Regular Article

The spin-1/2 square-lattice $J_1$-$J_2$ model: the spin-gap issue

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Received 31 August 2014 / Received in final form 7 October 2014
Published online 2 January 2015 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2015

Abstract. We use the coupled cluster method to high orders of approximation in order to calculate the ground-state energy, the ground-state magnetic order parameter, and the spin gap of the spin-1/2 $J_1$-$J_2$ model on the square lattice. We obtain values for the transition points to the magnetically disordered quantum paramagnetic phase of $J_2^3 = 0.454J_1$ and $J_2^2 = 0.588J_1$. The spin gap is zero in the entire parameter region accessible by our approach, i.e. for $J_2 \leq 0.49J_1$ and $J_2 > 0.58J_1$. This finding is in favor of a gapless spin-liquid ground-state in this parameter regime.

1 Introduction

The spin-1/2 quantum Heisenberg antiferromagnet with nearest-neighbor (NN), $J_1 > 0$, and next-nearest-neighbor (NNN) bonds, $J_2 \geq 0$, described by the Hamiltonian

$$H = J_1 \sum_{\langle ij \rangle} s_i \cdot s_j + J_2 \sum_{\langle\langle ij \rangle\rangle} s_i \cdot s_j$$

is one of the most challenging quantum spin models. The model was introduced more than 25 years ago in order to describe the breakdown of Néel antiferromagnetic (NAF) long-range order (LRO) in cuprate superconductors [1–3].

This model has since attracted much attention as a canonical model for studying frustration driven quantum phase transitions between semiclassical ground-state phases with magnetic LRO and magnetically disordered quantum phases (see, e.g., Refs. [1–41]). These studies demonstrate clearly that there is a magnetically disordered spin-rotation-invariant quantum phase in the region of strong frustration $0.44 \leq J_2/J_1 \leq 0.6$. Although the semi-classical ground-state phases of the model, namely the NAF LRO at $J_2/J_1 \leq 0.44$ and the collinear antiferromagnetic (CAF) LRO at $J_2/J_1 \geq 0.6$ are well-understood, an active controversial debate has started very recently regarding the nature of the intermediate quantum phase in this model and its quantum phase transitions [15,21,28–41]. Most of the papers argue that the transition at about $J_2/J_1 \approx 0.44$ is continuous, but that the transition at about $J_2/J_1 \approx 0.6$ is of first order (see, e.g., Refs. [4,5,11,13,28,31,41]). A particular focus of these recent papers has been on the existence of an excitation gap in the intermediate quantum phase. Those papers in favor of an excitation gap are given by references [25,29,31,36,38]. We remark that a finite gap between a spin-rotation-invariant singlet ground state and a magnetic triplet excitation (spin gap) would be in accordance with earlier results in favor of a valence-bond ground state breaking translational symmetry (see e.g., Refs. [3,6,8,11,15,17,21]). In contrast to these findings, there are several recent investigations reporting indications of a gapless spin liquid state [30,32,33,41]. Very recent calculations using density matrix renormalization group with explicit implementation of $SU(2)$ spin rotation symmetry in reference [35] have found a gapless spin liquid for $0.44 < J_2/J_1 < 0.5$ and a gapped plaquette valence bond phase for $0.5 < J_2/J_1 < 0.61$.

In addition to the basic theoretical interest in this frustrated quantum many-body model, we mention that interest in this model is motivated also by its relation to experimental studies of various magnetic materials, such as VOMoO$_4$ (Ref. [42]), Li$_2$VOSiO$_4$, and Li$_2$VOGeO$_4$ (Refs. [43–45]). However, none of these materials has as yet exhibited exchange parameters $J_1$ and $J_2$ suitable for a magnetically disordered phase at very low temperatures.

In this paper we focus on the calculation of the gap to triplet excitations (spin gap) using a very general ab initio many-body technique, the coupled-cluster method (CCM), that was successfully applied in various fields of many-body physics [46–48]. The spin gap can be calculated directly within the framework of the CCM by using an appropriate excited-state formalism [49–52].

2 Brief illustration of the coupled-cluster method

We illustrate here only some relevant features of the CCM. For more general information on the methodology of the
CCM, see, e.g., references [46–48, 50–54]. The CCM has recently been applied widely to frustrated quantum spin systems (see, e.g., Refs. [10, 16, 20, 21, 51, 52, 55–65]). In particular, the CCM has been applied to calculate the ground-state properties of model (1) in two recent publications [21, 58], although the spin gap of the model has never before been calculated by using the CCM.

We mention firstly that the CCM automatically yields results in the limit \( N \to \infty \). Here we follow references [21, 58] to calculate the ground-state properties of the model using the CCM. The starting point for a CCM calculation is the choice of a normalized reference (or model) state \(|\Phi\rangle\). We then define a set of mutually commuting multispin creation operators \( C^+_I \) with respect to this state, where the index \( I \) runs over a complete set of many-body configurations. For the system under consideration here we choose as CCM reference states the two-sublattice Néel state for small \( J_2/J_1 \) and for large \( J_2/J_1 \) one of two possible collinear striped states. Although these CCM reference states are magnetically ordered states, various applications of the CCM to one- and two-dimensional quantum spin systems demonstrate that high-order implementations of the CCM are appropriate to describe magnetically disordered ground-state phases (see e.g. Refs. [16, 20, 21, 51, 56–65]).

We perform a rotation of the local axis of the spins such that all spins in the reference state align along the negative \( z \) axis. In the rotated coordinate frame the reference state reads \(|\Phi\rangle = |1\rangle|1\rangle|1\rangle \ldots\), and we can treat each site equivalently. The corresponding multispin creation operators are written as \( C^+_I = s^+_i s^+_j s^+_k \ldots \), where the indices \( i, j, k, \ldots \) denote arbitrary lattice sites. The CCM parameterizations of the ket- and bra-ground states are given by:

\[
H|\Psi\rangle = E|\Psi\rangle, \quad \langle \tilde{\Psi}|H = E\langle \tilde{\Psi}|;
\]

\[
|\Psi\rangle = e^S|\Phi\rangle, \quad S = \sum_{I \neq 0} S_I C^+_I;
\]

\[
\langle \tilde{\Psi}| = \langle \Phi|\tilde{S}e^{-S}, \quad \tilde{S} = 1 + \sum_{I \neq 0} \tilde{S}_I C^+_I.
\] (2)

By using the Schrödinger equation, \( H|\Psi\rangle = E|\Psi\rangle \), we can write the ground-state energy as \( E_{GS} = \langle \Phi|e^{-S}He^S|\Phi\rangle \). The magnetic order parameter (sublattice magnetization) is given by:

\[
m_s = -\frac{1}{N} \sum_{i=1}^{N} \langle \tilde{\Psi}|s^+_i|\Psi\rangle,
\] (3)

where \( s^+_i \) is expressed in the rotated coordinate system. The ket-state and bra-state correlation coefficients are obtained by solving the CCM ket- and bra-state equations given by

\[
\langle \Phi|C^+_I e^{-S}He^S|\Phi\rangle = 0, \quad \forall I \neq 0.
\] (4)

\[
\langle \Phi|\tilde{S}e^{-S}[H, C^+_I]e^S|\Phi\rangle = 0, \quad \forall I \neq 0.
\] (5)

Each ket- or bra-state equation belongs to a certain creation operator \( C^+_I = s^+_i s^+_j s^+_k \ldots \), i.e. it corresponds to a certain set (configuration) of lattice sites \( i, j, k, \ldots \). For the problem at hand only those correlation coefficients \( S_I \) and \( \tilde{S}_I \) related to clusters with even numbers of spin flips are different from zero. The ket- or bra-ground states belong to total spin \( S^z = 0 \). We use an enlarged unit cell of four sites for the CCM calculations in order to allow for the possibility of CCM solutions that break translational symmetry, e.g., for gapped valence-bond ground states.

We use the well established LSUBn approximation scheme in order to truncate the expansion of \( S \) and \( \tilde{S} \) (cf., e.g., Refs. [21, 50, 54, 58, 59, 61–65]). Within the LSUBn scheme all multispin correlations over all distinct locales on the lattice defined by \( n \) or fewer contiguous sites are taken into account in the correlation operators \( S_I \) and \( \tilde{S}_I \). Although the ground state is not in the focus of the present paper, we present here results including the LSUB12 approximation, which goes beyond the LSUB10 approximation presented in references [21, 58]. This increase in the level of approximation yields an improvement of the accuracy of the ground-state data.

In order to calculate the gap to triplet excitations we follow references [51, 52], where the spin gap was calculated by CCM for the two-dimensional \( J-J' \) model [66–69] and the one-dimensional \( J_1-J_2 \) model [70], respectively. Both models exhibit a quantum phase transition from a gapless phase to a gapped valence-bond phase. It was found in references [51, 52] that the opening of the spin gap at the transition point to the valence-bond phase is well described by the CCM.

To obtain the excited state \(|\Psi_e\rangle\) from the ground state \(|\Psi\rangle\) (2) we apply an excitation operator \( X^e \) linearly to \(|\Psi\rangle\), such that

\[
|\Psi_e\rangle = X^e e^{S}|\Psi\rangle; \quad X^e = \sum_{I \neq 0} X^e_I C^+_I.
\] (6)

Using the Schrödinger equation, \( H|\Psi_e\rangle = E_e|\Psi_e\rangle \), we find that

\[
\Delta_e X^e|\Phi\rangle = e^{-S}[H, X^e]e^S|\Phi\rangle,
\] (7)

where \( \Delta_e = E_e - E_{GS} \) is the spin gap. Applying \( \langle \Phi|C_I \) to equation (7) we find

\[
\Delta_e X^e_I = \langle \Phi|C_I e^{-S}[H, X^e]e^S|\Phi\rangle,
\] (8)

which we solve in order to get \( \Delta_e \). The choice of configurations in the excitation operator is restricted to contain only those which change the total spin \( S^z \) by one. Hence, the choice of clusters for the excited-state is different from those for the ground state. For the excited state we use the same approximation scheme, LSUBn, as for the ground state thus achieving comparable accuracy for both the ground and the excited states. We find that for high orders of approximation the number of configurations for the excited state is larger than that for the ground state, i.e. the calculation of the excited state is more difficult computationally.

The LSUBn approximation becomes exact for \( n \to \infty \), and so we can improve our results by extrapolating the “raw” LSUBn data to \( n \to \infty \). There are well-tested extrapolation rules [16, 20, 21, 51, 52, 56–59, 61–65] for the
ground-state energy per spin $e_{GS} = E_{GS}(n)/N$, the magnetic order parameter $m_s(n)$, and the spin gap $\Delta_e(n)$. We use $e_{GS}(n) = a_0 + a_1(1/n)^2 + a_2(1/n)^3$ for the ground-state energy, $m_s(n) = b_0 + b_1(1/n)^{1/2} + b_2(1/n)^{3/2}$ for the magnetic order parameter, and $\Delta_e(n) = c_0 + c_1(1/n) + c_2(1/n)^2$ for the spin gap. Moreover, we know from references [19–21,58] that the lowest level of approximation, LSUB2, conforms poorly to these rules. Hence, as in previous calculations [19–21,58], we exclude LSUB2 data from the extrapolations.

3 Results and discussion

In order to obtain results for the ground-state energy per spin, $e_{GS}$, the magnetic order parameter (sublattice magnetization), $m_s$, and the spin gap, $\Delta_e$, we solved numerically up to 1,374,389 equations (for the LSUB12 approximation of the excited state starting with the Néel reference state) using the code of Farnell and Schulenburg [71]. Unfortunately, for the collinear striped reference state we can present LSUB12 data for the ground state only. For the spin gap we are limited to LSUB10, since the number of equations is higher (namely 2,266,307 for LSUB12 approximation of the excited state) than that for the Néel reference state. We find stable solutions for the CCM equations at given level of LSUB$n$ approximation for the pure Heisenberg antiferromagnet ($J_2 = 0$) and then we track this stable solution until it terminates at $J_2/J_1 = 0.59$ (LSUB4), 0.54 (LSUB6), 0.52 (LSUB8), 0.50 (LSUB10), 0.49 (LSUB12), respectively. We notice that this allows us consider a parameter region that is noticeably beyond the presumably continuous transition between the NAF phase and the quantum phase. On the other side of the phase diagram the transition to the quantum phase is likely first order, and, therefore, below the CCM transition point at $J_2^c/J_1 = 0.588$ (see below) the validity of the CCM data starting from the collinear striped reference state becomes questionable.

Although our paper focuses primarily on the calculation of the spin gap, we present firstly results for the ground-state energy per spin (Fig. 1) and the sublattice magnetization (Fig. 2). The results for the spin gap are then shown (Fig. 3). In all figures we show LSUB$n$ data for $n = 4, 6, 8, 10, 12$ as well as data extrapolated to $n = \infty$, where extrapolations including new LSUB12 data are labeled by ‘ex 4-12’ and the extrapolations without including LSUB12 data are labeled by ‘ex 4-10’. Note that the graphs for $e_{GS}$ and $m_s$ are very similar to those of reference [21], although LSUB12 data are now included for $J_2/J_1 \leq 0.49$ and $J_2/J_1 \geq 0.56$. The comparison of both curves (‘ex 4-12’ and ‘ex 4-10’, with and without LSUB12, respectively) allows us to form an impression on the accuracy of the extrapolated CCM data.

Taking into account the new LSUB12 data we find for the phase transition points between the semiclassical phases and the quantum phase $J_2^c = 0.454J_1$ and $J_2^c = 0.588J_1$, whereas the previous CCM values (without LSUB12 data) were $J_2^c = 0.447J_1$ and $J_2^c = 0.586J_1$ (see Ref. [21]). These results are in good agreement with the density matrix renormalization group data of reference [35]. Let us mention that many of the earlier attempts to determine the transition points obtained values $J_2^c \approx 0.4$ (or even smaller values) (see e.g., Refs. [4,5,11–14,18]). In view of our results at hand and other recent results [35,41] the transition point is rather at higher values $J_2^c \sim (0.44 \ldots 0.45)J_1$.

The ground-state energy $e_{GS}$ (Fig. 1) shows a monotonic increase with increasing $J_2$ for the Néel reference state. This figure shows also that there is a monotonic increase of $e_{GS}$ with decreasing $J_2$ (until about $J_2 \sim 0.57 \ldots 0.58J_1$, i.e., slightly beyond the transition point $J_2^c$), starting from the regime of large $J_2$ and using the collinear striped reference state. For values of $J_2 \lesssim 0.58J_1$, we see that there is a drastic downturn in $e_{GS}$, which indicates that the CCM using the collinear striped reference state becomes inappropriate beyond $J_2^c$.

The results for the magnetic order parameter $m_s$ shown in Figure 2 may be used in order to determine the above reported transition points, $J_2^c$ and $J_2^c$, at which the order parameter vanishes. Figure 2 shows continuous behavior for the order parameter near $J_2^c$ and near $J_2^c$. However, it is clear from this figure that the decrease...
the collinear striped state is used as the CCM reference state, we are not able to present LSUB12 data, since the number of equations which have to be solved within the excited-state formalism exceeds the available computational resources, see also main text.

Results for the spin gap \( \Delta \) are presented in Figure 3. These data show a monotonic behavior of \( \Delta \), with increasing level \( n \) of the CCM-LSUBn approximation for all values of \( J_2 \), which indicates that the extrapolation to \( n \to \infty \) as described in Section 2 is reasonable. A consistency check of our spin-gap data is also provided by our results within the semiclassical ground state phases with magnetic LRO, i.e. for for \( J_2 \leq J_2^c \) and \( J_2 \geq J_2^c \), where the spin gap has to be zero. Indeed, our extrapolated results for \( \Delta \) in the limit \( n \to \infty \) are very close to zero within the Néel phase. In the CAF phase we obtain a small finite \( \Delta \) (\( n = \infty \)), which indicates that extrapolations in this regime are less accurate\(^1\).

The most relevant and important results for the current discussion relating the “spin-gap issue” concern our results for the spin gap in the region \( J_2 > J_2^c \). As already mentioned above we compare two extrapolations labeled by ‘ex 4-12’ (including the LSUB12 data in the extrapolation) and by ‘ex 4-10’ (without including the LSUB12 data in the extrapolation). We can consider the difference between both extrapolations as a measure of the accuracy of the extrapolated spin-gap data. As for the ground-state energy and the magnetic order parameter both extrapolations for the spin gap almost coincide which yields evidence that the extrapolation works very well and our extrapolated data are almost converged, and likely higher-order approximations would not modify our results noticeably. We remark again that the Néel reference state allows us to find results up to \( J_2 = 0.49 J_1 \) for approximation level LSUB12 (and even larger values of \( J_2 \) for lower approximation levels, see Fig. 3), which is clearly inside the magnetically disordered quantum regime. We find that there is no significant increase of the LSUBn values for the spin gap in the parameter region accessible by using the Néel reference state for \( J_2 > J_2^c \), i.e. for a considerable region inside the magnetically disordered quantum phase. Also the extrapolated spin gap \( \Delta \) (\( n \to \infty \)) available until \( J_2 = 0.49 J_1 \) remains practically zero. Thus, our CCM results for the spin gap are in favor of a gapless spin liquid\(^2\) in accordance with references [32,33,35,40,41]. However, our results do not rule out the possibility that a spin gap occurs within the parameter region \( 0.49 \leq J_2 / J_1 \leq 0.59 \), as indicated by recent density matrix renormalization group calculations [35].

Let us briefly discuss these findings in relation to previous CCM results concerning a possible valence-bond solid ground state [21]. In reference [21] generalized susceptibilities related to possible valence-bond states were calculated. Starting from the pure Heisenberg model at \( J_2 = 0 \) (i.e., in the Néel phase), both susceptibilities for the columnar dimerized and plaquette valence-bond states grow monotonically with increasing \( J_2 \) (cf. Figs. 8 and 9 in Ref. [21]). These susceptibilities become very large, but remain finite in the region around \( J_2^c \); this behavior was interpreted as an indication for the emergence of a valence-bond solid phase. In view of our new CCM results for the spin gap a more consistent interpretation is that of enhanced dimer-dimer or/and plaquette-plaquette correlations which may be even critical (i.e. with a power-law decay without valence-bond LRO\(^2\)) in a small but finite region \( 0.454 \leq J_2 / J_1 \leq 0.49 \) (cf. also Refs. [35,40]). Note, however, that the results for the generalized susceptibilities presented in reference [21] are in accordance with a plaquette ordered quantum phase proposed in reference [35] for \( J_2 / J_1 \geq 0.5 \).

4 Summary

We have used a general ab initio many-body technique called the coupled-cluster method (CCM) to high orders of approximation in order to calculate ground-state properties and the triplet excitation gap of the square-lattice system \( S = 1/2 \) J1-J2 model. Our results for the transition points, \( J_2^c = 0.454 J_1 \) and \( J_2^c = 0.588 J_1 \), between the semiclassical ground-state phases with magnetic LRO and the intermediate magnetically disordered quantum phase are in

Fig. 3. Spin gap \( \Delta \), plotted as a function of the frustration parameter \( J_2 / J_1 \). Note the beyond \( J_2 / J_1 = 0.58 \), where the collinear striped state is used as the CCM reference state, we are not able to present LSUB12 data, since the number of equations which have to be solved within the excited-state formalism exceeds the available computational resources, see also main text.

\(^1\) The reason for lower accuracy of the spin gap using the collinear striped reference state might by attributed to the facts that (i) we do not have LSUB12 data for the extrapolation and (ii) \( J_1 \) bonds within a certain LSUBn approximation belong to contiguous sites, but \( J_2 \) bonds do not (this difference becomes relevant for larger \( J_2 \), i.e. in the CAF phase).

\(^2\) Typically, the term “spin liquid” is used for a state without any long-range order (magnetic, or more exotic valence-bond etc.), i.e. there is no spontaneously broken symmetry: Usually all correlations exhibit a fast exponential decay. However, a spin liquid might be “critical” or “near-critical”, see e.g., the discussion in references [35,40], i.e., it may exhibit a power-law decay or very slow exponential decay of correlation functions.
very good agreement with recent density matrix renormalization group calculations [35]. The direct calculation of the spin gap within the intermediate quantum phase, which is possible until \(J_2/J_1 = 0.49\), does not give any hint for an opening of a spin gap in this phase. Therefore, our results are in favor of a gapless spin-liquid quantum ground state in the region \(J_2^c < J_2 < 0.49 J_1\). However, a gapped phase for \(J_2/J_1 \gtrsim 0.5\) cannot be ruled out, because our CCM approach becomes inappropriate for \(0.49 \lesssim J_2/J_1 \lesssim 0.58\).

For the numerical calculation we used the program package “The crystallographic CCM” (D.J.J. Farnell and J. Schulenburg) [71].

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