Doping phase diagram of a Hubbard model for twisted bilayer cuprates

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We study the twisted Hubbard model of a cuprate bilayer at a fixed twist angle \( \theta = 53.13^\circ \) using the variational cluster approximation, a method that treats short-range dynamical correlations exactly. At intermediate interlayer tunneling, the phase difference \( \phi \) between the \( d \)-wave order parameters of two layers is \( \pi \) in the overdoped regime, while it is zero in the underdoped regime, close to the Mott phase. At strong interlayer tunneling, we observe a clear time-reversal symmetry breaking phase near optimal doping, in which the phase difference \( \phi \) changes continuously from 0 to \( \pi \). However, this phase has trivial topology. We also apply a cluster extension of dynamical mean field theory to the same problem, but fail to detect a time-reversal breaking phase with that method.

I. INTRODUCTION

The experimental discovery of correlated insulators and unconventional superconductivity in twisted bilayer graphene (TBG) [1, 2] has opened up the new field of twistronics [3, 4]. By twisting two graphene sheets by a small relative angle, a long-period moiré pattern forms in the bilayer. At special magic angles, the moiré band structure of TBG exhibits isolated flat bands near charge neutrality [5–7], which lead to a variety of strongly correlated phenomena. Following this discovery, various twisted van der Waals heterostructures have been constructed and investigated [8], including transition metal dichalcogenides [9–12], double bilayer graphene [13–15], and trilayer graphene [16–19].

Recently, twistronics concepts have been extended to high-temperature superconductors [20, 21], which are strongly correlated materials by themselves. This was motivated by the experimental realization of two-dimensional (2D) monolayer Bi\textsubscript{2}Sr\textsubscript{2}CaCu\textsubscript{2}O\textsubscript{8+\delta} (Bi2212), whose transition temperature is shown to be very close to that of bulk samples [22, 23]. It is theoretically predicted that, at large twist angles (close to \( 45^\circ \)), a fully gapped \( d + id \) superconducting phase emerges, which spontaneously breaks time-reversal symmetry (TRS) and is topologically nontrivial [20]. This TRS breaking superconducting phase is also predicted to be stable at small twist angle, due to the strong renormalization of Bogoliubov-de Gennes (BdG) quasiparticles near the nodes [21]. In order to determine the pairing symmetry of cuprate superconductors, \( c \)-axis twisted Josephson junctions, formed by stacking two Bi2212 crystals along the \( c \)-axis, have been realized [24–29]. However, most experimental works did not observe the angular dependence of the Josephson current [24, 25, 29]. Owed to the novel technique of van der Waals stacking, high-quality twisted Bi2212 Josephson junctions with an atomically sharp interface have been successfully fabricated recently [29, 30].

Previous theoretical work on twisted bilayer cuprates are mainly based on Bogoliubov-de Gennes mean-field theory [20, 21, 28, 31, 32], which does not take into account the effects of strong correlations. To overcome this, a twisted \( t-J \) model of cuprates has been proposed and studied within slave-boson mean-field theory [33], in which a topological-trivial time-reversal symmetry breaking superconductor is also found, but within a small range of twist angles around \( 45^\circ \), questioning the possibility of topological superconductors in this region. In spite of this work, the stability of the novel superconducting phases against doping has not been fully addressed before in the literature. In this paper, we will numerically study the twisted Hubbard model of bilayer cuprates using the variational cluster approach (VCA) and cluster dynamical mean field theory (CDMFT). These approaches have been successfully used in the past to study high-temperature superconductors and the Hubbard model at intermediate coupling is arguably a better representation of these materials. We will focus on a fixed twisted angle \( \theta = 53.13^\circ \), at which these cluster methods are easily applicable, and investigate the superconducting phase diagram as a function of doping for two different sets of interlayer tunneling.

This paper is organized as follows. In Sec. II, we introduce the Hubbard model for the twisted bilayer. In Sec. III, we review the variational cluster approximation (VCA) and present our main results obtained from this method, e.g., the phase diagram of bilayer as a function of hole doping. In Sect. IV, we present the corresponding results from cluster dynamical mean field theory (CDMFT).

II. MODEL

A. Hamiltonian

We assume that each of the two layers of the system can be described by the one-band Hubbard model (the sites correspond to the location of copper atoms). The bilayer is then described by the following tight-binding Hubbard model:

\[
H = H^{(1)} + H^{(2)} + H_\perp, \tag{1}
\]

where the intra-layer Hamiltonian \( H^{(l)} \) is

\[
H^{(l)} = \sum_{r \in \ell, \sigma} t_{r \ell} c_{r \ell, \sigma}^\dagger c_{r \ell, \sigma} + U \sum_{r} n_{r \uparrow} n_{r \downarrow} - \mu \sum_{r, \sigma} n_{r \ell, \sigma}, \tag{2}
\]

\[
H^{(1)} = \sum_{r \in \ell, \sigma} t_{r \ell} c_{r \ell, \sigma}^\dagger c_{r \ell, \sigma} + U \sum_{r} n_{r \uparrow} n_{r \downarrow} - \mu \sum_{r, \sigma} n_{r \ell, \sigma}, \tag{2}
\]
In order to simplify as much as possible our numerical work, we will restrict our analysis to a twist angle of \( \theta = 2 \arctan \frac{1}{2} = 53.13^\circ \). The unit cell of the twisted bilayer at that angle is illustrated on Fig. 1 and contains ten sites (five per layer).

B. Symmetries

The bilayer system is invariant under a \( \pi/4 \) rotation around the \( z \) axis (perpendicular to the bilayer plane) and under the \( \pi \) rotations \( C_x \), \( C_y \) and \( C_d \) illustrated on Fig. 2, which make up the \( D_4 \) point group, the same as for an isolated layer. Possible superconducting gap functions for this system should in principle be classified according to the irreducible representations of \( D_4 \). Table II shows the character table and the simplest gap functions associated with each irreducible representation. Representations \( B_1 \) and \( B_2 \) correspond to what is usually called \( d_{x^2-y^2} \) and \( d_{xy} \), respectively. Representations \( A_1 \) and \( A_2 \) correspond respectively to \( s \)-wave (or extended \( s \)-wave) and \( f \)-wave, and the two-dimensional representation \( E \) would correspond to (triplet)
call it the reduced Brillouin zone and its wave vectors are noted \( \tilde{k} \). The hopping matrix in \( H \) can be expressed as a \( 2L N_b \times 2L N_b \) matrix \( t(\tilde{k}) \), a function of \( \tilde{k} \), which is the sum of a \( \tilde{k} \)-independent part \( t_c \) and of the inter-cluster part \( V(\tilde{k}) \) : \( t(\tilde{k}) = t_c + V(\tilde{k}) \). The self-energy \( \Sigma_c(\omega) \) associated with the cluster Green function \( G_c(\omega) \) is thus defined by Dyson’s equation on the cluster:

\[
G_c^{-1} = \omega - t_c - \Sigma_c(\omega)
\]  

(4)

In CPT, the electron self-energy is approximated by that of the restriction \( H' \) of the Hamiltonian to the cluster. In the mixed momentum-cluster site basis, the electron Green function is then given by the following relation:

\[
G^{-1}(\tilde{k}, \omega) = \omega - t(\tilde{k}) - \Sigma_c(\omega) = G_c^{-1}(\omega) - V(\tilde{k})
\]  

(5)

We assume here that the chemical potential \( \mu \) is included in the hopping matrix \( t(\tilde{k}) \).

CPT is unable to describe broken symmetry states: it is not a self-consistent approach, nor is it based on a variational principle. The VCA adds a variational aspect to CPT: the cluster Hamiltonian \( H' \) is augmented by a certain number of Weiss fields:

\[
H' \rightarrow H' + \sum_a h_a \hat{O}_a
\]  

(6)

where the operators \( \hat{O}_a \) are defined on the cluster only, and possibly represent broken symmetries. These additional terms are in turn subtracted from \( V \), so that the original Hamiltonian \( H \) in unaffected. The values \( h_a \) of these Weiss fields are not arbitrary, but set by Potthoff’s variational principle: The following function:

\[
\Omega(h_a) = \Omega' - \frac{d\omega}{2\pi} \sum_{\tilde{k}} \ln \det \left[ 1 - V(\tilde{k}) G_c(\omega) \right]
\]  

(7)

should be stationary with respect to these fields \( h_a \). In that expression, \( \Omega' \) is the ground state energy of the cluster Hamiltonian \( H' \) and \( G_c(\omega) \) is the electron Green function derived from the cluster Hamiltonian \( H' \) that includes the Weiss fields \( h_a \hat{O}_a \).

In the problem at hand, it might seem natural to use the 10-site unit cell shown in Fig. 1 as the repeated cluster, especially since 10 sites is an easily manageable size for an exact-diagonalization solver. However, the set of numbered sites in Fig. 1 does not have the \( D_4 \) symmetry of the full Hamiltonian, and this complicates the VCA computations. We will rather use a slight refinement of the method described above, assuming that the repeated unit is a supercluster of 10 sites obtained by assembling an octagonal cluster of 8 sites and a point-like cluster of 2 sites, each delimited by a red dashed line in Fig. 1. The self-energy of the supercluster is then a direct sum of the self-energies of an 8-site and of a 2-site cluster. Otherwise, the method is unchanged from the general approach described above.

On the octagonal cluster, we will define Weiss fields associated with \( d \)-wave superconductivity on each layer. On
each layer of the lattice, we can define an operator field that describes d-wave superconductivity:

\[
\Delta^{(\ell)} = \sum_{\ell=1}^{2} \left\{ c_{\ell,\uparrow} c_{\ell+\mathbf{y}^{(\ell)},\downarrow} - c_{\ell,\downarrow} c_{\ell+\mathbf{x}^{(\ell)},\uparrow} - c_{\ell,\uparrow} c_{\ell+\mathbf{y}^{(\ell)},\downarrow} + c_{\ell,\downarrow} c_{\ell+\mathbf{x}^{(\ell)},\uparrow} \right\} \tag{8}
\]

where \( \mathbf{x}^{(\ell)} \) and \( \mathbf{y}^{(\ell)} \) are the orthogonal lattice vectors on layer \( \ell \). We can then add the following combinations to the cluster Hamiltonian:

\[
H' \rightarrow H' + \sum_{\ell=1,2} d^{(\ell)} \Delta^{(\ell)} + \text{H.c} \tag{9}
\]

where \( \Delta^{(\ell)} \) is a restriction to the cluster of the lattice operator (8) and \( d^{(\ell)} \) is a complex amplitude. The real and imaginary parts of \( d^{(\ell)} \) are then Weiss fields in the sense of the coefficients \( h_\alpha \) of Eq. (6). Because of overall phase symmetry, one can always assume that \( d^{(1)} \) is real, but we must assume in all generality that \( d^{(2)} \) is complex. The complex phase of \( d^{(2)} \) is then the relative phase \( \phi \) of the superconducting order parameters of the two layers, and a value other than zero or \( \pi \) would signal a spontaneous TRS breaking and possible topological properties.

We applied the VCA method on this cluster system, using the two sets of interlayer tunneling defined in Table I. In practice, this means computing the cluster Green function \( G_\ell(\omega) \) repeatedly while adjusting the Weiss fields \( d^{(\ell)} \) so as to make the Potthoff functional stationary (in fact, minimum). Once the stationary values are found, the Green function (5) can be used to compute the ground state average of any one-body operator, in particular the order parameter \( \Psi^{(\ell)} = \langle \Delta^{(\ell)} \rangle / N \) (\( N \) is the number of sites) on each layer. The electron density \( n \) can be likewise computed from the Green function (the chemical potential \( \mu \) is the actual control parameter that is varied).

Fig. 4 shows the order parameter \( \Psi^{(\ell)} \) as a function of electron density \( n \) for hole doping and the two sets of interlayer tunneling (intermediate and strong). We note the characteristic dome shape that is typically obtained in quantum cluster methods, qualitatively agreeing with the known properties of cuprates. The electron density computed from the Green function (5) has some systematic error, as can be seen from the fact that the order parameter vanishes not at \( n = 1 \), as it should from Mott physics, but at \( n = 1.006 \). The bottom layer of the figure shows the relative phase of the order parameters \( \Psi^{(2)} \) and \( \Psi^{(1)} \) (on the two layers). At intermediate inter-layer tunneling, this phase is 0 at low doping, which is the signature of the \( B_1 \) representation of Table II. Beyond about 7% doping, this phase switches to \( \pi \), a signature of the \( B_2 \) representation. There is thus a doping-induced transition of the bilayer superconducting state, which coincides with the passage from underdoped...
Figure 6. (Color online) Impurity models used in CDMFT. On the left: the 4-site cluster used for sites (2,3,4,5) and (7,8,9,10) of the unit cell, as labeled in Fig. 1. On the right, the 2-site cluster used for sites (1,6).

to overdoped, judging by the location of optimal doping on the upper panel of the figure.

For strong interlayer tunneling, the situation is different: an intermediate phase appears in which the relative complex phase of the two order parameters changes continuously from 0 to π. This intermediate phase breaks time reversal symmetry and corresponds roughly to a plex phase of the two order parameters changes continuously as the position of the minimum moves continuously from φ = ±π (B2 representation). Near φ = 0, the profile changes suddenly to one where the minimum is at φ = 0 (B1 representation). Note that the vertical scale is tiny (10−5), in multiples of t, which defines the energy unit here. This means that the energy difference between the two representations B1 and B2 might be just too small to be of consequence experimentally (∼10−2meV or ∼10−1K in terms of temperature), at an intermediate interlayer tunneling of V1 = 0.1.

On the bottom half of Fig. 5, the same type of data is shown at strong interlayer tunneling (V1 = 0.4). There the transition between B2 and B1 is gradual as the position of the minimum moves continuously from φ = ±π to φ = 0, with a spontaneous breaking of the symmetry. Even though this TRS breaking state is what we are looking for, such a strong value of interlayer tunneling is unrealistic.

Does this TRS breaking state have nontrivial topology? In a strongly correlated system, this question may be answered through the properties of the approximate interacting Green function (5) [40, 41]. The key idea is to define a “topological Hamiltonian” ħκ(κ) = −G−1(k, ω = 0), which can be diagonalized:

\[ h_\epsilon(k) |\alpha, \kappa) = \mu_\alpha(k) |\alpha, \kappa) \]  

One can then define a generalized Chern number just like in noninteracting systems:

\[ C_1 = \int \frac{d^2 k}{2\pi} \mathcal{F}_{xy}(k) \quad \mathcal{F}_{xy}(k) = \frac{\partial^2 \mathcal{A}_y}{\partial k_x} \frac{\partial^2 \mathcal{A}_x}{\partial k_y} \]  

with the Berry connection

\[ \mathcal{A}_j(k) = -i \sum_{\mu, \alpha} \langle \alpha, \kappa | \delta_{\kappa \mu} | \alpha, \kappa \rangle, \quad (j = x, y) \]

When applying this formula to the TRS states found by VCA, we find the topology to be trivial (the Chern number vanishes). This results from a compensation between different regions of the Brillouin zone, with opposite Berry curvature.

IV. RESULTS FROM CLUSTER DYNAMICAL MEAN FIELD THEORY

In order to test the robustness of our predictions, we have also studied the same system using cluster dynamical mean field theory (CDMFT) [42–45] with an exact diagonalization solver at zero temperature (or ED-CDMFT). Here the Weiss fields of VCA are replaced by a bath of uncorrelated orbitals whose parameters are determined self-consistently. Because the presence of this bath increases the size of the problem, the cluster cannot be as large as in VCA and typically contains no more than 4 sites.

Each cluster, together with the associated bath, defines an Anderson impurity model (AIM):

\[ H_{\text{imp}} = H_c + \sum_{\mu, \alpha} \theta_{\mu, \alpha} \left( c_\mu^\dagger a_\alpha + H.c. \right) + \sum_{\alpha \beta} \epsilon_{\alpha \beta} a_\alpha^\dagger a_\beta, \]  

where \( a_\alpha \) annihilates an electron in the bath orbital labeled \( \alpha \). The Nambu formalism must be used to incorporate pairing between bath sites, within the matrix \( \epsilon_{\alpha \beta} \). (i) For each iteration, the cluster Hamiltonian (13) is solved, i.e., the linearization solver at zero temperature (or ED-CDMFT). Here the Weiss fields of VCA are replaced by a bath of uncorrelated orbitals whose parameters are determined self-consistently.

The bath parameters \( \theta_{\mu, \alpha} \) and \( \epsilon_{\alpha \beta} \) are determined by an approximate self-consistent procedure, as proposed initially in [46], that goes as follows: (i) initial values of these parameters are chosen on the first iteration. (ii) For each iteration, the cluster Hamiltonian (13) is solved, i.e., the cluster Green function \( G_\epsilon(\omega) \) is computed. The latter can be expressed as

\[ G_\epsilon(\omega) = -\omega \Gamma(\omega) - \Sigma(\omega) \]  

where \( \Gamma(\omega) \) is the bath hybridization matrix:

\[ \Gamma_{ij}(\omega) = \sum_{\alpha, \alpha'} \theta_{ij, \alpha} \left( \frac{1}{\omega - \epsilon} \right)_{\alpha \alpha'} \theta_{ij, \alpha'}^* \]  

(iii) The bath parameters are updated, by minimizing the distance function:

\[ d(\epsilon, \theta) = \sum_{i \omega_n} W(i \omega_n) [G_\epsilon(i \omega_n)^{-1} - \tilde{G}(i \omega_n)^{-1}] \]
where $\tilde{G}(\omega)$, the projected Green function, is defined as

$$\tilde{G}(\omega) = \frac{1}{N} \sum_k G(k, \omega), \quad G(k, \omega) = \frac{1}{\omega - t_k - \Sigma_k(\omega)}.$$

(17)

Ideally, $\tilde{G}(\omega)$ should coincide with the impurity Green function $G_i(\omega)$, but the finite number of bath parameters does not allow for this correspondence at all frequencies, and so a distance function $d(\epsilon, \theta)$ is defined, with emphasis on low frequencies along the imaginary axis. The weight function $W(\omega)$ is where the method has some arbitrariness; in this work $W(\omega)$ is taken to be a constant for all Matsubara frequencies lower than a cutoff $\omega_c = 2t$, with a fictitious temperature $\beta^{-1} = t/50$. (iv) We go back to step (ii) and iterate until the bath parameters or the bath hybridization function $\Gamma(\omega)$ stop varying within some preset tolerance.

In the current problem, the 10-site unit cell was separated in three impurity problems: a four-site cluster on each layer (which together are equivalent to the 8-site cluster used in VCA in the last section), made respectively of the orbitals $(2,3,4,5)$ and $(7,8,9,10)$ as labeled on Fig. 1, and a two-site cluster made of orbitals $(1,6)$. These clusters are illustrated on Fig. 6. The 4-site cluster is connected to 8 uncorrelated bath orbitals, and contains 6 independent parameters: Two bath energies $\epsilon_{1,2}$, two hybridization $\theta_{1,2}$ and two pairing amplitudes $\Delta_{1,2}$ between bath orbitals, with signs appropriate for describing $d$-wave superconductivity. This way of parametrizing the bath is not the most general possible, but has been successfully used in the past [47–49]. The two-site cluster connects the two layers and also contains 6 bath parameters, except that the anomalous part is contained in the hybridization, i.e., it connects the bath sites to the cluster sites, not the bath sites themselves. In order to allow for a relative phase between the pairing on the two layers, the pairing bath parameters $\Delta_{1,2}$ on the square cluster of the second layer are allowed to take complex values, whereas those on the first layer are assumed to be real. Once a converged CDMFT solution is found, the same order parameters $\Delta^{(i)}$ as in the previous section are computed.

Fig. 7 shows the results of CDMFT applied to this system, for both intermediate (left) and strong (right) interlayer tunneling. The results depend on the initial set of bath parameters. On the top panels, the bath pairing parameters were initialized with opposite values on the two layers, whereas on the bottom panels, they were initialized with the same values. At intermediate interlayer tunneling ($V_1 = 0.1$), the order parameters stay opposite throughout the doping range if the bath pairings are initialized this way; in other words, if the system is primed in the $B_2$ representation, it will stay in that representation. At strong interlayer tunneling ($V_1 = 0.4$), this only occurs if doping is large enough. In other words, for doping 12% or less, the system primed in the $B_2$ representation will either not converge, or converge to a normal solution, indicating its incompatibility with the $B_2$ initial conditions. On the other hand, if the system is primed in the $B_1$ representation, then it stays in the $B_1$ representation, except that, at strong interlayer tunneling, it converges for larger values of doping, and converges to a normal solution at very small doping.
It is thus difficult to discriminate between the $B_1$ and $B_2$ representations within CDMFT, which does not have the fine energy resolution that VCA has. Nevertheless, we sense from the above results that the $B_1$ representation is preferred at low doping and the $B_2$ representation at higher doping, but a strong interlayer tunneling is needed for that. Also, despite allowing in principle for an arbitrary complex phase between the anomalous bath parameters of the two layers, only the phases $0$ and $\pi$ are found: no state with spontaneous breaking of time reversal is found in CDMFT. This may be related to the fact that the main 4-site impurity model in CDMFT is confined to each layer, i.e., the complex, twisted inter-layer structure has an impact only through the self-consistency relation. In studying such systems, it seems that the VCA is a better choice.

V. CONCLUSION

In a one-band Hubbard model for a cuprate bilayer twisted by an angle of $53.13^\circ$, the relative phase of the superconducting order parameter in the two layers depends on hole doping away from half-filling. In the underdoped regime, the relative phase vanishes, whereas it is $\pi$ in the overdoped regime. If the interlayer tunneling is strong, then there is an intermediate phase between those two in which this phase varies continuously from $0$ to $\pi$. Time reversal symmetry is broken in that intermediate phase, but the topology is trivial, at least as computed from the electron Green function. At intermediate interlayer tunneling, this TRS breaking phase does not exist.

It is possible that this TRS breaking phase survives at weaker interlayer tunneling if the twist angle is closer to $45^\circ$. A twist angle of $43.60^\circ$ corresponds to a unit cell of 58 copper sites [20] and might be amenable to a similar VCA study, albeit markedly more complex numerically. Work in this direction will be necessary in order to assess whether this putative phase is realistic in strong-coupling superconductivity.

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