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Stabilizing fluctuating spin-triplet superconductivity in graphene via induced spin-orbit coupling

Jonathan B. Curtis,[1, 2, *] Nicholas R. Poniatowski,[2] Yonglong Xie,[2] Amir Yacoby,[2] Eugene Demler,[3] and Prineha Narang[1, †]

[1] *College of Letters and Science, University of California, Los Angeles, CA 90095, USA*
[2] *Department of Physics, Harvard University, Cambridge, MA 02138, USA*
[3] *Institute for Theoretical Physics, ETH Zürich, 8093 Zürich, Switzerland*

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A recent experiment showed that proximity induced Ising spin-orbit coupling enhances the spin-triplet superconductivity in Bernal bilayer graphene. Here, we show that, due to the nearly perfect spin rotation symmetry of graphene, the fluctuations of the spin orientation of the triplet order parameter suppress the superconducting transition to nearly zero temperature. Our analysis shows that both Ising spin-orbit coupling and in-plane magnetic field can eliminate these low-lying fluctuations and can greatly enhance the transition temperature, consistent with the recent experiment. Our model also suggests the possible existence of a phase at small anisotropy and magnetic field which exhibits quasi-long-range ordered spin-singlet charge 4e superconductivity, even while the triplet 2e superconducting order only exhibits short-ranged correlations. Finally, we discuss relevant experimental signatures.

Introduction—Two-dimensional materials have emerged as a highly tunable platform for studying spin triplet superconductivity. Notable examples include rhombohedral trilayer graphene (RTG) [1], moiré graphene [2–4], and Bernal bilayer graphene (BBG) [5, 6]. In the case of BBG, superconductivity was initially found to emerge exclusively in the presence of an in-plane magnetic field exceeding the paramagnetic limit, consistent with a triplet order parameter [5]. A very recent experiment showed that introducing a layer of tungsten diselenide (WSe_2)—a semiconducting material with strong spin-orbit coupling—on top of BBG, as schematically depicted in Fig. 1(a), can stabilize superconductivity at zero magnetic field and enhance the critical temperature by an order of magnitude [6].

An important task prompted by this recent experiment is to identify the role of the proximity-induced spin-orbit coupling in promoting triplet superconductivity in BBG. One possibility is that spin-orbit coupling selects a particular symmetry-broken parent state from which the superconductivity emerges, and previous work [7–9] shows the intervalley coherent ground state is conducive to triplet pairing. Alternatively, the presence of WSe_2 can introduce virtual tunneling processes that enhance pair binding energies, boosting the critical temperature [10].

Here we present a mechanism by which proximity-induced spin-orbit coupling promotes triplet superconductivity. Specifically, we show that Ising spin-orbit coupling (defined below) strongly breaks the nearly perfect spin-rotation symmetry in graphene, suppressing fluctuations of the otherwise gapless modes corresponding to orientational fluctuations (in spin space) of the triplet order parameter. While triplet superconductivity is formally impossible in intrinsic graphene due to the Mermin-Wagner theorem, it is allowed via a BKT-type transition once spin-rotation symmetry is broken by the WSe_2. In fact similar ideas pertaining to transport were recently put forth in Ref. [11]. This situation, where induced Ising spin-orbit coupling promotes pure triplet superconductivity, is to be contrasted with the mixed-parity "Ising" superconductivity found in a variety of materials with strong, intrinsic Ising spin-orbit coupling [12, 13].

We further predict that any perturbation which selects a particular axis for the triplet spin orientation will stabilize superconductivity: in-plane magnetic field will suppress order-parameter fluctuations and stabilize a particular spin-polarized condensate [11], whereas Ising spin-orbit coupling stabilizes a unitary (spin nematic) triplet state. As our theory is based entirely on general symmetry arguments, it should be broadly applicable to many graphene-based triplet superconductors including BBG, RTG, and moiré systems.

Microscopic Model—We begin by considering a minimal BCS model for BBG consisting of two spin-degenerate [14] Fermi surfaces at valleys $\mathbf{K}$ and $\mathbf{K}'$ in the presence of an induced Ising spin orbit coupling of strength λ. This is depicted in Fig. 1(b) which shows the intervalley pairing and induced spin-orbit coupling in the Brillouin zone.

Electrons are described by a four-component spinor $\psi_{\mathbf{k}}$, with Pauli matrices σ_i (ρ_i) acting on the spin (valley) degree of freedom. To describe superconductivity this is further doubled into an eight-component Nambu spinor $\Psi_{\mathbf{k}} = \left(\psi_{\mathbf{k}},\ i\sigma_2\bar{\psi}^T_{-\mathbf{k}}\right)^T$, which captures pairing between Kramers degenerate states. The Bogoliubov-de Gennes Hamiltonian which describes triplet pairing is then written as

$$H_{\mathrm{BdG}} = \frac{1}{2}\sum_{\mathbf{k}} \Psi^\dagger_{\mathbf{k}} \begin{pmatrix} \xi_{\mathbf{k}} + \frac{1}{2}\lambda\rho_3\sigma_3 & \mathbf{d}\cdot\boldsymbol{\sigma} i\rho_2 \\ -\bar{\mathbf{d}}\cdot\boldsymbol{\sigma} i\rho_2 & -\xi_{\mathbf{k}} + \frac{1}{2}\lambda\rho_3\sigma_3 \end{pmatrix} \Psi_{\mathbf{k}}\,, \tag{1}$$

* joncurtis@ucla.edu
† prineha@ucla.edu

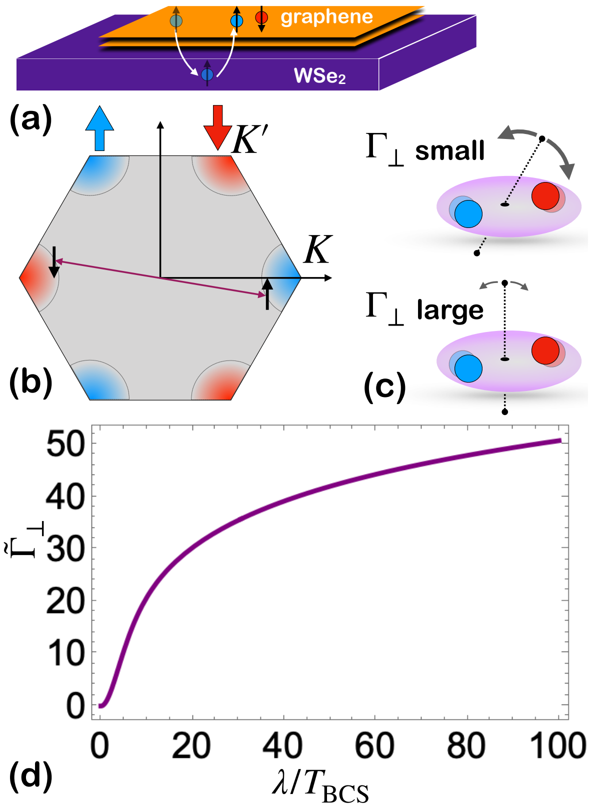

FIG. 1. Schematic overview of system considered. (a) Illustration of Bernal bilayer graphene (BBG) on top of WSe_2. Hybridization with the WSe_2 induces an Ising spin-orbit coupling in the graphene. (b) f-wave triplet pairing in the graphene Brillouin zone. Pairing is between $\mathbf{K}$ and $\mathbf{K}'$ so that electrons are in Kramers pairs with total momentum zero. Ising spin-orbit coupling locks the spin to each valley. (c) $\Gamma_\perp$ governs the scale of the d-vector fluctuations. For small $\Gamma_\perp$ fluctuations of the orientation of $\mathbf{d}$ are sizeable, whereas for larger $\Gamma_\perp$ the $\mathbf{d}$-vector is strongly pinned along the z-axis. (d) Calculation of dimensionless pinning energy $\tilde{\Gamma}_\perp = \Gamma_\perp/N(E_F)$ as a function of the induced Ising spin-orbit coupling λ, relative to the BCS pairing temperature.

with $\xi_{\mathbf{k}}$ the normal state electronic dispersion around the Fermi level at each valley and λ the induced Ising spin-orbit coupling. Superconductivity is captured by the order parameter $\mathbf{d}$ which in mean-field is given by $\mathbf{d} = g_f \sum_{\mathbf{k}} \langle \psi^T_{\mathbf{k}} \left(\rho_2 \sigma_2 \boldsymbol{\sigma}\right) \psi_{-\mathbf{k}} \rangle$. Here we focus in particular on pairing which is isotropic, valley-singlet, and spin-triplet [15], recently argued to be generically favorable in many graphene-based systems [9, 14, 16, 17]. In the case of more complex orbital or valley pairing textures, additional collective fluctuations are expected to appear [18]. [19]

Our crucial assumptions are (i) intrinsic graphene has $SO(3)$ spin-symmetry (while in theory this is not true, in practice this is a good approximation), (ii) other pairing interactions (e.g. singlet pairings) are negligible (while not crucial, this greatly simplifies our theory), and (iii) the normal state respects all symmetries (this assumption is crucial; we elaborate in the Supplemental Material [20] on what happens if the normal state is spin-polarized). In short, we expect our mechanism is not relevant to systems with a spin-polarized normal state, where $SO(3)$ spin-symmetry is already broken by the large exchange splitting. In these systems, triplet superconductivity may appear even without external field or induced spin-orbit coupling. We also note that the induced spin-orbit coupling $\lambda \sim 0.7$ meV inferred from experiment [6] could rival the Fermi energy of the small pockets, requiring a more detailed treatment in future work and possibly challenging (ii).

Using (1) and assumptions (i-iii) we project the Ising spin-orbit coupling on to the dominant triplet manifold and derive a Ginzburg-Landau free-energy for the triplet order parameter $\mathbf{d}$ near the critical point (see Supplemental Material for details). Within the weak-coupling BCS limit [21], we find

$$f = K|\nabla \mathbf{d}|^2 + r|\mathbf{d}|^2 + \Gamma_\perp |\mathbf{d}_\perp|^2 + \frac{1}{2}u|\mathbf{d}|^4 + \frac{1}{2}v(i\mathbf{d}\times\mathbf{d}^\star)^2, \tag{2}$$

with $r \sim \log(T/T_{\rm BCS})$ changing sign at the mean-field BCS temperature, and $\Gamma_\perp$ the spin-orbit induced pinning energy, with $|\mathbf{d}_\perp|^2 = |d_x|^2 + |d_y|^2$ (see discussion below). The nonlinearity u respects an enlarged $SU(3)$ symmetry while the nonlinearity $v = u$ further disfavors the spin-polarized condensate, reducing the symmetry to $SO(3)$.

Crucially, for nonzero λ we find a finite $\mathbf{d}$-vector pinning energy $\Gamma_\perp$ [22], which selects the z-axis as the favored orientation of the $\mathbf{d}$-vector, depicted in Fig. 1(c). This pinning energy, $\Gamma_\perp$ is plotted as a function of λ in Fig. 1(d). For small $\Gamma_\perp$, corresponding to intrinsic graphene, the actual T_c will be dramatically suppressed due to strong fluctuations, as we now show.

Goldstone Modes—We now explore the phase diagram of the Ginzburg-Landau functional in Eq. (2) as a function of temperature T and pinning energy $\Gamma_\perp$ by using a self-consistent large-N expansion in the number of spin components (see Supplemental Material), so we may tune the order parameter symmetry continuously and in a controlled way. Similar models have already been studied extensively in the context of BKT transitions in ultracold spinor condensates [23–25], where Monte Carlo studies have been conducted for a range of anisotropy and interaction parameters [25]. In this work, we will focus on the qualitative effect that Gaussian fluctuations have on the phase diagram, rather than quantitatively determine the phase diagram since there is still great debate about the model and parameters.

Since this is a Gaussian approximation, it does not include topological defects such as vortices and is expected to underestimate the role of fluctuations. In particular, this approach does not allow us to distinguish between long-range ordered phases and quasi-long-range ordered (QLRO) phases. However, it is known that in two-dimensions at finite-temperature phase fluctuations do not permit true long-range order, and thus when we discuss the symmetry breaking phases with $\langle d_z \rangle \neq 0$ it is understood that these are in reality QLRO. Nevertheless, this approach allows us to study the impact of spin-orbit coupling on fluctuations in a controlled way by finding the renormalized critical temperature T_c where the $\langle d_z \rangle$ QLRO condensate develops. We also find a small region of $4e$ QLRO superconductivity at small anisotropy which will be discussed later.

The quantity of interest is the condensate $\psi \sim \langle d_z \rangle$. Within mean-field theory, this exhibits a phase transition at $T_c = T_{\rm BCS}$. However, once order-parameter fluctuations are taken in to account [26, 27], we find the renormalized critical temperature T_c drops and is now given by

$$\log\left(\frac{T_c}{T_{\rm BCS}}\right) = 2\mathrm{Gi}_{(2)} \log \mathrm{Gi}_{(2)} + \mathrm{Gi}_{(2)} \log \tilde{\Gamma}_\perp . \tag{3}$$

Here $\tilde{\Gamma}_\perp = \Gamma_\perp / N(E_F)$ is the dimensionless pinning energy relative to the density-of-states.The important parameter here is the Ginzburg-Levanyuk number [26, 27] $\mathrm{Gi}_{(2)} = T_{\rm BCS}/E_F$, which measures the strength of superconducting fluctuations.

The Ginzburg-Levanyuk parameter is not known experimentally in BBG since it requires measuring the "pseudogap" temperature $T_{\rm BCS}$. Nevertheless, in other graphene-based superconductors, it has been argued that $\mathrm{Gi}_{(2)}$ may be quite large, with $T_{\rm BCS}/T_F \sim 0.1$ [28], and tunneling spectroscopy measurements have found evidence for a substantial pseudogap [29, 30]. In BBG, superconductivity occurs proximal to phases which seem to feature small Fermi surfaces with E_F as low as 0.6 meV (see Supplemental Material). Roughly estimating $T_{\rm BCS} \sim 0.5$ K [31], we then find $T_F \sim 7$ K, in line with strong-coupling $\mathrm{Gi}_{(2)} \sim 0.07$.

Intrinsic spin-orbit coupling in graphene is bounded by 40 μeV [32]; once placed on top of WSe_2 it is estimated to be of order 0.7 meV, corresponding to a near 20-fold increase in λ and 60-fold increase of $\Gamma_\perp$. *Conservatively*, this may then yield an increase in T_c by a factor of 33%, however since we have only gone to leading order in $1/N$, our approximations likely underestimate the size of the effect.

This is illustrated schematically in Fig. 2, which shows the phase diagram *before* including vortices, which are responsible for the BKT transition known to occur in all two-dimensional superfluids. We expect that including vortices will lead to a further suppression of the superfluid phase [25] down to the relevant BKT temperature based on the topological classification of the different defects. However, our estimates indicate that at least some part of the observed spin-orbit induced enhancement of T_c may be due to the suppression of soft triplet fluctuations.

Magnetic Field—In addition to spin-orbit coupling, we consider breaking the spin-rotation symmetry by applying an in-plane magnetic field, as in Ref. [5]. The relevant term in the Ginzburg-Landau expansion is not $\Gamma_\perp |\mathbf{d}_\perp|^2$, but rather

$$f_{\rm Zeeman} = -\mu_{\rm eff} H_\parallel \mathbf{e}_x \cdot (i\mathbf{d} \times \mathbf{d}^\star), \tag{4}$$

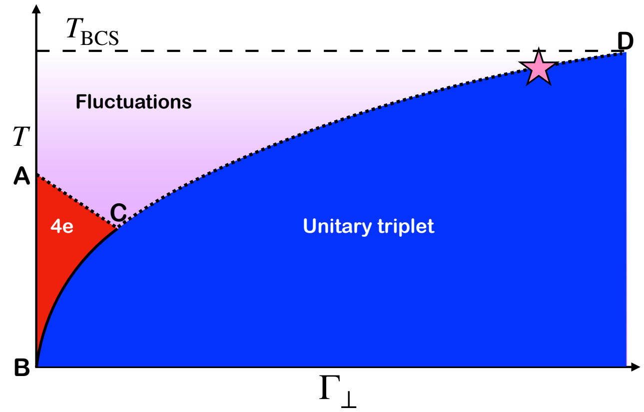

FIG. 2. Qualitative phase diagram of triplet superconductivity in graphene in the $T-\Gamma_\perp$ plane. Below $T_{\rm BCS}$ (dashed line) there are strong superconducting fluctuations. The transition to the QLRO unitary triplet phase occurs below the renormalized T_c (dashed line **C-D**), which after including vortices occurs via a $2e$ BKT transition. Near $\tilde{\Gamma}_\perp = 0$ Goldstone modes strongly suppress triplet order, but may allow for a QLRO $4e$ singlet phase below T^* (dashed line **A-C**), via a $4e$ BKT transition. Within the $4e$ QLRO phase there may be a second Ising transition which spontaneously breaks the residual $\mathbb{Z}_2$ symmetry (solid line **B-C**). In the presence of a substrate with large spin-orbit coupling, we reside at the star shown, where T_c is greatly enhanced.

where $H_{\rm ext}$ is the applied in-plane magnetic field that couples to the spin-polarized condensate, with effective magnetic moment of $\mu_{\rm eff}$.

We find $\mu_{\rm eff} \sim 2\mu_B N'(E_F)/\tilde{g}_f$ where $N'(E_F)$ is a measure of the particle-hole asymmetry and $\tilde{g}_f = N(E_F) g_f$ is the dimensionless BCS coupling constant in the relevant channel (see Supplemental Material). We estimate that for a 100 mT field, $\mu_{\rm eff} H_{\rm ext}/N(E_F) \sim \mathrm{Gi}_{(2)} \times \frac{2\mu_B H_{\rm ext}}{T_{\rm BCS}} \times \frac{1}{\tilde{g}_f} \sim 0.2$, assuming a typical BCS coupling constant of $\tilde{g}_f \sim 0.1$ [33].

In mean-field theory, this term slightly raises T_c and causes the transition to now enter into a fully spin-polarized condensate, analogous to the A_1 phase of ^{3}He [34]. To see this, change basis to the Zeeman sublevel condensates $\Psi = (\Psi_{+1}, \Psi_0, \Psi_{-1})^T$, whereupon the quadratic free energy is then characterized by the potential $V^{(2)} = \Psi^\dagger (r - \mu_{\rm eff} \mathbf{H}_\parallel \cdot \hat{\mathbf{S}})\Psi$. Near T_c the gain from Zeeman energy will dominate the cost and the condensate will always polarize.

Magnetic field can also suppress fluctuations by breaking spin-rotation symmetry, with all remaining neutral modes now acquiring a gap of order $|\mu_{\rm eff} H_{\rm ext}|$. We therefore expect similar behavior as the case of the Ising spin-orbit coupling, with the replacement of $\Gamma_\perp \to \mu_{\rm eff} H_{\rm ext}$. Comparing the pinning energies, we see that the spin-orbit interaction with $\Gamma_\perp \sim 30\, N(E_F)$ is roughly 150 times stronger than the pinning energy of the 100 mT field $gH_{\rm ext} \sim 0.2\, N(E_F)$. It is therefore plausible that there will be a significant increase in T_c when comparing the 0.7 meV spin-orbit interaction to the 100 mT magnetic field.

4e-Condensate—We now return to the existence of $4e$-pairing. One can form a charged $4e$ order parameter $\chi = \langle \mathbf{d}^2 \rangle$ which breaks $U(1)$ gauge symmetry but not $SO(3)$ rotation symmetry. Since this order is more robust against magnetic fluctuations it can appear at small $\Gamma_\perp$.

This is shown in the phase-diagram (greatly exaggerated in size) in Fig. 2. The boundary of this phase (see Supplemental Material) is found to appear at a temperature $T^\star$ given relative to T_c as

$$\log\left(\frac{T^\star}{T_c}\right) = \tilde{\Gamma}_c - \tilde{\Gamma}_\perp - \mathrm{Gi}_{(2)} \log\left(\frac{\tilde{\Gamma}_\perp}{\tilde{\Gamma}_c}\right). \qquad (5)$$

$T^* > T_c$ decreases with increasing $\Gamma_\perp$ before eventually joining the $2e$ phase in a multi-critical point at critical anisotropy $\tilde{\Gamma}_c = \frac{1}{2}\mathrm{Gi}_{(2)}$.

This phase has fluctuating triplet pairs which retain their phase coherence in spite of a random local spin orientation. Such phases have been proposed to be relevant to a number of graphene superconductors [35–37], as well as ultracold quantum gases [23, 25], and feature a flux quantum of $h/4e$ —half the usual value.

Cooling $T < T_c < T^*$ we expect a second transition into the $2e$ phase, shown in Fig. 2 as the blue region at lower temperatures below the red region. Because the $4e$ condensate has half flux-quantization, it only fixes the phase of the condensate modulo π, leaving a remnant $\mathbb{Z}_2$ symmetry which is broken at T_c. In reality however, this is quite subtle [25] and may be complicated by competing orders [38].

Experimental Signatures—It may be possible to tune the induced Ising spin-orbit coupling, and hence the pinning energy $\Gamma_\perp$, by adding spacer layers of an inert material or even by tuning the twist-angle between the BBG and WSe_2 [10] in order to confirm the dependence of T_c on $\Gamma_\perp$. Nernst effect measurements [39–42], and pair-tunneling probes [43], or proximity-induced collective modes [44] may help identify preformed pairs.

Using scanning SQUID [45], NV magnetometry [46, 47], or surface impedance [48] measurements it may be possible to directly identify the magnetic neutral modes. To this end, we calculate the dynamical magnetic susceptibility $\chi_\parallel(\omega, \mathbf{q} = 0)$ for in-plane fields in the normal-state above T_c (see Supplemental Material). Due to superconducting fluctuations (and neglecting $4e$ order) we find $\Im[\chi_\parallel(\omega)]/\omega \propto \mathrm{Gi}_{(2)}/(\tilde{r}_{\text{eff}} + \tilde{\Gamma}_\perp)^2$, with $r_{\text{eff}} \to 0$ at T_c. This produces a near-singular scaling of the magnetic noise as $T \to T_c^+$ (ultimately stopped by $\tilde{\Gamma}_\perp > 0$), manifesting clearly in the probes mentioned above.

Finally, tunneling spectroscopy can establish the existence of a pseudogap phase above the actual transition, which would confirm the presence of strong-fluctuations. This has already been accomplished in the case of twisted bilayer and trilayer graphene, and has indicated strong-coupling physics [28, 29].

Conclusion—We have presented evidence which supports the claim that graphene multilayers, and in particular BBG, may host triplet superconductivity. In its intrinsic form, graphene has almost perfect spin-rotation symmetry and this, combined with the two-dimensional nature of graphene, leads to remarkably strong fluctuation effects which shape the phase diagram. Both in-plane Zeeman field and induced Ising spin-orbit coupling suppress these fluctuations and can lead to an enhancement of superconductivity, consistent with experiments.

While our work is to some degree universally applicable to triplet pairing in graphene, there remain a number of complications due to the particular proximal isospin-polarized metals, or possible spin-polarized normal states. It therefore remains important to understand how these polarized metals couple to superconducting fluctuations. In particular, it is interesting to consider the coupling fluctuating triplet order to various density-wave orders, such as the intervalley coherent order [5, 6, 38], with the joint fluctuations of these phases perhaps explaining aspects of the system absent magnetic field. By expanding the symmetry group from $SO(3) \times U(1)$ to include other symmetries,such as valley $U(1)$, one might obtain a unified [49–51] model of both the isospin ordered and triplet paired states, similar to Refs. [52, 53]. Recent proposals for spontaneous spin-valley locking [54] may also naturally fall out of a higher-symmetry parent phase which spontaneously breaks $SO(3)$ before $U(1)$, though in this case the high-symmetry of graphene is again relevant, as there now should be neutral modes associated to fluctuations of the spin-valley-locking axis. Ultimately, graphene offer a unique opportunity to study superconductivity beyond mean-field and in the presence of strong correlations.

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[20] See [URL] for Supplemental Material, which includes References [55–57].

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