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Originally published:

March 2023

ACS Nano 17(2023)6, 5913-5920

DOI: <https://doi.org/10.1021/acsnano.2c12879>

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A van der Waals Heterostructure with an Electronically Textured Moiré Pattern: PtSe₂/PtTe₂

Jingfeng Li,¹ Mahdi Ghorbani-Asl,² Kinga Lasek,¹ Vimukthi Pathirage,¹ Arkady V. Krasheninnikov,^{2,3} Matthias Batzill^{1,*}

¹ Department of Physics, University of South Florida, Tampa, FL 33620, USA

² Helmholtz-Zentrum Dresden-Rossendorf, Institute of Ion Beam Physics and Materials Research, 01328 Dresden, Germany

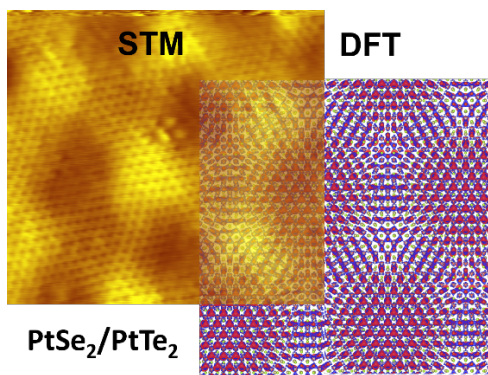
³ Department of Applied Physics, Aalto University, P.O. Box 11100, 00076 Aalto, Finland

* Corresponding author: Matthias Batzill; e-mail: mbatzill@usf.edu

Abstract:

The interlayer interaction in Pt-dichalcogenides strongly affects their electronic structures. The modulations of the interlayer atom-coordination in vertical heterostructures based on these materials are expected to laterally modify these interlayer interactions and thus provide an opportunity to texture the electronic structure. To determine the effects of local variation of the interlayer atom coordination on the electronic structure of PtSe₂, van der Waals heterostructures of PtSe₂ and PtTe₂ have been synthesized by molecular beam epitaxy. The heterostructure forms a coincidence lattice with 13-unit cells of PtSe₂ matching 12-unit cells of PtTe₂, forming a moiré superstructure. The interaction with PtTe₂ reduces the band gap of PtSe₂ monolayers from 1.8 to 0.5 eV. While the band gap is uniform across the moiré unit cell, STS and dI/dV mapping identify gap states that are localized within certain regions of the moiré unit cell. Deep states associated with chalcogen *p_z*-orbitals at binding energies of ~-2 eV also exhibit lateral variation within the moiré unit cell, indicative of varying interlayer chalcogen interactions. Density functional theory calculations indicate that local variations in atom coordination in the moiré unit cell causes variations in the charge transfer from PtTe₂ to PtSe₂ thus affecting the value of the interface dipole. Experimentally this is confirmed by measuring the local work function by field emission resonance spectroscopy, which reveals a large work function modulation of ~0.5 eV within the moiré structure. These results show that the local coordination variation of the chalcogen atoms in the PtSe₂/PtTe₂ van der Waals heterostructure induces a nanoscale electronic structure texture in PtSe₂.

TOC figure:



One of the most exciting features of van der Waals (vdW) heterostructures is the formation of moiré structures due to long-range coincidence lattices formed by the combination of materials with different lattice constants or rotation (twist) angles. Despite weak interlayer interactions, different relative atom positions within the moiré unit cell cause a periodic potential that modifies the electronic structure of the two-dimensional (2D) materials. These moiré patterns in vdW- heterostructures thus allow an additional degree of engineering of materials properties, which has been utilized to demonstrate new phenomena and potential applications. Some of the properties that have emerged due to moiré super-periodicities in vdW heterostructures are: (i) the lateral moiré superstructure gives rise to superlattice Dirac points and consequently Hofstadter butterfly states. This has been observed in h-BN/graphene vdW-heterostructures.^{1,2} (ii) Moiré structures formed by twisting two graphene lattices result in strongly correlated Mott insulator or superconducting states at precisely controlled twist angles,^{3,4,5} and (iii) in semiconducting transition metal dichalcogenide (TMD) heterostructures interlayer interactions induce local modulations of their band gaps^{6,7} that may cause flat bands,^{8,9} quantized confined states,¹⁰ and optical excitations of the so-called moiré excitons.^{11,12} Here we explore another periodic texturing in the electronic properties induced by moiré superlattice in vdW heterostructures in the form of lateral work function modulations. To accomplish strong work function modulations in the moiré structures, the interlayer charge redistribution must depend on local interlayer atom coordination within the moiré unit cell. This has been shown for monolayer materials deposited on crystalline supports such as NaCl,¹³ FeO,^{14,15} or Pb¹⁶- monolayers on silver, platinum, or silicon crystals, respectively. More prominently this effect has been also explored for graphene on Rh¹⁷ or h-BN on various transition metals (e.g. Rh,¹⁸ Ru,¹⁹ Cu,^{20,21} Ir,²²) where the varying coordination of the BN atoms with respect to the surface metals gives rise to strongly variable chemical interactions causing large corrugation of the h-BN layer and consequently modulations of interlayer dipoles and local work function.²³ Subsequently, it was also shown that the work function modulation of such a hex-BN monolayer on a metal substrate can be transferred further to a semiconducting MoSe₂ layer epitaxially grown on the hex-BN/metal substrate.²⁴ In this study we are aiming to establish such electronic modulations in a pure, potentially free-standing vdW system, *i.e.*, averting a 3D-metal substrate. In order to achieve the strongly modulated interlayer interactions that can cause the desired work function modulations, we explore the platinum dichalcogenide system, which has demonstrated extraordinary layer-dependencies of their electronic structure.^{25,26,27,28,29} PtSe₂ and PtTe₂ exhibit a band gap of 1.8 eV and 0.5 eV, respectively, as monolayers,³⁰ but become semi-metals for multilayer samples indicating the strong dependence of their electronic structure on interlayer interactions. This dependence on interlayer interaction makes them promising materials for studying effects of moiré structures on the electronic properties in vdW heterostructures. Moreover, the work function variation within a moiré unit cell is more pronounced for larger unit cells,²¹ which can be achieved by appropriate differences in the lattice constants. The larger lattice constants of tellurides versus selenides of the same transition metal makes their combination into heterostructures ideal for creating such moiré patterns. Here we show that the electronic structure of PtSe₂ monolayer grown on PtTe₂ is modulated by the varying interlayer interaction in the moiré pattern and the work function in PtSe₂/PtTe₂ heterostructure is indeed strongly modulated by close to 0.5 eV within the moiré unit cell. Thus, this demonstrates an electronically textured moiré-pattern in a pure vdW-heterostructure system.

Results and Discussion

PtSe₂ monolayers are synthesized on PtTe₂ ultrathin films by molecular beam epitaxy (MBE). Fig. 1 (a) shows a large-scale STM image of a few layer PtTe₂ film grown on a HOPG substrate. PtTe₂ films on HOPG

exhibits large flat terraces with only terrace steps of ~ 0.4 nm height, corresponding to the interlayer separation between PtTe₂ van der Waals layers. This film is used as a substrate for PtSe₂ growth. Interestingly, PtSe₂ grows in a Stranski-Krastanov-like growth mode, with the first monolayer wetting the surface and subsequent growth resulting in cluster formation, see supplemental information Fig. S1. While such a growth for 2D materials is unusual and its mechanism not well understood for this system, it facilitates the study of monolayer PtSe₂ on PtTe₂ films. Fig. 1(b) shows the deposition of close to one monolayer of PtSe₂ on the PtTe₂ substrate. The surface exhibits the same atomic height steps as for pure PtTe₂, suggesting that the step-structure of the underlying PtTe₂ substrate is not affected by the PtSe₂ growth. The surface now exhibits a moiré pattern whose periodic modulation is observed in the STM images. Figure 1(c) shows a high-resolution STM image of the moiré structure and its Fourier transformation with the corresponding Fourier-filtered images. From Fig. 1(c) the moiré unit cell is determined to consist of 13 PtSe₂ unit cells, which is consistent with a close coincidence lattice of $13 \times a_{\text{PtSe}_2}$ (13×0.38 nm = 4.94 nm) on $12 \times a_{\text{PtTe}_2}$ (12×0.41 nm = 4.92 nm). In addition to this majority moiré structure formed by van der Waals heterostructure of rotationally aligned PtSe₂ and PtTe₂ lattices, a slightly rotated structure is also observed. These two moiré structures are rotated by $\sim 11^\circ$ relative to each other as shown in Fig. 1(d). The rotation angles of the moiré unit cells are amplifying small rotations of the atomic lattices and an 11° rotation of the moiré unit cells corresponds to only a $\sim 1^\circ$ rotation of the atomic lattices (see Fig. S2) so that the grown PtSe₂ monolayer is still roughly aligned with the PtTe₂ substrate. The amplification of the rotation of the moiré structure also enables to identify the aligned from the rotated PtSe₂ monolayer. If the PtSe₂ monolayer is aligned with the PtTe₂ substrate the atomic unit cell of PtSe₂ is also aligned with the moiré unit cell, while if the PtSe₂ layer is rotated by $\sim 1^\circ$ relative to the PtTe₂ substrate than the moiré unit cell is rotated by $\sim 10^\circ$ relative to the PtSe₂ unit cell, which can be observed in atomic resolution images. Generally, the order and long-range coherence of the moiré structure in this PtSe₂/PtTe₂ system is less than in many of the related moiré patterns of monolayer films on metal supports discussed above. One possible reason is the lower synthesis temperature for this system, which is limited to ~ 300 °C. At higher growth temperatures an intermixing of the PtSe₂ with the PtTe₂ substrate is observed. The relatively low thermal stability of these phases has also been reported for pure PtTe₂ which results in easy loss of Te and transformation into different compositional phases upon vacuum annealing.³¹

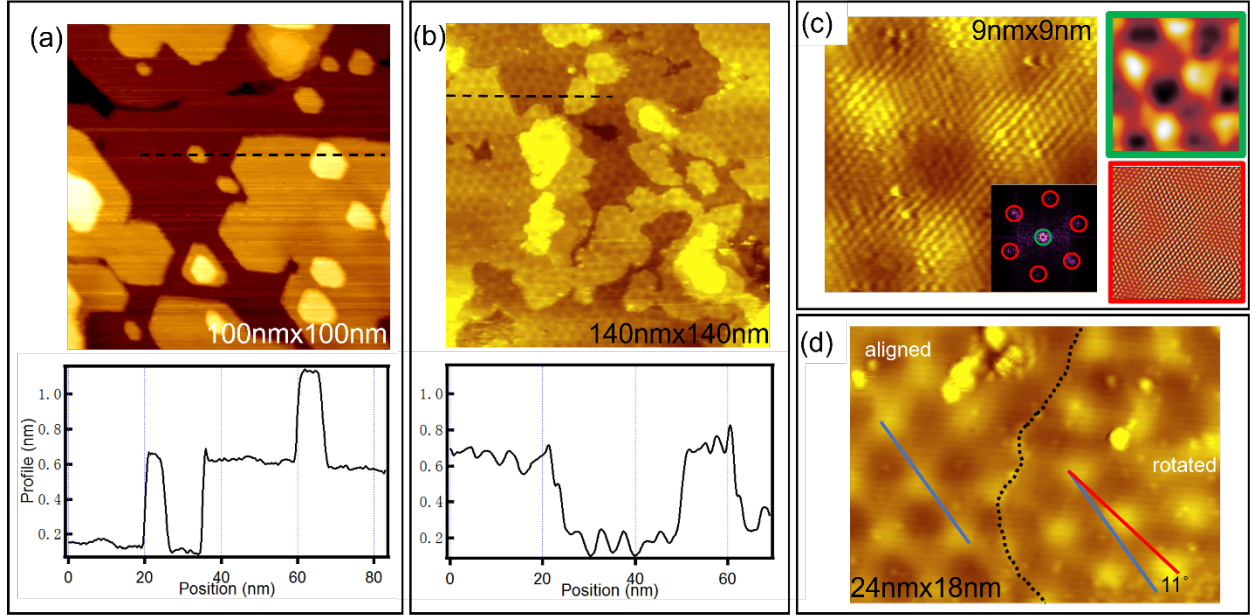


Fig. 1: STM characterization of the morphology of PtSe₂/PtTe₂ heterostructure. (a) PtTe₂ multilayers on HOPG, which serves as the substrate for PtSe₂ growth. The line profile along the dashed line is shown in the bottom panel and shows step heights corresponding to the interlayer separation of PtTe₂. (b) STM image of monolayer PtSe₂ on PtTe₂. The surface is uniformly covered with a moiré structure and the step heights correspond to that of the underlying PtTe₂ substrate. The black dashed lines indicate the location of the line profiles shown in the bottom panel. Tunneling condition $V_{bias}=3V$, $I_t=0.2nA$. (c) High-resolution STM image of the moiré structure and its Fourier transformations (a larger version of the Fourier transformation can be found in Fig. S3). The red and green circles in the Fourier transformation indicate the atomic- and moiré - periodicity, respectively, and the corresponding Fourier-filtered images are shown. Tunneling condition $V_{bias}=0.5V$, $I_t=0.1nA$. (d) Two observed moiré structures, separated by the dashed line. These two moiré structures originate from a small rotation of the PtSe₂ lattice with respect to the PtTe₂ substrate. The red and blue lines indicate the direction of the moiré unit cell, which illustrate the roughly 11° rotation between these two moiré unit cell directions. Tunneling condition $V_{bias}=3V$, $I_t=0.2nA$.

Interface states: PtSe₂ exhibits strong layer-dependent electronic properties. STS measurements for mono-, bi-, and tri-layer PtSe₂ exhibit band gaps of 1.8 eV, 0.6 eV, and 0 eV, respectively, as shown in Fig. 2(a) for PtSe₂ layers grown directly on the HOPG substrate. These extraordinarily strong variations are due to interlayer interactions in the Pt-dichalcogenide family. This raises the question of how the interlayer interactions between a PtSe₂ on a PtTe₂ substrate affect the electronic structure of a monolayer PtSe₂. The interlayer interactions are dominated by the frontier *p*-orbitals of the chalcogen atoms, whose overlap in the vdW gap gives rise to the closing of the band gaps in the pure materials. In the heterostructure interlayer interactions may be dominated by the 4*p*- and 3*p*- orbitals of Te- and Se- atoms, respectively. Thus, the interaction between the different chalcogen atoms and their spatially varying coordination within the moiré structure is anticipated to contribute to local electronic structure variation within the moiré unit cell. STS measurements of the electronic structure of monolayer PtSe₂ on PtTe₂ in different regions of the moiré unit cell are shown in Fig. 2(b). We consider three high symmetry sites of the moiré structure as follow: (i) PtSe₂ is in registry with PtTe₂, *i.e.*, the two structures form a 1T-stacking, and therefore denoted ‘1T-site’, (ii) sites where Se-atoms are on top of Te-atoms, denoted as ‘Se-Te site’, and (iii) Se atoms are over the Pt-sites in the PtTe₂ layer, and thus denoted ‘Se-Pt site’. These three sites

are illustrated in Fig. 2(e). It can be seen that similar to bi-layer PtSe₂, the band gap in monolayer PtSe₂ on PtTe₂ is also narrowed as compared to quasi-freestanding PtSe₂ monolayers grown on a HOPG substrate, but a band gap, similar to that observed for bilayer PtSe₂, of ~ 0.5 eV remains. This indicates that the interlayer interactions between PtSe₂ and PtTe₂ have a similar effect on the gap as in PtSe₂-PtSe₂ contacts. While the band gap narrowing of monolayer PtSe₂ on PtTe₂, compared to free-standing monolayer PtSe₂, is uniform across the moiré unit cell, there are variations in the local density of states (LDOS) within different regions of the moiré unit cell. Electronic states can be identified with a strong variation at -0.8 eV below the Fermi level, as reflected in the dI/dV map. These states at -0.8 eV binding energy are most pronounced at site (i) and absent at site (iii). We demonstrate its localization by comparing the topographic STM image with dI/dV mapping shown in Fig. 2(c) and (d), respectively. Comparison of the aligned with that of the slightly rotated PtSe₂ moiré structure shows that the states at -0.8eV show an even stronger localization at site (ii) in the moiré unit cell (see Fig. S5).

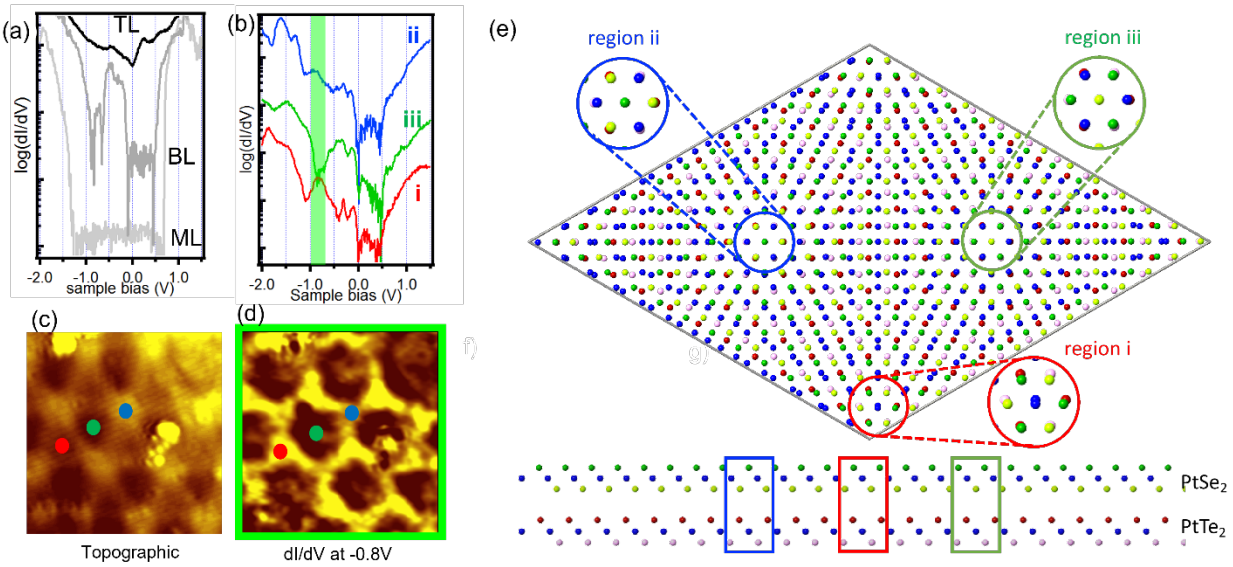


Fig. 2: Electronic structure of PtSe₂. (a) Shows layer dependence of the dI/dV spectra of PtSe₂ grown on a HOPG substrate. STM images of PtSe₂ islands and location of the STS spectra is shown in Fig. S4. The monolayer (ML) exhibits a large band gap of 1.8 eV, the bilayer (BL) has a band gap of 0.6 eV, and the trilayer (TL) is metallic. (b) Selected dI/dV spectra of PtSe₂-monolayer on PtTe₂ substrate at high symmetry sites of the moiré structure (set point for STS: $I=100$ pA, $V=0.6$ V). The dI/dV spectra are plotted on a log-scale, the same spectra in (a) and (b) can be found plotted on a linear scale in Fig. S6. The position at which the color-coded spectra were taken is also indicated in the STM image shown in (c). A dI/dV map taken at -0.8 eV of the same region is shown in (d), highlighting the localized distribution of the electronic state at -0.8 eV. The atomic structure model of the moiré unit cell is presented in (e), a detailed description of the three low symmetry sites can be found in the text.

Electronic states within an energy window for which the free standing PtSe₂ monolayer exhibits a band gap can be attributed to originating from the interlayer interaction with the PtTe₂ substrate. In addition to these ‘shallow’ states, we also observe lateral modulations of electronic states further away from the Fermi-level. These deep states are shown in Fig. 3 for a region that shows both moiré structures due to a PtSe₂ domain that is aligned with PtTe₂ and a domain with a slight rotation of the PtSe₂ layer with respect to PtTe₂. Electronic states at -1.6 eV and -2 eV are clearly discerned in the dI/dV spectroscopy. Based on

previous calculations, these states are associated with $p_{x,y}$ and p_z chalcogen orbitals, respectively.³² Modulations of these states are observed for both moiré structures but are more pronounced in the domains where the PtSe₂ is rotated with respect to the PtTe₂. The p_z state at -2 eV is observed in regions i and iii of the aligned moiré but is suppressed in region ii. This suggests that the states associated with p_z orbitals are altered if the chalcogen atoms in the two layers are on top of each other. In contrast, the states at -1.6 eV are associated with $p_{x,y}$ orbitals and those are less affected in the moiré structure. Interestingly the p_z states have been associated with a spin texture of PtSe₂ monolayers³² and thus their modulation in the moiré structure implies a local modulation also of this spin texture.

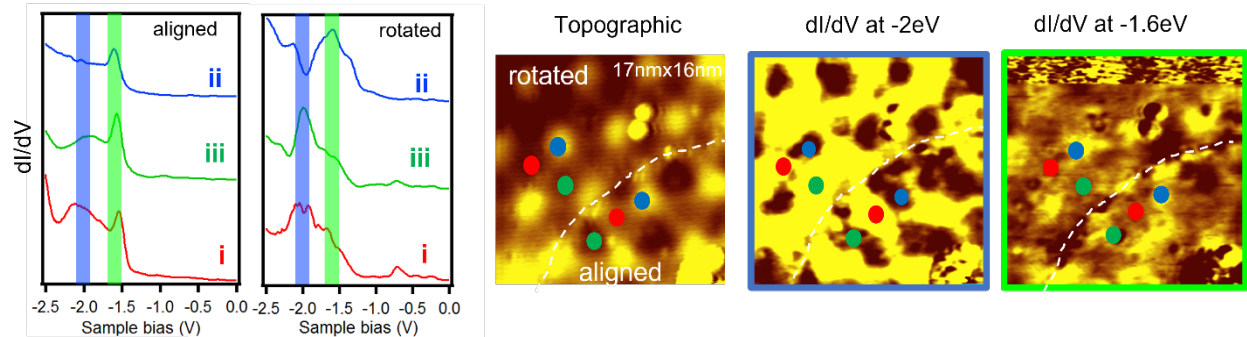


Fig. 3: dI/dV spectroscopy of deep states for different positions in the two moiré structures (set point for STS: $I=100\text{pA}$, 1V). For modulations of shallow states in the rotated domains, detailed spectra are shown in Fig. S5. Two main states can be identified at -1.6 eV and -2 eV, which may be attributed to $p_{x,y}$ and p_z derived selenium states, respectively. The STM image (tunneling condition: $V_{\text{bias}}=3\text{V}$, $I_t=0.2\text{nA}$) shows both aligned and rotated moiré domains. The domain boundary is indicated by the dashed line. The state at -2eV exhibits a stronger localization within regions i and iii, while it is absent in region ii. The state at -1.6 eV is less modulated within the moiré structure, supporting the assignment to $p_{x,y}$ derived states.

Work function: Determining work function variations in nanoscale structures by field emission resonance (FER) spectroscopy has been widely used in scanning tunneling microscopy.^{13,14,15,16,17,18,19,20,21,22,23,24,33,34,35} Briefly, in FER the bias voltage between an STM tip and the sample surface is chosen such that the Fermi level of the STM tip lies above the vacuum level of the sample. In this Fowler-Nordheim tunneling regime, the drop of the potential in the tunnel junction causes a potential well between the tip and the sample surface. The quantum confined image potential states in this quantum well are probed in FER spectroscopy. Local changes in the substrate work function modify the image potential states and thus a shift of the resonance states is a direct measure of the work function modulations along the surface. It is generally challenging to determine absolute values of the work function because the STM tip properties are not well-defined, but for a stable tip the relative changes for different substrate sites are given by the shift of the resonance states.

Figure 4 shows FERs at different positions along a line across the moiré unit cell. All FERs, apart from the lowest energy one, which is known to be unreliable,^{20,23,24,Error! Bookmark not defined.} shift approximately by the same amount. Reported evidence suggests that the second FER is the most relevant for measuring work function variations,^{Error! Bookmark not defined.,36} and therefore the reported values refer to that of the 2nd FER. The largest work function is observed for site ii which is 0.48 eV larger than for site iii. Site i is in between, with a work function 0.28 eV lower than site ii. Slightly less work function modulations are observed for the rotated moiré structure as shown in Fig. S7. This is a substantial work function difference and the first

reported for a transition metal dichalcogenide vdW heterostructure. Strong modulations of the work function imply surface dipoles that must originate from interlayer charge transfer in the PtSe₂/PtTe₂ heterostructure. To gain further insight in the experimentally observed work function modulations, density functional theory (DFT) calculation of the local electronic structure and charge transfer within the moiré structure have been performed, which is discussed next.

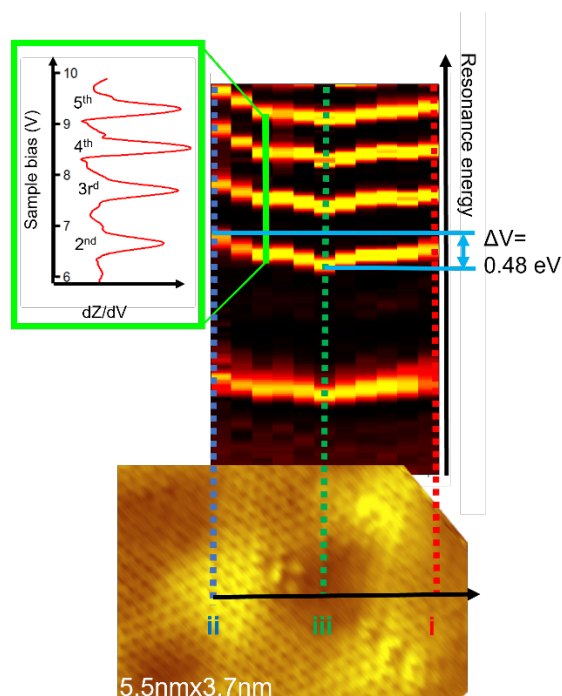


Fig. 4: Work function modulation within moiré unit cell measured by field emission resonance (FER) spectroscopy. FER spectra are measured along the moiré unit cell from regions ii-to-iii-to-i. One spectrum is shown as an example and the others are plotted as an intensity map to illustrate the variation of the resonances as a function of the position. The largest resonance energy difference is observed between regions ii and iii and is measured to be 0.48 eV. The strong variations of the FER at different sites of the moiré unit cell demonstrate the work function modulation within the moiré unit cell. The imaging conditions for the STM data are: $V_{bias}=0.5V$, $I_t=0.1nA$.

PtSe₂/PtTe₂ moiré structure and electronic properties from DFT calculations: To obtain deeper insights into the atomic structure of the interface, we performed extensive DFT calculations using an unsupported PtSe₂/PtTe₂ supercell as a model heterostructure, see the Methods section for detail. A supercell with a single PtTe₂ layer, rather than multilayers as in the experiments, is necessitated by the computational limitations imposed by the large number of heavy atoms in the structure. Test simulations on a smaller unit cell with rotated PtSe₂/PtTe₂ layers show that the band gap of monolayer PtSe₂ is not changed by the number of supporting PtTe₂ layers (Fig. S8). This illustrates that the monolayer PtTe₂ can be a reasonable model system to qualitatively describe the behavior in the experimental system. The computed relaxed crystal structure exhibits only very weak topographical buckling of the bilayer structure, implying that the observed apparent topography in STM images is likely associated with an electronic contrast, however, this needs to be verified by structural probes.

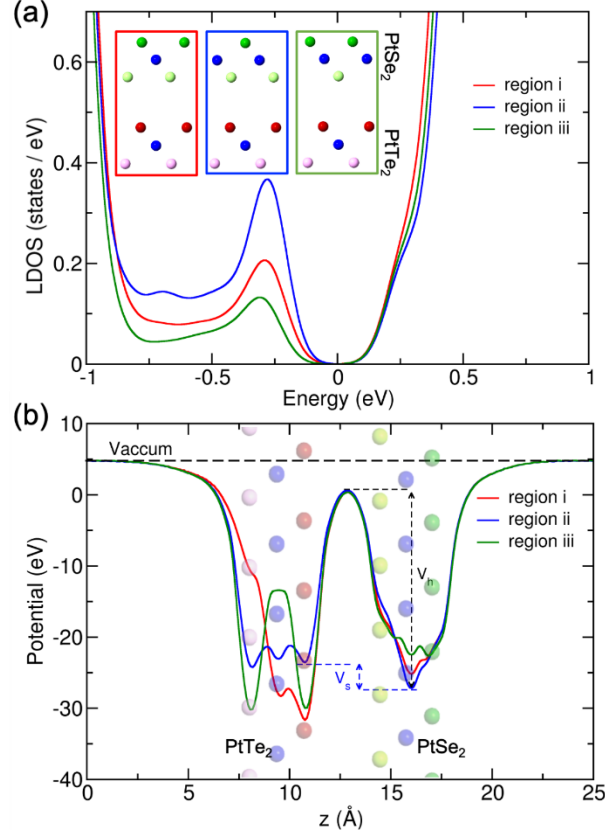


Fig. 5: (a) Local density of states for different regions in the moiré structures of the $\text{PtSe}_2/\text{PtTe}_2$ heterostructure. The LDOS is shown for PtSe_2 . (b) Electrostatic potential along the direction perpendicular to the heterostructure and averaged within planes parallel to the heterostructure.

Depending on the relative positions of the interface Se atoms in the PtSe_2 with respect to the interface Te atoms in the PtTe_2 monolayer, see Figure 2 (e), the Se-Te interatomic distances and angles are different in different regions of the moiré pattern. As a result, the local interactions between the layers is different giving rise to the variation of the local electronic structure. Figure 5 (a) shows LDOS at three moiré sites in the heterostructure. The calculated band gap edge energies of the $\text{PtSe}_2/\text{PtTe}_2$ heterostructure are almost unchanged for the different sites within the moiré structure with only small variations of less than 30 meV (see Fig. S9), and thus the ‘local’ band gap remains nearly constant within the moiré unit cell (note that the calculated band gap energy depends sensitively on inclusion of spin-orbit coupling in the calculations, see Table S1), the LDOS below the Fermi level (at about -0.3 eV) is larger in region ii, as compared to the other sites. The constant band gap is consistent with the experimental STS data. Also, the variation of the LDOS at the band edge is consistent with the STM apparent topographic appearance (see Fig. 2(c)) of the heterostructure indicating higher tunneling contrast in region ii than in the other two moiré regions. We further analyzed the modulation of the averaged electrostatic profile perpendicular to the heterostructure at different sites in the moiré structure, Figure 5 (b), where the differences are evident. The variation of the electrostatic potential at different sites is caused by the amount of charge transfer at the specific interface area. Our results indicate that the potential height V_h at the PtSe_2 layer is larger in region ii than in the other two regions. This is consistent with the calculated Bader charges

transferred from PtTe₂ to PtSe₂, which were found to be 4×10^{-3} , 7×10^{-3} and 2×10^{-3} electrons/atoms for regions i, ii and iii, respectively, see Figure 6. The charge transfer from PtTe₂ to PtSe₂ is related to the difference in the work functions, which can be described by the relation $Q \propto \Delta\varphi$, where Q represents the charge, and $\Delta\varphi$ is the difference in the work functions between the two monolayers, respectively.³⁷ These charge transfers can be associated with the experimentally measured work function modulations. Since the native PtSe₂ bilayer does not exhibit such work function modulations, such behavior in the heterostructure can be attributed solely to charge transfer-induced dipoles at the PtSe₂/PtTe₂ interface.

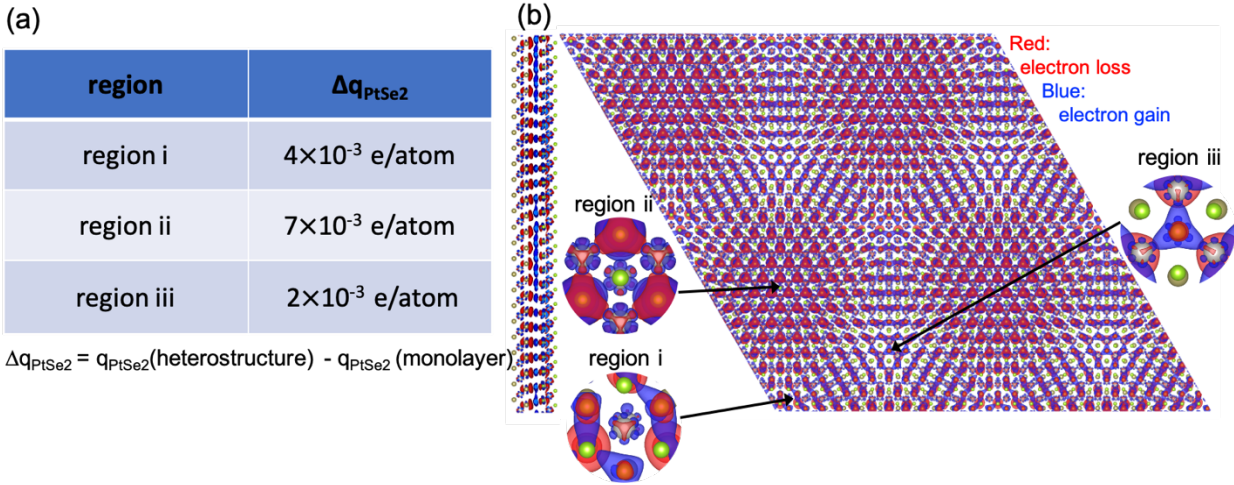


Fig. 6: (a) Average Bader charges at each region in the PtSe₂/PtTe₂ heterostructure with PtSe₂ being on top, as compared to the isolated PtSe₂ monolayer. (b) The charge transfer between PtSe₂ and PtTe₂ layers calculated as a difference between the charge in the heterostructure and the isolated monolayers. Blue and red represent charge accumulation and charge depletion. Note the variation of charge in different areas of the moiré pattern.

Conclusion

In conclusion, we have shown that the PtSe₂/PtTe₂ vdW heterostructure is a strongly electronically corrugated moiré system, as evident from the experimental STM/STS data. Even with relatively weak interlayer interactions in a pure vdW bilayer the varying interlayer atom configuration within the moiré unit cell have pronounced effects on the local electronic structure. STS show that the atom coordination between the two materials causes interlayer states that vary within the moiré structure, and their localization is imaged by dI/dV mapping. The varying interactions of the chalcogen *p*-orbitals in the PtSe₂/PtTe₂ moiré unit cell are giving rise to varying interface dipoles and thus different local work functions. This has been experimentally determined by FER spectroscopy and work function shifts close to 0.5 eV have been observed for the different local stacking configurations. The strong local modulation of the work function implies in-plane dipoles to permit the electrostatic potential offsets. Surface dipoles and work function variations have been shown to facilitate the alignment of molecular adsorbates on hex-BN/metal substrates,^{38,39,40,41,42} and the control over the adsorbate charge transfer within the moiré structure,^{43,44} and similar effects can be anticipated for this purely vdW heterostructure. Pt-

dichalcogenides have also shown promise for hydrogen evolution and oxygen reduction electrocatalysis.^{45,46,47} Work function engineering by the formation of vdW heterostructures is an approach to tune their surface chemical properties, and thus vdW heterostructures may be anticipated to exhibit varying and intriguing electrocatalytic properties as compared to their individual components. Moreover, the Pt-chalcogenides are known for their spin textures^{32,48} and magnetic properties^{49,50} and the interaction of the local spins with the periodic moiré -dipoles may give rise to magnetic properties modulations within the moiré structure. Moreover, STS studies have detected variation of p_z -orbitals, which have been associated with spin texture in Pt-dichalcogenides.³² This may indicate a lateral modulation of such spin textures within the moiré unit cell. Local magnetic properties may be studied by spin-resolved STS in the future.

Methods

Experimental: PtTe₂ and PtSe₂ are grown in dedicated tellurium and selenium growth chambers by co-deposition of Pt and chalcogens. First a PtTe₂ multilayer film is grown at 300 °C substrate temperature on a freshly cleaved and vacuum-annealed HOPG substrate.^{31, 51} Subsequently, a monolayer of PtSe₂ is grown on top of the PtTe₂ at growth temperature of 300 °C. For both compounds, Pt was evaporated in an e-beam evaporator from a 2mm solid Pt-rod. Tellurium was supplied from a Knudsen cell and selenium from a hot-wall valved cracker source. In either case, the chalcogen to Pt-flux ratio exceeded a 10:1 ratio. The prepared films were characterized in a closed-cycle low-temperature STM with a base temperature close to 20 K. The bias voltage was applied to the sample, so the unoccupied and occupied states were recorded under positive and negative bias, respectively. dI/dV spectra were recorded with a lock-in amplifier with a 30 mV modulation voltage and the feedback-loop turned off. The FER spectra were obtained from recording Z-V, with the feedback-loop on, as a function of applied bias voltage and dZ/dV was obtained by calculating the derivative numerically.

Computational: Density functional theory (DFT) calculations were performed using Vienna Ab initio Simulation Package, within the plane-wave projector augmented-wave (PAW) method.^{52,53} The exchange-correlation functional was employed in the generalized gradient approximation of Perdew-Burke-Ernzerhof.⁵⁴ An energy cutoff of 400 eV was set for plane-wave expansion of the supercell calculations. van der Waals interactions were taken into account using the many-body dispersion combined with fractional ionic atoms method.⁵⁵ The effect of spin-orbit coupling is included in the electronic structure calculations. The Brillouin zone of the supercells was sampled using Gamma-point approximation. The charge densities were illustrated using the VESTA package.⁵⁶ The PtSe₂/PtTe₂ heterostructure was constructed by using an interface consisting of 13 × 13 unit cells of PtSe₂ and 12 × 12 unit cells of PtTe₂ monolayer corresponding to a lattice mismatch of only 0.9%. The supercell has a lattice constant of $a=b=48.65 \text{ \AA}$ including 939 atoms.

Acknowledgement: Financial support from the National Science Foundation under award 2140038 is acknowledged. A.V.K. thanks the German Research Foundation (DFG) for the support through Project KR 4866/6-1 and the collaborative research center “Chemistry of Synthetic 2D Materials” SFB-1415-417590517. The authors further thank the HZDR Computing Center, HLRS, Stuttgart, Germany, and TU Dresden Cluster “Taurus” for generous grants of CPU time.

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