Theory of c-axis Josephson tunneling in $d_{x^2−y^2}$-wave superconductors

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The temperature and angular dependence of the c-axis Josephson current and the superfluid density in layered $d_{x^2−y^2}$-wave superconductors are studied within the framework of an extended Ambegaokar-Baratoff formalism. In particular, the effects of angle-dependent tunneling matrix elements and Andreev scattering at grain boundaries are taken into account. These lead to strong corrections of the low-temperature behavior of the plasma frequency and the Josephson current. Recent c-axis measurements on the cuprate high-temperature superconductors $HgBa_2CaCu_2O_{8+δ}$ and $Bi_2Sr_2CaCu_2O_{8+δ}$ can therefore be interpreted to be consistent with a $d_{x^2−y^2}$-wave order parameter.

Interfaces such as grain boundaries in the high-temperature (high-$T_c$) superconductors are known to have important theoretical, technical, and practical implications. [1] In a set of early experiments it was shown that the critical current between single-crystal surfaces depends sensitively on their relative orientation. [2,3] More precisely, when two such samples are matched along their crystal a-b planes, but twisted with respect to each other about the direction of their common c-axis, it was found that the critical current decreases rapidly as a function of the twist angle $α$. This strong angular dependence has since been observed frequently at grain boundaries in the high-$T_c$ cuprates. It is a consequence of the $d_{x^2−y^2}$-wave symmetry of the superconducting order parameter in these compounds. [1]

FIG. 1. Schematic representation of two single-crystal samples, matched along their a-b planes and twisted about the c-axis by an angle $α$.

In the recent literature, there have been theoretical as well as experimental puzzles regarding the precise nature of c-axis Josephson tunneling in the cuprate superconductors, particularly in $Bi_2Sr_2CaCu_2O_{8+δ}$. On the theoretical front, several scenarios have been considered to model the angular behavior of the Josephson current. [4–8] However, in these frameworks it appears difficult to reproduce the strong dependence on the twist angle $α$ which has been observed in experiment. On the experimental side, there is a recent controversy arising from a c-axis Josephson tunneling experiment in which two single crystals were twisted about the c-axis and subsequently joined together. [7] In contrast to earlier experiments [2,3], the authors found a critical current that is very small but independent of $α$, implying that the superconducting order parameter in $Bi_2Sr_2CaCu_2O_{8+δ}$ has a dominant s-wave component. [7] However, it appears that the tunneling area in this particular measurement may have been too large to guarantee homogeneity of the contact areas. Furthermore, the small magnitude of the observed Josephson current suggests a large contribution from the surface area, which in turn would not adequately reflect the symmetry of the bulk superconducting order parameter. [8]

We will analyze a set of more recent experiments, which measure the c-axis Josephson current in superconducting $Bi_2Sr_2CaCu_2O_{8+δ}$. [9,10] Two single-crystal whiskers of this compound with very small widths $Φ \sim 10\mu m$ were joined along their a-b planes. By varying their relative orientations the angular dependence of the Josephson current along the c-axis was detected. A clear fourfold symmetry with respect to the twist angle $α$ was observed, consistent with a $d_{x^2−y^2}$-wave superconducting order parameter for this compound. Surprisingly, however, the angular dependence turned out to be much more pronounced than one would have expected from previous theoretical treatments, based on the Ginzburg-Landau model and on the Ambegaokar-Baratoff formalism for superconductor-insulator-superconductor (S-I-S) junctions, which predict a simple $\cos (2α)$ dependence for $d_{x^2−y^2}$-wave superconductors. [11]

Here we propose an extension of the Ambegaokar-Baratoff treatment for the superfluid density and the Josephson current along the c-axis of layered unconventional superconductors, taking into account (i) the coherent component of the tunneling current, (ii) the $d_{x^2−y^2}$-wave symmetry of the order parameter, and (iii) the angular dependence of the transfer matrix element along the crystal c-direction.

Let us first discuss the issue of coherence. In contrast to the case of s-wave superconductivity, Josephson coupling between two unconventional superconductors with nodes in their gap functions is possible only if there is

75.50.+r, 74.25.Nf, 74.20.Mn, 74.72.Hs
a coherent component to the tunneling current. Hence, purely incoherent tunneling will not result in a Josephson coupling between two \(d_{x^2-y^2}\)-wave superconductors, as already noted in Refs. [4] and [12], and for the layered \(\kappa-(ET)_2\) salts in Refs. [13] and [14]. Coherent tunneling between adjacent layers with planar quasiparticle wave vectors \(\mathbf{k}\) and \(\mathbf{k}'\) can be defined by the condition \(\mathbf{k} \parallel \mathbf{k}'\). This is what Ambegaokar and Baratoff called “specular transmission”. [11,15] On the other hand, if the tunneling is incoherent \(\mathbf{k} \neq \mathbf{k}'\), i.e. \(\mathbf{k}\) and \(\mathbf{k}'\) are completely uncorrelated. For the sake of the following argument, let us consider only these two extreme cases. In real materials, coherent and incoherent processes are typically mixed, in which case only the coherent component is relevant. The Josephson coupling is proportional to \(\langle \Delta(\phi)\Delta(\phi') \rangle\), where \(\langle \cdot \cdot \cdot \rangle\) represents angular averages over \(\phi\) and \(\phi'\), which are the angles between the wave vectors \(\mathbf{k}\) and \(\mathbf{k}'\) and the crystal a-axis. Hence, when \(\mathbf{k}\) and \(\mathbf{k}'\) are uncorrelated \(\langle \Delta(\phi)\Delta(\phi') \rangle = \langle \Delta(\phi) \rangle \langle \Delta(\phi') \rangle = 0\). This argument applies not only to \(d_{x^2-y^2}\)-wave systems, but also to other unconventional superconductors. In contrast, there will be Josephson coupling between two \(s\)-wave superconductors even when the tunneling is completely incoherent, and therefore this is one of the important differences between conventional and unconventional superconductivity.

When the transport along the c-axis is due to coherent tunneling, the temperature dependence of the out-of-plane superfluid density \(\rho_{sc}\) is different from that of the in-plane superfluid density \(\rho_{s||}\). Recent calculations performed for anisotropic three-dimensional models have predicted a linear decrease with increasing temperature, both for \(\rho_{sc}\) and \(\rho_{s||}\). [16,17] However, within the tunneling model, the out-of-plane superfluid density for \(d_{x^2-y^2}\)-wave superconductors is given by

\[
\rho_{sc}(t) = \frac{\Delta(t)}{\Delta_0} \int_0^{\pi/2} d\phi \sin \phi \tanh \left( \frac{\Delta(t) \sin \phi}{2T} \right) \approx 1 - 1.645 \left( \frac{T}{\Delta_0} \right)^2 - 3.606 \left( \frac{T}{\Delta_0} \right)^3,
\]

where \(t = T/T_c\) is the reduced temperature and \(\Delta(t)\) is the amplitude of the superconducting gap function. Thus the tunneling model predicts a \(t^2\)-dependence at low temperatures, consistent with the out-of-plane superfluid density observed in YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) [18], Tl\(_2\)Ba\(_2\)CuO\(_6\) [19], and \(\kappa-(ET)_2\)Cu(N(CN))\(_2\)Br [13].

More recently, the out-of-plane superfluid density in Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\) was measured, using a sensitive microwave resonance technique. [20] Noting that the square of the Josephson plasma frequency is proportional to \(\rho_{sc}(t)\), it was observed that the low-temperature behavior of the c-axis superfluid density is proportional to \(t^5 \sim t^6\). A comparable high power-law has also been reported in HgBa\(_2\)CaCu\(_{1+\delta}\). [21] In order to describe this behavior, Xiang and Wheatley [22] introduced an angle-dependent tunneling matrix element proportional to \(\cos^2(2\phi)\), which vanishes along the nodal directions. This behavior is common to those high-\(T_c\) cuprates which do not contain Cu-O chains. It is caused by the overlap of Cu 4s and O 2p orbitals. [20,22] Introducing an angle-dependent tunneling matrix element into the formalism for the superfluid density leads to

\[
\rho_{sc}(t) = \frac{15\Delta(t)}{8\Delta_0} \int_0^{\pi/2} d\phi \sin^5 \phi \tanh \left( \frac{\Delta(t) \sin \phi}{2T} \right) \approx 1 - 3.606 \left( \frac{T}{\Delta_0} \right)^3 - 443.35 \left( \frac{T}{\Delta_0} \right)^6.
\]

While the leading power appears to be \(t^3\), comparison of the coefficients of the \(t^3\) and \(t^6\)-terms shows the \(t^6\)-power to be dominant at all experimentally relevant low temperatures.

![FIG. 2. Out-of-plane superfluid density as a function of temperature. The solid line includes the angular dependence of the tunneling matrix element along the c-direction, whereas the dashed line does not. The inset shows the low-temperature region.](attachment:image.png)
Baratoff formalism, and assuming that there is a component of coherent tunneling along the c-direction, one obtains

\[
I_c(T, \alpha) = \frac{\Delta_2^2(t)}{\Delta_0} \int_0^{2\pi} d\phi \sum_{n=0}^{\infty} \frac{T f_> f_<}{\sqrt{\omega_n^2 + \Delta_0^2} |f_>|^2 \sqrt{\omega_n^2 + \Delta_0^2} |f_<|^2},
\]

(3)

where \(\omega_n = 2\pi T(n + 1/2)\) is the Matsubara frequency, and \(t_\perp\) is the tunneling matrix element along the c-direction. The angular dependence of the gap function is contained in the factors

\[
\begin{align*}
  f_> & = \cos (2\phi), & f_< & = \cos (2(\phi + \alpha)), \\
  & \text{if } |\cos (2\phi)| > |\cos (2(\phi + \alpha))|, \\
  f_> & = \cos (2(\phi + \alpha)), & f_< & = \cos (2\phi), \\
  & \text{if } |\cos (2\phi)| < |\cos (2(\phi + \alpha))|.
\end{align*}
\]

(4)

(5)

The factor in the denominator \(1 + (\Delta_0/t_\perp)^2(|f_>| - |f_<|)^2\) includes the effect of Andreev reflections [23] at the grain boundary. If this factor is neglected, one obtains the usual Ambegaokar-Baratoff expression, with a dependence of the Josephson current on the twist angle very close to \(\cos(2\alpha)\). Furthermore, one notes that this correction of the Josephson current due to Andreev scattering vanishes if \(\alpha = 0\), i.e., when the two whiskers are perfectly aligned.

Taking into account the additional angular dependence of the tunneling matrix element in the above derivation leads to a slightly modified expression, analogous to the discussion of the superfluid density,

\[
I_c(T, \alpha) = \frac{\Delta_2^2(t)}{\Delta_0} \int_0^{2\pi} d\phi \sum_{n=0}^{\infty} \frac{T f_> f_<}{1 + (\Delta_0/t_\perp)^2(|f_>| - |f_<|)^2} \times \frac{15|f_> f_<|^2}{8\sqrt{\omega_n^2 + \Delta_0^2} |f_>|^2 \sqrt{\omega_n^2 + \Delta_0^2} |f_<|^2}.
\]

(6)

Let us first discuss the angular dependence of the zero-temperature limit of Eqs. 3 and 6, given by

\[
I_c(T = 0, \alpha) = \frac{1}{2\pi} \int_0^{2\pi} \frac{f_> f_<}{|f_>|^2} K \left(\sqrt{1 - \left(\frac{f_<}{f_>}\right)^2}\right) \frac{F(\phi)}{1 + (\Delta_0/t_\perp)^2(|f_>| - |f_<|)^2}.
\]

(7)

where \(K(k)\) is the complete elliptic integral of the first kind. Here \(F(\phi) = 1\) for the case of angle-independent tunneling along the c-axis (Eq. 3), and \(F(\phi) = |f_< f_>|^2\) when this angular dependence is included (Eq. 6).

Fig. 3 shows the angular dependence of the \(c\)-axis Josephson current for various values of the ratio of the gap amplitude to the tunneling matrix element \(\Delta_0/t_\perp\). Data from the recent cross-whisker measurements on \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}\) by Takano et al. [10] are also shown as filled circles. In Fig. 3(a) the tunneling element is assumed to be constant. Therefore the angular dependence is less pronounced than in Fig. 3(b) where an additional \(\cos^2(2\phi)\) modulation of \(t_\perp\) has been included. Comparing with the experimental data, both models appear to fit the strong angular dependence of the measured current reasonably well with \(\Delta_0/t_\perp \sim 10\) for \(F(\phi) = 1\), and \(\Delta_0/t_\perp \sim 20\) for \(F(\phi) = |f_< f_>|^2\). However, the rapid decay of \(I_c(T = 0, \alpha)\) appears to be better captured by the model which includes the additional angular modulation of the tunneling matrix element. In this comparison with the experiment we have assumed that the data point at \(\alpha = 0.488\pi\) which violates the otherwise monotonic behavior in \(I_c(T = 0, \alpha)\) is of extrinsic origin, perhaps related to inhomogeneities in the contact surfaces.

**FIG. 3.** Dependence of the zero-temperature \(c\)-axis Josephson current on the twist angle \(\alpha\). Solid lines: calculated \(I_c(T = 0, \alpha)\) according to Eq. 7, (a) with \(F(\phi) = 1\), and (b) with \(F(\phi) = |f_< f_>|^2\). From top to bottom the ratio of the gap amplitude to the \(c\)-axis tunneling matrix element is given by \(\Delta_0/t_\perp = 0, 1, 2, 5, 10, 20,\) and 50. Circles: data from Ref. 10.

The most important conclusion from the above considerations is that Andreev scattering has a crucial role in determining the strong angular dependence of the Josephson coupling across grain boundaries. As observed in Fig. 3, the additional factor \((\Delta_0/t_\perp)^2(|f_>| - |f_<|)^2\) leads to substantial deviations from the simple proportionality \(I_c(T, \alpha) \propto \cos(2\alpha)\), which one obtains without taking Andreev processes into account. This observation is also related to the question of whether these grain boundaries are S-N-S or S-I-S junctions. The present study indicates that they seem to be something in between.

Finally, let us use the above formalism to make some predictions regarding the temperature dependence of the \(c\)-axis Josephson tunneling current for various twist angles. From the discussion of the angular dependence it is expected that the current exhibits peaks at \(\alpha = 0\) and \(\pi/2\), and vanishes at \(\alpha = \pi/4\) and \(\alpha = 3\pi/4\). In anal-
ogy to the data presented in Ref. [10], we show in Fig. 4 the temperature dependence for selected twist angles in the region $\alpha \in [\pi/4, \pi/2]$. The ratio $\Delta_0/t_\perp$ was fixed according to the best fits obtained in Figs. 3(a) and (b). One observes in Fig. 4 that while the main temperature dependence is governed by the prefactor $\Delta^2(t) \sim (1 - t^\alpha)$ in Eqs. 3 and 6, the most noticeable differences between the two cases, $F(\phi) = 1$ vs. $F(\phi) = |f_{c}^{s}f_{s}^{c}|^2$, occur in the low-temperature regions. Moreover, they are more pronounced at large twist angles close to $\alpha = \pi/2$. Also, exactly at $\alpha = \pi/2$ the expression for the Josephson current reduces to $\rho_{sc}(t)$. Comparison of future experimental data with these curves will aid in distinguishing between the proposed models.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig4}
\caption{Temperature dependence of the c-axis Josephson current. From top to bottom the twist angle is $\alpha = \pi/2, 0.45\pi, 0.4\pi, 0.35\pi$, and $0.3\pi$. (a) $F(\phi) = 1$ and $\Delta/t_\perp = 10$. (b) $F(\phi) = |f_{c}^{s}f_{s}^{c}|^2$ and $\Delta/t_\perp = 20$.}
\end{figure}

In conclusion, we have studied the angular and temperature dependence of the superfluid density and the Josephson current across c-axis grain boundaries, such as the recently synthesized twisted cross-whisker junctions of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. For both quantities, $\rho_{sc}(t)$ and $I_c(T, \alpha)$, the inclusion of an additional $\cos^2(2\phi)$ angular modulation in the tunneling matrix element is important for accurate modeling of the low-temperature properties of nodal cuprate superconductors without Cu-O chains. The observed strong angular dependence of the Josephson current, deviating substantially from a simple $\cos(2\alpha)$ behavior, can be accounted for by Andreev scattering processes at the grain boundaries. Therefore the measured Josephson current across whisker surfaces in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ appears to be fully consistent with a $d_{x^2-y^2}$-wave symmetry of the superconducting order parameter.

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