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# Monolayer van der Waals kagome lattice for high-temperature flat-band physics

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## 1. Comparison with other 2D flat band systems

It is important to recognize that there have been long-standing efforts to realize 2D flat band lattice models using a variety of approaches, including self-assembled organic frameworks, artificially-assembled molecular lattices, and orbital decoration-based approaches. These efforts highlight the strong community-wide interest in realizing and exploring geometry-driven 2D flat band systems. Below, we briefly review these earlier platforms, discuss some of their intrinsic limitations, and compare them with the 2D vdW kagome-based flat band system introduced in this work.

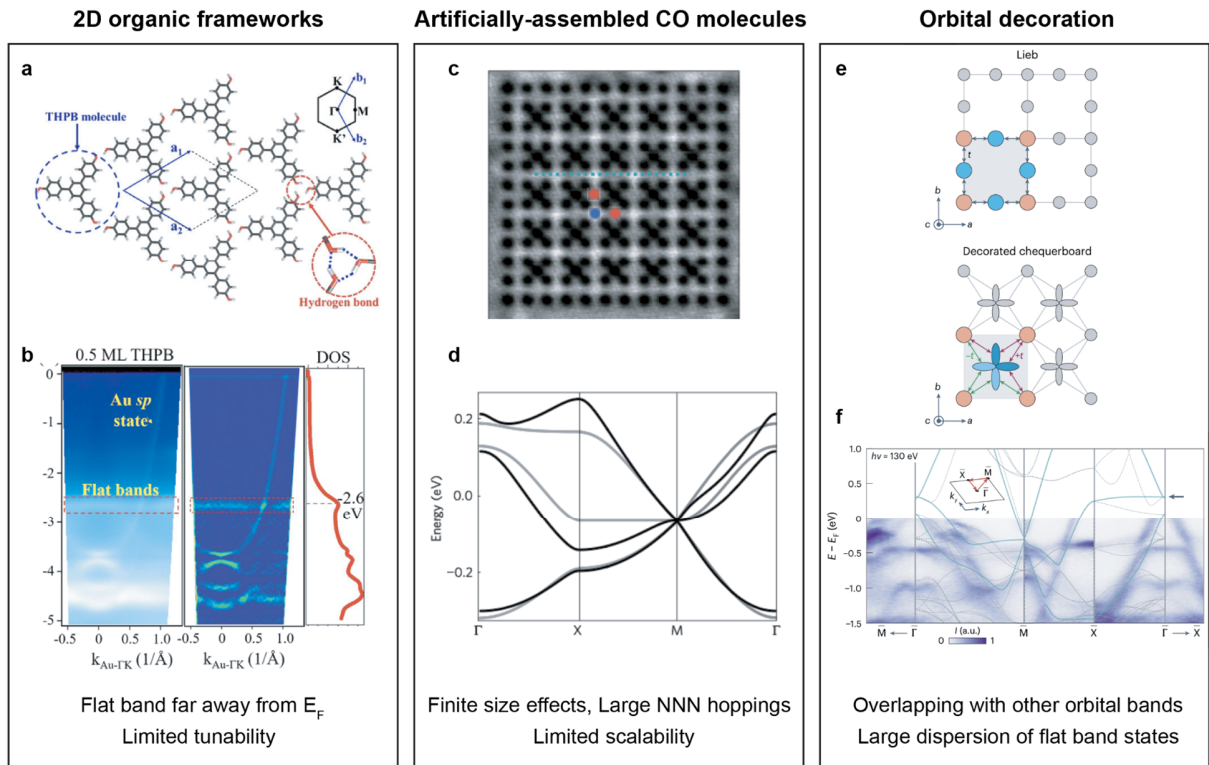


**Fig. S1. Various approaches toward geometry-driven 2D flat band systems.** **a,b**, Self-assembled organic frameworks (adapted from Ref. 1), **c,d**, Artificially assembled CO molecules (adapted from Ref. 2), **e,f**, Orbital decorated lattice (adapted from Ref. 3).

### A) Self-assembled organic frameworks

In pursuit of geometry-driven flat band systems, designs based on two-dimensional organic frameworks – such as metal-organic frameworks (MOFs), covalent organic frameworks (COFs), and hydrogen-bond organic frameworks (HOFs) – have been actively explored. In these approaches, organic molecules are deposited on metallic substrates, where they self-assemble into long-range ordered structures mimicking crystalline lattice geometries.

For example, a recent study in Ref. 1 explored 1,3,5-tris(4-hydroxyphenyl)benzene molecules deposited onto the Au(111) surface, which form a trigonal lattice network, as shown in Fig. S1a. The spatial distribution of density of states on this molecular trigonal lattice effectively realizes a kagome-like lattice, which can in principle host flat bands. However, from an electronic structure perspective, the realized organic framework is essentially a molecular insulator, with a giant bandgap of  $\approx 4\text{--}5$  eV, resulting in the flat band states located far away from the Fermi level at  $\approx -2.6$  eV (Fig. S1b). The electronic structure near the Fermi level is instead dominated by the highly dispersive Au *sp* bands from the substrate. Since the supporting substrate is typically essential for the formation of such 2D organic frameworks, isolating the organic framework and tuning its electronic structure to access low-energy flat band physics remains elusive. In sharp contrast, the vdW kagome lattice realized in our work offers more direct crystalline 2D flat band system, which can be readily integrated into 2D vdW heterostructure devices and whose flat band electronic structure can be directly tuned to the Fermi level to realize and explore flat band physics.

## **B) Artificially assembled molecular lattice**

As another pathway to mimic 2D lattice models, artificially-assembled molecular lattices have also been explored. In this approach, carbon monoxide (CO) molecules are artificially manipulated and positioned on a Cu(111) surface using a STM tip to create the desired lattice geometry (Fig. S1c). This platform has been used to simulate various lattice models, including hexagonal and Lieb lattices, as correctly noted by the reviewer.

However, because the CO molecules must be assembled one-by-one using the STM tip, this approach has inherently limited scalability. In practice, the lattice size remains restricted to only a few unit cells (for example,  $5\times 5$  unit cells in Ref. 2). This artificial CO lattice also introduces additional effects, such as finite size effects and large next-nearest-neighbor hoppings ( $t'/t \approx 0.6$ ). These result in a significant broadening of the flat band in CO-based lattice models, such that its bandwidth becomes comparable to that of the dispersive bands in the model (Fig. S1d). Altogether, while artificially assembled molecular lattices provide a proof-of-principle platform for simulating lattice models, their artificial nature and limited scalability make them fundamentally different from the intrinsic crystalline flat band material platform realized in our work.

### C) Orbital decoration-based approaches

Kagome lattice is not the only lattice geometry hosting 2D electronic flat bands. For example, Lieb lattice represents another 2D lattice geometry that realizes frustrated hopping in the real-space and flat bands in the momentum-space. However, materials that realize the Lieb lattice are much rarer than kagome lattice systems<sup>2,4</sup>, and the exploration of flat band physics based on a true Lieb lattice geometry is still at a relatively early stage.

Instead of studying a true Lieb lattice system, an interesting recent work explored a chequerboard lattice system,  $\text{Pd}_5\text{AlI}_2$ , where lattice decoration by multiple  $d$ -orbitals gives rise to effective Lieb-lattice-like hopping pathways, as illustrated in Fig. S1e. However, the presence of multiple  $d$ -orbital degrees of freedom inevitably results in strong overlap of the flat band with many other dispersive bands (Fig. S1f). This significantly limits both the tunability of the flat band and the isolation of intrinsic flat-band physics. Moreover, anisotropic hopping processes associated with these  $d$ -orbitals further distort the flat band dispersion, such that the “flat band” realized in  $\text{Pd}_5\text{AlI}_2$  is in fact highly dispersive with a bandwidth of  $\approx 1$  eV (Fig. S1f).

These limitations of the  $\text{Pd}_5\text{AlI}_2$  system stand in sharp contrast to the band structure realized in the  $\text{A}_2\text{T}_3\text{X}_4$  system, where a single-orbital-based flat band, emerging directly from the kagome lattice geometry without the need for orbital decoration, remains isolated in energy and features a strongly quenched dispersion of  $\approx 0.18$  eV.

In summary, the above approaches (self-assembled organic frameworks, artificially-assembled CO molecules, and orbital decoration) represent important earlier efforts toward 2D electronic flat band systems. However, these artificial or indirect approaches give rise to inevitable limitations in exploring flat band physics, including limited scalability, non-ideal flat band dispersion, overlap with other electronic states, and restricted tunability. In some sense, the fact that these previous studies have relied on such artificial or indirect realizations reflects the challenges in achieving a true crystalline, intrinsic 2D flat band platform. This is precisely what we accomplished in the present study via 2D  $\text{A}_2\text{T}_3\text{X}_4$ , opening a realistic pathway to explore long-envisioned emergent flat band phenomena based on frustrated lattice models.

## 2. Comparison with other vdW kagome system

Key properties of the  $A_2T_3X_4$  system established in the present study include:

- (i) Exfoliability down to the monolayer, enabling the realization of a true 2D kagome lattice.
- (ii) An isolated kagome flat band emerging just below the Fermi level.

These properties together establish  $A_2T_3X_4$  as a long-sought ideal 2D kagome platform.

Below, we discuss how these key features of  $A_2T_3X_4$  are distinguished from previously explored vdW kagome candidates, such as  $Nb_3(Cl,Br,I)_8$  and  $Pd_3P_2S_8$ . Specifically, these earlier systems either cannot be realized in the true monolayer limit or do not host kagome-derived flat bands at low energies.

### A) Non-kagome flat band in distorted vdW kagome lattice $Nb_3(Cl,Br,I)_8$

$Nb_3(Cl,Br,I)_8$  is a series of vdW kagome materials, where the kagome lattice composed of Nb exhibits pronounced trimerization distortion, realizing a breathing kagome geometry as shown in Fig. S2b. While such breathing kagome lattices can still host flat bands due to preserved mirror symmetries<sup>5</sup>,  $Nb_3(Cl,Br,I)_8$  does not realize these kagome-derived flat bands at low energies. Instead, its physics is dominated by localized molecular-orbital states confined within the Nb trimers, as detailed below.

To understand the low-energy physics of  $Nb_3(Cl,Br,I)_8$ , we first illustrate two complementary approaches for describing the electronic structure of breathing kagome lattice: one treats the breathing distortion as a perturbation to the ideal kagome model (Fig. S2a), and the other considers molecular orbitals within trimers (Fig. S2c). In the former, the breathing distortion opens a band gap at the Dirac point, while preserving the kagome flat band state. We note that this band gap is topologically trivial, making the Chern number of the lowest band  $C = 0$  (Fig. S2a). Meanwhile, in the molecular orbital description, the hopping  $t$  within the trimers gives rise to one eigenstate at energy  $-2t$  and two degenerate states at energy  $t$  (Fig. S2c).

In the case of  $Nb_3(Cl,Br,I)_8$ , the degree of trimerization is substantial, reaching ~16 % of the bond length of the undistorted kagome lattice. Accordingly, the DFT band structure of  $Nb_3(Cl,Br,I)_8$  lies close to the molecular orbital limit, as shown in Fig. S2b, although some dispersion remains due to inter-trimer hopping. Most importantly, comparison with the kagome bands in Fig. S2a clearly indicates that the low-energy quasi-flat band crossing the Fermi level in  $Nb_3(Cl,Br,I)_8$  originates from the topologically trivial band with  $C = 0$ . Summarizing the above

considerations, the low-energy flat band in  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$  represents a trivial molecular orbital state confined within the Nb trimers, different from topological flat bands of kagome origin. We note that the topologically trivial nature of the low-energy quasi-flat band in  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$  has been explicitly noted in several previous studies<sup>5,6</sup>.

In reality, the molecular flat band crossing the Fermi level in  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$  splits into spin-up and spin-down bands, developing localized cluster magnetic moments and opening a bandgap (Fig. S2d). In this context, the low-energy physics of  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$  is best described by the local moment magnetism in an effective triangle lattice (Fig. S2e), and does not reflect the topological kagome flat band physics we pursued in  $A_2T_3X_4$ .

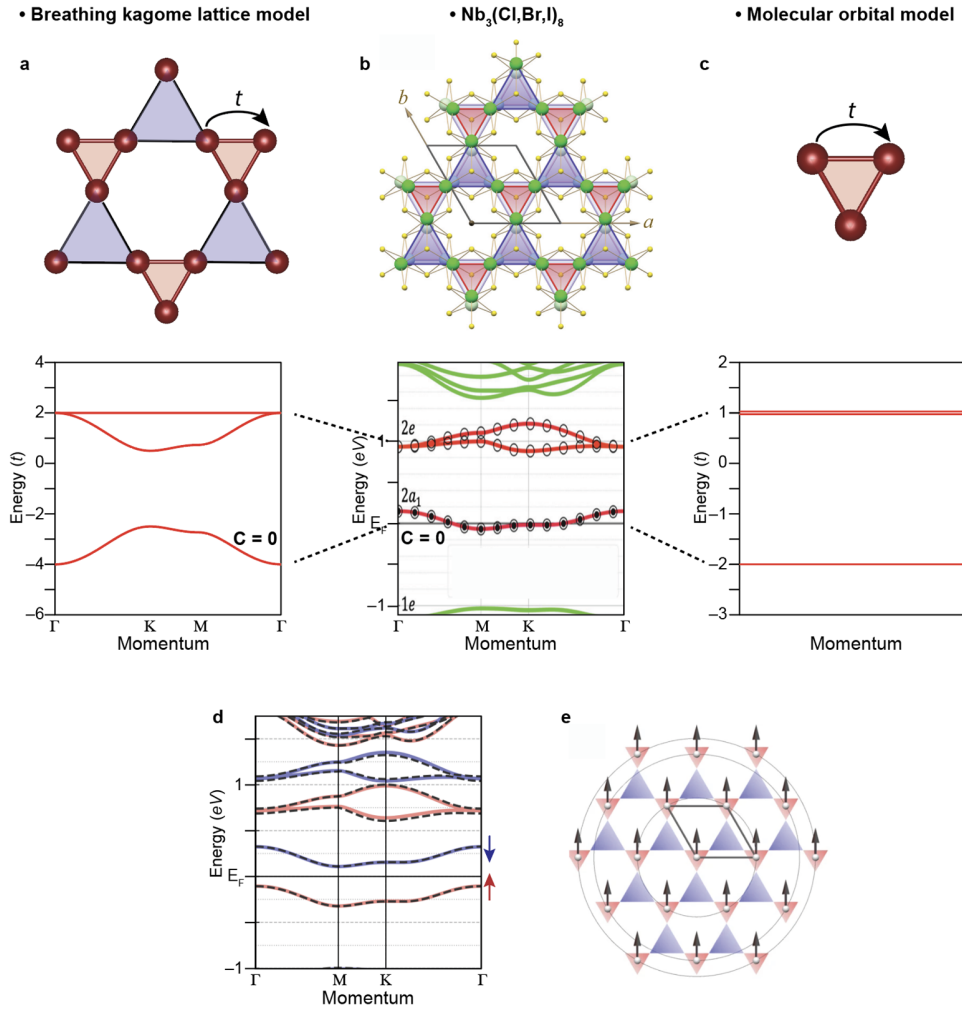


**Fig. S2 | Non-kagome flat band in the distorted vdW kagome lattice  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$ .** **a,c,** Two complementary descriptions of the electronic structure of  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$  based on a breathing kagome lattice model (a) and a trimer molecular orbital model (c). **b,** DFT band structure of  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$ . The low-energy flat band originates from a topologically trivial, molecular orbital state within the trimer, and is different from the topological flat band of kagome origin. **d,e,** Development of the local cluster moment in  $\text{Nb}_3(\text{Cl},\text{Br},\text{I})_8$ . Panels b,d,e are adapted from Ref. 7.

## B) Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> with mechanically interlocked vdW layers

Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> represents another vdW kagome candidates explored to date. Despite its vdW nature, however, the reliable exfoliation of isolated monolayer Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> remains infeasible. To understand the microscopic origin of this difficulty, we present the crystal structure of Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> in Fig. S3a. An important structural feature of Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> is that each kagome layer contains protruding sulfur atoms, as shown in Fig. S3a. This protrusion of sulfur atoms results in mechanical interlocking between adjacent vdW layers, making the reliable exfoliation of 2D limit Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> intrinsically challenging. Accordingly, previous exfoliation studies on Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> have reported that monolayer Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> appears only as a narrow step terrace attached to bulk crystals, with a lateral width below 1  $\mu\text{m}$  (Fig. S3b). Achieving monolayer Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> suitable for practical 2D devices or vdW heterostructures remains infeasible, which has precluded experimental exploration of ideal 2D kagome physics using Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub>.

To further support this point, we synthesized our own Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> crystals and performed systematic exfoliation tests. As shown in Fig. S3c, bulk Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> crystals exhibit a transparent red color, indicating a much larger band gap and a significantly more insulating character compared to the A<sub>2</sub>T<sub>3</sub>X<sub>4</sub> system. We note that this strongly insulating nature may explain why there have been no direct photoemission measurements to constrain the band structure in Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> thus far. Our exfoliation of Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> crystals onto more than twenty 1"×1" silicon substrates did not yield any monolayer or few layer flakes, again confirming intrinsic challenges in realizing 2D limit Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> due to structurally-interlocked vdW layers (Fig. S3b).

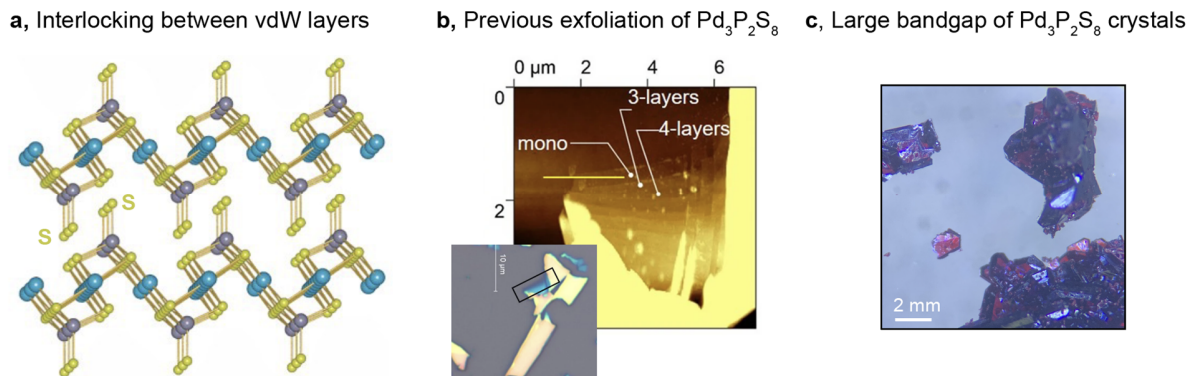


**Fig. S3 | Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> with mechanically interlocked vdW layers** **a**, Mechanical-interlocking between adjacent vdW layers of Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub>. This intrinsic structural feature makes exfoliation of 2D kagome layers less trivial. **b**, Previous exfoliation reports on a candidate vdW kagome system Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub>. Despite its vdW character, monolayer regions only exist locally as step terraces. **c**, Optical image of Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> crystals. Bulk Pd<sub>3</sub>P<sub>2</sub>S<sub>8</sub> crystals exhibit a transparent red color, reflecting a significantly larger band gap and a more insulating character compared to the A<sub>2</sub>T<sub>3</sub>X<sub>4</sub> system. Panel b is adapted from Ref. 8.

In summary, these earlier explorations of vdW kagome systems clearly reflect strong community-wide interest in realizing the 2D limit kagome platforms. However, as discussed above, existing vdW kagome candidates either do not host kagome-derived flat bands at low energies ( $\text{Nb}_3(\text{Cl}, \text{Br}, \text{I})_8$ ) or cannot be reliably realized in the true 2D limit ( $\text{Pd}_3\text{P}_2\text{S}_8$ ). By contrast, our work establishes  $A_2T_3X_4$  as a long-sought ideal 2D kagome system overcoming these limitations. The present study thus opens compelling opportunities to explore kagome lattice physics and long-sought geometry-driven flat band phenomena in the true 2D limit using  $A_2T_3X_4$ .

### 3. Experimental determination of flat band orbital parity via polarization-resolved ARPES

A key underlying mechanism enabling the realization of isolated single-orbital kagome bands in  $A_2T_3X_4$  is the strong crystal field splitting imposed by the surrounding  $X$  atoms in low-symmetry planar coordination. As discussed in Fig. 1c and Extended Data Fig. 1, this crystal field environment effectively isolates a single  $d_{xy}$  orbital degrees of freedom near the Fermi level, ultimately realizing a model single-orbital kagome dispersion in  $A_2T_3X_4$  without interference from other orbitals.

While the orbital character of the kagome flat band in  $A_2T_3X_4$  was assigned based on DFT in the Main text, the orbital parity of this band can also be experimentally inferred using polarization-resolved ARPES. Below, we develop a general framework to analyze the orbital parity of the kagome flat band from polarization-dependent ARPES matrix elements. Based on this framework, we experimentally determine that the kagome flat band in  $A_2T_3X_4$  possesses mirror-odd orbital parity, fully consistent with the  $d_{xy}$  orbital character inferred from DFT.

#### Framework for analyzing the orbital parity of kagome flat band using polarization-resolved ARPES

To start, we first note that in systems with multiple sublattice degrees of freedom – such as hexagonal and kagome lattices – the photoemission selection rules are governed by two main factors:

- 1) The local orbital character at each sublattice, and
- 2) Phase interference between photoelectrons from different sublattices.

In earlier ARPES studies of graphene and kagome systems, this phase interference effect has been widely exploited to determine the phase structure of Dirac electron wavefunctions. We now extend this framework to derive the photoemission selection rules for flat band electrons in the kagome lattice. We note that the overall sublattice interference formalism follows that developed in prior works, including (Ref. 9,10).

We start with the standard tight-binding Hamiltonian for the kagome lattice:

$$H(\mathbf{k})\psi(\mathbf{k}) = -t * \begin{pmatrix} 0 & \cos \mathbf{k} \cdot \mathbf{d}_{AB} & \cos \mathbf{k} \cdot \mathbf{d}_{AC} \\ \cos \mathbf{k} \cdot \mathbf{d}_{BA} & 0 & \cos \mathbf{k} \cdot \mathbf{d}_{BC} \\ \cos \mathbf{k} \cdot \mathbf{d}_{CA} & \cos \mathbf{k} \cdot \mathbf{d}_{CB} & 0 \end{pmatrix} \begin{pmatrix} \psi_A(k) \\ \psi_B(k) \\ \psi_C(k) \end{pmatrix} = E\psi(\mathbf{k})$$

where  $t$  is the hopping parameter,  $d_{pq}$  is the vector connecting  $p$  and  $q$  sublattices,  $\psi(\mathbf{k})$  is the wavefunction at the wavevector  $\mathbf{k}$ . Focusing on the  $\Gamma$  point ( $k = 0$ ), the secular equation is greatly simplified to

$$-t \begin{vmatrix} E/t & 1 & 1 \\ 1 & E/t & 1 \\ 1 & 1 & E/t \end{vmatrix} = 0$$

which has doubly degenerate roots at  $E/t = 1$  and a single root at  $E/t = -2$ . The flat band eigenstate corresponds to the former, indicating that, for the flat band electrons, the phase relationship between three kagome sublattices has to satisfy

$$\psi_A + \psi_B + \psi_C = 0.$$

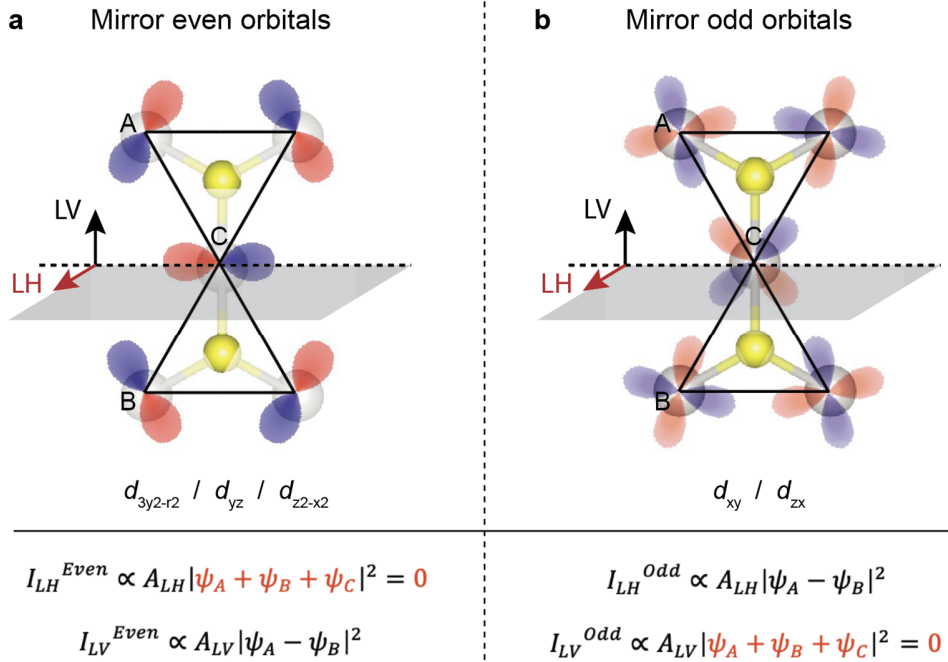


**Fig. S4 | Interference of photoelectrons from three kagome sublattices.** **a**, Case for orbitals with mirror even parity. In our local coordinate,  $d_{3y^2-z^2}$ ,  $d_{yz}$ , and  $d_{z^2-x^2}$  orbitals possess mirror even parity. **b**, Case for orbitals with mirror odd parity. In our local coordinate,  $d_{xy}$  and  $d_{zx}$  orbitals possess mirror odd parity.

Now we examine how this phase relationship determines the photoemission selection rules. For orbitals with even parity with respect to the scattering plane (Fig. S4a), the interference of photoelectrons from the three kagome sublattices under linear horizontal (LH) and linear vertical (LV) light polarizations can be approximated as:

$$I_{LH}^{even} \propto A_{LH} |\psi_A + \psi_B + \psi_C|^2$$

$$I_{LV}^{even} \propto A_{LV} |\psi_A - \psi_B|^2$$

where  $A_{LH}$  and  $A_{LV}$  are the LH and LV polarization components of the vector potential of incident radiation, respectively. Combined with the phase relationship established above ( $\psi_A + \psi_B + \psi_C = 0$ ), this implies that the kagome flat band intensity at  $\Gamma$  should vanish under LH polarization when the underlying orbital has mirror even parity. By contrast, for orbitals with mirror odd parity (Fig. S4b), the interference approximately becomes:

$$I_{LH}^{odd} \propto A_{LH} |\psi_A - \psi_B|^2$$

$$I_{LV}^{odd} \propto A_{LV} |\psi_A + \psi_B + \psi_C|^2$$

indicating that the photoemission intensity is now expected to vanish for the LV polarization channel. The resulting photoemission selection rules for kagome flat band electrons can thus be summarized as Table S1 below:

| Kagome flat band, $k = 0$ |    | Orbital symmetry                         |                          |
|---------------------------|----|------------------------------------------|--------------------------|
|                           |    | Even ( $d_{3y2-z2}, d_{yz}, d_{z2-x2}$ ) | Odd ( $d_{xy}, d_{zx}$ ) |
| Photon Polarization       | LH | $0$                                      | <i>finite</i>            |
|                           | LV | <i>finite</i>                            | $0$                      |

**Table. S1 | Photoemission selection rule for kagome flat band electrons at  $\Gamma$ .**

This framework thus demonstrates that, by analyzing polarization-dependence at the  $\Gamma$  point, the underlying orbital parity constituting the kagome flat band can be directly determined.

Based on this theoretical framework, in Fig. S5a,b, we compare ARPES spectra of bulk  $\text{Rb}_2\text{Pd}_3\text{S}_4$  measured using LV and LH polarizations. Notably, the flat band photoemission intensity at the  $\Gamma$  point is clearly suppressed for the LV polarization channel (Fig. S5a), while no such anomalous suppression is observed for the LH polarization channel (Fig. S5b). In light of the selection rules in Table S1, this behavior directly indicates that the low-energy flat band in

$\text{Rb}_2\text{Pd}_3\text{S}_4$  possesses mirror odd orbital parity. This is fully consistent with the  $d_{xy}$  orbital character originally inferred from DFT calculations, providing independent experimental support for our orbital character assignment.

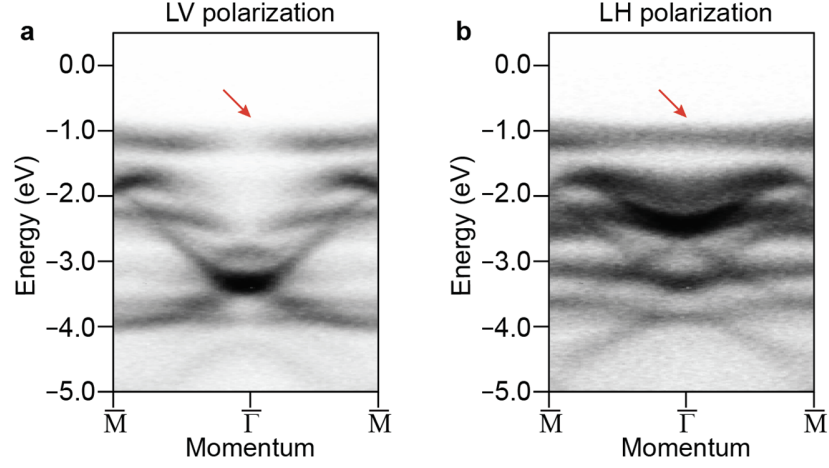


**Fig. S5 | Polarization-dependence of the kagome flat band intensity at  $\Gamma$  ( $k = 0$ ).** **a,b**, ARPES spectra of  $\text{Rb}_2\text{Pd}_3\text{S}_4$  measured under LV and LH polarizations, respectively. The anomalous suppression of the flat band intensity at  $\Gamma$  is observed for the LV polarization channel.

#### 4. Role of orbital degrees of freedom for the higher-binding-energy electronic structure

While we have primarily focused on the low-energy  $d_{xy}$  kagome bands in the Main text, other  $d$ -orbital degrees of freedom are also present in  $A_2T_3X_4$  and contribute to the electronic structure at higher binding energies. Although these states do not directly participate in the low-energy physics, these orbitals are also subject to the underlying kagome lattice symmetry and expected to give rise to characteristic kagome band features, including Dirac bands at K and flat bands. Below, we briefly present these higher-binding-energy kagome features observed in our nano-ARPES measurements and discuss their orbital origins based on orbital-projected DFT calculations.

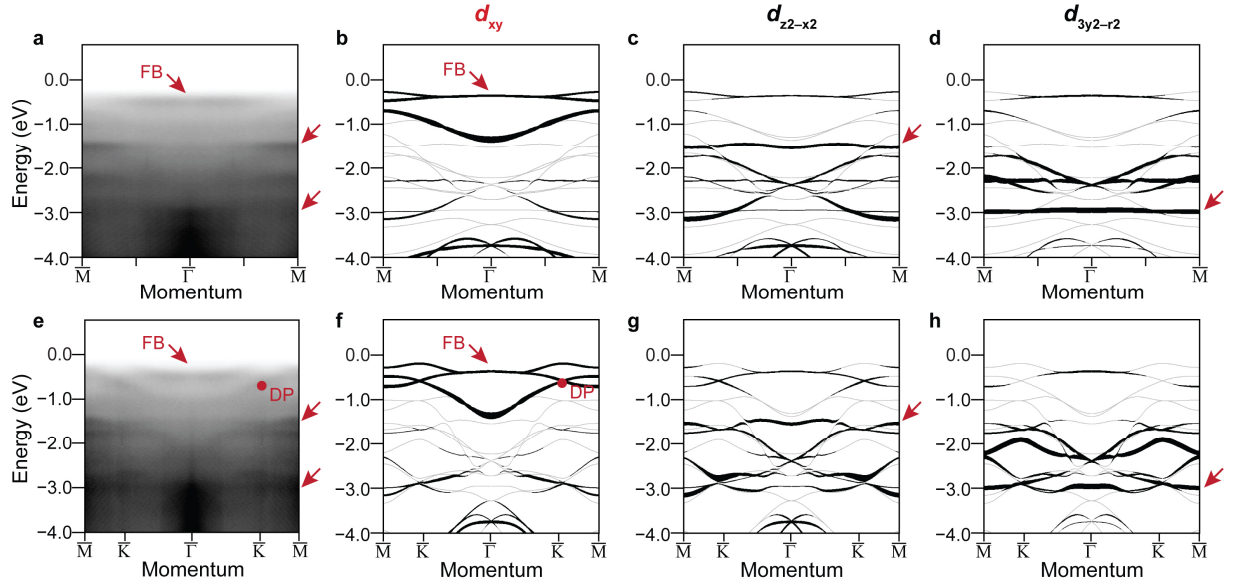


**Fig. S6 | Comparison between nano-ARPES spectra (a,e) and orbital-projected DFT dispersions (b–d,f–h) of monolayer  $\text{Cs}_2\text{Pd}_3\text{S}_4$ .** The nano-ARPES spectra are reproduced from Fig. 3h,i in the Main text. The DFT bands in b–d,f–h are vertically shifted to fit the experimental Fermi level.

Fig. S6 presents the nano-ARPES spectra of monolayer  $\text{Cs}_2\text{Pd}_3\text{S}_4$  (Fig. S6a,e), along with the projected DFT dispersions in  $d_{xy}$ ,  $d_{z^2-x^2}$  and  $d_{3y^2-z^2}$  orbital channels (Fig. S6b–d,f–h). Just below the Fermi level, an isolated flat band and Dirac band are clearly resolved in the nano-ARPES spectra (Fig. S6a,e). These low-energy bands are closely reproduced by the  $d_{xy}$  orbital-projected DFT calculations (Fig. S6b,f), allowing us to unambiguously assign these features to kagome bands derived from the isolated  $d_{xy}$  orbital degrees of freedom. At the same time, there also appear several additional weakly dispersive features in the monolayer ARPES spectra, most prominently around  $-1.5$  eV and  $-2.9$  eV (see red side arrows in Fig. S6a,e). Notably, these flat features are

also precisely reproduced by the  $d_{z^2-x^2}$  and  $d_{3y^2-z^2}$  projected DFT dispersions, respectively (Fig. S6c,d,g,h).

These analyses thus establish that, while the low-energy physics of the  $A_2T_3X_4$  vdW kagome system is dominated by an isolated  $T$ - $d_{xy}$  orbital degrees of freedom, other  $d$ -orbital states also contribute to the higher-energy-electronic structure by forming their respective kagome band manifolds, including flat bands.

## 5. Additional details of nano-ARPES measurements

In nano-ARPES experiments, the photon flux is intrinsically reduced compared to conventional ARPES due to the additional focusing optics. In our measurements, the  $A_2Pd_3S_4$  flakes of interest are further encapsulated beneath a graphene capping layer, which additionally attenuates the photoemission signal. Given these conditions, careful optimization of the experimental parameters is critical to maximize the detectable photoelectron yield in nano-ARPES measurements. In addition to identifying the optimal photon energy (98 eV) and photon polarizations (LH) that maximize the signal from  $A_2Pd_3S_4$ , we also explored different sample geometries and Brillouin zones (Fig. S7a) to further enhance the photoemission matrix elements associated with the  $d_{xy}$  kagome flat band of interest.

Fig S7b,c display the representative raw ARPES spectra of 2D  $Cs_2Pd_3S_4$ , measured in the 1st and 2nd Brillouin zones, respectively. It is apparent that the photoemission intensity from the kagome flat band is significantly enhanced in the 2nd Brillouin zone. However, at large momenta in the 2nd Brillouin zone ( $|k| > 1.0 \text{ \AA}^{-1}$ ), the kagome dispersions overlap with strong Dirac band intensity originating from graphene/graphite layers in the heterostructure. Taking these experimental considerations into account, we present in the Main text the dispersion measured in the 2nd Brillouin zone over the momentum range  $|k| \approx 0.0\text{--}1.0 \text{ \AA}^{-1}$ , symmetrized with respect to  $\Gamma$  to enable direct comparison with DFT calculations.

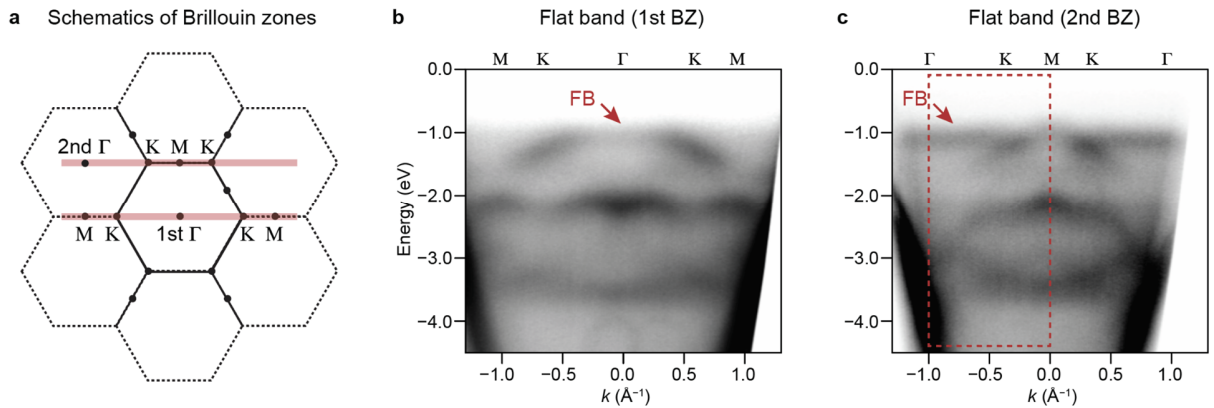


**Fig. S7 | Comparison of kagome flat band intensity between the 1st and 2nd Brillouin zones.** **a**, Schematic illustration of the Brillouin zones and momentum directions along which the flat band dispersions in b,c are measured. **b,c**, Nano-ARPES spectra of 2D  $Cs_2Pd_3S_4$  measured in the 1st and 2nd Brillouin zones, respectively. The data presented in the Main text Fig. 3 is from the 2nd Brillouin zone (marked with red-dashed rectangles in c), symmetrized with respect to  $\Gamma$  to avoid overlap with the Dirac band intensity at high momenta.

## 6. High-electron-density flat bands and high-temperature flat band phenomena

In this work, we establish a new class of 2D flat band systems based on the novel vdW kagome family  $A_2T_3X_4$ . This system adopts fundamentally different mechanisms to realize flat bands compared to existing Moiré-based flat band platforms, leading to more than two orders of magnitude enhanced flat band electron density. This key distinction opens an important prospect for realizing correlated and topological flat band phenomena at significantly enhanced energy/temperature scales, as directly supported by our exact diagonalization analysis. Below, we provide an intuitive discussion of how the increased flat band electron density in 2D vdW kagome system leads to enhanced energy scales for emergent flat band phenomena.

**A) Enhancement of overall energy scales:** The kinetic energy (bandwidth)  $W$ , interaction energy  $U$ , and their ratio  $U/W$  in Moiré and kagome flat band systems are explicitly compared in Table S1. With the substantially higher electron density  $n$  and reduced localization length scales  $a$ , both  $W$  and  $U$  are significantly enhanced in kagome systems, while the ratio  $U/W$  remains comparable. Since the ratio  $U/W$  primarily determines the nature of the correlated phases that may emerge, comparable  $U/W$  indicates that the 2D kagome system accesses strongly correlated regimes similar to those realized in Moiré systems. At the same time, the absolute magnitudes of  $U$  and  $W$  set the characteristic energy scales for many-body energy gaps. In nearly flat-band systems, such as 2D  $A_2T_3X_4$  and twisted bilayer  $\text{MoTe}_2$ , the many-body gaps are typically a fraction of the interaction scale  $U$ . Therefore, an order-of-magnitude enhancement in  $U$  naturally leads to correspondingly larger many-body gaps and higher characteristic temperatures. Our explicit exact diagonalization calculation on  $A_2T_3X_4$  kagome systems confirms this physical picture, revealing fractional Chern gaps that are an order of magnitude larger than those reported for twisted bilayer  $\text{MoTe}_2$ .

| 2D $A_2\text{Pd}_3\text{S}_4$ |                         | $t\text{-MoTe}_2$ |
|-------------------------------|-------------------------|-------------------|
| 6.2 Å                         | Localization length $a$ | 52 Å              |
| 140 meV                       | Flat band width $W$     | 10 meV            |
| 460 meV                       | Interaction energy $U$  | 19–35 meV         |
| 3.1                           | $U/W$                   | 1.9–3.5           |
| 14–29 meV                     | Fractional Chern gap    | 1–5 meV           |

**Table S2** | Comparison between energy scales in 2D kagome and Moiré flat band systems. The values for the  $t\text{-MoTe}_2$  systems are adapted from Ref. 11.

**B) Larger upper bound on the energy gap:** Another way to understand the role of higher electron density is considering a theoretical upper bound for the charge gap of topological states, including strongly correlated fractionalized phases. This bound, which is recently established in Ref. 12, relates the optical charge gap to band properties – such as the effective mass and Chern number – as well as to the electron density. Notably, this analysis shows that the upper bound on the many-body gap scales linearly with carrier density. For example, in Moiré superlattices with limited carrier density ( $n \approx 0.027 \text{ nm}^{-2}$ ), the attainable many-body gap is fundamentally restricted to below  $\sim 10 \text{ meV}$  by this upper bound. This emphasizes that the energy and temperature scales of many-body phenomena realizable in Moiré superlattices are indeed significantly constrained by the limited flat band electron density. By contrast, the kagome flat band system studied here significantly boosts the theoretical upper bound on the energy gap by hosting substantially higher electron density. An independent microscopic calculation is required for each material system to verify whether such an enhanced gap is realizable in practice, which is precisely what our exact diagonalization calculations demonstrate for  $\text{A}_2\text{Pd}_3\text{S}_4$ .

**C) *Heuristic length-scale comparison:*** More heuristic approach to understand different energy scales between Moiré and kagome flat band systems is following. Outside of solid-state physics, ultracold optical lattices provide another platform for quantum many-body physics. While optical lattices allow highly controlled design of lattice geometry and interactions, their characteristic length scale ( $a \approx 1000 \text{ nm}$ ) far exceeds that of solid-state systems ( $a \approx 1 \text{ nm}$ ). This difference in characteristic length scale is directly reflected in the corresponding energy and temperature scales, with quantum phenomena in optical lattices occurring at drastically lower temperatures than in solid state systems, typically in the nK– $\mu\text{K}$  range. From this perspective, Moiré superlattices with a characteristic length scale ( $a \approx 10 \text{ nm}$ ) emulate physics between true crystalline systems ( $a \approx 1 \text{ nm}$ ) and ultracold optical lattices ( $a \approx 1000 \text{ nm}$ ), and its temperature scale is also correspondingly reduced. By contrast, the vdW kagome system studied here realizes a flat band within an intrinsic crystalline platform, opening a fundamentally new opportunity to access flat band phenomena at enhanced energy and temperature scales.

## 7. Additional details of transport measurements

A key challenge in transport measurements of semiconducting 2D materials often arises from large contact resistance due to Schottky barriers at the electrode/channel interface. For instance, early transport studies on 2D semiconducting transition metal dichalcogenides encountered similar challenges, and optimized device fabrication strategies emerged only after several years of sustained effort. While a systematic investigation of optimal contact metals will ultimately be required for comprehensive transport characterization of 2D vdW kagome lattices, we find that few-layer graphite contacts provide moderate contact resistance on  $A_2T_3X_4$ , enabling initial transport characterization of its gate-tunable response.

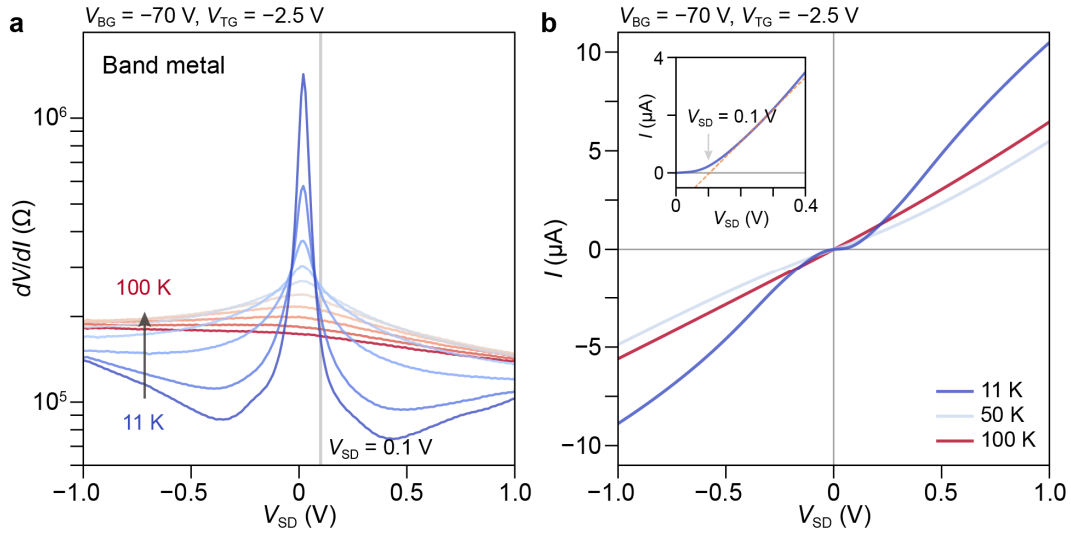


**Fig. S8 | Temperature-dependent differential resistance and current-voltage characteristics of the dual-gate transport device. a**, Differential resistance ( $dV/dI$ ) as a function of source-drain bias ( $V_{SD}$ ) measured at temperatures ranging from 11 K to 100 K in steps of 10 K. The measurements were performed in the band metal state at a global bottom-gate voltage  $V_{BG} = -70$  V and a top-gate voltage  $V_{TG} = -2.5$  V. The gray solid line indicates the DC bias  $V_{SD} = 0.1$  V applied for the lock-in measurements presented in Fig. 4. **b**, Corresponding current-voltage ( $I$ - $V$ ) characteristics at 11 K, 50 K, and 100 K. The inset provides a magnified view of the 11 K data, where the dashed orange line represents a linear fit, highlighting the recovery of the linear transport regime above  $V_{SD} = 0.1$  V.

To characterize contact properties, we measured the differential resistance of the two-terminal  $Rb_2Pd_3Se_4$  device as a function of source-drain bias in the band metal regime, as displayed in Fig. S8a. A large global bottom gate voltage ( $V_{BG} = -70$  V) effectively turns on the contact region, giving rise to ohmic contact with linear current-voltage ( $I$ - $V$ ) characteristics at high-temperatures,  $T > 50$  K (Fig. S8b). As the temperature is lowered, however, the differential resistance near zero source-drain bias sharply increases (Fig. S8a), which reflects the influence of

residual Schottky barrier and reduced thermal activation at low temperatures. To mitigate the Schottky contact resistance and access the intrinsic transport properties of the channel, we apply a finite DC bias voltage across the source and drain, similar to prior studies on semiconducting 2D material<sup>13</sup>. Specifically, based on the  $I$ - $V$  characteristics shown in the inset of Fig. S8b,  $V_{SD} = 100$  mV is chosen for all lock-in measurements, which represents the minimum bias required to reach the linear transport regime.

## 8. Tight-binding modeling and correlated states of $\text{Rb}_2\text{Pd}_3\text{S}_4$ and $\text{Cs}_2\text{Pd}_3\text{S}_4$

For the Hartree-Fock mean-field calculations and exact diagonalization (ED) analysis, we developed tight-binding models for  $\text{A}_2\text{Pd}_3\text{S}_4$ , as shown in Extended Data Fig. 10. Our model is based on a single-orbital tight-binding model on the kagome lattice, including hopping terms up to the seventh-nearest neighbors. Among the long-range hoppings, we note that the 3rd- and 6th-nearest-neighbor hoppings can occur along two distinct real-space paths, as illustrated in Fig. S9a. Taking this into account, we considered two different tight-binding models: In Model-I, we used a single parameter  $t_3$  ( $t_6$ ) to describe all 3rd (6th)-nearest-neighbor hoppings, while Model-II treated the two hopping paths using independent parameters,  $t_3$  and  $t_3'$  ( $t_6$  and  $t_6'$ ). The resulting band structures from two models are compared in Fig. S9b,c. While both models reproduce the topmost flat band with comparable accuracy, Model-II provides an improved fit to states at energies well below the flat band, thereby more faithfully capturing all three kagome band manifolds. Accordingly, all calculations presented in the Main text were performed using Model-II. The fitted tight-binding parameters are  $(t_1, t_2, t_3, t_3', t_4, t_5, t_6, t_6', t_7, \lambda_{\text{SOC}}) = (-0.1146, -0.0154, -0.0195, -0.0176, -0.0105, 0.0095, -0.0014, 0.0037, -0.0063, 0.0165)$  for  $\text{Rb}_2\text{Pd}_3\text{S}_4$  and  $(-0.1134, -0.0164, -0.0255, -0.0255, -0.0067, 0.0064, 0.0024, -0.0009, -0.008, 0.0167)$  for  $\text{Cs}_2\text{Pd}_3\text{S}_4$ , in eV units. At the same time, we use this Supplementary Section to compare results obtained from both models, demonstrating that the many-body phase diagram and fractional ground states obtained for  $\text{A}_2\text{Pd}_3\text{S}_4$  remain robust with respect to the detailed choice of tight-binding parameterization.

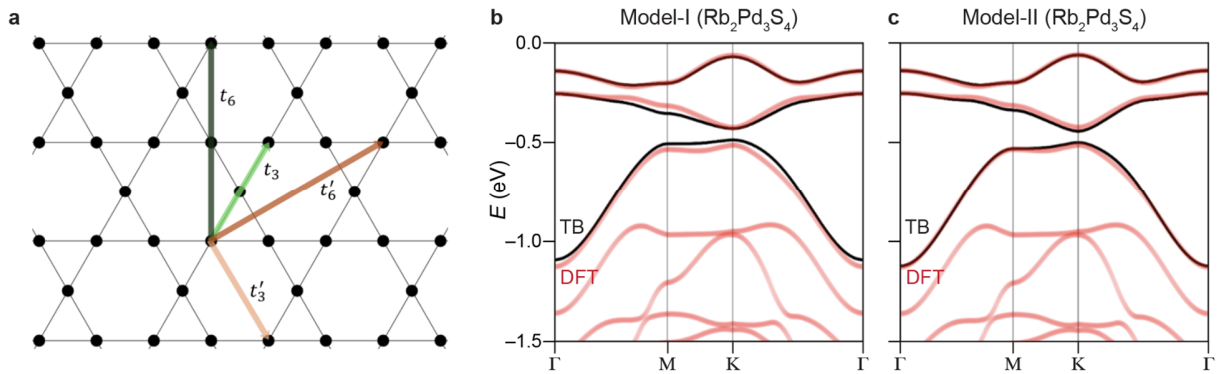


**Fig. S9. Tight-binding models for  $\text{Rb}_2\text{Pd}_3\text{S}_4$ .** **a**, Symmetry-inequivalent 3rd- and 6th-nearest-neighbor hoppings in the kagome lattice. **b,c**, Tight-binding models I and II (black lines) fit to the DFT dispersions (red lines).

**A) Many-body phase diagrams:** In Fig. S10a,b, we compare the Hartree-Fock calculations obtained from the Model-I (Fig. S9b) and Model-II (Fig. S9c) tight-binding parameters. It is apparent that, while the precise locations of the phase boundaries shift to smaller  $U$  for Model-II, the overall structure of the phase diagrams remains robust. In particular, the various types of noncollinear and stripe phases, as well as the predominant ferromagnetic phase, appear robust against the choice of the tight-binding model. This robustness of the phase diagram can be understood as follows: The differences in the band structure between two parameterizations are of order  $O(10)$  meV. Since the interaction scale relevant for the many-body phases is on the order of a few eV, the effect of the detailed tight-binding parametrization on the many-body phase diagram is expected to be negligible.

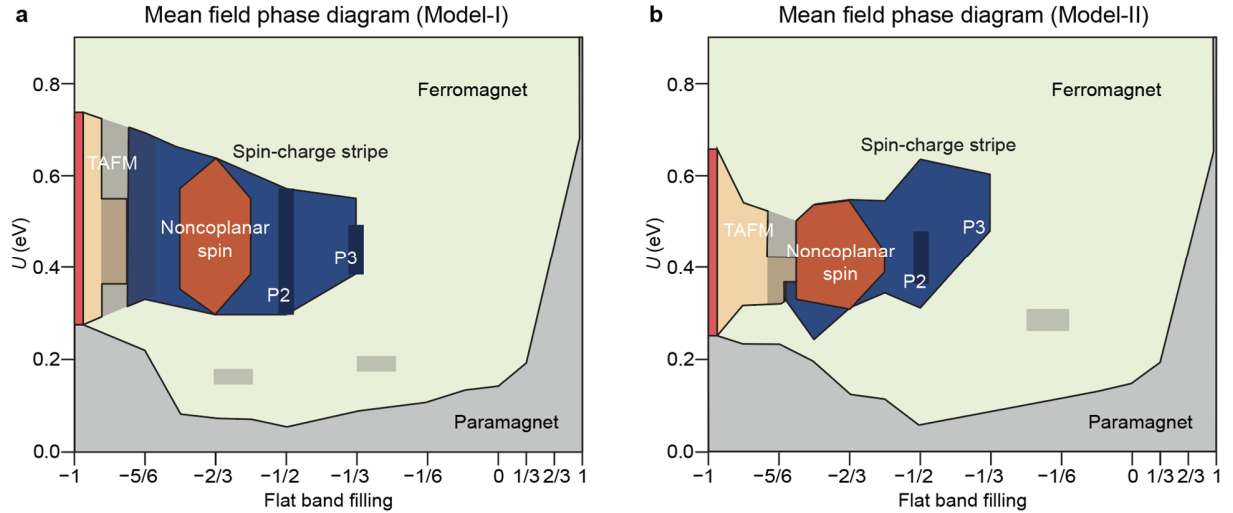


**Fig. S10. a,b,** Hartree-Fock phase diagram obtained using Model-I and Model-II tight-binding parameters for  $\text{Rb}_2\text{Pd}_3\text{S}_4$ , respectively.

**B) Fractional ground states:** In addition to the Hartree-Fock phase diagram, we also discuss exact diagonalization (ED) calculations for the kagome flat band. Before discussing the ED results for Model-I and Model-II, we first outline the details of our ED methodology.

In the ED calculations on our kagome system, band-mixing effects between the topmost flat topological band and the second-highest band should be taken into account, because the average band gap between the two is approximately 150 meV, which is smaller than the typical Coulomb interaction scale of about 450 meV. Since the gap between the topmost and the third band is approximately 600 meV, larger than the interaction scale, we expect the band mixing to be most significant between the topmost and second-highest bands. Therefore, we perform two-band

ED, incorporating configurations with at most one hole excited from the highest band to a lower band. This treatment captures the dominant band-mixing processes at leading order<sup>14–16</sup>. All ED calculations were performed on an  $N=21$  discretized momentum grid. The grid with  $k$ -points labeled are shown in Fig. S11.

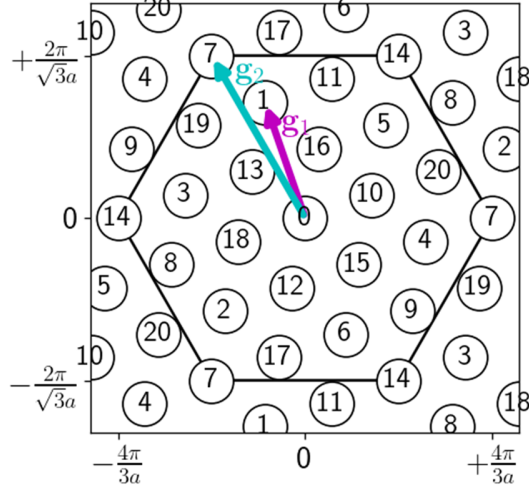


**Fig. S11.** Discretized momentum grid in the first Brillouin zone. In our momentum labeling, each sector is specified by  $k = k_1\mathbf{g}_1 + k_2\mathbf{g}_2$ .

In Fig. S12, we compare the ED calculations based on the Model-I and Model-II tight-binding band structures. For both models, the robust fractional ground states are observed at  $1/3$  filling, with energy gaps of approximately 20 meV for Model-I (Fig. S12c) and 14 meV for Model-II (Fig. S12d). For comparison, calculations without band mixing effects are also included for Model-I (Fig. S12a) and Model-II (Fig. S12b). These calculations are still performed within the two-band ED framework, but interband excitations from the lower band to the flat band are prohibited. Corresponding calculations for  $\text{Cs}_2\text{Pd}_3\text{S}_4$  are shown in Fig. S13 without (Fig. S13a) and with (Fig. S13b)) band mixing effects, yielding similar fractional ground states with a gap of approximately 29 meV (Fig. S13b).

These results together demonstrate that the fractional ground states in  $\text{A}_2\text{Pd}_3\text{S}_4$  remain robust against band mixing effects, detailed model parameterizations, and variations in the alkali metal elements. As a representative case, we present the results for  $\text{Cs}_2\text{Pd}_3\text{S}_4$  in Fig. 5f of the Main text.

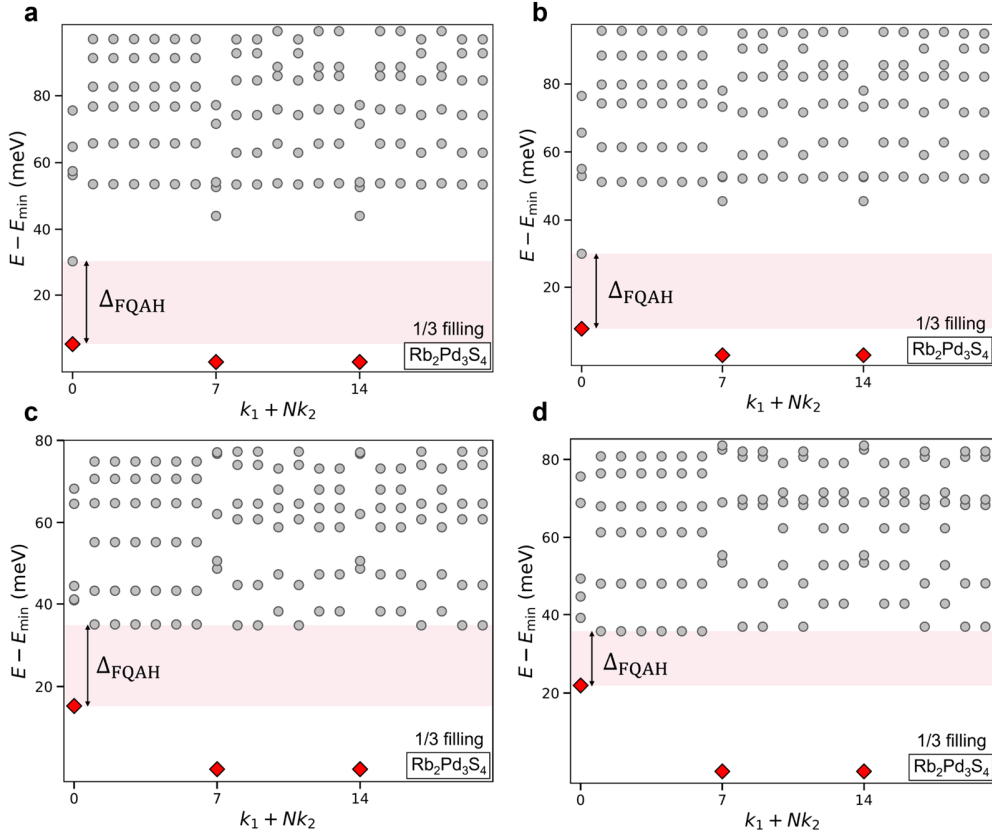


**Fig. S12.** **a,b**, Many-body energy spectra for  $\text{Rb}_2\text{Pd}_3\text{S}_4$  at 1/3-filling without band mixing, obtained using (a) Model-I and (b) Model-II. **c,d**, Corresponding spectra for  $\text{Rb}_2\text{Pd}_3\text{S}_4$  at 1/3-filling with band mixing, obtained using (c) Model-I and (d) Model-II. The three topological ground states are highlighted in red, and the energy gap separating them from the lowest excited state is denoted by  $\Delta_{\text{FQAH}}$ .

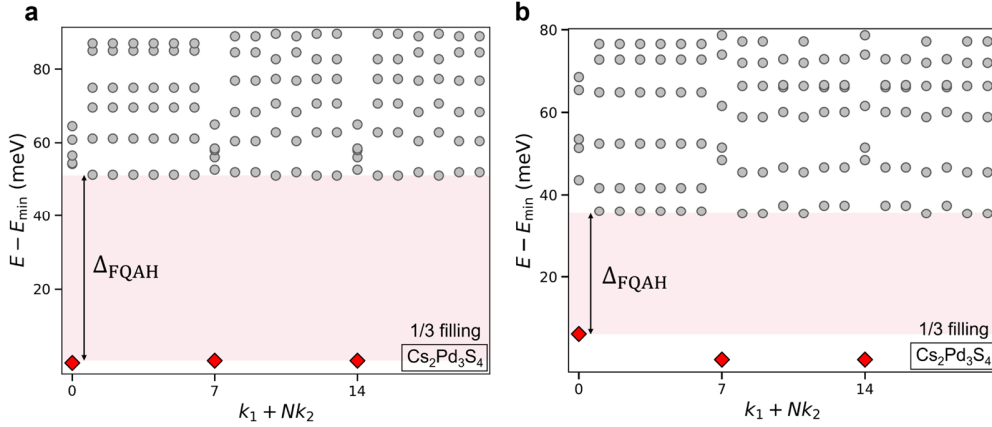


**Fig. S13.** **a,b**, Many-body energy spectra for  $\text{Cs}_2\text{Pd}_3\text{S}_4$  (a) without band mixing and (b) with band mixing. The three topological ground states are highlighted in red, and the energy gap separating them from the lowest excited state is denoted by  $\Delta_{\text{FQAH}}$ .

Before concluding this Section, we further note that the many-body phase diagram of  $A_2\text{Pd}_3\text{S}_4$  shown in Fig. S10 closely resembles that obtained for the ideal kagome lattice model<sup>17</sup>. This correspondence further indicates that the 2D  $A_2T_3X_4$  vdW kagome platform lies very close to the ideal kagome limit and can faithfully reproduce the essential physics of the model kagome lattice.

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