Exact quantum spin dynamics and what it teaches us

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UNIVERSITÄT BIELEFELD
Fakultät für Physik

Simplified remarks on typicality

Typicality bill of rights – FOR 2692

"All" non-equilibrium states relax equal

- C. Bartsch, J. Gemmer, Dynamical typicality of quantum expectation values, Phys. Rev. Lett., 102, 110403 (2009)
- P. Reimann, J. Gemmer, Why are macroscopic experiments reproducible? Imitating the behavior of an ensemble by single pure states, Physica A 552, 121840 (2020)
- T. Heitmann, J. Richter, D. Schubert, R. Steinigeweg, Selected applications of typicality to real-time dynamics of quantum many-body systems, Zeitschrift für Naturforschung A 75, 421 (2020)

"All" similar Hamiltonians produce equal time-evolution

- P. Reimann, Typical fast thermalization processes in closed many-body systems, Nat. Commun. 7, 10821 (2016)
- L. Dabelow, P. Reimann, Relaxation Theory for Perturbed Many-Body Quantum Systems versus Numerics and Experiment, Phys. Rev. Lett. 124, 120602 (2020)

"All" random states are equal(ly hot)

- A. Hams, H. De Raedt, Fast algorithm for finding the eigenvalue distribution of very large matrices, Phys. Rev. E 62, 4365 (2000)
- J. Schnack, J. Richter, R. Steinigeweg, Accuracy of the finite-temperature Lanczos method compared to simple typicality-based estimates, Phys. Rev. Research 2, 013186 (2020)

S. Lloyd@arXiv:1307.0378: Pure state quantum statistical mechanics and black holes, submitted to PRB in 1988 but rejected by one sentence referee report: "There is no physics."
Deutsch, Srednicki, Goldstein, Lebowitz, Tumulka, Zanghi, Popescu, Short, Winter, Sugiura, Shimizu, . . .

If states or Hamiltonians do not behave like this, they are considered an exception and called atypical.

If you do not want to be mainstream look for atypical stuff.

Yes, we can!

$$\begin{pmatrix} 3 & 42 & 4711 \\ 42 & 0 & 3.14 \\ 4711 & 3.14 & 8 \\ -17 & 007 & 13 \\ 1.8 & 15 & 081 \end{pmatrix}$$

1. A flash on magnetic molecules
2. Typicality approach to equilibrium
3. Stability of clock transitions
4. Synchronization of spins

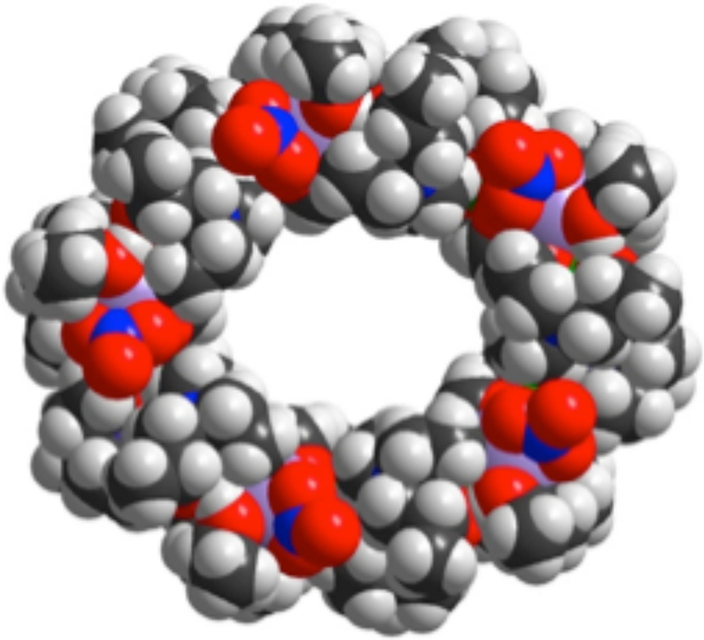
We are the sledgehammer team of matrix diagonalization.
Please send inquiries to jschnack@uni-bielefeld.de!

We investigate

magnetic molecules

J. Schnack, Contemporary Physics **60**, 127-144 (2019)
Jürgen Schnack, Magnetismus im Molekülmaßstab, Physik-Journal **4**, 37 (2017)

You have got a molecule!

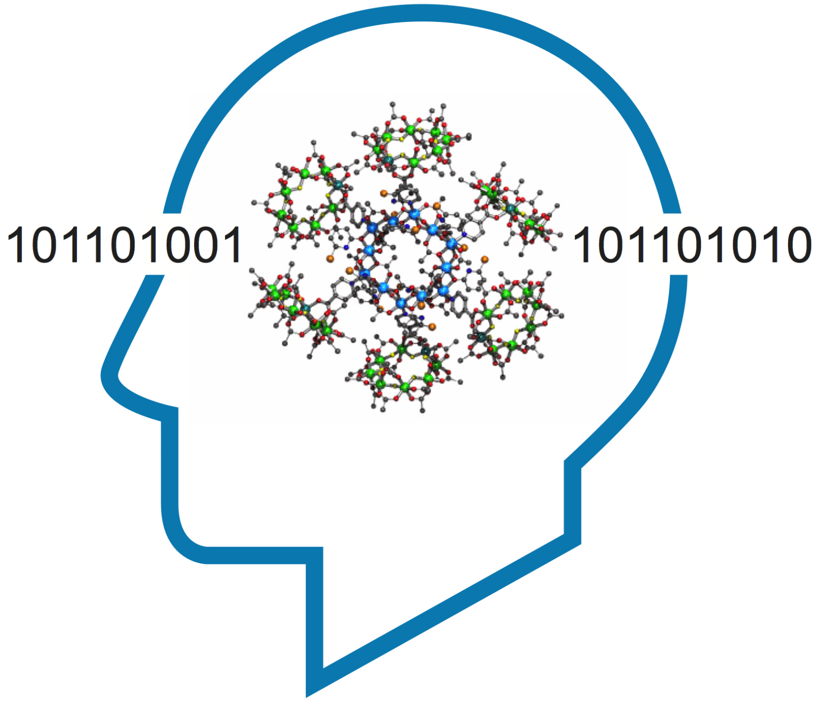


$$S = 60!$$

Congratulations!

Powell group: npj Quantum Materials **3**, 10 (2018)

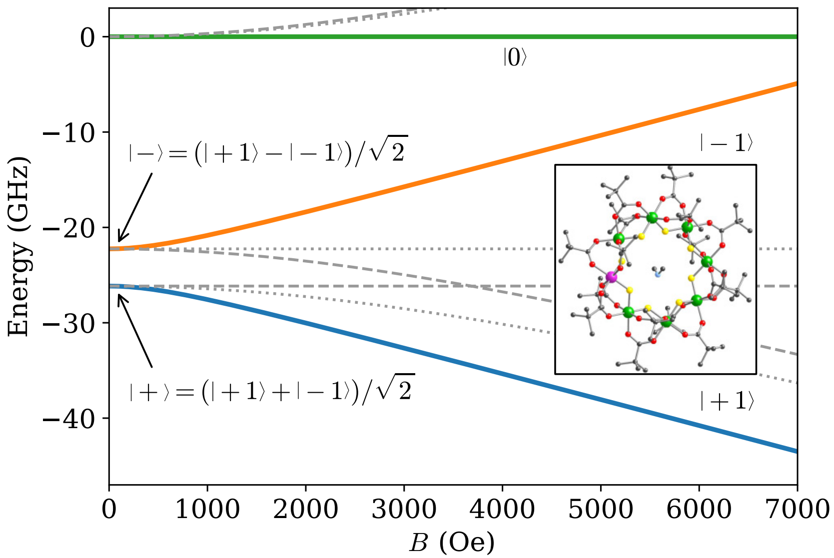
You want to build a quantum computer!



Very smart!

Wernsdorfer group: Phys. Rev. Lett. **119**, 187702 (2017)

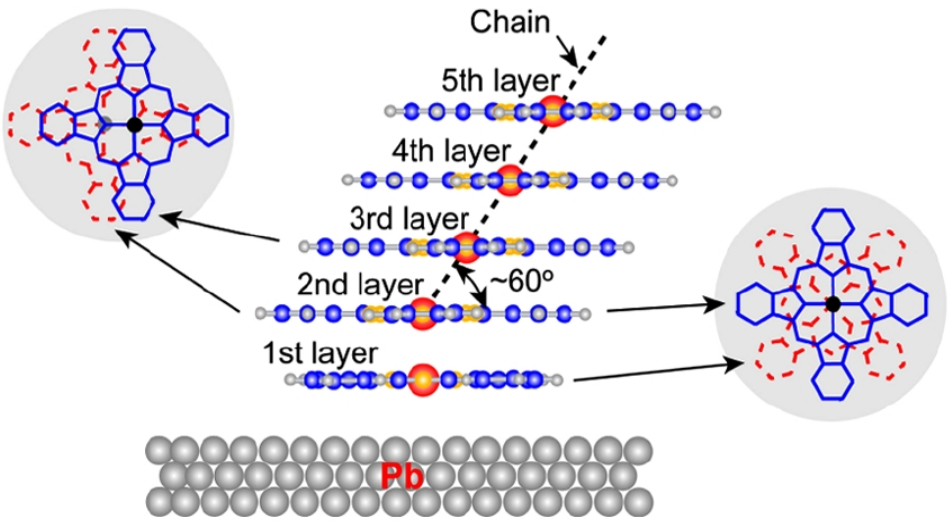
You want to achieve quantum coherence!



Desperately needed!

Friedman group: Phys. Rev. Research **2**, 032037(R) (2020)

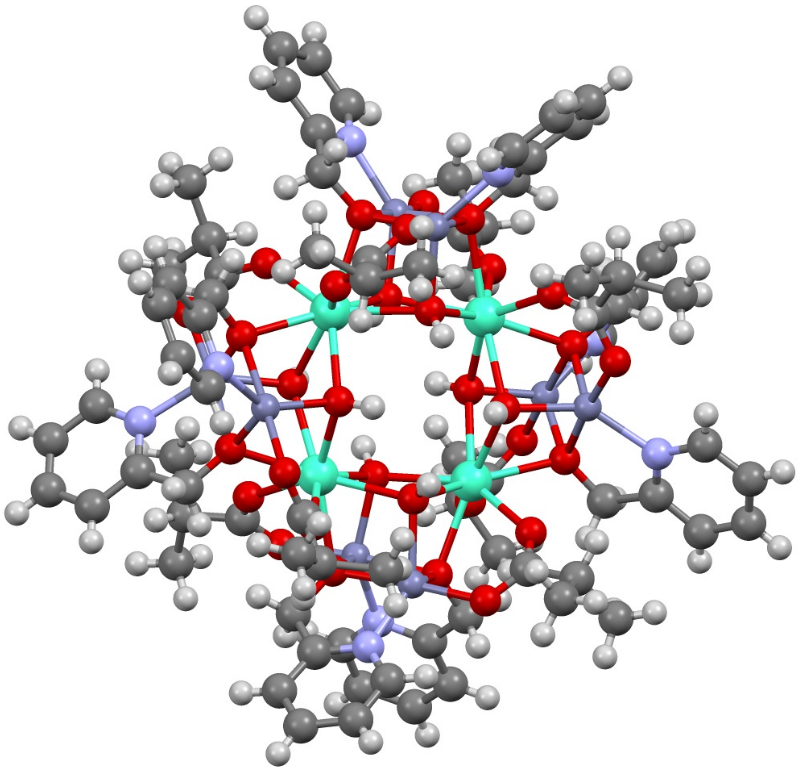
You want to deposit your molecule!



Next generation magnetic storage!

Xue group: Phys. Rev. Lett. **101**, 197208 (2008)

You want molecular magnetocalorics!



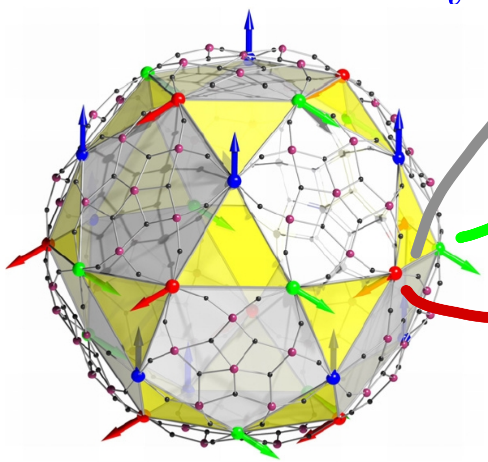
Cool!

Brechin group: Angew. Chem. Int. Ed. **51**, 4633 (2012)

You have got an idea about the modeling!

Heisenberg Zeeman

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



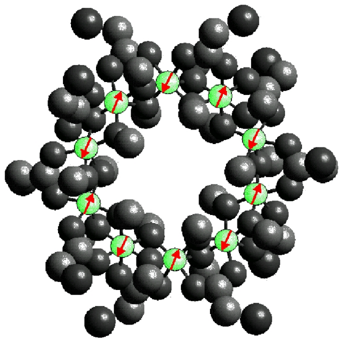
You have to solve the Schrödinger equation!

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

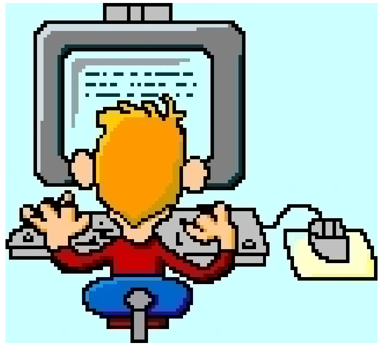
Eigenvalues E_n and eigenvectors $\mid \phi_n \,\rangle$

- 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!



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



$\mathrm{Fe}^{\mathrm{III}}_{10}$: $N = 10$, $s = 5/2$, $\dim(\mathcal{H}) = (2s+1)^N$

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

Can we evaluate the partition function

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

without diagonalizing the Hamiltonian?

Yes, with magic!

Typicality approach to molecular magnetism

Solution I: trace estimators

$$\text{tr}\left(\underset{\sim}{O}\right) \approx \langle\, r \,|\, \underset{\sim}{O} \,|\, r \,\rangle = \sum_{\nu} \langle\, \nu \,|\, \underset{\sim}{O} \,|\, \nu \,\rangle + \sum_{\nu \neq \mu} r_\nu r_\mu \langle\, \nu \,|\, \underset{\sim}{O} \,|\, \mu \,\rangle$$

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

- $|\, \nu \,\rangle$ some orthonormal basis of your choice; not the eigenbasis of $\underset{\sim}{O}$, 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

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

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

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

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!

Partition function I: simple approximation

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

$$O^{\mathrm{r}}(T,B) \approx \frac{\langle\, r \,|\, \underset{\sim}{O} e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle}{\langle\, r \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle} = \frac{\langle\, r \,|\, e^{-\beta \underset{\sim}{H}/2} \underset{\sim}{O} e^{-\beta \underset{\sim}{H}/2} \,|\, r \,\rangle}{\langle\, r \,|\, e^{-\beta \underset{\sim}{H}/2} e^{-\beta \underset{\sim}{H}/2} \,|\, r \,\rangle}$$

- 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???
- Typicality = any random vector will do: $|\, r \,\rangle \equiv (T = \infty)$

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

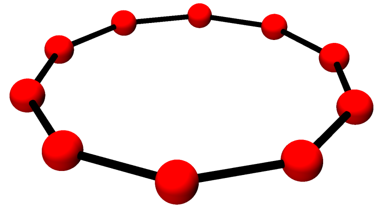
Partition function II: Finite-temperature Lanczos Method

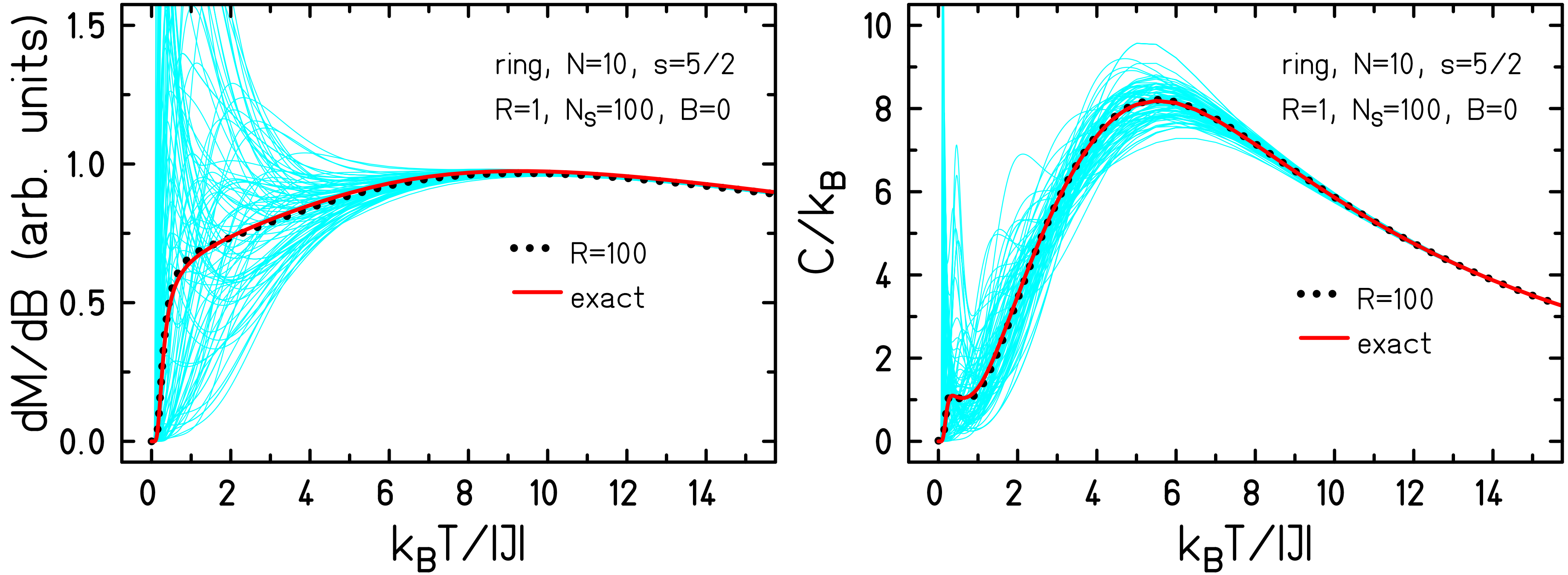
$$Z^{\text{FTLM}}(T,B) \approx \frac{1}{R}\sum_{r=1}^{R}\sum_{n=1}^{N_L} e^{-\beta\epsilon_n^{(r)}} |\langle\, n(r)\,|\, r\,\rangle|^2$$

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

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

FTLM 1: ferric wheel



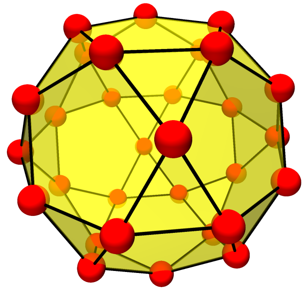


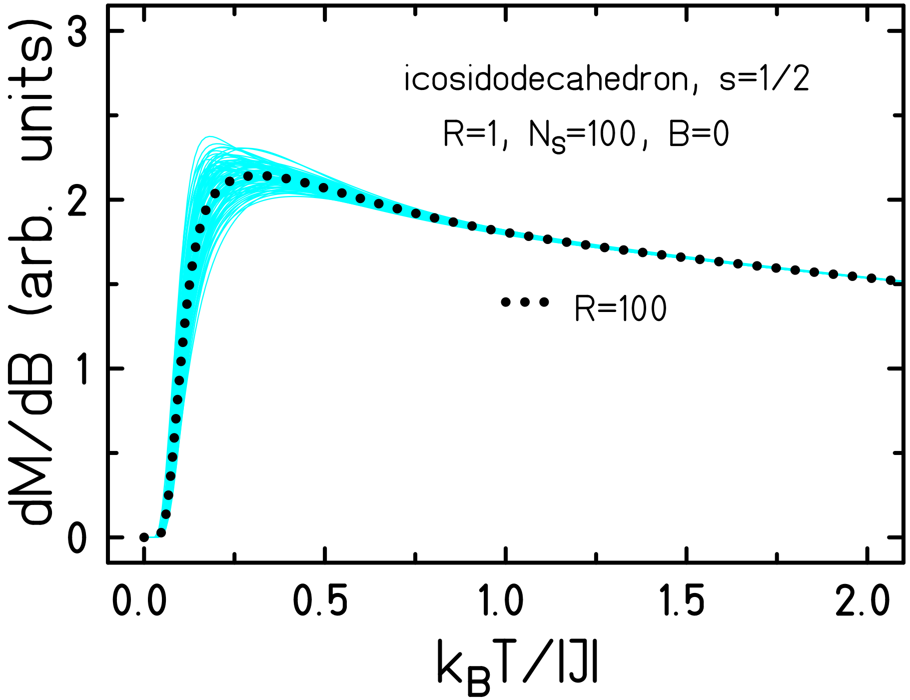
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020).

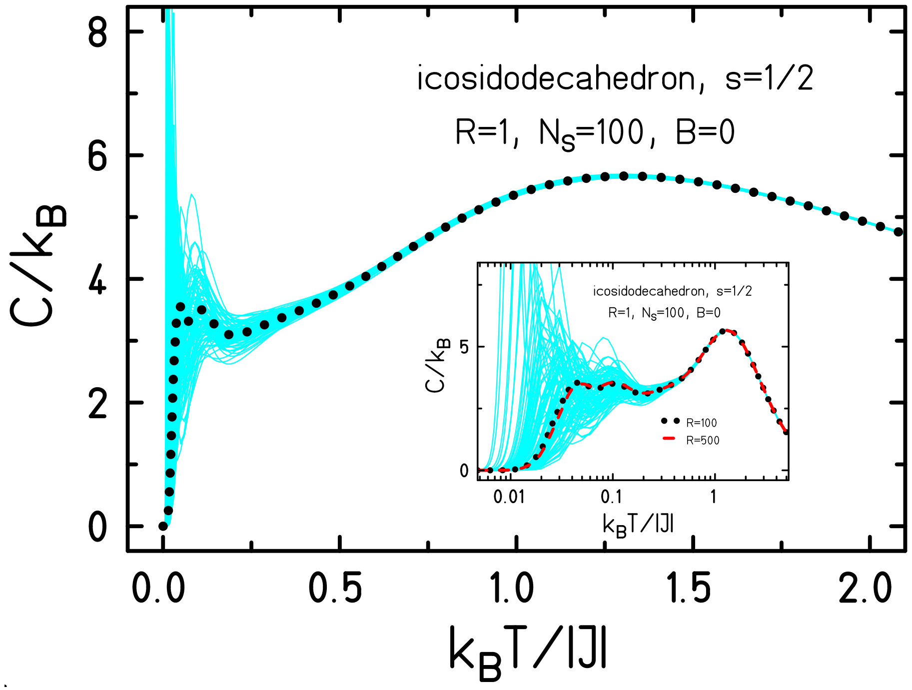
(2) SU(2) & D_2: R. Schnalle and J. Schnack, Int. Rev. Phys. Chem. **29**, 403 (2010).

(3) SU(2) & C_N: T. Heitmann, J. Schnack, Phys. Rev. B **99**, 134405 (2019)

FTLM 2: icosidodecahedron



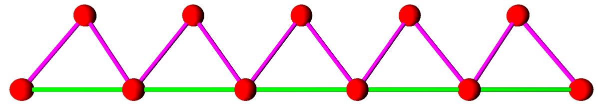


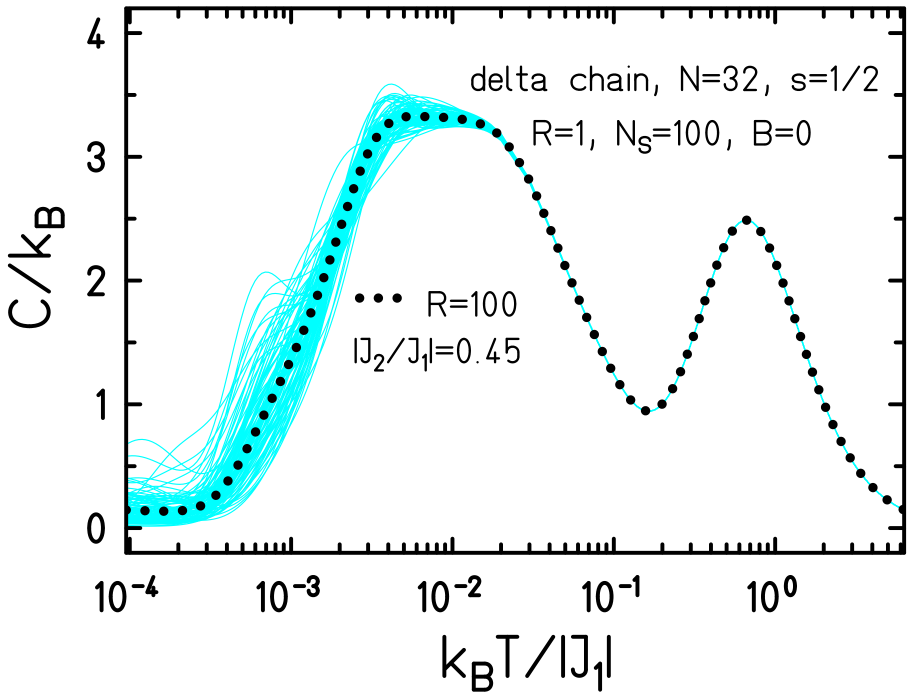


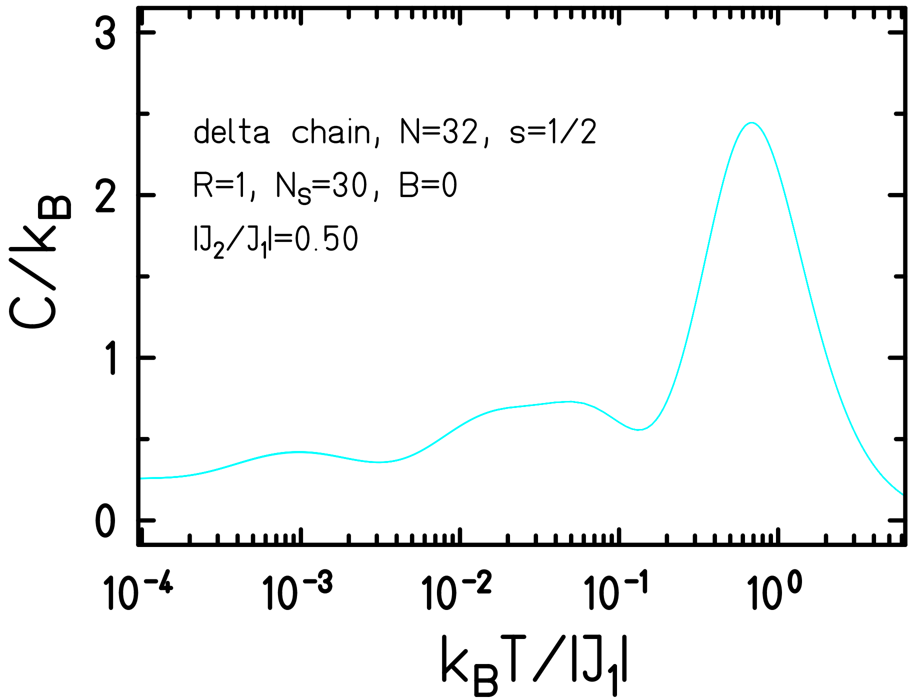
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020).

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

FTLM 3: sawtooth chain





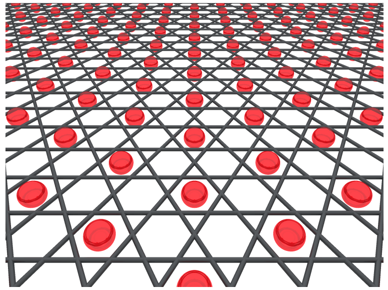


$|J_2/J_1| = 0.45$ – near critical, $|J_2/J_1| = 0.50$ – critical.

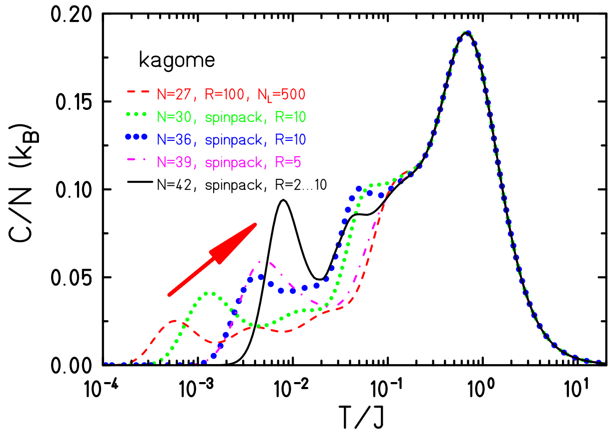
Frustration, technically speaking, works in your favour.

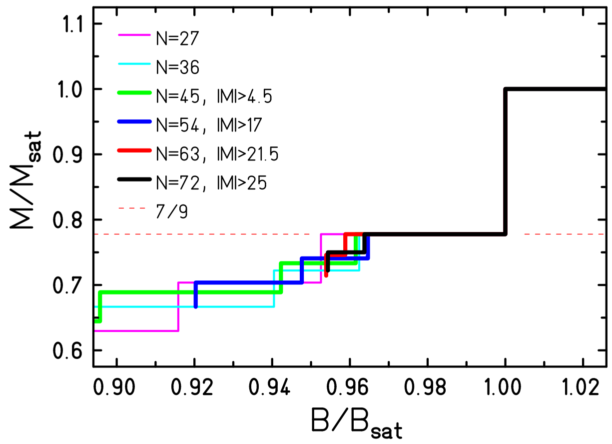
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020)

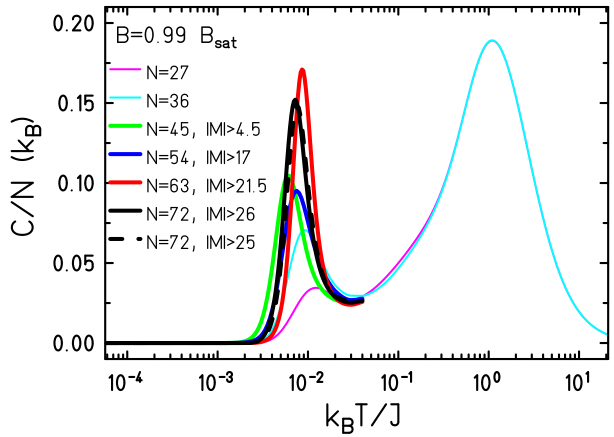
(2) J. Schnack, J. Richter, T. Heitmann, J. Richter, R. Steinigeweg, Z. Naturforsch. A 75, 465 (2020)



FTLM 4: kagome







Specific heat of kagome with $N = 42$ – role of low-lying singlets, and magnon crystalization at high field.

(1) J. Schnack, J. Schulenburg, J. Richter, Phys. Rev. B **98**, 094423 (2018)

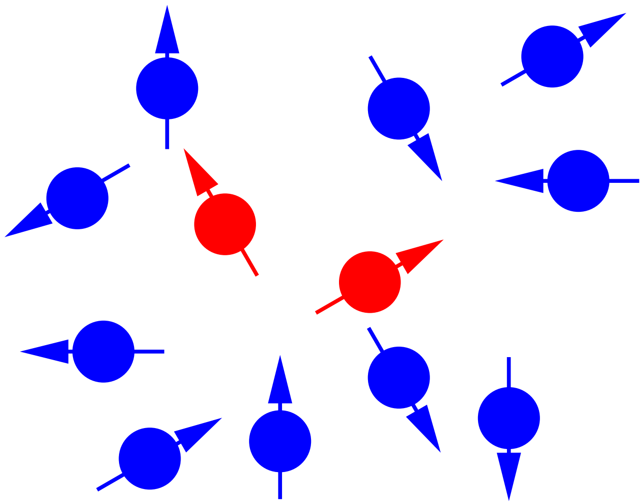
(2) J. Schnack, J. Schulenburg, A. Honecker, J. Richter, Phys. Rev. Lett. 125, 117207 (2020)

Stability of clock transitions

Decoherence is typical, clock transitions are atypical.

$\Rightarrow$ Patrick Vorndamme

Context



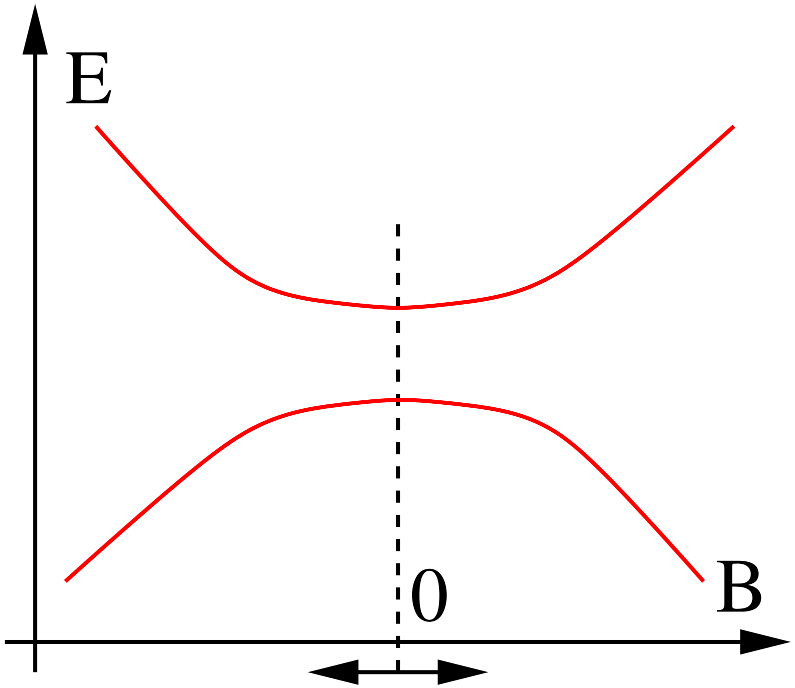
Investigation of decoherence of a subsystem if the combined system (including bath) is evolved via the time-dependent Schrödinger equation.

Employed measure of decoherence: reduced density matrix

$$\underset{\sim}{\rho}_{\text{system}} = \text{Tr}_{\text{bath}}\left(\underset{\sim}{\rho}\right)$$

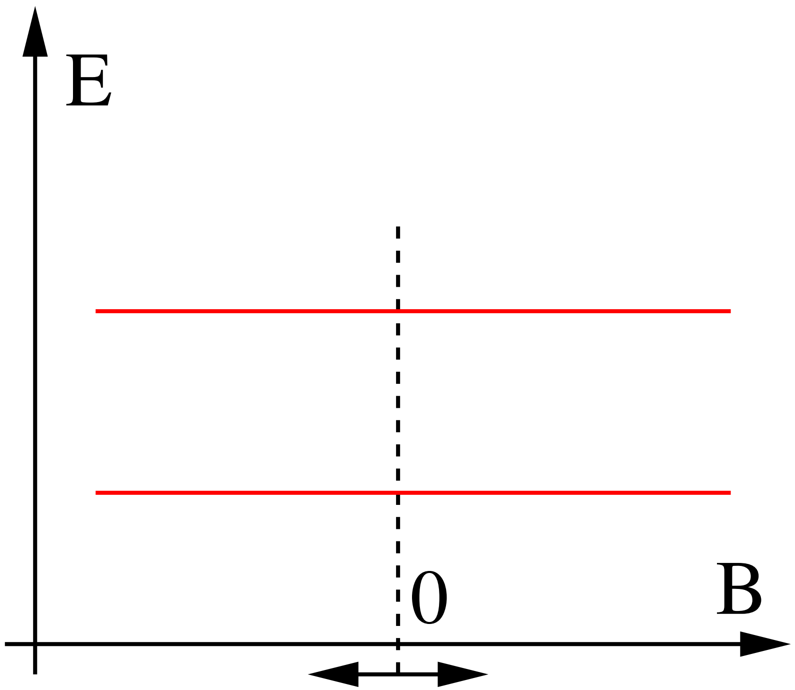
Typicality: unitary-time evolution of pure state approximates dynamics in environment.

Clock transitions

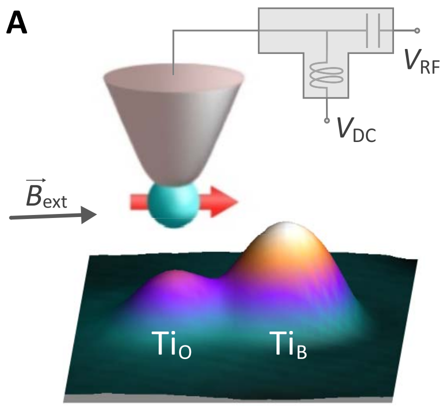


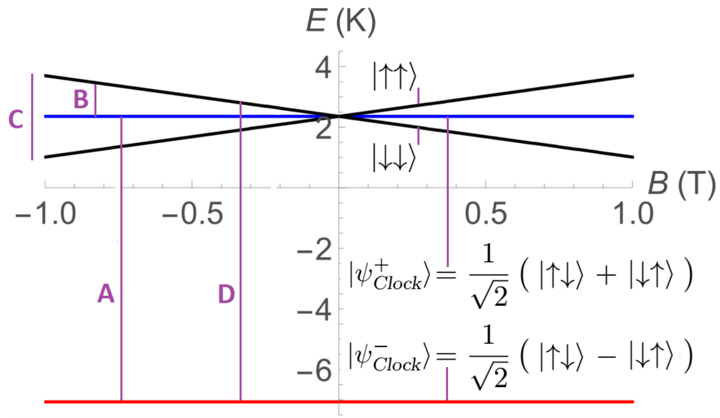
Fluctuations produce little effect on dynamics of superposition since ΔE of clock transition is independent of field at $B = 0$, at least to some order of a Taylor expansion.

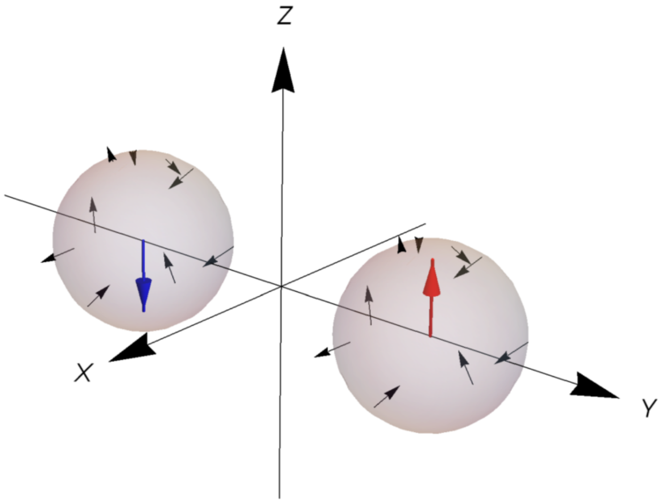
Perfect clock transitions

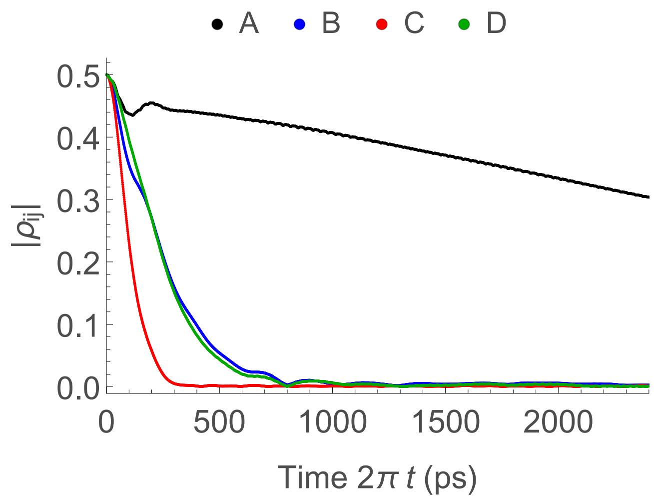


Fluctuations produce very small effect on superposition since ΔE of transition is *totally* independent of field.





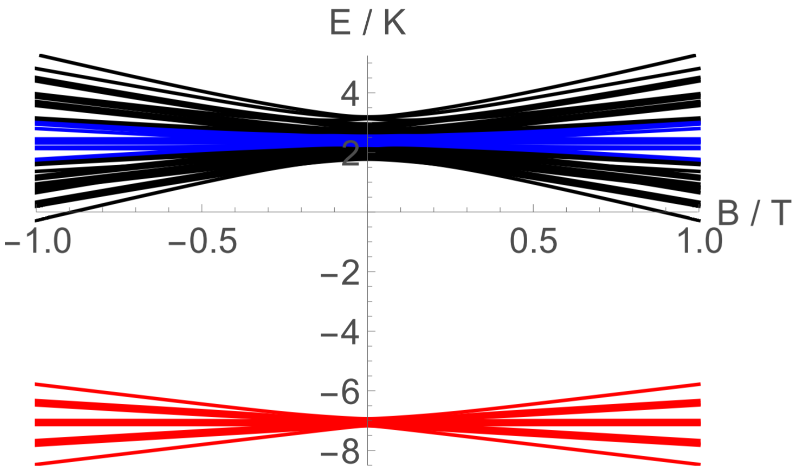




P. Vorndamme, J. Schnack, Phys. Rev. B 101, 075101 (2020)

Y. Bae, K. Yang, P. Willke, T. Choi, A. J. Heinrich, and C. P. Lutz, Sci. Adv. 4, eaau4159 (2018)

Decoherence of clock transitions III



Single-particle/mean-field picture only valid for small couplings to a few bath spins.

Initial product state entangles in the course of time. Eigenstates of the full Hamiltonian loose clock property.

P. Vorndamme, J. Schnack, Phys. Rev. B 101, 075101 (2020)

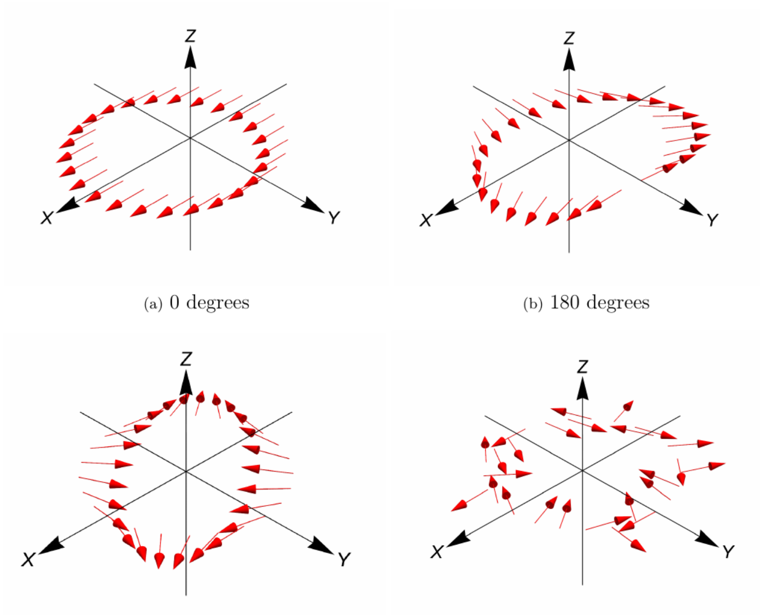
Synchronization of spins

Synchronization is atypical.

$\Rightarrow$ Patrick Vorndamme

Synchronization – Heisenberg model

$$\underset{\sim}{H} = -\sum_{j=1}^{N} J_j \, \vec{\underset{\sim}{s}}_j \cdot \vec{\underset{\sim}{s}}_{j+1} - \sum_{j=1}^{N} h_j \, \underset{\sim}{s}_j^z \, , \qquad |\,\psi(t=0)\,\rangle = \bigotimes_{j=1}^{N} \frac{1}{\sqrt{2}} \left(|\!\uparrow\rangle + e^{i\theta_j} \,|\!\downarrow\rangle \right)$$



(a) 0 degrees (b) 180 degrees

(c) 360 degrees (d) random directions

P. Vorndamme, H.-J. Schmidt, Chr. Schröder, J. Schnack, New J. Phys. **23**, 083038 (2021).

Synchronization – Heisenberg model with random Js

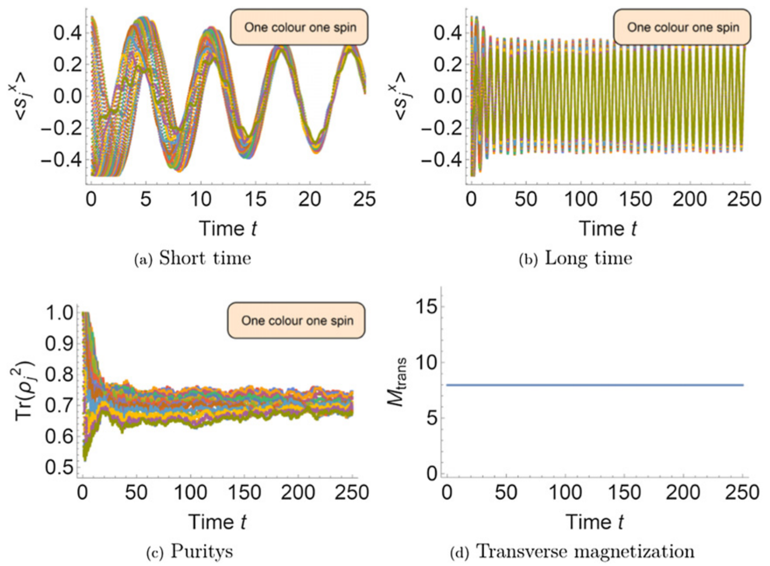


Figure 3. Time evolution of initial state $|\psi_B\rangle$ w.r.t. Hamiltonian equation (1) with isotropic Heisenberg interactions and $J_j \in [1.6, 2.4]$, $h_j = -1\ \forall j$, $N = 25$. The video for (a) (https://stacks.iop.org/NJP/23/083038/mmedia) are provided in the supplementary data.

- Spins synchronize under unitary time evolution!

- $\underset{\sim}{\rho}_j = \mathit{Tr}_{\bigotimes_{k\neq j} \mathscr{H}_k}\left(|\,\psi\,\rangle\langle\,\psi\,| \right)$,

  purity $\mathit{Tr}\left(\underset{\sim}{\rho}_j^2\right) := 1$ (not entangled), $= 0.5$ (maximally entangled).

Transient synchronization – XYZ model

$$\underset{\sim}{H}_{XYZ} = -J \sum_{j=1}^{N} \underset{\sim}{s}_{j}^{x} \underset{\sim}{s}_{j+1}^{x} - (J-\delta) \sum_{j=1}^{N} \underset{\sim}{s}_{j}^{y} \underset{\sim}{s}_{j+1}^{y} - (J-2\delta) \sum_{j=1}^{N} \underset{\sim}{s}_{j}^{z} \underset{\sim}{s}_{j+1}^{z} - h \sum_{j=1}^{N} \underset{\sim}{s}_{j}^{z} \ .$$

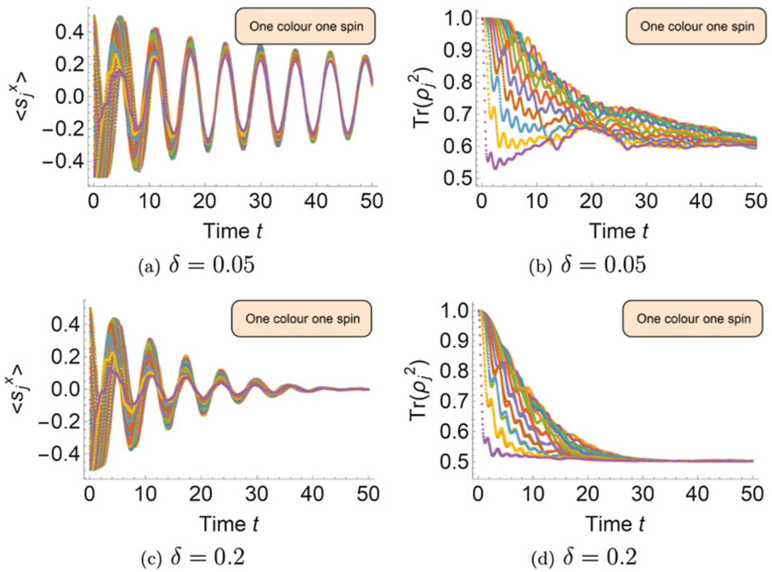
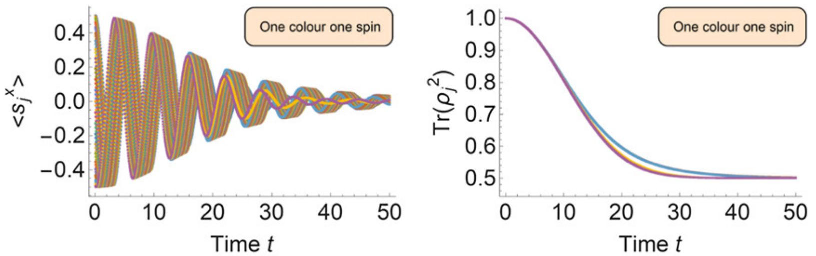


Figure 9. Time evolutions of initial state $|\psi_B\rangle$ w.r.t. Hamiltonian equation (14) for two values of δ, and $N = 24$, $J = 2$, $h = -1$. Videos of (a) and (c) are provided in the supplementary data.

P. Vorndamme, H.-J. Schmidt, Chr. Schröder, J. Schnack, New J. Phys. **23**, 083038 (2021).

No synchronization – XX model

$$\underset{\sim}{H}_{XX} = \quad -J \sum_{j=1}^{N} \left(\underset{\sim}{s}_j^x \underset{\sim}{s}_{j+1}^x + \underset{\sim}{s}_j^y \underset{\sim}{s}_{j+1}^y \right) - h \sum_{j=1}^{N} \underset{\sim}{s}_j^z \ .$$

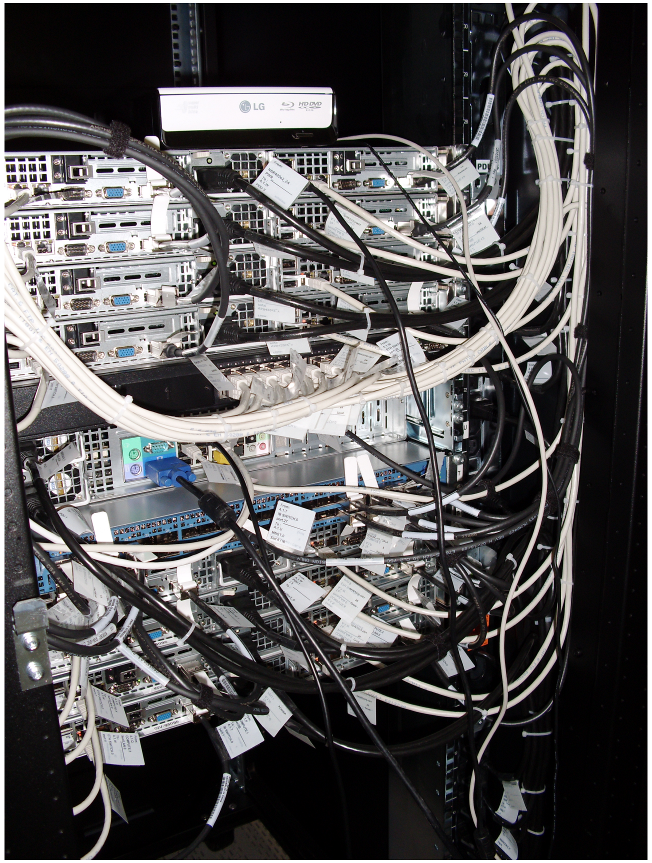


(a) Single spin expectation values (b) Puritys

Figure 10. Time evolution of initial state $|\psi_B\rangle$ w.r.t. Hamiltonian equation (15) with parameters $N = 24$, $J = 0.1$ and $h = -1$. The video of (a) is provided in the supplementary data.

P. Vorndamme, H.-J. Schmidt, Chr. Schröder, J. Schnack, New J. Phys. **23**, 083038 (2021).

Summary



- Magnetic molecules for storage, q-bits, MCE, and since they are nice.
- Typicality – potentially powerful concept: Fundamental Aspects of Statistical Mechanics and the Emergence of Thermodynamics in Non-Equilibrium Systems (FOR 2692).
- Synchronization of spin systems. SU(2) symmetry needed?

Many thanks to my collaborators







- C. Beckmann, M. Czopnik, T. Glaser, O. Hanebaum, Chr. Heesing, M. Höck, K. Irländer, N.B. Ivanov, H.-T. Langwald, A. Müller, H. Schlüter, R. Schnalle, Chr. Schröder, J. Ummethum, P. Vorndamme (Bielefeld)
- K. Bärwinkel, T. Heitmann, R. Heveling, H.-J. Schmidt, R. Steinigeweg (Osnabrück)
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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); J. Wosnitza (Dresden-Rossendorf); J. van Slageren (Stuttgart); R. Klingeler (Heidelberg); O. Waldmann (Freiburg); U. Kortz (Bremen)

Thank you very much for your attention.

The end.

Molecular Magnetism Web

www.molmag.de

Highlights. Tutorials. Who is who. Conferences.