

Molecular Magnetism: Questions, Tools, and Answers

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文部科学省

In late 20th century people coming from



transport theory



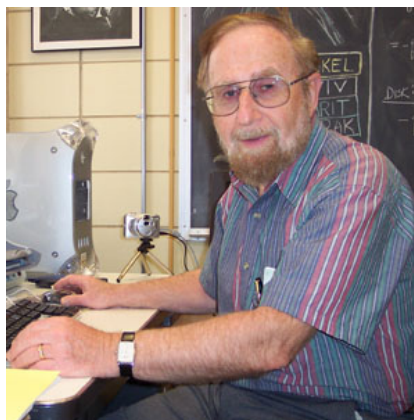
general relativity



nuclear physics



Schottky diodes

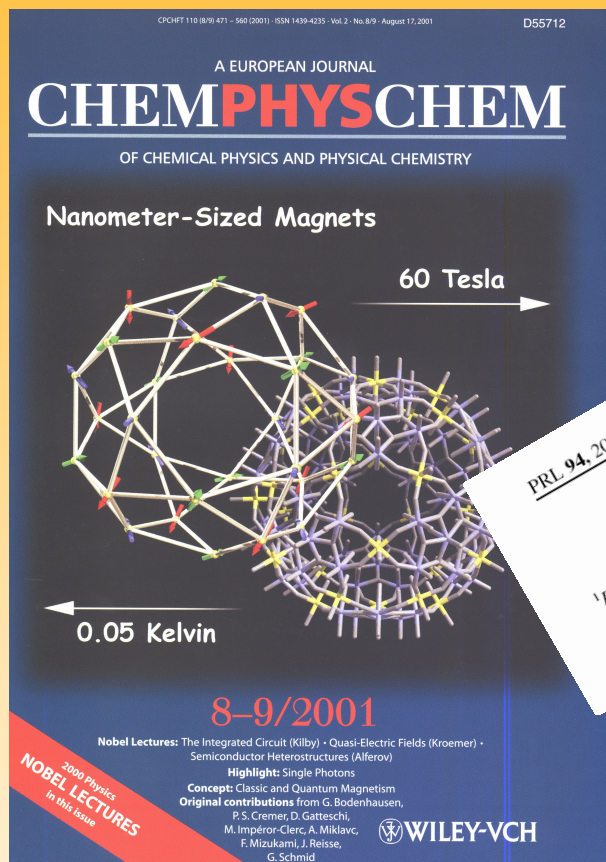


were triggered by a “magnetic” enthusiast.

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



PHYSICAL REVIEW LETTERS
 PRL 94, 207203 (2005)

Metamagnetic Phase Transition of the Antiferromagnet
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 Laboratory & Department of Physics and Astronomy, Iowa State
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 on
 Germany

Rotational modes in molecular magnets with antiferromagnetic Heisenberg exchange
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 Marshall Luban†
 Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011
 (Received 13 July 2000; published 12 December 2000)

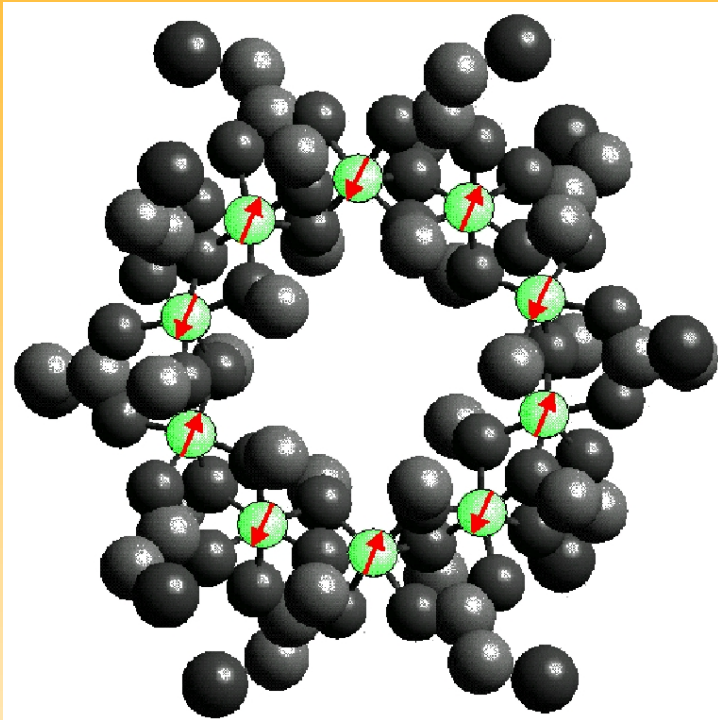
PHYSICAL REVIEW LETTERS
 VOLUME 88, NUMBER 16

Macroscopic Magnetization Jumps due to Independent Magnons in Frustrated Quantum Spin Lattices
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²Institut für Theoretische Physik, TU Braunschweig, Mendelssohnstrasse 3, D-38106 Braunschweig, Germany
³Universität Osnabrück, Fachbereich Physik, Barbarastrasse 7, D-49069 Osnabrück, Germany
 (Received 29 August 2001; published 8 April 2002)

Quantum numbers for relative ground states
 Klaus Bärwinkel,* Peter Hage,[†]
 Universität Osnabrück, Fac.
 (Received 21

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 spin rings

Contents for you today

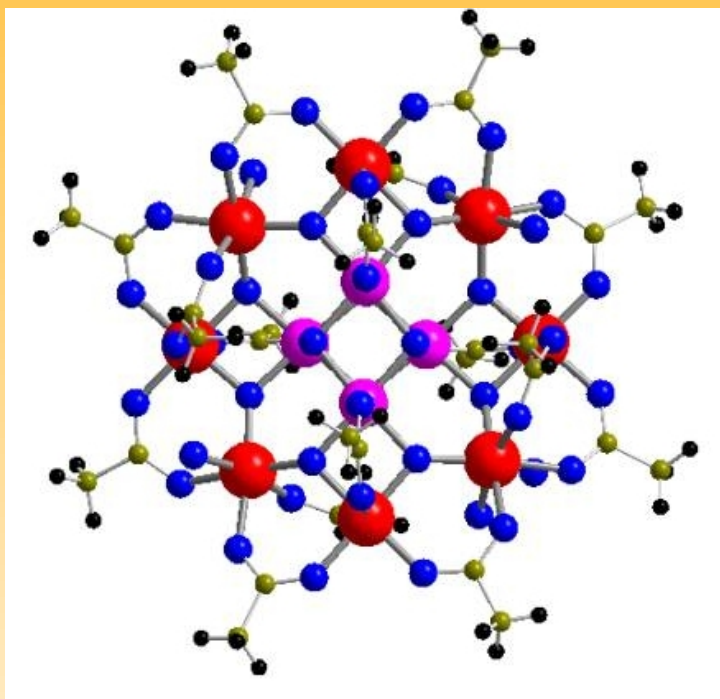


Fe_{10}

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

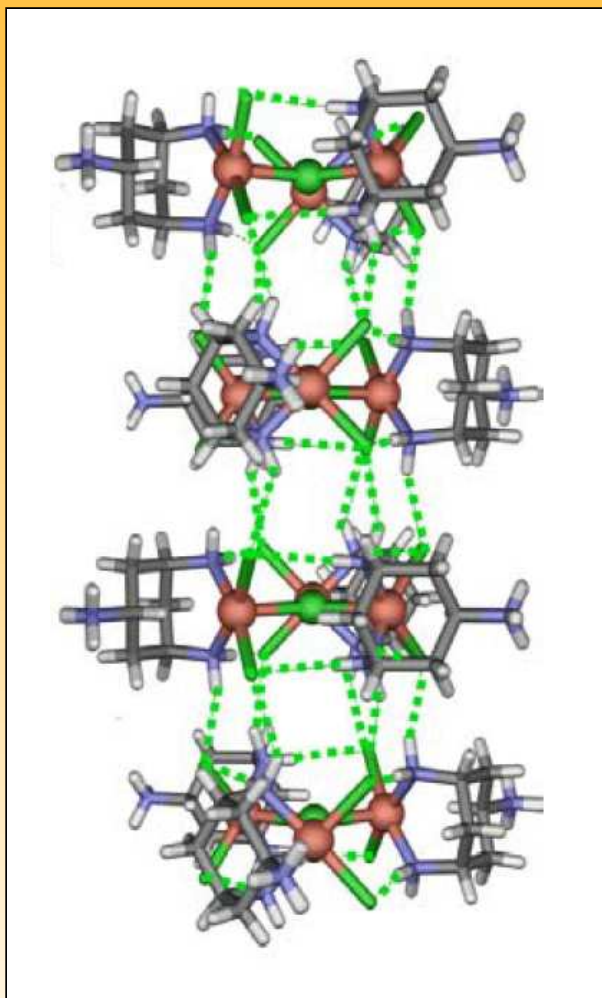
The beauty of magnetic molecules I



Mn₁₂

- 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.**

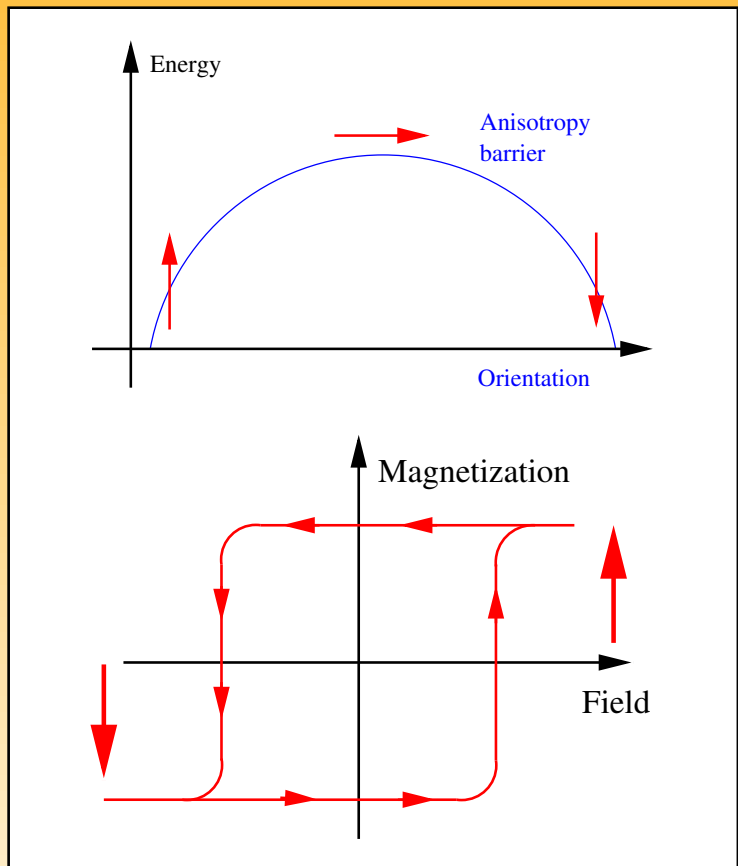
The beauty of magnetic molecules II



- 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)

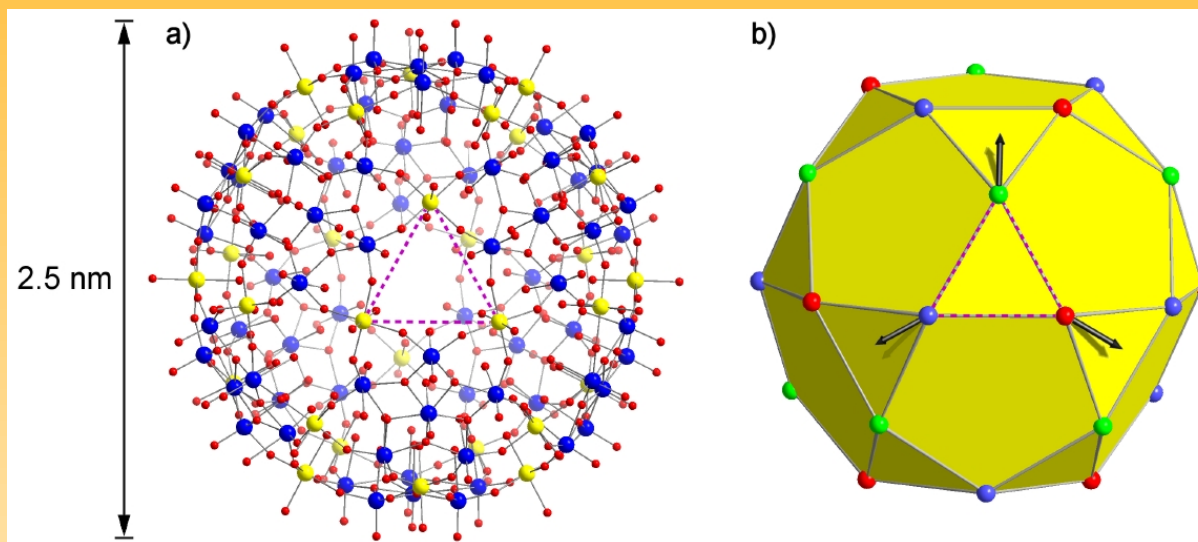
The beauty of magnetic molecules III



- 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

{Mo₇₂Fe₃₀} – 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

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

Exchange/Anisotropy
Dzyaloshinskii-Moriya
Zeeman

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

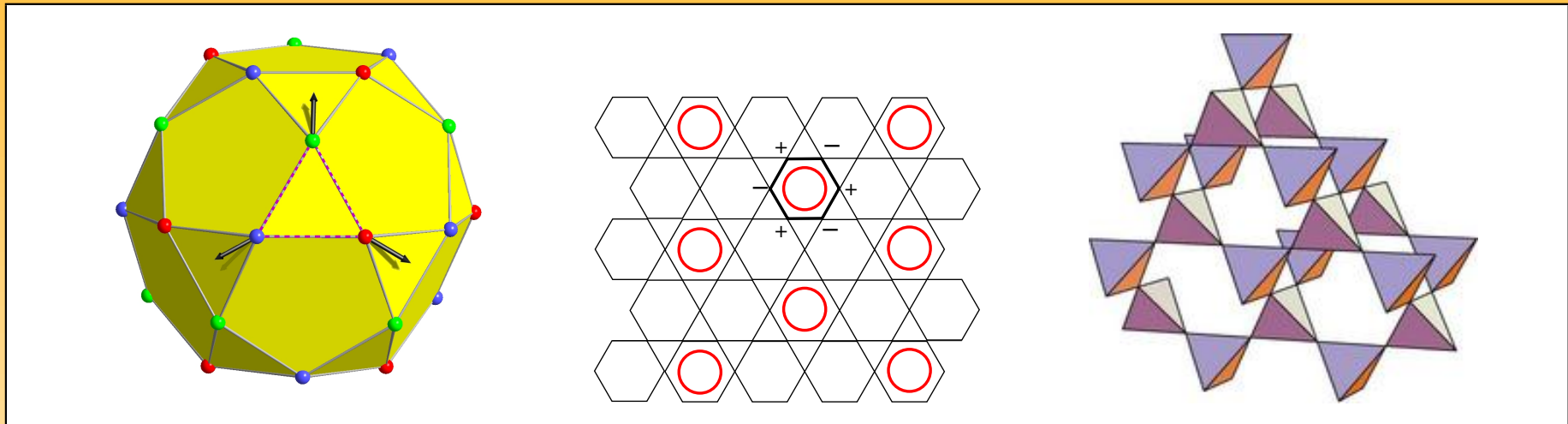
$$\tilde{H} = - \sum_{i,j} J_{ij} \vec{\tilde{s}}(i) \cdot \vec{\tilde{s}}(j) + g \mu_B B \sum_i^N \tilde{s}_z(i)$$

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



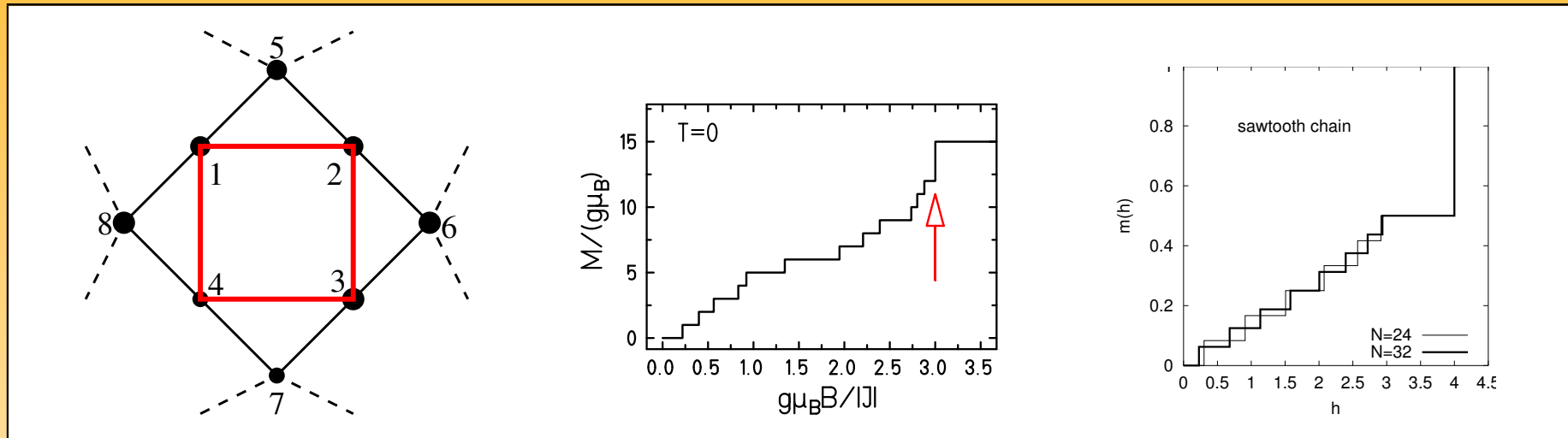
- Several frustrated antiferromagnets show an unusual behavior at the saturation field (1,2). AF = $\uparrow\downarrow$, saturation = $\uparrow\uparrow\uparrow\uparrow \dots$
- Example systems: icosidodecahedron, kagome lattice, pyrochlore lattice.

(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



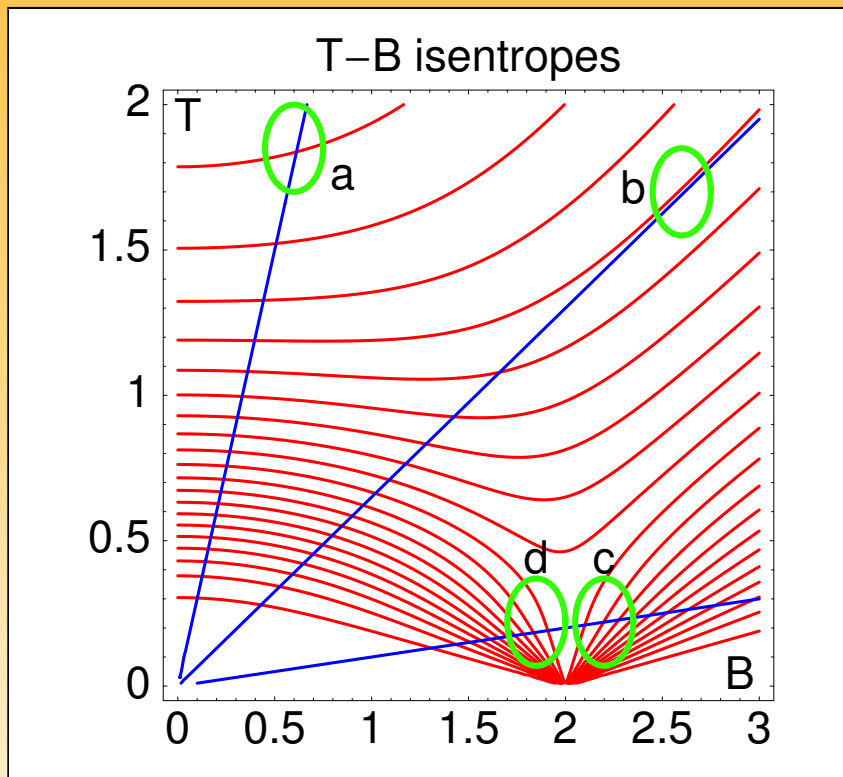
- Usually a magnetization curve is rather smooth.
- Unusually high magnetization jump at the saturation field.
- Many identical **localized independent magnons** flip their spins simultaneously.

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.

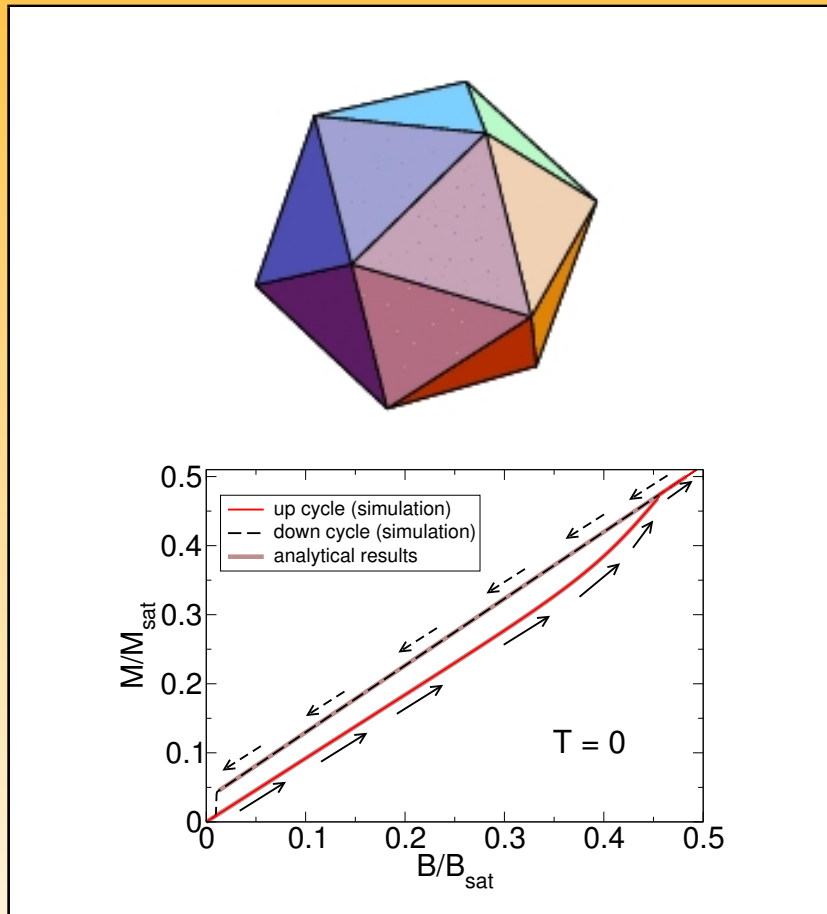
J. Schnack, J. Low Temp. Phys. **142** (2006) 279

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

Hysteresis without Anisotropy

Metamagnetic phase transition I

Hysteresis without anisotropy

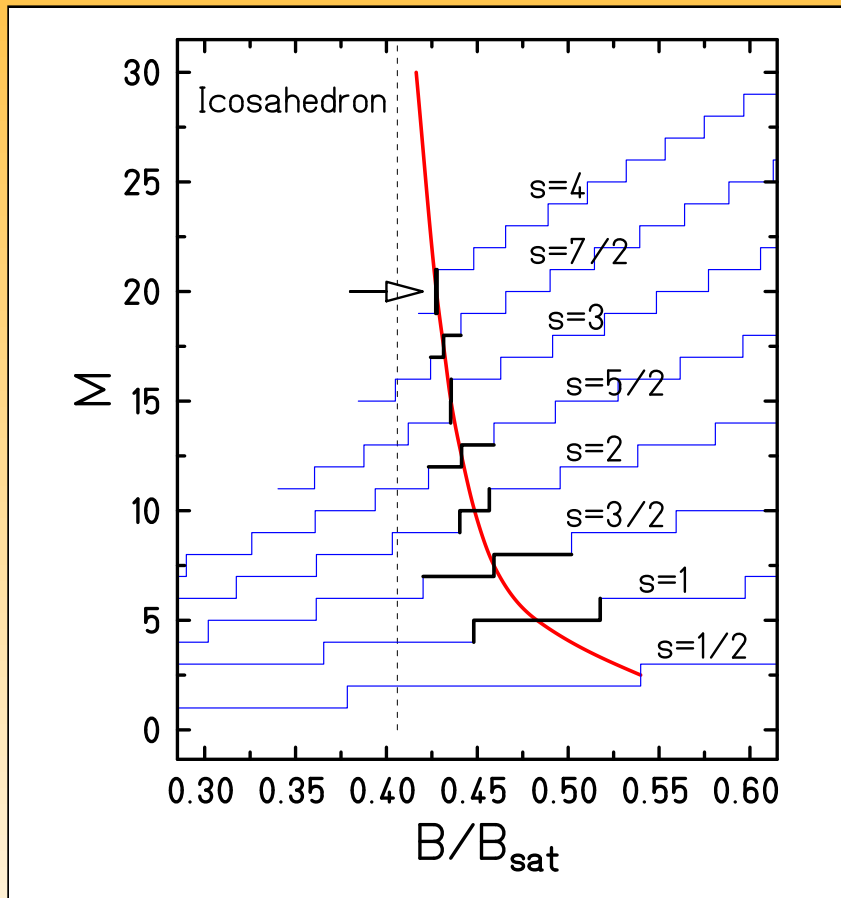


- Hysteresis is usually caused by anisotropy
- Hysteresis behavior of the classical isotropic Heisenberg icosahedron in an applied magnetic field.
- Classical spin dynamics simulations (thick lines + movie).
- Analytical stability analysis (grey lines).

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
⇒ 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

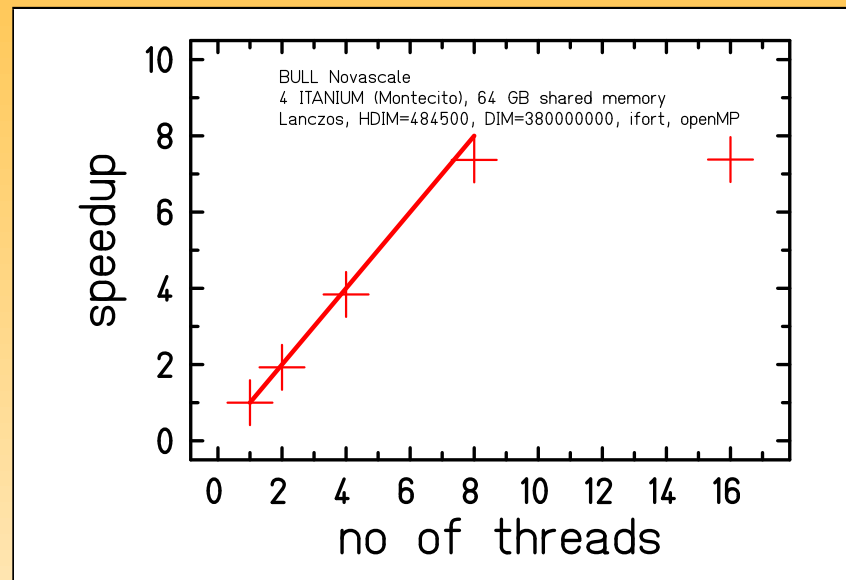
```

schnack@singlet:~/hpc
File Edit View Terminal Tabs Help
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
    
```


Parallelization issues I

Improvement of Lanczos diagonalization for very large system size:



- System size: $10^{10} \dots 10^{12}$ 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).

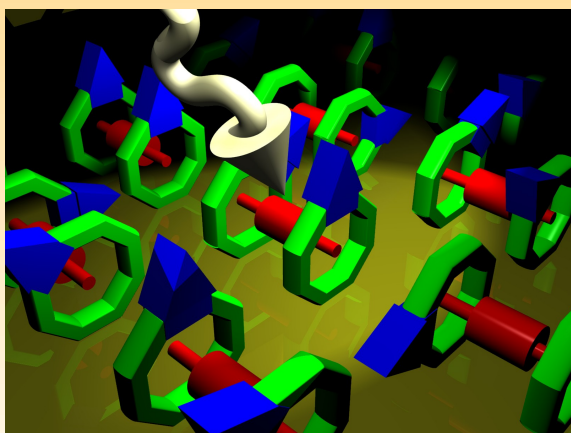
(1) J. Schnack, P. Hage, H.-J. Schmidt, arXiv:0706.3293v1 [cond-mat.str-el]



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 $10^{10} \dots 10^{12}$ 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