UNIVERSITÄT OSNABRÜCK

Selected small quantum systems: Theoretical techniques, fundamental properties, and thermodynamics

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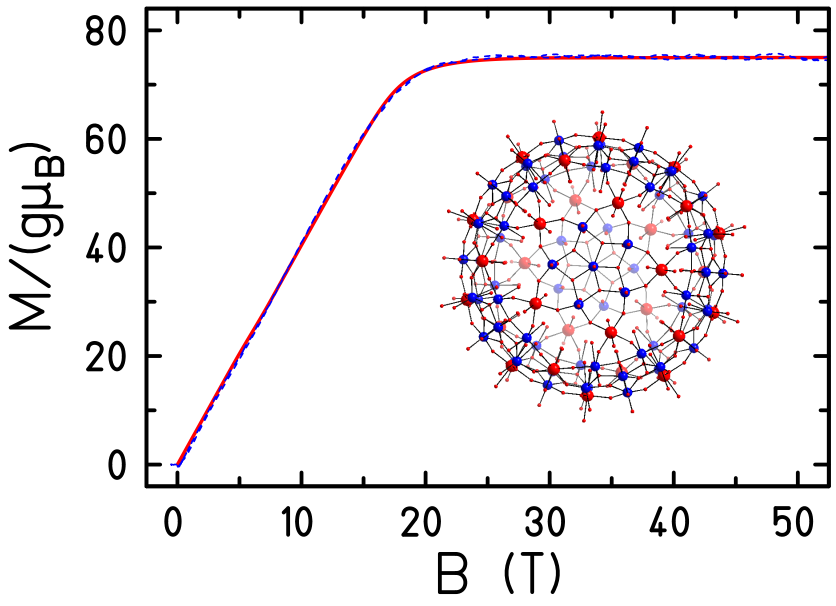

The picture on the title page displays magnetisation curves of the polyoxometalate $\{Mo_{72}Fe_{30}\}$ [MSS$^+$99]. The dashed blue curve depicts the experimental result and the solid red curve the theoretical result calculated with the help of an approximate Hamiltonian [LSM00].
The inset shows the structure of $\{Mo_{72}Fe_{30}\}$ which was synthesized in the group of Prof. Dr. A. Müller at the university of Bielefeld. Small red balls depict oxygen, big red balls depict iron and blue balls molybdenum. The structure is displayed viewing along a fivefold symmetry axis. The inset figure was produced by Paul Kögerler (Bielefeld).

Contents

1. Introduction

This collection of articles summarizes the author's work done after 1996. It is submitted to the Fachbereich Physik at the University of Osnabrück for the purpose of the author's Habilitation.

The first chapters, which precede the collection of articles, provide a short introduction of the fields the articles belong to. They also inform how the articles of the author contribute to research efforts of the specific field.

Three main fields are covered in this collection. All of them deal with small quantum systems: with characteristics of their ground states and low-lying excitations, and with their thermal properties. Due to experimental progress both in synthesis as well as in characterization small and mesoscopic systems attract more and more attention. A common characteristics of these systems is their finite particle number, not one or two and not infinity, which leads to deviations from the behaviour expected in the thermodynamics limit and often calls for an extension of physical concepts, like for instance the concept of phase transitions.

In addition these systems show all degrees of correlations induced by interparticle interactions. Weakly interacting particles like the recently investigated atomic vapours contained in magnetic traps may be rather well described as ideal quantum gases using standard methods, whereas the interacting spins of a magnetic molecule or interacting fermions in nuclei form highly correlated systems. In the latter case the partition function cannot be evaluated easily or even at all. One method to determine thermodynamic properties nevertheless is the method of thermostated dynamics, which was developed within the last fifteen years. While the method is successfully applied to classical systems as for instance noble gas clusters or classical spin systems, its extension to quantum systems is a matter of current research.

The following introduction is organized as follows. The second chapter presents the latest results on properties of magnetic molecules, the third chapter introduces to the physics of ideal Fermi or Bose gases contained in magnetic traps. The fourth and last chapter summarizes the effort made to uncover thermodynamic properties of interacting Fermi systems.

Throughout the article all operators are underlined with a tilde, e.g. $\underset{\sim}{H}$, expectation values are denoted by brackets, e.g. $\langle \underset{\sim}{H} \rangle$ or calligraphic letters, e.g. $\mathcal{H}$ and thermal averages are symbolized by double brackets like in $\langle\langle\ \underset{\sim}{H}\ \rangle\rangle$. If not needed explicitly $\hbar$ and k_B are sometimes dropped.

References to articles submitted for the purpose of the author's Habilitation are given in bold face letters.

2. Small magnetic molecules

The synthesis of molecular magnets has undergone rapid progress in recent years [SGC93, GCPS94, Gat94, CDGM96, MPPG98]. Each of the identical molecular units can contain as few as two and up to several dozens of paramagnetic ions (“spins”). The largest paramagnetic molecule synthesized to date, the polyoxometalate $\{\mathrm{Mo}_{72}\mathrm{Fe}_{30}\}$ [MSS$^+$99] contains 30 iron ions of spin $s = 5/2$. Although these materials appear as macroscopic samples, i.e. crystals or powders, the intermolecular magnetic interactions are utterly negligible as compared to the intramolecular interactions. Therefore, measurements of their magnetic properties reflect mainly ensemble properties of single molecules.

The properties of magnetic molecules are investigated with a variety of well-known techniques like electron parametric resonance (EPR) [BG90], NMR spin-lattice relaxation rate measurements [Mor56, LGBC97b, LGBC97a], **[FLB$^+$00]**, neutron scattering [BL89], magnetisation measurements [TDP$^+$94], torque magnetometry [WSK$^+$99], and calorimetry [GNS$^+$98].

Their magnetic features promise a variety of applications in physics, magneto-chemistry, biology, biomedicine and material sciences [Gat94, CDGM96]. The most promising progress is being made in the field of spin crossover substances using effects like “Light Induced Excited Spin State Trapping (LIESST)” [GHS94].

It appears that in the majority of these molecules the localized single-particle magnetic moments couple antiferromagnetically and the spectrum is rather well described by the Heisenberg model with isotropic next neighbour interaction [BG90, DGP$^+$93, CCF$^+$95, PDK$^+$97, WSK$^+$99]. Thus, the interest in the Heisenberg model, which is known already for a long time, but used mostly for infinite systems like chains or lattices, was renewed by the successful synthesis of magnetic molecules. Studying such spin arrays focuses on qualitatively new physics caused by the finite size of the system.

The work presented in this chapter was mainly done with K. Bärwinkel, H.-J. Schmidt, D. Mentrup (Universität Osnabrück), and M. Luban (Ames Lab & Iowa State University), the respective publications **[MSL99, BSS00b, FLB$^+$00, MSSL00, BSS00a, Sch00, SL01]** are presented in section A on page 36.

2.1 Heisenberg model

The Hamilton operator for the isotropic Heisenberg model including the interaction with an external magnetic field (Zeeman term) reads

$$\underset{\sim}{H} \;=\; -2\,J \sum_{(u,v)} \underset{\sim}{\vec{s}}(u) \cdot \underset{\sim}{\vec{s}}(v) + g\mu_B B \underset{\sim}{S}_z \ , \tag{2.1}$$

where J is the exchange interaction with units of energy, and $J < 0$ corresponds to antiferromagnetic, $J > 0$ to ferromagnetic coupling. The vector operators $\underset{\sim}{\vec{s}}(u)$ are the single-particle spin operators. The sum in Eq. (2.1) runs over all distinct interacting pairs (u, v) of spins at positions u and v. Since the Hamilton operator commutes with $\underset{\sim}{\vec{S}}^2$ and $\underset{\sim}{S}_z$, total spin S and total magnetic quantum number M are good quantum numbers. For spin rings the Hamiltonian (2.1) is also invariant under cyclic shifts which leads to another good quantum number, the translational quantum number k.

These symmetries of the isotropic Heisenberg Hamilton operator allow to decompose the Hilbert space $\mathcal{H}$ into a set of mutually orthogonal subspaces $\mathcal{H}(S, M, k)$ **[BSS00b]** in which a complete diagonalization can be performed either analytically [Kou97, Kou98], **[BSS00b]** or numerically [BF64, BJ83, FLMU91, Man91, DGP$^+$93, GJL94, FLS97, Wal00].

The achieved insight could be used to compare the quantum Heisenberg model to its classical counterpart. The classical Heisenberg model [LLB98, Sch99b] turns out to provide accurate quantitative results for static properties, such as magnetic susceptibility, down to thermal energies of the order of the exchange coupling [LLB98, Sch99b, MLS$^+$]. However, considerable care is required comparing dynamical properties which express themselves in time-dependent spin-spin correlation functions. Here the finite, and possibly rather small, dimension of the Hilbert space restricts the quantum dynamics, for instance to be recurrent **[MSL99, MSSL00]**.

2.2 Exact properties of antiferromagnetically coupled spin rings

The successful description of magnetic molecules by means of the Heisenberg model resulted in new efforts to find exact properties for instance for low-lying states and thus joined long-lasting investigations performed for infinite chains and lattices, see e.g. [BLP92, BLLP94, RIVR95, SGJ96, NRS98, TR99, WKS$^+$00]. Already 40 years ago a special class of spin arrays – bipartite spin systems – was found to possess strict ground state properties which are characterized by the sign rule of Marshall and Peirls [Mar55] and by the theorem of Lieb, Schultz and Mattis [LSM61, LM62]. But since many magnetic molecules are non-bipartite spin systems, the formulation of similar properties is highly desirable.

Looking at the properties of spin rings with constant, isotropic nearest-neighbour interaction one realizes that both the ground states as well as the first excited states share systematic properties which so far are understood only for Heisenberg spin rings of an even number of spin sites, i.e. bipartite rings. Rings with an odd number of sites, which are non-bipartite and thus can be characterized as frustrated [RIVR95], show unexpected properties in degeneracy and translational quantum number **[BSS00a, Sch00]**.

Knowing systematic rules for quantum numbers also of non-bipartite systems would be very useful for a comparison of theoretical results with measurements. One could employ knowledge about quantum numbers of ground and first excited states in order to understand the thermal behaviour of quantities like the magnetic susceptibility. In addition these exact values may help to improve low temperature approximations. Usually the high temperature behaviour of observables is well known, e.g. from classical spin dynamics [LLB98], but at low temperature such approximations are poor. The knowledge of ground and first excited states could already be sufficient for a considerable improvement.

Some of our results for ground state properties **[BSS00a]** are summarized in table 2.1. Without exception we find:

1. The ground state belongs to the subspace $\mathcal{H}(S)$ with the smallest possible total spin quantum number S; this is either $S = 0$ for $N \cdot s$ integer, then the total magnetic quantum number M is also zero, or $S = 1/2$ for $N \cdot s$ half integer, then $M = \pm 1/2$.
2. The restricted ground state within a subspace of constant total magnetic quantum number M belongs to $\mathcal{H}(S)$ with S attaining its smallest value $S = |M|$.
3. If $N \cdot s$ is integer, then the ground state is non-degenerate.
4. If $N \cdot s$ is half integer, then the ground state is fourfold degenerate.
5. If s is integer or $N \cdot s$ even, then the shift quantum number is $k = 0$.
6. If s is half integer and $N \cdot s$ odd, then the shift quantum number turns out to be $k = N/2$.
7. If $N \cdot s$ is half integer, then $k = \lfloor (N+1)/4 \rfloor$ and $k = N - \lfloor (N+1)/4 \rfloor$ is found. $\lfloor (N+1)/4 \rfloor$ symbolizes the greatest integer less or equal to $(N+1)/4$.

It appears that for the properties of the first excited state strict rules do not hold in general, but only for "high enough" N ($N > 5$). Then, as can be anticipated from table 2.1, we can conjecture that **[Sch00]**

- if N is even, then the first excited state has $S = 1$ and is threefold degenerate, and
- if N is odd and the single particle spin is half-integer, then the first excited state has $S = 3/2$ and is eightfold degenerate, whereas
- if N is odd and the single particle spin is integer, then the first excited state has $S = 1$ and is sixfold degenerate.

Except for small odd N also the cyclic shift quantum numbers k of the first excited state show an interesting regularity.

- For odd $N \geq 7$, k assumes a certain value for all integer spins and another value for all half-integer spins. We conjecture that the k quantum numbers for half-integer spins are $k = 3\lfloor (N+1)/4 \rfloor$ and $k = N - 3\lfloor (N+1)/4 \rfloor$. $\lfloor (N+1)/4 \rfloor$ symbolizes the greatest integer less or equal to $(N+1)/4$. For integer spins numerical data are poor, but it seems that k is as close as possible to $N/2$, i.e. $k = \lfloor N/2 \rfloor$ and $k = N - \lfloor N/2 \rfloor$.
- For even N the shift quantum number k is $N/2$, if $N/2$ is also even; if $N/2$ is odd, $k = 0$ for half-integer spin and $k = N/2$ for integer spin.

For spin-$\frac{1}{2}$-rings these properties can be derived using the Bethe ansatz [Bet31, Hul38, Kar94]. For rings of higher spin quantum number and $N > 4$ only numerical evidence could be collected so far.

Having evaluated the spectra of small Heisenberg rings with isotropic next-neighbour interaction one can approximate the infinite chain limit, which for the $s = \frac{1}{2}$ ground state is known as the Bethe-Hulthén limit [Bet31, Hul38]. Because the sequences converge rather slowly and energy eigenvalues can be evaluated only for some small N, the Levin u-sequence acceleration method [Lev73, Lub77] is employed, which leads to impressive estimates of the antiferromagnetic ground state energies as well as of the excitation gap for infinite rings or chains of larger spin quantum numbers **[Sch00]**.

2.3 Rotational modes

It is clear that even Heisenberg systems of relatively modest size pose a major theoretical challenge. A stunning example is provided by the recently synthesized molecular magnet [MSS+99] $\{Mo_{72}Fe_{30}\}$, where the 30 Fe^{3+} ions (spins 5/2) occupy the sites of an icosidodecahedron. The total dimension of the Hilbert space for this spin system is a staggering 6^{30}, namely of order Avogadro's number, precluding the unsophisticated calculation of the energy eigenvalues and eigenvectors on any imagined configuration of immense, ultra-fast computers.

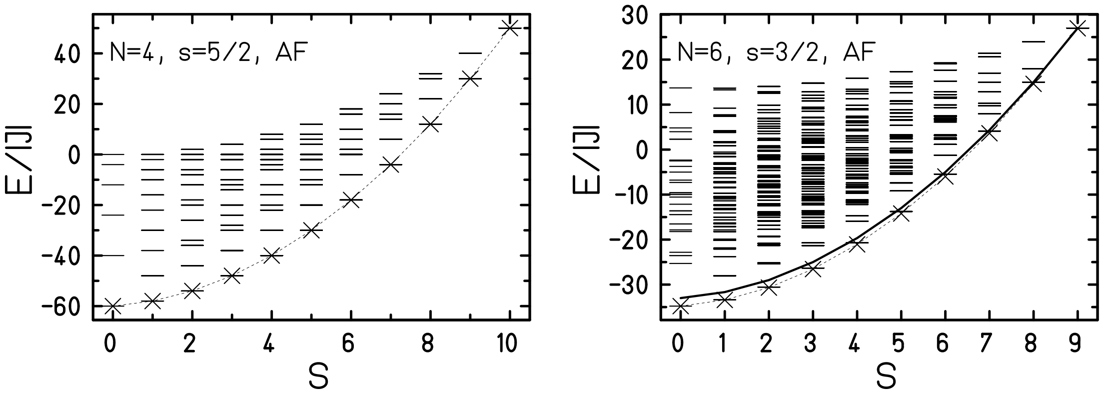

Figure 2.1: Energy spectra of antiferromagnetically coupled Heisenberg spin rings (horizontal dashes). The crosses connected by the dashed line represent the fit to the rotational band, which by definition matches both the lowest and the highest energies exactly. On the l.h.s the dashed line reproduces the exact rotational band, whereas on the r.h.s. it only approximates it, but to high accuracy. The solid line on the r.h.s. corresponds to the approximation following from the sublattice structure, i.e. using $D = 4$ **[SL01]**.

Major low-temperature properties of Heisenberg spin arrays in general and $\{Mo_{72}Fe_{30}\}$ in particular [MLS$^+$] can be understood employing the fact that all these systems possess a low-lying rotational band, i.e. , the minimal energies for each value of total spin S depend quadratically on S

$$E_{S,min} \approx -J\,[D(N,s)/N]\,S(S+1) + E_a \ . \tag{2.2}$$

While this result has previously been noted for special spin arrays like rings with an even number of spin sites (see Fig. 2.1) or triangular lattices [Kar94, BLP92, BLLP94, CCF$^+$96, JJL$^+$99, ACC$^+$00], we find that it also applies for rings with an odd number of sites as well as for all of the polytope configurations we have investigated (tetrahedron, cube, octahedron, icosahedron, triangular prism, and axially truncated icosahedron) **[SL01]**. The coefficient $D(N,s)$ in (2.2) can be approximated using the symmetry of the spin array [BLP92, BLLP94], **[SL01]**, which shows up for instance in the classical ground state configuration. This symmetry related coefficient D turns out to be $D = 4$ for rings of arbitrary, but even N [ACC$^+$00] and $D = 6$ for the icosidodecahedron, cube, and octahedron, independent on s **[SL01]**. The coefficient D reflects the high s limit for the given spin array, $D = D(N, \infty)$. Our investigations **[SL01]** have revealed that the specific $D(N,s)$ for any finite s is always a little larger than D, compare also r.h.s. of Fig. 2.1.

The concept of rotational bands is useful even in cases where $\underset{\sim}{\vec{S}}^2$ does not commute with the Hamilton operator, if the symmetry breaking terms are small and can be treated perturbatively. The eigenstates and eigenenergies of the unperturbed (rotationally symmetric) Hamilton operator can be classified by their total spin quantum number S, and symmetry breaking terms like on-site anisotropy or dipolar interactions will lead to energy corrections [JJL$^+$99].

2.4 Outlook

The rotational band can be understood as originating from interacting sublattice spins [BLP92, BLLP94]. The subdivision into sublattices is given by the symmetry of the spin array and expresses itself in the classical ground state. We conjecture that the picture of interacting sublattice spins holds even for moderate excitations off the rotational band. Therefore we extend the concept of the rotational band towards an approximate Hamilton operator [LSM00], see also [HgZ98],

$$\underset{\sim}{H}_1^{\text{eff}} = -\frac{D\,J}{N}\left[\underset{\sim}{\vec{S}}^2 - \sum_{j=1}^{N_{SL}} \underset{\sim}{\vec{S}}_j^2\right] , \tag{2.3}$$

where the sublattice spin quantum numbers are called S_j. The spin operators $\underset{\sim}{\vec{S}}^2$ and all $\underset{\sim}{\vec{S}}_j^2$ commute with each other and with $\underset{\sim}{H}_1^{\text{eff}}$. Thus the eigenvalues E_1^{eff} of $\underset{\sim}{H}_1^{\text{eff}}$ are easily evaluated and given in terms of the quantum numbers S and S_j. One realizes that higher lying bands are also parabolic and parallel to the ground state band. This is rather realistic for the second band and indeed observed in rings of even N [Wal00]. That the second band is parallel to the first one should be visible in inelastic neutron scattering as well as in EPR measurements [LSM00].

Figure 2.2 demonstrates for$\{\text{Mo}_{72}\text{Fe}_{30}\}$ that the agreement between the approximate quantum model and experiment is excellent. The thin curve displays the result of approximation (2.3) for $T = 4$ K. The dashed curve in Fig. 2.2 gives the experimental data obtained [MLS$^+$] using a pulsed field that led to an estimated effective spin temperature of approximately 4 K. We attribute the very small discrepancy in the slope of the magnetisation curve below the critical field to the fact that the classical coefficient $D = D(N, \infty)$ always underestimates the true coefficient $D(N, s)$ by a few percent **[SL01]**. In order to improve approximation (2.3) we use $D(N, s)$ in the following. We adjust $D(N, s)$ so that the resulting magnetisation curve (thick curve in Fig. 2.2) provides an optimal fit to the measured data. This is achieved by taking $D(N, s) = 6.23$ for $\{\text{Mo}_{72}\text{Fe}_{30}\}$. Thus the effective Hamiltonian becomes

$$\underset{\sim}{H}_2^{\text{eff}} = -J\,\frac{D(N,s)}{N}\left[\underset{\sim}{\vec{S}}^2 - \gamma\left(\underset{\sim}{\vec{S}}_A^2 + \underset{\sim}{\vec{S}}_B^2 + \underset{\sim}{\vec{S}}_C^2\right)\right] , \tag{2.4}$$

with $\gamma = 1.148$ in order to maintain the correct value of the largest energy eigenvalue. Finally, the ground state energy of $\{\text{Mo}_{72}\text{Fe}_{30}\}$ described by $\underset{\sim}{H}_2^{\text{eff}}$ is $E_0/k_B \approx -364$ K as compared to -305 K which follows from Eq. (2.3). We are currently attempting to calculate $D(30, 5/2)$ from first principles using DMRG techniques [Whi93].

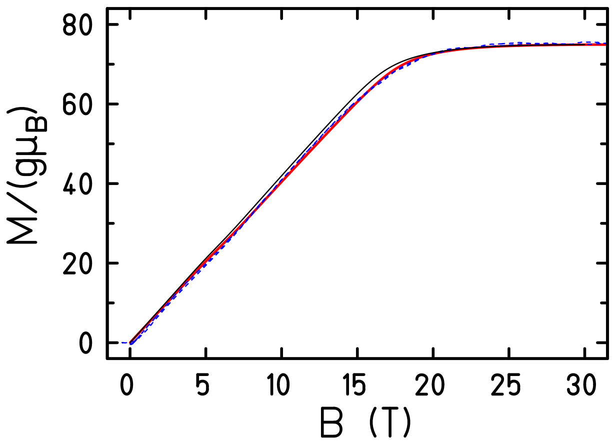

Figure 2.2: Magnetisation versus magnetic field: The thin curve displays the result of approximation (2.3) for $T = 4$ K. Experimental data using a pulsed field are given by the dashed curve. The pulsed field appears to have heated the spin system to approximately 4 K, while the cryostat temperature was 0.46 K. Not shown are error bars due to an uncertainty of ± 0.5 T for the data. The thick curve giving the result of the improved approximation, Eq. (2.4), taking $D(N, s) = 6.23$, also at 4 K, closely reproduces the measured values.

With the upcoming synthesis of yet more advanced and larger compounds with exotic properties it can be expected that the field of magnetic molecules will flourish and provide important challenges to the theoretical description of molecular magnetism.

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	$E_0/(NJ)$
	1	4	1	4	1	4	1	4	1	deg
	0	1/2	0	1/2	0	1/2	0	1/2	0	S
	1	1, 2	0	1, 4	3	2, 5	0	2, 7	5	k
$\frac{1}{2}$	4.0	3.0	2.0	2.236	1.369	2.098	1.045	1.722	0.846	$\Delta E/\vert J\vert$
	3	4	3	2	3	8	3	8	3	deg
	1	3/2	1	1/2	1	3/2	1	3/2	1	S
	0	0	2	0	0	1, 6	4	3, 6	0	k
1	4	2	3	2.612	2.872	2.735	2.834	2.773	2.819	$E_0/(NJ)$
	1	1	1	1	1	1	1	1	1	deg
	0	0	0	0	0	0	0	0	0	S
	0	0	0	0	0	0	0	0	0	k
1	4.0	2.0	2.0	1.929	1.441	1.714	1.187	1.540	1.050	$\Delta E/\vert J\vert$
	3	9	3	6	3	6	3	6	3	deg
	1	1	1	1	1	1	1	1	1	S
	1	0, 1, 2	2	2, 3	3	3, 4	4	4, 5	5	k
$\frac{3}{2}$	7.5	3.5	6	4.973	5.798	5.338	5.732	5.477	5.704††	$E_0/(NJ)$
	1	4	1	4	1	4	1	4	1	deg
	0	1/2	0	1/2	0	1/2	0	1/2	0	S
	1	1, 2	0	1, 4	3	2, 5	0	2, 7	5	k
$\frac{3}{2}$	4.0	3.0	2.0	2.629	1.411	2.171	1.117	1.838	0.938††	$\Delta E/\vert J\vert$
	3	16	3	8	3	8	3	8	3	deg
	1	3/2	1	3/2	1	3/2	1	3/2	1	S
	0	0, 1, 2	2	2, 3	0	1, 6	4	3, 6	0	k
2	12	6	10	8.456	9.722	9.045	9.630	9.263††	9.590††	$E_0/(NJ)$
	1	1	1	1	1	1	1	1	1	deg
	0	0	0	0	0	0	0	0	0	S
	0	0	0	0	0	0	0	0	0	k
2	4.0	2.0	2.0	1.922	1.394	1.652	1.091	1.431††	0.906††	$\Delta E/\vert J\vert$
	3	9	3	6	3	6	3	6	3	deg
	1	1	1	1	1	1	1	1	1	S
	1	0, 1, 2	2	2, 3	3	3, 4	4	4, 5	5	k
$\frac{5}{2}$	17.5	8.5	15	12.434	14.645	13.451	14.528†	13.848††	14.475††	$E_0/(NJ)$
	1	4	1	4	1	4	1	4	1	deg
	0	1/2	0	1/2	0	1/2	0	1/2	0	S
	1	1, 2	0	1,4	3	2, 5	0	2, 7	5	k

Table 2.1: Properties of ground and first excited state of antiferromagnetically coupled Heisenberg rings for various N and s: ground state energy E_0, gap ΔE, degeneracy *deg*, total spin S and shift quantum number k. † – O. Waldmann, private communication. †† – projection method [Man91].

3. Ideal Bose and Fermi systems

Ideal quantum gases are usually treated in the thermodynamic limit, i.e. infinitely spread at a certain density, since all applications which were important in the past, like the electron gas, phonons or photons, deal with huge particle numbers. Only the experimental attempts of the last years to investigate finite Fermi and Bose systems and to describe them in terms of thermodynamics called for new theoretical effort.

Interesting finite Fermi systems are for instance nuclei, which behave like a liquid drop and therefore can undergo a first order liquid-gas-like phase transition [JMZ84, GKM84, BLV84, BLV85, SW86, SCG$^+$89, PMR$^+$95]. On the low excitation site of the caloric curve the nuclear systems might be well described as an ideal Fermi gas in a common harmonic oscillator potential (shell model).

Small Bose systems became available through the development of traps. Here the focus is on the Bose-Einstein condensation which for instance could be found investigating dilute atomic vapours (e.g. alkali atoms) in magnetic traps [AEM$^+$95, DMA$^+$95, KvD96, BSH97]. Again the system can be well described as an ideal quantum gas contained in an external harmonic oscillator potential, for an overview see [DGPS99].

Rigorously speaking a phase transition towards the Bose-Einstein condensate does not happen in an external potential since the chemical potential is always non-zero for finite temperatures [Hua87]. Only for a vanishing potential (infinite volume limit) it is zero for all temperatures below T_c. It is also true that this phase transition does only occur for dimensions $d > 2$. Dealing with finite systems one has to relax the rigorous point of view and look for criteria which could identify a “smooth Bose-Einstein condensation”.

The investigated ideal quantum gases are characterized by a constant particle number and may be represented by the canonical or the micro-canonical ensemble [GH96, GH97] depending on whether heat exchange with the environment is possible or not.

The work presented in this chapter was done with H.-J. Schmidt (Universität Osnabrück), the respective publications **[SS98, SS99]** are presented in section B on page 37.

3.1 Canonical ensemble of fermions and bosons in harmonic traps

Although the micro-canonical, canonical and grand-canonical ensembles are connected by integral transformations and one would expect them to lead to the same results at least for large particle numbers, some observables like the fluctuation of the condensate fraction are rather sensitive to the choice of the ensemble. For the following investigations we choose the canonical ensemble, because the particle number is rather constant and heat exchange with the environment possible. This is of course already a simplification of real experiments, since the systems are not really in equilibrium, losing for instance particles due to evaporative cooling or creation of bound states.

The partition function of the canonical ensemble of non-interacting fermions and bosons in harmonic traps was already investigated in a book by Peter Landsberg [Lan61], but later forgotten and rediscovered [BF93, BLD97], **[SS98]**.

It turns out that the partition function Z_N for the canonical ensemble of N identical and non-interacting particles can be recursively built starting with the single-particle partition function

$$Z_N(\beta) = \frac{1}{N} \sum_{n=1}^{N} (\pm 1)^{n+1}\, Z_1(n\beta)\, Z_{N-n}(\beta) \ , \quad Z_0(\beta) = 1 \ , \ \beta = \frac{1}{k_B T} \ , \tag{3.1}$$

where the upper sign in the sum stands for bosons, the lower sign for fermions. Mean energy and specific heat can be derived by partial derivatives as usual. For the particle densities a similar recursion relation holds [BF93], **[SS98]**.

Regarding the question how to characterize the Bose-Einstein condensation in finite systems, a first natural criterion is the maximum in the specific heat [PP77, KT96], which is a kink in the free three-dimensional case because the mean energy jumps at T_c. Looking at figures 3.1 and 3.2 (upper right corner) one sees that the specific heat exhibits a clear maximum in the three-dimensional oscillator (Fig. 3.2) whereas it does not in the one-dimensional oscillator (Fig. 3.1). Moreover, the specific heat of N bosons in the one-dimensional oscillator is the same as the specific heat of N fermions in the same oscillator **[SS98]**! Therefore, although it seems to be experimentally favourable to investigate quasi one-dimensional Bose gases it is doubtful whether the effects unveil much about Bose-Einstein condensation [DGPS99]. The mere fact of a macroscopic occupation of the single-particle ground state [KvD96] is not so exciting from a theorists point of view since that happens also for distinguishable particles, see figures 3.1 and 3.2 (lower left) and **[SS98]**.

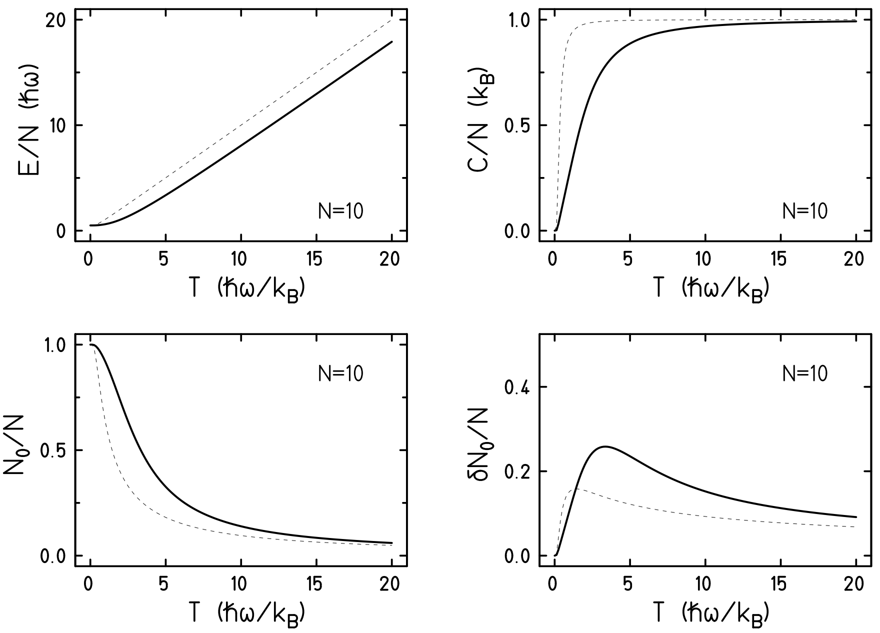

Figure 3.1: One-dimensional harmonic oscillator: mean energy, specific heat, ground state occupation number and its fluctuation for bosons (solid line) and distinguishable particles (dashed line) **[SS98]**.

3.2 Fermion-boson symmetry

A closer inspection of the canonical partition function uncovers a surprising symmetry property which connects fermions and bosons contained in harmonic oscillator potentials of odd space dimensions **[SS99]**,

$$Z_N^+(\beta) = (-1)^N Z_N^-(-\beta) \ , \tag{3.2}$$

where the partition function with the negative argument has to be understood as the analytic continuation into the region of negative temperatures. In thermodynamic mean values like mean energy or specific heat this symmetry shows up as

$$E_N^+(\beta) = -E_N^-(-\beta) \quad , \qquad C_N^+(\beta) = C_N^-(-\beta) \ . \tag{3.3}$$

A straight forward application of the above result is to calculate fermionic partition functions and mean values by evaluating the respective bosonic ones at negative temperatures and thereby to avoid the “sign problem”, which denotes the poor convergence of alternating sequences like fermionic partition functions due to massive cancellations in the summation.

More generally, the property (3.2) is related to the fact that the single-particle partition function has an analytic continuation to the whole β-axis where it is an odd function,

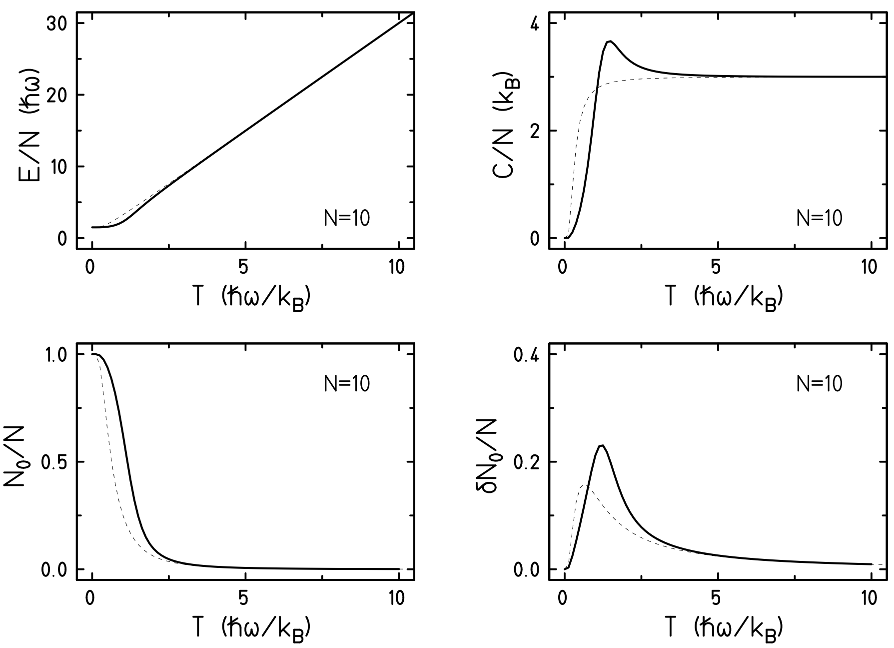

Figure 3.2: Three-dimensional harmonic oscillator: mean energy, specific heat, ground state occupation number and its fluctuation for bosons (solid line) and distinguishable particles (dashed line) **[SS98]**.

$Z_1(-\beta) = -Z_1(\beta)$. Thus the fermion-boson symmetry depends only on the oddness of Z_1 and not on the form of the single-particle Hamiltonian.

3.3 Outlook

The above presented recursion relation allows to evaluate properties of non-interacting Fermi and Bose gases starting from their single-particle partition function, which can be evaluated analytically in some and numerically in the other cases. But the atomic vapours, although dilute, consist of interacting bosons or fermions. A direct evaluation of the partition function, now for interacting particles, is no longer possible and approximations are used. Among them mean-field approximations (Gross-Pitaevskii) [Gro61, Pit61a, Pit61b, Gro63, DGPS99], quantum Monte-Carlo calculations [KDL97, GCL97], and path-integral methods [Cep95, BLD97, BDL97, FBDL99, TBLD00] are very popular. The next chapter describes an alternative approach which aims at an evaluation of thermodynamic mean values by time-averaging.

4. Thermostated quantum dynamics

Statistical properties of finite interacting systems are of great interest. The aim is to describe the behaviour of systems like atomic clusters, atomic vapours or atomic nuclei at finite temperatures and to investigate properties like the specific heat or phase transitions. These statistical properties are given by the partition function which for classical systems in the canonical ensemble reads

$$Z = \int \prod_{i=1}^{N} \mathrm{d}^3 x_i \, \mathrm{d}^3 p_i \, \exp\left\{ -\frac{H}{k_B T} \right\} , \tag{4.1}$$

where H denotes the Hamilton function, and for quantum systems

$$Z = \mathrm{tr}\left(\exp\left\{ -\frac{\underset{\sim}{H}}{k_B T} \right\} \right) , \tag{4.2}$$

where $\underset{\sim}{H}$ is the Hamilton operator. For realistic systems like atomic clusters or nuclei where the Hamilton function or operator contains a (two-body) interaction it is hard or impossible to evaluate the partition function especially for the quantum description.

Equations of motion for the investigated system are often much easier; either they are exactly known and can be integrated at least numerically as it is the case with the classical Hamilton's equation or they can be approximated with standard methods like Time-dependent Hartree-Fock (TDHF) or quantum molecular dynamics methods as it is the case on the quantum side. The idea then is to extract the desired thermodynamic quantities from the time evolution of the system. If the system is ergodic, ensemble averages can be replaced by time averages.

During the last decade a huge progress has been made on the classical side of the problem (see for instance [Nos84, Hoo85, KB90, KBB90, KBB91, Nos91]). To put it into a few words, the basic idea is to exploit the equipartition theorem and to use for instance the kinetic energy as a measure of the current temperature.

In quantum mechanics the problem is much more involved. No useful a-priori-relation between expectation values of observables and temperature like in the equipartition theorem can be exploited. Attempts have been made to derive a thermal non-linear Schrödinger equation which results in an ergodic wave function [Kus93], but this method needs thermodynamic relations as an input, which are not known for most systems and therefore itself a matter of investigation.

The first idea presented here is to couple an additional degree of freedom to the original system, which serves as a thermometer. This idea was already successfully applied in nuclear physics in order to determine the caloric curve of nuclei and to investigate the nuclear liquid-gas phase transition **[SF97]**. The second step is to construct a feedback algorithm in order to drive the system via complex time steps towards the desired temperature **[Sch98]**. A third ongoing project tries to develop a Nosé-like thermostat for quantum systems starting from previous work by Grilli and Tosatti [GT89].

The work presented in this chapter was done in part with H. Feldmeier (GSI, Darmstadt), the respective publications **[SF97, FS97, FNRS98, Sch98, Sch99a, FS00]** are presented in section C on page 38.

4.1 Molecular dynamics approach

In cases where the (canonical) partition function is not known the time–dependent Schrödinger equation cannot be solved either. The following time-dependent quantum variational principle (TDVP) [KK76, KS81]

$$\delta \int_{t_1}^{t_2} dt \, \langle\, Q(t) \,|\; i\frac{d}{dt} - \underset{\sim}{H} \;|\, Q(t) \,\rangle \quad = \quad 0 \tag{4.3}$$

allows to derive approximations to the time–dependent Schrödinger equation on the level of accuracy one needs or can afford. For the variation of the trial state $\langle\, Q(t) \,|$ in the complete Hilbert space the TDVP leads to the Schrödinger equation. Taking (anti-symmetrized) product states of wave packets, i.e. coherent states [KS85], as trial states leads to all kinds of quantum mechanical molecular dynamics models [Hel75, DPC86, Fel90, KD97]. One of the latter is the model of Fermionic Molecular Dynamics (FMD) [Fel90, FBS95], **[FS97, FS00]**, which describes many-fermion systems with Slater determinants of Gaussian wave packets with time-dependent width. In addition the trial states can be correlated in order to account for short-ranged repulsion **[FNRS98]**. Although the model was primarily developed to simulate nuclear reactions, which are non-equilibrium phenomena, it is also capable to address equilibrium situations.

It is a genuine feature of approximations derived from Eq. (4.3) that the resulting time evolution is in general non-unitary, i.e. $\langle\, \underset{\sim}{H} \,\rangle$ is a constant of motion but not the higher moments $\langle\, \underset{\sim}{H}^n \,\rangle\, , n > 1$. Strictly speaking, this improves the ergodicity of time-averages because the equations of motion are non-linear and chaotic, but unfortunately in a rather uncontrollable way.

Another important question is whether or under which circumstances one can expect that time-averages converge against the proper limit, since the quantum dynamics is restricted, see for instance [OR93, SF96], **[Sch99a]**.

Nevertheless, the idea to derive thermal properties from time-averages of an approximate quantum molecular dynamics is not only used in nuclear but also in atomic and plasma physics, for instance to describe hydrogen plasmas under high preasure, see e.g. [KTR94, ES97].

4.2 Phase transitions in nuclei

Mean-field models predict a first order phase transition for nuclear matter with a critical temperature which depends on the proton-neutron asymmetry [JMZ84, GKM84, BLV84, BLV85, SW86, SCG$^+$89]. A recent experimental attempt by the ALADIN group [PMR$^+$95] to deduce an equation of state, which relates the excitation energy of a hot nucleus to its temperature, has stimulated both, theoretical and experimental efforts in this field [NHW$^+$95, MS95, PN95, MGP$^+$96, PMR$^+$96, FBK$^+$96, XLTF96]. In the experiment excited projectile spectators were investigated in Au+Au collisions at a beam energy of $E/A = 600$ MeV. While the equation of state refers to a stationary system where liquid and vapour (evaporated nucleons) are in equilibrium, the experiment deals with an expanding source. This causes some uncertainties for the temperature, which is deduced from isotope ratios, since the system cools while it is expanding. Thus the temperature has to be understood as belonging to a transient equilibrium.

In molecular-dynamics calculations the finite system may be excited avoiding collective expansion, but similar problems arise when the phase transition sets in. Particles which escape from the nucleus cool down the residue and thermal equilibrium cannot be maintained. In order to avoid these difficulties, in the present simulations the excited nuclear system is confined by a wide container potential which is chosen to be a harmonic oscillator potential. Its frequency ω serves as a thermodynamic variable like the volume in the ideal gas case. Due to the containment evaporated nucleons cannot escape, but form a cloud of equilibrated vapour around the excited nucleus.

Thermodynamic relations are obtained by coupling the nuclear system to a reference system which serves as a thermometer, see Fig. 4.1. Both, the time-evolution of the nuclear system and of the thermometer are described by the Fermionic Molecular Dynamics (FMD) model. Assuming thermal equilibrium in the sense of ergodicity the temperature of the nuclear system is derived from the time-averaged energy of the thermometer and related to the excitation energy of the nucleus. The resulting caloric curve – evaluated for ^{24}Mg, ^{27}Al and ^{40}Ca (Fig. 4.2, l.h.s.) – shows the clear structure of a first-order phase transition and has a striking similarity with the experimentally determined one (Fig. 4.2, r.h.s.) **[SF97]**.

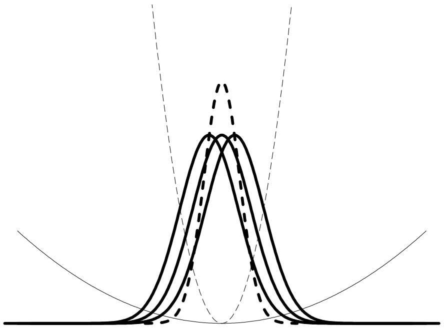

Figure 4.1: Sketch of the setup: the self-bound excited nuclear system is represented by Gaussian wave packets (solid lines) which are enclosed in a broad container potential. For the thermometer only one wave packet is shown (dashed line), it is contained in a different oscillator.

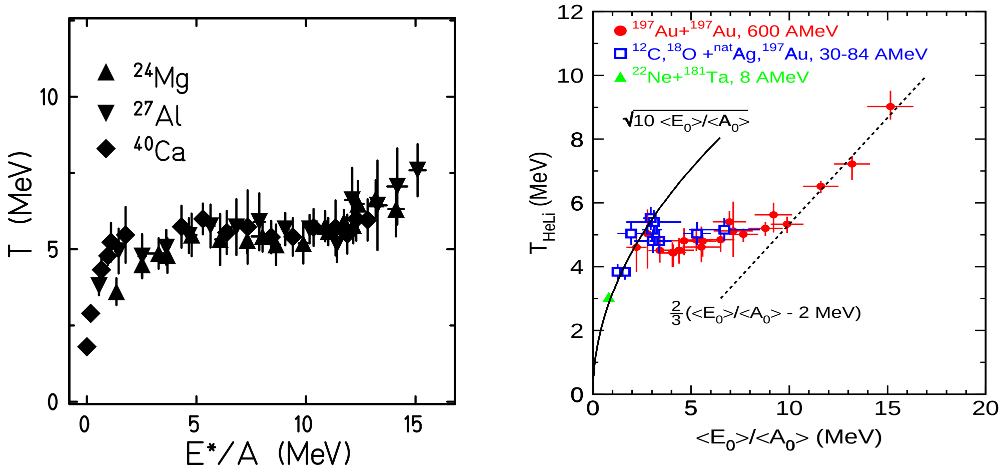

Figure 4.2: Caloric curve: l.h.s. FMD **[SF97]**, r.h.s. experimental values [PMR$^+$95, Poc97].

4.3 Thermostated dynamics

Thermostated dynamics is a very powerful method to determine thermal properties for classical systems [Nos84, Hoo85, KB90, KBB90, KBB91, Nos91]. In the simplest scheme the system is cooled or heated via pseudo-friction coefficients if the present temperature is too high or too low, respectively. Hamilton's equations of motion are

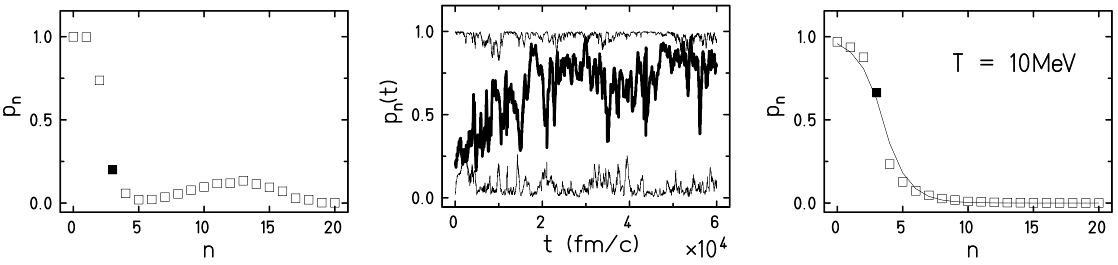

Figure 4.3: Occupation probabilities p_n; l.h.s.: distribution at $t = 0$; middle: time evolution $p_n(t)$ for $n = 0$ — top line, $n = 3$ — middle, $n = 6$ — bottom line; r.h.s.: time averaged distribution (symbols), the solid curve shows the canonical result expected for the desired temperature.

therefore extended in the following way

$$\frac{d}{dt}\vec{x}_k = \frac{\partial H}{\partial \vec{p}_k}\,, \quad \frac{d}{dt}\vec{p}_k = -\frac{\partial H}{\partial \vec{x}_k} - \vec{p}_k\,\zeta\,, \qquad k = 1,\dots,N \tag{4.4}$$

$$\frac{d}{dt}\zeta \propto \left(E_{kin}(N) - \frac{3\,N}{2}T\right)\,. \tag{4.5}$$

The last line, equation (4.5), shows nicely how the system is driven towards the temperature T via the coupling to the pseudo-friction coefficient ζ. It was shown that the resulting distributions are those of the canonical ensemble (see e.g. the report [Nos91]). In addition ergodicity can be improved by using more pseudo-friction coefficients and different couplings to the original system [KBB90].

The ansatz chosen to equilibrate a model quantum system **[Sch98]** is to describe the total system by a time-dependent state $|\,Q(t)\,\rangle$ consisting of the original system $|\,system(t)\,\rangle$ and the thermometer $|\,thermometer(t)\,\rangle$

$$|\,Q(t)\,\rangle = |\,system(t)\,\rangle \otimes |\,thermometer(t)\,\rangle\,, \tag{4.6}$$

and to install a feedback mechanism which uses the difference between the temperature T_{th} measured by the thermometer and the desired temperature T. Then the total system is evolved by a complex time step $d\tau$,

$$d\tau = dt - i d\beta\,, \quad d\beta \propto (T_{th} - T)/T_{th}\,, \quad |\,Q(t)\,\rangle \to |\,Q(t + d\tau)\,\rangle\,, \tag{4.7}$$

where the imaginary part is directly proportional to the temperature difference. As can be inferred from (4.7) $d\beta > 0$ results in cooling and $d\beta < 0$ in heating of the system.

The results of time averaging are compared to the canonical ensemble of the ideal Fermi gas enclosed in a common harmonic oscillator potential. It is demonstrated that the system is indeed ergodic, i.e. the time averaged occupation numbers coincide with those obtained in the canonical ensemble, see Fig. 4.3 and **[Sch98]**.

4.4 Outlook

Methods like the two introduced above suffer from the disturbance of the investigated system by the thermometer system. It is clear that the necessary interaction between system and thermometer introduces unwanted correlations especially at low temperatures. Therefore, in an ongoing project we try to construct a Nosé-like thermostat for quantum systems starting from previous work by Grilli and Tosatti [GT89].

The idea of Ref. [GT89] is to scale the coordinate and momentum operators by a time-dependent function s as follows

$$\underset{\sim}{\vec{x}} \rightarrow s\,\underset{\sim}{\vec{x}} \quad , \qquad \underset{\sim}{\vec{k}} \rightarrow \frac{\underset{\sim}{\vec{k}}}{s} \;, \tag{4.8}$$

which leaves the canonical commutation relations invariant, i.e. $[\underset{\sim}{\vec{x}}, \underset{\sim}{\vec{k}}] = [s\underset{\sim}{\vec{x}}, \underset{\sim}{\vec{k}}/s]$, and to replace the Hamilton operator $\underset{\sim}{H}$ by

$$\sum_{i=1}^{N} \frac{\underset{\sim}{\vec{k}}{}_i^2}{2\,m_i\,s^2} + \underset{\sim}{V}\left(\left\{s\,\underset{\sim}{\vec{x}}_i\right\}\right) + \frac{p_s^2}{2M} + T\ln s \;, \tag{4.9}$$

where s is an additional degree of freedom, known from the Nosé-method [Nos84, Nos91], which simulates the coupling to the infinite heat bath. The authors of Ref. [GT89] brought forward the argument that in principle such a scaling leads to the canonical ensemble for the original system assuming an ergodic micro-canonical ensemble for the combined system which includes the new degree of freedom s. In practice they had to supplement additional fluctuating forces of Langevin type with temperatures that needed adjustment during the time evolution.

In our attempt to investigate the method systematically we follow the time-evolution both by numerical exact integration of the Schrödinger equation and by means of Fermionic Molecular Dynamics. First results show, that the proposed method does not hold what the authors promise. Nevertheless, it is possible to exchange the proposed scaling by general unitary transformations which depend parametrically on the scaling variable s [SMF01].

A success of our effort would provide a very valuable method to investigate complex systems – bosons, fermions, molecules and clusters – at finite temperatures. A general technique like this would contribute in atomic, molecular, nuclear and solid state physics as well as in quantum chemistry.

Bibliography

[ACC$^+$00] Gian Luca Abbati, Andrea Caneschi, Andrea Cornia, Antonio C. Fabretti, and Dante Gatteschi, *[Fe(OCH3)2(dbm)]12: synthesis, solid-state characterization and reactivity of a new molecular ferric wheel.*, Inorg. Chim. Acta **297** (2000), 291.

[AEM$^+$95] M.H. Anderson, J.R. Ensher, M.R. Matthews, C.E. Wieman, and E.A. Cornell, *Observation of Bose-Einstein condensation in a dilute atomic vapour*, Science **269** (1995), 198.

[BDL97] Fons Brosens, Jozef T. Devreese, and L.F. Lemmens, *Confined harmonically interacting spin-polarised fermions*, Phys. Rev. E **57** (1997), 3871.

[Bet31] Hans Bethe, Z. Phys. **71** (1931), 205.

[BF64] J.C. Bonner and M.E. Fisher, *Linear magnetic chains with anisotropic coupling*, Phys. Rev. **135** (1964), A640.

[BF93] Peter Borrmann and G. Franke, *Recursion formulas for quantum statistical partition functions*, J. Chem. Phys. **98** (1993), 2484.

[BG90] A. Bencini and Dante Gatteschi, *Electron parametric resonance of exchange coupled systems*, Springer, Berlin, Heidelberg, 1990.

[BJ83] R. Botet and R. Jullien, *Ground-state properties of a spin-1 antiferromagnetic chain*, Phys. Rev. B **27** (1983), 631.

[BL89] E. Balcar and S.W. Lovesey, *Theory of magnetic neutron and photon scattering*, Clarendon, Oxford, 1989.

[BLD97] Fons Brosens, L.F. Lemmens, and Jozef T. Devreese, *Thermodynamics of coupled identical oscillators within the path integral formalism*, Phys. Rev. E **55** (1997), 227.

[BLLP94] Bernard Bernu, Philippe Lecheminant, Claire Lhuillier, and L. Pierre, *Exact spectra, spin susceptibilities, and order parameter of the quantum Heisenberg antiferromagnet on the triangular lattice*, Phys. Rev. B **50** (1994), 10048.

[BLP92] Bernard Bernu, Claire Lhuillier, and L. Pierre, *Signature of Néel order in exact spectra of quantum antiferromagnets on finite lattices*, Phys. Rev. Lett. **69** (1992), 2590.

[BLV84] P. Bonche, S. Levit, and D. Vautherin, *Properties of highly excited nuclei*, Nucl. Phys. A **427** (1984), 278.

[BLV85] P. Bonche, S. Levit, and D. Vautherin, *Statistical properties and stability of hot nuclei*, Nucl. Phys. A **436** (1985), 265.

[BSH97] C.C. Bradley, C.A. Sackett, and R.G. Hulet, *Bose-Einstein condensation of Lithium: Observation of limited condensate number*, Phys. Rev. Lett. **78** (1997), 985.

[BSS00a] Klaus Bärwinkel, Heinz-Jürgen Schmidt, and Jürgen Schnack, *Ground state properties of antiferromagnetic Heisenberg spin rings*, J. Magn. Magn. Mater. **220** (2000), 227.

[BSS00b] Klaus Bärwinkel, Heinz-Jürgen Schmidt, and Jürgen Schnack, *Structure and relevant dimension of the Heisenberg model and applications to spin rings*, J. Magn. Magn. Mater. **212** (2000), 240.

[CCF$^+$95] Andrea Caneschi, Andrea Cornia, Antonio C. Fabretti, Dante Gatteschi, and Wanda Malavasi, *Polyiron(III)-Alkoxo Clusters: A novel Trinuclear Complex and Its Relevance to the Extended Lattices of Iron Oxides and Hydroxides*, Inorg. Chem. **34** (1995), 4660.

[CCF$^+$96] Andrea Caneschi, Andrea Cornia, Antonio C. Fabretti, Simon Foner, Dante Gatteschi, Romano Grandi, and Luisa Schenetti, *Synthesis, crystal structure, magnetism, and magnetic anisotropy of cyclic clusters comprising six iron(III) ions and entrapping alkaline ions*, Chem. Eur. J. **2** (1996), 1379.

[CDGM96] E. Coronado, P. Delhaes, Dante Gatteschi, and J.S. Miller (eds.), *Localized and Itinerant Molecular Magnetism: From Molecular Assemblies to the Devices*, NATO Advanced Studies Institute, Series E: Applied Sciences, vol. 321, Dordrecht, Kluwer Academic, 1996.

[Cep95] David M. Ceperley, *Path-integrals in the theory of condensed helium*, Rev. Mod. Phys. **67** (1995), 279.

[DGP$^+$93] C. Delfs, Dante Gatteschi, L. Pardi, Roberta Sessoli, K. Wieghardt, and D. Hanke, *Magnetic properties of an octanuclear iron(III) cation*, Inorg. Chem. **32** (1993), 3099.

[DGPS99] Franco Dalfovo, Stefano Giorgini, Lev P. Pitaevskii, and Sandro Stringari, *Theory of Bose-Einstein condensation in trapped gases*, Rev. Mod. Phys. **71** (1999), 463.

[DMA$^+$95] K.B. Davis, M.-O. Mewes, M.R. Andrews, N.J. van Druten, D.S. Durfee, D.M. Kurn, and Wolfgang Ketterle, *Bose-Einstein condensation of a gas of sodium atoms*, Phys. Rev. Lett. **75** (1995), 3969.

[DPC86] S. Drożdż, M. Płoszajczak, and E. Caurier, *Variational approach to the Schrödinger dynamics in the Klauder's continuous representations*, Ann. Phys. **171** (1986), 108.

[ES97] Werner Ebeling and Friedemann Schautz, *Many particle simulations of the quantum electron gas using momentum-dependent potentials*, Phys. Rev. E **56** (1997), 3498.

[FBDL99] S. Foulon, Fons Brosens, Jozef T. Devreese, and L.F. Lemmens, *Confined harmonically interacting spin-polarized fermions in a magnetic field: Thermodynamics*, Phys. Rev. E **59** (1999), 3911.

[FBK$^+$96] P. Finocchiaro, M. Belkacem, T. Kubo, V. Latora, and A. Bonasera, *Second order phase transition: from infinite to finite systems*, Nucl. Phys. A **600** (1996), 236.

[FBS95] Hans Feldmeier, Konrad Bieler, and Jürgen Schnack, *Fermionic Molecular Dynamics for Ground States and Collisions of Nuclei*, Nucl. Phys. A **586** (1995), 493.

[Fel90] Hans Feldmeier, *Fermionic Molecular Dynamics*, Nucl. Phys. A **515** (1990), 147.

[FLB$^+$00] Y. Furukawa, Marshall Luban, Ferinando Borsa, D.C. Johnston, A.V. Mahajan, L.L. Miller, Detlef Mentrup, Jürgen Schnack, and A. Bino, *Spin dynamics of the magnetic cluster* $[Cr_4S(O_2CCH_3)_8(H_2O)_4](NO_3)_2H_2O$, Phys. Rev. B **61** (2000), 8635.

[FLMU91] Klaus Fabricius, Ute Löw, K.-H. Mütter, and Peer Ueberholz, *Complete Solution of the antiferromagnetic Heisenberg rings with* $N = 12$–16 *sites*, Phys. Rev. B **44** (1991), 7476.

[FLS97] Klaus Fabricius, Ute Löw, and Joachim Stolze, *Dynamic correlations of antiferromagnetic spin-*$\frac{1}{2}$ *XXZ chains at arbitrary temperature from complete diagonalization*, Phys. Rev. B **55** (1997), 5833.

[FNRS98] Hans Feldmeier, Thomas Neff, Robert Roth, and Jürgen Schnack, *A Unitary Correlation Operator Method*, Nucl. Phys. A **632** (1998), 61.

[FS97] Hans Feldmeier and Jürgen Schnack, *Fermionic Molecular Dynamics*, Prog. Part. Nucl. Phys. **39** (1997), 393.

[FS00] Hans Feldmeier and Jürgen Schnack, *Molecular Dynamics for Fermions*, Rev. Mod. Phys. **72** (2000), 655.

[Gat94] Dante Gatteschi, *Molecular Magnetism: a Basis for New Materials*, Adv. Mater. **6** (1994), 635.

[GCL97] Peter Grüter, David Ceperley, and Frank Laloë, *Critical temperature of Bose-Einstein condensation of hard-sphere gases*, Phys. Rev. Lett. **79** (1997), 3549.

[GCPS94] Dante Gatteschi, Andrea Caneschi, L. Pardi, and Roberta Sessoli, *Large clusters of metal ions: The transition from molecular to bulk magnets*, Science **265** (1994), 1054.

[GH96] Siegfried Grossmann and Martin Holthaus, *Microcanonical fluctuations of a Bose system's ground state occupation number*, Phys. Rev. E **54** (1996), 3495.

[GH97] Siegfried Grossmann and Martin Holthaus, *Fluctuations of the particle number in a trapped Bose-Einstein condensate*, Phys. Rev. Lett. **79** (1997), 3557.

[GHS94] Philipp Gütlich, Andreas Hauser, and Hartmut Spiering, *Thermisch und optisch schaltbare Eisen(II)-Komplexe*, Angew. Chem. **106** (1994), 2109.

[GJL94] O. Golinelli, Thierry Jolicœur, and R. Lacaze, *Finite-lattice extrapolations for a Haldane gap antiferromagnet*, Phys. Rev. B **50** (1994), 3037.

[GKM84] Alan L. Goodman, Joseph I. Kapusta, and Aram Z. Mekjian, *Liquid-gas phase instabilities and droplet formation in nuclear reactions*, Phys. Rev. C **30** (1984), 851.

[GNS$^+$98] A.M. Gomes, M.A. Novak, Roberta Sessoli, Andrea Caneschi, and Dante Gatteschi, *Specific heat and magnetic relaxation of the quantum nanomagnet* $Mn_{12}Ac$, Phys. Rev. B **57** (1998), 5021.

[Gro61] E.P. Gross, Nuovo Cimento **20** (1961), 454.

[Gro63] E.P. Gross, J. Math. Phys. **4** (1963), 195.

[GT89] M. Grilli and E. Tosatti, *Exact canonical averages from microcanonical dynamics for quantum systems*, Phys. Rev. Lett. **62** (1989), 2889.

[Hel75] Eric J. Heller, *Time-dependent approach to semiclassical dynamics*, J. Chem. Phys. **62** (1975), 1544.

[HgZ98] Christopher L. Henley and Nai gong Zhang, *Semiclassical eigenstates of four-sublattice antiferromagnets*, Phys. Rev. Lett. **81** (1998), 5221.

[Hoo85] William G. Hoover, *Canonical dynamics: Equilibrium phase-space distributions*, Phys. Rev. A **31** (1985), 1685.

[Hua87] Kerson Huang, *Statistical mechanics*, John Wiley & Sons, New York, 1987.

[Hul38] L. Hulthén, Arkiv Mat. Astron. Fysik **26A** (1938), no. 11.

[JJL$^+$99] M.-H. Julien, Z.H. Jang, Alessandro Lascialfari, Ferdinando Borsa, M. Horvatić, Andrea Caneschi, , and Dante Gatteschi, *Proton NMR for measuring quantum level crossing in the magnetic molecular ring Fe10*, Phys. Rev. Lett. **83** (1999), 227.

[JMZ84] H.R. Jaqaman, A.Z. Mekjian, and L. Zamik, *Liquid-gas phase transition in finite nuclear matter*, Phys. Rev. C **29** (1984), 2067.

[Kar94] Michael Karbach, *Finite-Size-Effekte im eindimensionalen Spin-$\frac{1}{2}$-XXZ-Modell*, Ph.D. thesis, Bergische Universität - Gesamthochschule Wuppertal, 1994.

[KB90] Dimitri Kusnezov and Aurel Bulgac, *Canonical ensembles averages from pseudomicrocanonical dynamics*, Phys. Rev. A **42** (1990), 5045.

[KBB90] Dimitri Kusnezov, Aurel Bulgac, and Wolfgang Bauer, *Canonical ensembles from chaos*, Ann. of Phys. **204** (1990), 155.

[KBB91] Dimitri Kusnezov, Aurel Bulgac, and Wolfgang Bauer, *Canonical ensembles from chaos II: Constrained dynamical systems*, Ann. of Phys. **214** (1991), 180.

[KD97] Dieter Kiderlen and Pawel Danielewicz, *Fragments in gaussian wave-packet dynamics with and without correlations*, Nucl. Phys. A **620** (1997), 346.

[KDL97] Steven E. Koonin, D.J. Dean, and K. Langanke, *Shell model Monte Carlo methods*, Phys. Rep. **278** (1997), 1.

[KK76] A.K. Kerman and Steven E. Koonin, *Hamiltonian formulation of time-dependent variational principles for the many-body system*, Ann. Phys. **100** (1976), 332.

[Kou97] Dimitris Kouzoudis, *Heisenberg $s = \frac{1}{2}$ ring consisting of a prime number of atoms*, J. Magn. Magn. Mater. **173** (1997), 259.

[Kou98] Dimitris Kouzoudis, *Exact analytical partition function and energy levels for a Heisenberg ring of $N = 6$ spin 1/2 sites*, J. Magn. Magn. Mater. **189** (1998), 366.

[KS81] Peter Kramer and Marcos Saraceno, *Geometry of the Time-Dependent Variational Principle in Quantum Mechanics*, Lecture Notes in Physics, no. 140, Springer, 1981.

[KS85] John R. Klauder and B.-S. Skagerstam, *Coherent states*, World Scientific Publishing, Singapore, 1985.

[KT96] Klaus Kirsten and David J. Toms, *Bose-Einstein condensation of atomic gases in a general harmonic-oscillator confining potential trap*, Phys. Rev. A **54** (1996), 4188.

[KTR94] D. Klakow, Christian Toepffer, and Paul-Gerhard Reinhard, *Hydrogen under extreme conditions*, Phys. Lett. A **192** (1994), 55.

[Kus93] Dimitri Kusnezov, *Quantum ergodic wave functions from a thermal non-linear Schrödinger equation*, Phys. Lett. A **184** (1993), 50.

[KvD96] Wolfgang Ketterle and N.J. van Druten, *Bose-Einstein condensation of a finite number of particles trapped in one or three dimensions*, Phys. Rev. A **54** (1996), 656.

[Lan61] Peter T. Landsberg, *Thermodynamics with quantum statistical illustrations*, Interscience Publishers, New York, London, 1961.

[Lev73] D. Levin, *Development of non-linear transformations for improving convergence of sequences*, Intern. J. Computer Math. B **3** (1973), 371.

[LGBC97a] Alessandro Lascialfari, Dante Gatteschi, Ferdinando Borsa, and Andrea Cornia, *Erratum: Spin dynamics in mesoscopic size magnetic systems: A 1H NMR study in rings of iron(III) ions [Phys. Rev. B 55, 14341 (1997)]*, Phys. Rev. B **56** (1997), 8434.

[LGBC97b] Alessandro Lascialfari, Dante Gatteschi, Ferdinando Borsa, and Andrea Cornia, *Spin dynamics in mesoscopic size magnetic systems: A 1H NMR study in rings of iron(III) ions*, Phys. Rev. B **55** (1997), 14341.

[LLB98] James H. Luscombe, Marshall Luban, and Ferdinando Borsa, *Classical Heisenberg model of magnetic ring clusters: Accurate approximations for correlation functions and susceptibility*, J. Chem. Phys. **108** (1998), 7266.

[LM62] Elliot H. Lieb and Daniel C. Mattis, *Ordering energy levels of interacting spin systems*, J. Math. Phys. **3** (1962), 749.

[LSM61] Elliot H. Lieb, T.D. Schultz, and Daniel C. Mattis, *Antiferromagnetic chain*, Ann. Phys. (N.Y.) **16** (1961), 407.

[LSM00] Marshall Luban, Jürgen Schnack, and Robert Modler, *Approximate quantum model of the Heisenberg molecular magnet* $\{Mo_{72}Fe_{30}\}$, Phys. Rev. Lett. (2000), submitted.

[Lub77] Marshall Luban, *Acceleration of convergence of the 1/d expansion for the critical temperature of the spherical model*, Prog. Theor. Phys. **58** (1977), 1177.

[Man91] Efstratios Manousakis, *The spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a square lattice and its application to the cuprous oxides*, Rev. Mod. Phys. **63** (1991), 1.

[Mar55] W. Marshall, *Antiferromagnetism*, Proc. Royal. Soc. A (London) **232** (1955), 48.

[MGP$^+$96] Luciano G. Moretto, Roberta Ghetti, Larry W. Phair, Kin Tso, and Gordon J. Wozniak, *Comment on "Probing the nuclear liquid-gas phase transition"*, Phys. Rev. Lett. **76** (1996), 2822.

[MLS$^+$] Achim Müller, Marshall Luban, Christian Schröder, Robert Modler, Paul Kögerler, Maria Axenovich, Jürgen Schnack, Paul C. Canfield, Sergei Bud'ko, and Neil Harrison, *Giant magnetic molecules: New vistas in mesoscopic magnetism*, in preparation.

[Mor56] T. Moriya, *Nuclear magnetic relaxation in antiferromagnets*, Prog. Theor. Phys. **16** (1956), 23.

[MPPG98] Achim Müller, F. Peters, M.T. Pope, and Dante Gatteschi, *Polyoxometalates: very large clusters - nanoscale magnets*, Chem. Rev. **98** (1998), 239.

[MS95] H. Müller and B.D. Serot, *Phase transitions in warm, asymmetric nuclear matter*, Phys. Rev. C **52** (1995), 2072.

[MSL99] Detlef Mentrup, Jürgen Schnack, and Marshall Luban, *Spin dynamics of quantum and classical Heisenberg dimers*, Physica A **272** (1999), 153.

[MSS$^+$99] Achim Müller, S. Sarkar, Syed Qaiser Nazir Shah, Hartmut Bögge, Marc Schmidtmann, Sh. Sarkar, Paul Kögerler, B. Hauptfleisch, A.X. Trautwein, and V. Schünemann, *Archimedean synthesis and magic numbers: "Sizing" giant molybdenum-oxide-based molecular spheres of the Keplerate type*, Angew. Chem. Int. Ed. Engl. **38** (1999), 3238.

[MSSL00] Detlef Mentrup, Heinz-Jürgen Schmidt, Jürgen Schnack, and Marshall Luban, *Transition from quantum to classical Heisenberg trimers: Thermodynamics and time correlation functions*, Physica A **278** (2000), 214.

[NHW$^+$95] Joseph B. Natowitz, Kris Hagel, Roy Wada, Z. Majka, P. Gonthier, J. Li, N. Mdeiwayeh, B. Xiao, and Y. Zhao, *Limiting temperatures of neutron rich nuclei: A possible interpretation of data from isotope yield ratios*, Phys. Rev. C **52** (1995), R2322.

[Nos84] Shuichi Nosé, *A unified formulation of the constant temperature molecular-dynamics methods*, J. Chem. Phys. **81** (1984), 511.

[Nos91] Shuichi Nosé, *Constant Temperature Molecular Dynamics Methods*, Prog. of Theor. Phys. Suppl. **103** (1991), 1.

[NRS98] N.B.Ivanov, Johannes Richter, and Ulrich Schollwöck, *Frustrated quantum Heisenberg ferrimagnetic chains*, Phys. Rev. B **58** (1998), 14456.

[OR93] Akira Ohnishi and Jørgen Randrup, *Statistical properties of antisymmetrized molecular dynamics*, Nucl. Phys. A **565** (1993), 474.

[PDK+97] Bernd Pilawa, R. Desquiotz, M.T. Kelemen, M. Weickenmeier, and A. Geisselman, *Magnetic properties of new* Fe_6 *(triethanolaminate(3-))*$_6$ *spin clusters*, J. Magn. Magn. Mater. **177** (1997), 748.

[Pit61a] Lev P. Pitaevskii, Zh. Eksp. Teor. Fiz. **40** (1961), 646.

[Pit61b] Lev P. Pitaevskii, Sov. Phys. JETP **13** (1961), 451.

[PMR+95] Josef Pochodzalla, Theo Möhlenkamp, Thorsten Rubehn, Andreas Schüttauf, Andreas Wörner, Erik Zude, Marieluise Begemann-Blaich, Th. Blaich, Claudius Gross, Hans Emling, A. Ferrero, G. Immé, Ileana Iori, Gerd J. Kunde, Wolf-Dietrich Kunze, Volker Lindenstruth, Uli Lynen, A. Moroni, Walter F.J. Müller, Burkhard Ocker, Giovanni Raciti, Hans Sann, Carsten Schwarz, Wolfgang Seidel, Volker Serfling, Joachim Stroth, Andrzej Trzcinski, Wolfgang Trautmann, Andrzej Tucholski, Giuseppe Verde, and Bogdan Zwieglinski, *Probing the nuclear liquid-gas phase transition*, Phys. Rev. Lett. **75** (1995), 1040.

[PMR+96] Josef Pochodzalla, Theo Möhlenkamp, Thorsten Rubehn, Andreas Schüttauf, Andreas Wörner, Erik Zude, Marieluise Begemann-Blaich, Th. Blaich, Hans Emling, A. Ferrero, Claudius Gross, G. Immé, Ileana Iori, Gerd J. Kunde, Wolf-Dietrich Kunze, Volker Lindenstruth, Uli Lynen, A. Moroni, Walter F.J. Müller, Burkhard Ocker, Giovanni Raciti, Hans Sann, Carsten Schwarz, Wolfgang Seidel, Volker Serfling, Joachim Stroth, Wolfgang Trautmann, Andrzej Trzcinski, Andrzej Tucholski, Giuseppe Verde, and Bogdan Zwieglinski, *Pochodzalla et al. reply*, Phys. Rev. Lett. **76** (1996), 2823.

[PN95] Gabor Papp and Wolfgang Nörenberg, *The path of hot nuclei towards multifragmemtation*, APH Heavy Ion Physics **1** (1995), 241.

[Poc97] Josef Pochodzalla, *The search for the liquid-gas phase transition in nuclei*, Prog. Part. Nucl. Phys. **39** (1997), 443.

[PP77] H.R. Pajkowski and R.K. Pathria, *Criteria for onset of Bose-Einstein condensation in ideal systems confined to restricted geometries*, J. Phys. A **10** (1977), 561.

[RIVR95] Johannes Richter, N.B. Ivanov, Andreas Voigt, and K. Retzlaff, *Ground-state phase relationships and level ordering for frustrated quantum antiferromagnets*, J. Low Temp. Phys. **99** (1995), 363.

[SCG^{+}89] Michael Schönhofen, Manuel Cubero, M. Gering, M. Sambataro, Hans Feldmeier, and Wolfgang Nörenberg, *The nuclear equation of state in effective relativistic field-theories and pion yields in heavy-ion collisions*, Nucl. Phys. A **504** (1989), 875.

[Sch98] Jürgen Schnack, *Molecular dynamics investigations on a quantum system in a thermostat*, Physica A **259** (1998), 49.

[Sch99a] Jürgen Schnack, *Thermodynamics of the harmonic oscillator using coherent states*, Europhys. Lett. **45** (1999), 647.

[Sch99b] Christian Schröder, *Numerische Simulationen zur Thermodynamik magnetischer Strukturen mittels deterministischer und stochastischer Wärmebadankopplung*, Ph.D. thesis, University of Osnabrück, 1999, download from http://elib.uni-osnabrueck.de/dissertations/physics/Chr.Schroeder/.

[Sch00] Jürgen Schnack, *Properties of the first excited state of nonbipartite Heisenberg spin rings*, Phys. Rev. B **62** (2000), 14855.

[SF96] Jürgen Schnack and Hans Feldmeier, *Statistical Properties of Fermionic Molecular Dynamics*, Nucl. Phys. A **601** (1996), 181.

[SF97] Jürgen Schnack and Hans Feldmeier, *The nuclear liquid–gas phase transition within Fermionic Molecular Dynamics*, Phys. Lett. B **409** (1997), 6.

[SGC93] Roberta Sessoli, Dante Gatteschi, and Andrea Caneschi, *Magnetic bistability in a metal-ion cluster*, Nature (London) **365** (1993), 141.

[SGJ96] Ulrich Schollwöck, O. Golinelli, and Thierry Jolicœur, *$S=2$ antiferromagnetic quantum spin chain*, Phys. Rev. B **54** (1996), 4038.

[SL01] Jürgen Schnack and Marshall Luban, *Rotational modes in molecular magnets with antiferromagnetic Heisenberg exchange*, Phys. Rev. B **63** (2001), 014418.

[SMF01] Jürgen Schnack, Detlef Mentrup, and Hans Feldmeier, *Critical revision of the Grilli-Tosatti quantum thermostat method*, Physica A (2001), in preparation.

[SS98] Heinz-Jürgen Schmidt and Jürgen Schnack, *Investigations on finite ideal quantum gases*, Physica A **260** (1998), 479.

[SS99] Heinz-Jürgen Schmidt and Jürgen Schnack, *Thermodynamic fermion-boson symmetry in harmonic oscillator potentials*, Physica A **265** (1999), 584.

[SW86] B.D. Serot and J.D. Walecka, *The relativistic nuclear many-body problem*, Adv. Nucl. Phys. **16** (1986), 1.

[TBLD00] J. Tempere, Fons Brosens, L.F. Lemmens, and Jozef T. Devreese, *Condensation and interaction range in harmonic boson traps: a variational approach*, Phys. Rev. A **61** (2000), 043605.

[TDP+94] Kingsley L. Taft, Christopher D. Delfs, Georgia C. Papaefthymiou, Simon Foner, Dante Gatteschi, and Stephen J. Lippard, *$[Fe(OMe)_2(O_2CCH_2Cl)]_{10}$, a molecular ferric wheel*, J. Am. Chem. Soc. **116** (1994), 823.

[TR99] P. Tomczak and Johannes Richter, *Ground state of the spin-1/2 Heisenberg antiferromagnet on a two-dimensional square-hexagonal-dodecagonal lattice*, Phys. Rev. B **59** (1999), 107.

[Wal00] Oliver Waldmann, *Symmetry and energy spectrum of high-nuclearity spin clusters*, Phys. Rev. B **61** (2000), 6138.

[Whi93] Steven R. White, *Density-matrix algorithms for quantum renormalization groups*, Phys. Rev. B **48** (1993), 10345.

[WKS+00] Ch. Waldtmann, H. Kreutzmann, Ulrich Schollwöck, K. Maisinger, and H.U. Everts, *Ground state and excitations of a one-dimensional Kagome-like antiferromagnet*, Phys. Rev. B **62** (2000), 9472.

[WSK+99] Oliver Waldmann, J. Schülein, R. Koch, Paul Müller, Ingo Bernt, Rolf W. Saalfrank, Hans Peter Andres, Hans U. Güdel, and P. Allenspach, *Magnetic anisotropy of two cyclic hexanuclear Fe(III) clusters entrapping alkaline ions*, Inorg. Chem. **38** (1999), 5879.

[XLTF96] H. Xi, W.G. Lynch, M.B. Tsang, and William A. Friedman, *Secondary decays and the helium lithium isotope thermometer*, Phys. Rev. C **54** (1996), R2163.

Publication list of Jürgen Schnack

The following list summarizes all publications of Jürgen Schnack published after his dissertation. Contributions to conference proceedings have been omitted. The order is chronological.

1. Hans Feldmeier and Jürgen Schnack, *Fermionic Molecular Dynamics*, Prog. Part. Nucl. Phys. **39** (1997), 393-442.
2. Jürgen Schnack and Hans Feldmeier, *The nuclear liquid–gas phase transition within Fermionic Molecular Dynamics*, Phys. Lett. B **409** (1997), 6-10.
3. Jürgen Schnack, *Molecular dynamics investigations on a quantum system in a thermostat*, Physica A **259** (1998), 49-58.
4. Hans Feldmeier, Thomas Neff, Robert Roth, and Jürgen Schnack, *A Unitary Correlation Operator Method*, Nucl. Phys. A **632** (1998), 61-95.
5. Heinz-Jürgen Schmidt and Jürgen Schnack, *Investigations on finite ideal quantum gases*, Physica A **260** (1998), 479-489.
6. Heinz-Jürgen Schmidt and Jürgen Schnack, *Thermodynamic fermion-boson symmetry in harmonic oscillator potentials*, Physica A **265** (1999), 584-589.
7. Jürgen Schnack, *Thermodynamics of the harmonic oscillator using coherent states*, Europhys. Lett. **45** (1999), 647-652.
8. Klaus Bärwinkel, Jürgen Schnack, and Uwe Thelker, *Quasi-Particle Picture for Monatomic Gases*, Physica A **262** (1999), 496-504.
9. Detlef Mentrup, Jürgen Schnack, and Marshall Luban, *Spin dynamics of quantum and classical Heisenberg dimers*, Physica A **272** (1999), 153-161.
10. Detlef Mentrup, Heinz-Jürgen Schmidt, Jürgen Schnack, and Marshall Luban, *Transition from quantum to classical Heisenberg trimers: Thermodynamics and time correlation functions*, Physica A **278** (2000), 214-221.

11. Y. Furukawa, Marshall Luban, F. Borsa, D.C. Johnston, A.V. Mahajan, L.L. Miller, Detlef Mentrup, Jürgen Schnack, and A. Bino, *Spin dynamics of the magnetic cluster* $[Cr_4S(O_2CCH_3)_8(H_2O)_4](NO_3)_2H_2O$, Phys. Rev. B **61** (2000), 8635-8638.
12. Klaus Bärwinkel, Heinz-Jürgen Schmidt, and Jürgen Schnack, *Structure and relevant dimension of the Heisenberg model and applications to spin rings*, J. Magn. Magn. Mater. **212** (2000), 240-250.
13. Klaus Bärwinkel, Heinz-Jürgen Schmidt, and Jürgen Schnack, *Ground state properties of antiferromagnetic Heisenberg spin rings*, J. Magn. Magn. Mater. **220** (2000), 227-234.
14. Hans Feldmeier and Jürgen Schnack, *Molecular Dynamics for Fermions*, Rev. Mod. Phys. **72** (2000), 655-688.
15. Jürgen Schnack, *Properties of the first excited state of nonbipartite Heisenberg spin rings*, Phys. Rev. B **62** (2000), 14855-14859
16. Jürgen Schnack and Marshall Luban, *Rotational modes in molecular magnets with antiferromagnetic Heisenberg exchange*, Phys. Rev. B **63** (2001), 014418 (7 pages).

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A. Small magnetic molecules – publications

1. Detlef Mentrup, Jürgen Schnack, and Marshall Luban, *Spin dynamics of quantum and classical Heisenberg dimers*, Physica A **272** (1999), 153-161.
2. Detlef Mentrup, Heinz-Jürgen Schmidt, Jürgen Schnack, and Marshall Luban, *Transition from quantum to classical Heisenberg trimers: Thermodynamics and time correlation functions*, Physica A **278** (2000), 214-221.
3. Klaus Bärwinkel, Heinz-Jürgen Schmidt, and Jürgen Schnack, *Structure and relevant dimension of the Heisenberg model and applications to spin rings*, J. Mag. Mag. Mater. **212** (2000), 240-250.
4. Klaus Bärwinkel, Heinz-Jürgen Schmidt, and Jürgen Schnack, *Ground state properties of antiferromagnetic Heisenberg spin rings*, J. Mag. Mag. Mater. **220** (2000), 227-234.
5. Y. Furukawa, Marshall Luban, F. Borsa, D.C. Johnston, A.V. Mahajan, L.L. Miller, Detlef Mentrup, Jürgen Schnack, and A. Bino, *Spin dynamics of the magnetic cluster* $[Cr_4S(O_2CCH_3)_8(H_2O)_4](NO_3)_2H_2O$, Phys. Rev. B **61** (2000), 8635-8638.
6. Jürgen Schnack, *Unexpected properties of the first excited state of non-bipartite Heisenberg spin rings*, Phys. Rev. B **62** (2000), 14855-14859.
7. Jürgen Schnack and Marshall Luban, *Rotational modes in molecular magnets with antiferromagnetic Heisenberg exchange*, Phys. Rev. B **63** (2001), 014418.

B. Small fermion and boson systems – publications

1. Heinz-Jürgen Schmidt and Jürgen Schnack, *Investigations on finite ideal quantum gases*, Physica A **260** (1998), 479-489.
2. Heinz-Jürgen Schmidt and Jürgen Schnack, *Thermodynamic fermion-boson symmetry in harmonic oscillator potentials*, Physica A **265** (1999), 584-589.

C. Thermostated quantum dynamics – publications

1. Hans Feldmeier and Jürgen Schnack, *Fermionic Molecular Dynamics*, Prog. Part. Nucl. Phys. **39** (1997), 393-442.
2. Jürgen Schnack and Hans Feldmeier, *The nuclear liquid–gas phase transition within Fermionic Molecular Dynamics*, Phys. Lett. B **409** (1997), 6-10.
3. Hans Feldmeier, Thomas Neff, Robert Roth, and Jürgen Schnack, *A Unitary Correlation Operator Method*, Nucl. Phys. A **632** (1998), 61-95.
4. Jürgen Schnack, *Molecular dynamics investigations on a quantum system in a thermostat*, Physica A **259** (1998), 49-58.
5. Jürgen Schnack, *Thermodynamics of the harmonic oscillator using coherent states*, Europhys. Lett. **45** (1999), 647-652.
6. Hans Feldmeier and Jürgen Schnack, *Molecular Dynamics for Fermions*, Rev. Mod. Phys. **72** (2000), 655-688.