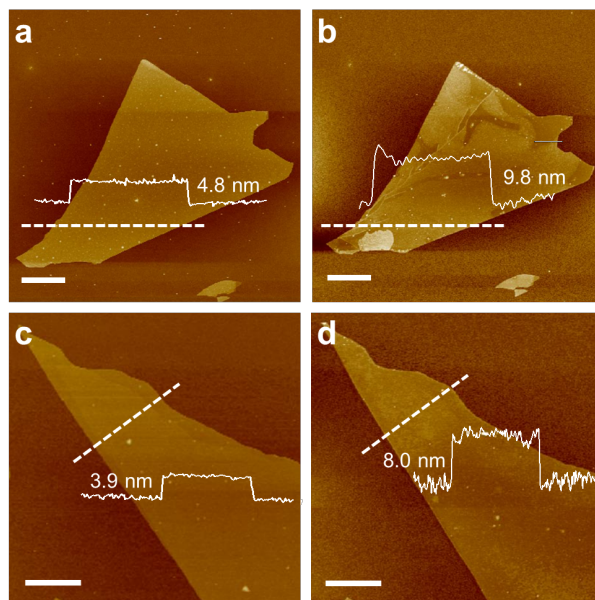


Molecular engineering stabilizes the commensurate charge density wave state in 1T-TaS₂/arginine hybrid superlattices

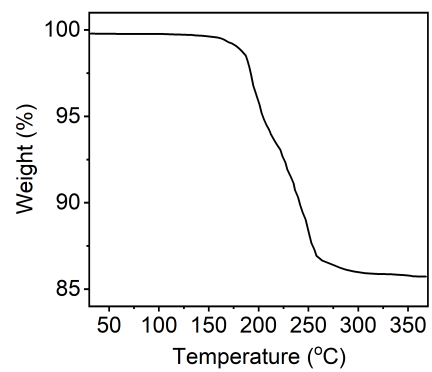
In the format provided by the
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Contents

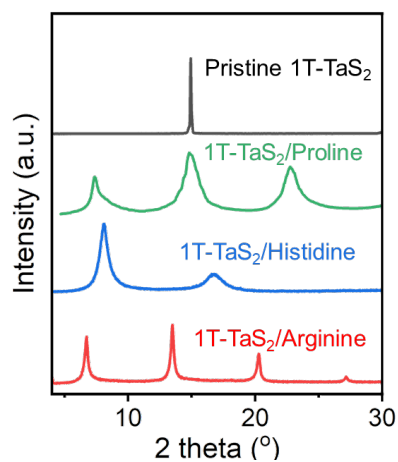
Supplementary Fig. 1	Structure characterization of 1T-TaS ₂ and 1T-TaS ₂ /Arg superlattice flakes
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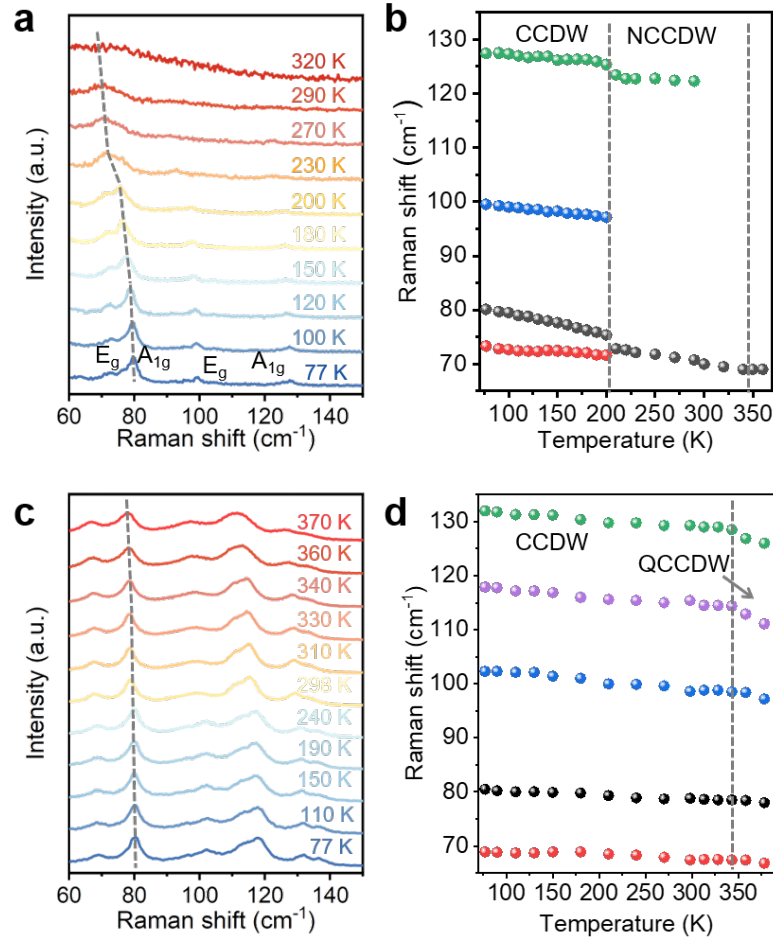
Supplementary Fig. 1 | Structure characterization of 1T-TaS₂ and 1T-TaS₂/Arg superlattice flakes. Representative AFM images of the same 1T-TaS₂ flakes before (a and c) and after (b and d) intercalation of Arg molecules, along with the height profiles of the corresponding linecuts in the AFM images. Scale bars, 2 μm .



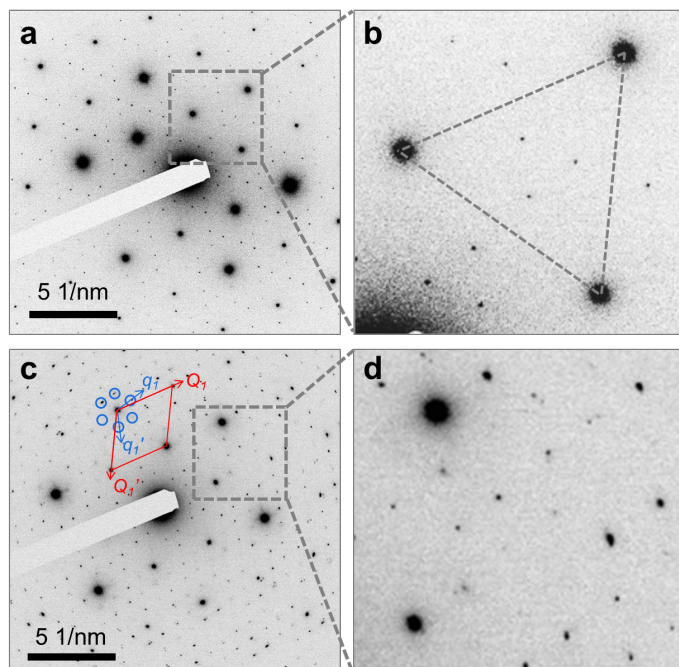
Supplementary Fig. 2 | TGA measurement of 1T-TaS₂/Arg superlattice, showing a typical mass loss of 14.1%.



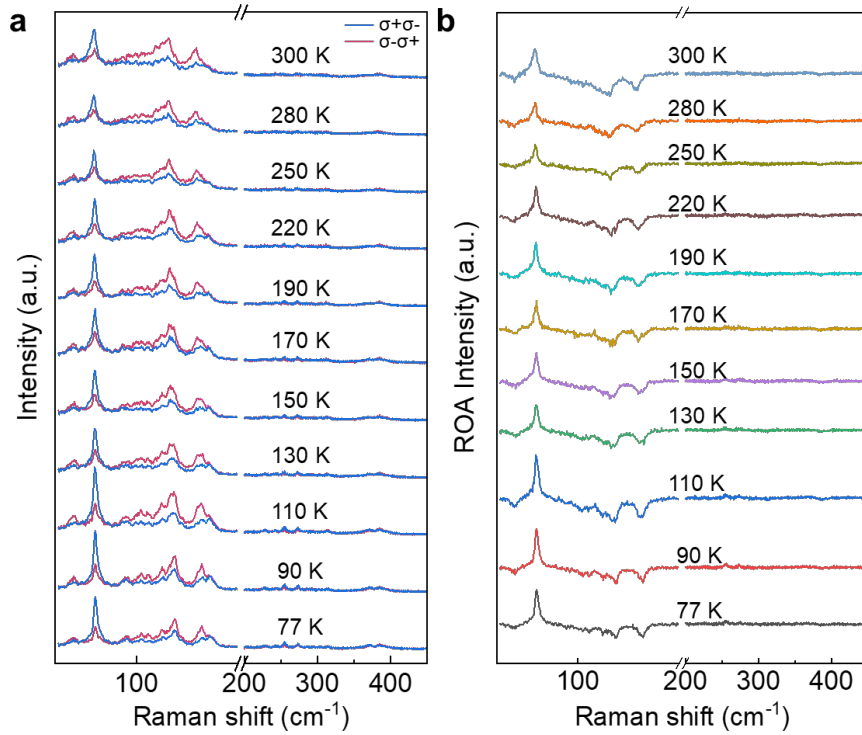
Supplementary Fig. 3 | Comparison of modulating effect of different molecules. XRD patterns of pristine 1T-TaS₂, 1T-TaS₂/Proline, 1T-TaS₂/Histidine, and 1T-TaS₂/Arginine, respectively.



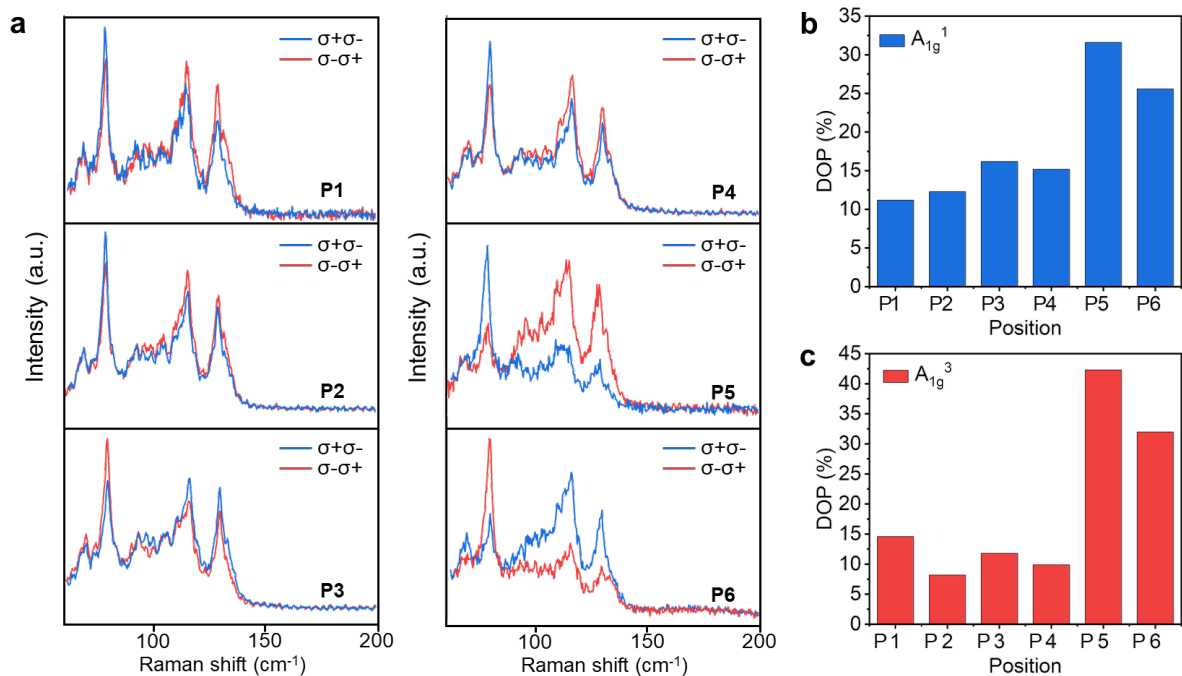
Supplementary Fig. 4 | Raman spectra of 1T-TaS₂ and 1T-TaS₂/Arg at different temperatures in the warming process. a,c, Raman spectra of 1T-TaS₂ (a) and Arg/1T-TaS₂ (c), respectively. b,d, Frequencies of discernible peaks are plotted as a function of increasing temperature. The phase transition temperatures are marked by dashed lines.



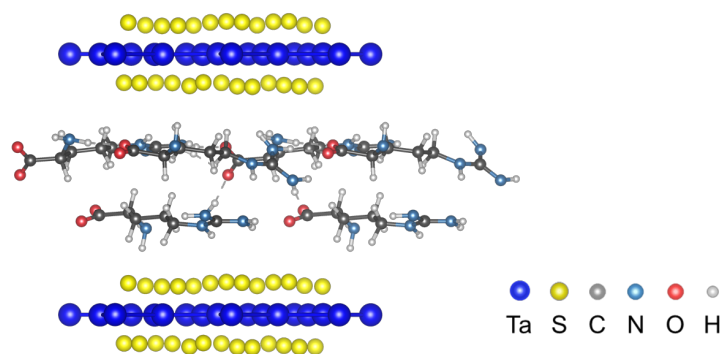
Supplementary Fig. 5 | SAED patterns of intrinsic 1T-TaS₂. Selected area electron diffraction (SAED) patterns of a 1T-TaS₂ flake at room temperature (a,b) and 77.3 K (c,d). A representative set of peaks is marked in blue circles.



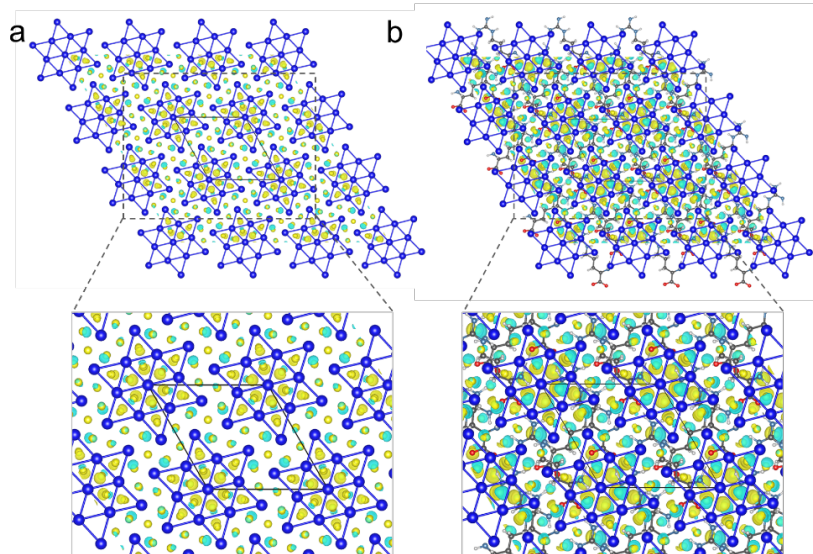
Supplementary Fig. 6 | ROA of 1T-TaS₂/Arg superlattice at different temperatures. **a**, Raman scattering spectra of Arg/1T-TaS₂ superlattice obtained in circularly polarized light in contrarotating $\sigma+\sigma-$ and $\sigma-\sigma+$ configurations, measured at different temperatures. **b**, ROA intensity of Arg/1T-TaS₂ at different temperatures obtained by subtracting $\sigma-\sigma+$ from $\sigma+\sigma-$. The ROA intensity retains with temperature decreasing.



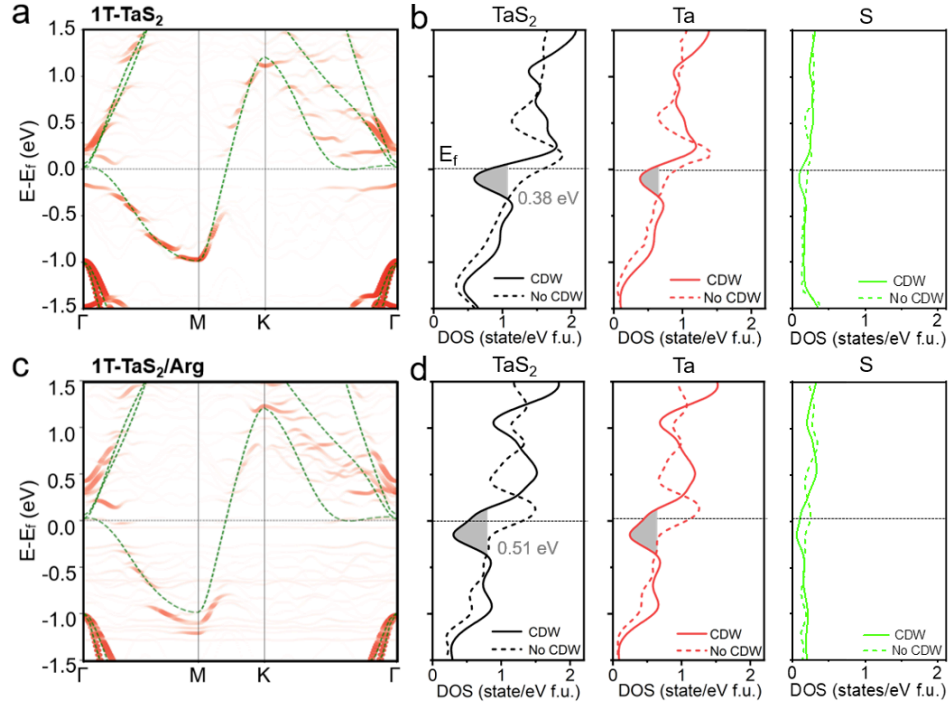
Supplementary Fig. 7 | DOP of A_{1g}^1 and A_{1g}^3 Raman modes for six different samples (P1-P6), demonstrating a broad distribution of possible overall ROA. a, ROA spectra of different 1T-TaS₂/Arg superlattice flakes (P1-P6) with $\sigma+\sigma^-$ and $\sigma-\sigma^+$ configuration lights at room temperature. **b**, **c**, DOP of A_{1g}^1 (b) and A_{1g}^3 (c) Raman modes of different flakes.



Supplementary Fig. 8 | Structure of 1T-TaS₂ upon arginine intercalation. Sideview of 1T-TaS₂/Arg, demonstrating the sulfur atom displacement in the superlattice.



Supplementary Fig. 9 | Charge-density difference (CDD) of 1T-TaS₂ and 1T-TaS₂/Arg. CDD of 1T-TaS₂ (a) and 1T-TaS₂/Arg (b). An absolute isosurface value of 0.018 e/bohr³ is adopted. The positive (yellow clouds) and negative (cyan clouds) CDDs signify the increase and decrease in electron density compared to that in the non-CDW phase, respectively.



Supplementary Fig. 10 | DFT-calculated unfolded band structures and density of states (DOS) near the Fermi level (E_f). **a,c**, Calculated unfolded band structures of **(a)** 1T-TaS₂ and **(c)** 1T-TaS₂/Arg. The green dashed lines represent the band structure of the primitive cell of undistorted non-CDW structure, while the band structures of the CDW phase are shown as red. The intensity is proportional to the unfolding spectral weight. **b,d**, DOS near E_f of pristine **(b)** 1T-TaS₂ and **(d)** intercalated 1T-TaS₂/Arg, including total TaS₂ DOS and projected DOS (PDOS). The solid lines represent CDW states, while the dashed lines represent non-CDW states. The energy axis of the DOS and PDOS panels align with the band-structure panels. The shaded regions highlight the DOS reconstruction induced by CCDW formation, including the suppression of spectral weight near E_f and redistribution of Ta-derived states, which is more pronounced in 1T-TaS₂/Arg than in pristine 1T-TaS₂. These near- E_f changes are treated only as indicators of electronic reconstruction, rather than as direct measures of the CDW gap or energy gain.