek, Phys. Rev. B **49**, 5065 (1994).

Partition function III: use symmetries if you can

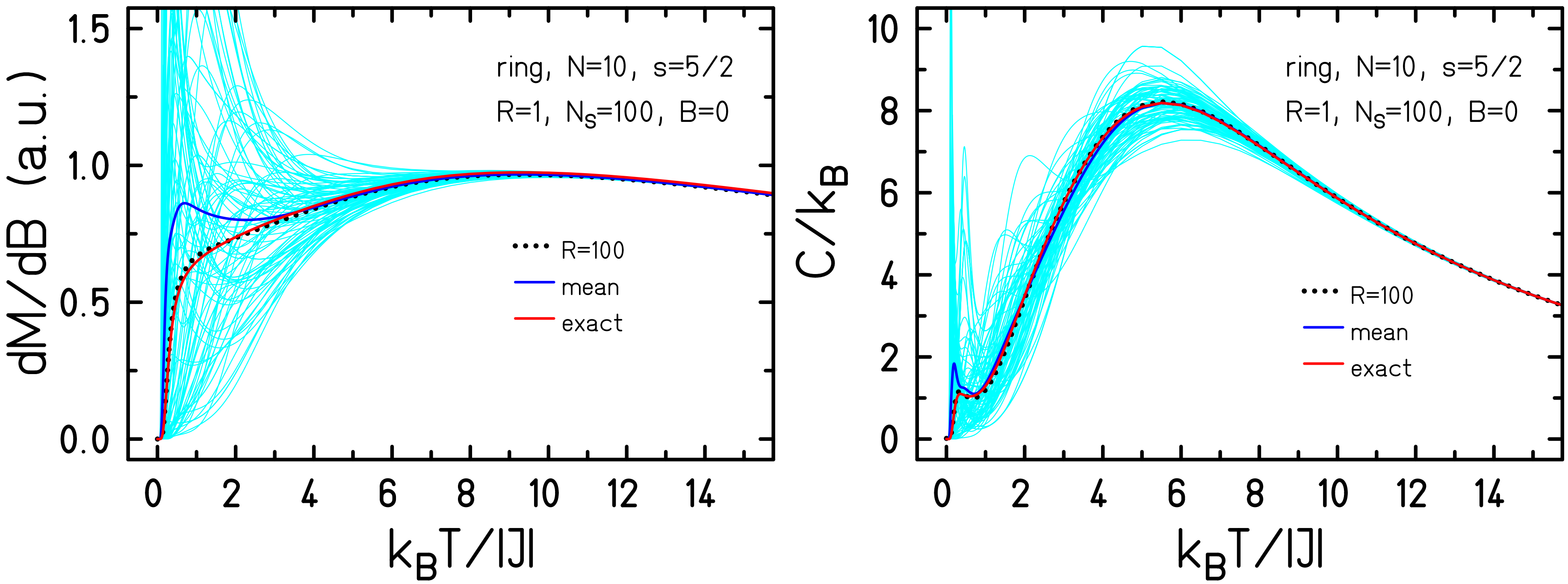
$$Z^{\text{FTLM}}(T,B) \approx \sum_{\gamma=1}^{\Gamma} \frac{1}{R} \sum_{r=1}^{R} \sum_{n=1}^{N_L} e^{-\beta\epsilon_n^{(r)}} |\langle\, n(r)\,|\,r\,\rangle|^2$$

- γ labels all the symmetries you use in your calculation (all irreducible representations).
- $|\,r\,\rangle$ is then drawn from the respective subspace with this symmetry.

J. Schnack and O. Wendland, Eur. Phys. J. B **78** (2010) 535-541

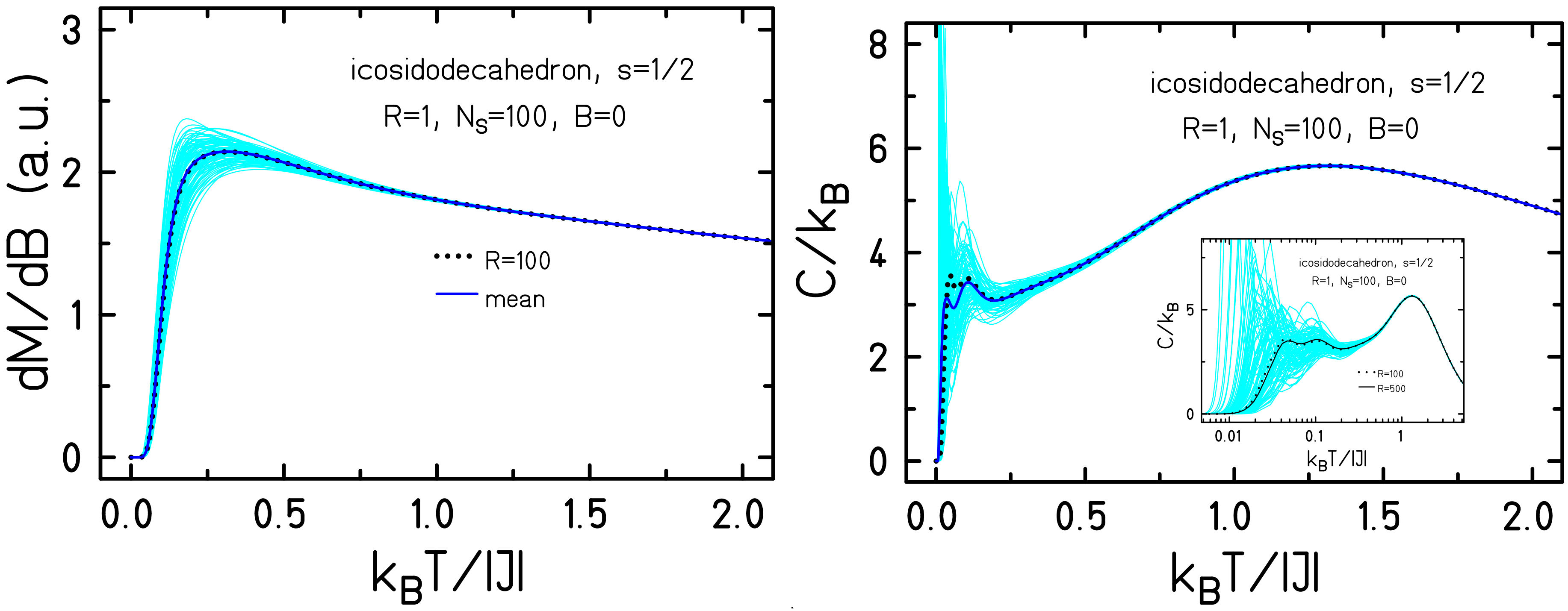
How good is it?

FTLM 1: ferric wheel



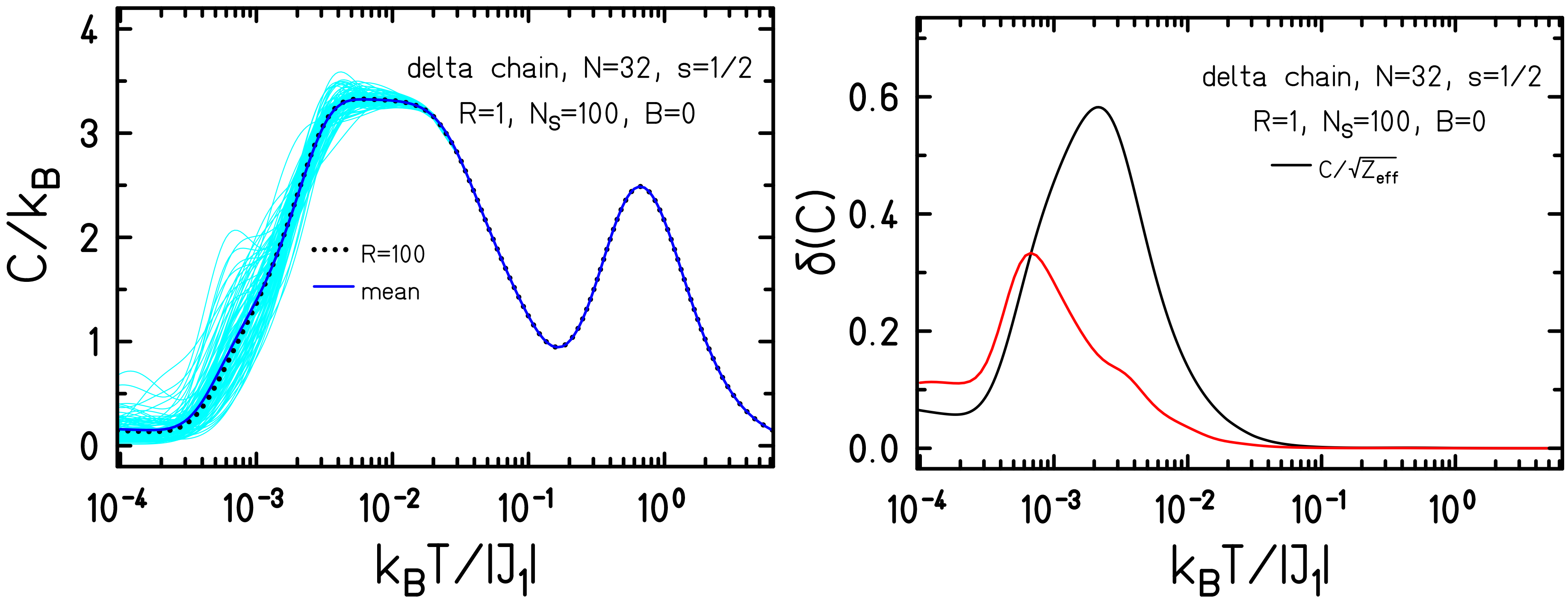
J. Schnack, J. Richter, R. Steinigeweg, arXiv:1911.08838

FTLM 2: icosidodecahedron



J. Schnack, J. Richter, R. Steinigeweg, arXiv:1911.08838

FTLM 3: sawtooth chain



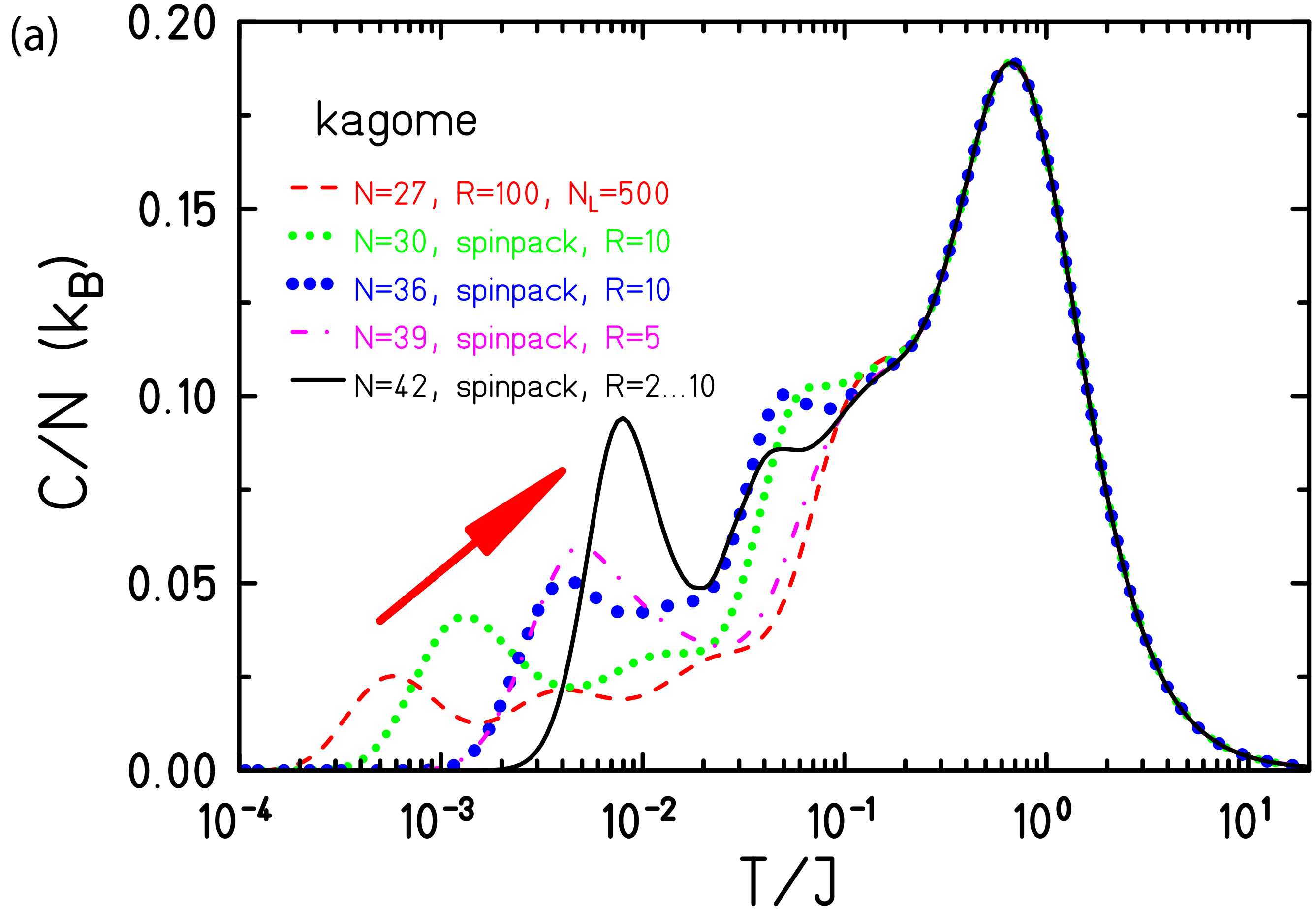
This is how one calculates Fe_6Gd_6 as approximation for $Fe_{10}Gd_{10}$.

Frustration, technically speaking, works in your favour.

J. Schnack, J. Richter, R. Steinigeweg, arXiv:1911.08838

What about the other magic numbers?

Magic 42



J. Schnack, J. Schulenburg, J. Richter, Magnetism of the $N = 42$ kagome lattice antiferromagnet, Phys. Rev. B **98**, 094423 (2018)

Magic 60


Prof. Annie Powell's Research Group
Team
Research
Publications
Teaching
Contact
Login
60 group members!

Magic 64

When I get older losing my hair
Many years from now
Will you still be sending me a Valentine
Birthday greetings bottle of wine

Will you still need me (for diagonalizing hamiltonians),
will you still feed me (with interesting problems)
When I'm sixty-four

Send me a postcard, drop me a line
Stating point of view
Indicate precisely what you mean to say
Yours sincerely, wasting away . . .

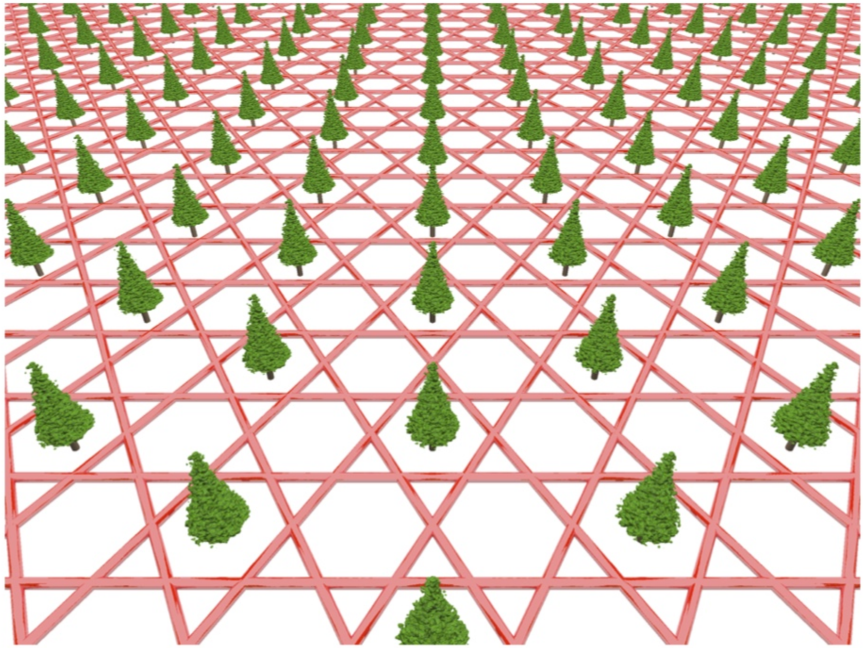
Magic 66

Ihr werdet euch noch wundern, wenn ich erst Rentner bin
Sobald der Stress vorbei ist, dann lang ich nämlich hin, oh ho, oh ho, oh ho
Dann back ich äußerst lässig, ein Supermolekül,
Das hat nen Spin von 70, das ist doch gar nicht viel, oh ho, oh ho, oh ho

Mit sechsundsechzig Jahren, da fängt das Leben an
Mit sechsundsechzig Jahren, da hat man Spaß daran
Mit sechsundsechzig Jahren, da kommt man erst in Schuss
Mit sechsundsechzig ist noch lange nicht Schluss

Magic 72

Merry Christmas
and a
Happy New Year



Magnon crystallization in the kagome lattice antiferromagnet, arXiv:1910.10448

Thank you very much for your attention.

The end.

Molecular Magnetism Web

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