

Supplementary Information for

**Local interface effects modulate global charge order
and optical properties of $1T$ -TaS₂/ $1H$ -WSe₂
heterostructures**

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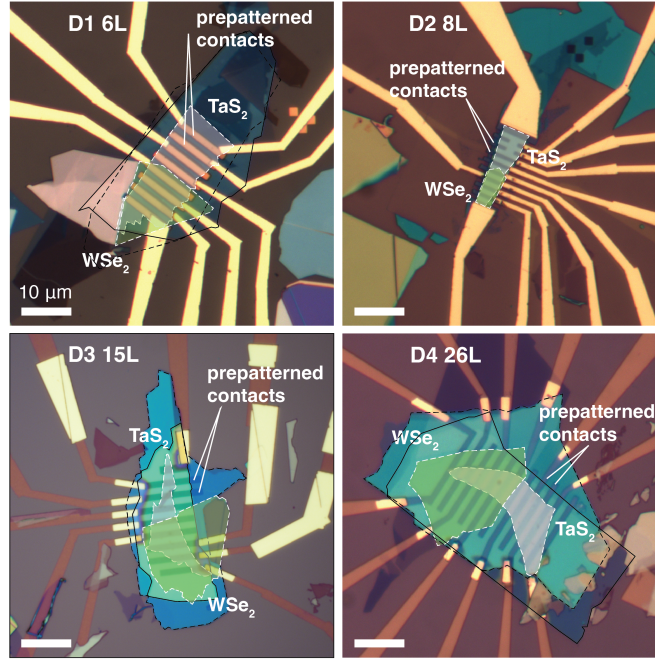
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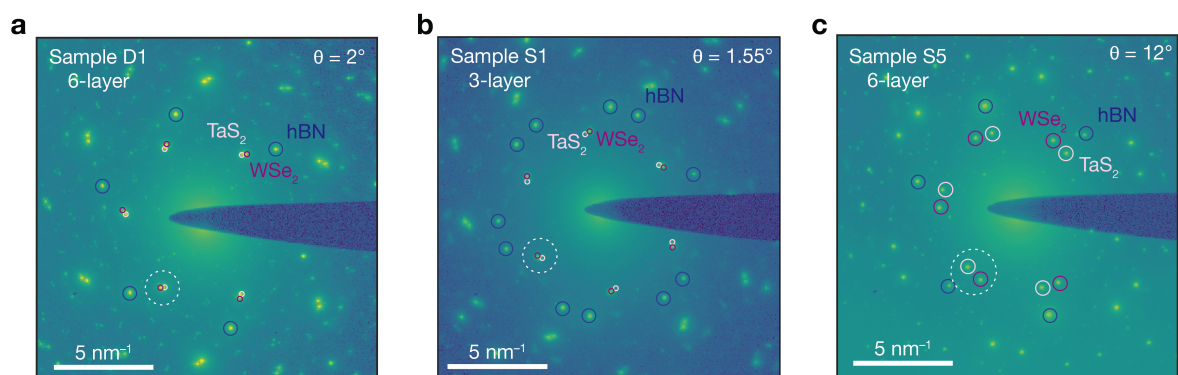
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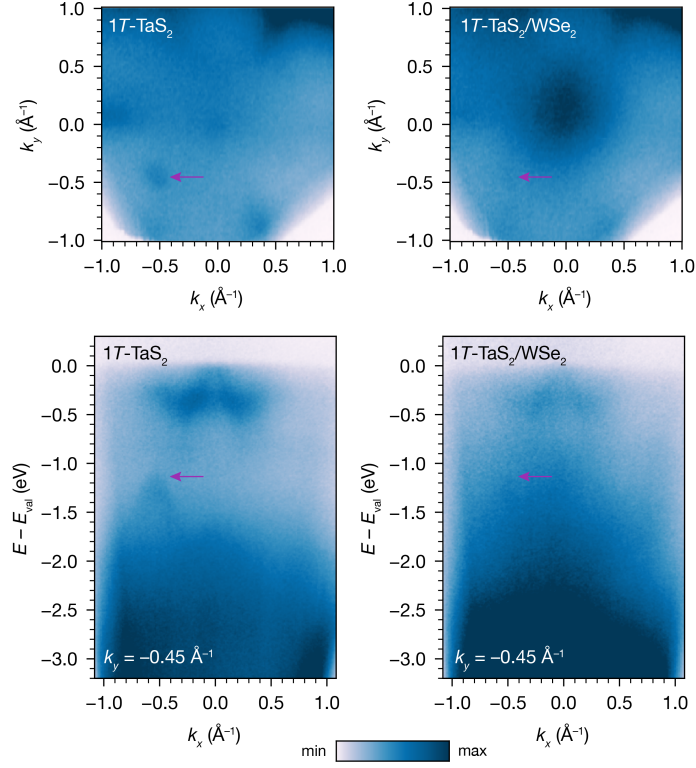
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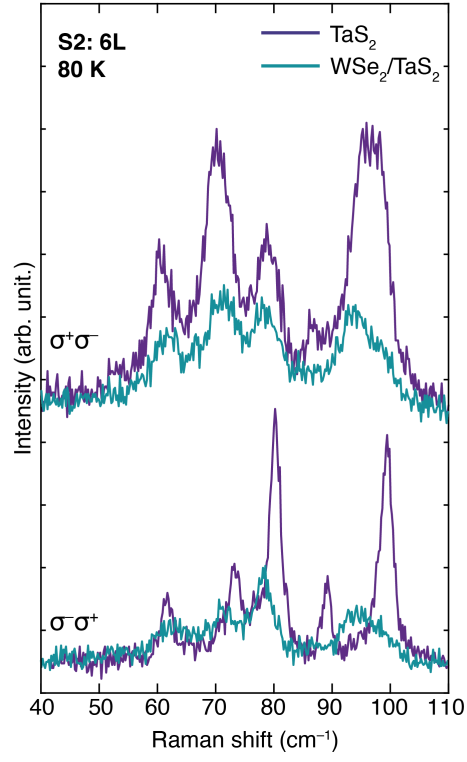
Supplementary Figure 1. Optical micrographs of the measured devices. Images of devices D1–D4 are shown. $1T$ –TaS₂ and $1H$ –WSe₂ flakes are false-colored white and green, respectively, and $1T$ –TaS₂ and $1H$ –WSe₂ flake outlines are marked with white dashed lines. Bottom and top hBN flake outlines are marked with dashed and solid black lines, respectively. Two prepatterned contacts on each device are indicated with white lines.



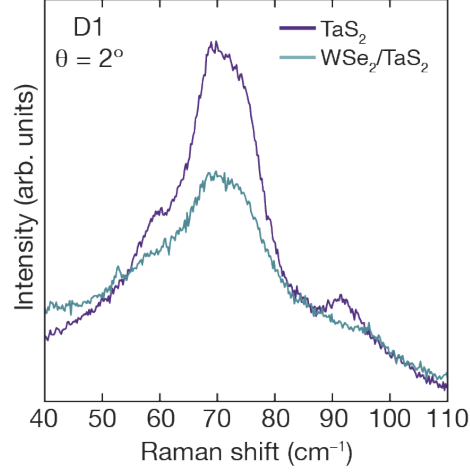
Supplementary Figure 2. Determining interlayer alignment of $1T$ -TaS₂ and $1H$ -WSe₂ using TEM diffraction. (a–c) TEM diffraction of samples D1 (a), S1(b) and S5 (c).



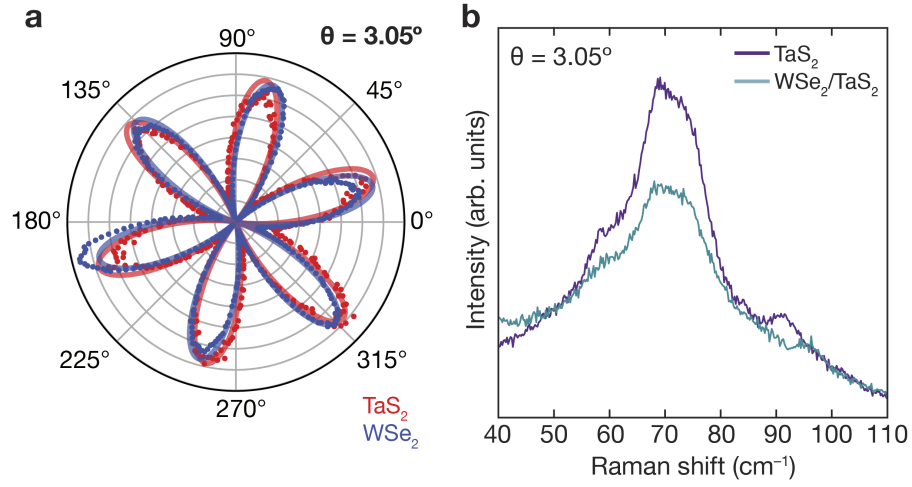
Supplementary Figure 3. Potential evidence of band folding observed in ARPES data. Top panels: Isoenergy cuts for regions R1 (standalone 1T-TaS₂) and R2 (heterostructure region), integrated over a 0.1 eV energy range. Bottom panels: Corresponding ARPES band dispersions centered at $k_y = -0.45$ eV, integrated from $k_y = -0.40$ eV to $k_y = -0.50$ eV. Arrows indicate possible CDW-related band-folding features.



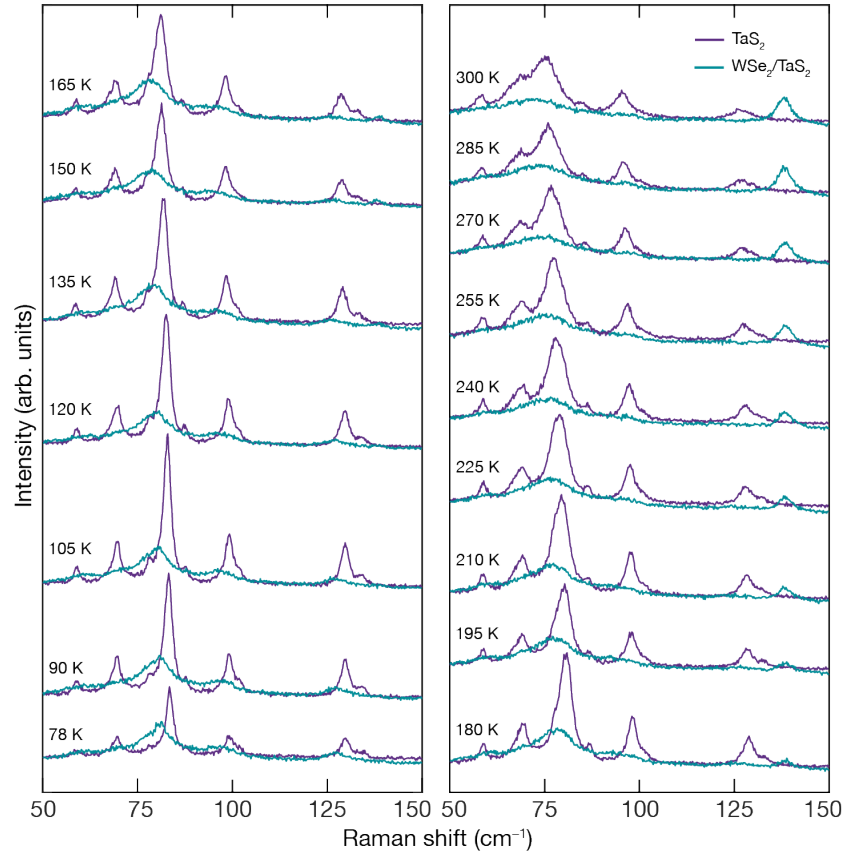
Supplementary Figure 4. Polarized Raman spectra of sample S2. Raman spectra in the cross-polarized configuration for sample S2 reveal modes with 1E_g symmetry. These results are consistent with preceding literature[1].



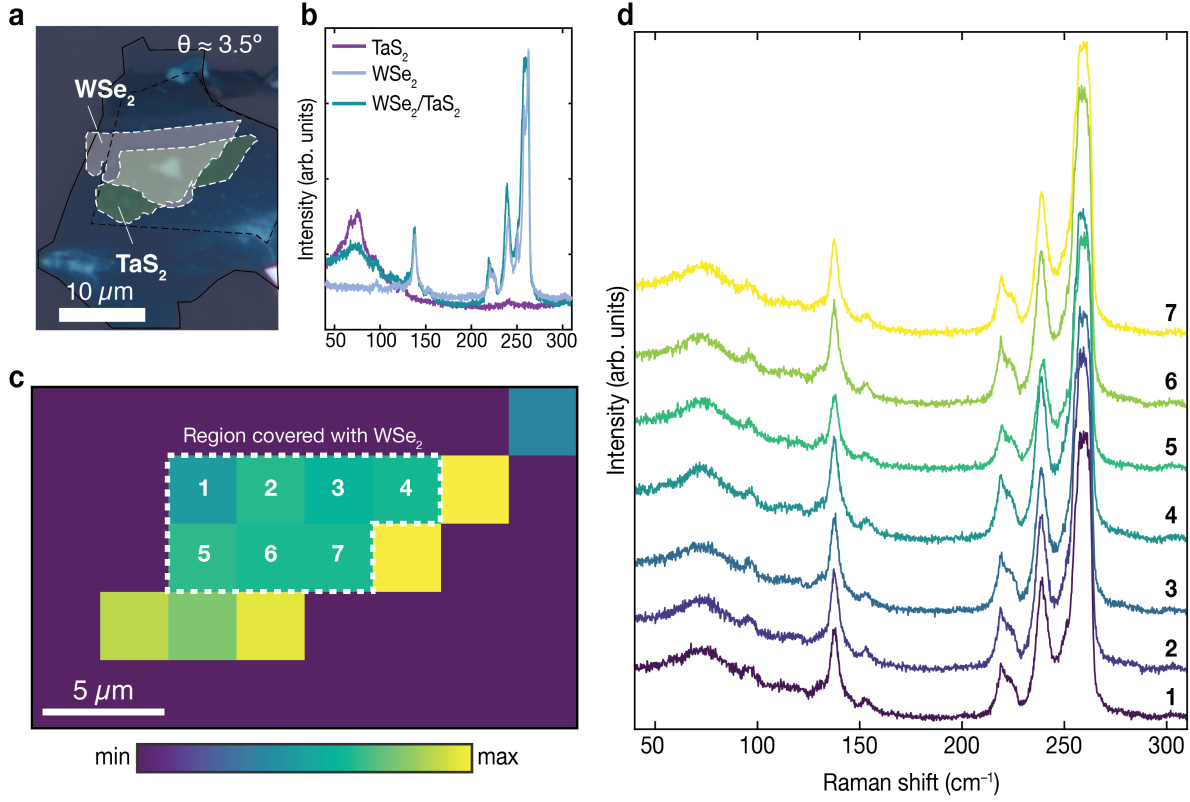
Supplementary Figure 5. Raman spectra of device D1. Raman spectra were measured for two distinct regions of device D1 fabricated from an nearly aligned heterostructure of 6-layer $1T$ -TaS₂ and monolayer $1H$ -WSe₂. Azimuthal alignment, derived from TEM diffraction, is denoted.



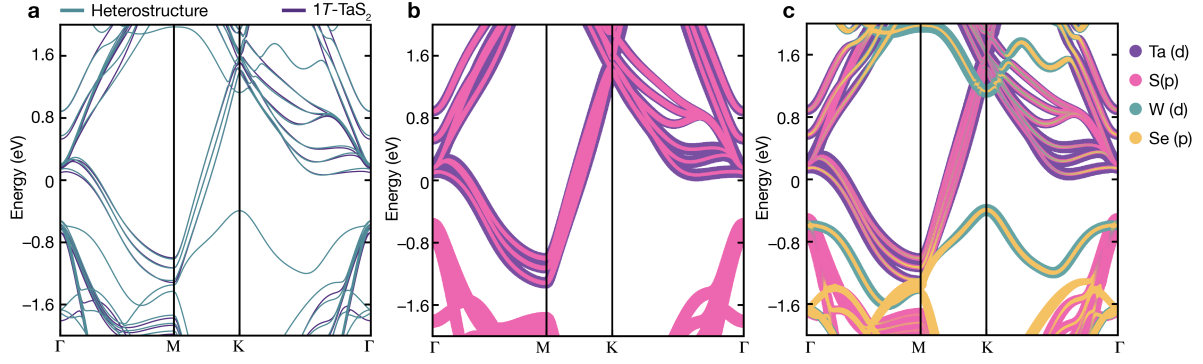
Supplementary Figure 6. CDW suppression in an nearly aligned heterostructure of 5-layer $1T$ - TaS_2 interfaced with $1H$ - WSe_2 . **(a)** Second-harmonic generation (SHG) measurement of the azimuthal alignment between $1T$ - TaS_2 and $1H$ - WSe_2 . **(b)** Raman spectra measured for the heterostructure and the $1T$ - TaS_2 -only regions.



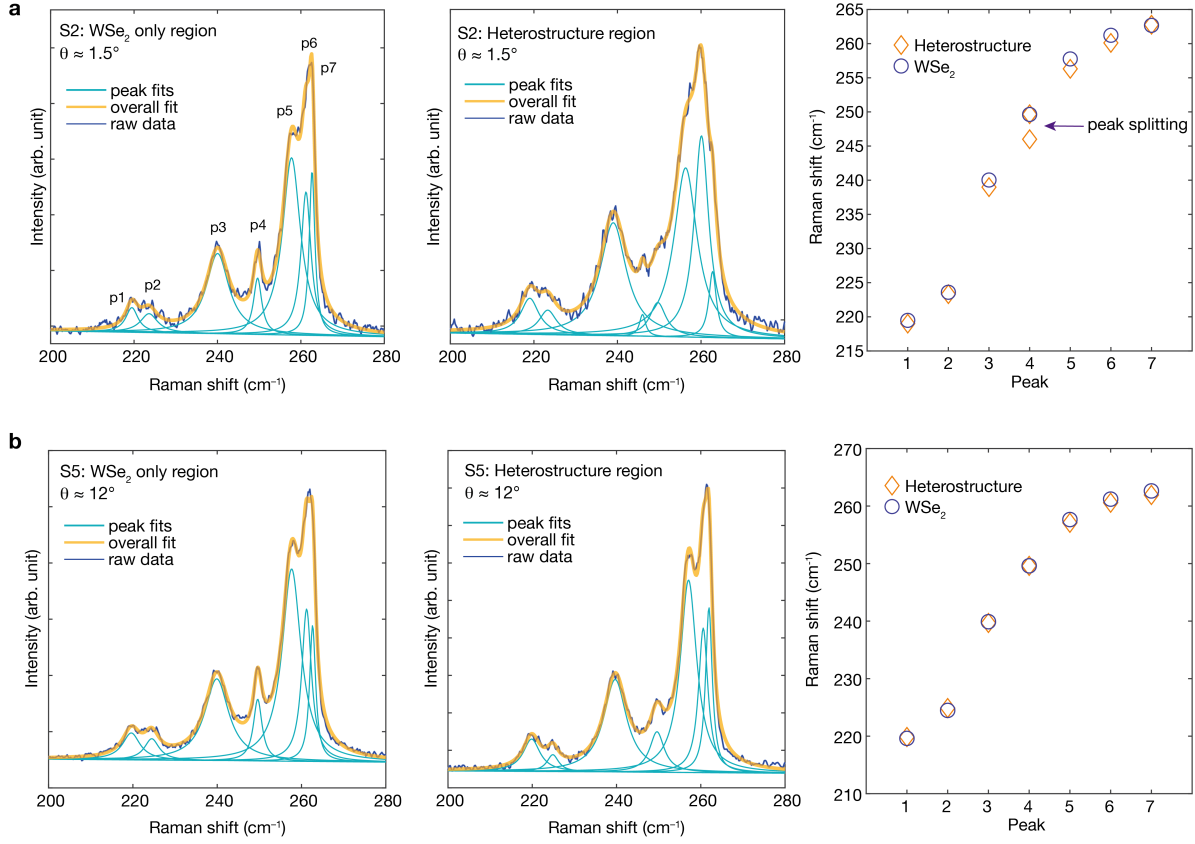
Supplementary Figure 7. Temperature-dependent Raman spectra for sample S1. Raman spectra were measured between 78 K and 300 K on two distinct regions of sample S1 fabricated from an nearly aligned heterostructure of 3-layer 1*T*-TaS₂ and monolayer 1*H*-WSe₂.



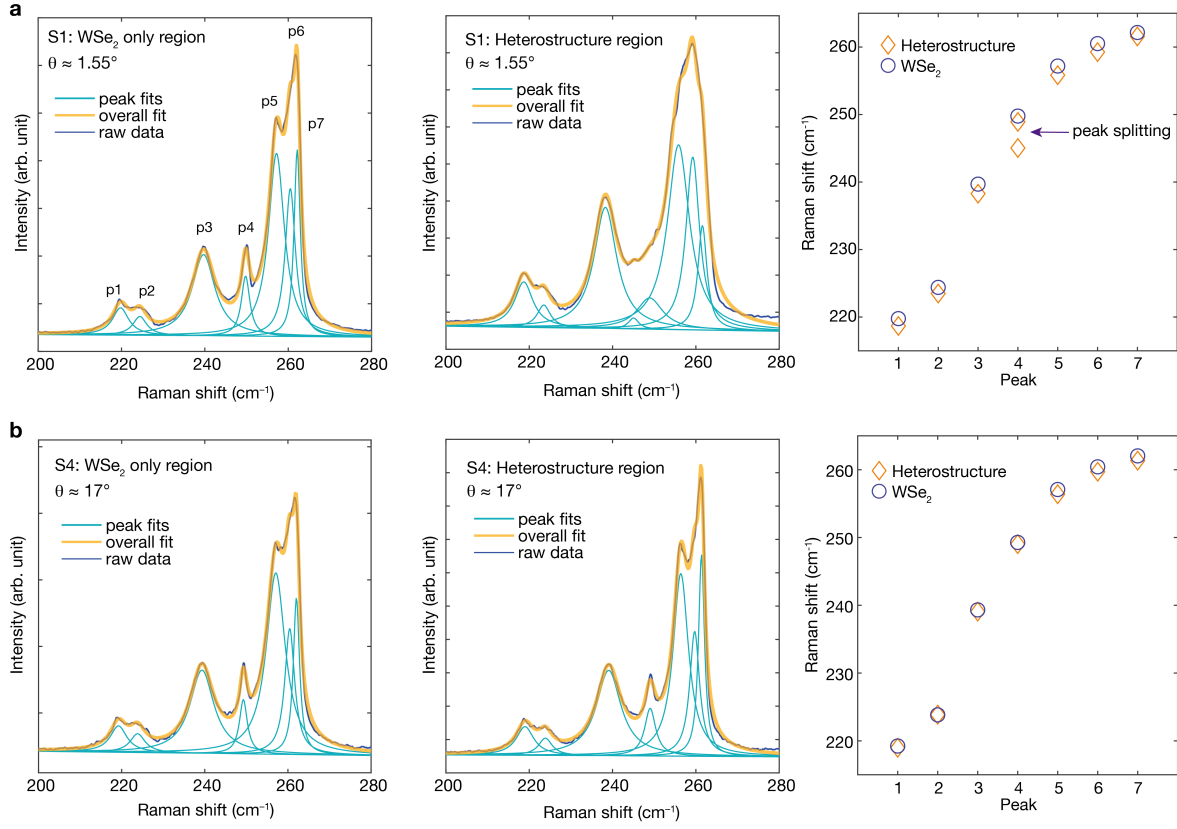
Supplementary Figure 8. Raman mapping of heterostructures. **(a)** Optical micrograph of the hBN/ $1H$ -WSe₂/3-layer $1T$ -TaS₂/hBN heterostructure. $1T$ -TaS₂ and $1H$ -WSe₂ flakes are false-colored white and green, respectively, and $1T$ -TaS₂ and $1H$ -WSe₂ flake outlines are marked with white dashed lines. Bottom and top hBN flake outlines are marked with dashed and solid black lines, respectively. **(b)** Representative Raman spectra from the standalone $1T$ -TaS₂, standalone $1H$ -WSe₂, and the heterostructure regions. **(c)** Raman intensity integrated between 50 and 100 cm⁻¹ across the heterostructure. The $1H$ -WSe₂-covered region is outlined with white dashed lines. **(d)** Raman spectra measured in the heterostructure region, with spectrum numbers corresponding to the positions labeled in (c).



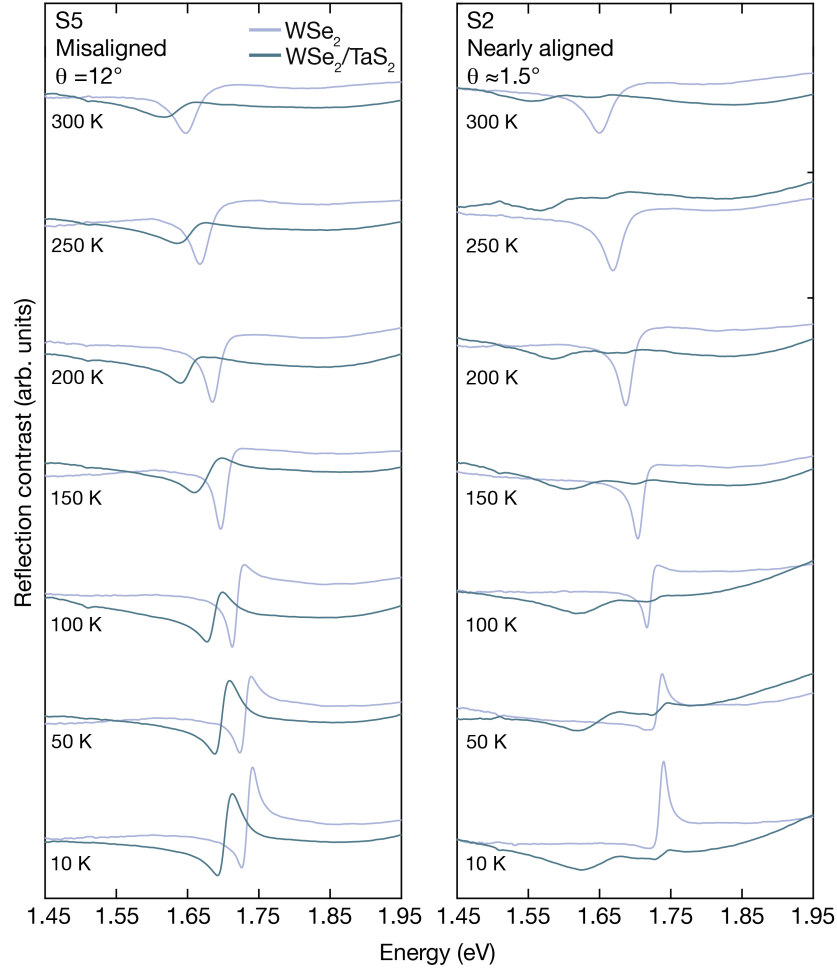
Supplementary Figure 9. Band structure calculations of the high-symmetry nearly aligned heterostructure and standalone $1T\text{-TaS}_2$. **(a)** Overlaid band structures of the high-symmetry 3-layer $1T\text{-TaS}_2$ and its heterostructure with monolayer $1H\text{-WSe}_2$. **(b,c)** Orbital-projected band structures of standalone high-symmetry 3-layer $1T\text{-TaS}_2$ **(b)** and its heterostructure with monolayer $1H\text{-WSe}_2$ **(c)**.



Supplementary Figure 10. Lorentzian fits for S2 and S5. **(a,b)** Lorentzian fits for the standalone $1H$ -WSe₂ and heterostructure regions, along with summarized peak positions for **(a)** S2, the nearly aligned sample, and **(b)** S5, the misaligned sample, each comprising 6-layer $1T$ -TaS₂ and monolayer $1H$ -WSe₂.



Supplementary Figure 11. Lorentzian fits for S1 and S4. **(a,b)** Lorentzian fits for the standalone $1H\text{-WSe}_2$ and heterostructure regions, along with summarized peak positions for **(a)** S1, the nearly aligned sample, and **(b)** S4, the misaligned sample, each comprising 3-layer $1T\text{-TaS}_2$ and monolayer $1H\text{-WSe}_2$.



Supplementary Figure 12. Temperature-dependent reflection contrast (RC). Temperature-dependent RC for the nearly aligned sample S2 and the misaligned sample S5, comprising 6-layer $1T$ -TaS₂ and monolayer $1H$ -WSe₂.

Supplementary Note 1: Quantifying RC broadening

To identify excitonic resonances of $1H$ -WSe₂, the experimentally obtained reflectance spectrum from the sample region (R_{sample}) and the substrate region ($R_{\text{substrate}}$) is used to calculate the reflection contrast according to the following formula:

$$RC = \frac{R_{\text{sample}} - R_{\text{substrate}}}{R_{\text{substrate}}} \quad (1)$$

RC provides the change in reflectance of the sample relative to the substrate and, on transparent substrates, is approximately proportional to the optical absorption, particularly when the RC is close to or below unity. For the multilayered stacks used here, RC features are strongly influenced by interference effects. To account for this, we use a transfer matrix model (TMM) under normal incidence [2] to model light propagation and interference at each interface across the layered structure in both the sample and substrate regions. Here, the dielectric function of monolayer $1H$ -WSe₂ near the excitonic resonance is approximated using a Lorentzian oscillator model [3]:

$$\varepsilon(E) = \varepsilon_b + \frac{A}{(E_0^2 - E^2 - iE\Gamma_{nr})} \quad (2)$$

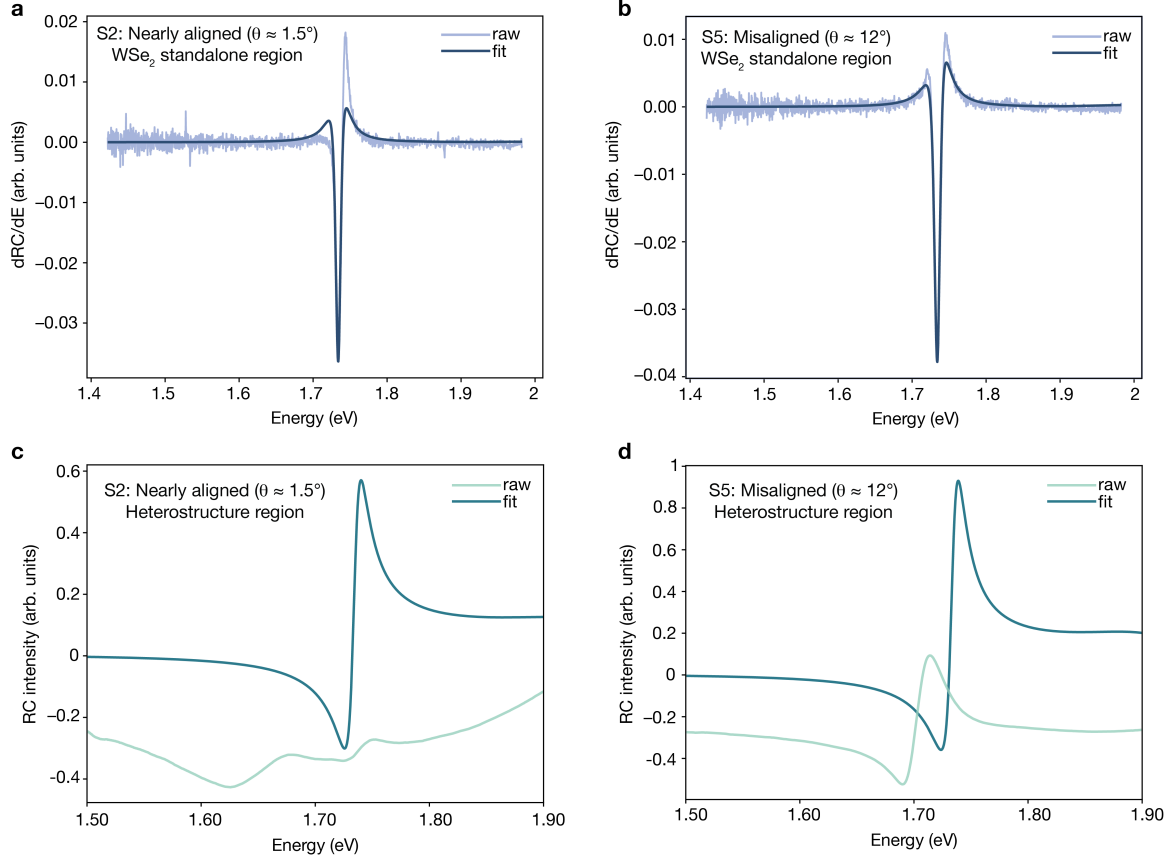
where ε_b is the background dielectric constant, and A , E_0 , and Γ_{nr} are the oscillator strength, resonance energy, and non-radiative broadening, respectively. The refractive indices (n) and extinction coefficients (k) of the other materials in the stack are taken from the literature [4]. The n , k values for $2H$ -TaS₂ are adopted from Ref. [5].

For $1H$ -WSe₂, the parameters of the 1s exciton from Equation 2 were iteratively optimized via least-squares fitting. Supplementary Figures 13a and 13b show such fits for the isolated $1H$ -WSe₂ regions of the nearly aligned sample S2 and misaligned sample S5, respectively. We applied a similar fitting procedure to the junction regions. While the fitting does not capture the background as accurately, the aligned sample shows comparatively broader features.

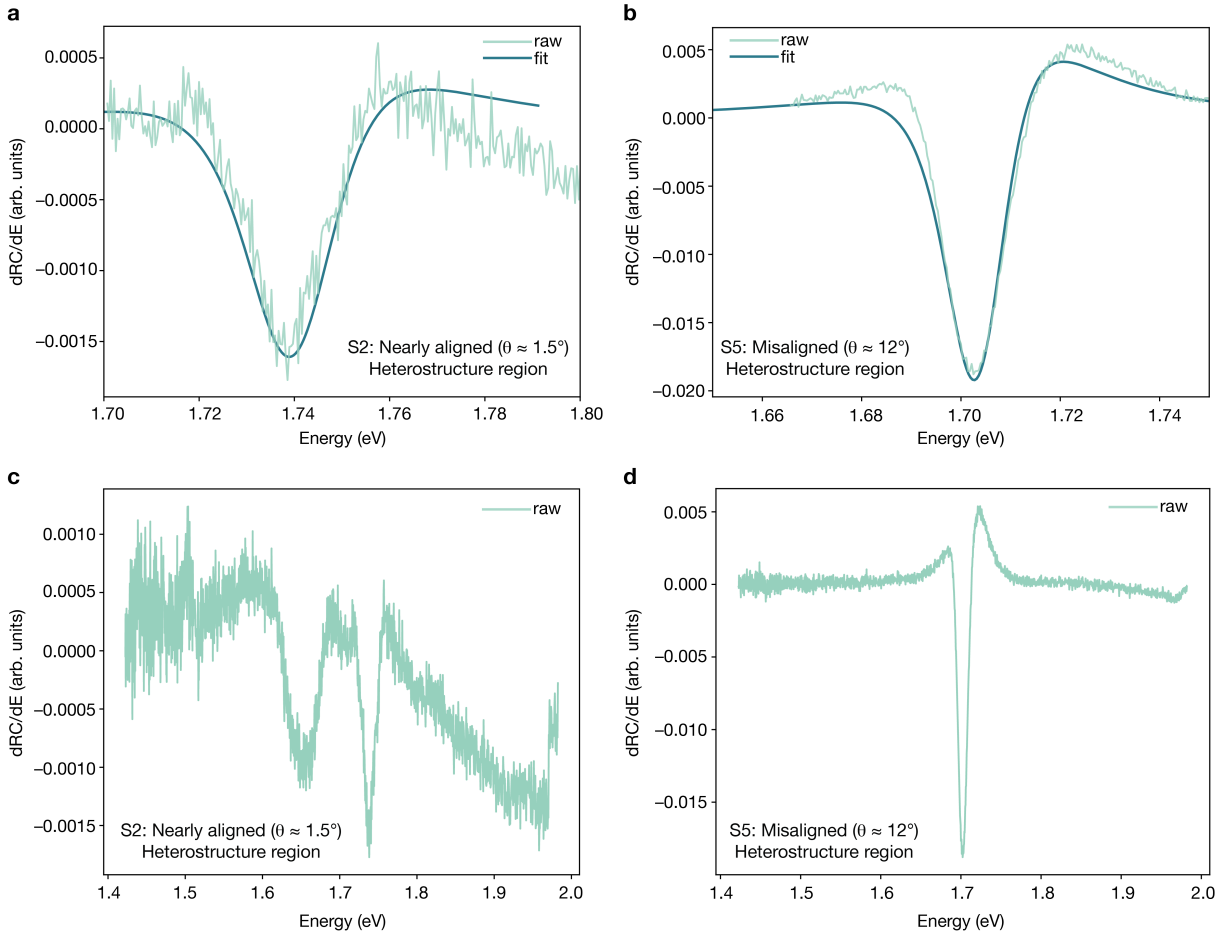
Alternatively, we compare the experimental RC spectra of the junction regions with calculated RC spectra based on noninteracting $2H$ -TaS₂ and $1H$ -WSe₂ layers, using the TMM model and fitted parameters from the isolated regions (Supplementary Figures 13c and 13d for S2 and S5, respectively). Broadening of the 1s exciton peak is evident for both S2 and S5.

Next, to quantify the broadening observed in the junction regions, the calculated RC spectrum is convolved with a Lorentzian of linewidth $\Delta\Gamma$ and shifted in energy to match the first derivative of the experimental RC spectra in magnitude and broadening (Supplementary Figures 14a and 14b). This additional broadening reflects the reduced exciton lifetime due to charge transfer across the junction, with $\tau = \hbar/\Delta\Gamma$ providing a lower bound assuming homogeneous broadening [6, 7]. Notably, we obtain broadening of a few meV for S2 and S5, similar to that observed in WS₂/graphite stacks [6, 7]. This is consistent with sub-ps charge transfer timescales in our heterostructures. The larger broadening in the aligned sample (by ~ 6 meV) suggests a relatively faster charge transfer rate, in addition to a stronger attenuation of the exciton oscillator strength.

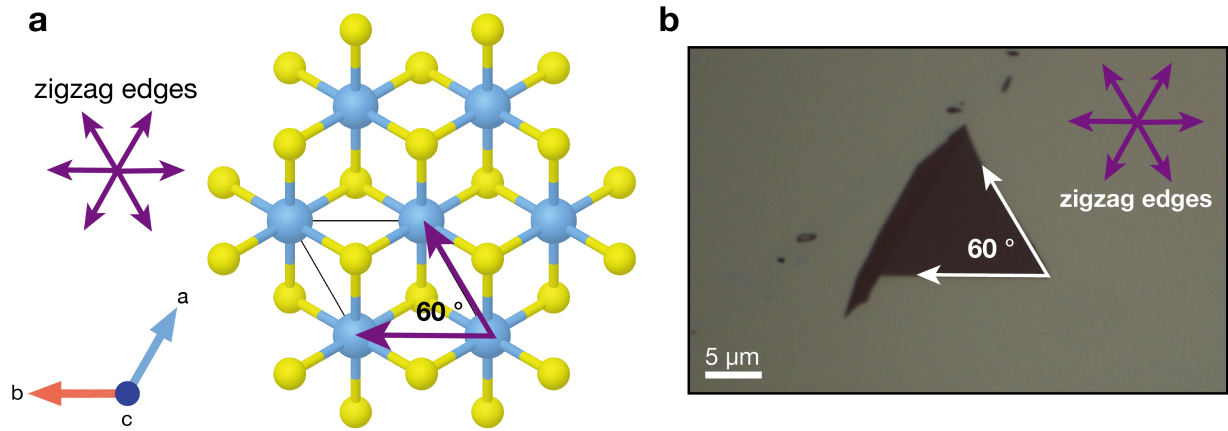
Finally, Supplementary Figures 14c and 14d show the raw RC spectra over an expanded energy range. Additional features are observed in the aligned sample S2, and these warrant further investigation to determine their origin.



Supplementary Figure 13. Reflection contrast (RC) analysis part 1. **(a,b)** Transfer matrix model (TMM) fits to measured RC spectra for the nearly aligned sample S2 **(a)** and the misaligned sample S5 **(b)**, each comprising 6-layer $1T$ - TaS_2 and monolayer $1H$ - WSe_2 . **(c,d)** Experimental RC spectra from the junction regions compared with calculated RC spectra assuming noninteracting layers for S2 **(c)** and S5 **(d)**.



Supplementary Figure 14. Reflection contrast (RC) analysis part 2. **(a,b)** First derivative of experimental RC spectra from the junction regions compared with simulated spectra incorporating additional broadening for S2 **(a)** and S5 **(b)**. **(c,d)** Raw RC data from **(a,b)** shown over an expanded spectral range for S2 **(c)** and S5 **(d)**.



Supplementary Figure 15. Cleavage behavior of transition metal dichalcogenides. **(a)** Crystal structure of 1T-TaS₂ with zigzag edges labeled. **(b)** 1T-TaS₂ flake with a well-defined 60° edge. Based on TEM studies in [8–11], the 60° angle identifies the zigzag direction. The zigzag direction of this flake, determined using optical microscopy, is consistent with the crystallographic edge orientation obtained from nanoARPES measurements.

References

- (1) Lacinska, E. M.; Furman, M.; Binder, J.; Lutsyk, I.; Kowalczyk, P. J.; Stepniewski, R.; Wysmolek, A. Raman Optical Activity of 1T-TaS₂. *Nano Letters* **2022**, *22*, 2835–2842.
- (2) Byrnes, S. J. Multilayer optical calculations, 2020.
- (3) Li, Y.; Chernikov, A.; Zhang, X.; Rigosi, A.; Hill, H. M.; van der Zande, A. M.; Chenet, D. A.; Shih, E.-M.; Hone, J.; Heinz, T. F. Measurement of the optical dielectric function of monolayer transition-metal dichalcogenides: MoS₂, MoSe₂, WS₂, and WSe₂. *Phys. Rev. B* **2014**, *90*, 205422.
- (4) Polyanskiy, M. RefractiveIndex.INFO - Refractive index database, <https://refractiveindex.info>, Accessed: 2025-03-24, 2011.
- (5) Munkhbat, B.; Wróbel, P.; Antosiewicz, T. J.; Shegai, T. O. Optical constants of several multilayer transition metal dichalcogenides measured by spectroscopic ellipsometry in the 300–1700 nm range: high index, anisotropy, and hyperbolicity. *ACS Photonics* **2022**, *9*, 2398–2407.
- (6) Hill, H. M.; Rigosi, A. F.; Raja, A.; Chernikov, A.; Roquelet, C.; Heinz, T. F. Exciton broadening in WS₂/graphene heterostructures. *Physical Review B* **2017**, *96*, 205401.
- (7) Rigosi, A. F.; Hill, H. M.; Li, Y.; Chernikov, A.; Heinz, T. F. Probing interlayer interactions in transition metal dichalcogenide heterostructures by optical spectroscopy: MoS₂/WS₂ and MoSe₂/WSe₂. *Nano Letters* **2015**, *15*, 5033–5038.
- (8) Husremović, S.; Groschner, C. K.; Inzani, K.; Craig, I. M.; Bustillo, K. C.; Ercius, P.; Kazmierczak, N. P.; Syndikus, J.; Van Winkle, M.; Aloni, S.; Taniguchi, T.; Watanabe, K.; Griffin, S. M.; Bediako, D. K. Hard ferromagnetism down to the thinnest limit of iron-intercalated tantalum disulfide. *Journal of the American Chemical Society* **2022**, *144*, 12167–12176.
- (9) Husremović, S.; Goodge, B. H.; Inzani, K.; Mier, A.; Ribet, S. M.; Bustillo, K. C.; Taniguchi, T.; Watanabe, K.; Ophus, C.; Griffin, S. M.; Bediako, D. K. Encoding multistate charge order and chirality in endotaxial heterostructures. *Nature Communications* **2023**, *14*, 6031.

- (10) Husremović, S. et al. Tailored topotactic chemistry unlocks heterostructures of magnetic intercalation compounds. *Nature Communications* **2025**, *16*, 1208.
- (11) Guo, Y.; Liu, C.; Yin, Q.; Wei, C.; Lin, S.; Hoffman, T. B.; Zhao, Y.; Edgar, J. H.; Chen, Q.; Lau, S. P.; Dai, J.; Yao, H.; Wong, H.-S. P.; Chai, Y. Distinctive in-plane cleavage behaviors of two-dimensional layered materials. *ACS Nano* **2016**, *10*, 8980–8988.