Origin of High-$T_c$ Superconductivity in Doped Hubbard Models and Their Extensions
– Roles of Uniform Charge Fluctuations –

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Doped Hubbard model is a simple model for the high-$T_c$ cuprate superconductors, while its ground state remains a challenge. Here, by performing state-of-the-art variational Monte Carlo calculations for the strong-coupling Hubbard model, we find evidences that the $d$-wave superconducting phase emerges always near the phase separation region and the superconducting order has one-to-one correspondence with the enhancement of charge compressibility. The order as well as the phase separation are vulnerable to realistic intersite Coulomb interaction while the superexchange interaction enhances both. An appropriate combination of these two widens the stable superconducting phase.

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The discovery of high-$T_c$ superconductivity in copper oxides [1] triggers studies of the superconductivity induced by the strong electronic correlations. After an enormous number of studies, the intrinsic phase diagram of the copper oxides is still not a completely resolved issue. Most of the superconducting copper oxides have the dome structure of the critical temperature $T_c$ as a function of the hole doping concentration $\delta$ centered at the optimum value $\sim 0.15$, after the quick disappearance of the antiferromagnetic order upon the doping to the Mott insulator of the mother materials. However, the multi-layer compound shows a wide coexistence region of the superconductivity and the antiferromagnetic order [2]. Recently the interface of La$_2$CuO$_4$/La$_{2-x}$Sr$_x$CuO$_4$, which is expected to realize purely two-dimensional superconductivity, has strikingly shown a pinning of $T_c$ at a constant value $\sim 40$K [3] in marked contrast with the dome structure in bulk, which supports that the intrinsic nature of the copper oxides is described by an extended region of the phase separation (PS), if the long-ranged Coulomb interaction is screened by the interlayer screening. At the interface, the phase separation may occur between layers. The intrinsic phase diagram of the copper oxides without impurity and long-ranged Coulomb effects is still an actively debated issue.

One of the most fundamental models to describe the high-$T_c$ superconductivity is the Hubbard model on the square lattice, which only considers the nearest-neighbor hopping $t$ and on-site Coulomb repulsion $U$ of electrons defined as follows:

$$H = -t \sum_{\langle \sigma \rangle} (c_{\sigma \alpha}^+ c_{\sigma \alpha} + \text{h.c.}) + U \sum_i n_{i \uparrow} n_{i \downarrow},$$

where we use the standard notation (see [4], S.1 for details of the models). A large number of theoretical works including analytical and numerical calculations have been devoted to the Hubbard model [5-10] (Detailed comparison of previous studies are shown in [4], S.2). Many of works suggest that the superconductivity appears near half band filling for sufficiently large $U/t$ [7-10]. However, numerically exact or high-precision calculations [3, 6, 10] do not necessarily show clear evidence of the high-$T_c$ superconductivity. Thus, the relation between strong electronic correlations and the high-$T_c$ superconductivity still remains an unresolved issue although there are many proposals for origin of the high-$T_c$ superconductivity [3, 5, 7-10].

In this Letter, we show a direct and quantitative one-to-one correspondence between superconductivity and enhanced uniform charge susceptibility, which clearly shows that the tendency for the PS is the origin of the $d$-wave superconductivity. The present result also offers an intriguing implication to the recent interface experiment [3]. We further reveal roles of intersite Coulomb repulsion $V$ that reduces both superconducting phase and uniform charge fluctuations as well as roles of superexchange interaction $J$ that enhances both of them.

To examine the origin of high-$T_c$ superconductivity in the Hubbard model [1], we employ many-variable variational Monte Carlo (mVMC) method [28] (for details and validity of the method, see [4], S.1 and S.3). This method enables us to perform high-precision calculations under spatial and temporal fluctuations of spin and charge on equal footings with a sufficient flexibility of wavefunctions, which are important in strongly correlated systems.

Figure 1 shows the doping dependence of several physical properties for $U/t = 10$; peak value of the spin structure factor $S(q_{\text{peak}})/N_s$, which is the square of the antiferromagnetic ordered moment, and average value of superconducting correlation $\langle D^2 \rangle$ at long distance with the $d_{x^2-y^2}$ symmetry, corresponding to the square of the superconducting order parameter. We also plot the condensation energy $\Delta E$. The details and definitions of physical quantities are shown in [4], S.1.

We find the $d_{x^2-y^2}$-wave superconducting phase only in the strong coupling region $U/t \gtrsim 6$, which is consistent
For previous studies\cite{5–7, 9, 11, 12, 15}, we note that the incommensurate spin orders or stripe phases are not found in the relevant doping region $\delta \lesssim 0.2$ even when we employ large sublattice structures. We also note that the charge structure factors have no significant peak at $q \neq 0$.

(b) Doping dependence of condensation energy $\Delta E$. The condensation energy is defined as $\Delta E = (E_{\text{SC}} - E_{\text{Normal}})/N$, where $E_{\text{SC}}$ ($E_{\text{Normal}}$) is the total energy of the superconducting phase (normal phase). The calculations are performed for sizes of $N_s = 12 \times 12$, $14 \times 14$, $16 \times 16$ on the square lattice, and we confirm that the finite-size effects are negligibly small. The shaded region denotes the PS region and the black dashed line represents the spinodal point. Details of PS are shown in the main text and Fig. 2. The superconducting phase without PS remains only in the yellow region. In the present plots and the plots in the later figures, the error bars indicate the estimated statistical errors of the Monte Carlo sampling (See S.1).

with previous studies\cite{5–7, 9, 11, 12, 15}. For instance, at $U/t = 10$, the $d$-wave superconductivity emerges for $\delta \lesssim 0.2$ as shown in Fig. 1. Both $\Delta E$ and $\overline{P}_{d_{2-z^2}}$ have dome structures around $\delta \sim 0.1$. The antiferromagnetic quantum critical point (AFQCP) where the antiferromagnetic spin fluctuations diverge, appears at $\delta \sim 0.18$. The $d$-wave superconductivity coexists with the antiferromagnetism in the ground state for $\delta \lesssim 0.18$. The coexistence has been theoretically studied before in several different contexts\cite{9, 11, 20, 30}. The coexistence is basically consistent with the multilayer cuprates\cite{2}, where the PS may be suppressed by the interlayer self-doping.

To examine the effects of charge fluctuations, the doping dependence of the chemical potential $\mu$ (see S.4, S.1 for the definition of $\mu$ and S.6 for the charge structure factor in PS region) is shown in Fig. 2, where the uniform charge susceptibility $\chi_c \equiv dn/d\mu$ monitors the charge fluctuation. The spinodal point of doping ($\delta_s$), where charge fluctuations diverge ($\chi_c^{-1} = 0$) is found to increase at larger $U$. Accordingly, the PS region becomes wider by increasing $U/t$. If we enforce the charge uniformity, superconducting correlation has the maximum around $\delta_s \sim 0.14$ (the spinodal point depicted by dashed black line in Fig. 1), for $U/t = 10$. This indicates that the enhanced charge fluctuations stabilize the superconducting phase around half filling.

However, if the long-range Coulomb interaction is suppressed as in the Hubbard model, the present result indicates that in a wide region of the nominal doping concentration, the system undergoes a real-space PS into the antiferromagnetic Mott insulator and the superconducting region with the pinned $T_c$. This prediction is in striking agreement with the recent interfacial superconductivity\cite{3}.

Here, to control the charge fluctuations, we introduce nearest-neighbor Coulomb interactions $V$ ($H_V = V \sum_{i,j} n_i n_j$), which indeed inevitably exit in real materials (see also S.4, S.1 for details of interac-
In reality, can be induced by the \( J \) notations are the same as Fig. 1. Single band framework \([31–33]\). Hybridizations in cuprate superconductors beyond the \( p \) significantly enhanced superconducting phase may be un-

ished, while the antiferromagnetic order of the PS, which corroborates this conclusion and \( V/t \) up to 20×20 lattices. Notations are the same as Fig. 4.

It is also an intriguing issue to examine whether the instability toward the phase separation at the wavenumber \( q = 0 \) can be converted into the instability toward charge ordering at nonzero \( q \) observed in some cases of the cuprates by employing a realistic off-site Coulomb interactions.

To further understand the interplay of spin fluctuations and the instability toward the PS, by keeping \( V = 0 \), we introduce the nearest-neighbor superexchange interactions \( J (H_J = J \sum_{(i,j)} S_i \cdot S_j, \text{ see } [1], S.1 \) for details of interactions) that does not follow the standard relation \( J_{\text{eff}} \sim 4t^2/U \). In reality, \( J \) can be induced by the \( d-p \) hybridizations in cuprate superconductors beyond the single band framework [31,33].

As illustrated in Fig. 4(a), finite \( J/t = 0.5 \) largely enhances the PS region, while the antiferromagnetic order does not change appreciably. Accompanied by the enhanced charge fluctuations, the condensation energy becomes an order of magnitude larger. Because the AFQCP is close to the spinodal point as shown in Fig. 4(b), this significantly enhanced superconducting phase may be understood from the synergetic effects of spin and charge fluctuations. We later emphasize the importance of short-ranged fluctuations. However, anyway, this phase is again preempted by the PS.

In addition to \( J/t \), we again add \( V \). As we see in Fig. 4(a), by increasing \( V/t \), locations of AFQCP do not change appreciably, while locations of the spinodal point rapidly approach half filling. In connection with the suppressed charge fluctuations, the condensation energy is significantly reduced, again suggesting the key role of the proximity of the PS in establishing high-\( T_c \) superconductivity. However, it is remarkable that, for the coexisting \( J \) and \( V \), the superconducting phase with a substantial condensation energy survives in a wide range (0.1 \( \lesssim \delta \lesssim 0.3 \) for \( V/t = 2 \)) outside the PS region.

The large condensation energy is ascribed mainly to two local sources: One is that the double occupancy \( D \) is
FIG. 5: (color online). Relation between the peak value of the superconducting correlation $\max(\bar{P}_{d_{z^2-r^2}})$ and the strength of enhancement of charge fluctuations characterized by $b$ in various cases, where $b$ is defined from $\chi_c^{-1} = a + b\delta$. Inset: Doping concentration dependence of $20\times \bar{P}_{d_{z^2-r^2}}$ (curves with symbols) and $0.1\times \chi_c^{-1}$ (lines passing crosses) for two examples with offset in the ordinate for clarity. The crosses represent the spinodal point $\chi_c \rightarrow \infty$. The slope of linear fitting of $\chi_c^{-1}$ gives $b$. The notation for the shaded zone is the same as Fig. II.

largely reduced in the superconducting phase than that in the normal phase, which leads to the gain in the onsite Coulomb energy. This is because, the $d$-wave pair prohibits the double occupation strictly by symmetry, which is particularly effective when $D$ remains not small in the normal phase (as around $\delta \sim 0.1$) (Fig. S6 in S.4 of [1] shows how the reduced $D$ in the superconducting state enhances $\Delta E$.) This mechanism cannot be captured by the $t$-$J$ model. If $J > 0$, the other source is the antiferromagnetic correlation $S_i \cdot S_j$: The superconducting order enhances the underlying nearest-neighbor “antiferromagnetic” correlations even when $J = 0$, which provides the energy gain immediately when $J > 0$ (See [1], S. 4). The long-range part of antiferromagnetic correlation does not directly contribute to this gain.

The strong coupling nature of high-$T_c$ superconductivity emerges not from the long-ranged part and the quantum criticality but rather from the local binding, as expected when approaching the regime of BEC. This local attractive interaction leads to Cooper pairing but does not necessarily lead to PS. This is because the PS signaled by the convex curve with a peak structure in the chemical potential as in Fig. 2 is mainly caused by the contribution of the kinetic energy part in the chemical potential, which is evidenced in Fig.S 8 of [1]. This peak in the kinetic energy is efficiently suppressed by $V$ rather independently of the emergence of the local attractive interaction. While $V$ suppresses PS, some choices of $V$ and $J$ largely strengthen the energy gain from $D$ because of the enhanced $D$ in the normal state. This is the reason why an appropriate combination of $V$ and $J$ stabilizes the high-$T_c$ superconductivity without PS in an extended region. It implies that the superconducting stability is not a universal property but largely relies on material details. It requires a reexamination of the conditions for the emergence of high-$T_c$ superconductivity. The necessity of both $V$ and $J$ also requires careful analyses how they are derived quantitatively from first principles.

To see relation between enhanced uniform charge fluctuations and the stability of superconductivity, we plot in Fig. III the relation between the maximum value of $\bar{P}_{d_{z^2-r^2}}$ and the measure for the enhanced charge fluctuation $b$ defined by $\chi_c^{-1} \equiv (d^2 E/d\delta^2)^{-1}$ diverges at $a + b\delta = 0$, which represent the spinodal point and the charge fluctuation becomes more enhanced in more wide-spread doping region for smaller $b$. The relation shows that a clear correlation between $\max(\bar{P}_{d_{z^2-r^2}})$ and $b$, indicating that the enhanced charge fluctuation stabilizes the superconductivity. In the inset, we plot the doping concentration dependence of the superconducting correlation and $\chi_c^{-1}$ for two typical examples. In addition to the correspondence between $\max(\bar{P}_{d_{z^2-r^2}})$ and $b$, in all the cases we studied, the peaks of $\bar{P}_{d_{z^2-r^2}}$ are located at the concentrations close to the spinodal points (crosses), indicating the one-to-one correspondence between $\bar{P}_{d_{z^2-r^2}}$ and the charge fluctuation.

To summarize, the origin of the high-$T_c$ superconducting phase in the doped Hubbard model is found primarily as arising from the phase separation instability. Controlling the charge fluctuation through the off-site interactions possibly by tuning dielectric constant offers a possible way to stabilize the high-$T_c$ superconducting phase. Though it is not so easy, an interesting future issue is to find a way to suppress the ratio of the off-site to on-site interactions by keeping a large on-site interaction in real materials with the help of $ab$ initio calculations [3]. In this respect, the recently studied interfacial superconductivity [3] offers a promising way and supports the relevance of the present phase diagram with an extended region of PS as the genuine one if the long-ranged Coulomb interaction is screened on a single layer by the capacitor formation with the neighboring metallic layers.

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Supplemental Materials

S.1 MODEL, METHOD, AND DEFINITIONS OF PHYSICAL QUANTITIES

We employ the standard Hubbard model on the square lattice, defined by the Hamiltonian

\[ H = -t \sum_{\langle i,j \rangle, \sigma} (c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.}) + U \sum_i n_{i\uparrow} n_{i\downarrow}, \]

where \( c_{i\sigma}^\dagger \) (\( c_{i\sigma} \)) is the creation (annihilation) operator on the \( i \)-th site with spin \( \sigma \) and \( n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma} \) is the number operator. The transfer integral \( t \) is only taken for nearest-neighbor sites. We take \( N_c = L \times L \) sites with periodic-periodic (PP) and antiperiodic-periodic (AP) boundary conditions. We define the doping rate \( \delta \) as \( \delta = 1 - N_c/N_s \), where \( N_s = \sum_{i,\sigma} n_{i\sigma} \). We add the off-site Coulomb and superexchange interactions defined as

\[ H_V = V \sum_{\langle i,j \rangle} n_{i\uparrow} n_{j\downarrow}, \]

\[ H_J = J \sum_i S_i \cdot S_j, \]

where \( S_i = 1/2 \sum_{\sigma,\sigma'} c_{i\sigma,\sigma'}^\dagger \sigma,\sigma' c_{i\sigma,\sigma'} \) and \( n_i = n_{i\uparrow} + n_{i\downarrow} \).

To study the ground-state of the doped Hubbard model, we employ a many-variable variational Monte Carlo (mVMC) method combined with the quantum-number projection. Our variational wave function is defined as

\[ |\psi\rangle = P_G P_J P_{d-h}^\text{ex} L^{K=0} L^{S=0} |\phi_{\text{pair}}\rangle, \] (S.1)

where \( P_G, P_J, P_{d-h}^\text{ex} \) are the Gutzwiller [1], Jastrow [2, 3], and doublon-holon correlation factors [4], respectively. The Gutzwiller factor punishes the double occupation of electrons on the same site through the variational parameters \( g \) defined as

\[ P_G = \exp(-g \sum_i n_{i\uparrow} n_{i\downarrow}). \]

The Jastrow factors are defined as

\[ P_J = \exp\left(-\frac{1}{2} \sum_{i,j} v_{ij} n_{i\sigma} n_{j\sigma}\right), \]

where the long-range part drives the distinction between the metal and insulator [5]. The doublon-holon correlation factors [4] are defined as

\[ P_{d-h}^\text{ex} = \exp\left[-\sum_{m=0}^{\ell=1,2} \sum_i \xi_{(m)}^{(\ell)} \sum_i \xi_{(m)}^{(\ell)}\right], \]

where \( \xi_{(m)}^{(\ell)} \) is a many-body operator which is diagonal in real-space representations. When a doublon (holon) exists at the \( i \)-th site and \( m \) holons (doublons) surround at the \( \ell \)-th nearest neighbor, \( \xi_{(m)}^{(\ell)} \) gives 1. Otherwise, \( \xi_{(m)}^{(\ell)} \) gives 0. The spin (momentum) quantum number projection operator \( L^{S=0} (L^{K=0}) \) restores SU(2) spin symmetry (translational symmetry) with the total spin \( S = 0 \) (total momentum \( K = 0 \)). These projections substantially improve the accuracy of cluster properties, make the size dependence smaller and the extrapolation to the thermodynamic limit easier [6].

The one-body part \( |\phi_{\text{pair}}\rangle \) is the generalized pairing wave function defined as

\[ |\phi_{\text{pair}}\rangle = \left[ \sum_{i,j=1}^{N_s} f_{ij} \xi_{i\uparrow}^{(1)} \xi_{j\downarrow}^{(1)} \right]^{N_s/2} |0\rangle, \] (S.2)

where \( f_{ij} \) denotes the variational parameters (Details of \( f_{ij} \), see [4, 7]). In this study, we allow \( f_{ij} \) to have \( 2 \times 2 \) sublattice structure or equivalently we have \( 2 \times 2 \times N_s \) independent variational parameters for one-body part. All the variational parameters are simultaneously optimized by using the stochastic reconfiguration method [5, 8]. The variational function \( |\psi\rangle \) in Eq. (S.1) can flexibly describe paramagnetic metals, the antiferromagnetic phase, and superconducting phases as well as their fluctuations and/or coexistence. It is important to fully optimize the long-range part of \( f_{ij} \) to realize states with strong fluctuations and well-developed short-ranged order as well as strongly renormalized metals as we detail later. Actually, by extending the \( 2 \times 2 \) sublattice structures of the variational parameters \( f_{ij} \), we confirmed that the accuracy of the energy is improved. Furthermore, by applying the power Lanczos method [9], we can also substantially improve the energy. However, through the careful examination of such extensions, we confirmed that estimates of the physical properties (superconducting correlations, antiferromagnetic correlations, etc.) change little. In addition, numerical cost of such extensions is demanding. Therefore, to perform the comprehensive calculations for the doped Hubbard with additional intersite interactions, we have used the present tractable variational wave functions. Nevertheless, we again emphasize that the estimates of the physical properties themselves are accurate enough and our conclusions do not change.

To discuss the condensation energy, we generate two different wave functions, i.e., normal and superconducting wave functions by choosing proper initial states. We employ the non-interacting Fermi sea for the normal state, and BCS \( d \)-wave superconductivity state for superconducting phase as the initial states [5]. By optimizing these initial states, we obtain normal and superconducting states. In the strong coupling region, the antiferromagnetic order appears near half filling as the normal state as a result of the optimization, although we do not assume the antiferromagnetic order as an initial state, which means that the paramagnetic normal state is unstable.
To determine the ground state of the doped Hubbard model, we calculate spin structure factor and equal-time superconducting correlation. The spin structure factor is defined as

$$S(q) = \frac{1}{3N_s} \sum_{i,j} \langle S_i \cdot S_j \rangle e^{iq(r_i-r_j)}$$,

and the equal-time superconducting correlations are defined as

$$P_\alpha(r) = \frac{1}{2N_s} \sum_{r_i} (\Delta^\dagger_\alpha(r_i) \Delta^\dagger_\alpha(r_i + r) + \Delta_\alpha(r_i) \Delta^\dagger_\alpha(r_i + r))$$.

In actual calculations, to reduce numerical cost, we restrict summation with respect to $r_i$ to $r_i = 0$. Superconducting order parameters $\Delta_\alpha(r_i)$ are defined as

$$\Delta_\alpha(r_i) = \frac{1}{\sqrt{2}} \sum_r f_\alpha(r) (c_{r_i \uparrow} c_{r_i + r \downarrow} - c_{r_i \downarrow} c_{r_i + r \uparrow})$$.

Here, $f_\alpha(r)$ is the form factor that describes the symmetry of the superconductivity. For $d_{x^2-y^2}$ superconductivity, we define

$$f_{d_{x^2-y^2}}(r) = \delta_{r_y,0}(\delta_{r_x,1} + \delta_{r_x,-1}) - \delta_{r_y,0}(\delta_{r_y,1} + \delta_{r_y,-1})$$,

where $\delta_{i,j}$ denotes the Kronecker’s delta and $r = (r_x, r_y)$. We define long-range average of the superconducting correlation as

$$\bar{P}_{d_{x^2-y^2}} = \frac{1}{M} \sum_{2<r=|r|<L-1} P_{d_{x^2-y^2}}(r)$$,

where $M$ is the number of vectors satisfying $2 < r < L - 1$. As shown in Fig. S.1, the criterion $r > 2$ is, within the present purpose, practically a sufficient probe to see whether the pairing order-parameter correlation is saturated to a nonzero value and offers a good measure for the square of the order parameter in the long-range ordered superconducting state.

We also calculate the chemical potential by using the relation

$$\mu(\bar{N}) = \{E(N_1) - E(N_2)\}/\{N_1 - N_2\}$$,

$E(N_1)$ is the total energy at filling $N_1$ and $\bar{N} = (N_1 + N_2)/2$. To reduce the finite-size effects, we perform calculation only at the electron densities that satisfy the closed-shell condition in the non-interacting case.

The nonzero condensation energy $\Delta E = (E_{SC} - E_{Normal})/N_s$ is defined when the superconducting (with energy $E_{SC}$) and normal states ($E_{Normal}$) exist as local minima. The normal state is not necessarily the paramagnetic state but can be another symmetry broken state such as the antiferromagnetically ordered state, if it has a lower energy than the paramagnetic state. It is remarkable that in the present calculation, if the superconducting state with a nonzero order parameter exists, it always has a normal state as local minima as well. The transition from the normal to the superconducting states by reducing the doping concentration from the overdoped region is always a weak first-order transition where the superconducting order parameter jumps from zero to a small nonzero value in the ground state. For instance, at $(V/t = 0, J/t = 0)$, $(V/t = 0, J/t = 0.5)$, $(V/t = 1, J/t = 0.5)$ and $(V/t = 2, J/t = 0.5)$, the superconducting state emerges as a metastable state at $\delta \sim 0.25, 0.33, 0.29, 0.28$ while it becomes the ground state only for $\delta \leq 0.22, 0.31, 0.27, 0.28$, respectively. The first-order jump decreases with the increase in $V/t$ suggesting an existence of the tricritical point at around $(V/t = 2, J/t = 0.5)$. Toward half filling, the order parameter of the superconducting state looks continuously going to zero, which is connected to the antiferromagnetic Mott insulator. Here, again the non-superconducting state continues to exist as a metastable excited state.

In connection with the experimental measurement of the condensation energy by the specific heat or the upper critical field, the present definition is not exactly identical to each other because the normal state in the experiment usually excludes the magnetic order as the normal state, for instance. This means that the experimental value overestimates the true condensation energy. However, the present definition certainly gives more useful criterion to determine whether the superconducting state is the true ground state or not.

The normal state is defined as the state that has vanishing superconducting order within the numerical accuracy. It does not exclude the possibility of a state with a tiny order parameter expected from the Kohn-Luttinger mechanism. In addition, the normal state we ob-
tained has a robust and developed superconducting correlation with the extended s-wave order parameter with the form factor \( \cos k_x + \cos k_y \), which scales to zero in the thermodynamic limit within the numerical accuracy.

Monte Carlo sampling of real space configurations of the electrons is employed to calculate physical quantities following the standard procedure [3]. The number of Monte Carlo samples for the calculation of physical quantities is typically 128 000. The statistical error of the Monte Carlo sampling estimated from a number of independent computations is indicated in the last parentheses in the numerical data as well as error bars in the plots in figures.

### S.2 COMPARISON WITH PREVIOUS STUDIES

In table S.1 we summarize the previous numerical studies on the doped Hubbard model. We summarize estimates of \( T_c \), region of superconducting (SC) phase, and antiferromagnetic (AF) phase. We also summarize information on phase separation (PS) and condensation energy \( \Delta E \).

In the first column, the results of the present study is summarized.

In the second column, we show several Monte Carlo i.e. auxiliary-field quantum Monte Carlo (QMC) and Gaussian-basis quantum Monte Carlo (GBMC) as well as path-integral renormalization group (PIRG) calculations. We note that these methods do not restrict the form of the wavefunction a priori and give the accurate estimates of the energy among various numerical schemes, if the interaction is from weak to intermediate coupling region \( U/t \lesssim 6 \). The accuracy of the PIRG has been benchmarked to be accurate [32] and applied to various cases [22, 24]. The GBMC has been benchmarked with the pre-projection method [14], which substantially relaxes the limitation and eliminates the origin of the errors (boundary terms) [35] and then gives good agreement with the QMC results. The (high-\( T_c \)) superconducting phase does not appear in the region of \( U/t \lesssim 6 \) in all of these methods. The absence is consistent with the present nVMC result, i.e., we confirm that the superconducting phase is not stabilized for \( U/t \lesssim 6 \) as shown in the first column. This is consistent with some other results [22, 24] as well as the CPMC studies [20].

At \( U/t = 4 \), the divergence of the compressibility is suggested at \( \delta \sim 0 \) [10, 29], which means that the phase separation is absent but the system is on the marginal quantum critical point [36, 37]. The absence of the phase separation or restriction at most to a tiny region \( \delta < 0.06 \) [10, 11, 12, 29, 32] is well consistent with the present study. The phase separation is clearly observed in a wide region of the doping concentration in the present study for the strong coupling region \( U/t > 6 \), which has not been well studied before in the quantitatively accurate methods.

In the third column, we mainly show the results obtained by dynamical mean-field theory (DMFT) calculations with cluster extension such as dynamical cluster approximation (DCA) and cellular DMFT (CDMFT). We also show the results of variational cluster approximations (VCA).

All of these works suggest that the \( d \)-wave superconducting phase appear around \( \delta \sim 0.1 \). The absence of the superconductivity for \( U \leq 6 \) observed in the present study is not consistent with DMFT and its extensions [12, 17, 21], which may be attributed to the overestimate of the superconductivity in DMFT because of the mean-field approximation. We note that the presence or absence of the superconductivity is determined only by the long-ranged part of the pairing correlation, while such spatial correlations and fluctuations are not captured by the DMFT.

Some works suggest that the first-order phase transition between two metal phases occurs, i.e., phase separation occurs between metals [12]. This type of phase separation is only found in DMFT calculations and not observed in other calculations such as VMC and constrained-path Monte Carlo (CPMC) as shown in the fourth and fifth columns.

In the fourth column, we show several previous VMC calculations. In the previous VMC calculations, the form of wave functions is limited and they use different wavefunctions to describe the Fermi liquid, antiferromagnetic phase, \( d \)-wave superconducting phase, and their coexistence phase [21], respectively. We obtain typically 5% lower energy compared to early VMC results [22]. For example, for \( U/t = 10 \), \( L = 10 \), \( \delta = 0.88 \), and AP boundary conditions, Yokoyama et al. [22] obtain \( E/N_s \sim -0.60t \) while we obtain \( E/N_s \sim -0.625t \). Recent VMC studies implemented a number of additional improvements to reach better accuracy, [24, 31] which are comparable to the present study in energy. In contrast to most of earlier studies, we employ flexible one-body part of the wave functions defined in Eq. (S.2). By optimizing the long-range part of \( f_{ij} \), this wave function can describe from insulators to antiferromagnetic metals, superconducting phases, strongly correlated metals and their competitions/coexistence on an equal footing in a single framework. It is important for VMC results to benchmark the accuracy by comparing with the available accurate results obtained without assuming biased forms of wavefunctions as those listed in the second column. By comparing with established results, we show in S.3 that our wave functions allow precise estimations of physical properties.

In the fifth column, the results of VMC and CPMC methods, which mainly study the normal state properties and instability toward PS, are shown. Neuscamman et al. [23] have used a variational wave function with a large number of variational parameters, which is similar
to ours. However, their estimate of the phase separation region in the doped Hubbard model extends to a larger doping concentration $\delta \sim 0.15$ even at $U/t = 4$. This contradicts other and present estimates. The reason for the overestimate of the phase separation in Ref. [25] is not clear enough for the moment. The CPMC studies also suggested the phase separation up to $\delta \sim 0.1$ at $U/t = 4$ [27] (or incommensurate antiferromagnetic order instead [28]). This has been criticized in Ref. [29] by taking into account the coexisting antiferromagnetic and BCS guiding functions, which give more or less the absence of the phase separation. Many works including numerically exact method such as QMC suggest that PS does not occur in the weak coupling region ($U/t \lesssim 8$) and our present work is consistent with them. Although Becca et al. [30] claim that PS does not occur even in the strong coupling region ($U/t = 10$) from the result of charge structure factor by using Green-function Monte Carlo (GFMC) method, the charge structure factor is not a proper quantity to detect the PS as we will show in S. 6. For the case with the next-neighbor-hopping $t' = -0.4t$, the phase separation is observed at strong coupling $U/t = 10$ [31].

Our result on the PS is consistent with the most of the former studies where the PS occurs in the strong coupling region. However, the relation between the PS and the superconductivity clarified as a key in the present work has not been well studied in the literatures.

Here, we mention about the previous studies on the extended Hubbard model. In the strong coupling region, effects of intersite interactions such as $V/t$ and $J/t$ are studied by using VMC and CDMFT [32, 40]. They showed that the intersite Coulomb interaction $V$ reduces the superconducting order parameter. However, they do
not study the competitions with other phases such as antiferromagnetic phase. Thus, it is not clear whether the superconducting phase is robust against intersite Coulomb interactions. By performing the high-precision calculations that treat the superconducting phase and antiferromagnetic phase or strongly correlated metal on an equal footing, we show that the superconducting phase becomes unstable for small $V (V/U = 0.1)$. This fragility of superconducting phase is not clarified in previous studies. In addition, we again note that the CDMFT often overestimates the stability of the superconducting phase because of its mean-field nature.

**S.3 BENCHMARK OF PRESENT MVMC METHOD**

To show the accuracy of the present mVMC method, we compare our results with those of the exact diagonalization (ED), auxiliary-field QMC, and GBMC for the Hubbard model on the square lattice, since they provide us in general with the best estimates of the energy as well as other physical properties. A weak point of the QMC and GBMC methods are that they are applicable only in the region up to the intermediate coupling. However, they give accurate energies and physical properties and are useful for the benchmark. In fact, the QMC is a numerically exact method within the statistical error and the GBMC is well established to give very good results of the spin structure factor are also well consistent with the QMC and GBMC calculations. These results also confirm the accuracy of our mVMC method. In large systems, our mVMC offers consistent results with the QMC and GBMC calculations. These results also confirm the accuracy of our mVMC method. We also note that the accuracy and applicability of the mVMC method in general have also been examined in the literature.[4][14][43].

**S.4 DETAILS OF CONDENSATION ENERGY**

In this section, we show the details of condensation energy, i.e., kinetic-energy gain $\Delta E_{\text{kin}}$ and potential-energy

| Physical Properties | mVMC(2 × 2) | ED |
|---------------------|-------------|----|
| $4 \times 4$ (PP), $n = 1$ | | |
| Energy per site | -0.8500(1) | -0.8513 |
| $S(q_{\text{peak}})/N_s$ | 0.0575(2) | 0.0569 |
| $q_{\text{peak}}$ | $(\pi, \pi)$ | $(\pi, \pi)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.2063(14) | -0.2063 |

| $4 \times 4$ (PP), $n = 0.625$ | | |
| Energy per site | -1.2190(1) | -1.2238 |
| $S(q_{\text{peak}})/N_s$ | 0.0130(1) | 0.01300 |
| $q_{\text{peak}}$ | $(\pi/2, \pi)$ | $(\pi/2, \pi)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.0704(5) | -0.0683 |

| $4 \times 4$ (AP), $n = 1$ | | |
| Energy per site | -0.9081(1) | -0.9120 |
| $S(q_{\text{peak}})/N_s$ | 0.0414(1) | 0.039698 |
| $q_{\text{peak}}$ | $(\pi, \pi)$ | $(\pi, \pi)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.1591(8) | -0.1537 |

| $4 \times 4$ (AP), $n = 0.75$ | | |
| Energy per site | -1.1504(1) | -1.1607 |
| $S(q_{\text{peak}})/N_s$ | 0.0179(2) | 0.0179 |
| $q_{\text{peak}}$ | $(\pi/2, \pi)$ | $(\pi/2, \pi)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.0944(7) | -0.0936 |

Table S II: $[U/t = 4]$ Comparison of Energy, peak value of spin structure factor $S(q_{\text{peak}})$, its wavenumber $q_{\text{peak}}$, and nearest-neighbor spin correlation $\langle S_i \cdot S_j \rangle$ between the exact diagonalization (ED) results and those of mVMC, where mVMC(2 × 2) means that the number of the variational parameters for $f_{ij}$ is $2 \times 2 \times N_s$. The parentheses denote the error bars in the last digit.
Table S III: $[U/t=10]$ Comparison of Energy, peak value of spin structure $S(q_{\text{peak}})/N_s$, its wavenumber $q_{\text{peak}}$, and nearest-neighbor spin correlation $\langle S_i \cdot S_j \rangle$. The method is the same as Table III. The parentheses denote the error bars in the last digit.

| Physical Properties | mVMC (2 x 2) | ED |
|---------------------|-------------|----|
| $4 \times 4$ (PP), $n = 1$ |            |    |
| Energy per site     | -0.43632(5) | -0.43931 |
| $S(q_{\text{peak}})/N_s$ | 0.0860(3) | 0.0835 |
| $q_{\text{peak}}$     | $(\pi, \pi)$ | $(\pi, \pi)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.3010(9) | -0.3057 |
| $4 \times 4$ (AP), $n = 1$ |            |    |
| Energy per site     | -1.0444(3) | -1.0564 |
| $S(q_{\text{peak}})/N_s$ | 0.01505(7) | 0.01508 |
| $q_{\text{peak}}$     | $(\pi/2, \pi)$ | $(\pi/2, \pi)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.0818(5) | -0.0754 |
| $4 \times 4$ (AP), $n = 1$ |            |    |
| Energy per site     | -0.9022(3) | -0.9255 |
| $S(q_{\text{peak}})/N_s$ | 0.0261(3) | 0.0216 |
| $q_{\text{peak}}$     | $(\pi, 0)$ | $(\pi, \pi/2)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.1087(15) | -0.1073 |
| $4 \times 4$ (AP), $n = 1$ |            |    |
| Energy per site     | -0.4422(1) | -0.4457 |
| $S(q_{\text{peak}})/N_s$ | 0.0852(2) | 0.0819 |
| $q_{\text{peak}}$     | $(\pi, \pi)$ | $(\pi, \pi)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.2994(17) | -0.3044 |
| $4 \times 4$ (AP), $n = 1$ |            |    |
| Energy per site     | -0.9232(3) | -0.9555 |
| $S(q_{\text{peak}})/N_s$ | 0.0261(3) | 0.0216 |
| $q_{\text{peak}}$     | $(\pi, 0)$ | $(\pi, \pi/2)$ |
| $\langle S_i \cdot S_j \rangle$ | -0.1087(15) | -0.1073 |

Table S IV: Comparison of total energy between mVMC results and those of numerically well benchmarked accurate methods. The parentheses denote the error bars in the last digit.

| QMC | GBMC | mVMC |
|-----|------|------|
| $8 \times 8$ (PP), $n = 50/64$ | $U/t = 4$ | -72.80(6) | -72.51(5) | -71.417(4) |
|      | $U/t = 6$ | -63.64(12) | -62.553(9) |
| $10 \times 10$ (PP), $n = 82/100$ | $U/t = 4$ | -109.7(6) | -107.51(1) |
|      | $U/t = 6$ | -92.07(22) | -91.91(1) |
| $12 \times 12$ (PP), $n = 122/144$ | $U/t = 4$ | -151.4(14) | -150.14(2) |

We also show the nearest-neighbor spin correlation $\Delta S$, which is defined as

\[ S_{nn} = \langle S_i \cdot S_j \rangle, \]

\[ \Delta S = (S_{nn,SC} - S_{nn,\text{Normal}}), \]  

where $i$ and $j$ represent the nearest neighbor sites.

Fig. S 2: Distance dependence of $d_{x^2-y^2}$-wave superconducting correlation $P_{d_{x^2-y^2}}(r)$ at $n = 10/16 = 0.625$ for PP boundary condition. For $U/t = 4$ and $U/t = 10$, mVMC well reproduces the exact values. In the present plots and the plots in the later figures, the error bars indicate the estimated statistical errors of the Monte Carlo sampling (See S. 1).

Fig. S 3: Superconducting correlation $P_{d_{x^2-y^2}}(r)$ for $d_{x^2-y^2}$-wave symmetry as a function of distance $r$ at $n = 12/16 = 0.75$ for $4 \times 4$ lattice with AP boundary condition. For both $U/t = 4$ and $U/t = 10$, mVMC well reproduces the exact values (ED).
and the loss in the kinetic energy in the superconducting state in comparison to the state with the antiferromagnetic correlations only. Because the energy gain arising from the short-range singlet correlation exists for finite $J$, total condensation energy becomes large compared to the simple Hubbard model. As shown in Fig. S4, short-range singlet correlation does not largely depend on interaction parameters.

In Fig. S5 we show the kinetic (potential) part of chemical potential $\mu_{\text{kin}} (\mu_U)$ for $U/t = 10$, defined as

$\mu_{\text{kin}}(\tilde{N}) = \{E_{\text{kin}}(N_1) - E_{\text{kin}}(N_2)\}/\{N_1 - N_2\}$,

$\mu_U(\tilde{N}) = \{E_U(N_1) - E_U(N_2)\}/\{N_1 - N_2\}$,

where $\tilde{N} = \{N_1 + N_2\}/2$. Kinetic part of chemical potential shows the convex doping dependence, while $\mu_U$ is nearly independent of the doping. This convex doping dependence of $\mu_{\text{kin}}$ suggests that PS is mainly caused by the kinetic energy.

A strong crossover from the states with the Mott proximity in the underdoped region to the overdoped region takes place in two-fold way: One is the charge instability represented by divergence of charge compressibility at $\delta = \delta_s$. The other is the magnetic instability repre-
In this section we examine the effects of the next-nearest-neighbor hopping. To directly compare with the case of $t'/t = 0$, we employ the same onsite Coulomb repulsion, i.e., $U/t = 10$. When the next-nearest-neighbor hopping $t' = -0.3t$ is present following the realistic parameter of the cuprate superconductors, the condensation energy is strongly suppressed as we see in Fig. S7(a). Concomitantly with this suppression, the phase separation also disappears as we see Fig. S7(b). The results are not well consistent with the experimental results of the hole doped copper oxides expected from the material dependence of the parameters in the following points: (1) The suppression of the superconductivity at larger $-t'/t$ does not follow the relation between the expected material dependence of $t'/t$ and the critical temperature $T_c$ [43]. (2) Wide antiferromagnetically ordered region is not consistent with a quick destruction of the antiferromagnetic order upon hole doping. The origin of the discrepancy is not clear at the moment. Possible origins are the following: (1) Realistic value of the onsite Coulomb repulsion is smaller than the present value $U/t = 10$. (2) A combination of $V$ and $J$ expected in the effective low-energy model is required to stabilize the superconductivity. (3) Single band models are not sufficient to reproduce the quantitative aspect of the copper oxides. (4) Small but finite impurities immediately destroy the antiferromagnetic order.

**S.5 RESULTS WITH NEXT-NEAREST-NEIGHBOR HOPPING $t' = -0.3t$**

In this section, we estimate the amplitude of the charge structure factor allowed in finite size systems when the phase separation occurs as a macroscopic phase. In the canonical ensemble, the charge structure factor $N(q) = \frac{1}{N_s} \sum_{i,j} \langle n_i n_j \rangle e^{i q \cdot (r_i - r_j)}$, at $q = 0$ must be zero because total charge should be conserved, while one may expect the growth of $N(q)$ at the lowest possible wavenumber as the signature of the Bragg peak at $q = 0$ expected for the phase separation region. However, we here show that the growth is in practice suppressed by the energy loss caused by the domain wall formation in numerically...
Here, we first roughly estimate the energy cost caused by the density modulation imposed in a metal with the period of system size (namely at the nonzero and lowest possible wavenumber in the periodic boundary condition) to simulate the energy cost by the domain wall formation between two different density phases. (Note that this estimate is valid if the density modulation from the uniform phase is small, which is justified later.) For this purpose, we consider the non-interacting Hamiltonian $H_0 = \sum_{k, \sigma} \epsilon_k c_k^\dagger c_k \sigma$, where $k$ is momentum vector and $\epsilon_k$ is band dispersion, respectively. The ground state of this Hamiltonian is Fermi-sea state (with of course uniform density), which is defined as $|\phi_0\rangle = \prod_{|k| < k_F, \sigma} c_{k\sigma}^\dagger |0\rangle$, where $k_F$ is Fermi wavenumber. Here, we calculate the energy loss in the charge-modulated (CM) phase $|\phi_{CM}\rangle$, which is defined as

$$|\phi_{CM}\rangle = \hat{\rho}|\phi_0\rangle, \quad \hat{\rho} = 1 + \gamma \hat{n}_q,$$

where $q$ is the wavenumber of charge modulation. For simplicity, we consider square lattice $[\epsilon_k = -2t'\cos k_x + \cos k_y]$, $q = (q_x = 2\pi/L, 0)$ ($L$ is the linear dimension of system), and half filling (see Fig. S11(a)). Note that $q$ is the lowest possible wavenumber of the density modulation. First, we calculate the local density at site $l$ as follows:

$$\langle c_l^\dagger c_l \rangle = \langle \phi_{CM}| c_l^\dagger c_l |\phi_{CM}\rangle / \langle \phi_{CM}|\phi_{CM}\rangle$$

$$= N_e / 2N_s + 2\gamma \cos \frac{2\pi l}{L} \left(1 + M|\gamma|^2\right),$$

where $N_e$ is number of total electrons and $M = \langle \phi_0| \hat{n}_q^\dagger \hat{n}_q |\phi_0\rangle = \sum_{k \in R, \sigma} \sim 2L$ (definition of $R$, see Fig. S11(a)). Therefore, by assuming $M|\gamma|^2 \ll 1$, amplitude of charge modulation $\eta$ is approximately given as

$$\eta \sim 2 \times \frac{2\gamma}{L},$$

where factor 2 comes from the spin degrees of freedom. Here, we define mean charge modulation $\bar{\eta}$ as

$$\bar{\eta} = \frac{1}{L} \times \int_0^L \eta \cos \frac{2\pi x}{L} dx = \frac{2}{\pi} \eta,$$

Then, the energy loss within the first-order with respect

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**Fig. S9:** (a) Doping dependence of condensation energy $\Delta E/t$ for $U/t = 10, t'/t = -0.3$. Broken red line represents the condensation energy for $U/t = 10, t'/t = 0$. (b) Doping ($\delta$) dependence of averaged $d_{x^2-y^2}$-wave superconducting correlations $\bar{P}_{x^2-y^2}$ and peak values of spin structure factors $S(q_{\text{peak}})$ for $U/t = 10$ and $t'/t = -0.3$. For comparison, we plot $\bar{P}_{x^2-y^2}$ of $U/t = 10, t'/t = 0$ by broken line.

**Fig. S10:** Doping dependence of chemical potential $\mu$ for $t'/t = 0$ and $t'/t = -0.3$.

accessible system sizes.
To $q$ is calculated as follows:

$$E_q = \frac{\langle \phi_{CM} | H_0 | \phi_{CM} \rangle}{\langle \phi_{CM} | \phi_{CM} \rangle}$$

$$= \sum_{k \in D, \sigma} \epsilon_k + \frac{|\gamma|^2}{1 + M|\gamma|^2} \left[ - \sum_{k \in R, \sigma} \epsilon_k + \sum_{k \in R, \sigma} \epsilon_{k+q} \right]$$

$$\sim \sum_{k \in D, \sigma} \epsilon_k + \frac{|\gamma|^2 q_x}{1 + M|\gamma|^2} \sum_{k \in R, \sigma} \frac{\partial \epsilon_k}{\partial k_x}.$$  \hspace{1cm} (S.12)

From this, we evaluate the energy loss arising from the density modulation, $\Delta E_{CM}$ as

$$\Delta E_{CM} = E_q - E_{q=0}$$

$$= \frac{|\gamma|^2 q_x}{1 + M|\gamma|^2} \sum_{k \in R, \sigma} 2t^* \sin k_x$$

$$\sim 16|\gamma|^2 t^*,$$  \hspace{1cm} (S.13)

where we again assume $M|\gamma|^2 \ll 1$.

If the energy loss $\Delta E_{CM}$ is smaller than the energy gain of the phase separation $\Delta E_{PS}$, the spatially inhomogeneous phase becomes stable. As shown in Fig. S 11(b), from the mVMC calculations for a typical case ($U/t = 10, V = J = 0$), we evaluate the energy gain by the phase separation with the amplitude 0.1 (in the unit of the doping concentration $\delta$) at most $5 \times 10^{-3} t$. Then we have roughly estimated the energy gain in the case of the density modulation $\bar{\eta}$ as $5 \times 10^{-3} t \times 0.1$, simply by approximating the curve in Fig. S 11(b) by a linear function. Thus, the condition that the spatially inhomogeneous phase becomes stable is given by

$$\frac{\Delta E_{CM}}{N_s} \sim \frac{16|\gamma|^2 t^*}{N_s} < \Delta E_{PS} \sim 5 \times 10^{-3} t \times \bar{\eta}.$$  \hspace{1cm} (S.14)

Given this condition is satisfied and by assuming that $t^*$ is the same as $t$, we can evaluate the maximally allowed amplitude of the charge modulation as

$$|\eta| < 0.03,$$  \hspace{1cm} (S.15)

in finite size systems. Thus, even when the phase separation is the correct solution in the infinite size system, the amplitude of charge structure factor $N(q)$ at the lowest possible wavenumber for $N_s = 16 \times 16 = 256$ is given as

$$N(q) = \frac{1}{N_s} \sum_{i,j} n_i n_j e^{iq(r_i-r_j)} = N_s \times |\eta|^2 \sim 0.2.$$  \hspace{1cm} (S.16)

Although the present estimate is rough, the order estimate of enhancement is expected to be correct. Around $q \sim 0$, we indeed see $N(q)$ in the order of 0.1, but it is buried in the background structure. Thus, it is difficult to see clear signature of the phase separation from $N(q)$ in available system size. In contrast to this, the doping dependence of the chemical potential $\mu$ offers a reliable estimation of phase separation region in the relatively small systems, because they can be correctly calculated by the uniform density state. Further analysis such as performing calculations for larger system size is intriguing issue but left for future study.
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