Frustration and magnetic orderings in the square-lattice Hubbard model with anisotropic next-nearest-neighbor hopping

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Abstract. Magnetic orders of the square-lattice Hubbard model with the anisotropic next-nearest-neighbor hopping parameters at half filling are studied by the variational cluster approximation based on the self-energy functional theory. We show that in the strong correlation region at zero temperature the disordered phase appears between the collinear-ordered and 120°-ordered phases due to frustration in the spin degrees of freedom of the model. We also show that the phase transitions to the two ordered phases from the disordered phase are of the first order.

1. Introduction

Physics of geometrical frustration in strongly correlated electron systems has attracted much attention. In particular, magnetic orders of the triangular-lattice Hubbard and related models have been one of the major issues in this field. Here, the presence of the spin-liquid phase has been of great interest. Experimentally, the organic charge-transfer salts \(\kappa\)-(BEDT-TTF)\(_2\)X are a good example of such systems: the compound with \(X=\text{Cu}[\text{N(CN)}_2]\text{Cl}\) displays a long-range antiferromagnetic order in its Mott-insulating phase [1, 2] and the compound with \(X=\text{Cu}_2(\text{CN})_3\) exhibits spin-liquid behaviors at low temperatures, where the bulk spin susceptibility is strongly suppressed by the effects of frustration in the spin degrees of freedom [3, 4].

The emergence of the spin-liquid phase has been studied using a number of theoretical methods for the isotropic and anisotropic triangular-lattice Hubbard models with hopping parameters \(t\) and \(t'\) and on-site Hubbard repulsion \(U\). The isotropic and anisotropic Heisenberg models have also been studied. In the isotropic case, the 120° ordered phase was shown to be stable for the Heisenberg model in the linear spin-wave theory (LSWT) [5, 6], but it was reported that a spin-liquid phase can exist in the model when the parameters are close to the metal-insulator transition [7, 8, 9]. In the anisotropic case, the existence of the spin-liquid phase was predicted to occur in the model with the anisotropic hopping parameters \(t'/t < 1\), which was not found by the LSWT [5, 6]; many numerical techniques were employed including the path-integral renormalization group (PIRG) method [10], dynamical mean-field theory (DMFT) [11], exact diagonalization (ED) method [12, 13], and the finite-temperature Lanczos method [14]. A wider range of \(t'/t\) was studied by the variational Monte Carlo (VMC) [15] method, which indicated possible existence of the spin-liquid phase in a large-\(U\) region. The variational cluster approximation (VCA) was also used to show that the metal-insulator transition from the
metallic phase to the nonmagnetic insulating phase occurs [16, 17]. The spin-liquid phase in the triangular-lattice Heisenberg model with the next-nearest-neighbor exchange couplings was also studied recently by VMC [18], where the spin-liquid phase appears between the 120° ordered phase and stripe antiferromagnetic ordered (or collinear ordered) phase.

In this paper, motivated by such developments in the field, we study the effects of frustration in the spin degrees of freedom on the magnetic behaviors of the square-lattice Hubbard model at half filling, where the anisotropic next-nearest-neighbor hopping parameters are included. We in particular focus on the region of strong electron correlations at a large $U/t$ value. We employ the method of VCA based on the self-energy functional theory (SFT) [19, 20, 21], which is useful for predicting the presence of the spontaneous symmetry breaking of the model. We will thereby show that the nonmagnetic disordered phase appears between the collinear-ordered and 120°-ordered phases and that the phase transitions to the two ordered phases from the disordered phase are of the first order.

2. Model and method

The Hubbard model we consider is defined on the square lattice with the isotropic nearest-neighbor hopping parameter $t_1$ and anisotropic next-nearest-neighbor hopping parameters $t_2$ and $t_2'$, as is illustrated in Fig. 1. The Hamiltonian is given by

$$H = -t_1 \sum_{\langle i,j_1 \rangle, \sigma} c_{i\sigma}^\dagger c_{j_1\sigma} - t_2 \sum_{\langle i,j_2 \rangle, \sigma} c_{i\sigma}^\dagger c_{j_2\sigma} - t_2' \sum_{\langle i,j_2' \rangle, \sigma} c_{i\sigma}^\dagger c_{j_2'\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow},$$

where $\langle i,j_1 \rangle$ indicates the nearest-neighbor bonds, and $\langle i,j_2 \rangle$ and $\langle i,j_2' \rangle$ indicate the next-nearest-neighbor bonds. $c_{i\sigma}$ is the annihilation operator of an electron with spin $\sigma$ at site $i$, and $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ is the number operator. $U$ is the on-site Coulomb repulsion. In this paper, we consider the case at half filling and the large $U$ region assuming a value $U/t_1 = 60$, where the Mott insulating phase is realized. We choose this value of $U/t$ because we are interested in the frustration in the spin degrees of freedom of the model and want to avoid the metal-insulator transition occurring at smaller $U/t$ values. We should moreover mention that the method of
VCA cannot be applied to the Hesenberg-like spin models. We in particular consider the case at $t_1 = t_2 = t$ and $t'_2 = t'$ [see Fig. 1(b)], including the two limiting cases at $t_1 = t_2 = t$ and $t'_2 = t' = 0$ [isotropic triangular lattice, see Fig. 1(a)] and at $t_1 = t_2 = t'_2 = t = t'$ [square lattice with the isotropic next-nearest-neighbor hopping parameters, see Fig. 1(c)]. It has been reported that the $120^\circ$ ordered phase is realized for the isotropic triangular lattice shown in Fig. 1(a) [7] and the collinear ordered phase is realized for the lattice shown in Fig. 1(c) [22]. In this paper, we will discuss how the above two ordered phases change when the hopping parameters $t_1 = t_2 = t$ and $t'_2 = t'$ are varied as $0 < t' < t$, which has not been studied so far.

We employ the method of VCA based on the self-energy functional theory [19, 20, 21]. The advantage of VCA is that the spontaneous symmetry breaking can be treated within the framework of the theory. The stability of the ground state of the original system can be examined via the calculation of the grand potential $\Omega$ of the system from the exact self-energy. The grand potential $\Omega$ is expressed as a functional of the self-energy $\Sigma$ based on the self-energy functional theory:

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

where $\Omega'$ is the exact grand potential of the reference system and $G_0$ is the noninteracting Green function. We use the exact self-energy $\Sigma'$ of the reference system, which is replaced by the Green function of the reference system $G'^{-1} = G_0^{-1} - \Sigma'$. To investigate the spontaneous symmetry breaking in VCA, we introduce the Weiss fields as variational parameters. In the present case, the Hamiltonian of the reference system is given by $H' = H + H_{\text{tri}} + H_{\text{col}}$ with

$$H_{\text{tri}} = h_{\text{tri}} \left[ \sum_{i \in A} e_A \cdot S_i + \sum_{i \in B} e_B \cdot S_i + \sum_{i \in C} e_C \cdot S_i \right],$$

$$H_{\text{col}} = h_{\text{col}} \sum_i e_i Q_{\text{col}} \cdot r_i (n_{i\uparrow} - n_{i\downarrow}),$$

where $e_A$, $e_B$ and $e_C$ are the unit vectors rotated each other by $120^\circ$ defined on the A, B and C sublattices, respectively, and $S_i$ is the spin operator at site $i$ [16, 17]. $h_{\text{tri}}$ and $h_{\text{col}}$ are the strengths of the Weiss fields for the $120^\circ$ ordered and collinear ordered states, respectively. We define $Q_{\text{col}}$ to be either $(\pi, 0)$ or $(0, \pi)$. We use the clusters of the size $L_c = 2 \times 3 = 6$ sites for calculating the $120^\circ$ ordered phase and $L_c = 2 \times 2 = 4$ sites for calculating the collinear ordered phase. Note that longer period phases such as the spiral phase mentioned in a different system [15] cannot be treated in the present approach.

3. Results of calculation

Figures 2 (a) and (b) show the calculated results for the grand potential $\Omega - \Omega_0$ (per site) as a function of the Weiss field, where $\Omega_0$ is the grand potential at zero Weiss fields. We find that the grand potentials of both the $120^\circ$ ordered and collinear ordered states have two minima at $h = 0$ and $h \neq 0$ and a maximum between the two minima, indicating that the stable point of $h_{\text{col}}$ and $h_{\text{tri}}$ changes discontinuously from zero to a finite value at the critical point of $t'/t$.

More quantitatively, we find that $h_{\text{tri}} \neq 0$ at $t'/t < 0.214$ and $h_{\text{col}} \neq 0$ at $t'/t > 0.589$, and that the nonmagnetic disordered phase appears at $0.214 \leq t'/t \leq 0.589$. These results indicate that both of the magnetic phase transitions from the disordered phase are discontinuous, or of the first order, with respect to the parameter $t'/t$.

Figures 2 (c) and (d) show the calculated ground-state energies $E = \Omega + \mu$ (per site) of the $120^\circ$ ordered and collinear ordered phases compared with the energy of the disordered phase as a function of $t'/t$. We find that the critical point of the $120^\circ$ ordered phase is located at $t'/t = 0.214$ and that of the collinear ordered phase is located at $t'/t = 0.589$, and the disordered
Figure 2. Calculated results for (a) the grand potential (per site) as a function of the Weiss field $h_{\text{tri}}$, (b) that of the Weiss field $h_{\text{col}}$, (c) the ground-state energy (per site) of the $120^\circ$ ordered phase compared with that of the disordered phase as a function of $t'/t$, and (d) that of the collinear ordered phase. In (a) and (b), the stationary points are marked by black dots. In (c) and (d), the inset enlarges the region near the critical point.

Finally, let us calculate the magnetic order parameters of the $120^\circ$ ordered and collinear ordered phases in VCA. The order parameter $M$ may be defined as the expectation value of the one-body operator $O$ as

$$
\langle O \rangle = \frac{1}{L_c} \sum_{\alpha, \beta} O_{\alpha \beta} \left< c_i^\dagger c_i \right> = \frac{1}{L_c} \sum_{Q} \int_C \frac{dz}{2\pi i} \text{Tr} \left[ O G(Q, z) \right],
$$

where $G$ is the one-particle Green function $G = (G^{-1}_0 - \Sigma')^{-1}$ calculated in VCA and the path $C$ in the contour integrals encloses the poles of the integrand on the real axis below the chemical potential. The one-body operator for the $120^\circ$ ordered phase is defined as $O_{\text{tri}} = \sum_i e_i \cdot S_i$ for the three sublattices $\alpha_i$ (A, B and C) of the triangular lattice. The one-body operator for the collinear ordered phase is defined as $O_{\text{col}} = \frac{1}{2} \sum_i e_i^Q \cdot r_i (n_i^\uparrow - n_i^\downarrow)$ with $Q = (\pi, 0)$ or $(0, \pi)$.

Figure 3 shows the calculated ground-state phase diagram as a function of $t'/t$, where the order parameters $M_{\text{tri}}$ for the $120^\circ$ ordered phase and $M_{\text{col}}$ for the collinear ordered phase are given. The $120^\circ$ ordered phase appears at $0 \leq t'/t \leq 0.214$, which vanishes abruptly at $t'/t = 0.214$. The collinear ordered phase appears at $0.589 \leq t'/t \leq 1.0$, which vanishes abruptly...
Figure 3. Calculated ground-state phase diagram of the square-lattice Hubbard model at half filling with the anisotropic next-nearest-neighbor hopping parameters, where the order parameters (or sublattice magnetizations) $M_{\text{tri}}$ and $M_{\text{col}}$ are shown as a function of $t'/t$.

at $t'/t = 0.589$. The disordered phase then appears in-between. The phase transitions to the two magnetic phases from the disordered phase is thus of the first order.

Note that the present Hubbard model in the strong correlation limit may be mapped onto the frustrated spin-1/2 Heisenberg model defined on the same lattices with the exchange coupling constants of $J_{ij} = 4t^2_{ij}/U$ for the bonds with the hopping amplitude $t_{ij}$. Assuming $J = 4t^2/U$ and $J' = 4t'^2/U$ at $U/t = 60$, we predict that the $120^\circ$ ordered phase appears at $0 \leq J'/J \leq 0.045$, the collinear ordered phase appears at $0.347 \leq J'/J \leq 1.0$, and the nonmagnetic disordered phase appears in-between, i.e., at $0.045 \leq J'/J \leq 0.347$. We note that the metastable magnetic phases exist at $0.214 \leq t'/t \leq 0.239$ (or $0.045 \leq J'/J \leq 0.057$) for the $120^\circ$ ordered phase and at $0.502 \leq t'/t \leq 0.589$ (or $0.252 \leq J'/J \leq 0.347$) for the collinear ordered phase. The corresponding Heisenberg-model calculations have not been carried out as far as we know, which should be done in future and compared with the present results.

4. Summary
We have employed the method of VCA based on SFT to study the effects of frustration in the spin degrees of freedom on the magnetic orders of the square-lattice Hubbard model at half filling with the anisotropic next-nearest-neighbor hopping parameters at zero temperature. We have focused in particular on the strong correlation region of the model and have shown that the disordered phase appears between the collinear-ordered and $120^\circ$-ordered phases. We have also shown that the phase transitions to the two ordered phases from the disordered phase are of the first order.

Although we have restricted ourselves to the study of the Hubbard model at $t_1 = t_2$ in this paper, the full magnetic phase diagram of the $t_1 - t_2 - t'_2$ Hubbard model in its entire parameter space needs to be clarified for future study, including the presence of the spin-liquid phase caused by the frustration in the spin degrees of freedom.

Acknowledgments
This work was supported in part by a Grant-in-Aid for Scientific Research (No. 26400349) from JSPS of Japan. T K acknowledges support from the JSPS Research Fellowship for Young
Scientists.

References
[1] Miyagawa K, Kawamoto A, Nakazawa Y and Kanoda K 1995 Phys. Rev. Lett. 75 1174.
[2] Lefebvre S, Wzietek P, Brown S, Bourbonnais C, Jérôme D, Mézière C, Fournigue M, and Batail P 2000 Phys. Rev. Lett. 85 5420.
[3] Shimizu Y, Miyagawa K, Kanoda K, Maesato M and Saito G 2003 Phys. Rev. Lett. 91 107001.
[4] Manna R S, Souza de M, Brühl A, Schlueeter A J and Lang M 2010 Phys. Rev. Lett. 104 016403.
[5] Merino J, McKenzie R H, Marston J B and Chung C H 1999 J. Phys.: Condens. Matter 11 2965.
[6] Trumper A E 1999 Phys. Rev. B 60 2987.
[7] Sahebsara P and Sénéchal D 2008 Phys. Rev. Lett. 100 136402.
[8] Yoshioka T, Koga A and Kawakami N 2010 Phys. Rev. Lett. 103 036401.
[9] Yang H-Y, Läuchli A M, Mila F and Schmidt K P 2010 Phys. Rev. Lett. 105 267204.
[10] Morita H, Watanabe S and Imada M 2002 J. Phys. Soc. Jpn. 71 2109.
[11] Kyung B and Tremblay A-M S 2006 Phys. Rev. Lett. 97 046402.
[12] Koretsune T, Motome Y and Furusaki A 2007 J. Phys. Soc. Jpn. 76 074719.
[13] Clay R T, Li H and Mazumdar S 2008 Phys. Rev. Lett. 101 166403.
[14] Kokalj J and McKenzie R H 2013 Phys. Rev. Lett. 110 206402.
[15] Tocchio L F, Feldner H, Becca F, Valenti R and Gros C 2013 Phys. Rev. B 87 035143.
[16] Laubach M, Thomale R, Hanke W and Li G arXiv:1401.8198.
[17] Yamada A 2014 Phys. Rev. B 89 195108.
[18] Kaneko R, Morita S and Imada M 2014 J. Phys. Soc. Jpn. 83 093707.
[19] Potthoff M, Aichhorn M and Dahnken C 2003 Phys. Rev. Lett. 91 206402.
[20] Dahnken C, Aichhorn M, Hanke W, Arrigoni E and Potthoff M 2004 Phys. Rev. B 70 245110.
[21] Potthoff M 2003 Eur. Phys. J. B 32 429 and 36 335.
[22] Nevidomskyy A H, Scheiber C, Sénéchal D and Tremblay A-M S 2008 Phys. Rev. B 77 064427.