nature portfolio

Peer Review File

Charge-spin dichotomy in the kagome metal $CsCr_3Sb_5$

Corresponding Author: Professor Takasada Shibauchi

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)
The manuscript by Onishi et al. investigates the electronic orderings of the kagome metal CsCr3Sb5 through elastoresistivity and linear dichroic (LD) measurements using a laser-based photoemission electron microscopy (laser-PEEM). Studies on this specific CsCr3Sb5 compound are interesting and timely. CsCr3Sb5 is closely related to the intensely studied kagome superconductor CsV3Sb5, but harbors stronger correlations presumably originating from flat electronic band close to EF. The experimental situation needs to be scrutinized. Previous works established a magnetic transition at T*=55K, but under hydrostatic pressure this transition seems to split. The second transition was previously suggested to be a charge density wave. In their work Onishi and collaborators suggest that (already) at ambient pressure there are two well separated electronic transitions in CsCr3Sb5. The first one is the magnetic transition at T*, of which they observe three separate domains in laser-PEEM experiments. The second one is a charge density wave that would appear at a slightly higher temperature of TCDW = 64K and would lead to observable electrical transport anisotropy.

Overall, the manuscript is well written, the data clearly presented, and references to the existing literature appropriate. However, the main conclusion of the work, i.e., that there is an electronic phase transition at T=64K corresponding to 4x1 charge ordering, is only weakly supported by the elastoresistance experiment. Given this, I suggest revising the manuscript and emphasizing the limitations of the current study, in particular that the interpretation of the elastoresistance data is not straightforward, i.e., that other interpretations are possible and likely.

The first part of the manuscript focuses on the Linear Dichroic PEEM experiment, which according to the authors probe mainly states near Γ. By varying the angle θ between the polarization E1 and the sample a-axis, the authors observe three domains, with a typical size of 10μm. Each domain has a different linear dichroic response that can be fitted to a cosinus with a distinct amplitude (I_fit) and angle offset (θ_fit). From a temperature dependence, the domains' structure seems to emerge around 55K, i.e. around T*. The authors' interpretation that these domains are associated to the T* transition seems very reasonable.

The second part of the manuscript focuses on the elastoresistance experiment. Technically, the authors used a modified Montgomery method that allows extracting the resistance anisotropy on a single sample as a function of anisotropic strain. This precludes the use of two samples as in the 'differential' method that can mix E2g and A1g signals (which recently was the origin of conflicting results in CsV3Sb5). Thus, the data is most probably robust. There are multiple interesting aspects in the reported data. First the 'nematic' E2g response shows an enhancement below 64K, before it peaks at around 55K. Second, the isotropic A1g coefficient also starts to deviate from a monotonic behavior below 64K and shows multiples anomalies, with (at least) three notable extrema.

There are multiple ways to interpret the E2g elastoresistance response. The authors' interpretation is that there is a second-order phase transition at 64K and a corresponding rotational symmetry breaking. Tentatively it would correspond to a charge-density wave suggested in prior XRD experiments. This explains the emerging anisotropy seen at that temperature in the electrical transport measurement under finite anisotropic strain (fig 3e) needed to partially detwin the CDW-related domains. This also fits with the emergence of hysteretic behavior below 65K or so.

However, I have difficulties with this interpretation. Assuming that there is a CDW order parameter that develops below 64K, and couples to E2g ϵ_xx-ϵ_yy strain, one expects an extremum of the corresponding elastoresistance coefficient at that temperature. The maximum sensitivity of the order parameter is right at the phase transition, and an extremum in m_E2g is simply expected from the strain dependence of the critical temperature TCDW. For instance, in CsV3Sb5 the m_E2g

coefficient peaks at T_CDW (refs 15,16 and ref 14 from some of the present authors). Similarly, in the case of BaNi2As2 that is discussed by the authors, the CDW transition is associated with a maximum of the m_B1g coefficient (refs 46,47). Here, however, 64K does only correspond to an enhancement of the elastoresistance coefficient, nothing more. In the absence of a clear thermodynamic signature of a phase transition, for example using specific heat, it is simply extremely speculative to associate this temperature with a rotational symmetry-breaking induced by a charge-density wave.

One other possibility is that there is no six-fold to two-fold symmetry breaking at 64K. It may be simply broken at T*, where the E2g elastoresistance coefficient is maximized. The fact that there is an enhancement of m_E2g below 64K alone does not indicate rotational symmetry-breaking. The same is true for the resistance anisotropy in Fig.3e, as it may be a simple consequence of the growing E2g elastoresistance. The growing elastoresistance may simply be a consequence of the emergence of E2g-symmetric fluctuations (and not of a phase transition) or, alternatively, signal a broadening of the T* transition that is induced by the finite anisotropic stress applied experimentally because the sample suffers from differential thermal expansion with the PZT piezoelectric.

The authors also report the symmetry-preserving m_A1g elastoresistance coefficient. This is very appreciable. The data show three local extrema (Fig. 3d) around 57K, 55K and 45K. One could tentatively associate these extrema to different instabilities, one being the magnetic T* transition. Like the E2g coefficient there is, however, only an enhancement of the response below 64K and no sharp anomaly at that temperature. Again, it is then surprising to associate this temperature with a CDW transition. Because the corresponding strain is symmetry-preserving, there is in principle no reason not to expect coupling between the CDW order parameter and the $\epsilon_{xx}+\epsilon_{yy}$ strain which in turns would imply a distinct anomaly at 64K beyond a simple enhancement.

Thus, different interpretations are possible. A clear assessment requires additional measurements that one can't reasonably ask the authors to provide in the present manuscript. Instead, my suggestion is to expose the different scenarios at hand in the manuscript, such that the reader has all information to interpret the present study.

Finally, I point out two details to the authors consideration:
* In some places 'uniaxial' or 'unidirectional' is incorrectly used. For instance, line 340 "under uniaxial strain" is incorrect, but instead "biaxial strain" could be used. Same thing for line 220 and "under unidirectional strain".
* Lines 268-271 "Such temperature dependence deviates from the Curie-Weiss divergence, excluding the existence of Fe-SC type of electronic nematicity in this material". If this is what is meant, I do not agree that not observing a Curie-Weiss diverging elastoresistance excludes electronic nematicity. Rather, it is surprising that the experimental situation is 'so' simple in the iron-based SCs. Even for a true electronic nematic instability a different temperature dependence of the 'nematic' elastoresistance can be observed if the electron-phonon coupling has a distinct temperature dependence. Alone a non-Curie-Weiss dependence alone is not a proof of the absence of electronic nematicity.

Reviewer #2

(Remarks to the Author)
Onishi and coworkers report on a study on the Cr Kagome homologue of the AV3Sb5 Kagome family, which is indeed gaining a rapidly strong interest owing to its different intriguing properties contrasting the V-based Kagome. At ambient pressure, so far only one phase transition featuring magnetic and structural properties at ~55 K had been reported. In their study, the authors could show two different distinct phase transitions at two different temperatures. The one already reported in the literature at 55K which they attribute to a spin density wave phase (SDW) and another at a distinct higher temperature at around 64K which they appoint to a charge density wave (CDW) phase. By using linear dichroism asymmetry in laser-assisted photoemission microscopy, they could demonstrate the rotational symmetry breaking at the SDW onset temperature. Indeed, they showed three distinct domains obeying a two-fold symmetry thus breaking the original six-fold symmetry. Moreover, by using elastoresistivity measurements and anisotropic strain-resistivity measurements, they could show two distinct phase transitions which they ascribe to separate CDW and SDW phase transitions. The authors argue that the phase transition at 55K should be ascribed to the SDW given that previous magnetic susceptibility measurements show an anomaly at the same transition temperature. Based on the possible electronic nesting nature of the 1x4 reported CDW away from the gamma, they assign the higher temperature transition to the charge order transition.

The manuscript and data presented by the authors are clear and well consistent with each other. While the observation of domain competition has been reported before (ref. 41), the imaging of these domain structure and the observation of an additional phase transition provide important new insight for understanding this novel material. Despite the limited but growing number of studies on this material and the current theoretical agenda, the theoretical claims and scenarios suggested by the authors are reasonable and offer new directions to explore in order to tap on these claims. The paper could be suitable for publication in Nature Communications once a few issues are addressed:

1) The authors argue that the 4x1 CDW sets in at 64 K, and that the SDW transition below 55 K is of 2x1 symmetry. Yet, superlattice peaks of 4x1 symmetry have been observed below 55 K (ref. 24, 31), but not above. This appears inconsistent with the claim of the authors, and needs to be addressed. In particular, the authors argue that e-ph coupling might be too weak to allow observation of the CDW above 55 K. But why is the observed period of the superstructure then 4x1 instead of 2x1 below 55 K?
2) What evidence besides the susceptibility and suggested nesting vectors do the authors have for the assignment of SDW and CDW transitions?
3) The hysteresis loops of the elastoresistivity curves are not centered around zero. Does this originate from the residual

strain the authors discuss?
4) What influence on the observed phase transitions does this residual strain has? Can the authors exclude with certainty that this residual strain is responsible for inducing the additional transition observed at 64 K?
5) In the supplement, section C, the authors discuss the photoemission matrix elements to originate from electron-phonon coupling. I believe this should read electron-photon coupling.
6) The authors discuss the derivation of the photoemission matrix elements in the limit of non-interacting electrons. However, they argue that CsCr3Sb5 shows strong electronic correlations. This appears inconsistent to me. What influence would the consideration of correlation effects have on their derivation?
7) In Fig. 1b, the a axis appears twice.
8) In Fig. 1c, the reciprocal axes are labelled q1, q2, but in the text they are referred to as a*.

Reviewer #3

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4

(Remarks to the Author)

This is a well-written manuscript that describes evidence for a phase transition at ~64K in single crystals of the kagome metal CsCr3Sb5. This identification relies on detailed analysis of linear dichroism microscopy and elastoresistivity measurements as functions of temperature.

The data at ambient pressure suggests this transition to be associated with a charge density wave and Fermi surface nesting around the M point of the Brillouin zone. In contrast, the authors identify the previously observed transition (~55K) as associated with a spin-density wave and nesting centered at the Gamma point. Although the 55K transition had been previously reported, and an additional feature at slightly higher T was seen under pressure, the current identification is well justified and would merit publication.

The work is timely, given the current interest in kagome metals. However, the current version of the manuscript would need clarification on several essential points, as follows:

1. The authors argue that the transition at 64 K deviates from the Curie-Weiss divergence, excluding electronic nematicity. However, the rotational symmetry breaking in the resistivity measurements could also be consistent with a nematic order. Moreover, kagome metals are known to support bond-density-wave orders, which may also lead to resistivity anisotropy. It would be helpful if the authors could discuss this issue and/or present more direct evidence for a genuine CDW.

2. What are the effects of disorder/impurities in the sample when measuring the elastoresistivity coefficients in Fig. 3, especially for temperatures between the two transition temperatures? The authors comment on the rounding of the coefficients near the transition as indicative of second-order behavior, but a similar effect would likely arise from inhomogeneities. It would be important for the authors to discuss the effects of sample quality and reproducibility when measuring multiple samples.

3. In Figs. 3d, e & g, there is a clear non-monotonic feature in the curves at around 8 K. Can the authors comment on such a deviation? Could it be associated with another kind of transition? An artifact of the measurement?

4. In Fig. 2f. the authors show a real space map of the angle theta_fit, which reveals domains with three preferred orientations. The authors interpret these domains as distinct density modulations at the three Cr sites, consistent with the breaking of C6 symmetry down to C2. Could the formation of such domains be associated with local uniaxial strain formation, which locally favors one orientation over others?

The evidence and discussion in the text are measured and appropriately non-conclusive. However, the tone of the abstract is significantly more definitive; I recommend revising it to better align with the main text.

Reviewer #5

(Remarks to the Author)
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Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have provided a careful response to my comments.

The arguments of the authors are reasonable. In particular, I agree that the absence of strong anomaly at 64K in the elastoresistance does not necessarily imply the absence of a CDW transition at that temperature. However, given the limitations of the elastoresistance technique that cannot access microscopic information, their interpretation remains hypothetical.

In this context, I believe that the revision of the manuscript with the addition of a discussion on alternative scenarios is particularly important. The point of the authors is now made clearer, and the readers will be able to judge by themselves. In turn, I believe that the manuscript is now ready for publication.

Reviewer #2

(Remarks to the Author)
The authors have addressed all comments appropriately. I can now support publication of the manuscript.

Reviewer #3

(Remarks to the Author)
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Reviewer #4

(Remarks to the Author)

The authors have thoroughly addressed all the issues raised by the referees and made the corresponding changes to the manuscript. These changes include important discussions of the possible scenarios underlying the new transition reported by the authors and the limitations of the present work. The new version not only provides important insights into this material behavior but also highlights outstanding issues that may motivate future work. Thus, I recommend the publication of the revised manuscript.

Reviewer #5

(Remarks to the Author)
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Reviewer #1 (Remarks to the Author):

The manuscript by Onishi *et al*. investigates the electronic orderings of the kagome metal $CsCr_3Sb_5$ through elastoresistivity and linear dichroic (LD) measurements using a laser-based photoemission electron microscopy (laser-PEEM). Studies on this specific $CsCr_3Sb_5$ compound are interesting and timely. $CsCr_3Sb_5$ is closely related to the intensely studied kagome superconductor CsV_3Sb_5, but harbors stronger correlations presumably originating from flat electronic band close to E_F. The experimental situation needs to be scrutinized. Previous works established a magnetic transition at T^*= 55 K, but under hydrostatic pressure this transition seems to split. The second transition was previously suggested to be a charge density wave. In their work Onishi and collaborators suggest that (already) at ambient pressure there are two well separated electronic transitions in $CsCr_3Sb_5$. The first one is the magnetic transition at T^*, of which they observe three separate domains in laser-PEEM experiments. The second one is a charge density wave that would appear at a slightly higher temperature of T_{CDW} = 64 K and would lead to observable electrical transport anisotropy.

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There are multiple ways to interpret the E_{2g} elastoresistance response. The authors' interpretation is that there is a second-order phase transition at 64 K and a corresponding rotational symmetry breaking. Tentatively it would correspond to a charge-density wave suggested in prior XRD experiments. This explains the emerging anisotropy seen at that temperature

in the electrical transport measurement under finite anisotropic strain (fig 3e) needed to partially detwin the CDW-related domains. This also fits with the emergence of hysteretic behavior below 65 K or so.

Reply) We sincerely thank the reviewer for the careful reading of our manuscript and for the constructive and insightful comments. We have revised the manuscript to clarify the interpretation of the elastoresistivity data and to explicitly discuss alternative scenarios, as suggested.

However, I have difficulties with this interpretation. Assuming that there is a CDW order parameter that develops below 64 K, and couples to $E_{2g}\ \epsilon_{xx} - \epsilon_{yy}$ strain, one expects an extremum of the corresponding elastoresistance coefficient at that temperature. The maximum sensitivity of the order parameter is right at the phase transition, and an extremum in $m_{E_{2g}}$ is simply expected from the strain dependence of the critical temperature T_{CDW}. For instance, in CsV_3Sb_5 the $m_{E_{2g}}$ coefficient peaks at T_{CDW} (refs 15,16 and ref 14 from some of the present authors). Similarly, in the case of $BaNi_2As_2$ that is discussed by the authors, the CDW transition is associated with a maximum of the $m_{B_{1g}}$ coefficient (refs 46,47). Here, however, 64 K does only correspond to an enhancement of the elastoresistance coefficient, nothing more. In the absence of a clear thermodynamic signature of a phase transition, for example using specific heat, it is simply extremely speculative to associate this temperature with a rotational symmetry-breaking induced by a charge-density wave.

Reply) As the reviewer correctly points out, when a transition temperature depends on strain, the elastoresistivity coefficient can contain a contribution proportional to $\partial\rho/\partial T$, reflecting the strain dependence of the transition temperature (refs 14 and 15 from the main text). More explicitly, $(\mathrm{d}\rho/\mathrm{d}\epsilon)_{T_{\mathrm{CDW}}} \propto (\partial\rho/\partial T)_{T_{\mathrm{CDW}}}(\mathrm{d}T_{\mathrm{CDW}}/\mathrm{d}\epsilon)$. In CsV_3Sb_5 and similarly in $BaNi_2As_2$, the peak in the elastoresistivity coefficient at T_{CDW} originates from the fact that $\partial\rho/\partial T$ itself exhibits a pronounced maximum at the transition. In the present case, however, as shown in Fig. R1, $\mathrm{d}\rho_{xx}/\mathrm{d}T$ exhibits a change in temperature dependence around 64 K but does not show a clear extremum at that temperature. Note that resistivity depends on both the number and scattering time of carriers, and that changes in these factors at a transition may cancel each other out. Thus, resistivity anomalies at the transition are sometimes very weak. Therefore, even if a transition exists near 64 K and its temperature depends on strain, one does not necessarily expect a pronounced peak in the elastoresistivity coefficient. Accordingly, the absence of a sharp extremum in $m_{E_{2g}}$ at ~ 64 K does not contradict the interpretation that a transition may occur near this temperature.

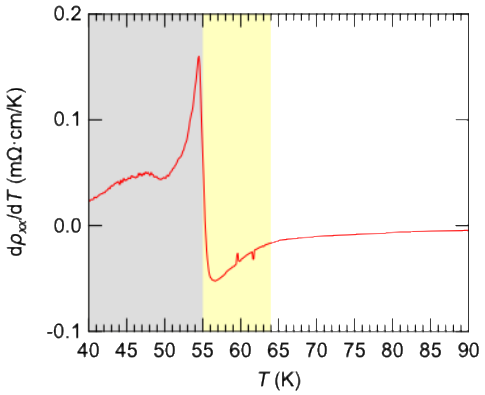

Fig. R1. Temperature dependence of $\mathrm{d}\rho_{xx}/\mathrm{d}T$ measured by four-terminal method.

One other possibility is that there is no six-fold to two-fold symmetry breaking at 64 K. It may be simply broken at T^*, where the E_{2g} elastoresistance coefficient is maximized. The fact that there is an enhancement of $m_{E_{2g}}$ below 64 K alone does not indicate rotational symmetry-breaking. The same is true for the resistance anisotropy in Fig.3e, as it may be a simple consequence of the growing E_{2g} elastoresistance. The growing elastoresistance may simply be a consequence of the emergence of E_{2g}-symmetric fluctuations (and not of a phase transition) or, alternatively, signal a broadening of the T^* transition that is induced by the finite anisotropic stress applied experimentally because the sample suffers from differential thermal expansion with the PZT piezoelectric.

Reply) We agree that elastoresistivity data alone cannot uniquely determine the microscopic origin of the anomaly at 64 K, and alternative scenarios must be considered. In this study, we identified three independent experimental features emerging around 64 K:

1. Enhancement of $m_{E_{2g}}$ with clear kink behaviors in both $m_{E_{2g}}$ and $m_{A_{1g}}$,
2. Emergence of in-plane resistivity anisotropy defined as $(\rho_{xx} - \rho_{yy})/(\rho_{xx} + \rho_{yy})$,
3. Onset of hysteresis in the strain–resistivity loops.

In particular, the hysteresis observed in Figs. 3b, 3c, 3f, and 3g is naturally interpreted as arising from strain-induced changes in the population of symmetry-related domains, implying the presence of a symmetry-broken phase.

Nevertheless, as pointed out by the reviewer, several alternative interpretations should be scrutinized. We have revised the manuscript to explicitly discuss the following scenarios:

(i) Long-range order.

The observations are consistent with rotational symmetry breaking. Since magnetic susceptibility shows an anomaly only at 55 K, a charge-related order remains a plausible candidate. Given that a 4×1 modulation has been reported previously and its onset temperature has not been clearly established, associating its development with ~ 64 K is reasonable. However, elastoresistivity alone cannot determine the modulation period, and other possibilities (including incommensurate order) cannot be excluded.

(ii) Short-range order.

A short-range CDW breaking rotational symmetry is another possibility. However, it would be more difficult to reconcile such a scenario with the observed hysteresis.

(iii) Fluctuation regime.

The enhancement of $m_{E_{2g}}$ below 64 K and its further increase toward 55 K may reflect a fluctuation regime. Although the temperature dependence does not follow a Curie–Weiss form such as $m_{E_{2g}} \propto 1/(T - \Theta)$, nematic fluctuations towards a possible nematic transition at 55 K cannot be excluded. In this fluctuation scenario, the anisotropy $(\rho_{xx} - \rho_{yy})/(\rho_{xx} + \rho_{yy})$ could arise from the elastoresistive response under residual strain due to differential thermal contraction between the sample and the PZT stack:

$$(\Delta\rho/\rho)_{xx} = \left(m_{A_{1g}} + m_{E_{2g}}\right)\epsilon_{xx}/2 + \left(m_{A_{1g}} - m_{E_{2g}}\right)\epsilon_{yy}/2,$$

$$(\Delta\rho/\rho)_{yy} = \left(m_{A_{1g}} - m_{E_{2g}}\right)\epsilon_{xx}/2 + \left(m_{A_{1g}} + m_{E_{2g}}\right)\epsilon_{yy}/2.$$

However, this scenario encounters difficulty when considering the results shown in Fig. R2 (Supplementary Fig. S5). Figure R2 compares the in-plane resistivity measured using the modified Montgomery method and the four-terminal method. In addition to the different measurement techniques, the crystallographic orientation relative to the piezo polling direction differs by 90 degrees between the two samples: in the modified Montgomery configuration the *a*-axis is parallel to the *y* direction, whereas in the four-terminal configuration the *a*-axis is parallel to the *x* direction.

If the system remains hexagonal between 64 K and 55 K and the anisotropy is solely due to residual strain in the laboratory *x*–*y* frame, one would expect similar temperature dependence of ρ_{xx} in both samples regardless of crystal orientation. Instead, below 64 K the temperature dependence of ρ_{xx} in the four-terminal sample resembles that of ρ_{yy} in the modified Montgomery sample rather than ρ_{xx}. This behavior is difficult to reconcile with a purely residual-strain scenario in a hexagonal phase.

Such behavior is more naturally understood if a rotational-symmetry-broken phase emerges in this temperature range. In that case, the measured resistivity reflects a combination of multi-domain contributions and intrinsic elastoresistive effects within each domain. While a fully quantitative analysis remains challenging, the comparison between the two configurations suggests that the anomaly near 64 K may not be straightforwardly attributed to a fluctuation in the hexagonal phase.

(iv) Broadening of the 55 K transition by residual strain.

As the reviewer points out, residual strain is inevitably present in the sample due to differential thermal contraction between the sample and the piezo stack. It cannot be excluded that spatial inhomogeneity of the strain experienced by the sample leads to broadening of the observed signals. Such inhomogeneity may depend on factors such as the sample shape and the thickness of the adhesive layer, and it is difficult to quantitatively evaluate these effects.

However, if the transition at T^* ~ 55 K were broadened up to around 64 K (i.e., over a temperature range of approximately ±10 K), one would expect the associated kinks or anomalies in the measured quantities to become substantially smeared. In contrast, this is not what we observe. For example, in Fig. 3e, the anisotropy $(\rho_{xx} - \rho_{yy})/(\rho_{xx} + \rho_{yy})$ shows a clear onset around 55 K. Similar well-defined features can also be identified in Figs. 3d, 3e, and 3g. These observations suggest that the anomaly near 64 K is unlikely to arise solely from broadening of the 55 K transition and that interpreting the 64 K anomaly as originating from a mechanism distinct from the T^* transition remains a reasonable scenario.

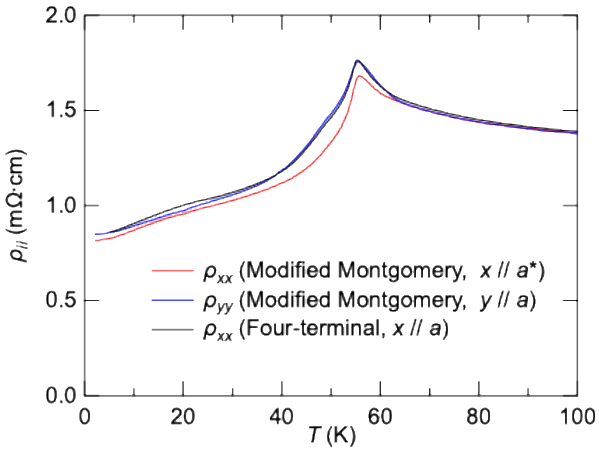

Fig. R2. Temperature dependence of in-plane resistivity measured by Modified Montgomery method and four-terminal method.

The authors also report the symmetry-preserving $m_{A_{1g}}$ elastoresistance coefficient. This is very appreciable. The data show three local extrema (Fig. 3d) around 57 K, 55 K and 45 K. One could tentatively associate these extrema to different instabilities, one being the magnetic T^* transition. Like the E_{2g} coefficient there is, however, only an enhancement of the response below 64 K and no sharp anomaly at that temperature. Again, it is then surprising to associate this temperature with a CDW transition. Because the corresponding strain is symmetry-preserving, there is in principle no reason not to expect coupling between the CDW order parameter and the $\epsilon_{xx} + \epsilon_{yy}$ strain which in turns would imply a distinct anomaly at 64 K beyond a simple enhancement.

Reply) We agree that $m_{A_{1g}}$ exhibits multiple extrema around 57 K, 55 K, and 45K. As discussed above regarding the contribution proportional to $\partial\rho/\partial T$, the positions of these extrema roughly correspond to those of extrema found in $\partial\rho/\partial T$ in Fig. R1. As the strong anomalies at 57 and 55 K in $\partial\rho/\partial T$ can appear when $\rho(T)$ has a peak at T^*, we do not consider these as signatures of two transitions. The origin of the weak anomaly at 45 K is not clear, but around this temperature the hysteresis in Fig. 1g shows a rapid decrease, suggesting that this may be related to some change in the domain scattering. We stress that there is a clear kink in the temperature dependence of elastoresistivity coefficients at 64 K, and the above discussions imply that the observed behaviors of $m_{A_{1g}}$ do not contradict the possibility of a transition near 64 K.

Thus, different interpretations are possible. A clear assessment requires additional measurements that one can't reasonably ask the authors to provide in the present manuscript. Instead, my suggestion is to expose the different scenarios at hand in the manuscript, such that the reader has all information to interpret the present study.

Reply) We thank the reviewer for this highly valuable suggestion. Following this advice, we have revised the manuscript to explicitly discuss multiple possible scenarios, as described above, so that readers can appropriately interpret the present results considering the current experimental limitations. We believe that this revision substantially improves the clarity and balance of the manuscript, and we appreciate the reviewer's constructive guidance.

Finally, I point out two details to the authors consideration:

* In some places 'uniaxial' or 'unidirectional' is incorrectly used. For instance, line 340 "under uniaxial strain" is incorrect, but instead "biaxial strain" could be used. Same thing for line 220 and "under unidirectional strain".

Reply) We thank the reviewer for pointing this out. We have corrected the wording accordingly.

* Lines 268-271 "Such temperature dependence deviates from the Curie-Weiss divergence, excluding the existence of Fe-SC type of electronic nematicity in this material". If this is what is meant, I do not agree that not observing a Curie-Weiss diverging elastoresistance excludes electronic nematicity. Rather, it is surprising that the experimental situation is 'so' simple in the iron-based SCs. Even for a true electronic nematic instability a different temperature dependence of the 'nematic' elastoresistance can be observed if the electron-phonon coupling has a distinct temperature dependence. Alone a non-Curie-Weiss dependence alone is not a proof of the absence of electronic nematicity.

Reply) We thank the reviewer for this important clarification. We agree that greater caution is required regarding the presence or absence of nematic order. We have revised the manuscript to explicitly mention that the enhancement of $m_{E_{2g}}$ below 64 K may be interpreted in terms of fluctuations, as discussed above.

Reviewer #2 (Remarks to the Author):

Onishi and coworkers report on a study on the Cr Kagome homologue of the AV_3Sb_5 Kagome family, which is indeed gaining a rapidly strong interest owing to its different intriguing properties contrasting the V-based Kagome. At ambient pressure, so far only one phase transition featuring magnetic and structural properties at ~55 K had been reported. In their study, the authors could show two different distinct phase transitions at two different temperatures. The one already reported in the literature at 55K which they attribute to a spin density wave phase (SDW) and another at a distinct higher temperature at around 64K which they appoint to a charge density wave (CDW) phase. By using linear dichroism asymmetry in laser-assisted photoemission microscopy, they could demonstrate the rotational symmetry breaking at the SDW onset temperature. Indeed, they showed three distinct domains obeying a two-fold symmetry thus breaking the original six-fold symmetry. Moreover, by using elastoresistivity measurements and anisotropic strain-resistivity measurements, they could show two distinct phase transitions which they ascribe to separate CDW and SDW phase transitions. The authors argue that the phase transition at 55K should be ascribed to the SDW given that previous magnetic susceptibility measurements show an anomaly at the same transition temperature. Based on the possible electronic nesting nature of the 1x4 reported CDW away from the gamma, they assign the higher temperature transition to the charge order transition.

The manuscript and data presented by the authors are clear and well consistent with each other. While the observation of domain competition has been reported before (ref. 41), the imaging of these domain structure and the observation of an additional phase transition provide important new insight for understanding this novel material. Despite the limited but growing number of studies on this material and the current theoretical agenda, the theoretical claims and scenarios suggested by the authors are reasonable and offer new directions to explore in order to tap on these claims. The paper could be suitable for publication in Nature Communications once a few issues are addressed:

Reply) We sincerely thank the reviewer for the careful reading of our manuscript and for the constructive and encouraging comments. We appreciate the positive assessment of the clarity and consistency of the data, as well as the thoughtful questions that helped us improve the manuscript. Below, we respond to each point in detail.

1) The authors argue that the 4x1 CDW sets in at 64 K, and that the SDW transition below 55 K is of 2x1 symmetry. Yet, superlattice peaks of 4x1 symmetry have been observed below 55 K (ref. 24, 31), but not above. This appears inconsistent with the claim of the authors, and needs to be addressed. In particular, the authors argue that e-ph coupling might be too weak to allow observation of the CDW above 55 K. But why is the observed period of the superstructure then 4x1 instead of 2x1 below 55 K?

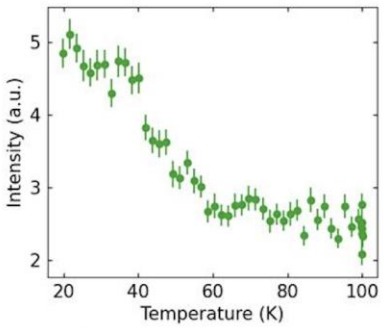

Fig. R3. Temperature dependence of X-ray diffraction intensity at (3, 0.25, 0) (Reprinted from ref. 31 by courtesy of W. Yao and P. Dai).

Reply) Regarding consistency with previous XRD studies, in Y. Liu *et al*. (ref. 24), 4×1 superlattice peaks were reported to be present at 50 K and absent at 70 K. However, the detailed temperature dependence between these values was not resolved. Therefore, whether the onset occurs at 55 K or at a higher temperature such as ~ 64 K remains experimentally undetermined in that work. In W. Yao *et al*. (ref. 31), the temperature dependence of the (3, 0.25, 0) peak intensity appears to show an onset around ~ 60 K (see Fig. R3), but it is not clearly established whether the true transition corresponds to 55 K or 64 K. Therefore, the existing XRD data do not strictly exclude a scenario in which a 4×1 CDW begins to develop near 64 K. Below 55 K the magnetic order has the 2x1 structure, but to detect this magnetic scattering is required and the structural XRD detect the 4x1 CDW order.

2) What evidence besides the susceptibility and suggested nesting vectors do the authors have for the assignment of SDW and CDW transitions?

Reply) In a rotational-symmetry-broken phase, the presence of multiple domains further complicates the situation, making it difficult to microscopically predict the relationship between the nesting vector and susceptibility (including the elastoresistivity coefficients). In the present study, our assignment of T_{CDW} ~ 64 K is based on a qualitative and consistent picture that considers previous XRD results, magnetic susceptibility and NMR measurements, as well as our PEEM observations.

At the same time, as pointed out in the previous comments, the available experimental evidence remains limited. We have therefore revised the manuscript to explicitly discuss alternative interpretations, including possibilities other than a CDW scenario.

3) The hysteresis loops of the elastoresistivity curves are not centered around zero. Does this originate from the residual strain the authors discuss?

Reply) We thank the reviewer for pointing this out. The offset of the elastoresistivity loops from zero strain is not due to residual strain, but simply because the applied voltage was swept asymmetrically from −30 V to 150 V, reflecting the

compliance characteristics of the piezo stack. As described in the Methods section, zero strain is defined at zero applied voltage in our measurements. We have revised the manuscript to clarify this point in the Methods.

4) What influence on the observed phase transitions does this residual strain has? Can the authors exclude with certainty that this residual strain is responsible for inducing the additional transition observed at 64 K?

Reply) In our elastoresistivity measurements, residual strain is inevitably present due to the difference in thermal contraction between the sample and the piezo stack. If the transition observed at 64 K were influenced by residual strain, two possibilities may be considered. One is that an unknown phase absent at zero strain is induced by strain. The other is that a phase transition already present at zero strain and its transition temperature shifts under strain. Although it is difficult to completely exclude these possibilities based solely on the elastoresistivity data, we consider our interpretation in terms of previously reported CDW and SDW transitions to be reasonable rather than invoking an entirely unknown strain-induced phase. Since this issue is important, we have revised the manuscript to explicitly discuss these possibilities.

As a further comment on the second scenario, where the transition temperature shifts under strain, we consider the possibility that the transition at 55 K under zero strain is shifted to 64 K due to residual strain. In this case, the transition temperature would change by nearly 10 K, implying a very strong strain sensitivity.

Although the temperature dependence of the thermal expansion of $CsCr_3Sb_5$ has not been reported so far, we estimate the magnitude of $\Delta L/L_{300\,\mathrm{K}}$ to be the order of the low 10^{-3} considering the isostructural CsV_3Sb_5 (ref. 15). Because the piezo stack has a negative thermal contraction along the polling direction on the order of $+10^{-3}$, the residual strain is expected to be on the order of low 10^{-3}.

If a transition temperature shift of approximately 10 K were caused by such strain, a simple estimate suggests that a strain of order 10^{-4} would change the transition temperature by nearly 1 K. This is a scale that can be swept by applying voltage to the piezo stack. Therefore, if such strong strain dependence existed, one would expect that sweeping strain near 64 K would drive the system to cross the phase boundary, resulting in a noticeable change in slope within each single elastoresistivity curve. However, as shown in Fig. R4 (Supplementary Fig. S6), no such behavior is observed in the experimental data. While the detailed strain dependence of the transition temperatures at 55 K and 64 K remains unknown,

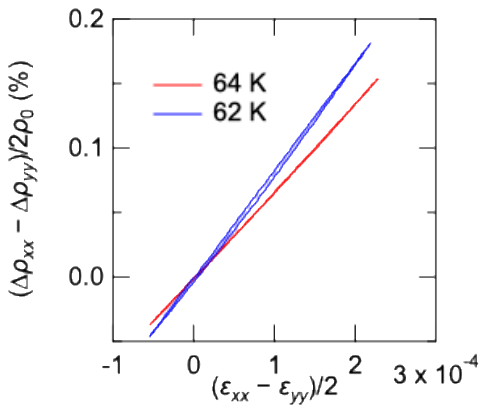

Fig. R4. Relative changes in the resistivity as a function of strain in the E_{2g} channel (modified Montgomery sample).

the results suggest that the strain sensitivity is unlikely to be large enough for residual strain to shift the 55 K transition all the way to 64 K.

5) In the supplement, section C, the authors discuss the photoemission matrix elements to originate from electron-phonon coupling. I believe this should read electron-photon coupling.

Reply) We thank the reviewer for pointing this out. We have corrected "electron–phonon coupling" to "electron–photon coupling" in the Supplementary Information III.

6) The authors discuss the derivation of the photoemission matrix elements in the limit of non-interacting electrons. However, they argue that $CsCr_3Sb_5$ shows strong electronic correlations. This appears inconsistent to me. What influence would the consideration of correlation effects have on their derivation?

Reply) We thank the reviewer for this important comment. The expectation of strong correlations in $CsCr_3Sb_5$ indeed implies that the removing of an electron may strongly perturb the system. Contrary to the non-interacting limit, it means that Ψ_i^{N-1} is not an eigen state of the (N-1) system. The overlap of Ψ_i^{N-1} with multiples eigen state of the (N-1) system would then lead to the observation of satellites in the photoemission spectra [Supplementary Ref. 4]. However, it does not affect the symmetry of the measured electronic state. In the Supplementary Section III, since our goal is to clarify the symmetry constraints on the polarization dependence of the photoemission matrix elements, we can overlook such effect of strong correlations on the photoemission spectra and simplify the derivation within the non-interacting picture. We added this clarification to the Supplementary Section III.

7) In Fig. 1b, the a axis appears twice.

Reply) We thank the reviewer for pointing this out. We have corrected the notation.

8) In Fig. 1c, the reciprocal axes are labelled q1, q2, but in the text they are referred to as a*.

Reply) We thank the reviewer for pointing this out. We have corrected the notation.

Reviewer #3 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reply) We thank the co-reviewer for their time and effort in evaluating our manuscript and appreciate their contribution to the review process.

Reviewer #4 (Remarks to the Author):

This is a well-written manuscript that describes evidence for a phase transition at ~ 64 K in single crystals of the kagome metal $CsCr_3Sb_5$. This identification relies on detailed analysis of linear dichroism microscopy and elastoresistivity measurements as functions of temperature.

The data at ambient pressure suggests this transition to be associated with a charge density wave and Fermi surface nesting around the M point of the Brillouin zone. In contrast, the authors identify the previously observed transition (~55 K) as associated with a spin-density wave and nesting centered at the Gamma point. Although the 55 K transition had been previously reported, and an additional feature at slightly higher *T* was seen under pressure, the current identification is well justified and would merit publication.

Reply) We sincerely thank the reviewer for the positive and encouraging assessment of our work. We are pleased that the identification of the ~64 K transition and its distinction from the previously reported ~ 55 K transition are viewed as well justified. We appreciate the reviewer's thoughtful evaluation and support for publication.

The work is timely, given the current interest in kagome metals. However, the current version of the manuscript would need clarification on several essential points, as follows:

1. The authors argue that the transition at 64 K deviates from the Curie-Weiss divergence, excluding electronic nematicity. However, the rotational symmetry breaking in the resistivity measurements could also be consistent with a nematic order. Moreover, kagome metals are known to support bond-density-wave orders, which may also lead to resistivity anisotropy. It would be helpful if the authors could discuss this issue and/or present more direct evidence for a genuine CDW.

Reply) We thank the reviewer for raising the possibility of nematic order and related scenarios.
As discussed in our response to Reviewer #1, although several features observed in the elastoresistivity measurements suggest the possibility of a transition at T_{CDW} ~ 64 K, the available experimental results remain limited, and it is important to consider alternative interpretations. In the revised manuscript, we have therefore expanded the discussion to explicitly address possibilities other than a CDW, including nematic order. We believe that this revision provides a more balanced interpretation of the present results.

2. What are the effects of disorder/impurities in the sample when measuring the elastoresistivity coefficients in Fig. 3, especially for temperatures between the two transition temperatures? The authors comment on the rounding of the coefficients near the transition as indicative of second-order behavior, but a similar effect would likely arise from inhomogeneities. It would be important for the authors to discuss the effects of sample quality and reproducibility when measuring multiple samples.

Reply) We thank the reviewer for this important comment. As pointed out, it is necessary to address sample quality and reproducibility across multiple samples.

As shown in Fig. R2 (Supplementary Fig. S5), the temperature dependence of the in-plane resistivity measured using the modified Montgomery method and the four-terminal method is similar for the two different samples used in this study. In both cases, the residual resistivity ratio $R_{100\,\mathrm{K}}/R_{0\,\mathrm{K}}$ is approximately 2.3. Although it is difficult to completely exclude possible effects of disorder or impurities, these results indicate that the two samples have comparable quality. Furthermore, the obtained $R_{100\,\mathrm{K}}/R_{0\,\mathrm{K}}$ value is comparable to or slightly higher than that reported in previous work (the original Y. Liu *et al*., where $R_{100\,\mathrm{K}}/R_{0\,\mathrm{K}} \sim 1.7$). We have revised the manuscript to clarify these points and to explicitly discuss the potential influence of sample quality.

3. In Figs. 3d, e & g, there is a clear non-monotonic feature in the curves at around 8 K. Can the authors comment on such a deviation? Could it be associated with another kind of transition? An artifact of the measurement?

Reply) As noted, a change in behavior below approximately 10 K is observed not only in $m_{E_{2g}}$ and $(\rho_{yy} - \rho_{xx})/(\rho_{yy} + \rho_{xx})$ measured using the modified Montgomery method, but also in $d(\Delta\rho_{xx}/\rho_{xx})/d\epsilon_{xx}$ measured using the four-terminal method. The fact that this feature appears in two different samples measured with different configurations suggests that it is unlikely to be a measurement artifact. At present, however, we do not have sufficient evidence to associate this feature with a specific phase transition. It appears to be distinct from the CDW and SDW transitions considering the discussion in this work. While we refrain from further speculation at this stage, this low-temperature feature may provide an interesting direction for future investigations.

4. In Fig. 2f. the authors show a real space map of the angle theta_fit, which reveals domains with three preferred orientations. The authors interpret these domains as distinct density modulations at the three Cr sites, consistent with the breaking of C6 symmetry down to C2. Could the formation of such domains be associated with local uniaxial strain formation, which locally favors one orientation over others?

Reply) As a general property of phases that break rotational symmetry, the formation of domains may indeed be influenced by local strain, which could locally favor one orientation over the others. Therefore, it is possible that the observed domains are related, at least in part, to local strain effects. However, establishing a direct relationship between the domain formation and local strain distribution would require additional spatially resolved strain measurements, which are beyond the scope of the present study. We consider this an interesting direction for future investigations.

The evidence and discussion in the text are measured and appropriately non-conclusive. However, the tone of the abstract is significantly more definitive; I recommend revising it to better align with the main text.

Reply) We thank the reviewer for the positive assessment of the main text and for the helpful suggestion regarding the abstract. We have revised the abstract to better align with the more measured and non-conclusive tone of the discussion in the main text.

Reviewer #5 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reply) We thank the co-reviewer for their time and effort in evaluating our manuscript and appreciate their contribution to the review process.