Phase diagrams of Heisenberg chains with different cell spins: role of the tree-site exchange interactions

Nedko Ivanov\textsuperscript{1,2} and Jürgen Schnack\textsuperscript{1}
\textsuperscript{1}Fakultät für Physik, Universität Bielefeld, Postfach 100131, D-33501 Bielefeld, Germany
\textsuperscript{2}ISSP, Bulgarian Academy of Sciences, Tzarigradsko chaussee 72, 1784 Sofia, Bulgaria
E-mail: ivanov@physik.uni-bielefeld.de

Abstract. Additional three-site exchange interactions, naturally arising, e.g., in the strong-coupling expansion of the two-band Hubbard model and in various effective spin Hamiltonians, may stabilize various exotic spin phases which are not typical for the pure Heisenberg models on frustrated lattices and/or with extra biquadratic exchange terms. The reason is hidden in some specific features of the isotropic three-site exchange interactions, such as the promotion of collinear spin configurations as well as the reinforcing tendency towards clustering of the quantum spins on the shortest exchange bonds. As a result, systems with integer and half-integer composite spins in the unit cell might be expected to support different quantum phase diagrams. In this work, we study this phenomenon by comparing the quantum phase diagrams of two Heisenberg chains with extra three-site exchange terms and two different alternating pairs of site spins $S$ and $\sigma$ [$S, \sigma = (1, \frac{1}{2})$ and $(\frac{3}{2}, \frac{1}{2})$], resulting in half-integer and integer cell spins $S + \sigma$, respectively.

1. Introduction
Quantum spin systems with competing exchange interactions present an excellent background for studying novel quantum states of matter such as the quantum spin ice, various spin nematic states, as well as different types of quantum spin liquids. Most of these systems are currently considered as real candidates for future applications in quantum computing and information technologies. The Heisenberg spin systems with three-site exchange couplings discussed below, which constitute a special class of magnetically frustrated systems, have been remaining scarcely explored [1, 2, 3, 4, 5, 6, 7], as opposed to those with extra biquadratic terms [8]. The large number of newly synthesized Heisenberg spin chains with alternating $S$ and $\sigma$ site spins $S > \sigma$ in the unit cell (for a review, see, e.g., Ref. [9]) offer very suitable platform where the effects of the frustrating three-site exchange interaction may be explored even in the extreme quantum case with site spins $\sigma = \frac{1}{2}$. Clearly, this is not the case with the standard uniform-spin models, since the square of the spin-$\frac{1}{2}$ matrix is a constant. Based on extended numerical density-matrix renormalization group (DMRG) and exact diagonalization (ED) calculations [5, 7], below we compare the quantum phase diagrams of this model for half-integer and integer cell spins. The Heisenberg chain with extra three-site exchange terms (see figure 1) is defined by the following Hamiltonian

$$\mathcal{H} = \sum_{n=1}^{L} J_1 S_{2n} \cdot (\sigma_{2n-1} + \sigma_{2n+1}) + J_3 \left[ (S_{2n} \cdot \sigma_{2n-1}) (S_{2n} \cdot \sigma_{2n+1}) + h.c. \right],$$  \hspace{1cm} (1)
First, let us briefly comment on the classical phase diagram of equation (1), which can be constructed by using the ground-state (GS) cluster configurations shown in figure 1. By fitting the directions of the sharing $\sigma$ spins of the FM and FiM three-spin clusters, we can obtain the macroscopic non-degenerate (up to a global rotation of spins) FM and FiM configurations corresponding to local minima of the classical energy (FM and FiM phases in figure 2). Using the two cluster configurations $D$, the same procedure now leads to a $2^L$-fold degenerate classical phase, denoted by $D$ in figure 2. At a classical level, the phase diagrams of both models are qualitatively indistinguishable. The established classical diagram has been independently confirmed by classical Monte Carlo simulations.

where $L$ stands for the number of elementary cells, each containing two different site spins ($S > \sigma$). In what follows we use the standard parameterization of the coupling constants $J_1 = J \cos t$ and $J_3 = J \sin t$ ($0 \leq t < 2\pi$), where $J = 1$ is used as an energy unit. Note that for extreme quantum systems with $\sigma = \frac{1}{2}$ and arbitrary $S$, the well-known biquadratic spin interactions $(S_i \cdot \sigma_j)^2$ here reduce to bilinear Heisenberg terms, so that the extra three-site term in equation (1) actually is a general form for the isotropic nearest-neighbor exchange interactions which include up to tree lattice sites.

2. Classical phase diagram

Figure 1. (left) The considered Heisenberg chains and (right) the optimal cluster states used as building blocks of the classical spin phases presented in figure 2.

Figure 2. Classical (inner circles) and quantum (outer circles) phase diagrams of the models with half-integer (left) and integer (right) cell spins $S + \sigma$. FM and FiM stand for, respectively, the ferromagnetic and ferrimagnetic phases. $P$ is a partially polarized magnetic state with a spatially modulated magnetization. $D$ denotes a $2^L$-fold degenerate classical phase composed of two types of collinear cluster spin configurations shown in figure 1. This phase contains both non-magnetic and magnetic ground-state configurations. $SL_1$ and $SL_2$ denote, respectively, gapless and gapped spin-liquid (SL) states characteristic, respectively, for the models with half-integer and integer cell spins. $N$ stands for a quasi-nematic state with short-range nematic order and spin-2 lowest-lying excitations. Here, $t_1 \approx 30^\circ$, $t_2 = 25.5^\circ$, and $t_3 = 132^\circ$. $t_F = \pi - \arctan \frac{1}{\sigma(2S+1)}$ is the exact FM phase boundary for arbitrary site spins $S$ and $\sigma$. 
3. Comparison of the quantum phase diagrams

3.1. Modulated non-Lieb-Mattis type magnetic states

The established partially-polarized magnetic states (see, e.g., the P sectors of the diagrams in figure 2) in both models do not exist in the classical phase diagram, so that their appearance is a pure quantum effect. Due to the frustration effect of the tree-site interaction, the Lieb-Mattis theorem [10] is not applicable, so that the so-called quantization of the unit-cell magnetic moment is destroyed, i.e., the magnetic moment per cell can take arbitrary values. This means, in particular, that in such systems the magnetic moment may change continuously to zero near the transition from magnetic to non-magnetic states (t₁ and t₂ points in figure 2), or to be spatially modulated, like in the P sectors of both models, which are close to the ferromagnetic phase boundary t_F. In the latter case, the local magnetization clearly shows an incommensurate modulation with long-distance periodicity depending on the distance from the boundary. As a matter of fact, these modulated states follow the rule q(S + σ − m₀) = integer, where q is the period of the modulated structure, and m₀ is the magnetic moment per unit cell [5, 7]. For t ≲ t_F, the modulated structure is characterized by (q, m₀) = (8, 9/8) and (3, 5/3) for the (1, 1/2) and (3/2, 1/2) models, respectively. Skipping further discussions on this interesting modulated ferrimagnetic states, we only mention that similar partially-polarized non-Lieb-Mattis-type phases have been earlier identified [11, 12] and studied [13] in other magnetically frustrated one-dimensional systems.

3.2. Doubly degenerated non-magnetic states

The DMRG analysis of the short-range correlations in open chains reveals regions of the parameter space in both models where in the GS a regular alternating-bond structure characterized by different values of the correlators 〈σ_{2n−1} · S_{2n}〉 = u and 〈S_{2n} · σ_{2n+1}〉 = v (u < v) appears. The uv (vu) “dimerized” GS |Ψ_L⟩ (|Ψ_R⟩) is stabilized in open chains with a σ spin on the left (right) end of the chain and corresponds to a uv (vu) dimerization of the local Hamiltonians h_n (see figure 3). The established uv structure of the GS is strongly pronounced in the middle of the SL regions in both models, see figure 2, where the values of u and v indicate the formation of nearly pure spin-1/2 and spin-2 (or spin-3) states of the nearest-neighbor spins in the systems with half-integer and integer cell spins S + σ, respectively. Very recently, it was also
Figure 4. (left) Finite-size scaling of the lowest triplet excitation in the gapless $SL_1$ phase of the $(S,\sigma) = (1,\frac{1}{2})$ chain (DMRG results for $t = 45^\circ$). (right) Extrapolated triplet gap vs $t$ in the gapped $SL_2$ phase of the $(S,\sigma) = (\frac{3}{2},\frac{1}{2})$ chain (DMRG results). Irrespective of the gap structure of the low-energy spectrum, the non-magnetic states in both models are doubly degenerate as a result of the dimerization.

3.3. Nematic-like phase
The three-site spin interaction term in equation (1) for every unit cell can be represented in the following symmetric form

$$V_n^{(3)} = \frac{1}{2} \left( S_{2n}\sigma_{2n}^\alpha + S_{2n}\sigma_{2n+1}^\beta + \sigma_{2n-1}\sigma_{2n+1}^\alpha + \sigma_{2n-1}\sigma_{2n+1}^\beta \right), \quad (2)$$

where $\alpha, \beta = x, y, z$. The two symmetric terms in the parentheses may be considered as tensor order parameters (up to some normalization factors) for on-site and bond nematic phases constructed from $S$ and $\sigma$ types spins, respectively. This representation of $V_n^{(3)}$ means that in systems with dominant isotropic three-site spin interactions nematic orders of different types could be stabilized. As a matter of fact, the numerical ED calculations concerning the low-lying excitations in periodic $L = 8$ chains, described by the Hamiltonian (1), point towards the formation of such a short-ranged nematic order in the subsystem of $S$ spins [6], the $N$ sector in figure 2. Information about the non-magnetic $N$ state in the $(1,\frac{1}{2})$ model can be extracted...
form the lowest-laying states in different total-spin ($S_t$) sectors. The established tower of well-separated lowest multiplets containing only even $S_t$ sectors is a special feature of the spectrum of this quasi-quadrupolar spin state. This spectrum structure is known as a fingerprint of the spin quadrupolar (i.e., nematic) order, unlike the Anderson tower of states [14] containing all $S_t$ sectors. The latter is a characteristic of the Néel order. In fact, Anderson towers of states have been observed even in some finite isotropic spin-S chains and magnetic molecules [15, 16, 17]. Interestingly, such regions with enhanced quadrupolar fluctuations do not appear on the phase diagram of the ($\frac{3}{2}, \frac{3}{2}$) model.

4. Summary
In the present work, based on extensive DMRG and ED numerical calculations, we compared the quantum phase diagrams of two Heisenberg chains with extra three-site spin exchange interactions and different (half-integer and integer) cell spins $S + \sigma$. We demonstrated that in the regions with dominating three-site spin interactions the models with half-integer and integer cell spins exhibit different non-magnetic phases. Nevertheless, the stabilization of these phases is closely related to some peculiarities of the three-site spin interaction, such as the support of collinear spin states, the pronounced tendency towards dimerizations of the nearest-neighbor exchange bonds, and the enhancement of quadrupolar spin fluctuations. We believe that the presented results will encourage future studies on real systems with dominating isotropic three-site exchange interactions.

Acknowledgments
We are grateful to Jörg Ummethum for adapting his DMRG code systems with three-site spin interactions. Support by the Bulgarian Science Foundation, Grant DN0818/14.12.2016, is gratefully acknowledged.

References
[1] Michaud F, Vernay F , Manmana S R and Mila F 2012 Phys. Rev. Lett. 108 127202
[2] Michaud F, Manmana S R and Mila F 2013 Phys. Rev. B 87 140404(R)
[3] Michaud F and Mila F 2013 Phys. Rev. B 88 094435
[4] Wang Z -Y, Furuya S C, Nakamura M and Komakura R 2013 Phys. Rev. B 88 224419
[5] Ivanov N B, Ummethum J. and Schnack J 2014 Eur. Phys. J. B 87 226
[6] Ivanov N B and Schnack J 2014 J. Phys.: Conf. Series 558 012015
[7] Ivanov N B, Petrova S I and Schnack J 2016 Eur. Phys. J. B 89 121
[8] Lhuillier C and Misguich G, Introduction to frustrated magnetism Materials, Experiments, Theory 2011 (Springer Series in Solid State Sciences vol 164) ed C Lacroix, P Mendels and F Mila (Berlin Heidelberg: Springer Verlag) chapter 2 pp 23-41
[9] Ivanov N B and Schollwöck U, Quantum Heisenberg ferrimagnetic chains Some Contemporary Problems of Condensed Matter Physics 2001 ed Vlaev S J and Gaggero-Sager L M (Nova Science Publishers Inc., Huntington, New York) chapter 5 pp 145-176
[10] Lieb E H and Mattis D C 1962 J. Math. Phys. 3 749
[11] Ivanov N B, Richter J 2004 Phys. Rev. B 69 214420
[12] Shimokawa T, Nakano H, 2013 J. Kor. Phys. Soc. (SI) 63 391
[13] Furuya Sh S, Giamarchi Th 2014 Phys. Rev. B 89 205131
[14] Anderson P W 1952 Phys. Rev. B 86 694
[15] Schnack J and Luban M 2000 Phys. Rev. B 63 014418
[16] Waldmann O 2001 Phys. Rev. B 65 024424
[17] Machens A, Konstantinidis N P, Waldmann O, Schneider I and Eggert S 2013 Phys. Rev. B 87 144409