Challenges with FTLM

Jürgen Schnack

Department of Physics – University of Bielefeld – Germany

http://obelix.physik.uni-bielefeld.de/∼schnack/

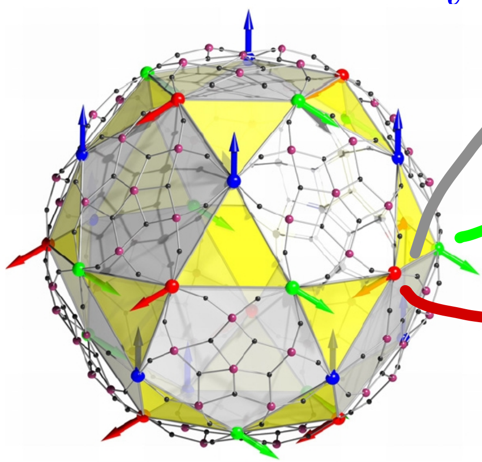
Jülich HPC flash seminar
FZ Jülich, Germany, 9 October 2025

Let’s calculate something!

You have got an idea about the modeling!

Heisenberg Zeeman

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



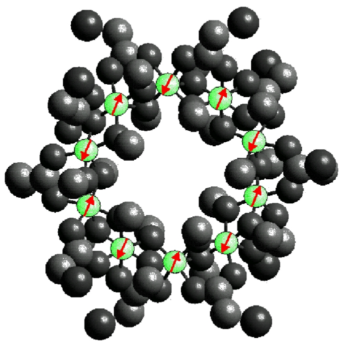
You have to solve the Schrödinger equation!

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

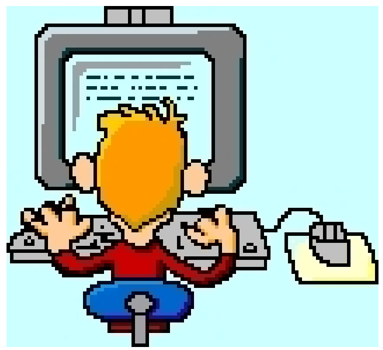
Eigenvalues E_n and eigenvectors $| \, \phi_n \, \rangle$

- 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$^{\text{III}}_{10}$: $N = 10$, $s = 5/2$, 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!

Solution I: trace estimators

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

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

- $|\, \nu \,\rangle$ 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

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

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

- 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).

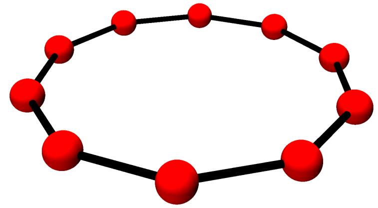
Partition function II: Finite-temperature Lanczos Method

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

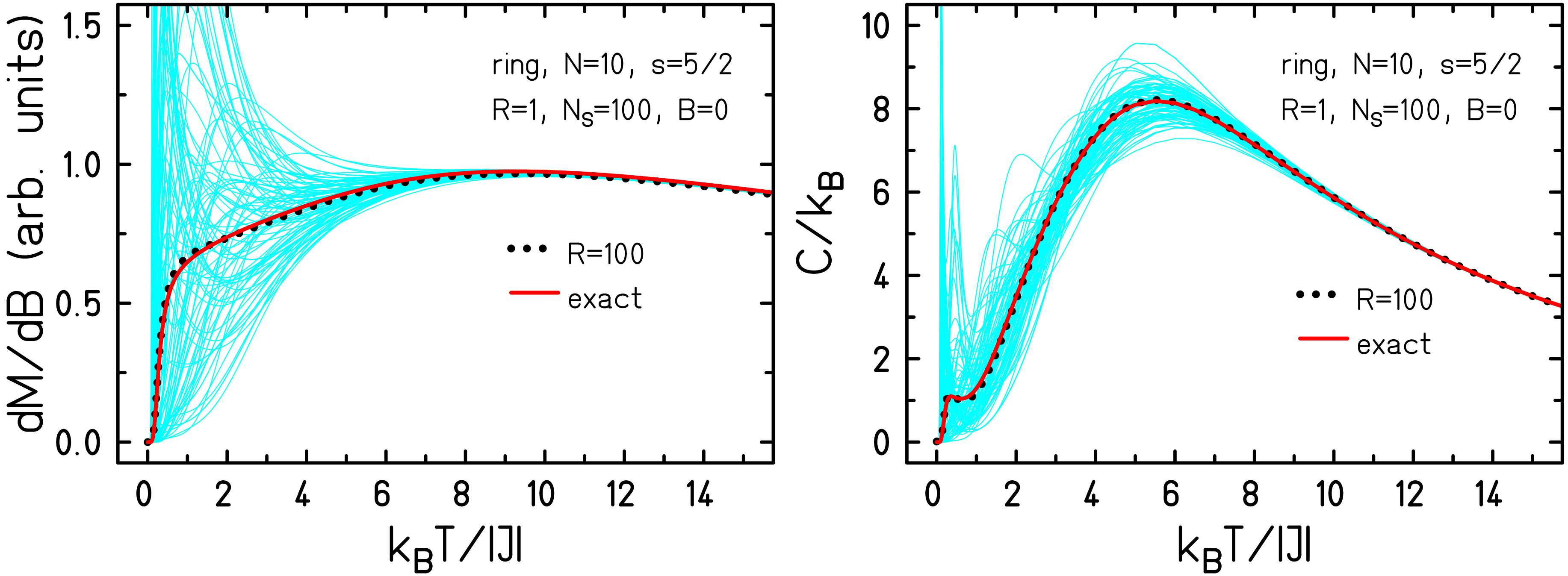
- Averaging over R random vectors is better.
- $|\, n(r) \,\rangle$ n-th Lanczos eigenvector starting from $|\, r \,\rangle$ (now normalized).
- Partition function replaced by a small sum: $R = 1 \dots 100$, $N_L \approx 100$.
- Use symmetries! Copy Hilbert subspaces!

$$\text{Tr}\left(\underset{\sim}{S}^z e^{-\beta \underset{\sim}{H}}\right) \approx \sum_{M=M_{\text{min}}}^{M_{\text{max}}} \frac{\text{dim}(\mathcal{H}(M))}{R} \sum_{r=1}^{R} \sum_{n=1}^{N_L} M e^{-\beta\epsilon_n^{(M,r)}} |\langle\, n(M,r) \,|\, M, r \,\rangle|^2$$

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



FTLM 1: ferric wheel

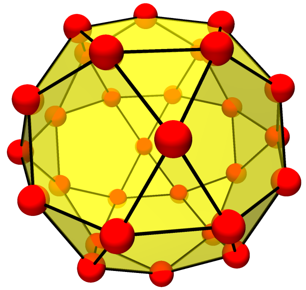


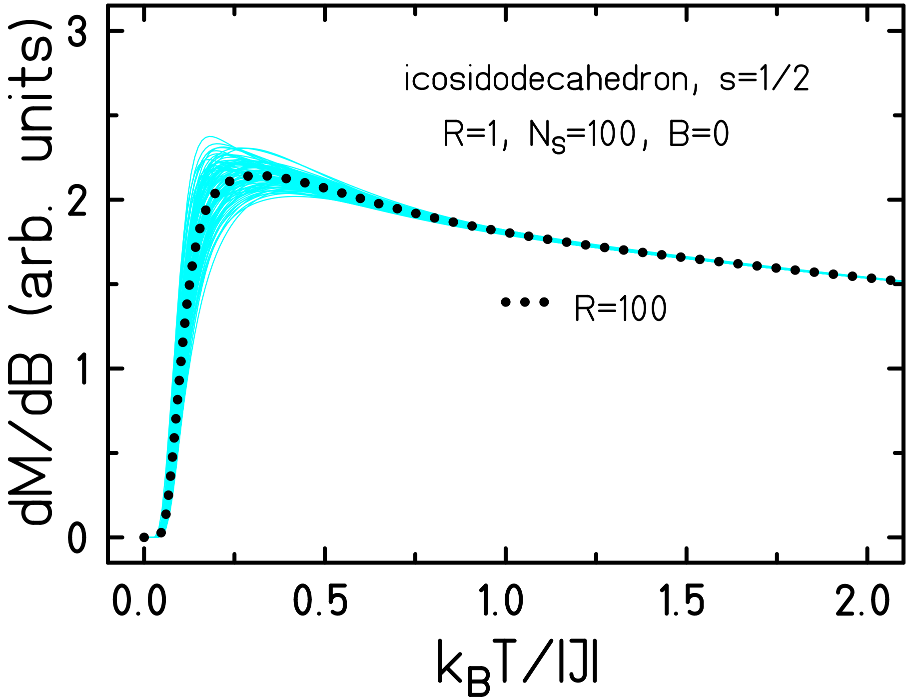
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020).

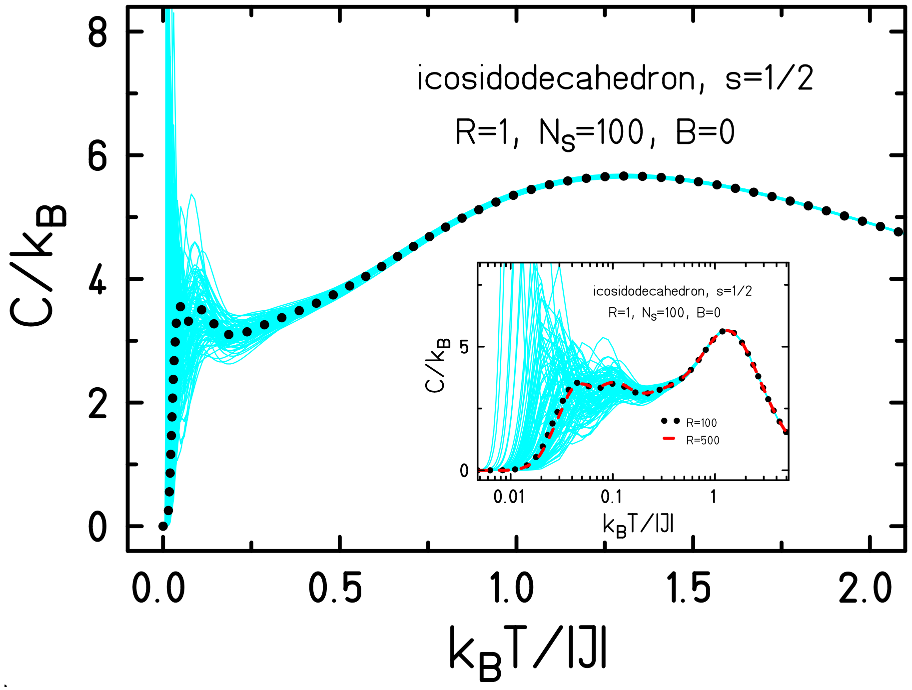
(2) SU(2) & D_2: R. Schnalle and J. Schnack, Int. Rev. Phys. Chem. **29**, 403 (2010).

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

FTLM 2: icosidodecahedron



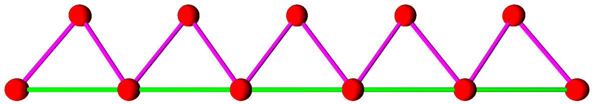


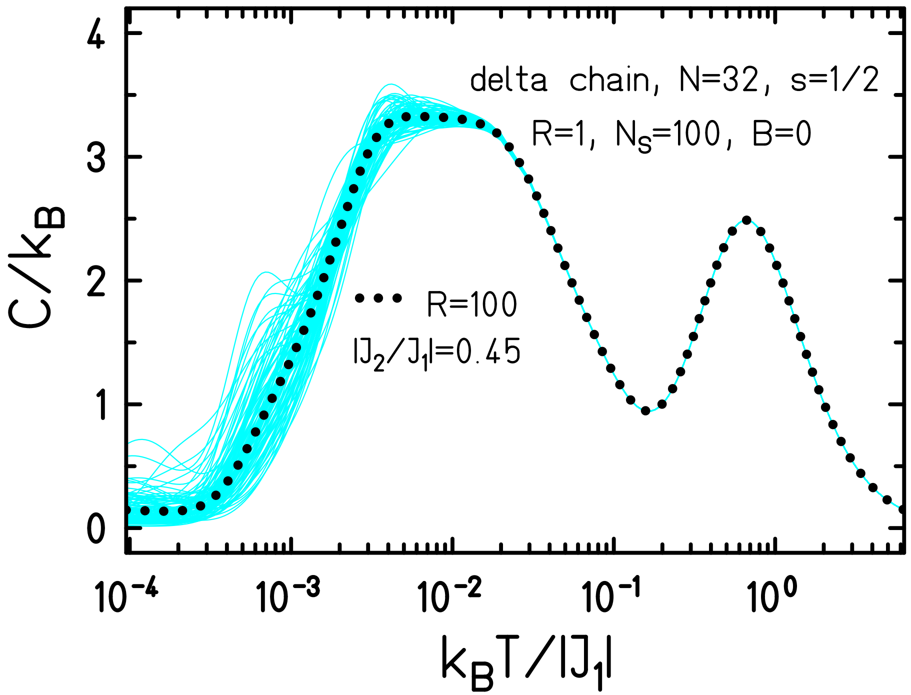


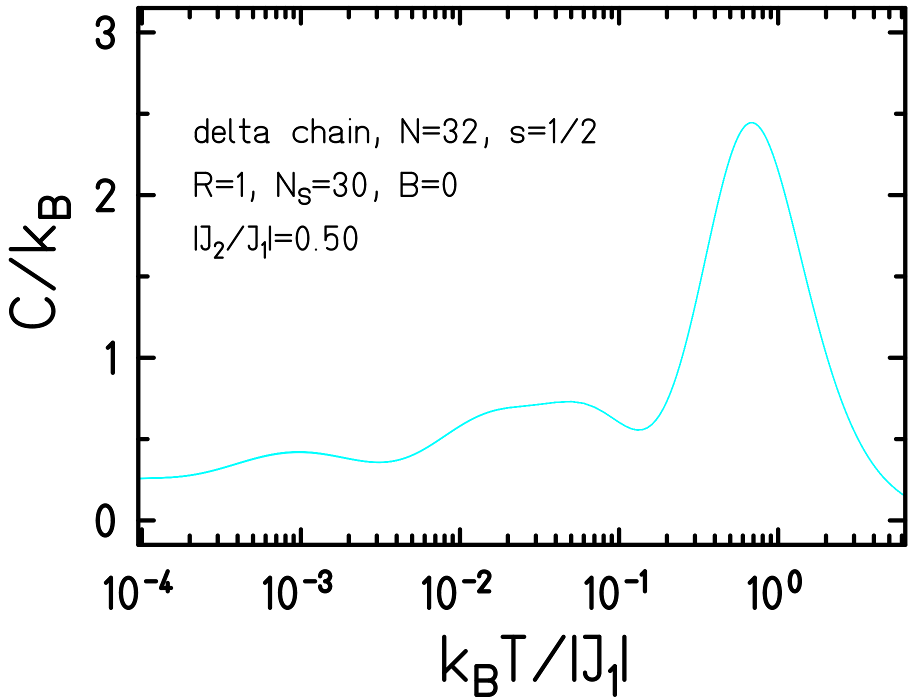
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020).

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

FTLM 3: sawtooth chain







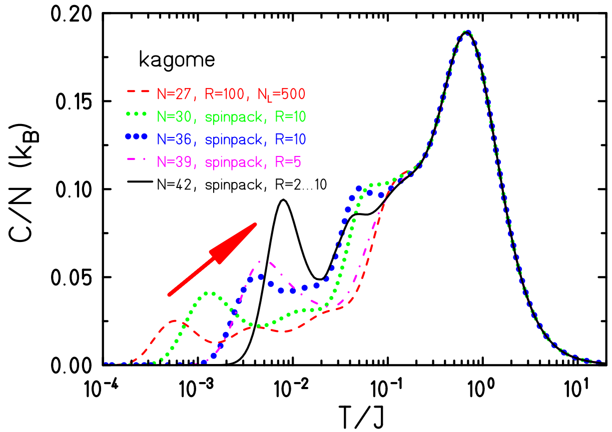
$|J_2/J_1| = 0.45$ – near critical, $|J_2/J_1| = 0.50$ – critical.

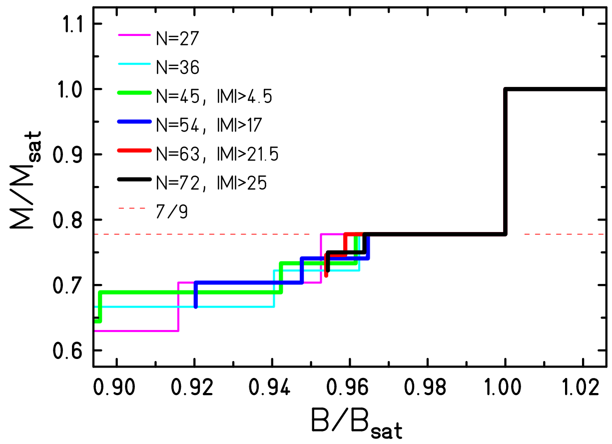
Frustration, technically speaking, works in your favour.

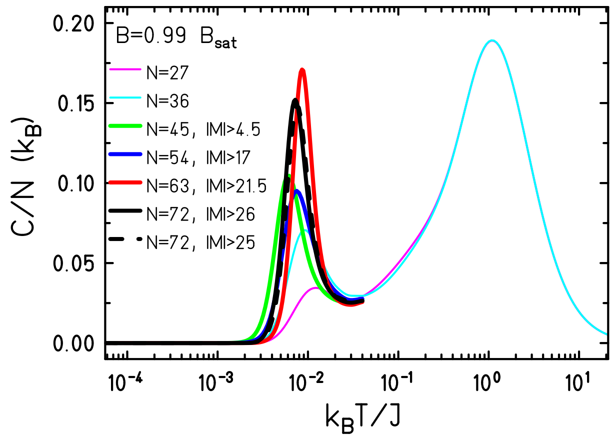
(1) J. Schnack, J. Richter, R. Steinigeweg, Phys. Rev. Research **2**, 013186 (2020)

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

FTLM 4: kagome (using spinpack)





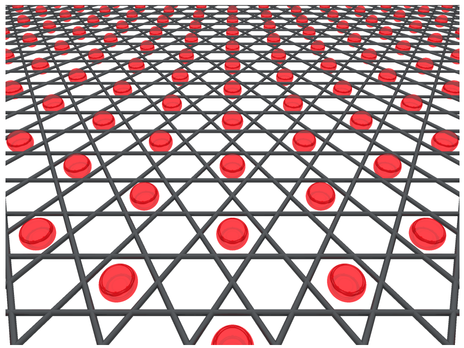


Specific heat of kagome with $N = 42$ – role of low-lying singlets

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

Magnon crystalization at high field.

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



. . . and many more results with Johannes Richter.

Anisotropic magnetic molecules

Symmetries will disappear!

Finite-temperature Lanczos Method III

$$\underset{\sim}{H} = \sum_{i<j} \underset{\sim}{\vec{s}}_i \cdot \mathbf{J}_{ij} \cdot \underset{\sim}{\vec{s}}_j + \sum_i \underset{\sim}{\vec{s}}_i \cdot \mathbf{D}_i \cdot \underset{\sim}{\vec{s}}_i + \mu_B B \sum_i g_i \underset{\sim}{s}_i^z$$

- Problem: for anisotropic Hamiltonians likely no symmetry left $\rightarrow$ accuracy drops (esp. for high T).
- Simple traces such as Tr $\left(\underset{\sim}{S}^z\right) = 0$ tend to be wrong for R not very big. More general, the magnetization vs temperature at small fields converges only very poorly.
- Why did this not happen before?

O. Hanebaum, J. Schnack, Eur. Phys. J. B **87**, 194 (2014)

Finite-temperature Lanczos Method IV

Employ very general symmetry (time-reversal invariance)

$$\vec{\mathcal{M}}(T,-\vec{B}) \;=\; -\vec{\mathcal{M}}(T,\vec{B})$$

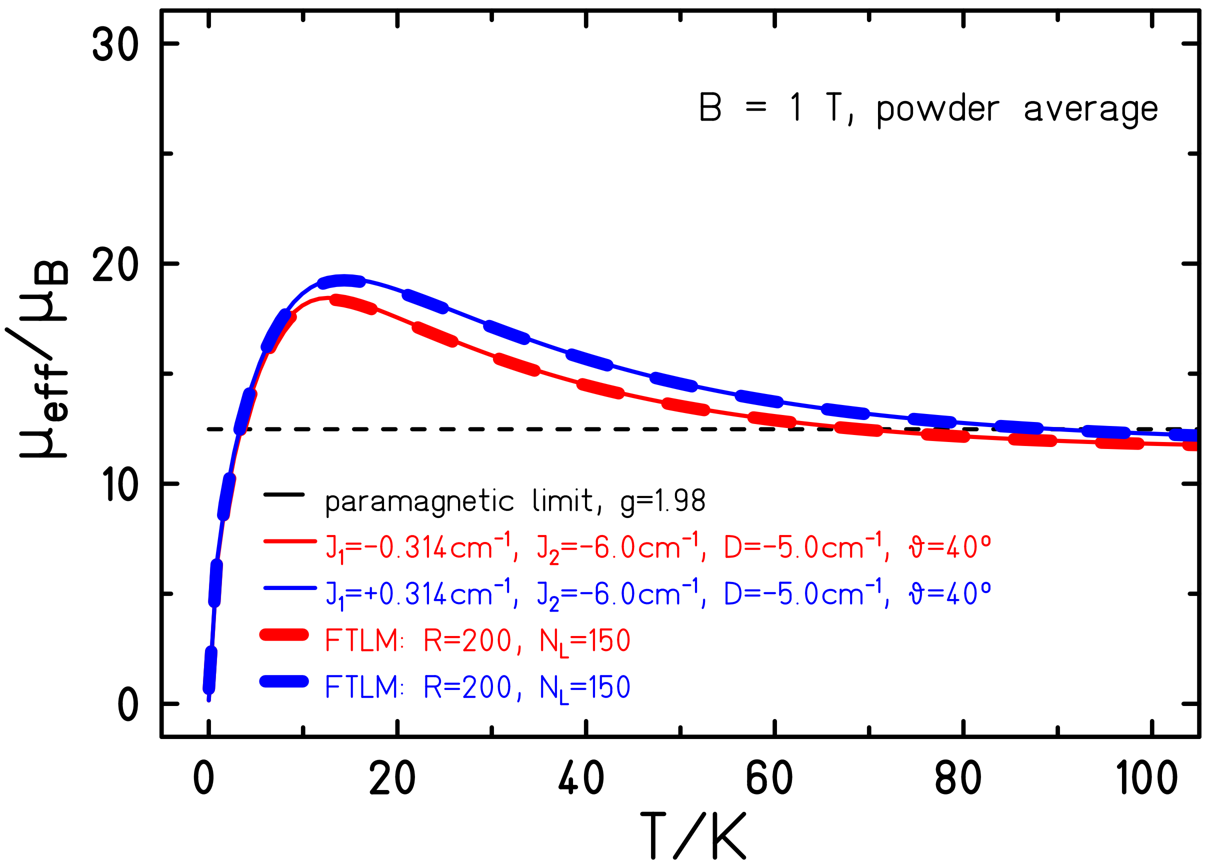
Use Lanczos energy eigenvector $|\,n(\nu)\,\rangle$ and time-reversed counterpart $|\,\tilde{n}(\nu)\,\rangle$

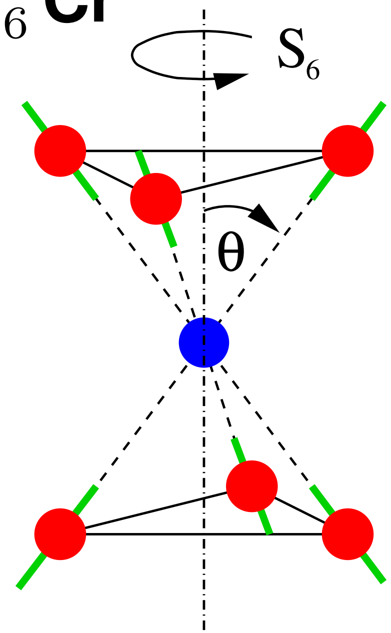
$$|\,n(\nu)\,\rangle = \sum_{\vec{m}} c_{\vec{m}}\,|\,\vec{m}\,\rangle \qquad , \qquad |\,\tilde{n}(\nu)\,\rangle = \sum_{\vec{m}} c^{*}_{\vec{m}}\,|\,-\vec{m}\,\rangle$$

- Restores $\vec{\mathcal{M}}(T,-\vec{B}) = -\vec{\mathcal{M}}(T,\vec{B})$ and (some) traces.
- More practical: use pairs of time-reversed random vectors; still accurate.

O. Hanebaum, J. Schnack, Eur. Phys. J. B **87**, 194 (2014)

Glaser-type molecules: $Mn_6^{III}Cr^{III}$





$s = 2$, $s = 3/2$

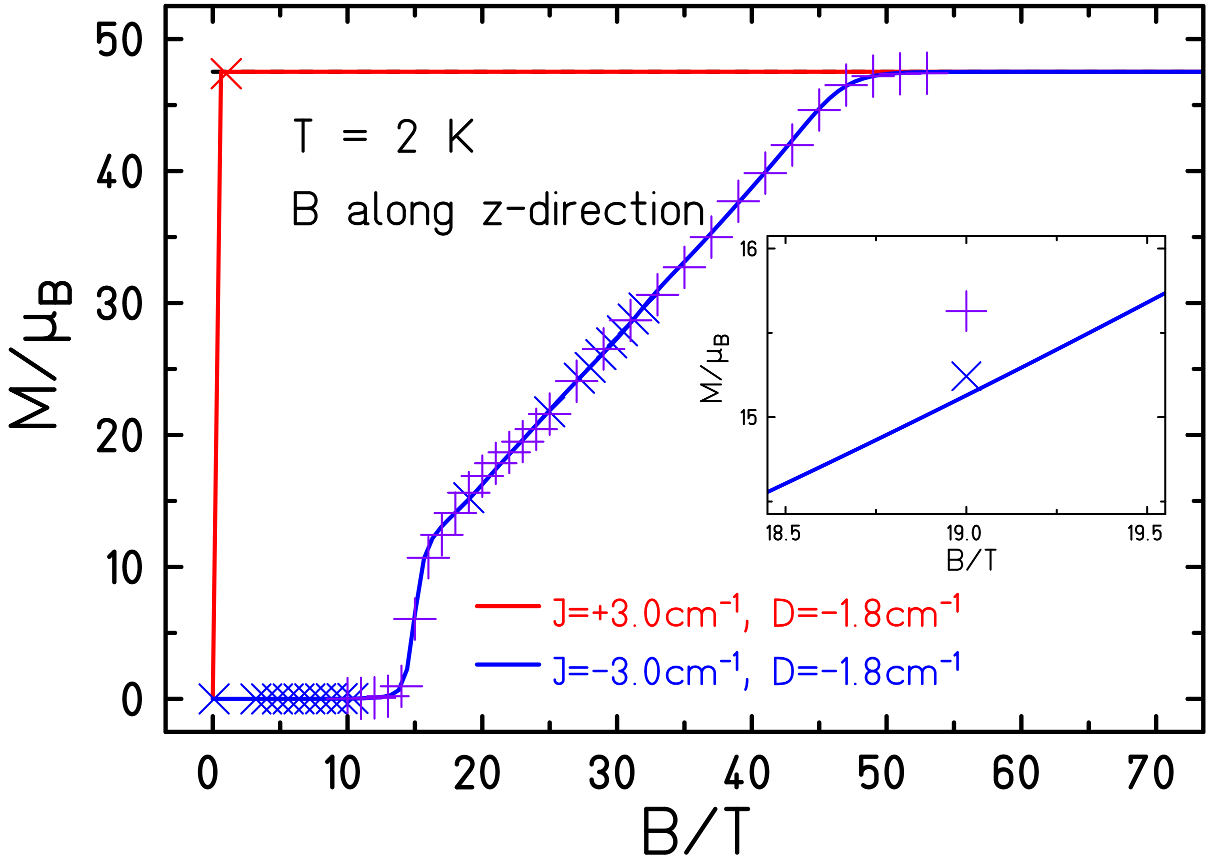
$\dim(\mathcal{H}) = 62,500$

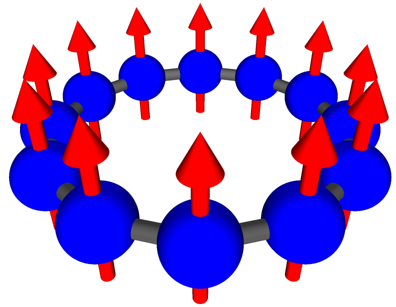
non-collinear easy axes

Hours compared to days, notebook compared to supercomputer!

O. Hanebaum, J. Schnack, Eur. Phys. J. B **87**, 194 (2014)

A fictitious $\mathrm{Mn}_{12}^{\mathrm{III}}$ – M_z vs B_z





$s = 2$

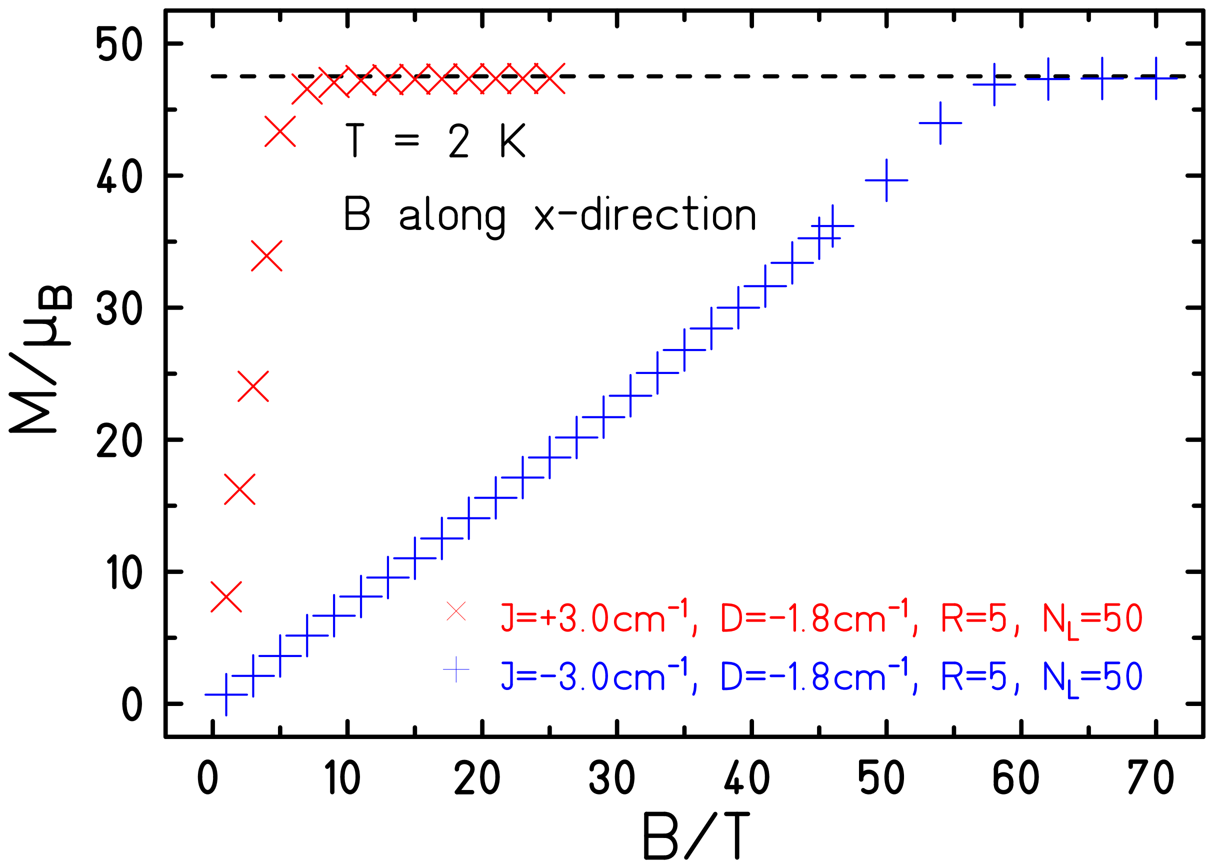
$\dim(\mathcal{H}) = 244,140,625$

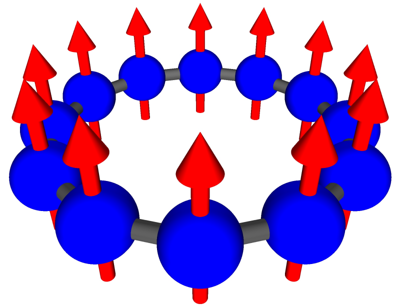
collinear easy axes

A few days compared to *impossible*!

O. Hanebaum, J. Schnack, Eur. Phys. J. B **87**, 194 (2014)

A fictitious Mn_{12}^{III} – M_x vs B_x





No other method can deliver these curves!

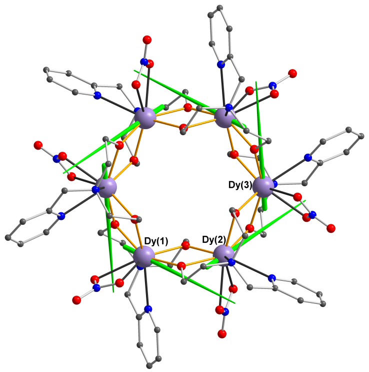
O. Hanebaum, J. Schnack, Eur. Phys. J. B **87**, 194 (2014)

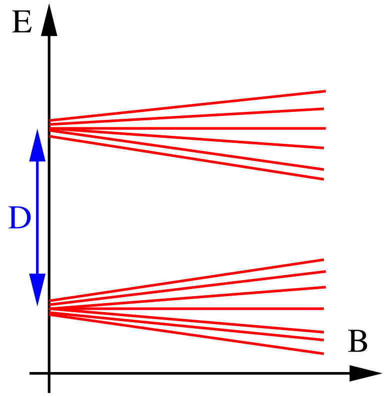
VERY anisotropic magnetic molecules

Let’s drive the problem and the method to their extrems!

sFTLM* I

$$\underset{\sim}{H} = \sum_{i<j} \underset{\sim}{\vec{s}}_i \cdot \mathbf{J}_{ij} \cdot \underset{\sim}{\vec{s}}_j + \sum_{i} \underset{\sim}{\vec{s}}_i \cdot \mathbf{D}_i \cdot \underset{\sim}{\vec{s}}_i + \mu_B\, B \sum_i g_i\, \underset{\sim}{s}_i^z$$





- Very strong alternating easy axes with $D \approx -20$ K. $J \approx 0.1$ K and dipolar interaction.
- $\rightarrow$ Density of states $=$ bumpy road.
- sFTLM*: “s” might be symmetric, strange or super, not yet clear. It was called “L” in (1).
- Alternative approach: OFTLM.

(1) M. Aichhorn, M. Daghofer, H. G. Evertz, and W. von der Linden, Phys. Rev. B **67**, 161103(R) (2003).

sFTLM II – the problem

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

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

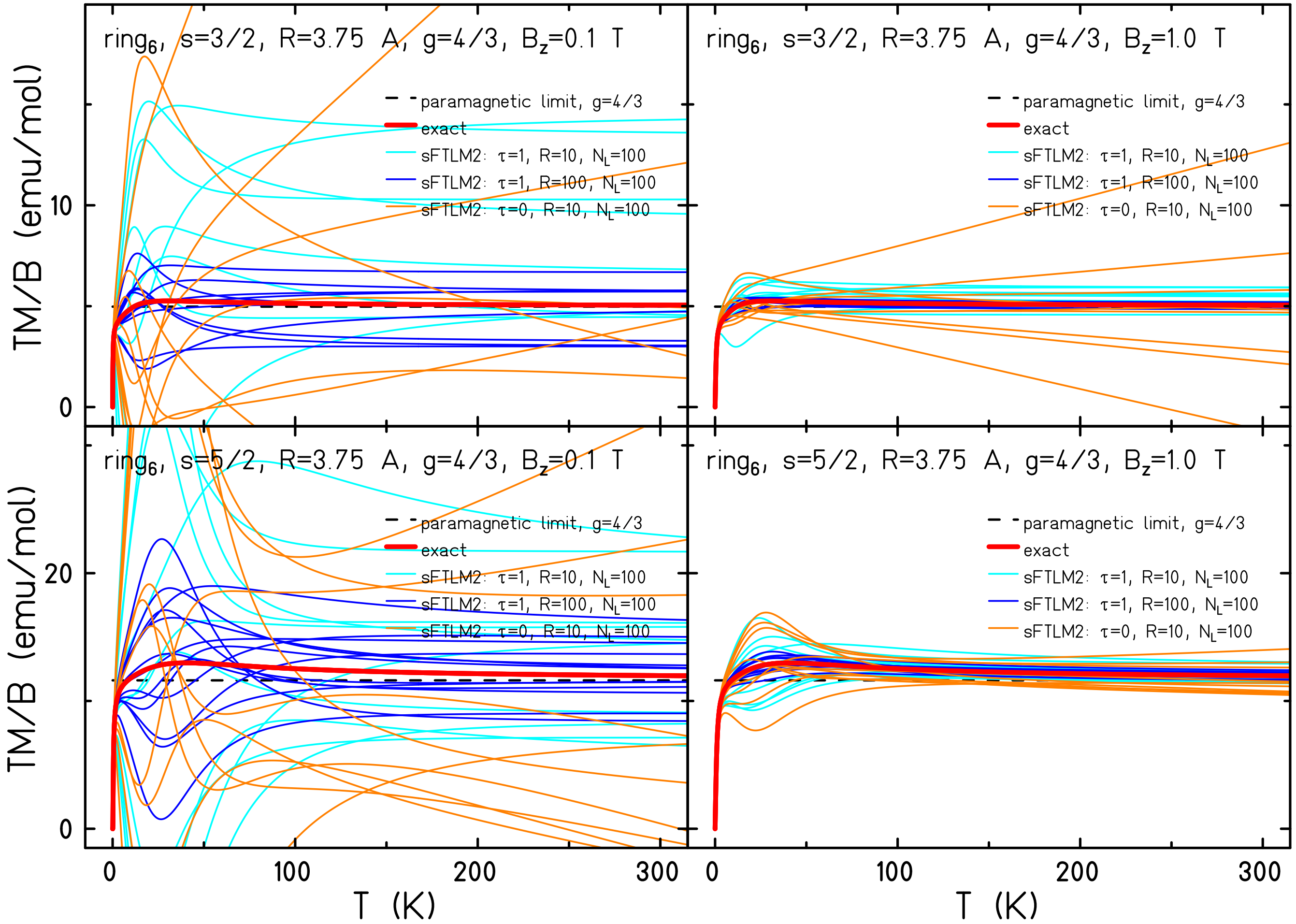
The traces are the same, but the approximations are not for $[\underset{\sim}{Q}, \underset{\sim}{H}] \neq 0$.
Symmetric version should be better (1), but needs much more resources (2).

$$\text{Tr}\left(\underset{\sim}{Q}e^{-\beta\underset{\sim}{H}}\right) \approx \frac{\dim(\mathcal{H})}{R} \sum_{r=1}^{R} \sum_{n=1}^{N_L} e^{-\beta\epsilon_n^{(r)}} \langle\, r \,|\, \underset{\sim}{Q} \,|\, n \,\rangle\langle\, n \,|\, r \,\rangle$$

$$\text{Tr}\left(e^{-\beta\underset{\sim}{H}/2}\underset{\sim}{Q}e^{-\beta\underset{\sim}{H}/2}\right) \approx \frac{\dim(\mathcal{H})}{R} \sum_{r=1}^{R} \sum_{m,n=1}^{N_L} e^{-\beta(\epsilon_m^{(r)}+\epsilon_n^{(r)})/2} \langle\, r \,|\, m \,\rangle\langle\, m \,|\, \underset{\sim}{Q} \,|\, n \,\rangle\langle\, n \,|\, r \,\rangle$$

(1) M. Aichhorn, M. Daghofer, H. G. Evertz, and W. von der Linden, Phys. Rev. B **67**, 161103(R) (2003).
(2) I. Rousochatzakis, S. Kourtis, J. Knolle, R. Moessner, and N. B. Perkins, Phys. Rev. B **100**, 045117 (2019).

sFTLM III – large fluctuations due to very different energy scales



We will get there, whatever it takes!

Summary

- FTLM is a phantastic method that delivers quasi-exact equilibrium observables for systems with Hilbert-(sub)-spaces with dimensions up to 10^{11}.
- Frustrated spin systems can show very unusual and exciting magnetocaloric properties.
- Improvement of FTLM for very anisotropic spin systems such as toroidal magnetic molecules is on its way.

Thank you very much for your attention.

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)
- J. Richter, J. Schulenburg (Magdeburg); B. Lake (HMI Berlin); B. Büchner, V. Kataev, H.-H. Klauß (Dresden); A. Powell, C. Anson, W. Wernsdorfer (Karlsruhe); J. Wosnitza (Dresden-Rossendorf); J. van Slageren (Stuttgart); R. Klingeler (Heidelberg); O. Waldmann (Freiburg); U. Kortz (Bremen)

Molecular Magnetism Web

www.molmag.de

Highlights. Tutorials. Who is who. Conferences.