Chiral $d + is$ superconducting state in the two dimensional $t$-$t'$ Hubbard model

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Applying the recently developed variational approach to Kohn-Luttinger superconductivity to the $t$-$t'$ Hubbard model in two dimensions, we have found, for sizeable next-nearest neighbor hopping, an electron density controlled quantum phase transition between a $d$-wave superconducting state close to half filling and an $s$-wave superconductor at lower electron density. The transition occurs via an intermediate time reversal breaking $d + is$ superconducting phase, which is characterized by nonvanishing chirality and density-current correlation. Our results suggest the possibility of a bulk time reversal symmetry breaking state in overdoped cuprates.

As demonstrated by Kohn and Luttinger (KL), the generic ground state of a clean degenerate electron system should be superconducting. Direct application of the KL approach to the $t$-$t'$ Hubbard model in two dimensions has shown that, at sufficiently weak coupling when no particle-hole instabilities develop, the phase diagram of the model in the $t'/t$ versus electron density, $\rho$, plane, exhibits several superconducting phases of different symmetry. Similar conclusions have been reached previously in mean field studies of the $t$-$t'$-J model and more recently making use of the renormalization group method and fluctuation exchange approximation. Thus, by tuning one control parameter ($t'/t$ or $\rho$), a quantum phase transition between two different superconducting states can be realized. The purpose of this paper is to study such transitions within a simple but realistic microscopic model.

As a particular example motivated by the physics of high temperature superconductors, we have chosen to study the two-dimensional $t$-$t'$ Hubbard model with the bare single particle dispersion $\epsilon_{\mathbf{k}} = -2t(\cos k_x a + \cos k_y a) + 4t' \cos k_x a \cos k_y a$ where $a$ is the in-plane lattice constant. This is the minimal model including electron correlations and reproducing the shape of the Fermi surface observed by angle resolved photoemission spectroscopy in the cuprates. In a previous study, we have shown that close to half filling $d$-wave is the dominant pairing symmetry in a broad range of $t'/t$ and that, under reducing the electron concentration, different pairing symmetries start to dominate. Here we have solved the gap equation at $\rho = 0.45$, in which case a density-controlled transition between the $d$-wave and $s$-wave pairing states was predicted to occur in the vicinity of $\rho \approx 0.7$. This choice of $t'/t$ is motivated by the observation that at smaller ratios of $t'/t$, the transition either happens at small electron fillings, or is located close to the Van Hove density in which case our method does not apply. Moreover, according to a recent study, TI and Hg based materials do indeed support sizeable next nearest neighbor hopping.

We address this problem within the recently developed variational approach to superconductivity. The main idea of Ref. [10] is as follows: Performing a canonical transformation which eliminates the scattering processes to first order in $U/t$ (where $U$ is the local Hubbard interaction), we have constructed an effective Hamiltonian which leads to an interaction in the Cooper channel of the form $V_{\mathbf{k}p} = U + U^2 \chi_1(\mathbf{k} + \mathbf{p}, \epsilon_{\mathbf{p}} - \epsilon_{\mathbf{k}})$, where $\chi_1(q, \omega)$ is the real part of the particle-hole susceptibility $\chi(q, \omega) = L^{-1} \sum_{\mathbf{k}} \langle f_{\mathbf{k}+\mathbf{q}} f_{\mathbf{k}} \rangle / (\epsilon_{\mathbf{k}+\mathbf{q}} - \epsilon_{\mathbf{k}} - i\delta)$ and $L$ is the number of lattice sites. Superconductivity is realized if a nontrivial order parameter $\Delta_{\mathbf{k}}$ which solves the gap equation

$$\Delta_{\mathbf{k}} = -\frac{1}{L} \sum_{\mathbf{p}} V_{\mathbf{k}p} \Delta_{\mathbf{p}} \frac{\tanh(\frac{E_p}{2T})}{2E_p}$$

(1)

can be found. In Eq. (1) we introduced the quasiparticle energy $E_p = (\epsilon_{\mathbf{p}}^2 + |\Delta_{\mathbf{p}}|^2)^{1/2}$, where $\epsilon_{\mathbf{p}} = \epsilon_{\mathbf{p}} - \mu$ and $\mu$ is the chemical potential.

All calculations reported in this paper were performed for moderate interaction strength $U/t = 7$. Strictly speaking, this is outside the region of validity of our weak-coupling approach. The need for such values of $U$ is due to the finite system sizes, typically up to 512 × 512, which we can study within a reasonable time. At smaller values of $U$, the finite size effects become appreciable. However, we do not believe the large values of $U$ are a serious drawback of our calculations. In fact, in the $d$-wave state the value of $U$ is just a multiplicative factor entering $V_{\mathbf{k}p}$ (since the $d$-wave state is not affected at all by the constant term $U$ in $V_{\mathbf{k}p}$). On the other hand, although both terms in $V_{\mathbf{k}p}$ do affect the $s$-wave pairing state, we have checked a posteriori that our solutions in the $s$-wave channel exhibit very small on-site pairing amplitudes, thus providing a nontrivial self-consistency check of the calculation.

At $\rho = 0.685$ we have solved the gap equation at $T = 0$ by an iteration method. The calculation was performed many times, starting from a random complex vector $\Delta_{\mathbf{k}}$. We have found three types of solutions: pure $d$-wave, pure $s$-wave, and $d + is$-type solutions which can be written as $\Delta_{\mathbf{k}} = d_{\mathbf{k}} + is_{\mathbf{k}}$ with real $d_{\mathbf{k}}$ and $s_{\mathbf{k}}$. As a next step, we have required that the self-consistent solution is one of the above three types (which speeded up the calculation considerably) and in this way we calculated the data presented in Fig. [11]. Note that pure $s$-wave and $d$-wave solutions are stable for all studied densities, whereas the $d + is$-type solution can be stabilized only in the interval...
0.655 < \rho < 0.715.

We have calculated the BCS condensation energy $E_{\text{cond}} = -\sum_{\mathbf{k}} (E_{\mathbf{k}} - |\xi_{\mathbf{k}}|^2)/2E_{\mathbf{k}}$ as a function of $\rho$ for all three types of solutions. The results are plotted in Fig. 1b. We find that the $d + is$ state is the true ground state in the whole region of its stability. Thus, at weak coupling, the quantum phase transition between the $s$-wave state at low electron density and the high-density $d$-wave state occurs via a mixed symmetry state $d + is$, in agreement with a model calculation for an isotropic Fermi surface and separable interactions.\textsuperscript{11} Within our mean field like approach, both $s/d + is$ and $d/d + is$ transitions are of second order. Field theoretic studies suggest that for the $d/d + is$ transition this is valid also beyond mean field.\textsuperscript{11,12}

It is worth pointing out that the complex admixture increases the condensation energy substantially, up to 30 per cent close to the crossing point of pure $s$-wave and $d$-wave states. Since the condensation energy is determined by the typical value of $|\Delta_{\mathbf{k}}|$ close to the Fermi energy and, according to Fig. 1b, the maximal value of $|\Delta_{\mathbf{k}}|$ is approximately the same in the mixed and pure states close to the crossing point, the density of states in the $d + is$ state must be substantially suppressed in the subgap region. This is indeed found in an explicit calculation, see Fig. 2. Note that a small true gap is opened in the subgap region.

![Fig. 1](image1.png)

**Fig. 1:** (a) Maximal value of $|\Delta_{\mathbf{k}}|$ at the Fermi surface. Triangles: pure $d$-wave state. Squares: pure $s$-wave state. Circles: the same for $|d_{\mathbf{k}}|$ and $|s_{\mathbf{k}}|$ in the $d + is$ state. (b) Condensation energy per lattice site (in units of $E/t$) calculated as functions of the electron density $\rho$ for $t'/t = 0.45$ and $U/t = 7$ on special lattices\textsuperscript{10} with $L = 512 \times 512$ (a,d) and $L = 256 \times 256$ (c). (b) was calculated by extrapolating the results in (a) to 4096 × 4096.

![Fig. 2](image2.png)

**Fig. 2:** Density of states per lattice site (in units of $t$) for $\rho = 0.69$ in the pure $d$-wave, pure $s$-wave, and in the $d + is$ state (dashed, thin solid, and thick solid lines, respectively). Calculated following Ref.\textsuperscript{10} for the same parameters as Fig. 1. The finite weight at low energies is due to the finite width of the $\delta$ functions, $\gamma/t = 2 \times 10^{-4}$.

![Fig. 3](image3.png)

**Fig. 3:** Magnitudes of the real (left panel) and imaginary (right panel) parts of the pairing function, $|D_{ij}|$ and $|S_{ij}|$, in the chiral $d + is$ state. The middle of the square corresponds to $i = j$. Calculated for $\rho = 0.69$ and the same parameters as in Fig. 1b.
d + is state, whereas both the d-wave and the s-wave states are gapless.

In order to get further insight into the nature of the d + is state, let us consider the singlet pairing function \( F_{ij} = \sum_\sigma \sigma (c_{i \sigma} - c_{j \sigma}) \). In the d + is state we can write \( F_{ij} = D_{ij} + i S_{ij} \) where \( D_{ij} = L^{-1} \sum_k (d_k / E_k) \cos k \cdot R_{ij} \), \( S_{ij} = L^{-1} \sum_k (s_k / E_k) \cos k \cdot R_{ij} \), and \( R_{ij} \) is the vector connecting lattice sites \( i \) and \( j \). Note that \( D_{ij} \) and \( S_{ij} \) are real and even in \( i, j \). In Fig. 3 we plot \( |D_{ij}| \) and \( |S_{ij}| \) in the chiral state. We find that \( |D_{ij}| \) is peaked for \( j - i = (1, 0) \), \( (2, 0) \), and symmetry related points, whereas \( |S_{ij}| \) peaks for \( j - i = (3, 0) \), \( (3, 1) \), \( (2, 2) \), and symmetry related points. This means that the d-wave and s-wave states do not compete for the same Cooper pairs, allowing for a coexistence of both states in the d + is state.

The temperature dependence of the d-wave and s-wave parts of the gap in the d + is state is shown for \( \rho = 0.7 \) in Fig. 1. Two critical temperatures \( T^d_c \) and \( T^{d+is}_c \) can be defined, the mixed symmetry state being realized at \( T < T^{d+is}_c \), and the pure d-wave state occurring at \( T^{d+is}_c < T < T^d_c \). Collecting the data for different \( \rho = \text{const} \) together we obtain the phase diagram Fig. 1. Note that the region of stability of the d + is state shrinks with increasing temperature, due to the lower entropy in the mixed state than in the pure states.

The complex order parameter in the d + is state implies that time reversal symmetry is broken. Moreover, the point group symmetry is reduced from \( C_{4v} \) down to \( C_{2v} \), i.e. the 90° rotations and reflections across the diagonals, \( m' \), do not belong any more to the symmetry operations of the d + is state. A physical order parameter which distinguishes the two possible time-reversal related states \( d + is \) and \( d - is \) can be constructed as an odd function of the local spin operator \( S_i = 2^{-1} \hat{\sigma}_{\alpha\beta} c_{i\alpha}^\dagger c_{i\beta} \) (\( \hat{\sigma} \) is the vector of Pauli matrices) or of the charge current \( j_{ij} = -i d_{ij} \sum_\sigma (c_{i\sigma}^\dagger c_{j\sigma} - c_{i\sigma} c_{j\sigma}) \) defined on the bond \( ij \).

However, because of spin rotation invariance of the d + is state, we have \( S_i = 0 \), while the unbroken \( m \) reflection symmetry, i.e. \( x \rightarrow -x \), forces \( j_{ij} = 0 \). Therefore more complicated functions of \( S_i \) and \( j_{ij} \) have to be considered.

Let us start by taking the simplest order parameter made up solely of the spin operators. This must necessarily be a third power of \( S_i \). The spin chirality \( E_{ijk} = \langle S_i, (S_j \times S_k) \rangle \), where \( i, j, \) and \( k \) are lattice sites, is a natural candidate for such an order parameter, since it is spin rotation invariant. Very recently, the \( d + id_{xy} \) RVB spin liquid has been characterized by \( E_{ijk} \) and in what follows we show that this order parameter is useful in our case as well, if the lattice points \( i, j, \) and \( k \) are chosen in such a way that a nonzero value of \( E_{ijk} \) is not prohibited by the remaining \( C_{2v} \) symmetry.

In a singlet superconducting state, the chiral order parameter can be evaluated as

\[
E_{ijk} = -\frac{3}{4} \Im \left[ G_{ij} F_{jk} F_{ki}^* + G_{jk} F_{ki} F_{ij}^* + G_{ki} F_{ij} F_{jk}^* \right],
\]

where \( G_{ij} = \langle c_{i\sigma}^\dagger c_{j\sigma} \rangle = L^{-1} \sum_k f_k \cos k \cdot R_{ij} \) is independent of \( \sigma \) and an even function of \( i \) and \( j \). For conve-

nience let us introduce the following abbreviated notation for the neighborhood of the lattice point \( \mathbf{R} \) (see inset to Fig. 11): 0 = \( \mathbf{R} - \mathbf{x} + \mathbf{y} \), 1 = \( \mathbf{R} - \mathbf{x} \), 2 = \( \mathbf{R} \), 3 = \( \mathbf{R} + \mathbf{y} \), and 4 = \( \mathbf{R} + \mathbf{x} \), where \( \mathbf{x} \) and \( \mathbf{y} \) are unit lattice vectors in the \( x \) and \( y \) direction, respectively. Consider first the triangle 123. Since \( G_{12} = G_{23}, \ S_{12} = S_{23}, \ D_{12} = -D_{23}, \) and \( D_{13} = 0 \), making use of Eq. (2), one finds readily that \( E_{123} = (3/2) D_{12} (S_{12} G_{13} - S_{13} G_{12}) \). In Fig. 1 we plot \( E_{123} \) as a function of doping.

Let us point out that the chiral order parameter provides also a direct measure of broken spatial symmetries. In fact, the nonvanishing value of \( E_{123} \) is a direct consequence of broken \( m' \) symmetry, since in a \( m' \) symmetric system \( E_{123} = E_{321} = -E_{123} \). Moreover, a clockwise rotation of the lattice around the point 2 leads to \( E_{123} \rightarrow E_{231} \), and a straightforward calculation shows \( E_{321} = -E_{231} \). On the other hand, under the \( m \) reflection we have \( E_{123} \rightarrow E_{231} = E_{123} \), consistent with the

\[
\begin{array}{c}
\text{(a)}
\end{array}
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\text{FIG. 4: (a) Maximal amplitudes of the real and imaginary parts of \( \Delta_k \) at the Fermi surface in the d + is state, (b) specific heat \( c_V \) per lattice site, and (c) normalized penetration depth and spin susceptibility as functions of temperature. Calculated for \( \rho = 0.7 \) (and the same parameters as in Fig. 1) on a special lattice with \( L = 256 \times 256 \). (b) and (c) were calculated by extrapolating the data from (a) to a lattice 1024 × 1024.}
\end{array}
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hand, nonvanishing surface currents have been predicted recently (for a symmetry classification of superconducting states coexisting with a chiral translationally invariant pseudogap phase of nonsuperconducting origin). The crucial difference of our calculation with respect to Ref. 21 is that in our case no net currents are flowing, \( \langle j_{ij} \rangle = 0 \). This makes it possible that a translationally symmetric chiral state is realized also in a one band model.

Our weak coupling approach, which should be reliable sufficiently away from half filling, therefore predicts a bulk chiral \( d + is \) state in overdoped cuprates with sizeable next-nearest neighbor hopping. Next we address the question about experimental observability of such a state. Upon cooling, two phase transitions should be visible. Figure 4b shows that this is indeed the case but the anomaly of the specific heat at \( T_d^{d+is} \) is small, since the quasiparticles are already frozen out to a large extent at \( T_d^{d+is} \). This is expected to be a generic result which holds except for the case when \( T_d^{d+is} \sim T_c^{d+is} \). On the other hand, within BCS theory the in-plane penetration depth is given by

\[
\lambda = \frac{\epsilon^2}{\varepsilon_0 a^2 \hbar^2 c^2} \sum_{\mathbf{k}} \left( \frac{\partial f(\mathbf{E}_k)}{\partial \mathbf{k}} \right)^2 \frac{\partial f(\mathbf{E}_k)}{\partial \mathbf{k}},
\]

where \( \lambda \) is the interlayer distance. In Fig. 4c we show explicitly that \( T_d^{d+is} \) should be observable in the temperature dependence of \( \lambda \), and also of the spin susceptibility \( \chi \). Note also the activated behavior at low temperatures which is consistent with the full gap.

Direct tests of time-reversal symmetry breaking are more involved, since in a perfect sample \( \langle j_{ij} \rangle = 0 \) and therefore no spontaneous magnetic fields develop. Moreover, since the photoemission experiments test only the diagonal part of the Nambu-Gorkov electron Green’s function, the broken parity in our \( d + is \) state in overdoped cuprates with sizeable next-nearest neighbor hopping should be observable next-nearest neighbor hopping. Next we address the question about experimental observability of such a state. Upon cooling, two phase transitions should be visible. Figure 4b shows that this is indeed the case but the anomaly of the specific heat at \( T_d^{d+is} \) is small, since the quasiparticles are already frozen out to a large extent at \( T_d^{d+is} \). This is expected to be a generic result which holds except for the case when \( T_d^{d+is} \sim T_c^{d+is} \). On the other hand, within BCS theory the in-plane penetration depth is given by

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Recently a spontaneous splitting of zero bias conductance peaks in a tunneling experiment on overdoped cuprates has been interpreted in terms of a bulk time reversal symmetry breaking superconducting state. However, this experiment measures the properties of a \( [110] \) oriented surface of a \( d \)-wave superconductor and in this geometry, time reversal symmetry may be broken in the vicinity of the surface even in absence of such symmetry breaking in the bulk. Therefore phase sensitive experimental tests of the putative bulk chiral \( d + is \) state at

invariance of the \( d + is \) state under the \( m \) reflection.

Let us turn to order parameters based on current operators. The simplest order parameter of this type is

\[
B_{ijk} = \frac{1}{2} (n_i j_{jk} + j_k n_i) = t_{jk} \text{Im}(F_{ij}^* F_{ik}),
\]

and the equality applies to a general singlet superconductor. For a \( d + id \) superconductor, this specializes to \( B_{ijk} = t_{jk} (D_{ij} S_{ik} - S_{ij} D_{ik}) \). An example of \( B_{ijk} \) is shown in Fig. 5. The nonvanishing density-current correlation \( B_{ijk} \) suggests that potential scattering on imperfections such as impurities or surfaces will, in general, cause finite currents and magnetic fields in the vicinity of these defects. For scattering on nonmagnetic impurities, such currents have been explicitly calculated only recently (for a \( d + id \)) superconductor\(^{29}\). On the other hand, nonvanishing surface currents have been predicted already by Anderson and Morel\(^{30}\). In our case the surface currents should be most pronounced for \([110]\)-like surfaces, while for \([100]\)-like surfaces the unbroken \( m \) symmetry forbids the formation of such currents. This is fully consistent with the predictions of Ginzburg-Landau theory\(^ {26}\).

Next let us consider, in analogy to spin chirality, the third power of local current operators, \( P_{ijk} = (j_{ij} j_{jk} j_{ki}) \). Note that, being an expectation value of a Hermitian operator \( j_{ij} j_{jk} j_{ki} \), the order parameter \( P_{ijk} \) is a real quantity. A nonzero value of \( P_{ijk} \) means that the product of three current operators along a closed loop is different for current operators ordered clockwise and anticlockwise around the loop. Therefore we call \( P_{ijk} \) the current chirality. A lengthy but straightforward calculation shows that, in a singlet superconductor, \( P_{ijk} = -(32/3) t_{ij} t_{jk} t_{ki} E_{ijk} \). Thus, up to a trivial multiplicative constant, the spin and current chiralities are equal. Making use of the symmetry properties of \( E_{ijk} \) we find that the four possible current chiralities which can be defined on an elementary plaquette 0123 of the square lattice satisfy \( P_{013} = P_{012} = -P_{032} = -P_{123} \). The resulting current chirality pattern is shown in the inset of Fig. 3. Note the similarity of Fig. 3 with the distribution of currents in a chiral state of a three-band model introduced by Varma as a description of the pseudogap phase of the cuprates\(^ {21}\). This similarity is consistent with the recent symmetry classification of superconducting states coexisting with a chiral translationally invariant pseudogap phase of nonsuperconducting origin\(^ {22}\). The crucial difference of our calculation with respect to Ref. 21 is that in our case no net currents are flowing, \( \langle j_{ij} \rangle = 0 \). This makes it possible that a translationally symmetric chiral state is realized also in a one band model.

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large doping should avoid geometries with in-plane edges. We propose to measure the current-phase relation $I(\phi)$ of $c$-axis Josephson junctions between a conventional superconductor and overdoped cuprates. Assuming that the Josephson coupling is due to a finite $s$ component of the cuprates, from the relative sign of the first and second harmonics of $I(\phi)$ one can determine the relative phase of the $d$ and $s$ components of the order parameter.

In conclusion, applying the recently developed variational approach to KL superconductors, we have shown that for sufficiently large next-nearest-neighbor hopping $t'/t$, the Hubbard model predicts a change of pairing symmetry from $d$-wave to $s$-wave with overdoping, in agreement with a previous calculation. The quantum phase transition between the two superconducting states occurs via an intermediate chiral $d + is$ phase. We have shown that the density-current correlation and the spin and current chiralities serve as local measures of the broken time reversal and parity symmetry in the $d + is$ state, and we calculated their spatial distribution. Our results suggest the possibility of a bulk time reversal symmetry breaking state in overdoped cuprates with sizeable next-nearest neighbor hopping. We have calculated thermodynamic quantities which might be useful for experimental tests of the putative $d + is$ pairing state.

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