# Molecular Magnetism: Questions, Tools, and Answers

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Paralleles Kolloquium

RWTH, Aachen, June 4th 2007

Universität Bielefeld
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文部科学省

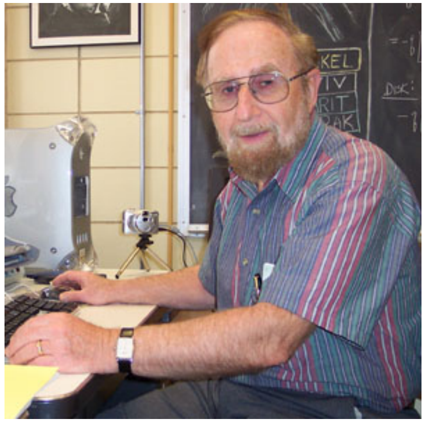
# In late 20th century people coming from

transport theory

general relativity

nuclear physics

Schottky diodes



were triggered by a “magnetic” enthusiast.

# Meanwhile a big collaboration has been established

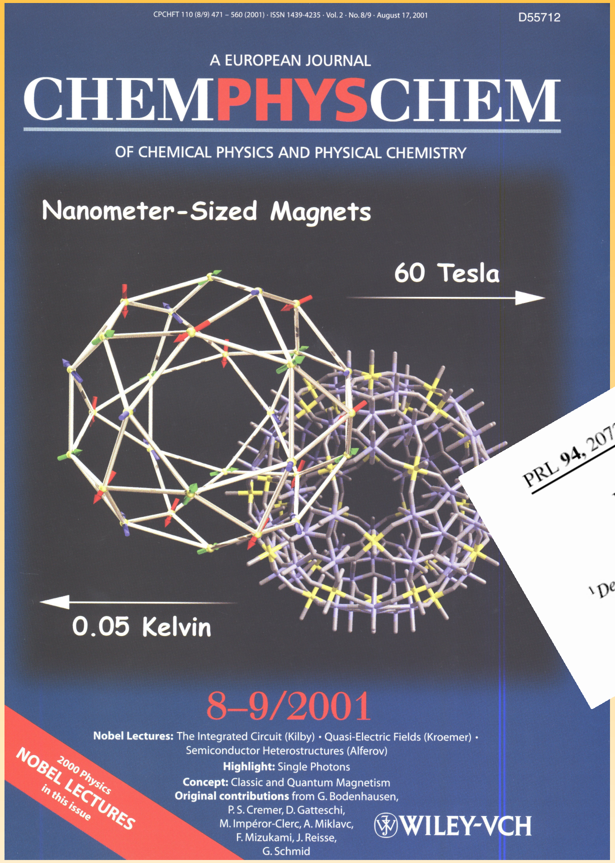
- T. Englisch, T. Glaser, A. Müller (U. Bielefeld) & Chr. Schröder (FH Bielefeld);

- 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



CPCHFT 110 (8/9) 471 – 560 (2001) · ISSN 1439-4235 · Vol. 2 · No. 8/9 · August 17, 2001

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CHEMPHYSCHEM

OF CHEMICAL PHYSICS AND PHYSICAL CHEMISTRY

Nanometer-Sized Magnets

8–9/2001

**Nobel Lectures:** The Integrated Circuit (Kilby) · Quasi-Electric Fields (Kroemer) · Semiconductor Heterostructures (Alferov)

**Highlight:** Single Photons

**Concept:** Classic and Quantum Magnetism

**Original contributions** from G. Bodenhausen, P. S. Cremer, D. Gatteschi, M. Impéror-Clerc, A. Miklavc, F. Mizukami, J. Reisse, G. Schmid

2000 Physics NOBEL LECTURES in this issue

WILEY-VCH

PRL 94, 207203 (2005) PHYSICAL REVIEW LE

week ending 27 MAY 2005

**Metamagnetic Phase Transition of the Antifer**

Christian Schröder,[1,*] Heinz-Jürgen Schmidt,[2] Jürge

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(Received 23 January 2005; published 23 Ma

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PHYSICAL REVIEW B, VOLUME 63, 014418

**Rotational modes in molecular magnets with antiferromagnetic Heisenberg exchange**

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Ames Laboratory & Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011

(Received 13 July 2000; published 12 December 2000)

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VOLUME 88, NUMBER 16 PHYSICAL REVIEW LETTERS

**Macroscopic Magnetization Jumps due to Independent Mag in Frustrated Quantum Spin Lattices**

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[2]Institut für Theoretische Physik, TU Braunschweig, Mendelssohnstrasse 3, D-38106 Braunschweig, Germany
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(Received 29 August 2001; published 8 April 2002)

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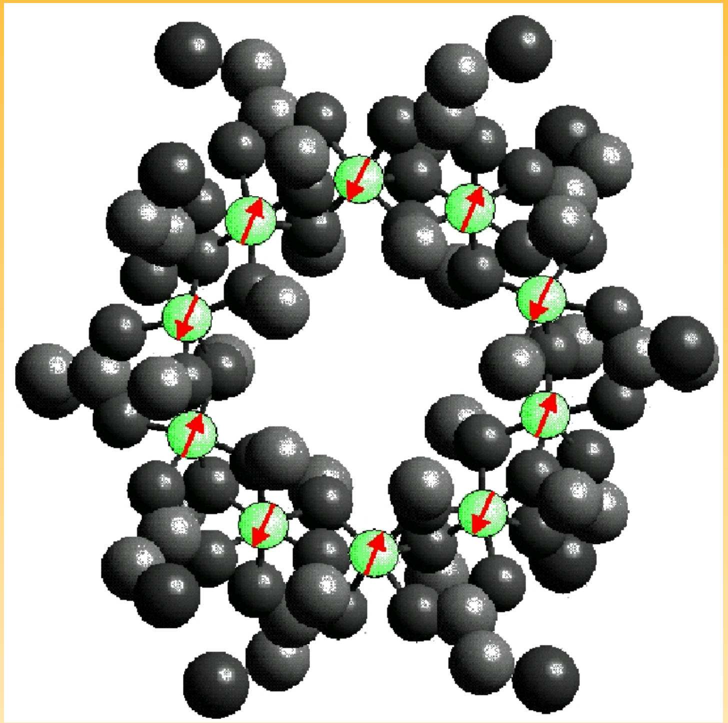
PHYSICAL REVIEW B 68, 05

**Quantum numbers for relative ground states** spin rings

Klaus Bärwinkel,* Peter Hage,†
Universität Osnabrück, Fac
(Received 24

# Contents for you today

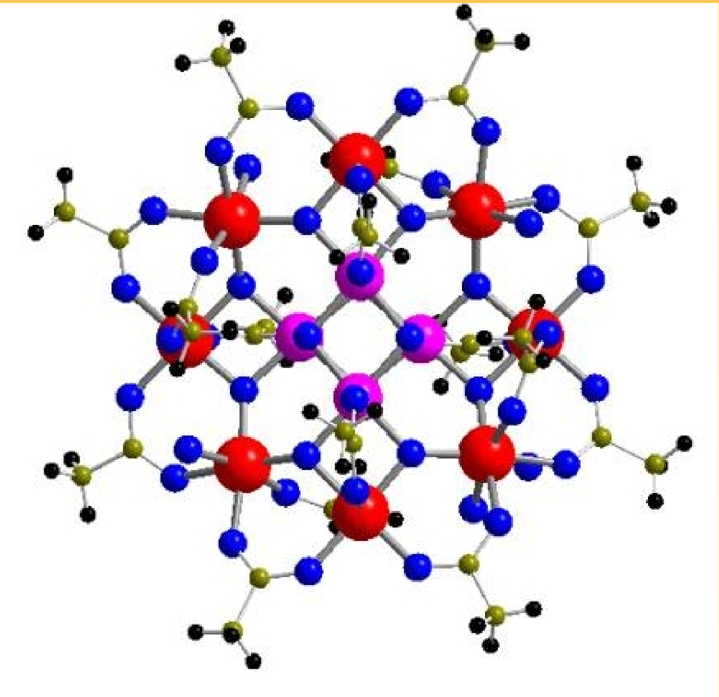


Fe$_{10}$

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

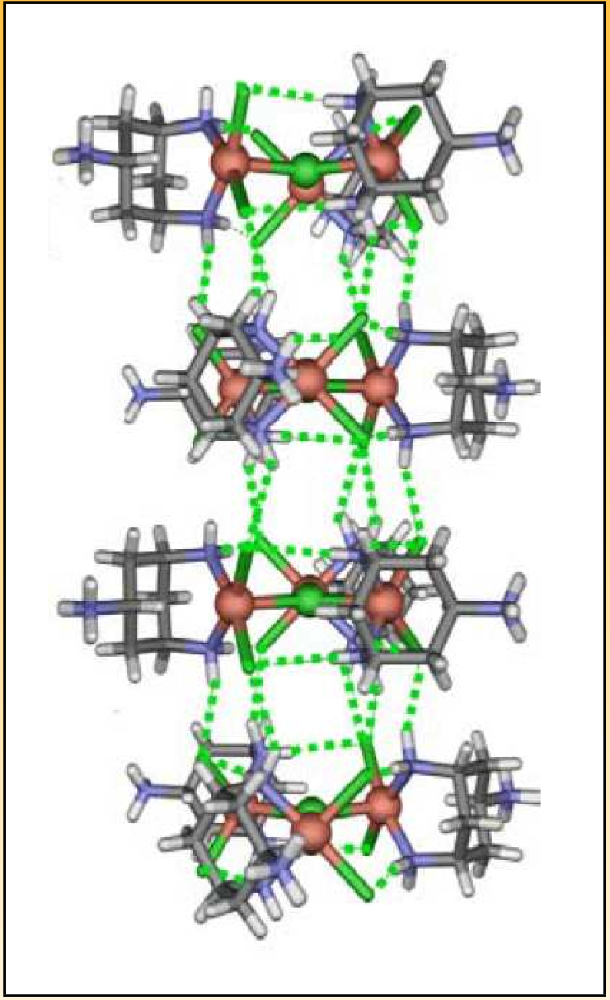
# The beauty of magnetic molecules I



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.

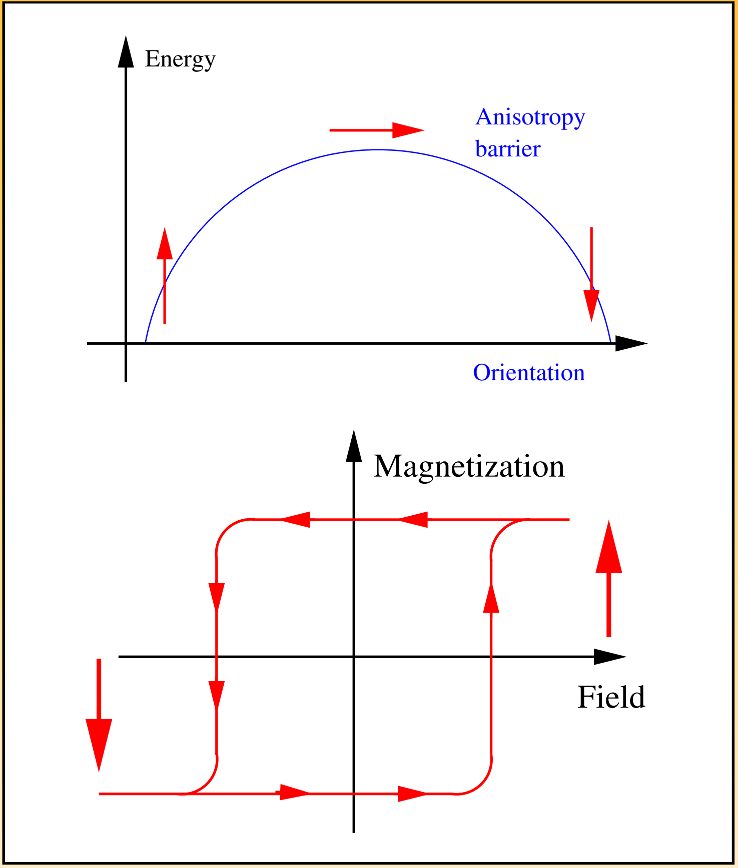
# The beauty of magnetic molecules II



- Dimers (Fe$_2$), tetrahedra (Cr$_4$), cubes (Cr$_8$);
- Rings, especially iron rings (Fe$_6$, Fe$_8$, Fe$_{10}$, $\dots$);
- 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



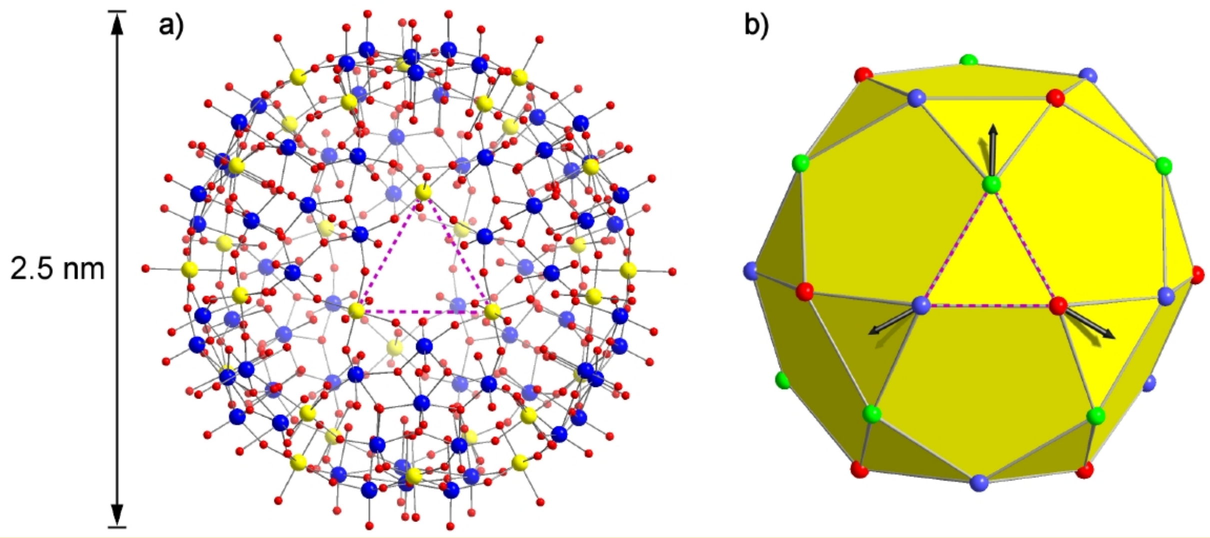
- Single Molecule Magnets (SMM): magnetic molecules with large ground state moment; e.g. $S = 10$ for $\mathrm{Mn}_{12}$ or $\mathrm{Fe}_8$

- 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
# $\{\text{Mo}_{72}\text{Fe}_{30}\}$ – 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

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

Exchange/Anisotropy Dzyaloshinskii-Moriya Zeeman

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

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

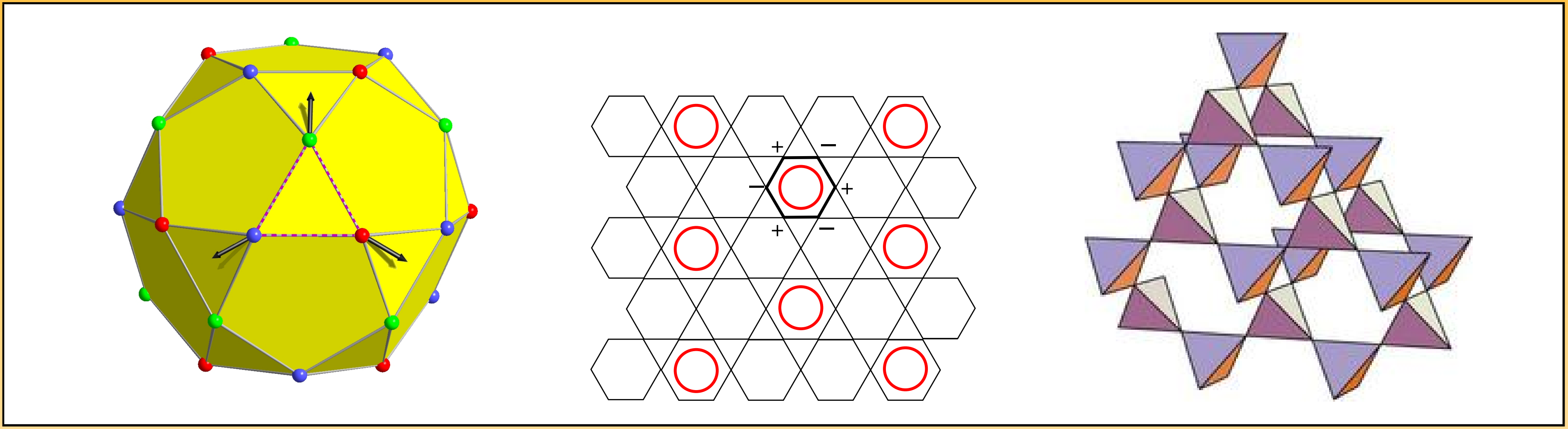
Heisenberg Zeeman

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

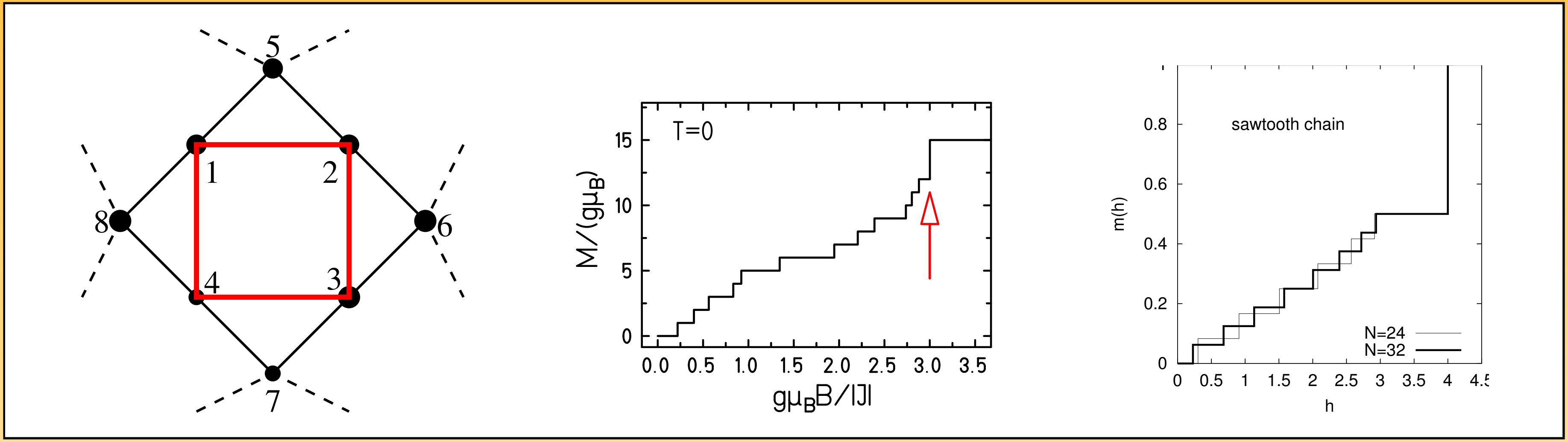


- Several frustrated antiferromagnets show an unusual behavior at the saturation field (1,2). AF $=\uparrow\downarrow$, saturation $=\uparrow\uparrow\uparrow\uparrow\ldots$.

- 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)

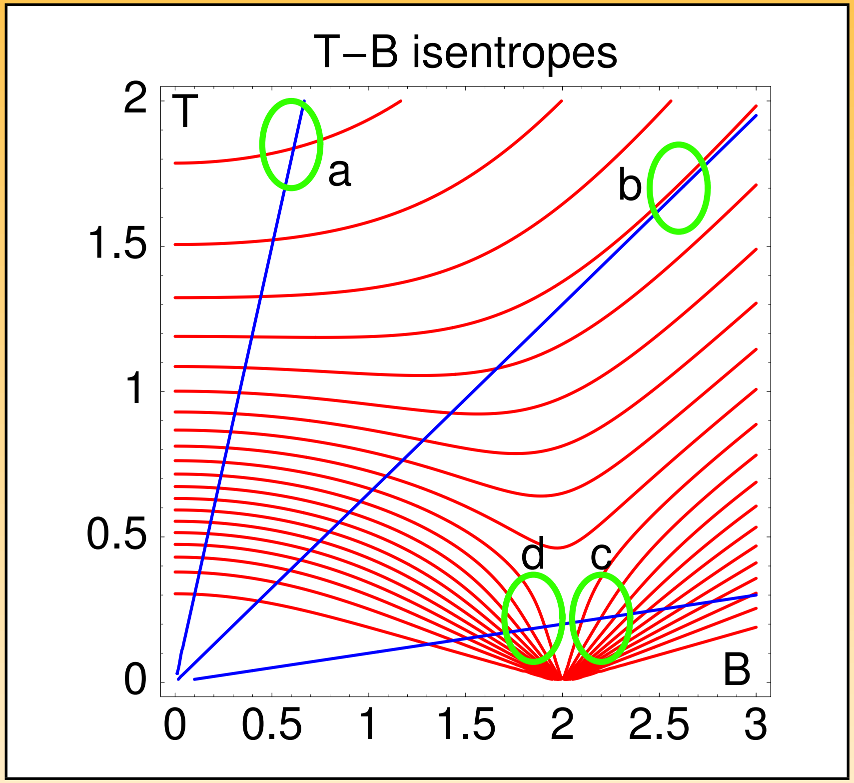
# Giant magnetization jumps in frustrated antiferromagnets II
# Magnetization jumps due to independent magnons



- 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.

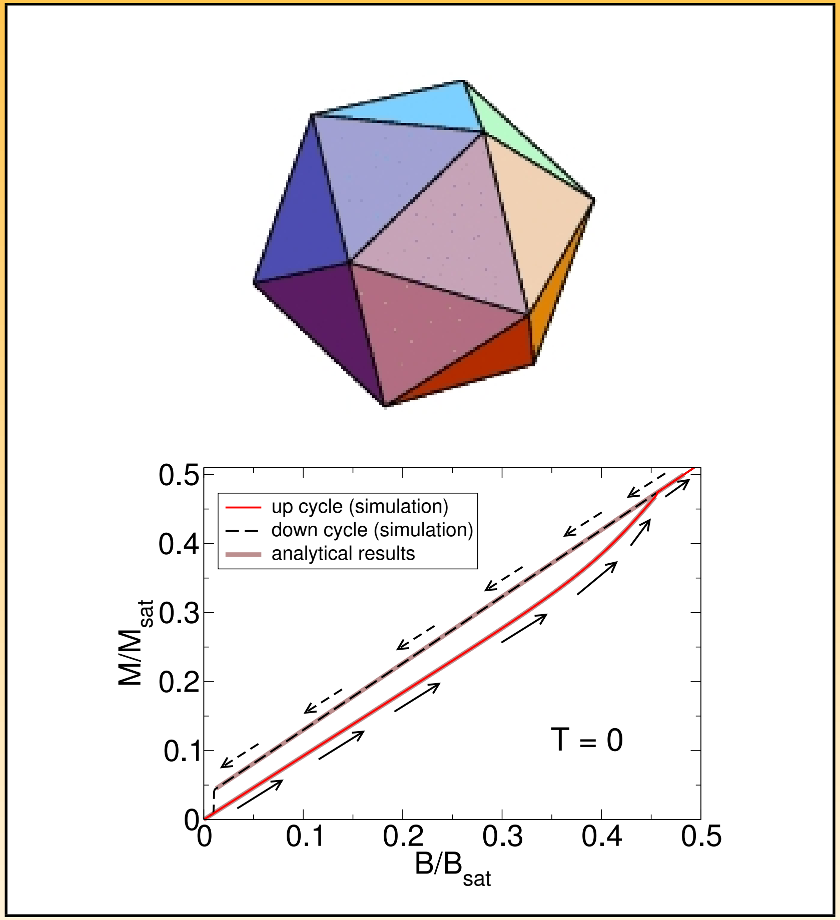
Case (d) does not occur for a paramagnet.

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

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

# Hysteresis without Anisotropy

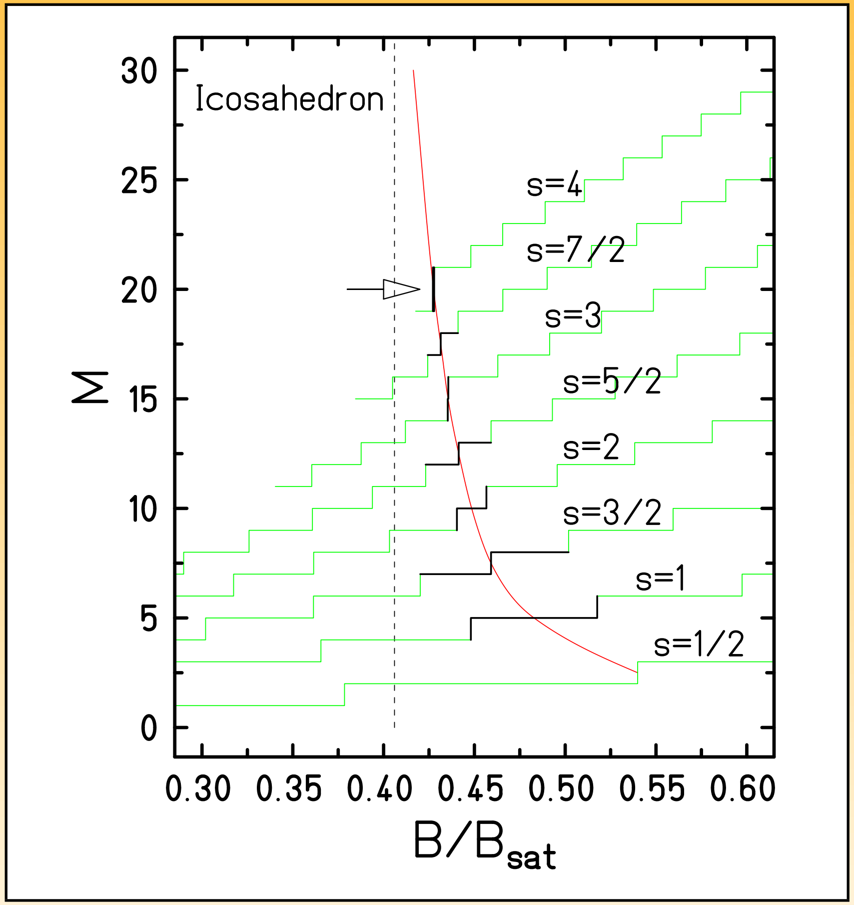
# Metamagnetic phase transition I
# Hysteresis without anisotropy



- 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

# My hardware I

1. hardware

(Runs with fuel . . . )

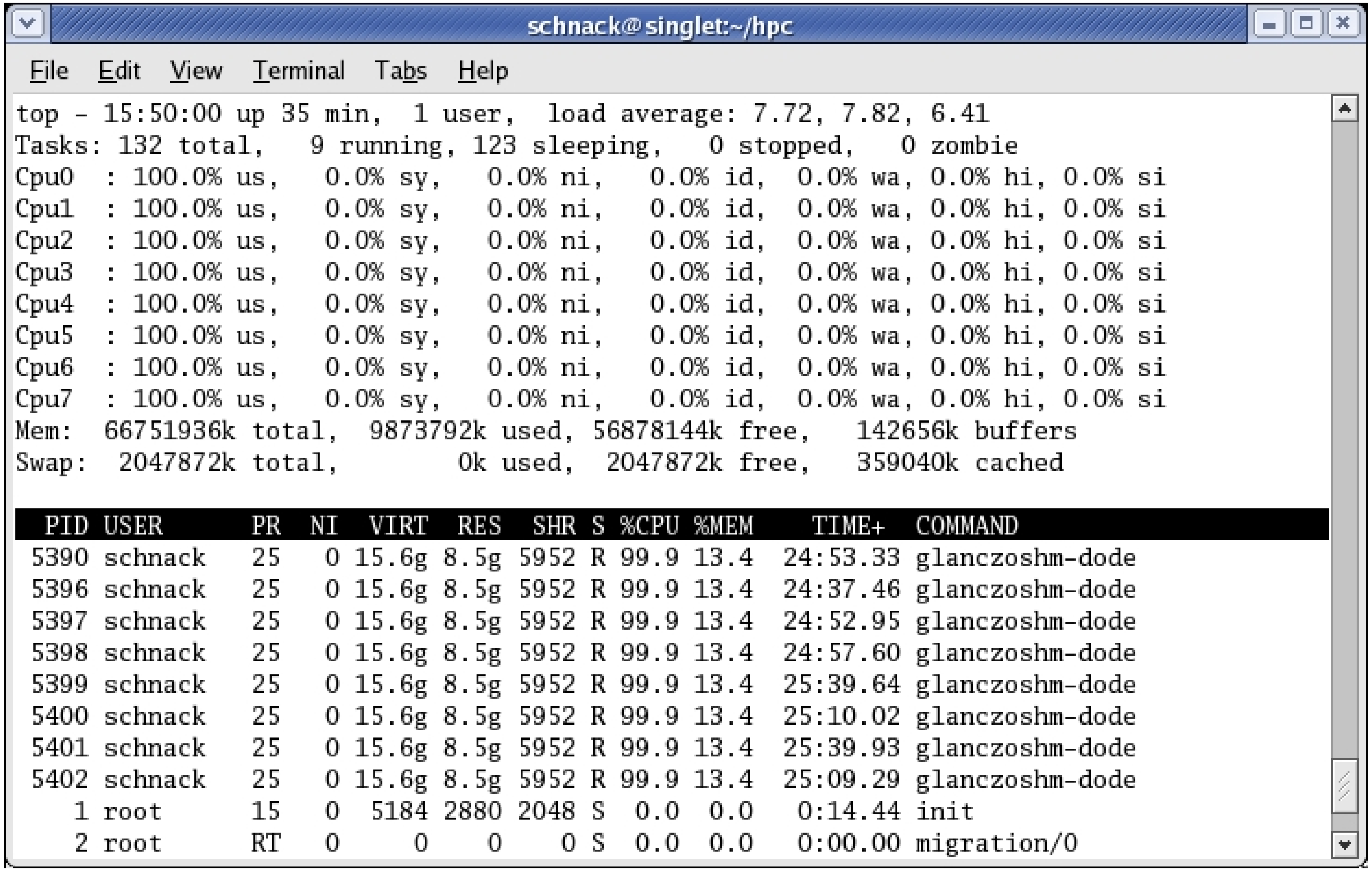
# My hardware II

2. Espresso

# My hardware III

- 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 ;-) )

# My hardware IV



```
top - 15:50:00 up 35 min,  1 user,  load average: 7.72, 7.82, 6.41
Tasks: 132 total,   9 running, 123 sleeping,   0 stopped,   0 zombie
Cpu0  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Cpu1  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Cpu2  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Cpu3  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Cpu4  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Cpu5  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Cpu6  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Cpu7  : 100.0% us,  0.0% sy,  0.0% ni,  0.0% id,  0.0% wa, 0.0% hi, 0.0% si
Mem:  66751936k total,  9873792k used, 56878144k free,   142656k buffers
Swap:  2047872k total,        0k used,  2047872k free,   359040k cached

  PID USER      PR  NI  VIRT  RES  SHR S %CPU %MEM    TIME+  COMMAND
 5390 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  24:53.33 glanczoshm-dode
 5396 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  24:37.46 glanczoshm-dode
 5397 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  24:52.95 glanczoshm-dode
 5398 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  24:57.60 glanczoshm-dode
 5399 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  25:39.64 glanczoshm-dode
 5400 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  25:10.02 glanczoshm-dode
 5401 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  25:39.93 glanczoshm-dode
 5402 schnack   25   0 15.6g 8.5g 5952 R 99.9 13.4  25:09.29 glanczoshm-dode
    1 root      15   0  5184 2880 2048 S  0.0  0.0   0:14.44 init
    2 root      RT   0     0    0    0 S  0.0  0.0   0:00.00 migration/0
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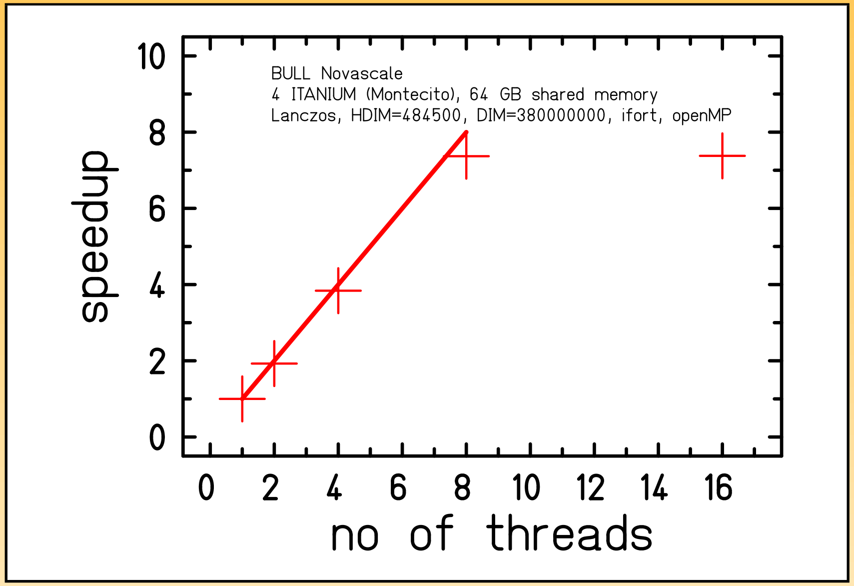
# Parallelization issues I



**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, in preparation

# Parallelization issues II

- Goals:
  Numerically exact treatment of small quantum systems;
  Ground states, spectroscopic data;
  Time evolution (e.g. TDMRG by U. Schollwöck, RWTH)
  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.

# 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