nature portfolio

Peer Review File

Electron Correlation and Incipient Flat Bands in the Kagome Superconductor $CsCr_3Sb_5$

Corresponding Author: Professor Lexian Yang

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
This manuscript by Yidian Li et al. presents an ARPES study of the newly discovered Kagome superconductor $CsCr_3Sb_5$, revealing strong electron correlations and incipient flat bands near the Fermi level. While the study offers intriguing results, several points need improvement before publication:
1） While the study observes a transition at 55 K and attributes it to a density wave, there is limited discussion on the specific nature of this density wave. Is it a charge density wave, a spin density wave, or something else? Further analysis and discussion are needed to clarify this aspect.
2） The study primarily focuses on the low-temperature electronic structure. Investigating the temperature dependence of the flat band, particularly across the density wave transition, could provide crucial insights into its role in the electronic behavior of $CsCr_3Sb_5$.
3） Visualizing the flat band in the ARPES data (Fig. 3 in the main text and Figs. 4 and 7 in the supplementary) is challenging. Consider using the second-derivative to enhance the visibility of the flat band and illustrate its distribution in momentum space. This would allow readers to better assess the flat band features.
4） The statement "With increasing binding energy, the pocket around Γ shrinks and line-like features can be resolved, forming a kagome-shape structure in the momentum space" requires clarification." Specifically, identify which bands refer to these "line-like features".
5） The manuscript states, "These features are in overall consistent with the calculation in Fig. 1f, as confirmed by the comparison between the integrated energy distribution curve (EDC) and the calculated density-of-states (DOS) in Fig. 2h." Explain more clearly how comparing the density of states validates the calculated band structure. While the DOS provides information about the overall distribution of electronic states, it doesn't directly confirm the specific band dispersions.
6） In Figure 3, the red arrows indicating the flat band in the experimental data point to different energy positions, while in the theoretical calculation, they appear at the same energy. Provide a brief explanation for this discrepancy.
7） Typo's: Line 89: Figs.1d → Fig.1d.

Reviewer #2

(Remarks to the Author)
The paper reports ARPES results on CsCr3Sb5. CsCr3Sb5 is a compound of active current interest and the reported results are one of the first ones to be reported. The measurements are likely technically challenging due to small single crystal sizes. The results are compared to theoretical calculations. Both theory and experiment seem technically correct to me. Concerning theory besides information about the photoemission one might desire the information about the orbital occupancies and renormalizations from the self energy for the paper to be self contained.

The main problem of the paper is that from the data one cannot infer that much information. Namely, what is most clearly resolved is the less correlated Sb band (for this DMFT seems to agree; the authors could explain why there is renormalization ; is this due to the Cr weight of the band or is there a shift of the onsight energy?) Less clearly, but present in the data is the incoherent weight close to the Fermi level, where the flat bands are expected. In contrast to calculations the flat band occurs somewhat below the Fermi level (the paper should in a more clear way discuss whether in this aspect theory and DMFT calculations differ). The precision of the paper is sufficient only to document this incoherent weight but is insufficient to characterize the electrons there in any way.

One does not know how renormalized the bands are nor can one get the information about the lifetime. (Concerning this point, the paper does write: "Interestingly, the flat bands above EF in Fig. 3a are pushed closer to EF and strongly renormalized, leaving the incipient flat band with prominent spectral...", I wonder how this is consistent with their observation of a flat band below 80 meV,
and how can they infer strong renormalization if the dispersion cannot be clearly seen at the precision of the experiments (that are, admittedly, challenging). Is according to experiments the flat band pushed below Fermi level or not? In DFT calculations there are several bands above the Fermi level. If one broadens these spectral functions with the impurity scattering /experimental resolution and multiplies with the Fermi functions is the DFT consistent with the experiment or there is an actual shift of the flat bands below the Fermi level?

The only more detailed information about the spectra is inferred from the widths of the laser ARPES MDCs that document a growth above 55K (see . The paper tells that this "can explain semiconducting-like behavior". In reality, what one sees from the figure is that there is little evolution of the MDC below 45K (likely because the width is instrumental resolution/impurity scattering limited there?) and above 65K, and in between there is a jump. This jump might be more related to the change of the dispersion upon CDW transition rather than a change of the scattering rate. The paper could also be more clear about which particular band this change is about. Also, precisely at the Fermi level, the analysis of the MDC widths is complicated by the Fermi functon(see for instance
Phys. Rev. Lett. 131, 236502 for a recent example). . How does the width behave at higher binding energies?

In summary, the paper reports one of the first ARPES experiments on the compound. It does show signal of the flat band close to the Fermi level. The information one can infer from the signal directly is however somewhat limited and one should more critically compare the theory with experiment (including the experimental resolution artefacts) to be able to judge what those experiments really tell.

I do not recommend the publication in the present form.

Reviewer #3

(Remarks to the Author)
Superconductivity in highly correlated kagome systems has been a topic of theoretical interest for years, with experimental realization achieved in CsV_3Sb_5, which exhibits superconductivity, charge-density-wave (CDW) orders, and nonmagnetic behavior with weak electron correlations. In contrast, $CsCr_3Sb_5$, where V is replaced with Cr, features strong electron correlations, frustrated magnetism, and characteristic flat bands near the Fermi level. A comparative study of the electronic structures of these two materials offers a deeper understanding of correlated kagome systems.

The authors conducted transport measurements, ARPES (angle-resolved photoemission spectroscopy), and DFT+DMFT (density functional theory + dynamical mean field theory) calculations to investigate the electronic structure of $CsCr_3Sb_5$. Their findings indicate that Cr 3d electrons exhibit incoherence due to strong electron correlations driven by Hund's coupling. The study also reveals the presence of an "incipient flat band" near the Fermi level (E_x) at the Brillouin zone (BZ) center. Additionally, a scattering rate increase at 55 K was observed, potentially linked to CDW transition behavior. The authors propose that $CsCr_3Sb_5$ is a Hund's metal with an incipient flat band near E_x, which could underlie the novel properties of this material.

$CsCr_3Sb_5$ offers a promising platform for studying correlation effects in kagome metals, particularly exotic phenomena such as CDW and superconductivity. The ARPES measurements conducted by the authors covered the entire 3D BZ, mapping the electronic structure in detail, albeit with data quality that is high but not exceptional. Before the manuscript can be considered for publication in Nature Communications, several questions and concerns need to be addressed:

Comparison with CsV_3Sb_5: $CsCr_3Sb_5$ introduces three additional electrons per kagome cell, effectively shifting the bands downward under a rigid band approximation. In both experimental and calculated data, what are the major differences in band structure and Fermi surface (FS) topology between the two materials? The calculated FS topology (Fig. 1f, g) and experimental data (Fig. 2a, Fig. 3) suggest a three-dimensional character for $CsCr_3Sb_5$, unlike the primarily two-dimensional nature of CsV_3Sb_5. Additionally, the DFT+DMFT calculations (Fig. 2e) highlight the correlation effects. How do the renormalization factors differ between these materials, and what is the primary driving force behind these differences (e.g., Hund's coupling)?

Orbital-Selective Band Shift: The experimental data show a downshift of the flat band below EF, whereas other bands at similar energies (Fig. 3a) remain largely unaffected. Only the band near the BZ center experiences this pronounced effect. Could the authors discuss the cause of this orbital-selective band shift?

CDW Features: While the material undergoes a CDW transition, as noted in prior reports, the authors did not observe band folding, shadow bands, or gap openings. What could account for this discrepancy? Could it result from mixing of bulk and surface states? Alternatively, could surface reconstruction or surface disorder below 55 K (as observed through techniques like LEED or STM) explain the absence of these features?

Comparison with Other ARPES Data: Another ARPES study (arXiv:2406.05293v2) presents visually higher-quality data. What are the major differences between the findings of these two reports?
Minor Points: In Fig. 2(e-g), the directions should be explicitly labeled, as currently only k//k_{//}k// is indicated. To improve clarity, overlaying DFT calculations with experimental data would better illustrate their consistency.

In summary. The authors performed detailed measurements and theoretical analyses of $CsCr_3Sb_5$. However, until the issues outlined above are resolved, the manuscript is not yet suitable for publication in Nature Communications in its current form.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
This manuscript has been significantly improved by the authors' careful response to the reviewers' feedback. They have effectively addressed all my concerns and included new data and analyses. I am now satisfied with the quality of this paper and confidently recommend it for publication in Nature Communications.

Reviewer #4

(Remarks to the Author)
The authors of the manuscript report on temperature-dependent ARPES measurements on the kagome CsCr3Sb5 and discuss the electronic properties of the system in the context of Hund's metal with an incipient flat band near the Fermi surface supported by DFT and DFT+DMFT calculations. This is an interesting work, which, in view of the present discussion on this kagome material, brings some valuable information to the community studying these materials.
There are, in my opinion, still a few issues that the authors should further clarify before recommending the paper for publication.

1) As reviewer 1 points out, it is unclear what information the authors extract from the transition at T=55K with their measurements. The authors refer to the literature in term of an ' intertwined spin and charge density wave'. What does this exactly mean? And what signatures of it are the authors seeing in their experiments? Here the authors should state what they can interpret from their observations. Along the manuscript they only mention a CDW but not a spin density wave, do they find signatures for such a spin density wave? This would be a very useful piece of information.

2) the ARPES data look very broad (not only near Ef) and interpretation of them seems to strongly rely on the calculations. While the information of incoherent versus coherent spectral weight as a function of temperature plays a role (see next comments), could the authors include further comments on the quality of their samples?

3) For the discussion, the authors strongly rely on DMFT calculations performed by one of the authors in a previous publication, nevertheless, more information on these calculations should be included in the manuscript. Are these charge self-consistent calculations? at which temperatures in the QMC calculations are they performed? the discussion of cohernet vs. ncoherent spectral weight needs some further elaboration. How are the calculations treated in the CDW phase? S hybridizes strongly with Cr, how is this considered in the calculations?

After the authors have clarified these points, in my opinion the manuscript is suitable for publication.

Version 2:

Reviewer comments:

Reviewer #4

(Remarks to the Author)
The authors have satisfactorily replied to all the points raised in my previous report and I recommend publication of the manuscript in Nat. Comm.

Reply to Reviewer #1

Reviewer's Comment:

This manuscript by Yidian Li et al. presents an ARPES study of the newly discovered Kagome superconductor $CsCr_3Sb_5$, revealing strong electron correlations and incipient flat bands near the Fermi level. While the study offers intriguing results, several points need improvement before publication:

Authors' response:

We thank the reviewer for reviewing our manuscript and acknowledging that "the study offers intriguing results". We also appreciate his/her pertinent comments, which greatly helped us improve our manuscript. In the revised manuscript, we have addressed all points raised by the reviewer. Below are our point-by-point responses to the reviewer's comments:

Reviewer's Comment:

1） While the study observes a transition at 55 K and attributes it to a density wave, there is limited discussion on the specific nature of this density wave. Is it a charge density wave, a spin density wave, or something else? Further analysis and discussion are needed to clarify this aspect.

Authors' response:

We thank the reviewer for this insightful discussion. The nature of the density wave has been discussed in Nature 632, 1032 (2024). The density wave state is an intertwined charge-density wave and spin-density wave. The charge order is observed in the X-ray diffraction measurements and the spin-density wave is identified by the magnetic susceptibility and NMR experiments. In our work, the density wave transition is observed in the resistivity and magnetic susceptibility measurements. A sudden increase of the carrier scattering rate is also revealed by our temperature-dependent ARPES experiment across the density wave transition.

In the revised manuscript, we made the following changes of the manuscript to further address the nature of the density-wave:

(1) In line 4 of paragraph 3 in page 4, we added the sentence "Previous X-ray scattering and nuclear magnetic resonance experiments revealed an intertwined CDW and spin-density wave (SDW) below 55 K."

(2) In line 8 of paragraph 1 in page 9, we added the sentence "and the competition between density-

wave and pressurized superconductivity".

Reviewer's Comment:

2) The study primarily focuses on the low-temperature electronic structure. Investigating the temperature dependence of the flat band, particularly across the density wave transition, could provide crucial insights into its role in the electronic behavior of $CsCr_3Sb_5$.

Authors' response:

We thank the reviewer for this helpful suggestion. We have conducted temperature-dependent experiments across the density-wave transition as shown in Fig. R1. We do not observe a clear change of the flat band position at the Γ point, except for a slight suppression of its spectral weight. At $k_{||}$ = 1.0 Å^{-1}, there may be a slight shift of the EDC peak. However, this possible shift is smeared by the thermal effect and weak intensity of the flat band. No correlation between the flat band shift and the density-wave transition is observed [also see arXiv: 2406.05293 (2024)]. Following the reviewer's suggestion, we include the temperature-dependent measurement in the Supplementary Information.

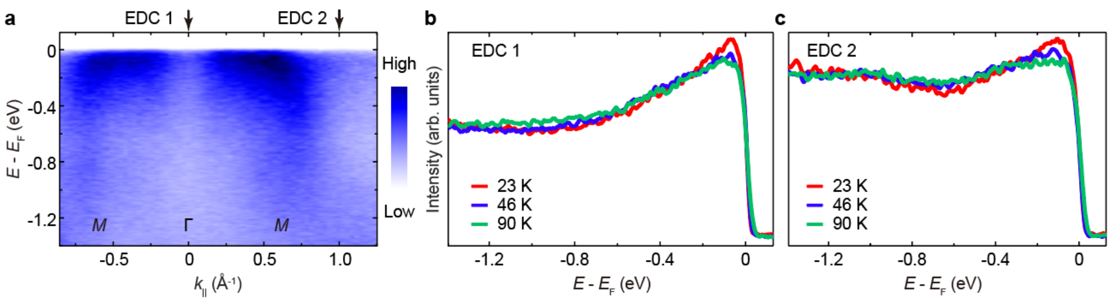

Fig. R1. **a**, Band structure along the Γ*M* direction. **b**, **c**, Temperature-dependent energy-distribution curves (EDCs) at (**b**) the Γ point (EDC 1) and (**c**) $k_{||}$ = 1.0 Å^{-1} (EDC 2). Data were collected using 62 eV linear-vertically (LV) polarized photons.

Reviewer's Comment:

3) Visualizing the flat band in the ARPES data (Fig. 3 in the main text and Figs. 4 and 7 in the supplementary) is challenging. Consider using the second-derivative to enhance the visibility of the flat band and illustrate its distribution in momentum space. This would allow readers to better assess the flat band features.

Authors' response:

We thank the reviewer for this suggestion. However, due to the complex band structure and intrinsically incoherent 3*d* electronic states, the second-derivative with respect to the energy axes does not work very well to visualize the flat band, especially when the flat band is near E_F (the Fermi-Dirac distribution contributes strong artificial features near E_F).

The flat band is better visualized in our experiments with higher photon-energies. As shown in Fig. R2, using photons at 200 eV, a flat band is clearly resolved near 80 meV below E_F in the raw data and EDCs, consistent with the results in the main text. In the revised manuscript, we include Fig. R2 in the Supplementary Information to support our discussion.

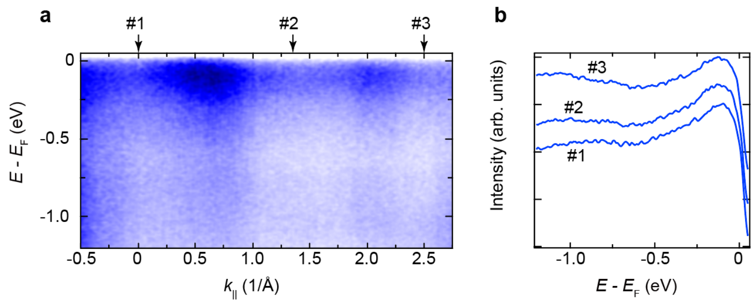

Fig. R2. Observation of the flat band using photons at 200 eV. **a**, ARPES intensity map along $\bar{\Gamma}\bar{M}$. **b**, EDCs at the momentum positions marked in **a**. The flat band can be clearly resolved directly in the ARPES intensity map and EDC peaks.

Reviewer's Comment:

4) The statement "With increasing binding energy, the pocket around $\bar{\Gamma}$ shrinks and line-like features can be resolved, forming a kagome-shape structure in the momentum space" requires clarification." Specifically, identify which bands refer to these "line-like features".

Authors' response:

We thank the reviewer for this helpful suggestion. The "kagome-shape structure in the momentum space" is indicated by dashed lines in Fig. 2d. It is formed by the dispersive bands near $\bar{K}$, as indicated by the black arrow in Fig. R3.

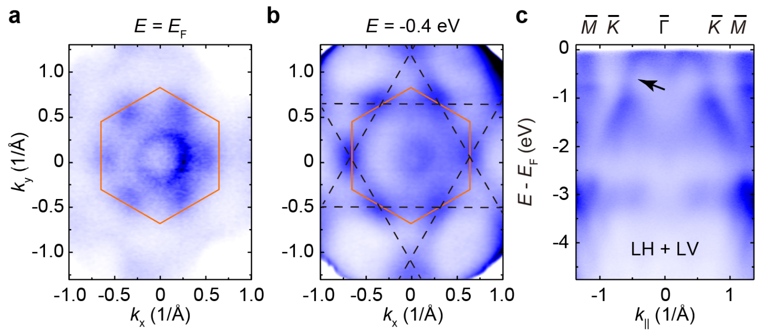

Fig. R3. **a**, **b**, Constant-energy contours at different binding energies. **c**, Band dispersion measured along $\bar{\Gamma}\bar{K}\bar{M}$.

Following the suggestion of the reviewer, in the revised manuscript, we change this sentence to "With increasing binding energy, the circular electron pocket around the $\bar{\Gamma}$ point shrinks and a star-of-David-shape pattern can be resolved at E = -0.4 eV, as indicated by the black dashed lines (Fig. 2d)" in line 9 of paragraph 2 in page 5. We also added a black arrow in Fig. 2d to indicate the band that forms the star-of-David-shape pattern.

Reviewer's Comment:

5） The manuscript states, "These features are in overall consistent with the calculation in Fig. 1f, as confirmed by the comparison between the integrated energy distribution curve (EDC) and the calculated density-of-states (DOS) in Fig. 2h." Explain more clearly how comparing the density of states validates the calculated band structure. While the DOS provides information about the overall distribution of electronic states, it doesn't directly confirm the specific band dispersions.

Authors' response:

We thank the reviewer for this comment. We are sorry for the misleading statement here. "These features" refer to the overall distribution of electronic states in a large energy range, including the dispersive bands near E_F, the M-shape feature around -1.5 eV, and weakly dispersive bands around -3 eV. They contribute peaks in the integrated EDCs, which are captured by the calculated DOS. In the revised manuscript, we change this sentence to "These features contribute peaks in the integrated energy distribution curve (EDC) that is in overall consistency with the calculated density-of-states (DOS), as compared in Fig. 2h" in line 5 of paragraph 3 in page 5.

Reviewer's Comment:

6） In Figure 3, the red arrows indicating the flat band in the experimental data point to different energy positions, while in the theoretical calculation, they appear at the same energy. Provide a brief explanation for this discrepancy.

Authors' response:

We thank the reviewer for his/her careful review of our manuscript. We place the arrows slightly away from the experimental feature to avoid possible obstruction of the data. We are sorry that the arrows were not aligned. In the revised manuscript, we have aligned the arrows in the experimental data.

Reviewer's Comment:

7） Typo's: Line 89: Figs.1d → Fig.1d.

Authors' response:

We thank the reviewer for pointing out this typo. We have corrected it in the revised manuscript.

Again, we greatly thank the reviewer #1 for his/her helpful discussions and suggestions. We hope our reply can relieve his/her concerns and look forward to his/her final recommendation.

Reply to Reviewer #2

Reviewer's Comment:

The paper reports ARPES results on CsCr3Sb5. CsCr3Sb5 is a compound of active current interest and the reported results are one of the first ones to be reported. The measurements are likely technically challenging due to small single crystal sizes. The results are compared to theoretical calculations. Both theory and experiment seem technically correct to me.

Authors' response:

We thank the reviewer for acknowledging the interest of our work. We are pleased that the reviewer concludes "*the reported results are one of the first ones*" and "*both theory and experiment seem technically correct to me*". The comments and suggestions have greatly helped us improve our manuscript. However, after carefully reading the reviewer's report, we think there may be some misunderstandings of our work. Below are our detailed responses to the reviewer's comments:

Reviewer's Comment:

Concerning theory besides information about the photoemission one might desire the information about the orbital occupancies and renormalizations from the self energy for the paper to be self contained.

The main problem of the paper is that from the data one cannot infer that much information.

Authors' response:

We thank the reviewer for this comment. However, with due respect, we do not agree that "*from the data one cannot infer that much information*".

(1) By directly comparing the DFT calculation with our ARPES data in Supplementary Fig. 4, we estimate the overall renormalization factor to be about 1.4.

(2) It is challenging to directly estimate the orbital occupancies from ARPES experiments, especially based on the intrinsically incoherent electronic states. However, theoretical calculations including the Hund's coupling have estimated the orbital occupancies [for details, please see arXiv: 2401.16770 (2024) by Yilin Wang who is one of the authors of the current work], as shown in Table R1. We emphasize that the overall agreement between our ARPES experiment and DMFT calculation supports the theoretical calculation. The slight difference in the experimental and calculated Fermi level makes minor changes to the orbital occupancy.

TABLE I. Occupancy of Cr d-orbitals of $CsCr_3Sb_5$ from DFT and DFT+DMFT calculations, without and with one-hole doping. The DFT+DMFT calculations are performed at U=5.0, J_H =0.88 eV and T = 100 K.

	d_{xz}	d_{yz}	$d_{x^2-y^2}$	d_{z^2}	d_{xy}	total
DFT, no-doping	0.967	0.826	0.767	0.899	1.037	4.496
DFT, one-hole	0.904	0.840	0.728	0.917	1.060	4.449
DMFT, no-doping	0.908	0.837	0.845	0.831	0.868	4.289
DMFT, one-hole	0.890	0.827	0.811	0.828	0.872	4.228

Table R1. Calculated orbital occupancy of Cr *d* orbitals. The table is from arXiv: 2401.16770 (2024).

Based on the above argument, we conclude that our experiments and calculations do provide important information regarding the band renormalization and orbital occupancy. In addition to these results, we would like to further address the main findings of our work:

(1) As "*one of the first ones*", we characterize the basic kagome electronic structure of $CsCr_3Sb_5$ (including the flat band, van Hove singularity, and Dirac dispersion) that is "*a compound of active current interest*". In particular, compared to the extensively studied sibling compound CsV_3Sb_5, $CsCr_3Sb_5$ exhibits strong electron correlation, magnetism, and unconventional superconductivity under pressure. The characterization of its electronic structure lays down the foundations for understanding its novel properties and is of broad interest to the community.

(2) We reveal the important role of Hund's coupling in the electronic properties of $CsCr_3Sb_5$, which identifies $CsCr_3Sb_5$ as an interesting correlated Hund's metal and provides an ideal platform to investigate the interplay between Hundness and kagome physics.

(3) The cooperation between the incipient flat band and strong Hund's coupling is expected to play an important role in the novel properties of $CsCr_3Sb_5$, including the enhanced effective mass, the intrinsically incoherent electronic states, and the unconventional superconductivity. The physics is similar to some iron- and nickel-based superconductors, which will help construct a universal physical picture of unconventional superconductivity in multi-orbital systems with flat bands and strong correlation.

Reviewer's Comment:

What is most clearly resolved is the less correlated Sb band (for this DMFT seems to agree; the authors could explain why there is renormalization; is this due to the Cr weight of the band or is there a shift of the onsight energy?)

Authors' response:

We thank the reviewer for this discussion. In a correlated material such as $CsCr_3Sb_5$, the electron correlation of 3*d* electrons can naturally induce renormalization of the *p* electrons through *p*-*d* electron interaction or hybridization. As shown in Fig. 3a and Supplementary Fig. 6, our orbital-projected DFT calculation suggests that the Sb *p* orbital has a mixed *d* orbital component. Therefore, we conclude that the renormalization of the Sb *p* orbital band is due to the *d* orbital weight instead of a shift of the on-site energy.

In the revised manuscript, we added the sentence "We notice that the electron band around $\bar{\Gamma}$ has mixed *p* and *d* orbital components, suggesting a *p*-*d* interaction (Supplementary Information)." in line 6, paragraph 2, page 6 to clarify this point.

Reviewer's Comment:

Less clearly, but present in the data is the incoherent weight close to the Fermi level, where the flat bands are expected. In contrast to calculations the flat band occurs somewhat below the Fermi level (the paper should in a more clear way discuss whether in this aspect theory and DMFT calculations differ).

Authors' response:

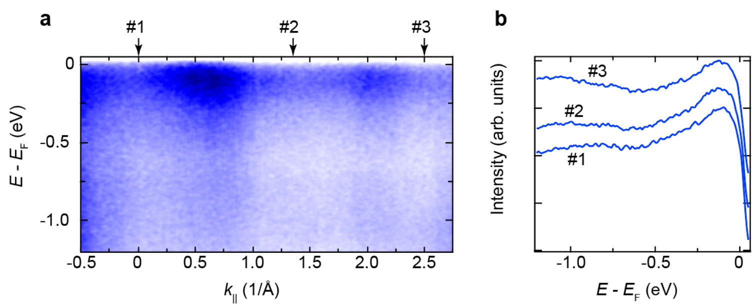

Fig. R4 (same as Fig. R2). **a**, ARPES intensity map along $\bar{\Gamma}\bar{M}$. **b**, EDCs at the momentum positions marked in **a**. The flat band can be resolved directly in the ARPES intensity map and EDC peaks.

We thank the reviewer for this comment. Before responding to this comment, we show in Fig. R4 that the flat band is clearly resolved using photons at 200 eV.

The incoherence of Cr *d* bands (including the flat band) is mainly due to the strong correlation enhanced by Hund's coupling, as evidenced by our DMFT calculation. The difference between ARPES data and DMFT calculations regarding the position of the flat band may be mainly due to the following reasons:

(1) With a flat band just at the Fermi level, The Fermi-Dirac distribution and Gaussian convolution accounting for the energy resolution will broaden and shift the peak position, as shown in Fig. R5.

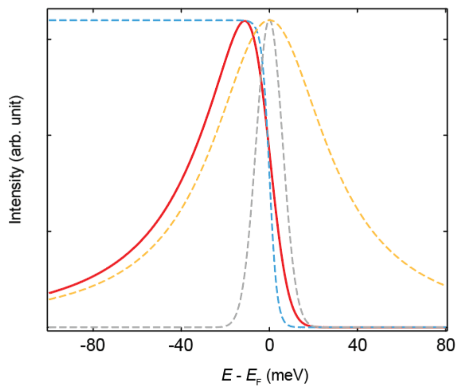

Fig. R5. The simulation of the thermal effect and energy resolution effect in a flat band just at the Fermi level. The flat band is represented by the yellow dashed line. With the thermal (blue dashed line) and resolution (gray dashed line) effects considered, the peak position shifts below E_F.

(2) It is likely that there is a self-doping effect and/or surface effect that raise the Fermi level so that the flat band locate below E_F. Similar effects have been observed in vanadium-based compound CsV_3Sb_5 [see, e.g. Phys. Rev. B 106, L121112 (2022)]. Actually, it is commonly observed that the Fermi level is different between ARPES experiments and theoretical calculations.

(3) The on-site Coulomb interaction and Hund's coupling strongly affect the position of the flat band (Supplementary Fig. 8) [arXiv:2401.16770 (2024)]. While the DMFT calculation with $U = 5$ eV and $J_H = 0.88$ eV shows an overall agreement with our ARPES data, it is likely that the correlation effect is even stronger, which may further modulate the position of the flat band.

We would like to point out that similar results have been presented in other ARPES experiments on $CsCr_3Sb_5$ [arXiv: 2406.05293 (2024) and arXiv: 2406.17769 (2024)]. Following the comments of the reviewer, we add the sentences "It is likely that E_F of $CsCr_3Sb_5$ crystals are slightly raised up by self-doping or surface effects and the flat bands, similar to the situation in CsV_3Sb_5. Therefore, the flat band can be experimentally observed by ARPES experiment" in line 11 of the last paragraph in page 6 to address this point.

Reviewer's Comment:

The precision of the paper is sufficient only to document this incoherent weight but is insufficient to characterize the electrons there in any way. One does not know how renormalized the bands are nor can one get the information about the lifetime. (Concerning this point, the paper does write: "Interestingly, the flat bands above EF in Fig. 3a are pushed closer to EF and strongly renormalized, leaving the incipient flat band with prominent spectral...", I wonder how this is consistent with their observation of a flat band below 80 meV, and how can they infer strong renormalization if the dispersion cannot be clearly seen at the precision of the experiments (that are, admittedly, challenging).

Authors' response:

We thank the reviewer for this discussion. However, with due respect, we do not agree that "*The precision of the paper is sufficient only to document this incoherent weight but is insufficient to characterize the electrons there in any way*".

(1) Regarding the band renormalization and electron correlation:

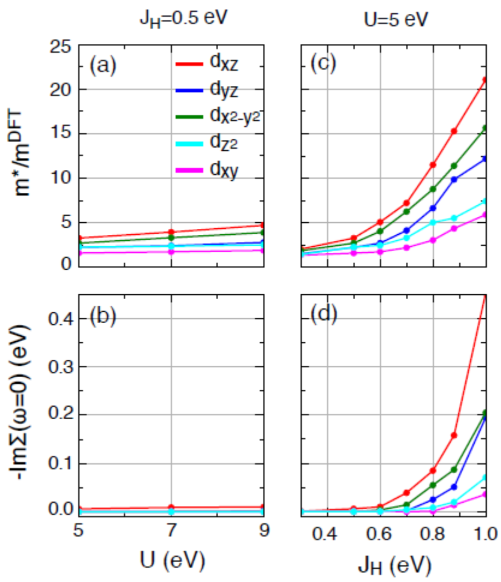

Fig. R6. The effect of on-site Coulomb interaction U and Hund's coupling J_H in the effective mass (band renormalization) and imaginary part of electron self-energy of $CsCr_3Sb_5$. Data are from arXiv: 2401.16770 (2024).

We have shown in the Supplementary Information that the overall renormalization factor of the band structure is about 1.4 (Supplementary Fig. 4). We emphasize that the electron correlation in $CsCr_3Sb_5$ is not merely manifested by the renormalization of the band width. As shown in Fig. R6, the renormalization effect is strongly enhanced by Hund's coupling [arXiv:2401.16770 (2024)]. In particular, the electron self-energy is also strongly enhanced by Hund's coupling, which is the intrinsic reason for the incoherence of Cr *d* bands. Consistently, the observation of the flat band near E_F is also a manifestation of electron correlation since the on-site Coulomb interaction and Hund's coupling strongly modulate the flat bands (Please also see our response about the difference of the flat band position in the experiment and calculation in page 9 and page 12). The comparison between our ARPES experiment and DMFT calculation with different Hund's coupling (Supplementary Information) well supports the strong renormalization of the band structure, consistent with other ARPES study of this compound [arXiv:2406.05293v2].

(2) Regarding "*the information about the lifetime*":

Although the *d*-orbital bands are not clearly resolved due to the strong correlation enhanced by Hund's coupling, our experiment provides qualitative information that the *d* orbital electrons are strongly scattered with short lifetimes. By comparing with the DMFT calculation, our experiment further supports the calculation about the lifetime in a semi-quantitative way [see the calculation of the imaginary part of electron self-energy (related to electron lifetime) in arXiv: 2401.16770 (2024)]. Moreover, we can clearly resolve the Sb *p* orbital band crossing E_F and track the evolution of the scattering rate (or lifetime) of Sb *p* electrons. Therefore, our work does provide information about the carrier lifetime.

Reviewer's Comment:

Is according to experiments the flat band pushed below Fermi level or not? In DFT calculations there are several bands above the Fermi level. If one broadens these spectral functions with the impurity scattering /experimental resolution and multiplies with the Fermi functions is the DFT consistent with the experiment or there is an actual shift of the flat bands below the Fermi level?

Authors' response:

(1). Is according to experiments the flat band pushed below Fermi level or not?

Yes, the flat band is pushed below the Fermi level (Please also see our reply to the reviewer's comment about the position of the flat band).

(2). In DFT calculations there are several bands above the Fermi level. If one broadens these spectral functions with the impurity scattering /experimental resolution and multiplies with the

Fermi functions is the DFT consistent with the experiment or there is an actual shift of the flat bands below the Fermi level?

No, the DFT calculation cannot reproduce the experimental results even after considering the scattering/resolution/thermal effects (Supplementary Fig. 4). Even considering the rising of the Fermi level in experiments, the flat bands are still above the Fermi level in the DFT calculation. Electron correlation is necessary to reproduce the experimentally observed band structure, including the overall band dispersions, *d* electron incoherence, and flat band.

Reviewer's Comment:

The only more detailed information about the spectra is inferred from the widths of the laser ARPES MDCs that document a growth above 55K (see . The paper tells that this "can explain semiconducting-like behavior". In reality, what one sees from the figure is that there is little evolution of the MDC below 45K (likely because the width is instrumental resolution/impurity scattering limited there?) and above 65K, and in between there is a jump. This jump might be more related to the change of the dispersion upon CDW transition rather than a change of the scattering rate. The paper could also be more clear about which particular band this change is about.

Authors' response:

We thank the reviewer for this comment. With due respect, however, we do not agree with the reviewer that " *the widths of the laser ARPES MDCs that document a growth above 55K*" is "*the only more detailed information*" we can infer from the spectra (please also see our responses to the reviewer's other comments).

(1) We agree with the reviewer that the MDC shows little evolution below 45K and above 65K. The width is not instrumental resolution limited since the momentum resolution of our laser-ARPES is about 0.01 $Å^{-1}$, much smaller than the MDC width. On the other hand, while the contribution from the impurity scattering cannot be completely ruled out, it does not affect our conclusion about the sudden increase of scattering rate across the density-wave transition.

(2) Please note that the resistivity value does not change much between 2 K and 120 K. In particular, the resistivity change is only about 0.2 mΩ cm (Fig. 1e) above 60 K and below 40 K. The resistivity tends to remain at a nearly constant level at high temperatures. While we do not expect to fully explain the resistivity curve by the temperature dependence of carrier scattering rate (we are trying

to understand the peculiar semiconducting behavior above the density-wave transition), the temperature evolution of the MDC width qualitatively fits to the temperature evolution of the resistivity, particularly the sudden jump near the transition temperature.

(3) As shown by both synchrotron-based (Figs. 4a, b and Fig. R1) and laser-based (Fig. 4d) ARPES data at low and high temperatures, there is a minor change of the band dispersion across the density-wave-like transition. In particular, we fit the band dispersions measured using laser-ARPES at different temperatures. As shown in Fig. R7, no clear change in the band dispersion is observed.

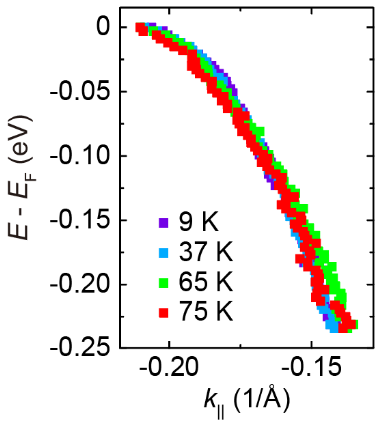

Fig. R7. Band dispersion of Sb *p*-band around $\bar{\Gamma}$ extracted by fitting the MDCs. Data were measured by laser-ARPES at selected temperatures.

(3) More importantly, the band dispersions remain crossing the Fermi level above the transition temperature. In the energy band theory, the system should be metallic at high temperatures. However, the resistivity measurement suggests a semiconducting behavior above the transition temperature (Fig. 1e). This contradiction can NOT be understood by the change of dispersions. Other effects beyond the energy band theory should be considered.

(4) The enhanced carrier scattering can induce insulating or semiconducting behaviors. For example, in Kondo systems, the fluctuation of local moments at high temperatures induces insulating resistivity [Rev. Mod. Phys. 66, 755 (1984)]. The physics in $CsCr_3Sb_5$ is similar to the Kondo systems [arXiv: 2401.16770 (2024)]: Hund's coupling induces the localization of the magnetic moments and intrinsically incoherent Cr 3*d* states. In the paramagnetic state above the transition temperature, the fluctuation of local spin moments further enhances the scattering rate of electrons (including the Sb *p* electrons due to the *p*-*d* interaction), as observed in our temperature-dependent measurements, which will play an important role in the resistivity of the system.

Based on the above facts and arguments, we believe that the observed sudden increase of the scattering rate is related to the semiconducting behavior above the transition temperature. To relieve the concern of the reviewer, in the revised manuscript, we changed the sentence "*which can explain the semiconducting behavior of the system at high temperatures*" to "which is related to the semiconducting behavior of the system at high temperatures" in line 8 of paragraph 2 in page 4. We also changed "which is relevant to the semiconducting-like transport property" to "which is argued to be relevant to the semiconducting-like transport property" in the abstract.

In line 5 of the third paragraph in page 7, we have mentioned that the laser-ARPES measures the electron band around $\bar{\Gamma}$ (due to its limited momentum detection range). The analysis of the MDC width was done for this Sb *p* orbital band. In the revised manuscript, we have more explicitly pointed out that the MDC width of Sb *p* orbital band was tracked in Fig. 4.

Reviewer's Comment:

Also, precisely at the Fermi level, the analysis of the MDC widths is complicated by the Fermi functon(see for instance Phys. Rev. Lett. 131, 236502 for a recent example). . How does the width behave at higher binding energies?

Authors' response:

We agree with the reviewer that the MDC analysis can be complicated by the Fermi function. However, as shown in Phys. Rev. Lett. 131, 236502, the effect is particularly significant for heavy bands near the Fermi level and for $\Delta E/k_BT$>>1, where ΔE is the energy resolution and k_BT is the thermal energy. In our case, neither the band in analyses is a heavy band nor the condition of $\Delta E/k_BT$>>1 is fulfilled (the energy resolution of our Laser-ARPES is about 3 meV and we focuses on the temperatures near the density-wave transition). Moreover, this method has been widely used to explain the transport behavior [Science 285, 2110 (1999), Physica B: Condensed Matter 351, 250 (2004)], extract electron-self-energy, estimate the electron-phonon and electron-electron interaction strength [Science 285, 2110 (1999), Phys. Rev. Lett. 108, 187001 (2012), Phys. Rev. B 82, 195132 (2010)], and estimate the mean free path [PRL 96, 017005 (2006)] in the literature.

We also fit the MDCs at about 30 meV below E_F as shown in Fig. R8. A similar enhancement of the scattering rate is observed. Therefore, we believe that our method is well justified.

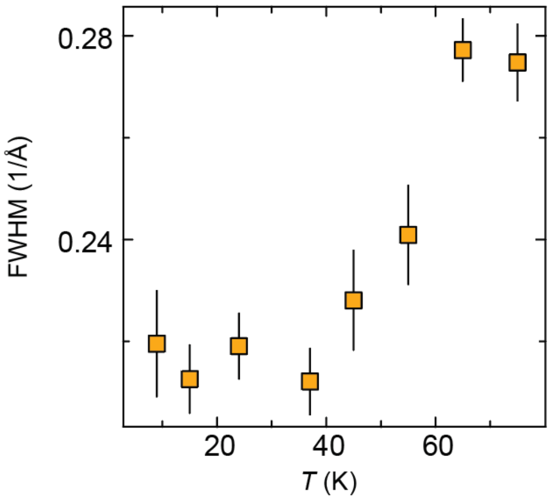

Fig. R8. Temperature evolution of the width of the MDC at 30 meV below E_F.

We thank the reviewer again for his/her helpful comments and discussions that greatly help us improve our manuscript. In our response letter and revised manuscript, we have addressed all the questions. We hope our responses can relieve the concerns of the reviewer and sincerely ask him/her to reconsider his/her recommendation.

<u>**Reply to Reviewer #3**</u>

Reviewer's Comment:

Superconductivity in highly correlated kagome systems has been a topic of theoretical interest for years, with experimental realization achieved in CsV_3Sb_5, which exhibits superconductivity, charge-density-wave (CDW) orders, and nonmagnetic behavior with weak electron correlations. In contrast, $CsCr_3Sb_5$, where V is replaced with Cr, features strong electron correlations, frustrated magnetism, and characteristic flat bands near the Fermi level. A comparative study of the electronic structures of these two materials offers a deeper understanding of correlated kagome systems.

The authors conducted transport measurements, ARPES (angle-resolved photoemission spectroscopy), and DFT+DMFT (density functional theory + dynamical mean field theory) calculations to investigate the electronic structure of $CsCr_3Sb_5$. Their findings indicate that Cr 3d electrons exhibit incoherence due to strong electron correlations driven by Hund's coupling. The study also reveals the presence of an "incipient flat band" near the Fermi level (E_x) at the Brillouin zone (BZ) center. Additionally, a scattering rate increase at 55 K was observed, potentially linked to CDW transition behavior. The authors propose that $CsCr_3Sb_5$ is a Hund's metal with an incipient flat band near E_x, which could underlie the novel properties of this material.

$CsCr_3Sb_5$ offers a promising platform for studying correlation effects in kagome metals, particularly exotic phenomena such as CDW and superconductivity. The ARPES measurements conducted by the authors covered the entire 3D BZ, mapping the electronic structure in detail, albeit with data quality that is high but not exceptional. Before the manuscript can be considered for publication in Nature Communications, several questions and concerns need to be addressed:

Authors' response:

We sincerely appreciate the time and effort the reviewer has devoted to carefully reviewing our work. We also thank the reviewer for abstracting our manuscript and acknowledging the comprehensiveness of our work. The pertinent and insightful comments have greatly helped us improve our manuscript. Below are our point-by-point responses to the reviewer's comments:

Reviewer's Comment:

Comparison with CsV_3Sb_5: $CsCr_3Sb_5$ introduces three additional electrons per kagome cell, effectively shifting the bands downward under a rigid band approximation. In both experimental *and calculated data, what are the major differences in band structure and Fermi surface (FS) topology between the two materials? The calculated FS topology (Fig. 1f, g) and experimental data (Fig. 2a, Fig. 3) suggest a three-dimensional character for $CsCr_3Sb_5$, unlike the primarily two-dimensional nature of CsV_3Sb_5. Additionally, the DFT+DMFT calculations (Fig. 2e) highlight the correlation effects. How do the renormalization factors differ between these materials, and what is the primary driving force behind these differences (e.g., Hund's coupling)?*

Authors' response:

We thank the reviewer for this discussion.

(1) The major difference in the band structure:

From the theoretical perspective, as compared in Figs. 1f and g, the basic band structures of the two compounds are similar. However, due to the additional electrons introduced by Cr atoms, the Fermi level of $CsCr_3Sb_5$ is raised close to the flat bands. Meanwhile, due to the electron correlation effect, the band width is narrower in $CsCr_3Sb_5$.

From the experimental perspective, the electron correlation effect in $CsCr_3Sb_5$ is further enhanced by Hund's coupling, inducing incoherent electronic states. Meanwhile, the flat bands are pushed below the Fermi level, forming the incipient flat band.

(2) Comparison between the Fermi surface of the two compounds:

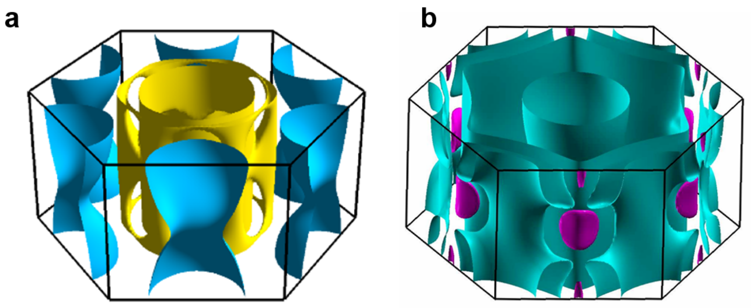

Fig. R9. Comparison between the calculated Fermi surfaces of (a) $CsCr_3Sb_5$ and (b) CsV_3Sb_5.

Due to the different Fermi level positions, the Fermi surface is different in the two compounds. According to the calculation, there are three and two bands crossing the Fermi level in $CrCr_3Sb_5$ and CsV_3Sb_5, respectively (Figs. 1f and g in the main text). Therefore, the calculated Fermi surface is different in the two compounds as compared in Fig. R9. In $CsCr_3Sb_5$, there are two hole pockets and one electron pocket around $\bar{\Gamma}$, together with an electron pocket around $\bar{M}$. By contrast, there is one electron pocket and a large hole pocket around $\bar{\Gamma}$, together with a pocket-like feature near $\bar{K}$ in CsV_3Sb_5. In the experiment, the Fermi surface of $CsCr_3Sb_5$ mainly consists of an electron pocket around $\bar{\Gamma}$ and spectral weight distribution near $\bar{M}$, while the experimental Fermi surface of CsV_3Sb_5 consists of complex warped star-of-David features in addition to the electron pocket around $\bar{\Gamma}$. The Fermi surfaces of the two compounds are clearly different.

(3) Three-d*imensionality of two compounds:*

Regarding the three-dimensionality of the calculated Fermi surfaces. We think it is quite similar in the two compounds, as shown in Fig. R9. This similarity can also be seen from the calculated band structure along ΓA (Figs. 1f and g in the main text).

(4) The difference in the renormalization factor:

CsV_3Sb_5 is generally recognized as a weak-correlated material with negligible band renormalization effect. The overall renormalization factor of $CsCr_3Sb_5$ is about 1.4 (Supplementary Fig. 4). This difference is mainly induced by the relatively stronger on-site electron correlation of Cr 3*d* electrons.

(5) *Primary driving force behind these differences:*

Apart from a relatively stronger correlation of Cr 3*d* electrons, as pointed out by the reviewer, Hund's coupling also plays an important role in the electronic correlation of $CsCr_3Sb_5$. It enhances electron self-energy and effective mass, modulates the position of the flat band, and induces an orbital-dependent correlation effect [arXiv: 2401.16770 (2024)].

Reviewer's Comment:

Orbital-Selective Band Shift: The experimental data show a downshift of the flat band below EF, whereas other bands at similar energies (Fig. 3a) remain largely unaffected. Only the band near the BZ center experiences this pronounced effect. Could the authors discuss the cause of this orbital-selective band shift?

Authors' response:

We thank the reviewer for this discussion. The electron correlation enhanced by the Hund's coupling renormalizes the overall band structure. For the dispersive band, particularly the weakly correlated *p*-bands, this renormalization is manifested by the change of band width. For the correlated flat band, this renormalization effect is mainly observed as a "band shift" (Supplementary Fig. 8). Experimentally, the "band shift" is easier to be resolved than the band width renormalization.

Moreover, the Hund's coupling also induces an orbital-dependent correlation effect. That is, the flat band is most strongly affected. A similar orbital-dependent correlation effect induced by the strong Hund's coupling has been revealed in other ARPES studies of $CsCr_3Sb_5$ [arXiv: 2406.05293 (2024)] and $Cs(V_{1-x}Cr_x)_3Sb_5$ [arXiv: 2407.03147 (2024)].

Reviewer's Comment:

CDW Features: While the material undergoes a CDW transition, as noted in prior reports, the authors did not observe band folding, shadow bands, or gap openings. What could account for this discrepancy? Could it result from mixing of bulk and surface states?

Alternatively, could surface reconstruction or surface disorder below 55 K (as observed through techniques like LEED or STM) explain the absence of these features?

Authors' response:

We thank the reviewer for raising this interesting question. The absence of the CDW/SDW-related band reconstruction (folding, shadow bands, or gap opening) can be understood for the following reasons:

(1) According to previous X-ray diffraction experiments, the system forms a four-fold CDW [Nature 632, 1032–1037 (2024)]. Therefore, the wavevector of the density wave is small compared to the BZ. The band folding may be complicated and difficult to resolve from the main bands.

(2) The electronic states are intrinsically incoherent due to the electron correlation effect enhanced by the strong Hund's coupling. While we believe that there are band folding and/or gap opening related to the CDW/SDW formation in $CsCr_3Sb_5$, it is extremely challenging to resolve these fine features related to the density-wave states.

(3) As pointed out by the reviewer, the mixing of bulk and surface states may affect the CDW-related band reconstruction, as already noticed in AV_3Sb_5 [see, e.g. Phys. Rev. B 107, 245143 (2023)].

We believe the reviewer raised an important question that deserves further experimental investigation. Surface-sensitive experiments such as LEED and STM, as proposed by the reviewer, will certainly deepen our understanding of the novel properties of $CsCr_3Sb_5$. We look forward to these experimental breakthroughs in the future.

Reviewer's Comment:

Comparison with Other ARPES Data: Another ARPES study (arXiv:2406.05293v2) presents visually higher-quality data. What are the major differences between the findings of these two reports?

Authors' response:

We thank the reviewer for comparing our work with other ARPES data. We believe the visually different data quality is mainly due to the color scale used in the data plot. Fig. R10 compares our data with those in arXiv: 2406.05293 in the same color scale. We can safely conclude that the data quality in the two experiments is comparable [the flat band seems to be better resolved in our experiment (Fig. R2)].

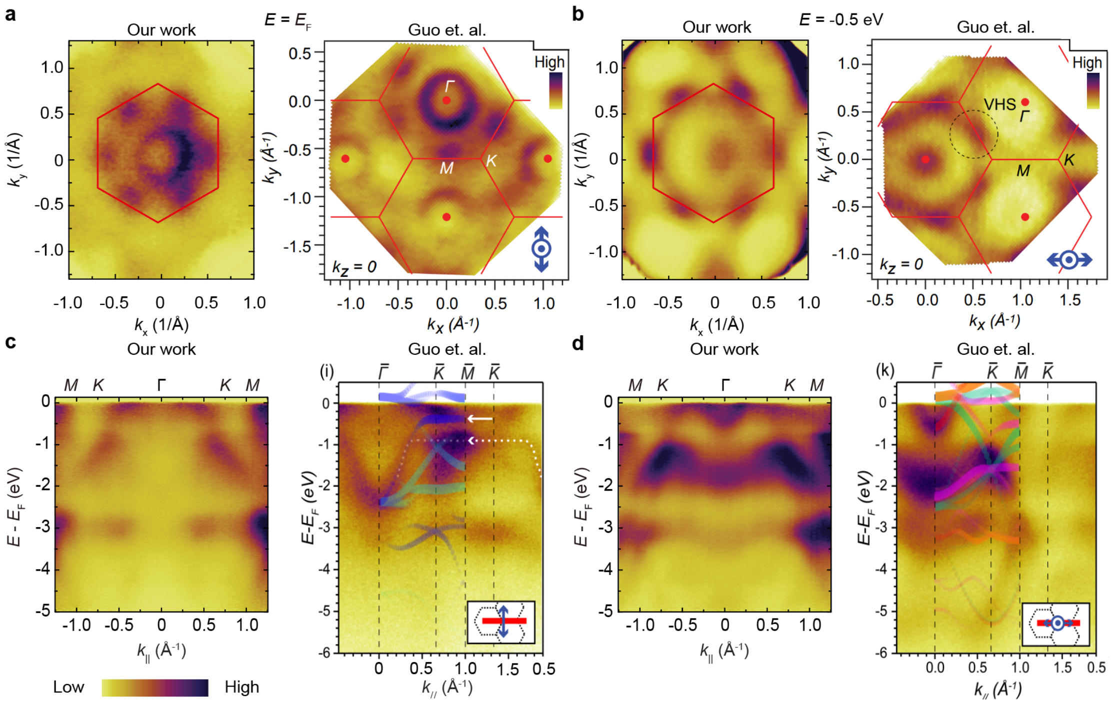

Fig. R10. Comparison between our work and Guo et. al. [arXiv:2406.05293v2] in the same colormap.

The major results in our work and arXiv:2406.05293v2 are similar and consistent. The main differences between the two works are as follows:

(1) With the help of DMFT calculation, our work reveals strong Hund's coupling semi-quantitatively, while Guo et al. discussed the effect of Hund's coupling mainly in a qualitative way. By polarization-dependent experiments, Guo et al. discussed the orbital-dependent correlation effect in detail.

(2) We reveal the enhancement of the carrier scattering rate above the density-wave transition, while Guo et. al. focused on the temperature dependence of the flat band.

We emphasize again that the two works are in overall agreement with each other and both provide important insights into the electronic properties of $CsCr_3Sb_5$.

Reviewer's Comment:

Minor Points: In Fig. 2(e-g), the directions should be explicitly labeled, as currently only k//k_{//}k// is indicated. To improve clarity, overlaying DFT calculations with experimental data would better illustrate their consistency.

Authors' response:

We thank the reviewer for this helpful suggestion. In the revised manuscript, we have added labels of high-symmetry directions. The ARPES data with the calculated results overlaid are shown in Supplementary Fig. 4, which shows the overall consistency between the experiment and calculation, as suggested by the reviewer.

Reviewer's Comment:

In summary. The authors performed detailed measurements and theoretical analyses of $CsCr_3Sb_5$. However, until the issues outlined above are resolved, the manuscript is not yet suitable for publication in Nature Communications in its current form.

Authors' response:

We sincerely thank the reviewer for his/her insightful suggestions and comments. With his/her help, the manuscript is now greatly improved. We believe that we have properly addressed all the concerns of the reviewer in our response letter and the revised manuscript that is now ready for timely publication. We look forward to the reviewer's further comments and his/her final recommendation for the publication of our manuscript.

List of changes (highlighted in yellow in the revised manuscript)

1. In the abstract, we changed the sentence "play an important role in the unconventional superconductivity" to "play an important role in the electronic properties and unconventional superconductivity".
2. In the abstract, we changed the sentence "which is relevant to the semiconducting-like transport property" to "which is argued to be relevant to the semiconducting-like transport property".
3. In line 8 of paragraph 2 in page 4, we changed the sentence "which can explain the semiconducting behavior" to "which is related to the semiconducting behavior".
4. In line 4 of paragraph 3 in page 4, we added the sentence "Previous X-ray scattering and nuclear magnetic resonance experiments revealed an intertwined CDW and spin-density wave (SDW) below 55 K."
5. In line 1 of paragraph 1 in page 5, we changed "Figs.1d" to "Fig.1d".
6. In line 9 of paragraph 2 in page 5, we changed the sentence "With increasing binding energy, the pocket around $\bar{\Gamma}$ shrinks and line-like features can be resolved, forming a kagome-shape structure in the momentum space (Fig. 2d)." to "With increasing binding energy, the circular electron pocket around the $\bar{\Gamma}$ point shrinks and a star-of-David-shape pattern can be resolved at -0.4 eV, as indicated by the black dashed lines in Fig. 2d".
7. In line 5 of paragraph 3 in page 5, we changed the sentence "These features are in overall consistent with the calculation in Fig. 1f, as confirmed by the comparison between the integrated energy distribution curve (EDC) and the calculated density-of-states (DOS) in Fig. 2h" to "These features contribute peaks in the integrated energy distribution curve (EDC) that is in overall consistency with the calculated density-of-states (DOS), as compared in Fig. 2h".
8. In line 6 of paragraph 2 in page 6, we added the sentence "We notice that the electron band around $\bar{\Gamma}$ has mixed *p* and *d* orbital components, suggesting a *p*-*d* interaction (Supplementary Information)."
9. In line 3 of paragraph 3 in page 6, we change the sentence "This flat band close to E_F may dominate the transport properties of $CsCr_3Sb_5$" to "This flat band close to E_F may play an important role in the transport properties of $CsCr_3Sb_5$".
10. In line 6 of paragraph 3 in page 6, we change the sentence "suggests that the DFT-calculated flat bands are pushed closer to E_F and the experimental E_F is slightly raised up" to "suggests

that the DFT-calculated flat bands are pushed closer to E_F (Supplementary Information)".

11. In line 11 of the last paragraph in page 6, we added the sentences "It is likely that E_F of $CsCr_3Sb_5$ crystals are slightly raised up by self-doping or surface effects and the flat bands, similar to the situation in CsV_3Sb_5. Therefore, the flat band can be experimentally observed by ARPES experiment."
12. In line 5 of paragraph 2 in page 7, we change the sentence "the calculation with $U = 5$ eV and $J_H = 0.5$ eV" to "the calculation with different Hund's coupling".
13. In line 5 of paragraph 3 in page 7, we change the sentence "The electron band around $\bar{\Gamma}$" to "The Sb *p* orbital band around $\bar{\Gamma}$".
14. In line 1 of paragraph 1 in page 8, we change the sentence "By fitting the MDCs at E_F to Lorentzians" to "By fitting the MDCs of the Sb *p* orbital band at E_F to Lorentzians".
15. In line 8 of paragraph 1 in page 9, we added the sentence "and the competition between density-wave and pressurized superconductivity."
16. We updated Fig. 2 by labeling the high-symmetry directions.
17. We included new sessions in the Supplementary Information (IX: flat band measured at 200 eV; X: Temperature dependence of the flat band) to support our discussion.
18. We properly cited the three other ARPES studies of $CsCr_3Sb_5$ and $Cs(V_{1-x}Cr_x)_3Sb_5$.

<u>Reply to Reviewer #4</u>

<u>Reviewer's Comment:</u>

The authors of the manuscript report on temperature-dependent ARPES measurements on the kagome CsCr3Sb5 and discuss the electronic properties of the system in the context of Hund's metal with an incipient flat band near the Fermi surface supported by DFT and DFT+DMFT calculations. This is an interesting work, which, in view of the present discussion on this kagome material, brings some valuable information to the community studying these materials.

There are, in my opinion, still a few issues that the authors should further clarify before recommending the paper for publication.

<u>Authors' response:</u>

We sincerely thank the reviewer for his/her time and efforts in reviewing our manuscript. We appreciate that the reviewer acknowledges our work as "an interesting work" that "brings some valuable information to the community studying these materials". In response to the reviewer's comments, we have carefully addressed all the concerns in the revised manuscript. Below are our point-by-point responses to the reviewer's comments:

<u>Reviewer's Comment:</u>

1) As reviewer 1 points out, it is unclear what information the authors extract from the transition at T=55K with their measurements. The authors refer to the literature in term of an ' intertwined spin and charge density wave'. What does this exactly mean? And what signatures of it are the authors seeing in their experiments? Here the authors should state what they can interpret from their observations. Along the manuscript they only mention a CDW but not a spin density wave, do they find signatures for such a spin density wave? This would be a very useful piece of information.

<u>Authors' response:</u>

We thank the reviewer for this discussion.

(1) The term "intertwined spin and charge density wave" refers to coexisted CDW and SDW that can interplay with each other. It was identified in Nature 632, 1032 (2024). The SDW was detected through magnetic susceptibility and NMR experiments, while the CDW was observed in X-ray diffraction measurements. Similar intertwined density-wave states have also been revealed in other correlated materials such as nickelate superconductors. Interestingly, nickelate

superconductor exhibit properties analogous to $CsCr_3Sb_5$, including flat band, strong Hund's coupling, and superconductivity under pressure [Nat. Commun. 11, 6003 (2020); Nature 631, 531 (2024)].

(2) In our transport measurements, the intertwined density wave orders are manifested by the abrupt changes in resistivity and magnetic susceptibility measurements, consistent with the previous study [Nature 632, 1032 (2024)].

(3) In our ARPES experiments, we did not observe clear band reconstruction related to the density wave transition, as we have explicitly claimed in the manuscript. We emphasize that the major scope of the current work is to study the correlated kagome electronic structure of this interesting material. While the connection between the electronic structure and the density-wave transition is important, it remains a topic for future investigation, as we have proposed in our manuscript (line 2, the first paragraph of page 8). **Nevertheless, this result is understandable based on the following facts (please also see our previous reply to the comments of reviewer #3)**: (a) In strongly correlated materials such as cuprate superconductors and Hund's metal $La_3Ni_2O_7$, it is also challenging to resolve the band reconstruction related to the charge order or density waves using ARPES [Rev. Mod. Phys. 93, 025006 (2021); Nat. Commun. 15, 4373 (2024)]. (b) According to previous X-ray diffraction experiments, the system forms a four-fold CDW below the transition temperature [Nature 632, 1032–1037 (2024)]. Therefore, the wavevector of the density wave is small compared to the Brillouin zone. The band folding may be complicated and difficult to resolve from the main bands. (c) The Cr 3*d* electronic states of $CsCr_3Sb_5$ are intrinsically incoherent due to the electron correlation effect enhanced by the strong Hund's coupling, making it extremely challenging to resolve the fine features related to the density-wave transition.

(4) Regarding "*what they can interpret from their observations*": Although we did not observe the change of the band structure, the density-wave transition is indeed manifested in our temperature-dependent experiment by the enhancement of the carrier scattering with increasing temperature across the critical temperature (Fig. 4g of the main text). This observation (a) not only confirms the density-wave transition near 55 K (b) but also is relevant to the semiconducting-like behavior at high temperatures. (c) Moreover, according to the previous theoretical study [arXiv: 2401.16770], it is consistent with a Kondo-like behavior by screening

the local moments at low temperatures.

Finally, with due respect, we would like to point out that there might be a misunderstanding regarding the comment that "*Along the manuscript they only mention a CDW but not a spin density wave*". In the manuscript we refer the transition near 55 K as a "density wave" that consists both CDW and SDW.

We have discussed our main findings and their significance in the "discussion" section. Following the comment of the reviewer, we added the sentences "With decreasing temperature, the system is expected to undergo an incoherence-to-coherence crossover by screening the local moments [45], and the relatively coherent flat bands can be observed near E_F, which is also consistent with our observation of the reduced scattering rate with decreasing temperature." in line 10 of paragraph 3 in page 8.

Reviewer's Comment:

2) the ARPES data look very broad (not only near Ef) and interpretation of them seems to strongly rely on the calculations. While the information of incoherent versus coherent spectral weight as a function of temperature plays a role (see next comments), could the authors include further comments on the quality of their samples?

Authors' response:

We thank the reviewer for this comment regarding the sample quality.

Generally speaking, understanding the experimental results by theoretical calculation is necessary and important. In particular, it has been commonly shown that the interpretation of ARPES data on the topological materials and kagome materials strongly relies on the calculations. In kagome materials, the existence of the flat band is guaranteed by their structural geometry, which must be evidenced by theoretical calculation. Otherwise, the observed features are groundless.

In $CsCr_3Sb_5$, the broad ARPES data are primarily attributed to its strongly correlated nature (please also see our previous reply to the comments of reviewer #3), as evidenced by other independent studies [arXiv: 2406.05293, arXiv: 2406.17769]. The samples used in our experiments were provided by the group that first synthesized $CsCr_3Sb_5$ and discovered its pressurized superconductivity. Rigorous characterization of the samples guaranteed their quality [Nature 632, 1032 (2024)], which is also confirmed by the following experiments:

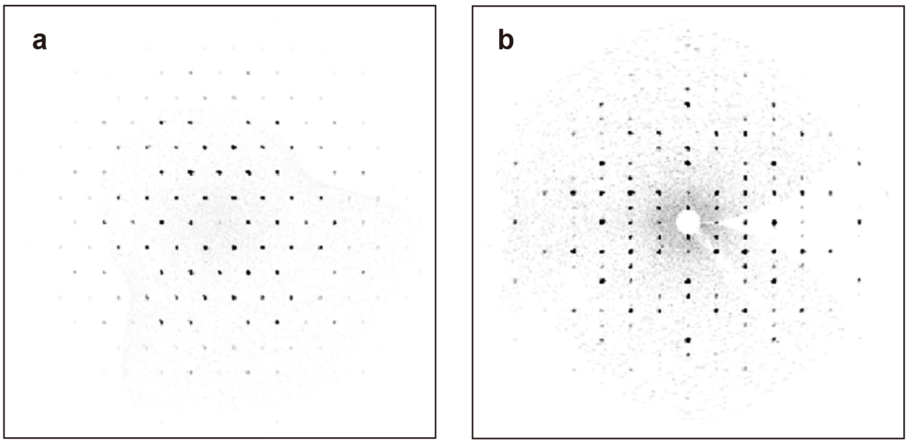

Fig. R1 | Single crystal X-ray diffraction (XRD) of $CsCr_3Sb_5$. Patterns were measured along the (**a**) (hk2) plane, and (**b**) (0kl) plane.

(1) Single crystal XRD experiments reveal shape diffraction peaks (Fig. R1), confirming the high quality of the samples in our experiment.

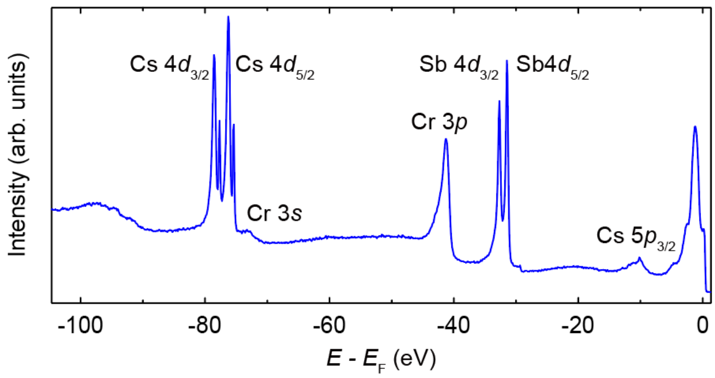

Fig. R2 (the same as Supplementary Fig. 3) | Core-level photoemission spectra of $CsCr_3Sb_5$. Data were measured with 160 eV photons.

(2) We observe sharp peaks from Cs, Cr, and Sb atoms in the core-level photoemission experiment (Fig. R2), confirming the proper composition of our samples and its high quality.

To relieve the concern of the reviewer, we include a new section "Single crystal X-ray diffraction measurements of $CsCr_3Sb_5$" in the Supplementary Information to show the quality of our samples.

Reviewer's Comment:

3) For the discussion, the authors strongly rely on DMFT calculations performed by one of the authors in a previous publication, nevertheless, more information on these calculations should be included in the manuscript. Are these charge self-consistent calculations? at which temperatures in the QMC calculations are they performed? the discussion of cohernet vs. ncoherent spectral weight *needs some further elaboration. How are the calculations treated in the CDW phase? Sb hybridizes strongly with Cr, how is this considered in the calculations?*

Authors' response:

We thank the reviewer for this comment.

We have mentioned in the method part that the DMFT calculations were performed with full charge self-consistency. Here are more details about the calculation:

The DFT+DMFT calculations are performed with full charge self-consistency using the EDMFTF code developed by Haule et al. based on Wien2k package. The temperature for CTQMC is 100 K. We choose an energy window of -10 eV to 10 eV, including both the correlated Cr 3*d* orbitals and other non-correlated orbitals such as Sb *p* orbitals, so the strong hybridization effect between the Sb *p* and Cr 3*d* orbitals has been captured in the calculations. For example, the strong hybridization will lead to ~4.3 electrons in Cr 3*d* orbitals instead of the nominal 4 electrons. The DFT+DMFT calculations are performed in the non-CDW state, i.e., the ideal crystal structure without any distortion. This is justified by the experimental result that the electronic structure shows minor change across the density-wave transition.

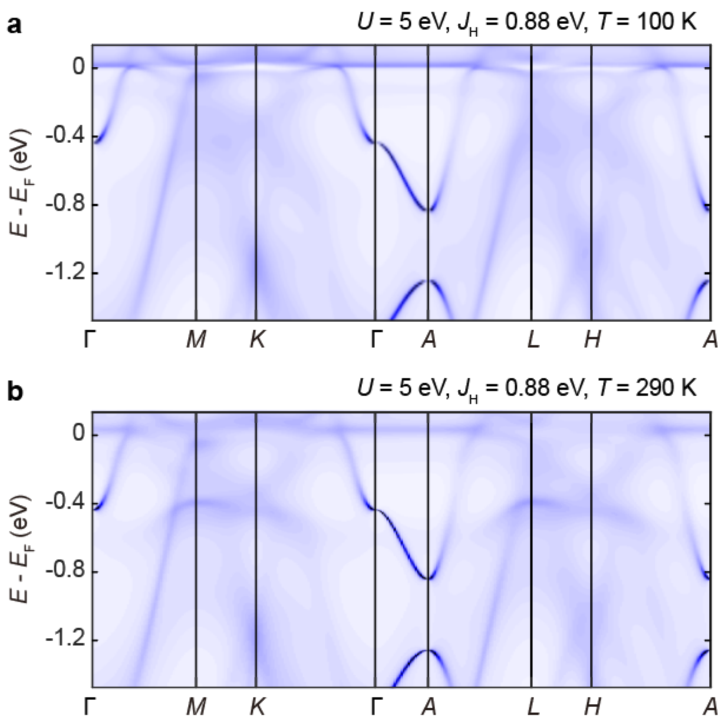

Fig. R4 | DFT+DMFT calculation of the electronic structure, with (**a**) on-site Coulomb interaction $U = 5$ eV, Hund's coupling $J_H = 0.5$ eV, temperature $T = 100$ K, and (**b**) $U = 5$ eV, $J_H = 0.88$ eV, $T = 290$ K.

Regarding the coherent vs. incoherent spectral weight, Hund's correlation leads to incoherent spectra at high temperatures due to strong fluctuations of local Cr 3*d* moments. As the temperature decreases, the system is expected to undergo an incoherence-to-coherence crossover by screening the local moments. Therefore, the relatively coherent flat bands can be observed near E_F, as shown in the DMFT calculations at different temperatures (Fig. R4), which is similar to the Kondo screening physics.

Based on the above discussions, in line 10 of paragraph 3 in page 8, we added the sentences "With decreasing temperature, the system is expected to undergo an incoherence-to-coherence crossover by screening the local moments [45], and the relatively coherent flat bands can be observed near E_F, which is also consistent with our observation of the reduced scattering rate with decreasing temperature."

To relieve the concern of the reviewer, we add more details in the "method" section about DFT+DMFT calculations. We also include a new section "DFT+DMFT calculation of the electronic structure at different temperatures." in the Supplementary Information to support our discussions.

Reviewer's Comment:

After the authors have clarified these points, in my opinion the manuscript is suitable for publication.

Authors' response:

Once again, we sincerely thank the reviewer for his/her valuable discussions and suggestions, which have significantly improved the quality of our manuscript. We hope our responses have addressed his/her concerns and look forward to the final recommendation.

List of changes (highlighted in yellow in the revised manuscript)

1. In line 10 of paragraph 3 in page 8, we added the sentences "With decreasing temperature, the system is expected to undergo an incoherence-to-coherence crossover by screening the local moments, and the relatively coherent flat bands can be observed near E_F, which is also consistent with our observation of the reduced scattering rate with decreasing temperature."
2. We add more details in the "method" section about DFT+DMFT calculations.
3. We include a new section "Single crystal X-ray diffraction measurements of $CsCr_3Sb_5$" in the Supplementary Information to support our discussions on samples quality.
4. We include a new section "DFT+DMFT calculation of the electronic structure at different temperatures." in the Supplementary Information to support our discussions regarding the coherent-to-incoherent crossover.