Supplementary materials for Emergent flat band electronic structure in a VSe_2 / Bi_2Se_3 heterostructure

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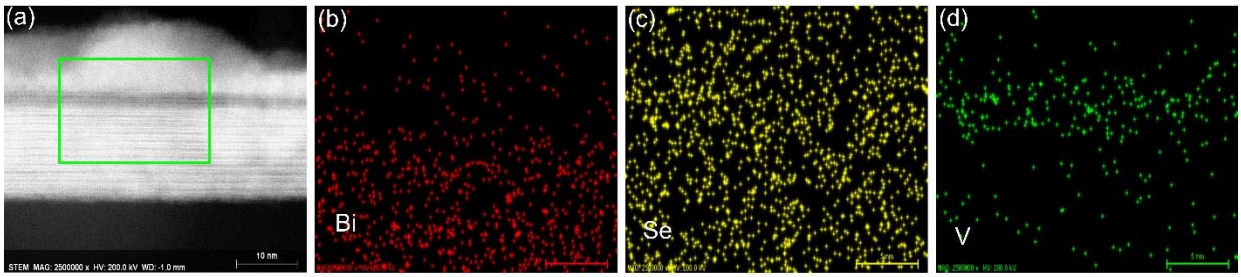
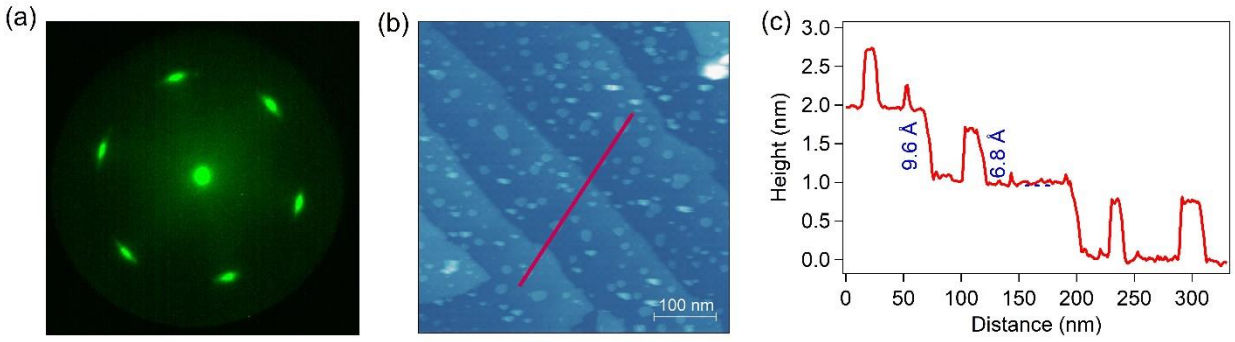
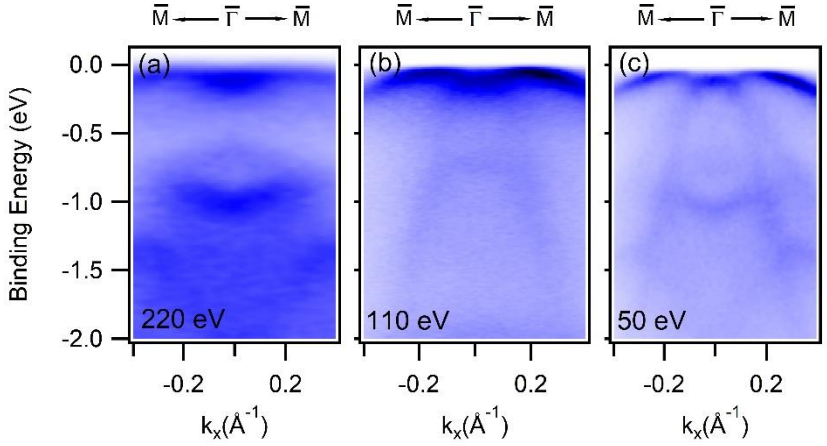


Supplementary Fig. 1a HAADF STEM cross-section image of 2 ML VSe_2 / 12 QL Bi_2Se_3 heterostructure. **b-d** STEM-EDX elemental distribution map of the green area in **a** for Bi, Se, and V, respectively. Bi, Se, and V atoms are found to be accumulated within the Bi_2Se_3 and VSe_2 layers as expected.

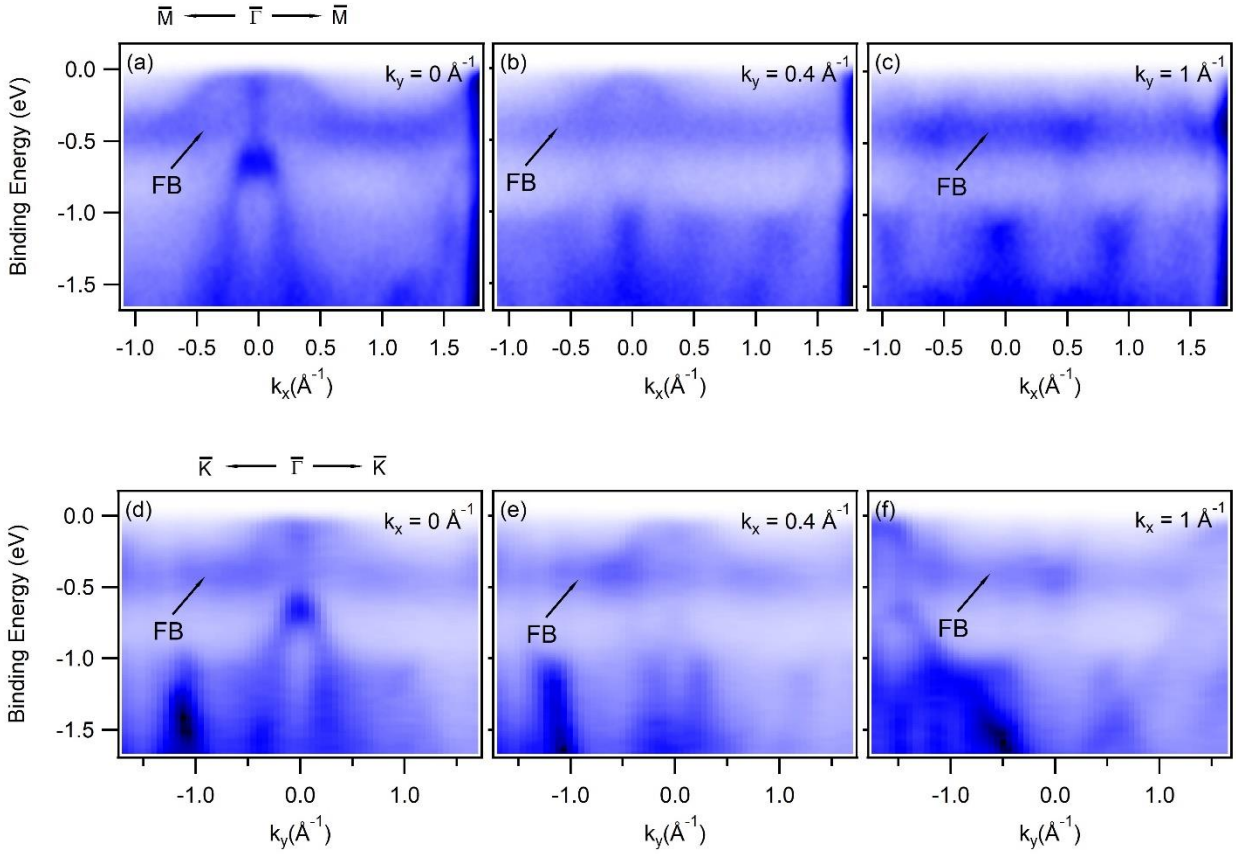


Supplementary Fig. 2 a μLEED pattern of the 0.3 ML VSe_2 / Bi_2Se_3 sample taken with 25 eV electron energy. The pattern exhibits six-fold crystal symmetry and the diffraction pattern does not show two distinct spots expected to be originating from the different in-plane lattice parameters of VSe_2 and Bi_2Se_3. This indicates the in-plane relaxation of Bi_2Se_3 and VSe_2. It is also evident that the VSe_2 overlayer has its axis aligned with the Bi_2Se_3 axis Furthermore, stretched spots along the rotational direction show the presence of VSe_2 domains oriented at slightly different directions to the Bi_2Se_3 lattice. **b** A large scale room temperature STM surface topography of the 0.3 ML VSe_2

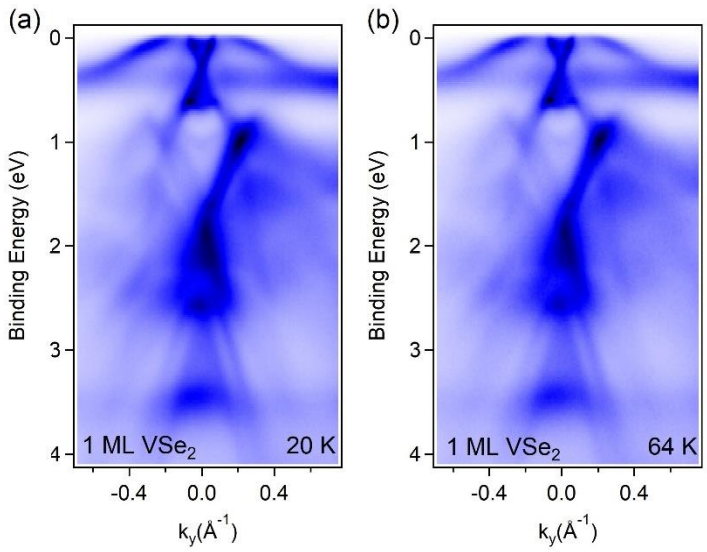
/ Bi_2Se_3 sample (at sample bias -1.2 V, set point 1 nA). The plot of height versus distance along the red line in **b** reveals that the heights are 6.8 Å for VSe_2 and 9.6 Å Bi_2Se_3.



Supplementary Fig. 3 a-c Photon energy dependent ARPES maps of a 3 ML VSe_2 / 12 QL Bi_2Se_3 heterostructure. The spectra were recorded with 220 eV, 110 eV, and 50 eV photons, respectively at 10 K and along the $\bar{\Gamma} - \bar{M}$ direction of the BZ.



Supplementary Fig. 4 a-f ARPES maps of a 1 ML VSe_2 /12 QL Bi_2Se_3 heterostructure at different k_x and k_y. The spectra were recorded with 169 eV photons at 10 K. The flat band is present at any spectrum taken at different k_x or k_y for the fixed direction at one of them. This shows that the flat band covers the entire BZ.



Supplementary Fig. 5 a, b ARPES maps of the ~1 ML VSe_2 / 12QL Bi_2Se_3 sample at 20 K and 64 K, respectively. V 3d orbital of VSe_2 disperses within the 0.45 eV energy range from the E_F as consistent with earlier reports[1-2]. The V 3d orbital overlaps with the flat band without inducing a prominent modification on its dispersionless feature. Furthermore, ARPES maps taken at 20 K and 64 K does not exhibit significant modifications on the nature of the band structures. **Note:** ARPES maps were conducted at BL1 beamline of the HISOR[3], Japan, with 50 eV photon energy along the $\bar{\Gamma} - \bar{M}$ direction in the BZ. Fermi edge is adjusted with regard to the edge of the photoemission intensity.

Supplementary References:

1. Strocov, V. N. et al. Three-dimensional electron realm in VSe_2 by soft X-ray photoelectron spectroscopy: origin of charge-density waves. Phys. Rev. Lett. **109**, 086401 (2012).

2. Wong, P. K. J. et al. Evidence of Spin Frustration in a Vanadium Diselenide Monolayer. Adv. Mater. **31**, 1901185 (2019).

3. Iwasawa, H. et al. Rotatable high-resolution ARPES system for tunable linear-polarization geometry. J. Synchrotron Radiat. **24**, 836–841 (2017).