

Supplemental Materials for "Bulk Superconductivity driven by Disorder-Induced Delocalization"

Lu Chen^{1,2,3,*}, Sae-Hee Ryu^{4,*}, Avior Almoalem³, Yuanqi Lyu², Koh Yamakawa², Luke Pritchard Cairns², Ryan Day², Ehud Altman², Daniel Podolsky⁵, Dung-Hai Lee^{1,2}, Vidya Madhavan³, Eli Rotenberg⁴ & James G. Analytis^{1,2,6,7}

¹*Materials Science Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

²*Department of Physics, University of California, Berkeley, California 94720, USA*

³*Department of Physics, University of Illinois Urbana-Champaign 61801, USA*

⁴*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

⁵*Physics Department, Technion, Haifa 32000, Israel*

⁶*CIFAR Quantum Materials, CIFAR, Ontario, Toronto M5G 1M1, Canada*

⁷*Kavli Energy NanoScience Institute, Berkeley, California 94720, USA*

S1 Temperature-dependent Hall resistivity

The field dependence of in-plane electrical Hall resistivity $\rho_{xy}(H)$ for $4Hb\text{-Ta}(\text{S}_{1-x}\text{Se}_x)_2$ $x = 0\%$, $x = 0.25\%$, and $x = 1\%$ samples measured at selected temperatures are plotted in Fig. S1. For $x = 0\%$ sample, $\rho_{xy}(H)$ is linear-in-field above $H \sim 1\text{ T}$ at all measured temperatures and has a non-linear field dependence below $\sim 1\text{ T}$. For $x = 0.25\%$ and $x = 1\%$ samples, $\rho_{xy}(H)$ shows a stronger non-linear field dependence below $\sim 30\text{ K}$, clearly suggesting multiple bands participate in $\rho_{xy}(H)$. ρ_{xy} of 0% sample shows a sign change from negative to positive as temperature increases above 35 K [18], consistent with a CCDW transition that happens around 30 K (as shown in Fig. 1 (c)). Above 35 K , ρ_{xy} is dominated by a hole band, while it is dominated by an electron band below 35 K . Compared to the 0% sample, 0.25% and 1% samples are dominated by the hole band across the whole measured temperature range. The changes in the sign and field dependence of ρ_{xy} reveal that the underlying CCDW phases - modulated by Se concentration - strongly alter the Fermi surface structure and related band parameters.

S2 Chemical origin of Se substitution for core-level spectroscopy

Fig. S2 shows x-ray photoelectron spectroscopy (XPS) results for Se and S atoms in $4Hb\text{-Ta}(\text{S}_{1-x}\text{Se}_x)_2$. Although the concentration is low, it is possible to observe Se $3d$ core electrons for both T - and H -terminated surfaces in Fig. S2 (a,b), respectively. These spectra consist of two spin-orbit split doublets, whose intensities are proportional to their proximity to the surface, owing to the short escape depth of the electrons. Thus in (a) we assign the stronger, blue doublet to Se in the outermost surface layer with $1T$ termination, while in (b) the stronger, blue doublet is assigned to $1H$ termination.

The energy shift between the H and T is due to their chemical state difference and can be explained by the charge transfer from T to H layers. However, there are also final state relaxation contributions to the absolute binding energies that relate to the local electronic screening in each layer. The data also clearly show a proportional growth of spectral weight as Se concentration increases, verifying our stated Se concentrations.

The simple 2-component fitting model employed hides the complexity of the distribution of chemical shifts expected from the CDWs in each layer through the inclusion of Gaussian broadening. The buried T -layer's Se doublet in (b) is much broader than in the exposed T layer seen in (a). This indicates a larger spread of charge density among atomic sites in the buried layers, either due to enhanced CDW strength, or due to enhanced interactions with H layers above and below. Further investigation is needed to understand this.

Fig.S2 (c,d) show corresponding spectra for S $2p$. Like Se $3d$, these also show a partitioning of the spectrum into (to lowest order) two doublets corresponding to T - and H -layer S atoms with their intensities ordered by their surface proximity. From this we can conclude that the Se and S atoms sit in chemically identical states, with identical chemical shifts. This confirms that the Se occupies S sites, and substitutes into the T and H layers roughly equally.

Compared to Se, the S statistics are much better and we could discern additional fine structure, such as a small splitting of the T -layer's peaks. Furthermore, the spectra are well-fitted when the H layers include a tail towards high binding energy, modelled using the Shirley function. This reflects the fact that H layers are metallic, with the tail representing energy loss of the outgoing electrons to Fermi liquid excitations. For the H surfaces, we found, as for Se, that the buried T S atoms have a broader energy distribution. This was represented by a broad doublet (light blue) combined with two sharper peaks (darker blue).

S3 H-layer Fermi surfaces and charge-transfer analysis

Fig. 3(c,d) show the Fermi surfaces of the H -surface layers for the different sample classes. These Fermi surfaces are well-reproduced by the TB calculation for monolayer $1H$. They are qualitatively similar to those of the buried H -layers in Fig. 3 (a,b), but are stronger and sharper due to the lack of attenuation through overlaying layers, and differ in details such as the topology around a saddle point Van Hove Singularity (VHS, marked V in the figure) and overall dimensions. These differences are understood within a simple charge transfer model illustrated in Fig. S3, in which each T -layer donates a fixed negative charge $-Q$ to its adjacent H layers. Due to the surface termination, the surface T and H layers will have net charge $\pm Q$ while deeper layers will have net charge $\pm 2Q$. Therefore the charge transfer difference between surface and subsurface H determines Q and the overall charge state of all layers.

Density Functional Theory (DFT) calculations for the SOD reconstruction predict the T -

layer to host a narrow half-filled flat band derived from d_{z^2} orbitals at E_F . The flat band holds exactly $1 e^-$ per SOD cluster (13 Ta atoms), or equivalently $1/13 \approx 0.077 e^-$ per Ta atom in the 1×1 unit cell. Quantifying the charge transfer between the T and H layers is therefore important for determining the occupancy of this band. To estimate the charge content in our bulk layers, we begin by estimating the interlayer charge transfer Q using Luttinger's theorem, by comparing the Luttinger area of surface and buried H layer Fermi contours. Previously, Q was estimated by comparing the H -layer Fermi surface of $4Hb$ -TaS₂ to that of $2H$ -TaS₂ [22]. In contrast, our measurement compares two side-by-side spots on the same $4Hb$ -TaS₂ crystal — one H-terminated (surface H -layer) and one T-terminated (subsurface H -layer) — which eliminates random measurement errors, and systematic errors from sample quality, sample-to-sample differences in band structure, lattice constant, and work function.

To quantitatively analyze the H -layer Fermi surfaces, we fitted the experimental data using a tight-binding (TB) model based on the three-band parametrization of Liu *et al.* [48]. The fitting was performed using the H -terminated surface of the 0% Se sample measured at $T = 46$ K, which is nominally above the low-temperature 3×3 CDW transition of the H layer in $4Hb$ -TaS₂. The model does not include the 3×3 or $\sqrt{13} \times \sqrt{13}$ R13.9° reconstructions and is therefore used only to describe the underlying unreconstructed H -layer Fermi surface and estimate charge transfer on a per-Ta basis relative to the (1×1) unit cell.

In Fig. 3(a,d), the TB models have been adjusted to the different charge levels in the (a) subsurface and (d) surface H layers. This amounts to a relative shift of E_F in (d) being 16 meV lower than in (a). This energy shift is corroborated by a similar bandwidth change ΔW in the bandstructure slices, which is similarly narrower in (d) than in (a). Furthermore, the shape of the bands near point V shows that in (a) the VHS is nearly at E_F while in (d) the VHS has been pushed above E_F . However, calculation of the charge transfer based on the measurement of ΔW or the position of V is complicated because the bottom of the band and the VHS are influenced by the CDWs that are not included in the TB model. Therefore, we optimize the fits using features at the Fermi surface that are least affected by the CDWs.

Namely, due to mirror symmetry in the central plane of the H -layer, the out-of-plane spins are completely decoupled eigenstates, resulting in two bands of opposite spin having six symmetry-protected crossings C. The bands at C are typically strong and sharp, are protected from splitting, and are not strongly affected by the CDWs. Denoting the distance between opposite C points as $\overline{CC}$, we derive from the TB model the rate of charge accumulated by the H -layer for a given change in $\overline{CC}$. Between Fig. 3(a) and Fig. 3(d), we determine $\Delta\overline{CC} = 0.011 \text{ \AA}^{-1}$, corresponding to $Q = 0.021 e^-$ per Ta atom for the 0%I sample. Using the algebra summarized in Table S1, the bulk T -layer filling is $1 - 2 \times 13Q = 0.46 e^-/\text{SOD}$ for 0%I samples, rising to $0.70 e^-/\text{SOD}$ for 1%M samples. In both cases the flat band retains the majority of its charge, confirming that the T -layer is not depopulated. Note that for the 0%I sample, we are assuming that there is no defect-induced charge transfer as discussed for the 0%M sample in Section S4. This is established by the lack of observable S vacancies in Fig. 4 (a,d).

By focusing on a convenient metric—the distance between protected band crossings C in Fig. S3(b), and by analysis of our TB model’s density of states vs. energy and band slope at the crossing points, we determine the empirical relationship:

$$Q [e^-/1 \times 1] = 1.92 \times \Delta\overline{CC} [\text{\AA}^{-1}], \quad (3)$$

where $\Delta\overline{CC}$ is the change in distance between opposite band crossings C. The coefficient 1.92 is derived from the TB band slope $dE/dk = 1.08 \text{ eV \AA}$ at the crossing and the TB density of states $dN/dE = 3.55 \text{ e}^-/\text{eV}$ per Ta atom at E_F , giving $\Delta k \times (dE/dk) \times (dN/dE) = (\Delta\overline{CC}/2) \times 1.08 \times 3.55 = 1.92 \Delta\overline{CC}$. To assess what fraction of the flat band in the $\sqrt{13}$ -reconstructed surface T -layer is transferred, we multiply Q by 13 to convert to e^-/SOD ; complete depopulation of the flat band would correspond to $13Q = 1 \text{ e}^-/\text{SOD}$. These values of $\overline{CC}$ and Q are summarized for 0%I, 1%M, and 0%M samples in Table S1. These confirm the presence of charge transfer from T to H layers. However, the values of Q are substantially smaller than those reported earlier [23; 22], with values in the range $0.15 < 13Q < 0.4 \ll 1$, demonstrating that the flat band is far from completely depopulated by charge transfer alone, consistent with the residual d_{z^2} spectral weight observed in the 0%I samples (Sec. S5).

The smallest charge transfer and highest T flat band occupation were observed for the 1%M sample. This result suggests a small charge-doping effect of Se substitution, which would act to reduce the interlayer charge transfer Q . A similar trend with Se doping was previously observed for much higher Se concentrations [49].

Supplementary Table S1: Extracted charge transfer parameters and layer fillings for the three sample classes. Q is the charge transferred per 1×1 unit cell across a single $1T/1H$ interface; $q_{d,H}$ is the additional H -layer charge from native S vacancies. The $1T$ filling rows are per SOD cluster (13×13 unit cell), where the half-filled flat band holds $1 \text{ e}^-/\text{SOD}$. See Sec. S3 and Sec. S4 for full details.

Parameter	Symbol	Units	0%I (Pristine)	1%M (Se-doped)	0%M (Defective)
$\overline{CC}$ distance ($1T$ -surface)		$\text{\AA}^{-1}$	1.120	1.115	1.031
$\overline{CC}$ distance ($1H$ -surface)		$\text{\AA}^{-1}$	1.131	1.121	1.046
Interface transfer	Q	$e^-/1 \times 1$	0.021	0.012	0.030
Bulk $1H$ charge transfer	$q_{HB} (= 2Q)$	$e^-/1 \times 1$	0.042	0.023	0.060
H-layer defect charge	$q_{d,H}$	$e^-/1 \times 1$	0	0	0.20

S4 Metallic 0% Se samples and the role of native disorder

Fig. S4 shows a third, anomalous class of samples that have no Se, and yet display the metallic windmill states in the T -surface. Such 0%M samples are a minority of the 0% samples examined. We have found by cleaving samples multiple times that the metallic or insulating character of the T layer is consistent throughout a given crystal, therefore the metal/insulator distinction is probably due to small variations of growth conditions affecting each entire crystal and not due to inhomogeneities within a given crystal.

The possibility of seeing windmill states without Se doping suggests that this transition can occur also for other sources of disorder. It also reconciles the variety of different spectra in the literature claimed for 0% Se doping which have seen either surfaces similar to our 0%*I* samples [22] or surfaces similar to our 0%*M* samples [23]. What sets our results apart is that we have baseline insulating 0%*I* samples for quantitative comparison. The Fermi surface dimensions in both the surface and next-deep *H*-layers show a significant accumulation of electrons in the Ta *d*-bands of the 0%*M* samples, evidenced by both qualitative assessment of topology (wider dog's bones) and quantitative analysis (reduced $\overline{CC}$ distance) compared to the standard 0%*I* samples. Since XPS uncovered no dopants or contaminants (see Sec. S2), the natural explanation for this charge accumulation is S vacancies. Based on the cross-sample $\overline{CC}$ comparison and Eq. 3, we estimate that *H*-layers in 0%*M* samples carry an excess of around 0.20 electrons per Ta atom (1×1 unit cell) relative to the 0%*I* samples, corresponding to an S vacancy density of around 5% (each S vacancy donates 2 electrons, with 2 S sites per Ta).

Sulfur vacancies can in principle add additional defect charge terms $q_{d,T}$ or $q_{d,H}$ to the *T*- or *H*-layers. However, it has been argued that this excess charge resides in the *H*-layer rather than the *T*-layer. First-principles calculations have shown that S vacancies in the *T*-layer ($V_{S,1T}$) distort the local CDW structure and suppress the flat band locally without doping the surrounding *T*-layer [50], so $V_{S,1T}$ defects contribute negligibly to the bulk electron count; accordingly we set $q_{d,T} = 0$ in our model. The present data provide the first experimental evidence that S vacancies in the *H*-layer ($V_{S,1H}$) directly accumulate charge in the *H*-layer: no other plausible source of excess electrons such as impurity elements is apparent in our XPS survey spectra, and the reduced size *H*-layer central Fermi surface of the 0%*M* samples cannot be explained by the small intrinsic charge transfer Q alone. This is consistent with the theoretical expectation that $V_{S,1H}$ defects locally suppress the $T \rightarrow H$ charge transfer, returning electrons to the system and raising the net *H*-layer filling [50]. The intrinsic $T \rightarrow H$ charge transfer Q is found to be $\sim 40\%$ larger for 0%*M* compared to the 0%*I* samples (Table S1). This shows that while local charge transfer is suppressed near defects [50], it is not globally suppressed by defects.

S5 Polarization-dependent ARPES of occupied d_{z^2} states in insulating 1*T* layers

The bound states associated with the insulating 1*T*-layer state in the 0%*I* samples are visible as broad spectral features and a high diffuse background within ~ 100 meV below E_F , seen in Fig. 3(a) for 72 eV, as well as in Fig. S5(a) for 102 eV light. These states resemble those seen in [22], but here we report that these states have little or no spectral weight at E_F . The momentum distribution of these states is made clear with a larger energy integration window, see Fig. S5(b) showing the rotation of the features consistent with the chiral SOD reconstruction.

In order to investigate the orbital character of these *T*-layer states, we switched the x-ray polarization from *p*-polarized (light polarized in the scattering plane) to *s*-polarized (light polarized perpendicular to the scattering plane, and parallel to the k_y direction). According to the basic

theory of ARPES [51], orbitals that are even with respect to the xz scattering plane will be visible in p -polarization but suppressed in s -polarization, namely d_{z^2} , d_{xz} , and $d_{x^2-y^2}$. The opposite polarization dependence holds for d_{xy} and d_{yz} orbitals that are odd with respect to the scattering plane. The low-energy states of the T -layer are well-known to be d_{z^2} -derived, centered on Γ and localized to the central Ta atom of the SOD clusters, while the low-energy states of the H -layer are located away from Γ and are derived from a hybrid combination of even and odd d_{xy} , $d_{x^2-y^2}$, and d_{z^2} orbitals, the latter contributing weakly away from Γ . Thus we expect the T -layer states, if any, to be present in p but completely absent in s polarization, while the H -layer states to be partially present in both p and s , depending on the details of the hybridization.

Fig. S5 compares the ARPES Fermi Surface and low-lying band structure for (a,b) p -polarization and (c) s -polarization. These states show that all traces of the diffuse states near Γ are extinguished in s -polarization, consistent with the expectation that these are d_{z^2} -derived states associated with the SOD Ta atoms. The remaining states in (b) are retained in (c), but their intensities are suppressed according to the relative contribution of even $d_{z^2}/d_{x^2-y^2}$ orbitals (that are suppressed in (c)) and d_{xy} (that are not suppressed in (c)). These results strongly support the assignment of the diffuse states to d_{z^2} orbitals of the central Ta in the SOD clusters.

S6 STM evidence for superconductivity in fragmented CDW regions

Additional STM measurements are shown in Supplementary Fig. S6. Each row in Supplementary Fig. S6 corresponds to a different local region of the pristine $4Hb$ -TaS₂ sample.

S7 Thermal Conductivity

To understand how lattice disorder connects to the apparent electronic emergence, we also study the thermal conductivity and thermal Hall conductivity in the $1T$, $2H$ and $4Hb$ -Ta(S_{1-x}Se_x)₂ systems. Such measurements can be used to analyze the Wiedemann-Franz law, which states that at sufficiently low temperatures all metals should satisfy $\kappa/\sigma T = L_0$, where $L_0 = 2.44 \times 10^{-8} \text{V}^2/\text{K}^2$ is the Lorenz number. The Wiedemann-Franz law demonstrates that for normal metals the majority carriers of charge and entropy should be electrons at temperatures where phonons are frozen out. Deviations from the Wiedemann-Franz law can therefore indicate the presence of neutral excitations, such as phonons that do not carry charge, yielding Lorenz ratios substantially larger than L_0 , as well as unconventional electronic transport arising from strong scattering or incoherent quasiparticle dynamics.

Figure S7 (a) shows the evolution of the thermal conductivity for $1T$ -, $2H$ - and $4Hb$ -Ta(S_{1-x}Se_x)₂ series, where phonons are the dominant heat carriers [52]. The features are consistent with the temperature dependence of a conventional phonon-dominated κ_{xx} , in which increasing thermal excitation of phonons initially enhances the thermal conductivity until higher-momentum

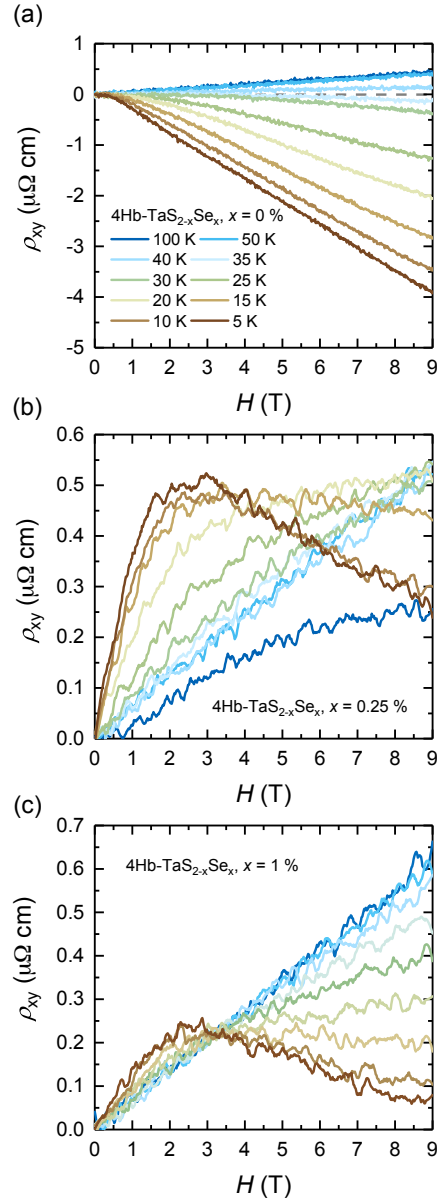
states are populated and Umklapp scattering sets in, suppressing κ_{xx} and producing the characteristic phonon peak [53]. In pristine $4Hb$ -TaS₂, the charge density wave transition at ~ 30 K will also create additional scattering of phonons from electrons.

The thermal conductivity of the pristine $4Hb$ -TaS₂ is second only to the $1T$ - structure, indicating the pristine $4Hb$ -TaS₂ has a slightly higher lattice disorder than $1T$ -TaS₂. As the Se concentration grows, the thermal conductivity is suppressed in harmony with the increasing electrical resistivity due to an increased scattering of both phonons and electrons by impurities. The calculated Lorenz ratio $L_{xx} = \kappa_{xx}/(T\sigma_{xx})$, shown in Fig. S7 (b), exceeds the Sommerfeld value L_0 . Deviations of L_{xx} from L_0 reflect the relative efficiency of heat transport compared to charge transport. Although both phononic and electronic contributions to κ_{xx} are reduced upon Se substitution, the deviation of L_{xx} from L_0 becomes more pronounced in the $x = 1\%$ sample, indicating that disorder suppresses electrical conductivity more strongly than total thermal conductivity. These results are consistent with phonon-dominated heat transport, where disorder disproportionately degrades charge transport relative to heat transport.

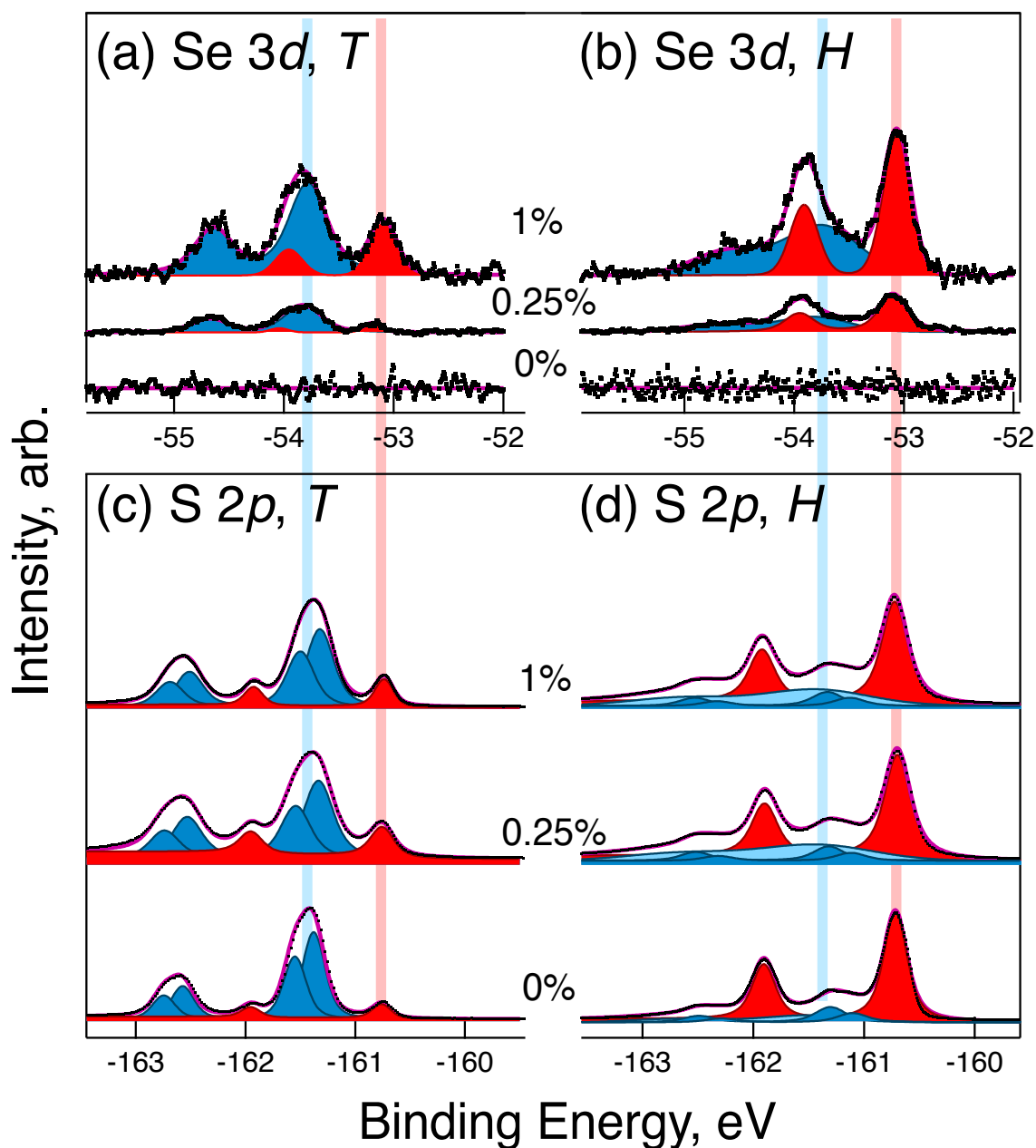
The most striking effect is found in studying the Wiedemann-Franz law in the thermal Hall effect. As shown in Figure S7(c), the pristine $4Hb$ -TaS₂ system has a substantial thermal Hall signal, which can be attributed to the existence of high-mobility electrons. (Note that the thermal Hall signal in $1T$ - is almost absent, consistent with its insulating behavior.) Upon adding Se, the thermal Hall effect is strongly suppressed and vanishes nearly immediately at $x = 1\%$, indicating a severe reduction of the contribution of mobile electrons in the heat transport channel. Notably, the $0\%I$ samples look very consistent with the Wiedemann-Franz law at low temperatures, as do $2H$ -TaS₂ samples, consistent with the conclusion that most of the conduction arises from H -layer (as shown in Fig. S7 (d)). However, in $1\%M$ and $0.25\%M$ samples the Wiedemann-Franz law is strongly violated below the 3×3 CDW transition. This suggests additional multi-band effects come into play as the T -layer and H -layer become coupled. The violation of Wiedemann-Franz is not explicit evidence of Mott delocalization but of changes to the coupling of electrons and phonons.

We believe this to be more than the effect of just disorder. The comparison to the $2H$ -system is quite instructive here. $2H$ -TaS₂ has a thermal conductivity and thermal Hall conductivity about 3 to 4 times smaller than the pristine $4Hb$ -TaS₂, suggesting that the main difference is due to greater scattering in the $2H$ - structures. However, when Se is added, the $4Hb$ -Ta(S_{1-x}Se_x)₂ has a thermal Hall effect that is much more suppressed than the thermal conductivity, suggesting electrons are scattered much more strongly than would be expected from simple disorder effects. This implication is even clearer when studying the Lorenz ratio L_{xy} from the thermal Hall effect (as shown in Fig. S7(d)). In the $x = 0\%$ system, L_{xy} approaches L_0 at low temperatures, just as it does in $2H$ -TaS₂. However, upon adding Se, the Lorenz ratio strongly diverges from L_0 , reaching nearly $L_{xy}/L_0 \approx 45$ at $x = 0.25\%$ around T_c . The deviation from the Wiedemann-Franz law suggests that the new electrons entering the Fermi surface form a highly incoherent and strongly scattered metallic state. Such scattering may arise from low-energy inelastic processes, including

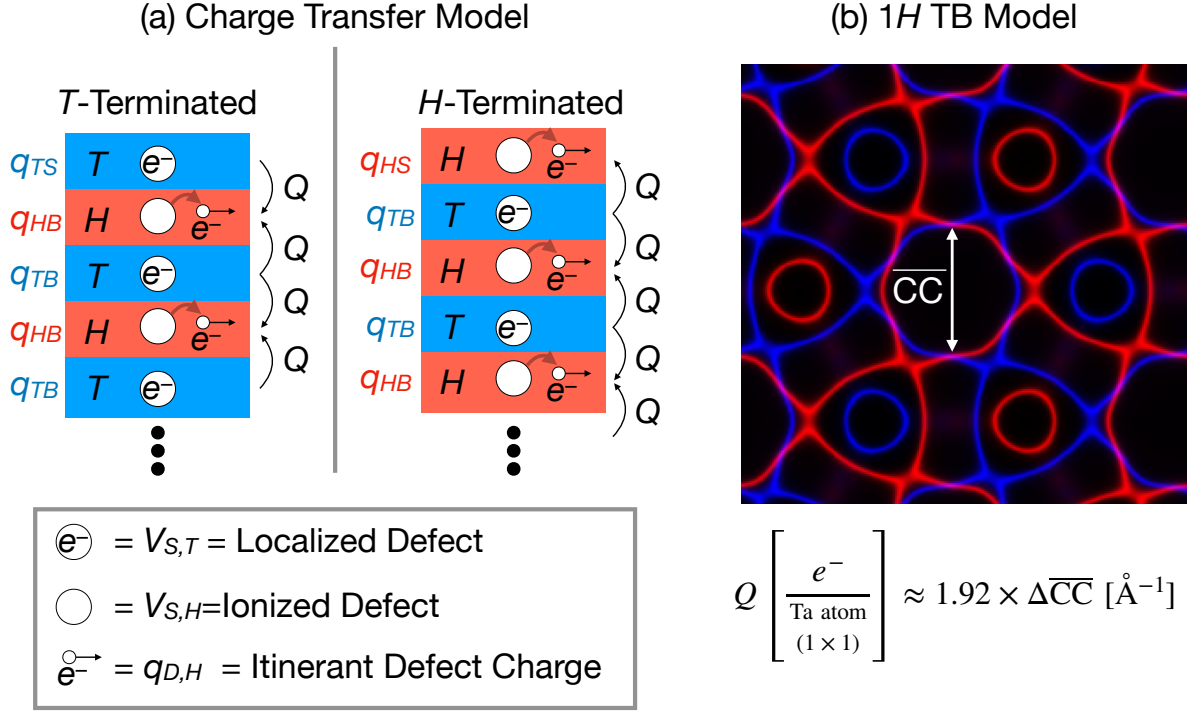
superconducting fluctuations near T_c , as well as multiband effects.



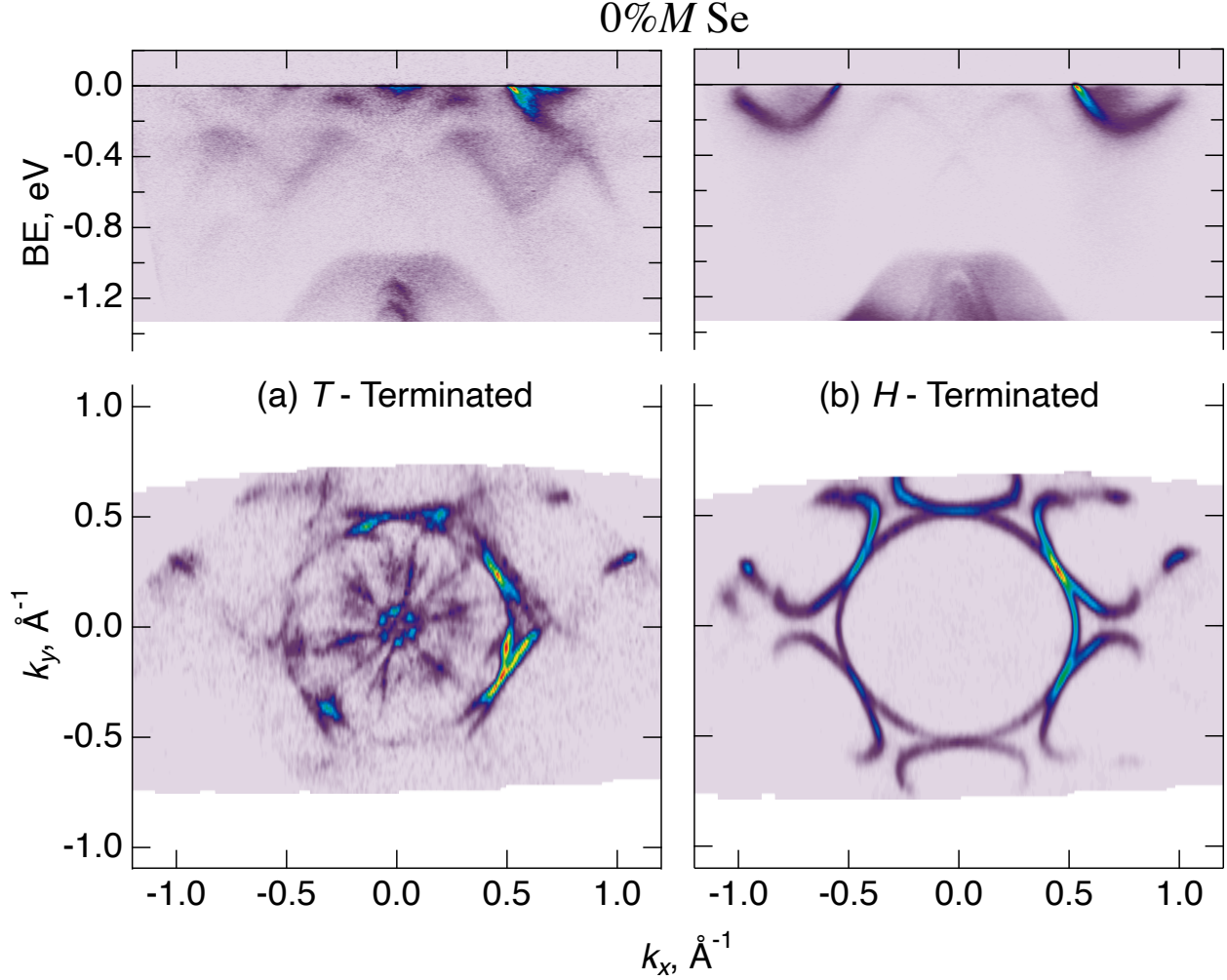
Supplementary Figure S1: Field dependence of in-plane electrical Hall resistivity $\rho_{xy}(H)$ for $4\text{Hb-Ta}(\text{S}_{1-x}\text{Se}_x)_2$ (a) $x = 0\%$, (b) $x = 0.25\%$, and (c) $x = 1\%$ samples measured at several selected temperatures up to $H = 9$ T. The $x = 0\%$ sample shows nearly a linear-in-field dependence in $\rho_{xy}(H)$, while the $x = 0.25\%$ and $x = 1\%$ samples show a non-linear field dependence at low temperature and gradually evolve toward a linear-in-field behavior as temperature increases. $\rho_{xy}(H)$ clearly shows a sign change from negative to positive around 35 K in the $x = 0\%$ sample, which is tied to the low-temperature CCDW transition, while it remains positive within the measured temperature range for the $x = 0.25\%$ and $x = 1\%$ samples.



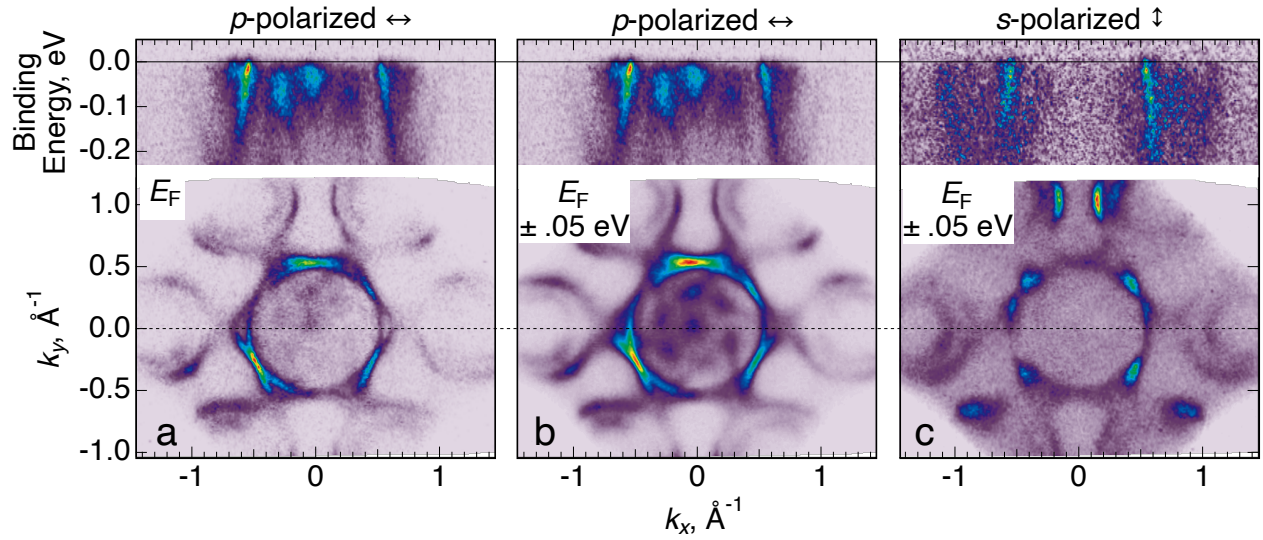
Supplementary Figure S2: (a,b) Se 3d and (c,d) S 2p x-ray photoelectron spectra for *T*- and *H*-terminated surfaces respectively, and for Se concentrations of 0%, 0.25%, and 1.0%. The spectra are comprised of spin-orbit split doublets (red or blue) each of which represents a distinct atomic site near the surface. The 0% sample was of the insulating type designated 0%*I*. The Se spectra are presented after subtraction of a smooth background for clarity. The binding energy ranges (a-d) are plotted on the same scales, so that the leading binding energies for H and T core level peaks may be directly compared for Se 3d and S 2p electrons.



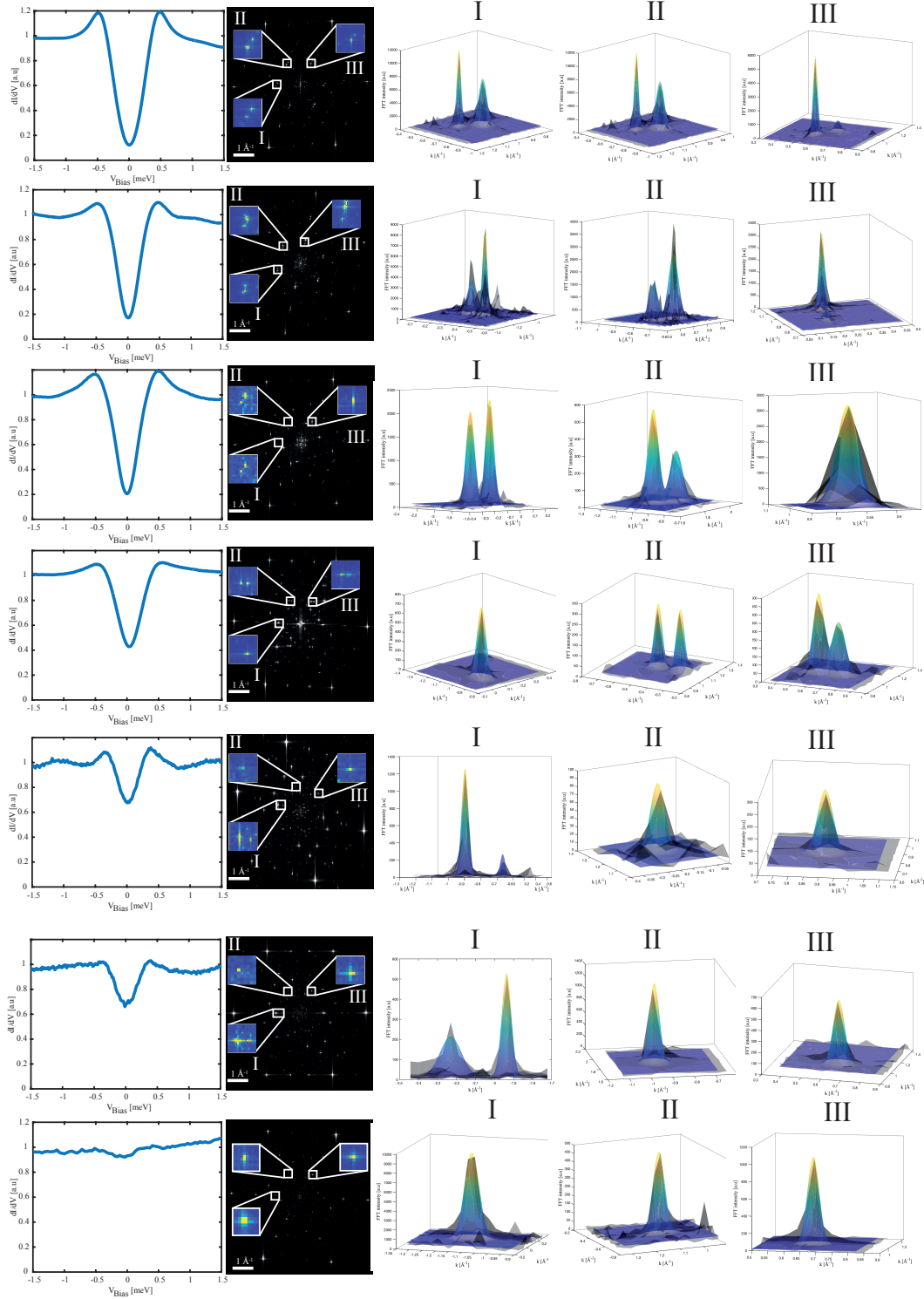
Supplementary Figure S3: Charge transfer model and $\overline{CC}$ metric. (a) Schematic of the layer-resolved charge transfer in $4Hb\text{-TaS}_2$ for T - and H -terminated surfaces. Each T -layer donates charge Q per interface to each adjacent H -layer; surface layers receive Q while bulk layers receive $2Q$ due to two adjacent interfaces. The layer fillings q_{TS} , q_{TB} , q_{HS} , and q_{HB} are defined relative to the isolated-layer reference and are summarized in Table S1. When present, negative or ionized S vacancies are indicated by large white circles. (b) Calculated spin-up (red) and spin-down (blue) Fermi contours of the $1H$ TB model, with the $\overline{CC}$ metric indicated. An empirical rule for estimating the charge transfer Q is included.



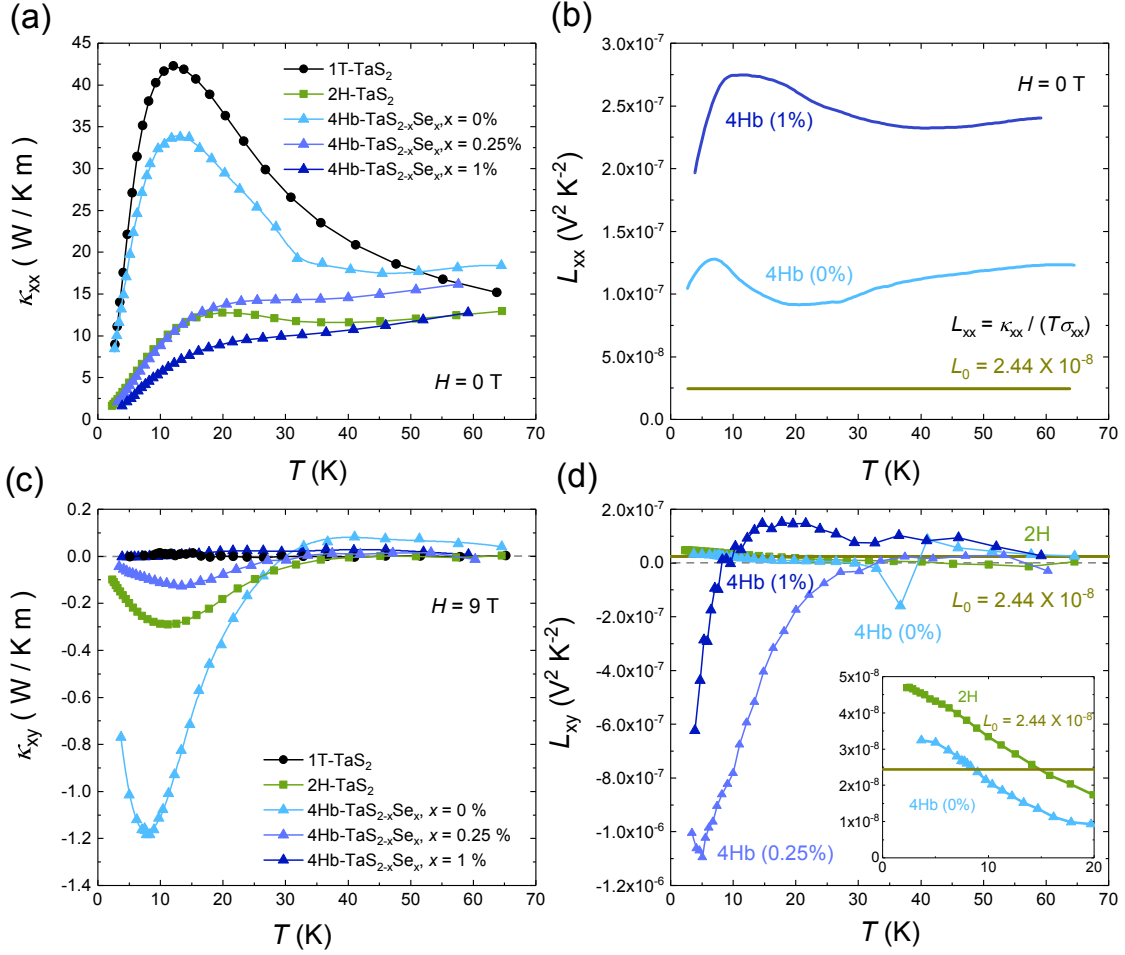
Supplementary Figure S4: **ARPES evidence for a delocalization transition in 4Hb-Ta(S_{1-x}Se_x)₂ with no Se.** (a,b) Data are the same as those shown in Fig. 3 of the main text for T - and H -terminated surfaces, but on a sample with 0% Se substitution containing intrinsic disorder, presumably due to S vacancies. While such samples were less commonly found, it is likely that quench cooling or cleaving conditions can create enough disorder on the surface as to delocalize the flat band on the T -layer.



Supplementary Figure S5: **ARPES evidence for occupied d_{z^2} orbitals in insulating T surface layers of 0% Se.** (a) Bandstructure along $k_y=0$ (upper) and Fermi surface (lower) of 0% I sample, collected at 102 eV with p -polarized light. (b) same, with integration window ± 50 meV to highlight the concentration of diffuse states just below E_F . (c) same as (b) but collected with s -polarized light. The states near Γ are suppressed due to d_{z^2} orbital character, while the remaining states from the underlying H -layer are preserved due to their partial d_{xy} character.



Supplementary Figure S6: Additional STM measurements showing the correlation between CDW fragmentation and superconducting gap formation at different locations of the 1H layer in pristine 4Hb-TaS₂ samples. Each row shows (left) the local tunneling spectrum dI/dV , (middle) the corresponding Fourier transform of the CDW order, and (right) enlarged views of the CDW Bragg peaks labeled I, II, and III. Regions with stronger CDW peak splitting, which are indicative of more fragmented SOD order and enhanced domain-wall density, exhibit larger superconducting gaps. In contrast, regions with sharper and unsplit CDW peaks show strongly reduced or absent superconducting gaps.



Supplementary Figure S7: Thermal conductivity, thermal Hall conductivity, and quenched disorder in 4Hb-Ta(S_{1-x}Se_x)₂. (a) Comparison of thermal conductivity κ_{xx} of 1T-, 2H- and 4Hb-TaS₂ structures, illustrating a dramatic drop in the phononic thermal conductivity peak and thus an increase in phonon scattering when 1% Se is added in 4Hb-TaS₂. (b) Calculated Lorenz number $L_{xx} = \kappa_{xx}/(T\sigma_{xx})$ in 4Hb-Ta(S_{1-x}Se_x)₂ $x = 0\%$ and 1% samples. Lorenz number expected of a conventional metal L_0 is shown as the horizontal dark yellow line. The deviation from the Wiedemann-Franz law becomes more pronounced in the $x = 1\%$ sample, suggesting the electrons are much more strongly scattered due to enhanced interactions. (c) Thermal Hall conductivity κ_{xy} of 1T-TaS₂, 2H-TaS₂, 4Hb-Ta(S_{1-x}Se_x)₂ $x = 0\%$, $x = 0.25\%$, and $x = 1\%$ samples as a function of temperature in a magnetic field of $H = 9$ T. In 4Hb-Ta(S_{1-x}Se_x)₂, κ_{xy} is mainly contributed by electrons. The amplitude of κ_{xy} is strongly suppressed as Se concentration increases, which further illustrates the enormous effect of electron scattering on adding Se. κ_{xy} is almost completely suppressed at $x = 1\%$, similar to the 1T-TaS₂ case where the electrons are completely localized. (d) Calculated Lorenz number $L_{xy} = \kappa_{xy}/(T\sigma_{xy})$ in 2H-TaS₂, 4Hb-Ta(S_{1-x}Se_x)₂ $x = 0\%$, $x = 0.25\%$, and 1% samples in a magnetic field of $H = 9$ T. The WFL is mostly fulfilled in 2H-TaS₂ and pristine 4Hb-Ta(S_{1-x}Se_x)₂ (as shown in the main panel (d) and inset zoom-in at low temperature), while it is strongly violated in the Se-substituted 4Hb-Ta(S_{1-x}Se_x)₂ samples.