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Functional Renormalization Group Study of Superconductivity in Rhombohedral Trilayer Graphene

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We employ a functional renormalization group approach to ascertain the pairing mechanism and symmetry of the superconducting phase observed in rhombohedral trilayer graphene. Superconductivity in this system occurs in a regime of carrier density and displacement field with a weakly distorted annular Fermi sea. We find that repulsive Coulomb interactions can induce electron pairing on the Fermi surface by taking advantage of momentum-space structure associated with the finite width of the Fermi sea annulus. The degeneracy between spin-singlet and spin-triplet pairing is lifted by valley-exchange interactions that strengthen under the RG flow and develop nontrivial momentum-space structure. We find that the leading pairing instability is *d*-wave-like and spinsinglet, and that the theoretical phase diagram versus carrier density and displacement field agrees qualitatively with experiment.

Introduction—Recent experiments have demonstrated that graphene multilayers can exhibit rich strongcorrelation physics [1–18], including broken spin/valley flavor symmetries and superconductivity, when the layers are twisted to magic angles that yield extremely flat moiré superlattice bands [19–23]. Twist angle changes during processing make systematic studies more difficult, however, and devices inevitably have some twistangle disorder [24]. For this reason, the recent observation [25, 26] of superconductivity and broken flavor symmetries in untwisted rhombohedral trilayer graphene (RTG) has been a pleasant surprise. In the regime of displacement field and carrier density where strong correlations have been observed, the normal state is a twodimensional hole gas with a distorted annular Fermi surface, which has been precisely characterized using quantum oscillation measurements [25, 26] that take advantage of the exceptional sample perfection. The strongest superconductivity appears as an instability of a flavorsymmetric paramagnetic normal state and has an inplane critical magnetic field that seems most compatible with spin-singlet pairing [27, 28].

Several ideas have already been explored in connection with superconductivity in RTG [29–35]. Conventional acoustic-phonon-mediated attraction [29] can explain superconductivity only if direct Coulomb interactions between electrons do not play a significant role. The experimental observation of a nearly temperature-independent resistivity up to 20 K suggests that electron-phonon interactions are relatively weak [25], however, arguing against this mechanism. Inter-valley coherence fluctuation mediated pairing was proposed as a possibility [30, 31, 33], motivated by the experimental observation that superconductivity is proximal to a phase transition to a partially isospin polarized (PIP) phase [30]. However, the observation of a sudden jump in quantum oscillation frequencies between superconducting (SC) and PIP phases [25] indicates that the phase transition is first order and therefore

that these critical fluctuations will not be strong. Since superconductivity in RTG is in the clean limit, the Kohn-Luttinger (KL) mechanisms [36] has also been considered as a possibility [32, 35]. It was demonstrated that superconductivity can arise from the combination of annular Fermi surfaces and long-range Coulomb repulsion in RTG when a random phase approximation (RPA) is employed [32, 37].

In this Letter, we apply the functional renormalization group (FRG) method to investigate the pairing mechanism and symmetry in RTG. We start with longrange bare Coulomb interactions, and include a valleyexchange interaction that is ~ 100 times weaker than intra- and inter-valley interactions because the Fermi surfaces are small in size compared to the RTG Brillouin zone. We find that the valley-exchange interaction is enhanced under the RG flow and develops nontrivial momentum-space structures that cannot be represented by a simple momentum independent inter-valley Hund's coupling [30, 31]. The enhanced valley-exchange interaction breaks valley-SU(2) symmetry and lifts the degeneracy between spin-singlet and spin-triplet pairing. The renormalized pairing interactions develop features at cross-annulus momentum-transfers due to inter-Fermi surface particle-hole fluctuations. For experimentally relevant ratios of the annulus radii, these features favor dwave spin-singlet pairing and *p*-wave spin-triplet pairing on the inner and outer Fermi surfaces, respectively. The competition between pairing and particle-hole channel instabilities is sensitive to the Fermi-level density of states as well as the precise annulus shape.

Model— We employ the well established six-band continuum model detailed in the Supplementary Information (SI) [39] that accounts for long-range Coulomb interactions, gate and dielectric screening, and all pertinent details of the low-energy electronic structure. In the following calculations, we choose the displacement field-induced electrostatic potential $\Delta_d = 30$ meV and



FIG. 1. (a) Patching scheme for the annular Fermi surfaces (solid curves). The momentum space around each valley is divided into 48 patches. The circles on the Fermi surfaces specify the wavevectors at which we evaluate 4PVs. The dotted curves identify the wavevectors at which energy maxima occur along radial directions, and separate patches that belong to the inner and outer Fermi surfaces in our FRG calculations. (b) Diagrammatic representation of the one-loop RG flow equation for 4PVs, including particle-particle (PP), exchange (EX) and forward scattering (FS) contributions. Here the temperature T is the RG flow time, and the slashes on the propagators denote temperature derivatives [38].

carrier density $n_e = -1.75 \times 10^{12} \text{ cm}^{-2}$ as representative [39]. The corresponding annular Fermi surfaces are illustrated in Fig. 1(a). To capture the competition between superconductivity, inter-valley coherence, and spin-polarized half metals in RTG, we employ the temperature-flow FRG scheme because it properly accounts for small-q particle-hole fluctuations [38, 45, 46]. The technical details of the present study are similar to earlier works [38, 47–49], especially those in multi-orbital systems with more than one Fermi surface [50–52]. To implement numerical FRG calculations, we set the initial temperature to $T_0 \sim 11600 \text{ K}$ [39] and employ the N-patch scheme [38, 53] illustrated in Fig. 1(a).

The one-loop FRG flow equations for 4-point vertices (4PVs) with spin-SU(2) symmetry are depicted diagrammatically in Fig. 1(b). These diagrams are similar to KL-mechanism diagrams [36], previously employed to study pairing instabilities in graphene-based systems [32, 35, 54–61]. The essential difference between the KL diagram and the FRG flow equation is that the former focuses only on the irreducible pairing vertex while the latter retains all 4PVs on an equal footing, making it possible to explore competing orders. Moreover, particle-hole fluctuations that do not develop instabilities can upon reducing temperature still give progressively larger contributions to the RG flow of the pairing vertex, leading to an enhancement of the pairing instability. Appealing to standard rescaling and power count-



FIG. 2. (a) Schematic diagrams of $u_{a,t,e}$. (b)-(c) Renormalized pairing interactions at temperature T = 70 mK, including (b) $u_t(n_1, \bar{n}_1; \bar{n}_2)$ and (c) $u_e(n_1, \bar{n}_1; n_2)$. The bottom-left and top-right 24×24 blocks are scatterings on the inner and outer Fermi surfaces, respectively. The remaining blocks are scatterings between the inner and outer Fermi surfaces.

ing arguments [47, 49], we focus on the zero-frequency 4PVs with momenta on the Fermi surface. We therefore choose the wavevectors marked by circles in Fig. 1(a) to represent each patch and approximate the 4PVs by $u(\mathbf{k}_1, \mathbf{k}_2; \mathbf{k}_3) = u(n_1, n_2; n_3)$ for all wavevectors \mathbf{k}_i in the same patch n_i [62]. The fourth wavevector is determined by momentum conservation. As shown in Fig. 2(a), the 4PVs can be classified as intra-valley (u_a) , inter-valley (u_t) , or valley-exchange (u_e) using the definitions

$$u_{a}(n_{1}, n_{2}; n_{3}) = u(Kn_{1}, Kn_{2}; Kn_{3}),$$

$$u_{t}(n_{1}, n_{2}; n_{3}) = u(Kn_{1}, K'n_{2}; K'n_{3}),$$

$$u_{e}(n_{1}, n_{2}; n_{3}) = u(Kn_{1}, K'n_{2}; Kn_{3}),$$

(1)

where the patch indices are associated with valleys. For example, n_2 in u_a and u_t are on K- and K'-valley Fermi surfaces, respectively. Time-reversal symmetry requires $u(\tau_1 n_1, \tau_2 n_2; \tau_3 n_3) = u^*(\bar{\tau}_1 \bar{n}_1, \bar{\tau}_2 \bar{n}_2; \bar{\tau}_3 \bar{n}_3)$, where \bar{n} denotes the opposite patch number of n on the same Fermi surface. Using these conventions, the valley index can be removed from the FRG flow equations (see SI [39]).

Renormalized pairing interaction— The initial values of the 4PVs are obtained by patch averaging bare Coulomb interactions using band eigenstates to calculate wave function overlap factors (see Sec. IV in SI [39]). For opposite-valley electron pairing, the relevant 4PVs are $u_t(n_1, \bar{n}_1; \bar{n}_2)$ and $u_e(n_1, \bar{n}_1; n_2)$, where the outgoing momenta belong to K and K' valleys. The initial values of u_e are much weaker (less than 1%) than those of u_t because the magnitude of the inter-valley momentumtransfer is much larger than the typical size of the annular Fermi surfaces in RTG [39]. As shown in Figs. 2(b) and (c), u_t and u_e flow to comparable magnitudes as temperature reduced to T = 70 mK. The effective pairing interactions are

$$V_{s,t} = u_t(n_1, \bar{n}_1; \bar{n}_2, n_2) \pm u_e(n_1, \bar{n}_1; n_2, \bar{n}_2), \quad (2)$$

where the upper (lower) sign is for spin-singlet (spintriplet) pairing, and the fourth patch index is restored



FIG. 3. (a) Temperature flows of pairing interaction in channels distinguished by gap function symmetries. (b) Magnitudes of spin-singlet $d \pm id$ and spin-triplet $p \pm ip$ gap functions on the Fermi surfaces. (c) Angular momentum amplitudes Δ_m of the gap function on the inner (left) and outer (right) Fermi surfaces. (b)-(c) are calculated at T = 70 mK. (d) Spin-singlet d-id and spin-triplet p-ip gap functions on the K-valley Fermi surfaces, where the length and direction of arrows specify gap amplitude and phase. (e) Schematic diagram of the inner-, inter-, and outer-Fermi surface scatterings that constitute the total pairing interaction $u_t(n_1, \bar{n}_1; \bar{n}_2, n_2)$. Similar definitions are used for $u_e(n_1, \bar{n}_1; n_2, \bar{n}_2)$. (f)-(g) Contributions from the three types of scatterings illustrated in (e) to pairing interactions in (f) spin-singlet d-wave and (g) spin-triplet p-wave channels.

based on momentum conservation. The results shown in Figs. 2(b) and (c) indicate that the inner Fermi surface prefers spin-singlet pairing because u_t and u_e share similar momentum-space structure, resulting in $|V_s| > |V_t|$. In contrast, scattering across the annular Fermi surfaces and on the outer Fermi surface prefers spin-triplet pairing because u_t and u_e possess opposite momentum-space structures, leading to $|V_s| < |V_t|$.

Pairing symmetry— The gap function at T_c is specified by the solution of the linearized gap equation. For the present study, we find that the spin-singlet and spin-triplet gap functions are mainly determined by the momentum-space structures of $V_{s,t}$ and can be well approximated by the eigenvectors of the largest magnitude negative eigenvalues of $V_{s,t}$ (see details in the SI [39]). Figure 3(a) plots several of the lowest eigenvalues of $V_{s,t}$ as a function of T. We find that the strongest spin-singlet and spin-triplet channels are respectively d-wave-like and p-wave-like, and that both channels have doubly degenerate eigenvectors. Figure 3(b) plots the magnitudes of the chiral $d \pm id$ and $p \pm ip$ combinations of these eigenvectors, which are stablized below the critical temperatures [39]. Note that the leading pairing instability is spin-singlet d-wave-like because it possesses the lowest eigenvalue as shown in Fig. 3(a). The deep minima in the gap function on the outer Fermi surface in Fig. 3(b) are due to strong mixing between different angular momentum channels. The terminology used to distinguish p- and d-wave pairings is justified by the Fourier coefficients of both gap functions given in Fig. 3(c), where one angular momentum is always dominant. Figure. 3(d) illustrates the d-idand p - ip gap structures, which are largest on the inner Fermi surface for spin-singlet *d*-wave pairing and are comparable in magnitude on the inner and outer Fermi surfaces for spin-triplet *p*-wave pairing.

Figure 3(f)-(g) decompose the spin-singlet d-wave (V_s^d)

and spin-triplet p-wave (V_t^p) pairing interactions into contributions from inner-, inter-, and outer-Fermi surface scatterings illustrated in Fig. 3(e) (details in SI [39]). We find that V_s^d is dominated by scattering on the inner Fermi surfaces while the strongest contribution to V_t^p is the inter-Fermi surface scattering, which explains the comparable pairing amplitudes on the annular Fermi surfaces in the spin-triplet p-wave channel. The ultimate pairing symmetry depends on the competition between all three types of scattering, which depends in turn on the Fermi surface shape as we show later.

Pairing mechanism— Figure 4(a) and (b) depicts the EX diagram contributions to u_t and u_e , and the associated inter-valley $(\Pi_{KK'}^{ph})$ and intra-valley (Π_{KK}^{ph}) particlehole susceptibilities are plotted in Figs. 4(c) and (d). By comparing Figs. 4(c)-(d) with Figs. 2(b)-(c), we find that the momentum-space structure of these susceptibilities is responsible for the momentum-space structure developed in the pairing interactions under FRG, which in turn control the pairing symmetry discussed earlier. For RTG, the momentum-space structure of $\Pi_{KK'}^{ph}$ and Π^{ph}_{KK} arises primarily from inter-Fermi surface nesting, as illustrated in Figs. 4(d) and (e). In fact, the hightemperature initial stage of the RG flow is dominated by FS processes that screen the long-range Coulomb interaction. The EX enhancement becomes important in the intermediate temperature regime, giving rise to the momentum-space structure of the paring interactions, which is then amplified by the PP diagram upon further reducing temperature [39]. Therefore, we argue that the KL-like finite angular momentum pairing we have found stems from particle-hole fluctuations that are enhanced by inter-Fermi surface nesting.

It is instructive to analyze the pairing symmetry using the simplified circular Fermi surface model shown in Fig. 4(f) for which the electron energy spectrum is valley



FIG. 4. (a)-(b) EX diagram contributions to $u_t(n_1, \bar{n}_1; \bar{n}_2, n_2)$ and $u_e(n_1, \bar{n}_1; n_2, \bar{n}_2)$, where blue (red) solid lines denote K(K')-valley propagators. (c)-(d) Particle-hole susceptibilities associated with (a)-(b), calculated at T = 70 mK. (e) Three equivalent inter-Fermi-surface nesting wavevectors $\mathbf{k}_{25} - \mathbf{k}_1 \approx \mathbf{k}_4 - \mathbf{k}_9 \approx \mathbf{k}_{22} - \mathbf{k}_{17}$, at which Π_{KK}^{ph} peaked: $(n_1, n_2) = (1, 25), (9, 4),$ and (17, 22) marked by stars in (d). (f) Circular annular Fermi surfaces: $k_{F1,2}$ are the two Fermi wavevectors. $\theta_{1,2}$ are the angles spanned by $k_{F2} - k_{F1}$ on the inner and outer Fermi surfaces.

independent [39]. Therefore, $\Pi_{KK}^{ph}(\boldsymbol{k}_{n_1} - \boldsymbol{k}_{n_2}) = \Pi_{KK}^{ph}(\theta)$ and $\Pi_{KK'}^{ph}(\boldsymbol{k}_{n_1} + \boldsymbol{k}_{n_2}) = \Pi_{KK}^{ph}(\pi + \theta)$, where θ denotes the angle spanned by $\boldsymbol{k}_{n_1} - \boldsymbol{k}_{n_2}$ on the Fermi surfaces. The π -phase difference between Π_{KK}^{ph} and $\Pi_{KK'}^{ph}$ is distorted by trigonal warping in Figs. 4(c) and (d). Performing Fourier series expansion in θ ,

$$-\Pi^{ph}_{KK}(\theta) = \sum_{m} A_m \cos(m\theta).$$
(3)

Since the momentum-space structures of the pairing interactions are primarily determined by those of the particle-hole susceptibilities, we approximate $\delta u_t(\theta) \propto$ $-\langle u_t \rangle^2 \Pi^{ph}_{KK'}(\theta)$ and $\delta u_e(\theta) \propto -\langle u_a \rangle \langle u_e \rangle \Pi^{ph}_{KK}(\theta)$, where $\langle u_{a,t,e} \rangle$ denote their averaged values over θ [39]. Combining with Eqs. (2)-(3), the singlet and triplet pairing interactions in the *m*-wave channel $V_{s,t}^m \propto [(-1)^m \pm 1] A_m$. As shown in Fig. 4(f), inter-Fermi surface nesting enhances $-\Pi_{KK}^{ph}(\theta)$ around θ_1 and θ_2 on the inner and outer Fermi surfaces. For this study, $\theta_1 \sim \pi/2$, leading to $-A_2 > |A_1|$, and hence preferring spin-singlet d-wave (m=2) pairing on the inner Fermi surface. In contrast, $\theta_2 < \pi/6$, which results in $A_1 > |A_2|$ and prefers spintriplet p-wave (m=1) pairing. Inter-Fermi surface scattering always prefers spin-triplet *p*-wave pairing because $-\Pi_{KK}^{ph}(\theta)$ exhibits a maximum at $\theta = 0$, as indicated in Fig. 4 (d). Overall, the competition between d-wave singlet and *p*-wave triplet pairings in RTG is sensitive to the shape of the annular Fermi surfaces.

Phase diagram— Quantitative prediction of superconducting critical temperature T_c from FRG calculations stands as a challenging issue [44,47]. Here we provide crude T_c estimates by solving linearized gap equations with an effective interaction (see details in Sec. VI of SI

TABLE I. Matrix structures and symmetries under timereversal \mathcal{T} and inversion C_2 operations of the order parameters for Pomeranchuk instability (PI), valley polarization (VP), ferromagnetism/anti-ferromagnetism (FM/AFM), and inter-valley charge/spin coherence (IVC/IVS). Here FM and AFM are distinguished by identical and opposite spin polarizations in two valleys, $s_{x,y,z}$ and $\tau_{x,y,z}$ are Pauli matrices in spin and valley subspaces, and \mathcal{K} denotes complex conjugate.

Particle-hole instability	PI	VP	FM	AFM	IVC	IVS
Order parameter	$s_0 \tau_0$	$s_0 \tau_z$	$s_z \tau_0$	$s_z \tau_z$	$s_0 au_{x,y}$	$s_z \tau_{x,y}$
$\mathcal{T} = i s_y au_x \mathcal{K}$	\checkmark	×	×	\checkmark	\checkmark	×
$C_2 = s_0 \tau_x$	\checkmark	×	\checkmark	×	×	×



FIG. 5. Phase diagrams as function of (a) n_e and (b) Δ_d . The inserts depict the annular Fermi surfaces for several typical values of n_e and Δ_d that are associated with the SC, IVS and FM states. These results are obtained by choosing dielectric constant $\epsilon = 4$ and gate-sample distance $d_s = 40$ nm.

[39]). For the electron pairing channel, the linearized gap equation is

$$\Delta_{s,t}(n) = -\sum_{n_1} \prod_{KK}^{pp} (n_1) V_{s,t}(n_1, \bar{n}_1; \bar{n}, n) \Delta_{s,t}(n_1), \quad (4)$$

where $\Delta_{s,t}$ are spin-singlet and spin-triplet order parameters, $\Pi_{KK}^{pp}(n_1)$ denotes the particle-particle susceptibility, and the renormalized pairing interactions $V_{s,t}$ are given by Eq. (2). Since both Π_{KK}^{pp} and $V_{s,t}$ are temperature dependent, T_c is estimated for each channel as the temperature at which the corresponding eigenvalue of $-\Pi_{KK}^{pp}V_{s,t}$ equals 1.

We have also explored the competition between pairing and particle-hole channel instabilities. Table. I summarizes several typical particle-hole channel instabilities (see details in Sec. VI of SI [39]). As shown in Fig. 5(a), for large hole densities that are associated with relatively thick annular Fermi seas, spin-singlet *d*-wave pairing is the leading instability. The corresponding T_c exhibits a dome-like behavior *vs.* hole density that is qualitatively consistent with experimental observations [25]. We note that experimental signatures of a SC state do appear to be present in the longitudinal resistivity at hole densities that exceed the region that can currently be identified as in the SC dome [25] The data seem to be consistent with the notion that superconductivity survives to higher hole densities, as in our calculations, albeit with substantially decreased T_c . By decreasing the hole density and moving the Fermi level toward the VHS (see Fig. 5(a)), the PIP state seen in experiment appears consistent with an IVS state emerging first and then being replaced by the FM state. In our calculations, these two states compete closely when the Fermi level of the system approaches the VHS. Since the averaged value of u_e is positive, IVS and FM always dominate over IVC and AFM (see SI [39]). Similar behavior is revealed in the phase diagram $vs. \Delta_d$ for a given n_e in Fig. 5(b). The qualitative features of these phase diagrams are therefore in good agreement with experimental observations [25].

Discussion— Numerical estimates of superconducting $T_c's$ depend on model parameters, such as the dielectric constant ϵ (see SI [39]). One general trend is that stronger Coulomb interaction results in higher T_c , contradictory to the acoustic-phonon-mediated superconductivity [29]. The experimental phase diagram [25] shows that the PIP (IVS) state emerges at a larger hole density than in our theoretical estimation. If the stability region of the IVS state is expanded to agree with experiment, it intervenes before the peak of the superconducting dome is reached. This shift would avoid the region in the phase diagram where the *p*-wave spin-triplet pairing state becomes nearly degenerate with or even dominates over the d-wave spin-singlet pairing [39]. In fact, an accurate determination of the phase boundary is obviously very demanding and beyond the scope of the present FRG study. The sudden jumps of the $T'_c s$ for the IVS and FM states shown in Fig. 5 arise from the fact that the particlehole susceptibilities possess maxima at finite temperatures due to proximity to the VHS [39]. Such a jump in T_c for the IVS state is consistent with experimental observation of the jump in quantum oscillation frequency between SC and PIP phases [25]. We have explored the effects of changing other model parameters [39]. The resulting phase diagrams share similar qualitative features with Fig. 5.

The present study explains the spin-singlet SC dome observed in RTG in terms of inter-Fermi-surface nesting that leads to *d*-wave singlet pairing. We find that the Fermi surface geometry plays the key role in determining pairing symmetry and predict that the strongest *d*-wave spin-singlet pairing emerges when the ratio of the enclosed areas of the inner and outer Fermi surfaces is $r = S_{in}/S_{out} \approx 0.176$, close to the value of 0.2 measured near the SC dome [25, 39]. Larger values of *r* obtained by increasing Δ_d or n_e lead to first-order spin/valley ferromagnetic phase transitions that reconstruct the Fermi surface [63], removing the annulus and preempting the pairing instability.

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