

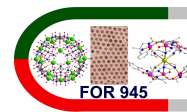
Quasi exact few-body quantum magnetism

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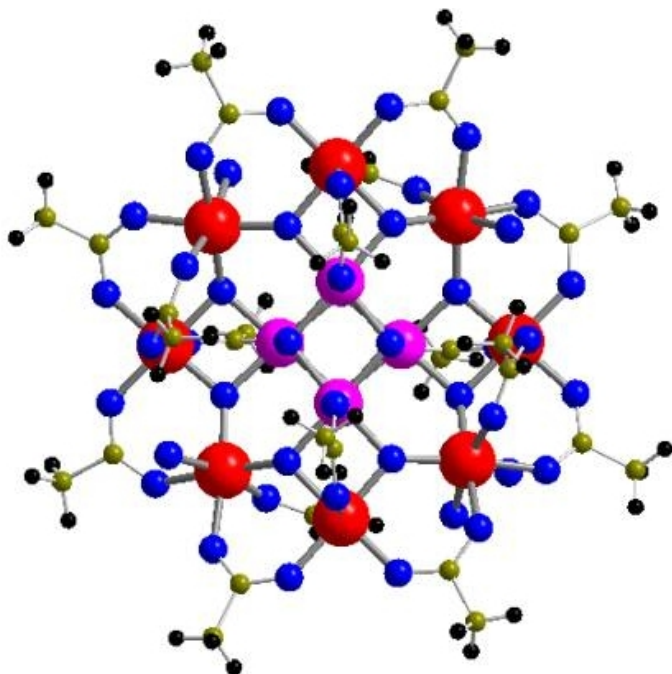
<http://obelix.physik.uni-bielefeld.de/~schnack/>

Quantum Chemistry Colloquia
Chemistry, TU Berlin, 12. 5. 2017



Beauty of Magnetic Molecules

The beauty of magnetic molecules I

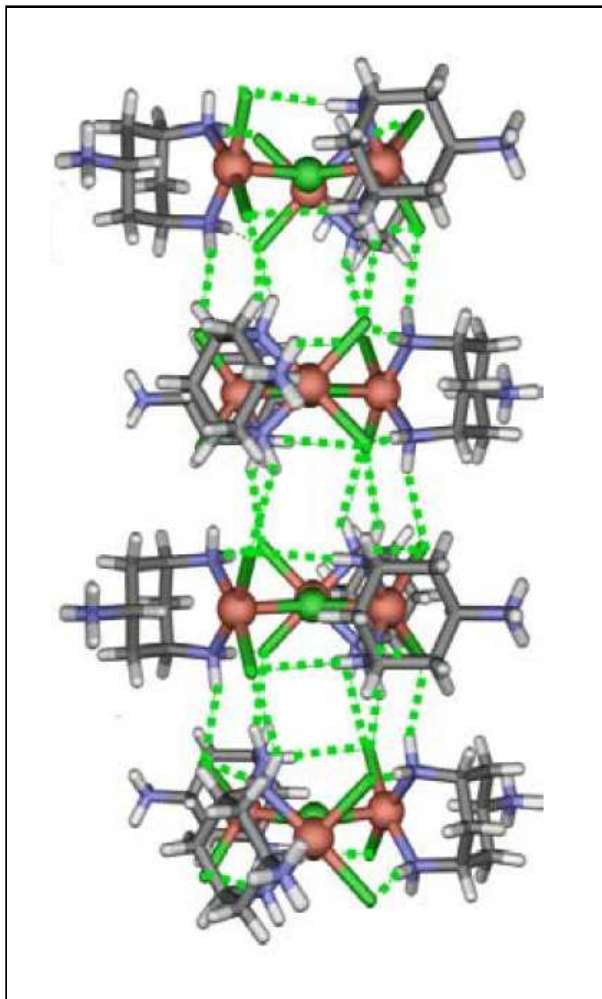


Mn₁₂

- Inorganic or organic macro molecules, e.g. polyoxometalates, 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);
- Single spin quantum number $1/2 \leq s \leq 7/2$;
- Intermolecular interaction relatively small, therefore measurements reflect the thermal behaviour of a single molecule.

Magnetism goes Nano, Ed. Stefan Blügel, Thomas Brückel, and Claus M. Schneider, FZ Jülich, Institute of Solid State Research, Lecture Notes **36** Jülich 2005

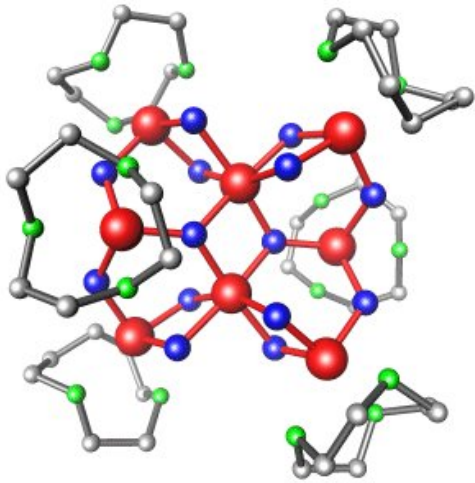
The beauty of magnetic molecules II



- Dimers (Fe_2), tetrahedra (Cr_4), cubes (Cr_8);
- Rings, especially iron rings (Fe_6 , Fe_8 , Fe_{10} , ...);
- 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:

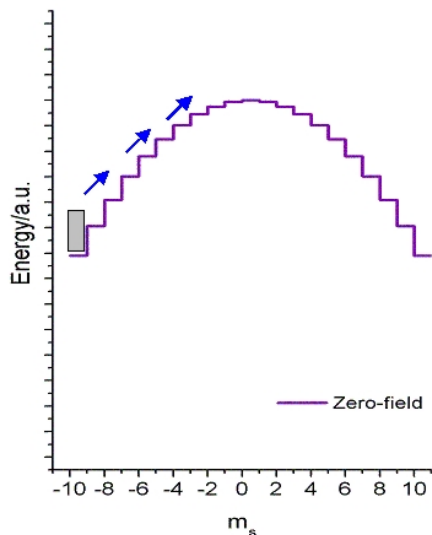
J. Schnack, H. Nojiri, P. Kögerler, G. J. T. Cooper, L. Cronin, Phys. Rev. B 70, 174420 (2004)

The beauty of magnetic molecules III



- Single Molecule Magnets (SMM): magnetic molecules with large ground state moment;
- Example: $S = 10$ for Mn_{12} or Fe_8 ;
- Anisotropy dominates approximate single-spin Hamiltonian:

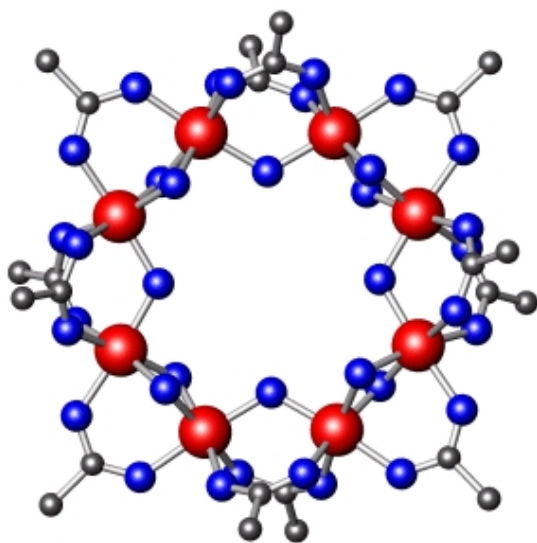
$$\underline{H} = -D\underline{S}_z^2 + \underline{H}', \quad [\underline{S}_z, \underline{H}'] \neq 0$$



- Single molecule shows: metastable magnetization, hysteresis, ground state magnetization tunneling, thermally and phonon assisted tunneling.
- **Today's major efforts: improve stability of magnetization; investigate on surfaces.**

The beauty of magnetic molecules IV

Why magnetic molecules?



Cr₈

- Interacting spin system largely decoupled from remaining degrees of freedom;
- Transition few-spin system \Rightarrow many-spin system, contribution to understanding of bulk magnetism; investigation of frustration effects;
- Transition quantum spin system ($s = 1/2$) \Rightarrow classical spin system ($s_{\text{Fe}} = 5/2$, $s_{\text{Gd}} = 7/2$);
- Easy to produce, single crystals with $> 10^{17}$ identical molecules can be synthesized and practically completely characterized;
- **Speculative applications: magnetic storage devices, magnets in biological systems, light-induced nano switches, displays, catalysts, qubits for quantum computers.**

Contents for you today



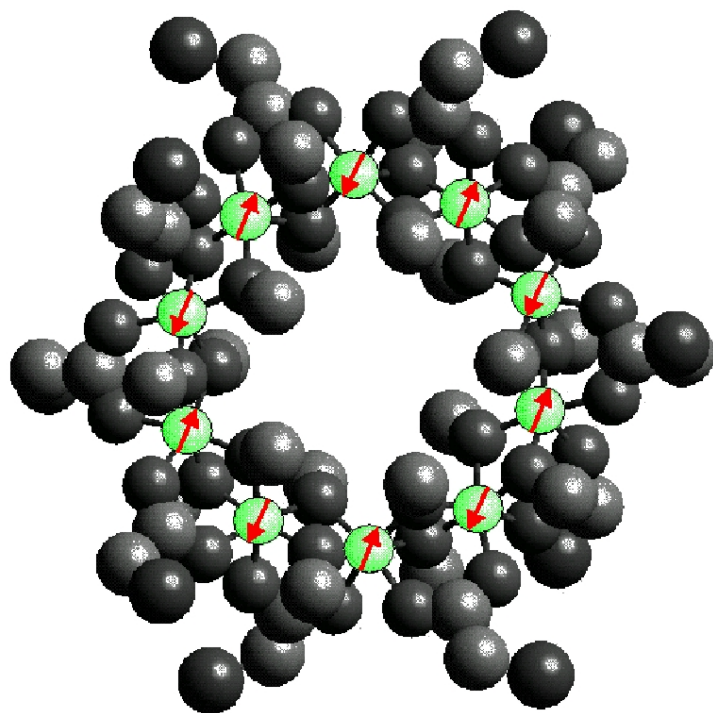
$$\begin{pmatrix} 3 & 42 & 4711 \\ 42 & 0 & 3.14 \\ 4711 & 3.14 & 8 \\ -17 & 007 & 13 \\ 1.8 & 15 & 081 \end{pmatrix}$$

1. Beauty of magnetic molecules ✓
2. **Some basic theory**
3. Finite-Temperature Lanczos
4. **QMC: intermolecular interactions**
5. NRG: deposited molecules

We are the sledgehammer team of matrix diagonalization.
Please send inquiries to jschnack@uni-bielefeld.de!

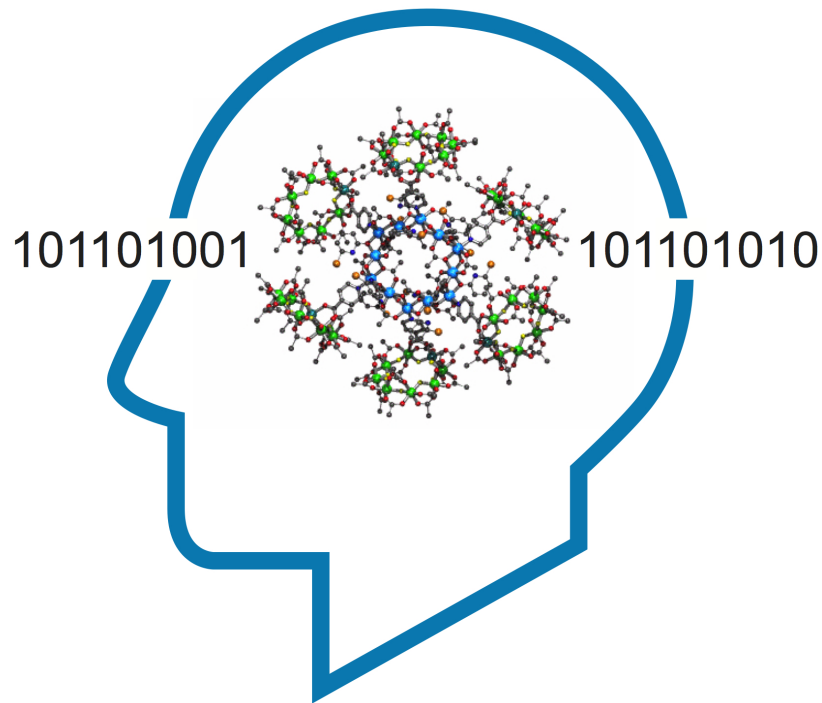
Some basic theory

You have got a molecule!



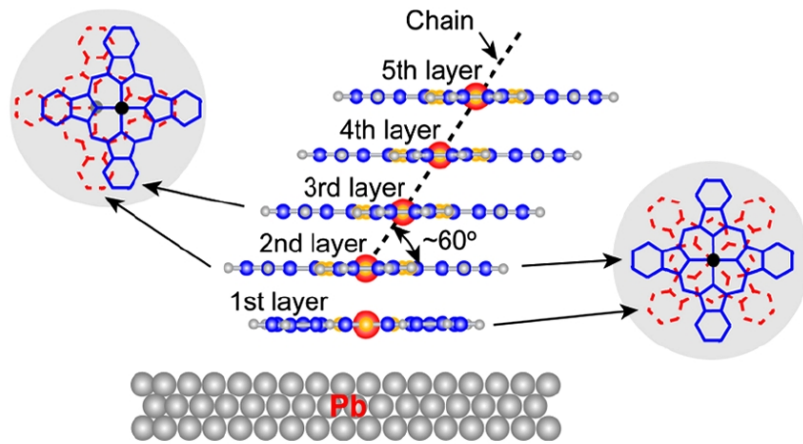
Congratulations!

You want to build a quantum computer!



Very smart!

You want to deposit your molecule!



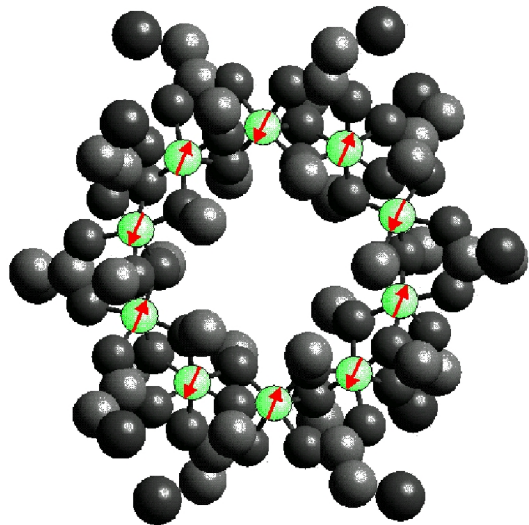
Next generation magnetic storage!

You have got an idea about the modeling!

$$\tilde{H} = -2 \sum_{i < j} J_{ij} \vec{\tilde{s}}(i) \cdot \vec{\tilde{s}}(j) + g \mu_B B \sum_i^N s_z(i)$$

Heisenberg

Zeeman



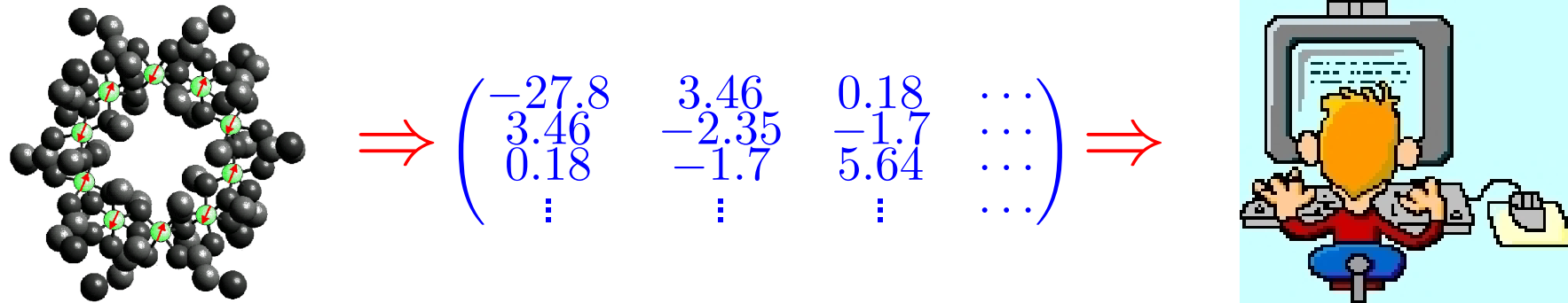
You have to solve the Schrödinger equation!

$$\underline{H} | \phi_n \rangle = E_n | \phi_n \rangle$$

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).

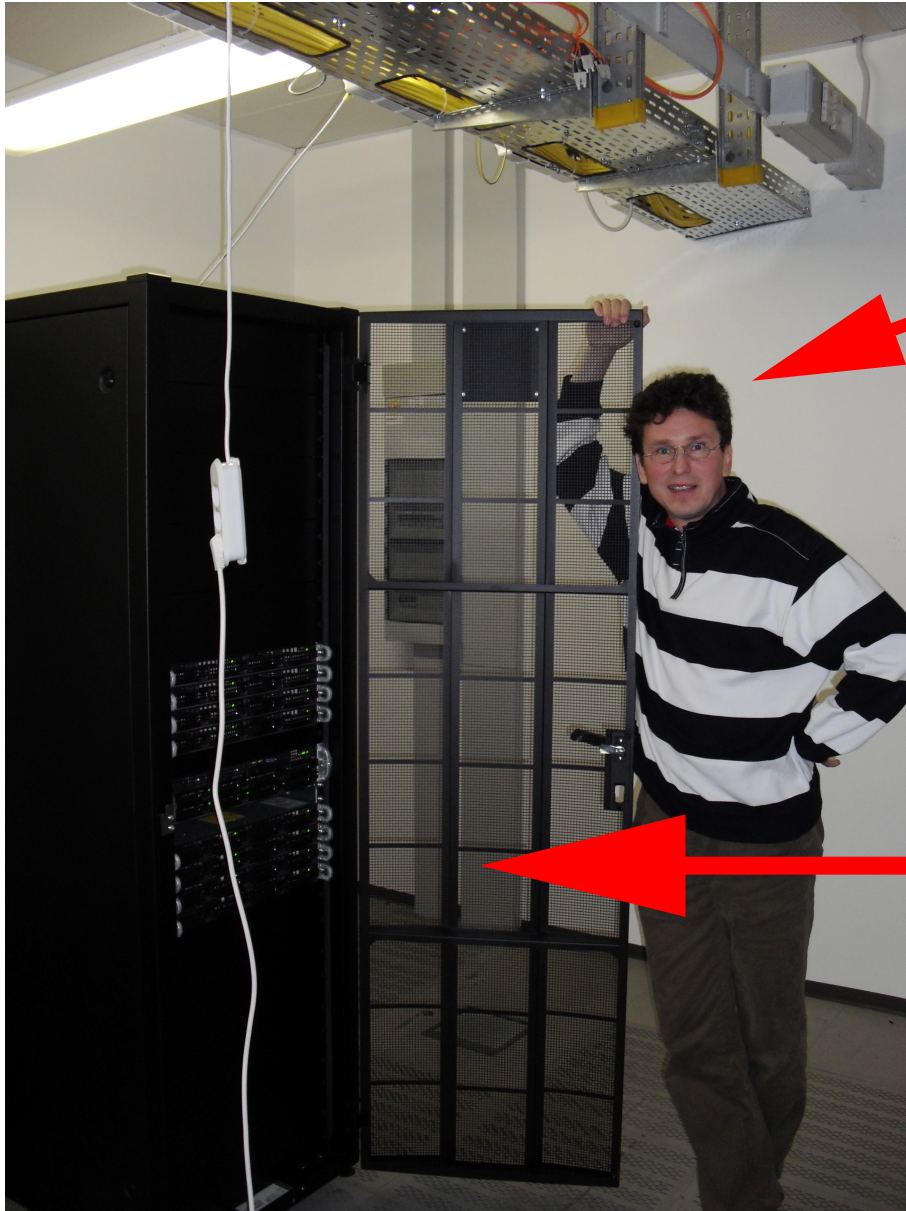
In the end it's always a big matrix!



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

Dimension=**60,466,176**. Maybe **too** big?

Thank God, we have computers



“Espresso-doped multi-core”

128 cores, 384 GB RAM

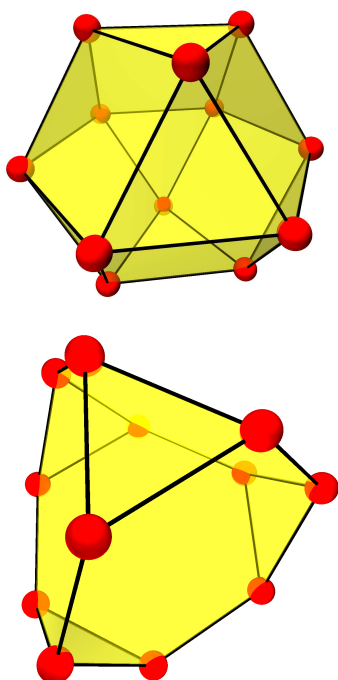
... but that's not enough!

Complete diagonalization: SU(2) & point group symmetry

Quantum chemists need to be much smarter since they have smaller computers!

- (1) D. Gatteschi and L. Pardi, *Gazz. Chim. Ital.* **123**, 231 (1993).
- (2) J. J. Borrás-Almenar, J. M. Clemente-Juan, E. Coronado, and B. S. Tsukerblat, *Inorg. Chem.* **38**, 6081 (1999).
- (3) B. S. Tsukerblat, *Group theory in chemistry and spectroscopy: a simple guide to advanced usage*, 2nd ed. (Dover Publications, Mineola, New York, 2006).

Irreducible Tensor Operator approach



Spin rotational symmetry SU(2):

- $\underline{H} = -2 \sum_{i < j} J_{ij} \underline{\tilde{S}}_i \cdot \underline{\tilde{S}}_j + g\mu_B \underline{\tilde{S}} \cdot \underline{B}$;
- Physicists employ: $[\underline{H}, \underline{S}_z] = 0$;
- Chemists employ: $[\underline{H}, \underline{S}^2] = 0, [\underline{H}, \underline{S}_z] = 0$;

Irreducible Tensor Operator (ITO) approach;
Free program MAGPACK (2) available.

- (1) D. Gatteschi and L. Pardi, *Gazz. Chim. Ital.* **123**, 231 (1993).
- (2) J. J. Borrás-Almenar, J. M. Clemente-Juan, E. Coronado, and B. S. Tsukerblat, *Inorg. Chem.* **38**, 6081 (1999).
- (3) B. S. Tsukerblat, *Group theory in chemistry and spectroscopy: a simple guide to advanced usage*, 2nd ed. (Dover Publications, Mineola, New York, 2006).

Idea of ITO

$$\begin{aligned}
 \underline{H}_{\text{Heisenberg}} &= -2 \sum_{i < j} J_{ij} \vec{s}_i \cdot \vec{s}_j \\
 &= 2\sqrt{3} \sum_{i < j} J_{ij} T_{\vec{s}}^{(0)}(\{k_m\}, \{\bar{k}_m\} | k_i = k_j = 1)
 \end{aligned}$$

Irreducible Tensor Operator approach

- Express spin operators and functions thereof as ITOs;
- Use vector coupling basis $|\alpha S M\rangle$ and recursive recoupling.

(1) Gatteschi, Tsukerblat, Coronado, Waldmann, ...
 (2) R. Schnalle, Ph.D. thesis, Osnabrück University (2009)

Point Group Symmetry

$$|\alpha' S M \Gamma\rangle = \mathcal{P}^{(\Gamma)} |\alpha S M\rangle = \left(\frac{l_{\Gamma}}{h} \sum_R \left(\chi^{(\Gamma)}(R) \right)^* \mathcal{G}(R) \right) |\alpha S M\rangle$$

Method:

- Projection onto irreducible representations Γ of the point group (1,2);
- No free program, things are a bit complicated (3,4).

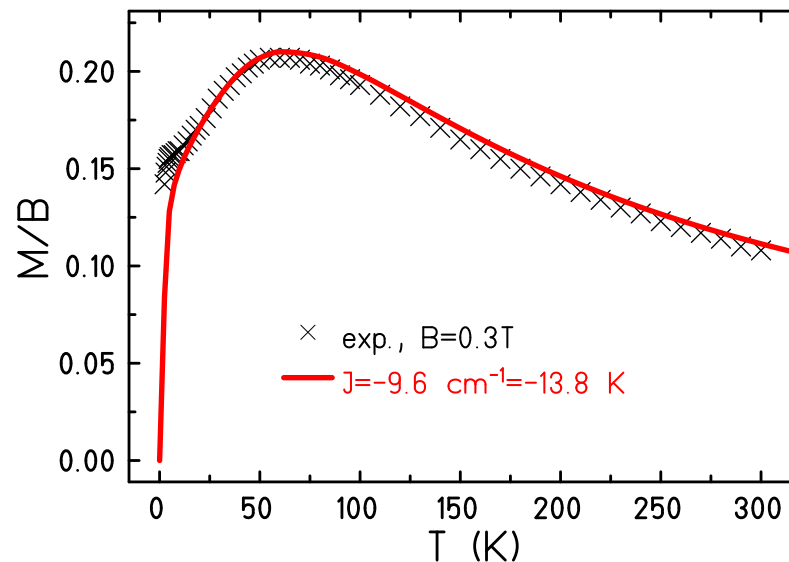
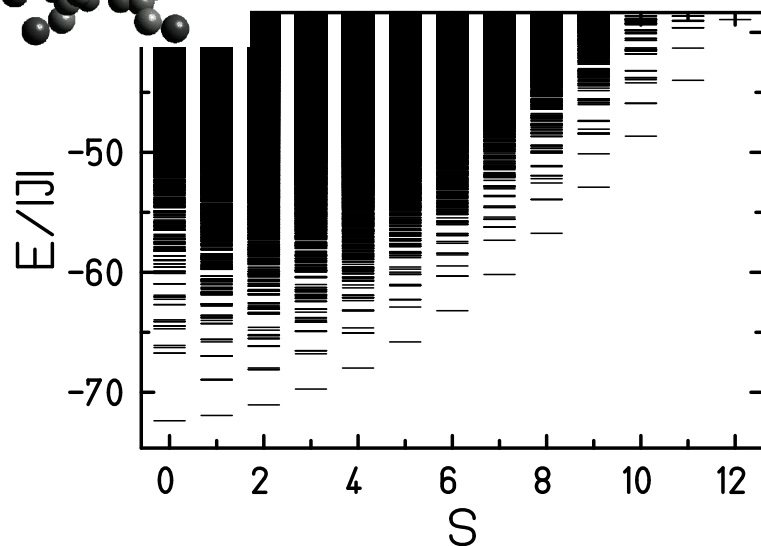
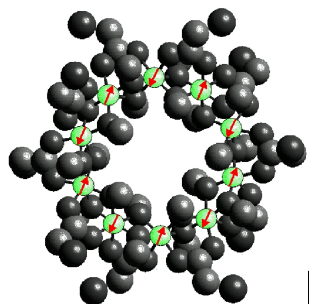
(1) M. Tinkham, *Group Theory and Quantum Mechanics*, Dover.

(2) D. Gatteschi and L. Pardi, *Gazz. Chim. Ital.* **123**, 231 (1993).

(3) O. Waldmann, *Phys. Rev. B* **61**, 6138 (2000).

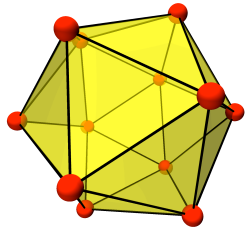
(4) R. Schnalle and J. Schnack, *Int. Rev. Phys. Chem.* **29**, 403-452 (2010) \Leftarrow contains EVERYTHING.

Example: Fe₁₀

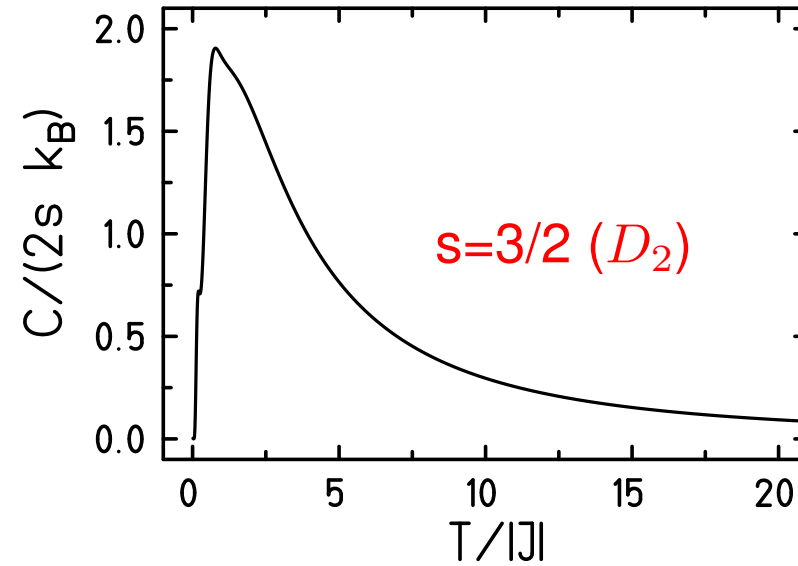
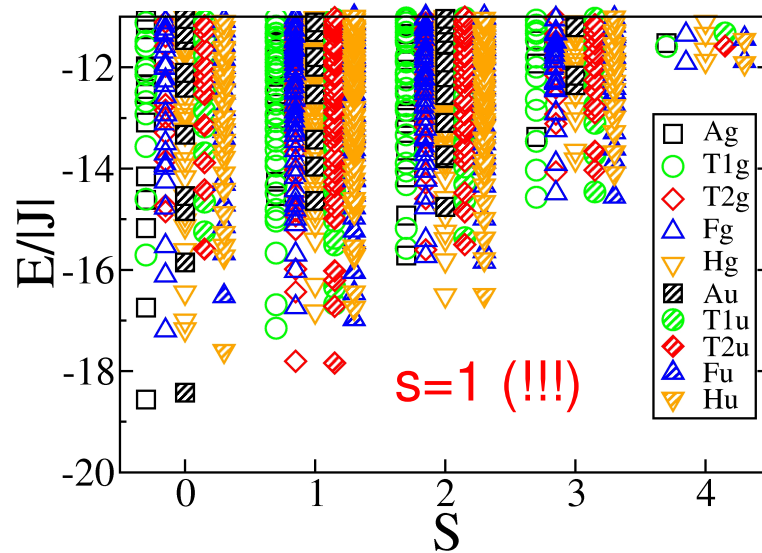


Spin ring, $N = 10$, $s = 5/2$, Hilbert space dimension 60,466,176; symmetry D_2 (1).

- (1) R. Schnalle and J. Schnack, *Int. Rev. Phys. Chem.* **29**, 403-452 (2010).
- (2) C. Delfs *et al.*, *Inorg. Chem.* **32**, 3099 (1993).



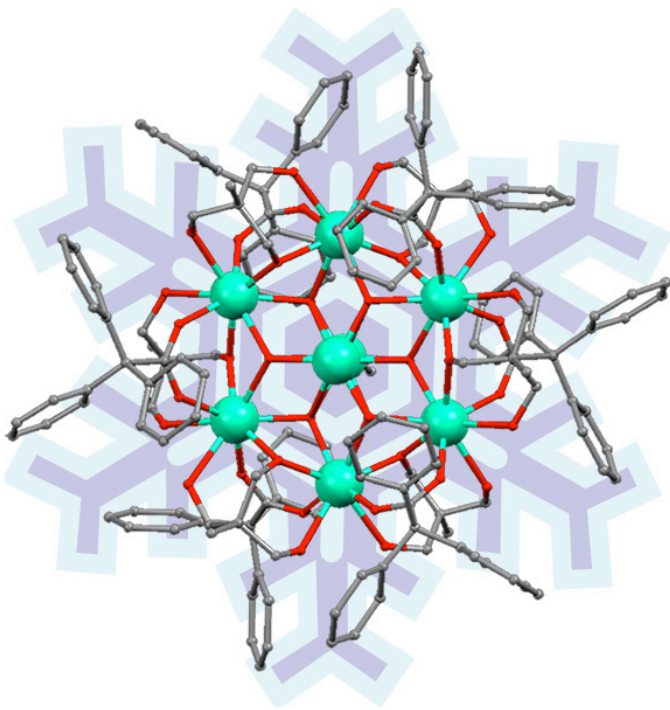
Example: Icosahedron



Icosahedron, $s = 3/2$, Hilbert space dimension 16,777,216; symmetry I_h ; Evaluation of recoupling coefficients for $s = 3/2$ in I_h **practically impossible** (1).

(1) R. Schnalle and J. Schnack, Int. Rev. Phys. Chem. **29**, 403-452 (2010).

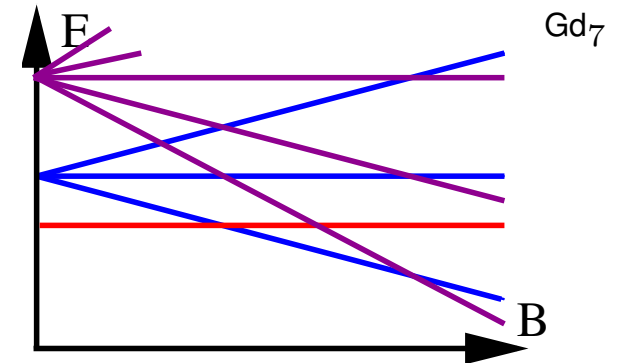
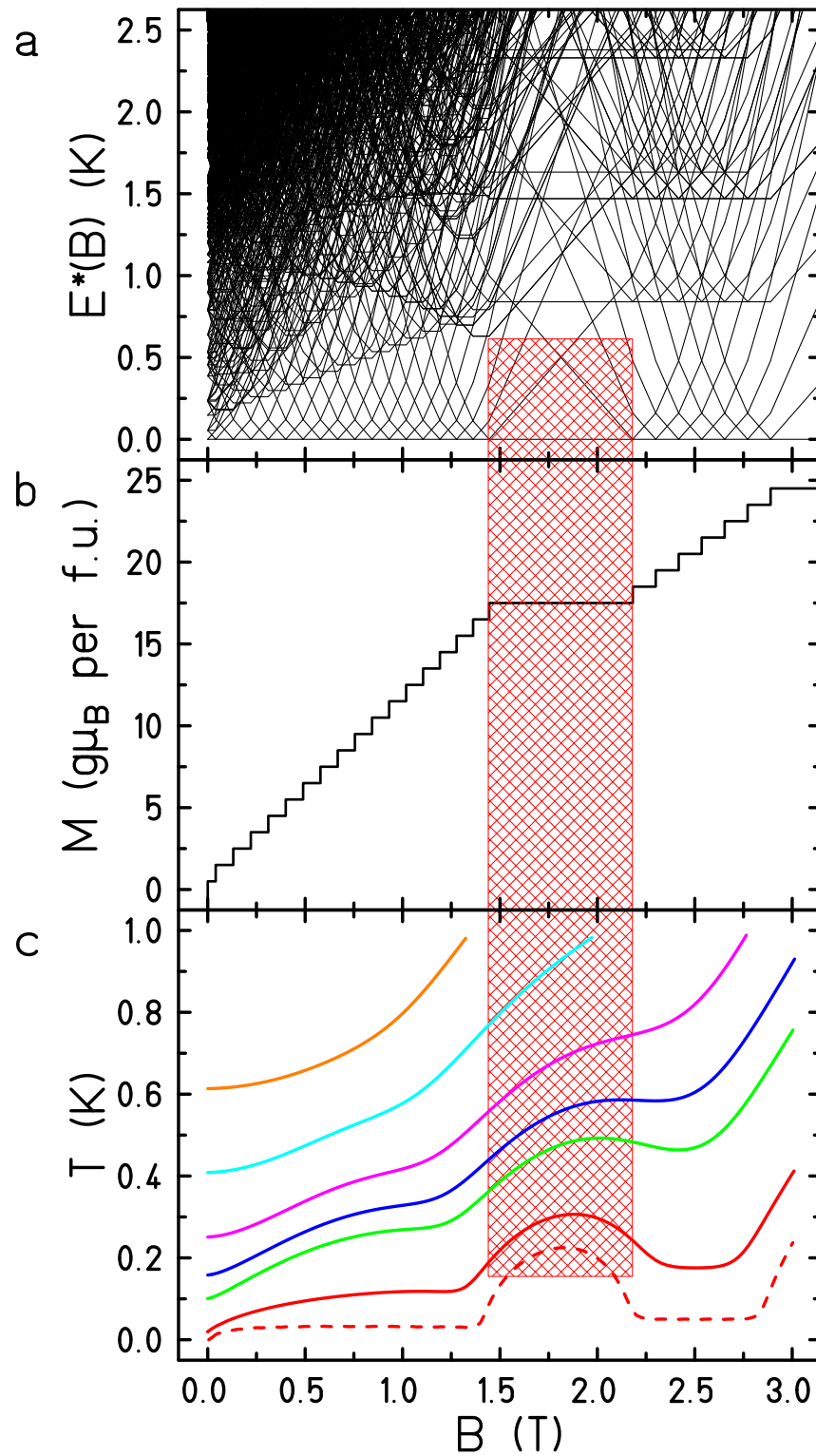
Gd₇ – Magnetocalorics



- Often magnetocaloric observables not directly measured, but inferred from Maxwell's relations.
- First real cooling experiment with a molecule.
- $$\underline{H} = -2 \sum_{i < j} J_{ij} \vec{\tilde{s}}_i \cdot \vec{\tilde{s}}_j + g \mu_B B \sum_i^N \tilde{s}_i^z$$

 $J_1 = -0.090(5) \text{ K}, J_2 = -0.080(5) \text{ K}$
 and $g = 2.02$.
- **Very good agreement down to the lowest temperatures.**

J. W. Sharples, D. Collison, E. J. L. McInnes, J. Schnack, E. Palacios, M. Evangelisti, Nat. Commun. **5**, 5321 (2014).



What if your molecule is
BIGGER?

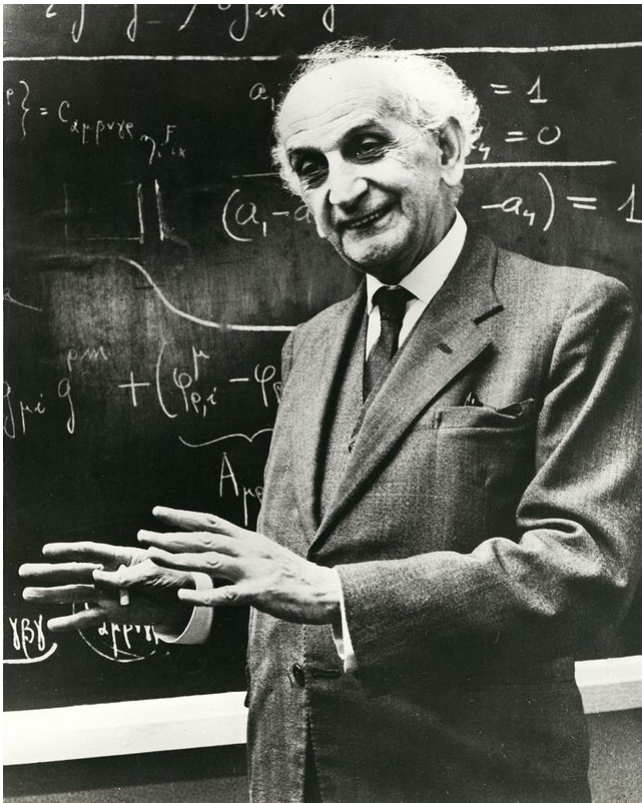


SuperMUC @ LRZ!

Finite-temperature Lanczos Method

(Good for dimensions up to 10^{10} .)

Lanczos – a Krylov space method



- Idea: exact diagonalization in reduced basis sets.
- But which set to choose???
- Idea: generate the basis set with the operator you want to diagonalize:
 $\{ |\phi\rangle, \tilde{H} |\phi\rangle, \tilde{H}^2 |\phi\rangle, \tilde{H}^3 |\phi\rangle, \dots \}$
- But which starting vector to choose???
- Idea: almost any will do!
- Cornelius Lanczos (Lánczos Kornél, 1893-1974)

(1) C. Lanczos, J. Res. Nat. Bur. Stand. **45**, 255 (1950).

Finite-temperature Lanczos Method I

$$Z(T, B) = \sum_{\nu} \langle \nu | \exp \left\{ -\beta \tilde{H} \right\} | \nu \rangle$$

$$\langle \nu | \exp \left\{ -\beta \tilde{H} \right\} | \nu \rangle \approx \sum_n \langle \nu | n(\nu) \rangle \exp \left\{ -\beta \epsilon_n \right\} \langle n(\nu) | \nu \rangle$$

$$Z(T, B) \approx \frac{\dim(\mathcal{H})}{R} \sum_{\nu=1}^R \sum_{n=1}^{N_L} \exp \left\{ -\beta \epsilon_n \right\} |\langle n(\nu) | \nu \rangle|^2$$

- $|n(\nu)\rangle$ n-th Lanczos eigenvector starting from $|\nu\rangle$
- Partition function replaced by a small sum: $R = 1 \dots 10, N_L \approx 100$.

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

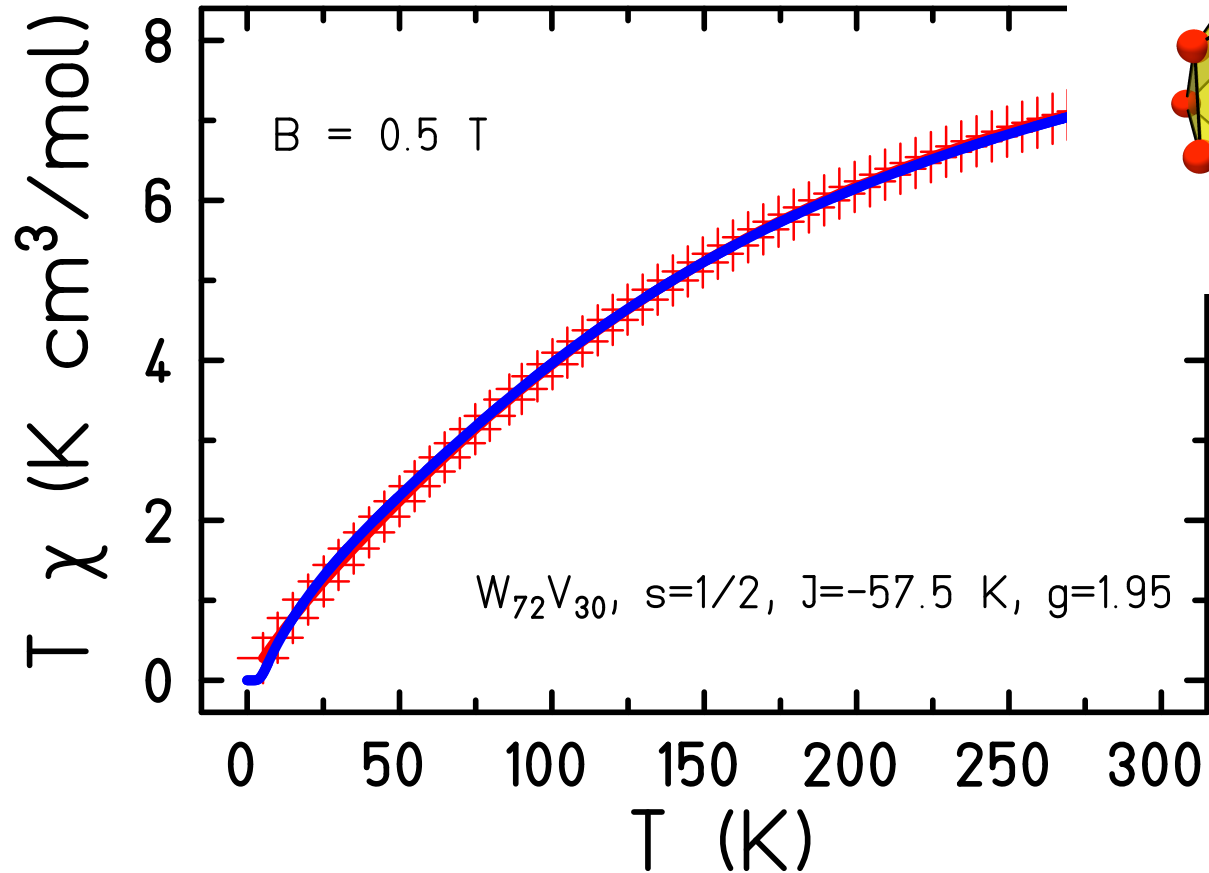
Finite-temperature Lanczos Method II

$$Z(T, B) \approx \sum_{\Gamma} \frac{\dim(\mathcal{H}(\Gamma))}{R_{\Gamma}} \sum_{\nu=1}^{R_{\Gamma}} \sum_{n=1}^{N_L} \exp\{-\beta\epsilon_n\} |\langle n(\nu, \Gamma) | \nu, \Gamma \rangle|^2$$

- Approximation better if symmetries taken into account.
- Γ denotes the used irreducible representations; often this is just the \mathcal{S}^z symmetry, i.e. $\Gamma \equiv M$

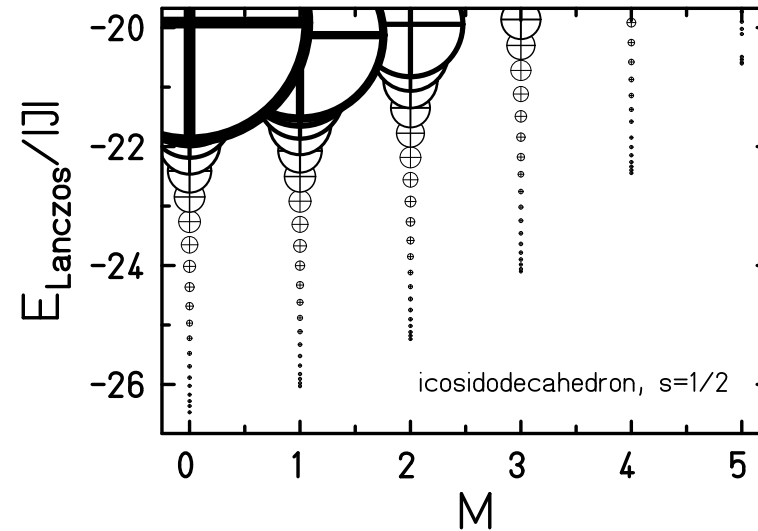
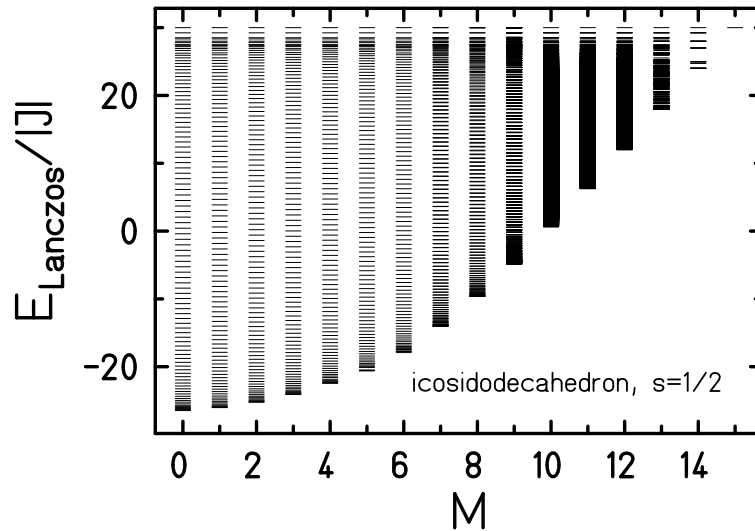
J. Schnack and O. Wendland, Eur. Phys. J. B **78** (2010) 535-541

Icosidodecahedron $s = 1/2$



Exp. data: A. M. Todea, A. Merca, H. Bögge, T. Glaser, L. Engelhardt, R. Prozorov, M. Luban, A. Müller, Chem. Commun., 3351 (2009).

Icosidodecahedron $s = 1/2$



- The true spectrum will be much denser. This is miraculously compensated for by the weights.

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

Finite-temperature Lanczos Method III

$$\tilde{H} = -2 \sum_{i < j} \vec{\tilde{S}}_i \cdot \mathbf{J}_{ij} \cdot \vec{\tilde{S}}_j + \sum_i \vec{\tilde{S}}_i \cdot \mathbf{D}_i \cdot \vec{\tilde{S}}_i + \mu_B B \sum_i g_i \tilde{S}_i^z$$

- **Problem:** for anisotropic Hamiltonians no symmetry left
→ accuracy drops (esp. for high T).
- Simple traces such as $\text{Tr} \left(\tilde{S}^z \right) = 0$ tend to be wrong for R not very big.

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

Finite-temperature Lanczos Method IV

Employ very general symmetry (time-reversal invariance)

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

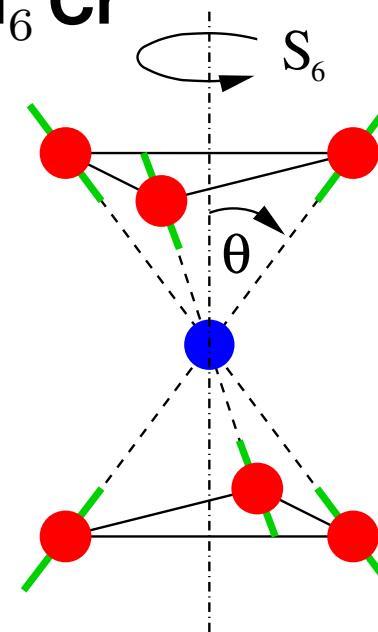
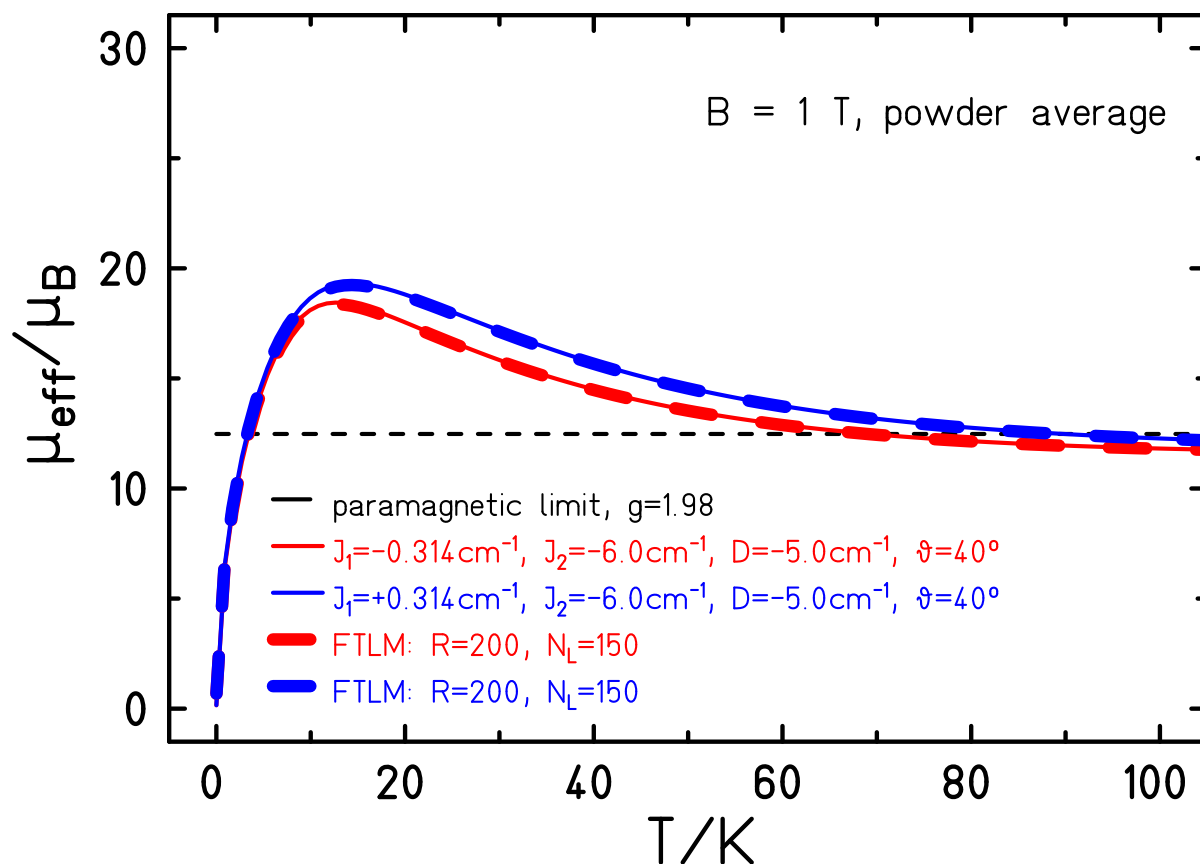
Use Lanczos energy eigenvector $|n(\nu)\rangle$ and time-reversed counterpart $|\tilde{n}(\nu)\rangle$

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

- 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: $\text{Mn}_6\text{Cr}^{\text{III}}$

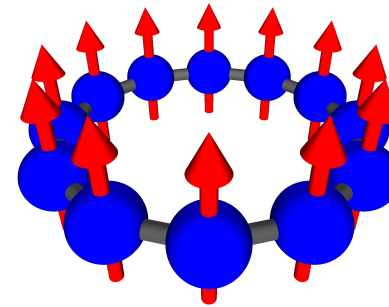
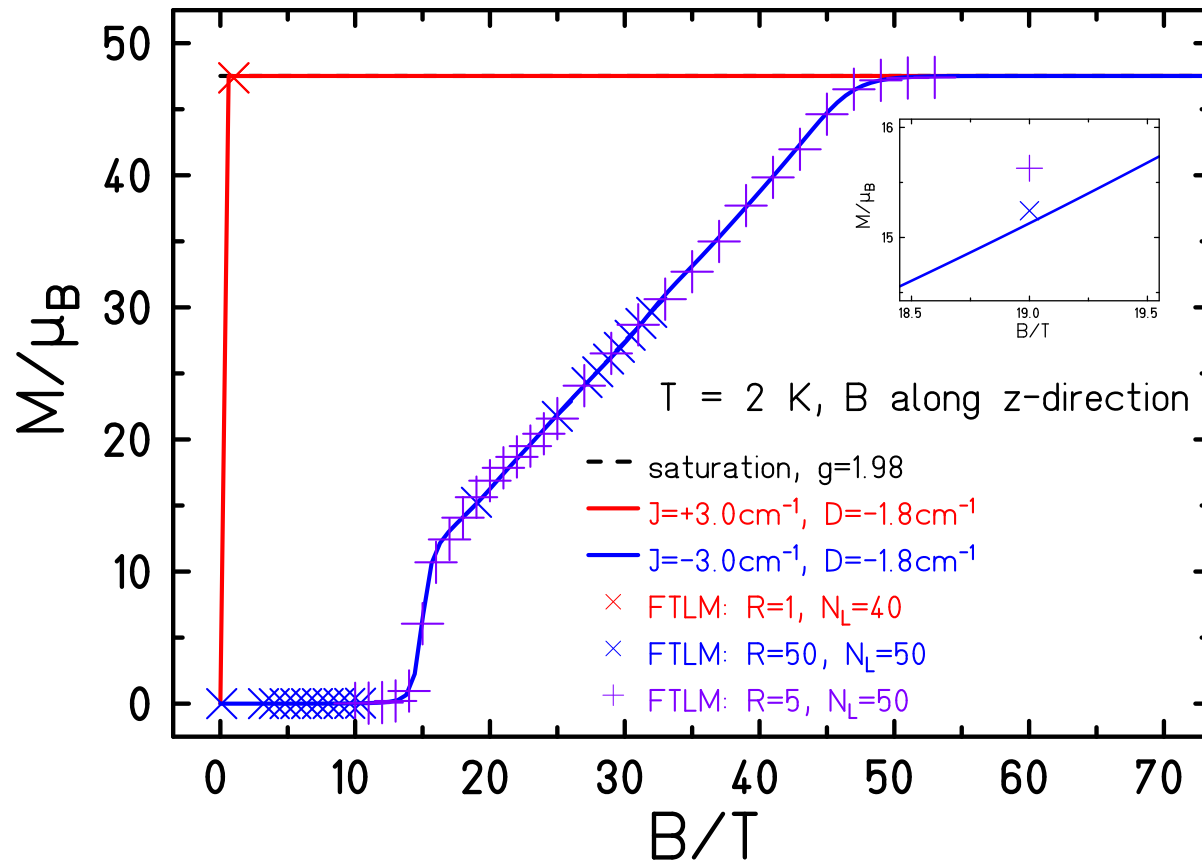


$s = 2, s = 3/2$
 $\dim(\mathcal{H}) = 62,500$
 non-collinear easy axes

Hours compared to days, notebook compared to supercomputer!

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

A fictitious $Mn_{12}^{III} - M_z$ vs B_z



$s = 2$

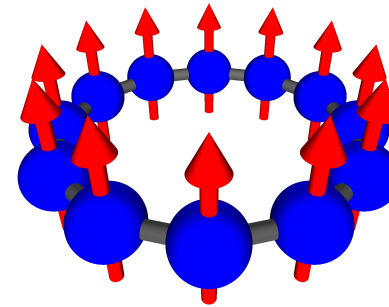
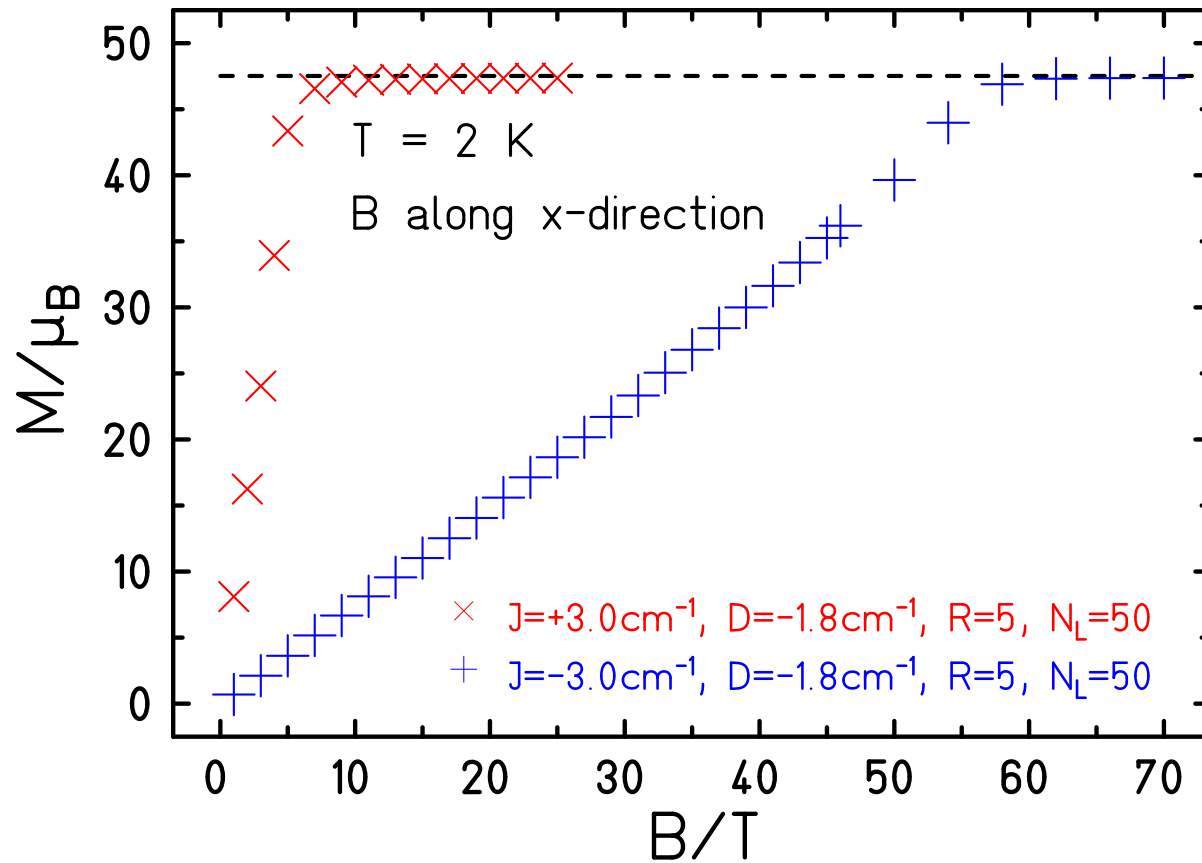
$\dim(\mathcal{H}) = 244, 140, 625$

collinear easy axes

A few days compared to *impossible!*

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

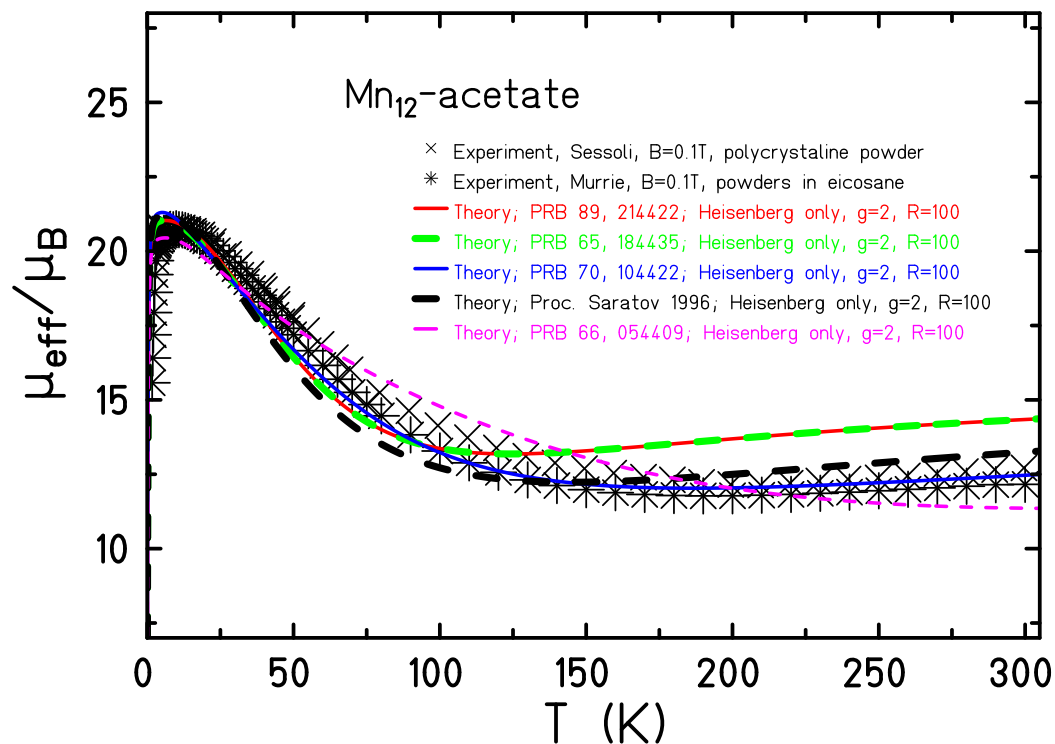
A fictitious $\text{Mn}_{12}^{\text{III}} - M_x$ vs B_x



No other method can deliver these curves!

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

Effective magnetic moment of Mn₁₂-acetate



We can check DFT parameter predictions for large molecules! **Normally!**

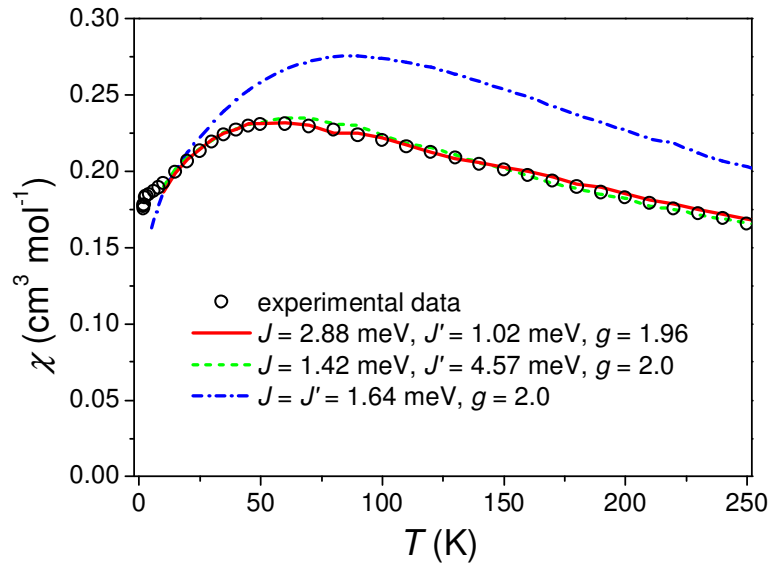
O. Hanebaum, J. Schnack, Phys. Rev. B **92** (2015) 064424



⇒ S. Ghassemi Tabrizi, A. V. Arbuznikov, and M. Kaupp, J. Phys. Chem. A **120**, 6864 (2016).

Intermolecular interactions

Quantum Monte Carlo



Chopped (sliced) partition function:

- $$Z(T, B) = \sum_{\nu} \langle \nu | \exp \left\{ -\beta \tilde{H} \right\} | \nu \rangle$$

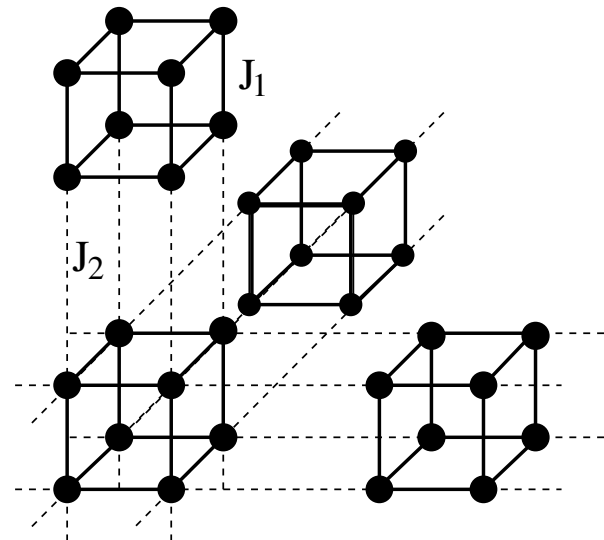
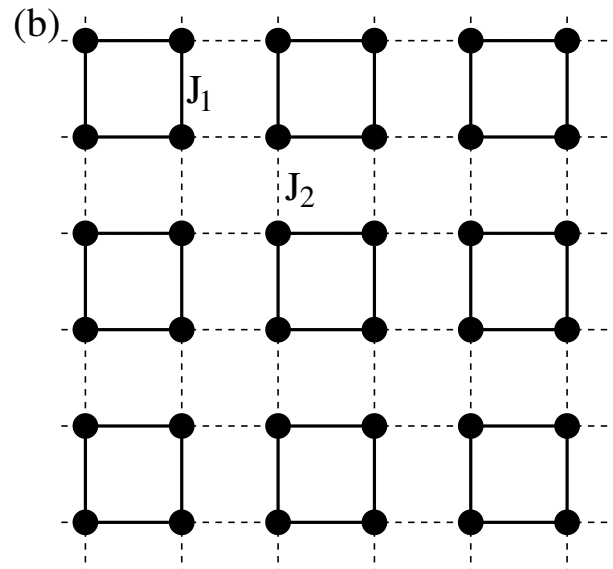
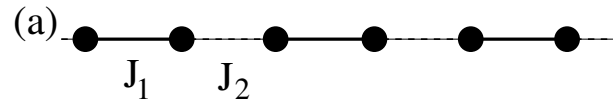
$$= \sum_{\nu} \langle \nu | \left[\exp \left\{ -\beta \tilde{H} / m \right\} \right]^m | \nu \rangle$$

$$= \sum_{\nu, \alpha, \beta, \dots} \langle \nu | \exp \left\{ -\beta \tilde{H} / m \right\} | \alpha \rangle \langle \alpha | \dots$$

$$\approx \sum_{\nu, \alpha, \beta, \dots} \langle \nu | \left\{ 1 - \beta \tilde{H} / m \right\} | \alpha \rangle \langle \alpha | \dots$$
- Bad/no convergence for frustrated systems (negative sign problem).

- (1) A. W. Sandvik and J. Kurkijärvi, Phys. Rev. B **43**, 5950 (1991).
- (2) A. W. Sandvik, Phys. Rev. B **59**, R14157 (1999).
- (3) L. Engelhardt and M. Luban, Phys. Rev. B **73**, 054430 (2006); L. Engelhardt *et al.*, Phys. Rev. B **79**, 014404 (2009).
- (4) J. Ummethum *et al.*, Phys. Rev. B **86**, 104403 (2012).

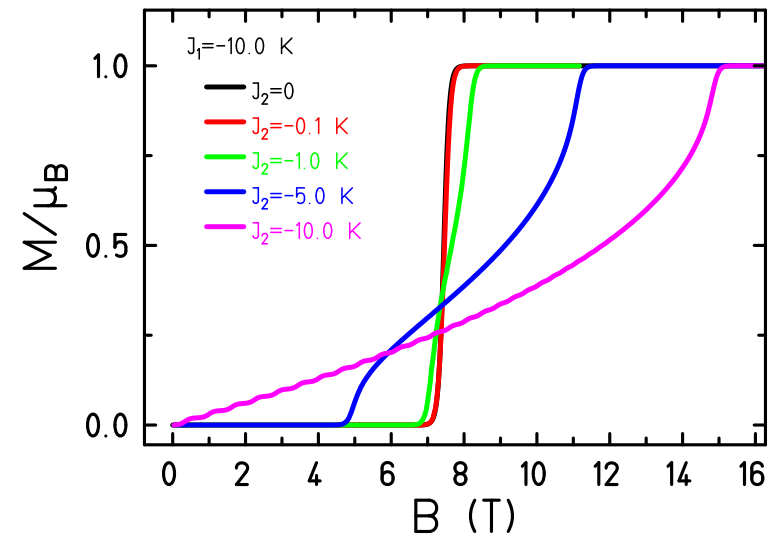
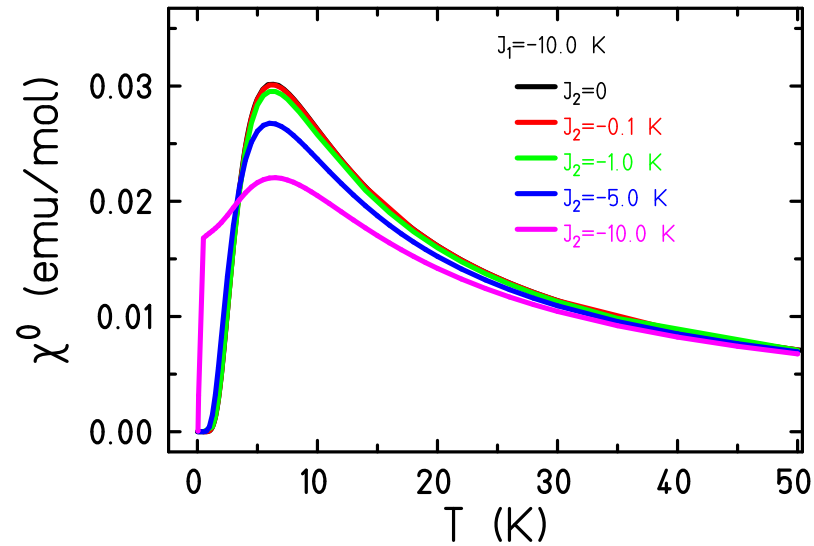
Intermolecular interactions – systems



QMC investigation: Dimers in 1-d, squares in 2-d, cubes in 3-d.
 All interactions antiferromagnetic, all spins $s = 1/2$. $N = 100 \dots 1000$. PBC.

(1) J. Schnack, Phys. Rev. B **93**, 054421 (2016).

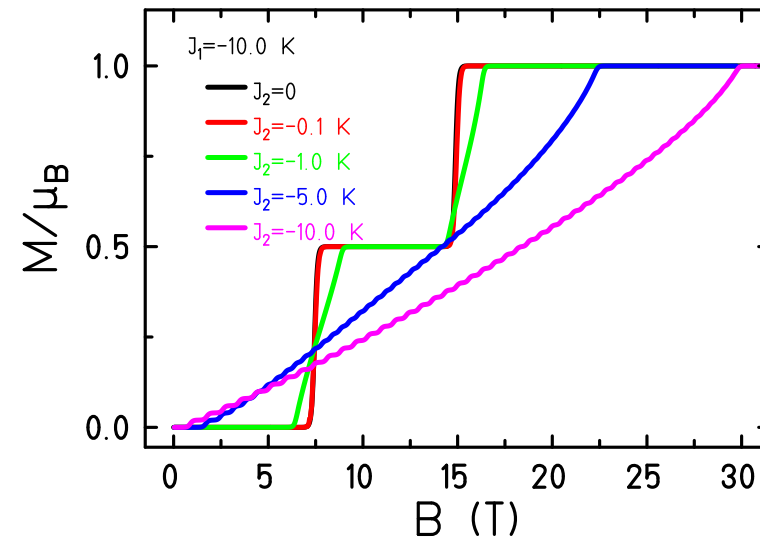
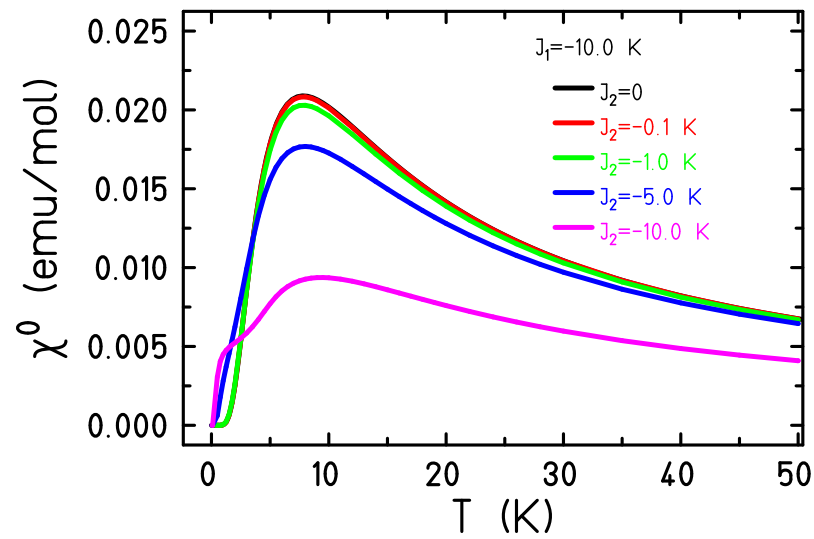
Intermolecular interactions – 1-d



Gradually af dimers turn into gapless af chain.
Even at 50 % gap still large.

(1) J. Schnack, Phys. Rev. B **93**, 054421 (2016).

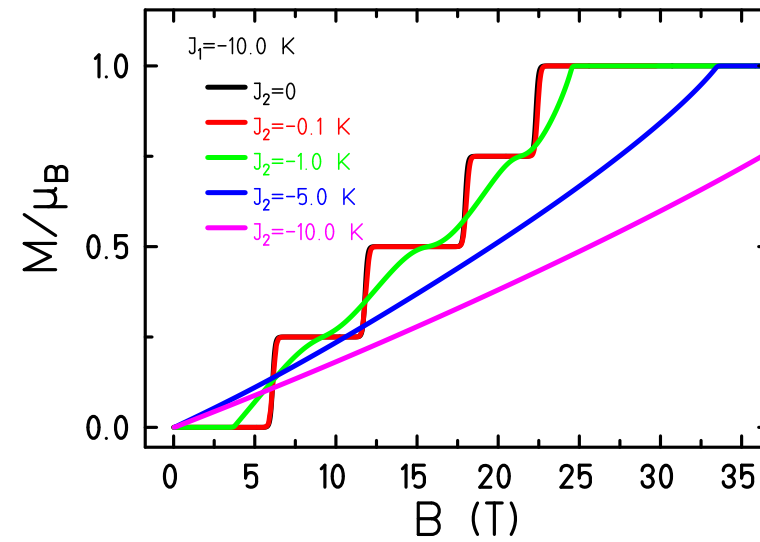
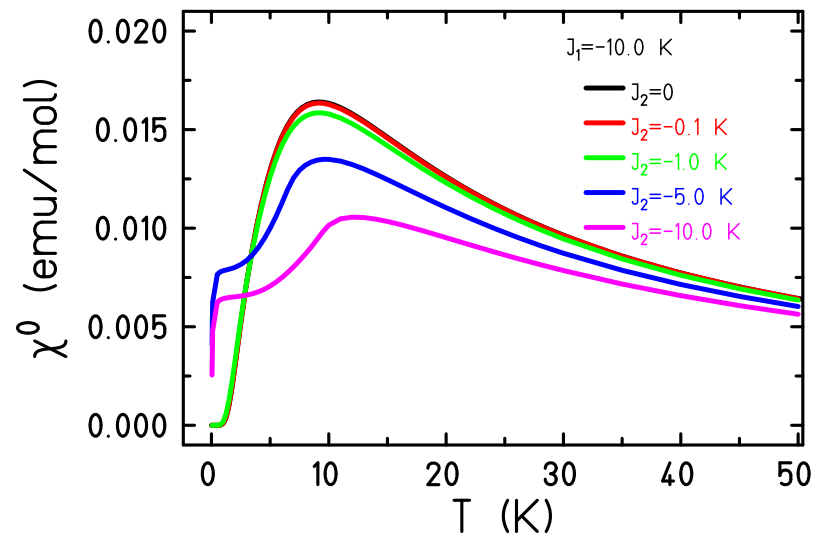
Intermolecular interactions – 2-d



The two magnetization steps vanish more rapidly with intermolecular interactions in 2-d.

(1) J. Schnack, Phys. Rev. B **93**, 054421 (2016).

Intermolecular interactions – 3-d



In 3-d 10 % intermolecular interactions sufficient to wash out magnetization steps.

(1) J. Schnack, Phys. Rev. B **93**, 054421 (2016).

Take home:

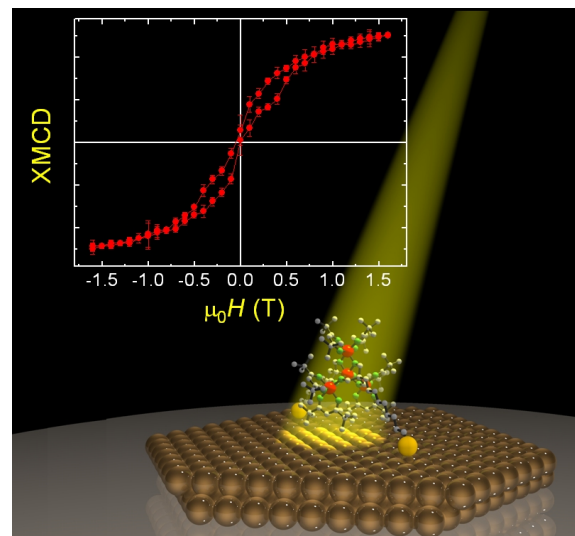
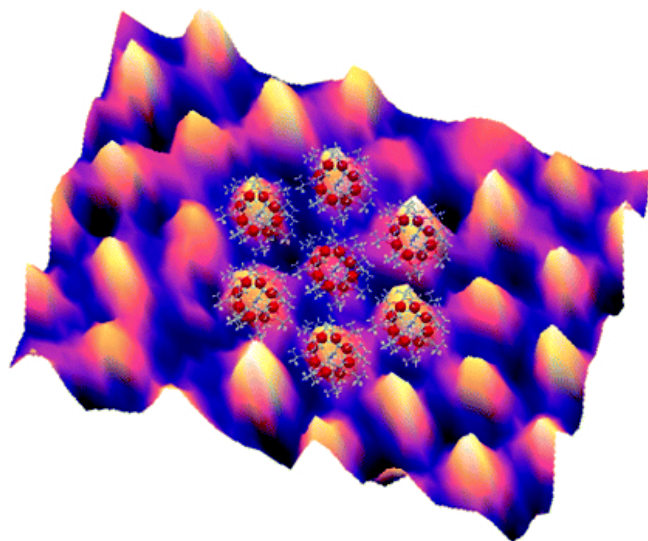
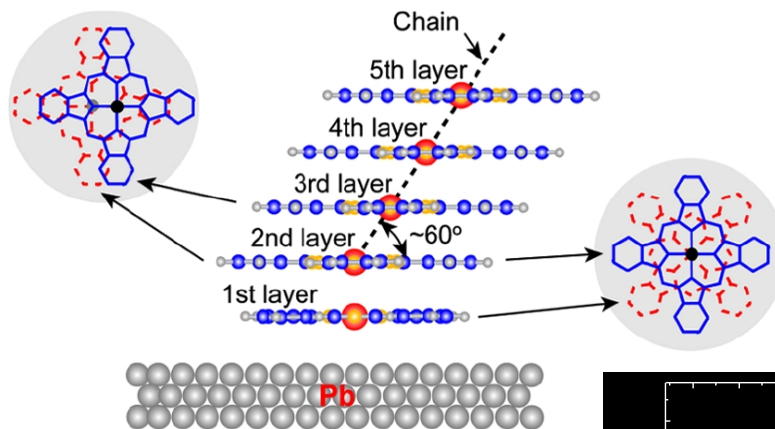
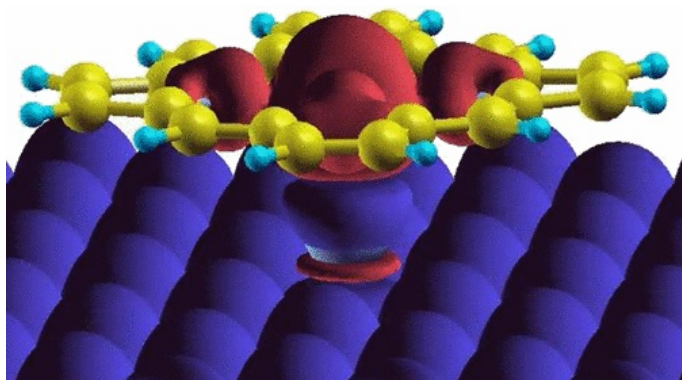
10 % intermolecular interactions in 3-d is enough

(...to spoil static molecular magnetic observables.)

Numerical Renormalization Group calculations

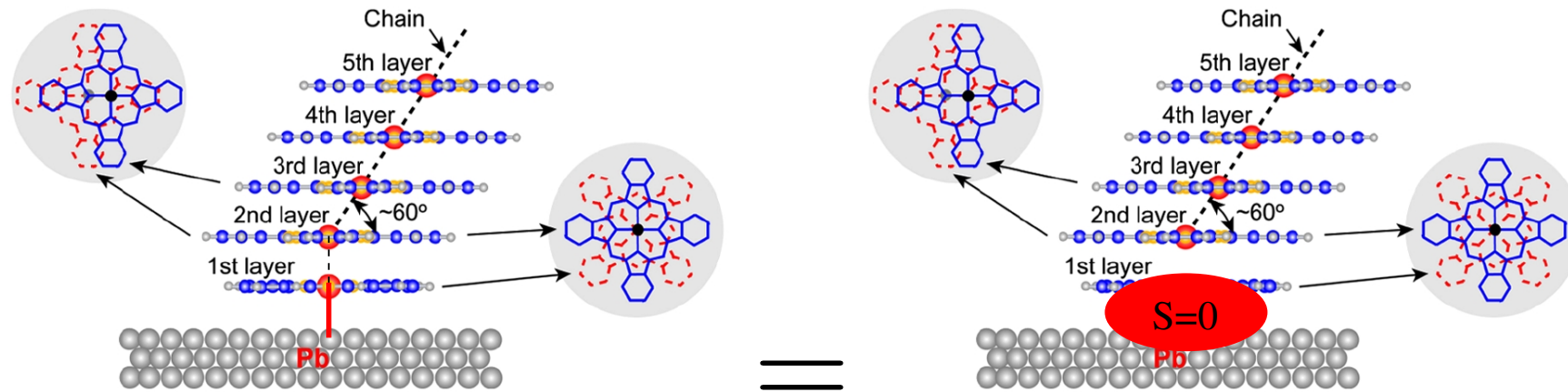
(Good for deposited molecules.)

You want to deposit a molecule



M. Bernien *et al.*, Phys. Rev. Lett. **102**, 047202 (2009); A. Ghirriet *et al.*, ACS Nano, **5**, 7090-7099 (2011); X. Chen *et al.*, Phys. Rev. Lett. **101**, 197208 (2008); M. Mannini *et al.*, Nature Materials **8**, 194 - 197 (2009).

Physical example (ICMM 2010)

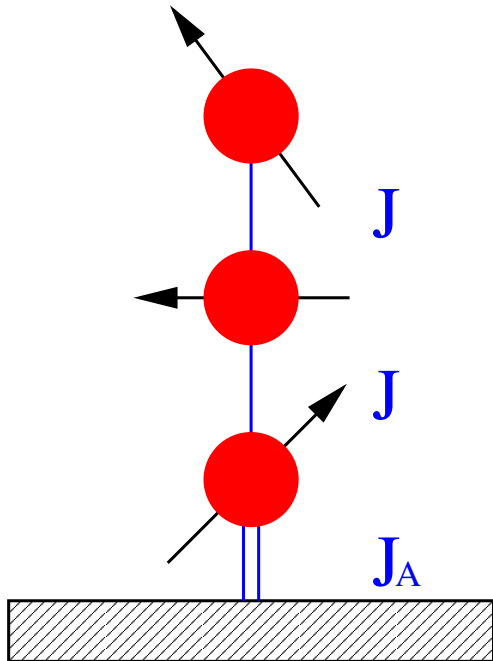


Stack of deposited Cobalt phthalocyanine (CoPc) molecules;
 Co^{2+} with spin $s = 1/2$.

Under which circumstances is the picture of total screening correct?

X. Chen *et al.*, Phys. Rev. Lett. **101**, 197208 (2008).

NRG – minimal model (**already an approximation!**)



- $\tilde{H} = \tilde{H}_{\text{electrons}} + \tilde{H}_{\text{coupling}} + \tilde{H}_{\text{impurity}}$

$$\tilde{H}_{\text{electrons}} = \sum_{i \neq j, \sigma} t_{ij} d_{i\sigma}^\dagger d_{j\sigma} + g_e \mu_B B \tilde{S}^z$$

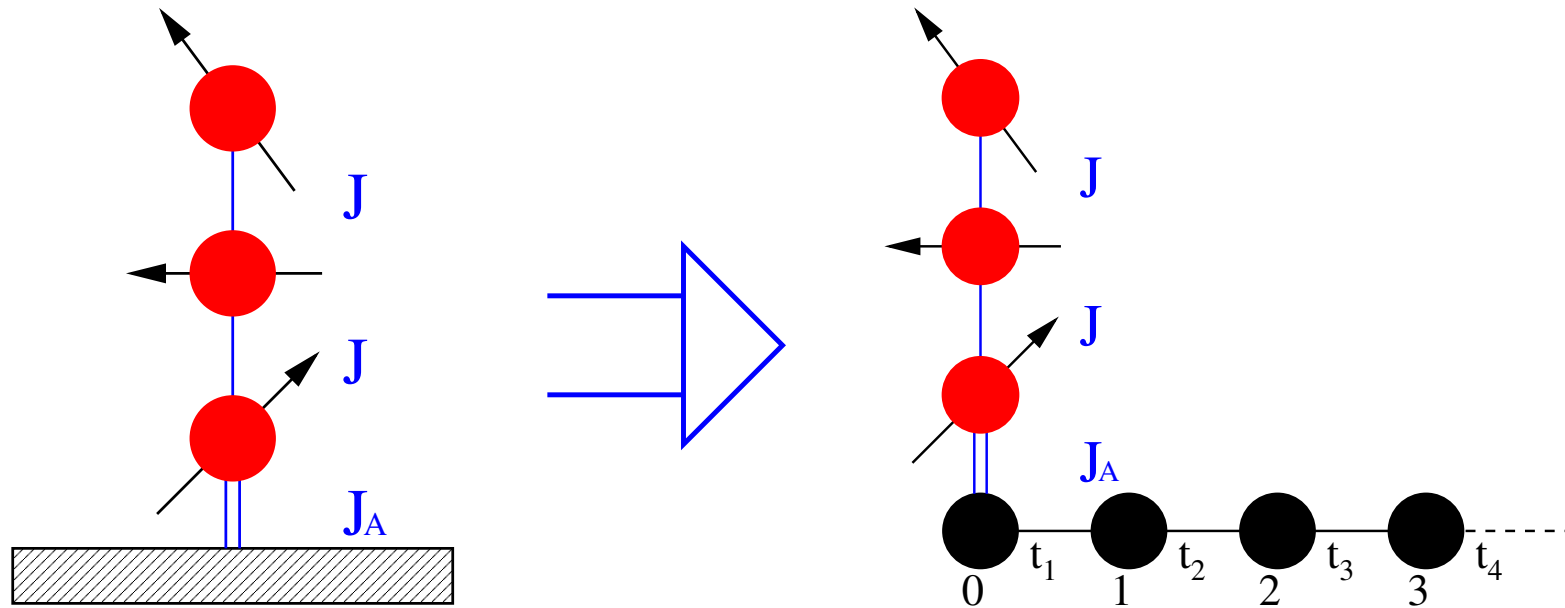
$$\tilde{H}_{\text{coupling}} = -2J_A \tilde{\mathbf{S}} \cdot \tilde{\mathbf{s}}_0 \quad , \quad \tilde{\mathbf{s}}_0 - \text{spin density at contact}$$

- $\tilde{H}_{\text{impurity}} = \text{Hamiltonian of your molecule!}$

- **NRG** \equiv construction of a small (!) effective model in order to evaluate properties of the deposited cluster, the impurity (3).

(1) K. G. Wilson, Rev. Mod. Phys. **47**, 773 (1975)
 (2) M. Höck, J. Schnack, Phys. Rev. B **87**, 184408 (2013)
 (3) *Impurity* is a technical term in this context and not an insult to chemists.

NRG in a cartoon



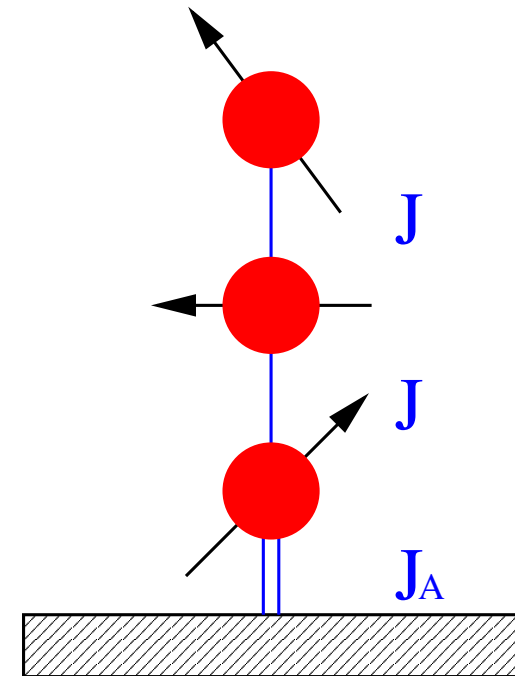
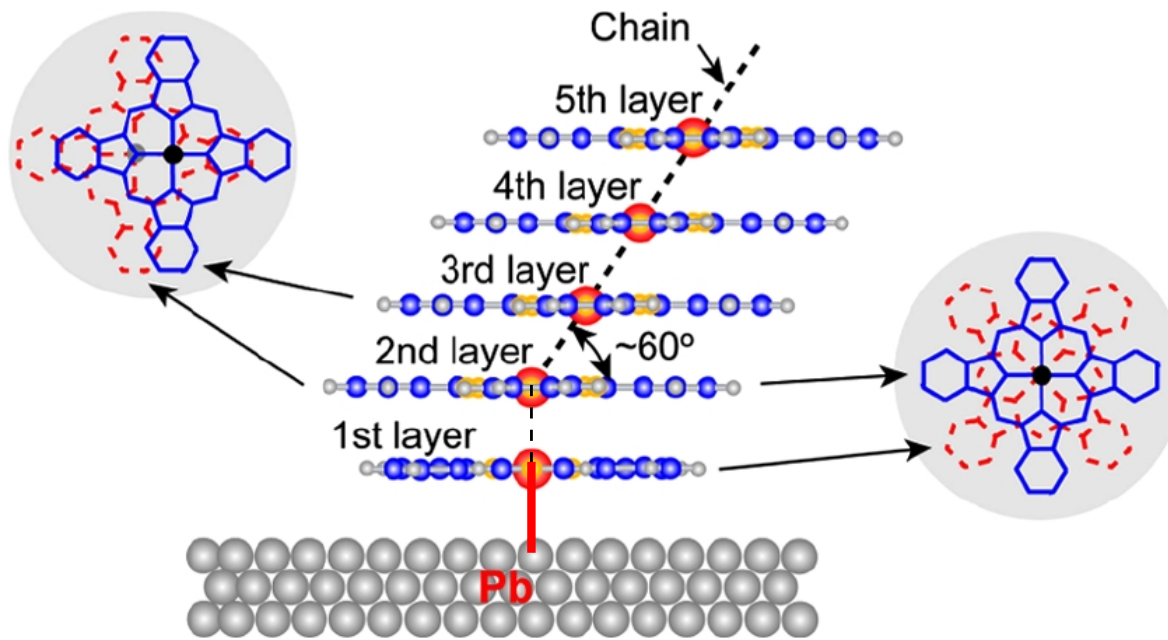
Metallic surface is replaced by semi-infinite Hubbard chain;

Parameters of the chain: hopping matrix elements and on-site energies;

Stepwise enlargement of the chain ($t_1 > t_2 > t_3 \dots$);

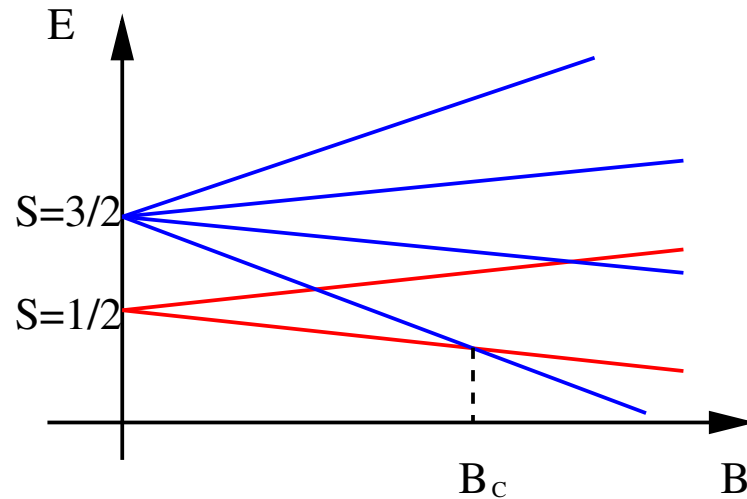
Truncation of basis set when matrices grow too big.

Once more: deposited chain

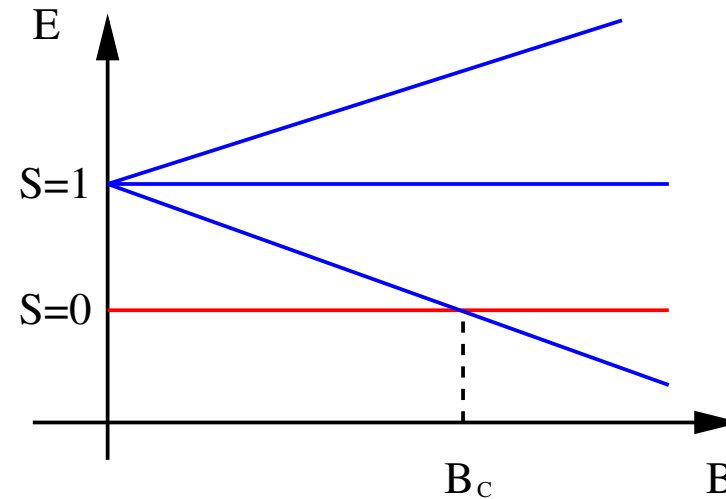


X. Chen *et al.*, Phys. Rev. Lett. **101**, 197208 (2008).

Energy levels of limiting cases for deposited trimer



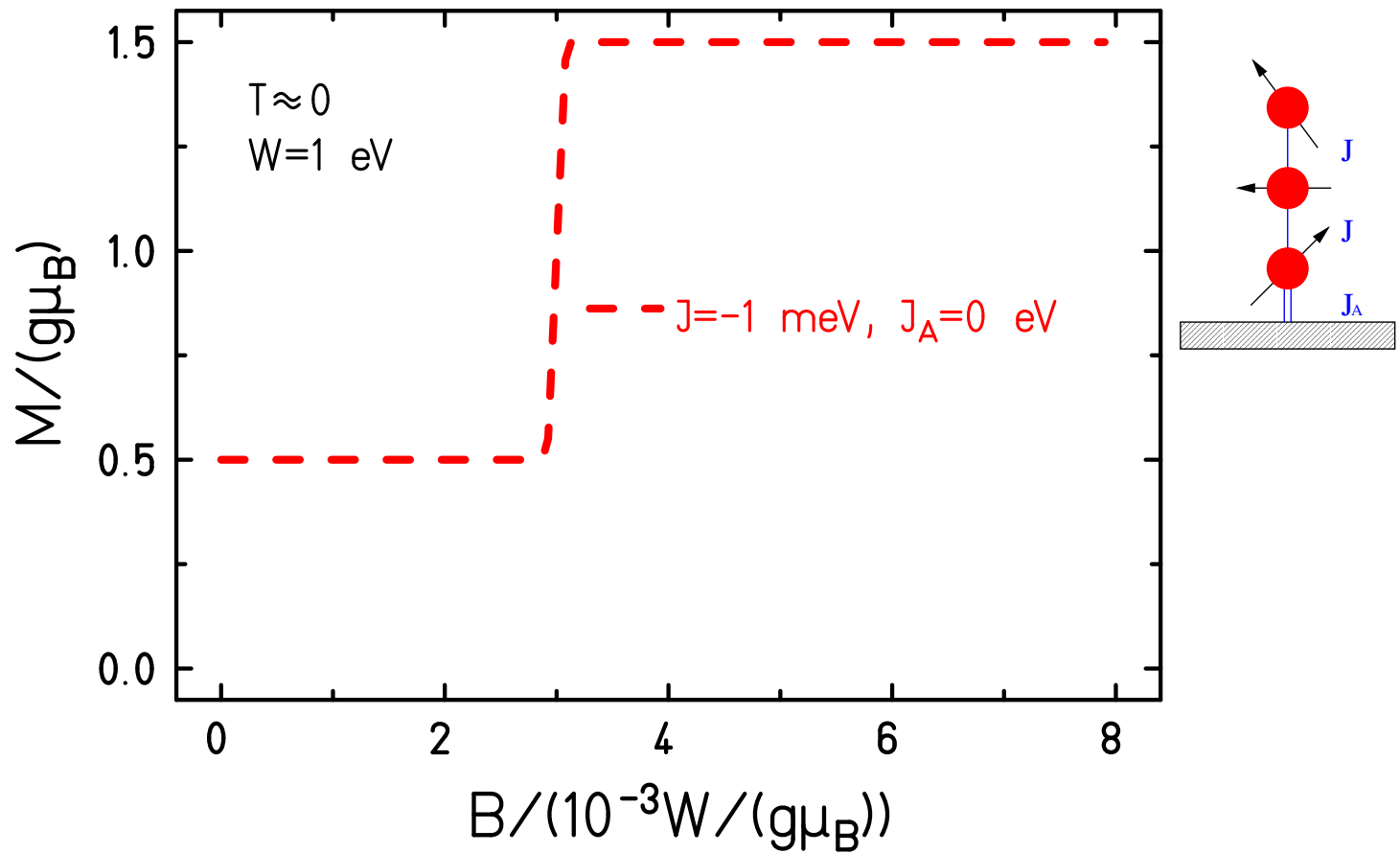
• energy levels of a trimer



• energy levels of a dimer

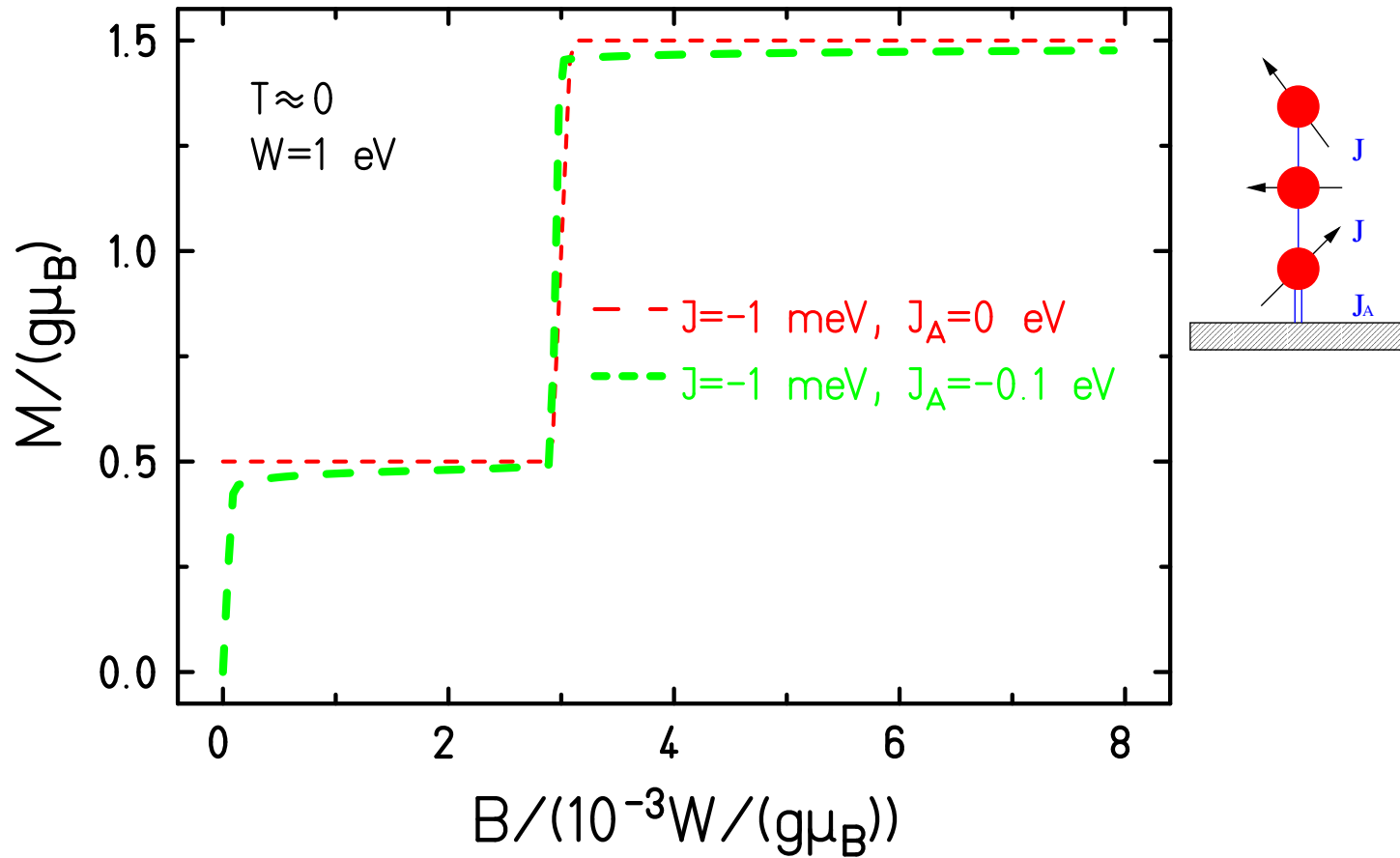
Magnetization curves different; could be seen in XMCD.
 NRG calculates observables also between limiting cases
 and can thus tell under which circumstances a limiting case applies.

Increasing coupling to the substrate



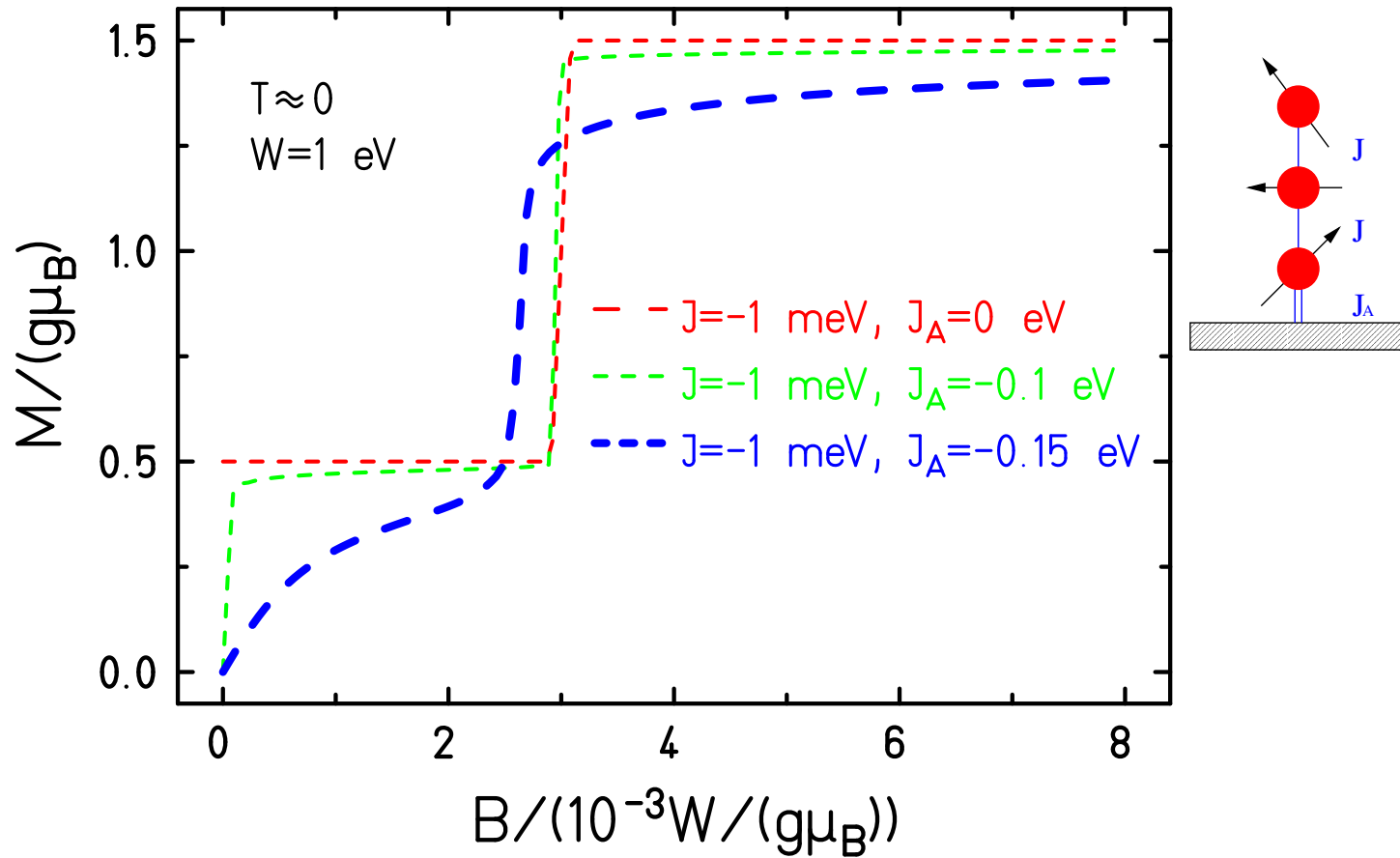
H.-T. Langwald and J. Schnack, submitted; arXiv:1312.0864.

Increasing coupling to the substrate



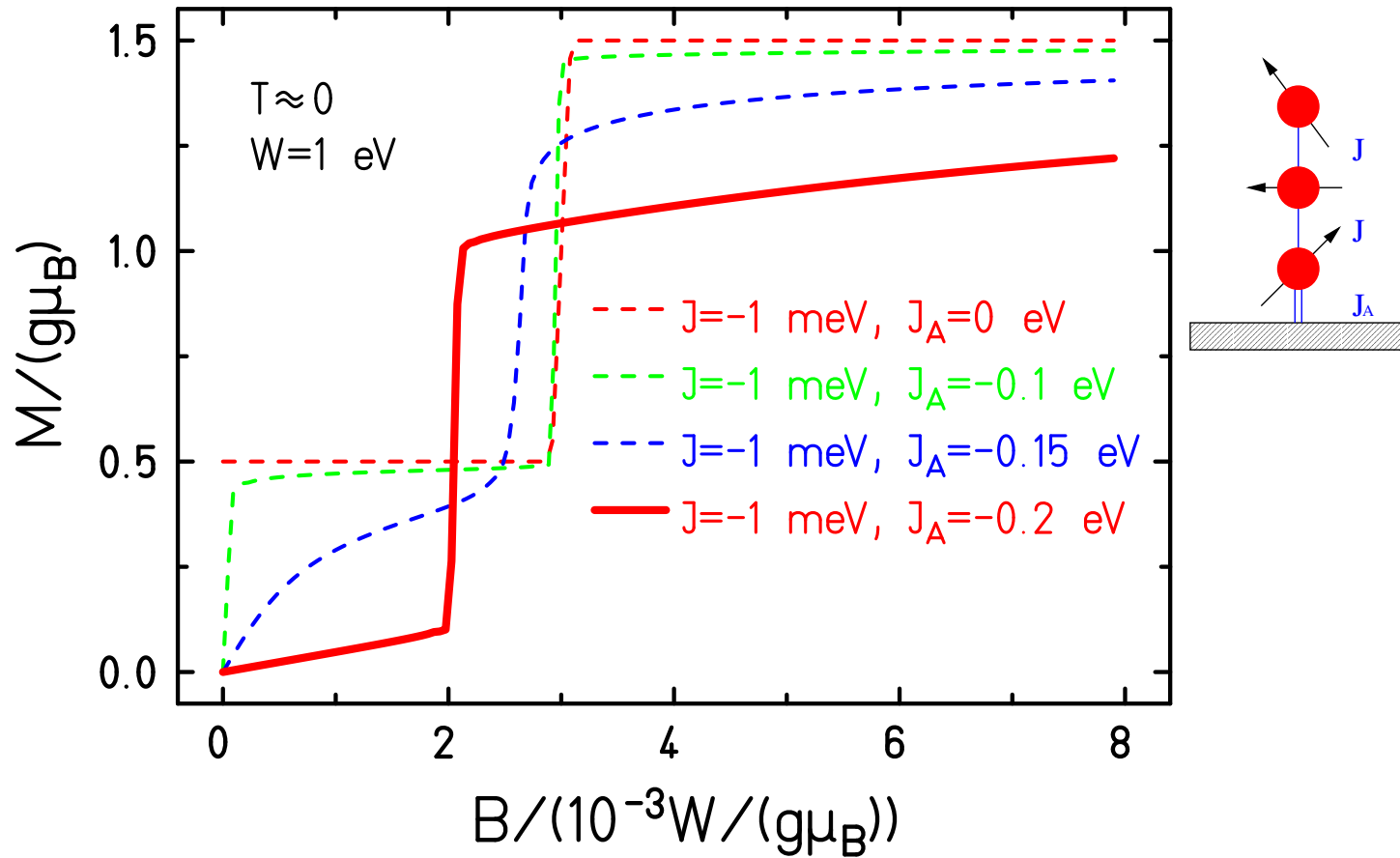
H.-T. Langwald and J. Schnack, submitted; arXiv:1312.0864.

Increasing coupling to the substrate



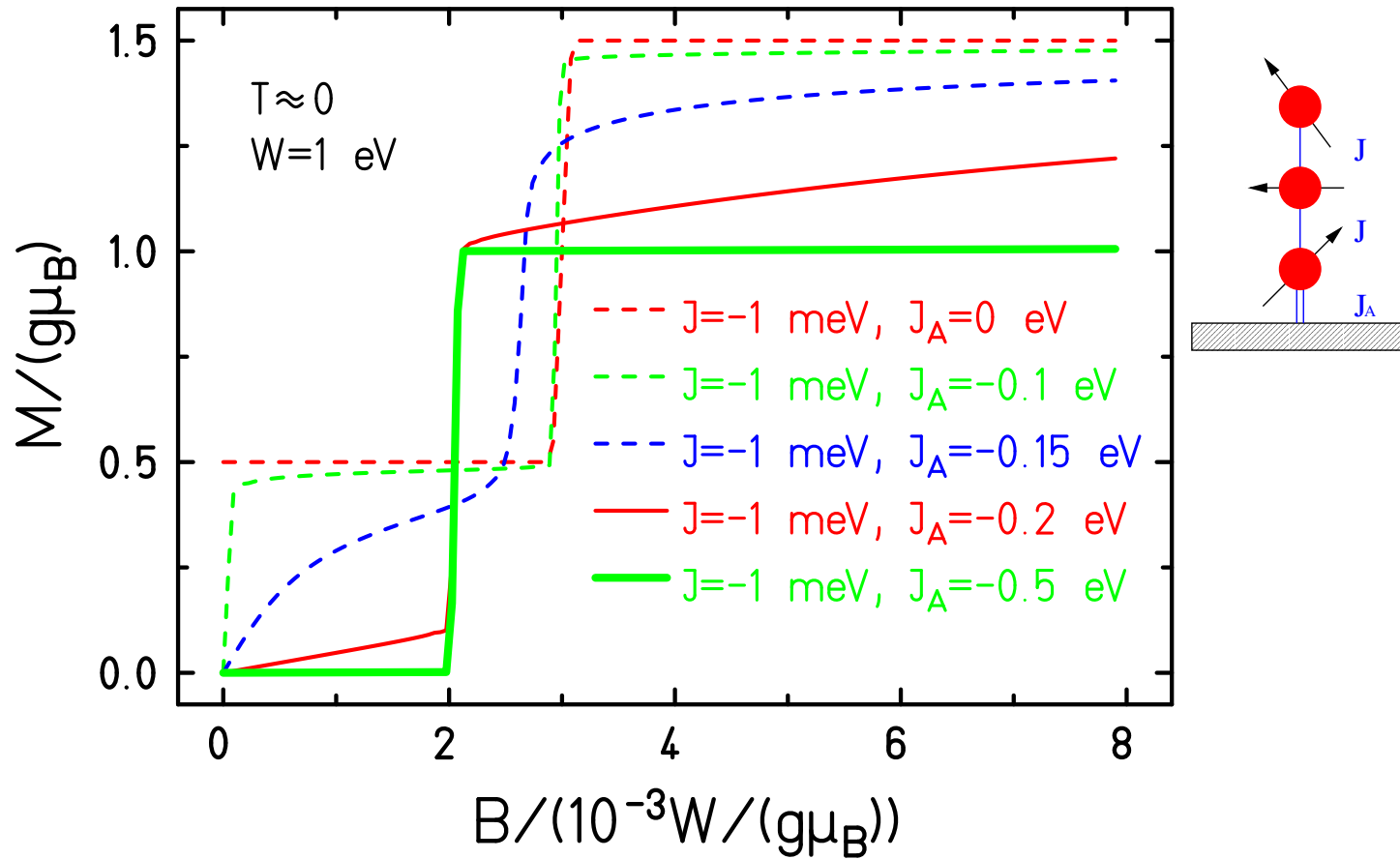
H.-T. Langwald and J. Schnack, submitted; arXiv:1312.0864.

Increasing coupling to the substrate



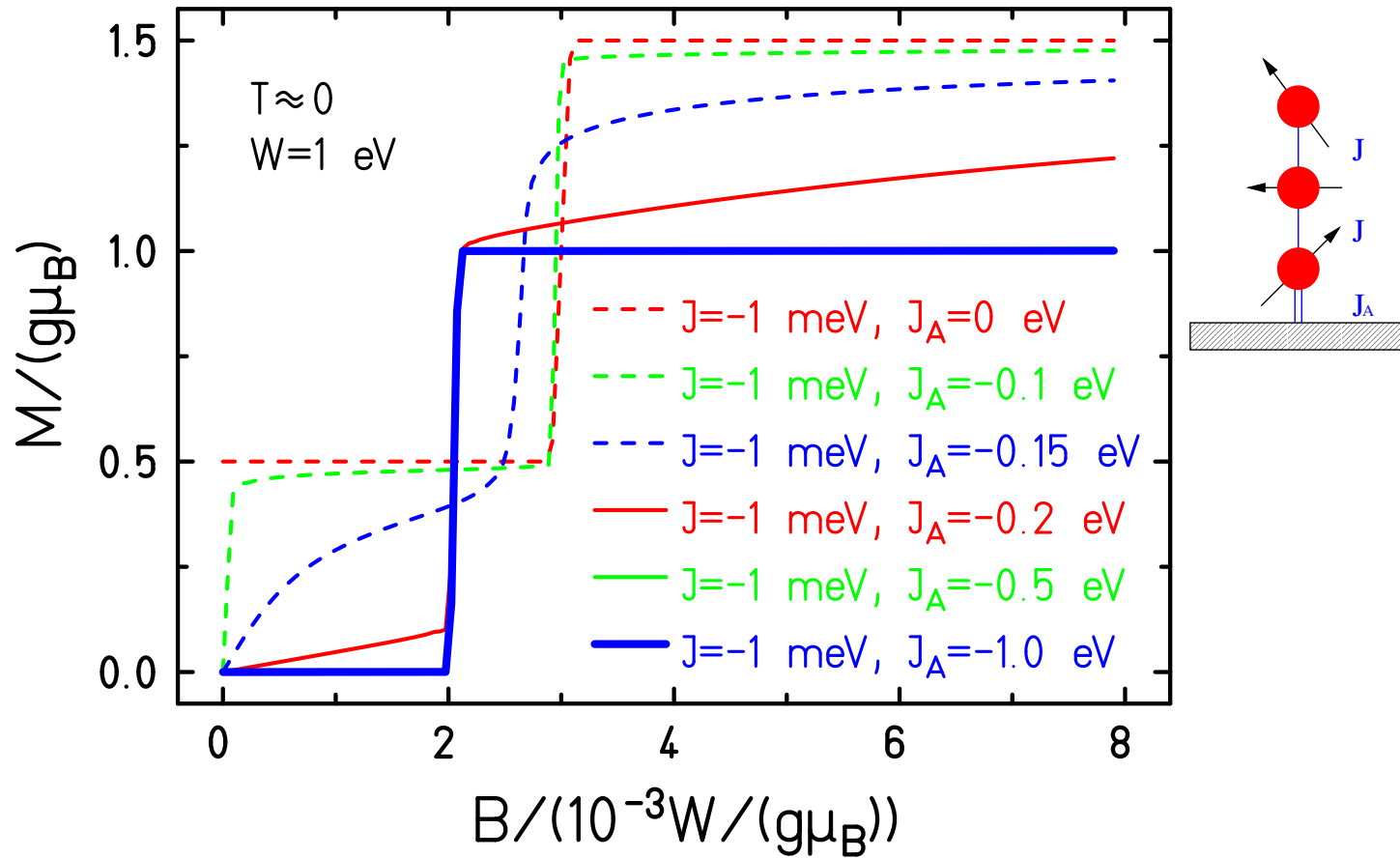
H.-T. Langwald and J. Schnack, submitted; arXiv:1312.0864.

Increasing coupling to the substrate



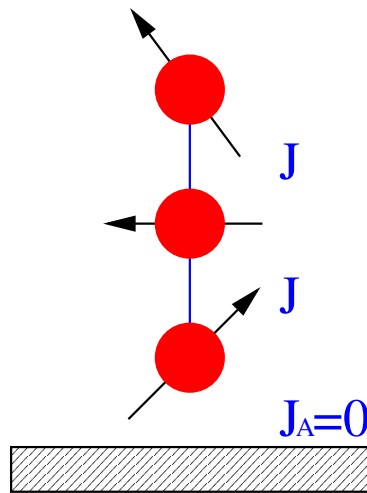
H.-T. Langwald and J. Schnack, submitted; arXiv:1312.0864.

Increasing coupling to the substrate

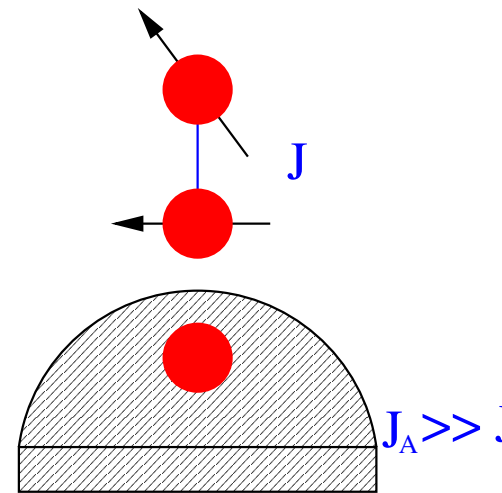


H.-T. Langwald and J. Schnack, submitted; arXiv:1312.0864.

Weak vs. strong coupling



- weak coupling limit:
unperturbed molecule (trimer)
- $|J_A| \lesssim 0.1W$



- strong coupling limit:
effective remainder (dimer)
- $|J_A| \gtrsim 0.5W$

Inbetween: no simple characterization + further sequential screening possible

Summary: theory methods

- **Complete diagonalization:** exact; spectra, transitions, observables, time-evolution; Dimension of largest Hilbert space $< 10^5$.
- **Finite Temperature Lanczos Method (FTLM):** pseudo-spectrum, low-lying levels good, transitions, observables, time-evolution; DoH $< 10^{10}$.
- **Quantum Monte Carlo (QMC):** observables; bad/no convergence for competing interactions (frustration) due to negative sign problem; otherwise HUGE systems possible.
- **Density Matrix Renormalization Group (DMRG):** low-lying target states, correlation functions, short time evolution, maybe thermodynamics; best for 1-d; HUGE systems possible.
- **Numerical Renormalization Group (NRG):** Kondo or Anderson impurity problems; logarithmic discretization of density of states of conduction electrons; observables as function of T and B .

Many thanks to my collaborators worldwide

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Thank you very much for your
attention.

The end.

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