FOR2692 meets SFB/TRR288: typicality and exact quantum dynamics for equilibrium and non-equilibrium properties

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Colloquium of SFB/TRR 288, Frankfurt/Mainz/Karlsruhe
Online, 23 April 2021

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, . . .

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. Spin-phonon issues

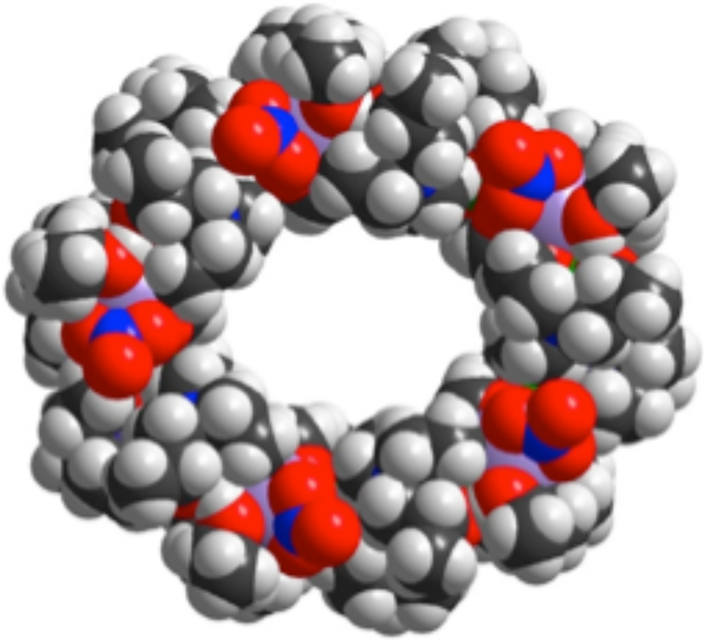
We are the sledgehammer team of matrix diagonalization.
Please send inquiries to jschnack@uni-bielefeld.de!

We investigate

magnetic molecules

J. Schnack, Contemporary Physics **60**, 127-144 (2019)

You have got a molecule!

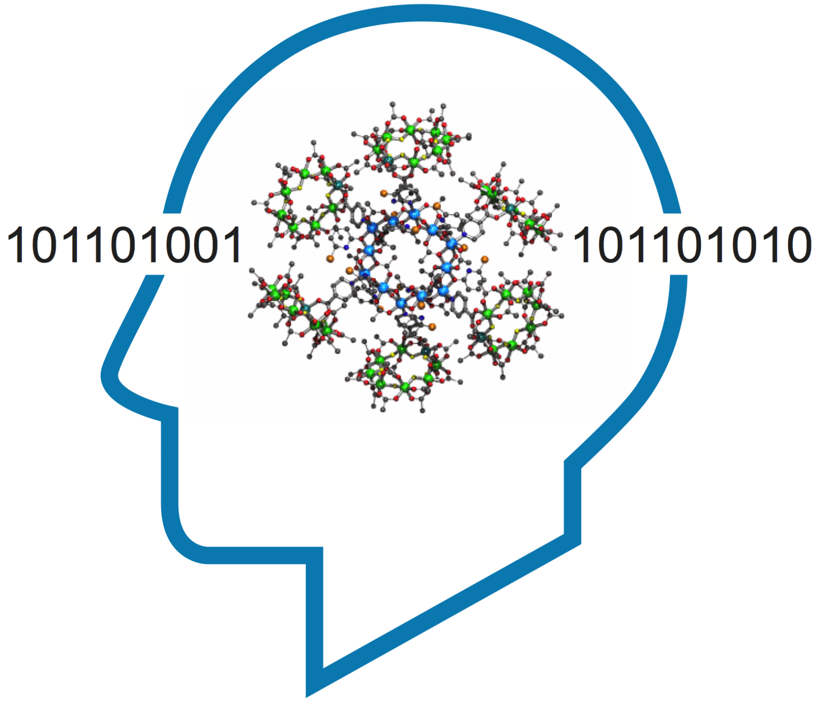


$$S = 60!$$

Congratulations!

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

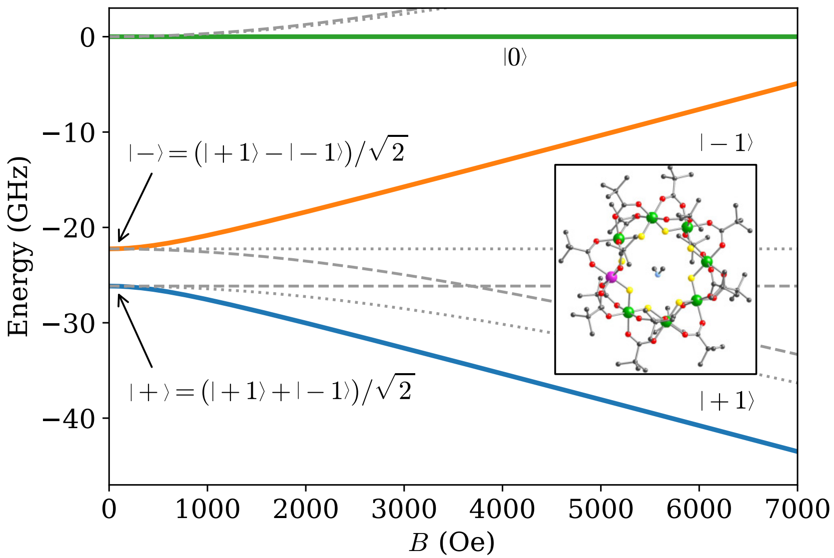
You want to build a quantum computer!



Very smart!

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

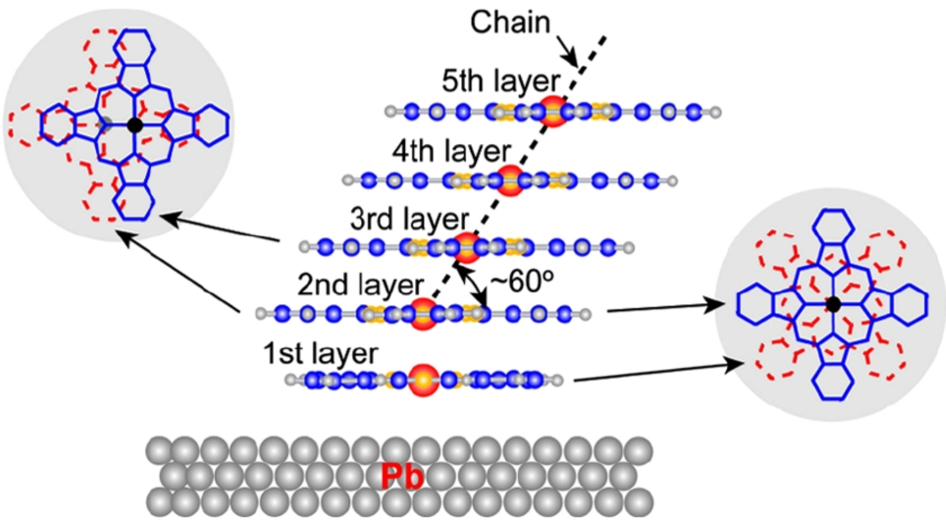
You want to achieve quantum coherence!



Desperately needed!

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

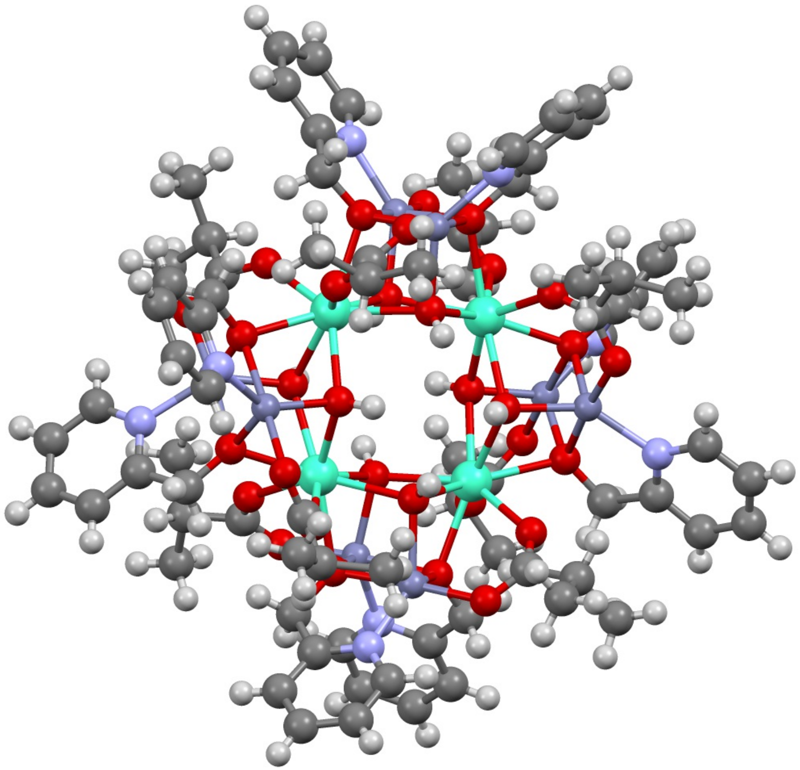
You want to deposit your molecule!



Next generation magnetic storage!

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

You want molecular magnetocalorics!



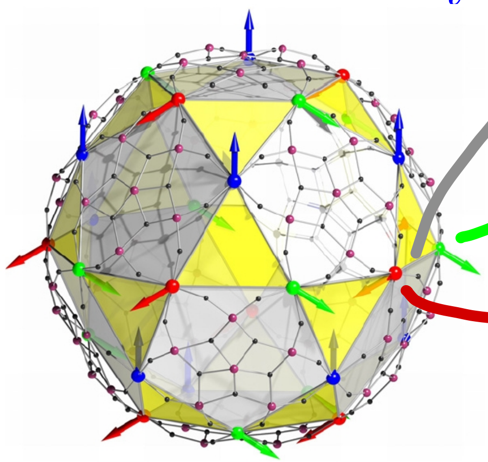
Cool!

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

You have got an idea about the modeling!

Heisenberg **Zeeman**

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



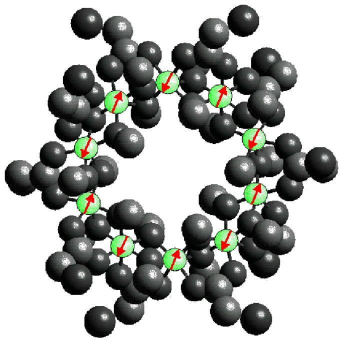
You have to solve the Schrödinger equation!

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

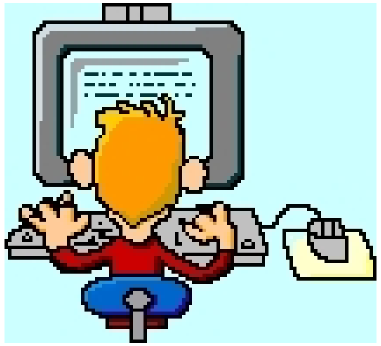
Eigenvalues E_n and eigenvectors $\mid \phi_n \rangle$

- 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!



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



Fe^{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

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

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

- 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).

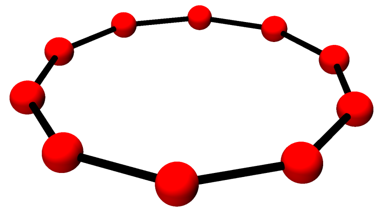
Partition function II: Finite-temperature Lanczos Method

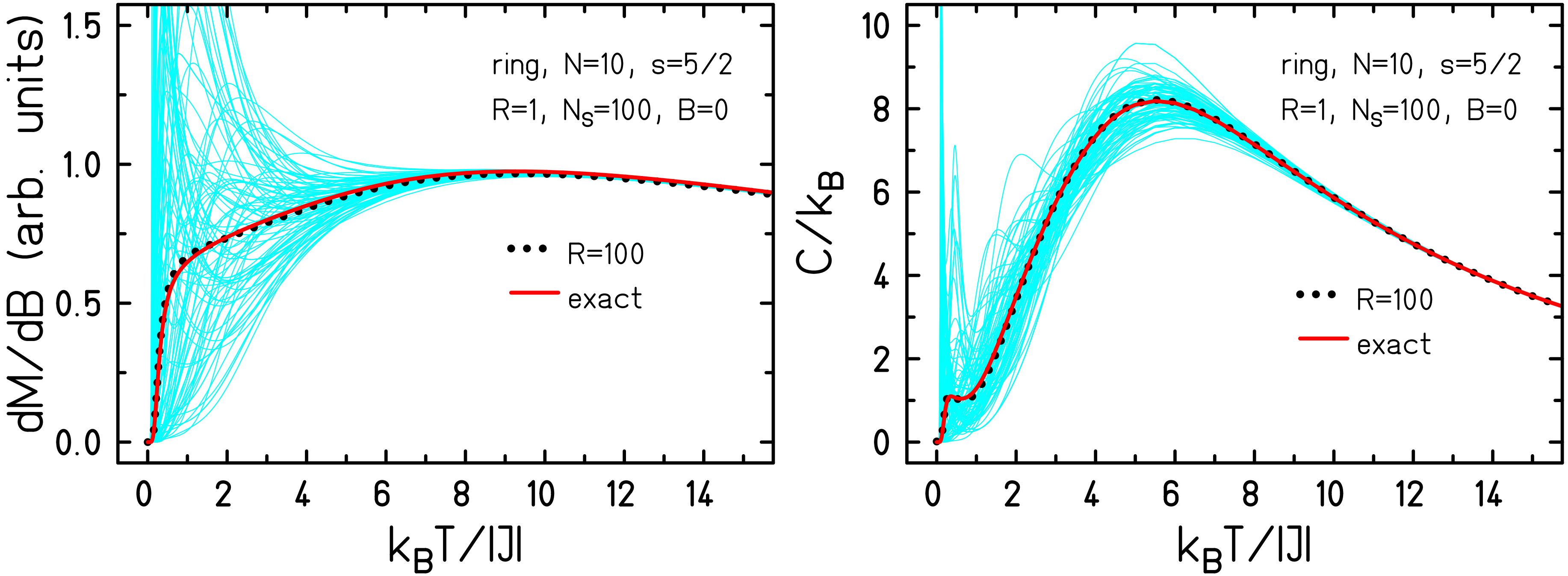
$$Z^{\text{FTLM}}(T,B) \;\approx\; \frac{1}{R}\sum_{r=1}^{R}\sum_{n=1}^{N_L} e^{-\beta\epsilon_n^{(r)}} |\langle\, n(r)\,|\, r\,\rangle|^2$$

- 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: $R = 1\ldots 100, N_L \approx 100$.
- Use symmetries!

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

FTLM 1: ferric wheel



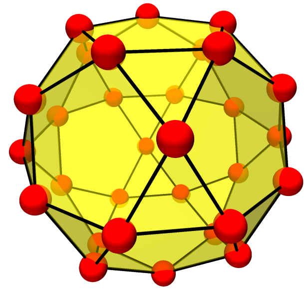


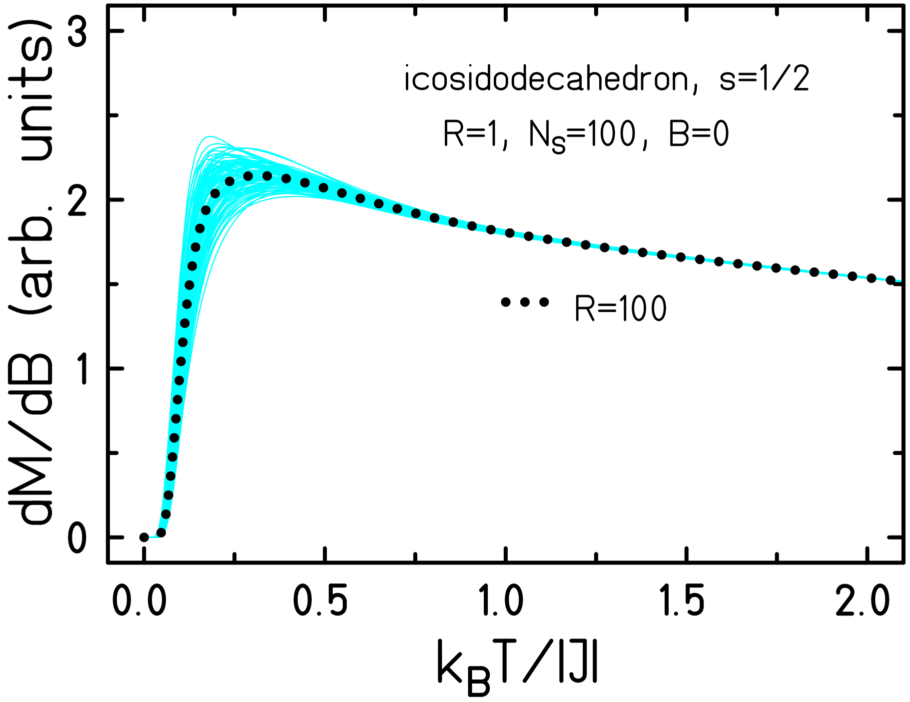
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020).

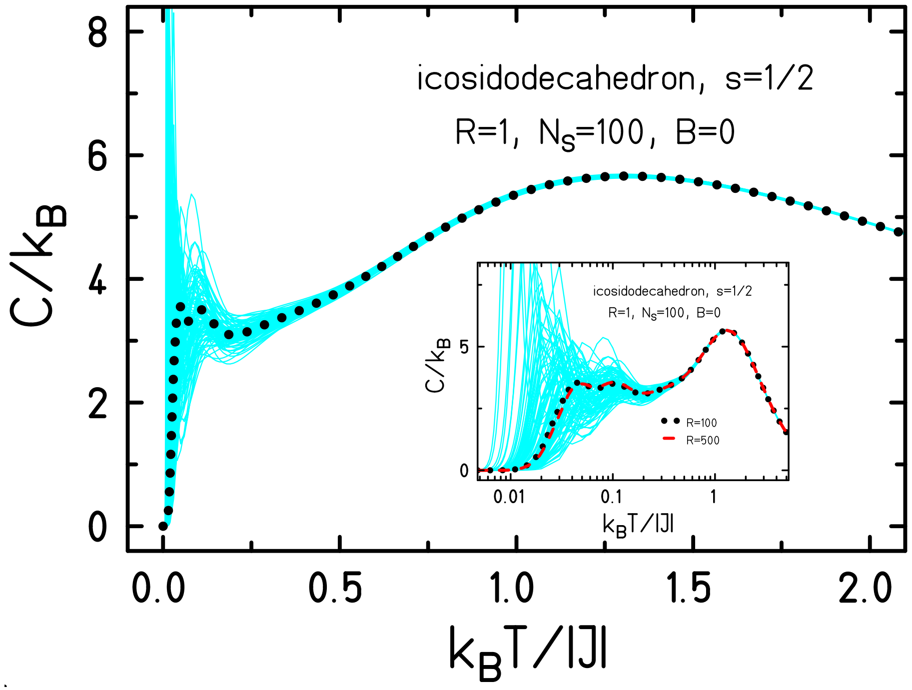
(2) SU(2) & D_2: R. Schnalle and J. Schnack, Int. Rev. Phys. Chem. **29**, 403 (2010).

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

FTLM 2: icosidodecahedron



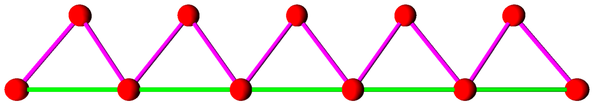


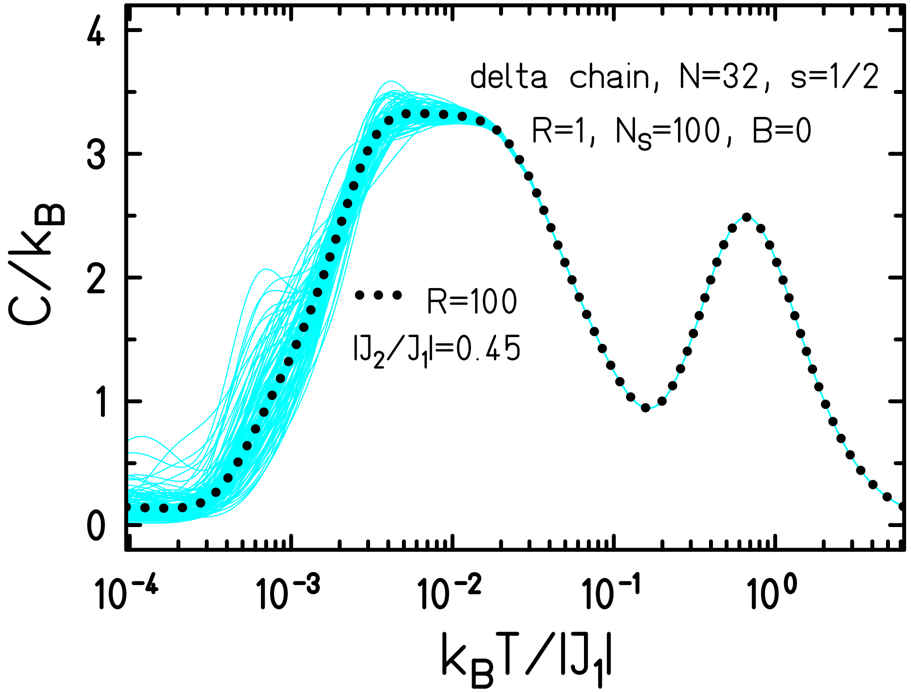


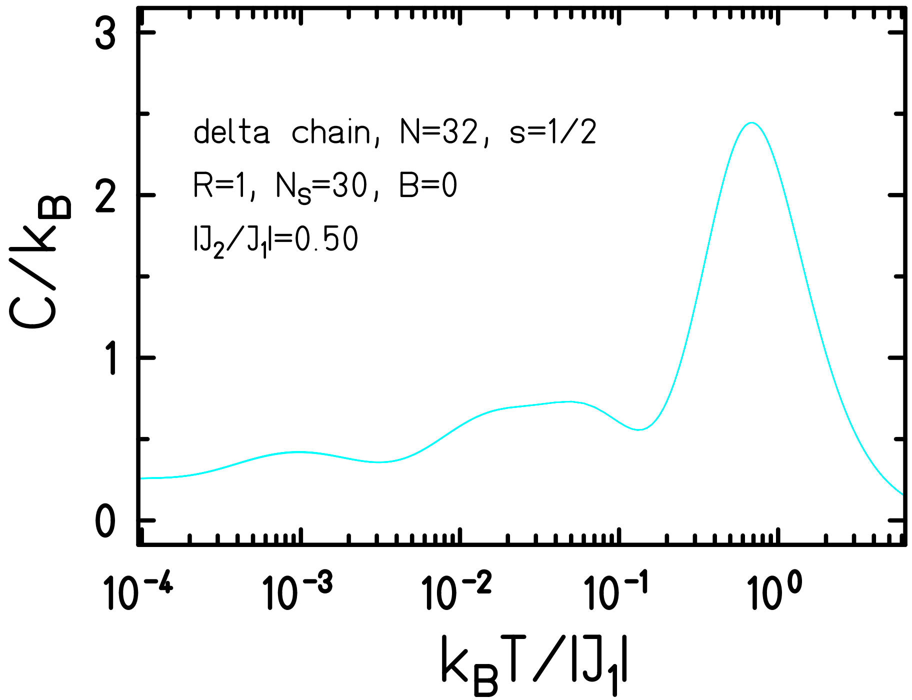
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020).

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

FTLM 3: sawtooth chain





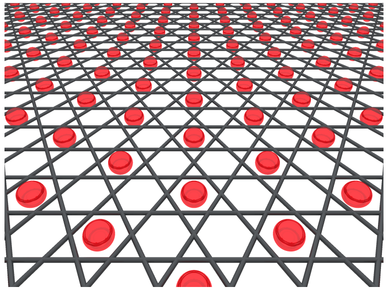


$|J_2/J_1| = 0.45$ – near critical, $|J_2/J_1| = 0.50$ – critical.

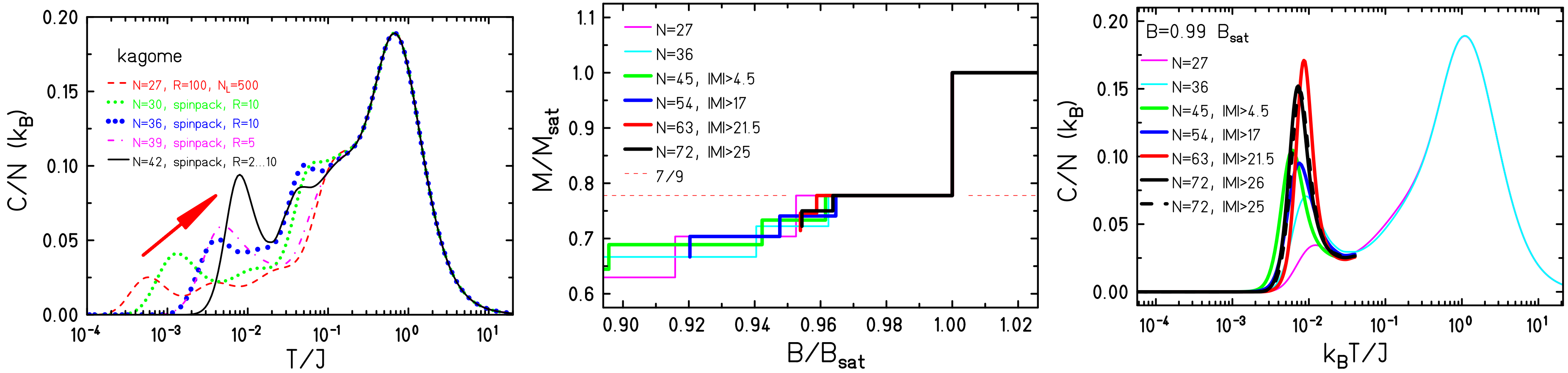
Frustration, technically speaking, works in your favour.

(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



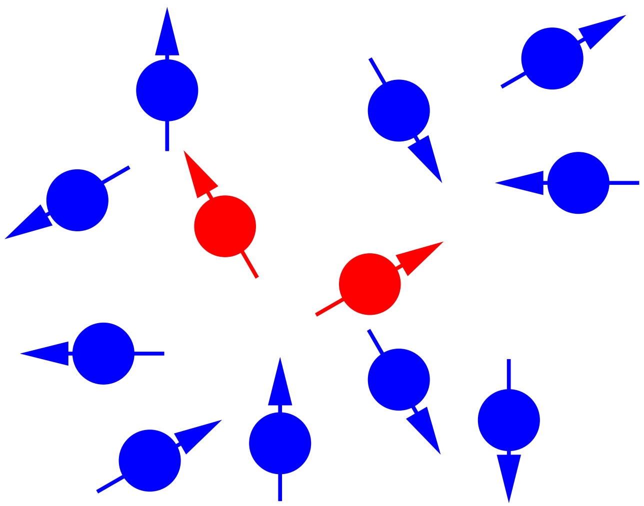
Specific heat of kagome with $N = 42$ – role of low-lying singlets, and magnon crystalization at high field.

(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

Context



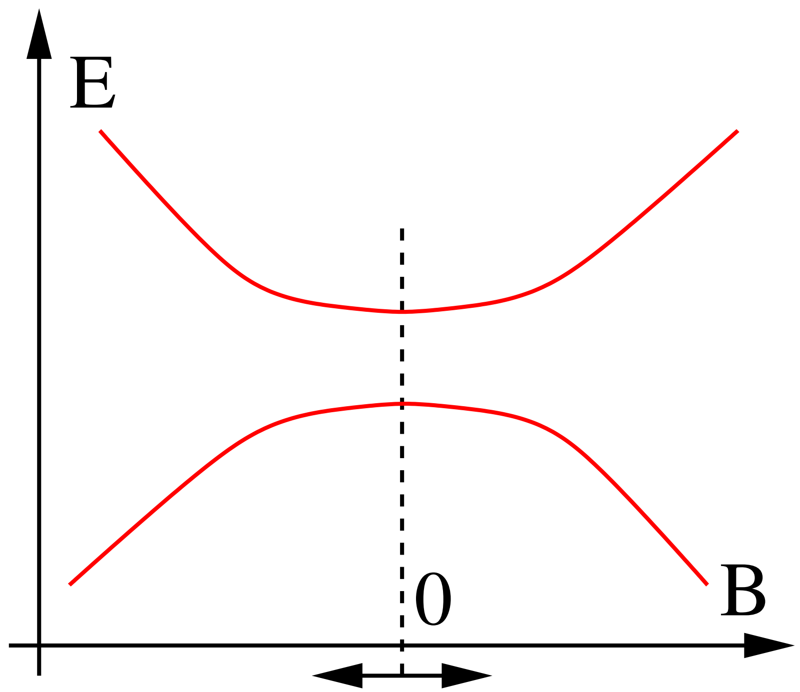
Investigation of decoherence of a subsystem if the combined system (including bath) is evolved via the time-dependent Schrödinger equation.

Employed measure of decoherence: reduced density matrix

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

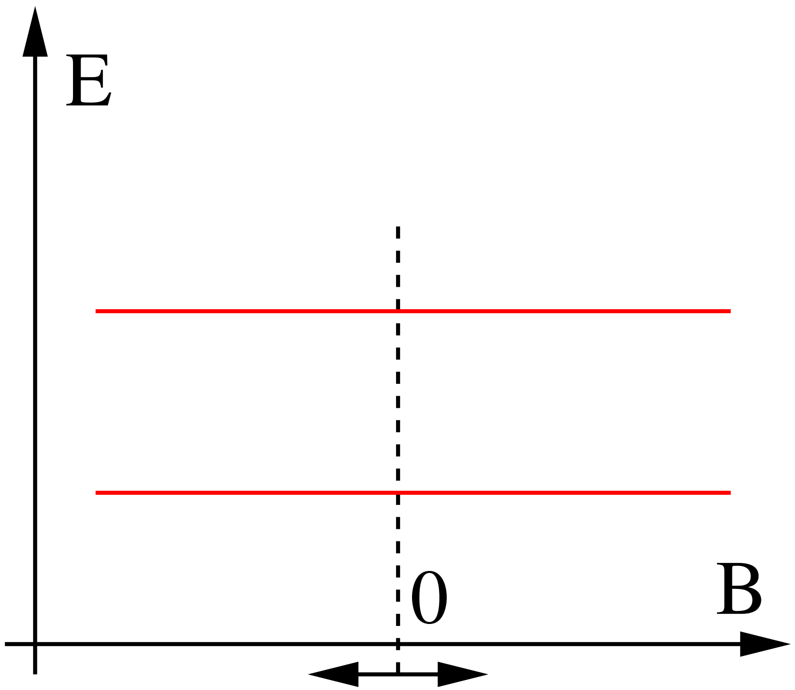
Typicality: unitary-time evolution of pure state approximates dynamics in environment.

Clock transitions

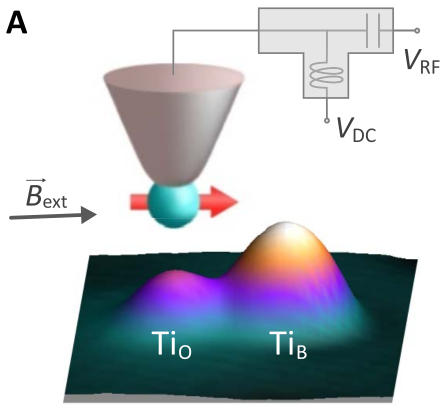


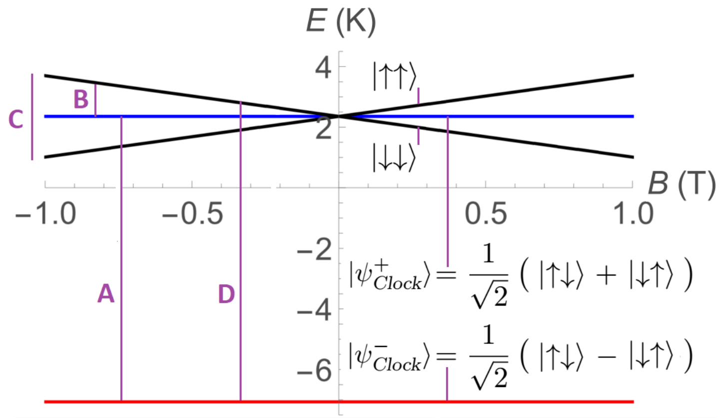
Fluctuations produce little effect on dynamics of superposition since ΔE of clock transition is independent of field at $B = 0$, at least to some order of a Taylor expansion.

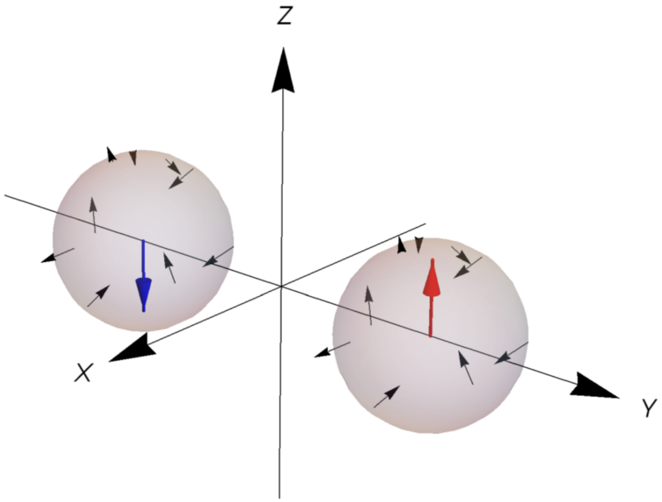
Perfect clock transitions

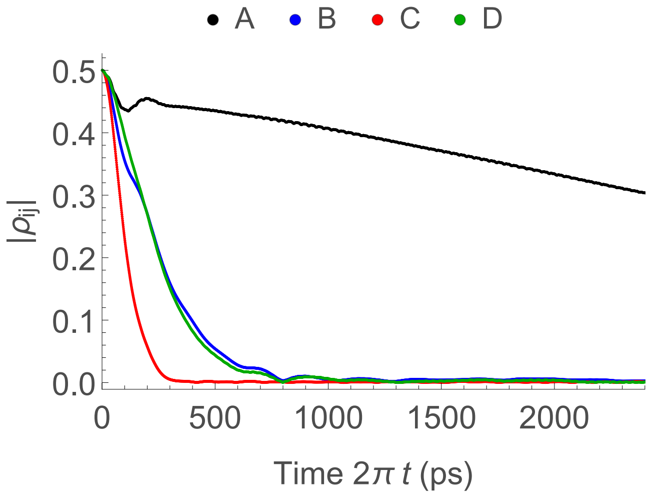


Fluctuations produce very small effect on superposition since ΔE of transition is *totally* independent of field.





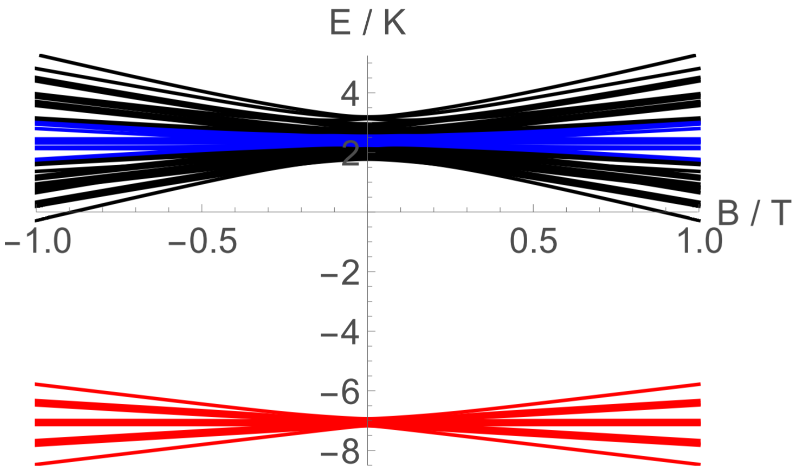




P. Vorndamme, J. Schnack, Phys. Rev. B 101, 075101 (2020)

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)

Spin-phonon interaction

Model Hamiltonian (effective, spin-only, bilinear)

$$\underset{\sim}{H} = \sum_{i,j} \underset{\sim}{\vec{s}}(i) \cdot \mathbf{J}_{ij} \cdot \underset{\sim}{\vec{s}}(j) + \mu_B \vec{B} \cdot \sum_{i}^{N} \mathbf{g}_i \cdot \underset{\sim}{\vec{s}}(i)$$

Exchange/Anisotropy Zeeman

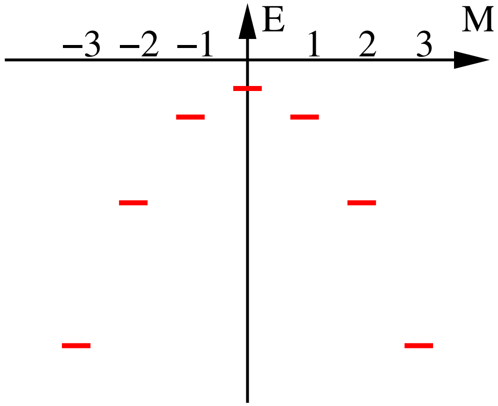
$\mathbf{J}_{ij}$: Heisenberg exchange, anisotropic exchange, and single-ion anisotropy.

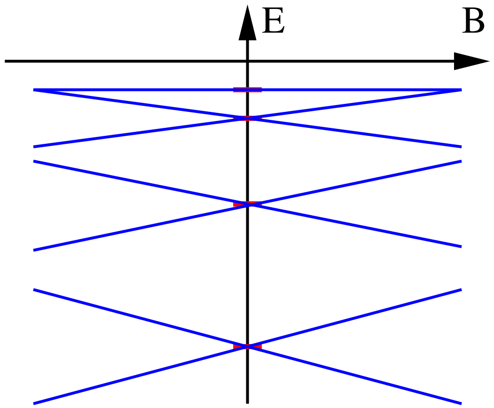
Isotropic Heisenberg Hamiltonian

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

Heisenberg Zeeman

Single-ion anisotropy – single spin I





$\underset{\sim}{H} = D(\underset{\sim}{s}^z)^2 + g\mu_B B \underset{\sim}{s}^z$

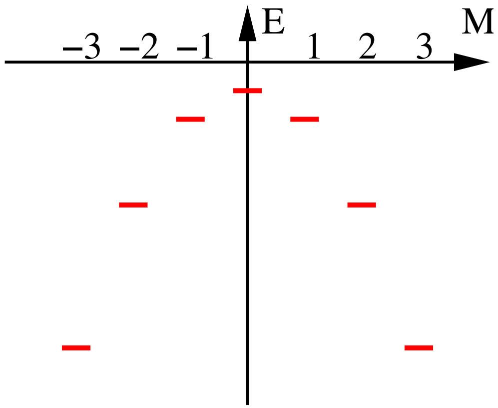
$D < 0$ easy axis, $D > 0$ hard axis;

eigenvectors: $\mid s, m \,\rangle$

eigenvalues: $E_m = Dm^2 + g\mu_B Bm \; , \; m = -s, \dots, s$

IMPORTANT: $[\underset{\sim}{H}, \underset{\sim}{s}^z] = 0 \Rightarrow$ level crossings at $B = 0$

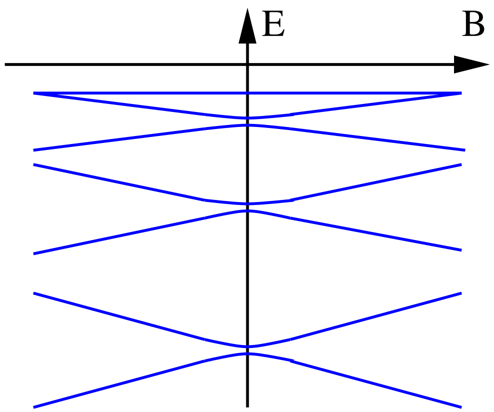
Single-ion anisotropy – single spin II



$$\underset{\sim}{H} = D(\underset{\sim}{s}^z)^2 + E\left\{(\underset{\sim}{s}^x)^2 - (\underset{\sim}{s}^y)^2\right\} + g\mu_B B \underset{\sim}{s}^z$$

$|E| < |D|$ – major axes of the anisotropy tensor;

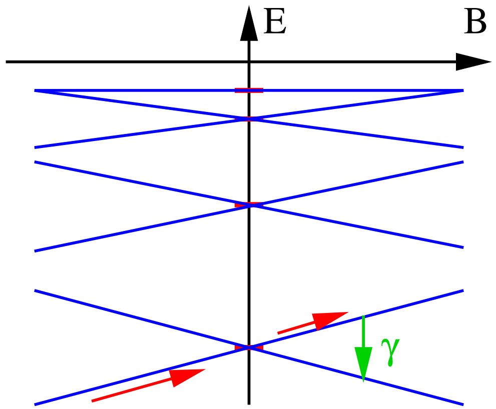
NO LONGER eigenvectors: $|\, s, m\, \rangle$

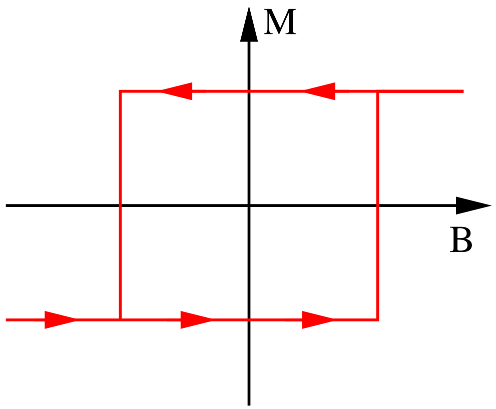


eigenvalues are more complicated functions of $\vec{B} = B\vec{e}_z$: $E_\mu(B)$

IMPORTANT: $[\underset{\sim}{H}, \underset{\sim}{s}^z] \neq 0 \Rightarrow$ avoided level crossings at $B = 0$ for integer spins
(otherwise Kramers degeneracy)

Bistability – uniaxial system – $\underset{\sim}{S}^z$-symmetry





Goal: single-molecule magnets (SMM)

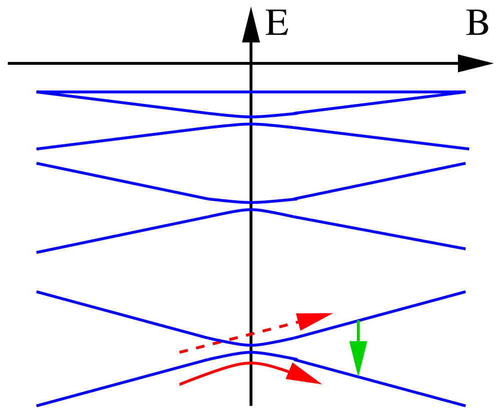
$\underset{\sim}{H} = \sum_i D_i (\underset{\sim}{s}_i^z)^2 + \mu_B B \sum_i g_i \underset{\sim}{s}_i^z + \underset{\sim}{H}_{\text{ferro int}}$

IMPORTANT: $[\underset{\sim}{H}, \underset{\sim}{S}^z] = 0 \Rightarrow$ level crossings at $B = 0$

$\Rightarrow$ low-temperature TIME-DEPENDENT hysteresis

Side remark: For macroscopic systems in the ferromagnetic phase the relaxation time is HUGE, that's why we don't experience it.

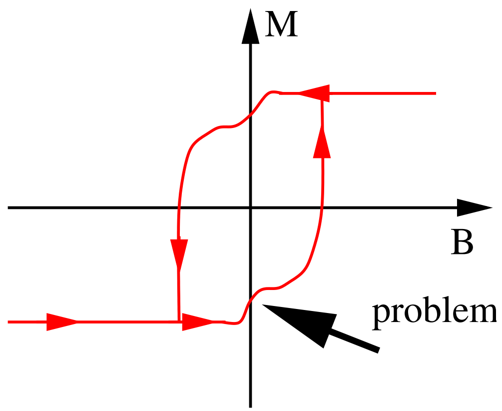
Bistability – general system – NO $\underset{\sim}{S}^z$-symmetry



$$\underset{\sim}{H} = \sum_i \underset{\sim}{\vec{s}}_i \cdot \mathbf{D}_i \cdot \underset{\sim}{\vec{s}}_i + \mu_B B \sum_i g_i \underset{\sim}{s}_i^z + \underset{\sim}{H}_{\text{ferro int}}$$

$\mathbf{D}_i$ individual anisotropy tensors

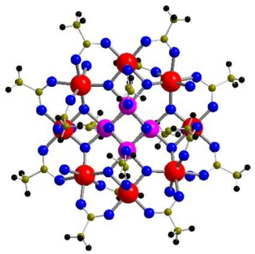
$\Rightarrow$ low-temperature TIME-DEPENDENT hysteresis closes at $B = 0$ – not bistable & bad for storage



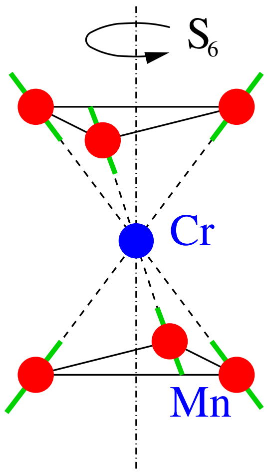
REASON: branching at avoided level crossings; strong dependence on tunneling gap and $\dot{B}$;

slow change of $B \Rightarrow$ system follows ground state, compare Landau-Zener-Stückelberg or slow/fast train at switch

Bistability – state of the art



Today's major goals:

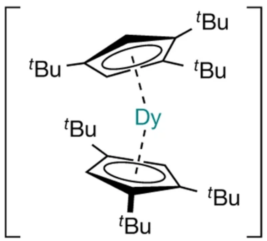


ferromagnetic spin-spin interaction

uniaxial anisotropy tensors

symmetry that does not permit E-terms

PERSISTENT PROBLEM: phonons



Nick Chilton, Thorsten Glaser, Jeff Long, Alessandro Lunghi, Mark Murrie, Frank Neese, Stefano Sanvito, Roberta Sessoli, Richard Winpenny, Yan-Zhen Zheng, . . .

Spin-phonon interaction – DFT view of the problem

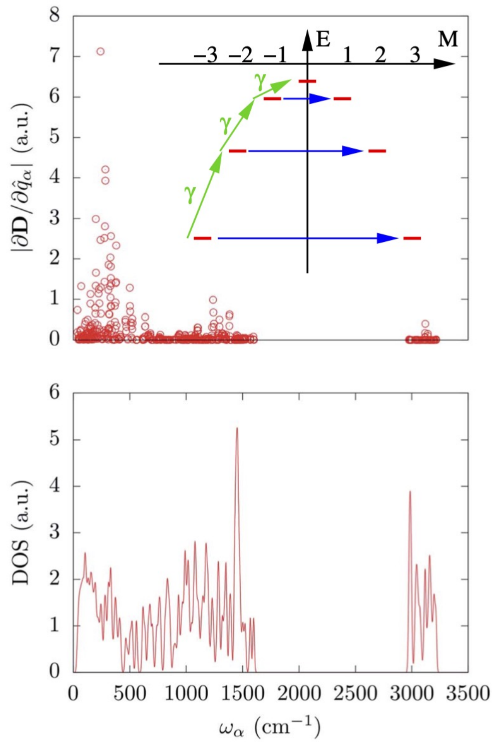


Fig. 2 Top panel: calculated spin-phonon coupling coefficients projected onto the normal modes basis set and displayed as a function of the modes frequency. Bottom panel: DFT calculated density of states for the Γ-point normal modes of vibration.

Calculate structure by means of DFT (1)

Calculate phonon density of states by means of DFT + molecular dynamics (1,2,3)

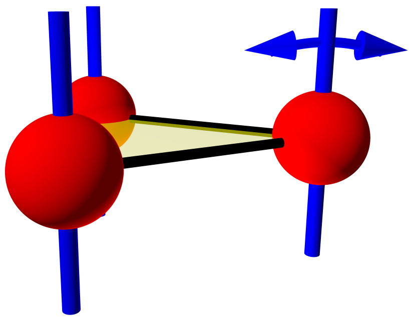
Calculate coupling coefficients from DFT (2,3,4)

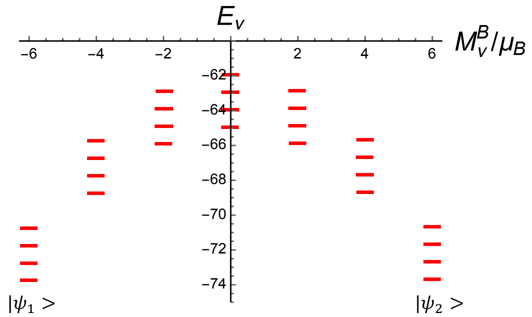
Perturbation picture: set up rate equations for phonon transitions between eigenstates of unperturbed spin Hamiltonian (2,3)

ADVANTAGE: many realistic phonons

(1) A. V. Postnikov, J. Kortus, and M. R. Pederson, physica status solidi (b) **243**, 2533 (2006).
(2) A. Lunghi and S. Sanvito, Science Advances **5**, eaax7163 (2019).
(3) A. Albino, S. Benci, L. Tesi, M. Atzori, R. Torre, S. Sanvito, R. Sessoli, and A. Lunghi, Inorg. Chem. **58**, 10260 (2019); ⇒ figure.
(4) D.A.S. Kaib, S. Biswas, K. Riedl, S.M. Winter, R. Valentí, Phys. Rev. B **103**, L140402 (2021).

Spin-phonon interaction – our question





Can phonons induce a tunnel splitting?

Know that non-collinear easy axes produce tunnel splitting

Set up special phonon modes that tilt easy axes in plane with C_3 axis out of uniaxial alignment

ADVANTAGE: quantum many-body solution for spins and phonons

$\Rightarrow$ correlated spin-phonon states:

$$\Psi_\nu = \sum c^{\nu}_{m_1,m_2,m_3,n_1,n_2,n_3} \,|\, m_1, m_2, m_3, n_1, n_2, n_3 \,\rangle$$

(1) K. Irländer and J. Schnack, Phys. Rev. B **102**, 054407 (2020).

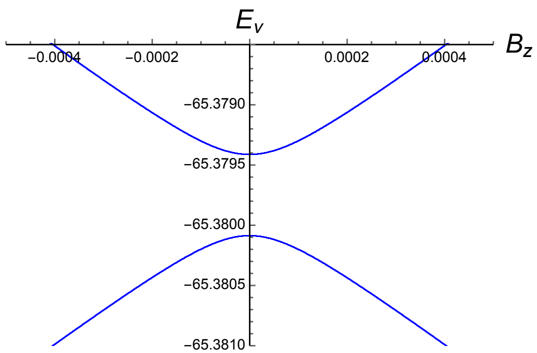
Spin-phonon interaction – Hamiltonian

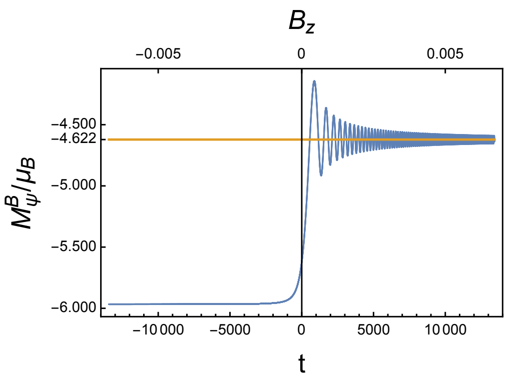
$$
\begin{aligned}
\underset{\sim}{H} &= -2J\left(\underset{\sim}{\vec{s}}_1\cdot\underset{\sim}{\vec{s}}_2+\underset{\sim}{\vec{s}}_2\cdot\underset{\sim}{\vec{s}}_3+\underset{\sim}{\vec{s}}_3\cdot\underset{\sim}{\vec{s}}_1\right)\\
&+\underset{\sim}{\vec{s}}_1\cdot\mathbf{D}_1(\underset{\sim}{\theta}_1)\cdot\underset{\sim}{\vec{s}}_1+\underset{\sim}{\vec{s}}_2\cdot\mathbf{D}_2(\underset{\sim}{\theta}_2)\cdot\underset{\sim}{\vec{s}}_2+\underset{\sim}{\vec{s}}_3\cdot\mathbf{D}_3(\underset{\sim}{\theta}_3)\cdot\underset{\sim}{\vec{s}}_3\\
&+\omega_1\left(\underset{\sim}{a}_1^\dagger\underset{\sim}{a}_1+\frac{1}{2}\right)+\omega_2\left(\underset{\sim}{a}_2^\dagger\underset{\sim}{a}_2+\frac{1}{2}\right)+\omega_3\left(\underset{\sim}{a}_3^\dagger\underset{\sim}{a}_3+\frac{1}{2}\right)\\
&+g\mu_B\cdot\vec{B}\cdot\left(\underset{\sim}{\vec{s}}_1+\underset{\sim}{\vec{s}}_2+\underset{\sim}{\vec{s}}_3\right)
\end{aligned}
$$

$$
\mathbf{D}_i(\underset{\sim}{\theta}_i) = D\,\vec{e}_i(\underset{\sim}{\theta}_i,\phi_i)\otimes\vec{e}_i(\underset{\sim}{\theta}_i,\phi_i)
$$

$$
\underset{\sim}{\theta}_i = \theta_{i,0}+\alpha\left(\underset{\sim}{a}_i^\dagger+\underset{\sim}{a}_i\right)\,, \qquad \theta_{i,0}=0,
$$
i.e., zero mean tilt

Spin-phonon interaction – our result (applies to integer spins)





Can phonons induce a tunnel splitting?

$\Rightarrow$ Yes, they can!

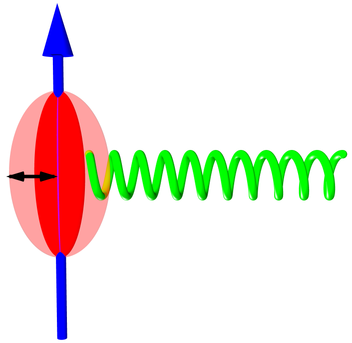
Ground state, practically, does not contain any phonons, nevertheless tunneling occurs. Coupling to zero-point motion suffices (2).

BAD NEWS: It is not enough to cool quantum devices, you have to prevent the coupling to disturbing sources at all.

Side remark: result probably already known in field of vibronic coupling (Atanasov, Shrivastava, Tsukerblat, Coronado).

(1) K. Irländer and J. Schnack, Phys. Rev. B **102**, 054407 (2020).
(2) F. Ortu *et al.*, Dalton Trans. **48**, 8541 (2019).

SUSY spin-phonon interaction (applies to integer spins)



$$\underset{\sim}{H} = D(\underset{\sim}{s}^z)^2 + E\left\{(\underset{\sim}{s}^x)^2 - (\underset{\sim}{s}^y)^2\right\} + g\mu_B B \underset{\sim}{s}^z + \underset{\sim}{H}_{\text{HO}}$$

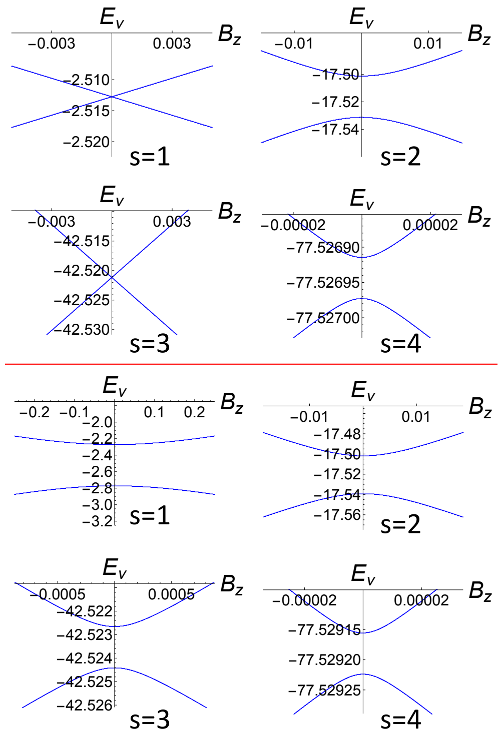
Special phonons that modify only:

L: $E = \alpha\left(\underset{\sim}{a}^\dagger + \underset{\sim}{a}\right)$ or Q: $E = \alpha\left(\underset{\sim}{a}^\dagger + \underset{\sim}{a}\right)^2$

L: tunneling gap for even s, no gap for odd s.

This is not Kramers, but related to another symmetry.

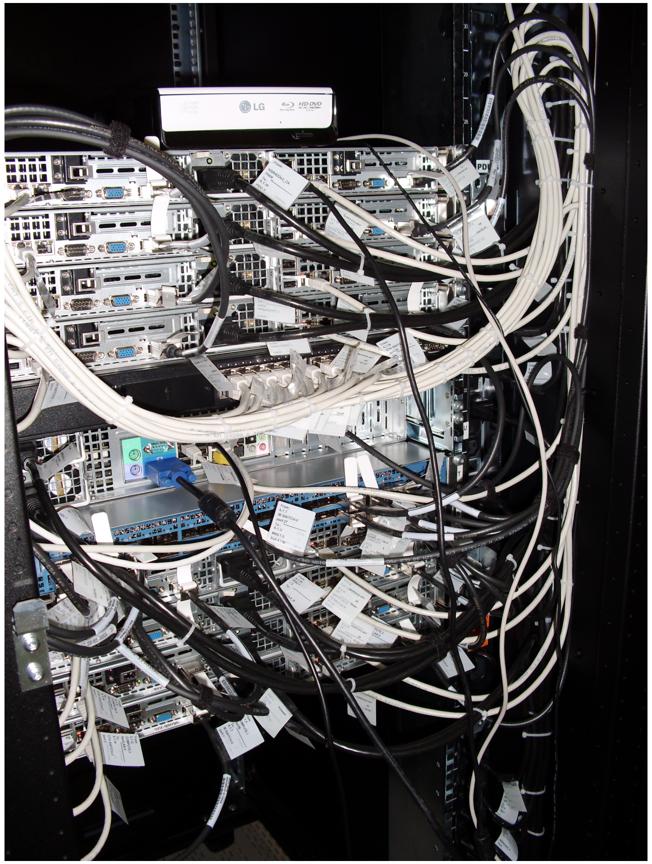
Q: tunneling gap for all s.



RESULT: very interesting behavior; there are some phonons that do not produce a tunneling gap thanks to the way they couple. SUSY at work.

(1) K. Irländer, H.-J. Schmidt, J. Schnack, Eur. Phys. J. B **94**, 68 (2021)

Summary



- Magnetic molecules for storage, q-bits, MCE, and since they are nice.
- SMM challenges: quantum tunneling and phonons
- Magnetism is much richer and more complicated than shown here. Talk focused on 3d ions with weak spin-orbit interaction.
- Typicality is a powerful approach.
- ED, HTE, CMC, QMC, FTLM, DMRG, DDMRG, thDMRG, DFT for magnetic molecules.

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Thank you very much for your attention.

The end.

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