Quantum J_1-J_2 Antiferromagnet on a Stacked Square Lattice: Influence of the Interlayer Coupling on the Ground-State Magnetic Ordering

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Using the coupled-cluster method and the rotation-invariant Green's function method, 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 $J_2 < J_{c_1}$ and large $J_2 > J_{c_2}$ 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 $J_\perp \sim (0.2\text{–}0.3)J_1$.

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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 past 15 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 $J_{c_1} < J_2 < J_{c_2}$, where $J_{c_1} \approx 0.4$ and $J_{c_2} \approx 0.6$. The ground state (GS) at low $J_2 < J_{c_1}$ exhibits semiclassical Néel magnetic LRO with the magnetic wave vector $\mathbf{Q}_0 = (\pi, \pi)$. The GS at large $J_2 > J_{c_2}$ shows so-called collinear magnetic LRO with the magnetic wave vectors $\mathbf{Q}_1 = (\pi, 0)$ or $\mathbf{Q}_2 = (0, \pi)$. These two collinear states are characterized by a parallel spin orientation of nearest neighbors in the vertical (horizontal) direction and an antiparallel spin orientation of nearest neighbors in the horizontal (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]. 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 $J_\perp/J_1 \sim 0.07$ 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

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

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) [5,17,25–28]. 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. [6,20–24]. The starting point for the CCM calculation is the choice of a reference state $|\Phi\rangle$. For $|\Phi\rangle$ 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 $|\Phi\rangle = |\downarrow\rangle|\downarrow\rangle|\downarrow\rangle \ldots$. Note that in this new

frame the Hamiltonian is modified, and $|\Phi\rangle = |\downarrow\rangle|\downarrow\rangle|\downarrow\rangle\ldots$ 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 $S = \sum_{I\neq 0}\mathcal{S}_I C_I^+$. The C_I^+ represent a set of multispin creation operators $C_I^+ = s_i^+, s_i^+ s_j^+, s_i^+ s_j^+ s_k^+, \ldots$. The application of all of the C_I^+ on $|\Phi\rangle$ creates a complete set of states, which may contribute to $|\Psi\rangle$. The correlation operator S contains the coefficients $\mathcal{S}_I$, 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^z.

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 [20,21,23], in which in the correlation operator S all multispin 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 multispin 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 by exploiting lattice symmetry and conservation laws. In the CCM-LSUB8 approximation, we have finally 25 953 (43 070) fundamental configurations for the Néel (collinear) reference state. To solve the set of the corresponding ket equations, we use parallel computing [29].

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 [23] $M(n) = c_0 + c_1(1/n)^{c_2}$, 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 [22].

Next we give a brief illustration of the spin-rotation-invariant Green's function method [25,30]. More details can be found in Refs. [5,17,26,28]. Considering the equations of motion for the commutator Green's function $\langle\langle s_{\mathbf{q}}^+; s_{\mathbf{q}}^-\rangle\rangle_\omega$ and supposing spin-rotation invariance, i.e., $\langle s_m^z\rangle \equiv 0$, we get $\omega^2\langle\langle s_{\mathbf{q}}^+; s_{\mathbf{q}}^-\rangle\rangle_\omega = \langle[i\dot{s}_{\mathbf{q}}^+, s_{\mathbf{q}}^-]_-\rangle + \langle\langle -\ddot{s}_{\mathbf{q}}^+; s_{\mathbf{q}}^-\rangle\rangle_\omega$. To treat the operator $\ddot{s}_{\mathbf{q}}^+$ containing products of three spin operators along nearest-neighbor sequences, a decoupling procedure in the spirit of Ref. [25] is performed. For example, the operator product $s_A^- s_B^+ s_C^+$ is replaced by $\eta_{A,B}\langle s_A^- s_B^+\rangle s_C^+ + \eta_{A,C}\langle s_A^- s_C^+\rangle s_B^+$, where A, B, and C represent spin sites. The introduction of vertex parameters $\eta_{\gamma,\mu}$ is aimed to improve the approximation and to fulfill fundamental constraints like the sum rule.

By analogy with Refs. [5,26], we use four different vertex parameters, namely, $\eta_{1\|}$ related to the correlator $c_{1,0,0}$, $\eta_{1\perp}$ related to $c_{0,0,1}$, η_2 commonly related to $c_{2,0,0}$, $c_{2,1,0}$, $c_{2,2,0}$, $c_{1,0,1}$, $c_{1,1,1}$, and $c_{0,0,2}$, and η_3 related to $c_{1,1,0}$. The correlators are defined as $c_{k,l,m} \equiv c_{\mathbf{R}} = \langle s_{\mathbf{0}}^+ s_{\mathbf{R}}^-\rangle = 2\langle \mathbf{s_0}\cdot\mathbf{s_R}\rangle/3$, with the lattice vector $\mathbf{R} = k\mathbf{a}_1 + l\mathbf{a}_2 + m\mathbf{a}_3$, and have to be determined self-consistently. Performing the approximations mentioned above, we obtain $\langle\langle s_{\mathbf{q}}^+; s_{\mathbf{q}}^-\rangle\rangle_\omega = m_{\mathbf{q}}/(\omega^2 - \omega_{\mathbf{q}}^2)$, where for $m_{\mathbf{q}}$ and $\omega_{\mathbf{q}}^2$ explicit equations can be given. The equation for $\omega_{\mathbf{q}}^2$ 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 $\chi_{\mathbf{q}}^{+-} = -\lim_{\omega\to 0}\langle\langle s_{\mathbf{q}}^+; s_{\mathbf{q}}^-\rangle\rangle_\omega$ has to be isotropic in the limit $\mathbf{q} \to \mathbf{0}$ [17,26,28]. The remaining two equations are obtained as follows: First, we use the relation $\eta_3 = (\eta_2 e^{-J_2} + J_2\eta_{1\|})(1 + J_2)^{-1}$, 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 $e_0^{\text{input}} = 3J_1 c_{1,0,0} + 3J_2 c_{1,1,0} + 3J_\perp c_{0,0,1}/2$ as an additional input. For the stacked HAFM considered, we make the ansatz $e_0^{\text{input}}(J_2, J_\perp) = f_1(J_\perp) + f_2(J_2)$ (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 [25,26,28,30], the correlation function $\langle \mathbf{s_0}\cdot\mathbf{s_R}\rangle$ at $T = 0$ is given by

$$\langle \mathbf{s_0}\cdot\mathbf{s_R}\rangle = \frac{3}{2N}\sum_{\mathbf{q}\neq\mathbf{Q}_j}\frac{m_{\mathbf{q}}}{2\omega_{\mathbf{q}}}e^{-i\mathbf{q}\cdot\mathbf{R}} + \frac{3}{2}\sum_{\mathbf{Q}_j} C_{\mathbf{Q}_j} e^{-i\mathbf{Q}_j\cdot\mathbf{R}}. \quad (2)$$

The second term (condensation part) describes LRO, where the sum runs over different nonequivalent magnetic wave vectors $\mathbf{Q}_j$ taking into account the possibility to have degenerate GSs. For model (1), we have $\mathbf{Q}_0 = (\pi, \pi, \pi)$ for the Néel phase and $\mathbf{Q}_1 = (\pi, 0, \pi)$ or $\mathbf{Q}_2 = (0, \pi, \pi)$ for the collinear phase. Magnetic LRO is accompanied by a diverging static susceptibility $\chi_{\mathbf{q}}^{+-}$ at $\mathbf{q} = \mathbf{Q}$, giving an additional equation for $C_{\mathbf{Q}}$. Note that for the collinear phase both condensation terms are equal; i.e., $C_{\mathbf{Q}_1} = C_{\mathbf{Q}_2}$. The order parameter M can be calculated by $M^2 = 3|C_{\mathbf{Q}}|/2$. That way, the order parameter is linked to the long-range behavior of the correlation functions because M is nonzero if $\lim_{|\mathbf{R}|\to\infty}\langle \mathbf{s_0}\cdot\mathbf{s_R}\rangle$ 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 functions of $J_\perp$. To determine these transition points, we calculate the order parameters for various $J_\perp$ to find those values $J_2 = \alpha_{\text{Neel}}(J_\perp)$ and $J_2 = \alpha_{\text{coll}}(J_\perp)$ where the order parameters vanish. In Fig. 1, we present some typical curves showing the order parameters versus J_2 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 is typical for second-order transitions. Note, however, that there are arguments [3,9] that the transition from the collinear phase to the quantum paramagnetic phase should be first-order. The order parameters are monotonously increasing with $J_\perp$, and the transition points α_{Neel} and α_{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 α_{Neel} and α_{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 is also observed in the classical model and in the 3D quantum J_1-J_2 model on the body-centered cubic lattice [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 e_0^{input} used as an input is a smooth function of J_2, whereas a first-order GS transition is characterized by a kink in e_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 $J_2 \approx 0.5$ 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 $J_2 = 0.5$, 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 [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 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

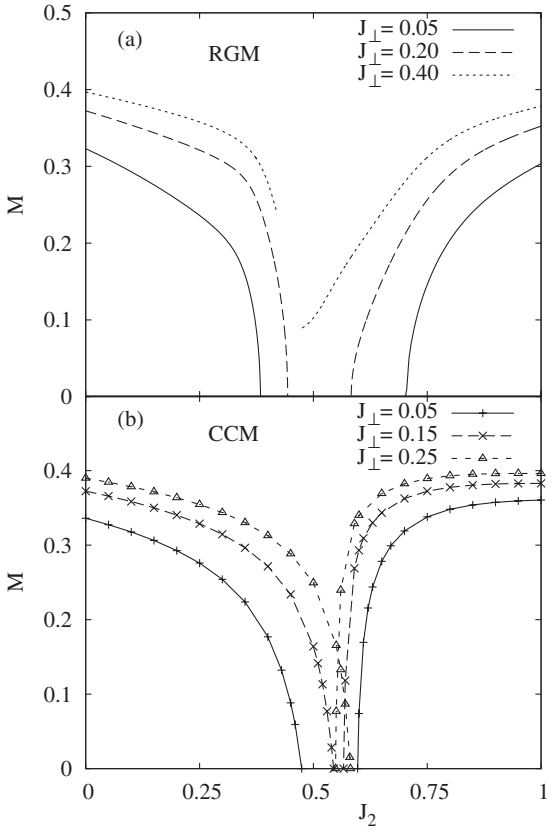

FIG. 1. Magnetic order parameter M versus J_2 for various strengths of the interlayer coupling $J_\perp$. (a) RGM, (b) CCM.

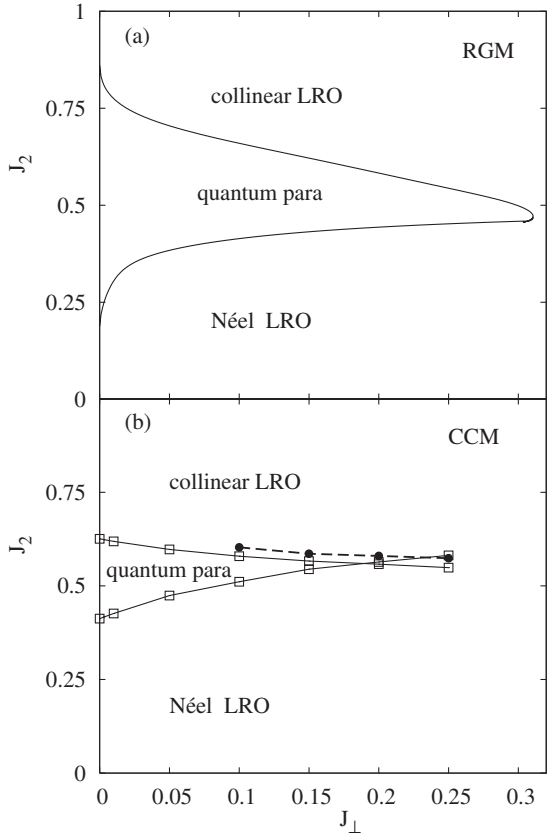

FIG. 2. Ground-state phase diagram. (a) RGM, (b) CCM. The solid lines show those values of J_2 where the order parameters vanish. The dashed line in (b) represents those values of J_2 where the two energies calculated for the Néel and collinear reference states become equal.

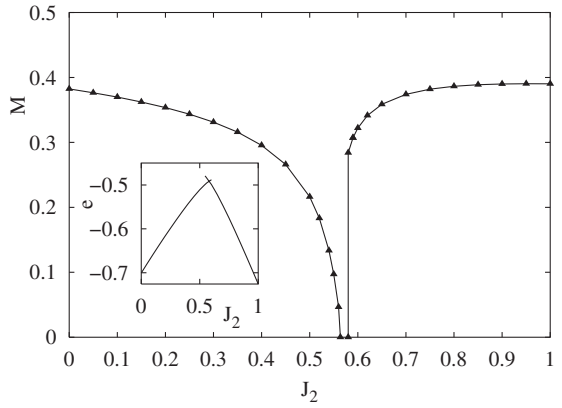

FIG. 3. CCM results for the energy per spin e for both reference states (inset) and the order parameter M for $J_\perp = 0.2$. Both quantities are obtained by extrapolation of the raw LSUBn results to the limit $n \to \infty$ as explained in the text. The energies calculated with the Néel and collinear 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.

comparison of the energies. Provided that the CCM equations converge for the Néel and the collinear reference states far enough beyond those points where the order parameters vanish, we can determine the point where both energies become equal. For the considered LSUBn approximations, this happens for $J_\perp \gtrsim 0.1$. In the inset in 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 a dashed line.

We obtain that both transition points α_{coll} and α'_{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 α_{Neel} and α'_{coll} become equal, and one has a direct first-order transition between the two semiclassically long-range ordered phases.

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