

# van Hove Singularity-Driven Emergence of Multiple Flat Bands in Kagome Superconductors

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The newly discovered Kagome superconductors  $AV_3Sb_5$  ( $A=K, Rb$  and  $Cs$ ) continue to bring surprises in generating unusual phenomena and physical properties, including anomalous Hall effect, unconventional charge density wave, electronic nematicity and time-reversal symmetry breaking. Here we report an unexpected emergence of multiple flat bands in the  $AV_3Sb_5$  superconductors. By performing high-resolution angle-resolved photoemission (ARPES) measurements, we observed **four branches of flat bands that span over the entire momentum space**. The appearance of the flat bands is not anticipated from the band structure calculations and cannot be accounted for by the known mechanisms of flat band generation. It is intimately **related to the evolution of van Hove singularities**. It is for the first time to observe such emergence of multiple flat bands in solid materials. Our findings provide new insights in revealing the underlying mechanism that governs the unusual behaviors in the Kagome superconductors. They also provide a new pathway in producing flat bands and set a platform to study the flat bands related physics.

The Kagome lattice, with corner-sharing triangle networks, engenders characteristic electronic structures with van Hove singularities (vHs) at the Brillouin zone boundary, Dirac cone at the zone corner and a flat band through the whole momentum space[1, 2]. Such unique electronic structures facilitate the exploration for a plethora of novel phenomena such as quantum spin liquid phase, non-trivial topology, superconductivity and other correlated phenomena[1, 1–11, 11–14]. The newly discovered Kagome superconductors  $AV_3Sb_5$  ( $A=K, Rb$  and  $Cs$ )[15, 16] have immediately attracted tremendous interests because they generate abundant quantum states and physical properties including the giant anomalous Hall effect without long-range magnetic order[17, 18], unconventional charge and pair density waves[19–22], electronic nematicity[23], time-reversal symmetry breaking[24, 25] and possible unconventional superconductivity[21, 24, 26–29]. Understanding of these novel quantum phenomena requires thorough investigations on the electronic structures of  $AV_3Sb_5$  systems.

On the other hand, the flat band systems have emerged as a fertile playground in condensed matter physics exemplified by the twisted bilayer graphene[30–32]. The flat bands are characterized by quenched kinetic energy, singular density of states, electron correlation and localization. Plenty of Coulomb interaction-driven many-body states are expected in flat band systems, such as ferromagnetism[33, 34], superconductivity[35, 36], Wigner crystal[37]

and quantum Hall states[38–41]. Theoretically, the flat bands are predicted mainly based on two origins: the localized atomic orbitals and some peculiar lattice geometries[33, 34, 36, 42–45]. However, the experimental observation of flat bands becomes possible only recently and its realization is limited to a few systems such as heavy fermion materials[46], Kagome compounds[47, 48] and twisted bilayer graphene[30, 49, 50].

We carried out angle-resolved photoemission (ARPES) measurements by using our lab-based laser ARPES system (see Methods). We systematically measured  $\text{AV}_3\text{Sb}_5$  ( $\text{A}=\text{K}, \text{Rb}$  and  $\text{Cs}$ ) (abbreviated as KVS, RVS and CVS, respectively) and Ti-substituted  $\text{CsV}_{3-x}\text{Ti}_x\text{Sb}_5$  ( $x=0.04, 0.15$  and  $0.27$ ) (abbreviated as CVS(0.04), CVS(0.15) and CVS(0.27), respectively) samples (see Methods).  $\text{AV}_3\text{Sb}_5$  exhibit charge density wave (CDW) transitions at  $\sim 80$  K in  $\text{KV}_3\text{Sb}_5$ ,  $\sim 102$  K in  $\text{RbV}_3\text{Sb}_5$  and  $\sim 93$  K in  $\text{CsV}_3\text{Sb}_5$ [15, 16, 51–53]. The Ti-substitution in  $\text{CsV}_{3-x}\text{Ti}_x\text{Sb}_5$  introduces holes and suppresses the CDW transition[53]. The  $\text{CsV}_{3-x}\text{Ti}_x\text{Sb}_5$  ( $x=0.04$ ) exhibits a CDW transition at  $\sim 63$  K while CDW transition is undetectable in the  $\text{CsV}_{3-x}\text{Ti}_x\text{Sb}_5$  ( $x=0.15$  and  $x=0.27$ ) samples[53].

Figure 1 shows band structure of  $\text{CsV}_3\text{Sb}_5$  measured along the  $\bar{\Gamma}$ – $\bar{\text{M}}$  high-symmetry direction at 20 K. In addition to the original  $\alpha$  band around  $\bar{\Gamma}$  and  $\beta$ ,  $\gamma$  and  $\delta$  bands around  $\bar{\text{M}}$ [16, 52], four dispersionless features are clearly observed that span the entire measured momentum space, as marked by arrows and labelled as FB1–FB4 in Fig. 1a. These features show up more clearly in the second-derivative images in Fig. 1b. Their energy positions are at the binding energies of  $\sim 70$  meV (FB1),  $\sim 200$  meV (FB2),  $\sim 550$  meV (FB3) and  $\sim 700$  meV (FB4). The presence of these flat bands can also be observed in the photoemission spectra (energy distribution curves, EDCs) (Fig. 1c) and gets more evident in the corresponding second-derivative EDCs (Fig. 1d).

The observation of these multiple flat bands are unexpected because no such bands are present in the calculated band structures [15, 52]. In our ARPES measurements with  $\sim 15 \mu\text{m}$  spot size, we observed various regions with different intensities of the folded bands (Fig. S1 in Supplementary Materials) related to the CDW order and surface reconstructions of different cleaved surfaces [21, 52, 54]. However, the flat bands can be observed both in regions with strong band foldings and in regions with little band foldings (Fig. S1 in Supplementary Materials), indicating that they are not induced by the bulk CDW or surface reconstructions. These flat bands are robust and can be observed in measurements under different polarization

geometries (Fig. S2 in Supplementary Materials). Furthermore, they are observed not only along  $\bar{\Gamma}$ - $\bar{M}$  direction, but also along  $\bar{\Gamma}$ - $\bar{K}$  and other directions (Fig. S3 in Supplementary Materials), thus covering the entire two-dimensional Brillouin zone.

The flat bands are ubiquitous in the  $AV_3Sb_5$  ( $A=K, Rb$  and  $Cs$ ) compounds and show a peculiar doping dependence in  $CsV_{3-x}Ti_xSb_5$  ( $x=0\sim 0.27$ ). Fig. 2 shows the band structures measured at 20 K along the  $\bar{M}$ - $\bar{\Gamma}$ - $\bar{M}$  direction in  $KV_3Sb_5$  (Fig. 2a),  $RbV_3Sb_5$  (Fig. 2b),  $CsV_3Sb_5$  (Fig. 2c),  $CsV_{3-x}Ti_xSb_5$  ( $x=0.04$ ) (Fig. 2d),  $CsV_{3-x}Ti_xSb_5$  ( $x=0.15$ ) (Fig. 2e) and  $CsV_{3-x}Ti_xSb_5$  ( $x=0.27$ ) (Fig. 2f) samples. The corresponding second-derivative images are shown in Fig. 2g-2l. The EDCs for different samples at a particular momentum away from the main bands are presented in Fig. 2m, with contributions mainly from the flat bands. The corresponding second-derivative EDCs are presented in Fig. 2n. The multiple flat bands are clearly observed in all the parent  $AV_3Sb_5$  ( $A=K, Rb$  and  $Cs$ ) compounds with similar energy scales. Upon hole doping through Ti-substitution in  $CsV_{3-x}Ti_xSb_5$ , the FB1 and FB2 bands remain pronounced with only a slight energy position variation due to hole doping-induced chemical potential shift. In contrast, the FB3 and FB4 bands exhibit a dramatic change with the hole doping. With increasing hole doping, the FB3 and FB4 bands get weaker in the spectral intensity, shrink in their energy separation and eventually merge into a single flat band FB34 at the high doping in the CVS(0.27) sample. The quantitative energy position variation of the flat bands in these samples is summarized in Fig. 2o. It has been found that the hole doping in  $CsV_{3-x}Ti_xSb_5$  suppresses the CDW transition which is completely undetectable in the high doping CVS(0.27) sample[53]. The observed doping evolution of the FB3 and FB4 flat bands is closely related to the doping evolution of the CDW transition in  $CsV_{3-x}Ti_xSb_5$ .

In order to further check on the relation between the flat bands and the CDW transition, we carried out temperature-dependent measurements on  $RbV_3Sb_5$  across the CDW transition temperature of  $\sim 102$  K. Fig. 3 shows the band structures of  $RbV_3Sb_5$  measured along the  $\bar{M}$ - $\bar{\Gamma}$ - $\bar{M}$  direction at different temperatures. The corresponding EDCs and the second-derivative EDCs at a particular momentum are presented in Fig. 3b and 3c, respectively. As seen in Fig. 3, the FB1 and FB2 flat bands are observed in the entire temperature range of 20 $\sim$ 170 K. Their energy positions show little temperature dependence when crossing the CDW transition. On the other hand, the FB3 and FB4 flat bands exhibit a dramatic evo-

lution with temperature. With the increasing temperature, the spectral intensity of these two flat bands gets weaker, the energy separation between them gets smaller, and eventually these two flat bands FB3 and FB4 merge into one flat band FB34 at temperatures above the CDW transition temperature of  $\sim 102$  K. The energy position evolution with temperature for the flat bands is summarized in Fig. 3d. The temperature dependence of the flat bands shows a strong resemblance to the doping dependence shown in Fig. 2. These strongly indicate that the origin of the FB3 and FB4 flat bands is closely related to the CDW transition.

In order to understand the origin of the flat bands, we compared their energy positions with those original bands and found that the emergence of the flat bands is closely related to the van Hove singularities at the  $\bar{M}$  point. Fig. 4a shows the band structure of  $\text{CsV}_3\text{Sb}_5$  measured along  $\bar{\Gamma}$ - $\bar{M}$  direction at 20 K in the CDW state. This measurement covers both the flat bands and the original bands at the  $\bar{M}$  point, making their direct comparison possible. As seen from the band structure calculations in Fig. 4b, in the CDW state, three van Hove singularities (vHs1, vHs2 and vHs3) are present at  $\bar{M}$ . Each van Hove singularity splits in energy and forms an upper branch (vHs1u, vHs2u and vHs3u) and a lower branch (vHs1l, vHs2l and vHs3l). The observed bands at  $\bar{M}$  in Fig. 4a show a good correspondence to those in Fig. 4b, as labelled on the right of Fig. 4a. Fig. 4c shows the second-derivative EDCs measured at  $k_{\parallel} = -0.27$   $1/\text{\AA}$  in Fig. 4a highlighting the contribution of the flat bands and at  $k_{\parallel} = +0.66$   $1/\text{\AA}$  in Fig. 4a emphasizing the contribution of the original bands at  $\bar{M}$ . The energy positions of the flat bands exhibit a good correspondence to those of the van Hove singularity bands. When it comes to the  $\text{CsV}_{3-x}\text{Ti}_x\text{Sb}_5$  ( $x=0.27$ ) sample with high hole-doping without CDW transition, there remains three van Hove singularities expected at  $\bar{M}$  (Fig. 4e). However, the vHs3 does not split although vHs1 and vHs2 keep splitting at  $\bar{M}$ . The expected three bands can be clearly observed in Fig. 4d at  $\bar{M}$  as marked on its right side. Fig. 4f shows the second-derivative EDCs measured at  $k_{\parallel} = -0.27$   $1/\text{\AA}$  and at  $\bar{M}$  from Fig. 4d. Again, the energy positions of the flat bands shows an excellent agreement with those of the van Hove singularity bands. These results strongly indicate that the emergence of the flat bands is intimately linked to the band structures at  $\bar{M}$ , particularly the van Hove singularities.

Flat bands systems, constituted by localized or dressed heavy electrons with large effective masses, are an ideal platform to explore for correlated electronic phenomena, such as

magnetism and unconventional superconductivity [30, 31, 55]. There are several mechanisms that can produce the dispersionless extension of spectral intensity in the momentum space. When electrons are localized within atoms and the electron hopping is limited, a flat band can be formed, such as the **4f electrons in heavy Fermion** materials (Fig. 4g) [55]. In some special lattice geometries, like in **Kagome lattice** (Fig. 4h), the flat band can be formed due to destructive interference [31]. In addition, we note that there are two other ways to produce flat band-like features. In the case of **electron coupling with a bosonic mode, a flat band-like feature may appear at the mode energy  $\Omega_0$**  (Fig. 4i, see Fig. S4 in Supplementary Materials) [56]. In the case of **electron-mode coupling with the presence of a vHs, a flat band-like feature may emerge at  $\Omega_0 + E$  with  $E$  being the binding energy of the vHs** (Fig. 4j, see Fig. S5 in Supplementary Materials) [57].

Our measured results provide important information on understanding the origin of the flat bands. Since multiple flat bands are observed in  $\text{AV}_3\text{Sb}_5$  and their energies coincide with those of vHs, this rules out the possibility that these flat bands come from impurity bands [58–60]. This can also exclude the possibility of localized atomic orbital bands (Fig. 4g). One possibility is whether these flat bands may come from the **scattering of electrons at the vHs by impurities or disorders**. To the best of our knowledge, such phenomena have not been observed in the previous ARPES measurements of materials with vHs [47, 61]. Although the cleaved sample surface consists of different regions, like Cs-terminated, half Cs-terminated and Sb-terminated in  $\text{CsV}_3\text{Sb}_5$  [21], we always observe similar flat bands, even in regions with strong band foldings and in regions with little band foldings (Fig. S1 in Supplementary Materials). These results indicate that the flat bands we observed are more likely attributed to the intrinsic bulk properties of  $\text{AV}_3\text{Sb}_5$  materials.

Our observed flat bands are different from the Kagome lattice-induced ones (Fig. 4h) which are  $\sim 1$  eV away from the Fermi level [62]. Electron-mode coupling can produce flat band-like feature (Fig. 4i). However, our observation of four flat bands is unlikely due to such electron-mode coupling because no collective excitations with similar energy scales have been observed in  $\text{AV}_3\text{Sb}_5$ . Furthermore, in the electron-mode coupling picture (Fig. 4i), the spectral weight of the flat band-like feature decays rapidly with the momentum moving away from the main band. This is inconsistent with our observation that the spectral weight of the flat bands changes little in the wide momentum space. In the case of electron-mode coupling with a coexisting vHs, the flat band-like feature can also occur near the energy

position of the vHs (Fig. 4j). This scenario closely connects the flat band-like feature with the vHs which is consistent with our observations. But similar to the electron-mode coupling case, the flat band-like feature loses its spectral weight rapidly with the momentum moving away from the vHs. This is not consistent with the nearly uniform momentum distribution of the flat bands we observed.

In summary, we have discovered the emergence of multiple flat bands in  $AV_3Sb_5$ . These observations are unexpected and the origin of these flat bands can not be understood by the known mechanisms of flat band generation. The occurrence of these flat bands is clearly associated with the van Hove singularities which may be further related to their singular density of states (DOS). The van Hove singularities have been reported in a number of systems, but the vHs-driven generation of the flat bands has been observed for the first time in  $AV_3Sb_5$ . These indicate that some unusual mechanisms are at play in  $AV_3Sb_5$ , like the discovery of the unexpected anomalous Hall effect in this system. Our work provides a new paradigm to generate flat bands and will stimulate further efforts to understand its origin and other unusual phenomena in Kagome superconductors.

## Methods

**Growth and characterization of single crystals.** High quality single crystals of  $AV_3Sb_5$  ( $A=K, Rb$  and  $Cs$ ) and  $CsV_{3-x}Ti_xSb_5$  ( $x=0.04, 0.15$  and  $0.27$ ) were grown from modified self-flux methods [15, 53]. The crystals were characterized by X-ray diffraction and measurements of the magnetic susceptibility and electrical resistance [52, 53].  $AV_3Sb_5$  exhibit CDW transitions at  $\sim 80$  K ( $KV_3Sb_5$ ),  $\sim 102$  K ( $RbV_3Sb_5$ ) and  $\sim 93$  K ( $CsV_3Sb_5$ ) [15, 16, 51–53], respectively. The Ti-substitution for V in  $CsV_{3-x}Ti_xSb_5$  crystal has been confirmed by the scanning transmission electron microscopy, electron energy-loss spectroscopy and scanning tunneling microscope [53].  $CsV_{3-x}Ti_xSb_5$  ( $x=0.04$ ) exhibits a CDW transition at  $\sim 63$  K while no CDW transition is observed in the  $CsV_{3-x}Ti_xSb_5$  ( $x=0.15$  and  $x=0.27$ ) samples [53].

**High resolution ARPES measurements** High-resolution angle-resolved photoemission measurements were carried out on our lab system equipped with a Scienta DA30L electron energy analyzer [63, 64]. We use ultraviolet laser as the light source that can provide a photon energy of  $h\nu=6.994$  eV with a bandwidth of  $0.26$  meV. The energy resolution was set at  $\sim 2.5$  meV. The angular resolution is  $\sim 0.3^\circ$ , corresponding to a momentum resolution of

$\sim 0.0041/\text{\AA}$  for the photon energy of 6.994 eV. The laser spot size is set at  $\sim 15 \mu\text{m}$ . The Fermi level is referenced by measuring on a clean polycrystalline gold that is electrically connected to the sample. The sample was cleaved *in situ* and measured in vacuum with a base pressure better than  $5 \times 10^{-11}$  Torr.

**Band structure calculations.** First-principles calculations are performed by using the Projected Augmented Wave Method (PAW) density functional theory (DFT), as implemented in the Vienna Ab Initio Simulation Package (VASP) [65–67]. A  $2 \times 2 \times 1$  supercell is constructed to describe the TrH CDW phase of  $\text{AV}_3\text{Sb}_5$ . The crystal structures are relaxed by using the Perdew-Burke-Ernzerhof (PBE) functional [68] and zero damping DFT-D3 van der Waals correction [69] until the forces are less than 0.001 eV/ $\text{\AA}$ . The cutoff energy of the plane wave basis is set at 600 eV and the energy convergence criterion is set at  $10^{-7}$  eV. The corresponding Brillouin zones are sampled by using a  $16 \times 16 \times 10$  (for primitive cell) and a  $8 \times 8 \times 10$  (for supercell)  $\Gamma$  centered  $\mathbf{k}$ -grid. The effective band structure is calculated by the band-unfolding method [70, 71] proposed by Zunger et al. with BandUP code [72, 73]. The  $2 \times 2 \times 1$  TrH CDW order reconstruction is an input in our band-unfolding calculation. All the DFT calculations are *ab-initio* without any adjustable parameters except for the standard exchange-correlation functional and pseudopotentials.

**Spectral function simulation of the electron-mode coupling.** We performed a simulation of the **single-particle spectral function considering the electron coupling with a bosonic mode**. The purpose here is to illustrate the emergence of the flat band-like feature in the case of electron-mode coupling with a coexisting vHs. We follow the fashion of the Migdal theory [74] and calculate the Migdal electron-mode self-energy  $\Sigma_{ep}$ .

$$\Sigma_{ep}(\mathbf{k}, i\omega_n) = \frac{1}{N} \sum_{\mathbf{q}} |g_{\mathbf{k}, \mathbf{q}}|^2 \left( \frac{b(\Omega_{\mathbf{q}}) + f(\epsilon_{\mathbf{k}+\mathbf{q}})}{i\omega_n + \Omega_{\mathbf{q}} - \epsilon_{\mathbf{k}+\mathbf{q}}} + \frac{1 + b(\Omega_{\mathbf{q}}) - f(\epsilon_{\mathbf{k}+\mathbf{q}})}{i\omega_n - \Omega_{\mathbf{q}} - \epsilon_{\mathbf{k}+\mathbf{q}}} \right), \quad (1)$$

For simplicity, we considered a case of Einstein mode with constant frequency  $\Omega_{\mathbf{q}} \equiv \Omega_0$  and homogeneous electron-mode coupling constant  $g_{\mathbf{k}, \mathbf{q}} \equiv g$ . Here,  $b(\Omega)$  is the Bose-Einstein distribution function, and  $f(\epsilon_{\mathbf{k}})$  is the Fermi-Dirac distribution. The spectral function is given by  $A(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im} G(\mathbf{k}, i\omega_n \rightarrow \omega + i\delta)$ , where the Green function  $G(\mathbf{k}, i\omega_n) = (i\omega_n - \epsilon_{\mathbf{k}} - \Sigma_{ep})^{-1}$ . In the simulation, we take the case of square lattice, and the dispersion  $\epsilon_{\mathbf{k}} = 2t(\cos k_x + \cos k_y)$ . The cut is along the high symmetry line  $\Gamma - \text{X} - \text{M}$ . The vHs



at  $X = (\pi, 0)$  is placed at the energy of  $-0.4\text{ eV}$ . The mode frequency  $\Omega_0$  is set to be  $30\text{ meV}$  and the coupling constant is  $g = 0.2$ . The simulated results are shown in Fig. S5 in Supplementary Materials and in Fig. 4j.

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### Author Contributions

X.J.Z., H.J.G., L.Z. and H.L.L. conceived this project. H.L.L. performed ARPES experiments and analyzed the ARPES data. Z.Z., H.T.Y., C.M.S. and H.J.G. contributed to crystal growth of pure  $\text{CsV}_3\text{Sb}_5$  and Ti-doped  $\text{CsV}_3\text{Sb}_5$ . H.X.L. and Y.G.S. contributed to crystal growth of  $\text{KV}_3\text{Sb}_5$ ,  $\text{RbV}_3\text{Sb}_5$  and pure  $\text{CsV}_3\text{Sb}_5$ . Y.P.H., Y.H.G., J.P.H., K.J. and Z.Q.W. contributed to DFT calculations and theoretical analysis. F.J. and Q.M.Z. contributed to Raman measurements and analysis. L.Z., H.C., C.H.Y., T.M.M., X.L.R., B.L., Y.J.S., Y.W.C., H.Q.M., G.D.L. and X.J.Z. contributed to the development and maintenance of the ARPES systems and related software development. H.L.L., F.F.Z., F.Y., S.J.Z., Q.J.P. and Z.Y.X. contributed to the maintenance of  $7\text{ eV}$  laser system. H.L.L., L.Z. and X.J.Z. wrote this paper with information from all co-authors. All authors participated in discussion and comment on the paper.

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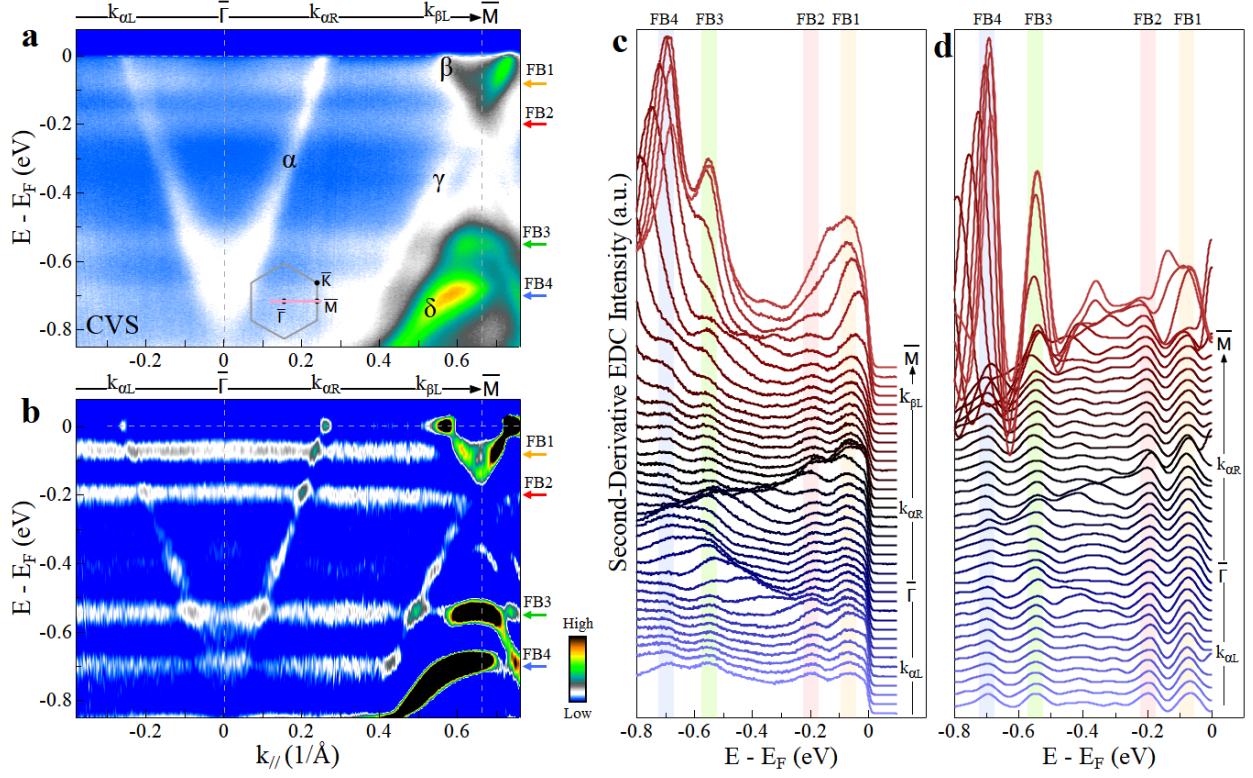


FIG. 1: **Observation of multiple flat bands over the entire momentum space in  $\text{CsV}_3\text{Sb}_5$ .** **a**, Band structure measured at 20 K along the  $\bar{\Gamma}$ - $\bar{M}$  direction. The location of the momentum cut is marked as a solid pink line in the inset. **b**, The corresponding second-derivative image with respect to energy obtained from (a). The four arrows on the right side of (a) and (b) are guides to the eye for the observed flat bands. **c**, The corresponding photoemission spectra (energy distribution curves, EDCs) from the band structure in (a). The momentum range where the EDCs are extracted is marked as a black arrow on the top of (a). The EDCs extracted at the Fermi momenta ( $k_{\alpha L}$ ,  $k_{\alpha R}$  and  $k_{\beta L}$ ) and high-symmetry points ( $\bar{\Gamma}$  and  $\bar{M}$ ) are marked. Four vertical shaded regions at the binding energies of 70 meV (FB1), 200 meV (FB2), 550 meV (FB3) and 700 meV (FB4) are guides to the eye for the flat bands. **d**, The corresponding second derivative EDCs from the second derivative image in (b). The momentum range where the second derivative EDCs are extracted is marked as a black arrow on the top of (b).

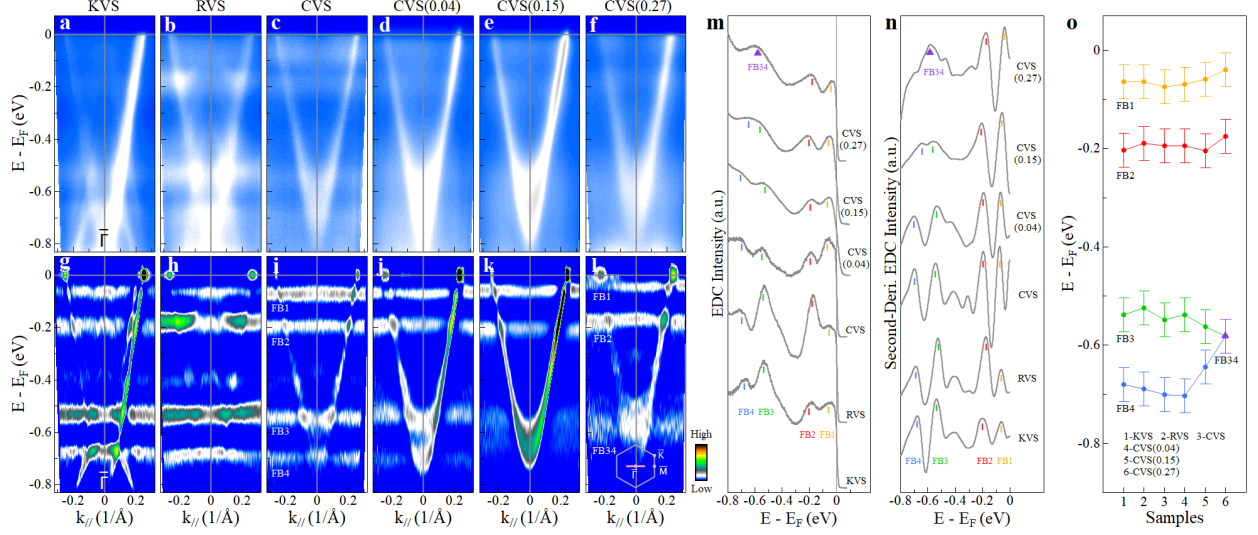


FIG. 2: Ubiquitous flat bands in  $AV_3Sb_5$  ( $A=K, Rb$  and  $Cs$ ) and their doping dependence. **a-f**, Band structures measured at 20K along the  $\bar{M}-\bar{\Gamma}-\bar{M}$  direction in  $KV_3Sb_5$  (KVS) (a),  $RbV_3Sb_5$  (RVS) (b),  $CsV_3Sb_5$  (CVS) (c),  $CsV_{3-x}Ti_xSb_5$  ( $x=0.04$ ) (CVS(0.04)) (d),  $CsV_{3-x}Ti_xSb_5$  ( $x=0.15$ ) (CVS(0.15)) (e) and  $CsV_{3-x}Ti_xSb_5$  ( $x=0.27$ ) (CVS(0.27)) (f). The location of the momentum cuts for (a)-(f) is marked as a solid pink line in the inset of (l). **g-l**, The corresponding second-derivative images with respect to energy obtained from (a)-(f). **m**, EDCs extracted at  $k_{||} = -0.27$   $1/\text{\AA}$  from (a)-(f). This momentum point is chosen to highlight the contribution from the flat bands because it is away from the original and folded bands. FB1-FB4 and FB34 mark the position of the flat bands. In each EDC, the position of the flat bands are marked by bars or triangles. **n**, Second derivative EDCs extracted at  $k_{||} = -0.27$   $1/\text{\AA}$  from (g-l). **o**, Sample-dependent and doping-dependent energy positions of the observed flat bands. Samples 1-6 represent KVS, RVS, CVS, CVS(0.04), CVS(0.15) and CVS(0.27), respectively, as marked on the bottom. The error bars reflect the uncertainty in determining the energy positions of the flat bands.

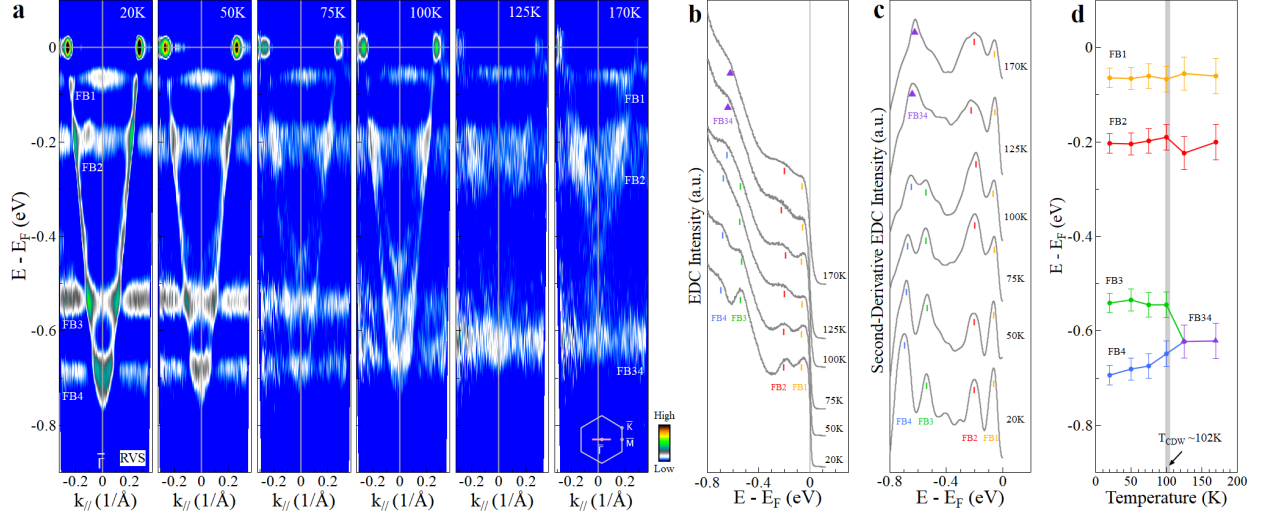


FIG. 3: **Evolution of the flat bands with temperature in  $\text{RbV}_3\text{Sb}_5$ .** **a**, Band structures measured at different temperatures from 20 K to 170 K along the  $\bar{M}-\bar{\Gamma}-\bar{M}$  direction. The location of the momentum cut is marked as a solid pink line in the inset of the right-most panel. These are second derivative images with respect to energy obtained from the original data. **b**, The EDCs at  $k_{//} = +0.30$  1/Å measured at different temperatures extracted from the original data. **c**, The corresponding second-derivative EDCs at  $k_{//} = +0.30$  1/Å measured at different temperatures extracted from (a). FB1-FB4 and FB34 indicate the energy positions of the flat bands. In each EDC, the positions of the flat bands are marked by bars or triangles in different colors. **d**, Temperature-dependent energy positions of the flat bands. The error bars reflect the uncertainty in determining the energy positions of the flat bands.

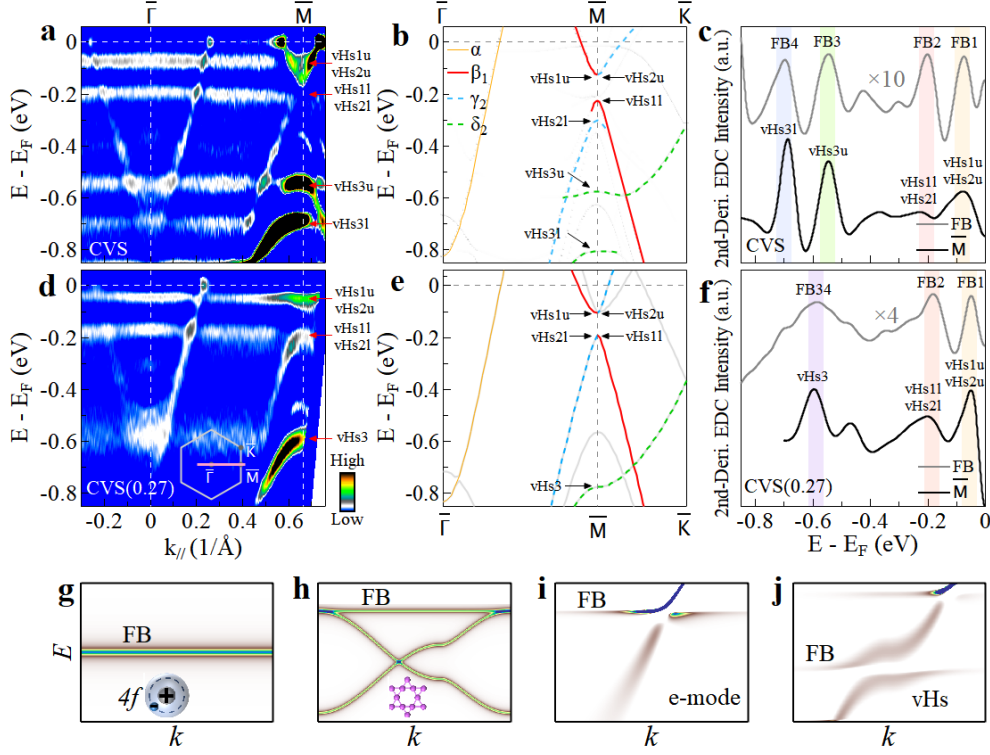


FIG. 4: **The relation between the flat bands and the van Hove singularities and the origins of flat bands.** **a and d**, Band structures measured at 20 K along the  $\bar{\Gamma}$ - $\bar{M}$  direction in  $\text{CsV}_3\text{Sb}_5$  (a) and  $\text{CsV}_{3-x}\text{Ti}_x\text{Sb}_5$  ( $x=0.27$ ) (d). The location of the momentum cuts is marked as a solid pink line in the inset of (d). These are second-derivative images with respect to energy obtained from original data. **b and e**, Calculated band structures along the  $\bar{\Gamma}$ - $\bar{M}$ - $\bar{K}$  direction in  $\text{CsV}_3\text{Sb}_5$  with the reconstructed TrH crystal structure[75] (b) and with pristine crystal structure (e) at  $k_z = \pi/c$  without spin-orbit coupling. The van Hove singularities (vHs1, vHs2 and vHs3) are marked by arrows which are associated with the  $\beta_1$ ,  $\gamma_2$  and  $\delta_2$  bands, respectively, that are highlighted by colored lines. When a van Hove singularity splits at  $\bar{M}$ , the two branches are labelled with suffixes u (up) and d (down) such as vHs3u and vHs3d. **c**, Second-derivative EDCs at  $k_{//} = +0.30 \text{ 1/\AA}$  (grey line) contributed mainly from the flat bands and at  $\bar{M}$  (black line) contributed mainly from the van Hove singularities, extracted from (a). For comparison, the second-derivative EDC at  $k_{//} = +0.30 \text{ 1/\AA}$  is multiplied by 10 in intensity. **f**, Same as (c) but extracted from (d). **g**, Flat bands in heavy fermion materials. **h**, The flat band in Kagome lattice. **i**, The flat band-like feature in electron-mode coupling [56]. This is the EDC second derivative image; the original image is shown in Fig. S4 in Supplementary Materials. **j**, The flat band-like feature coming from van Hove singularity in an electron-mode coupling system. This is the EDC second derivative image; the original image is shown in Fig. S5 in Supplementary Materials.