Phase Diagram of the Frustrated Square-lattice Hubbard Model in a Strong Correlation Regime

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Variational cluster approximation is used to study the frustrated Hubbard model at half filling defined on the two-dimensional square lattice with anisotropic next-nearest-neighbor hopping parameters. We present the ground-state phase diagram of the model in a strong correlation regime at zero temperature. We show that the magnetic phases with Néel, collinear, and spiral orders appear in relevant parameter regions, and in an intermediate region between these phases, the quantum disordered phase caused by the geometrical frustration in the spin degrees of freedom emerges.

1. Introduction

The effect of geometrical frustration in strongly correlated electron systems has been one of the major issues of condensed matter physics. In particular, a spin liquid state caused by the frustration has been interpreted as an exotic state of matter, where the magnetic long-range order is destroyed, yielding a quantum paramagnetic (or nonmagnetic) state at zero temperature\(^1\) or even exotic mechanisms of high-temperature superconductivity.\(^2\) The Hubbard, Heisenberg, and related models defined on the two-dimensional square and triangular lattices with the geometrical frustration have been studied in this respect to find the novel quantum disordered states by means of a variety of theoretical methods.

In the square-lattice cases, the \(J_1-J_2\) Heisenberg model with the nearest-neighbor \((J_1)\) and next-nearest-neighbor \((J_2)\) exchange interactions have been studied for more than two decades.\(^3\)\(^{-}\)\(^{32}\) At \(J_2 = 0\), where the frustration is absent, the model is known to have the Néel-type antiferromagnetic long-range order. With increasing \(J_2\), the frustration increases, but at \(J_2 = J_1\), the model again has the ground state with the collinear antiferromagnetic long-range order. The strongest frustration occurs around \(J_2/J_1 = 0.5\), where a nonmagnetic disordered states such as a valence bond state\(^4\)\(^{-}\)\(^{8}\),\(^10\)\(^{-}\)\(^{16}\),\(^18\),\(^22\),\(^29\) and a spin-liquid state\(^12\),\(^24\)\(^{-}\)\(^{28}\) have been suggested to appear, the region of which has recently been studied further in detail.\(^31\),\(^32\)

The \(t_1-t_2\) Hubbard model with the nearest-neighbor \((t_1)\) and next-nearest-neighbor \((t_2)\) hopping parameters has also been studied, where it has been shown that the ground state is the Néel order at a small \(t_2/t_1\) region and a collinear order around \(t_2 = t_1\)\(^{33}\),\(^34\) and that the quantum disordered state appears between these ordered states.\(^35\),\(^36\)

In the triangular-lattice cases, the anisotropic \(J-J'\) triangular Heisenberg model has been studied. In the isotropic case \((J = J')\), the \(120^\circ\) spiral ordered phase is known to be stable.\(^37\) In the anisotropic case, the Néel order is realized when \(J'/J\) is small and the spiral order is realized around \(J'/J = 1\)\(^{38}\)\(^{-}\)\(^{49}\) and between these phases, a dimer ordered phase\(^39\) or a spin-liquid phase\(^47\),\(^48\) have been predicted to appear. The anisotropic \(t-t'\) triangular Hubbard model has also been studied\(^50\)\(^{-}\)\(^{54}\) and a quantum disordered state has also been observed between the Néel and spiral phases.\(^51\),\(^52\),\(^54\)

Recently, the magnetic orders in the triangular-lattice Heisenberg model with the nearest-neighbor \((J_1)\) and next-nearest-neighbor \((J_2)\) exchange interactions have also been studied, where the quantum disordered phase is shown to appear between the spiral and collinear phases.\(^55\)\(^{-}\)\(^{59}\)

In this paper, motivated by the above developments in the field, we study the frustrated square-lattice Hubbard model at half filling with the isotropic nearest-neighbor and anisotropic next-nearest-neighbor hopping parameters and clarify the appearance of possible magnetic orderings and emergence of quantum disordered phase. The search is done for a wide parameter space including the square, crossed-square, and triangular lattices. We use the variational cluster approximation (VCA) based on the self-energy functional theory (SFT),\(^60\)\(^{-}\)\(^{62}\) which enables us to take into account the quantum fluctuations of the system, so that we can study the effect of geometrical frustration on the spin degrees of freedom and determine the critical interaction strength for the spontaneous symmetry breaking of the model. We in particular focus on the strong correlation regime at zero temperature of which not much is known so far, and compare our results with those of the Heisenberg model for which many studies have been accumulated.

We will thereby show that the magnetic phases with the Néel, collinear, and spiral orders appear in relevant regions of the parameter space of our model and that the quantum disordered phase caused by the effect of...
frustration emerges in a wide parameter region between the ordered phases obtained. The orders of the phase transitions will also be determined. We will summarize our results as a ground-state phase diagram in a full two-dimensional parameter space. This phase diagram will make the characterization of the quantum disordered phase more approachable although it is beyond the scope of the present paper.

2. Model and method

We consider the frustrated Hubbard model defined on the two-dimensional square lattice at half filling as illustrated in Fig. 1. The Hamiltonian is given by

\[ H = -t_1 \sum_{\langle i,j \rangle} \sum_{\sigma} c_{i\sigma}^\dagger c_{j\sigma} - t_2 \sum_{\langle \langle i,j \rangle \rangle} \sum_{\sigma} c_{i\sigma}^\dagger c_{j\sigma}^\dagger + t'_2 \sum_{\langle \langle i,j \rangle \rangle^\prime} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} - \mu \sum_{i,\sigma} n_{i\sigma}, \]  

(1)

where \( c_{i\sigma}^\dagger \) is the creation operator of an electron with spin \( \sigma \) at site \( i \) and \( n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma} \). \( \langle i,j \rangle \) indicates the nearest-neighbor bonds with an isotropic hopping parameter \( t_1 \), and \( \langle \langle i,j \rangle \rangle \) and \( \langle \langle i,j \rangle \rangle^\prime \) indicate the next-nearest-neighbor bonds with anisotropic hopping parameters \( t_2 \) and \( t'_2 \), respectively [see Fig. 1(a)]. \( U \) is the on-site Coulomb repulsion between electrons and \( \mu \) is the chemical potential maintaining the system at half filling. In the large-\( U \) limit, the model can be mapped onto the frustrated spin-1/2 Heisenberg model

\[ H = J_1 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J'_2 \sum_{\langle\langle i,j \rangle\rangle^\prime} \mathbf{S}_i \cdot \mathbf{S}_j \]  

(2)

in the second-order perturbation of the hopping parameters. We define the spin-1/2 operator \( \mathbf{S}_i = c_{i\uparrow}^\dagger \sigma_{\alpha\beta} c_{i\beta} \), where \( \sigma_{\alpha\beta} \) is the vector of Pauli matrices. The exchange coupling constants are given by \( J_1 = 4t_1^2/U \), \( J_2 = 4t_2^2/U \), and \( J'_2 = 4t'_2^2/U \) for the lattice shown in Fig. 1(a).

Because in this paper we are interested in the geometrical frustration in the spin degrees of freedom of the model and want to compare our results with those of the Heisenberg model for which related studies have been accumulated, we restrict ourselves to a large-\( U \) region assuming a value \( U/t_1 = 60 \), so that we can preclude the Mott metal-insulator transition. We treat a wide parameter space of \( 0 \leq t_2/t_1 \leq 1 \) and \( 0 \leq t'_2/t_1 \leq 1 \), including three limiting cases: (i) at \( t_2 = t'_2 = 0 \) [square lattice, see Fig. 1(b)], where the Néel order is realized, (ii) at \( t_2 = t'_2 = t_1 \) [crossed square lattice, see Fig. 1(c)], where the collinear order is realized, and (iii) at \( t_2 = t_1 \) and \( t'_2 = 0 \) [triangular lattice, see Fig. 1(d)], where the 120° spiral order is realized. We will calculate how the above three ordered phases change when the hopping parameters are varied in the ranges \( 0 \leq t_2 \leq t_1 \) and \( 0 \leq t'_2 \leq t_1 \).

We employ the VCA, which is a quantum cluster method based on the SFT\(^{60–62} \) where the grand potential \( \Omega \) of the original system is given by a functional of the self-energy. By restricting the trial self-energy to that of the reference system \( \Sigma' \), we obtain the grand potential in the thermodynamic limit as

\[ \Omega[\Sigma'] = \Omega' + \text{Tr} \ln (G_0^{-1} - \Sigma')^{-1} - \text{Tr} \ln G', \]  

(3)

where \( \Omega' \) and \( G' \) are the exact grand potential and Green function of the reference system, respectively, and \( G_0 \) is the noninteracting Green function. The short-range electron correlations within the cluster of the reference system are taken into account exactly.

The advantage of the VCA is that the spontaneous symmetry breaking can be treated within the framework of the theory. Here, we introduce the Weiss fields.
for magnetic orderings as variational parameters. The Hamiltonian of the reference system is then given by
\[ H' = H + H_N + H_C + H_S \]
\[ H_N = h_N' \sum_i e^{iQ_N \cdot r_i} S_i^z \]
\[ H_C = h_C' \sum_i e^{iQ_C \cdot r_i} S_i^z \]
\[ H_S = h_S' \sum_i e_{a_i} \cdot S_i \]
where \( h_N', h_C', \) and \( h_S' \) are the strengths of the Weiss fields for the Néel, collinear, and spiral orders, respectively. The wave vectors are defined as \( Q_N = (\pi, \pi) \) for the Néel order and \( Q_C = (0, 0) \) or \( (0, \pi) \) for the collinear order. For the spiral order, the unit vectors \( e_{a_i} \) are rotated by 120\(^\circ\) to each other, where \( a_i = (1, 2, 3) \) is the sublattice index of site \( i \). The variational parameter is optimized on the basis of the variational principle \( \partial \Omega / \partial h' = 0 \) for each magnetic order. The solution with \( h' \neq 0 \) corresponds to the ordered state.

We use a 12-site cluster shown in Fig. 2 as the reference system. This cluster is convenient because we can treat the two-sublattice states (Néel and collinear states) with an equal number of up and down spins, and at the same time the three-sublattice state (spiral state) with an equal number of three sublattice sites. Note that longer period phases such as a spiral phase mentioned in a different system\(^{12}\) cannot be treated in the present approach.

3. Results of calculations

First, let us present the entire phase diagram of our model in Fig. 3, where the result for our Hubbard model in the \( (t_2/t_1, t_2'/t_1) \) plane as well as the same result converted to the Heisenberg-model parameters \( (J_2/J_1, J_2'/J_1) \) are shown. We find three ordered phases: the Néel ordered phase around \( (t_2/t_1, t_2'/t_1) = (0, 0) \), the collinear ordered phase around \( (t_2/t_1, t_2'/t_1) = (1, 1) \), and the spiral ordered phase around \( (t_2/t_1, t_2'/t_1) = (1, 0) \) and \( (0, 1) \). The quantum disordered phase, which is absent in the classical system, appears in an intermediate region between the three ordered phases. As shown below, the phase transition to the collinear phase is of the first order (or discontinuous) and the phase transitions to the Néel and spiral phases are of the second order (or continuous). This phase diagram is determined based on the calculated ground-state energies \( E = \Omega + \mu \) (per site) and magnetic order parameters \( M \) (per site) defined as \( M_N = (2/L_c) \sum_i e^{iQ_N \cdot r_i} \langle S_i^z \rangle \) for the Néel order, \( M_C = (2/L_c) \sum_i e^{iQ_C \cdot r_i} \langle S_i^z \rangle \) for the collinear order, and \( M_S = (2/L_c) \sum_i e_{a_i} \cdot \langle S_i \rangle \) for the spiral order, where \( \langle \cdot \rangle \) stands for the ground-state expectation value and \( L_c \) is the number of sites in the reference system. In the following, we will circumstantiate the obtained phases, in particular along the lines (i), (ii), and (iii) drawn in Fig. 3(a), whereby we will discuss some details of our calculated results and compare with other studies.

Along the line (i): The results are shown in the left panel of Fig. 4, where we assume \( t_2 = t_2' \). At \( t_2 = 0 \), the ground state is the Néel order, and with increasing \( t_2 \), the energy of the Néel order gradually approaches the energy of the disordered state. At \( t_2/t_1 = 0.73 \), the energy of the Néel order continuously reaches the energy of the disordered state and the Néel order disappears. The calculated order parameter indicates the continuous phase transition. At \( t_2/t_1 = 1 \), on the other hand, the ground state is the collinear order. The ground-state energy of the collinear order increases with decreasing \( t_2 \) and at \( t_2/t_1 = 0.79 \), it crosses that of the disordered state, resulting in a discontinuous phase transition as the calculated order parameter indicates. The disordered state thus appears at \( 0.73 < t_2/t_1 < 0.79 \), which corresponds to the region \( 0.53 < J_2/J_1 < 0.63 \) in the Heisenberg-model parameters. In comparison with previous studies on the \( J_1-J_2 \) square-lattice Heisenberg model, which have estimated the transition point between the Néel and disordered phases to be at \( J_2/J_1 = 0.40 - 0.44 \)\(^{17,22,24,31,32}\), our result slightly overestimates the stability of the Néel order. This overestimation may be caused by the cluster.
geometry used in our calculations; if we use the $2 \times 2$ site cluster as the reference system, the transition occurs at $J_2/J_1 = 0.42^{36}$ which is in good agreement with the previous studies. The transition point between the collinear and disordered phases, on the other hand, has been estimated to be at $J_2/J_1 = 0.59 - 0.62^{17,22,24,32}$ which is in good agreement with our result.

Along the line (ii): The results are shown in the middle panel of Fig. 4, where we assume $t_2' = 0$. With increasing $t_2$ from $t_2 = 0$ at which the ground state is the Néel order, the energy of the Néel order gradually approaches the energy of the disordered state, and at $t_2/t_1 = 0.88$, the Néel order disappears continuously. The calculated order parameter indicates the continuous phase transition. At $t_2/t_1 = 1$, on the other hand, the ground state is the spiral order although the energy difference between the spiral and disordered states is very small [see the inset of Fig. 4c)] due to the strong geometrical frustration of the triangular lattice. With decreasing $t_2$ from $t_2/t_1 = 1$, the ground-state energy of the spiral order increases gradually and approaches the energy of the disordered state, and at $t_2/t_1 = 0.89$, the spiral order disappears continuously, in agreement with the calculated order parameter. Thus, the disordered phase appears in a very narrow region $0.88 < t_2/t_1 < 0.89$. The corresponding Heisenberg-model parameters where the Néel and spiral orders disappear are around $J_2/J_1 = 0.79$. The previous studies for the anisotropic triangular-lattice Heisenberg model done by the coupled-cluster and exact-diagonalization methods have given the value around $J_2/J_1 = 0.80 - 0.87$ for the transition point, which is in good agreement with our result.

Along the line (iii): The results are shown in the right panel of Fig. 4, where we assume $t_2 = t_1$. At $t_2' = 0$, the ground state is the spiral order although the energy difference from the disordered state is very small [see the inset of Fig. 4e)]. With increasing $t_2'$, the energy of the spiral order gradually approaches the energy of the disordered state, and at $t_2'/t_1 = 0.34$, the spiral order disappears continuously, in agreement with the calculated order parameter. On the other hand, with decreasing $t_2'$ from $t_2'/t_1 = 1$ at which the collinear order is stable, the ground-state energy of the collinear order increases and crosses the energy of the disordered state at $t_2'/t_1 = 0.59$. The transition is thus discontinuous, in agreement with the calculated order parameter. The disordered state therefore appears at $0.34 < t_2'/t_1 < 0.59$, which corresponds to the region $0.11 < J_2/J_1 < 0.34$ if we use the Heisenberg-model parameters. To our knowledge, no comparable calculations have been made for the frustrated Heisenberg model in this parameter region.

4. Summary

In summary, we have used the VCA based on the SFT to study the two-dimensional frustrated Hubbard model at half filling with the isotropic nearest-neighbor and anisotropic next-nearest-neighbor hopping parameters. We have in particular focused on the effect of geometrical frustration on the spin degrees of freedom of the
model in the strong correlation regime at zero temperature, and have investigated the magnetic orderings and emergence of the quantum disordered phase in a wide parameter space including the square, crossed-square, and triangular lattices.

We have thereby presented the ground-state phase diagram of the model, which includes the magnetic phases with the Néel, collinear, and spiral orders. We have also shown that the quantum disordered phase caused by the effect of frustration emerges in a wide parameter region between the three ordered phases obtained and that the phase transition from the Néel and spiral orders to the disordered phase is continuous (or second-order transition), whereas the transition from the collinear order to the disordered phase is discontinuous (or first-order transition). We have compared our results with the results of the corresponding Heisenberg-model calculations that have been made so far and found that the agreement is good whenever the comparison is possible. We hope that our results for the phase diagram will encourage future studies on the characterization of the quantum disordered state obtained.

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