

Robust Orbital-Selective Flat Bands in Layered Transition-Metal Oxyhalides at Room Temperature

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Flat electronic bands, which amplify electron correlations by quenching kinetic energy, provide an ideal foundation for exotic quantum phases. However, prevailing strategies—including geometrically frustrated lattices, moiré superlattices, and heavy-fermion systems—often suffer from inherent trade-offs among robustness, tunability, and orbital control, thereby limiting their broad applicability. Here, we report the experimental discovery of intrinsic orbital-selective flat bands in the layered transition-metal oxyhalide family MOX_2 ($M = \text{Nb, Ta}$; $X = \text{Cl, Br, I}$), directly observed by angle-resolved photoemission spectroscopy at room temperature. Importantly, this materials platform exhibits pronounced tunability: The flat-band bandwidth and correlation strength can be systematically tuned via halogen substitution, while the Fermi level is controllable through electrostatic gating and surface charge transfer. Remarkably, the flat band remains stable upon dimensional reduction, persisting from the bulk crystal down to the few-layer limit. Our theoretical analysis reveals that this flat band transcends simple lattice geometry, originating instead from an orbital-driven synergetic Su-Schrieffer-Heeger-Lieb mechanism: a cooperative interplay between quasi-one-dimensional $M-d_{z^2}$ Su-Schrieffer-Heeger chains and a two-dimensional $M-d_{x^2-y^2}$ Lieb-like sublattice, reinforced by Peierls dimerization. Together, these findings establish layered transition-metal oxyhalides as a versatile materials-by-design platform for systematically engineering intrinsic flat bands via chemical, electronic, and dimensional control, which provides a robust pathway to exploring room-temperature flat-band physics.

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Flat electronic bands (FBs) have emerged as a central motif in quantum materials research, providing a unifying platform where strong electron-electron correlations intertwine with a nontrivial topology [1–8]. Within these FBs, the suppression of electronic kinetic energy amplifies

Coulomb interactions, so that even moderate on-site repulsion can drive the system far from conventional Fermi-liquid behavior [9]. Indeed, the dominance of the on-site Coulomb interaction U over electron kinetic energy t offers a universal mechanism for correlation effects, giving rise to unconventional superconductivity [10–15], Mott phase [16–20], non-Fermi-liquid transport [21,22], and topologically ordered phases hosting integer or fractional excitations, as first realized in Landau levels [23–25] and more recently in moiré materials [26–31]. Known routes to realizing FBs can be broadly grouped into three categories: (i) geometrically frustrated lattices, where quantum destructive interference suppresses hopping [32–36], as experimentally observed in kagome [22,37–40], Lieb [41], and pyrochlore [21,42] lattices; (ii) moiré superlattices, which flatten bands via momentum-space band folding

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[43–46]; and (iii) correlated heavy-fermion systems, where orbital localization narrows the bandwidth [47–50]. Despite their distinct microscopic origins, these approaches share the essential feature of quenching kinetic energy to amplify electron correlations.

However, all established strategies face intrinsic trade-offs between robustness, tunability, and orbital selectivity. moiré FBs engineered via momentum-space folding as well as quantum destructive interference-induced FBs are vulnerable to broadening from interlayer coupling or disorder, often requiring cryogenic stabilization [7,14,36,51]. Correlation-driven FBs in heavy-fermion systems, though profoundly localized, suffer from the extreme atomic localization of f orbitals, which severely limits fine-tunability [8,9,48]; furthermore, the dual nature of f orbitals—partially localized and strongly hybridized with conduction d electrons—blurs the distinction between localized and itinerant states. Symmetry-protected FBs demand preservation of delicate crystalline symmetries that can be fragile to disorder or strain in realistic settings [5,52]. The trilemma of achieving strong orbital selectivity, room-temperature stability, and multiparameter control has hindered the development of tunable FB platforms for correlated-electron studies. Consequently, an ideal FB material that simultaneously fulfills these requirements has yet to be realized.

In this work, we resolve this challenge by identifying an intrinsic orbital-selective FB mechanism in the layered van der Waals transition-metal oxyhalides MOX_2 ($M = \text{Nb}, \text{Ta}$; $X = \text{Cl}, \text{Br}, \text{I}$). Their crystal structure—featuring Peierls-dimerized chains and a Lieb-like lattice—selectively quenches kinetic energy in $M d_{z^2}$ orbitals through bond-length disproportionation and electron localization. This yields a momentum-independent isolated FB across the material family, as directly observed by angle-resolved photoemission spectroscopy (ARPES). Crucially, this platform offers multidimensional tunability: the FB bandwidth and correlation strength can be modulated via halogen substitution ($\text{Cl} \rightarrow \text{Br} \rightarrow \text{I}$), while the Fermi level can be tuned through carrier regulation. Owing to their intrinsic two-dimensional nature and weak interlayer coupling, the FB persists from bulk crystals down to few-layer flakes and remains stable at room temperature without the need for moiré engineering. Density functional theory (DFT) calculations attribute its microscopic origin to the hybridization between the quasi-1D $M d_{z^2}$ Su-Schrieffer-Heeger (SSH) chain and the 2D Lieb-like O (p_x)- M ($d_{x^2-y^2}$)- X (p_y) network which is further reinforced by Peierls distortion. The flat bands do not involve all Nb d orbitals collectively but emerge selectively from specific orbitals whose lattice connectivity supports the cooperative SSH-Lieb mechanism. The orbital selectivity originates purely from the geometric orbital-lattice connectivity of the SSH-Lieb network, distinct from correlation-driven differentiation in a many-body sense. This orbital selectivity

confines electrons to the flat band, highlighting their localized nature, while the orbital geometry provides an additional degree of freedom to facilitate flat-band formation and tune correlation strength [53]. By combining orbital selectivity, room-temperature stability, and multiparameter tunability within an intrinsic material platform, MOX_2 establishes a new paradigm for engineering correlated quantum matter beyond the constraints of existing FB systems.

Figure 1(a) shows that NbOCl_2 crystallizes in the monoclinic space group C_2 (Supplemental Material Fig. S1 [54]), forming a quasi-2D framework with weak van der Waals stacking along the a axis [55–58]. Within each layer, edge-sharing NbOCl_2 -distorted octahedra form chains along the b axis through O bridges and connect along the c axis via Cl bridges [Fig. 1(b)], giving rise to a strong in-plane anisotropy. The lattice exhibits two cooperative structural distortions: Along the c axis, Peierls distortion dimerizes the Nb chains into alternating short (d_1) and long (d_2) Nb–Nb bonds [Fig. 1(c)]; along the b axis, Nb ions undergo a collective off-center displacement toward one bridging oxygen, producing alternating Nb–O bond lengths [Fig. 1(b)]. These coupled distortions break inversion symmetry, stabilize a low-energy polar configuration, and generate a robust in-plane ferroelectric polarization at room temperature [59], accompanied by pronounced second-harmonic generation and other optical responses in such a system [57,58,60,61]. Owing to the exceptionally weak interlayer coupling, large-area monolayer and few-layer flakes can be readily obtained by mechanical exfoliation and retain their structural and electronic integrity at room temperature over extended periods, making them ideal for surface-sensitive probes [57].

Examining the lattice geometry projected onto the b - c plane reveals a network closely resembling a Lieb lattice. Although the chlorine atoms in NbOCl_2 are slightly displaced from the atomic plane, their bonding environment allows the system to be effectively modeled as a Lieb-like lattice [Fig. 1(b)], capturing the essential symmetry and hopping pathways. Figure 1(d) shows the contrast between the kagome lattice and the Lieb lattice, where corner-sharing triangles in the kagome lattices yield FBs at the band edges, whereas Lieb lattices host them near the spectrum center in idealized tight-binding models. For Nb^{4+} ($4 d^1$), the effects of the crystal field lower the d_{z^2} orbital relative to the other d orbitals, making it the main contributor to the states near E_F [Fig. 1(e)] [58]. In the absence of lattice distortion, the configuration of the d_{z^2} electrons would produce a simple metallic state with one itinerant electron in the orbital d_{z^2} . However, Peierls dimerization along the Nb–Cl–Nb chains splits the d_{z^2} manifold into bonding (BB) and antibonding (AB) states, thereby opening a gap and driving a metal-insulator transition. The corresponding schematic density of states shows a fully occupied BB and an empty AB, consistent with the insulating ground state [57]. This orbital-lattice

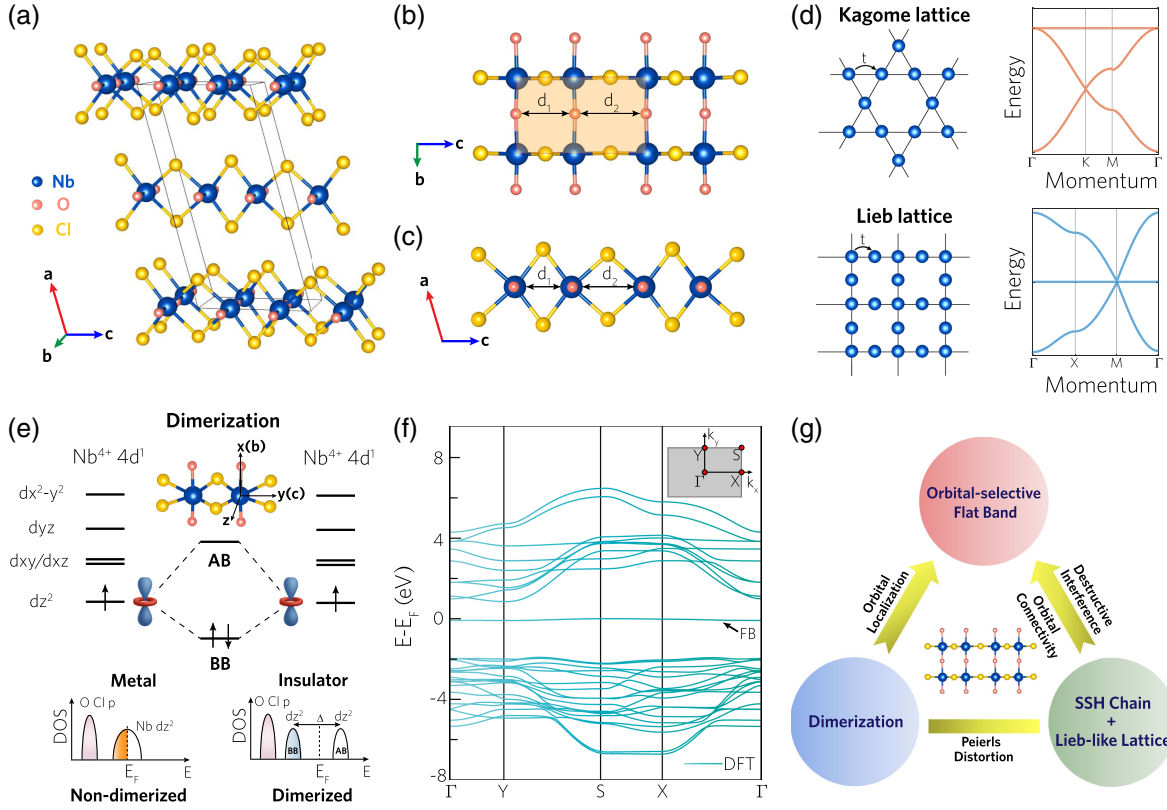


FIG. 1. Crystal structure, cooperative Peierls distortions, and emergence of an orbital-selective flat band in NbOCl_2 . (a)–(c) Crystal structure of NbOCl_2 viewed along various crystallographic directions. Nb (blue), O (red), and Cl (yellow) atoms form quasi-2D layers stacked along the a axis. Within each layer, alternating Nb–Nb bond lengths d_1 and d_2 highlight pronounced dimerization. (d) Schematic lattices and tight-binding band structures of kagome and Lieb systems, where symmetry-protected flat bands arise from destructive interference. Owing to distinct lattice geometries, the flat-band positions differ between these two systems. NbOCl_2 adopts a related Lieb-like lattice configuration. (e) Orbital energy levels of Nb^{4+} ($4d^1$) atoms and bonding configurations in the dimerized state. (f) DFT band structure of the monolayer dimerized NbOCl_2 . The rectangular BZ is defined with k_x along the Γ –X direction (parallel to the b axis) and k_y along the Γ –Y direction (parallel to the c axis). The narrow, well-isolated FB near the Fermi level remains robust in the ultrathin limit, with the Fermi level inside the gap between the FB and dispersive bands. (g) Schematic of the synergetic SSH-Lieb mechanism for flat-band formation. The cooperative interplay between quasi-1D SSH chains—driven by Peierls dimerization to enhance orbital localization—and 2D Lieb-like lattice interference—which quenches kinetic energy—stabilizes the robust orbital-selective flat band.

reconstruction provides a natural multicomponent route to kinetic-energy quenching, as further corroborated by DFT and Wannier-function analysis in the following text. Figure 1(f) presents the DFT band structure for dimerized monolayer NbOCl_2 , with the rectangular Brillouin zone (BZ) defined by k_x along the Γ –X direction (parallel to the b axis) and k_y along the Γ –Y direction (parallel to the c axis). The calculations reveal a narrow, well-isolated FB predominantly derived from d_{z^2} orbitals across the entire BZ. Here, the energy is referenced to the valence-band maximum, which is set to 0 eV, placing the FB at the top of the valence manifold, while the Fermi level lies inside the band gap between the FB and the higher-lying conduction bands, consistent with previous theoretical work [62]. The persistence of this FB in the monolayer DFT calculation underscores its intrinsic nature. Figure 1(g) summarizes the synergetic mechanism identified in this work: Peierls

dimerization structurally suppresses interdimer d_z hopping to enhance quasi-one-dimensional orbital localization, while the hybrid SSH-Lieb orbital connectivity enforces two-dimensional destructive interference. This synergetic interplay, as detailed below via Wannier analysis, stabilizes an orbital-selective flat band that is both structurally and electronically robust, providing a natural platform for exploring correlation-driven physics.

To directly probe the electronic structure and verify the predicted orbital-selective flat band, ARPES measurements were performed on high-quality NbOCl_2 single crystals at room temperature to avoid charging effects. Figure 2(a) shows constant energy contours acquired with linear horizontal (LH) polarized light at $h\nu = 66$ eV, spanning binding energies from $E_B = 2.2$ to 7.5 eV. The pronounced anisotropy in spectral weight along the Γ –X and Γ –Y directions reflects the material’s low crystallographic

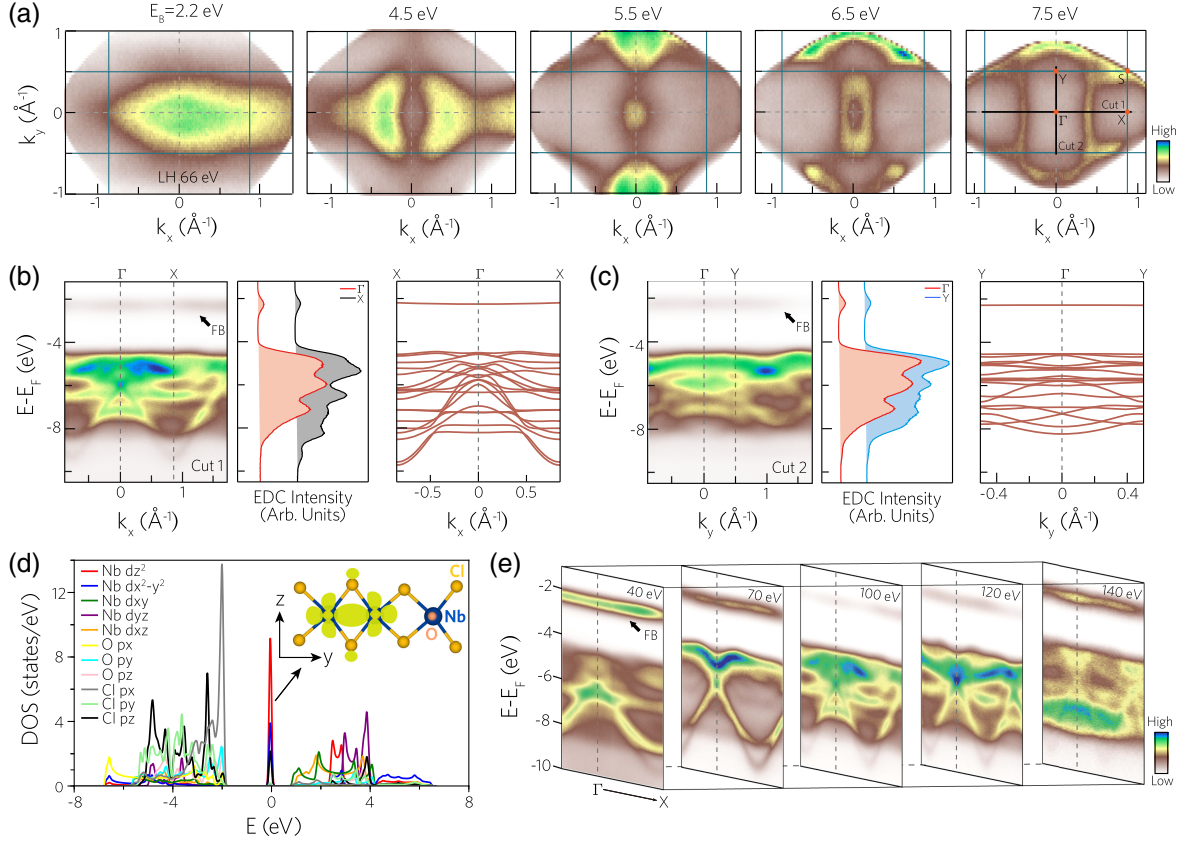


FIG. 2. Momentum-independent flat band and its orbital character in NbOCl_2 characterized by ARPES and DFT. (a) Constant-energy contours of NbOCl_2 measured at room temperature ($T = 300$ K) using photon energy $h\nu = 66$ eV, shown at representative binding energies $E_B = 2.2, 4.5, 5.5, 6.5$, and 7.5 eV. High-symmetry points of the BZ are marked by orange circles. Momentum cuts (cut 1 and cut 2) indicated by black lines are chosen for detailed analysis. (b) Band structures along the Γ -X direction (cut 1) recorded at $h\nu = 100$ eV. The middle shows the corresponding EDCs integrated at Γ and X points. The right presents the DFT band structure along the same path for comparison. (c) The same as (b), but along the Γ -Y direction (cut 2). (d) Orbital-resolved PDOS for monolayer NbOCl_2 . The inset shows the real-space charge density of the FB's wave function from DFT. (e) Photon-energy-dependent ARPES spectra along the Γ -X direction, taken at $h\nu = 40, 70, 100, 120$, and 140 eV, revealing the k_z dependence of the valence bands.

symmetry. High-symmetry points (blue circles) and momentum cuts (cut 1 and cut 2, gray lines) were selected for detailed band dispersion analysis. Figure 2(b) presents the band dispersion along the Γ -X direction (cut 1), measured at $h\nu = 100$ eV. A remarkably narrow, nearly dispersionless feature appears near $E_B \sim 2.2$ eV, denoted as the FB. Energy distribution curves (EDCs) integrated at Γ and X points show clear peaks at this energy, confirming the FB's momentum-independent character. DFT calculations for monolayer NbOCl_2 along the same path (right) reproduce the FB alongside more dispersive valence-band states. In the theoretical band-structure plots, the energy axis is referenced to the valence-band maximum, which is set to 0 eV, whereas all ARPES spectra are referenced to the Fermi level. For a direct comparison, a rigid energy alignment is applied between the theoretical and experimental spectra, without altering the relative band dispersions. The apparent absence of conduction band states in ARPES reflects the insulating nature of NbOCl_2 and the known tendency of semilocal DFT to underestimate band

gaps. With this energy referencing, the DFT calculations show excellent agreement with the experimental observations. Figure 2(c) shows the momentum cut along the Γ -Y direction (cut 2) and the corresponding EDCs at the Γ and Y point. The FB retains its extreme flatness, while the valence bands below exhibit reduced dispersion and narrower bandwidth compared to those along the Γ -X direction, indicating anisotropic conductivity [59,63]. This is likely originated from the anisotropic Cl- and O-mediated hopping pathways and the different electronegativity between Cl and O atoms. Corresponding DFT calculations [Fig. 2(c), right] also confirm the flatness feature for the d_{z^2} -dominated FB along the Γ -Y direction, underscoring its isotropic localization in the whole momentum space. To further clarify the orbital character of the FB, Fig. 2(d) shows the orbital-resolved projected density of states (PDOS) for monolayer NbOCl_2 . The FB near the valence-band maximum (VBM) is dominated by Nb d_{z^2} orbitals, with a finite admixture of $d_{x^2-y^2}$ and Cl/O p states, because the low-symmetry environment breaks the strict

orthogonality between d_{z^2} and $d_{x^2-y^2}$ orbitals, allowing Cl-mediated hybridization that imparts a finite $d_{x^2-y^2}$ component near E_F . The inset displays the real-space charge density of the FB's wave function from DFT, highlighting their strong confinement around dimerized Nb sites and the toroidal in-plane character of the d_{z^2} orbital. Photon-energy-dependent measurements [Fig. 2(e)] trace the Γ -X dispersion over a wide k_z range (40–140 eV). While the FB intensity is stronger at lower photon energies, its binding energy remains fixed with negligible k_z dependence, confirming its intrinsic two-dimensional nature in NbOCl₂. In contrast, the spectral weight of some deeper valence bands varies with photon energy due to matrix element effects. Additional photon-energy-dependent constant energy contours and momentum cuts showing the invariance of the flat-band binding energy across multiple k_z values are provided in Supplemental Material Fig. S2 [54]. Remarkably, these ARPES measurements reveal that the flat band remains entirely dispersionless along all three momentum directions (k_x , k_y , and k_z). Furthermore, temperature-dependent ARPES and related EDC analyses (Supplemental Material Fig. S3 [54]) indicate that the flat-band energy position and spectral weight exhibit negligible variation across the studied temperature range, persisting even through the reported ferroelectric-antiferroelectric crossover [64], thereby supporting its exceptional room-temperature stability.

To assess the universality of this orbital-selective FB mechanism, we extended our investigation to the broader transition-metal oxyhalide family, synthesizing the 5d analog TaOCl₂ [65] and the halogen series NbOBr₂ and NbOI₂ [60,66]. Figure 3 presents a comparative spectroscopic study of the entire family. Figures 3(a) and 3(b) directly contrast the electronic structures of NbOCl₂ and TaOCl₂ along high-symmetry paths. Similar measurements for NbOI₂ are provided in Supplemental Material Fig. S4(a) [54]. Across all compounds, ARPES spectra acquired at $h\nu = 70$ eV combine momentum cuts obtained with LH and linear vertical (LV) polarizations. The experimental results show remarkable consistency with the corresponding monolayer DFT band calculations. Nearly momentum-independent FBs are clearly resolved across the entire BZ. Figures 3(c) and 3(d) show the momentum cuts along the Γ -X direction and the Γ -Y directions for all four compounds under LH polarization, respectively. In NbOCl₂, TaOCl₂, and NbOBr₂, the flat band remains extremely flat along both directions. In contrast, in NbOI₂, the FB remains nearly flat along Γ -X but becomes more dispersive along the Γ -Y direction, as further evidenced by the EDC analysis in Fig. 3(e). Notably, NbOI₂ displays a multiband feature near the FB along Γ -Y indicated by the red arrows. To elucidate the microscopic origin of this feature, we performed orbital-projected DFT calculations [Supplemental Material Fig. S4(b) [54]]. These calculations reveal that the feature arises from the

coexistence of two orbitally distinct bands: an uppermost flat band corresponding to the Nb d_{z^2} orbital and a band immediately below derived from the I p_x orbital [Supplemental Material Figs. S4(e) and S4(h) [54]].

In order to probe the orbital symmetry, we also performed polarization-dependent measurements on these materials (Supplemental Material Figs. S5–S7 [54]). A universal trend is observed: The FB intensity is maximized under LH polarization but strongly suppressed under LV polarization near the Γ point in both directions (Supplemental Material Fig. S5 [54]). While the spectral intensity for the FB is similarly prominent along the Γ -X direction, it decreases toward and beyond the Y point along the Γ -Y direction [Figs. 3(d) and 3(e)]. These trends indicate that the FB is derived primarily from the even-parity d_{z^2} orbital. Given the near-rotational symmetry of the d_{z^2} state in the $x-y$ plane, comparable intensities at X and Y would be expected if the d_{z^2} orbital were the only contributor. The observed anisotropy-enhanced spectral weight of the FB at X versus its suppression at Y indicates a minor admixture of the $d_{x^2-y^2}$ orbital, which redistributes the matrix elements, which is consistent with the orbital-resolved PDOS as shown in Fig. 2(d) and Supplemental Material Fig. S8 [54]. Furthermore, the full set of EDC stacks and Lorentzian fits of the peak positions across Γ -X and Γ -Y show negligible peak shifts (Supplemental Material Fig. S9 [54]). Extending this analysis to NbOBr₂ and NbOI₂, we summarized the extracted bandwidths in Fig. 3(f). The bandwidths for NbOCl₂, TaOCl₂, and NbOBr₂ are below 75 meV, in agreement with theoretical predictions (approximately 80 meV) [62]. However, the FB in NbOI₂ displays a larger bandwidth compared to the other compounds, reaching approximately 240 meV along the Γ -Y direction [Fig. 3(f)]. This evolution is governed by the halogen p orbitals: The diffuse I $5p$ orbitals possess a much larger spatial extent and lower electronegativity compared to Cl $3p$ or Br $4p$. This significantly enhances the p - d hybridization along the Peierls-type chain. Despite the bandwidth enhancement in NbOI₂ along the Γ -Y direction, the extremely narrow bandwidth of the FB in the other compounds gives rise to a pronounced peak in the density of states [Fig. 2(d) and Supplemental Material Fig. S8 [54]], highlighting the potential role of strong electron correlations. Importantly, the FB in the present system remains strictly dispersionless along all measured momentum directions (Supplemental Material Fig. S7 [54]), in sharp contrast to FBs originating from geometry-frustrated lattices such as the kagome lattice. In kagome systems, flat bands are protected by destructive interference only in the ideal nearest-neighbor tight-binding limit; in real materials, longer-range hopping, interlayer coupling, or hybridization with dispersive bands generally lifts the exact flatness, resulting in direction-dependent dispersion [38]. This reflects the fact that the corresponding eigenstates are no longer strictly compact localized states. By contrast, the

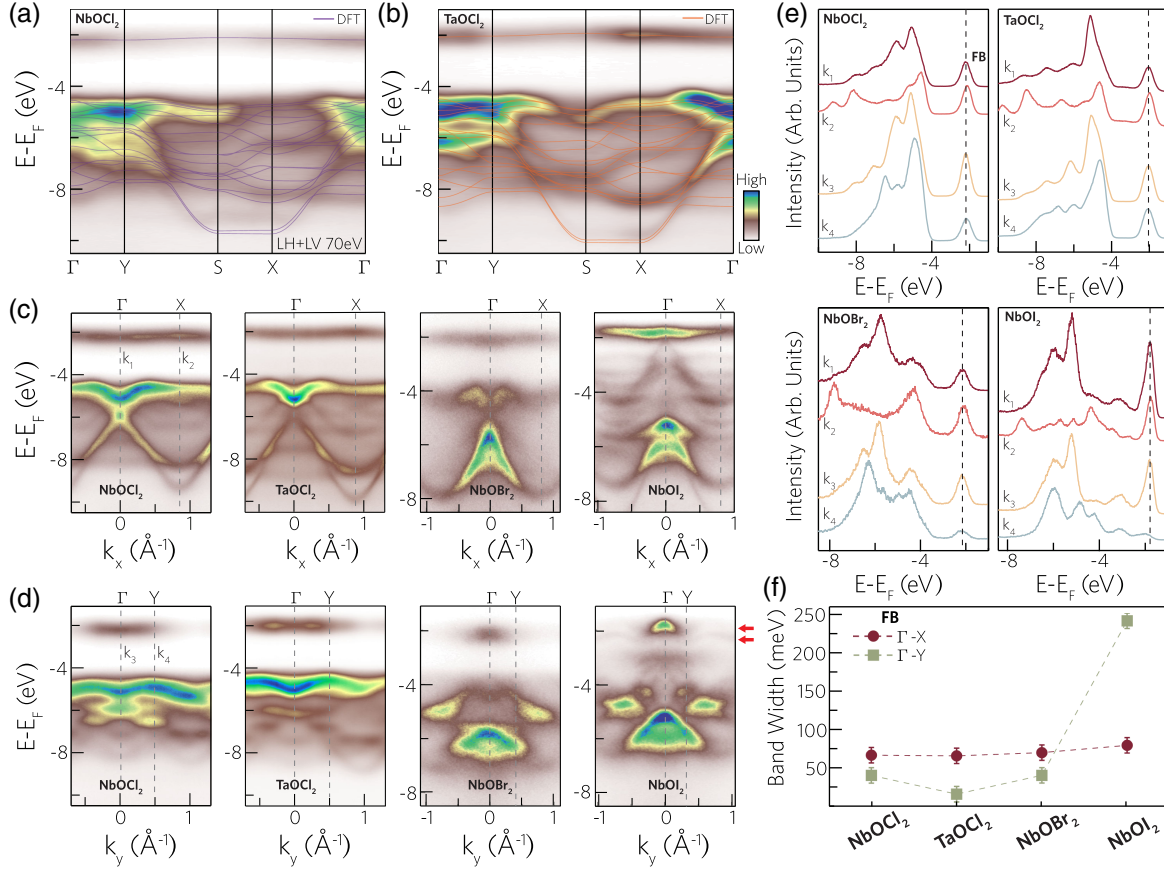


FIG. 3. Universality and tunability of orbital-selective flat bands across the transition-metal oxyhalide family. (a),(b) Experimental band structures of NbOCl₂ (a) and TaOCl₂ (b) measured along high-symmetry directions, overlaid with DFT calculations (colored lines). The ARPES spectra were obtained using combined LH and LV polarizations at $h\nu = 70$ eV to access the full band manifold. (c), (d) High-resolution momentum cuts along the Γ -X (k_x) direction (c) and Γ -Y (k_y) direction (d) for NbOCl₂, TaOCl₂, NbOBr₂, and NbOI₂. Vertical dashed lines indicate the momenta (k_1 – k_4) at which the EDCs in (e) were extracted. The flat-band dispersion is observed across the series, with a pronounced increase in bandwidth along Γ -Y for NbOI₂. Notably, a distinct multiband feature is observed near FB in NbOI₂ along Γ -Y, marked by red arrows. Further analysis is provided in Supplemental Material Fig. S4 and Sec. IV [54]. (e) Corresponding EDCs for the four compounds, with the FB peak positions marked by vertical dashed lines. (f) Summary of the FB bandwidths extracted from EDC stacks along both directions. Details are provided in Supplemental Material Fig. S9 and Sec. VIII [54]. While the bandwidth along Γ -X (dark red circles) remains nearly constant across the family, a substantial enhancement along Γ -Y (olive green squares) is observed for NbOI₂, demonstrating the effective modulation of correlation strength via the spatial extent of the halogen orbitals.

robust momentum-independent flatness observed here indicates a distinct physical mechanism beyond pure geometric destructive interference.

Regarding energy positions relative to E_F , the FB in TaOCl₂ is slightly closer to the Fermi level (approximately 2.05 eV) than in NbOCl₂ (approximately 2.2 eV). In NbOBr₂, the FB position is similar to NbOCl₂, whereas in NbOI₂, the FB lies closer to E_F (approximately 1.8 eV), suggesting an enhanced tunability of the flat band and electrical conductivity within this material family. Another notable feature in NbOI₂ is the band crossing and gap closure between the FB and the underlying valence band. This behavior can be traced to the evolution of the halogen's chemical properties from Cl to I. First, the

substantially lower electronegativity of iodine compared to chlorine raises the on-site energy of the I 5*p* orbitals, positioning them much closer to the Nb 4*d* states. Second, the significantly larger spatial extent of the I 5*p* orbitals results in much stronger *p*-*d* hybridization with the Nb 4*d* orbitals. This enhanced hybridization not only increases the overall bandwidth, but also further shifts the top of the iodine-derived *p* bands upward through level repulsion [Supplemental Material Fig. S4(b) [54]]. These combined effects—elevated orbital energies and hybridization-induced broadening—cause the valence bands below the FB in NbOI₂ to shift upward by approximately 2 eV relative to NbOCl₂, thereby bridging the preexisting energy gap. Taken together, these comparative results demonstrate

that the orbital-selective FB mechanism is robust across the MOX_2 family, while the degree of flatness and the relative band position can be systematically tuned via orbital extent.

Beyond static chemical substitution, dynamic control of the electronic states is crucial for functional quantum devices. We, therefore, explored the active tunability of the flat bands in $NbOCl_2$ via electrostatic gating (Supplemental Material Fig. S10 [54]) and surface doping (Supplemental Material Fig. S11 [54]). Transport measurements on back-gated $NbOCl_2$ field-effect transistors reveal nearly linear output characteristics at 200 and 295 K, confirming high-quality Ohmic contacts [Supplemental Material Fig. S10(b) [54]]. Consistent with the large effective mass and orbital localization expected from a flat-band system, the extracted field-effect mobility is relatively low ($\mu_{FE} \approx 0.016 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$). Despite this, the system exhibits clear gate tunability. Temperature-dependent conductivity follows a strict Arrhenius law ($R^2 > 0.997$), indicating thermally activated transport. Crucially, as the gate voltage V_g increases from 10 to 50 V, the activation energy E_a decreases monotonically from 121 to 110 meV [Supplemental Material Fig. S10(c) [54]]. Conversely, applying a negative gate voltage tunes the Fermi level toward the middle of the band gap, suppressing the channel current to a negligible level. This modulation demonstrates that the Fermi level can be electrostatically tuned between the midgap and the mobility edge, confirming that $NbOCl_2$ is a gate-tunable correlated insulator. To visualize the evolution of the flat band under carrier injection, we also performed *in situ* rubidium (Rb) surface deposition (Supplemental Material Fig. S11 [54]). Unlike the simple rigid-band shifts typically observed in conventional semiconductors, electron doping in $NbOCl_2$ induces a complex spectral reconstruction [Supplemental Material Figs. S11(a) and S11(b) [54]]. As the Rb dosage increases [Supplemental Material Fig. S11(c) [54]], we observe the flat band moves toward the Fermi level E_F and a spectral weight transfer from the intrinsic flat band (P_1) to a new emergent chemically induced state (P_2) at lower binding energy [Supplemental Material Figs. S11(d) and S11(e) [54]]. Notably, in the heavily doped sample, the emergence of this new state in the EDCs extends the spectral weight to very shallow binding energies, indicating a significant enhancement in the system's metallicity [red line in Supplemental Material Fig. S11(d) [54]]. This renormalization of surface electronic states highlights the strong susceptibility of the orbital-selective flat bands to carrier density, further establishing transition-metal oxyhalides as a practical materials-design platform for engineering tunable correlated physics in quantum materials.

It has been theoretically predicted that the electronic properties of $NbOCl_2$ remain nearly unchanged from the three-dimensional bulk down to the two-dimensional

limit—an exceptionally rare behavior among layered materials [67–69]. Having confirmed the universality of the orbital-selective FB in bulk samples, we next experimentally examine its persistence in the few-layer regime. Notably, this material family has been shown to exhibit good environmental stability under ambient conditions, with Raman signatures remaining unchanged over extended periods of air exposure [57,70], ensuring that the intrinsic physical properties are preserved during device fabrication and measurement. Figure 4 shows that the FB in $NbOCl_2$ is remarkably robust upon thickness reduction. For micro-ARPES measurements on few-layer samples, we fabricated graphene/ $NbOCl_2$ /hBN heterostructures in which mechanically exfoliated few-layer $NbOCl_2$ flakes were encapsulated by graphene and supported by hBN with patterned Au contacts on a SiO_2/Si substrate [Fig. 4(a)]. In this device configuration, graphene serves primarily as a protective capping layer and as an electrically conductive layer to mitigate charging effects during photoemission measurements. The optical image [Fig. 4(b)] reveals the morphology of the stacking heterostructure, while atomic force microscopy confirms a representative flake thickness of approximately 2 nm [three layers; Fig. 4(c)]. ARPES spectra along the Γ -X direction for the few-layer device [Fig. 4(d)] reveal a sharp, dispersionless feature at approximately 2 eV below the Fermi level, as pointed out by the orange arrow. For comparison, graphene alone [Fig. 4(e)] has been measured to show no such FB feature and confirm its origin from $NbOCl_2$. Bulk $NbOCl_2$ samples measured under identical conditions [Fig. 4(f)] exhibits an almost identical FB. EDCs at the Γ point [Fig. 4(g)] further highlight the close similarity between few-layer (purple) and bulk (red) spectra, with only minimal energy shifts. This negligible change from bulk to few-layer suggests that the exceptional properties of $NbOCl_2$, including its strong optical responses that are likely preserved even in the monolayer limit [57]. While our data conclusively demonstrate the persistence of the flat band in few-layer flakes, its stability in the strict monolayer limit is strongly suggested by the extreme 2D nature of the electronic structure observed in bulk [Fig. 2(e)] and the excellent agreement with monolayer DFT calculations. The negligible evolution from bulk to trilayer further supports the notion that the monolayer will host the same robust flat band. In practical terms, this means that devices based on relatively easy-to-fabricate few-layer flakes can already deliver performance comparable to that of ideal monolayers, substantially lowering the barrier for applications.

Having established the good agreement between ARPES measurements and DFT calculations (Figs. 2 and 3), we now turn to reveal the microscopic origin of the FB. Earlier theoretical studies highlighted two essential ingredients: (i) orbital mixing between Nb and Cl atoms and (ii) the critical role of Peierls distortion in driving the band-gap

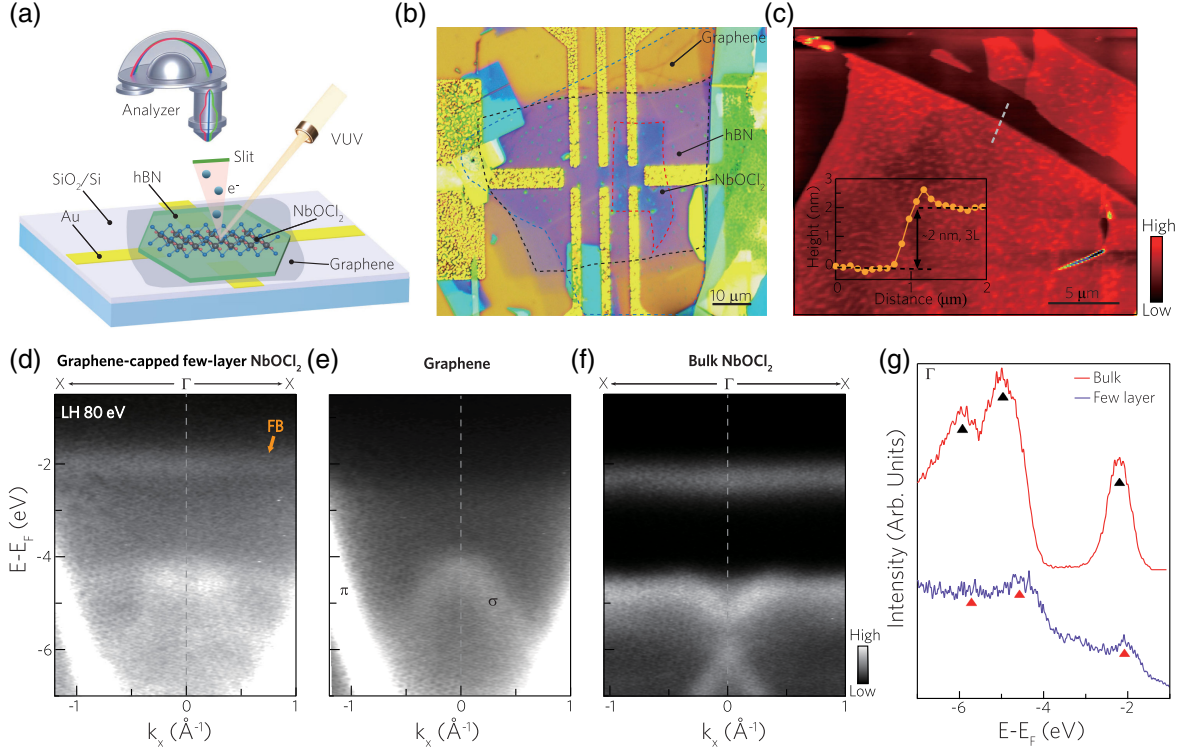


FIG. 4. Persistence of the flat band in few-layer NbOCl₂ revealed by ARPES. (a) Schematic of the micro-ARPES setup for NbOCl₂/graphene heterostructures, with few-layer NbOCl₂ encapsulated by graphene and supported on hBN and Au contacts on a SiO₂/Si substrate. (b) Optical image of the device, showing patterned Au electrodes, hBN support, graphene encapsulation, and NbOCl₂ flakes. (c) Atomic force microscopy image of the NbOCl₂ flake with height profile (inset), confirming a thickness of approximately 2 nm (three layers). The position of the profile is marked by the gray dashed line. (d)–(f) ARPES dispersions along Γ -X for graphene-capped few-layer NbOCl₂ (d), bare graphene (e), and bulk NbOCl₂ (f), measured with photon energy $h\nu = 80$ eV under LH polarization. The FB in the few-layer NbOCl₂ sample is marked by an orange arrow in (d). (g) EDCs at the Γ point comparing few-layer (purple) and bulk (red) NbOCl₂, showing that the flat band persists in reduced thickness.

opening and emergent chirality [59,62,68,71]. Compared to the previous works, we propose here a unified microscopic model that captures the interplay between distinct orbital connectivities and the effective dimensionality of the hopping processes. The full Wannierization and tight-binding analysis reveal that the flat-band formation in NbOCl₂ arises from a hybrid SSH-Lieb network. This mechanism uniquely exploits the synergy between quasi-1D orbital chains and the destructive interference of a 2D Lieb-like sublattice. First, to explore the influence of the dimerization process, we compare a 2×1 Peierls-distorted supercell in real space with a 2×1 nondistorted supercell generated by doubling the unit cell along the y axis—the direction of the Peierls distortion. This construction allows a direct comparison between distorted and undistorted states. We further investigate the evolution of lattice distortion by gradually shifting atomic coordinates of each atom between the nondimerized high-symmetry lattice (A) and dimerized low-symmetry lattice (B), following the relation $F = xA + yB$ with $x + y = 1$ as defined in Ref. [72]. The two limited cases $x = 1, y = 0$ and $x = 0, y = 1$ correspond to the undistorted and fully distorted

structures, respectively, while intermediate values represent virtual snapshots of the dimerization process (Supplemental Material Fig. S12 [54]). Following the progression of the Peierls distortion, we can map the gap-opening mechanism. This interpolation reveals that even a modest distortion ($x = 0.75, y = 0.25$) breaks the $-y/y$ mirror symmetry in real space and induces a small gap along the Y - S path of the BZ between two bands near the Fermi level [Supplemental Material Fig. S12(b) [54]]. From this point onward, the gap-opening process proceeds inexorably, culminating in the fully developed dimerized lattice (B). This analysis establishes that the gap formation is driven predominantly by the emergence of Nb atom dimerization. Throughout the A - B evolution, the most pronounced macroscopic consequence is this symmetry-breaking gap opening, whereas the overall topology of the band structure remains largely preserved.

We next investigate the orbital character of the A and B phases to evaluate the crucial role of the orbital hybridization in driving the FB formation. Figures 5(a) and 5(b) present the orbital-projected band structures for both phases, revealing that the 28 bands above and below the

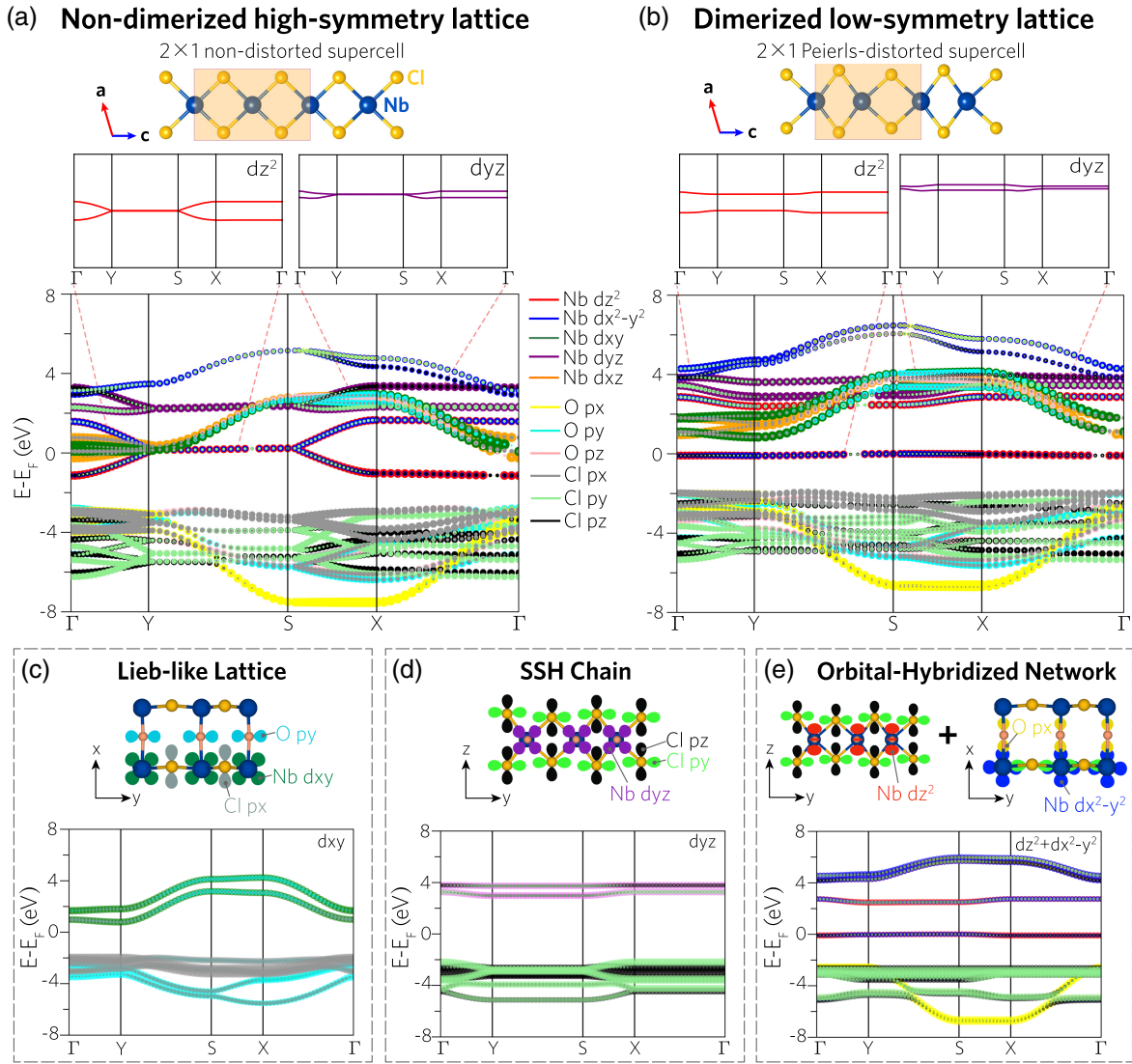


FIG. 5. Microscopic origin of the orbital-selective flat band arising from the synergy between SSH-Lieb physics and Peierls distortion. (a) Orbital-projected band structure of monolayer NbOCl_2 in the nondimerized high-symmetry lattice. The color scale indicates relative contributions from Nb d and Cl/O p orbitals. The inset highlights the dispersive nature of Nb d_{z^2} and d_{yz} bands in certain momentum regions. (b) The same as in (a) but for the dimerized low-symmetry lattice. Because of the Peierls distortion, the d_{z^2} and d_{yz} -derived bands exhibit enhanced flatness relative to the nondimerized state but open up a band gap with different sizes, indicating an origin predominantly rooted in orbital character rather than structural effects alone. (c)–(e) Schematic orbital-lattice building blocks and corresponding tight-binding band models for representative orbital connectivity in the dimerized low-symmetry lattice. (c) The Lieb-like sublattice, primarily formed by Nb d_{xy} , Cl p_x , and O p_y orbitals. (d) The quasi-1D SSH chain, originating from the Nb d_{yz} , Cl p_y , and p_z orbitals. (e) The hybridized SSH-Lieb network responsible for the flat band at the VBM. This cooperative interplay between the Peierls-dimerized Nb d_{z^2} chain (SSH physics) and the O-Nb $d_{x^2-y^2}$ -Cl Lieb-like sublattice combines 1D confinement with 2D destructive interference to stabilize the robust orbital-selective flat band.

band gap derive primarily from Nb- d , Cl- p , and O- p states. Focusing on the FB at the VBM, there is a clear orbital mixing: Although the main contribution arises from Nb- d_{z^2} (red) orbitals, some weight also comes from Nb- $d_{x^2-y^2}$ (blue) and Cl- p_z (black), with a minor contribution from Cl- p_y (lime). Moreover, closer inspection shows that another set of conduction bands also opens a gap as the

system evolves from state A to state B . These states are predominantly derived from Nb- d_{yz} orbitals (purple). By comparing these two distinct gap-opening processes, we can gain insight into the formation of the FB. A minimal tight-binding model is developed using hoppings derived from Wannierization of the Nb d_{z^2} and d_{yz} DFT bands. This model includes two Wannier functions, d_{z^2} (d_{yz}), centered

on each Nb atom, Nb₁ and Nb₂, of the 2 × 1 supercell. The tight-binding Hamiltonian is constructed as

$$\begin{pmatrix} E_1 + 2t_{01} \cos(k_y(R + r_j - r_i)) & t_a e^{ik_y(R+r_j-r_i)} + t_b e^{ik_y(R+r_j-r_i)} \\ t_a e^{-ik_y(R+r_j-r_i)} + t_b e^{-ik_y(R+r_j-r_i)} & E_2 + 2t_{02} \cos(k_y(R + r_j - r_i)) \end{pmatrix}, \quad (1)$$

where $\mathbf{r}_i$ and $\mathbf{r}_j$ are the atomic coordinates and $\mathbf{R}$ is a lattice translation. E_1 (E_2) denotes the on-site energy for atom 1 (2). The parameter t_{01} (t_{02}) describes the intercell hopping between atom 1 (2) and its copy in the next cell. In the off-diagonal terms, t_a and t_b represent the intracell interactions between atom 1 and atom 2. Applying this Hamiltonian to the *A* scenario reveals that $t_{01} = t_{02}$ and $t_a = t_b$ due to symmetry, resulting in a closed gap for both orbital types. In contrast, in the *B* case, $t_{01} = t_{02}$ since the two atoms host the same type of orbitals, but $t_a < t_b$ due to dimerization. The computed band structures are shown in the small panels in Figs. 5(a) and 5(b). This analysis maps the complex multiorbital problem onto an SSH-like Hamiltonian, where the gap-opening mechanism originates from the Peierls distortion. Crucially, the orbital geometry of the d_{yz} state, confined to the $y-z$ plane, prevents electron hopping along the x direction, thereby realizing an effective dimensionality reduction. In this picture, these two bands with extreme flatness may be viewed as a quasi-one-dimensional SSH chain of Nb- d_{yz} and d_{z^2} orbitals mediated by Cl atoms, whose band structure is gapped out and flattened due to symmetry breaking. According to the SSH model, the band gap magnitude is $2 \times |t_a - t_b|$ for both orbital types. The Nb d_{z^2} and d_{yz} orbitals exhibit substantially different band gaps of 2.31 and 0.37 eV, respectively. This pronounced disparity identifies a connectivity-driven renormalization as the actual mechanism behind the Peierls-type metal-insulator transition. While Peierls dimerization provides the structural trigger acting uniformly on the lattice, SSH physics governs the orbital-dependent electronic response. The magnitude of the gap is governed by how each orbital is embedded in the lattice: The d_{yz} orbital hosts a nearly isolated SSH chain with minimal band splitting, whereas the d_{z^2} orbital's gap is strongly amplified by its hybridization with the Lieb-like O-Nb-Cl network.

To this end, we performed a full Wannierization, including all the Nb- d , Cl- p , and O- p orbitals and extracted all the most meaningful hopping parameters between atoms in the supercell. Our Wannierization analysis decomposes the complex electronic structure of NbOCl₂ into three independent orbital-lattice modules [Figs. 5(c)–5(e)]. We first classify the Nb d orbitals by symmetry into t_{2g} (d_{xy} , d_{yz} , d_{xz}) and e_g (d_{z^2} , $d_{x^2-y^2}$) manifolds. Within the t_{2g} set, the d_{xy} and d_{xz} orbitals each couple to a single O- p and Cl- p orbital, forming two distinct three-site clusters: O(p_y)-Nb(d_{xy})-Cl(p_x) and O(p_z)-Nb(d_{xz})-Cl(p_x). Each cluster maps onto a separate Lieb-like sublattice [Fig. 5(c)].

Crucially, the calculated band structures reveal that the Lieb-like sublattice alone—even under Peierls distortion—fails to achieve global flatness. While it exhibits dispersionless features in specific momentum regions, it retains significant dispersion along other directions. This inability to generate a fully isolated flat band demonstrates that destructive interference in 2D Lieb-like sublattice is insufficient on its own, thereby pointing to the indispensable role of the 1D SSH chain physics in fully quenching the kinetic energy and realizing an isolated orbital-selective flat band. Consistent with previous predictions, the remaining d_{yz} orbital forms an SSH chain along the y axis, mediated by Cl p_y and p_z orbitals [Fig. 5(d) and Supplemental Material Fig. S13 [54]]. Along the orthogonal x direction, the orientation of the d_{yz} orbital precludes overlap with neighboring oxygen orbitals, while at the same time it fails to hybridize with the other t_{2g} states that possess finite spatial extension along x direction (Supplemental Material Table S1 and Fig. S13 [54]). With both direct and indirect hopping channels blocked along the x direction, d_{yz} electrons are confined into quasi-one-dimensional chains, effectively realizing an SSH model. This reduction in dimensionality, driven by orbital geometry rather than structural constraints alone, emerges from our DFT analysis of NbOCl₂ as a natural extension of the SSH model beyond its conventional one-dimensional setting. In particular, the directional anisotropy of the d_{yz} orbital enforces quasi-1D confinement even within a 2D lattice, demonstrating that orbital shape can itself generate low-dimensional electronic behavior in higher-dimensional crystals. This orbital-geometry-driven blocking of hopping not only stabilizes the d_{yz} flat band, but also establishes a general principle of effective dimensional reduction.

However, this straightforward picture, valid for t_{2g} orbitals, becomes considerably more intricate for the e_g orbitals and plays a pivotal role in understanding the FB at the VBM. The Wannierization reveals a significant on-site hybridization between the Nb e_g orbitals, with an order of 1 eV. This strong coupling between these e_g states is enabled by the distorted octahedral coordination of Nb. Furthermore, in contrast to the d_{yz} orbital, the toroidal in-plane shape of the Nb- d_{z^2} orbital allows for hopping along the x -axis mediated by the O- p_x orbitals (Supplemental Material Table S1 [54]). Our Wannier-based analysis uncovers the indispensable role of O orbitals in forming the O-Nb-Cl network and the hybridization between Nb- d_{z^2} and $d_{x^2-y^2}$ orbitals. As a result, the underlying Lieb-like

lattice formed by the $O(p_x)$ - $Nb(d_{x^2-y^2})$ - $Cl(p_y)$ triangle responsible for the destructive interference becomes intricately intertwined with the quasi-one-dimensional SSH chain, formed by $Nb-d_{z^2}$ orbitals via $Cl p_y$ and p_z states [Fig. 5(e)]. The manifestation of this hybridized SSH-Lieb network gives rise to the FB at the VBM, further reinforced by the Peierls-distortion-induced dimerization. This hopping analysis forms the basis for constructing the tight-binding models for each unique sublattice [Figs. 5(c)–5(e)]. The validity of this hybrid SSH-Lieb model is also illustrated by the halogen-dependent evolution of the electronic structure. Specifically, the comparison between $NbOCl_2$ and $NbOI_2$ highlights a unique decoupling of the electronic mechanisms along orthogonal directions. Despite the obvious dispersion acquired along Γ - Y due to the heavy halogen influence, the FB remains exceptionally flat along the Γ - X direction in $NbOI_2$ [Fig. 3(f) and Supplemental Material Fig. S4 [54]]. This behavior confirms our theoretical picture: Unlike the SSH chain direction (Γ - Y), the dispersion along Γ - X is mediated primarily by the Lieb-like sublattice. Since the oxygen atom remains unchanged across the halogen series and retains its $2p$ orbital character, the chemical environment along this axis is robust. More importantly, the flatness along Γ - X is protected by the destructive interference inherent to the Lieb lattice geometry. This geometric localization mechanism operates independently of the hopping strength along the orthogonal SSH chains. Thus, even when the heavy iodine atoms enhance the hopping and bandwidth along Γ - Y , the Lieb-symmetry-protected flatness along Γ - X remains strictly preserved. This independent tunability experimentally confirms that 1D SSH physics and 2D Lieb physics coexist within the same crystalline lattice, dictated by distinct orbital connectivities, as discussed above. By selectively modulating the bandwidth and orbital hybridization along the Γ - Y axis via halogen substitution while preserving the robust flatness along the Γ - X axis, the transition-metal oxyhalide family effectively functions as an orbital-selective anisotropic quantum simulator. This realization demonstrates that orbital geometry can be exploited to engineer and tune low-dimensional physics and correlation effects in higher-dimensional materials. Collectively, these orbital-lattice configurations effectively explain the emergence of robust orbital-selective FB and other multidimensional experimental insights. To further quantitatively validate our model through the orbital selections derived by the Wannierization, we performed the Slater-Koster parametrization of each d - p interaction. The results summarized in Supplemental Material Table S1 [54] quantify the interactions of $Nb d - O p$ and $Nb d - Cl p$ and show complete consistency with our tight-binding models, with finite overlaps taking place exclusively in the orbital clusters mentioned above. Our theoretical framework is broadly applicable, because it relies on orbital characterization and Slater-Koster overlaps that can extend

to the entire family of transition-metal oxyhalides. Beyond its generality, the conceptual novelty of this model paves the way for the realization of flat-band systems via sublattice hybridization and orbital-geometry-driven dimensional reduction, combined with the dimerization process.

By integrating ARPES and transport measurements as well as theoretical modeling, we have established the transition-metal oxyhalides (MOX_2) as a versatile platform hosting intrinsic orbital-selective flat bands at room temperature. Our analysis identifies a novel microscopic origin rooted in a hybrid SSH-Lieb network, where the co-operative interplay between quasi-1D orbital chains and a 2D Lieb-like sublattice enforces kinetic-energy quenching. This mechanism is experimentally validated by the universal polarization selectivity and the distinct halogen-dependent bandwidth evolution, which demonstrates the unique decoupling of 1D and 2D physics in this material system. Crucially, we demonstrate that these flat bands are not only robust from the bulk to the few-layer limit, but also tunable: Electrostatic gating and surface doping effectively modulate the Fermi level, confirming the system as a gate-tunable correlated insulator. These findings articulate a new design principle for engineering tunable correlated phases in room-temperature van der Waals materials, offering a pathway to explore flat-band physics without relying on moiré superlattices or extreme conditions.

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X. L., L. Z., G. S., and R. C. proposed and designed the research. X. L., D. O., and S. P. carried out the ARPES experiments at the National Synchrotron Light Source II and Solaris National Synchrotron Radiation Centre, with support from A. K. K., A. R., E. V., N. O., R. K., and D. W. X. L. analyzed the data. X. L., W. M.-K., and E. L. grew the single crystals under the supervision of X. R. X. L. prepared the few-layer samples and performed transport measurements with the help of S. P., J. L., and Q. S. L. Z. and G. S. performed DFT calculations. All authors contributed to writing the paper and participating in discussions and comments on the paper. X. L., L. Z., G. S., and R. C. oversaw the project.

The authors declare no competing interests.

DATA AVAILABILITY

The data are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX: METHODS

1. Sample growth and characterization

Single crystals of the transition-metal oxyhalide family MOX_2 ($M = \text{Nb, Ta}$; $X = \text{Cl, Br, I}$) were synthesized via chemical vapor transport. For NbOCl_2 and TaOCl_2 , high-purity Nb_2O_5 (Ta_2O_5 ; 99.99%), Nb (Ta ; 99.99%), and NbCl_5 (TaCl_5 ; 99.99%) powders were mixed in a stoichiometric molar ratio of 1:1:2. For NbOBr_2 and NbOI_2 , elemental bromine (Br_2) and iodine (I_2) were used as halogen sources. All starting materials were sealed in evacuated quartz tubes and placed in a horizontal three-zone furnace following the procedure in Refs. [57,60,65,66]. Rectangular-shaped crystals were obtained with a typical growth duration lasting from three weeks to one month. Phase purity and composition were confirmed by energy-dispersive x-ray spectroscopy. Polarization-resolved Raman spectra (Supplemental Material Fig. S1 [54]) further corroborate phase purity and provide vibrational fingerprints that validate the crystallographic orientation.

2. Device fabrication

The device used for micro-ARPES measurements was fabricated by assembling a van der Waals heterostructure consisting of few-layer NbOCl_2 , graphene, and hBN on a SiO_2 (300 nm)/Si substrate with prepatterned Au electrodes. Few-layer NbOCl_2 flakes were mechanically

exfoliated inside a glove box and transferred onto hBN using a polymer-based dry transfer technique. A graphene layer was subsequently stacked on top of NbOCl_2 to provide encapsulation and environmental protection. Electrical contacts were defined by depositing Ti (5 nm)/Au (30 nm) electrodes on SiO_2 (300 nm)/Si substrate using standard photolithography methods. The entire stacking and transfer procedure was carried out under inert conditions to minimize exposure of NbOCl_2 to ambient moisture and oxygen. The resulting graphene/ NbOCl_2 /hBN heterostructures enabled stable and high-quality micro-ARPES measurements. The sample thickness was determined by atomic force microscopy (AFMWorkshop HR) measurements, using a silicon probe in tapping mode.

3. ARPES measurements

ARPES experiments were carried out at beam lines URANOS of the SOLARIS National Synchrotron Radiation Centre and the National Synchrotron Light Source II ESM beam lines of Brookhaven National Laboratory. Both beam lines were equipped with Scienta DA30 electron analyzers. The results were reproduced at both facilities. The angular and total energy resolutions were set to 0.1° and less than 20 meV. For the measurements of the few-layer samples at ESM, microfocused synchrotron radiation was employed with a beam spot approximately $10 \times 5 \mu\text{m}^2$. The flake positions were identified by real space mapping. Bulk crystals were *in situ* cleaved prior to measurement. Critically, all samples—both bulk and exfoliated few-layer flakes—were measured at room temperature ($T = 300 \text{ K}$) under ultrahigh vacuum conditions (better than $9 \times 10^{-11} \text{ Torr}$). The Fermi level was calibrated using a clean polycrystalline Au reference in electrical contact with the sample.

4. DFT calculations

First-principles calculations were performed in the framework of density functional theory (DFT), as implemented in the QUANTUM-ESPRESSO package [73,74]. Projected augmented wave pseudopotentials were selected for all the calculations, in the generalized gradient approximation based on the Perdew-Burke-Ernzerhof (PBE) scheme. The plane wave basis set was truncated using a cutoff energy of 50 Ry for the plane waves and 500 Ry to represent the charge density in all calculations. The Brillouin zone was sampled by $16 \times 8 \times 1$ Monkhorst-Pack k -point grid. Considering the z axis as the stacking direction for the bulk, the supercell size was kept fixed along that direction for all the analyzed monolayer structures. An approximately 13 Å vacuum space was added along the out-of-plane direction, proven to be sufficient to prevent periodic replicas from interacting with each other. For all the considered structures, the atomic positions were optimized using the conjugate gradient algorithm in the

PBE [75] scheme, with a convergence criterion such that the total energy difference between consecutive structural optimization steps was less than 10^{-5} eV and all components of the forces acting on the atoms must be less than 10^{-4} eV/Å. Wannierization was carried out in the Wannier90 [76] framework by considering a total of 28 Wannier functions including every Nb-*d*, O-*p*, and Cl-*p* orbital per formula unit and a Monkhorst-Pack *k*-point grid of $12 \times 12 \times 1$ points, proved sufficient to appropriately reproduce the monolayer's *ab initio* band structure. Finally, the tight-binding parameters were extracted from the Wannier Hamiltonian by means of the PYTB code [77], which was used to perform tight-binding models for each of the individual sublattice.

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