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Phys. Rev. B 92, 184502 — Published 3 November 2015
DOI: [10.1103/PhysRevB.92.184502](10.1103/PhysRevB.92.184502)
Macroscopic character of composite high temperature superconducting wires

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(Dated: October 14, 2015)

The “d-wave” symmetry of the superconducting order in the cuprate high temperature superconductors is a well established fact, and one which identifies them as “unconventional.” However, in macroscopic contexts – including many potential applications (i.e. superconducting “wires”) – the material is a composite of randomly oriented superconducting grains in a metallic matrix, in which Josephson coupling between grains mediates the onset of long-range phase coherence. (See, e.g.,[1–5].) Here, we analyze the physics at length scales large compared to the size of such grains, and in particular the macroscopic character of the long-range order that emerges. While XY-superconducting glass order and macroscopic d-wave superconductivity may be possible, we show that under many circumstances – especially when the d-wave superconducting grains are embedded in a metallic matrix – the most likely order has global s-wave symmetry.

Classification of phases:
The anomalous average of the spin-singlet electron pair-annihilation operator which characterizes the superconducting state is

\[ \langle \psi_\uparrow(\mathbf{r})\psi_\downarrow(\mathbf{r}) \rangle \equiv \langle \psi_\uparrow(\mathbf{r})\psi_\downarrow(\mathbf{r}) \rangle / \sqrt{2}, \]  

(1)

where \( \psi_\sigma(\mathbf{r}) \) creates an electron with spin polarization \( \sigma \) at position \( \mathbf{r} \), and \( \langle \cdot \rangle \) represents the equilibrium average over all thermal and quantum fluctuations.

Above the critical temperature, \( T_c \), in the normal metal phase, \( \langle \psi_\uparrow(\mathbf{r})\psi_\downarrow(\mathbf{r}) \rangle = 0 \), while \( \langle \psi_\uparrow(\mathbf{r})\psi_\downarrow(\mathbf{r}) \rangle \neq 0 \) for all \( T < T_c \). In pure crystals this quantity is only a function of \( \mathbf{r} - \mathbf{r}' \), so it is convenient to introduce its Fourier transform \( \phi(\mathbf{p}) \). Possible superconducting phases were classified in Refs.[5,7]. In particular, in s-wave superconductors, \( \langle \phi(\mathbf{p}) \rangle \) is invariant under all symmetry transformations of the crystal and \( \langle \phi(\mathbf{r}) \rangle \neq 0 \), while for other forms of singlet order, \( \langle \phi(\mathbf{p}) \rangle \) changes sign under certain symmetry transformations, and consequently \( \langle \phi(\mathbf{r}) \rangle = 0 \). For example, in a d-wave superconductor, \( \langle \phi(\mathbf{p}) \rangle \) (as well as \( \langle \phi(\mathbf{r}) \rangle \)) changes sign under rotation by \( \pi/2 \), as shown schematically in Fig. 2.

In a disordered system, \( \langle \phi(\mathbf{r}, \mathbf{r}') \rangle \) is a sample specific random quantity which does not posses any spacial symmetry. As a result, in any singlet superconducting phase in a disordered system, since no symmetry prevents it, generically \( \langle \phi(\mathbf{r}) \rangle \neq 0 \).

In this article we assume that the disorder ensemble preserves a group of translational and rotational symmetries statistically (i.e. on average). We will show below that after averaging over the realizations of the disorder, several superconducting phases emerge that can be precisely characterized.

The internal symmetry of the superconducting pairs (for example, s-wave or d-wave) refers to the transformation properties of the quantity

\[ \Phi(\mathbf{r} - \mathbf{r}') = \langle \phi(\mathbf{r}, \mathbf{r}') \rangle. \]  

(2)

under rotation, where the underline indicates a quantity that has been averaged over configurations of the quenched variables, e.g. the size, shape, orientation, and location of the superconducting grains as well as over realizations of the disordered potential in the metal.

However, the internal structure of \( \Phi(\mathbf{r}) \) is not directly measurable. It is therefore reasonable to define the symmetry of the superconducting state in terms of “phase sensitive measurements.” Definitionally, this refers to any measurement of the relative phase of the order parameter at two macroscopically separated locations on the surface of the system. For instance, this can be measured in a SQUID consisting of Josephson junctions at two surface positions connected by an external (macroscopic) conventional superconducting wire loop. In an s-wave state, the phase difference around the loop is zero independent of the relative orientation of the two surfaces; consequently in equilibrium there is no magnetic flux through the SQUID. By contrast, in a d-wave state, a phase difference of \( \pi \) is induced in a “corner SQUID” in which the loop connects portions of the surface approximately at right angles to each other, so there is a half quantum of magnetic flux through the SQUID in equilibrium. At the same time, if the SQUID loop connects nearly parallel portions of surface of a d-wave state (either on the same or opposite sides of the system), no equilibrium flux is induced. Similar analysis can be used to give a phase sensitive definition of other possible pairing symmetries.

The appropriate order parameter characterization of a superconducting glass state is a bit more subtle: while \( \langle \phi(\mathbf{r}, \mathbf{r}') \rangle \neq 0 \), its local phase varies randomly as a function of position, and correspondingly its configuration (or spatial) average vanishes. However, this state is sharply distinguished from the normal state by the existence of a non-zero Edwards-Anderson type order parameter

\[ M(\mathbf{r} - \mathbf{r}') \equiv \langle |\phi(\mathbf{r}, \mathbf{r}')| \rangle, \]  

(3)

i.e. the glass state has \( \langle \phi(\mathbf{r}, \mathbf{r}') \rangle = 0 \) but \( M \equiv M(\mathbf{0}) > 0 \). Another feature of a glassy state, reflecting the existence of random variations of the phase of \( \langle \phi(\mathbf{r}, \mathbf{r}') \rangle \), is that there are local equilibrium currents, \( \langle J(\mathbf{r}) \rangle \neq 0 \), and...
associated spontaneous breaking of time-reversal symmetry. It is easy to see that the configuration (or spatial) average current must vanish, $\langle \vec{J}(r) \rangle = \vec{0}$. Instead, an appropriate tensor order parameter is

$$
\tau_{ab} \equiv \langle J_a(\vec{0}) \rangle \langle J_b(\vec{0}) \rangle.
$$

A complete classification of the various broken symmetry phases in disordered superconductors is not currently available. However, in terms of the various quantities introduced above, we can define the phases which we will encounter in our discussion:

1) In the “normal” state at elevated temperatures, no symmetries are broken, and hence all the order parameters vanish.

2) In a “s-wave superconducting” state, $\Phi(r)$ is non-zero, while $\tau_{a,b} = 0$ – i.e. there are no equilibrium currents in the bulk, and $\Phi(r)$ is invariant under all symmetry transformations, i.e. it is rotationally invariant. In phase sensitive measurements, no equilibrium flux is induced in a SQUID, regardless of its geometry.

3) In a “d-wave superconducting” state, $\tau_{a,b} = 0$, $\Phi(r)$ is non-zero for non-zero $|r|$ and changes sign under rotation by $\pi/2$ about some axis, but is invariant under rotation by $\pi$. Consequently, $\Phi(\vec{0}) = 0$ and a half flux quantum is induced in equilibrium in a suitable corner SQUID.

4) In a “superconducting XY glass” state, $\Phi(r)$ vanishes but both $M$ and $\tau_{a,b}$ are non-zero. It is widely accepted, but still not completely settled that such a state exists in three dimensions below a non-zero glass transition temperature. There is also the possibility of a partially ordered glass phase with $M(r) = 0$ but $\tau_{a,b} \neq 0$, but whether this arises in generic models is still being debated.\textsuperscript{9,10}

5) It is possible to have a phase of coexisting uniform and glassy order – in such a phase both $\Phi$ and $\tau_{a,b}$ are non-zero. Depending on the behavior of $\Phi(r)$ under symmetry transformations (“rotations”), such a phase can still be classified as s-wave or d-wave etc.

2b) The most unexpected new state we identify here is what we will henceforth refer to as a “globally s-wave superconducting” state. Macroscopically, this is simply an unfamiliar limit of a s-wave state – one in which locally (i.e. in each superconducting grain) the order parameter has d-wave symmetry, but globally it is s-wave. Spectroscopically, such a state can reflect its microscopic origins as a d-wave superconductor, but from the viewpoint of macroscopic phase-sensitive measurements, it has s-wave symmetry. Moreover, time-reversal symmetry is unbroken, $\tau_{ab} = 0$.

The effective Hamiltonian: Below the bulk transition temperature, in a system composed of grains of size large compared to the coherence length, there is a well developed magnitude of the order parameter on each grain. We will consider the case of most relevance to the cuprates, in which the order parameter on each grain transforms according to a one-dimensional non-trivial (d-wave) representation of the point group. The only important low energy degree of freedom is the overall phase of the superconducting order parameter on each grain, which we will designate $\theta_j$.

There is a degree of arbitrariness in the definition of $\theta_j$; we choose a convention such that when $\theta_j = 0$, the order parameter on the grain is real and is positive in some particular crystallographic direction. Moreover we assume that the grains are sufficiently large that we can neglect quantum fluctuations of the order parameter. In this case, the macroscopic properties of the system can be captured by the phenomenological model

$$
H = -\sum_{ij} J_{ij} \cos(\theta_i - \theta_j),
$$

where the Josephson coupling $J_{ij}$ between grains $i$ and $j$ is real.

On general phenomenological grounds, we can separate the contributions to $J_{ij}$ into three pieces,

$$
J_{ij} = \eta_i \eta_j J_{ij}^{(1)} + \eta_i J_{ij}^{(2)} + J_{ij}^{(3)}.
$$

Here, $J_{ij}^{(a)} \geq 0$ for all $a$, while $\eta_i = \pm 1$ and $\eta_j = \pm 1$ are random variables with vanishing mean which determine the sign of the corresponding contributions to $J_{ij}$. The magnitude of each contribution to $J_{ij}$ is characterized by its mean, $\bar{J}_{ij}^{(a)} \equiv \sum_j J_{ij}^{(a)}$. To be explicit, we will always consider this model in three spatial dimensions, although it need not be isotropic.

For grains of conventional superconductors, under most circumstances, the Josephson coupling would be positive, which is to say that $J_{ij}^{(1)} \approx J_{ij}^{(2)} \approx 0$; in this limit, the model is equivalent to an XY ferromagnet which has a single phase transition at $T_c \sim J^{(3)}$, where physically the “ferromagnetic phase” corresponds to a statistically uniform s-wave superconducting phase. (A mean-field estimate yields $T_c^{(MF)} = J^{(3)}/2$.)

In any case in which the crystalline axes of d-wave superconducting grains are embedded into a disordered metal with random orientations, the sign of $J_{ij}$ is random, which means that $\bar{J}_{ij}^{(3)} = 0$. The two remaining terms in Eq 6 have very different character: The term proportional to $J_{ij}^{(1)}$ has its sign determined by a product of quantities that depend on the properties of each grain separately (which we will see is roughly related to the shape of the grains). Conversely, the sign of the term proportional to $J_{ij}^{(2)}$ is determined by a joint property of the pair of grains (which we will see is related to the relative orientation of their crystalline axes). In the limit
$J^{(1)} = 0$ (and $J^{(3)} = 0$), this problem is a version of the standard model of an XY spin-glass.\textsuperscript{17} For $J^{(2)} = 0$ (and $J^{(3)} = 0$), this problem is a version of the well known Mattis model\textsuperscript{11}.

Let us redefine the zero of phase on each grain separately to introduce “Mattis-transformed” phases

$$\tilde{\theta}_j \equiv \theta_j + \pi(1 - \eta_j)/2.$$  \hfill (7)

In terms of these transformed variables, the form of $H$ is unchanged, but the role of the different distributions is interchanged such that $\tilde{J}^{(1)} \to J^{(3)}$, $\tilde{J}^{(2)} \to J^{(2)}$, and $\tilde{J}^{(3)} \to J^{(1)}$. That the pure Mattis model is transformed in this way into a pure ferromagnetic XY model reflects the well known fact that this model introduces disorder without frustration.

**Conjectured phase diagrams:** In Fig. 1a we show a conjectured phase diagram for the model in Eq. 5 under the conditions that $J^{(3)} = 0$ as a function of the dimensionless temperature, $T/J$, and the relative magnitude of the Mattis and spin-glass type couplings, $0 \leq \tilde{J}^{(2)}/\tilde{J} \leq 1$, where $\tilde{J} \equiv \sqrt{J^{(1)}/2} + (J^{(2)})^2$. We have labeled the ordered state at small $\tilde{J}^{(2)}/\tilde{J}$ “globally s-wave”; as we will show below, in the present context this corresponds to the state defined in 2h), above. (It is the state in which the Mattis transformed phases are uniformly (“ferromagnetically”) ordered.) The “XY glass phase” is defined in 4), above. The intermediate state, in the present context, has coexisting global s-wave and XY glass order, corresponding to 5), above. Formally, via the transformation in Eq. 7, the same phase diagram applies to the problem (which has been studied in the spin-glass literature\textsuperscript{16}) of an XY spin glass with an excess of ferromagnetic interactions ($J^{(1)} = 0$, while $J^{(2)}$ and $J^{(3)} \neq 0$).

The arguments leading to this phase diagram along the edges are as follows:

i) For $J^{(2)} = 0$ the Mattis transformed problem is equivalent to an XY ferromagnet with some randomness in the magnitude of the exchange couplings. Thus, we conclude that there is a single phase transition with $T_c \sim J^{(1)}$ to a phase with long-range superconducting order.

ii) Decades of work has still not resulted in a well established understanding of even the most basic features of the XY spin-glass in $d = 3$. It is well accepted that there is a thermodynamic transition to a XY glass phase in large enough $d$. (See for example Ref.\textsuperscript{9}.\textsuperscript{9}) However, while it is widely believed that this conclusion applies in $d = 3$ (but not in $d = 2$), it remains some uncertainty concerning this conclusion.\textsuperscript{10}. Numerical experiments certainly reveal that if there is a transition at $T_{glass} \propto J^{(2)}$, the proportionality constant must be small (i.e. the spin-glass transition temperature is at least an order of magnitude smaller than for the corresponding model without frustration). Another intensely debated issue is whether there exists an intermediate partially ordered phase with $M = 0$ but $\tau_{ab} \neq 0$. We have drawn the phase boundaries in the limit $J^{(1)} \to 0$ in Figs. 1a and b under the assumption that there is a finite $T_{glass}$ in $d=3$. If it turns out that $T_{glass} = 0$, the lines marking the boundaries of the various glassy phases should be reinterpreted as crossover lines below which relaxation rates become extremely small. Conversely, if there are two transitions, then the lines should be interpreted as the mean of the two transitions. Even if there is a transition, it is not clear whether the spin-glass phase would have a non-zero critical current. Moreover, in a glassy state, thermodynamic and transport quantities (including any “apparent critical current”) would be time dependent. This is very different from the “globally s-wave superconducting state” where the critical current is finite and time independent. Approaching the glass phase from above ($T > T_{glass}$), the tendency to a state with non-zero $\tau_{ab}$ should give rise to a growing paramagnetic response - related to the so-called “Wohleben effect.”\textsuperscript{18,19}

We can also analyze the effect of moving in slightly from the edges of the phase diagram. Adding a small excess of ferromagnetic couplings in an XY spin-glass is not thought\textsuperscript{16} to fundamentally affect the nature of the glass phase, so one does not expect to encounter any new phases moving in from the right edge of the phase diagram into the regime in which $J^{(2)} \gg J^{(1)} > 0$. Similarly, at non-zero $T$, the same is true near the left edge of the phase diagram where $J^{(1)} > J^{(2)} > 0$ and $T > J^{(2)}$.

We note that strictly speaking, in the framework of Eq. 5, the “globally s-wave” state exists only at non-vanishing temperatures. To see this, we consider the problem in terms of the Mattis transformed variables, so we consider the situation at low temperature, deep in the uniformly (ferromagnetically) ordered phase. Even though $J^{(2)} \ll J^{(1)}$, there is some non-vanishing probability (proportional to $J^{(2)}/J$) to find some pairs of nearest grains coupled by a strong negative (frustrating) Josephson coupling. Consequently, in the ground-state, there will be a small vortex loop enclosing this bond. It is easy to see that two such double-degenerate defects interact as dipoles, so they will in turn freeze into a dipolar glass phase at a temperature which is roughly proportional to their concentration, i.e. at least to first approximation $T_{glass} \sim (J^{(2)})/(J^{(1)})$. Thus, at low enough temperatures and non-zero $J^{(2)}$ there occurs a phase with globally s-wave superconducting order coexisting with time-reversal symmetry breaking vortex glass order. At $T > T_{glass}$ the thermal fluctuations restore time reversal symmetry, leaving the globally s-wave state.

Quantum fluctuations of the order parameter, which are not included in the model in Eq. 5, produce effects similar to non-zero $T$; quantum tunnelling between the two degenerate vortex loop states of an isolated defect thus effectively restore time reversal symmetry. This makes it possible for the globally s-wave state to exist for small enough $J^{(2)}$ even at $T = 0$ as indicated by the dotted line in Fig. 1a. It is not currently known how the superconducting phases merge at higher temperatures and intermediate $J^{(2)}/J$, and indeed it is likely non-universal. We have shown with dashed lines one possible, particularly simple completion involving a single tetra-critical
We begin by considering a single isolated grain embedded in a perfectly disordered normal metal matrix. It is important to emphasize that, whether or not the order parameter inside the grain has d-wave character, the superconductor-metal interface breaks the point-group symmetry so a finite value of $\langle \phi (\mathbf{r}, \mathbf{r}') \rangle$ is induced in the neighboring metal. We will call the quantity $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ the local d-wave component. It is this component which survives in the normal metal on distances larger than $l$, while the local s-wave and other-wave components decay exponentially at large distances.

To obtain the requisite boundary condition for $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ at the normal metal-superconductor interface, one has to match its values in the superconductor and in the normal metal close to the interface. In particular, its sign reflects the sign of $\langle \phi (\mathbf{p}_L) \rangle$ in the grain’s bulk for $\mathbf{p} = \mathbf{p}_L$ normal to the interface ($|p_l| = p_F$). Thus, the sign of $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ changes along the boundary.

The key consequence is illustrated graphically in Fig. 2, which is a schematic of a grain of a d-wave superconductor embedded in a metallic matrix. At distances from the grain larger than $l$ but small compared to the size of the grain, the sign of $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ is dominated by the closest interface, and so it can be positive or negative depending on the orientation of that interface. However, far from the grain, the value of $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ gets contributions from Cooper pairs that have scattered from all parts of the grain’s surface and then diffused to the observation point $\mathbf{r}$. Thus its sign is determined by the sign of $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ averaged over the entire surface of the grain.

In other words at large distances, the sign of the anomalous average is fully determined by the geometry of the grain. Qualitatively level the sign of $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ is determined with the sign of $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ by the interference between the Cooper pairs traveling to the observation point $\mathbf{r}$ along different diffusive trajectories, as illustrated by the two representative paths shown as the dashed lines in Fig. 2.

To compute the Josephson coupling between a pair of grains formally requires the solution of this problem in the presence of two superconducting grains as a function of their relative phases. However, it is straightforward to understand the sign of the Josephson coupling by overlaying the patterns $\langle \phi (\mathbf{r}, \mathbf{r}) \rangle$ produced by each grain individually. As a result, for a pair of grains of comparable characteristic size $R$ separated by a distance $L$, we find that for $L > R$, the sign of $J_{ij}$ is determined by a product of single grain characteristics $1^{2-14}$. i.e. it appears as a contribution to $J_{ij}^{(1)}$ while for $L < R$, its sign is determined by mutual aspects of the shape and orientation of the two grains – i.e. it appears as a contribution to $J_{ij}^{(2)}$. Other than this, the considerations controlling the magnitude of the Josephson coupling are not substantially different from those in conventional S-N-S junctions. For instance, for $L \gg R \gg l$

$$J_{ij} = \eta_i \eta_j J_{ij}^{(1)} \sim \eta_i \eta_j \left( \frac{G D R}{|\mathbf{r}|^3} \right) e^{-|\mathbf{r}|/\xi_{T}} \quad (8)$$

where $D$ is the electron diffusion coefficient, $\xi_T = \sqrt{D/T}$ the coherence length of the surrounding metal, and $G$ is the characteristic conductance of the grain. In $d = 3$, generally $G \propto R$ so the term in parentheses scales as $J_{ij} \sim R^2/|\mathbf{r}|^3$.

Importantly for present purposes, pairs of randomly oriented grains with $R \gtrsim L$ contribute to $J^{(2)}$ while those with $L \gtrsim R$ contribute to $J^{(1)}$. Consequently, for fixed
size grains, the material can be tuned across the phase diagram in Figs. 1a & b by varying the concentration of grains, as indicated.

Identification of “globally s-wave” order: It should now be apparent that the global s-wave symmetry of the superconducting order refers to the symmetry of the induced order in the metallic host produced by the embedded d-wave grains. Even though the superconductivity originates within the grains, the relative phase of the order parameter from grain to grain is determined by the condition that the phase be constant throughout the metal. Another way to obtain an intuitive understanding is to imagine replacing the metal by an s-wave superconductor. Now, consider the effect this has on the phase of the superconducting order in each (d-wave superconducting) grain. It is clear that in this case, barring an accidental degeneracy, the phase of the d-wave order parameter on each grain will be locked to that of the surrounding metal – either with the same phase or with a phase-shift of $\pi$ depending on the shape of the grain. If we now imagine continuously decreasing the strength of the intrinsic s-wave order in the matrix, by adiabatic continuity we would approach the situation we have discussed here.

Further implications: In addition to its fundamental interest, the present results suggest new strategies for making better practical wires. Firstly, to avoid the various detrimental effects of frustration (and glassy phases), one would like to insure that $J^{(1)} > J^{(2)}$; this is accomplished by insuring that the separation, $L$, between the grains is larger than or comparable to their characteristic radius, $R$. However, it is also desirable that the magnitude of the Josephson couplings be as large as possible. At the very least, this implies that we would like $L < \xi_T$ corresponding to the temperature (less than the bulk $T_c$) at which the wires are to be used. Moreover, even when this inequality is satisfied, the coupling between two neighboring grains scales as $J \sim R^2/\lambda^3$. These considerations suggest that improved wires can be obtained by reducing the grain size and simultaneously increasing the concentration of grains subject to the condition $L/R \gtrsim 1$. In particular, there are likely regimes in which increasing $L$ causes a transition from the spin-glass to the globally s-wave regime, and correspondingly an increasing magnitude and decreasing time dependence of the critical current. One can readily make an estimate of the critical current density in the “globally s-wave” state is $I_c \sim GDR/\lambda^3$. We are not aware of any systematic theoretical studies of the critical current density in the in the superconducting XY glass phase. Indeed, as with other properties of a glass phase, the critical current density is surely time dependent.

Of course, this strategy has its limits – in order that the grains have undiminished local superconducting order, it is necessary that $R \gg \xi_0$, and that $R$ is large enough that quantum fluctuations of the superconducting phase are negligible. Subject to this, for the cuprates, it would be particularly interesting to explore the situation in which the London penetration depth, $\lambda$, is large compared to the grain size, $\lambda > L \sim R \gg \xi_0$, in which case the superfluid density of the wire would be homogeneous and isotropic, thus potentially mitigating some of the undesirable consequences of the quasi-2d nature of the cuprates.

Finally, another, rather obvious way to avoid frustration is to purposely construct “cigar-shaped” grains, which are then oriented in the composite. If this is done from a melt, it means that this must be a nematic suspension of the superconducting grains dissolved in a liquid metal. A wire made in this way would be globally d-wave.

![Fig. 2](image_url)

**Fig. 2.** Schematic representation of the sign of the anomalous average of the pair creation operator produced in the surrounding disordered metal by the proximity effect coupling to a grain of a d-wave superconductor. The symbol inside the grain represents the structure of $\langle \phi(p) \rangle$ inside the grain. The dashed lines represent typical pair diffusion paths the contribute to the proximity effect.

**ACKNOWLEDGMENTS**

We would like to acknowledge useful discussions with M. Beasley, M. Gingras, D. Huse, K. Moler and A. P. Young. We especially thank Aharon Kapitulnik for pointing out the relevance of the present theoretical considerations to HTC wires. This research was supported in part by Department of Energy under grant number DE-AC02-76SF00515 at Stanford (S.A.K.)

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As is conventional, we will ignore the dynamical fluctuations of the electromagnetic gauge fields, as issues related to Elizer’s theorem and gauge invariance are not important in the present context. In the limit of infinite on-site repulsion, typically considered in studies of the “$t-J$ model,” there are local kinematic constraints which cause $\phi(r,r')$ to vanish when $r$ is taken to represent the location of a particular Wannier state, independent of the symmetry of the pairing. In such a case, care must be taken to properly define $\phi$ as a suitably course-grained quantity, but this does not affect any of our conclusions.