

Supplementary Information for
Spontaneous Excitonic Mass Generation in Graphene Monolayers

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I. Isolation and identification of graphene monolayers

Graphene flakes were mechanically exfoliated onto clean $\text{SiO}_2/\text{Si}^{++}$ substrates using Scotch tape. Graphene monolayers were identified by optical contrast (Fig. S1a) and Raman spectroscopy (Fig. S1b), showing the characteristic features of monolayer graphene, a G peak at 1580 cm^{-1} and a 2D peak at 2675 cm^{-1} , with sharp full width at half maximum (FWHM) of 16 and 26 cm^{-1} ,

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6 respectively. Separately, for each graphene device, atomic force microscope (AFM) confirms
 7 surface cleanliness and monolayer thickness (~ 0.4 nm), as shown in Figs. S1c and S1d.

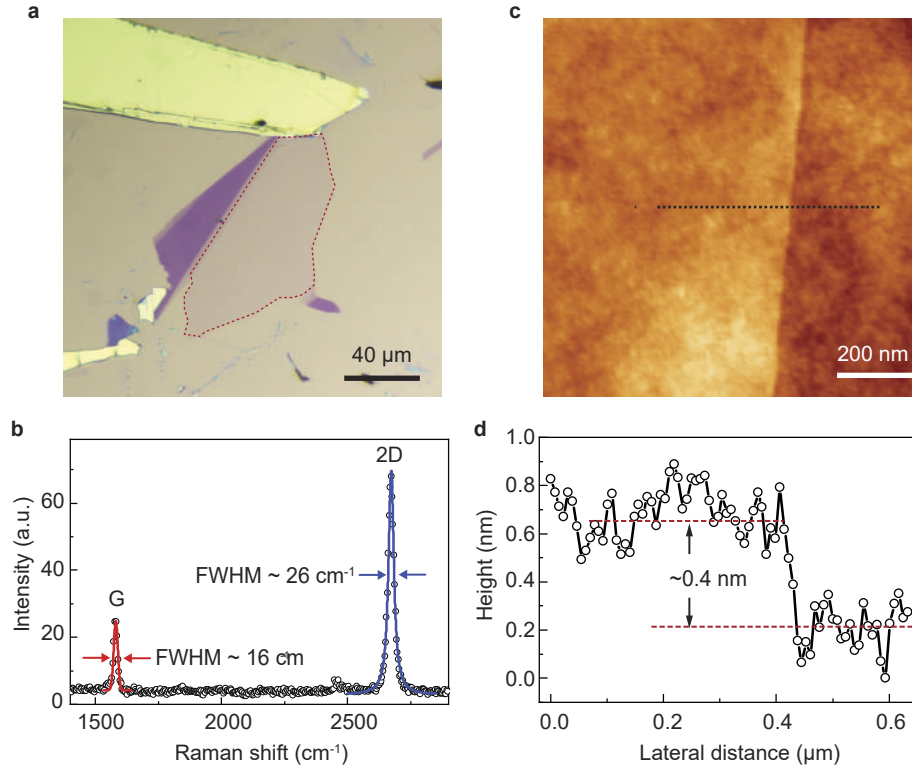


Fig. S1. **Monolayer graphene.** **a**, Optical micrograph of the graphene flake (red outline). **b**, Raman spectrum (532 nm excitation, 0.5 mW, 1800 lines/mm grating), showing the characteristic G and 2D peaks fitted with Lorentzian functions (red: G peak, blue: 2D peak). **c,d**, AFM topography with line profile across the graphene edge.

8 II. Dirac velocity extraction

9 The Dirac velocity is one of the fundamental parameters in graphene, quantifying the kinetic
 10 energy scale. From the linear band dispersion at low energy $E = \hbar v k$, the carrier density of
 11 occupied electron states up to the Fermi level becomes

$$n(E_D) = \frac{(E_F - E_D)^2}{\pi(\hbar v)^2}. \quad (\text{S1})$$

12 In this study, the Dirac velocity has been extracted independently from gate voltage dependent tun-
 13 nelling spectroscopy and LL spectroscopy in magnetic fields. For the gate voltage dependent spec-
 14 troscopy, we have assumed a parallel-plate capacitor model. The charge carrier density induced by
 15 a gate voltage V_G may be estimated by $n = \gamma|V_G - V_0|$, where $\gamma = \epsilon_r \epsilon_0 / t \approx 7.2 \times 10^{10} \text{ cm}^{-2} \text{ V}^{-1}$
 16 is the geometric capacitance per unit area, as determined by the dielectric constant ($\epsilon_r = 3.9$) and

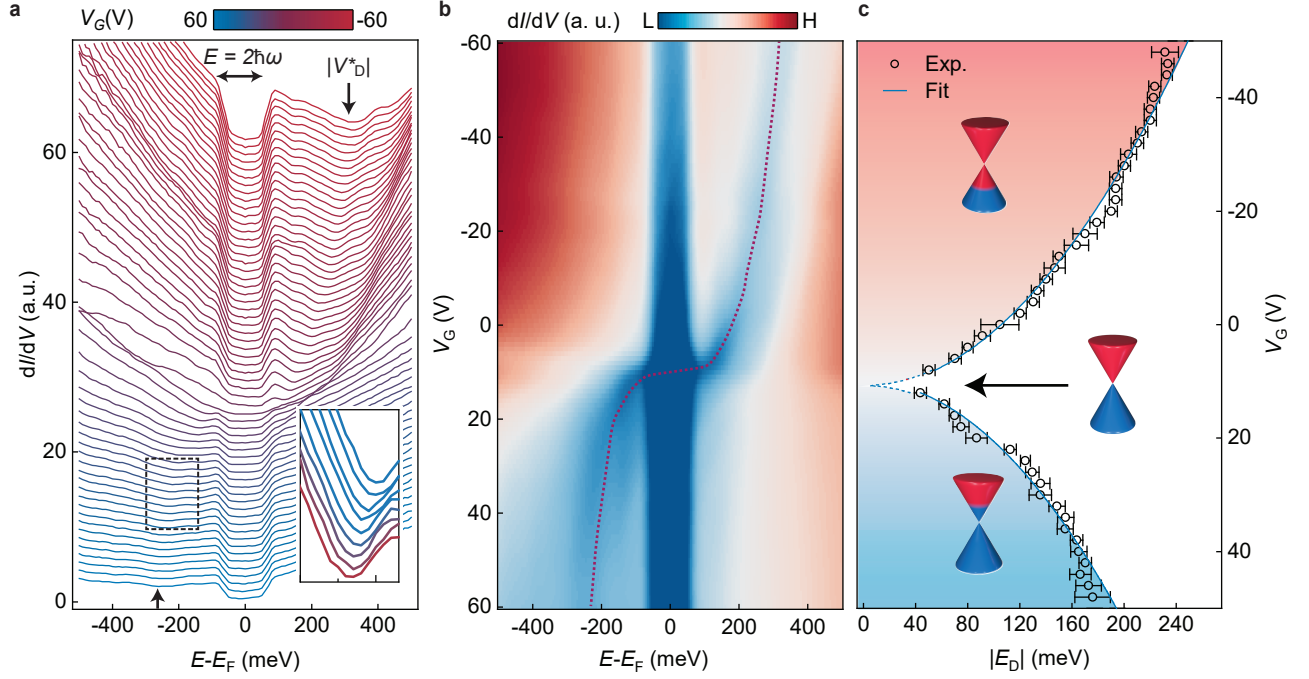


Fig. S2. **Gate-voltage-dependent spectroscopy of graphene.** **a**, dI/dV spectra of pristine graphene measured at gate voltage ranging from -60 V to 60 V, same data as Fig. 1d of the main text. **b**, dI/dV intensity map as a function of energy and gate voltage. **c**, Extracted Dirac point energy as a function of gate voltage. The solid curve is fit to Eq. S2, yielding $V_0 = -10$ V and $v = 1 \times 10^6$ m/s.

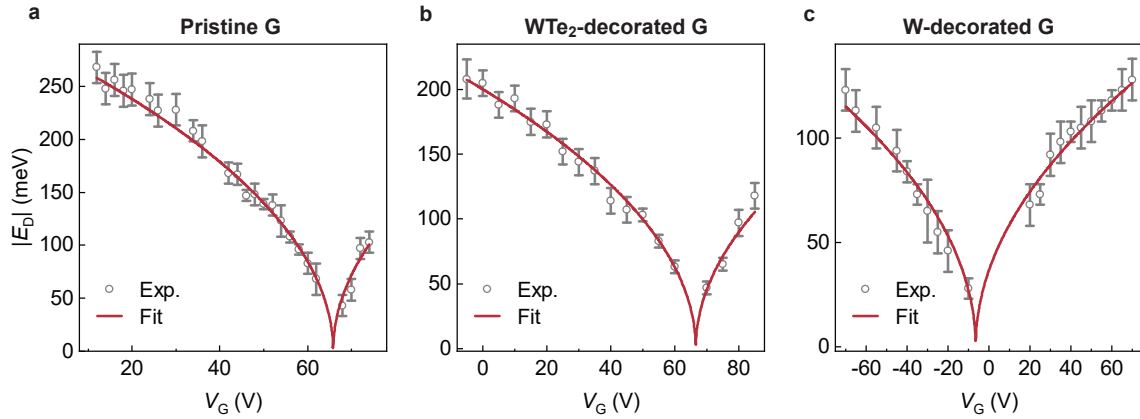


Fig. S3. **Dirac velocity of graphene.** Extracted Dirac point energy E_D as a function of gate voltage V_G for (a) Pristine graphene, (b) WTe_2 -decorated graphene, and (c) W -decorated graphene. Error bars represent uncertainties in the extraction of E_D . Solid blue lines are fits to the Eq. S2, from which we extract both the velocity v and charge neutrality condition $V_G = V_0$.

17 thickness ($t = 300$ nm) of the SiO_2 layer. Here, V_0 denotes the gate voltage at which graphene
 18 reaches charge neutrality ($E_F = E_D$). Therefore, the gate-induced energy shift of the Dirac point
 19 seen in tunneling spectroscopy (Fig. 1d,e main text and Fig. S2), is given by

$$|E_F - E_D| = \hbar v \sqrt{\pi \gamma |V_G - V_0|}, \quad (\text{S2})$$

20 Fits to Eq. S2 yield the Dirac velocity. Fits to spectra in pristine graphene are shown in Fig. S2
 21 and Fig. S3a, and for decorated graphene in Figs. S3b and S3c.

22 Independent estimates of v have been obtained from Landau level spectroscopy. Massless Dirac
 23 fermions in a magnetic fields occupy to a spectrum of Landau levels (LL), described by

$$E_n = E_0 + \text{sgn}(n) v \sqrt{2e\hbar B |n|}, \quad (\text{S3})$$

24 Here, E_0 is the energy of the zeroth LL, which depends on the residual doping in the system and
 25 does not depend on the magnetic field strength. As such, the Dirac velocity v can be extracted
 26 from the slope of the Landau-level energy shift in magnetic field, and the LL index n .

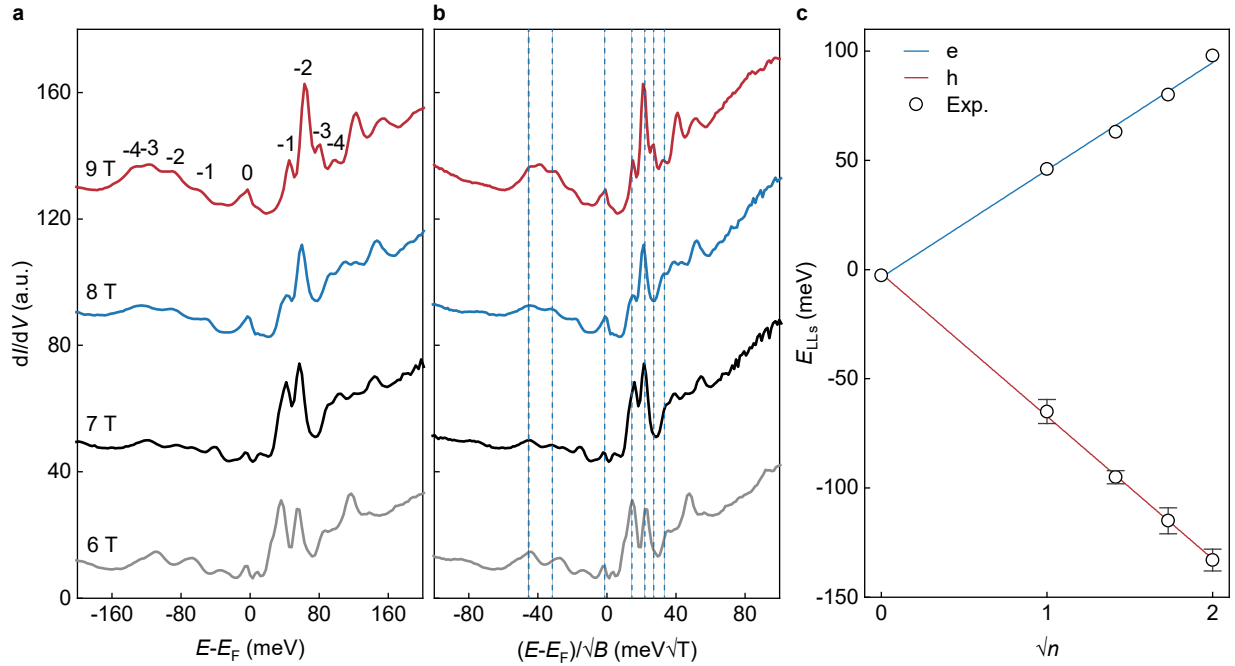


Fig. S4. **Landau level spectroscopy of decorated graphene.** **a**, Tunnelling spectra of WTe₂-decorated graphene acquired under magnetic fields. The zeroth Landau level is identified as the peak near the Fermi level that is independent of magnetic fields. The spectra were measured with a tunnelling setpoint of $V_B = 300$ mV and $I = 1$ nA, using a lock-in modulation of $V_m = 5$ mV at a frequency of 836 Hz. **b**, Corresponding spectra plotted as a function of energy normalized by $\sqrt{B}$. Vertical dashed lines indicate the identified LLs. **c**, Extracted Landau level energies as a function of $\sqrt{|n|}$. Solid lines are fits to Eq. S3, yielding $v \approx 0.46 \times 10^6$ m/s and 0.60×10^6 m/s for the electron and hole bands.

27 Figure S4 shows LL spectroscopy measurements under magnetic fields. LLs identified and
 28 assigned at moderately high magnetic field where they are spectrally distinct (Fig. S4a), and
 29 plotted in (Fig. S4b) as normalized by $\sqrt{B}$ in energy. Fits to Eq. S3 yield Dirac velocities $v \approx$
 30 0.46×10^6 m/s and 0.60×10^6 m/s for electron and hole bands, respectively, for the data shown

31 (WTe₂/graphene), and confirms the significant reduction of the Dirac velocity induced by metallic
 32 decoration. This velocity agrees well with that obtained for the same sample ($v \approx 0.7 \times 10^6$ m/s)
 33 by gate-voltage dependent spectroscopy.

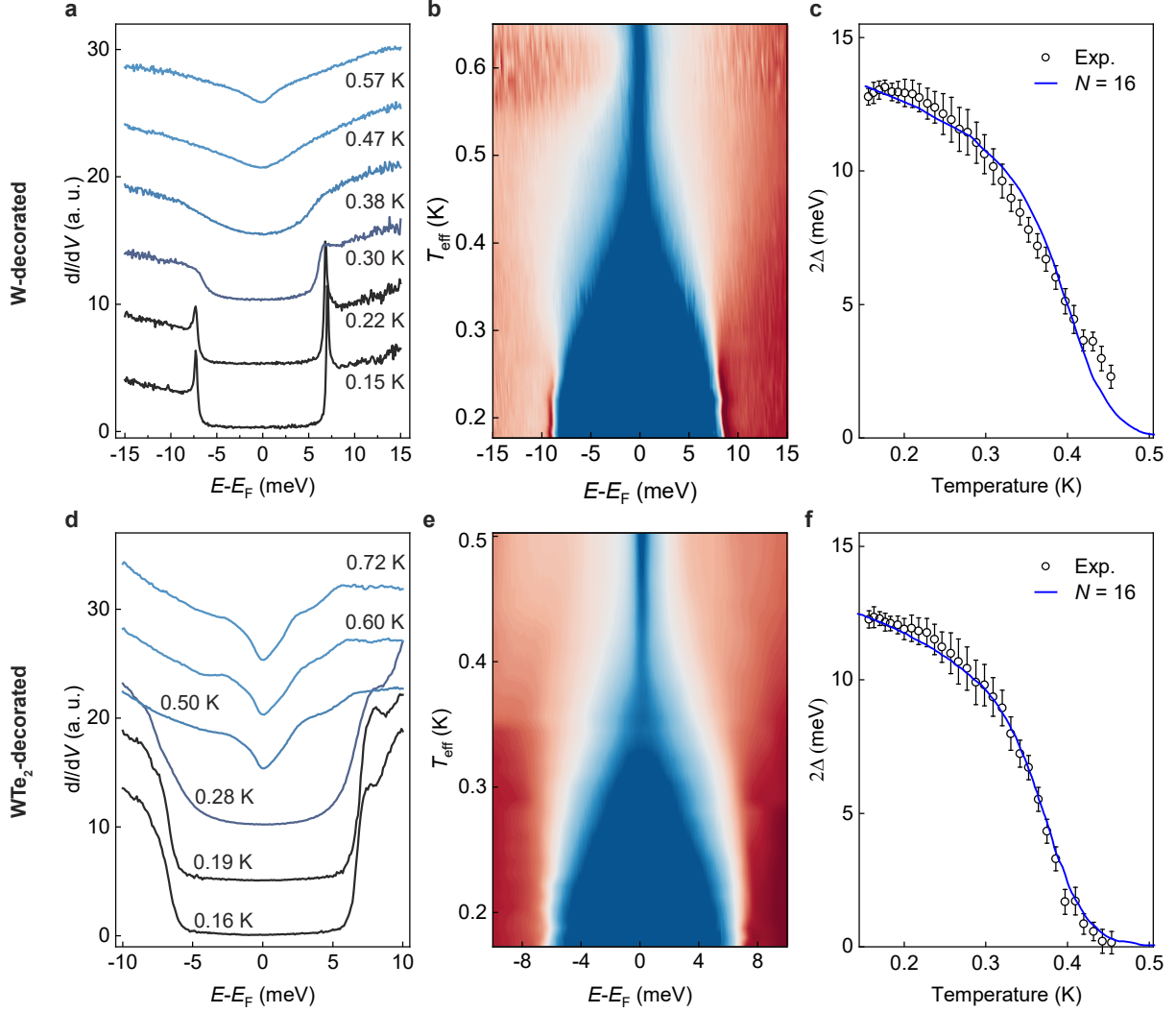


Fig. S5. **Excitonic condensation in decorated graphene monolayers.** **a**, dI/dV spectra of W-decorated graphene acquired at charge neutrality under different temperatures exhibiting a hard spectral gap, centered at E_F , fingerprint of the exciton condensate. **b**, Corresponding dI/dV intensity map as a function of energy and temperature, from which the spectra in **a** were extracted. **c**, Gap size as a function of temperature extracted from the data shown in **(b)**. The blue line is a fit using the finite-size 2D XY model. **d-f**, Corresponding data and fit for WTe₂-decorated graphene.

34 III. Additional data of the temperature dependence of spectral gap

35 Additional data sets of excitonic condensation in decorated graphene are shown in Fig. S5,
 36 comparing WTe₂ and W. In both systems, we observe consistent behavior in gap size ($2\Delta = 12 \sim$

14 meV) and critical temperature $T_c = 0.4 \sim 0.5$ K, with a gap ratio $\Delta/k_B T_c \geq 100$.

IV. Additional data showing the magnetic field anisotropy of the gap suppression

Figure S6 plots the magnetic-field response of the insulating state for both out-of-plane and in-plane field orientations for both the W/graphene and WTe₂/graphene systems. While the gap magnitude is suppressed in both out-of-plane and in-plane magnetic fields, it closes substantially more slowly in in-plane fields, and remains finite up to the maximum field that can be applied in our STM system ($B = 4$ T).

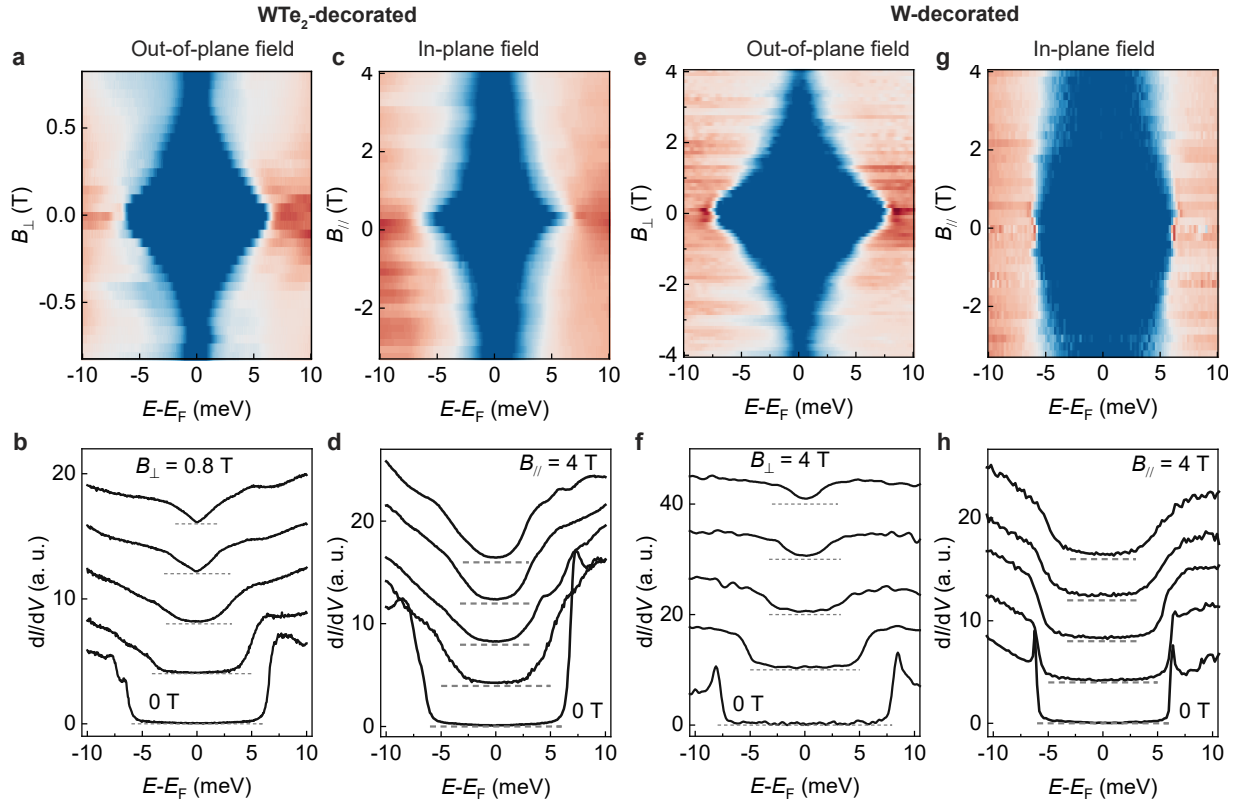


Fig. S6. **Magnetic field anisotropy of the insulating state.** **a-d**, dI/dV intensity map and representative spectra of WTe₂-decorated graphene measured under different out-of-plane and in-plane magnetic fields. **e-h**, Corresponding data for W-decorated graphene. The data for out-of-plane fields (**a** and **b**) are the same as those shown in Fig. 3c and Fig. 3d of the main text.

V. Additional data on the doping dependence of the gap

Figure S7 plots additional data of the doping dependence of the spectral gap in decorated graphene, further confirming the strong susceptibility to charge doping. In both WTe₂- and W-decorated graphene, the spectral gap emerges at charge neutrality despite the distinct charge-

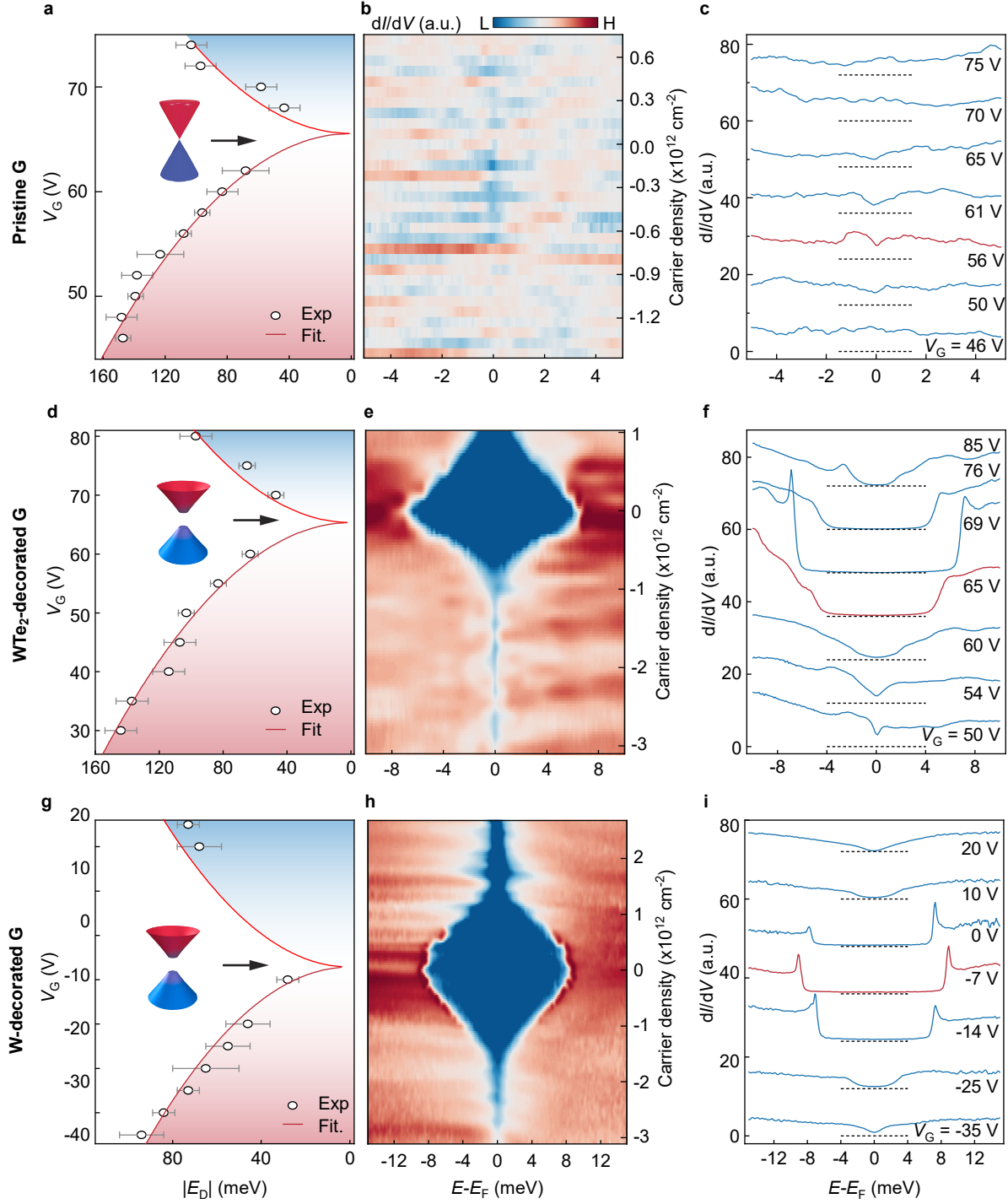


Fig. S7. **Charge doping dependence of the excitonic insulating state in WTe_2 -decorated graphene monolayers.** **a**, Dirac point energy (E_D) as a function of gate-voltage. **b**, dI/dV intensity maps as functions of gate voltage and energy. **c**, dI/dV spectra at different charge doping densities extracted from the corresponding maps in **b**. **d-i**, Corresponding data for WTe_2 -decorated (**d-f**) and W-decorated (**g-i**) samples, respectively, showing the gap opening at the charge neutrality for decorated samples.

transfer effects associated with the two types of metal islands. It is worth noting that, for both pristine and WTe_2 -decorated graphene, the charge-neutrality points are located at $V_0 > 60$ V, indicating that graphene is heavily *p*-doped. The difference of the charge-neutrality-point in gate voltage may suggest weak coupling and limited charge transfer between graphene and the WTe_2 islands. In contrast, after W deposition, the charge-neutrality point shifts to ~ -10 V, indicating substantial charge transfer from the W clusters. This charge transfer compensates the hole doping induced by the underlying substrate, resulting in mild *n*-doping of graphene.

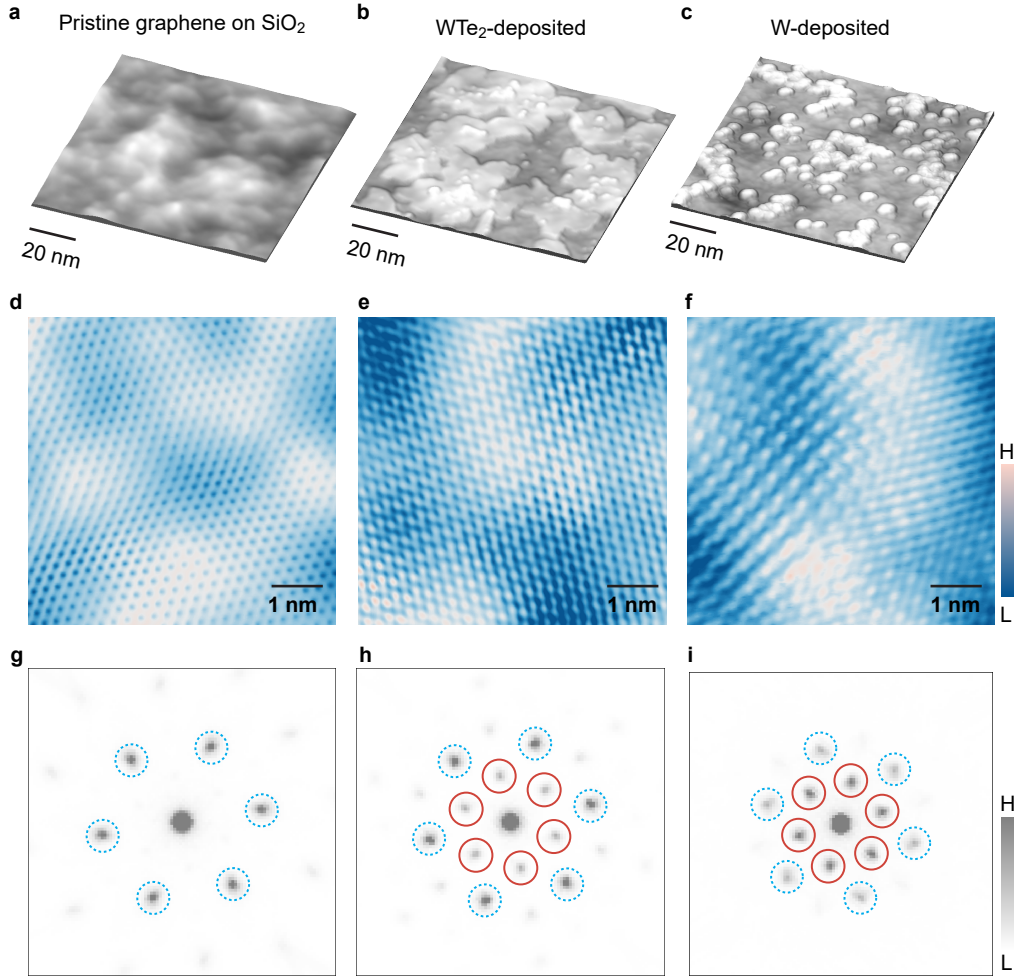


Fig. S8. STM Topography of graphene. **a-f**, STM topography images of graphene acquired before and after metal deposition, including large-scale (**a-c**) and atomic-resolution (**d-f**) images. The intact graphene lattice is preserved following decoration. **g-i**, Corresponding fast Fourier transform (FFT) images of (**d-f**). The intensity peaks corresponding to the graphene lattice and CDW modulations are indicated by dashed blue and solid red circles, respectively. STM images in (**a-c**) were acquired at $V_B = 1.5$ V, $I = 100$ pA, and 20 mV, 1.0 nA for (**d-f**).

VI. Charge density modulation in decorated graphene

Figure S8 shows representative STM topography images of graphene before and after decoration with metallic islands or clusters. Pristine graphene (Fig. S8a) exhibits an atomically clean surface with corrugations (RMS ~ 0.3 nm) originating from the underlying substrate, in agreement with previous reports [1, 2]. In WTe₂-decorated samples, as shown in Fig. S8b, the as-grown WTe₂ islands have lateral dimensions of a few tens of nanometers. In W-decorated samples (Fig. S8c), W clusters with sizes below 10 nm are uniformly distributed across the surface. In regions between the islands or clusters, the graphene lattice remains intact and atomically clean (Figs. S8d-f).

In both decorated samples, additional superstructures are observed beyond the atomic lattice, as further confirmed by the corresponding Fourier components in Figs. S8h and S8i. In contrast, such features are absent in pristine graphene (Fig. S8g). The wavevectors of these superstructures coincided with $\vec{q} = \vec{K}' - \vec{K}$. This modulation is consistent with the charge-density-wave-like spatial signature expected from intervalley coherence, and thus provides real-space evidence supporting the intervalley excitonic pairing mechanism discussed in the main text.

VII. Coulomb gap in decorated graphene

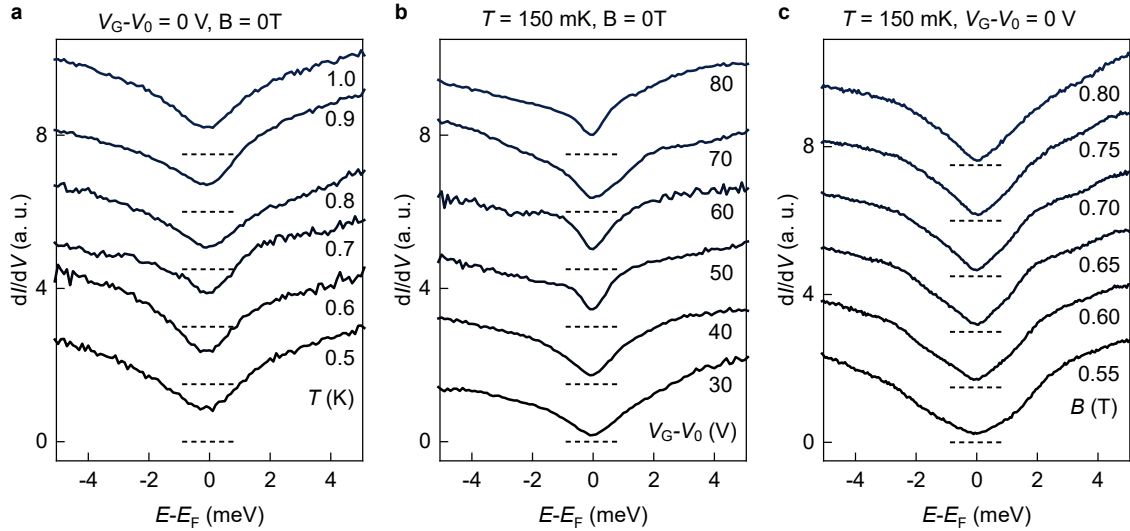


Fig. S9. **Coulomb gap in decorated graphene.** a,b dI/dV spectra of W-decorated graphene measured elevated temperatures (a), in the electron-doped regime (b) and high magnetic fields, showing a linear suppression in the DOS at E_F .

Throughout key datasets in Fig. 2 and Fig. 3 of the main text, we observe a soft pseudogap like suppression in LDOS centred at the Fermi level. In particular, this signature remains as a residual gap after suppression of the large spectral gap with temperature, doping, and magnetic field. Such

signature is known to arise from Coulomb gap and thus is a key manifestation of strong Coulomb interactions in this system. In the presence of long-range disorder, strong Coulomb interactions can produce a pronounced suppression of the density of states (DOS) near the Fermi level, known as a Coulomb gap. In decorated graphene, in addition to screening, the metallic islands or clusters inevitably introduce spatially inhomogeneous random potentials, which enhance localization and promote interaction-driven depletion of the low-energy DOS. Additional data of this Coulomb gap and taken at higher spectral resolution shown in Fig. S9 exhibiting a robust V-shaped suppression centered at E_F . This feature persists over a wide range of doping and temperature, consistent with the formation of a Coulomb gap [2, 3] and thus should not be confused with the excitonic mass gap.

VIII. Calibration of the STM junction temperature

At ultralow temperatures, the effective electronic temperature of tunnelling-junction can include contributions from electronic heating beyond the lattice temperature. Prior to measurement on graphene, we have calibrated against the spectral gap of the layered 2D superconductor PdTe₂. The crystal was cleaved in ultra-high vacuum and immediately transferred into the STM for subsequent measurements. An atomic-resolution topographic image is shown in the inset of Fig. S10b.

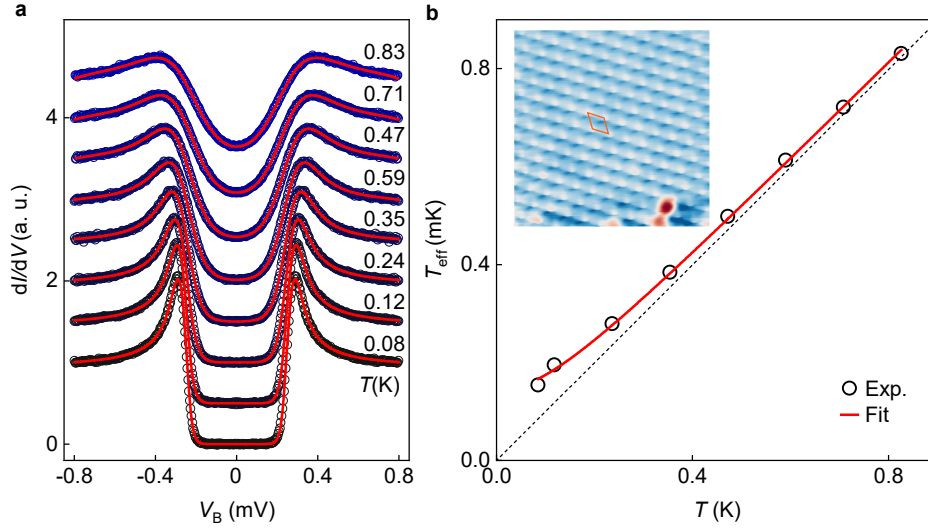


Fig. S10. STM junction temperature calibration. **a**, Superconducting spectra of PdTe₂ measured at STM bath temperatures (T) from 0.08 to 0.8 K. Solid lines are fits to Maki theory, from which the effective temperature of the tunnelling junction is extracted. The spectra were acquired with at tunnelling setpoint of $V_B = 1$ mV and $I = 1$ nA, using a lock-in modulation of $V_m = 2$ μ V at a frequency of 836 Hz. **b**, Extracted effective temperatures as a function of bath temperature T . The solid line is a fit to Eq. S6, yielding a minimum effective temperature of $T_0 = 143$ mK. The dashed line, $T_{\text{eff}} = T$, is shown for reference. Inset: atomic-resolution topography of PdTe₂ (5×5 nm²), acquired at $V_B = 5$ mV and $I = 100$ pA.

Spectra of the superconducting gap are plotted in Fig. S10a, where red solid lines show fits to Maki theory [4–6]. In the absence of external magnetic field, the superconducting density of states (DOS) can be expressed as

$$\rho(E) = \text{Re}\left(\frac{u}{\sqrt{u^2 - 1}}\right), \quad (\text{S4})$$

where u is defined by the nonlinear equation,

$$u = \frac{E}{\Delta} + \zeta \frac{u}{1 - u^2}. \quad (\text{S5})$$

with superconducting gap Δ and pair-breaking parameter ζ . Temperature enters the analysis by convolving Eq. S4 with the derivative of the Fermi-Dirac distribution, allowing us to extract a superconducting gap Δ , pair-breaking parameter ζ , and effective temperature T_{eff} . Figure S10b presents the extracted effective temperatures as a function of the STM bath temperatures (T). The solid line shows a fit to a voltage noise model [7, 8],

$$T_{\text{eff}} = \sqrt{T^2 + T_0^2}, \quad (\text{S6})$$

yielding a minimum effective temperature $T_0 = 143$ mK.

IX. Theory of exciton formation and condensation

A. Scales

In the theory supplement we use electromagnetic CGS units, and sometimes set $\hbar = 1$. Pristine graphene with chemical potential at the Dirac point has only one spatial scale, which is the lattice spacing $a \sim 1$ Å. The corresponding momentum and energy scales are, $\Lambda = \hbar/a$ and $E_\Lambda = v_\Lambda \Lambda \sim 7$ eV [9], respectively. Due to the electron-electron Coulomb interaction the Dirac velocity depends on the energy E [9], as

$$v^E = v_\Lambda \left[1 + \frac{\alpha_\Lambda}{4} \ln \left(\frac{E_\Lambda}{E} \right) \right], \quad (\text{S7})$$

where,

$$\alpha_\Lambda = \frac{e^2}{\varepsilon_r \hbar v_\Lambda} \quad (\text{S8})$$

and ε_r is the dielectric constant. When the energy $E \rightarrow 0$ the velocity is expected to be diverging ($v \rightarrow \infty$) in unscreened graphene.

In metal-decorated graphene with the chemical potential at the Dirac point, the Coulomb interaction is screened at distances smaller than the screening length l_s , the corresponding momentum

114 is $\Lambda_s = \hbar/l_s$. Hence the Fermi-Dirac velocity measured at energy smaller than $E_s \sim v_\Lambda \Lambda_s$ be-
 115 comes energy independent

$$v = v_\Lambda \left[1 + \frac{\alpha_\Lambda}{4} \ln \left(\frac{E_\Lambda}{E_s} \right) \right] \quad (\text{S9})$$

116 The corresponding fine structure constant,

$$\alpha = \frac{e^2}{\varepsilon_r \hbar v} \quad (\text{S10})$$

117 is that of Eq. (1) of the main text used to calculate the data shown in Fig. 2h.

118 In our decorated devices the Dirac velocity measured within $|E_D| \leq 200$ meV is reduced
 119 compared to the pristine case roughly by factor of 1/2. From here, using Eqs. S7 and S9 we
 120 estimate the screening length and the screening energy cutoff

$$\begin{aligned} l_s &\sim 1 \text{ nm} \\ E_s &\sim 0.5 \text{ eV}. \end{aligned} \quad (\text{S11})$$

121 Of course this is a very crude estimate since the dependencies on energy in Eqs. S7 and S9 are
 122 only logarithmic. What is important, compared to the pristine graphene, in the metal decorated
 123 graphene there is an additional spatial scale, l_s , and a related energy scale, E_s .

124 **B. Effective Hamiltonian of electron-hole interaction**

125 Since the long range Coulomb interaction is screened an electron attracts a hole only at short
 126 distances. Hence we can approximate the electron-hole attraction by δ -function and the Hamilto-
 127 nian of the pair reads

$$H = v \boldsymbol{\sigma}_1 \cdot \mathbf{p}_1 + v \boldsymbol{\sigma}_2 \cdot \mathbf{p}_2 - g \delta(\mathbf{r}_1 - \mathbf{r}_2) \quad (\text{S12})$$

128 Here the subscript 1 corresponds to an electron and the subscript 2 corresponds to a hole. The
 129 coupling constant can be expressed as,

$$g \sim \int_0^{l_s} \frac{e^2}{\varepsilon_r r} 2\pi r dr = \frac{e^2}{\varepsilon_r} 2\pi l_s = \frac{2\pi v \alpha}{\Lambda_s} \quad (\text{S13})$$

130 where α is that of Eq. 1 used in the main text and as Eq. S10.

C. Critical coupling g_c , resonances in electron-hole scattering as precursor to exciton condensation.

Unlike the weak coupling BCS problem here we are dealing with a strong coupling problem. This implies that for exciton condensation the coupling constant must be larger than some critical value, $g > g_c$. If $g < g_c$ graphene is in the usual semimetallic phase, but the electron-hole scattering must manifest a scattering resonance as a precursor to the condensation. We stress again that there is no similar effect in the weak coupling BCS. In this section we consider the electron-hole scattering at $g < g_c$.

Diagrams for the electron hole scattering amplitude with total momentum zero are shown in Fig. S11. Unlike in standard many body technique we show the hole moving in the same direction

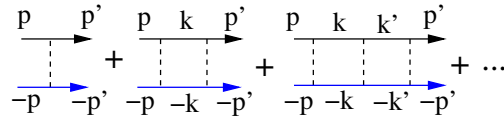


Fig. S11. Electron-hole scattering diagrams

as electron (blue lines). In the ladder intermediate states in Fig. S11 one has to account only positive energy states since the negative energy states are Pauli blocked. Because of Pauli matrices the summation of the ladder is not quite straightforward. Let define scattering angles as

$$\begin{aligned}
 \mathbf{p} &= (p, 0) \\
 \mathbf{p}' &= p(\cos\phi, \sin\phi) \\
 \mathbf{k} &= k(\cos\theta, \sin\theta) \\
 \mathbf{k}' &= k'(\cos\theta', \sin\theta')
 \end{aligned} \tag{S14}$$

and calculate first angular factors in the first, second, third, etc, order amplitudes

$$\begin{aligned}
 f_1 : \quad & \cos^2(\phi/2) = \frac{1}{2}(1 - \cos\phi) \\
 f_2 : \quad & \int \cos^2(\theta/2) \cos^2((\theta - \phi)/2) \frac{d\theta}{2\pi} = \frac{1}{4} \left(1 + \frac{1}{2} \cos\phi \right) \\
 f_3 : \quad & \int \frac{1}{4} \left(1 + \frac{1}{2} \cos\theta' \right) \cos^2((\theta' - \phi)/2) \frac{d\theta'}{2\pi} \\
 & = \frac{1}{8} \left(1 - \frac{1}{4} \cos\phi \right) \\
 & \dots \\
 f_n : \quad & = \frac{1}{2^n} \left(1 - \frac{1}{(-2)^{n-1}} \cos\phi \right)
 \end{aligned} \tag{S15}$$

147

Hence the full scattering amplitude is

$$\begin{aligned}
F(\epsilon, \phi) &= F_s(\epsilon) + F_p(\epsilon) \cos \phi \\
F_s(\epsilon) &= \frac{-g/2}{1 + \frac{g}{2}I} \\
F_p(\epsilon) &= \frac{g/2}{1 - \frac{g}{4}I} \\
I &= \int_0^{2\Lambda_s} \frac{1}{\epsilon - 2vk + i0} \frac{kdk}{2\pi}, \tag{S16}
\end{aligned}$$

148

where ϵ is the total energy of the pair, $\epsilon \ll 2v\Lambda_s$. The integral I is negative, therefore the interaction enhances the s -wave amplitude and suppresses the p -wave one. The upper limit of the integration is taken as $2\Lambda_s$, given that the Fourier transform of the interaction is

149

150

$$U_q = \begin{cases} g = \frac{2\pi e^2/\epsilon_r}{\Lambda_s} & q < \Lambda_s \\ g \frac{\Lambda_s}{q} = \frac{2\pi e^2/\epsilon_r}{q} & q > \Lambda_s \end{cases} \tag{S17}$$

151

This yields the δ -function approximation at $q < \Lambda_s$ and the unscreened Coulomb's law at $q > \Lambda_s$.

152

Therefore, the actual integration in the diverging part of I is

$$I = \int_0^{\Lambda_s} \frac{1}{k} kdk + \int_{\Lambda_s}^{\infty} \frac{\Lambda_s^2}{k^2} \frac{1}{k} kdk \approx \int_0^{2\Lambda_s} \frac{1}{k} kdk \tag{S18}$$

153

This justifies the upper limit of the integration.

154

Divergence of the zero energy s -wave scattering amplitude, $F_s(\epsilon = 0) = \infty$, indicates the onset of exciton condensation. Using Eq. S16 one finds the condition of the onset

155

$$1 - \frac{g}{8\pi v} \int_0^{2\Lambda_s} dk = 1 - \frac{g\Lambda_s}{4\pi v} = 0, \tag{S19}$$

156

that gives the critical value

$$g_c = \frac{4\pi v}{\Lambda_s} \tag{S20}$$

157

Comparing this with $g \sim \frac{2\pi v\alpha}{\Lambda_s}$ (Eq. S12), we find the critical value of the “fine structure constant”,

158

$\alpha_c \sim 2$.

159

As we already pointed out at $g < g_c$ the system is a normal Dirac liquid (semimetal) with electron-hole scattering resonance at a positive energy. Indeed, evaluation the integral I at $0 < \epsilon \ll 4v\Lambda_s$ gives the following scattering amplitude.

161

$$F_s = \frac{-g/2}{1 - \frac{g}{g_c} - \frac{g\epsilon}{16\pi v^2} \left[\ln \left(\frac{4v\Lambda_s}{\epsilon} \right) + i \right]} \propto \frac{1}{e - \epsilon_0 + i\Gamma/2} \tag{S21}$$

Here ϵ_0 is position of the resonance and Γ is its width, Assuming that $\delta g = g_c - g \ll g_c$ the values are

$$\begin{aligned}\epsilon_0 &= \frac{4v\Lambda_s}{\ln(g_c/\delta g)} \times \frac{\delta g}{g_c} \\ \Gamma &= \frac{2\epsilon_0}{\ln(g_c/\delta g)} \ll \epsilon_0\end{aligned}\tag{S22}$$

The resonance is a precursor to the exciton condensation. It is worth noting that the existence of the resonance is essentially based on the contact attractive interaction. This is underlined by the fact that resonance energy ϵ_0 , Eq. S22, is proportional to the screening cutoff Λ_s . The unscreened long-range attractive Coulomb interaction results in the Rutherford scattering of massless particles that does not have any resonances, see Ref. [10].

D. Exciton condensation at $g > g_c$

At $g > g_c$ the normal Fermi-Dirac liquid is unstable with respect to the exciton condensation. Here we consider only the $\mu = 0$ case. Let us look for the condensate wave function in the BCS-like form,

$$\begin{aligned}\Psi &= \Pi_k (u_k + v_k b_{-\mathbf{k}}^\dagger c_{\mathbf{k}}^\dagger) |0\rangle \\ u_k^2 + v_k^2 &= 1.\end{aligned}\tag{S23}$$

Here $|0\rangle$ is the “physical vacuum”, that is, the normal Fermi-Dirac sea filled by electrons up to the Dirac point ($\mu = 0$). In essence the wave function Eq. S23 is a straightforward extension of the “Cooper-like” problem considered in the previous section. The standard calculation gives the energy that is the expectation value of Eq. S12 over Ψ .

$$\begin{aligned}E &= \sum_{\mathbf{k}} 2\eta_{\mathbf{k}} v_{\mathbf{k}}^2 - \frac{g}{2} \sum_{\mathbf{k}, \mathbf{k}'} u_{\mathbf{k}} v_{\mathbf{k}} u_{\mathbf{k}'} v_{\mathbf{k}'} \\ \eta_{\mathbf{k}} &= v_{\mathbf{k}}\end{aligned}\tag{S24}$$

Here, based on the scattering resonance analysis we consider only s-wave, $v_{\mathbf{k}}$ is independent of the $\mathbf{k}$ -direction. Therefore, because of the first of Eqs. S15, $g \rightarrow g/2$. This is a consequence on linear (“ultrarelativistic”) dispersion. The energy (Eq. S24) must be minimized with Lagrange multiplier $\lambda_{\mathbf{k}}$ that provides the normalization $u_{\mathbf{k}}^2 + v_{\mathbf{k}}^2 = 1$. Variation with respect to $u_{\mathbf{k}}, v_{\mathbf{k}}$ gives

$$\begin{aligned}(\epsilon_p - \eta_p)u_p - \Delta v_p &= 0 \\ -\Delta u_p + (\epsilon_p + \eta_p)v_p &= 0\end{aligned}\tag{S25}$$

181 Here $\epsilon_p = \eta_p + \lambda_p$. These Eqs. result in relations that are similar to the BCS one

$$\begin{aligned}\epsilon_p &= \sqrt{\eta_p^2 + \Delta^2} \\ \Delta &= \frac{g}{2} \sum_k u_k v_k\end{aligned}\tag{S26}$$

182 Equation for the gap is

$$1 = \frac{g}{4} \sum_k \frac{1}{\epsilon_k} = \frac{g}{4} \int_0^{2\Lambda_s} \frac{1}{\epsilon_k} \frac{d^2 k}{(2\pi)^2}\tag{S27}$$

183 Consistently with the scattering problem this equation has solution only at $g > g_c$, see Eq. S20. At
184 $\delta g = g - g_c \ll g_c$ the solution is

$$\Delta = 2\Lambda_s v \frac{\delta g}{g_c} = \Delta = 2E_s \frac{\delta g}{g_c}\tag{S28}$$

185 The exciton condensate filling factor is

$$v_p^2 = \frac{1}{2} \left(1 - \frac{\eta_p}{\epsilon_p} \right)\tag{S29}$$

186 At large p the filling factor is $v_p^2 \approx \frac{\Delta^2}{4\eta_p^2} \propto \frac{1}{p^2}$. Therefore, the total density of exciton condensate is
187 logarithmically enhanced

$$n_e = \int v_p^2 \frac{d^2 p}{(2\pi)^2} \approx \frac{\Delta^2}{8\pi} \ln \left(\frac{2v\Lambda_s}{\Delta} \right)\tag{S30}$$

188 The total energy density (S24) is

$$\begin{aligned}E &= \frac{1}{2\pi v^2} \int_0^\infty \left[\eta \left(1 - \frac{\eta}{\sqrt{\eta^2 + \Delta^2}} \right) - \frac{\Delta^2}{2\sqrt{\eta^2 + \Delta^2}} \right] \eta d\eta \\ &= -\frac{\Delta^3}{12\pi v^2}\end{aligned}\tag{S31}$$

189 While, technically, the analysis in the present section is similar to that in the BCS problem,
190 physically the situation is very different. The major difference is the quantum phase transition
191 from normal Fermi-Dirac liquid at $g < g_c$ to the excitonic condensate at $g > g_c$. There is nothing
192 like this in BCS. We do not know the value of the coupling constant g . Comparing the measured
193 gap $\Delta \approx 7\text{meV}$ with Eqs. S11 and S28 we conclude that the supercriticality is $\delta g/g_c \sim 0.01$.

194 E. Superfluid stiffness near zero temperature

195 Using the energy density (Eq. S31) we can estimate stiffness of the condensate. The stiffness
 196 is important because it determines the critical temperature. Let us perform twist of the condensate,
 197 $\Delta \rightarrow \Delta e^{iq \cdot r}$. The twist results in the lower cutoff in all equations of the previous section, $k_{\min} \sim$
 198 $q/2$. Hence, $\Delta \rightarrow \sqrt{\Delta^2 - v^2 q^2/4}$. Hence the total energy (Eq. S31) is changed as $-\frac{\Delta^3}{12\pi v^2} \rightarrow$
 199 $-\frac{(\Delta^2 - v^2 q^2/4)^{3/2}}{12\pi v^2} \approx -\frac{\Delta^3}{12\pi v^2} + \frac{\Delta}{32\pi} q^2$. On the other hand, the stiffness ρ is defined as $\delta E = \frac{\rho}{2} q^2$.
 200 Hence, the stiffness is

$$\rho \approx \frac{\Delta}{16\pi} \quad (\text{S32})$$

201 We stress again that this is the stiffness near $T = 0$.

202 F. Temperature dependence of the excitonic gap, $\Delta/T_c \geq 100$ ratio.

203 In an infinite system, the BKT transition is sharp, whereas it becomes a smooth crossover in a
 204 finite system. To better capture the temperature evolution of the spectral gap, we take into account
 205 the finite size of the graphene regions between metal islands. At low temperatures, the spectral
 206 gap is approximately proportional to the condensate stiffness. Hence, the temperature evolution
 207 of the spectral gap follows directly from that of the stiffness $\rho(T)$. We find that the temperature
 208 dependence of the spectral gap is well reproduced by Monte Carlo simulations of finite clusters
 209 within the 2D XY model, using a cluster size of $N = 16$ [10], as shown in Fig. S5. This cluster
 210 size is consistent with our samples: the separation between metallic islands is $R \sim 10\text{--}30$ nm and
 211 the effective screening length is $l_s \sim 1$ nm (Eq. S11), corresponding to an expected cluster size of
 212 $N \sim R/l_s \sim 20$.

213 For a 2D BKT transition, the transition temperature satisfies the Nelson–Kosterlitz (NK) rela-
 214 tion [11]

$$k_B T_{\text{BKT}} = \frac{\pi}{2} \rho(T_{\text{BKT}}), \quad (\text{S33})$$

215 where $\rho(T_{\text{BKT}})$ is the condensate stiffness at the transition. Together with Eq. S32, this gives

$$k_B T_{\text{BKT}} \approx \frac{1}{32} \Delta(T_{\text{BKT}}). \quad (\text{S34})$$

216 This relation yields transition temperatures of $T_{\text{BKT}} \approx 0.4$ K for WTe₂-decorated graphene and
 217 $T_{\text{BKT}} \approx 0.44$ K for W-decorated graphene (dashed lines in Figs. S12), respectively. Despite the
 218 different metal species, both systems exhibit a similarly large gap-to-critical-temperature ratio,
 219 $\Delta/k_B T_c \approx 180$, suggesting a common underlying pairing mechanism.

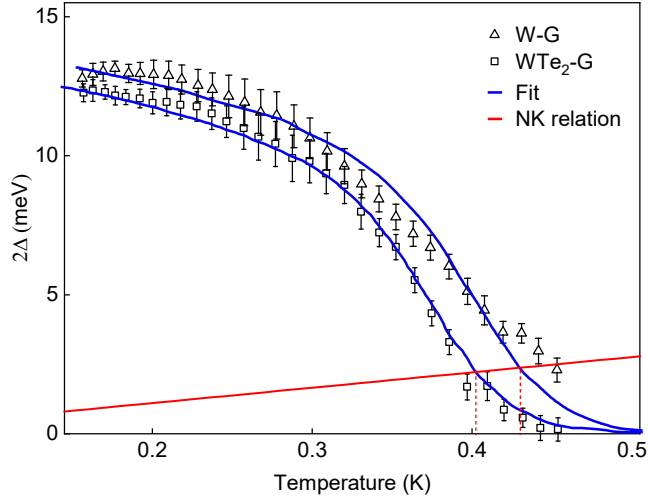


Fig. S12. Comparison of the temperature evolution of the spectral gap in the two systems, combining the data from Figs. S5c and S5f. The red solid line shows the NK-relation function (Eq. S34), and the red dashed lines indicate the estimated transition temperatures of $T_{\text{BKT}} = 0.40$ K and 0.44 K for WTe₂-decorated and W-decorated graphene, respectively.

G. Magnetic field dependence of the excitonic gap

Equation S27 for the gap contains integration over all momenta from zero to $2\Lambda_s$. However, in an out-of-plane magnetic field the electron/hole orbit is limited by magnetic length $l_B = \sqrt{\frac{\hbar}{|eB|}} \approx \frac{26\text{nm}}{\sqrt{B(\text{T})}}$. Therefore electron and hole separated by a distance larger than l_B do not interact because the interaction is contact. This implies that in a magnetic field the integration over momenta must be limited by $k_{\text{min}} \approx 1/(2l_B)$. This is very similar to our calculation of the superfluid stiffness. Hence, Eq. S27 is replaced by

$$1 = \frac{g_c + \delta g}{8\pi v} \int_{k_{\text{min}}}^{2\Lambda_s} \frac{k dk}{\sqrt{k^2 + \Delta^2/v^2}} \quad (\text{S35})$$

From here we find

$$\Delta(B) = \sqrt{\Delta_0^2 - v^2 \hbar |eB|/4} \quad (\text{S36})$$

where Δ_0 is the gap (Eq. S28) at zero magnetic field. Fits of data with this formula are presented in panels e and f of Fig.3 in the main text. We stress again that the suppression of the gap by magnetic field is due to the contact electron-hole attraction. With a long range electron-hole attraction an out-of-plane magnetic field enhances the gap.

H. CDW: intervalley pairing

The analysis above is equally applicable to the intervalley and to the intravalley pairing. Yet, the observed CDW points at the intervalley pairing. To discuss the CDW we return to the sublattice basis, whereby low-energy Dirac theory for pristine graphene is

$$H_0(\mathbf{k}) = v_F(k_x\sigma_x\tau_z + k_y\sigma_y). \quad (\text{S37})$$

Recasting the excitonic gap in sublattice basis, we add the simplest correction that: (i) gaps the spectrum (as observed), (ii) preserves point-group symmetries (trivial ordering, A_1), and (iii) has total momentum $\mathbf{Q} = \mathbf{K}$, i.e. is a charge density wave. The following structure achieves this

$$\delta H = \Delta\sigma_x\tau_x. \quad (\text{S38})$$

Note some other symmetry based properties of this gap structure: (i) preserves time-reversal symmetry (TRS) $\Theta = \tau_x\mathcal{K}$ (simplest case), (ii) preserves particle-hole symmetry (PHS) $\mathcal{P} = \sigma_z\mathcal{K}$. Since this preserves the anti-unitary PHS, i.e.

$$\text{PHS: } H_0(\mathbf{k}) + \delta H \rightarrow -H_0^*(-\mathbf{k}) - \delta H^* \quad (\text{S39})$$

then the LDOS satisfies

$$\rho(\mathbf{r}, \omega) = \rho(\mathbf{r}, -\omega). \quad (\text{S40})$$

Importantly, this implies that the excitonic charge density wave does not exhibit *contrast inversion* in STM imaging, which is often attributed to charge density wave orderings. STM measurements confirm the presence of a CDW (Figure 4 in the main text). These findings are all consistent with the excitonic order of Eq. S38.

I. Doping dependence of the excitonic gap, catalysis of exciton condensation by phonons

Doping away from the Dirac point strongly suppresses, as shown in Fig. 2e of the main text. Over a range of $\mu_c \sim \pm 70 \text{ meV} \sim 10\Delta$ in electrochemical potential. This is in disagreement with basic calculation which suggests that $\mu_c \sim \Delta$. Below we address this apparent disagreement considering electron-phonon coupling; phonons enhance the excitonic pairing, providing an attractive potential for $|\mu| \lesssim \mu_c$, with a crude estimate $\mu_c \approx 85 \text{ meV}$, which we detail below.

Graphene has a phonon mode with energy $\Omega_0 \approx 170 \text{ meV}$ at the K -point [12]. This mode

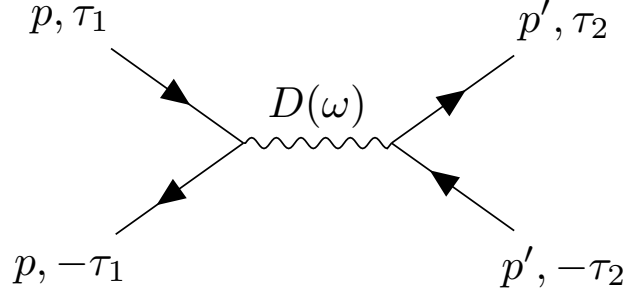


Fig. S13. Phonon contribution to excitonic pairing: the direct channel is attractive

belongs to A_1 , a type of in-plane breathing mode, with kinetic structure

$$\delta H_{\text{el-ph}} = g u_{\mathbf{k}} \sigma_x \tau_x. \quad (\text{S41})$$

Here $u_{\mathbf{k}}$ is the displacement/phonon field and g is the electron-phonon coupling, taken to be constant here.

Importantly, this electron-phonon coupling directly promotes the excitonic *intervalley* ordering we are interested in; integrating out the phonon, under the assumption it is static (since Ω_0 is already large compared to all other scales), we arrive

$$\delta V_{\text{el-ph}} = \frac{g^2}{\omega^2 - \Omega_0^2} (\sigma_x \tau_x) \otimes (\sigma_x \tau_x). \quad (\text{S42})$$

This provides attraction for excitons in the direct/ s -channel, which is depicted in Fig. S13. Note that using the language of Quantum Electrodynamics this is the “annihilation mechanism of attraction” that contributes to the energy of positronium [13]. Of course in the case of the positronium the annihilation goes via a virtual photon instead of the phonon. The annihilation mechanism does not contribute to the intravalley pairing and it does contribute to the intervalley pairing that we observe.

We see that phonon-mediated pairing is attractive for excitonic frequencies $\omega < \Omega_0$, and repulsive thereafter. Taking on-mass-shell condition, then we may equate $\omega = 2\mu$, since to excite an exciton requires 2μ in energy. We hence conclude, crudely, that for $2\mu \lesssim \Omega_0$ the phonon provides attraction, stabilising the excitonic pairing, and hence, the excitonic gap should only close for $\mu > \mu_c \approx \Omega_0/2 = 85 \text{ meV}$. Thus electron-phonon coupling in conjunction with the excitonic condensation provides an explanation for the anomalously large stability of the gap against doping μ .

The phonon does not constitute the primary pairing mechanism, but is understood to enhance the pairing strength and, in particular, the stability of the excitonic phase under doping.

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