Spatially Indirect Exciton Condensation in Superconductor Bilayers

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We look at the possibility of spatially indirect exciton condensation in electrically isolated superconducting bilayers. We find that in a mean-field approximation bilayers can have superconducting and excitonic order simultaneously if repulsive interactions between layers are sufficiently strong. The excitonic order implies interlayer phase coherence, and can be conveniently studied in a representation of symmetric and antisymmetric bilayer states. When both orders are present we find several solutions of the mean-field equations with different values of the the symmetric and antisymmetric state pair amplitudes. The mixed state necessarily has non-zero pair amplitudes for electrons in different layers in spite of the repulsive interlayer interactions, and these are responsible for spatially indirect Andreev reflection processes in which an incoming electron in one layer can be reflected as a hole in the opposite layer. We evaluate layer diagonal and off-diagonal current-voltage relationships that can be used to identify this state experimentally.

I. INTRODUCTION

Superconductivity is perhaps the most spectacular example of collective behavior of electrons. The microscopic BCS\textsuperscript{1,2} theory of the phenomena explains superconductivity in terms of bound (Cooper) electron pairs due to attractive interactions, mediated in the best understood case by phonons. Condensation of Cooper pairs gives rise to a non-Fermi-liquid state with spontaneously broken $U(1)$ symmetry characterized by a homogenous complex order parameter $\Delta$. One experimental manifestation of superconductivity is a non-Ohmic junction resistance with a normal metal. At subgap values of the bias voltage on the interface, the only way for a single electron to penetrate inside a superconductor is to form a Cooper pair. Therefore, by charge conservation, transmission through the interface should be accompanied by reflection of a hole, a process known as Andreev\textsuperscript{3} reflection. Studies of Andreev reflection can be used to probe\textsuperscript{4,5} a state of interest and continue to gather both theoretical\textsuperscript{6–13} and experimental\textsuperscript{14–17} attention.

In recent years experimentalists have uncovered examples of both electron-electron (Cooper) and electron-hole (excitonic) pairing in atomically thin two-dimensional materials\textsuperscript{18–23}. In general Cooper pairing is driven by attractive interactions, and electron-hole pair formation by repulsive interactions between electrons in different bands. Exciton condensates\textsuperscript{24–26} are coherent states of electrons and holes bound into pairs by the Coulomb interaction, and are described by a mean-field theory that is identical to BCS theory apart from a particle-hole transformation. In bulk materials the concept of exciton condensation is partly ambiguous because of the strong interactions between subsystems located in different layers that separately have nearly perfectly conserved particle number. Interlayer exciton condensation breaks the independent gauge invariance of the individual layers, i.e. when the exciton order parameter $x$ is nonzero, there is spontaneous phase coherence between layers and no longer can perform gauge transformation independently. There are still only a few examples of convincing experimental evidence for exciton condensation in systems without a strong external magnetic field, but this situation seems likely to change.

Recently, superconductivity has been observed in magic angle twisted bilayer graphene (tBG)\textsuperscript{32,33}, a two-dimensional system with a moiré superlattice that yields extremely flat conduction and valence bands. The strongest superconductivity appears when the valence band is doped away from half filling. Although several models\textsuperscript{34–38} for its superconductivity have been proposed, the mechanism remains unknown. Twisted bilayers are the simplest examples of a rich variety of graphene multilayer moiré superlattice systems that have received experimental attention recently\textsuperscript{39–42}. The motivation for this paper is to consider the possibility of realizing, in this flexible family of strongly-correlated electron systems, an exotic state in which interlayer exciton condensation and superconductivity occur simultaneously. Our target system is two twisted bilayers separated by a hexagonal boron nitride tunnel barrier. The superconductivity of the individual twisted bilayers is already established. Here we ask the following questions: i) can interlayer exciton condensation\textsuperscript{43} occur in principle between two-dimensional electron systems that are superconducting ii) if so, how would such a state be most unambiguously detected?}

So far research on the coexistence of two phases has been scarce\textsuperscript{44} partially because of the lack of a physical system with a potential to have both. In section II we describe the model we study and relationships between its order parameters. In section III we describe the phases we have identified and phase diagrams as a function of the strength of the interlayer repulsive
interaction strength \(g_x\) and the intralayer attractive interaction strength \(g_s\). In the section IV we propose an Andreev drag measurement which can be used to identify states with both types of order, taking advantage of the possibility of separate contacting to individual layers. This method of probing bilayer exciton condensates has been very successfully exploited in quantum Hall excitonic superfluids. Finally, in section V we discuss prospects for observing these states and the importance of deviations from the highly symmetric point that we considered in our explicit calculations.

II. MODEL

We consider the simplest possible model that can have both interlayer exciton condensation and s-wave superconductivity. The model assumes that both the attractive intralayer and repulsive interlayer interactions are momentum-independent, includes a layer degree of freedom \((l = t, b)\), and assumes either valley or spin singlet superconductivity that is described by making a particle-hole transformation for one spin or valley in each layer. The Hamiltonian

\[
H = \sum_{pl\sigma} \xi_{pl} c_{l\sigma}^\dagger(p)c_{l\sigma}(p) + \frac{\lambda_x}{S} \sum_{pp'q} c_{t\sigma}^\dagger(p+q)c_{l\sigma}(p') c_{l\sigma}(p) + \frac{\lambda_x}{S} \sum_{pp'q} c_{l\sigma}^\dagger(p+q)c_{b\sigma}(p') c_{b\sigma}(p),
\]

where \(c_{l\sigma}^\dagger\) creates (annihilates) a fermion in layer \(l = t, b\) with pseudospin \(\sigma = \uparrow, \downarrow\), and \(S\) is the area of the sample. In this way, we will focus on the case of layer-independent bands, \(\xi_{pl} \equiv \xi_p\), and reserve discussion of deviations from this symmetric limit to the end of the paper. (As we explain in Section V coexistence does not occur for \(\xi_{pl} \equiv -\xi_{pb}\).) The pseudospin label refers to real spin in the case of spin-singlet superconductors and to valley in the case of valley-singlet superconductors. Below we refer to this degree of freedom as spin for simplicity.

Below we characterize the ordered states by their mean-field Hamiltonians, and allow an exciton order parameter for each spin species,

\[
x_\sigma = \frac{\lambda_x}{S} \sum_p \langle c_{l\sigma}(p)c_{l\sigma}^\dagger(p) \rangle,
\]

and superconducting order parameters both within and between layers,

\[
\Delta_{\sigma\sigma'}^{tt} = -\frac{\lambda_{tt}}{S} \sum_p \langle c_{l\sigma}(p)c_{l\sigma'}(-p) \rangle,
\]

where \(\lambda_{tt} = \lambda_{bb} = \lambda_t\) and \(\lambda_{tb} = \lambda_{bt} = \lambda_b\). The ordered state is therefore characterized by six complex self-energies that vanish by symmetry if no symmetries are broken.

In a single-band superconductors, the pair potential can be chosen to be real because of global gauge invariance. It follows that one real number, the gap, fully characterizes the superconducting phase. In the present two-band system, we start with four complex pair potentials. If we also allow exciton particle-hole pairing for both spins, the ordered state is characterized by 12 real numbers. Since only three phases can be chosen at will by exploiting the conservation of the number of particles of particles of each spin in each layer, the energy can depend not only on absolute values of the order parameters, but also on their phases, or, more specifically, on gauge invariant combinations of such phases.

In the absence of interlayer superconducting coupling, there is only one gauge invariant combination of the phases: \(\psi_t - \psi_b - \psi_{\uparrow} - \psi_{\downarrow}\), where \(\psi_{\sigma = \uparrow, \downarrow}\) is the phase of the corresponding exciton pair potential and \(\psi_{l = t, b}\) is the phase of the intralayer superconducting pairing potential. This combination changes sign under flip of layer. If we assume that the ground state preserves spin and layer invariance, we conclude that \(\psi_t - \psi_b - \psi_{\uparrow} - \psi_{\downarrow}\) = \(\pi n\), where \(n\) is an integer. In the gauge \(\psi_b = 0, \psi_t = 0\), \(\psi_{\uparrow} = 0\), an even \(n\) will correspond to a state with two superconducting gaps having the same sign, while an odd \(n\) will correspond to a state with superconducting gap that changes sign under the layer flip.

To extend this analysis to the case with interlayer pairing, we identify four gauge invariant quantities that do not transform to themselves under layer or spin inversion: \(x_{\uparrow}\Delta_{tt}^{\uparrow\downarrow} \Delta_{tt}^{\downarrow\uparrow}, x_{\downarrow}\Delta_{tt}^{\downarrow\uparrow} \Delta_{tt}^{\uparrow\downarrow}, x_{\downarrow}\Delta_{tt}^{\uparrow\downarrow} \Delta_{tt}^{\downarrow\uparrow}\), and \(x_{\downarrow}\Delta_{tt}^{\downarrow\uparrow} \Delta_{tt}^{\uparrow\downarrow}\) and require that their values do not change under these transformations.

This condition restricts the absolute values of all self-energies \(|\Delta_{\sigma\sigma'}| = |\Delta_{\sigma\sigma'}| = |\Delta_d|, |x_{\uparrow}| = |x_{\downarrow}| \equiv x, |\Delta_{tt}^{\uparrow\downarrow}| = |\Delta_{tt}^{\downarrow\uparrow}| = \Delta_t\) as well as two gauge invariant combinations of phases

\[
\psi_t - \psi_b = \psi_{\uparrow} - \psi_{\downarrow} - 2\pi m,
\]

\[
\psi_t - \psi_b = \psi_{\uparrow} + \psi_{\downarrow} - 2\pi k,
\]

where \(\psi_{\uparrow}\) is the phase of \(\Delta_{tt}^{\uparrow\downarrow}\) and \(\psi_{\downarrow}\) is the phase of \(\Delta_{tt}^{\downarrow\uparrow}\). (\(d, i\) and \(i\) in \(\Delta_d\) and \(\Delta_t\) are intended to suggest direct and indirect pairing.) The system has one more independent gauge invariant combination of phases \(\psi_t + \psi_b - \psi_{\uparrow} - \psi_{\downarrow}\). Unlike those we considered previously, this combination transforms to itself under layer or spin flip, and therefore should be determined by energy minimization.

Using global gauge invariance related to conserved particle numbers for each spin and valley \(\psi_t, \psi_{\uparrow}\) and \(\psi_{\downarrow}\) can be chosen to be 0. Then because of \(\psi_{\uparrow} = 2\pi m\) and \(\psi_{\downarrow} = (\psi_t - \psi_b)/2 = \pi k\). In what follows, we use this gauge. This leaves us with mean-field Hamiltonians that depend only on four parameters, three absolute values and a single phase, whose values can be determined by solving self-consistent field equations.
III. PHASE DIAGRAM

We saw previously that layer and spin symmetries restrict the value of $\psi_-$, but not $\psi_+$. In this section, we allow both phases to take any value demonstrate explicitly that energies are minimized when spin/layer symmetries are respected: the four allowed values of $\psi_-$ follow from the self-consistency equations. The mean-field version of Eq. (1) can be written as

$$H_{mf} = \sum_p \Psi(p) H(p) \Psi(p),$$

where $\Psi(p) = (c_{i\uparrow}, c_{i\downarrow}, c_{b\uparrow}, c_{b\downarrow})$ is a vector in an extended Nambu space and the Hamiltonian matrix is

$$
\begin{pmatrix}
\xi_p & \Delta_i e^{i(\psi_+ + \psi_-)} & -\Delta_i e^{-i(\psi_+ + \psi_-)} & -\xi_p \\
\Delta_i & -\xi_p & -\xi_p & \Delta_i \\
-\xi_p & \Delta_i & -\xi_p & -\xi_p \\
\Delta_i e^{-i(\psi_+ - \psi_-)} & -\xi_p & -\xi_p & \Delta_i \\
\end{pmatrix}
$$

The negative eigenvalues of the matrix have the form $\epsilon_{\pm} = -\left(\eta^2 \pm 2x^2\right)^{1/2}$, where $\eta^2 = \lambda_x x^2 + \Delta_i^2 + \Delta_d^2$ and $\lambda_s^2 = x^2 \Delta_d^2 \sin^2(\psi_-) + 2\Delta_i \Delta_s \cos(\psi_+) \cos(\psi_-) + \Delta_d^2 \Delta_s^2 \cos^2(\psi_+ + \pi) + \Delta_i^2$. Stable phases of the system are determined by minimization of the energy density. At energy extrema,

$$E = \frac{2\lambda_s^2}{|\lambda_s^2|} - \frac{2\lambda_s^2}{\lambda_x} + \lambda_x^2 + \frac{1}{\lambda_x} \int d\xi \nu(\xi) \epsilon_b n_b(\xi),$$

where the sum is over quasiparticle bands $b$, $\nu(\xi)$ is the quasiparticle density-of-states per spin and per layer, and $n_b(\xi)$ is the Fermi occupation factor. At zero temperature, only quasiparticle states with negative energies are filled. Extrema of the energy are also extrema of Eq. (8), which we vary first with respect to the phases $\psi_-$ and $\psi_+$. We conclude that energy minima occur at extrema of $\alpha^2$, and that these occur (independent of $\xi_p$) when

$$\sin(\psi_+) = 0 \text{ and } \sin(\psi_-) = 0$$

or

$$\cos(\psi_+) = 0 \text{ and } \cos(\psi_-) = 0.$$  

The former condition is consistent with the conditions we derived for a mirror- and spin-symmetric state with $\Delta_0 \neq 0$. We will call it a parallel phase. The latter condition defines the antiparallel phase. We will see that the latter is consistent only with $\Delta_0 = 0$. Note that each condition in fact corresponds to two different states. Namely, (9) can be satisfied with $(\psi_1, \psi_2) = (\pi, \pi)$, and $(0, 0)$, that typically have different energies when interlayer superconducting order is present. Now we consider the antiparallel and parallel phases separately.

A. Antiparallel phase

We consider first extrema in which the phases $\psi_1$ and $\psi_2$ differ by $\pi$. The main conclusion of this subsection is that for any sufficiently smooth density of states, the energy of the antiparallel phase is always higher than that of a pure superconducting phase. In other words this solution of the mean-field equations corresponds to a local energy maximum not an energy minimum, and can therefore be discarded.

In the antiparallel phase, variation of the energy density in Eq. 8 with respect to $\Delta_i$ combines with Eq. (10) to yield the self-consistency equation

$$\Delta_i = -\frac{\lambda_x \Delta_i}{4} \int \nu(\xi) \left( \frac{1}{\epsilon_+} + \frac{1}{\epsilon_-} \right).$$

This equation clearly cannot be satisfied at any $\lambda_x > 0$. We conclude that $\Delta_i = 0$ in the antiparallel phase. The quasiparticle band energies are therefore

$$\pm \epsilon_{\pm} = \pm \sqrt{\lambda_x^2 + \Delta_d^2} \pm |x|$$

The form of the band energies allows us to identify the antiparallel phase as an exciton condensate (spontaneous interlayer phase coherent state) formed on top of superconductors within each of the layers. With $\Delta_i$ eliminated, the system of gap equations reduces to:

$$x = \lambda_x N(\sqrt{x^2 - \Delta_d^2}),$$

$$\Delta_d = -\frac{\lambda_x \Delta_d}{2} \int \frac{\nu(\xi)}{\sqrt{\lambda_x^2 + \Delta_d^2}} + \frac{\lambda_x \Delta_d}{2} \int \frac{\nu(\xi)}{\sqrt{\lambda_x^2 - \Delta_d^2}}$$

That is to say that the exchange self-energy is contributed by wavevectors that have different occupation numbers for the two orthogonal quasiparticle layer spinors. In Eq. (13) $N(\epsilon) \equiv 1/2 \int_{-\infty}^{\infty} d\xi \nu(\xi)$. A transition between pure superconductivity and an antiparallel phase would occur at zero temperature if the
latter phase were lower in energy. We find that

\[ E_m - E_{sc} = \frac{2}{|\lambda_s|} \left( \Delta_{d_m}^2 - \Delta_0^2 \right) + 2 \int_{-\mu}^{\Lambda} \nu(\xi) \left( \sqrt{\xi^2 + \Delta_{d_m}^2} - \sqrt{\xi^2 + \Delta_0^2} \right) \]

where we distinguish the superconducting gap \( \Delta_{d_m} \) in the mixed phase from the superconducting gap \( \Delta_0 \) in the pure superconducting phase, and have used a self-consistency equation to reexpress the last term. Since the energy of the pure superconducting phase is minimized by \( \Delta_0 \), we can conclude that the sum of the first two lines in Eq. 15 is non-negative. Provided that the density of states does not significantly vary on the scale of \( \sqrt{x^2 - \Delta_{d_m}^2} \), the third line can be approximated by

\[ 2\nu_0 \Delta_{d_m}^2 \sinh^{-1} \left( \frac{\sqrt{x^2 - \Delta_{d_m}^2}}{\Delta_{d_m}} \right) \],

where \( \nu_0 \) is the constant density-of-states, and the full energy difference is positive. Hence, at least for a sufficiently smooth density of states, the antiparallel phase will never be thermodynamically stable. We now analyze the parallel phase.

**B. Parallel phase**

The parallel phase is defined by the conditions \( \sin(\psi_+) = 0, \sin(\psi_-) = 0 \), and \( \psi_i, \psi_b \), are equal to \( 0 \). This condition, however, leaves an ambiguity since the \( \cos(\psi_+) \cos(\psi_-) \) present in the dispersion relation can be either +1 or \(-1\). We distinguish these possibilities by introducing \( f = 0, 1 \), such that \( \cos(\psi_+) \cos(\psi_-) = (-1)^f \). The quasiparticle energies can be expressed as:

\[ \epsilon_\pm = \sqrt{(\xi \pm x)^2 + (\Delta_d \pm (-1)^f \Delta_i)^2} \].

We can interpret this phase as two superconductors: one with Cooper pairs formed out of symmetric combinations of layers, another – from antisymmetric combinations. Indeed, the mean-field Hamiltonian matrix (6) is block-diagonal in this basis. Note here that even though the transformation to the symmetric and antisymmetric states block-diagonalizes \( H(p) \), energy density \( E \) (8) will still have terms that couple the two unless \( |\lambda_s| = \lambda_x \). Solutions with \( f = 0/1 \) then correspond to phases with in which either symmetric or antisymmetric superconducting gaps are larger. Variation of the full energy density (8) with respect to \( \Delta_i \) yields

\[ \frac{\delta E}{\delta \Delta_i} = -\frac{4\Delta}{\lambda_x} - \Delta_i \int \nu(\xi) \left( \frac{1}{\epsilon_-} + \frac{1}{\epsilon_+} \right) \]

\[ + (-1)^f \Delta_d \int \nu(\xi) \left( \frac{1}{\epsilon_-} - \frac{1}{\epsilon_+} \right) \]

One conclusion we can immediately draw is that the \( \Delta_i = 0 \) point is not an energy extremum whenever both
exciton condensates and superconductivity are present. Note that the superconducting gap of the symmetric quasiparticles is \( \Delta_d + (-1)^f \Delta_i \), while for the antisymmetric quasiparticles is \( \Delta_a = (-1)^f \Delta_i \). Because the exciton condensate breaks the symmetry between quasiparticle bands, one should expect that \( \Delta_a \) condensate breaks the symmetry between quasiparticle states summarized in Fig. 1 exists for \( c \) and \( f \). The mixed phase allows the in-layer interaction to be repulsive, the BCS instability present within each layer together with interlayer coherence induces a response in the interlayer Cooper channel.

If we write the self-consistency equations in terms of \( \Delta_+ = \Delta_d + (-1)^f \Delta_i \), \( \Delta_- = \Delta_d - (-1)^f \Delta_i \), and \( x \) we obtain,

\[
\frac{1}{\lambda_+ - \lambda_-} \left( \lambda_+ \Delta_+ + \lambda_- \Delta_- \right)
= \frac{\Delta_+}{2} \int \frac{d\xi \nu(\xi)}{\sqrt{(\xi + x)^2 + \Delta_+^2}},
\]

\[
\frac{1}{\lambda_+ - \lambda_-} \left( \lambda_+ \Delta_- - \lambda_- \Delta_+ \right)
= \frac{\Delta_-}{2} \int \frac{d\xi \nu(\xi)}{\sqrt{(\xi - x)^2 + \Delta_-^2}},
\]

\[
\frac{4}{\lambda_+ - \lambda_-} x = \int \frac{d\xi \nu(\xi)(x - \xi)}{\sqrt{(\xi - x)^2 + \Delta_+^2}} + \int \frac{d\xi \nu(\xi)(x + \xi)}{\sqrt{(\xi + x)^2 + \Delta_-^2}}.
\]

Here \( \lambda_+ \equiv (\lambda_a + \lambda_x)/2 \) and \( \lambda_- \equiv (\lambda_a - \lambda_x)/2 \). Note that even though the mean-field Hamiltonian matrix is block diagonal, the energy density has a term of the form \( \Delta_+ \Delta_- \) because \( \lambda_a \neq \lambda_x \). We solve this system numerically for the constant density-of-states case. The mixed state solution summarized in Fig. 1 exists for \( \nu(0) g_x \geq 1 \).

In the numerical calculation, to treat both \( f = 0 \) and \( f = 1 \) minima on the equal footing, we allowed the interlayer superconducting gap \( \Delta_i \) to take negative values. Then negative value of \( \Delta_i \) corresponds to the \( f = 1 \) minimum while positive values will correspond to the \( f = 0 \) minimum. Our calculation shows that \( f = 1 \) minimum tend to be more energetically favorable for larger values of intralayer attraction, which has a simple physical meaning: for \( |\lambda_a| \approx \lambda_x \) the symmetric and the antisymmetric superconductors are effectively decoupled and thus larger gap correspond to a larger effective Fermi energy. Along the first order phase boundary in \( g_x/g_s \) space separating mixed and pure superconducting phases, the two states have identical energies. It follows that the differential energy changes \( dE_{sc} \) in the superconducting phase and \( dE_{mix} \) in the mixed phase should be the same.

\[
\frac{dg_x}{dg} = \frac{\partial E_{sc}}{\partial g_x} = \frac{\partial E_{mix}}{\partial g_s}.
\]

Partial derivatives in the formula will be proportional.
to order parameters in each phase: $\partial E_{sc,mix}/\partial g_s = -2\Delta^2_{sc,mix}/g_s^2$, $\partial E_{sc}/\partial g_x = 2(x^2 - \Delta^2_x)/g_x^2 \approx 2\mu^2/g_x^2$. Formation of the mixed state accompanied by the decrease in the superconducting gap at least for large enough $|g_s|$. Therefore, $\frac{dg_s}{dg_x} < 0$ – for larger attractive interaction $g_s$ transition will happen at a larger critical $g_x$ in agreement with Fig. 1.

IV. NON-LOCAL ANDREEV REFLECTION

Having established that an exotic mixed state with both excitonic and Cooper pair order can exist, we now discuss Andreev reflection experiments in separately contacted bilayers that can be used to detect their presence. Consider metallic leads separately connected to the top and the bottom layers of our system. An electron incident on the interface between the top lead and the top layer has equal weight in symmetric and antisymmetric quasiparticle channels $|t\rangle = (|+\rangle + |-\rangle)2^{-1/2}$. Since the parallel state does not couple $|+\rangle$ with $|-\rangle$, we can consider their transmission contributions separately, at least if disorder is neglected. Incoming quasiparticles are reflected with probabilities $A_{tt}$ and $A_{tb}$ as holes, and with probabilities $B_{tt}$ and $B_{tb}$ as electrons, in the top and bottom layers respectively. The calculation of these reflection probabilities is discussed below.

If we apply $V$ voltage across the top part of the interface, the top and bottom layer differential conductances will be connected to reflection coefficients in a normal fashion. The total currents in the top and bottom layers are

$$I_{t/b} = e\nu_0 \int_0^{eV} d\omega \sigma_{t/b}(V)$$

where

$$\sigma_t(V) \equiv d_{tt}/dV = e^2 v_F \nu_0 (1 - B_{tt}(V) + A_{tt}(V)),$$

$$\sigma_b(V) \equiv d_{tb}/dV = e^2 v_F \nu_0 (A_{bb}(V) - B_{bb}(V)),$$

$e$ is an electron charge, and $v_F\nu_0$ is the product of the Fermi velocity and the density of states. Note that the combination $A_{tb} - B_{tb}$ of cross-layer reflection probabilities can be measured experimentally. We derive expressions for the scattering amplitudes in the Appendix using the Bogoliubov-de-Gennes equations. The amplitude to reflect an electron as a hole in either symmetric or antisymmetric states, at least in the limit $x \ll \mu$, around 1 at $\omega = 0$ similarly to a single N-S junction. However, the amplitude to reflect an incoming electron as an electron is no longer negligible even in the $\Delta_t \ll \mu$ case. The electron reflection amplitude at $\omega = 0$ is given by $b_k \approx x/(2\mu \pm x)$. Because $b_+$ and $b_-$ have opposite signs, the probability to reflect an electron to the bottom layer $B_{tb} = |b_+ - b_-|^2/4$ is substantially higher than the probability of the corresponding Andreev process, which is almost zero. For the the same reason electron reflection in the top layer is unimportant for small $\omega$ and instead the Andreev process dominates (Fig. 2). When energy is equal to the $+ or -$ gap and the gap $\Delta_\perp/\mu \ll 1$, the corresponding momenta in the leads and device match, giving rise to peaks in Andreev reflection and minima in electron reflection. As a result the Andreev process is dominant over electron reflection in the bottom layer. Altogether, we expect $A_{tb} - B_{tb}$ to have a $\mu$-like shape, with negative values around $\omega = 0$ and positive peaks at the superconducting gaps (Fig. 2).

If we follow the standard practice of modelling interface disorder by a delta-function potentials $v_f(\delta(z))$, $\langle v \rangle = (v_t + v_b)/2$ will play a role identical to that of the disorder in a regular superconductor by damping Andreev reflection at subgap energies and increasing electron-electron reflection in both symmetric and antisymmetric channels. The dimensionless quantity controlling its importance is $\langle v \rangle/v_F$, where $v_F$ is the Fermi velocity. Any difference $\Delta v = (v_t - v_b)/2$ between layers in the disorder parameter values will couple the symmetric and antisymmetric quasiparticle channels. The importance of these corrections is however also controlled by $\Delta v/v_F$, and for values $\Delta v/v_F < 0.5$ there is no qualitative change in the $\mu$-like shape of the differential conductance (Fig. 3). Sufficiently large values of $\Delta v/v_F$ raise the low-energy part of the differential conductance to positive values.

FIG. 3. Influence of interface disorder on the difference between hole (Andreev process) and electron interlayer reflection coefficients $A_{tb} - B_{tb}$. Disorder is modeled by a delta-function potential at the interface $\nu_0\delta(z)$. On each subplot, the ratio of $\langle v \rangle = (v_t + v_b)/2$ to the Fermi momentum is kept fixed. $\Delta v/p_F = (v_t - v_b)/(2p_F)$ is swept from -1.0 (red) to +0.8 (black) with step 0.1.
V. DISCUSSION

In recent years experimenters have established moiré heterojunctions as attractive platforms for new types of two-dimensional electron ground states. In this article, we have explored the phase diagram of bilayers with attractive effective interactions (λx) within each layer and repulsive interactions (λx) between layers. Two-dimensional bilayers with repulsive interlayer interactions can have excitonic insulator ground states that are counterflow superfluids and have spontaneous interlayer phase coherence. Our work is motivated by the discovery of superconductivity, and hence attractive effective interactions, in graphene bilayers and trilayers. We therefore address the possibility of states that have both superconductivity and counterflow superfluidity, and study how the occurrence of such exotic states would be manifested by generalized Andreev effects in separately contacted bilayers.

With this goal we have performed mean-field calculations for a model bilayer Hamiltonian with two identical layers, and attractive intralayer and repulsive interlayer interactions. We have neglected tunneling τ between the layers, assuming that the bilayers are separated by dielectric layers. Our calculations show that a mixed phase with both an interlayer exciton condensate and superconductivity appears over a wide range of model parameters. Interlayer coherence couples the two superconducting order parameters with energy extrema occurring when the pair amplitudes are in phase (parallel) and out of phase (antiparallel). We find that the energy of the antiparallel phase is higher than that of the pure superconducting phase for any sufficiently smooth density of states. In the parallel phase superconducting state, interlayer coherence appears as a strong-coupling instability of states with superconductivity in each layer, and is established when gx ≡ ν(εF)λx > 1. In twisted bilayer graphene we estimate that λx ≈ e2/(4πkF) ≈ 100 meVnm2, and that the density of states is around ν(εx) ≈ 10−2 nm−2 meV−1, implying that |gx| ≤ 1 is within reach. Since the superconducting transition temperature in tBG is ~ 1K, the value we have chosen for |gx| ≤ 1 is not unrealistic. After interlayer coherence is established, further λx increases cause the superconducting order parameters to decay. Note that self-consistent solution with both exciton order parameter x and direct intralayer pairing Δd nonzero also have non-zero interlayer pairing Δs.

As a convenience, we have limited our considerations to the case of two completely identical layers. This condition is however not important for the stability of the phase. Consider for example the case in which the Fermi energies in the two layers are slightly different: μi=μ±Δμ. This change induces a perturbation to the mean field Hamiltonian matrix of the form −τzσzΔμ, where τz and σz are the z-Pauli matrices acting on layer and Nambu indices correspondingly. After transformation to +/− basis the perturbation acquire form iτyσzμ. As a result, there will be no first-order contribution to the quasiparticle energies. At second order, only matrix elements between positive and negative energy states will contribute. The leading order correction to the quasiparticle energies will be small, ~ (Δμ2/√Δ2 + x2). We conclude that small deviations from the perfect layer symmetric case do not destroy the state, although they will change the mean-field-equation solutions quantitatively.

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Because Andreev reflection in symmetric and antisymmetric states cancel each other at $V = 0$, it is instrumental to consider also electron-electron reflection. It is still true however that Bogoliubov-de-Gennes equation consists of two uncoupled systems of equations. We can constrain ourselves to solving only one of them:

$$i\partial_t f = -\left(\frac{1}{2m} \frac{\partial^2}{\partial z^2} + \mu(z)\right)f + \Delta(z)\phi$$

$$i\partial_t \phi = \left(\frac{1}{2m} \frac{\partial^2}{\partial z^2} + \mu(z)\right)\phi + \Delta(z)f$$

The only difference from the classical\cite{3,53} setup is that $\mu(z)$ is a function of a coordinate to take into account the presence of the interlayer coherence inside the system. Fourier transform with respect to the time $f(t) = \int e^{-i\omega t} f_\omega$.
The last piece is the reflected hole. Now boundary conditions read:

\[ \frac{1}{2m} \partial_z^2 f = -(\omega + \mu(z)) f + \Delta(z) \phi \]  
\[ \frac{1}{2m} \partial_z^2 \phi = (\omega - \mu(z)) \phi - \Delta(z) f \]

Or, alternatively,

\[ \frac{1}{2m} \partial_z^2 \begin{pmatrix} f' \\ f \end{pmatrix} = \begin{pmatrix} -\omega + \mu(z) & \Delta(z) \\ -\Delta(z) & \omega - \mu(z) \end{pmatrix} \begin{pmatrix} f' \\ f \end{pmatrix} \]

Let's assume that \( \Delta \) changes abruptly from 0 to \( \Delta_0 \) and similarly \( \mu \) changes from \( \mu \) to \( \mu + x \):

\[ \mu(z) = \mu + \theta(z)x \]
\[ \Delta(z) = \theta(z)\Delta, \]

where \( x \) can be both positive and negative. We solve it by writing it as a system of the first order equations

\[ \frac{1}{2m} \partial_z \begin{pmatrix} f' \\ f \end{pmatrix} = \begin{pmatrix} 0 & 0 & -\omega + \mu \\ 0 & 0 & -\Delta \\ 1/2m & 0 & 0 \end{pmatrix} \begin{pmatrix} f' \\ f \end{pmatrix} \]

Eigenvalues of this matrix are \( \pm \frac{i}{\sqrt{2m}} (\mu + \sqrt{\omega^2 - \Delta^2})^{1/2} \), \( \pm \frac{i}{\sqrt{2m}} (\mu - \sqrt{\omega^2 - \Delta^2})^{1/2} \). A general solution for \( \omega < \Delta \) will be

\[ \begin{pmatrix} f' \\ f \end{pmatrix} = C e^{i\sqrt{2m} \mu + \omega \sqrt{\Delta^2 - \omega^2}} \begin{pmatrix} \sqrt{\frac{\omega + \sqrt{\Delta^2 - \omega^2}}{\Delta^2}} \\ \sqrt{\frac{-\omega + \sqrt{\Delta^2 - \omega^2}}{\Delta^2}} \\ \frac{D e^{iz \sqrt{2m} (\mu + i\sqrt{\Delta^2 - \omega^2})^{1/2}}} {\sqrt{\frac{\omega + i\sqrt{\Delta^2 - \omega^2}}{\Delta^2}}} \end{pmatrix} \]

where \( C \) and \( D \) are constants.

I will denote in what follows \( \sqrt{\frac{\omega + \sqrt{\Delta^2 - \omega^2}}{\Delta^2}} = u \) and \( \sqrt{\frac{-\omega + \sqrt{\Delta^2 - \omega^2}}{\Delta^2}} = v \) and \( k_{sC} = \sqrt{2m (\mu + i\sqrt{\Delta^2 - \omega^2})^{1/2}} \), \( k_{sD} = \sqrt{2m (\mu - i\sqrt{\Delta^2 - \omega^2})^{1/2}} \). On the left hand side equations for a hole and an electron decouple and as a solution I get instead:

\[ \begin{pmatrix} f' \\ f \end{pmatrix} = e^{i\sqrt{2m} \mu + \omega N} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \]
\[ + b e^{-i\sqrt{2m} \mu + \omega N} \begin{pmatrix} 1 \\ 0 \end{pmatrix} + a \begin{pmatrix} 0 \\ 1 \end{pmatrix} e^{i\sqrt{2m} \mu + \omega N} \]

The last piece is the reflected hole. Now boundary conditions read:

\[ 1 + b = \frac{C}{\sqrt{2}} \left( \frac{1}{k_{sc}} + \frac{D}{\sqrt{2}} \frac{1 - k_{sc}}{k_{sc}} \right) \]
\[ a = \frac{C}{\sqrt{2}} \left( \frac{1}{k_{sc}} + \frac{D}{\sqrt{2}} \frac{1 - k_{sc}}{k_{sc}} \right) \]
\[ k_{c} (1 - b) = \frac{C}{\sqrt{2}} \left( \frac{1}{k_{sc}} + \frac{D}{\sqrt{2}} \frac{1 - k_{sc}}{k_{sc}} \right) \]
\[ k_{h} a = \frac{C}{\sqrt{2}} \left( \frac{1}{k_{sc}} + \frac{D}{\sqrt{2}} \frac{1 - k_{sc}}{k_{sc}} \right) \]

from which we conclude that

\[ 2 = \frac{C u}{\sqrt{2}} \left( 1 + \frac{k_{sc}}{k_{c}} \right) + \frac{D v}{\sqrt{2}} \left( 1 - \frac{k_{sc}}{k_{c}} \right) \]
\[ 0 = \frac{C v}{\sqrt{2}} \left( 1 - \frac{k_{sc}}{k_{h}} \right) + \frac{D u}{\sqrt{2}} \left( 1 + \frac{k_{sc}}{k_{h}} \right) \]

hence

\[ \frac{D}{C} = -u \frac{1 - k_{sc}}{k_{c}}, \]

\[ \frac{D}{C} = -v \frac{1 - k_{sc}}{k_{h}}. \]
Thus, we expect the Andreev process to be dominant at frequencies corresponding to one of the gaps.

Green’s function, introduce a set of additional functions:

\[ C = \frac{2^{3/2}u}{u^2 (1 + \frac{k_{eC}}{k_h})(1 + \frac{k_{eD}}{k_h}) - u^2 (1 - \frac{k_{eC}}{k_h})(1 - \frac{k_{eD}}{k_h})} \]  

\[ D = \frac{2^{3/2}v}{v^2 (1 + \frac{k_{eC}}{k_h})(1 + \frac{k_{eD}}{k_h}) - v^2 (1 - \frac{k_{eC}}{k_h})(1 - \frac{k_{eD}}{k_h})} \]  

Expressions for \( a \) and \( b \):

\[ a = \frac{2uv (\frac{k_{eD}}{k_h} + \frac{k_{eC}}{k_h})}{u^2 (1 + \frac{k_{eC}}{k_h})(1 + \frac{k_{eD}}{k_h}) - u^2 (1 - \frac{k_{eC}}{k_h})(1 - \frac{k_{eD}}{k_h})} \]  

\[ b = \frac{u^2 (1 + \frac{k_{eC}}{k_h})(1 - \frac{k_{eC}}{k_h}) - u^2 (1 - \frac{k_{eC}}{k_h})(1 - \frac{k_{eD}}{k_h})}{u^2 (1 + \frac{k_{eC}}{k_h})(1 + \frac{k_{eD}}{k_h}) - u^2 (1 - \frac{k_{eC}}{k_h})(1 - \frac{k_{eD}}{k_h})} \]  

It is clear that a drastic change of the potential on the interface will lead to an increase in electron-electron scattering. Let us explore the low energy properties of both \( a \) and \( b \). Because at \( \omega = 0 \): \( k_e = k_h = p_F \), where \( p_F \) is a Fermi momentum inside the metallic lead, \( u = e^{i\pi/4},v = e^{3i\pi/4} \). As a result, at \( \omega = 0 \):

\[ a = -\frac{i\sqrt{\mu_N} (\sqrt{\mu - i\Delta + \sqrt{\mu - i\Delta}})}{\mu_N + \sqrt{\mu + i\Delta} \sqrt{\mu - i\Delta}} \]  

\[ b = \frac{(\sqrt{\mu_N} - \sqrt{i\Delta + \mu})(\sqrt{\mu_N} + \sqrt{\mu - i\Delta})}{\mu_N + \sqrt{\mu + i\Delta} \sqrt{\mu - i\Delta}} \]  

Because \( a \) for symmetric and antisymmetric states are approximately equal to each other whenever \( \mu \gg \Delta \), their difference is typically small is this condition is satisfied. Surprisingly, electron-electron scattering amplitude do not vanish in the \( \Delta_{\mu_N} \to 0 \) limit, because the effective Fermi energies inside and outside the system are different. When the \( \omega \) is close to the gap, \( u \approx v, k_{eC} \approx k_{eD} \), and if the gap is much smaller than \( \mu_N, k_e \approx k_h \), therefore \( b \approx 0, a \approx 1 \). Thus, we expect the Andreev process to be dominant at frequencies corresponding to one of the gaps.

**VIII. APPENDIX: GREEN’S FUNCTION FORMALISM**

In this appendix we present equations of motion for zero-temperature Green’s function and derive an important symmetry of self-energy in a way different from the one we have taken in the main text. In addition to a normal Green’s function, introduce a set of additional functions:

\[ iF^l_{\alpha\sigma}(p,t_1 - t_2) = \langle T \left( c_{p\sigma}(t_1)c_{p\sigma}^\dagger(t_2) \right) \rangle e^{-i2\mu t_1} \]  

\[ iF^l_{\alpha\sigma}(p,t_1 - t_2) = \langle T \left( c_{p\sigma}(t_1)c_{-p\sigma}^\dagger(t_2) \right) \rangle e^{i2\mu t_1} \]  

\[ iH^l_{\alpha}(p,t_1 - t_2) = \langle T \left( c_{p\sigma}(t_1)c_{p\sigma}^\dagger(t_2) \right) \rangle e^{i(\mu - \mu)} t_1 \]  

\[ iD^b_{\alpha\sigma}(p,t_1 - t_2) = \langle T \left( c_{p\sigma}(t_1)c_{-p\sigma}^\dagger(t_2) \right) \rangle e^{i(\mu + \mu)} t_1 \]  

\[ iD^b_{\alpha\sigma}(p,t_1 - t_2) = \langle T \left( c_{p\sigma}(t_1)c_{p\sigma}^\dagger(t_2) \right) \rangle e^{i(\mu + \mu)} t_1 \]
Averages are taken over ground states with fixed and, in general, different number of particles of each kind. Because of this, averages are no longer dependent on the time difference. To mitigate this problem, averages are multiplied by an exponential with the difference between energies on the left and the right. I will only consider spatially uniform solutions and without spontaneous magnetization.

First, look at the equation of motion for the normal and the anomalous Green’s functions:

\[
\left(\frac{i}{\partial t} - \epsilon_p t\right) G_{\uparrow \uparrow}(p, t) = F_{\uparrow \uparrow}^{\uparrow \uparrow}(p, t)\Delta_{\uparrow \uparrow}^t + D_{\uparrow \uparrow}^{bt}(p, t)\Sigma_{\uparrow \uparrow}^{tb} + H_{\uparrow \uparrow}^{bt}(p, t)X_{\uparrow \uparrow}^{bt} + \delta(t)
\]

\[
\left(\frac{i}{\partial t} - \epsilon_p t - 2\mu t\right) F_{\uparrow \uparrow}^{\uparrow \uparrow}(p, t) = \Delta_{\uparrow \uparrow}^t G_{\uparrow \uparrow}(-p, t) + H_{\uparrow \uparrow}^{bt}(-p, t)\Sigma_{\uparrow \uparrow}^{bt} - D_{\uparrow \uparrow}^{bt}(p, t)X_{\uparrow \uparrow}^{bt},
\]

where I defined self-energies:

\[
\Delta_{\uparrow \sigma \sigma'}^t = -\frac{i}{S} \sum V_{\uparrow \downarrow}(q) F_{\sigma \sigma'}^{\uparrow \downarrow}(-p - q, 0+)
\]

\[
X_{\sigma}^{\mu'} = \frac{i}{S} \sum V_{\sigma}^{\mu'}(q) H_{\sigma \sigma'}^{\mu}(p - q, 0+)
\]

\[
\Sigma_{\sigma \sigma'}^{\mu} = -\frac{i}{S} \sum V_{\sigma}^{\mu}(q) D_{\sigma \sigma'}^{\mu}(p + q, 0+).
\]

Note here that positive \( V_{\uparrow \downarrow} \) means repulsion within the layer and similarly for \( V_{\sigma}^{\mu'} \). The other two equations are:

\[
\left(\frac{i}{\partial t} - \epsilon_p - (\mu_\downarrow - \mu_\uparrow)\right) H_{\uparrow \downarrow}^{bt}(p, t) = \Delta_{\uparrow \downarrow}^t D_{\downarrow \uparrow}^{bt}(-p, t) + X_{\uparrow \downarrow}^{bt} G_{\downarrow \uparrow}^{bt}(p, t) + \Sigma_{\downarrow \uparrow}^{bt} F_{\downarrow \uparrow}^{bt}(-p, t)
\]

\[
\left(\frac{i}{\partial t} + \epsilon_p - (\mu_\downarrow + \mu_\uparrow)\right) D_{\downarrow \uparrow}^{bt}(p, t) = H_{\downarrow \uparrow}^{bt}(p, t)\Delta_{\downarrow \uparrow}^t + G_{\downarrow \uparrow}^{bt}(-p, t)\Sigma_{\downarrow \uparrow}^{bt} - F_{\downarrow \uparrow}^{bt}(p, t)X_{\downarrow \uparrow}^{bt}
\]

These equations can be either derived through equations of motion for operators, or diagrammatically. Equations (55)-(60) constrain the form of the mean-field hamiltonian. Indeed, consider a case without spin symmetry breaking: \( X_{\uparrow \downarrow}^{bt}e^{-i\psi_\uparrow} = X_{\downarrow \uparrow}^{bt}e^{-i\psi_\downarrow}, \Delta_{\downarrow \uparrow}^t = -\Delta_{\uparrow \downarrow}^t, G_{\downarrow \uparrow} = G_{\uparrow \downarrow}. \) The latter means that each term in (54) must be invariant under spin flip:

\[
H_{\uparrow \downarrow}^{bt}e^{i\psi_\uparrow} = H_{\downarrow \uparrow}^{bt}e^{i\psi_\downarrow}
\]

\[
D_{\downarrow \uparrow}^{bt}(p, t)\Sigma_{\downarrow \uparrow}^{bt} = D_{\uparrow \downarrow}^{bt}(p, t)\Sigma_{\uparrow \downarrow}^{bt}
\]

\[
F_{\downarrow \uparrow}^{bt}(p, t) = -F_{\uparrow \downarrow}^{bt}(p, t)
\]

From (55) it follows then that

\[
\frac{\tilde{\Sigma}_{\downarrow \uparrow}^{bt}}{\Sigma_{\downarrow \uparrow}^{bt}} = -\frac{H_{\uparrow \downarrow}^{bt}(-p, t)}{H_{\uparrow \downarrow}^{bt}(-p, t)} = e^{i(\psi_\downarrow - \psi_\uparrow + \pi)}
\]

Finally, since \( \Sigma_{\downarrow \uparrow}^{tb} \propto (c_{\downarrow \uparrow}c_{\uparrow \downarrow}), \Sigma_{\downarrow \uparrow}^{tb} = -\Sigma_{\uparrow \downarrow}^{bt}. \)

\[
\frac{\tilde{\Sigma}_{\downarrow \uparrow}^{bt}}{\Sigma_{\downarrow \uparrow}^{bt}} = e^{i(\psi_\downarrow - \psi_\uparrow)}
\]

Note that we did not imply anything about layer symmetry.