Quantum \( J_1-J_2 \) antiferromagnet on the stacked square lattice: Influence of the interlayer coupling on the ground-state magnetic ordering

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Using the coupled-cluster method (CCM) and the rotation-invariant Green’s function method (RGM), we study the influence of the interlayer coupling \( J_\perp \) on the magnetic ordering in the ground state of the spin-1/2 \( J_1-J_2 \) frustrated Heisenberg antiferromagnet \((J_1-J_2 \) model) on the stacked square lattice. In agreement with known results for the \( J_1-J_2 \) model on the strictly two-dimensional square lattice \( (J_\perp = 0) \) we find that the phases with magnetic long-range order at small \( J_2 < J_{c1} \) and large \( J_2 > J_{c2} \) are separated by a magnetically disordered (quantum paramagnetic) ground-state phase. Increasing the interlayer coupling \( J_\perp > 0 \) the parameter region of this phase decreases, and, finally, the quantum paramagnetic phase disappears for quite small \( J_\perp \approx 0.2 - 0.3 J_1 \).

The properties of the frustrated spin-1/2 Heisenberg antiferromagnet \((HAFM)\) with nearest-neighbor \( J_1 \) and competing next-nearest-neighbor \( J_2 \) coupling \((J_1-J_2 \) model) on the square lattice have attracted a great deal of interest during the last fifteen years (see, e.g., Refs. 1–12 and references therein). The recent synthesis of layered magnetic materials\(^{13,14}\) which can be described by the \( J_1-J_2 \) model has stimulated a renewed interest in this model. It is well-accepted that the model exhibits two magnetically long-range ordered phases at small and at large \( J_2 \) separated by an intermediate quantum paramagnetic phase without magnetic long-range order \((LRO)\) in the parameter region \( J_{c1} < J_2 < J_{c2} \), where \( J_{c1} \approx 0.4 \) and \( J_{c2} \approx 0.6 \). The ground state \((GS)\) at low \( J_2 < J_{c1} \) exhibits semi-classical Néel magnetic LRO with the magnetic wave vector \( Q_0 = (\pi, \pi) \). The GS at large \( J_2 > J_{c2} \) shows so-called collinear magnetic LRO with the magnetic wave vectors \( Q_1 = (\pi, 0) \) or \( Q_2 = (0, \pi) \). These two collinear states are characterized by a parallel spin orientation of nearest neighbors in vertical \((\text{horizontal})\) direction and an antiparallel spin orientation of nearest neighbors in horizontal \((\text{vertical})\) direction. The properties of the intermediate quantum paramagnetic phase are still under discussion, however, a valence-bond crystal phase seems to be most favorable.\(^{2,4,8,9}\)

The properties of quantum magnets strongly depend on the dimensionality.\(^{15,16}\) Though the tendency to order is more pronounced in three-dimensional \((3d)\) systems than in low-dimensional ones, a magnetically disordered phase can also be observed in frustrated 3d systems such as the HAFM on the pyrochlore lattice\(^{16}\) or on the stacked kagomé lattice.\(^{17}\) On the other hand, recently it has been found that the 3d \( J_1-J_2 \) model on the body-centered cubic lattice does not have an intermediate quantum paramagnetic phase.\(^{18,19}\) Moreover, in experimental realizations of the \( J_1-J_2 \) model the magnetic couplings are expected to be not strictly 2d, but a finite interlayer coupling \( J_\perp \) is present. For example, recently Rosner et al.\(^{14}\) have found \( J_\perp/J_1 \approx 0.07 \) for \( Li_2VOSiO_4 \), a material which can be described by a square lattice \( J_1-J_2 \) model with large \( J_2 \).\(^{13,14}\)

This motivates us to consider an extension of the \( J_1-J_2 \) model, namely the \( J_1-J_2 \) spin-1/2 HAFM on the stacked square lattice described by the Hamiltonian

\[
H = \sum_n \left( J_1 \sum_{\langle ij \rangle} \mathbf{s}_{i,n} \cdot \mathbf{s}_{j,n} + J_2 \sum_{\langle ij \rangle} \mathbf{s}_{i,n} \cdot \mathbf{s}_{j,n} \right) + J_\perp \sum_{i,n} \mathbf{s}_{i,n} \cdot \mathbf{s}_{i,n+1},
\]

where \( n \) labels the layers and \( J_\perp \geq 0 \) is the interlayer coupling. The expression in brackets represents the \( J_1-J_2 \) model of the layer \( n \) with intralayer couplings \( J_1 = 1 \) and \( J_2 \geq 0 \). The main problem we would like to study is the influence of \( J_\perp \) on the existence of the intermediate quantum paramagnetic GS phase. Note that the exact diagonalization widely used for the study of the strictly 2d \( J_1-J_2 \) model, see, e.g., Refs. 2–4, is not appropriate for the 3d problem under consideration. Therefore, we use the coupled-cluster method \((CCM)\)\(^{6,20–24}\) and the rotation-invariant Green’s function method \((RGM)\).\(^{5,17,26,28–30}\) Both methods have been successfully applied to quantum spin systems in arbitrary dimension and are able to deal with frustration.

Let us briefly illustrate some basic features of the CCM. For more details the reader is referred to Refs. 6 and 20–24. The starting point for the CCM calculation is the choice of a reference state \((\Phi)\). For \((\Phi)\) of the considered spin system we choose the two-sublattice Néel state for small \( J_2 \) but a collinear state for large \( J_2 \). To treat each site equivalently 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, i.e., in the rotated coordinate frame we have \((\Phi) = |\downarrow\rangle |\downarrow\rangle |\downarrow\rangle \ldots \). Note that in this new frame the Hamiltonian is modified,
and $|\Phi\rangle=|\downarrow\downarrow\rangle|\downarrow\rangle \ldots$ is not an eigenstate of this modified Hamiltonian, see, e.g. Refs. 20, 21, 23. For the ket GS $|\Psi\rangle$ with $H|\Psi\rangle=E|\Psi\rangle$ an exponential ansatz $|\Psi\rangle=e^{S}|\Phi\rangle$ is used, where the correlation operator $S$ is given by $S=\sum_{f\neq j} S_{C_f^+}^j$. The $C_f^+$ represent a set of multi-spin creation operators $C_f^+=s_i^+, s_i^+s_j^+, s_i^+s_j^+s_k^+, \ldots$. The application of all the $C_f^+$ on $|\Phi\rangle$ creates a complete set of states, which may contribute to $|\Psi\rangle$. The correlation operator $S$ contains the coefficients $S_j$ which are determined by requiring that the expectation value of $H$ is a minimum. The order parameter $M$ is given by the expectation value of $s_i^+s_i^-$. 

For the considered quantum many-body model it is necessary to use approximations in order to truncate the expansion of $S$. We use the well elaborated LSUBn scheme\textsuperscript{20,21,23} in which in the correlation operator $S$ all multi-spin correlations over all distinct locales on the lattice defined by $n$ or fewer contiguous sites are taken into account. For example, within the LSUB4 scheme one includes multi-spin creation operators of one, two, three or four spins distributed on arbitrary clusters of four contiguous lattice sites. The number of these fundamental configurations can be reduced exploiting lattice symmetry and conservation laws. In the CCM-LSUB8 approximation we have finally 25953 (43070) fundamental configurations for the Néel (collinear) reference state. To solve the set of the corresponding ket equations we use parallel computing.\textsuperscript{25}

Since the LSUBn approximation becomes exact for $n \to \infty$, it is useful to extrapolate the ‘raw’ LSUBn data to $n \to \infty$. An appropriate extrapolation rule for the order parameter of systems showing a GS order-disorder transition is the ‘leading power-law’ extrapolation\textsuperscript{23} $M(n)=c_0+c_1(1/n)^2$, where the results of the LSUB4,6,8 approximations are used for the extrapolation. For the GS energy per spin $e(n)=a_0+a_1(1/n^2)+a_2(1/n^4)$ is a reasonable extrapolation ansatz.\textsuperscript{22}

Next we give a brief illustration of the spin-rotation-invariant Green’s function method.\textsuperscript{26,27} More details can be found in Refs. 5,17,28 and 30. Considering the equations of motion for the commutator Green’s function $\langle i[s_i^+ s_i^-]_\omega \rangle$ and supposing spin inversion, i.e. $\langle s_i^+ \rangle=0$, we get $\omega^2 \langle i[s_i^+ s_i^-]_\omega \rangle=\langle [i\not{s}_i^+ s_i^-]_\omega \rangle+\langle [i\not{s}_i^- s_i^+]_\omega \rangle$. To treat the operator $\not{s}_i^+$ containing products of three spin operators along nearest-neighbor sequences, a decoupling procedure in the spirit of Ref. 26 is performed. For example, the operator product $\not{s}_A^+ \not{s}_B^+ \not{s}_C^+$ is replaced by $\eta_{A,B} \langle \not{s}_A^+ \not{s}_B^+ \rangle+\eta_{A,C} \langle \not{s}_A^+ \not{s}_C^+\rangle+\eta_{B,C} \langle \not{s}_B^+ \not{s}_C^+\rangle$, where $A,B,C$ represent spin sites. The introduction of vertex parameters $\eta_{\rho,\mu}$ is aimed to improve the approximation and to fulfill fundamental constraints like the sum rule.

By analogy with Refs. 5 and 28 we use four different vertex parameters, namely $\eta_{\parallel}$ related to the correlator $c_{1,0,0}$, $\eta_{\perp}$ related to $c_{0,0,1}$, $\eta_2$ commonly related to $c_{2,0,0}$, $c_{2,1,0}$, $c_{2,2,0}$, $c_{1,0,1}$, $c_{1,1,1}$, $c_{0,0,2}$, and $\eta_3$ related to $c_{1,1,0}$. The correlators are defined as $c_{k,l,m}=\langle \not{s}_i^+ \not{s}_j^- \rangle=2\langle \not{s}_R \otimes \not{s}_L \rangle/3$ with the lattice vector $\not{R}=ka_1+la_2+ma_3$ and have to be determined self-consistently. Performing the approximations mentioned above we obtain $\langle \not{s}_A^+ \not{s}_B^- \rangle_\omega=m_\omega/(\omega^2-\omega_0^2)$, where for $m_\omega$ and $\omega_0^2$ explicit equations can be given. The equation for $\omega_0^2$ contains the four vertex parameters and the nine correlators mentioned above. The correlators can be expressed by the Green’s function using the spectral theorem. To determine the four vertex parameters we use the sum rule $c_{0,0,0}=1/2$ and require that the static susceptibility $\chi_{\omega}^+ = -\lim_{\omega \to 0} \langle \not{s}_A^+ \not{s}_B^- \rangle_\omega$ has to be isotropic in the limit $q \to 0$.\textsuperscript{17,28,30} The remaining two equations are obtained as follows: First we use the relation $\eta_3 = \langle (\not{s}_A^+ \not{s}_B^-) | (1+\not{s}_2) \rangle^{-1}$ which was successfully applied in Ref. 5 to the 2d $J_1-J_2$ model. This relation interpolates between the two limiting cases $J_2 \to 0$ and $J_2 \to \infty$ and takes care of the relation $\lim_{J_2 \to 0} c_{1,0,0} = \lim_{J_2 \to \infty} c_{1,1,0}$. Finally we use, following Ref. 5, an approximative expression for the GS energy per spin $e_{\text{GS}}^{\text{input}} = 3J_1c_{1,0,0}+3J_2c_{1,1,0}+3J_1c_{0,1,2}$ as an additional input. For the stacked HAFM considered we make the ansatz $e_{\text{GS}}^{\text{input}} (J_2,J_\perp)=f_1(J_\perp)+f_2(J_2)$ (note that $J_1=1$). To fix $f_2$ we use the exact diagonalization result for the GS energy of the finite 2d $J_1-J_2$ model ($J_\perp=0$) of $N=32$ spins, i.e. we set $f_2(J_2)=e_0^{N=32}(J_2,J_\perp=0)$. To fix $f_1$ we use the GS energy of the unfrustrated stacked square lattice $e_0^{\text{SW}}(J_2=0,J_\perp)$ calculated by linear spin-wave theory and set $f_1(J_\perp)=e_0^{\text{SW}}(J_2=0,J_\perp)-e_0^{N=32}(J_2=0,J_\perp=0)$ this way taking into account the effect of the interlayer coupling and a finite-size correction.

To discuss GS magnetic order-disorder transitions we consider the magnetic order parameter. In the RGM scheme\textsuperscript{26–28,30} the correlation function $\langle \not{s}_R \otimes \not{s}_L \rangle$ at $T=0$ is given by

$$\langle \not{s}_R \otimes \not{s}_L \rangle = \frac{3}{2N} \sum_{q \neq Q} \frac{m_\omega e^{-iqR}}{2\omega} + \frac{3}{2} \sum_{Q_j} C_{Q_j} e^{-iQ_j R}. \quad (2)$$

The second term (condensation part) describes LRO, where the sum runs over different nonequivalent magnetic wave vectors $Q_j$ taking into account the possibility to have degenerate GSs. For model (1) we have $Q_0=\pi \perp \pi$ for the Néel phase and $Q_1=(\pi,0,\pi)$ or $Q_2=(0,0,\pi)$ for the collinear phase. Magnetic LRO is accompanied by a diverging static susceptibility $\chi_{\omega}^+$ at $q=Q$ giving an additional equation for $C_Q$. Note that for the collinear phase both condensation terms are equal, i.e. $C_{Q_1}=C_{Q_2}$. The order parameter $M$ can be calculated by $M^2=3|C_Q|^2/2$. That way, the order parameter is linked to the long-range behavior of the correlation functions because $M$ is nonzero if $\lim_{R \to \infty} \langle \not{s}_R \otimes \not{s}_L \rangle$ remains finite.

As in the 2d case the GS of the stacked model is characterized by two magnetically long-range ordered phases, namely a Néel phase for small $J_2$ and a collinear phase for large $J_2$. For not too large $J_\perp$ both magnetic phases are separated by a magnetically disordered quantum paramagnetic phase, where the phase transition points are func-
tions of $J_\perp$. To determine these transition points we calculate the order parameters for various $J_\perp$ to find those values $J_\perp = \alpha_{\text{Néel}}(J_\perp)$ and $J_\perp = \alpha_{\text{coll}}(J_\perp)$ where the order parameters vanish. In Fig. 1 we present some typical curves showing the order parameters versus $J_\perp$ for some values of $J_\perp$. Both approaches lead to qualitatively comparable results. The magnetic order parameters of both magnetically long-range ordered phases vanish continuously as it is typical for second-order transitions. Note, however, that there are arguments\textsuperscript{3,9} that the transition from the collinear phase to the quantum paramagnetic phase should be of first order. The order parameters are monotonously increasing with $J_\perp$, and the transition points $\alpha_{\text{Néel}}$ and $\alpha_{\text{coll}}$ move together. In Fig. 2 we present these transition points in dependence on $J_\perp$. Close to the strictly 2d case, i.e. for small $J_\perp \ll 1$, the influence of the interlayer coupling is largest. For a characteristic value of $J_\perp^* \approx 0.31$ (0.19) for the RGM (CCM) approach the transition points $\alpha_{\text{Néel}}$ and $\alpha_{\text{coll}}$ meet each other.

For larger $J_\perp$ exceeding $J_\perp^*$ we have a direct first-order transition between both types of magnetic LRO as it is also observed in the classical model and in the 3d quantum $J_1$-$J_2$ model on the body-centered cubic lattice.\textsuperscript{18,19}

However, the description of this first-order transition is not possible within the RGM approach. The reason is that the approximative expression for the GS energy per spin $\epsilon_0^{\text{input}}$ used as an input is a smooth function of $J_\perp$, whereas a first-order GS transition is characterized by a kink in $\epsilon_0$. As a consequence we find that there is no solution of the system of coupled RGM equations for parameter values being close to a first-order transition, i.e. for $J_\perp \approx 0.5$ and $J_\perp > J_\perp^*$. The order parameter curve for $J_\perp = 0.4$ depicted in Fig. 1(a) indeed shows a small region slightly below $J_\perp = 0.5$, where no solution exists.

In contrast to the RGM the CCM approach starts with two different reference states (Néel and collinear) related to the two types of magnetic LRO. Though we start our CCM calculation with a reference state corresponding to semiclassical order, one can compute the GS energy also in parameter regions where semiclassical magnetic LRO is destroyed, and it is known\textsuperscript{6,22–24} that the CCM yields precise results for the GS energy beyond the transition from the semiclassical magnetic phase to the quantum paramagnetic phase. The necessary condition for the

![FIG. 1: Magnetic order parameter $M$ versus $J_2$ for various strengths of the interlayer coupling $J_\perp$. (a): RGM, (b): CCM.](image1)

![FIG. 2: Ground-state phase diagram. (a): RGM, (b): CCM. The solid lines show those values of $J_2$ where the order parameters vanish. The dashed line in (b) represents those values of $J_2$ where the two energies calculated for the Néel and collinear reference state become equal.](image2)
Both quantities are obtained by extrapolation of the 'raw' LSb results to the limit n \to \infty as explained in the text. The energies calculated with Néel and collinear reference states become equal at \( J_2 \approx 0.58 \) indicating a first-order transition. For the order parameter \( M \) we take that value calculated with the reference state of lower CCM energy.

The convergence of the CCM equations is a sufficient overlap between the reference state and the true GS. Hence we can add to the above discussion of the order parameters a comparison of the energies. Provided that the CCM equations converge for the Néel and the collinear reference state far enough beyond those points where the order parameters vanish we can determine the point where both energies become equal. For the considered LSb approximations this happens for \( J_\perp \gtrsim 0.1 \). In the inset of Fig. 3 we show the energies versus \( J_2 \) for \( J_\perp = 0.2 \) calculated by extrapolation. The corresponding points \( J_2 = \alpha'_{\text{coll}}(J_\perp) \) where both energies meet are shown in Fig. 2 as dashed line.

We obtain that both transition points \( \alpha_{\text{coll}} \) and \( \alpha'_{\text{coll}} \) are close to each other and show a similar dependence on \( J_\perp \). Second, we find that at least for \( J_\perp \gtrsim 0.1 \) the energy obtained with the Néel reference state is lower than that obtained with the collinear reference state even for \( J_2 \) values where the Néel order parameter is already zero but the collinear order parameter is still finite. Thus, this energetic consideration leads to the following sequence of zero-temperature transitions: Second-order transition from Néel LRO to a quantum paramagnetic phase at \( J_2 = \alpha_{\text{Neel}} \) and then a first-order transition from the quantum paramagnetic phase to collinear LRO at \( J_2 = \alpha'_{\text{coll}} > \alpha_{\text{coll}} > \alpha_{\text{Neel}} \). This behavior is illustrated in Fig. 3, where the order parameter \( M \) is shown versus \( J_2 \) for fixed \( J_\perp = 0.2 \). For a certain value of \( J_\perp \approx 0.23 \) both transition points \( \alpha_{\text{Neel}} \) and \( \alpha'_{\text{coll}} \) become equal, and one has a direct first-order transition between the two semiclassically long-range ordered phases.

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