Unconventional Superconductivity in Magic-Angle Twisted Trilayer Graphene

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Magic-angle twisted trilayer graphene (MATTG) recently emerged as a highly tunable platform for studying correlated phases of matter, such as correlated insulators and superconductivity. Superconductivity occurs in a range of doping levels that is bounded by van Hove singularities which stimulate the debate of the origin and nature of superconductivity in this material. In this work, we discuss the role of spin-fluctuations arising from atomic-scale correlations in MATTG for the superconducting state. We show that in a phase diagram as function of doping (ν) and temperature, nematic superconducting regions are surrounded by ferromagnetic states and that a superconducting dome with Tc ≈ 2 K appears between the integer fillings ν = −2 and ν = −3. Applying a perpendicular electric field enhances superconductivity on the electron-doped side which we relate to changes in the spin-fluctuation spectrum. We show that the nematic unconventional superconductivity leads to pronounced signatures in the local density of states detectable by scanning tunneling spectroscopy measurements.

INTRODUCTION

Since the discovery of superconductivity and correlated insulating states in magic-angle twisted bilayer graphene (MATBG) [1–2], twisted van der Waals materials have become indispensable for the design of novel quantum materials at will [3]. In the quickly developing field of twistronics [4], tremendous theoretical [5–26] and experimental [27–46] efforts have been undertaken to unravel the nature of strong correlations [27–32, 37, 40–46], orbital ferromagnetism [27, 38, 39, 72] and magnetic field induced Chern correlated insulators [27–32, 37, 40–46], with preliminary evidence for possible superconductivity also present in twisted-double bilayer graphene [49–51], ABC trilayer graphene aligned to hexagonal boron nitride [74] and twisted transition metal dichalcogenides [58]. Very recently, another graphitic moiré system that features reproducible, highly tunable superconductivity (as well as correlated insulators) has been discovered: magic-angle twisted trilayer graphene (MATTG) [53–55], where the twist angle alternates by +θ and −θ between each graphene layer [see Fig. 1(a)].

Experiments on MATTG find superconductivity at doping levels between integer fillings of ν = −2 and ν = −3, where ν corresponds to the number of electrons per moiré unit cell relative to charge neutrality [53–55]. Additionally, Refs. 53 and 55 report superconductivity between fillings of ν = 2 and ν = 3, which is much weaker in Ref. 54 but can be stabilized upon application of a perpendicular displacement field. Furthermore, displacement fields are found to give rise to additional superconducting features between fillings of ν = 1 and ν = 2 and between ν = −1 and ν = −2 [53]. This demonstrates that (i) superconductivity in MATTG seems to preferentially appear in between integer fillings of the flat bands and that (ii) superconductivity in MATTG can be readily tuned through a perpendicular displacement field, which makes MATTG a particularly attractive platform for studying strongly correlated physics.

The coexistence of correlated insulating and superconducting states in MATTG has further elicited questions about their intrinsic relationship in graphene-based moiré materials. In particular, the question of whether the superconducting states are of unconventional nature and driven by electron-electron interaction, or conventional and mediated by electron-phonon coupling, is still in-
tensely debated at the moment even for MATBG [24, 32, 48]. The aim of this work is to shed light on this controversial question by presenting how unconventional superconductivity in MATTG can arise from spin fluctuation exchange on the atomic scale. Starting from a fully atomistic tight-binding description of the system, we investigate the effect of long-ranged electron-electron interactions on the phase diagram of MATTG. To this end, we derive a microscopic pairing interaction $\hat{\Gamma}_2$ in the fluctuation exchange approximation (FLEX) and solve the non-linear Bogoliubov-de Gennes equations as function of filling and temperature. Our results indicate that spin-singlet superconductivity can be driven by magnetic fluctuations in between integer fillings. At the same time, superconductivity is shown to depend sensitively on the value of the carbon $p_z$ Hubbard-$U$, which is influenced by experimental details such as environmental screening and the application of fields, and that it can be moved from one between-integer filling to another flexibly. For a suitably chosen interaction strength and without a displacement field, we find superconductivity is strongest in between fillings of $\nu = -2$ and $\nu = -3$ (with critical temperature $T_c \approx 2$ K) as well as around $\nu = +2 + \delta$, while being surrounded by ferromagnetic, semi-metallic or metallic states. We show that, for fixed interaction strength, a perpendicular electric field weakens the superconducting dome with on the hole-doped side, but enhances superconductivity at doping levels around $\nu = +2 + \delta$. Hereby we demonstrate that correlated and superconducting features driven by electronic interactions in MATTG are highly tunable by a perpendicular electric field which corroborates recent experimental findings [53, 55], in particular those of Ref. [54]. We then analyze the nature of the superconducting state further and show that according to our atomistic calculations MATTG hosts unconventional, nematic $d$-wave superconductivity that displays clear signatures of $C_{3z}$ symmetry breaking in the local density of states.

RESULTS

Atomic, Electronic and Magnetic Structure.--- The atomic structure of MATTG is constructed from three stacked sheets of graphene, where the outer layers ($l = 1, 3$) are perfectly aligned and the middle layer ($l = 2$) is twisted by $\pm \theta$ relative to the encapsulating layers, as schematically depicted in Fig. 1(a). Different regions in the moiré unit cell of MATTG can be labeled according to the stacking sequence of the trilayer. At the center of rotation, the stacking is of AAA type, where carbon atoms of all layers reside vertically displaced on top of each other, as shown in the inset of Fig. 1(a). In between the AAA regions, the local geometry successively changes from ABA to domain wall (DW) and BAB stacking. The atomic structure described is invariant under the mirror reflection symmetry $\sigma_h$ that exchanges the upper and lower layer ($1 \leftrightarrow 3, 2 \leftrightarrow 2$) and under three-fold rotations around the $\hat{z}$ axis described by the point group $C_{3z}$.

To account for lattice reconstruction effects, which have been shown to be important in the field of twistronics [47], we first relax the positions of MATTG using classical force fields (see Methods). According to the reflection symmetry $\sigma_h$, we find that the central layer remains flat, while the atoms of the outer layers undergo significant out-of-plane displacements, which is in good agreement with other work [75, 76]. In fact, recent results [75] indicates that perfect alignment between the outer layers, as considered in this study, is energetically favoured over a relative shift between the outer layers, i.e. AAB/BAA stacking, and hence should be the most relevant stacking configuration that is naturally realized in experimental samples.

To model the electronic structure of MATTG we use a microscopic tight-binding (TB) parametrization for the $p_z$ orbitals of the carbon atoms [77] combined with $ab$ initio density functional theory (DFT) simulations (see Supplementary Methods and Methods). To match the dispersion obtained from DFT, we complement the TB model with an additional onsite potential of $-35$ meV acting on the middle layer. As a result the flat bands intersect with the Dirac cone below the Dirac point, which was also noted in previous work for MATTG by Lopez-Bezanilla and Lado [76]. As shown in Fig. 1(b), we find that for $\theta = 1.61^\circ$, the low-energy electronic structure of MATTG consists of a set of flat bands (similar to MATBG), which are intersected by a Dirac cone with a large Fermi velocity [75, 76, 78] compared to the flat band kinetic energy scales. The twist angle investigated here is very close to the magic angle of $1.54^\circ$ which exhibits the smallest flat band width [79].

In MATBG, long-ranged electron-electron interactions were found to strongly alter the electronic structure, which has important consequences for the observation of broken symmetry phases. MATTG is an ostensibly similar system, and therefore, we also treat the long-ranged part of the electron interactions using a self-consistent Hartree theory in the atomistic tight-binding framework (see Supplementary Methods and Methods) [25, 80]. Through the Hartree potential, the electronic structure of MATTG in the normal state acquires a filling dependence, which shifts the electronic bands to higher or lower energies depending on the doping level. We show in Fig. 1(b) that the dispersion of the flat bands of MATTG in the normal state is indeed very sensitive to long-ranged electron interactions: removing electrons lowers the K-point energies of the flat bands relative to the $\Gamma$-point, which is similar to the doping dependence of MATBG [5, 80], and is accompanied by a global shift of the flat bands relative to the Dirac cone to more negative energies. In contrast, adding electrons increases the K-point energies of the flat bands relative to the $\Gamma$-point, and shifts the whole flat band manifold to higher energies relative to the Dirac cone. The Dirac cone with its...
FIG. 1. Unconventional superconductivity in magic-angle twisted trilayer graphene (MATTG). (a) The atomic structure of MATTG consists of three superimposed graphene sheets. While the outer graphene sheets are aligned (AA stacked) and untwisted, the inner layer (black) is twisted at an angle $\theta$ ($-\theta$) relative to the lower (upper) sheet. (b) Band structure of MATTG at $\theta = 1.61^\circ$. Long-range electron-electron interactions are taken into account through Hartree corrections that lead to a doping-dependent band structure. The flat band dispersion is strongly affected by the filling factor $\nu$ (blue: $\nu = +3$, black: $\nu = 0$, red: $\nu = -3$), whereas the Dirac cone remains unaffected. (c) Local density of states (LDOS) in the outer layer of MATTG for $\nu = -2.5$. The left panel clearly shows that the flat bands are mostly localized in the AAA regions. The right panel shows the spatial distribution of the LDOS in the top layer at energies of the remote valence band (RV$_1$), remote conduction band (RC$_1$), valence flat band (VFB) and conduction flat band (CFB), reflecting the $C_3$ symmetry of the non-interacting Hamiltonian. (d) Phase diagram of MATTG for $\theta = 1.61^\circ$ around $\nu = -2.5$. Our numerical calculations reveal a superconducting dome driven by low-energy AFM spin-fluctuations that ranges from $\nu = -2.35$ to $\nu = -2.7$ with an upper critical temperature of $T_c \approx 2$ K. The average amplitude of the order parameter $|\Delta|$ is reduced with increasing temperature and vanishes towards the integer fillings $\nu = -3$ and $\nu = -2$, where ferromagnetic phases (FM) dominate. At high temperatures, the system remains paramagnetic (PM). (e,f) Magnetic correlations in MATTG at zero (left) and nonzero $\Delta_D = \pm 30\text{ meV}$ (right) perpendicular displacement field. The lower panel displays the critical Hubbard interaction strength $U_c$ needed for the onset of magnetic order as a function of filling. The type of magnetic order is color-coded through an order parameter that continuously interpolates between AFM and FM order (red: FM, yellow: AFM, for a definition see Supplemental Methods). The upper panels display a sketch which is confirmed quantitatively for filling between $\nu = -3$ and $\nu = -2$ in (d) of parameter regions that can host unconventional SC driven by AFM spin-fluctuations for $U \approx 5\text{ eV}$ (dashed black horizontal line). As magnetic interactions can only provide the pairing glue for superconductivity as long as the system remains paramagnetic, i.e. $U < U_c$, this mechanism supports SC at $\nu = -2 - \delta$ and around $\nu = +2 + \delta$. Screening effects caused by the dielectric environment may change the position of the superconducting domes to different fillings (blue to green SC domes). Applying an electric field to the sample (panel (f)) enhances superconducting regions at the electron-doped side at $\nu = +2 + \delta$.

large Fermi velocity is insensitive to long-ranged electron interactions.

The local density of states in the outer layer of MATTG, as shown in Fig. 1 (c) for a doping level of $\nu = -2.5$, exhibits large peaks in the AAA regions for energies within the range of the flat bands (the conduction flat band CFB and valence flat band VFB). This behaviour is consistent with the earlier prediction that the flat-band physics in MATTG is similar to that of MATBG [79]. Although the flat bands have their largest spectral weight in the central layer, significant weight is distributed on the outer layers such that the van Hove singularities associated with the flat bands should be observable in scanning tunneling microscopy (STM) experiments. This is shown in the left panel of Fig. 1 (c), where the flat bands give rise to $C_3$ symmetric signatures in the LDOS. At larger energies (remote conduction band RC$_1$ and remote valence band RV$_1$), the states become more delocalized [see right panel of Fig. 1 (c)].

Having captured the effect of long-range electron-
electron interactions in the doping-dependent band structure, we study the influence of the remaining short-ranged terms, i.e., a repulsive Hubbard-$U$, by calculating the atomistic susceptibility $\chi_0$ in the magnetic channel. To this end, we employ the random phase approximation (RPA) in its static, long-wavelength limit [8, 9] (see Methods). As the effect of the Hartree potential on the band structure of MATTG is very similar to the MATBG case, the consequence we expect is a broader range of twist angles showing correlated states driven by short-range interactions [8]. For given filling and temperature $(\nu, T)$, the magnetic susceptibility $\chi_0$ contains information about (i) the critical interaction strength $U_\nu$, that is the minimal value of $U$ needed to drive the system from the paramagnetic regime ($U < U_\nu$) into magnetic order ($U \geq U_\nu$) and (ii) the type of magnetic order depending on the distribution of magnetic moments in the moiré unit cell.

In Figure 1(e) we show $U_\nu$ as a function of filling at $T = 1.3 \, \text{K}$. The color maps indicate the type of magnetic ordering [yellow: antiferromagnetic (AFM) and red: ferromagnetic (FM)]. A precise definition of the order parameter that continuously interpolates from FM to AFM order is given in the Supplemental Methods. We find that small values of $U_\nu$ are observed at (or close to) the integer fillings $\nu = \pm 3$, $\pm 2, \pm 1$ driving FM order. Small values of $U_\nu$ indicate that the system is very susceptible to this kind of magnetic order as already a small interaction value $U$ is sufficient to trigger the magnetic instability. Interestingly, for $\nu = -3, -2, \pm 1$ these dips in $U_\nu$ are surrounded by AFM regions exhibiting a much larger $U_\nu$. Such behaviour was previously observed in MATBG [7] and indicates that, depending on the value of the Hubbard-$U$, these AFM instabilities may not be strong enough to actually occur, which opens the door for possible spin-fluctuation mediated superconductivity.

To investigate the influence of an external, perpendicular electric field, we set the onsite energies of the outer layers to $\Delta_D = \pm 30 \, \text{meV}$, which models the presence of a displacement field similar in magnitude (see Supplementary Methods) to that applied in the experiments of Ref. [5] to achieve their highest superconducting transition temperatures. In Figure 1(f) we show the same RPA analysis for this additional interlayer potential. In contrast to what was found without an electric field, we now observe small values of $U_\nu$ close to $\nu = \pm 2$ electrons. We observe an increase in the small value of $U_\nu$ at $\nu = +1$, and the value at $\nu = +3$ remains relatively unchanged by the displacement field. Moreover, on the hole-doped side $U_\nu$ is increased strongly almost over the whole doping range, which is where the lowest values of $U_\nu$ were observed without a displacement field. Interestingly, now an AFM region which is surrounded by FM regions emerges around $\nu = +2.5$. Within this AFM region, $U_\nu$ is large, which yields a paramagnetic phase with AFM fluctuations for $U < U_\nu$.

**Spin-fluctuation induced Superconductivity.**—To model unconventional superconductivity in MATTG, we assume that Cooper pairs are formed due to intricate effects arising from microscopic electron-electron interactions. Hence, this approach differs from the conventional BCS theory with electron-phonon coupling as the predominant pairing mechanism. In the vicinity of magnetic (semi-metallic or metallic) instabilities described above, a spin-polarized electron may travel through the graphene lattice and polarize the other electrons' spin around it such that an attractive interaction arises between the electrons and they finally form a bound state [81]. Technically, we capture this effect of transverse and longitudinal spin-fluctuations by the microscopic pairing vertex $\Gamma_2$, which we derive using the the fluctuation-exchange approximation (FLEX). We then solve the non-linear Bogoliubov-de-Gennes (BdG) equations self-consistently, using the full atomistic FLEX pairing vertex to obtain information about the nature of the superconducting state (for details see Method section and Supplementary Methods).

When MATTG is doped near $\nu = -2.5$ in the absence of a displacement field, our unconventional BCS theory reveals nematic superconductivity that originates from AFM spin-fluctuation exchange. As shown in Fig. 1(d), as a function of filling and temperature, the system features a superconducting dome enclosed by FM instabilities at integer fillings $\nu = -2$ and $\nu = -3$. The transition temperature of the superconducting phase is substantially influenced by the spin-fluctuation spectrum. As AFM tendencies are weakened with increasing temperature and FM instabilities dominate around the integer fillings, the superconducting region is effectively confined between $\nu = -2$ and $\nu = -3$ with an upper critical temperature of $T_c \approx 2 \, \text{K}$. For even larger temperatures, the flat bands of the system are no longer resolved due to temperature broadening and the system continuously returns to ordering tendencies inherited from the untwisted system [9]. This can be clearly recognized by the FM ordering tendencies disappearing at high $T$.

The driving force behind this superconducting phase originates from low-energy AFM spin-fluctuations [81] that can provide an attractive potential for electrons in MATTG. The non-uniform real-space profile of the spin-mediated pairing vertex $\Gamma_2$ in the moiré unit cell (see Supplementary Methods) shows that these attractive components are strongest on nearest-neighbor bonds in the single graphene sheets of MATTG, thus suggesting in-plane Cooper pairs. The low-energy spin-fluctuations are strengthened in the vicinity of the magnetic instability, i.e., when the value of the repulsive Hubbard-$U$ is slightly below the critical interaction strength $U \lesssim U_c$. In this parameter regime, the system shows no magnetic order, but the attractive interactions mediated by the spin fluctuations can provide the pairing glue for unconventional spin-singlet superconductivity between the integer fillings. At the same time, singlet Cooper pairs may not be relevant close to the FM instabilities as the effective interaction $\Gamma_2$ is purely repulsive in the singlet channel.

The only free parameter in our approach is the value...
FIG. 2. Properties of the superconducting order parameter in MATTG for \( \nu = -2.5 \) and \( T = 0.2 \) K. (a) Spatially resolved atomistic gap \( |D(r_i)|/\max(|D(r_i)|) \) in the outer layer of MATTG. Our analysis reveals three degenerate real-valued ground states that break \( C_{3z} \) rotational symmetry on the moiré scale along the nematic axis \( C_2 \) (black arrow). (b) Layer-wise representation of the order parameter field. The value of \( |D(r_i)| \) is larger by a factor of \( \sim 10 \) in the middle layer compared to the outer layers with most weight being concentrated in the AAA regions. The phase of the superconducting gap is shifted by \( \pi \) between single layers. Additionally to the nematicity on the moiré scale, we find strong local atomic-scale nematicity in the orientation of the \( d \)-wave components \( \tau(r_i) \) (black streamlines). The moiré nematicity is clearly visible due to the emergence of a vortex-antivortex pattern near the AAA regions. (c) Local density of states (LDOS) in the superconducting state \( \Delta_3 \) from panel (a) in the AAA region of the outer layer of MATTG. Peaks at selected energies (1)-(4), which correspond to flat bands being gapped by the order parameter, show clear signatures of \( C_{3z} \) symmetry breaking. In subpanel (2) the influence of the \( C_2 \) nematic axis is visible: For low energies, quasiparticles are forbidden to occupy the AAA regions along the direction of \( C_2 \) due to the presence of the superconducting condensate, but accumulate on both sides where the gap amplitude vanishes. This leads to a strong degree of \( C_{3z} \) symmetry breaking as visualized by the color code in the main panel. (d) Strength of the superconducting order and its projections on the honeycomb \( d \)-wave components as a function of the Hubbard-\( U \). The \( d \)-wave order parameter increases exponentially as function of interaction strength \( U \). The \( s^+ \) projection has negligible weight. (e) Free energy of all complex superpositions of the real-valued irreducible representations \( d_{xy} \) and \( d_{xy-2y} \). The minima in free energy occur for the real-valued nematic solutions (and their \( U(1) \) transformations) only.

of the Hubbard-\( U \), which cannot easily be extracted from first-principles due to the large number of atoms in the moiré unit cell. Besides, transport measurements are very sensitive to the dielectric environment which may screen the interactions more strongly. This can be achieved by, for example, varying the distance between the MATTG sample and the metallic gate\( s \) [32, 67], using a dielectric substrate with larger dielectric constant [82] or placing an AB stacked graphene bilayer in the immediate vicinity of the sample [31]. In our study, we adopt \( U = 5.1 \) eV, which is a realistic value for graphene-based materials obtained from first-principles calculations [83, 84]. Choosing this particular value of the Hubbard interaction supports spin-fluctuation induced superconductivity for \( \nu = -2 - \delta \) on the hole-doped side and \( \nu = +2 + \delta \) on the electron-doped side, which is schematically visualized by the blue superconducting (SC) domes in the top panel of Fig. 1 (e). Based on our argument, we propose that the superconducting instabilities can shift to different fillings if the dielectric screening reduces the Hubbard-\( U \) to smaller values. For example, if the interaction strength is screened to \( U = 4.4 \) eV as visualized by the green dashed line in Fig. 1 (e), superconducting domes shift towards AFM regions in the phase diagram where \( U \leq U_c \). As can be seen in the top panel of the same Figure, this results in the formation of two superconducting domes (green) at fillings \( \nu = -1 - \delta \) on the hole doped side and at \( \nu = +1 - \delta \) on the electron doped side.

As recent experiments have demonstrated the displacement field tunability of the superconducting phase [53, 54], we further investigate the system in the presence of an electric field \( \Delta_D = \pm 30 \) meV, with the results shown in Fig. 1 (f). We observe that the spin-fluctuation spectrum undergoes major changes such that superconductivity is enhanced at the electron-doped side and a superconducting dome spreads over almost the entire filling range between \( \nu = +2 \) and \( \nu = +3 \), which is in agreement with recent experimental findings [53, 54]. This highly tunable filling dependence of the superconducting domes under the influence of an electric field is caused by modifications in the low-energy bandstructure of MATTG [70].
and hence the spin-fluctuation spectrum. As depicted in Fig. 1(e,f), FM, semi-metallic or metallic states move to the electron side at \( \nu = \pm 2, \pm 3 \) and the vacated phase space in between is taken over by superconducting domes driven by AFM fluctuations in the paramagnetic phase. We relate these findings to those of the experimental reports \([53, 54]\) in more detail in the discussion section below.

**Nematic superconducting order.**— Next, we analyze the nature of the superconducting spin-singlet order parameter \( \Delta \) that is obtained from our atomistic BCS theory for unconventional superconductivity (see Method section for details). We concentrate on \( U = 5.1 \text{ eV}, \ T = 0.2 \text{ K} \) and \( \nu = -2.5 \), firmly placing the system in a superconducting state. Here, the superconducting order shows clear nematic signatures of \( C_{3z} \)-symmetry breaking on the atomic (carbon-carbon) bond scale and the moiré length scale, see Fig. 2(a,b). First, the spatially resolved order parameter amplitude \( |\Delta(\mathbf{r})| \) obtained from averaging the order field \( \Delta = \Delta_0 \) over nearest-neighbor bonds, and shown in Fig. 2(a), depicts clear signatures of \( C_{3z} \) symmetry breaking on the moiré scale. The nematic superconducting ground-state is three-fold degenerate consisting of three order parameters with nematic axis \( C_2 \) varying by rotation around 120°. All three states are degenerate in free energy, thus breaking the original \( C_{3z} \) symmetry of the Hamiltonian spontaneously. Furthermore, we find that the order parameter is completely real-valued and thus restores time-reversal symmetry (TRS) in contrast to any chiral \( d_{x^2-y^2} \pm id_{xy} \) superconducting state. In fact, we find that complex-linear combinations of the \( d \)-wave components \( \Delta_{ij} = \cos(\theta) f_{d_{x^2-y^2}} + \sin(\theta) e^{i\phi} f_{d_{xy}} \) are energetically disfavored [see Fig. 2(e)]. The amplitude distribution of the order parameter is strongly enhanced in the AAA regions along the nematic \( C_2 \) axis in the middle layer of MATBG and is a factor of \( \sim 10 \) smaller in the outer two layers as depicted in Fig. 2(a). At the same time, the order parameter vanishes in the ABA and BAB regions as expected due to the lack of states in the non-interacting Hamiltonian.

In addition, we characterize the superconducting order parameter on the atomic scale, where the symmetry is given by the \( D_{6h} \) point group of the single graphene sheets. As the FLEX pairing vertex \( \Gamma_2 \) is most attractive on nearest-neighbor bonds, we project the gap onto the complete basis set \( f_\eta \) spanned by the irreducible representations of \( D_{6h} \), which consists of the extended s-wave \( \eta = s^+ \) and two \( d \)-wave components \( d_{xy} \) and \( d_{x^2-y^2} \), see Methods. Our analysis reveals that the real-valued order parameter shows nematic d-wave characteristics with vanishing s-wave amplitude, similar to atomic calculations in MATBG \([7, 85]\). To this end, we define a real-valued two-component vector for each carbon atom \( \mathbf{r}_j \): \( \boldsymbol{\tau}(\mathbf{r}_j) = \sum \Delta_{ij} (f_{d_{x^2-y^2}}^j, f_{d_{xy}}^j)^T \) that captures the spatially varying orientation in the \( d \)-wave components and is displayed as streamlines in Fig. 2(b). There exists a local \( d \)-wave nematicity on the carbon-carbon bond scale that forms a vortex-antivortex structure close to the AAA and ABA/BAB regions and is aligned to the \( C_2 \) nematic axis on the moiré scale. Interestingly, the phase of \( \tau \), from the middle to the outer layers of MATTG is shifted by \( \pi \), which is a consequence of the interlayer repulsion in the normal-state Hamiltonian. This \( \pi \)-locked Josephson coupling was previously also observed between the two graphene sheets in MATBG and is expected to stabilize the nematic phase. \([7, 85]\).

The averaged amplitude of the nematic superconducting order parameter \( |\Delta| = \langle |\Delta(\mathbf{r})| \rangle \) is sensitive to the choice of the Hubbard-\( U \) with respect to the critical interaction strength \( U_c \) predicted by the RPA analysis. In Figure 2(d), we show that for \( \nu = -2.5 \) and \( T = 0.2 \text{ K} \) nematic superconductivity is only present if \( U < U_c \approx 0.1 \text{ eV} \) in our approximation. Approaching the magnetic instability \( U \to U_c \), the overall amplitude of the pairing interaction \( \Gamma_2 \) increases and the gap parameter grows exponentially as function of interaction strength and density of states \( \rho(\epsilon) \) on the Fermi surface \( \propto \exp \left( -1/(\rho(\epsilon)|\Gamma_2|) \right) \) as expected in a weak-coupling theory. This also emphasizes the sensitivity of superconductivity to screening the interactions or to changes in the spin-fluctuation spectrum, as for example by a displacement field as demonstrated in Fig. 1(e,f).

The nematic properties of the superconducting state lead to clear signatures of \( C_{3z} \)-symmetry breaking in the local density of states (LDOS). Figure 2(e) depicts the LDOS at the AAA region in the outer layer of MATTG. We find that the superconducting state gaps the flat bands in MATTG, while the highly dispersive Dirac cone remains partially ungapped (within the energy resolution 0.01 meV of our calculations) as each Dirac cone shows two nodes on the Fermi surface, see Fig. 2(f). For lower energies \( E \ll |\Delta| \), quasi particle excitations are forbidden in the AAA regions along the nematic axis \( C_2 \) due to the large gap amplitude in the superconducting condensate as shown in the sub panel (2) of Fig. 2(e). For larger energies [sub panels (1, 3, 4)] the system continuously goes back to a spectral weight distribution similar to the normal-state Hamiltonian shown in Fig. 1 with a lower degree of \( C_{3z} \) symmetry breaking. To analyze the behavior of the Dirac cone and the flat bands in more detail, in Fig. 2(a) we show the density of states (DOS) in the superconducting phase either for the full system (black thick line) and restricted to the central layer (thin grey line). In the BCS formalism, condensation of Cooper pairs occurs on the energy scale of \( |\Delta| \), whereas electronic excitations are described in terms of fermionic quasiparticles with energy \( \pm E_{\alpha,n,k} \approx \pm \sqrt{(\epsilon_{\alpha,n,k} - \mu)^2 + |\Delta_{n,k}|^2} \) that are shifted relative to the energies in the normal state \( \epsilon_{\alpha,n,k} \) by the order parameter amplitude (in momentum-band space) \( |\Delta_{n,k}| \), resulting in a particle-hole symmetric DOS. In the DOS of all layers, we find separate features of the Dirac cone (highlighted by the red line) and the flat bands. While the flat bands are fully-gapped (black line), the Dirac cone remains partially ungapped and leads to
FIG. 3. Quasiparticle density of states (DOS) in the superconducting phase of MATTG for $\nu = -2.5$ and $T = 0.2 \, \text{K}$. (a) The density of states in the superconducting state captures fermionic quasiparticle excitations with energies $\pm E_{n,k} \approx \pm \sqrt{(\epsilon_{n,k} - \mu)^2 + |\Delta_{n,k}|^2}$ that result in the DOS being particle-hole (ph) symmetric. In the DOS of all layers (black line), we find clear and separate features of the Dirac cone (highlighted by the red line) and the flat bands: While the flat bands are fully-gapped on an energy scale of $\sim 1 \, \text{meV}$ (black line), the Dirac cone leads to separate linear signatures $\sim |E|$ (green dashed line) in the DOS. The partial DOS of the middle layer of MATTG (grey line) shows no contribution from the Dirac cone's DOS $\sim |E|$, but contains contributions of the flat bands only. This is in agreement with the Dirac cone having dominant spectral weight in the outer layers, whereas the flat bands dominate in the middle layer. (b,c) Quasiparticle energy landscape $E(k)$ as function of momentum in the Brillouin zone of MATTG. In the superconducting phase, the Dirac cones at $K$ and $K'$ each show two nodes in the quasiparticle spectrum reflecting the $C_{3z}$ symmetry breaking nature of the nematic superconducting phase in momentum space. These nodes become distinguishable by the dark blue regions in (c) indicating the vanishing gap amplitude $|\Delta_{n,k}| \rightarrow 0$ as the quasiparticle energies approach $E(k) \rightarrow 0$. (d) Schematic sketch of the Dirac cone in the normal-state (upper panel) and superconducting phase (lower panel). In the non-interacting case $\bar{\Delta} = 0$ and at filling $\nu = -2.5$, the chemical potential (yellow plane) cuts the Dirac cone at energies higher than the Dirac point such that the tip of the Dirac cone is mirrored by ph-symmetry and the Fermi surface consists of a ring (black line). In the superconducting phase, the quasiparticle spectrum shows two nodes at this Fermi surface (green arrows), but is gapped away from these points.

First, the flat bands of MATTG are fully-gapped on an energy scale of $\sim 1 \, \text{meV}$ (black line) corresponding to the average gap amplitude $|\Delta|$. Although the gap can reach values of $10 \, \text{meV}$ in the AAA regions where it is strongly amplified, the DOS is sensitive only to the spatially averaged value of the gap. Comparing this to the density of states of only the middle layer (thin grey line) illustrates that the Dirac cone has dominant weight on the outer layers only: The linear contribution to the density of states of all layers is not present in the central layer and a true gap of $\sim 1 \, \text{meV}$ opens up. This shows that the dominant contribution to the flat bands resides in the central layer.

Next, we analyze the band splitting of the Dirac cones by calculating the quasiparticle spectrum $E(k)$ throughout the Brillouin zone of MATTG, see Fig. 3 (b). The Dirac cones located at $K$ and $K'$ each show two nodes in the quasiparticle spectrum that reflect the $C_{3z}$ symmetry breaking of the nematic superconducting state in momentum space. In Figure 3 (c), these nodes become visible by the dark blue regions indicating the vanishing gap amplitude $|\Delta| \rightarrow 0$ as the quasiparticle energies approach $E(k) \rightarrow 0$. In Figure 3 (d) we sketch the situation close to the Dirac cone for the chosen filling of $\nu = -2.5$. The chemical potential (yellow plane) cuts the Dirac cone at energies higher than the one of the Dirac point such that the tip of the Dirac Cone is mirrored due to the particle-hole symmetry of the quasiparticle spectrum and the Fermi surface consists of a ring (black line). In the superconducting phase, the particle-hole symmetric quasiparticle spectrum shows two nodes on this Fermi surface [green arrows in panel (d)]. Away from these nodes, the Dirac cone is slightly gapped. Collectively, this leads to a linearly increasing DOS around the Fermi-level, in contrast to the constant contributions that would be present if the Dirac Cones remained completely ungapped.

Since the flat bands (Dirac Cone) can be labelled according to the symmetric (anti-symmetric) irreducible representation of the mirror reflection symmetry $\sigma_h$, the different contributions to the DOS suggest that that superconductivity in MATTG is mainly driven by the flat bands, whereas superconductivity in the Dirac Cone is only induced by proximity to the flat bands. This is also reflected in the real-space properties of the order parameter $\Delta$. Since the Dirac Cone belongs to the anti-symmetric irreducible representation it only has spectral weight on the outer layers ($l = 1, 3$), where superconductivity is significantly suppressed relative to the middle layer. Meanwhile, the superconducting state is fully gapped in the central layer, which must originate from the flat bands. We hence argue that quasiparticle excitations in the Dirac Cone are only possible due to proximity...
to the flat bands.

**DISCUSSION**

Our work emphasizes the role of spin-fluctuation exchange in the formation of superconducting instabilities in MATTG. By including both long-ranged (Hartree) and short-ranged (Hubbard-\(U\)) electron-electron interactions in our microscopic theory, we find that without a displacement field, ferromagnetic ordering dominates around integer fillings \(\nu = -3, -2, 1\) and even at relatively small interaction strengths \((U \approx 3 - 4 \text{ eV})\). In between these integer fillings stabilizing antiferromagnetic order requires a larger interaction strength \((U \approx 4 - 5 \text{ eV})\). Intriguingly, estimating \(U\) from the monolayer graphene case puts the interaction strength right at the border of these different ordering tendencies. This allows superconductivity mediated by spin fluctuations in between integer fillings for values smaller than the critical interaction required to stabilize AFM order. This superconductivity is bounded by the above mentioned stronger ferromagnetic order when approaching integer fillings, naturally restricting the region of superconductivity to dome structures in a \(\nu - T\) phase diagram.

At zero electric field, choosing, e.g., \(U = 5.1 \text{ eV}\) we find the strongest superconductivity for filling between \(\nu = -3\) and \(\nu = -2\) caused by low-energy anti-ferromagnetic spin-fluctuations in the paramagnetic phase. In this regime, superconductivity occurs within a characteristic dome shape in the \(\nu - T\) phase diagram exhibiting a critical temperature of \(T_c \approx 2 \text{ K}\). This is also where Refs. \[53\] and \[54\] find clear signatures of a superconducting state. A weaker superconducting feature is predicted around \(\nu = +2\), which, however, within our approach is not surrounded by ferromagnetic order. In Ref. \[53\] superconductivity was reported at a similar filling, while Ref. \[54\] only finds very weak indications of superconductivity around that filling without displacement field.

Including a displacement field, ferromagnetic order is weakened at the hole-side and strengthened at the electron side at least for integer fillings of \(\nu = +2\) and \(+3\), which we attribute to dramatic changes in the spin-fluctuation spectrum. Superconductivity is strengthened in between these fillings in agreement with Ref. \[54\]. The additional superconducting features, which appear only in Ref. \[53\] at fillings between \(\nu = +1\) and \(+2\) and in between \(\nu = -1\) and \(-2\) deserve further study. At values of \(U\) for which we can robustly argue for superconductivity between \(\nu = \pm 2\) and \(\pm 3\), antiferromagnetic order has already taken over at filling between \(\nu = \pm 1\) and \(\pm 2\). This might hint towards an insufficiency of our mean-field approach, overestimating the strength of antiferromagnetic order due to the absence of quantum fluctuations and neglecting interchannel feedback. From a methodological point of view, it would be conducive to study MATTG using unbiased methods like the functional renormalization group or tensor product states, although this may require fundamental advance to treat the large moiré unit cell without restricting the analysis to the low-energy bands.

As next steps, we suggest the experimental scrutiny of (i) the filling-dependence of the superconducting phase when tuning the electronic interactions by screening \[32\] and of (ii) the emergent nematicity. The latter should yield clear signatures of \(C_{3z}\)-symmetry breaking in the LDOS being accessible within STM measurements. Since recent experimental work \[55\] suggests that the superconducting phase might be of non-spin-singlet type close to the filling \(\nu = -2.4\), this also deserves further theoretical investigation in our approach as ferromagnetic spin-fluctuations, which surround the SC dome in our phase diagram, are known to drive spin-triplet SC phases \[88\]. Revealing the intrinsic interplay between spin-singlet and spin-triplet phases is an exciting avenue of future research.

**METHODS**

**Moiré Structure.**— The commensurate moiré unit cell of twisted trilayer graphene (TTG) can be defined using the same convention as twisted bilayer graphene (TBG) \[77\]. We start from perfectly aligned, AAA stacked trilayer graphene with a carbon atom of each layer residing at the origin of the \(x - y\) plane, and twist the middle layer anti-clockwise with respect to the encapsulating layers about the \(z\)-axis. This creates a structure with a single moiré pattern because of the alignment of the encapsulating layers.

For atomistic methods, commensurate moiré unit cells must be constructed. Following Ref. \[77\] commensurate moiré structures are defined by two integers \(n\) and \(m\) which specify the twist angle, \(\theta\), via

\[
\cos \theta = \frac{n^2 + 4nm + m^2}{2(n^2 + nm + m^2)}.
\]

The corresponding triangular moiré lattice has a length scale that is determined through the twist angle via

\[
L(\theta) = \frac{a_0}{2 \sin(\theta/2)},
\]

where \(a_0\) is the lattice constant of graphene. For our simulations, we use \(n, m = 20, 21\) (\(\theta = 1.61^\circ\)) which leads to a commensurate unit cell of size \(L(\theta) = 8.59\text{ nm}\) that contains \(N = 7566\) carbon sites.

We relax the atomic positions of TTG using classical force fields implemented in LAMMPS \[87\]. For the intralayer potential we use AIREBO-morse \[88\] and for the interlayer potential we use Kolmogorov-Crespi potential \[89\]. We take the lattice parameter of graphene to be \(a_0 = 2.42\ \text{Å}\).

**Atomistic Modelling & Hartree Calculations.**— In real space, the atomistic tight binding Hamiltonian
takes the form
\[ H^0 = \sum_{ij,\sigma} H^0_{ij} c^\dagger_{i\sigma} c_{j\sigma}, \]  
\[ H^0_{ij} = t(r_i - r_j). \]  
(3)

The operator \( c^\dagger_{i\sigma} \) annihilates (creates) an electron at site \( r_i \) with spin \( \sigma \). The \( p_z \) electrons are coupled via Slater-Koster hopping parameters [77]:
\[ t(d) = t_\parallel(d) + t_\perp(d) \]
\[ t_\parallel(d) = V_{pp\pi}^0 \exp \left(-\frac{|d| - a_{cc}}{\delta_0}\right) \left[ 1 - \left(\frac{d \cdot e_z}{|d|}\right)^2 \right] \]  
\[ t_\perp(d) = V_{pp\sigma}^0 \exp \left(-\frac{|d| - d_0}{\delta_0}\right) \left(\frac{d \cdot e_z}{|d|}\right)^2. \]  
(4)

Here, \( e_z \) is a unit vector which points perpendicular to the graphene sheets, \( a_{cc} = 1.362 \text{ Å} \) is the vertical spacing of graphite, \( \delta_0 = 0.184 \text{ Å} \) is the transfer integral decay length, and \( a_{cc} = a_0/\sqrt{3} \) is the distance between two nearest neighboring carbon atoms (in pristine graphene). The term \( V_{pp\pi}^0 = 0.48 \text{ eV} \) describes the interlayer hopping while \( V_{pp\sigma}^0 = -2.7 \text{ eV} \) models the interlayer hopping.

To obtain good agreement with DFT calculations for the electronic structure at charge neutrality, we add an additional onsite energy term through
\[ H^\Delta_{12} = -35 \text{ meV}, \]  
(5)
where this term only exists on the inner, twisted layer (2), as indicated by the subscript of the Hamiltonian.

We performed self-consistent Hartree tight-binding calculations following the method outlined in Ref. [80]. Full details of the method have been outlined in the Supplementary Methods for completeness. We utilise a 8×8 regular grid in the Brillouin zone to calculate the electron density and a 11×11 set of moiré unit cells to converge the Hartree potential in real space. An on-site interaction of 17 eV is used with a 1/|r| potential with a dielectric constant of 1 for all other interactions. We work in the limit of zero temperature and employ a linear mixing scheme to iteratively converge the set of equations.

The full tight binding Hamiltonian consists of the following contributions
\[ \hat{H} = \hat{H}^0 + \hat{H}^H + \hat{H}^\Delta + \hat{H}^E, \]
\[ \hat{H} = \sum_{ij,\sigma} H^0_{ij} c^\dagger_{i\sigma} c_{j\sigma}. \]  
(6)

The hopping terms are included through \( \hat{H}^0 \). Hartree interactions are included through \( \hat{H}^H \), the additional on-site potential energy is included through \( \hat{H}^\Delta \), and finally an electric field is included through \( \hat{H}^E \). Further details of the Hartree contributions and the inclusion of the electric field can be found in the Supplementary Methods.

**Density Functional Theory.** — Density Functional Theory (DFT) calculations were carried out using ONETEP [90] on large twist angle TTG structures, see Supplementary Methods for results. We utilised the Perdew-Burke-Ernzerhof exchange-correlation functional [91] with projector-augmented-wave pseudopotentials [92], a kinetic energy cutoff of 800 eV, and a minimal basis of four non-orthogonal generalized Wannier functions per carbon atom. Due to the metallic nature of TTG, we use the ensemble-DFT approach [93]. Our calculations are converged such that the total energy change between iterations is less than 25 meV.

**Magnetic Susceptibility.** — To account for magnetic fluctuations, we add a Hubbard interaction acting on the graphene \( p_z \) orbitals in the Hamiltonian \( \hat{H} \):
\[ \hat{H}^U = \hat{H} + \hat{H}^l, \quad \hat{H}^l = \sum_{i\sigma} \delta n_{i\sigma} n_{i\sigma}. \]  
(7)

To treat the four-fermion term \( \hat{H}^l \), we employ the random phase approximation in the static, long-wavelength limit \( q = (q_x, q_y) \to 0 \) as presented in Ref. [9]. To this end, we calculate the atomistic magnetic susceptibility \( \chi_0 \):
\[ \chi_0 = \hat{\chi}_0(q = q_0 = 0) = \frac{T}{N_k} \sum_{k, k_0} \tilde{G}(k, k_0) \cdot \tilde{G}(k, k_0)^T. \]  
(8)

The Green’s function \( \tilde{G}(k, k_0) = G_{ij}(k, k_0) \) as a function of Matsubara frequency \( k_0 \), moiré momentum \( k \) is given by
\[ \tilde{G}(k, k_0) = (i k_0 \mathbb{I} - \hat{H}(k) + \mu \mathbb{I})^{-1}, \]  
(9)

with \( \hat{H}(k) \) the “non-interacting” tight-binding Hamiltonian including the Hartree corrections and \( \mu \) is the chemical potential corresponding to the filling factor used in the Hartree potential.

We find that the critical Hubbard-\( U \) needed for the onset of magnetic ordering is given by \( U_c = -1/\lambda_0 \), with \( \lambda_0 \) being the lowest eigenvalue of \( \chi_0 \). The magnetic ordering is proportional to the corresponding eigenvector \( \vec{\chi}_0 \). For numerical evaluation of the Matsubara sum in Eq. (8), we use the exact same frequency grid presented in Ref. [8] and thus are able to take into account the effect of low-temperature instabilities. We sample the moiré Brillouin zone with \( N_k = 24 \) points for the RPA simulations.

**Fluctuation-Exchange approximation.** — To account for pairing instabilities mediated by charge- and spin-fluctuation exchange, we derive a microscopic pairing interaction \( \hat{\Gamma}_2 \) in the fluctuation-exchange approximation (FLEX) that incorporates effects of transverse and longitudinal spin-fluctuations. In terms of the full atomistic RPA susceptibility \( \chi_0 \), the scattering between Cooper pairs in the singlet channel is described by the pairing vertex [81]
\[ \hat{\Gamma}_2(q) = U \mathbb{I} - \frac{U^2 \chi_0(q)}{1 + U \chi_0(q)} + \frac{U^3 \chi_0^2(q)}{1 - U^2 \chi_0^2(q)}. \]  
(10)

For diagrammatic details on the derivation of the FLEX pairing vertex, the reader may refer to the Supplementary Methods. In the static, long-wavelength limit \( q = \)}
(\mathbf{q}, g_0) \to 0$, the spatial profile of the long-ranged interaction vertex strongly depends on the magnetic fluctuations predicted by the RPA analysis, see Supplementary Methods. The latter limit proves to contain the relevant physics when starting with local repulsive interactions. The RPA susceptibility $\tilde{\chi}_0$ predicts spin correlations at length scales intermediate to the carbon-carbon bond scale and moiré length scale, thus being described by orderings at $g = 0$. The system hence shows the same ordering tendencies in all moiré unit cells with variable orderings at $q$.

The resulting mean-field Hamiltonian can be rewritten in the Nambu spinor basis $\psi_k = (\hat{c}_{k\uparrow}^\dagger \; \hat{c}_{k\downarrow}^\dagger)^T$. The full $2N$-dimensional Hamiltonian for the spin-dependent p$_z$ orbitals of the carbon atoms is of Bogoliubov-de Gennes (BdG) form

$$H_{\text{MF}} = \sum_k \psi_k^\dagger \begin{pmatrix} \hat{H}(k) & \hat{\Delta}(k) \\ \hat{\Delta}^\dagger(k) & -\hat{H}^\dagger(-k) \end{pmatrix} \psi_k + \text{const.} \quad (12)$$

The BdG bilinear form is diagonalized by a block-structured unitary transform $\hat{U}_k$

$$H_{\text{MF}} = \sum_k \psi_k^\dagger \begin{pmatrix} \hat{H}(k) & \hat{\Delta}(k) \\ \hat{\Delta}^\dagger(k) & -\hat{H}^\dagger(-k) \end{pmatrix} \psi_k = \sum_k \left( \hat{U}_k \psi_k \right)^\dagger \begin{pmatrix} \hat{E}_k & 0 \\ 0 & -\hat{E}_k \end{pmatrix} \left( \hat{U}_k \psi_k \right) \quad (13)$$

$$\hat{U}_k = \begin{pmatrix} \hat{u}_k & -\hat{\bar{v}}_k \end{pmatrix} \quad \text{and} \quad \hat{u}_k^\dagger \hat{u}_k = \mathds{1}.$$ 

The matrices $\hat{u}_k$ ($\hat{\bar{v}}_k$) are $N \times N$ matrices, which describe the particle (hole) amplitudes of the fermionic Bogoliubov quasiparticles $\gamma_k$ with energies $\pm \hat{E}_k$. The latter are defined as

$$\begin{pmatrix} \gamma_{k\uparrow} \\ \gamma_{l-k\downarrow} \end{pmatrix} = \begin{pmatrix} \hat{u}_k^\dagger & \hat{v}_k^\dagger \\ -\hat{\bar{v}}_k^\dagger & \hat{u}_k^\dagger \end{pmatrix} \begin{pmatrix} c_{k\uparrow} \\ c_{l-k\downarrow} \end{pmatrix}. \quad (14)$$

Together with Eq. (11), this yields a set of non-linear equations that needs to be solved self-consistently:

$$\langle c_{n\uparrow}(k)c_{l\downarrow}(-k) - c_{l\downarrow}(k)c_{n\uparrow}(-k) \rangle_{\text{MF}} = \sum_n \left( u_{n,i,k\uparrow} v_{n,j,k\downarrow}^\ast + v_{n,j,k\uparrow} u_{n,i,k\downarrow}^\ast \right) \tanh \left( \frac{E_{n,k}}{2T} \right). \quad (15)$$

To this end, we start with an initial guess $\Delta_{ij}^{\text{init}}$ and iterate until convergence is achieved using a linear mixing $\Delta_{ij}^{n+1} = (1 - \alpha)\Delta_{ij}^n - \alpha\Delta_{ij}^{n-1}$ to avoid bipartite solutions in the fixed point iteration. In all of our calculations, we set the relative error for convergence of $\Delta$ to $\epsilon = 10^{-6}$ and set the mixing parameter to $\alpha = 0.2$.

The only account for the gap parameter $\Delta$ at the Fermi surface of the Brillouin zone. This approximation may not change the underlying physics significantly as our microscopic interaction $\Gamma_2$ in the static, long-wavelength limit ($q \to 0$) carries no momentum dependence and the size of the Brillouin zone of MATTG is drastically reduced due to the large real-space unit cell. Still, we checked that our results do not change qualitatively when taking a dense mesh with up to 24 $k$-points in the BZ into account. Furthermore, we track the free energy $F$ of the system for different initial guesses and during each self-consistency run to ensure proper convergence of the BdG algorithm, see Supplementary Methods.

To determine the local amplitude of the superconducting state from the bond-related order field $\Delta = \Delta_{ij}$, we introduce a three-dimensional vector $\hat{\Delta}(\mathbf{r}_i) = (\Delta_{i,j+1}, \Delta_{i,j+2}, \Delta_{i+1,j})^T$ that contains the superconducting bonds to all (three) neighboring sites of the carbon atom located at $\mathbf{r}_i$. The norm of this order parameter field yields the lattice-resolved amplitude $|\Delta| = ||\Delta(\mathbf{r}_i)||$ follows from averaging this quantity over all sites in the moiré unit cell. Here, we only take the $l = 1, 2, 3$ nearest neighbor carbon-carbon bonds $\delta_l$ into account. This is equivalent to considering all superconducting bonds $|\Delta| \approx ||\Delta||$ as nearest-neighbor sites are strongly favoured in terms of attractive interaction generated by the spin-fluctuations exchange mechanism, see Supplementary Methods.

**Symmetry classification of the gap parameter and LDOS.**— To characterize the superconducting order parameter with respect to different pairing channels on the atomic carbon-carbon bond scale, we project the order parameter onto the complete basis set formed by the irreducible representations of the $D_{3h}$ point group

$$\Delta_\eta(n) = \sum_l f_\eta(\delta_l) (c_{n\uparrow}c_{n+\delta_l\downarrow} - c_{n\downarrow}c_{n+\delta_l\uparrow}), \quad (16)$$

where $\eta$ denotes different spin-singlet pairing channels: the extended s-wave $s^+$ and two d-wave components $d_{xy}$ and $d_{xz-y^2}$. The coefficients $f_\eta(\delta_l)$ are form factors that correspond to the pairing channel and are obtained by symmetrizing the bond functions $\delta_l$ with the irreducible representations of the point group $D_{3h}$ of graphene. The form factors are: $s^+ = (1,1,1)/\sqrt{3}$, $d_{xy} = (2,-1,-1)/\sqrt{6}$ and $d_{xz-y^2} = (0,1,-1)/\sqrt{2}$. Here, we only take the $l = 1, 2, 3$ nearest neighbor carbon-carbon bonds $\delta_l$ into account, as they are dominant in terms of attractive interaction strength, see Supplementary Methods. The local density of states (LDOS) in the

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**Note:** The content above is a natural representation of the text, extracted from a scientific document. It includes the mathematical expressions and equations as they would appear in the original document, with proper alignment and formatting. The equations are numbered for reference. The text is structured in a coherent manner, maintaining the logical flow of the argument. Any mathematical symbols and notations have been translated to plain text where possible, ensuring that the natural representation is as faithful as possible to the original content.
superconducting phase is given by

\[ \rho_i(\omega) = \sum_{k,n} |u_{ni,k}|^2 \delta(\omega - E_{n,k}) + |v_{ni,k}|^2 \delta(\omega + E_{n,k}). \]

To this end, we assume that the gap does not change significantly when calculated at different points in the (mini)-Brillouin zone (BZ) such that \( \Delta(k) \approx \Delta(k = 0) \). Applying this \( \Gamma \)-point approximation, we diagonalize the Nambu Hamiltonian Eq. \((12)\) for up to 100 \(\times\) 100 points in the BZ of MATTG. To speed up the convergence, we use an adaptive momentum mesh that allows for finer sampling around the \(K\) and \(K'\) points. The \(\delta\)-function in Eq. \((17)\) is approximated by a Lorentzian kernel with broadening \(\eta = 0.03\) meV. The density of states (DOS) is obtained from the LDOS by adding up the contribution of all sites. To capture the degree of \(C_{3z}\) symmetry breaking in the LDOS in the nematic superconducting phase, we compute the energy-resolved anisotropy \(\zeta^{C_{3z}}(E)\) by comparing the LDOS at each site \(\bar{\rho}(E)\) with its counterpart \(R_\varphi \rho_i(E)\) resulting from rotation around \(\varphi \in \{120^\circ, 240^\circ\}\):

\[ \zeta(E) = \frac{1}{2} \sum_{\varphi \in \{2\pi/3, 4\pi/3\}} \frac{|R_\varphi \bar{\rho}(E) - \bar{\rho}(E)|}{||\bar{\rho}(E)||}. \]

Note that the nematic superconducting order parameter breaks \(C_{3z}\) symmetry on the microscopic (graphene) sublattice and moiré scale. Since the latter is easier to access experimentally, we smear the LDOS in real-space with a Gaussian envelope of width \(\sigma = a_0\) such that \(\zeta^{C_{3z}}(E)\) only captures the degree of \(C_{3z}\) symmetry breaking on the moiré scale.

DATA AVAILABILITY STATEMENT

Data are available from the corresponding authors upon reasonable request.

CODE AVAILABILITY STATEMENT

Simulation Codes are available from the corresponding authors upon reasonable request.

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COMPETING INTERESTS

The authors declare no competing interests.

AUTHOR CONTRIBUTIONS

A.F., Z.A.H.G. and L.K. performed atomistic modeling. D.M.K. and L.K. conceptualized and supervised the project. All authors contributed to interpreting the results and writing the manuscript.

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Supplemental Material
Unconventional Superconductivity in Magic-Angle Twisted Trilayer Graphene

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SUPPLEMENTARY METHODS

Structural Relaxations In Fig. 1 we display the out-of-plane corrugation that occurs upon relaxing MATTG at 1.61°. At this angle, which is close to the magic angle of TTG, the z-displacements exhibit the largest effects as the unit cells are not large enough for significant in-plane relaxations to occur [1]. We find that the inner, twisted layer essentially does not undergo any out-of-plane relaxations. The outer layers undergo significant relaxations to minimise the energy of the structure. In the AAA regions of the moiré unit cell (corners of the plot), an interlayer spacing of almost 3.6 Å is reached; whereas in the ABA (or ACA) regions of the moiré unit cell, the interlayer spacing is closer to 3.35 Å. These interlayer spacings reflect the equilibrium bilayer stacking of AA and AB bilayer graphene, respectively.

![Supplementary Figure 1](image_url)

Supplementary Figure 1. Out-of-plane corrugation effects of TTG at 1.61°. (a) Outer layer above the middle, twisted layer. The lower layer essentially has an identical relaxation pattern. (b) Inner, twisted layer. The inner layer is taken as the origin of the z-axis.

Density Functional Theory In Fig. 2, we show the band structures of TTG at 3.15° and 2.45° from DFT. The low-energy electronic structure consists of a set of flat bands which intersect a Dirac cone with a large Fermi velocity. Overall, we find good agreement between the DFT electronic structure and the atomistic tight binding model, which shall be described in the next section. The main difference between the methods is where the flat bands intersect the Dirac cone. In the DFT calculations, the flat bands intersect the Dirac cone at lower energies than the Dirac point, but in the tight binding calculations they intersect at higher energies than the Dirac point. To improve the model’s accordance with both our DFT simulations and the ones from Ref. 2, we include an additional onsite potential energy that only acts on the middle layer of the system −35 meV.

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Atomistic Hartree Theory  
The method outlined here follows that of Ref. 3. Also see Ref. 4 for a similar method, or Refs. 5, 6 for continuum model approach, or Ref. 7 for a orbital approach.

The long-ranged electron interaction contribution to the Hamiltonian can be included through

$$H_{ii}^H = \int dr \phi_z^2(r - \tau_i) V_H(r),$$  \hspace{1cm} (1)

where $\phi_z(r)$ is the $p_z$ orbitals of the carbon atoms and $V_H(r)$ is the Hartree potential of these orbitals. The Hartree potential is calculated from the convolution of the electron density $n(r)$ and the screened interaction $W(r)$, given by

$$V_H(r) = \int dr' W(r - r')[n(r') - n_0(r')],$$  \hspace{1cm} (2)

where $n_0(r)$ is a reference electron density of the uniform system. The electron density is determined through

$$n(r) = \sum_{nk} f_{nk} |\psi_{nk}(r)|^2$$  \hspace{1cm} (3)

where $\psi_{nk}(r)$ is the Bloch eigenstate of the tight binding model, with subscripts $n$ and $k$ denoting the band index and crystal momentum, respectively; $N_k$ is the number of k-points in the summation of the electron density, and $f_{nk} = 2\Theta(\varepsilon_F - \varepsilon_{nk})$ is the spin-degenerate occupancy of state $nk$ (where $\varepsilon_F$ is the Fermi energy and $\varepsilon_{nk}$ is the eigenvalue of that state). Inserting the Bloch states in Eq. (3) gives

$$n(r) = \sum_j n_j \chi_j(r),$$  \hspace{1cm} (4)

where $\chi_j(r) = \sum_R \phi_z^2(r - \tau_j - R)$ (with $R$ denoting the moiré lattice vectors) and the total number of electrons on the $j$-th $p_z$-orbital in the unit cell being determined by $n_j = \sum_{nk} f_{nk} |c_{nkj}|^2 / N_k$, with $c_{nkj}$ denoting the coefficients of the eigenvectors of the tight binding model.

The reference density is taken to be that of a uniform system, $n_0(r) = \bar{n} \sum_j \chi_j(r)$, where $\bar{n}$ is the average of $n_j$ over all atoms in the unit cell, which is related to the filling per moiré unit cell $\nu$ through $\bar{n} = 1 + \nu / N$, where $N$ is the total number of atoms in a moiré unit cell [4]. This reference density is taken to prevent overcounting the intrinsic graphene Hartree contribution which should be included in the hopping parameters.

In our atomistic model, we neglect contributions to the electron density from overlapping $p_z$-orbitals that do not belong to the same carbon atom, which is equivalent to treating $\phi_z^2(r)$ as a delta-function. Therefore, we calculate the Hartree on-site energies using

$$H_{ii}^H = \sum_{jR} (n_j - \bar{n}) W_{Rij},$$  \hspace{1cm} (5)

where $W_{Rij} = V_0 / |(R + \tau_j - \tau_i)|$, where $V_0$ is the Coulomb potential energy scale with a dielectric constant of 1. If $R = 0$ and $i = j$, we set $W_{0,ii} = 17$ eV [8].

Supplementary Figure 2. DFT band structure along the high symmetry path of the moiré Brillouin zone.
To obtain a self-consistent solution of the equations, we use a $8 \times 8$ $k$-point grid to sample the first Brillouin zone to converge the electron density, and we sum over a $11 \times 11$ supercell of moiré unit cells to converge the onsite energy. Linear mixing of the electron density is performed with a mixing parameter of 0.1 or less. Typically, the Hartree potential converges to an accuracy of better than 0.1 meV per atom within 100 iterations (see Fig. 3).

As mentioned in the main text, we find strong similarities between the effect of Hartree interactions in TTG and TBG [3–7]. This is perhaps not surprising as TTG can be mapped onto a system that is TBG (at a different twist angle) and a graphene layer [9], such that we might expect similar behaviour from the constitutive parts. We find the flat bands are quite sensitive to the Hartree interactions, while the Dirac cone with large Fermi velocity is completely insensitive to these interactions. Upon electron doping (as seen in the main text and later on), the flat band electronic structure strongly distorts, with the states at the edge of the Brillouin zone increasing in energy relative to the states at the centre. The converse is true for hole doping.

These strong band deformations arise because of the strongly peaked local density of states (LDOS) in the AAA regions, with the strongest weights in the inner, twisted layer. Upon removing (adding) electrons, they are practically only taken from (added to) the AAA regions. This localisation of the flat band LDOS gives rise to a strongly varying Hartree potential, as was also found in TBG [3–7]. In Fig. 4 (c,d) we show that the Hartree potential is strongly peaked in the AAA regions when 3 electrons are removed from 1.61° TTG. This Hartree potential substantially varies with doping level but does not change significantly with twist angle, as summarised in Fig. 4 (a,b).

In Fig. 4 (a) we plot how the layer-dependent constant contribution to the Hartree potential ($\Delta_i$) changes with doping level for several twist angles. Interestingly, we find that the outer layers (1/3) have a zero constant contribution ($\Delta_{1/3} = 0$) for all doping levels and twist angles. However, the central layer (2) has a constant contribution which changes significantly with doping level, but hardly changes with twist angle. When doping within the flat bands, we find that the constant contribution increases when electrons are added, but it becomes more negative when electrons are removed. This occurs because of electrons mainly being added/removed from the central layer. The change in this constant contribution is approximately linear ($\Delta_2 \approx \Delta_i \nu$) when doping inside of the flat bands. Doping the system outside of the flat bands causes the magnitude of the constant contribution to decrease, which reflects the fact that electrons are now being added/removed from the outer layers where the Dirac cone with large Fermi velocity resides.

In Fig. 4 (b) we plot the scale of the cosine contribution to the Hartree potential, as calculated by

$$V_l(\nu) = \frac{\sum_v V_H(\nu, \tau_u) \cdot v_c(\tau_u)}{\sum_j v_c(\tau_{ij}) \cdot v_c(\tau_{ij})},$$

where

$$v_c(\tau_u) = \sum_j \cos(b_j \cdot \tau_u).$$

Here $b_j$ is the three shortest reciprocal moiré lattice vectors [3] and note the summations only run over the atoms in layer $l$. The scale of the cosine contribution to the Hartree potential is largest in the inner layer (2), as this is where electrons are mainly being added/removed from. Doping the TTG inside of the flat bands causes an approximately linear change on the cosine contribution to the Hartree potential ($V_l \approx V_l' \nu$), which is similar to TBG [3–7]. Upon doping outside of the flat bands, the cosine contribution stops changing significantly as electrons are no longer being added/removed from the peaked LDOS AAA regions.

**Supplementary Figure 3.** Convergence of the self-consistent Hartree calculations at a twist angle of 1.6°. The doping level is set to $\nu = +3$ electrons and the self-consistent calculation is based off the charge density which comes out of the non-interacting tight-binding model. The relative error to the converged value is shown as a function of number of supercells. We assume $15 \times 15 = 225$ supercells to be sufficient for a converged result as the slope of the line has almost decayed to zero.
Supplementary Figure 4. Hartree potential dependence on twist angle and doping level for the inner and outer layers. (a) Constant contribution to the Hartree potential on each layer as a function of doping level for a number of twist angles. Legend shows the layer (1/3 or 2) and the twist angle convention used for the symbols. (b) Cosine contribution to the Hartree potential on each layer as a function of doping level for a number of twist angles. (c) Hartree potential on the outer layers for 1.61° at ν = −3. (d) Hartree potential on the inner layer for 1.61° at ν = −3.

In Fig. 5, the dependence of the Hartree potential on doping and dielectric environment of a 2° structure is shown. As seen in Fig. 5 (a), for the larger dielectric constant of 4, which models the hexagonal boron nitride (hBN) substrate, the constant contribution to the Hartree potential reduces on the middle layer by 65%. Whereas, as seen in Fig. 5 (b), the cosine contribution is significantly more robust with respect to the dielectric environment, where a maximum of a 30% reduction in the Hartree potential is observed. This insensitivity of the Hartree potential to the dielectric environment was also found in tBLG [3], where it was suggested to occur because of the already large internal dielectric constant of tBLG.

The above Hartree potential can be well parameterised by the following equation

$$H_{ii}^H \approx \Delta_i'\nu + V_i'\nu \sum_j \cos(b_j \cdot \tau_{il}), \quad (8)$$

where $\Delta_i'$ is the constant contribution on layer $l$, $V_i'$ is the scale of the Hartree potential on layer $l$, $\nu$ is the doping level, $b_j$ are the three shortest reciprocal lattice vectors, and $\tau_{il}$ are the atomic positions of carbon atom $i$ in layer $l$. A similar form was used to parameterise the Hartree potential of TBG [3]. Note that the AAA regions have to reside at the corners of the moiré unit cell for this equation to work - otherwise one must rigidly shift the potential such that the peaks of the potential occur in the AAA region. This allows one to efficiently investigate other doping levels and twist angles that are not calculated self-consistently, as the Hartree potential varies smoothly in $\nu$-space and does not change significantly in $\theta$-space close to the magic angle. Note that these Hartree theory calculations were not performed with the intrinsic symmetric polarisation potential.

In the RPA calculations described later, we take $\Delta_1' = 5$ meV, $V_{1/3}' = 4$ meV and $V_2' = 6$ meV. These values are based off the dielectric environment of air, but as we have shown in Fig. 5, there is little change to the cosine contribution parameters when taking into account the dielectric environment of hBN. The constant contribution is more sensitive to the dielectric environment. As the constant contribution only shifts the flat bands, but the cosine contribution causes significant deformations, the cosine parameters are the more important contribution. Therefore, the employed parameters should still provide reliable comparisons against experiments. In Ref. 10 a similar approach
Supplementary Figure 5. Hartree potential dependence on doping level and dielectric constant for the inner and outer layers. All figures are for a twist angle of 2°. (a) Constant contribution to the Hartree potential on each layer as a function of doping level for several dielectric constants, where the legend shows the layer (1/3 or 2) and the dielectric constant (1 or 4) convention used for the symbols. (b) Cosine contribution to the Hartree potential on each layer as a function of doping level for a number of twist angles. (c) Hartree potential on the outer layers for 2° at \( \nu = -3 \) with dielectric constant of 4. (d) Hartree potential on the inner layer for 2° at \( \nu = -3 \) with dielectric constant of 4.

was taken for TBG and excellent agreement was found with experiments, which suggests that it should work for TTG too.

Electric Field We include an electric field through

\[
H_{iii}^E = \pm \Delta_D,
\]

where \( \Delta_D \) is the additional potential that electrons feel on the outer layers of TTG, the sign of which depends on the layer and direction of the field.

We performed self-consistent Hartree calculations with \( \Delta_D = 10 \) meV to investigate the effect of screening from TTG has on the perpendicular electric field. We found that at this electric field strength, the intrinsic Hartree potential is not significantly effected other than an additional layer-dependent onsite potential which screens the perpendicular electric field. Therefore, we can use the Hartree potential in the absence of a field, and screen the field by a constant.

To extract an effective dielectric constant which screens a perpendicular field, we calculate the following \( \epsilon_r = 2\Delta_D/(\Delta_1 - \Delta_3 + 2\Delta_D) \), where \( \Delta_{1/3} \) are the layer-dependent onsite Hartree potentials. For a twist angle of 2° and doping level of \( \nu = 3 \) we find a dielectric constant of \( \epsilon_r = 3.35 \), and at charge neutrality we find \( \epsilon_r = 1.87 \). Overall, we find TTG has a dielectric constant of approximately 2-3 for screening perpendicular electric fields. This value of the perpendicular effective dielectric constant is not far from the value of 3-4 for a trilayer graphene system obtain from DFT calculations of Ref. 11.

With a dielectric constant of 3 and an onsite potential energy of \( \Delta_D = \pm 30 \) meV, the corresponding displacement field is approximately 0.25 Vnm\(^{-1}\). In the experiments of Ref. 12, the optimal electric field corresponds to a 0.5 Vnm\(^{-1}\) displacement field. This is within a factor of 2 of our calculations, which is reasonable agreement.

Full Atomistic Hamiltonian As discussed in the Methods section of the main text, the full tight binding Hamiltonian consists of the following contributions:

\[
\hat{H} = \hat{H}^0 + \hat{H}^H + \hat{H}^\Delta + \hat{H}^E, \quad H = \sum_{ij\sigma} H_{ij} c_i^{\dagger} c_j^\sigma, \quad (10)
\]
In Fig. 6 we display some additional band structure figures to that shown in the main text for different twist angles. Specifically, we show a calculation with a self-consistent Hartree potential and an intrinsic-symmetric polarisation potential, but without an electric field. We can clearly see that the Hartree potential causes significant band distortions for all twist angles, with the relative distortions at smaller twist angles being more pronounced.

Supplementary Figure 6. Band structure along high symmetry path for several twist angles and doping levels, but without an applied electric field.

We show the Fermi contour of MATTG in Fig. 7 for a set of three fractional fillings of the moiré bands: \( \nu \in \{-2.3, -2.5, -2.7\} \). Nesting is only supported at a vector \( q \) close to \( \Gamma \) that is incommensurate with the Brillouin zone. Thus, magnetic ordering at \( q \neq 0 \) is unlikely to occur. As expected from the symmetry analysis presented in the manuscript, the Fermi surface possesses a six-fold rotational symmetry. Contrary to TBG, twisted trilayer graphene is not two-fold symmetric \( C_2(\hat{y}) \) around the axis \( \hat{z}(\hat{y}) \) lying within the middle graphene sheet, but only exhibits the mirror reflection symmetry \( \sigma_h \). Hence, the symmetry of Fermi surface is reduced with respect to TBG.

**Magnetic RPA** To study magnetism, we add a Hubbard interaction acting on the graphene \( p_z \) orbitals in the Hamiltonian \( H \):

\[
H^U = H + H^I, \quad H^I = \sum_{i\sigma} U n_{i\sigma} n_{i\bar{\sigma}}.
\]

We employ the well established method of static, long-wavelength magnetic random phase approximation to treat the four-fermion term \( H_I \) as presented in Ref. 13. To analyze magnetic ordering tendencies, we calculate the magnetic susceptibility \( \hat{\chi}_0 \):

\[
\hat{\chi}_0 = \hat{\chi}_0(q = q_0 = 0) = \frac{T}{N_k} \sum_{k, k_0} \hat{G}(k, k_0) \circ \hat{G}(k, k_0)^T.
\]

The Green’s function \( \hat{G}(k, k_0) = G_{ij}(k, k_0) \) as a function of Matsubara frequency \( k_0 \), moiré momentum \( k \) is given by

\[
\hat{G}(k, k_0) = (ik_0 \mathbb{I} - \hat{H}(k) + \mu \mathbb{I})^{-1},
\]

Supplementary Figure 7. Fermi contours for three fractional fillings of the moiré bands. At the fillings shown in (a),(b),(c), magnetic ordering at \( q \neq 0 \) is unlikely due to the approximate nesting vector being incommensurate with the Brillouin zone. The Fermi surface is six-fold rotational symmetric with respect to the \( \Gamma \) point of the Brillouin zone as expected from the symmetry analysis presented in the manuscript.
Supplementary Figure 8. Fermi contours for half-integer filling values of the moiré flat bands. (a-g) Filling range in integer steps from $\nu = -3.5$ to $\nu = +3.5$.

with $H(k)$ the “non-interacting” tight-binding Hamiltonian including the Hartree corrections and $\mu$ the chemical potential corresponding to the filling factor used in the Hartree potential.

From the extended Stoner criterion [14], we find that the critical Hubbard-$U$ needed for the onset of magnetic ordering is given by $U_c = -1/\lambda_0$, with $\lambda_0$ the lowest eigenvalue of $\hat{\chi}_0$. The magnetic ordering is proportional to the corresponding eigenvector $\vec{v}_0$. For numerical evaluation of the Matsubara sum in Eq. (12), we use the exact same frequency grid presented in Ref. 10 and thus are able to take into account the effect of low-temperature instabilities consistently by using the temperature-dependent number of frequencies presented in Table I. We sample the moiré Brillouin zone with $N_k = 24$ points for the RPA simulations. To show that this resolution in $N_k$ is sufficient, we performed a convergence analysis at the exemplary filling of $\nu = -2.5$ and temperature $T = 1.32$ K (see Fig. 9).

Supplementary Table I. Inverse temperatures, temperatures and number of Matsubara frequencies in the RPA summations used for the simulation of the superconducting dome. The spacing of $\beta$ (and $T$) is chosen to be logarithmic. The parameters $T = 1.32$ K and $N_\omega = 500$ were used for Figs. 1 (c) and (d) of the main text.

| $\beta$ (eV$^{-1}$) | 50000 | 32374 | 20961 | 13572 | 8788 | 5690 | 3684 | 2385 | 1544 | 1000 |
|---------------------|-------|-------|-------|-------|------|------|------|------|------|------|
| $T$ (K)             | 0.23  | 0.36  | 0.55  | 0.86  | 1.32 | 2.04 | 3.15 | 4.87 | 7.52 | 11.6 |
| $N_\omega$          | 1200  | 966   | 780   | 620   | 500  | 500  | 500  | 500  | 500  | 500  |

The three types of magnetic ordering are shown in Fig. 10. In Figure 1 (d,e,f) of the main text, we do not differentiate between the two types of antiferromagnetic ordering [Fig. 10 (a,b)] as the consequences for superconductivity are very similar. For nonzero electric displacement field, i.e. $\Delta_D = \pm 30$ meV, the types of ordering do not differ
Supplementary Figure 9. Convergence of the magnetic susceptibility. The critical interaction strength \( U_c \) is shown as a function of number of sampling points \( N_k \) in the Brillouin zone for \( \nu = -2.5 \) and \( T = 1.32 \) K. The region shaded in blue indicates what results we accept as converged. The relative error of \( U_c \) to the value at \( N_k = 486 \) is annotated. Note that the type of ordering is the same AFM ordering for all BZ samplings.

Supplementary Figure 10. Magnetic orderings found for MATTG. Linecuts of the leading magnetic order parameter along the diagonal of the rhombus shaped unit cell. Only two layers are shown as the bottom layer contribution of the order parameter is equivalent to the top layer contribution for all ordering shown. Panel (a) shows the first type of antiferromagnetic (AFM) order that is a carbon-scale AFM order modulated in each layer and on the moiré scale. Panel (b) shows AFM order on the carbon scale stronger modulation on the moiré scale such that it exhibits a node in the top layer ABA/BAB regions. Panel (c) shows ferromagnetic (FM) order with some reminiscent ferrimagnetism in the ABA/BAB regions. In every case, the middle layer (panel titled “tA”) has a much larger amplitude than the outer layers (“A”).

Qualitatively, despite the breaking of \( C_{2z} \) symmetry. Fig. 10 shows that the weight of the magnetic instabilities is mainly in the middle layer in the AAA regions, which is a manifestation of the weight of the electronic wavefunctions mostly being centered around these points (see Fig. 1 (c) of the main text). For the \( \nu - T \) region where we determine the superconducting gap (Fig. 1 (d) of the main text), Fig. 11 shows \( U_c \) (a) and the type of magnetic order (b) using the order parameter

\[
\zeta = \frac{1}{\sqrt{N}} \left| \sum_i v_i^0 \right|,
\]

where \( N \) is the number of carbon atoms per unit cell.

Pairing Vertex in fluctuation-exchange approximation (FLEX) To account for pairing instabilities mediated by charge- and spin-fluctuation exchange, we derive a microscopic pairing interaction \( \hat{\Gamma}_2 \) in the fluctuation-exchange approximation (FLEX) that incorporates effects of transverse and longitudinal spin-fluctuations, see Fig. 13. In terms of the full atomistic RPA susceptibility \( \chi_0 \), the scattering between spin-singlet Cooper pairs is described by the pairing vertex [15]

\[
\hat{\Gamma}_2(q) = U \mathbb{I} - \frac{U^2 \chi_0(q)}{1 + U \chi_0(q)} + \frac{U^3 \chi_0^2(q)}{1 - U^2 \chi_0^2(q)}.
\]

In the static, long-wavelength limit (\( q \to 0 \)), we hence fall back to the computation of the atomistic susceptibility matrix, see paragraph ‘Magnetic RPA’. The real-space dependence of the interaction vertex \( \hat{\Gamma}_{2,ij} \) is shown in Fig. 14. Fixing one of the two real-space indices to either the AAA, ABA or DW regions, the interaction profile throughout the unit cell strongly depends on the spin-fluctuation spectrum predicted by the RPA analysis. For filling ranges dominated by AFM fluctuations, the interaction vertex is staggered throughout the moiré unit cell of
Supplementary Figure 11. Magnetic instabilities of MATTG between filling factors $\nu = -2, -3$. The left panel (a) emphasizes the regions where superconducting order by a spin fluctuation mechanism is possible by graying out the regions where $U_c \geq U = 5.11 \text{eV}$. On the right hand side (b), we plot the magnetic order parameter $\zeta$ that smoothly interpolates between AFM ($\zeta = 0$) and FM ($\zeta = 1$) order.

Supplementary Figure 12. Real-space profile of the effective pairing vertex $\hat{\Gamma}_2$ in the middle (upper panels) and outer layers (lower panels) of MATTG. (a,b) Close to an antiferromagnetic (AFM) instability, the pairing vertex is staggered in real-space: Strong on-site repulsion inherited from the repulsive Hubbard-$U$ is followed by nearest-neighbor attraction. Real-space profiles are shown starting from an atom located in the AAA, ABA or DW region of MATTG, respectively. The inset shows the interlayer component, which happens to be an order of magnitude smaller than comparable intralayer terms. The overall amplitude of the interaction is enhanced in the vicinity of the magnetic phase: As $U \to U_c$ the amplitude of the pairing interaction increases notably in the AAA regions of the middle layer as depicted in sub panels (a) and (b). The amplitude in the outer layer is merely affected. (c) Close to a ferromagnetic (FM) magnetic instability at, the spin-fluctuation mediated pairing vertex is purely repulsive throughout the moiré unit cell.

MATTG: While strong on-site repulsion is inherited from the initial Hubbard-$U$ term, AFM spin-fluctuation exchange generates attractive terms living on the nearest-neighbor bonds in the single graphene sheets. In contrast, the effective spin-singlet interaction is mostly repulsive in the vicinity of FM ordering tendencies.

We stress that the interlayer component of the interaction is an order of magnitude smaller than comparable interlayer terms. This leads to the conclusion, that on the atomistic scale, in-plane Cooper pairs are strongly favoured by spin-fluctuation exchange. These observations are not surprising as the interlayer coupling between the single graphene sheets in the non-interacting Hamiltonian is supressed by the same amount as expected for van der Waals heterostructures as MATTG.

The overall amplitude of the effective pairing vertex $\hat{\Gamma}_2$ increases in the vicinity of the magnetic instability, i.e. if $U \to U_c$. As the system shows an increased susceptibility for magnetic fluctuations in the middle layer of MATTG (especially in the AAA regions), the divergence of the interaction is first observed there. To calculate a meaningful $\nu - T$ phase diagram at fixed Hubbard interaction $U$ as shown in the manuscript, we hence circumvent the divergence...
Supplementary Figure 13. Diagrams contributing to the transverse/longitudinal spin-fluctuation mediated pairing interaction $\hat{\Gamma}_2(q)$ in the spin-singlet channel. Upper Row: Transverse Spin-fluctuations $\hat{\Gamma}_2^t(q)$. The momentum transfer occurring in the polarization function in RPA is given by $q_t = k_1 + k_2$ due to momentum conservation. Lower Row: Longitudinal Spin-Fluctuations $\hat{\Gamma}_2^l(q)$. The momentum transfer occurring in the polarization function in RPA is given by $q_t = k_1 - k_2$. Only an even number of particle-hole bubbles is allowed in the diagrammatic expansion in order to preserve the spin in the upper and lower leg of the pairing interaction. The diagrams that are resummed in the longitudinal channel are connected to the particle-hole susceptibility describing screening effects of the bare Coulomb interaction.

Supplementary Figure 14. Pairing vertex as a function of broadening. Average (a) and minimum (b) value of the attractive interaction on nearest-neighbor bonds originating from the effective spin-mediated pairing vertex $\hat{\Gamma}_2$ as function of Hubbard-$U$ for different choices of the broadening parameter $\eta$.

for values $U < U_c$ at certain fillings by applying a Lorentzian broadening to the interaction vertex

$$\hat{\Gamma}_2^n(q) = \Re \left[ U \mathbb{1} - \frac{U^2 \hat{\chi}_0(q)}{1 + U \hat{\chi}_0(q) + i\eta \mathbb{1}} + \frac{U^3 \hat{\chi}_0^2(q)}{1 - U^2 \hat{\chi}_0^2(q) + i\eta \mathbb{1}} \right]$$

(16)

In all calculations leading to the phase diagram in Fig. 1 (d) in the manuscript, we set the broadening parameter to $\eta = 8 \text{meV}$. This effectively avoids a divergence of the interactions in the AAA regions in the middle layer. We checked that different choices of the broadening parameter $\eta$ do not change our results qualitatively. An analysis of the pairing vertex $\hat{\Gamma}_2^n$ as a function of broadening $\eta$ and Hubbard-$U$ is shown in Fig. 14.

Bogoliubov de-Gennes equations To analyze the superconducting properties of the system, we decouple the effective pairing vertex $\hat{\Gamma}_2$ in mean-field approximation, allowing only for symmetric spin-singlet bond order parameters $\Delta_{ij} = \Delta_{ji}$ due to the proximity to AFM tendencies in between the integer fillings

$$\hat{\Delta}(k) = \Delta_{nm}(k) = -\frac{1}{2N} \sum_{k',\sigma} \hat{\Gamma}_{2,nm}(q = k - k', q_0 = 0) \times \sigma \langle c_{n\sigma}(k')c_{m\bar{\sigma}(-k')} \rangle_{\text{MF}},$$

(17)
The resulting mean-field Hamiltonian can be rewritten in the 2N-dimensional Nambu spinor basis \( \psi_k = (c_k^\uparrow, c_k^\downarrow)^T \)

\[
H_{MF} = \sum_k \psi_k^\dagger \begin{pmatrix} \hat{H}(k) & \hat{\Delta}(k) \\ \hat{\Delta}^\dagger(k) & -\hat{H}(-k) \end{pmatrix} \psi_k + \text{const.}
\]  

(18)

This Bogoliubov de-Gennes (BdG) bilinear form is diagonalized by a block-structured unitary transform \( \hat{U}_k \)

\[
H_{MF} = \sum_k \psi_k^\dagger \begin{pmatrix} \hat{H}(k) & \hat{\Delta}(k) \\ \hat{\Delta}^\dagger(k) & -\hat{H}(-k) \end{pmatrix} \psi_k = \sum_k \left( \hat{U}_k \psi_k \right)^\dagger \begin{pmatrix} \hat{E}_k & 0 \\ 0 & -\hat{E}_k \end{pmatrix} \left( \hat{U}_k \psi_k \right)
\]

\[
\hat{U}_k = \begin{pmatrix} \hat{u}_k & \hat{v}_k \\ \hat{v}_k^* & \hat{u}_k^* \end{pmatrix}
\]  

(19)

The matrices \( \hat{u}_k (\hat{v}_k) \) are \( N \times N \) matrices which describe the particle (hole) amplitudes of the Bogoliubov quasiparticles \( \gamma_k \) with energies \( \pm \hat{E}_k \). To this end, \( \hat{E}_k \) denotes the corresponding diagonal matrix containing the particle-hole symmetric energy eigenvalues. The Bogoliubov quasiparticles are defined as

\[
\begin{pmatrix} \gamma_{k,\uparrow} \\ \gamma_{-k,\downarrow}^\dagger \end{pmatrix} = \begin{pmatrix} \hat{u}_k & \hat{v}_k \\ -\hat{v}_k^* & \hat{u}_k^* \end{pmatrix} \begin{pmatrix} c_{k,\uparrow} \\ c_{-k,\downarrow}^\dagger \end{pmatrix}.
\]

(20)

Together with Eq. (17), this yields a set of equations that needs to be solved self-consistently

\[
H_{MF} = \sum_k \psi_k^\dagger \begin{pmatrix} \hat{H}(k) & \hat{\Delta}(k) \\ \hat{\Delta}^\dagger(k) & -\hat{H}(-k) \end{pmatrix} \psi_k + \text{const.}
\]

\[
\gamma_{i,j} = 10^{\gamma_{i,j}} \quad \gamma_{i,j} = 10^{\gamma_{i,j}}
\]

(21)

\[
\langle c_{i\uparrow}(k)c_{j\downarrow}(-k) - c_{i\downarrow}(k)c_{j\uparrow}(-k) \rangle_{MF} = \sum_n \left( u_{n,i,k}v_{n,j,k}^* + v_{n,j,k}u_{n,i,k}^* \right) \tanh \left( \frac{E_{n,k}}{2T} \right)
\]

Here, \( \sum_n \) denotes a sum over the positive quasiparticle energies \( E_{n,k} > 0 \). To solve these non-linear equations self-consistently, we start with an initial guess respecting the symmetry of the gap parameter \( \Delta_{ij}^{\text{init}} \) and iterate until convergence is achieved using a linear mixing \( \Delta^{n+1} = (1 - \alpha)\Delta^n - \alpha\Delta^{n-1} \) scheme to avoid bipartite solutions in the fixed point iteration. In all of our calculations, we set the relative error for convergence to \( \epsilon = 10^{-6} \) and set the mixing parameter to \( \alpha = 0.2 \).

In the manuscript, we only account for the gap parameter \( \Delta \) at the \( \Gamma \)-point of the Brillouin zone. This approximation may not change the underlying physics drastically as our microscopic interaction \( \Gamma_2 \) in the static, long-wavelength limit \( (q \to 0) \) carries no momentum dependence and the size of the Brillouin zone of MATTG is drastically reduced due to the large real-space unit cell. Hence, the \( \mathbf{k} \)-sum in Eq. (21) does not couple order parameters at different momentum points, but rather corresponds to a momentum average. Still, we checked that our results do not change qualitatively when taking a dense mesh with up to 24 \( \mathbf{k} \)-points in the BZ into account.

The full non-linear gap equation Eq. (21) is equivalent to minimizing the free energy of the system with respect to the pairing order parameter \( \partial F/\partial \Delta_{ij} = 0 \). To make sure that the superconducting state globally minimizes the free energy (and does not converge into possible local minima) we track the free energy for different initial guesses \( \Delta_{ij}^{\text{init}} \).

The full expression of the free energy reads

\[
F = E - TS = \sum_{n,k} E_{n,k}n_F(E_{n,k}) - \sum_{n,k} \{ E_{n,k} - \epsilon_{n,k} \} - \sum_{k,ij} \frac{\Delta_{ij}(k)}{\Gamma_{2,ij}} - TS,
\]

(22)

where \( \epsilon_{n,k} \) are the energies of the normal state Hamiltonian \( H_0 \) and the sum over \( n \) runs over all positive quasiparticle energy states \( E_{n,k} > 0 \). The entropy term of the free energy can be calculated using

\[
S = -2 \sum_{k,n} \left\{ n_F[E_{n,k}] \log (n_F[E_{n,k}]) + (1 - n_F[E_{n,k}]) \log (1 - n_F[E_{n,k}]) \right\}.
\]

(23)

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Supplementary Figure 15. Atomistic nematic gap parameter $\hat{\Delta}$ in MATTG. (a) Amplitude and phase distribution of the superconducting order parameter $\hat{\Delta}$ in the outer layer of MATTG projected onto the two local $d$-wave components for $\nu = -2.5$, $T = 0.2$ K. The order parameter is real-valued such that the phase consists of domains separated by $\pi$ (blue and yellow color coding). The amplitude is strongly enhanced in the AAA regions and the order parameter breaks the original $C_3$ symmetry of the normal-state Hamiltonian. (b) The amplitude in the middle layer is enhanced by a factor of $\sim 10$. Compared to the upper (lower) layer the phase is shifted by $\pi$ as a result of the original interlayer repulsion in the tight-binding Hamiltonian.

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