Letter Editors' Suggestion

Inhomogeneity-induced time-reversal symmetry breaking in cuprate twist junctions

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The lowest-order Josephson coupling, $J_1(\theta)\cos(\phi)$, between two d-wave superconductors with a phase difference ϕ across the junction vanishes when their relative orientation is rotated by $\theta = \pi/4$. However, in the presence of inhomogeneity, $J_1(\mathbf{r})$ is nonzero locally, with a sign that fluctuates in space. We show that such a random J_1 generates a global second-harmonic Josephson coupling, $J_2\cos(2\phi)$, with a sign that favors $\phi = \pm\pi/2$, i.e., spontaneous breaking of time-reversal symmetry. The magnitude of J_2 is substantially enhanced if the spatial correlations of $J_1(\mathbf{r})$ extend over large distances, such as would be expected in the presence of large-amplitude twist-angle disorder or significant local electronic nematicity. We argue that this effect likely accounts for the recent observations in twisted Josephson junctions between high-temperature superconductors.

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Twisting and stacking two-dimensional quantum materials has proven to be an extremely powerful tool, both in creating new interesting materials [1–3] and in probing their properties [4,5]. In particular, it has long been proposed that measuring the Josephson coupling between two d-wave superconductors as a function of the twist angle θ between them can reveal the pairing symmetry. Early experiments in the cuprate high-temperature superconductors [6] did not observe the predicted angle dependence of the Josephson coupling, in apparent contradiction to the known d-wave order parameter symmetry. In a notable set of recent experiments [7], a $|\cos(2\theta)|$ dependence was finally observed, possibly resolving this puzzle. However, still more recent experiments [8] have not reproduced this result. The source of these discrepancies remains an open question.

Here, we assume that there is an extrinsic explanation for this apparent non-d-wave behavior, and focus on the experiments [7] that show the expected angle dependence. In particular, for $\theta = \pi/4$, the lowest-order Josephson coupling J_1 vanishes by symmetry. It was recently predicted [9] that the second-order Josephson coupling J_2 (corresponding to an interplane coherent tunneling of two Cooper pairs) favors a spontaneously broken time-reversal symmetry (TRS) state, where the phase difference between the two superconductors is $\pm\pi/2$. Indeed, the experiment in Ref. [7] found evidence that at $\theta = \pi/4$, there is a substantial second-order Josephson coupling, revealed by measuring additional half-integer Shapiro steps. The precise microscopic mechanism of this second-order coupling remains to be clarified. Also still to be explored experimentally is the theoretical proposal that this could serve as a platform to realize chiral topological superconductivity [9–13], although the gap of the resulting state may be very small [14].

However, as we will discuss, the extreme anisotropy of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212)—the cuprate superconductor used in the twist-junction experiments—poses a significant quantitative difficulty with the intrinsic mechanism for generating J_2. Specifically, an estimate based on measured quantities in bulk crystals implies an intrinsic value of J_2 that is too small to explain the experiments.

In this Letter, we propose an alternative mechanism for the generation of J_2, a form of "order from disorder" driven by the effect of local spatial symmetry breaking. In Fig. 1(a) we schematically show a sample consisting of two d-wave superconductors rotated with respect to each other by an angle θ. We consider the case where each superconductor contains N layers, and we label them by the index n which extends from $-N+1$ to N. We neglect the effect of inhomogeneity on all layers but those with $n = 0, 1$ (which are separated by the twist junction), and assume that the Josephson coupling per unit area between these layers has the form

$$J_1 = \overline{J_1(\theta)} + \delta J_1(\boldsymbol{r}). \tag{1}$$

Here $\overline{J_1(\theta)}$ respects the d-wave symmetry so that it vanishes at $\theta = \pi/4$, while the random in sign quantity δJ_1 is generated by a local, sample-specific point-group-symmetry breaking, such that $\overline{\delta J_1(\boldsymbol{r})} = 0$. Here, the overline denotes configuration averages over a random ensemble of $\delta J_1(\boldsymbol{r})$.

The fluctuating component $\delta J_1(\boldsymbol{r})$ may arise from different physical sources. One possible source is spatial variations of the twist angle $\delta\theta(\boldsymbol{r})$ or any other form of disorder (including potential disorder) that breaks the local point group symmetries. A local admixture of an s-wave gap could also arise from "electron nematic domains." We will comment on these possibilities further in the discussion section below.

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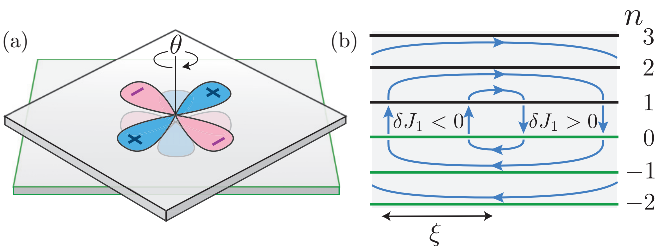

FIG. 1. (a) A Josephson junction made of two d-wave superconductors with a relative twist angle θ. For $\theta = \pi/4$, the lowest-order Josephson coupling J_1 vanishes by symmetry. (b) Cross section of the junction region. The layers of the two superconductors are labeled by n. The twist junction is between the $n = 0$ and $n = 1$ layers. The Josephson coupling between these layers is given by Eq. (1), and its spatial average vanishes at $\theta = \pi/4$. The Josephson coupling $\delta J_1(\boldsymbol{r})$ is correlated over a length ξ. The ground state breaks time-reversal symmetry spontaneously, and is characterized by circulating persistent current loops, illustrated by blue arrows.

We will characterize the fluctuations of J_1 by a correlation function

$$\overline{\delta J_1(\boldsymbol{r})\delta J_1(\boldsymbol{r}')} = \overline{\delta J_1(\boldsymbol{r})^2}\, f\left(\frac{|\boldsymbol{r}-\boldsymbol{r}'|}{\xi}\right). \tag{2}$$

Here, $f(\boldsymbol{x})$ is a dimensionless function normalized such that $f(\boldsymbol{0}) = 1$,[1] and $\int d^2x f(\boldsymbol{x}) = 1$,[2] and ξ is the correlation length of $\delta J_1(\boldsymbol{r})$.

The local interlayer superconducting phase difference between the $n = 0$ and $n = 1$ layers, $\phi(\boldsymbol{r}) = \phi_1(\boldsymbol{r}) - \phi_0(\boldsymbol{r})$, exhibits spatial fluctuations

$$\phi(\boldsymbol{r}) = \bar{\phi} + \delta\phi(\boldsymbol{r}), \tag{3}$$

which are correlated with the fluctuations of the critical current density $\delta J_1(\boldsymbol{r})$. It is easy to show that at $\theta = \pi/4$ the ground state of the system corresponds to $\bar{\phi} = \pm\pi/2$. Indeed, for $\bar{\phi} = \pm\pi/2$, the system gains energy linearly in $\delta\phi$ by adjusting the phase difference across the junction to the sign of the local Josephson coupling $\delta J_1(\boldsymbol{r})$. The energy cost of the resulting in-plane currents is only quadratic in $\delta\phi$. This simple argument also implies the violation of the time-reversal symmetry, and existence of *local* circulating supercurrents and associated magnetic fields, illustrated in Fig. 1(b). Time-reversal symmetry is broken in an interval of twist angles near $\theta = \pi/4$.

Importantly, in the cuprates, the phase stiffness is strongly anisotropic, with the c-axis stiffness much smaller than the in-plane one. As a result, there is a characteristic length in the problem (treating, for simplicity, the case of $N = 1$—the case of $N \gg 1$ is discussed below)

$$\ell^2 = \frac{\kappa}{\left(\overline{\delta J_1^2}\right)^{1/2}}, \tag{4}$$

where κ is the in-plane phase stiffness. There are two regimes depending on ξ/ℓ: In the case $\xi \gg \ell$ the system breaks into large domains with $\phi \approx 0$ or π, separated by domain walls. Time-reversal symmetry is broken in the vicinity of the domain walls where the phase twists from 0 to π. In this case, the critical current through the junction is of the order of $(\overline{\delta J_1^2})^{1/2}$. Below we show that in the opposite limit, $\xi_{sc} \ll \xi \ll \ell$ (where ξ_{sc} is the superconducting coherence length), the energy-phase relation has the form

$$E(\bar{\phi}) = -\overline{J_1(\theta)}\cos(\bar{\phi}) - J_2\cos(2\bar{\phi}), \tag{5}$$

where J_2 increases with ξ [see Eqs. (13) and their derivation below]. For $\xi \sim \ell$, we obtain

$$|J_2| \sim \left|J_2^{\max}\right| \sim \sqrt{\overline{\delta J_1^2}}. \tag{6}$$

Thus, our mechanism is capable of producing a J_2 that is as large as the magnitude of the local (random) Josephson coupling, given that the correlation length of the local Josephson coupling is sufficiently large.

In the presence of an in-plane magnetic field $B_\parallel$, another length scale arises, $l_B = \Phi_0/(B_\parallel w)$, where w is the smaller of the perpendicular penetration depth and the thickness of the device, and Φ_0 is the superconducting flux quantum. As long as l_B is larger than ξ, we expect the system to behave essentially as a Josephson junction with a spatially uniform J_1 and J_2. In particular, at $\theta = \pi/4$, the expected Fraunhofer pattern will reflect the doubled periodicity of the energy-phase relation. At higher fields (such that $l_B < \xi$) a more complicated interference pattern will directly reflect the inhomogeneity of the Josephson coupling [15].

Estimate of the intrinsic J_2. As a first exercise, we estimate the intrinsic second-harmonic Josephson coupling J_2 generated by the coherent tunneling of two Cooper pairs, neglecting the effects of inhomogeneity, and argue that the intrinsic J_2 is too small to account for the experiment of Ref. [7].

The measured values of the in-plane and out-of-plane penetration depths for optimally doped Bi-2212 are $\lambda_\parallel \sim 200$ nm and $\lambda_\perp \sim 100$ μm, respectively [16], corresponding to an anisotropic superfluid density $\kappa_\perp/\kappa_\parallel = (\lambda_\parallel/\lambda_\perp)^2 \sim 4 \times 10^{-6}$. This large anisotropy reflects the extremely two-dimensional (2D) electronic structure. To estimate the consequences of this for the expected ratio of J_2/J_1, we assume that the interlayer tunneling is not momentum conserving (the "incoherent tunneling model" [17]), which reproduces the approximately $\cos(2\theta)$ dependence of J_1 observed in the experiments.

For $\theta \sim 0$, $J_1(0)$ (which has units of energy per unit area) is determined from an appropriate average of the interlayer hopping $t_\perp$ by an Ambagoakar-Baratoff-like relation

$$J_1(0) = g_1|t_\perp|^2\frac{\nu^2}{k_F^2}|\Delta|, \tag{7}$$

where ν is the density of states at the Fermi energy, k_F is the Fermi wave vector, $|\Delta|$ is the size of the maximal gap, and g_1 is a dimensionless factor that depends, among other things, on the structure of the tunneling matrix elements in momentum space [17,18]. Similar reasoning gives for the intrinsic

[1] The normalization is necessary for the equation to hold when $\boldsymbol{r} = \boldsymbol{r}'$.

[2] When normalized, integrating $\overline{\delta J_1(\boldsymbol{r})\delta J_1(\boldsymbol{r}')}$ over $\boldsymbol{r}'$ is equal to $\xi^d\overline{\delta J_1(\boldsymbol{r})^2}$ in d dimension (here, $d = 2$). This is the correct disorder strength since site $\mathbf{r}$ is correlated to an "effective" region of size ξ^d.

second-order coupling

$$J_2 = g_2 |t_\perp|^4 \frac{\nu^3}{k_F^4}, \tag{8}$$

where g_2 is another dimensionless factor, and presumably J_2 is approximately independent of θ.

To relate these J_1 and J_2 to measured quantities, we assume that the coupling between planes in a bulk crystal is the same as for the twist junction with $\theta \sim 0$, so that we can identify $\kappa_\perp = J_1(0)d$, where d is the interbilayer distance. [For further consistency checks regarding the estimate of J_1 and J_2, see Supplemental Material (SM) [19].] Moreover, the in-plane superfluid stiffness empirically satisfies $\kappa_\parallel = g_\parallel T_c/d$ where $g_\parallel$ is of order one. Therefore

$$\left(\frac{J_2}{J_1(0)}\right) = \left(\frac{g_\parallel g_2}{g_1^2}\right)\left(\frac{\xi_{\rm sc}}{k_F d^2}\right)\left(\frac{T_c}{|\Delta|}\right)\left(\frac{\kappa_\perp}{\kappa_\parallel}\right). \tag{9}$$

Taking into account the fact that in optimally doped cuprates T_c and $|\Delta|$ are of the same order of magnitude, and so are $1/k_F$, d, and $\xi_{\rm sc}$, and ignoring the unknown numerical prefactors, we get that the intrinsic value of J_2/J_1 should be of the order of 10^{-6}–10^{-5}. In contrast, the measured value of J_2/J_1 reported in Ref. [7] is $\sim 10^{-2}$. Thus, unless for some reason g_1 is anomalously small, or g_2 unexpectedly large, the intrinsic effect is too small to account for the cuprate experiment.

Inhomogeneity-induced J_2. We use the following model to describe two layered superconductors coupled by a twisted Josephson junction,

$$\mathcal{H} = \sum_n \int d^2r \left[\frac{\kappa}{2}(\nabla\phi_n)^2 - \mathcal{J}^{(n)} \cos(\phi_n - \phi_{n-1})\right], \tag{10}$$

where $\phi_n(\boldsymbol{r})$ is the superconducting phase in layer n at position $\boldsymbol{r} = (x, y)$, and $\kappa = \kappa_\parallel d$ is the phase stiffness in the plane. Each superconductor contains N layers, and the sum over n extends from $-N+1$ to N. For $n \neq 1$, the interplane coupling per unit area is $\mathcal{J}^{(n)} = J_1(0)$, related to the three-dimensional phase stiffness by $J_1(0) = \kappa_\perp/d \equiv J_\perp$. For $n = 1$, the Josephson coupling is $\mathcal{J}^{(1)} = J_1(\boldsymbol{r})$, given by Eq. (1).

Note that we ignore magnetic screening throughout the analysis, neglecting the coupling of the superconducting phase to the electromagnetic field. This assumption is justified as long as the thickness of the system is much smaller than $\lambda_\perp$, and the lateral size is smaller than the Josephson penetration depth of the twist junction.

We analyze the system at $T = 0$, minimizing the energy (10). A similar analysis was performed in a 3D case in Ref. [20]; here, we generalize the analysis for the case of layered superconductors and a finite correlation length of the local Josephson coupling $\delta J_1(\boldsymbol{r})$.

For $\xi \ll \ell$ [where ℓ is defined in Eq. (4) for $N = 1$ and in Eq. (14) below for $N \to \infty$], the superconducting phase is nearly uniform within each of the two superconductors. We therefore write the superconducting phase as $\phi_n(\boldsymbol{r}) = s_n\bar{\phi}/2 + \delta\phi_n(\boldsymbol{r})$, where $s_n = 1$ for $n > 0$ and -1 otherwise ($\bar{\phi}$ is the average phase difference), and expand the energy up to second order in $\delta\phi_n(\boldsymbol{r})$. Minimizing the resulting expression, we obtain the energy-phase relation of the junction per unit area given by Eq. (5) with the second-order Josephson coupling is given by

$$J_2 = -\int \frac{d^2q}{(2\pi)^2} \frac{|\delta J_1(\boldsymbol{q})|^2}{\kappa q^2 + J_\perp \frac{1-e^{-\eta_q}+\alpha_q(1-e^{\eta_q})}{1+\alpha_q}}. \tag{11}$$

(See Supplemental Material [19] for further details.) Here, $\delta J_1(\boldsymbol{q})$ is the Fourier transform of $\delta J_1(\boldsymbol{r})$, $\eta_{\boldsymbol{q}} = \cosh^{-1}[1 + \kappa q^2/(2J_\perp)]$, and

$$\alpha_{\boldsymbol{q}} = e^{-2\eta_q(N-1)} \frac{J_\perp(e^{\eta_q} - 1) - \kappa q^2}{J_\perp(1 - e^{-\eta_q}) + \kappa q^2}. \tag{12}$$

Crucially, the sign of J_2 is *negative*. Therefore, at twist angle $\theta = \pi/4$ [such that $J_1(\theta) = 0$], the ground state corresponds to $\phi = \pm\pi/2$. Time-reversal symmetry is broken in a range of twist angles around $\pi/4$, such that $|J_2| > |J_1(\theta)|/4$. We provide explicit expressions for J_2 in two cases:

$$J_2 \approx -\frac{\overline{\delta J_1^2}\xi^2}{2\pi\kappa} \begin{cases} \ln\left(1 + \frac{1}{\xi}\sqrt{\frac{\kappa}{J_1(0)}}\right), & N \to \infty, \\ \ln\left(\frac{\kappa}{\left(\overline{\delta J^2}\right)^{1/2}\xi^2}\right), & N = 1. \end{cases} \tag{13}$$

The expressions are valid if ξ is sufficiently short: $\xi \ll \ell$, such that $\delta\phi_n(\boldsymbol{r}) \ll \pi$. Beyond this regime ($\xi > \ell$), the perturbative calculation breaks down [see discussion around Eq. (4)]. The length scale ℓ can be identified as the value of ξ where $J_2 \sim (\overline{\delta J_1^2})^{1/2}$. Using Eq. (13), this gives Eq. (4) for $N = 1$ (up to the logarithmic factor, which is of the order of unity in this case), whereas for $N \to \infty$ we find

$$\ell^2 \sim \frac{\kappa J_1(0)}{\overline{\delta J_1^2}}. \tag{14}$$

For $N = 1$, the derivation of Eq. (13) contains a subtlety—the integral in Eq. (11) is infrared divergent. This logarithmic divergence occurs also for any finite N, and signals a breakdown of the perturbative treatment in δJ_1. A more accurate treatment (see Supplemental Material [19]) reveals that the divergence is cut off by an emergent length scale,[3] of the order of ℓ^2/ξ, which leads to Eq. (13).

Importantly, comparing Eq. (13) to Eq. (9), we find that the inhomogeneity-induced J_2 differs from the intrinsic one by a factor of

$$\frac{J_2}{J_2^{\rm int}} \sim \frac{\overline{\delta J_1^2}}{J_1^2(0)}\left(\frac{\xi}{\xi_{\rm sc}}\right)^2, \tag{15}$$

where we have dropped all factors of order one.

Discussion. The vanishing of $J_1(\theta)$ as $\theta \to \pi/4$ and the existence of a higher-order Josephson coupling J_2 that is nonvanishing under the same conditions follow from the d-wave symmetry of the superconducting (SC) order. There are, generically, both intrinsic and extrinsic (disorder-related) contributions to $J_2(\theta)$. However, given the extremely small c-axis superfluid density in the particular material of interest (Bi-2212), the observed [7] magnitude $|J_2(\pi/4)/J_1(0)| \sim 10^{-2}$ is notably large. We have estimated that the intrinsic contribution is of order $|J_2^{\rm int}(\pi/4)/J_1(0)| \sim 10^{-5}$–$10^{-6}$ [Eq. (9)].

[3]To be exact, a length scale of the order $\ell^2/\xi \times [\ln(\ell/\xi)]^{-1/2}$; see SM [19].

What we have shown is that an extrinsic mechanism—one that derives from a fluctuating in space but random in sign first-order coupling $J_1(\boldsymbol{r})$—can produce an effect of the requisite magnitude but only under rather extreme circumstances, i.e., when $\overline{\delta J_1^2}/J_1^2(0)$ is sufficiently large *and* the correlation length ξ is relatively long. For instance, we can explain the observed value of $|J_2/J_1(0)|$ if we assume $\overline{\delta J_1^2}/J_1^2(0) = 0.1$ and $\xi/\xi_{sc} \sim 10^2$–10^3.

The existence of a nonvanishing $\delta J_1(\boldsymbol{r})$ when globally $\theta = \pi/4$ requires a local breaking of mirror symmetries (where the mirror plane is perpendicular to the system). Twist-angle disorder leads to such symmetry breaking. However, to produce an effect of the requisite magnitude observed in Ref. [7], either the distribution of the twist angles should be a substantial fraction of $\pi/4$, or their correlation length should be larger than the sample size (about 10 μm). Some source of substantial shear-strain disorder might also be relevant, but there would need to be some reason to believe that such strain would strongly perturb the local pairing symmetry.

We thus consider the most likely origin of the requisite disorder is to be pinned domains of an otherwise intrinsic electron-nematic order [21]. There exists strong—although not universally accepted—evidence of a tendency toward nematic order in the cuprates [22–26]. In particular, in Bi-2212, there is direct evidence from scanning tunneling microscopy (STM) [25,27–30] of local nematic order which is seen strongly and primarily at energies of order of the gap maximum (i.e., at energies at which the density of states in the superconducting state exhibits a maximum). This implies that the local nematicity has a strong effect on the local superconducting order parameter. Moreover, the fact that signatures of nematic symmetry breaking remain strong when STM features are spatially averaged over the field of view suggests a correlation length larger than the field of view, i.e., $\xi_{nem} > 100a$ (where a is the lattice spacing). In addition, a recent STM study [29] on a related material (Bi-2201) also provides evidence of long-range nematic correlations [31].

There are several testable consequences of various aspects of the above line of reasoning:

(1) An extrinsic effect depends strongly on ξ and $\overline{\delta J_1^2}$, which could well depend on details of sample preparation. This, in turn, might explain the already mentioned fact that the twist-angle dependence is not universally observed [8].

(2) The time-reversal breaking phase should have spontaneous circulating currents, as shown in Fig. 1(b). The expected order of magnitude of the in-plane magnetic moment is $m_\parallel \sim (2e/\hbar)\,(\overline{\delta J_1^2})^{1/2}\xi^3 d$. Taking $(\overline{\delta J_1^2})^{1/2} \sim 0.3\,J_1(0) \sim 1.5\times10^{-5}$ meV/nm^2, $\xi = 200$ nm, and $d = 1.5$ nm gives $m_\parallel$ of the order of a few μ_B. The associated magnetic fields should be observable near the edges of the system, and have a random sign.

(3) From STM [27] and other [32] studies, it appears that the nematicity in Bi-2212 vanishes (or at least becomes much weaker) for hole doping concentrations larger than a critical value, $p^\star \approx 0.19$. Thus, a correspondingly strong doping dependence of the magnitude of J_2 would be expected if electron nematicity plays a role in the effect [33].

From a symmetry point of view, the state of the system at $\theta = \pi/4$ is, as has been previously noted [9], a $d + id$ superconductor. In the absence of disorder (assuming that J_2 is generated by the intrinsic mechanism), this state should be fully gapped—although for numbers relevant to Bi-2212 this induced nodal gap Δ_{ind} would likely be immeasurably small. For the extrinsic case, there is an interesting question of principle whether this state is gapped or gapless. Potential disorder is expected to suppress the gap. The gap is further reduced by local Doppler shifts of the quasiparticle energies (Volovik effect [34]) due to the presence of equilibrium currents, $\delta E(\boldsymbol{r}) = v_F\, j_s(\boldsymbol{r})/\kappa$. If this energy shift is larger than $\Delta_{ind}(\boldsymbol{r})$, the gap closes.

Finally, we note that the present considerations are not confined to the cuprates. In less anisotropic systems, there may be circumstances in which the intrinsic effect dominates, and in which the resulting state at $\theta = \pi/4$ is fully gapped, as suggested in Ref. [9]. It is also worth noting that very similar considerations apply to a junction between an unconventional superconductor (e.g., a d-wave superconductor) and a conventional s-wave superconductor, without need for twist-angle engineering.

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