Emergent flat band electronic structure in a VSe$_2$/Bi$_2$Se$_3$ heterostructure

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Flat band electronic states are proposed to be a fundamental tool to achieve various quantum states of matter at higher temperatures due to the enhanced electronic correlations. However, materials with such peculiar electronic states are rare and often rely on subtle properties of the band structures. Here, by using angle-resolved photoemission spectroscopy, we show the emergent flat band in a VSe$_2$/Bi$_2$Se$_3$ heterostructure. Our photoemission study demonstrates that the flat band covers the entire Brillouin zone and exhibits 2D nature with a complex circular dichroism. In addition, the Dirac cone of Bi$_2$Se$_3$ is not reshaped by the flat band even though they overlap in proximity of the Dirac point. These features make this flat band distinguishable from the ones previously found. Thereby, the observation of a flat band in the VSe$_2$/Bi$_2$Se$_3$ heterostructure opens a promising pathway to realize strongly correlated quantum effects in topological materials.
The physics of solids is largely determined by their energy band structures. Therefore, the investigation and control of distinct electronic band dispersions assume a great length to understand and discover new states of the matter. One of the exotic electronic states is a type of flat band, which is predicted to host high-temperature superconductivity\(^1-4\), fractional quantum Hall effect\(^5,6\), and ferromagnetism\(^7-9\). In superconductors, a flat band can boost the coupling constant and the transition temperature \((T_c)\) as a result of an enhanced density of states at the Fermi level \((E_F)\)\(^10,11\). This mechanism was utilized to explain the unexpected superconductivity in rhombohedral graphite and twisted graphene\(^12-14\). Other examples of flat band materials are Kagome lattices in which the flat band stems from destructive quantum interference due to the frustrated lattice geometry\(^15\). The Kagome-type flat bands have been observed in FeSn and Fe\(_2\)Sn\(_2\) by angle-resolved photoemission spectroscopy (ARPES) and in Co\(_3\)Sn\(_2\)S\(_3\) by scanning tunneling spectroscopy (STS)\(^16-19\). However, the complexity of the electronic structure in the photoemission data and the lack of momentum resolution in STS make the observations elusive. Computational efforts have also been made focusing on designing flat band transition metal dichalcogenides (TMDs) through the formation of the Moiré superlattices, which could support strongly correlated physics at higher temperatures\(^15-17\). All these previous works conclude that the flat band media could be fertile to many novel states of the matter. However, the limited number of the materials with such non-trivial bands hinders future studies.

Motivated by earlier studies, we investigate the surface electronic structure of VSe\(_2\) TMD grown on the surface of Bi\(_2\)Se\(_3\) topological insulator (TI) and show the emergence of a flat band in the electronic states. This flat band covers the entire \(k_x-k_y\) plane of the Brillouin zone (BZ) and displays dispersionless behavior along the \(k_z\) direction as well. Furthermore, circular dichroism ARPES (CD-ARPES) measurements reveal that the CD signal of the flat band reverses the sign at several points within the BZ. Another notable observation is that the VSe\(_2\) overlayer and the emergence of the flat band do not reshape the Dirac cone of Bi\(_2\)Se\(_3\) in the vicinity of the Dirac point (DP) unlike the case of transition metal doping which opens a large gap at the DP\(^18\). We also observe Moiré patterns in VSe\(_2\) domains of monolayer (ML) thickness and stripe-type patterns in bare Bi\(_2\)Se\(_3\) through scanning tunneling microscopy (STM). Further elucidations on the crystalline and chemical properties of the system are provided by scanning transmission electron microscopy (STEM), and microspot low-energy electron diffraction (μLEED). Our results demonstrate a rich physics in this system and suggest a large family of materials as possible emergent flat bands and thus will motivate future studies based on heterostructure formed by other quantum materials such as superconductors.

Results

Structural properties. Bi\(_2\)Se\(_3\) and VSe\(_2\) are layered materials with their atomic stacking geometry shown in Fig. 1a. The layers in each compound are separated by van der Waals (vdW) gaps with weak covalent out-of-plane bonds connecting the layers. These properties allow for the formation of well-ordered VSe\(_2\)/Bi\(_2\)Se\(_3\) heterostructure despite the large in-plane lattice mismatch of around 20% between the two materials\(^19,20\). Figure 1b, c depict the relevant core-levels of such structures 0 ML, 0.3 ML, 2 ML, and 3 ML VSe\(_2\) on 12 quintuple layer (QL) Bi\(_2\)Se\(_3\). Upon deposition of the VSe\(_2\), the Bi 5d peaks of Bi\(_2\)Se\(_3\) located at 25.1 and 28 eV remain at the same binding energies, indicating the absence of V metals at the interface and/or in the bulk (Fig. 1b)\(^21\). This also shows that VSe\(_2\) surface deposition does not modify the chemical potential of Bi\(_2\)Se\(_3\), seen as the absence of an electron or hole doping effect. Compared with pristine Bi\(_2\)Se\(_3\), the Se 3d peak, however, appears at 0.1 eV higher binding energy for VSe\(_2\) grown sample. The difference in binding energy is possibly related to the charge density wave (CDW) phase of VSe\(_2\)\(^14\). Furthermore, V 2p\(_{1/2}\) and 2p\(_{3/2}\) peaks of VSe\(_2\) shown in Fig. 1c are located at 513 and 520.6 eV binding energies corresponding to \(t\)-\(\delta\) oxidation states being in agreement with the recent report\(^20\).

To further explore the system, we show a high-angle annular dark-field (HAADF)-STEM cross-section image of a 3 ML VSe\(_2\)/12 QL Bi\(_2\)Se\(_3\) heterostructure in Fig. 1d. Bi\(_2\)Se\(_3\) and VSe\(_2\) exhibit regular atomic layers with smooth interfaces and vdW gaps marked with red arrows in Fig. 1d. On the other hand, the interface spacing between the Bi\(_2\)Se\(_3\) and VSe\(_2\) layers smaller comparably than the vdW gaps which could strongly modify the local electronic structure. Furthermore, the STEM energy dispersive X-ray spectroscopy elemental maps presented in Supplementary Fig. 1 show the atomic distribution of Bi in the Bi\(_2\)Se\(_3\) layers, V in VSe\(_2\) layers, and Se across the heterostructure as expected.

To study the local crystal structure, Fig. 1e, f show the STM image of Bi\(_2\)Se\(_3\) and VSe\(_2\) regions of a 0.3 ML VSe\(_2\)/12 QL Bi\(_2\)Se\(_3\) sample, respectively. The Bi\(_2\)Se\(_3\) surface has a stripe-like pattern similar to Cs and Fe doped Bi\(_2\)Se\(_3\)\(^22\). The STM image of the VSe\(_2\) domains presented in Fig. 1f exhibits a Moiré pattern with \(\sim 2 \text{ nm} \times \text{2 nm}\) superstructure. This differs from the previous studies conducted on VSe\(_2\)/graphene\(^19\). Moiré pattern can be formed by a small misfit between the in-plane lattice parameters of the film and the underlying material or the relative rotation of two layers to each other, or both. By contrast, the lattice mismatch between the VSe\(_2\) and Bi\(_2\)Se\(_3\) is quite large (about 20%). Unfortunately, we cannot make a quantitative analysis for precise determination of the in-plane lattice parameters or the atomic displacement due to the limitation in our STM data taken at room-temperature experiment. However, similar Moiré pattern formation is also observed on ML MoSe\(_2\) grown on a graphene substrate whose origin is attributed to the lattice mismatch between the multiple unit cells of the two materials\(^23\). Thereby, the Moiré pattern in VSe\(_2\) could be formed due to the small mismatch between four-five-unit cells of Bi\(_2\)Se\(_3\) (4\(\delta\)\(\alpha\)\(_\text{Ga}\) = 16.56 Å or 5\(\delta\)\(\alpha\)\(_\text{Ga}\) = 20.7 Å) and five-unit cells of VSe\(_2\) (5\(\delta\)\(\alpha\)\(_\text{Se}\) = 16.8 Å or 6\(\delta\)\(\alpha\)\(_\text{Se}\) = 20.16 Å) for the rotationally aligned lattice geometry. Alternatively, the Moiré pattern could form be formed by the rotational misalignments of Bi\(_2\)Se\(_3\) and VSe\(_2\) atomic lattices. Moreover, the details of the STM data reveal that the layer height is 6.8 Å for VSe\(_2\) (Supplementary Fig. 2) being in the line with the recent findings\(^24\).

Observation of the flat band in a VSe\(_2\)/Bi\(_2\)Se\(_3\) heterostructure.

To examine the band structure, the binding energy vs. \(k_z\) plots are given in Fig. 2a for 12 QL Bi\(_2\)Se\(_3\) and in Fig. 2b–d for various thickness of VSe\(_2\) on 12 QL Bi\(_2\)Se\(_3\). Bi\(_2\)Se\(_3\) exhibits the typical band structure with the linear Dirac surface states (DSSs) forming the Dirac cone with the Dirac point (DP) at 0.36 eV below \(E_F\)\(^25\). Upon deposition of 0.3 ML VSe\(_2\) on the surface of Bi\(_2\)Se\(_3\), a flat band at 0.47 eV binding energy and with a \(\sim 0.18\) eV bandwidth emerges in the surface electronic structure (Fig. 2b). The flatness of the band is well distinguished in the ARPES maps where bulk bands and the DSS of Bi\(_2\)Se\(_3\) strongly disperse as a function of \(k_z\), while the flat band retains dispersionless across the \(\Gamma - M\) high-symmetry lines. VSe\(_2\) growth also induces the well-known M-state quantization\(^26\) of the bulk valence band of Bi\(_2\)Se\(_3\) shown in Fig. 2b. The flat band can be resolved in 1 ML and, less intense, in 2 ML VSe\(_2\) (Fig. 2c, d) and further increasing the thickness of VSe\(_2\) to 3 ML leads to disappearance of the flat band from the ARPES map (Fig. 2e). On the other side, the thicker VSe\(_2\) grown on a Bi\(_2\)Se\(_3\) exhibits the same electronic feature with the one grown on a highly ordered pyrolytic graphite (HOPG) substrate.
(Fig. 2f), indicating that the Bi$_2$Se$_3$ sublayer plays a crucial role in the formation of the new electronic states. It is also noticeable that the nature of the flat band stays unchanged with increasing VSe$_2$ thickness as its binding energy and dispersionless character remain the same. The only observable evolution is in the spectral intensity which gradually decreases with increasing VSe$_2$ thickness. This emphasized in Fig. 2g displaying the energy distribution curves (EDCs) taken at band at different VSe$_2$ thicknesses. It is also found that the DP of Bi$_2$Se$_3$ does not experience an energy shift upon surface deposition of VSe$_2$. This is correlated with the atomic number (Z-contrast).

Another interesting observation is that the flat band overlaps with the lower branch of the Dirac cone in the vicinity of $k_z = 0$ Å$^{-1}$ without inducing any appreciable change in its spectral shape. This can be even better seen in the films with the thicker VSe$_2$ coverage confirming that the flat band, DSSs, and the dispersive V 3$d$ state of VSe$_2$ coexist in the surface electronic structure (Fig. 2b-d and Supplementary Fig. 5). It is also found that the DP of Bi$_2$Se$_3$ does not experience an energy shift upon surface deposition of VSe$_2$. This indicates the absence of any band bending effect which is consistent with the behavior of Bi 5$d$ core levels presented in Fig. 1b.

To further investigate the flat band, $k_x$, $k_y$, intensity plots at $E_F$ and at the binding energy of the flat band ($E_{FB}$) for a 1 ML VSe$_2$/12 QL Bi$_2$Se$_3$ sample are shown in Fig. 2i, j, respectively. The Fermi surface is dominated by a flower-like electron pocket formed by the V 3$d$ orbitals of VSe$_2$ centered at Γ point as similar to the earlier observation for VSe$_2$ grown on different substrates. The constant energy cut at $E_{FB}$ is instead quite featureless. Besides, a weak residual of the start-like features of VSe$_2$ no new, distinct dispersion is seen along any direction of the BZ. This indicates that the flat band fills the entire BZs of Bi$_2$Se$_3$ and VSe$_2$ as depicted by blue and red hexagons in Fig. 2i, j, respectively. This can be also seen in Supplementary Fig. 4 where the spectra taken along the different directions in the BZ for fixed $k_x$ and $k_y$ momentum all show the existence of the flat band. Such electronic state spread over a large momentum area can significantly enhance the electronic correlation yielding quantum effects at very high-temperatures. It is also worth noting that the LEED pattern of the sample shows stretched diffraction spots along the rotational direction indicating the presence of the rotationally misaligned VSe$_2$ domains (Supplementary Fig. 2a) with respect to each other and to the Bi$_2$Se$_3$ substrate. The rotational misfit of ±3° estimated from µLEED pattern, however, is too small for a band to span whole BZ and to induce a fully occupied constant energy counter in the momentum space.

In Fig. 2k, we also present an ARPES map covering two BZ centers. The flat band connects two Γ points to each other. Another intriguing realization in this spectra is that the Bi$_2$Se$_3$ and VSe$_2$ shares the same in-plane lattice constants. This is consistent with our LEED measurements, which does not show distinct diffraction patterns arising from VSe$_2$ and Bi$_2$Se$_3$ (Supplementary Fig. 2a). This is supported with the EDC taken at the Fermi level and given on the top of the spectra in Fig. 2k. Two zone centers are separated with $k_z = 1.9$ Å$^{-1}$ yielding a 4.03 Å in-plane lattice parameter. This is smaller than in-plane lattice...
energy counters at the corresponding EDCs obtained along the 220 eV photon energy along the photons. A Tougaard background is subtracted from each spectrum. The corresponding ARPES maps are presented in Supplementary Fig. 3.

structure of 0.3, 1, 2, and ML VSe₂/12 QL Bi₂Se₃ heterostructures, respectively. A corresponding ARPES map can be found in Supplementary Fig. 4.

ky QL Bi₂Se₃ heterostructure are given in Fig.3. In the plot of momentum distribution curves obtained at various kz directions to distinguish the two-dimensional (2D) bands from the dispersions can be also extracted. This method allows studying the dispersion of the energy bands along the kz (out-of-plane) direction to distinguish the two-dimensional (2D) bands from the dispersive bulk bands. Such spectra acquired at varying photon energies (from 45 to 120 eV with 5 eV steps) for a 1 ML VSe₂/12 QL Bi₂Se₃ heterostructure are given in Fig. 3. In the plot of kₜ vs. kₓ dispersion at Eₚ, DSSs marked with dashed black lines exhibit no kₓ dependence (Fig. 3a). Similar spectrum at Eₚ given in Fig. 3b shows that a high spectral intensity along the kₓ = 0 Å⁻¹ originates from the bottom of the Dirac cone of Bi₂Se₃. Away from the kₓ = 0 Å⁻¹, the plot has non-vanishing spectral intensity contributed by the flat band. This can be better seen in the momentum distribution curves obtained at various kₓ points (Fig. 3c) in which each spectrum exhibits always finite density of states along the kₓ momentum direction. This implies the dispersionless nature of the flat band along the kₓ momentum direction. To further validate this observation, we present the binding energy-kₓ plots along the kₓ = ±0.25 Å⁻¹ in Fig. 3d, e, respectively. The plots clearly show that the flat band at 0.47 eV binding energy is kₓ independent confirming its non-bulk derived nature. We should also note that the M-shape bulk band located in the vicinity of 1 eV binding energy exhibits a nearly non-dispersive feature along the kₓ as shown in Fig. 3d, e. To further reveal the details of the flat band, Fig. 3f depicts the EDCs taken at different kₓ points. One can see that the EDC of the flat band does not exhibit a kₓ-dependent evolution in the binding energy and bandwidth, providing a signature that it has 2D nature and originates from single type of atomic orbital.

Photon energy-dependent electronic structure. In ARPES experiments, by recording the electronic structure with a wide photon energy range, a kₓ vs. k_y or binding energy vs. k_z dispersions can be also extracted. This method allows studying the dispersion of the energy bands along the k_z (out-of-plane) direction in the vicinity of the interface and induce new electronic states.

Circular dichroism ARPES. CD-ARPES has gained great attention due to its feasibility to investigate the helical spin–orbit texture in topological surface states. The principle of the method is the spectral weight differences in ARPES arising from the opposite helicity of the circularly polarized lights. CD-ARPES is then obtained from \[\left| \frac{I_{\text{RCP}} - I_{\text{LCP}}}{I_{\text{RCP}} + I_{\text{LCP}}} \right| \] where I_{\text{RHP}} and I_{\text{LHP}} are photoemission intensities for right hand circular polarized (RCP) and left hand circular polarized (LCP) lights, respectively. Thus, we have recorded the band structure of 0.3 ML VSe₂/12 QL Bi₂Se₃ sample with RCP and LCP, shown in Fig. 4a, b, respectively. The corresponding CD-ARPES is presented in a binary color map in Fig. 4c (red: negative-CD and blue: positive-CD). The bulk bands of Bi₂Se₃ dispersing below 0.8 eV binding energy show a strong CD signal as seen in Fig. 4a–c. CD signal of the DSSs exhibits a spectral weight switching from the −kₓ to +kₓ regions when changing the excitation energy from RCP to LCP. For clarity, the DC signal vs. kₓ is plotted in Fig. 4d at 0.1 eV
binding energy where the CD is positive for left and negative for the right side of the Dirac cone, marked with vertical arrows. Further away from the $k_y = 0$ Å$^{-1}$, the plot in Fig. 4d still shows non-zero CD. This is likely originating from the $V_{3d}$ orbitals, which dominate the density of states at the $E_F$ for VSe$_2$.

To investigate the dichroism effect in the flat band, the CD at $E_{FB}$ is also plotted as a function of $k_y$ in Fig. 4e and it exhibits sign inversions at $k_y = 0$ Å$^{-1}$ and $k_y = \pm 0.5$ Å$^{-1}$, and the maxima at $k_y = \pm 0.25$ Å$^{-1}$. This shows that similar to the DSSs, the CD in the flat band also exhibits helical texture where opposite $k_y$ momentums have opposite signs of the CD. Notably, zero CD signal is also observed as white color in the CD-ARPES along the $k_y = 0$ Å$^{-1}$. This depicts the nodal line, which was proposed to be the characteristic feature of the 2D electronic structure. In particular, the CD signal of the DSSs depends on the incident photon energy assigning it to the final state effect in the photoemission process. This was discussed in ref. with details where they propose the non-trivial connection between the spin–orbit texture and the CD signal. Thereby, the helical CD texture and the nodal line band suggest that the flat band could be topologically non-trivial.

Temperature-dependent electronic structure. To further study the nature of the flat band in detail, we present the temperature-dependent electronic structure of a 1 ML VSe$_2$/12 QL Bi$_2$Se$_3$ heterostructure. a, b $k_y-k_z$ dispersions at the $E_F$ and the $E_{FB}$, respectively. Dashed red lines in a mark the DSSs. c, MDCs at different $k_y$ points. d, e Binding energy vs. $k_y$ maps at $k_y = \pm 0.25$ Å$^{-1}$, respectively. Dashed cyan colored lines in d and e represent the dispersion of the flat band along the $k_y$ direction. f EDCs at various $k_y$ points to study the spectral shape of the flat band. ARPES maps for the plots were conducted along the $\Gamma-K$ direction in the BZ. Data were conducted from the 1 ML VSe$_2$/12 QL Bi$_2$Se$_3$ heterostructure at 10 K.
dependent ARPES maps of 1 ML VSe$_2$/12 QL Bi$_2$Se$_3$ in Fig. 5a-d and 0.3 ML VSe$_2$/12 QL Bi$_2$Se$_3$ in Fig. 5f-j for 10, 100, 200, and 300 K, respectively. For both samples, the temperature does not induce a prominent change on the nondispersive nature of the flat band. This can be seen in the EDCs obtained along the $k_y = -0.25$ Å$^{-1}$ from the ARPES maps (Fig. 5e–j). It is clear from the data that temperature does not affect the line shape or the binding energy of the flat band. At any given temperature, the flat band is located at 0.47 eV. Only noticeable change is the broadening of all spectral features with increasing temperature as can be expected from phonon contributions to the total lifetime. On the other side, these data constitute a strong indication that the binding energy of the flat band should change if it is formed by the impurities or disorders. This can be found in a recent study where the disorder induced kink like states in Bi$_2$Se$_3$ can be diminished by lightly annealing the sample$^{31}$. The temperature-dependent ARPES data also show that the flat band is not due to surface impurities. It is expected that the impurities will segregate into the bulk of Bi$_2$Se$_3$ and locate in sublattices with increasing temperature$^{32}$. This will significantly modify the chemical potential of the system. A number of ARPES studies have been devoted to studying the effects of the adatoms on the band structure of Bi$_2$Se$_3$ and the only change found has been electron or hole doping$^{18,26,32}$. No experimental work has shown indication of any new states or flat like bands induced by surface impurities on the surface electronic structure of a TI. Furthermore, resonant ARPES experiments also revealed that the transition metal impurity states evolve onto the surface states rather than forming a flat band$^{33,34}$. Finally, it is worth mentioning that the ARPES maps taken at room temperature can be compared with the room-temperature STM data presented in Fig. 1f, leading to the conclusion that the Moiré pattern and the flat band electronic states coexist in our system.

Discussion
Here we present a detailed ARPES study on a VSe$_2$/Bi$_2$Se$_3$ system and showed unexpected formation of a flat band on the surface electronic structure on this heterostructure. One of the first scenario to consider as the origin of the flat band is the surface V impurities. Even though the theory predicts nearly nondispersive features induced by impurities or disorders in TIs$^{31,35}$, experimental studies shows that these states evolve onto the surface states rather than being flat in the momentum space$^{33,34}$. Also, in contrast to our results, these states are expected to strongly modify the Dirac cone and open an energy gap at the DP$^{31,35}$. Another scenario to consider would be the existence of superlattices as seen in $\sqrt{3} \times \sqrt{3}$ silicone superstructure by STS where the local density of states forms the electronic Kagome lattice$^{36}$. Interface dislocation or strain can also flatten the original bands by introducing pseudo-magnetic field term to the Hamiltonian in Moiré superstructures$^5$. Furthermore, a pronounced band flattening in this scenario requires superstructure patterns with at least a few tens of nanometers periodicity which is much larger than one observed in the present case. However, in contrast to our observations, the flat band discussed within the superlattice frameworks is dispersionless only in the BZ of the superstructure$^{17,36}$.

On the other hand, our results show the emergent character of our flat band, which is likely due to the formation of the interface in the heterostructure between Bi$_2$Se$_3$ sublayer and VSe$_2$ top layer. This conclusion is supported by thickness and photon energy-dependent photoemission data showing that the flat band can be resolved with higher photon energies on thicker VSe$_2$ films. As a first example of the dispersionless electronic excitation in a topologically non-trivial band structure, our results could open a new pathway in the critical field of experimental realization and control of novel quantum effects.

Methods
Synthesis. Molecular beam epitaxial growth (MBE) technique was employed to grow VSe$_2$/Bi$_2$Se$_3$ and Bi$_2$Se$_3$ samples in a custom ultrahigh vacuum system located at the ESM beamline of NSLS-II. Se and Bi sources (5 N) were evaporated from the
ceramic crucibles while the e-beam evaporation method was used for V (99.8% purity) source. All samples were grown on Al2O3(0001) substrates at 235 °C. Before the growth, native substrate oxidation was first degassed at 520 °C for 15 min, and then flashed at 850 °C for 5 min. An ML VSe2 was grown on a freshly cleaved HOPG substrate at 235 °C after annealing the substrate at 350 °C for 2 h. Sample thicknesses were estimated within a 15% error bar by using a quartz thickness monitor and X-ray photoemission spectroscopy. Samples for ARPES and µLEED experiments were capped with 20 nm amorphous Se film before being removed from the MBE chamber.

Core-level spectroscopy. Core-levels were recorded at 21-ID-1 ESM beamline of National Synchrotron Light Source II (NSLS-II) by using a DA30 Scienta electron spectrometer at 10 K sample temperature.

Angle-resolved photoemission spectroscopy. ARPES experiments were performed at 21-ID-1 ESM beamline of NSLS-II by using a DA30 Scienta electron spectrometer. The pressure in the photoemission chamber was $1 \times 10^{-10}$ Torr and samples were kept at 15 K during the experiment by a closed-cycle He cryostat. The energy resolution in the ARPES experiments was better than 15 meV with a spot size of ~20 µm. Before the ARPES experiments, samples were annealed at 220 °C for 30 min to remove the Se capping layer. The angle between the light and the surface normal of the sample is 55° at the normal emission during the ARPES experiments. The films were grounded with a tantalum clip. A part of the ARPES experiments was conducted at the linear undulator beamline at the Hiroshima Synchrotron Radiation Center BL-1 (Supplementary Fig. 5). Photon energy is converted to $k_f$ momentum space by using the free electron final state approximation $hk_f = \sqrt{2m(E_{\text{kin}}+E)}$, where $m_e$ is the free electron mass, $E_{\text{kin}}$ is the kinetic energy of a photoelectron, and $E$ is the inner potential taken as 11.8 eV for Bi2Se3.17

TEM and STM microscopy. HAADF-STEM images were acquired with Hitachi HD2700C dedicated STEM with a probe Cs corrector operating at 200 kV at room temperature. Samples were prepared using the in-situ lift-out method on the FEI Helios 600 Nanolab dual-beam FIB. Final milling was completed at 2 keV. STM (Omicron VT-STM-XA 650) experiments were performed in an ultrahigh vacuum (UHV) system with a base pressure of 2 × 10⁻¹⁰ Torr at room temperature. All the STM images were observed in constant current mode using Pt/Ir tips. All bias values in the text refer to the bias applied to the sample. The STM images were analyzed using Gwyddion-2.55 software package. HAADF-STEM and STM experiments were conducted at the Center for Functional Nanomaterials, Brookhaven National Laboratory. Samples for STM were transferred with a vacuum suitcase.

Low-energy electron diffraction. µLEED experiment was performed at X-ray photoemission electron microscopy/low-energy electron microscopy (XPEEM/LEEM) endstation of the ESM beamline (21-ID-2).

Data availability

The data that support the findings of this study are available from the corresponding author upon request.

Received: 25 November 2020; Accepted: 12 December 2020; Published online: 20 January 2021

References

1. Heikkilä, T. & Volokov, G. E. Flat Bands as a Route to High-Temperature Superconductivity in Graphite (ed. Esquinazi, P.) 123–144 (Springer, Switzerland, 2016).
2. Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).
3. Heikkilä, T. T., Kopnin, N. R. & Volokov, G. E. Flat bands in topological media. JETP Lett. 94, 233–239 (2011).
4. Tang, E. & Fu, L. Strain-induced partially flat band, helical snake states and interface superconductivity in topological crystalline insulators. Nat. Phys. 10, 964969 (2014).
5. Tang, E., Mei, J. W. & Wen, X. G. High-temperature fractional quantum Hall states. Phys. Rev. Lett. 106, 236802 (2011).
6. Sun, K., Gu, Z., Katsura, H. & Das Sarma, S. Nearly flatbands with nontrivial topology. Phys. Rev. Lett. 106, 236803 (2011).
7. Guo, H. M. & Franz, M. Topological insulator on the kagome lattice. Phys. Rev. B 80, 113102 (2009).
8. Liu, Z. et al. Flatbands and emergent ferromagnetic ordering in Fe2Se5 kagome lattices. Phys. Rev. Lett. 121, 96401 (2018).
9. Kang, M. et al. Dirac fermions and flat bands in the ideal Kagome metal FeSn3. Nat. Mater. 19, 163–169 (2020).
10. Yin, J. et al. Negative flat band magnetism in a spin–orbit–coupled correlated kagome magnet. Nat. Phys. 15, 443–448 (2019).
11. Kopnin, N. B., Heikkilä, T. T. & Volokov, G. E. High-temperature surface superconductivity in topological flat-band systems. Phys. Rev. B 83, 220503(R) (2011).
12. Kopnin, N. B., Jía, M., Harju, A. & Heikkilä, T. T. High-temperature surface superconductivity in rhombohedral graphite. Phys. Rev. B 87, 140503 (2013).
13. Marchenko, D. et al. Extremely flat band in bilayer graphene. Sci. Adv. 4, 11 (2018).
14. Volokov, G. E. Graphene, graphene and the flat band superconductivity. JETP Lett. 107, 516–517 (2018).
15. Wu, F., Lovorn, T., Tutuc, E. & MacDonald, A. H. Hubbard model physics in transition metal dichalcogenide moiré bands. Phys. Rev. Lett. 121, 026402 (2018).
16. Naik, M. H. & Jain, M. Ultraflatbands and shear solitons in moiré patterns of twisted bilayer transition metal dichalcogenides. Phys. Rev. Lett. 121, 266401 (2018).
17. Bi, Z., Yuan, N. F. Q. & Fu, L. Designing flat bands by strain. Phys. Rev. B 100, 035448 (2019).
18. Yilmaz, T. et al. Distinct effects of Cr bulk doping and surface deposition on the chemical environment and electronic structure of the topological insulator Bi2Se3. Appl. Surf. Sci. 407, 371–378 (2017).
19. O' Connor, P. et al. Unique gap structure and symmetry of the charge density wave in single-layer VSe2. Phys. Rev. Lett. 121, 196402 (2018).
20. Liu, Z. L. et al. Epitaxially grown monolayer VSe2: an air-stable magnetic two-dimensional material with low work function at edges. Sci. Bull. 63, 419 (2018).
21. Walsh, L. A. et al. Interface chemistry of contact metals and ferromagnets on the topological insulator Bi2Se3. J. Phys. Chem. C 124, 23551 (2020).
22. Shokri, R. Coexistence of impurity-induced quasi-one-dimensional electronic structure and topological surface states of Bi2Se3. J. Appl. Phys. 119, 058304 (2016).
23. Ugeda, M. M. et al. Giant bandgap renormalization and excitonic effects in a monolayer transition metal dichalcogenide semiconductor. Nat. Mater. 13, 1091–1095 (2014).
24. Duviir, G. et al. Emergence of a metal-insulator transition and high-temperature charge-density waves in VSe2 at the monolayer limit. Nano Lett. 18, 5432–5438 (2018).
25. Xia, Y. et al. Observation of a large-gap topological-insulator class with a single Dirac cone on the surface. Nat. Phys. 5, 398 (2009).
26. Bianchi, M., Hatch, R. C., Mi, J., Iversen, B. R. & Hofmann, P. Simultaneous quantization of bulk conduction and valence states through adsorption of nonmagnetic impurities on Bi2Se3. Phys. Rev. Lett. 107, 086802 (2011).
27. Zangwill, A. Physics at Surfaces p. 21 (Cambridge Univ. Press, Cambridge, 1988).
28. Feng, J. et al. Electronic structure and enhanced charge-density wave order of monolayer VSe2. Nano Lett. 18(7), 4493 (2018).
29. Wang, Y. & Gedik, N. Circular dichroism in angle-resolved photoemission spectroscopy of topological insulators. Phys. Stat. Solid. 7, 64–71 (2013).
30. Scholz, M. R. et al. Reversal of the circular dichroism in angle-resolved photoemission from Bi2Te3. Phys. Rev. Lett. 110, 216801 (2013).
31. Miao, L. et al. Observation of a topological insulator Dirac cone reshaped by non-magnetic impurity resonance. npj Quantum Mater. 3, 29 (2018).
32. Schlenk, T. et al. Controllable magnetic doping of the surface state of a topological insulator. Phys. Rev. Lett. 110, 126804 (2013).
33. Yilmaz, T. et al. Photon energy and polarization-dependent electronic structure of Cr-doped Bi2Se3. Phys. Rev. Mater. 4, 024201 (2020).
34. Sánchez-Barriga, F. et al. Nonmagnetic band gap at the Dirac point of the magnetic topological insulator (Bi1−xMnx)2Se3. Nat. Commun. 7, 10559 (2016).
35. Yilmaz, T. et al. Gap-like feature observed in the non-magnetic topological insulators. J. Phys. Condens. Matter 32, 145503 (2020).
36. Li, Z. et al. Realization of flat band with possible nontrivial topology in electronic Kagome lattice. Sci. Adv. 4, eaau4511 (2018).
37. Bianchi, M. et al. Coexistence of the topological state and a two-dimensional electron gas on the surface of Bi2Se3. Nat. Commun. 1, 128 (2010).
advisory committee of HISOR) Proposal number 19BG041). T.Y. thanks Professor A.V. Balatsky for useful discussions.

**Author contributions**

T.Y. conceived and designed the experiments. T.Y. prepared the samples and performed the photoemission experiments with the help from K. Kaznatcheev, E.V., and B.S. E.F.S. and K.S. performed the ARPES experiments at HISOR. X.T. conducted the STM measurements. Z.D. and J.T.S. performed μLEED measurements. S.H. and K. Kisslinger performed HAADF-STEM experiments. T.Y. analyzed the experimental results and wrote the manuscript with contribution from E.F.S., B.S., K. Kaznatcheev, and E.V.

**Competing interests**

The authors declare no competing interests.

**Additional information**

Supplementary information is available for this paper at [https://doi.org/10.1038/s43246-020-00115-w](https://doi.org/10.1038/s43246-020-00115-w).

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Peer review information Primary handling editor: John Plummer

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