Dynamics of Small Magnetic Systems

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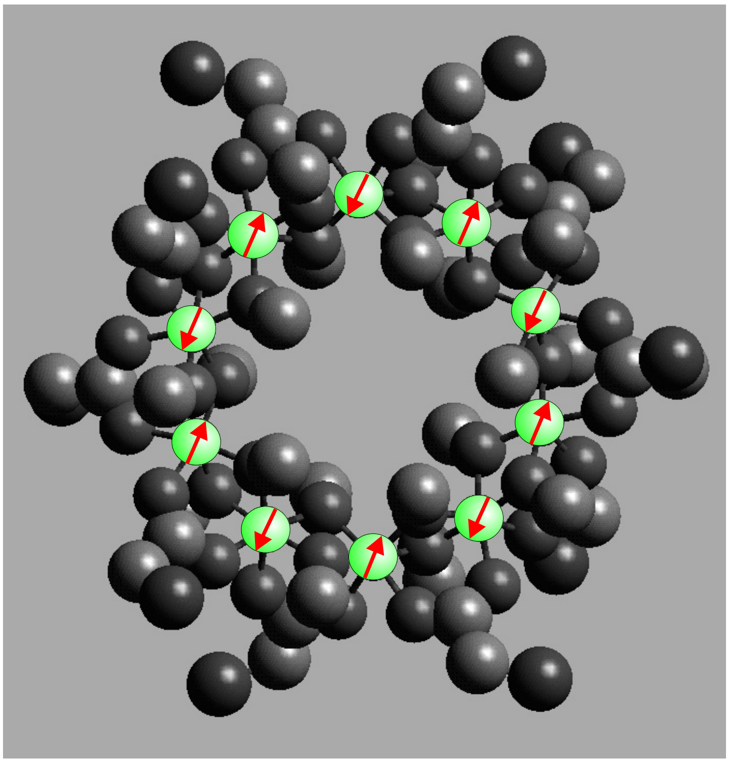

http://obelix.physik.uni-osnabrueck.de/∼schnack/

I am a bloody beginner in spin dynamics

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Origin and other interests

- nuclear physics (Gesellschaft für Schwerionenforschung, Darmstadt)
- molecular dynamics for fermions (FMD), dynamics at intermediate energies - fragmentation reactions
- thermostated molecular dynamics (Nosé-Hoover)
- ideal Fermi and Bose gases

Motivated for spins by Chr. Schröder[a]

- thermostated classical spin dynamics (Nosé-Hoover thermostat modified by Bulgac and Kusnezov)
- classical equations of motion

$$\frac{d}{dt}\vec{S}^\alpha = \frac{\partial H}{\partial \vec{S}^\alpha} \times \vec{S}^\alpha \; , \; H = -J_c \sum_{\alpha=1}^{N} \vec{S}^\alpha \cdot \vec{S}^{\alpha+1} - \mu_c \vec{B} \cdot \sum_{\alpha=1}^{N} \vec{S}^\alpha$$

- coupling of a deterministic or a stochastic thermostat
- time averaging to determine thermodynamic properties like susceptibility, spin-spin-correlation functions etc.
- classical treatment works astonishingly well
- WHY? WHAT ABOUT QUANTUM MECHANICS?

[a] university of Osnabrück, now Philips research

Small Magnetic Molecules

Properties

- new class of nanometer-size magnetic materials;
- molecules host from two up to thirty interacting paramagnetic ions;
- large number of nonmagnetic organic ligands;
- week intermolecular interactions;
- measurements on a bulk sample reflect intramolecular interactions only;
- well described in the Heisenberg model, additional terms like single side anisotropy rather small;
- speculations about applications range from mesoscopic magnets in biological systems, computer displays, photonic switches to catalysts

Structure

- simple clusters like a dimer (Fe_2) or a tetrahedron (Cr_4)
- magnetic rings, especially iron rings (Fe_6, Fe_8, Fe_{10}, ...) and others (Cr_8, Cu_6, Cu_8)
- complex clusters (Mn_{12})
- spin quantum number: s(Cu)=1/2, s(Cr)=3/2, s(Fe)=5/2

Research

Interesting obeservables

- molecular structure
- appropriate Hamilton operator and its parameters
- magnetisation, susceptibility, specific heat
- NMR, neutron scattering $\Leftrightarrow$ spin-spin-correlation function

Some groups in the field

- Prof. Dr. Dante Gatteschi, university of Florence
- Dr. Bernd Pilawa, Universität Karlsruhe
- Dr. Oliver Waldmann, Universität Erlangen-Nürnberg
- Prof. Dr. Achim Müller, Universität Bielefeld
- Ames Lab, Iowa: Prof. Dr. Marshall Luban, ...
- Gruppe Makroskopische Systeme und Quantentheorie, Universität Osnabrück

Contents

Heisenberg Model

Hamilton operator

$$\underset{\sim}{H} = \underset{\sim}{H}_0 + \underset{\sim}{H}_F = - \sum_{x,y}^{N} J(x,y)\ \vec{\underset{\sim}{s}}(x) \cdot \vec{\underset{\sim}{s}}(y) - \sum_{x}^{N} \mu\, B\, \underset{\sim}{s}^3(x)$$

$$\underset{\sim}{H}_0 = - \sum_{x,y} J(x,y) \left\{ \underset{\sim}{s}^3(x) \underset{\sim}{s}^3(y) + \frac{1}{2} \left[\underset{\sim}{s}^+(x) \underset{\sim}{s}^-(y) + \underset{\sim}{s}^-(x) \underset{\sim}{s}^+(y) \right] \right\}$$

spin operators

$$\left[\underset{\sim}{s}^a(x), \underset{\sim}{s}^b(y) \right] = i\, \epsilon_{abc}\, \underset{\sim}{s}^c(x)\, \delta_{xy} \quad , \quad \underset{\sim}{s}^\pm(x) = \underset{\sim}{s}^1(x) \pm i\, \underset{\sim}{s}^2(x)$$

product basis

$$\underset{\sim}{s}^3(x)\ |\, m_1, \dots, m_x, \dots, m_N\, \rangle = m_x\ |\, m_1, \dots, m_x, \dots, m_N\, \rangle$$

Method: decompose the Hilbert space into mutually orthogonal subspaces invariant w.r.t. $\underset{\sim}{H}$!

Symmetry about the 3-axis

$$\left[\underset{\sim}{H}, \underset{\sim}{S}^3\right] = 0 \quad , \quad \underset{\sim}{S}^3 = \sum_x \underset{\sim}{s}^3(x)$$

Eigenvalues M

$$M = -S_{\text{max}}, -S_{\text{max}} + 1, \ldots, S_{\text{max}} \quad \text{with} \quad S_{\text{max}} = \sum_{x=1}^{N} s(x)$$

Subspaces $\mathcal{H}(M)$

$$\dim\left(\mathcal{H}(M)\right) = \frac{1}{(S_{\text{max}} - M)!} \left[\left(\frac{d}{dz}\right)^{S_{\text{max}} - M} \prod_{x=1}^{N} \frac{1 - z^{2s(x)+1}}{1 - z}\right]_{z=0}$$

Subspaces $\mathcal{H}(M)$ for equal $s(x) = s$

$$\begin{aligned} \dim\left(\mathcal{H}(M)\right) \quad &= \sum_{n=0}^{[(S_{\text{max}} - M)/(2s+1)]} (-1)^n \binom{N}{n} \\ &\times \binom{N - 1 + (S_{\text{max}} - M) - n(2s + 1)}{N - 1} \end{aligned}$$

Problem equivalent to that of scoring sum M in a throw with N dice of $(2s + 1)$ faces.

Relevant Dimension II

Rotational symmetry

either all $s(x) = s$ or $B = 0$

$$\left[\underset{\sim}{H}, \vec{\underset{\sim}{S}}\right] = 0 \quad \& \quad \left[\vec{\underset{\sim}{S}}, \underset{\sim}{S}^3\right] = 0$$

Construction of $\mathcal{H}(S, M)$

magnon vacuum state spans $\mathcal{H}(S = S_{\text{max}}, M = S_{\text{max}})$

$$|\,\Omega\,\rangle = |\,m_1 = s(1), m_2 = s(2), \dots, m_N = s(N)\,\rangle$$

consider decrement in M

$$\underset{\sim}{S}^{-}\,|\,\Omega\,\rangle \in \mathcal{H}(M = S_{\text{max}} - 1) \text{ with } S = S_{\text{max}}$$

The orthogonal subspace belongs to $S = S_{\text{max}} - 1$. Proceeding one finds that each $\mathcal{H}(M)$ can be decomposed into orthogonal subspaces

$$\mathcal{H}(M) = \mathcal{H}(M, M) \oplus \underset{\sim}{S}^{-}\mathcal{H}(M + 1)$$

diagonalization necessary only in the subspaces $\mathcal{H}(S, S)$

$$\dim\left(\mathcal{H}(S, S)\right) = \dim\left(\mathcal{H}(M = S)\right) - \dim\left(\mathcal{H}(M = S + 1)\right)$$

Relevant Dimension III

Cyclic shift symmetry

all $s(x) = s$ and $J(x,y) = J(|x-y|)$; cyclic shift operator

$$\underset{\sim}{T} \mid m_1, \dots, m_{N-1}, m_N \rangle \quad = \quad \mid m_N, m_1, \dots, m_{N-1} \rangle$$

$$\left[\underset{\sim}{H}, \underset{\sim}{T}\right] = 0 \quad \& \quad \left[\underset{\sim}{T}, \underset{\sim}{\vec{S}}\right] = 0$$

Eigenvalues of $\underset{\sim}{T}$

$$z = \exp\left\{-i\frac{2\pi k}{N}\right\} \quad , \quad k = 0, 1, \dots, N-1$$

Construction of $\mathcal{H}(S, M, k)$

The subspaces $\mathcal{H}(S, M, k)$ are constructed using cycles and keeping track of proper cycles as well as epicycles.[a]

[a]K. Bärwinkel, H.J. Schmidt, J. Schnack, *Structure and relevant dimension of the Heisenberg model and applications to spin rings* J. Magn. Magn. Mater. (to be published 1999)

Relevant dimensions

		N							
		2	3	4	5	6	7	8	9
s	$\frac{1}{2}$	1	2	3	5	9	14	28	48
	$\frac{1}{2}$	**1**	**1**	**1**	**1**	**2**	**2**	**4**	6
	1	1	3	6	15	40	105	280	750
	1	**1**	**1**	**2**	**3**	8	15	37	84
	$\frac{3}{2}$	1	4	11	36	120	426	1505	5300
	$\frac{3}{2}$	**1**	**2**	**4**	8	23	61	192	590
	2	1	5	17	70	295	1260	5620	25200
	2	**1**	**2**	5	14	53	180	712	2800
	$\frac{5}{2}$	1	6	24	120	609	3150	16576	88900
	$\frac{5}{2}$	**1**	**2**	7	24	105	450	2085	9884

Table 1: Relevant dimension assuming only invariance with respect to rotations (upper rows) and assuming also invariance with respect to cyclic shifts (lower rows). The highlighted cases can be solved analytically.

Example $N = 6$, $s = 1/2$

$M = 3$: maximum dimension 1

$$|\,\Omega\,\rangle = |+{+}{+}{+}{+}{+}\rangle$$

$M = 2$: maximum dimension 1

$|-{+}{+}{+}{+}{+}\rangle$ generates proper cycle of dimension 6

$M = 1$: maximum dimension 2

$|-{-}{+}{+}{+}{+}\rangle$ generates proper cycle of dimension 6

$|-{+}{-}{+}{+}{+}\rangle$ generates proper cycle of dimension 6

$|-{+}{+}{-}{+}{+}\rangle$ generates epicycle of dimension 3

$M = 0$: maximum dimension 2

$|-{-}{-}{+}{+}{+}\rangle$ generates proper cycle of dimension 6

$|-{-}{+}{-}{+}{+}\rangle$ generates proper cycle of dimension 6

$|-{+}{-}{-}{+}{+}\rangle$ generates proper cycle of dimension 6

$|-{+}{-}{+}{-}{+}\rangle$ generates epicycle of dimension 2

Classical limit

Aim

- understand limits and applicability of classical spin dynamics
- here: limit of high spin quantum number s

Idea

$$\underset{\sim}{H} = \frac{J}{\hbar^2}\, \underset{\sim}{\vec{s}}_1 \cdot \underset{\sim}{\vec{s}}_2 = \frac{J}{2\hbar^2} \left(\underset{\sim}{\vec{S}}^2 - \underset{\sim}{\vec{s}}_1^{\,2} - \underset{\sim}{\vec{s}}_2^{\,2} \right) \quad ; \quad \underset{\sim}{\vec{S}} = \underset{\sim}{\vec{s}}_1 + \underset{\sim}{\vec{s}}_2$$

$$\underset{\sim}{\vec{\epsilon}}_n = \frac{\underset{\sim}{\vec{s}}_n}{\sqrt{\hbar^2\, s(s+1)}} \quad , \quad \left[\underset{\sim}{\epsilon}_{n\,x}, \underset{\sim}{\epsilon}_{n\,y} \right] = \frac{i}{\sqrt{s(s+1)}}\, \underset{\sim}{\epsilon}_{n\,z} \overset{s\to\infty}{\longrightarrow} 0$$

$$H_c = J_c\, \vec{e}_1 \cdot \vec{e}_2 \quad , \quad J_c = J\, s(s+1)$$

Procedure

- in order to gain the high spin limit, spectra for different s have to be mapped onto the same energy interval;
- compared quantities: density of states, autocorrelation function

$$\begin{aligned} \langle\langle\, \underset{\sim}{\vec{s}}_1(t) \cdot \underset{\sim}{\vec{s}}_1(0)\, \rangle\rangle &= \frac{1}{Z} \mathrm{tr} \left\{ \underset{\sim}{\vec{s}}_1(t) \cdot \underset{\sim}{\vec{s}}_1(0)\, e^{-\beta \underset{\sim}{H}} \right\} \\ Z &= \mathrm{tr} \left\{ e^{-\beta \underset{\sim}{H}} \right\} \end{aligned}$$

Spin Dimer I

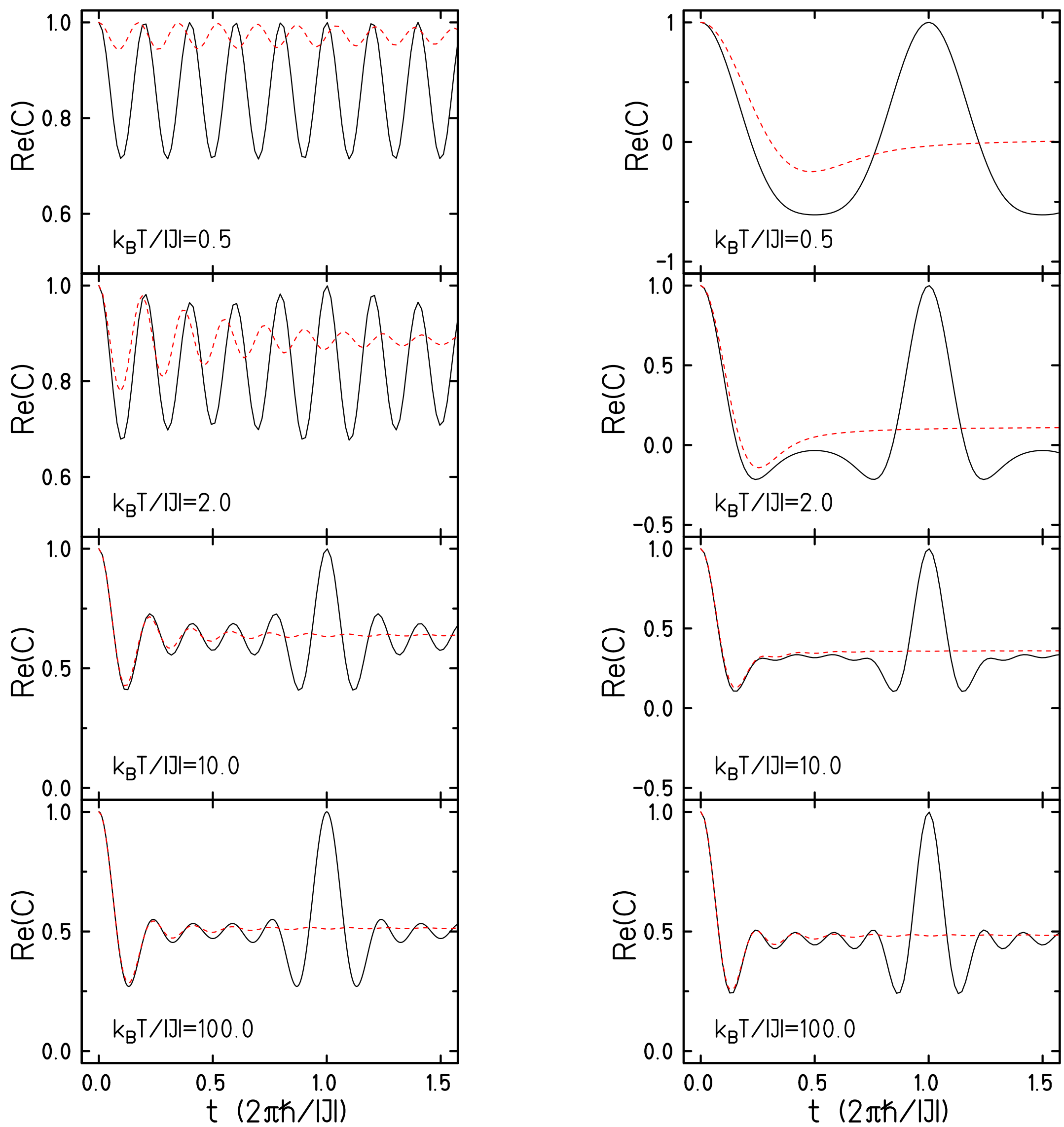

Figure 1: Normalized autocorrelation function $\mathrm{Re}\,(C(t))$ for a spin-$\frac{5}{2}$-dimer for four different temperatures (solid lines). The left panels display our results for the ferromagnetic dimer, the right panels the antiferromagnetic case. The dashed lines show the classical result.

Spin Dimer II

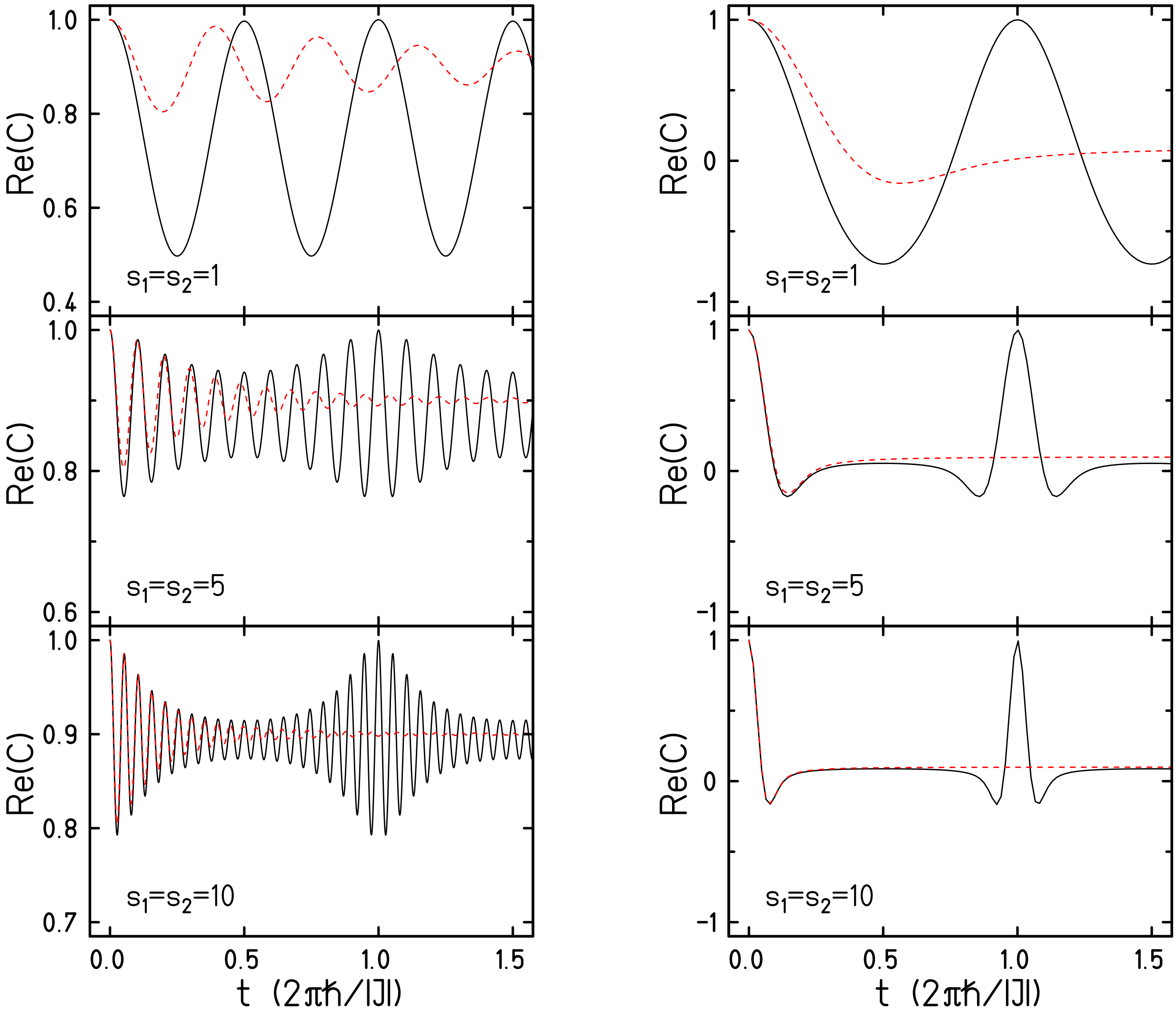

Figure 2: Normalized autocorrelation function for three different spins at the temperature $k_B\,T/(J\,s\,(s+1)) = 0.2$. The left panels displays the ferromagnetic dimer, the right panels the antiferromagnetic one. The solid lines show the quantum result, the dashed lines the classical.

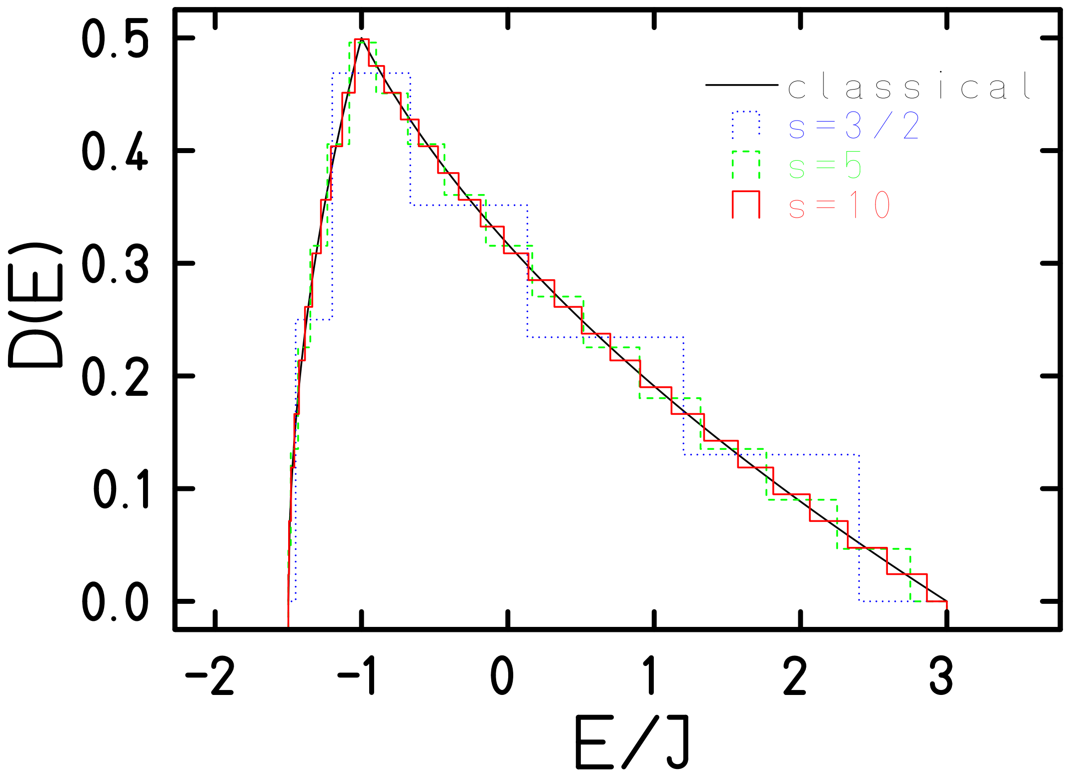

Figure 3: Normalized density of states for quantum Heisenberg trimers (dashed) and the classical counterpart (solid).

Classical density of states

$$D_c(E) = \begin{cases} \frac{1}{2J_c}\sqrt{\frac{2E}{J_c}+3} & : \quad -\frac{3}{2}\,J_c \le E \le -J_c \\ \frac{1}{4J_c}(3-\sqrt{\frac{2E}{J_c}+3}) & : \quad -J_c < E \le 3J_c \\ 0 & : \quad \text{else} \end{cases}$$

AF Heisenberg Rings I

Hamilton operator

$$\underset{\sim}{H} = -2\,J \sum_{x=1}^{N} \underset{\sim}{\vec{s}}(x) \cdot \underset{\sim}{\vec{s}}(x+1) \ , \quad \forall x : s(x) = s \ , \quad J < 0$$

Symmetries

- rotational symmetry: $\underset{\sim}{\vec{S}}$, $\underset{\sim}{S}^3$, quantum numbers S and M;
- cyclic shift symmetry: $\underset{\sim}{T}$, quantum number k;
- spin flip symmetry: $\underset{\sim}{C}$ changes all m_x into $-m_x$, quantum number π;

Ground state properties

1. ground state belongs to subspace $\mathcal{H}(S)$ with the smallest possible total spin quantum number S;
2. if Ns integer, then the ground state is non-degenerate,
3. if Ns half integer, then the ground state is fourfold degenerate,
4. if s integer or Ns even, then the translational shift quantum number is $k = 0$;
5. if s is half integer and Ns odd, then $k = N/2$;
6. if Ns is half integer, then $k = \lfloor (N+1)/4 \rfloor$ and $k = N - \lfloor (N+1)/4 \rfloor$; $\lfloor \cdot \rfloor$ greatest integer less or equal;

s		$N=2$	3	4	5	6	7	8	9	10	
	$\frac{1}{2}$	1.5	0.5	1	0.747	0.934	0.816	0.913	0.844	0.903	$E/(NJ)$
		1	4	1	4	1	4	1	4	1	deg
		1	1, 2	0	1, 4	3	2, 5	0	2, 7	5	k
		−		+		−		+		−	π
	1	4	2	3	2.612	2.872	2.735	2.834	2.773	2.819	$E/(NJ)$
		1	1	1	1	1	1	1	1	1	deg
		0	0	0	0	0	0	0	0	0	k
		+	−	+	−	+	−	+			π
	$\frac{3}{2}$	7.5	3.5	6	4.973	5.798	5.338	5.732	5.477		$E/(NJ)$
		1	4	1	4	1	4	1	4		deg
		1	1, 2	0	1, 4	3	2, 5	0	2, 7		k
		−		+		−					π
	2	12	6	10	8.456	9.722	9.045	9.630			$E/(NJ)$
		1	1	1	1	1	1	1			deg
		0	0	0	0	0	0	0			k
		+	+	+	+	+					π
	$\frac{5}{2}$	17.5	8.5	15	12.434	14.645	13.451				$E/(NJ)$
		1	4	1	4	1	4				deg
		1	1, 2	0	1,4	3	2, 5				k
		−		+							π

Summary

1. Relevant dimensions in the Heisenberg model[a]
2. Classical limit for spin dimers and trimers[b]
3. General properties of the antiferromagnetic ground state of small Heisenberg rings[c]

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[a] K. Bärwinkel, H.-J. Schmidt, J. Schnack, *Structure and relevant dimension of the Heisenberg model and applications to spin rings*, J. Magn. Magn. Mater. (to be published 1999)

[b] D. Mentrup, J. Schnack, M. Luban, *Spin dynamics of quantum and classical Heisenberg dimers*, Physica A **272** (1999) 153

[c] K. Bärwinkel, H.-J. Schmidt, J. Schnack, *Ground state properties of antiferromagnetic Heisenberg spin rings*, to be submitted to Phys. Rev. B