Superconductivity in twisted double bilayer graphene stabilized by WSe$_2$

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Identifying the essential components of superconductivity in graphene-based systems remains a critical problem in two-dimensional materials research. This field is connected to the mysteries that underpin investigations of unconventional superconductivity in condensed-matter physics. Superconductivity has been observed in magic-angle twisted stacks of monolayer graphene but conspicuously not in twisted stacks of bilayer graphene, although both systems host topological flat bands and symmetry-broken states. Here we report the discovery of superconductivity in twisted double bilayer graphene (TDBG) in proximity to WSe$_2$. Samples with twist angles 1.24° and 1.37° superconduct in small pockets of the gate-tuned phase diagram within the valence and conduction band, respectively. Superconductivity emerges from unpolarized phases near van Hove singularities and next to regions with broken isospin symmetry. Our results show the correlation between a high density of states and the emergence of superconductivity in TDBG while revealing a possible role for isospin fluctuations in the pairing.

Superconductivity in graphene heterostructures is consistently correlated with flat electronic bands, whether induced by a moiré potential, as in stacks of graphene monolayers with alternating twists $^{1-5}$ such as magic-angle twisted bilayer graphene (TBG), or by a gate voltage, as in rhombohedral trilayer graphene $^6$ or Bernal bilayer graphene (BBG)$^7$. Beyond the connection between flat bands and superconductivity $^8$, little is agreed upon in terms of the pairing mechanism or symmetry in each system or about the characteristics of the band structure that are crucial to the emergence of superconductivity $^9$. Introducing a sheet of tungsten diselenide (WSe$_2$) next to graphene has been reported to stabilize superconductivity in both TBG and BBG $^9$, thus enhancing the critical temperatures and fields and extending the parameter range over which superconductivity emerges. However, the mechanism behind this effect remains a subject of debate.

There is a puzzling contrast between the consistent observation of superconductivity in TBG $^{10,20}$ but not, so far, in twisted double bilayer graphene (TDBG), which has two twisted Bernal bilayers $^{21-26}$. The two systems have similar moiré bandwidths $^{27}$, similar signatures of strong interactions including correlated insulators and broken-symmetry states $^{28,29}$, and both have moiré bands that are understood to be topological, with Chern number $|C|=1$ for the first moiré band of TBG and $|C|=2$ for TDBG $^{30,31}$. One difference is the lack of $C_2$ symmetry (twofold rotation) in TDBG, whereas $C_2$ symmetry is preserved in the family of alternating-twist graphene monolayer stacks. Nevertheless, superconductivity has been predicted in TDBG by several theoretical works $^{32-34}$.

The high tunability of TDBG would make it a powerful experimental probe of the mechanisms underlying graphene superconductivity, as it has top and back gates to control the band structure through the vertical displacement field $D$ independently of the band filling $\nu$. Moreover, the flexibility of the TDBG platform leaves the apparent absence of superconductivity anywhere in its gate-tuned phase diagram particularly surprising and has led to speculation about a possible role of $C_2$ symmetry in stabilizing the superconducting state in moiré systems $^{35}$. After initial reports of zero-resistance states in the TDBG conduction band $^{21,22}$, it is...
now believed that the low resistance observed in those experiments reflected reduced scattering due to broken spin or valley symmetries\(^\text{14}\).

Here, we show that TDBG bounded on one side by few-layer WSe\(_2\) (Fig. 1a) does exhibit superconductivity, but the parameter range over which it is observed is qualitatively different from that in TBG. We report data for samples with twist angles 1.37° and 1.24°, referred to hereafter as D1 and D2 (see Supplementary Sections A–C for device details and characterization of the twist angle). In D1, superconductivity is observed in a small gate-defined pocket close to filling \(\nu \approx 1\) in the conduction band, near the so-called halo regions where broken-symmetry states are consistently reported\(^\text{18,35}\). In D2, the superconducting pocket is in the valence band near \(\nu \approx -3\), where no correlation-induced modifications have been reported. In contrast to several other van der Waals structures, superconductivity in TDBG emerges from isospin (spin and/or valley) unpolarized bands for both twist angles, but the superconducting pockets are localized to gate voltages that place the Fermi energy next to a van Hove singularity and close to phases with a tendency toward isospin polarization.

**Transport characterization of superconductivity**

Figure 1a illustrates the sample structure, including the TDBG and WSe\(_2\) layers with dual electrostatic gates. The devices were patterned into Hall bars, and lock-in measurements of the longitudinal (\(R_{xx} \equiv dV_{xx}/dI\)) and transverse (\(R_{xy} \equiv dV_{xy}/dI\)) differential resistance were made in a dilution refrigerator, using top gate \(V_{tg}\) and back gate \(V_{bg}\) voltages to set \(vD\). Except where noted, measurements were made in a mixing chamber at a temperature below 10 mK (Methods). The resulting resistivity maps are shown in Fig. 1b,c for D1 and D2, respectively, and are broadly consistent with published data for TDBG samples with positive \(\nu\) and antisymmetrized Hall filling \(\nu\) \((f)\) at \(B = \pm 0.8\) T along the white dashed line in b. Superconductivity (SC, shaded blue) occurs immediately next to van Hove singularities, where \(V_{tg}\) diverges and changes sign (vHS, shaded orange).

**Comparison of \(\nu\) to \(d\) conditions and obtained along the white dashed line in e.**
gate-voltage locations of the low-resistance features are adjacent to van Hove singularities that accompany the transition between opposite carrier types. These van Hove singularities are identified by locations where the Hall coefficient \( R_H = R_{11}/B \) goes through zero and the Hall density \( n_H = 1/(eR_H) \), where \( e \) is the electron charge, diverges and changes sign. For easier interpretation, we report here the Hall filling \( \nu_H = 4n_H/n_c \), which is the Hall density normalized by the measured carrier density at a nanoampere-scale critical current with a factor of four to account for spin and valley degeneracy (Fig. 1f; see Supplementary Section D for \( \nu_H \) over the full range of gate voltages).

Figure 2 illustrates the fragility of the low-resistance state to the direct current \( I_{dc} \), and to \( B_\perp \), confirming the presence of superconductivity (Supplementary Section E). We focus primarily on the stronger state in D1 (Fig. 2a). Figure 2b,c contrasts the \( I_{dc} \) breakdown of the \( R_{11} = 0 \) state within the superconducting pocket. Compare the sharp peaks in \( dV/dI \) at a nanoampere-scale critical current with the nearly flat \( dV/dI \) at nearby locations in \( \nu, D \). The temperature dependence of the \( \nu_H \) traces, obtained by integrating \( dV/dI \) at the red marker in Fig. 1f, shows the classic evolution of two-dimensional superconductors from step-like transitions at low temperatures to ohmic dependence above 100 mK (Fig. 2d). At this specific \( \nu, D \) setting, a Berezinski–Kosterlitz–Thouless (BKT) analysis of the evolution of the \( \nu_H \) power law near the critical current indicates a BKT transition temperature of \( 64 \pm 5 \) mK, where \( \nu_H \) is obtained at the upper inset.

The clearest demonstration of superconductivity is found in the \( B_\perp \) dependence of \( dV/dI \) for D1 (Fig. 2e). The repeated collapse and revival of the critical current with \( B_\perp \), also referred to as a Fraunhofer pattern, results from quantum interference, like that for a superconducting quantum interference device, of transport through Josephson junctions and is commonly used as a confirmation of superconductivity in TDBG experiments. The characteristic field scale of the fluctuations, \( \Delta B \approx 2-3 \) mT, is consistent with a domain size \( \sqrt{\phi_0/\Delta B} = 0.7 \mu m \) that is close to the Hall-bar width of 1 \( \mu m \), where \( \Phi_0 = h/(2e) \) is the superconducting flux quantum. Equivalent data for D2 (Fig. 2f) lacks the Fraunhofer modulations of D1, reminiscent of other low-\( T_c \) graphene superconductors. The absence of clear signatures of phase coherence leaves open the possibility of alternative explanations for the low-resistance state in D2, but its sensitivity to \( B_\perp < 5 \) mT is hard to reconcile with the symmetry-breaking mechanism proposed in ref. 24.

Moreover, the abrupt rise in \( dV/dI \) due to only a few nanoamperes of direct current is difficult to explain quantitatively by Joule heating. Figure 3 shows how the metrics of superconductivity vary with the band filling for D1 (see Supplementary Sections F–H for additional details and similar characterizations in D2). At each value of \( \nu \), the collapse of superconductivity with \( B_\perp \) (Fig. 3a) can be used to determine a critical out-of-plane magnetic field \( B_{\perp,\text{c}} \), defined as the field where the resistance rises to one half of its normal-state value. Measuring \( B_{\perp,\text{c}} \) as a function of temperature enables the extrapolation to a value \( B_{\perp,\text{c}}^0 \) at zero temperature (Fig. 3d). To the extent that Bardeen–Cooper–Schrieffer theory applies in this system, a Ginzburg–Landau coherence length \( \xi_G = \sqrt{\phi_0/(2\pi B_{\perp,\text{c}}^0)} \) may be determined. \( \xi_G \) is around 100 nm across most of the superconducting dome, comparable to the mean free path \( l_m \approx 200 \) nm, which can be estimated from the onset of Shubnikov–de Haas oscillations around 400 mT (Supplementary Section K).

Consistent with the two-dimensional nature of the superconductivity in TDBG, the in-plane critical fields \( B_{\parallel,\text{c}} \) are an order of magnitude greater than the out-of-plane fields \( B_{\perp,\text{c}} \) for both devices (Supplementary Section H). Conventional superconductivity, with spin-singlet Cooper pairs, is destroyed when the Zeeman energy induced by the in-plane magnetic field exceeds the superconducting gap, leading to the Pauli (Chandrasekhar–Clogston) limit field, \( B_p = 1.76k_B T_c^2 g^2 B_{\parallel,\text{c}}^0/\mu_B \) above which superconductivity is expected to vanish. Here, \( g \) is the Landé factor, \( \mu_B \) is the Bohr magneton, \( k_B \) is the Boltzmann constant and \( T_c^2 \) is the critical temperature at \( B = 0 \), as mapped out in Fig. 3b.

Figure 3c shows the collapse of superconductivity with \( B_{\perp,\text{c}} \). For quantitative analysis, \( B_{\perp,\text{c}} \) is defined by where \( R_{11} \) reaches a threshold of half of its normal-state resistance. The temperature dependence of \( B_{\perp,\text{c}} \) can be fitted to the phenomenological relation \( T/T_c^2 = 1 - (B_{\perp,\text{c}}/B_{\parallel,\text{c}}^0)^2 \) commonly used for Pauli-limited superconductivity (see, for example, Fig. 3e), and the resulting \( B_{\perp,\text{c}}^0 \) compared to the value of \( B_p \) assuming \( g = 2 \). The ratio \( \text{PVR} \equiv B_{\perp,\text{c}}^0/B_p \) is between 2 and 3 across most of the dome (Fig. 3c) for D1 but is less than 1 for D2 at optimal doping (Supplementary Section H). Recent studies of superconductivity in the family of magic-angle twisted graphene monolayers have identified a similar resilience against pair-breaking by an in-plane magnetic field, which depends strongly on the number of layers.
Superconductivity and broken-symmetry phases

The conditions under which superconductivity appears in TDBG offer an insightful comparison with other graphene systems, with or without a superlattice. An apparently unifying feature of graphene-based superconductivity in all known systems is the proximity to symmetry-broken phases. In most systems, for example, twisted graphene stacks\(^7\) and the SC2 phase of rhombohedral trilayer graphene\(^8\), superconductivity emerges directly from a phase in which two out of the four isospin components are occupied, as determined by magnetoresistance measurements in the normal phase, once superconductivity has been suppressed\(^9\).\(^10\).

In TDBG, we find instead that superconductivity emerges out of isospin-unpolarized bands, adjacent to regions with a tendency toward isospin polarization. Figure 4 highlights Hall measurements in TDBG that illustrate this correlation. The Hall filling \(\nu_H\) departs from the total filling \(\nu\) by ±1, ±2 or ±3 when isospin-symmetry breaking leaves the corresponding number of bands completely filled (therefore, not contributing to \(\nu_H\)), so the subtracted Hall filling \(\nu_H - \nu\) probes band polarization\(^9\).\(^10\).

Figure 4a,b shows \(\nu_H - \nu\) for the D1 conduction band at \(B_z = \pm 0.8\) T, with the dashed blue perimeter in Fig. 4a outlining the region where superconductivity would appear at \(B_z = 0\). As seen by the white colour in Fig. 4a and shown quantitatively in Fig. 4b, this region is isospin unpolarized, with \(\nu_H - \nu = 0\). However, it is immediately adjacent to a degeneracy-2 isospin-ferromagnetic phase (IF2; \(\nu_H - \nu = -2\)) at slightly higher doping, separated by a van Hove singularity where the Hall filling and the density of states diverge\(^10\).

To infer the broken symmetry of the IF2 state in D1, we studied the effect of \(B_z\) on the resistance of the correlated insulator at half-filling. If the \(\nu = 2\) insulator separates bands with different spin polarizations, its resistance would be expected to rise with \(B_z\). Figure 4c compares \(R_n\) at \(B_z = 0\) and \(B_z = 5\) T for the line cut in Fig. 4b. In contrast to many previous reports\(^9\),\(^10\),\(^19\)\(^-\)\(^23\), the \(R_n\) peak at \(\nu = 2\) in D1 is not affected by \(B_z\), indicating that the IF2 halo is not spin-polarized in this sample (Supplementary Section I).

Figure 4d,e shows \(\nu_H - \nu\) for the D2 valence band, highlighting the effect of \(B_z\) on the isospin order in this sample. At \(B_z = 0\), \(\nu_H - \nu\) transitions directly between 0 and 4 via a van Hove singularity for \(D < 0\) (Fig. 4d). At \(B_z = 3\) T, lobes of an IF2 isospin ferromagnet appear around \(D = \pm 0.1\) V nm\(^{-1}\), with hints of the state already visible at \(B_z = 0\) for \(D = \pm 0.1\) V nm\(^{-1}\). The emergence of the IF2 phase with \(B_z\) is mapped out in Fig. 4c, for line cuts at \(D = \pm 0.08\) and \(\pm 0.08\) V nm\(^{-1}\) (Supplementary Section J). No similar behaviour was seen in the D1 valence band (Supplementary Section D).

Our measurements collectively demonstrate that superconductivity in TDBG appears in isospin-unpolarized phases, near transitions to a polarized phase. For D1, the nearby IF2 phase is reached by a small change in gate voltage, whereas in D2, the addition of Zeeman energy via \(B_z\) achieves the effect. That IF2 phases and superconducting pockets are both localized to small fractions of the \((v, D)\) plane and that the two are consistently next to each other provides compelling evidence of a correlation between them. Based on this correlation, it is plausible that a particular feature of the electronic state—such as a diverging density of states or isospin fluctuations\(^15\)\(^-\)\(^40\)—may give rise to competing tendencies toward singlet Cooper pairing and isospin ferromagnetism, with the dominant phase determined by the band filling and displacement field.

The absence of superconductivity near the halos in D2 could then be explained by a stronger tendency to isospin polarization, consistent with the extension of the IF2 region down to \(\nu = 0.5\) in D2 but only to \(\nu = 1.5\) in D1 (Supplementary Section D).

That superconductivity occurs only for a single sign of the displacement field in the conduction band but the opposite in the valence band is evidence that WSe$_2$ plays a critical role in the stabilization of superconductivity. The sign of \(D\) in both cases allows the respective band to be pulled into proximity with the WSe$_2$ layer. However, the precise nature of this role remains unclear. An intriguing proposal in ref. \(^41\) suggests that virtual tunnelling into WSe$_2$ reduces the native Coulomb repulsion between electrons in a Cooper pair, thereby enabling other pairing mechanisms to stabilize superconductivity. Alternatively, it has been suggested that an Ising spin–orbit interaction induced by proximity to WSe$_2$ may stabilize zero-field superconductivity in BBG\(^16\). Unfortunately, our attempts to quantify spin–orbit coupling directly in D1 and D2 were inconclusive. Sample disorder prevented us from resolving possible spin-splitting in the Fermi surface, the technique that has been employed in other systems such as BBG on WSe$_2$ (refs. \(^19\),\(^42\); Supplementary Section K).
At the same time, several features of our data are consistent with the presence of a spin–orbit interaction induced by WSe$_2$. First, the violation of the Pauli limit seen in D1 is consistent with Cooper pairing in a superconducting state modified by spin–orbit coupling\cite{16,15}. Further, the Ising spin–orbit interaction couples the spin and valley degrees of freedom and may tip the balance between nearly degenerate symmetry-breaking orders\cite{13,14}, favouring, for example, a spin-valley-locked state\cite{15} that would be consistent with the apparent lack of spin polarization in the vicinity of $\nu=2$ in D1.

In D2, the need for higher $B$, to form the Ising phase at $D=-0.08$ V nm$^{-1}$, compared to $D=+0.08$ V nm$^{-1}$, can also be interpreted as a weakening of spin polarization when the valence band is pulled closer to WSe$_2$.

Our findings highlight the remarkable impact of proximal substrate interfaces and the moiřé potential in establishing new ground states in van der Waals heterostructures. They add to the growing body of evidence in graphitic systems suggesting a correlation—but not overlap—between electronic states with a tendency toward isospin polarization and those conducive to superconductivity. The localization of superconductivity to specific regions of the TDBG phase diagram encourages further investigations, both experimental and theoretical, to isolate the key elements of the electronic structure responsible for this phenomenon. Identifying these factors would have broad implications for our understanding of superconductivity in graphitic van der Waals heterostructures.

### Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41563-023-01653-7.

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Methods
Sample fabrication
Samples of WSe₂/AB–AB stacked TDBG were assembled using the cut-and-stack method. Hexagonal boron nitride (hBN), few-layer tungsten diselenide (WSe₂) and graphene flakes were first mechanically exfoliated onto clean SiO₂ (300 nm)/Si substrates. Bilayer graphene flakes were identified using optical contrast and cut into halves using a sharp ~1-μm-diameter tungsten dissecting needle. A stamp made of poly(bisphenol A carbonate) film laminated over a polydimethylsiloxane hemisphere on a glass slide attached to a micromanipulator was used for the dry transfer process. At a temperature of 90 °C, the top hBN and WSe₂ were picked up. The first half of the bilayer graphene flake was picked up at 50 °C, and the stage was rotated by θ = 1.4°, slightly overshooting to achieve the target angle. The remaining half of the bilayer graphene flake was picked up at the same temperature. Finally, the bottom hBN was picked up at 100 °C. For device D1, the entire stack, hBN/WSe₂/TDBG/hBN, was dropped onto a gold bottom gate pre-patterned on a clean SiO₂/Si substrate. For device D2, a graphite bottom gate was picked up at 100 °C, and the stack, hBN/WSe₂/TDBG/hBN/graphite, was dropped onto a SiO₂/Si substrate with alignment markers (Supplementary Section B). The top gate was first defined on the stack using electron-beam lithography, followed by electron-beam evaporation of Cr/Au (5 nm/50 nm). Edge contacts were made by dry-etching the stack using a CHF₃/O₂ plasma (40 sccm/4 sccm, 4 Pa chamber pressure and 60 W radiofrequency power), followed by evaporation of Cr/Au (8 nm/70 nm). The device was shaped into a Hall bar using a final CHF₃/O₂ plasma etch. A chamber pressure of at least 5 × 10⁻⁷ torr was maintained during the evaporation of chromium and gold at rates of 0.5 and 1.0 Å s⁻¹, respectively.

Transport measurements
The experiment was carried out in a Bluefors LD dilution refrigerator with a base temperature below 10 mK, as measured by the factory-supplied RuO₂ sensor attached to the mixing chamber plate. Throughout this paper, the temperatures cited are those of the mixing chamber. The effective electron temperature followed the mixing chamber temperature, at least down to 50 mK (Supplementary Section L). The base electron temperature was below 30 mK. The cryostat was equipped with a three-axis vector magnet.

The transport measurements were carried out using a lock-in amplifier with an a.c. bias, and the voltage was measured, as indicated in Fig. 1. Measurements in the superconducting state used an a.c. bias of 0.1 nA, whereas measurements in a finite magnetic field used an a.c. bias of 5 nA, unless otherwise stated. All measurements were performed at the base temperature. The tilt angle between the normal of the sample plane and the out-of-plane axis of the vector magnetic was kept <1°.

Top and bottom gate voltages were used to independently tune the carrier density n and electric displacement field D according to the equations: n = (C bg V bg + C w V w)/e and D = (C bg V bg − C w V w)/2ε0, where C bg and C w are, respectively, the bottom and top gate capacitances per unit area, e is the elementary charge and ε0 is the vacuum permittivity. C bg and C w were found using the slope of the Hall density, n H = 1/eR xy, where R xy = (R x [B z > 0] − R x [B z < 0])/(2|B|) is the antisymmetrized Hall resistance and e is the electron charge.

The device twist angle θ was estimated from the density corresponding to complete filling of the moiré miniband n(ν = ±ε) = 80θ/√3a₂, where a = 0.246 nm is the lattice constant of graphene. In our devices, ν = ±ε corresponded to n = ±3.6 × 10¹² cm⁻² and n = ±4.4 × 10¹² cm⁻², which translates to twist angles θ = 1.24° and θ = 1.37° for devices D1 and D2, respectively.

Critical temperature and magnetic fields
The critical temperature and magnetic fields were determined based on a percentage resistance threshold, α × 100%. The portion of R xx(z) (where z represents the temperature T or applied magnetic field B) when the superconducting sample returns to its normal state was fitted to r(z) = Az + C. The critical z value at which R xx(z) intersects ar(z) was then used to obtain the critical temperature and magnetic fields. A 50% threshold was used, unless otherwise specified.

Data availability
Source data are provided with this paper. All other data are available from the corresponding author upon reasonable request.

Code availability
Codes used for data analysis in this study are also available from the corresponding author upon reasonable request.

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Author contributions
R.S. fabricated the devices, with help from M.K. R.S. performed the measurements. R.S., M.K. and J.F. interpreted the data and wrote the paper. J.F. supervised the experiment. K.W. and T.T. provided the hBN crystals.

Competing interests
The authors declare no competing interests.

Additional information
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