Revealing the Charge Density Wave Proximity Effect in Graphene on 1T-TaS$_2$

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Abstract

The proximity-effect, a phenomenon whereby materials in close contact appropriate each other’s electronic-properties, is widely used in nano-scale devices to induce electron-correlations at heterostructure interfaces. Layered group-V transition metal dichalcogenides host charge density waves and are expected to induce CDWs in a thin proximal 2D metal such as graphene. Thus far, however, the extremely large density of states of the TMDs compared to graphene have precluded efforts to unambiguously prove such proximity induced charge density waves (CDW). Here, using scanning tunneling microscopy (STM) and spectroscopy (STS), we report the first
conclusive evidence of a CDW proximity effect between graphene and the commensurate CDW in 1T-TaS$_2$ (TaS$_2$ for brevity). We exploit the Mott gap of 1T-TaS$_2$ to scan the sample at bias voltages wherein only the graphene layer contributes to the STM topography scans. Furthermore, we observe that graphene modifies the band structure at the surface of TaS$_2$, by providing mid-gap carriers and reducing the strength of electron correlations there. We show that the mechanism underlying the proximity induced CDW is well-described by short-range exchange interactions that are distinctly different from previously observed proximity effects.

**Introduction**

The isolation and manipulation of atomically thin materials provides a ready-made two dimensional electron system$^1$ whose properties can be tuned by external knobs, such as stress or substrate morphology$^{2-6}$, leading to the emergence of correlated electron phases. Distinct from these external knobs, a very powerful approach to manipulate electron correlations is by contact proximity effects. It is well known that proximitizing materials that host correlated electron phases with a normal metal, induces correlations in the metal$^7$. This is a direct consequence of the quantum mechanical properties of electrons in solids; specifically, the non-local nature of electrons. As quantum particles do not have a well-defined position, electronic states cannot abruptly change from one type of ordering to another at the interface between two materials. Consequently, correlated states persist into the normal metal where scattering events begin to destroy the coherence (and vice versa). In the case of 2D materials where scattering is reduced due to their atomically sharp interfaces, proximity-effects are particularly robust allowing correlated states to persist over long distances. The discovery of graphene and other 2D materials, together with the technology enabling 2D heterostructures has led to the
observation of strong proximity effects at the atomic limit including proximity induced superconductivity, magnetism\textsuperscript{8} and spin-orbit effects\textsuperscript{9-16}.

The CDW state is yet another correlated electron state which is driven by electron phonon coupling\textsuperscript{17} (EPC) or alternatively via electron correlations\textsuperscript{18}. It has been observed in systems such as unconventional superconductors\textsuperscript{19} and many 2D transition-metal dichalcogenide (TMDs)\textsuperscript{20}. However, identifying a proximity effect between a CDW material and a normal metal has been elusive. This is primarily due to screening in 3D metals and interface defect scattering. The use of 2D materials that were stacked in an inert atmosphere to avoid interface damage and contamination, together with sensitive local probes providing direct access to the charge distribution in graphene, has made it possible to overcome these hurdles\textsuperscript{21,22}.

In this work, we present evidence of the emergence of a CDW in graphene produced by its proximity to the commensurate CDW (CCDW) in a TaS\textsubscript{2} substrate. While there are several recent experiments which have studied vertical heterostructures of graphene and a TMD with STM\textsuperscript{22-24}, it has proven difficult to experimentally isolate the electronic states in the graphene layer from those belonging to the TMD substrates making it difficult to definitively prove a proximity induced CDW in graphene. In this work, by using the coexisting CDW and Mott gap in TaS\textsubscript{2}, we can isolate the electronic states in the graphene layer and establish the presence of the proximity induced CDW. Using scanning tunneling microscopy (STM) and spectroscopy (STS) we show that the charge density modulation of the CCDW in TaS\textsubscript{2} persists within the contacted graphene layer. Additionally, we find that the Mott gap in TaS\textsubscript{2} is reduced due to its close contact with the Dirac carriers in graphene. By comparing with first-principle calculations based on density-functional theory (DFT), we find that in addition to a global
charge-transfer between the two surfaces caused by the work-function difference, the proximity induced CDW in graphene which is characterized by local charge modulation and lattice distortion is described by a novel mechanism of short-range exchange interactions mediated by second-order local electron hopping, which is distinctly different from superconducting, magnetic and spin-orbit proximity effects.

Results and discussion

Scanning tunneling microscopy of graphene covered TaS$_2$.

**Figure 1** Scanning tunneling microscopy of graphene covered TaS$_2$; (a) Schematic of the device and STM measurement setup. (b, left) Cartoon showing a single star-of-David (red) in TaS$_2$. The black arrows denote the displacement of Ta atoms towards a central Ta atom. (b, right) The transition temperatures between the incommensurate CDW (ICCDW), nearly commensurate CDW (NCCDW) and commensurate CDW (CCDW) phases of TaS$_2$. (c) Cartoon showing a monolayer graphene layer on top of TaS$_2$. Here, the angle between the graphene layer and TaS$_2$ is 13.9° to form a commensurate unit cell (cyan) for DFT calculations. (d) An STM topography image of graphene covered TaS2 measured at 77 K using a tunneling set point of
I=150 pA, Vb=250 mV. Each bright spot corresponds to a star of David with a measured CDW wavelength of 1.2 nm. This can also be seen in the 2D-FFT of the topography data (inset). (e) An STM topography image and its 2D FFT (inset) of a bulk TaS2 sample measured at 77 K using a tunneling set point of I=150 pA, Vb=500 mV.

TaS2 is comprised of hexagonal layers of tantalum atoms coordinated octahedrally by sulfur atoms. Below 550 K a high temperature metallic phase transits into an incommensurate CDW phase, followed by a nearly commensurate CDW (NCCDW) below 350 K, and a CCDW below 180 K21 (Fig. 1b, right). The CDW unit cell consists of a 13-atom cluster where 12 atoms displace from their high-temperature equilibrium positions toward the central, 13th Ta atom forming a $\sqrt{13} \times \sqrt{13}$ reconstructed supercell (Fig. 1b, left) also known as a David star (DS) structure. The CDW in TaS2 is driven by strong electron-phonon coupling which critically suppresses an acoustic phonon mode along the $\Gamma \rightarrow M$ direction25,26 (Supplementary Fig. 4) leading to the static displacement of the lattice with the wavevector $Q_{\text{CDW}}$, corresponding to the $\sqrt{13} \times \sqrt{13}$ CDW. This soft phonon mode at the Kohn anomaly wavevector, $Q_{\text{CDW}}$, consists primarily of longitudinal vibrations of Ta atoms with a minor contribution from transverse vibrations of S atoms relative to the phonon propagating direction $Q_{\text{CDW}}$. The DS atomic arrangement involves the local lattice contraction around the center of the star, in which the bond lengths between Ta ions are shorter than those between Ta ions outside the star.

We studied vertical heterostructures consisting of thin TaS2 flakes (7-50 nm) covered by monolayer graphene (Fig. 1a). The samples were assembled inside a dry argon-filled glovebox (<0.1 ppm O2, H2O)21,27 to avoid oxidation of the air sensitive surface of TaS2 and subsequently cooled to 77 K inside a home-built STM28,29. At this temperature TaS2 is in the CCDW regime. The STM topography (Fig. 1d) measured on a graphene-covered sample using a tunneling set-point of $V_b = 250$ mV, I=150 pA shows a triangular array of tall spots. The lattice spacing for this triangular array is about 1.2 nm as measured directly from the
topography image and confirmed with its 2D-FFT (Fig. 1d, inset). This spacing is equal to the expected CDW wavelength ($L_{CDW}$) in TaS$_2$ indicating that each tall spot corresponds to one DS. The topography of the graphene covered sample thus reflects the distinct arrangement of the DS clusters which are the hallmark of the CDW in TaS$_2$\textsuperscript{30}. The topography is identical to that measured on a bulk TaS$_2$ sample without a graphene cover under similar tunneling conditions (Fig. 1e). This is also true for typical negative bias setpoints of $V_b < -300$ mV. Therefore, for sufficiently large bias voltages of either sign, there is no noticeable difference in the STM topography of TaS$_2$ with or without a graphene cover. This is not surprising given the strong CDW in TaS$_2$ and the relatively small density of states in graphene compared to TaS$_2$.

Interfacial charge transfer and reduction of on-site Coulomb repulsion

**Figure 2** Effects of interaction between graphene and TaS$_2$. (a) and (b) are the measured $dI/dV$ curves on bulk TaS$_2$ and graphene/TaS$_2$ respectively. The positions of the Hubbard band peaks and the broad features originating from the CDW distortion are labelled. The dashed lines in (b) indicate the bias voltages at which topography scans (g-i) were measured. (c) The calculated DOS for bulk TaS$_2$. (d) The calculated DOS for the heterostructure decomposed into the graphene DOS (blue curve) and TaS$_2$ (red curve) DOS respectively. The graphene DOS is multiplied by a factor (39) before adding the TaS$_2$ DOS (black curve) for comparison with experiment. This accounts for the smaller tip-graphene separation. See SI for how the factor was estimated.
A map of dI/dV (V_b) vs position across 8 DSs in the graphene/TaS2 sample. The intensity of both the Hubbard bands is highest at the center of the DSs. (f) Three representative spectra from the colorml(e) demonstrate this intensity variation. (g-i) STM topography scans measured at bias voltages of -300 mV, -150 mV and +300 mV respectively. These energies are indicated by dashed lines in (b). The tunneling resistance (R_t) was kept constant at 1 GΩ for all scans. The small lateral mismatch between scans is a result of thermal drift of the sample. All scale bars are 2 nm.

The similarity between the topography scans of TaS2 with or without a graphene cover might suggest that there is little or no interaction between the two materials. Our spectroscopy data shows that this is not the case.

The band structure of bulk TaS2 consists of an isolated, half-filled, spin-degenerate, flat band at the Fermi level (E_F) (Supplementary Figure 5). In the CCDW phase, the flat band splits into an occupied spin up lower Hubbard band (LHB) and empty spin down upper Hubbard band (UHB) with a Mott gap of ~0.5 eV in between (ab initio calculation Fig. 2c). The other bands corresponding to the CDW distortion appear at energies below the LHB (CDW-VB) and above the UHB (CDW-CB) respectively. These spectroscopic features were observed in our experimentally measured dI/dV spectrum (Fig. 2a) on a bulk TaS2 sample and agree with previous reports20,31-33.

The dI/dV spectrum measured on the graphene covered sample (Fig. 2b) similarly shows peaks at ~ -270 meV and +70 meV which are identified as the LHB and UHB. This identification is supported by the intensity variation of these features as a function of distance from SD centers (Fig. 2e,f). However, the Mott gap between the two peaks appears to be absent. Instead, we observe a small but finite density of states. These states presumably belong to the graphene layer which is a gapless semi-metal whose low-energy dispersion consists of Dirac cones which touch at the charge neutrality point (CNP). Our ab initio calculations for monolayer graphene on monolayer TaS2 (Fig. 2d), decomposed into the graphene DOS and TaS2 DOS, reveal that the CNP of graphene is about 0.3 eV above E_F of the system, indicating that it is
hole doped. This kind of interfacial charge transfer is likely due to the differences in work function between the graphene ($\Phi_G \sim 4.6$ eV$^{34}$) and TaS$_2$ ($\Phi_{TaS_2} \sim 5.2$ eV$^{35}$).

Another notable difference is that the separation between the Hubbard band peaks ($\sim 338$ mV) is smaller by about 31% compared to the bare TaS$_2$ sample ($\sim 491$ mV). We attribute this to the reduction of the on-site Coulomb repulsion ($U$) in the graphene covered TaS$_2$. Graphene is known to be a semi-metal with high-mobility itinerant carriers. These highly mobile electrons from graphene somewhat suppress the localized picture of the narrow Hubbard d-bands in TaS$_2$ and thus reduce the on-site $U$ value of Ta. Comparing the $ab$ $initio$ calculated band structures (supplementary Fig. 6) for different $U$ values with the STS results, suggests that the graphene layer screens the Coulomb interaction in TaS$_2$ and the Hubbard $U$ of Ta is lowered from 2.27 eV to 1.70 eV. Therefore, we use a phenomenological value $U=1.70$ eV for our $ab$ $initio$ calculations (Fig. 2d) which reasonably reproduces the trend observed in the STS measurements.
Proximity induced CDW in graphene.

**Figure 3** | Modelling co-tunneling and electrostatic screening

(a) A schematic cross section of the tunneling junction. The mean graphene-TaS$_2$ separation is ~ 0.55 nm. Co-tunneling into the graphene and TaS2 layers was estimated by measuring the tunneling current decay constant $z_0$. (b) The integrated DOS of the graphene (blue curve) and the TaS2 (red curve) in the energy window $[E_F, E_F + eV_b]$ calculated using the DOS from DFT calculations. The integrated DOS for TaS2 is about an order of magnitude larger than that of graphene for large $V_b$. For $V_b$ inside the Mott gap, the graphene DOS dominates. (c) A schematic which considers the screening of the CDW in TaS$_2$ by the electrons in graphene. The bottom subplot shows the electron density ($\rho$) in the TaS$_2$ as a function of position. The resulting electric potential ($U(r)$) denoted by the orange line, tunes the local charge density of the graphene layer. This is denoted by the shift of the charge neutrality point of graphene’s Dirac cones (blue) at two representative positions. The Fermi level is denoted by the dashed black line. The red squiggly lines denote the Hubbard bands of the TaS$_2$. The hatched region shows the states within the energy window $[E_F, E_F + eV_b]$ for the two positions. There are more states available for tunneling in the graphene above the center of the DSs compared to the sides. This should result in a triangular CDW like pattern in topography measured at these tunneling conditions. This is at odds with the observed hexagonal honeycomb pattern. (d) A $dI/dV$ spectrum measured on a bilayer graphene-TaS$_2$ area which shows that the CNP is clearly around 0.3 eV. The Hubbard bands are greatly suppressed compared to the monolayer graphene-TaS$_2$ owing to the increased separation of the topmost graphene layer from the TaS$_2$, additional screening, and the lack of direct hybridization between the two materials.

In addition to the local height variation in the sample, STM topography also depends on the total number of states in the energy window $[E_F, E_F + eV_b]$. For CDW materials, the local
density of states variation dominates the topography signal. Since the integrated density of states of TaS$_2$ is orders of magnitude larger than that of the graphene (Fig. 3b) for large $V_b$, we estimate that there is significant tunneling current into both the graphene and TaS$_2$, despite the larger $z$ separation between the tip and TaS$_2$ layer (Fig. 3a). Consequently, the topography scans measured on the graphene covered sample resemble that of TaS$_2$ as shown for $V_b$=-300 mV (Fig. 2g) and $V_b$=+300 mV (Fig. 2i). Note that these two energy windows include the LHB and UHB of the TaS$_2$ respectively.

Surprisingly, the topography pattern measured at $V_b$=-150 mV (Fig. 2h), which is in the Mott gap of TaS$_2$, is hexagonal instead of triangular, but with the same periodicity as the CDW. A hexagonal lattice is the anti-lattice of a triangular lattice. This energy window includes only the states belonging to graphene since the TaS$_2$ layer is gapped (Fig. 3b) and thus reflects the electronic density redistribution in graphene due to its proximity to TaS$_2$. This observation suggests that the electrons in the graphene layer also form a CDW-like pattern which is out-of-phase with the underlying TaS$_2$ CDW. We call this the proximity induced CDW in graphene. The hexagonal pattern was observed for multiple energy windows inside the TaS$_2$ Mott gap but never outside it (see Supplementary Fig. 9).

Note that this out-of-phase CDW cannot be explained by a simple screening picture (Fig. 3c). To wit: if one models the CDW in TaS$_2$ as a periodic modulation of electron density ($\rho(r)$) which peaks at the center of the DSs (Fig. 3c, bottom), then the electrostatic potential($U(r)$) near the surface of TaS$_2$ will also peak near the center of the DSs (orange line in Fig. 3c). A graphene layer placed on top of TaS$_2$ will experience this spatially varying potential leading to spatial doping variation, i.e., shifting of the local CNP. This CNP variation is in addition to the average shift of the CNP (~ 0.3 eV) due to work function difference between
the two materials as discussed previously. If we consider an energy window in the Mott gap of the \( \text{TaS}_2 \), then there are more graphene states (hatched regions in Fig. 3c) near the center of the SD compared to the region between neighboring DSs. This should result in a triangular CDW pattern in STM topography, which is in-phase with the underlying \( \text{TaS}_2 \) CDW. This is contrary to our observation of a hexagonal pattern, thus ruling out this simple picture.

We also studied an area of the sample covered with bilayer graphene. In this area the topography showed almost no CDW pattern. dI/dV spectra (Fig. 3d) showed a bilayer graphene-like V shape with a pronounced dip near 0.3 V, indicating the CNP. Very weak features corresponding to the position of the Hubbard bands were also seen. These observations indicate that the proximity induced CDW relies on very short-range coupling between the graphene and \( \text{TaS}_2 \).

Modeling the proximity induced CDW.

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**FIGURE 4** Ab initio band structure calculations. (a) Top view of G/TaS\(_2\) heterostructure. Blue, brown, and yellow spheres indicate C, Ta, and S atoms, respectively. Black, blue, and brown rhombuses show the 5x5 graphene/\( \sqrt{13} \times \sqrt{13} \) TaS\(_2\) supercell, graphene 1x1 unit cell, and 1T-TaS\(_2\) 1x1 unit cell, respectively. The graphene and TaS\(_2\) layer are twisted by \(~13.9^\circ\) in this CDW.
phase. (b) Side view of graphene/TaS$_2$ heterostructure overlaid with the charge density map (bubble-shaped color contour indicating the electron density) corresponding to the states at the two crossing points of the Dirac cone and Lower Hubbard band. The slight overlap between graphene and TaS$_2$ electron clouds gives rise to the interlayer coupling and proximity effect. Here, blue, red, and green spheres represent C, Ta, and S atoms, respectively. (c) GGA+U band structure of $\sqrt{3} \times \sqrt{3}$ CDW reconstructed TaS$_2$ with U=2.27 eV (d) GGA+U band structure of graphene/TaS$_2$ using a phenomenological value U = 1.70 eV. Owing to the charge transfer from graphene to TaS$_2$, the Fermi level (zero energy) moves from the lower Hubbard band to the upper Hubbard band, and the graphene-associated Dirac point at the K-point of the superstructure Brillouin zone is shifted to ~0.3 eV above the Fermi level (E$_F$) indicating hole doping. (e-g) The integrated DOS in the energy range from E$_F$ to -300 mV, -200 mV and +300 mV respectively. DSs are overlaid for clarity. At -300 mV and +300 mV the DSs form a triangular lattice while at -200 mV they form a hexagonal lattice.

In addition to the global charge transfer between the graphene and TaS$_2$, we obtain local charge modulations and lattice distortions (see supplementary Fig. 8) mediated by the interfacial orbital hybridizations (Fig. 4b) through the crossing states of the graphene Dirac cone with the TaS$_2$ Hubbard bands (Fig 4d). Since different carbon atoms in the heterostructure unit-cell have a different registration with respect to the underlying CDW SD, it is natural to expect that the hybridization between graphene p$_z$-bands and TaS$_2$-d bands varies as a function of position within the supercell (Fig 4a). This can be seen in the calculated charge density map (Fig. 4b). Moreover, the local density of states is significantly affected by the energy-dependent interlayer hybridizations as can be seen in the calculated band structure (Fig. 4d), particularly within or outside the Mott gap of TaS$_2$. Thus, we calculate the total density of states in the energy windows [E$_F$, E$_F$ + eV$_b$] for V$_b$=-0.3 V, -0.2 V and +0.3 V (Fig. 4e-g). They show triangular, hexagonal and triangular CDW patterns below, within, and above the Mott gap region, respectively, demonstrating the inverted behavior of the in-gap states. These results are in good agreement with our topography scans (Fig. 2g-i) and further support the out-of-phase proximity induced CDW in the graphene layer.
We believe that the proximity induced CDW is a novel effect driven by periodically varying band hybridization. This can be modeled using a simplified Hamiltonian of the graphene/TaS$_2$ system given by:

\[
H = H_d + H_c + H_t,
\]

\[
H_d = \sum_{\langle i,j \rangle, \sigma} -t_{ij}^d d_\sigma^\dagger d_j, + h.c. - \sum_{\langle i',j' \rangle, \sigma} (\Delta_{d}^{CDW}(i',j'))^* d_{i', \sigma}^\dagger d_{j', \sigma} + h.c. + \sum_{\langle i',j' \rangle} |\Delta_{d}^{CDW}(i',j')|^2,
\]

\[
H_c = \sum_{\langle i,j \rangle, \sigma} -t_{ij}^c c_{i, \sigma}^\dagger c_{j, \sigma} + h.c. = \sum_{k, \sigma} (\epsilon_k - \mu) c_{k, \sigma}^\dagger c_{k, \sigma},
\]

\[
H_t = -t \sum_{l, \sigma} c_{l, \sigma}^\dagger d_{l, \sigma} + h.c.,
\]

where $H_d$ ($H_c$) stands for the simplified Hamiltonian of the 1T-TaS$_2$ (graphene) layer, respectively, and $H_t$ describes a weak charge transfer (hopping) term between these two layers (we neglect a small mismatch in spatial locations between the nearest-neighbor sites of the corresponding layer). The insulating TaS$_2$ layer shows CDW order with the order parameter $\Delta_{d}^{CDW}(i',j') \equiv \sum_{\sigma} \langle d_{i', \sigma}^\dagger d_{j', \sigma} \rangle$ where $i', j'$ are sites within the CDW unit cell. The graphene layer has the linear Dirac dispersion: $(\epsilon_k - \mu) \approx \hbar v_F |k - k_F|$ where $v_F$, $k_F$ are the Fermi velocity and Fermi wavevector respectively. Here, $i, j$ refer to the nearest-neighbor sites of the corresponding lattices, and $t_{ij}^{d(c)}$ refers to the nearest-neighbor tight-binding hopping terms on the 1T-TaS$_2$ (graphene) layer, respectively.

Via the second order perturbation in the $H_t$ term of the CDW unit cell, the following exchange term $H^{(2)}_t$ is generated:

\[
H^{(2)}_t = t^2 \sum_{\langle l', \sigma \rangle} c_{l', \sigma}^\dagger d_{l', \sigma}^\dagger d_{l', \sigma}^\dagger c_{l', \sigma} + h.c.
\]
A simple mean-field decoupling of $H_t^{(2)}$ in terms of $\Delta_{d}^{CDW}(i',j')$ (considering only $\sigma = \sigma'$ and assuming spin-isotropic CDW order $(d_{j',\sigma}^{+}d_{j',\sigma}) = (d_{j',\sigma}^{+}d_{j',\sigma})$) gives $H_t^{(2)} \rightarrow H_{t_2}^{MF}$ with:

$$H_{t_2}^{MF} \approx -t^2/2 \sum_{(i',j'),\sigma} (\Delta_{d}^{CDW}(i',j'))^* c_{i',\sigma}^{+} c_{j',\sigma} - t^2/2 \sum_{(i',j'),\sigma} (\Delta_{d}^{CDW}(i',j'))^* (c_{i',\sigma}^{+} c_{j',\sigma}) + h. c.,$$

where the mean-field decoupling term $(c_{i',\sigma}^{+} c_{j',\sigma}) d_{i',\sigma}^{+} d_{j',\sigma})$ in $H_{t_2}^{MF}$ is neglected since we expect $|\langle c_{i',\sigma}^{+} c_{j',\sigma} \rangle| \ll |\langle d_{i',\sigma}^{+} d_{j',\sigma} \rangle|$. The CDW proximity effect is manifested in $H_{t_2}^{MF}$ as a weak CDW order $\sum_{\sigma} (c_{i',\sigma}^{+} c_{j',\sigma})$ is induced on the graphene layer by the second order charge transfer between the two layers with the following identification:

$$\Delta_{c}^{CDW}(i',j') \equiv -1/2 \sum_{\sigma} \langle c_{i',\sigma}^{+} c_{j',\sigma} \rangle^* = -t^2/2 (\Delta_{d}^{CDW}(i',j'))^*,$$

or equivalently, $\sum_{\sigma} (c_{i',\sigma}^{+} c_{j',\sigma}) = t^2 \Delta_{d}^{CDW}(i',j')$. Via the above identification, the Hamiltonian $H_{t_2}^{MF}$ can be expressed as:

$$H_{t_2}^{MF} = \sum_{(i',j'),\sigma} \Delta_{c}^{CDW}(i',j') c_{i',\sigma}^{+} c_{j',\sigma} + h. c. + 2|\Delta_{c}^{CDW}(i',j')|^2,$$

which leads to $\Delta_{c}^{CDW}(i',j') = -1/2 \sum_{\sigma} \langle c_{i',\sigma}^{+} c_{j',\sigma} \rangle^* \text{ identified above via minimizing the free energy associated with } H_{t_2}^{MF} \text{ with respect to } \Delta_{c}^{CDW}(i',j'). \text{ Note that from above derivations, we indeed find that } |\langle c_{i',\sigma}^{+} c_{j',\sigma} \rangle| \sim t^2 |\langle d_{i',\sigma}^{+} d_{j',\sigma} \rangle| \ll |\langle d_{i',\sigma}^{+} d_{j',\sigma} \rangle|, \text{ as expected.}$

Note also that the CDW order parameters induced on graphene layer shows the opposite sign with respect to that on 1T-TaS$_2$ layer, consistent with the hole-like (particle-like) CDW intensity on graphene (1T-TaS$_2$) layer obtained from DFT calculations, respectively.

Meanwhile, the opposite sign of the proximity induced CDW on the graphene layer with
respect to the CDW on the TaS$_2$ layer as predicted within our mean-field theory is in perfect agreement with the “out-of-phase” relation between CDW patterns observed inside and outside of Mott gap by our STM measurement (see Fig. 2 g-i and corresponding text above). This agreement provides a strong case for CDW proximity in graphene. We emphasize here that the above mechanism based on charge transfer is distinct from all the previously realized proximity effects, including superconducting, magnetic, and spin-orbit proximity effects.

**Conclusions**

In summary, we demonstrate, by STM/STS measurements and DFT calculations, the existence of a proximity induced CDW in graphene. We propose a model based on the short-range exchange interaction between carriers in graphene and the CDW hosted by the TaS$_2$ crystal which qualitatively captures the main features of the CDW proximity effect. Concomitant with the proximity induced CDW in graphene, which is out-of-phase with that in TaS$_2$, we observe a substantial reduction in the Mott gap of the TaS$_2$ crystal, indicating the presence of proximity-induced mid-gap carriers which screen the Mott-Hubbard interaction. These observations open intriguing possibilities for engineering correlations and manipulating charge carriers in heterostructures. They suggest the prospect of non-linear electronic devices based on sliding and pinning of the CDW induced within the graphene layer and low power switches based on controlling correlation gaps through contact. Additional measurements including spin-resolved STM/STS, Raman spectroscopy, and electronic transport will further help elucidate the effects proposed in this work.
Methods

Sample fabrication and STM measurements
Samples were fabricated by mechanical exfoliation of graphene and separately TaS$_2$ flakes inside an argon-filled glovebox. TaS$_2$ flakes were exfoliated from a bulk 1T-TaS$_2$ crystal (2D semiconductor or grown by iodine chemical vapor transport) and transferred onto a passivated SiO$_2$-capped degenerately doped Si wafer. The graphene and 1T-TaS$_2$ flakes were aligned vertically and brought into close contact with micromanipulators under an optical microscope and then heated to promote adhesion. Standard electron beam lithography and electrode deposition (4-5nm Ti/40-50nm Au) were used to make electrical contact to the sample. After removing the PMMA mask the resulting heterostructure was annealed (180–220°C) in hydrogen/argon (10% : 90%) to remove polymer residues followed by AFM tip sweeping. STM and STS were performed using a homebuilt STM at 77 K in high-vacuum < $10^{-5}$ Torr. To locate the micron size samples we employed a technique using the STM tip (mechanically cut Pt/Ir) as a capacitive antenna$^{29}$. Two such samples were measured by STM using several mechanically cut Pt/Ir wire tips. Additionally, we also studied a TaS$_2$ flake transferred to a pre-patterned gold electrode using PDMS polymer. This sample was transferred to the STM system with < 15 mins of exposure to the atmosphere.

Ab initio calculation of band structure
We performed first-principle electronic structure calculations using the projector augmented wave (PAW) approach within the framework of density functional theory (DFT) as implemented in the Vienna ab initio Simulation Package (VASP)$^{36-38}$. The exchange correlation is described in the Perdew-Burke-Ernzerhof (PBE) form of generalized gradient approximation (GGA)$^{39,40}$. To take the strong correlation of Ta d-electrons into consideration, we perform generalized-gradient approximation plus on-site U (GGA+U) calculations with U=2.27eV for
bare, monolayer TaS$_2$ in accordance with previous DFT calculations$^{30,41}$. The lattice structure is theoretically optimized with the atomic forces converged within 0.01 eV/Ang.

Next, we calculated the band structure of a monolayer TaS$_2$ covered with graphene again using a GGA+U scheme (Fig. 2d). To enable this calculation, we considered a twist angle of 13.9 degrees between the graphene and TaS$_2$ layers. This forms a commensurate structure (Fig. 1d) because $5 \times a_G \approx L_{CDW}$, where $a_G=0.246$ nm is the lattice constant of graphene. We note however, that the main features of our results are insensitive to the choice of commensurate structures (Supplementary Figure 8). Furthermore, we chose a reduced value of U=1.7 eV to reproduce the observed spacing between the Hubbard bands.

The calculated DOS of the heterostructure is shown in Fig. 2d and is decomposed into the states from graphene(blue) and TaS$_2$(red) layers. However, we cannot directly compare their sum to the measured dI/dV because the tunneling current has a greater contribution from the graphene than the TaS$_2$ due to the exponential decay of tunneling with tip-sample distance. We therefore calculate a weighted sum (black) of the projected DOS of graphene and TaS$_2$ which takes this into account (see Supplementary Fig. 11 for details). This is in good qualitative agreement with the measured dI/dV curve (Fig. 2b). The transfer of electrons from the graphene layer to the TaS$_2$ layer also explains the shift of $E_F$ from the edge of the LHB in the TaS$_2$ to the edge of the UHB in graphene/TaS$_2$.

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Acknowledgements

NT and EYA acknowledge support from the Department of Energy grant DOE-FG02-99ER45742 and the Gordon and Betty Moore Foundation EPiQS initiative grant GBMF9453; MAA was supported by the National Science Foundation grant EFRI 1433307; GL was supported by Rutgers University; CJW was supported by The National Research Foundation of Korea (NRF), grant No. 2016K1A4A4A019222028 and 2020M3H4A2084417; SWC was supported by the Betty Moore Foundation’s EPiQS grant GBMF6402 and Rutgers University, C.-H. C. was supported by MOST (Grant NO.: 107-2112-M-009-010-MY3, 110-2112-M-A49-018-MY3) and the NCTS of Taiwan, R.O.C., J.H.T acknowledges support from the Ministry of Science and Technology, Taiwan under grant: MOST 109-2112-M-007 -034 -MY3, and from NCHC, CINC-NTU, AS-iMATE-109-13, and CQT-NTHU-MOE, Taiwan.
Supplementary information for “Revealing the Charge Density Wave Proximity Effect in Graphene on 1T-TaS$_2$”

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Sample fabrication and AFM scans

**Supplementary Figure 1: Graphene/TaS2 sample fabrication** (a-d) Schematic showing Graphene/TaS2 sample fabrication using a PMMA based wet transfer method. Steps a-c done in argon-filled glovebox. a) TaS2 exfoliated onto SiO2 chip and Graphene exfoliated onto PMMA membrane. Brought into alignment with micromanipulators in optical microscope setup. b) TaS2 and graphene brought into contact with micromanipulators. c) PMMA membrane melted onto SiO2 chip. d) Gold electrode added using electron beam lithography. (e) Optical microscope image of completed sample. (f) AFM topography scan of sample collected just before loading into the STM.

Samples were prepared in an argon filled glovebox using an optical microscope setup and remote-controlled micromanipulators in a process following previous work[1]. First, graphite was exfoliated onto a polymethyl methacrylate (PMMA) membrane using tape, and an appropriate graphene flake was found using an optical microscope. Likewise, a bulk flake of TaS2 was exfoliated onto a Silicon Dioxide (SiO2) chip and an appropriate flake was found (Appropriate flakes are >20 μm along longest axis). This was done inside an Argon filled glovebox to prevent the oxidation of the TaS2 flake. Using an optical microscope and remote-controlled micromanipulators, graphene and TaS2 flakes were aligned (Supplementary Figure 1a) and brought into direct contact (Supplementary Figure 1b). Using a stage heater, the PMMA membrane was melted (T < 70 C) onto the SiO2 chip (Supplementary Figure 1c), ensuring good contact between the graphene and TaS2 flakes.

Since the Graphene cover protects TaS2 from oxidation, the SiO2 chip with the Graphene/TaS2/PMMA combination was moved out of the glovebox to form a sample contact electrode(50 nm Au/4 nm Ti) using standard electron beam lithography, deposition, and liftoff techniques [1][2] (Supplementary Figure 1d).
Supplementary Figure 2: Residue removal via AFM sweeping. Resist residues (dirt) left on the sample surface post annealing were swept to the edges of the sample using repeated scanning with an AFM tip in contact mode. (a) shows the sample before the sweeping steps and (b) shows the same area after a few contact mode scans.

The final Graphene/TaS$_2$ (G/TaS$_2$) heterostructure goes through a 2-stage cleaning process. First, we follow a standard procedure [1] of annealing the heterostructure in forming gas (10% Hydrogen in Argon) for over 12 hours at $T=230$ C. This removes the bulk of the resist residues. Next, we use an AFM contact mode topography sweeping technique on the Graphene surface [3] to move any dirt and residues to the edges of the graphene surface (Supplementary Figure 2).

Finally, the heterostructure is loaded into a homebuilt STM/STS system [4] [5] and a simple capacitive technique [6] is used to identify and scan the graphene surface.

A bare TaS$_2$ device was also made inside the glovebox using a peel-off process (Supplementary Figure 3a-b). Prepattered electrodes were made on a Silicon wafer using a standard lithography technique. TaS$_2$ was exfoliated on a PDMS piece inside the glovebox. A suitable flake was identified under the optical microscope and aligned with an edge of the prepattered electrode. Upon contact, the TaS$_2$ flake sticks to the rough SiO$_2$ substrate and detaches from the PDMS. Electrical contact is established via the overlap between the TaS$_2$ flake, and the electrode as seen in the optical microscope image (Supplementary Figure 3c). The sample is then loaded onto a sample flag inside the glovebox. It is then taken out of the glovebox, wire bonded, transferred to the STM and pumped down to high vacuum. The total exposure time is <15 min. The sample shows a CCDW pattern in STM topography at 77 K (Supplementary Figure 3d). The surface contamination is due to partial oxidation of the surface after a brief exposure to air during the sample-loading process.
Supplementary Figure 3: Bare TaS$_2$ sample (a) TaS$_2$ exfoliated on PDMS and aligned with the edge of a pre-patterned Au/Ti electrode on a Silicon chip. (b) The TaS$_2$ is brought into contact with the electrode and retracted slowly, leaving the TaS$_2$ flake on the Silicon wafer. (c) Optical microscope image of the sample. (d) Example topography scan measured at Vb=500 mV and I=150 pA at 77K showing a CCDW pattern. The surface contamination is due to partial oxidation of the surface after a brief exposure to air during the sample-loading process.
Band structure of 1T-TaS$_2$

Kohn Anomalies and CDW Supercell

Supplementary Figure 4: Kohn anomaly and CDW supercell (a) Phonon dispersion of undistorted 1T-TaS$_2$. The Kohn anomaly wavevectors are marked by blue circle and yellow rhombus. Different broadening values $\sigma$ indicate the temperature effect. (b) Kohn anomaly wavevectors and Brillouin zone (BZ) of $1 \times 1$ (black) and $\sqrt{13} \times \sqrt{13}$ (red) structures. The blue Kohn anomaly wavevector $q$ along $\Gamma M$ locates at the zone centers of the $\sqrt{13} \times \sqrt{13}$ CDW BZs, whereas the yellow Kohn anomaly wavevector along $\Gamma K$ does not fit the $\sqrt{13} \times \sqrt{13}$ CDW BZs. (c) The $\sqrt{13} \times \sqrt{13}$ CDW BZs are rotated to form the commensurate BZs. (d) Ta atoms vibrate parallel to the Kohn anomaly wavevector $q$, while S atoms vibrate perpendicularly to $q$.

The calculated phonon dispersion for the unreconstructed 1T-TaS$_2$ in Supplementary Figure 3a shows two Kohn anomalies, phonon modes whose frequencies drop to (or below) zero, indicating a lattice instability. These Kohn anomalies can be stabilized without negative frequency by using higher broadening ($\sigma$) values. This corresponds to the removed CDW phase at higher temperatures observed in experiments. The blue Kohn anomaly instability at wavevector $q$ coincides with the wavevector $Q_{\text{CDW}}$ of the $\sqrt{13} \times \sqrt{13}$ CDW reconstruction, suggesting electron-phonon coupling as a possible origin for the CDW. Supplementary Figure 3b demonstrates that the blue Kohn anomaly wavevector $q$ along $\Gamma M$ locates at the zone centers of the $\sqrt{13} \times \sqrt{13}$ CDW BZs. The good match in the periodicity indicates the tendency from 1x1 unit cell toward the $\sqrt{13} \times \sqrt{13}$ CDW lattice. Although the q vector of 1x1 unit cell matches the period of $\sqrt{13} \times \sqrt{13}$ BZ, it is not possible for the 1x1 BZ to match the non-integer $\sqrt{13} \times \sqrt{13}$ BZ. This suggests a $\sim 13.9^\circ$ rotation of the $\sqrt{13} \times \sqrt{13}$ BZ as shown in Supplementary Figure 3c. The rotated $\sqrt{13} \times \sqrt{13}$ supercell thus commensurate with the 1x1 unit cell as observed and discussed in
this work experimentally and theoretically. On the other hand, the yellow Kohn anomaly does not fit the BZ of possible minimum supercells and thus is not observed. The blue soft phonon mode at the Kohn anomaly wavevector $q$ contains mainly longitudinal vibration of Ta atoms with minor transverse vibration of S atoms relative to the phonon propagating direction $q$ as depicted in Supplementary Figure 3d.

**CDW Induced Isolated Flat Band**

$\sqrt{13} \times \sqrt{13}$ TaS$_2$

without

CDW distortion

$\sqrt{13} \times \sqrt{13}$ TaS$_2$

with

CDW distortion

![Lattice relaxation](image)

Supplementary Figure 5: Evolution of band structure due to CDW distortion. Evolution of the GGA band structure (bottom) as the lattice relaxes (from left to right) from the undistorted lattice structure (shown top, left) to the $\sqrt{13} \times \sqrt{13}$ distortion (top, right).

The undistorted 13 Ta atoms within the $\sqrt{13} \times \sqrt{13}$ supercell (top-left panel), the same size as the CDW unit cell (top-right panel), each contribute one conduction electron to the undistorted band structure (bottom-left panel). As the lattice is allowed to relax, 6 of these Ta-associated bands are pushed below the Fermi level while 6 are pushed to higher energies, leaving a single, flat, isolated band at the Fermi level (bottom-right panel). This band is associated with an electron state localized at the center of the Stars of David in the CDW reconstructed structure (top-right panel). As a flat, isolated band, the electronic kinetic energy (and kinetic energy spread) is low, allowing interparticle interactions to dominate and correlated electronic phases can emerge.
In the 1T-TaS$_2$ monolayer, the DFT calculation shows an exchange gap at the Fermi level, even without including a Hubbard U. However, this gap is much smaller than that measured experimentally. To reproduce experimental observations, a Hubbard U is added to the calculation to encompass Coulomb repulsion between electrons in neighboring CDW stars. Increasing the value of U enhances the gap at the Fermi level. The value of U=2.27eV, previously determined from linear response theory, is used in the calculations given in the main text.
CDW, Exchange, and Mott gaps

![Graph showing band structures](image)

**Supplementary Figure 7**: (a) GGA band structure of undistorted 1T-TaS\(_2\) shows no band gap. (b) After relaxing the lattice, a CDW gap opens with an in-gap, half-filled, isolated flat band stuck at \(E_F\). (c) Adding spin splits the isolated flat band into two spin-polarized bands separated by an exchange gap. (d) Increasing the on-site Hubbard \(U\) expands the exchange gap into the experimentally observed Mott gap. (e) With the addition of graphene, the Hubbard \(U\) and the associated Mott gap are reduced in size due to screening from the graphene layer.

The outline of the emergence of the insulating state of 1T-TaS\(_2\) begins with the CDW lattice reconstruction which isolates a narrow, flat band at the Fermi level within the CDW-induced gap. Including spin freedom and exchange interactions, the flat band splits into an exchange gap at the Fermi level. Adding a Hubbard \(U\), enhances the gap into a Mott gap which reproduces experimental observations. Finally, adding a graphene layer on top causes a shift of the 1T-TaS\(_2\) bands toward lower energies and adds a Dirac cone, centered at the K-points of the superstructure Brillouin zone. Additionally, the graphene layer is found to reduce the spacing between the Upper and Lower Hubbard bands of 1T-TaS\(_2\) which is captured by using a smaller value of Hubbard \(U=1.70\text{eV}\).
Band structure of Graphene/1T-TaS2

Comparing DFT Unit Cells

Supplementary Figure 8: a) Top view of G/TaS2 heterostructure with the 5x5 G/$\sqrt{13} \times \sqrt{13}$ TaS2 R13.9° supercell containing 89 atoms with one star used in the DFT calculation (reproduced from main text Fig. 2d). b) DFT calculated charge transfer of the graphene layer based the 5x5 G/$\sqrt{13} \times \sqrt{13}$ TaS2 R13.9° supercell shows a local modulation of doping with the periodicity of the CDW in TaS2. Color scales range from -4 $10^{-4}$e/Å$^2$ (red) to -2 $10^{-4}$e/Å$^2$ (green). c) Out-of-plane displacement of Graphene induced by TaS2 CDW based on the 5x5 G/$\sqrt{13} \times \sqrt{13}$ TaS2 R13.9° supercell. d) Top view of G/TaS2 heterostructure with the 8x8 G/$\sqrt{39} \times \sqrt{39}$ TaS2 R16.5° supercell containing 245 atoms with 3 stars used in the DFT calculation. e) DFT calculated charge transfer of the graphene layer based the 8x8 G/$\sqrt{39} \times \sqrt{39}$ TaS2 R16.5° supercell shows a local modulation of doping with the periodicity the CDW in TaS2. Color scales range from -5 $10^{-4}$e/Å$^2$ (red) to -3 $10^{-4}$e/Å$^2$ (light blue). f) Out-of-plane displacement of Graphene induced by TaS2 CDW based on the 8x8 G/$\sqrt{39} \times \sqrt{39}$ TaS2 R16.5° supercell.

We have repeated the DFT calculations based on the minima 5x5 G/$\sqrt{13} \times \sqrt{13}$ TaS2 R13.9° supercell containing 89 atoms with 1 CDW star as shown in the main text (reproduced here in Supplementary Figure 8a). As shown there, the largest induced local charge transfers and lattice displacements in graphene are located around the center of the TaS2 CDW star (reproduced in S8b and S8c, respectively). Overall, the induced charge modulations and lattice distortions in graphene match well with the TaS2 CDW structure as observed in our STM measurement. Furthermore, we perform the same calculations for an even larger 8x8 G/$\sqrt{39} \times \sqrt{39}$ TaS2 R16.5° supercell containing 245 atoms with 3 TaS2 CDW stars as shown in Supplementary Figure 8d. Fig. S8e illustrates the charge transfer in the graphene layer induced by TaS2 showing a local modulation of doping with the periodicity the same as the CDW in TaS2. The charge modulation obtained from this large supercell not only agrees with our experimental findings but also consists with Fig. 1(c) (reproduced in S8b) based on the minima supercell. Similar displacement behavior can be seen in Fig. S8f that graphene distorts upward around the center of the CDW stars while distorts downward otherwise. We note that the vertical displacement above 0.1pm is generally larger than the horizontal displacement below 0.1pm. We can draw several important conclusions from these results.
With these 2 calculations showing the same trend, we thus are confident that our DFT accuracy is good enough to give a consistent picture of the lattice distortion in graphene as induced by TaS2 CDW. Additionally, as DFT calculations show the same charge modulation and lattice distortion trend in graphene for both the minima 5x5 G/√13 × √13 TaS2, R13.9° supercell and the large 8x8 G/√39 × √39 TaS2 R16.5° supercell, we are confident to conclude that the CDW-shape charge density distribution and lattice distortion in graphene is insensitive to the supercell size adopted in DFT calculations.

Further, we can conclude that the twist angle used in the calculation (13.9°) is not a requirement for the CDW proximity effect observed in the DFT calculation. The fact that the CDW proximity effect is seen in both DFT calculations and STM measurements for multiple twist angles, leads us to believe that the CDW proximity effect is not twist-angle dependent.

**Additional bias dependent topography**

![Supplementary Figure 9: Bias dependent topography scans on Graphene/TaS2 sample in the bias range [-300 mV, +300 mV]. Top row shows the measured dI/dV spectrum and the observed lattice pattern for various bias ranges. The middle row shows the evolution of the topography for negative biases while the bottom row shows the same for positive biases. The CDW pattern appears triangular for all biases outside the mott gap and hexagonal within the band gap. There is a narrow transition regime where it is difficult to differentiate the two. The topography signal is weak for |bias|<90 mV. All scans were recorded while keeping the junction resistance R= 1 giga Ohm.](image)

Since we see the Hubbard bands in the Graphene/1T-TaS2 spectra, it is safe to assume that the Mott gap in the 1T-TaS2 survives when covered with Graphene. If we now set our bias set point inside this gap (\( V_b < (LHB_{center} + LHB_{width}) \approx -272 + 102 = -170 \text{ mV} \)), then there are no states originating from the TaS2 inside the bias window. As a result, the topography signal should only measure the states in the Graphene layer.

For any bias outside this narrow range, there are always TaS2 states available to tunnel into. As a result, the topographic contribution from the Graphene cannot be isolated and substantial direct tunneling into
the TaS2 can coexist. For positive biases, the typical total number of states in the bias window for TaS2 is 40-60 times larger than that in Graphene. Therefore, a large fraction of the total tunneling current goes directly into the TaS2. For negative biases the situation is far better.

We therefore study how the topography changes as a function of bias set-point. These are the main observations:

1. For $V_b < -210$ mV and for $V_b > 0$ mV we see a triangular CDW lattice. This is expected because the bias windows contain one of the Hubbard bands. We know from Partial LDOS calculations and from the experiment that the Hubbard bands have a higher weight near the centers of the stars-of-David compared to the sides resulting in a triangular pattern.

2. For $-150$ mV < $V_b < 0$ we observe a weak hexagonal lattice. A hexagonal lattice can be thought of as the inverse of a triangular lattice. From the argument above, we know that the topography in this bias range only consists of direct tunneling into the Graphene layer.

From the second-order perturbation model as well as from DFT, we showed that the CDW order parameter should be out of phase in the Graphene layer compared to the CDW order parameter in the TaS2 layer. As a result, we can expect a lower density of states in the Graphene directly on top of the SD centers compared to the sides. This would correspond to a hexagonal lattice in topography.

**Chemical potential variation/screening model**

![Graphene Dirac cone and LDOS](image)

**Supplementary Figure 10: Expected effects of screening.** (a) A schematic of the Graphene Dirac cone located on top of the central Ta atom in a SD (right) and away from the central Ta atom (left). The local chemical potential is modulated by the underlying CDW by an amount $U(r)$ reaching a maximum amplitude $\phi$. The excess negative charge density near the central Ta atom locally shifts the Dirac point higher in energy. The hatched areas show the occupied states in a small energy window $eV_b$ below the Fermi level ($E_F$) located near the UHB. There are more occupied states in the Graphene near the center of the SD than away from it. (b) The same is shown as a local density of states plot where the curves represent the Graphene states near the center of the SD (red) and far away from it (blue). The shaded area represents the excess occupied states in a small energy window $eV_b$ below the Fermi level ($E_F$).

The electrons in Graphene can respond to an underlying external potential and partially screen it. This locally changes the chemical potential of Graphene. The charge density wave in 1T-TaS2 should act as a
very weak periodic potential $U(r)$ for the Graphene layer in contact with it. Note that this potential is expected to fall off quickly with distance from the surface because the negative CDW charge is balanced by the positive charge of the TaS2 lattice.

Let’s assume that there is no hybridization between the Graphene and TaS2 bands. The average doping of the Graphene layer (~0.3 eV) is set by the differences in the work function of Graphene and TaS2. The CDW potential should locally modulate this average doping level. The Graphene close to the SD centers should have excess hole doping compared to the Graphene away from the SD center in response to the excess negative charge density there (Supplementary Figure 10a). This would lead to the Dirac point moving up in energy by an amount $\phi$. For a small bias window ($eV_b$) in the Mott gap of TaS2, this leads to an excess of occupied states near the center of the SD (Supplementary Figure 10b). STM topography scans measured at this bias should show a CDW-like pattern in Graphene which peaks on top of the SD center. This should result in a triangular CDW pattern which is in-phase with the TaS2 CDW. This does not match the observed STM topography. This means that local chemical potential variation/screening cannot explain the proximity induced CDW in Graphene. This points to the role played by band hybridization as the dominant mechanism.

**Effects of direct tunneling into TaS2 layer**

![Diagram](image)

Supplementary Figure 11: parallel tunneling into TaS2 layer. (a) Cartoon showing the tunneling junction. The STM tip is at a distance $z_G$ away from the Graphene layer and $z_{TaS2}$ away from the TaS2 layer. The two layers are separated by approximately 0.55 nm. Tunneling current can flow between the tip and both layers but is exponentially suppressed with increasing distance. (b) Extracting the tunneling current decay constant $z_0$ using IZ spectroscopy. The current vs z displacement is plotted on a log-linear plot. The slope of the line is extracted using a linear fit (red line).

STM tunneling current is proportional to the number of available states in the bias window and decays exponentially with z distance [$I(z, V_b) \propto e^{-\frac{z}{z_0}} \int_0^{eV_b} d\epsilon \text{DOS}(\epsilon)$]. For Graphene on TaS2, we can estimate the tunneling current which flows into the Graphene layer and TaS2 layer using this simple picture as
shown in Supplementary Figure 11a. The total tunneling current can be expressed as a sum of these currents as, \( I \approx I_G + I_{TaS2} \) where,

\[
I_G \propto e^{-\frac{z_G}{z_0}} \int_{0}^{eV_b} d\epsilon \text{DOS}_G(\epsilon)
\]

and,

\[
I_{TaS2} \propto e^{-\frac{z_{TaS2}}{z_0}} \int_{0}^{eV_b} d\epsilon \text{DOS}_{TaS2}(\epsilon)
\]

Therefore,

\[
\frac{dI}{dV_b} \propto e^{-\frac{z_{TaS2}}{z_0}} \text{DOS}_{TaS2}(\epsilon) + e^{-\frac{z_G}{z_0}} \text{DOS}_G(\epsilon)
\]

\[
\propto \text{DOS}_{TaS2}(\epsilon) + e^{-\frac{z_{TaS2}-z_G}{z_0}} \text{DOS}_G(\epsilon)
\]

Since the tip is closer to the Graphene compared to the TaS2 by \( z_{TaS2} - z_G \approx 0.55 \) nm, the current flowing into the Graphene is higher by a factor \( \exp \left( \frac{z_{TaS2}}{z_0} - \frac{z_G}{z_0} \right) = \exp \left( \frac{5.56}{1.55} \right) \approx 39 \). Here, \( z_0 = 0.155 \) nm is a function of the tunneling barrier height and was determined experimentally using IZ-spectroscopy (Supplementary Figure 11b).

On the other hand, the total density of states in the TaS2 layer is typically higher than that in Graphene depending on the energy. For instance, according to the calculated DOS, for a bias voltage of -350 mV, there are about 3.19 states in the TaS2 for every state in Graphene. As a result, the net tunneling current flowing into the Graphene layer is about 39/3.19=12.2 times that of TaS2 for this bias. For large bias voltages of either sign, the DOS of TaS2 is significantly larger than that of the Graphene layer. At these bias values, direct tunneling into the TaS2 states cannot be ignored and significantly affects the STM topography scans.

On the other hand, the expected spectra are given by, \( \frac{dI}{dV} \propto \text{DOS}_{TaS2} + 39 * \text{DOS}_{Graphene} \). Note that this scaled sum is significantly different from the naïve direct sum: \( \frac{dI}{dV} \propto \text{DOS}_{TaS2} + \text{DOS}_{Graphene} \). We plot both the direct sum and scaled sum in Main figure 2e. The scaled sum qualitatively matches the experimentally observed spectrum (Main Figure 2f) much better than a direct sum.