Molecular Magnetism: Questions, Tools, and Answers

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DFG

文部科学省

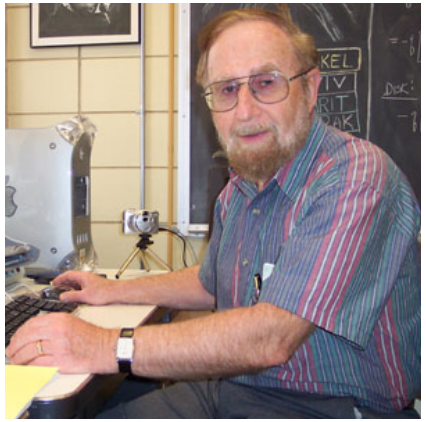
In late 20th century people coming from


transport theory


general relativity

nuclear physics


Schottky diodes



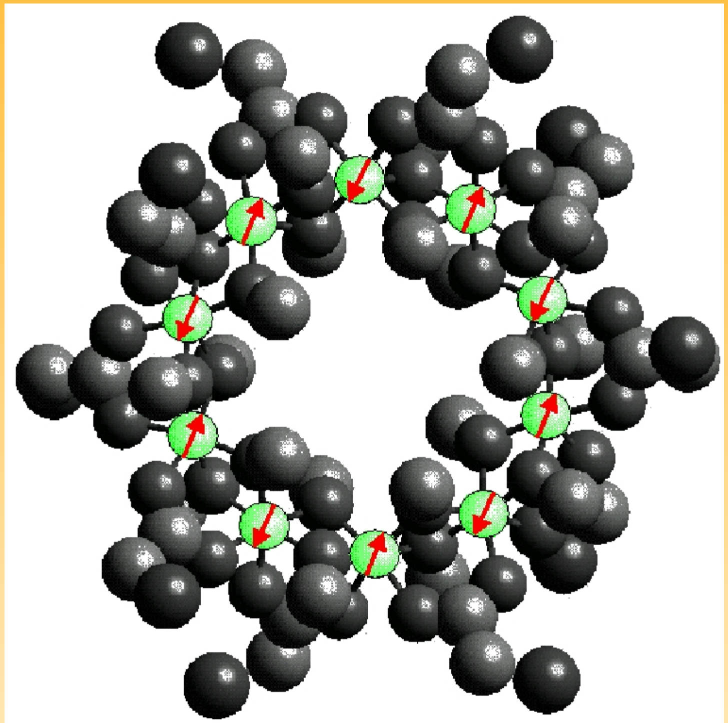
were triggered by a “magnetic” enthusiast.

Meanwhile a big collaboration has been established

- T. Englisch, T. Glaser, A. Müller (U. Bielefeld) & Chr. Schröder (FH Bielefeld);
- K. Bärwinkel, H.-J. Schmidt, M. Allalen, M. Brüger, D. Mentrup, D. Müter, M. Exler, P. Hage, F. Hesmer, K. Jahns, F. Ouchni, R. Schnalle, P. Shchelokovskyy, S. Torbrügge & M. Neumann, K. Küpper, M. Prinz (U. Osnabrück);
- M. Luban, D. Vaknin (Ames Lab, USA); P. Kögerler (RWTH, Jülich, Ames)
  J. Musfeld (U. of Tennessee, USA); N. Dalal (Florida State, USA);
  R.E.P. Winpenny (Man U, UK); L. Cronin (U. of Glasgow, UK);
  H. Nojiri (Tohoku University, Japan); A. Postnikov (U. Metz)
- J. Richter, J. Schulenburg, R. Schmidt (U. Magdeburg);
  S. Blügel (FZ Jülich); A. Honecker (U. Göttingen);
  E. Rentschler (U. Mainz); U. Kortz (IUB); A. Tennant, B. Lake (HMI Berlin);
  B. Büchner, V. Kataev, R. Klingeler (IFW Dresden)

. . . and various general results could be achieved

Contents for you today

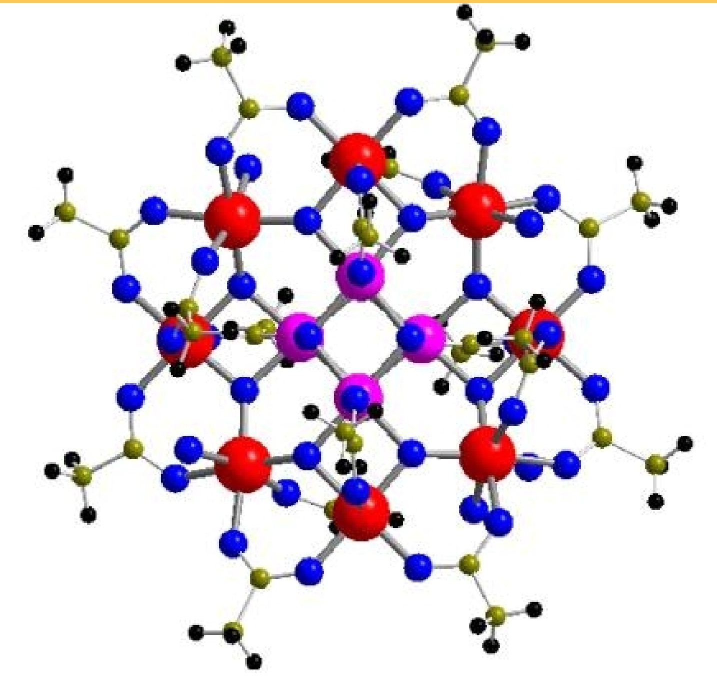


Fe_{10}

1. The suspects: magnetic molecules
2. The thumbscrew: Heisenberg model
3. Giant magnetization jumps in frustrated antiferromagnets
4. Hysteresis without anisotropy
5. My hardware & parallelization issues

Magnetic Molecules

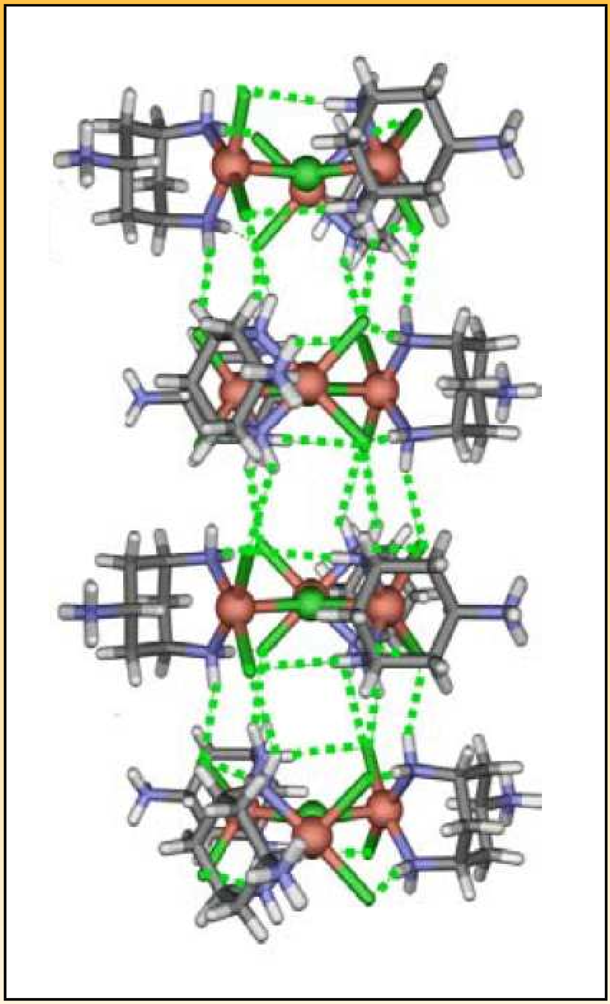
The beauty of magnetic molecules I



Mn_{12}

- Inorganic or organic macro molecules, where paramagnetic ions such as Iron (Fe), Chromium (Cr), Copper (Cu), Nickel (Ni), Vanadium (V), Manganese (Mn), or rare earth ions are embedded in a host matrix;
- Pure organic magnetic molecules: magnetic coupling between high spin units (e.g. free radicals);
- Spin = magnetic moment ("compass needle"): Molecule has magnetic properties.
- Speculative applications: magnetic storage devices, magnets in biological systems, light-induced nano switches, displays, catalysts, transparent magnets, qubits for quantum computers.

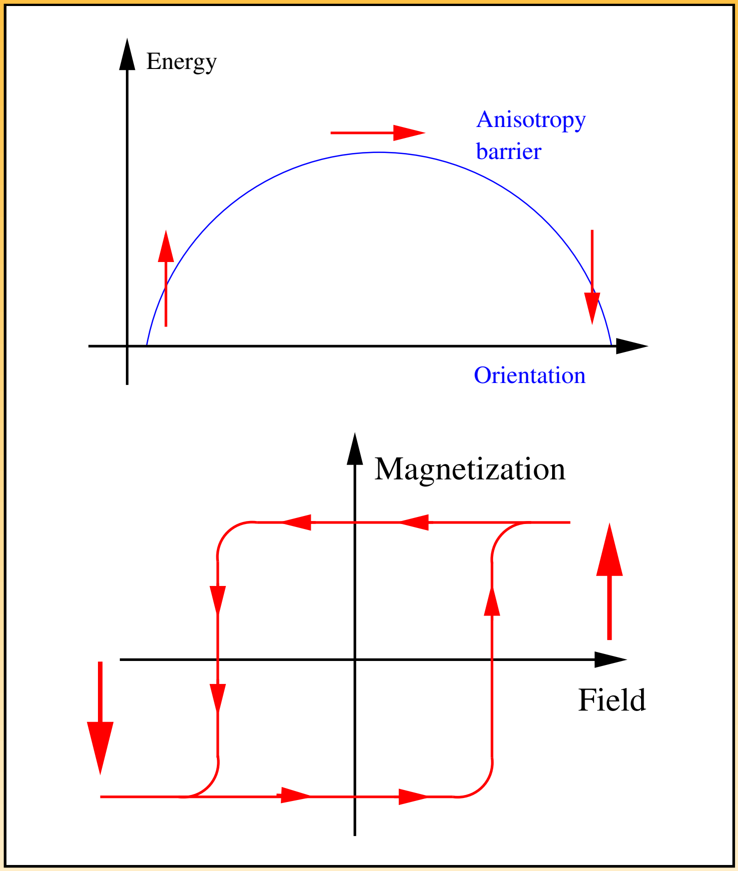
The beauty of magnetic molecules II



- Dimers (Fe_2), tetrahedra (Cr_4), cubes (Cr_8);
- Rings, especially iron and chromium rings (order from The Manchester Magic Ring Factory, Brunswick Street, Manchester, M13 9PL, UK);
- Complex structures (Mn_{12}) – drosophila of molecular magnetism;
- "Soccer balls", more precisely icosidodecahedra (Fe_{30}) and other macro molecules;
- Chain like and planar structures of interlinked magnetic molecules, e.g. triangular Cu chain:

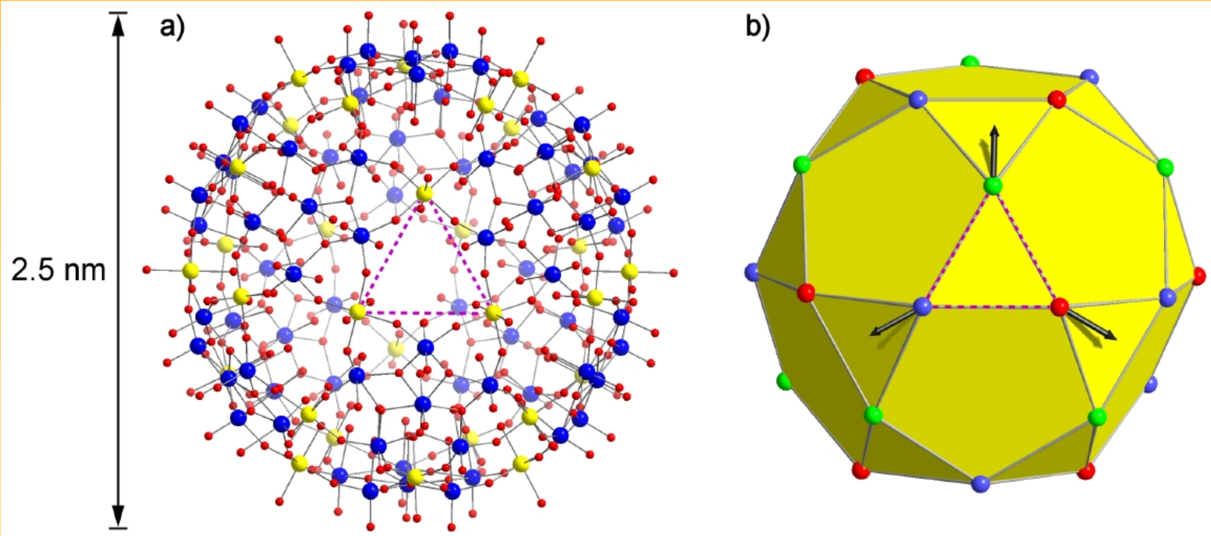
  J. Schnack, H. Nojiri, P. Kögerler, G. J. T. Cooper, L. Cronin, Phys. Rev. B 70, 174420 (2004)

The beauty of magnetic molecules III



- Single Molecule Magnets (SMM): magnetic molecules with large ground state moment; e.g. $S = 10$ for Mn_{12} or Fe_8
- Anisotropy barrier dominates behavior (as in your hard drive);
- Single molecule is a magnet and shows metastable magnetization and hysteresis; but also magnetization tunneling.
- Today's major efforts: improve stability of magnetization; investigate on surfaces.

The beauty of magnetic molecules IV
$\{$Mo$_{72}$Fe$_{30}\}$ – a giant magnetic Keplerate molecule



- Structure: Fe - yellow, Mo - blue,
  O - red;
- Exciting magnetic properties (1).
- Quantum treatment very complicated, dimension of Hilbert space $(2s+1)^N \approx 10^{23}$ (2).

(1) A. Müller *et al.*, Chem. Phys. Chem. **2**, 517 (2001) , (2) M. Exler and J. Schnack, Phys. Rev. B **67**, 094440 (2003)

Numerics

Model Hamiltonian – Heisenberg-Model

$$\underset{\sim}{H} = \sum_{i,j} \underset{\sim}{\vec{s}}(i) \cdot \mathbf{J}_{ij} \cdot \underset{\sim}{\vec{s}}(j) + \sum_{i,j} \vec{D}_{ij} \cdot \left[\underset{\sim}{\vec{s}}(i) \times \underset{\sim}{\vec{s}}(j)\right] + \mu_B\, B \sum_{i}^{N} g_i\, \underset{\sim}{s}_z(i)$$

Exchange/Anisotropy Dzyaloshinskii-Moriya Zeeman

Very often anisotropic terms are utterly negligible, then . . .

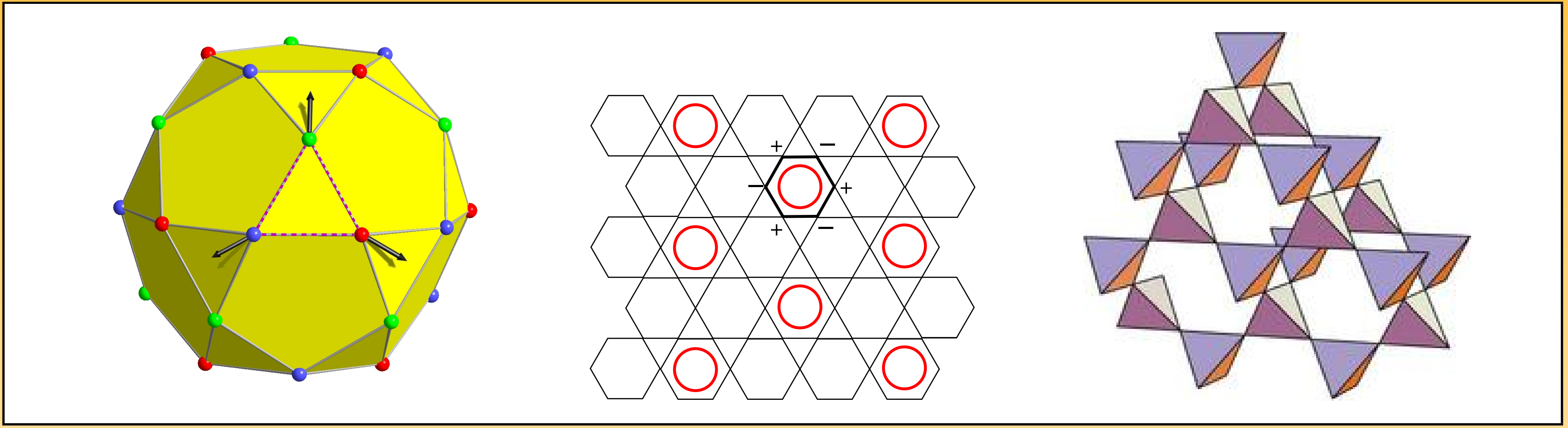
$$\underset{\sim}{H} = -\sum_{i,j} J_{ij}\, \underset{\sim}{\vec{s}}(i) \cdot \underset{\sim}{\vec{s}}(j) + g\, \mu_B\, B \sum_{i}^{N} \underset{\sim}{s}_z(i)$$

Heisenberg Zeeman

The Hamilton operator is represented as a matrix whose eigenvalues and eigenvectors have to be computed. Maximum size $\approx 30,000 \times 30,000$ complex$*16$.

Giant Magnetization Jumps

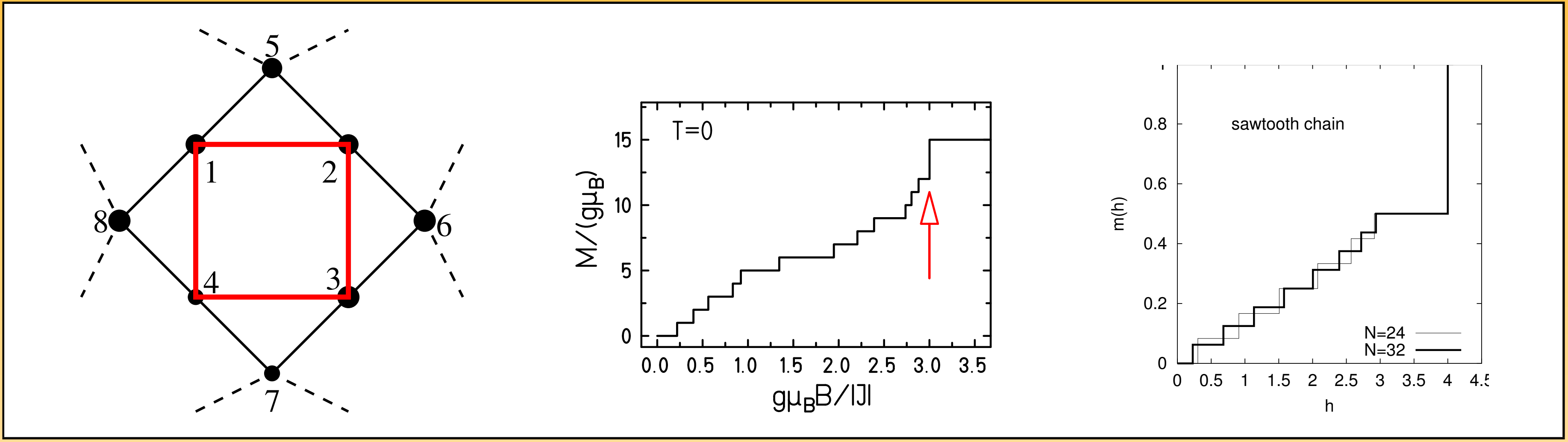
Giant magnetization jumps in frustrated antiferromagnets I
Systems



- Several frustrated antiferromagnets show an unusual behavior at the saturation field (1,2). AF $=\uparrow\downarrow$, saturation $=\uparrow\uparrow\uparrow\uparrow\ldots$.
- Example systems: icosidodecahedron, kagome lattice, pyrochlore lattice.

(1) J. Schnack, H.-J. Schmidt, J. Richter, J. Schulenburg, Eur. Phys. J. B **24**, 475 (2001)
(2) J. Schulenburg, A. Honecker, J. Schnack, J. Richter, H.-J. Schmidt, Phys. Rev. Lett. **88**, 167207 (2002)

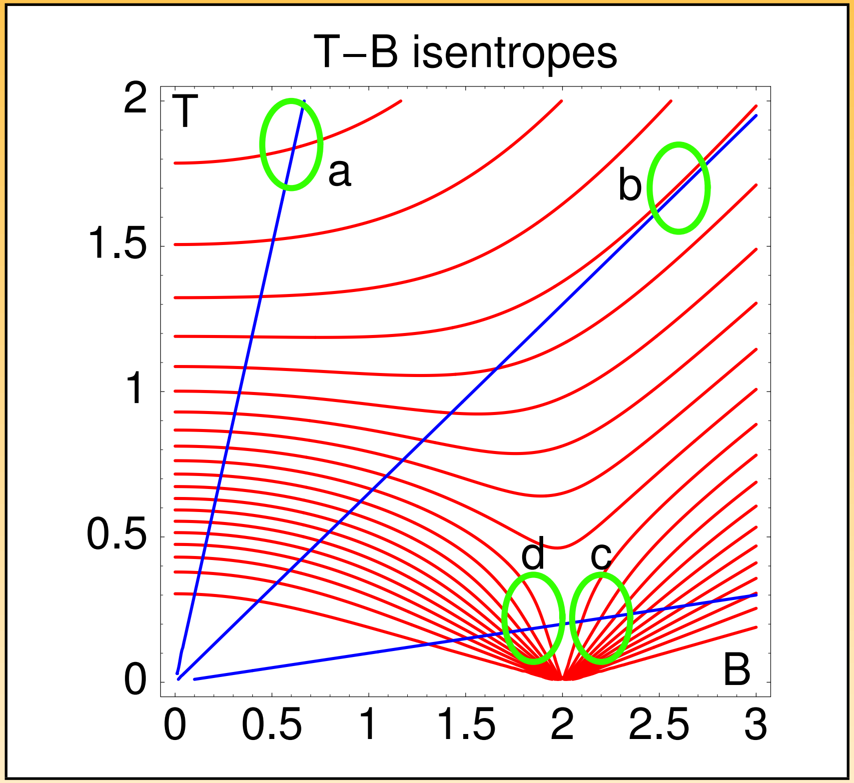
Giant magnetization jumps in frustrated antiferromagnets II
Magnetization jumps due to independent magnons



- Usually a magnetization curve is rather smooth.
- Unusually high magnetization jump at the saturation field.
- Many identical localized independent magnons flip their spins simultaneously.

J. Schulenburg, A. Honecker, J. Schnack, J. Richter, H.-J. Schmidt, Phys. Rev. Lett. **88**, 167207 (2002)
J. Richter, J. Schulenburg, A. Honecker, J. Schnack, H.-J. Schmidt, J. Phys.: Condens. Matter **16**, S779 (2004)

Giant magnetization jumps in frustrated antiferromagnets III
Giant magnetocaloric effect



blue lines: ideal paramagnet,
red curves: af dimer

Magnetocaloric effect, i.e. temperature change when changing the applied magnetic field:

(a) reduced,

(b) the same,

(c) enhanced,

(d) opposite

when compared to an ideal paramagnet.

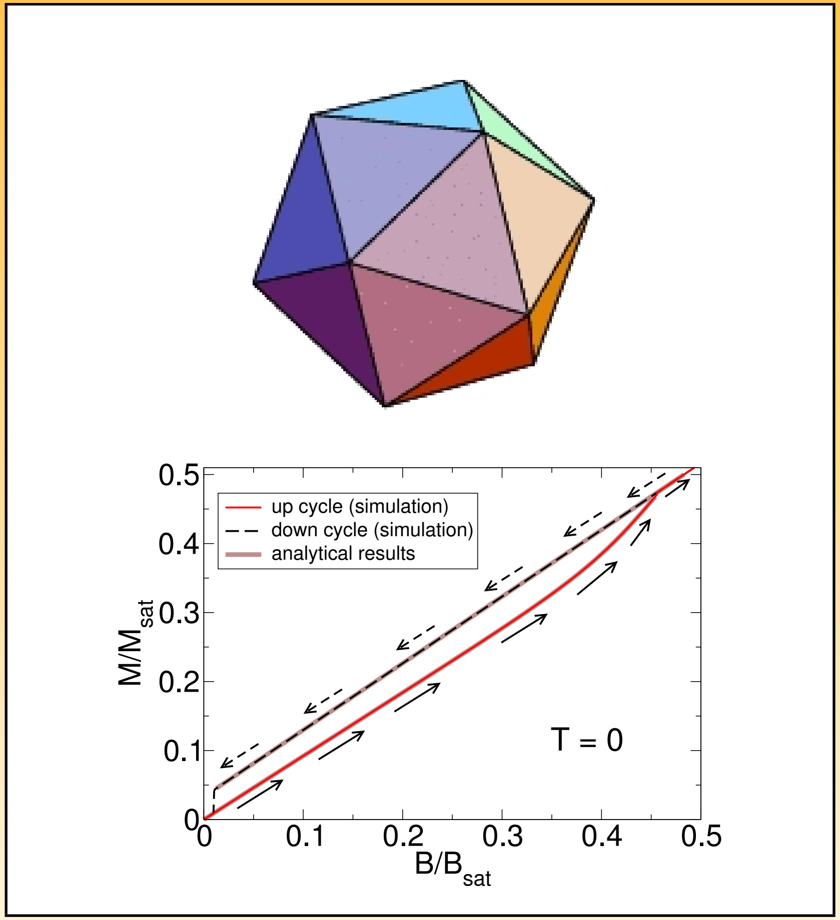
Case (d) does not occur for a paramagnet.

J. Schnack, J. Low Temp. Phys. **142** (2006) 279

J. Schnack, R. Schmidt, J. Richter, cond-mat/0703480

Hysteresis without Anisotropy

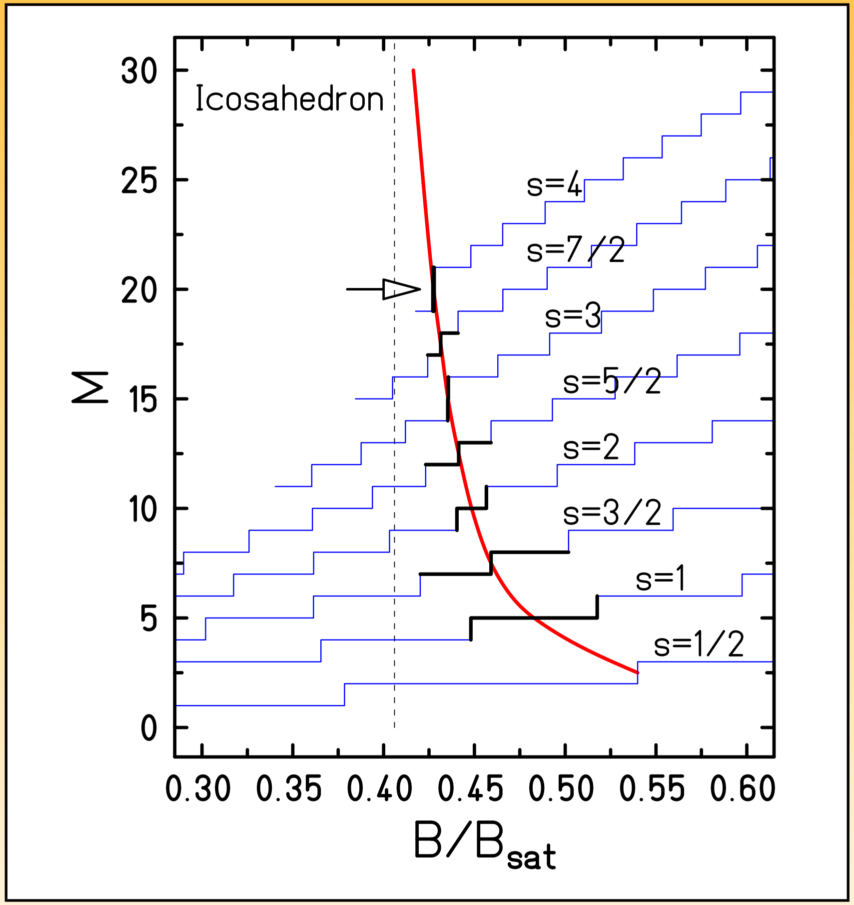
Metamagnetic phase transition I
Hysteresis without anisotropy



- Hysteresis is usually caused by anisotropy
- Hysteresis behavior of the classical isotropic Heisenberg icosahedron in an applied magnetic field.
- Classical spin dynamics simulations (thick lines + movie).
- Analytical stability analysis (grey lines).

C. Schröder, H.-J. Schmidt, J. Schnack, M. Luban, Phys. Rev. Lett. **94**, 207203 (2005)

Metamagnetic phase transition II
Quantum icosahedron



- Quantum analog:
  Non-convex minimal energy levels
  $\Rightarrow$ magnetization jump of $\Delta M > 1$.
- Lanczos diagonalization for various s.
- True jump of $\Delta M = 2$ for $s = 4$.
- Polynomial fit in $1/s$ yields the classically observed transition field.
- Numerics: Lanczos with vectors of lengths up to 1,342,275,012 used!

My hardware

&

Parallelization issues

Supercomputer 1st kind

Supercomputer
1st kind
(but . . .)

Fuel not compatible with the Kyoto protocol

2. Espresso

(Only 3 diagonalizations per gallon!)

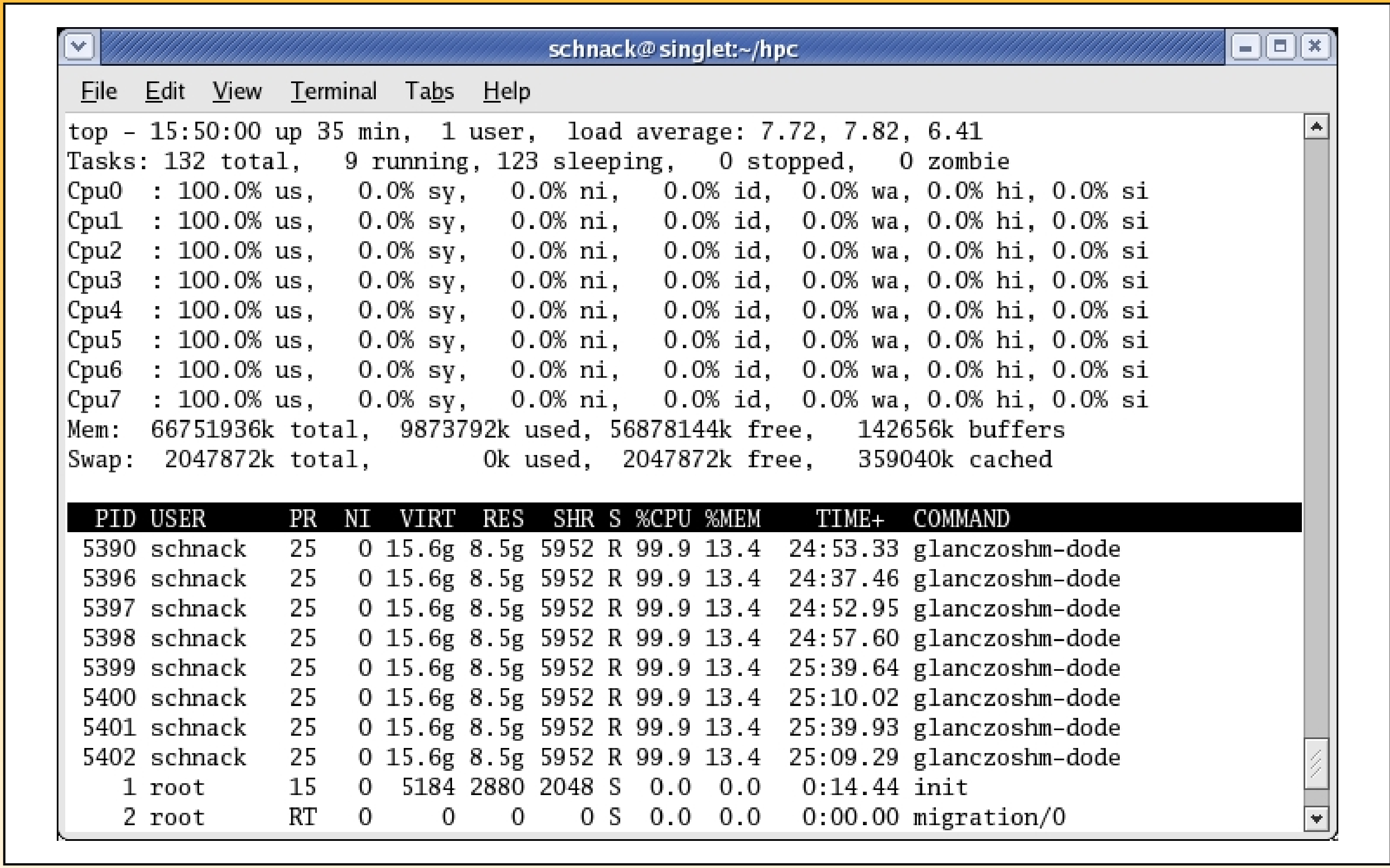
Supercomputer 2nd kind

- BULL NovaScale Server 3045:

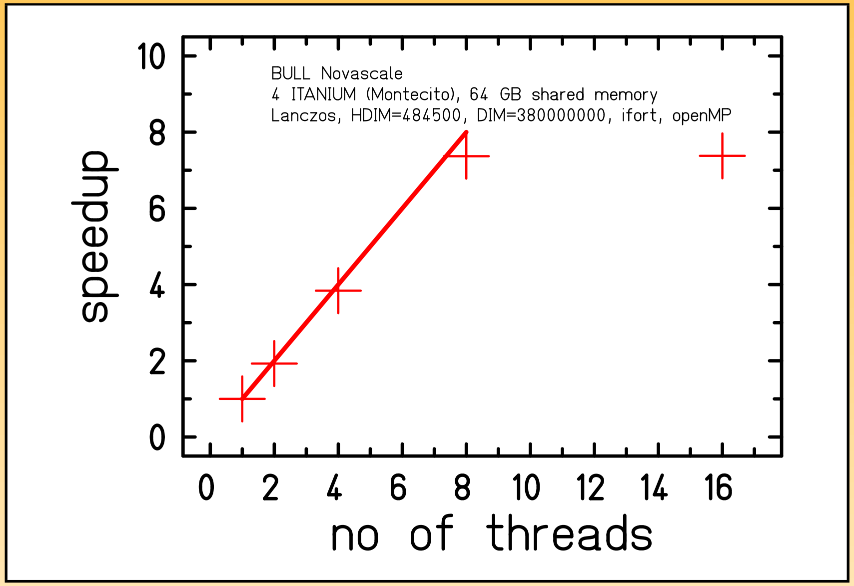
- Future: wide open
  8 ITANIUM TUKWILA (a 4 cores),
  512 GB RAM
  (an amazing computer power)

- Now:
  4 ITANIUM MONTECITO (a 2 cores),
  64 GB RAM
  (already an amazing computer power,
  but one can get used to it ;-))

Supercomputer 2nd kind



Parallelization issues I



Improvement of Lanczos diagonalization for very large system size:

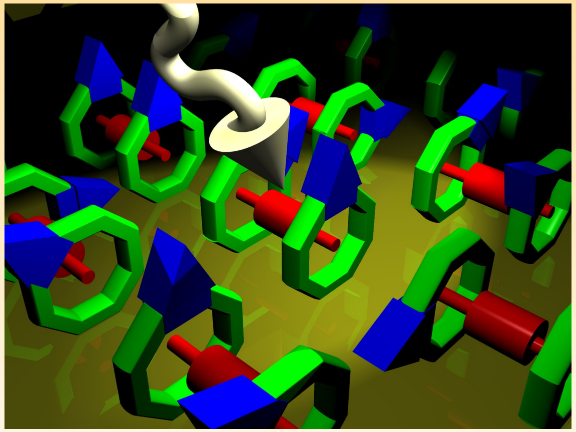
- System size: $10^{10} \ldots 10^{12}$ entries per vector;
- Rearrangement of loops – outer loop writes (1);
- Evaluation of matrix elements "on the fly", no storage;
- Analytical basis encoding in subspaces – faster than searching (1).

(1) J. Schnack, P. Hage, H.-J. Schmidt, arXiv:0706.3293v1 [cond-mat.str-el]

Parallelization issues II

- Goals:
  Numerically exact treatment of small quantum systems;
  Ground states, spectroscopic data;
  Time evolution (including heat bath coupling)
  Thermodynamics, statistics.
- Methodical problems:
  openMP parallelization of exact diagonalization (INTEL currently improves MKL, Ben Bennett),
  openMP parallelization of approximate methods (Lanczos, Arnoldi, DMRG),
  Numerical accuracy of vectors with $10^{10} \ldots 10^{12}$ entries.

Future HPC projects: The Manchester Gymwheel



- Structure:
  Two Fe_8 rings glued together by 4 rungs;

- Numerical demands:
  Size of Hilbert space ($s = 5/2$):
  $(2s+1)^N = 6^{16} = 2,821,109,907,456$
  Size of subspaces:
  dim$(\mathcal{H}(M=0)) = 163,112,472,594$
  If Manchester Computing hands over its BULL cluster, this might work!

- Workaround:
  Use more symmetries or treat similar system with ($s = 3/2$) instead.

Summary

There is a big demand
for fast and accurate numerics
in the theory of magnetism.

And, the end is not in sight, . . .

. . . , however, this talk is at its end!

Thank you very much for your attention.

German Molecular Magnetism Web

www.molmag.de

Highlights. Tutorials. Who is who. DFG SPP 1137