Toroidal magnetic molecules stripped to their basics

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Irz

Imagine . . .

Imagine someone tells you that

toroidal magnetic molecules

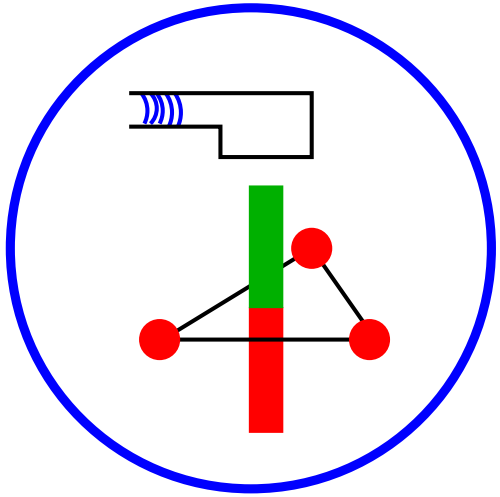
are superb building blocks
of quantum devices.

Would you buy one?

Or would you first check such molecules?

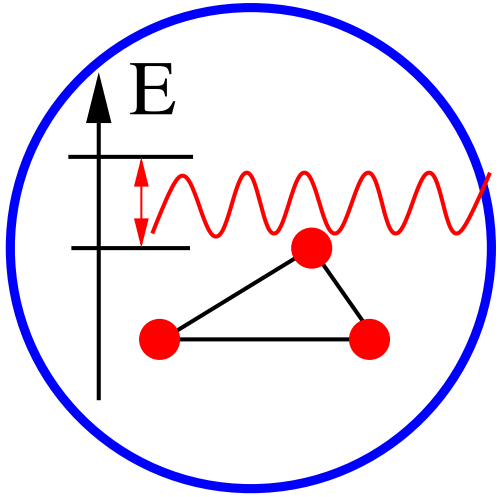
And if, what would you investigate?

Quantum devices – figures of merit



Memory unit

- requires bistability
- problem 1: quantum tunneling
- problem 2: stability against field fluctuations



Q-bit

- requires coherence
- problem decoherence

Yes, we can!

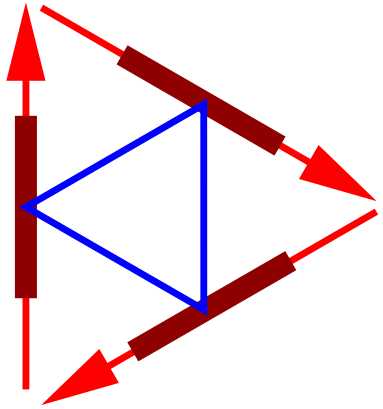
$$\begin{pmatrix} 3 & 42 & 4711 \\ 42 & 0 & 3.14 \\ 4711 & 3.14 & 8 \\ -17 & 007 & 13 \\ 1.8 & 15 & 081 \end{pmatrix}$$

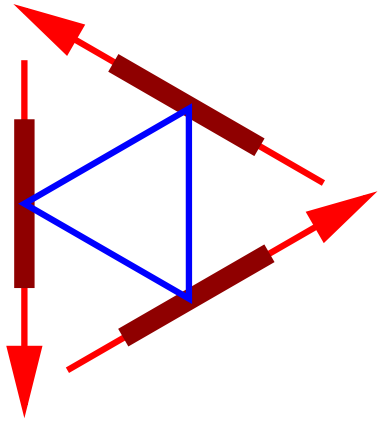
1. Toroidal magnetic molecules
2. Bistability, tunneling, and stability
3. Clock transitions and decoherence

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

Toroidal magnetic molecules

Torodial magnetic molecules I





Model Hamiltonian I

$$\underset{\sim}{H} = -2 \sum_{i<j} J_{ij} \vec{\underset{\sim}{s}}_i \cdot \vec{\underset{\sim}{s}}_j + D \sum_i \left(\vec{\underset{\sim}{s}}_i \cdot \vec{e}_i^{\,3} \right)^2 + \mu_B \, g \, \vec{B} \cdot \sum_i \vec{\underset{\sim}{s}}_i$$

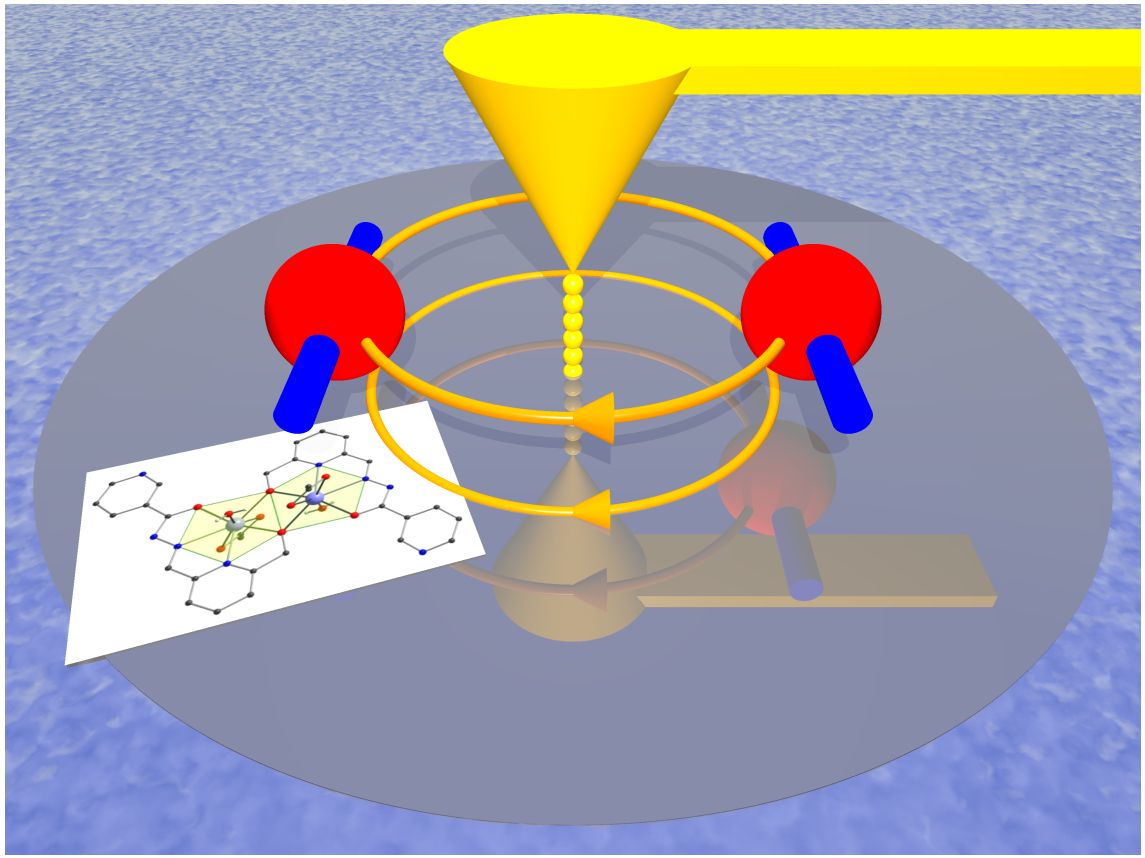
Toroidal magnetic moment

$$\vec{\underset{\sim}{\tau}} = \sum_i \vec{r}_i \times \vec{\underset{\sim}{s}}_i$$

Classical ground states with vanishing moment, but non-vanishing toroidal moment possible (easy axes $D < 0$ & weak exchange $|J_{ij}| \ll |D|$).

J. Tang, I. Hewitt, N. T. Madhu, G. Chastanet, W. Wernsdorfer, C. E. Anson, C. Benelli, R. Sessoli, and A. K. Powell, Angew. Chem. Int. Ed. **45**, 1729 (2006).
A. Soncini and L. F. Chibotaru, Phys. Rev. B **77**, 220406 (2008).
D. Pister, K. Irländer, D. Westerbeck, and J. Schnack, Phys. Rev. Research **4**, 033221 (2022).

Torodial magnetic molecules II – hypothetical switching

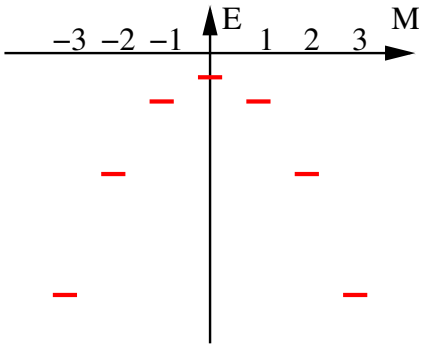


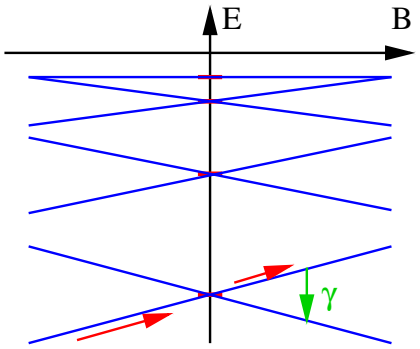
D. Pister, K. Irländer, D. Westerbeck, and J. Schnack, Phys. Rev. Research **4**, 033221 (2022).

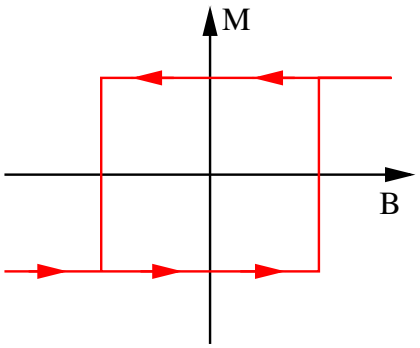
Bistability, tunneling, and stability against field fluctuations

(Remember what we know from SMMs!)

Single-ion anisotropy and bistability I – good SMM







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

$D_i < 0$ collinear easy axes

eigenvectors: $|\, M, \alpha \,\rangle$

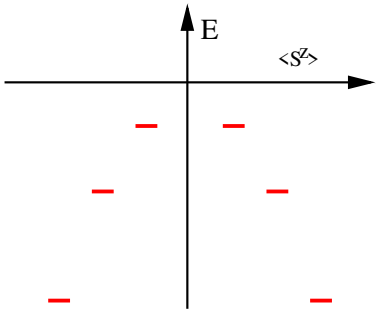
low-lying eigenvalues: $E_M = DM^2 + g\mu_B BM$

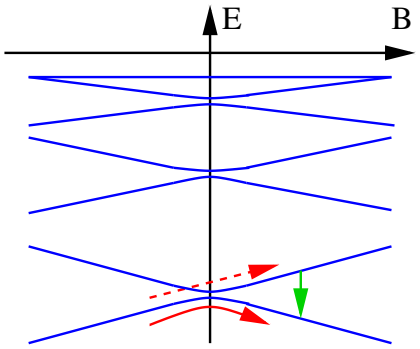
(strong exchange limit)

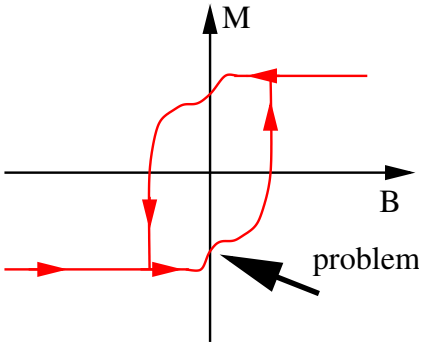
IMPORTANT: $[\underset{\sim}{H}, \underset{\sim}{S}^z] = 0$ since all D tensors aligned!!!

$\Rightarrow$ level crossings at $B = 0$
$\Rightarrow$ good hysteresis

Single-ion anisotropy and bistability II – bad/no SMM







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

$\mathbf{D}_i$ individual non-collinear anisotropy tensors

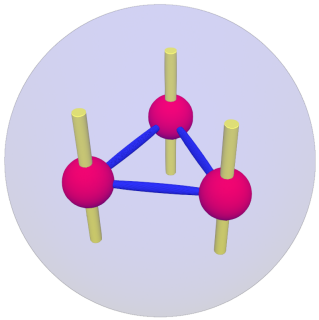
NO LONGER eigenvectors: $|\, M, \alpha \,\rangle$

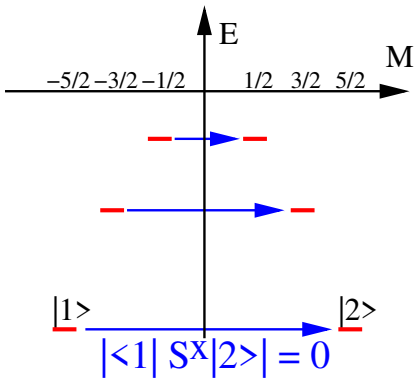
low-lying eigenvalues only approx. parabola (if at all)

IMPORTANT: $[\underset{\sim}{H}, \underset{\sim}{S}^z] \neq 0$

$\Rightarrow$ avoided level crossings at $B = 0$ for integer spins
$\Rightarrow$ poor/no hysteresis – not bistable & bad for storage

Single-ion anisotropy and bistability III – stability

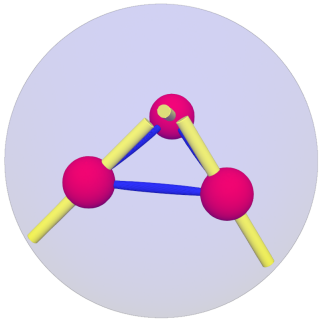


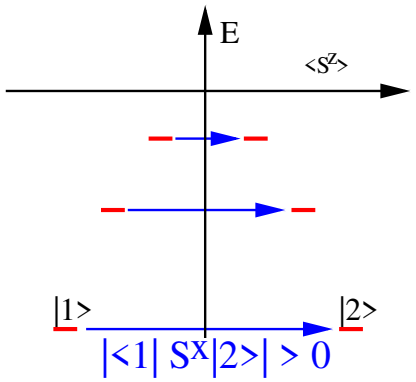


Collinear easy axes:

⇒ No tunneling gap

⇒ No transition matrix elements





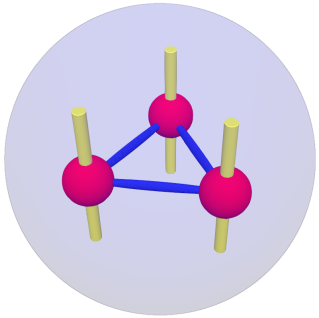
Non-collinear easy axes:

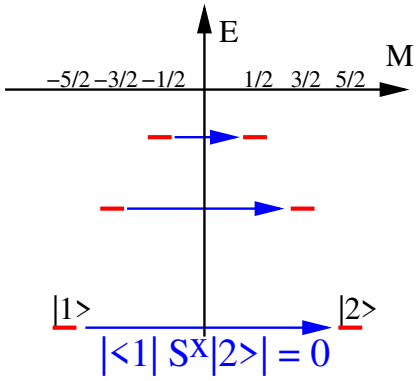
⇒ Tunneling gap for integer spin

⇒ (large) Transition matrix elements (1)

(1) K.-A. Lippert, C. Mukherjee, J.-P. Broschinski, Y. Lippert, S. Walleck, A. Stammler, H. Bögge, J. Schnack, and T. Glaser, Inorg. Chem. **56**, 15119 (2017).

Single-ion anisotropy and bistability IV – stability

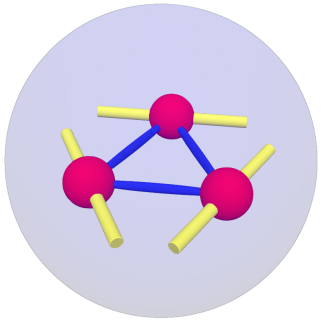


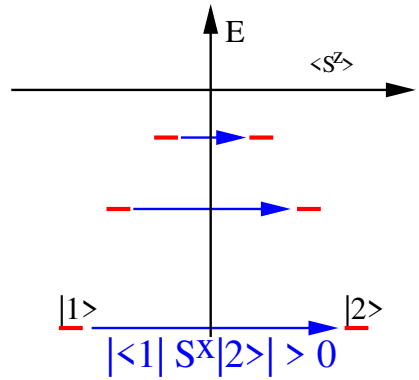


Collinear easy axes:

⇒ No tunneling gap

⇒ No transition matrix elements





Non-collinear easy axes:

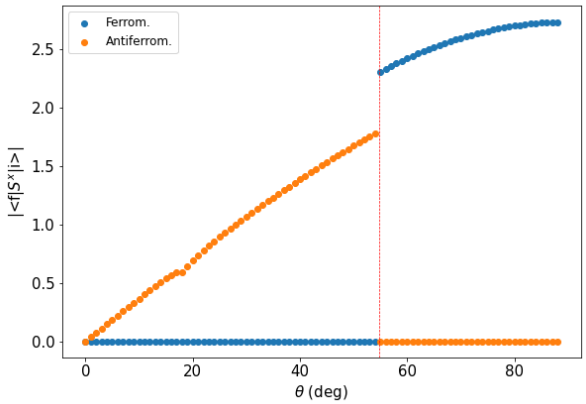
⇒ Tunneling gap for integer spin

⇒ (large) Transition matrix elements

Toroidal moments are here!

Toroidal magnetic moments – There is hope!

Trimer s=2.5



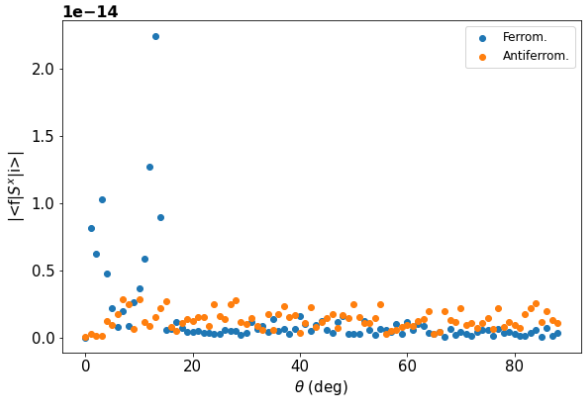
For gapped systems:

(1) Gap shrinks with increasing anisotropy!

(2) Gap shrinks with increasing spin!

(for all systems we investigated so far)

Hexagon s=1.5



Transition matrix element:

(1) Vanishes for certain canting angles ($N = 2, 3, 4$)!

(2) Vanishes completely ($N = 6$)!

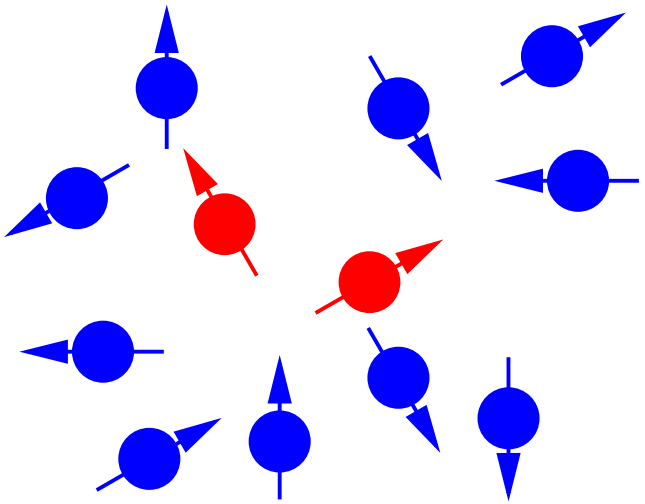
(work in progress)

$\Rightarrow$ Larger rings ($N > 4$) with larger spins might be preferential.

Master Theses of Daniel Pister and Jonas Waltenberg, Bielefeld University

Decoherence of (toroidal) clock transitions

Context



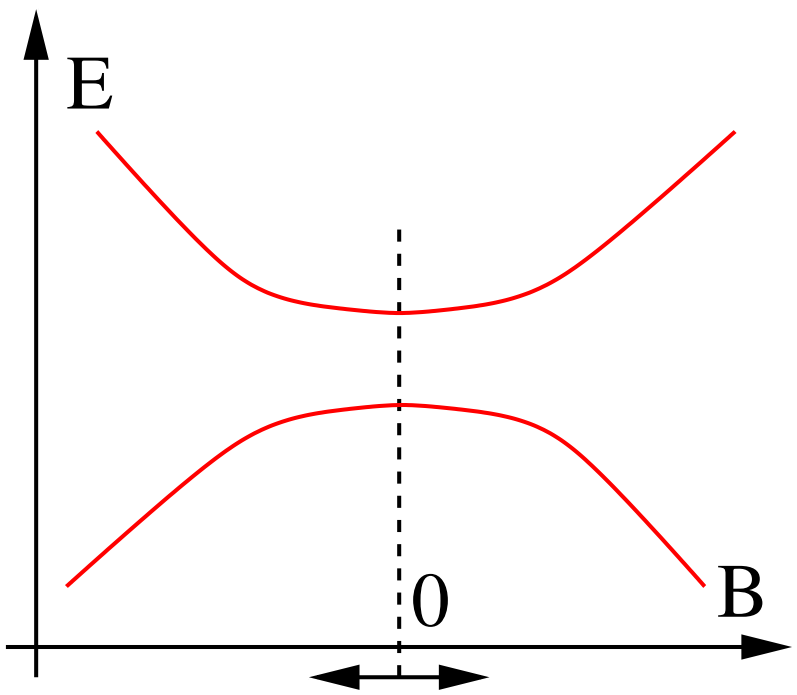
Investigation of decoherence of a subsystem if the combined system (including bath) is evolved via the time-dependent Schrödinger equation.

Employed measure of decoherence: reduced density matrix

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

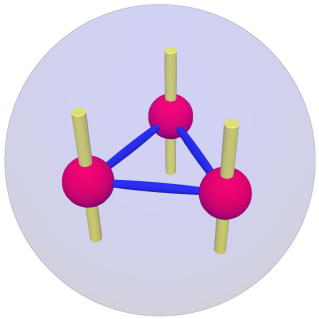
Typicality: unitary-time evolution of pure state approximates dynamics of density matrix.

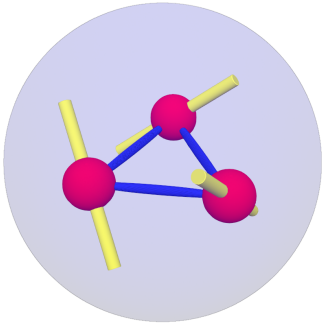
Concept of clock transitions

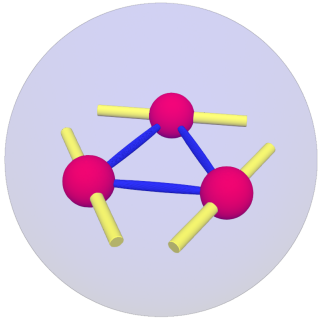


Fluctuations of B produce little effect on dynamics of superposition since ΔE of clock transition is independent of field at $B = 0$, at least to some order of a Taylor expansion.

Clock transitions with toroidal magnetic molecules







Model Hamiltonian II

$$\underset{\sim}{H} = -2 \sum_{i<j} J_{ij} \underset{\sim}{\vec{s}}_i \cdot \underset{\sim}{\vec{s}}_j + D \sum_i \left(\underset{\sim}{\vec{s}}_i \cdot \vec{e}_i^{\,3} \right)^2 + \mu_B \, g \, \vec{B} \cdot \sum_i \underset{\sim}{\vec{s}}_i + \underset{\sim}{H}_{\text{int}} + \underset{\sim}{H}_{\text{bath}}$$

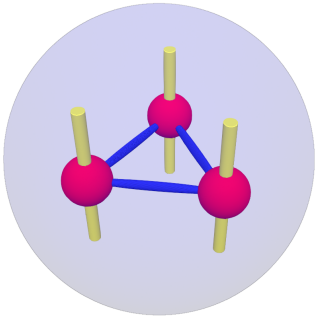
Reasonable parameters: weak J, strong D.
Dipolar interactions with and among $8 \ldots 10$ bath spins.

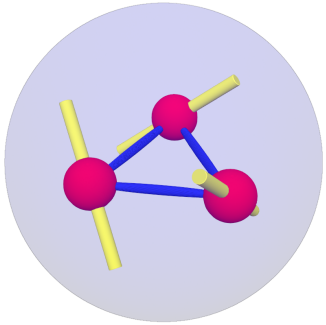
Investigation as function of tilt angle

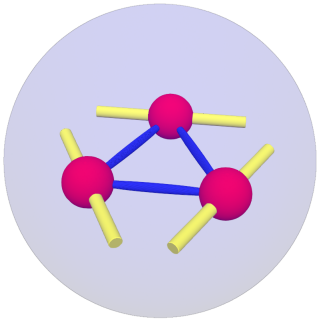
- various clock transitions of the spectrum,
- various arrangements of the decohering bath.

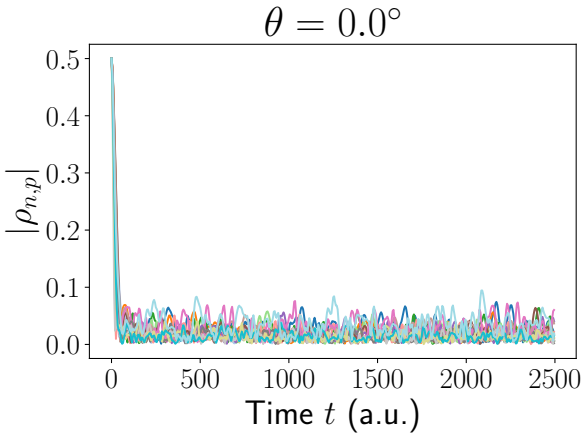
K. Irländer, J. Schnack, Phys. Rev. Research **5**, 013192 (2023).

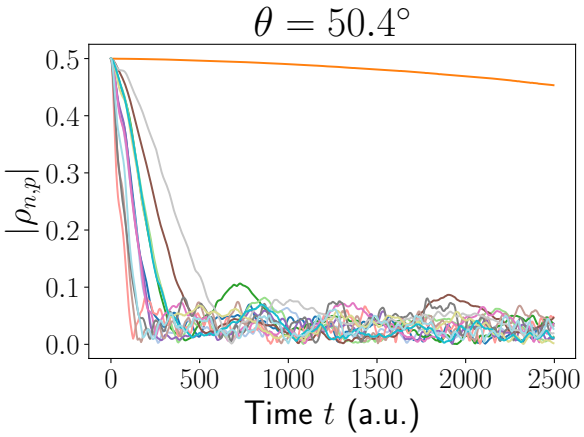
Clock transitions with toroidal magnetic molecules

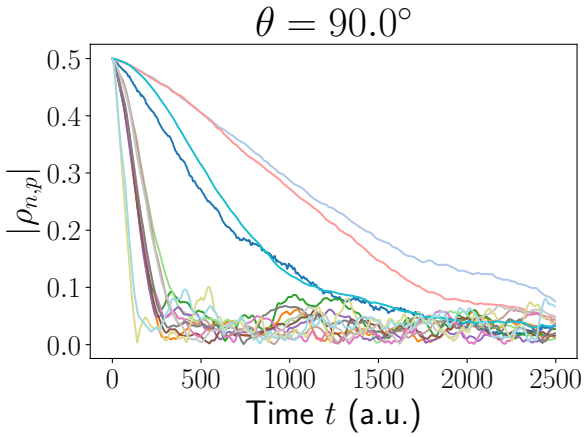






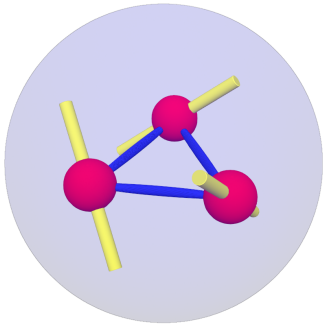


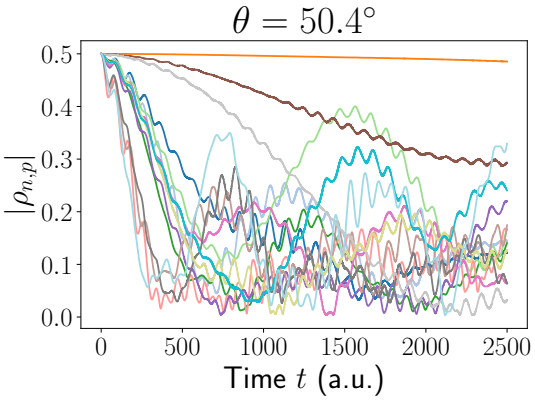


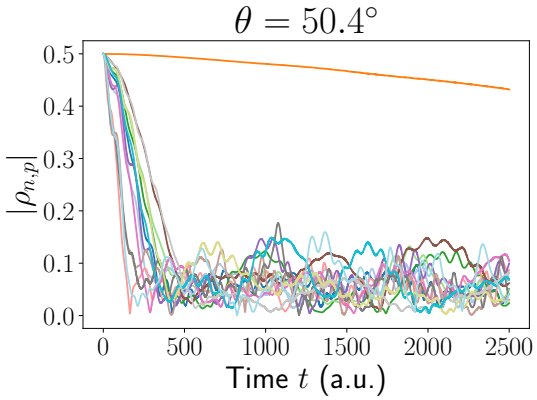


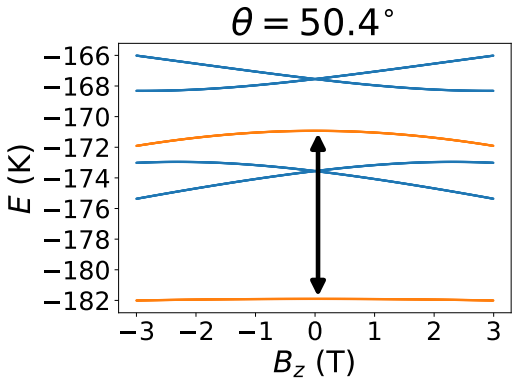
K. Irländer, J. Schnack, Phys. Rev. Research **5**, 013192 (2023).

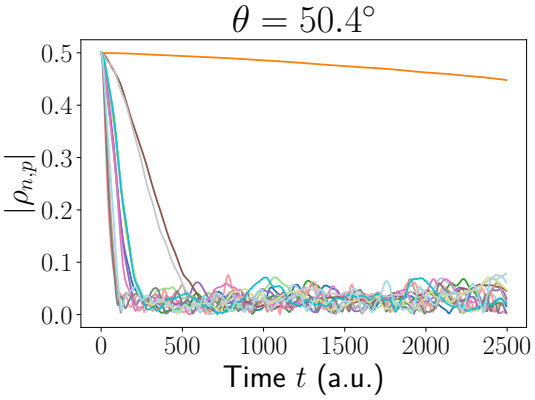
Clock transitions with toroidal magnetic molecules

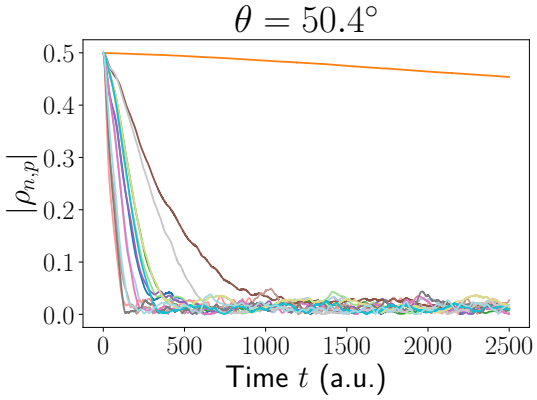








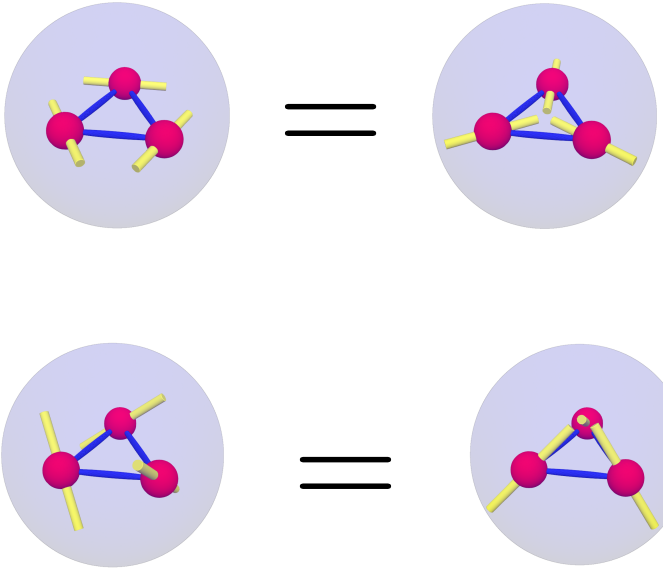




Decoherence as function of size of the bath (4, 6, 8, 10).

K. Irländer, J. Schnack, Phys. Rev. Research **5**, 013192 (2023).

Decoherence of toroidal magnetic molecules

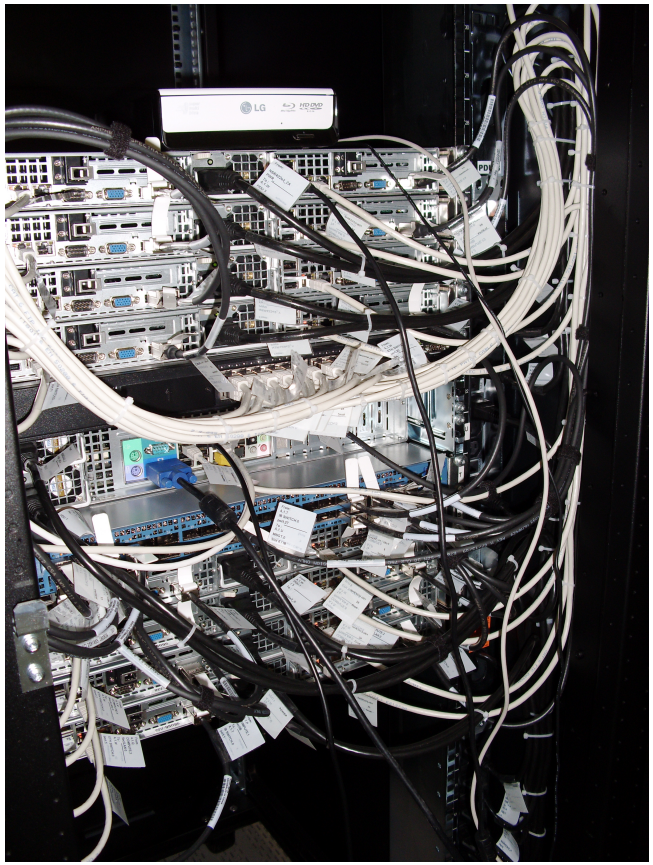


- Toroidal structure irrelevant, i.e. not correlated with desired properties (for Heisenberg interactions and non-collinear easy axes).
- Canted, near orthogonal anisotropy axes optimal in our example, i.e., they show longest coherence.
- Dipolar interactions between system spins do not alter the picture.

D. Pister, K. Irländer, D. Westerbeck, and J. Schnack, Phys. Rev. Research **4**, 033221 (2022).

K. Irländer, J. Schnack, Phys. Rev. Research **5**, 013192 (2023).

Summary ⇒ To-Do-List



- Toroidal magnetic molecules: perspectives not clear (to me).
- Relaxation measurements needed!
- Decoherence measurements needed!
- More theory needed to e.g. assess the influence of anisotropic interactions.

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, J. Waltenberg, D. Westerbeck (Bielefeld)
- K. Bärwinkel, T. Heitmann, R. Heveling, H.-J. Schmidt, R. Steinigeweg (Osnabrück)
- M. Luban (Ames Lab); 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 Cergy-Pontoise); E. Garlatti, S. Carretta, G. Amoretti, P. Santini (Parma); A. Tennant (ORNL); Gopalan Rajaraman (Mumbai); M. Affronte (Modena)
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The end.

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