Symmetric finite-temperature Lanczos method for anisotropic magnetic molecules

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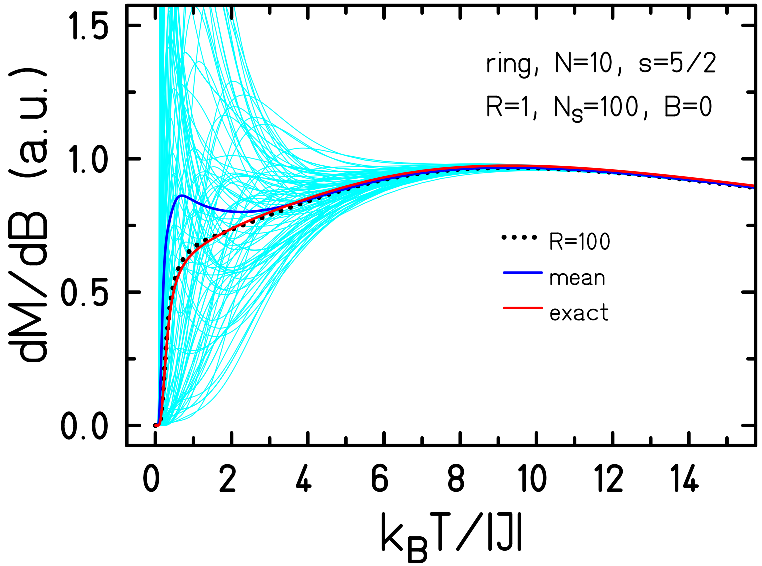
Finite-temperature Lanczos method in one slide

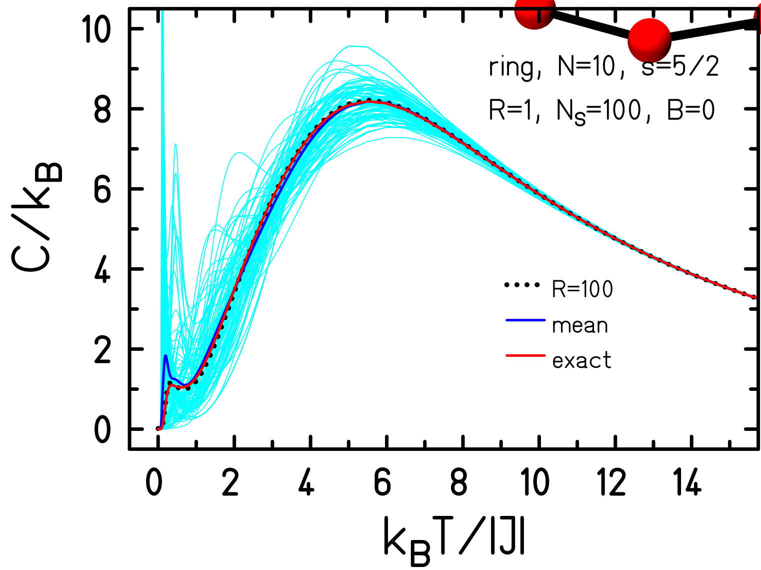
$$Z(T,B) \approx \langle\, r \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle \approx \sum_{n=1}^{N_L} e^{-\beta\epsilon_n^{(r)}} |\langle\, n(r) \,|\, r \,\rangle|^2$$

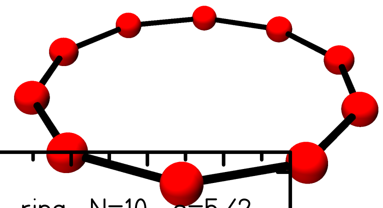
$$O^{\mathrm{r}}(T,B) \approx \frac{\langle\, r \,|\, \underset{\sim}{O} e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle}{\langle\, r \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r \,\rangle}$$

- $|\, r \,\rangle$ is a random vector. Any random vector will do: $|\, r \,\rangle \equiv (T = \infty)$
- $e^{-\beta \underset{\sim}{H}} = \sum_{n=1}^{N_L} |\, n(r) \,\rangle e^{-\beta\epsilon_n^{(r)}} \langle\, n(r) \,|$ is the spectral representation in the Krylov space of dimension N_L grown from seed $|\, r \,\rangle$.

(1) J. Jaklic and P. Prelovsek, Phys. Rev. B **49**, 5065 (1994).
(2) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020).





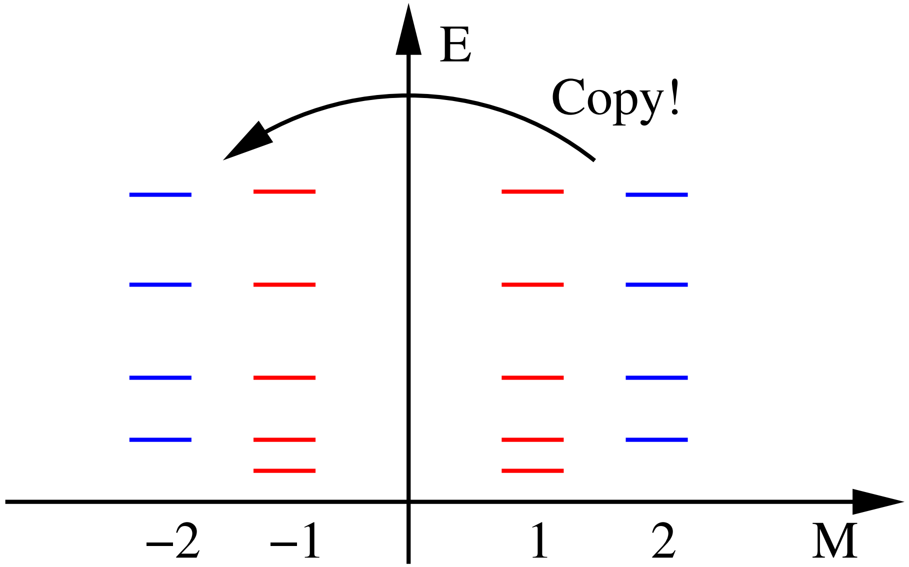


$$O^{\text{FTLM}}(T,B) \approx \frac{\langle\, r_1 \,|\, \underset{\sim}{Q} e^{-\beta \underset{\sim}{H}} \,|\, r_1 \,\rangle + \langle\, r_2 \,|\, \underset{\sim}{Q} e^{-\beta \underset{\sim}{H}} \,|\, r_2 \,\rangle + \cdots}{\langle\, r_1 \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r_1 \,\rangle + \langle\, r_2 \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r_2 \,\rangle + \cdots} \qquad \text{(correct)}$$

$$O^{\text{mean}}(T,B) \approx \frac{1}{R}\left(\frac{\langle\, r_1 \,|\, \underset{\sim}{Q} e^{-\beta \underset{\sim}{H}} \,|\, r_1 \,\rangle}{\langle\, r_1 \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r_1 \,\rangle} + \frac{\langle\, r_2 \,|\, \underset{\sim}{Q} e^{-\beta \underset{\sim}{H}} \,|\, r_2 \,\rangle}{\langle\, r_2 \,|\, e^{-\beta \underset{\sim}{H}} \,|\, r_2 \,\rangle} + \cdots \right. \qquad \text{(wrong)}$$

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

Why is FTLM so good?



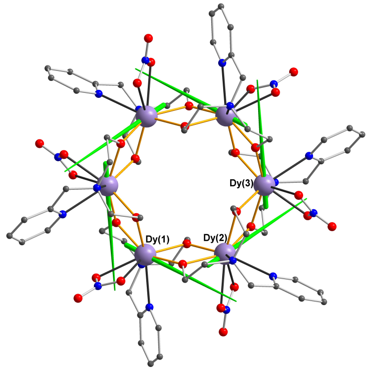
$\underset{\sim}{S}^z$–symmetry is used, spectra are generated in orthogonal subspaces $\mathcal{H}(M \geq 0)$

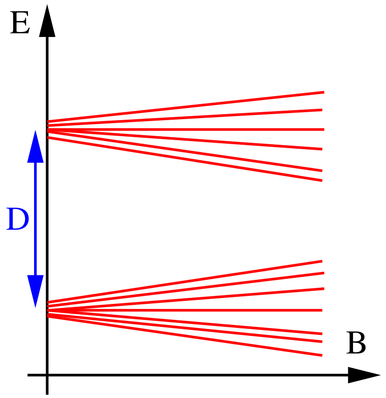
spectra for subspaces $\mathcal{H}(M < 0)$ are taken as copies

What would happen if you would generate the spectra for subspaces $\mathcal{H}(M)$ and $\mathcal{H}(-M)$ in independent random processes?

Effective model for Dy_6

$$\underset{\sim}{H} = \sum_{k<\ell} \underset{\sim}{\vec{j}}_k \cdot \mathbf{J}_{k\ell} \cdot \underset{\sim}{\vec{j}}_\ell + \sum_k \underset{\sim}{\vec{j}}_k \cdot \mathbf{D}_k \cdot \underset{\sim}{\vec{j}}_k + \mu_B \vec{B} \cdot \sum_k g_k \underset{\sim}{\vec{j}}_k$$

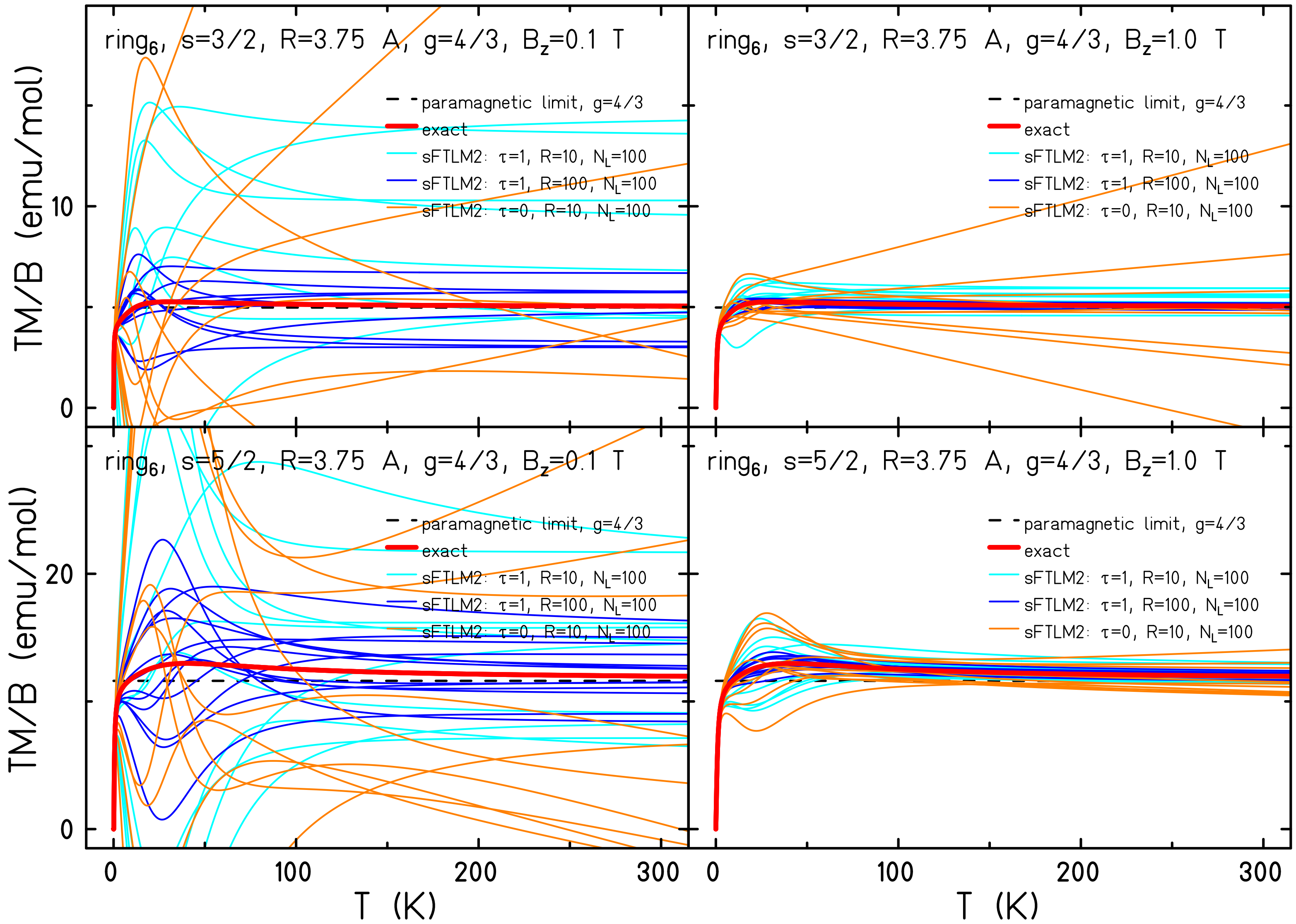




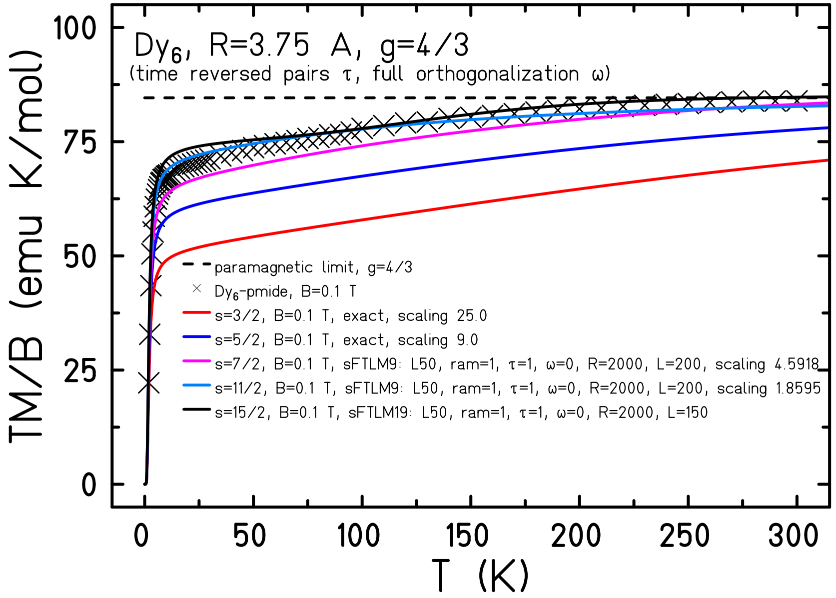
- Very strong alternating easy axes with $D \approx -20$ K. $J \approx -0.02$ K and (stronger) dipolar interaction.
- Hamiltonian has no symmetries! Anisotropy is dominant.
- dim$\mathcal{H} = 16,777,216 \quad \Rightarrow \quad$ FTLM!

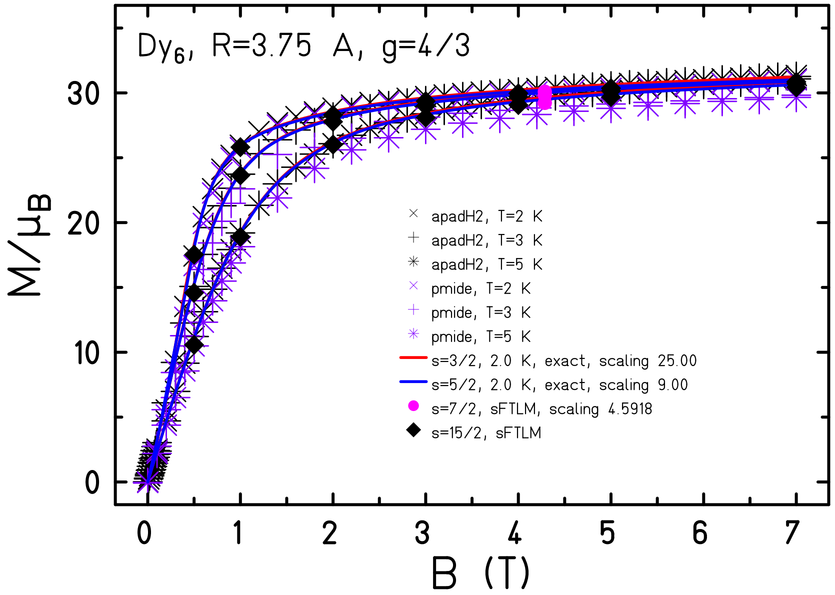
Warning! Method is approximate and holds only for small enough B since spin and orbital angular momentum have got different g_k.

Problem – FTLM converges badly for anisotropic models



Dy_6 – results





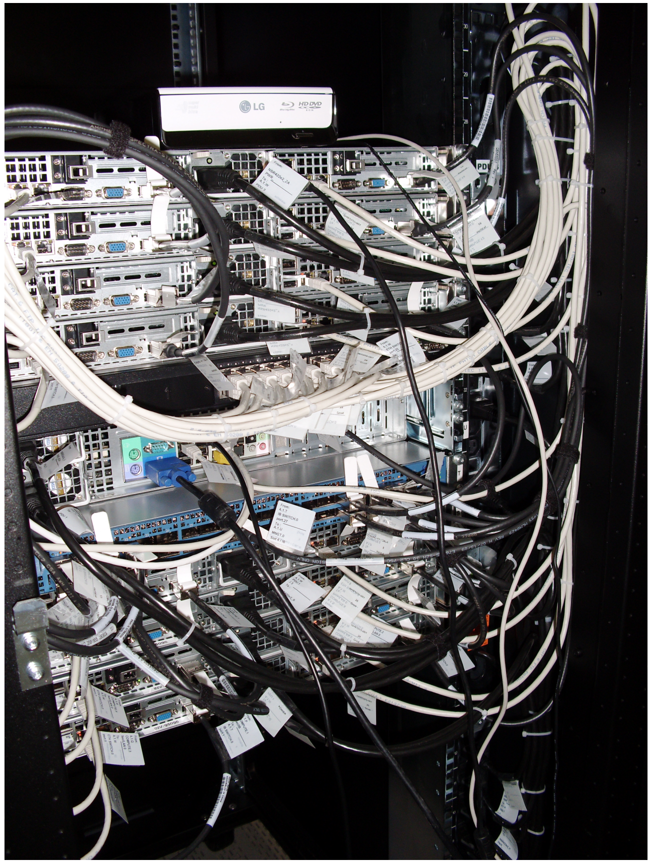
1. Use pairs of time-reversed random vectors (1)

2. Use symmetric version of FTLM (2):

$$\text{Tr}\left(\underset{\sim}{Q} e^{-\beta \underset{\sim}{H}}\right) = \text{Tr}\left(e^{-\beta \underset{\sim}{H}/2} \underset{\sim}{Q} e^{-\beta \underset{\sim}{H}/2}\right) \approx \langle\, r \,|\, e^{-\beta \underset{\sim}{H}/2} \underset{\sim}{Q} e^{-\beta \underset{\sim}{H}/2} \,|\, r \,\rangle \neq \langle\, r \,|\, e^{-\beta \underset{\sim}{H}} \underset{\sim}{Q} \,|\, r \,\rangle$$

(1) O. Hanebaum, J. Schnack, Eur. Phys. J. B **87**, 194 (2014).
(2) M. Aichhorn, M. Daghofer, H. G. Evertz, and W. von der Linden, Phys. Rev. B **67**, 161103(R) (2003).
(3) D. Westerbeck, Ph.D. thesis, Bielefeld University (2025).

Summary + To-Do-List



- Spin models are always approximate in case of strong spin-orbit interaction.
  Actually, they are always approximate (cf. e.g. Oliver Waldmann).

- Multicenter j-spin models allow accurate modelling and effective understanding of various aspects of Dy_6.

- How far does this description hold?

- FTLM for anisotropic models has to be further improved to converge with less random vectors.

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