Peer Review File

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Manuscript Title: Discovery of charge density wave in a kagome lattice antiferromagnet

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:

Referee #1 (Remarks to the Author):

In this manuscript, Xiaokun Teng et al. report a combined study of magnetization, transport, neutron scattering, STM and ARPES on a Kagome antiferromagnet, FeGe. The authors demonstrate the short-range CDW, antiferromagnetic order, and van Hove singularities in FeGe. The authors also show that the CDW order is coupled with the magnetic order in this material. I agree that the authors show CDW order in the non-superconducting Kagome antiferromagnet, FeGe. However, in comparison with the superconducting AV3Sb5 materials, I didn't find there is enough solid and new physics, and I am not convinced this manuscript deserves publication in Nature. Below are my detailed comments.

1. The authors think that correlation in FeGe is due to the flat band. Why no flat band is observed in their ARPES and dI/dV spectrum data?
2. The CDW in AV3Sb5 is usually long-range ordered. Why the CDW in FeGe is short-range ordered?
3. How can the author exclude that the CDW is not induced by the phonon softening in FeGe?
4. Why the CDW order can not be seen in the STM topography, but only appears in the dI/dV map? Can the authors see the CDW in the STM topography taken with low-bias voltages?
5.In the ARPES data, the authors showed several van Hove Singularities. However, why no clear van Hover Singularity features can be seen in the dI/dV spectrum shown in the inset of Fig. 3g?
6. Can the authors also detect the CDW gap in the ARPES measurements?
7. The authors use the chiral flux phase of the circulating currents to explain the phenomena in FeGe. Since the STM experiments can be directly performed on the Kagome layer, is there any direct evidence for the circulating currents in the Kagome layer?
8. Line 127, it should be "by the white square in Fig. 2f….."

Referee #2 (Remarks to the Author):

I have read the manuscript by Teng et al. with much interest. The authors report the coexistence and interplay between antiferromagnetism and charge-density-waves in a kagome metal.

The material studied here, FeGe, has clearly a rich physics, which the authors have studied using a suite of advanced probes. The collection of data and information on this system is impressive, and experiments have clearly been performed to high standards and in a very comprehensive fashion. As a result, the study encloses a wealth of information about the electronic and magnetic structure of this system, which enabled the authors to draw connections between the fermiology and the symmetry-broken phases which include a combination of charge and magnetic order.

I believe the results are of broad relevance for the quantum matter community, and as such I am supportive of this manuscript being featured in Nature. I find that the observed phenomenology is sufficiently unique and exciting as to meet the novelty criteria of this journal (and agree with the authors' closing statement that "Regardless of the theoretical explanation of these experimental results, we have identified a kagome metal system in FeGe where strong electron correlations lead to a rich interplay of magnetic order, topology, CDW, and AHE"). At the same time, the findings presented here are interpreted within a framework which is not fully supported by the available evidence.

The main issue is the interpretation of the anomalous Hall effect (AHE) as arising from a chiral flux phase that is closely associated to a time-reversal-symmetry breaking charge-density-wave. If I understand correctly, the Hall data indicate that below the spin-flop field, the Hall resistivity is linearly proportional B and therefore there is no AHE, while above the spin-flop field, the linear behavior extrapolates to a finite value at zero field, suggesting the presence of an AHE arising from a field-induced ferromagnetic moment which is not ascribed to the canted magnetic moments but rather to a magnetic flux generated by a chiral CDW. This behavior arises only below the CDW transition; therefore, the authors conclude that the CDW must be in some way involved. The interpretation proposed by the authors is then that there are two chiral CDWs in neighboring planes whose opposite fluxes are perfectly compensated at low fields and remain compensated up to a spin-flop field of several Teslas, even though the magnetic moments are progressively tilted away from the c-axis (making the AFM order slightly uncompensated as reflected in the increasing magnetic moment shown in Extended Data Figure 2c). Then, at the spin-flop field, this balance between the CDW fluxes is suddenly removed, and there is a net flux that develops along the c-axis, breaking time-reversal symmetry and generating an AHE. While this interpretation is somewhat plausible, it rests on a series of hypothesis which are not experimentally validated, first and foremost the time-reversal-symmetry breaking nature of the CDW, which is not assessed directly (for instance, by means of a comparison between spin-flip and non-spin-flip neutron diffraction measurements). Furthermore, the susceptibility data as a function of external magnetic field display a shoulder before the spin-flop field, an effect which becomes the more prominent the lower the temperature (i.e. deeper into the CDW phase). This enhanced susceptibility seems to indicate the rise of the c-axis magnetization ahead of the transition, yet the crossover in the Hall resistivity seems to be very sharp, occurring precisely at the spin-flop field. How is such a behavior explained by a chiral flux phase which is only activated in the spin-flop phase?

While the one above is the major item that caught my attention, I made a few extra notes about some passages that I could not entirely understand and I was hoping the authors could clarify:

- Top of page 3: "There is not yet an example of a coupled CDW and magnetic order that emerges within an established magnetic phase where the CDW appears as a consequence of magnetism.". The authors should clarify what they mean by "CDW appears as a consequence of magnetism". There is no doubt that these two orders are coupled, as chiefly demonstrated by the modification to the ordered moment that occurs as the CDW develops. However, what hard evidence do the authors have that the CDW is a consequence of magnetism? One could perhaps say the two have a common origin in the fermiology (AFM order from a flat band instability and CDW from a van Hove singularity as proposed here) but I do not see a direct link, or perhaps I missed this while reading the manuscript.

- Middle of page 3: “When flat bands are near the Fermi level, EF, strong electron correlations can induce magnetic order, as has been found in kagome lattice FeSn.”. To the best of my understanding, this scenario is still the subject of speculations. That the origin of AFM order is a Stoner instability created by the flat bands has not been confirmed experimentally, nor theoretically. Some recent studies have proposed such a connection, but the evidence remains inconclusive (primarily because it is hard to ‘catch’ a flat band in the act of creating an instability, and DFT-LDA is known to have severe limitations in describing the effect of exchange fields and consequently the position of spin-polarized bands – in particular flat bands)

- Top of page 4: “Since AFM order in FeGe likely arises from electronic correlations of the flat electronic bands near the Fermi level…”. This is related to the previous point, but here this hypothesis (that the flat bands drive AFM order) is applied to FeGe. Are there previous theoretical works supporting this scenario? Note that the works cited here do not refer to FeGe but to other compounds (some of which, like CoSn, do not exhibit magnetic order).

- Bottom of page 8: “one can consider a nesting scenario where vHS2 at M below EF nests with vHS3 at L above EF…”. While the presence of a vHS near the Fermi energy is a telltale of a possible strongly nested Fermi surface in hexagonal systems, the ultimate assessment for such a scenario comes from the shape of the Fermi surface itself. If nesting conditions are present, one expects nearly flat Fermi contours connecting the vHS at the M-points, as observed in some of the AV3Sb5 compounds. It would be helpful if the authors could show a non-projected version of the Fermi surface in 4a and discuss how close the Fermi contours are to the ideal nesting conditions.

- Middle of page 9: “The energy splitting of the majority and minority bands increases with decreasing temperature and shifts the electronic bands such that the nesting of vHSs in Fig. 4a to form CDW becomes possible.”. This is an interesting possibility which can be tested theoretically via spin-polarized LDA+U calculations and experimentally by performing ARPES measurements at different temperatures (say between T_CDW and room temperature). Without the support of either pieces of evidence, however, this scenario remains purely hypothetical and the interpretation of the role of AFM order-induced Zeeman splitting is not conclusive (although somewhat plausible). I think a clarification of this aspect will be particularly important.

- Extended Data Figure 3. It would be very helpful if the authors could show the Hall resistivity data down to low temperature in panel c, where the AHE signal deviates significantly from zero. Additionally, I was expecting to see signatures of the change in slope of rho_xy vs B at the spin-flop field in panel e, but this is only visible for the large jump that takes place for the AFM phase. Could the authors show an additional panel that focuses on the AFM+CDW region, using a color map that makes the transition in rho_xy more apparent?

In sum, I very much enjoyed reading this manuscript and I thought the experimental information is remarkably extensive, which has taught me a lot of interesting things about this new material. This kagome compound is clearly a very exciting, new playground to explore the interplay between correlation phenomena from a different angle, especially when it comes to the CDW/AFM interplay. At the same time, I cannot recommend this paper for publication in its current form for the reasons I outlined above, but I hope the authors can address my questions and, most importantly, provide new information in support of their proposed interpretation of the origin of the AHE.

Referee #3 (Remarks to the Author):

In this manuscript, the authors report an intriguing set of experimental observations in their hexagonal FeGe single crystals. The central results are about a new phase below T ~ 100K that has pronounced signatures in physical properties (magnetic susceptibility, electric transport, and specific heat). Using single-crystal neutron diffraction, the authors attribute the phase to a charge-density-wave (CDW) order, whose propagating wave vectors are also seen with Fourier-transform scanning tunneling spectroscopy. A highlight is that the wave vectors are the same as the CDW wave vectors in the AV3Sb5 family, which shares the same layered kagome crystal structure as hexagonal FeGe and is currently under intense research. Based on this commonality, the authors argue that peculiarities of electronic structure and correlations in kagome metals may be at the origin of the intriguing physical properties of FeGe, in particular what they call an emergent anomalous Hall effect (AHE), which has also been claimed in the AV3Sb5 family. Finally, since the CDW wave vectors appear to connect van Hove singularities in FeGe that presumably follow from antiferromagnetic (AFM) order formation below 410K, it is suggested that the CDW is strongly coupled to the AFM order, which makes it unique among other CDW examples in correlated metals.

To my knowledge, the 100K transition has not been reported in hexagonal FeGe. The manuscript is well-written with a broad vision. Conceptual connections (and contrasts) are made to the spin-charge stripes in high-Tc cuprate superconductors, and especially to a topical 'chiral flux phase' in AV3Sb5, which features time-reversal symmetry breaking due to inter-atomic circulating currents (similar physics has been proposed for explaining the pseudogap phase of high-Tc cuprates). A charge order with emergent orbital magnetization may indeed produce magnetoelectric transport consequences such as AHE, so the whole story sounds physically plausible, and can be anticipated to intrigue several major research communities. The collaboration also involves well-known research groups. With such a story, the manuscript will almost certainly make an impact.

These merits of the manuscript could warrant publication in Nature, given that the experiments and interpretation are technically sound. But this is where I see major issues, which occur at different levels. I try to indicate below how the issues might be overcome if the authors firmly believe in what they claim.

1. There is a stark discrepancy between the current study and a previous one in their experimental magnetometry data: between Fig.1f of the present manuscript, and Fig.2 of Beckman et al., Physica Scripta. Vol. 6, 151-151 (1972). The latter data were re-plotted in Ref.[24], yet upon citing this reference and discussing the result in Fig.1f, the authors did not mention the discrepancy. The issue

cannot be overlooked. Hexagonal FeGe was first studied at low T and in B fields over half a century ago, quite carefully indeed by researchers in Sweden including M. Richardson, who first reported the growth method [48] used by the present study. Richardson was on the 1972 paper mentioned above, and the most important feature around T=100K (of χ_{perp}) in the present Fig.1f is completely absent in the previous data. Importantly, all the other features seem to correspond. So, the discrepancy concerning the 100K anomaly must be due to a difference in either the purity of samples, or the way how the measurements were done.

If it is related to sample quality, I am afraid that the present samples are problematic, e.g., contaminated by impurity phases that can inter-grow with hexagonal FeGe. Some candidates can be found in these recent papers: [doi:10.1016/j.jssc.2018.10.030], [doi:10.1039/d0dt03923c], and some of them even have a magnetic transition near 100K. I should say that I still do not consider impurity to be the likely explanation, since all other aspects of the present data set (especially specific heat) are in support of the 100K transition being genuine to hexagonal FeGe. But, in case there is a problem with the samples, essentially all conclusions of the present manuscript would be invalid.

To show that the above discrepancy is not related to sample quality, the authors should try to reproduce the lack-of-100K-anomaly χ_{perp} data previously reported by Beckman et al. There is one possible way out: while the c-axis is unique, there are still two high-symmetry choices for the in-plane field direction that are mutually orthogonal. The authors should precisely define their in-plane field direction, and I am guessing that another in-plane direction would produce the result of Beckman et al., i.e., no anomaly at 100K. And if that turns out true, this statement in the manuscript is false: "the reduction of susceptibility is consistent with gapping density of states at the Fermi level by the CDW." It brings me to my next criticism, namely, if the transition at 100K leaves a highly anisotropic signature in the magnetic susceptibility, it is more likely to be magnetic than CDW.

2. I am unconvinced by the "CDW" interpretation of the 100K order even if it is genuine to FeGe. The key support for the CDW interpretation is this sentence: "Since these peaks persist to large Q-positions in higher zones where the Fe2+ magnetic form factor drops off dramatically, they must be nonmagnetic and associated with lattice distortions induced by a CDW or lower crystalline symmetry of unknown origin (Figs. 2f, 2k)."

I do believe that the H,K=integer, L=half-integer diffraction peaks (as shown in Fig.2e) are magnetic. However, those peaks are clearly still present in the "higher zones", which shows that the Fe2+ form factor alone is not enough to fully suppress magnetic scattering in those zones. Extended Data Fig.6g tells the same: neglecting the strong peaks around Q=3-4A^-1, it is not obvious at all that the magnetic intensity drops strongly up to Q=8A^-1. It means that due to the crystal structure, the form factor does not predominantly determine magnetic peak intensities, and that the structure factor also matters a lot. While the "CDW" peaks in Extended Data Fig.6h show an increasing trend towards larger Q, the overall intensities are weaker than the magnetic peaks in Extended Data Fig.6g. In other words, there is still 'room' for the new peaks below 100K to be magnetic.

The array of "CDW" peaks in Fig.2f actually have peculiar behaviors. As an example, I consider H=half-integer peaks in rows of the same integer L. At L=0 we essentially see no intensity, which implies that atoms move only in the c direction upon entering the "CDW" phase. The L=-1, -2, -3, and

-4 rows of (H half-integer) peaks are common in that, in each row, the intensities monotonically *decrease* towards higher H. I cannot easily understand this by CDW, but it would be natural if the underlying order is actually magnetic. Both the form factor and the "S_perpQ" spin-polarization factor (by virtue of the magnetic scattering intensity being zero at L=0) would be able to explain the decrease in each row with increasing H.

The bottom line here is that the magnetic form factor argument is weak for proving a CDW origin. The STS data do not help much either, because when the system is already ordered magnetically for in-plane q=0 (below 410K), having an extra magnetic order at H or K=0.5 will lead to a second-order effect that looks like a (weak) charge order: the product of two time-reversal-odd order parameters is time-reversal-even (as a CDW should be). To make the CDW case stronger, the authors should consider improving their experimental evidence:

1) A displacive model calculation can be done for the "CDW" neutron diffraction intensities (it does not have to be a full refinement of the distorted structure), to see how much of a (2x2) lattice distortion is needed to produce observed signal magnitudes. As explained above, I expect the atomic displacements to be along c, which should strongly affect tunneling currents in STM measurements. With that estimate, they would hopefully be able to find corresponding signatures with an STM tip (distance to top atoms of the same element).

2) A non-resonant X-ray diffraction experiment should be ideal for revealing a CDW order. The authors seem to have in-house access to a variable-T X-ray diffractometer, and should attempt such measurements below 100K.

3) A spin-polarized neutron diffraction measurement of several of the "CDW" peaks should show that they are all non-spin-flip and hence due to coherent nuclear scattering. Or, the authors can perform unpolarized neutron diffraction up to much higher Q, where no magnetic peaks are seen above 100K. If they are able to show that the peaks at half-integer H,K become visible below 100K at those high Q, it would be strong evidence for CDW.

3. I believe that the AHE interpretation is problematic. The AHE magnitude is extracted from the linear extrapolation of high-field \rho_{xy} data back to zero field for taking the intercept. In Fig.1k, the 180K data indeed look like a spin-flop transition, namely, the Hall coefficient changes abruptly across the transition, resulting in \rho_{xy} vs. B linear relationships on both sides of the transition. As both lines would go through the origin (under extrapolation to B=0), a jump must occur at the transition field.

However, the 10K data in Fig.1k show no jump, but only a change in slope around 7T. One might still argue that it is because the AHE's non-zero intercept cancels the spin-flop jump, but the argument is based on a coincidence. With curiosity I found additional data plotted in Extended Data Fig.3d-e (I am thankful that the authors are careful with showing raw data). To me, the jump disappears immediately below "T_CDW", which would mean that the coincidental cancellation holds true at every temperature below "T_CDW". That is very unlikely, and I think a more plausible explanation is that there is no AHE of the claimed type at all, and that the transition at 100K is of a magnetic origin. In particular, I suspect that the "spin-flop" transition becomes something else below "T_CDW" - I see a discontinuity in the magnetization data at "T_CDW" near 7.5T in Fig.1i. That would explain why no jump in \rho_{xy} is observed below ~100K. An investigation of the field-hysteresis behavior for temperatures both above and below 100K might be useful in this regard.

4. I find several seemingly important statements which I cannot agree as they are phrased at present. They do not necessarily affect the validity of the whole paper but should still be revised.

(Page 7) "Therefore, while the CDW order clearly enhances the ordered moment of the collinear A-type AFM phase, we do not observe any effect of the CDW on the formation of the canted AFM structure."

The canted AFM structure develops at much lower temperatures than the CDW. In other words, there is no experimental comparison between canted AFM with/without CDW. How do the authors come to this conclusion?

(Page 9) "The energy splitting of the majority and minority bands increases with decreasing temperature and shifts the electronic bands such that the nesting of vHSs in Fig. 4a to form CDW becomes possible."

I think such nesting would be spin-flip nesting, and more likely to (if not can only) promote spin- rather than charge-density waves. It is another reason to suspect that the order below 100K is magnetic.

(Page 10) "Importantly, we observe no AHE in FeGe in the low field regime even from the circulating currents. This indicates that the circulating currents are locked to the ordered moments in the low field range, clearly different from AV3Sb5 where a small field already induces an AHE without magnetic order."

A net circulating-current magnetization at high fields could at most be an add-on response to the spin part of the magnetization, which could enhance the Hall signal, and when the spin magnetization goes to zero (extrapolation to zero field from the spin-flopped phase), the circulating-current magnetization also goes to zero. I hence do not think the authors have managed to explain the origin of AHE, even if I were to fully agree with their cartoon in the last panel of Fig.1.

Calling such an effect "AHE" is potentially misleading, because AHE in the strict sense implies spontaneous breaking of time-reversal symmetry. Here, the time-reversal symmetry is in the first place broken by the applied c-axis field, i.e., the observed effect is a non-linear Hall effect rather than AHE. I understand that the terminology was (problematically) introduced first in some of the AV3Sb5 studies and the current authors are not the ones to blame, but if their work becomes published in a prestigious journal like Nature, this is a good opportunity to help correct misconceptions in some communities especially for the non-specialists.

5. (Minor) I was confused by some (likely) typos:

Fig.1k caption, \rho_xx should be \rho_xy

Text page 4, a reference to Fig. 2d should be Fig. 2c?

Text page 5, "... reminiscent of the out-of-plane magnetic susceptibility (Fig.1i) ..." I see nothing similar, the 2-4T part of Fig.1i is completely white in color. Perhaps they mean in-plane instead of out-of-plane, and Fig.1f instead of Fig.1i?

(There could be more as I did not note all of them.)

In summary, the claims of the manuscript are novel and of broad interest. But I have concerns about the soundness of the experimental data and interpretation, and cannot recommend the work for publication. If, after further investigations, the authors conclude that the order below 100K is genuine to hexagonal FeGe, but it is something other than CDW, or that the AHE interpretation has to be altered, the finding could still be considered a nice discovery (a new order in a half-century old kagome metal is still exciting). Such result may warrant publication in a less selective journal.

Referee #4 (Remarks to the Author):

In this paper, the authors report the coexistence of CDW and antiferromagnetic phase. The key point here is the COUPLING between CDW and magnetic order, which had never been reported in any material system. If this could be confirmed, the paper would definitely deserve to be published in Nature. However, based on the results in the paper, I can only confirm the coexistence of CDW and antiferromagnetic phase. The coupling between CDW and magnetic order cannot be demonstrated by the current experimental results. If the evidence of coupling could be provided, I would support the publication in Nature.

Figure 1 shows the transport and magnetic measurement results, which clearly demonstrates that something happens at T-CDW. Figure 2 and Figure 3 show the neutron scattering and STM data, which clearly demonstrate lattice distortion at T-CDW and the CDW gap. The ARPES data in Figure 4 clearly shows the vHSs in this material, which provides importance information for understanding the coexistence of CDW and AFM in this material. These experimental results convincingly confirm a CDW phase in the AFM system. However, to confirm the coupling, the authors need to show the CDW state can be tuned by manipulating the magnetic states, and vice versa.

One more minor point, what generates the peak at ~ 20 K in the magnetic susceptibility vs T curve in figure 1f? Is it due to the magnetic frustration?

Author Rebuttals to Initial Comments:

Replies to comments made by the referees

Referee #1

"In this manuscript, Xiaokun Teng et al. report a combined study of magnetization, transport, neutron scattering, STM and ARPES on a Kagome antiferromagnet, FeGe. The authors demonstrate the short-range CDW, antiferromagnetic order, and van Hove singularities in FeGe. The authors also show that the CDW order is coupled with the magnetic order in this material. I agree that the authors show CDW order in the non-superconducting Kagome antiferromagnet, FeGe. However, in comparison with the superconducting AV3Sb5 materials, I didn't find there is enough solid and new physics, and I am not convinced this manuscript deserves publication in Nature. Below are my detailed comments."

Reply: We appreciate these comments from the referee, especially for the recognition that we have shown that the CDW order is coupled with the magnetic order. As we have clearly stated in the paper, we are not aware of any Kagome lattice material where there is an interplay between magnetic order and charge order. As far as we know, there has not been any charge order found in any other magnetically ordered Kagome lattice materials. The AV_3Sb_5 family of materials also does not exhibit any explicit magnetism. Therefore, we respectfully disagree that our work does not have "enough solid and new physics". We have revised our manuscript based on the suggestions by all the referees and hope that the referee can appreciate better the importance of the present work.

"1. The authors think that correlation in FeGe is due to the flat band. Why no flat band is observed in their ARPES and dI/dV spectrum data?"

Reply: We thank the referee for this question. In our detailed ARPES work on this material, flat bands across the Brillouin zone have indeed been observed as shown below. This will be reported separately. STM on the other hand is not a decisive way to determine flat band, as flat band is defined in momentum space.

Redacted Figure + Text

"2. The CDW in AV3Sb5 is usually long-range ordered. Why the CDW in FeGe is short-range ordered?"

Reply: The CDW order is potentially due to nesting of van Hove singularities (vHs). As shown in our temperature dependence of the ARPES data overlaid with the formation of the magnetic moment (m^2), the flat band and vHs in FeGe are changing as a function of temperature. Specifically, the observed portion of the flat band is shifting away from E_F while the vHS is shifting towards E_F as temperature is lowered between AFM and CDW ordering temperatures. If this interpretation is correct, the imperfect nesting condition of vHs occurring near 100 K may be the reason for the formation of short range CDW order. Regardless the microscopic origin of the short-range CDW order, we would argue that this experimental fact further suggests that this system is interesting and different from that of AV3Sb5.

Redacted Figure + Text

[redacted]

"3. How can the author exclude that the CDW is not induced by the phonon softening in FeGe?"

Reply: We have carried out inelastic neutron scattering experiment to study spin waves and phonons on the systems. From our preliminary data above and below CDW temperature shown below, we find no evidence of an acoustic phonon softening, similar to AV_3Sb_5. However, inelastic neutron scattering data is beyond the scope of present discovery paper and will be published after detailed analysis is carried out in the future.

[redacted]

“4. Why the CDW order can not be seen in the STM topography, but only appears in the dI/dV map? Can the authors see the CDW in the STM topography taken with low-bias voltages?”

Reply: For correlated materials, it is not surprising that the CDW order is more often detected in dI/dV maps. Primary examples are cuprate superconductors (4a CDW order is mainly reported by dI/dV maps in many papers). We do can detect CDW order with some of other bias voltages as proposed by the reviewer. We show a topographic data taken at -30mV and its Fourier transform below. Signatures of 2x2 charge order are marked by red circles in the Fourier transform data. We have included this data as Extended Data Figure 11 in the revised draft.

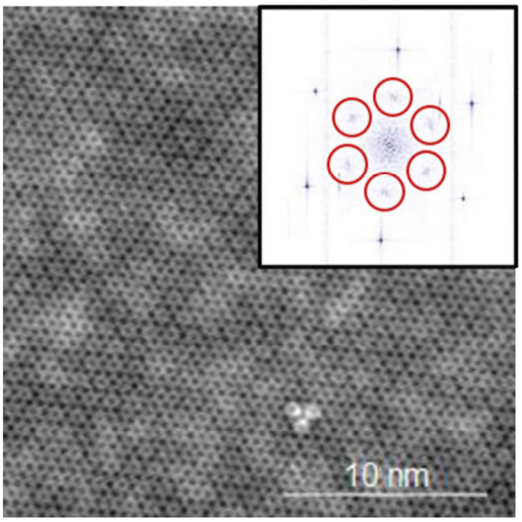

Fig A4: STM topographic data taken at -30mV and its Fourier transform.

"5.In the ARPES data, the authors showed several van Hove Singularities. However, why no clear van Hover Singularity features can be seen in the dI/dV spectrum shown in the inset of Fig. 3g?"

Reply: STM is not an ideal technique to detect van Hove singularity for 3D materials. Van Hove singularity in 2D features a divergent peak in LDOS. For 3D materials, van Hove singularity position has a *kz* dispersion over a large energy range (300meV for such as AV_3Sb_5), and the divergent peak is largely smeared out. In STM literature of AV_3Sb_5, there is no substantial peak that can be ambiguously identified as van Hove singularity (and many of the claimed van Hove singularities only reference to k_z=0 bands are outside the energy range of +-50meV). There are spectral peaks outside +-50meV for FeGe, but for the reasons of 3D kz dispersion, we don't think there is strong enough evidence to claim any of these as a key signature of van Hove singularity. ARPES in this regard is more rigorous for discussing the existence of van Hove singularity with an excellent k_z resolution.

"6. Can the authors also detect the CDW gap in the ARPES measurements?"

Reply: Yes, we indeed observed the CDW gap. Below we show the energy distribution curves taken where the vHS band crosses the Fermi level, divided by the Fermi Dirac function convolved with the energy resolution. A suppression of the spectral weight is observed below T_{CDW}, with an energy scale consistent with that observed by STM. More detailed momentum-dependence and temperature dependence will be reported separately.

Redacted Figure + Text

[redacted]

" 7. The authors use the chiral flux phase of the circulating currents to explain the phenomena in FeGe. Since the STM experiments can be directly performed on the Kagome layer, is there any direct evidence for the circulating currents in the Kagome layer?"

Reply: The direct STM evidence for chiral flux phase is still unknown theoretically and is experimentally under discussion. Such STM evidence have been discussed in AV_3Sb_5 system while the neutron discovery of magnetic coupling of CDW in the current work is unprecedented. Therefore, we would like to focus on the neutron evidence here in this work and discuss the STM evidence in a separate follow up work.

One piece of evidence that invited the theoretical discussion of chiral flux phase is the STM observation of magnetic field switching of chirality of the charge order in AV_3Sb_5. In FeGe, the spin flop transition magnetic field is beyond our STM instrumentation capability. However, the internal magnetism is also expected to polarize the chirality of charge order within each kagome lattice layer in FeGe. In this regard, we have detected the chirality switch of the charge order with neighboring kagome layers, as shown below:

[redacted]

Redacted Figure + Text

[redacted]

In addition, Spin polarized STM has also been performed in FeGe to support the chirality switch between c-axis antiferromagnetically ordered kagome layers. Some of such evidence is shown below, which will be discussed in a separate follow up work:

Redacted Figure + Text

[redacted]

These follow up STM work essentially support our neutron discovery of magnetic coupling of charge order, and the chiral flux phase is certainly one plausible interpretation for these collective observations.

“8. Line 127, it should be “by the white square in Fig. 2f.....””

Reply: We thank the referee for this comment, and this has been corrected.

Referee #2

“I have read the manuscript by Teng et al. with much interest. The authors report the coexistence and interplay between antiferromagnetism and charge-density-waves in a kagome metal.

The material studied here, FeGe, has clearly a rich physics, which the authors have studied using a suite of advanced probes. The collection of data and information on this system is impressive, and experiments have clearly been performed to high standards and in a very comprehensive fashion. As a result, the study encloses a wealth of information about the electronic and magnetic structure of this system, which enabled the authors to draw connections between the fermiology and the symmetry-broken phases which include a combination of charge and magnetic order.

I believe the results are of broad relevance for the quantum matter community, and as such I am supportive of this manuscript being featured in Nature. I find that the observed phenomenology is sufficiently unique and exciting as to meet the novelty criteria of this journal (and agree with the authors' closing statement that "Regardless of the theoretical explanation of these experimental results, we have identified a kagome metal system in FeGe where strong electron correlations lead to a rich interplay of magnetic order, topology, CDW, and AHE"). At the same time, the findings presented here are interpreted within a framework which is not fully supported by the available evidence."

Reply: We thank the referee for the recognition of the comprehensive and high quality of our work, and also that our results are of broad relevance for the readership of Nature. We especially thank the referee for the appreciation of our results for their experimental values independent of theoretical interpretation.

"The main issue is the interpretation of the anomalous Hall effect (AHE) as arising from a chiral flux phase that is closely associated to a time-reversal-symmetry breaking charge-density-wave. If I understand correctly, the Hall data indicate that below the spin-flop field, the Hall resistivity is linearly proportional B and therefore there is no AHE, while above the spin-flop field, the linear behavior extrapolates to a finite value at zero field, suggesting the presence of an AHE arising from a field-induced ferromagnetic moment which is not ascribed to the canted magnetic moments but rather to a magnetic flux generated by a chiral CDW. This behavior arises only below the CDW transition; therefore, the authors conclude that the CDW must be in some way involved. The interpretation proposed by the authors is then that there are two chiral CDWs in neighboring planes whose opposite fluxes are perfectly compensated at low fields and remain compensated up to a spin-flop field of several Teslas, even though the magnetic moments are progressively tilted away from the c-axis (making the AFM order slightly uncompensated as reflected in the increasing magnetic moment shown in Extended Data Figure 2c). Then, at the spin-flop field, this balance between the CDW fluxes is suddenly removed, and there is a net flux that develops along the c-axis, breaking time-reversal symmetry and generating an AHE. While this interpretation is somewhat plausible, it rests on a series of hypothesis which are not experimentally validated, first and foremost the time-reversal-symmetry breaking nature of the CDW, which is not assessed directly (for instance, by means of a comparison between spin-flip and non-spin-flip neutron diffraction measurements). Furthermore, the susceptibility data as a function of external magnetic field display a shoulder before the spin-flop field, an effect which becomes the more prominent the lower the temperature (i.e. deeper into the CDW phase). This enhanced susceptibility seems to indicate the rise of the c-axis magnetization ahead of the transition, yet the crossover in the Hall resistivity seems to be very sharp, occurring precisely at the spin-flop field. How is such a behavior explained by a chiral flux phase which is only activated in the spin-flop phase?"

Reply: These are excellent questions asked by the referee. First of all, we agree with the referee that our interpretation of the data is only one possibility and we do not (and cannot) claim that this is a unique interpretation of the data. We also agree with the referee that we have not carried out neutron diffraction work under field to figure out whether the spin flop transition is a first order phase transition. If we assume that the spin-flop transition is a first order transition below and above the CDW temperature, and occurs only above the dashed line in Fig. 2c when moments are saturated, then the initial rise in magnetization at fields below spin-flop transition is likely due to uneven polarization of the flux phase due to applied field. Since the magnetic moment from Fe along the c-axis produces much large molecular field than that from the flux phase, the system overall is still antiferromagnetic and therefore there is no AHE. After the spin-flop transition, the molecular field from Fe moment is mostly parallel to the FeGe plane, and therefore the effect of uneven polarization of the flux phase becomes much larger, and therefore the AHE. Of course, we agree with the referee and editor that this is only one plausible interpretation of the data. We have considerably tuned down this claim, and made absolutely clear in the paper that this is only a hypothesis. In the coming months, we will do neutron scattering experiments under a magnetic field, and sort out how spin-flop transition occurs below and above CDW. We trust that these future experiments will allow us to gradually sort out what is happening in the system.

"While the one above is the major item that caught my attention, I made a few extra notes about some passages that I could not entirely understand and I was hoping the authors could clarify:

- Top of page 3: "There is not yet an example of a coupled CDW and magnetic order that emerges within an established magnetic phase where the CDW appears as a consequence of magnetism.". The authors should clarify what they mean by "CDW appears as a consequence of magnetism". There is no doubt that these two orders are coupled, as chiefly demonstrated by the modification to the ordered moment that occurs as the CDW develops. However, what hard evidence do the authors have that the CDW is a consequence of magnetism? One could perhaps say the two have a common origin in the fermiology (AFM order from a flat band instability and CDW from a van Hove singularity as proposed here) but I do not see a direct link, or perhaps I missed this while reading the manuscript."

Reply: We thank the referee for these comments. What we meant was that the magnetic order occurs at a temperature much higher than the CDW temperature in FeGe. This is much different from the cuprates and nicklates, where CDW ordering temperature occurs above or at the magnetic ordering temperature. Regarding CDW appearing as a consequence of magnetism, our understanding of the system is exactly as the referee has suggested, namely that the two may have a common origin in the electronic structure. In our picture consistent with our data (shown below), the kagome flat band initially at E_F in the paramagnetic state demonstrated by first principle calculations (10.1103/PhysRevB.102.125130) possibly induced the A-type antiferromagnetic order via a combination of high density of states and nesting. With decreasing temperature, the flat band splits under exchange splitting. The spin minority portion shifts up and brings to near the Fermi level the vHS. However, before we can definitively prove this, we have softened the language to suggest strong coupling instead of consequence.

[redacted]

Redacted Figure + Text

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" - Middle of page 3: "When flat bands are near the Fermi level, EF, strong electron correlations can induce magnetic order, as has been found in kagome lattice FeSn.". To the best of my understanding, this scenario is still the subject of speculations. That the origin of AFM order is a Stoner instability created by the flat bands has not been confirmed experimentally, nor theoretically. Some recent studies have proposed such a connection, but the evidence remains inconclusive (primarily because it is hard to 'catch' a flat band in the act of creating an instability, and DFT-LDA is known to have severe limitations in describing the effect of exchange fields and consequently the position of spin-polarized bands – in particular flat bands)"

Reply: We agree largely with the comments raised by the referee here. As one can see from temperature dependence of band structure shown above in Fig. B1, we have preliminary evidence of shifting of the flat band with the ordering of the magnetic moment as temperature decreases, thus providing tentative evidence for the scenario suggested by the referee. Nevertheless, we agree we should soften the claim, and revised the sentence in question to soften the claim. "When flat bands are near the Fermi level, E_F, strong electron correlations may induce magnetic order, as has been suggested in kagome lattice FeSn."

"- Top of page 4: "Since AFM order in FeGe likely arises from electronic correlations of the flat electronic bands near the Fermi level…". This is related to the previous point, but here this hypothesis (that the flat bands drive AFM order) is applied to FeGe. Are there previous theoretical works supporting this scenario? Note that the works cited here do not refer to FeGe but to other compounds (some of which, like CoSn, do not exhibit magnetic order)."

Reply: We agree with the referee that the written statement is a bit of speculative as written. It is known from first principles calculations (both DFT and DMFT) on FeGe (arXiv: 2203.01930), the kagome flat bands are at E_F in the paramagnetic phase and are split in the AFM phase. Also, in our temperature dependence of the ARPES data, we find clear evidence of a shift of the flat band away from E_F with decreasing temperature. This is consistent with above mentioned scenario where we only observe the portion of the flat band below E_F. With that said, we agree with the referee that we cannot prove that the flat bands are causing the AFM, but only that our data are consistent with this interpretation.

"- Bottom of page 8: "one can consider a nesting scenario where vHS2 at M below EF nests with vHS3 at L above EF…". While the presence of a vHS near the Fermi energy is a telltale of a possible strongly nested Fermi surface in hexagonal systems, the ultimate assessment for such a scenario comes from the shape of the Fermi surface itself. If nesting conditions are present, one expects nearly flat Fermi contours connecting the vHS at the M-points, as observed in some of the AV3Sb5 compounds. It would be helpful if the authors could show a non-projected version of the Fermi surface in 4a and discuss how close the Fermi contours are to the ideal nesting conditions."

Reply: We appreciate the referee for this suggestion. In the revised Figure 4, we changed the Fermi surface to non-projected flattened version to better illustrate possible vHS nesting scenario. We reproduced it here, where we can see nearly flat Fermi contours connecting the vHSs at the M points, similar to what has been reported for AV_3Sb_5.

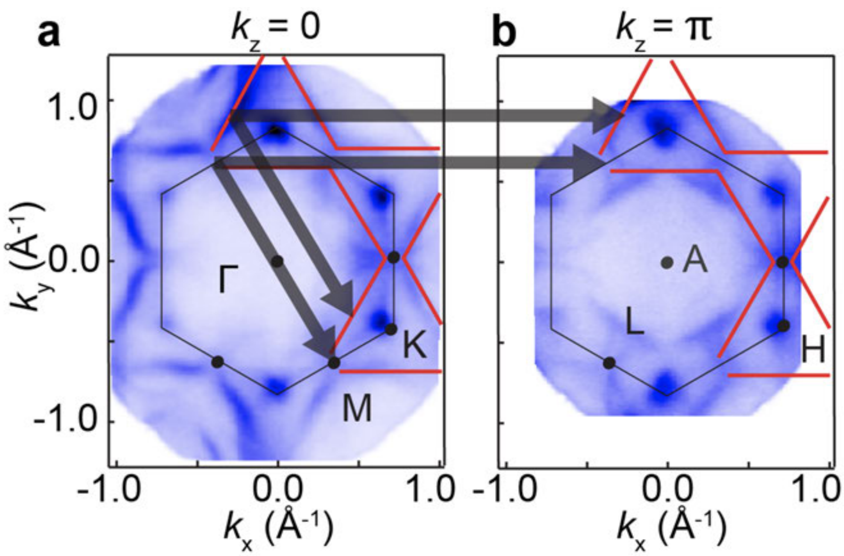


Fig. B2: Fermi surface measured at the kz=0 and π planes, showing quasi-flat pieces of the Fermi surface connected to the vHS at the M points.

"- Middle of page 9: "The energy splitting of the majority and minority bands increases with decreasing temperature and shifts the electronic bands such that the nesting of vHSs in Fig. 4a to form CDW becomes possible.". This is an interesting possibility which can be tested theoretically via spin-polarized LDA+U calculations and experimentally by performing ARPES measurements at different temperatures (say between T_CDW and room temperature). Without the support of either pieces of evidence, however, this scenario remains purely hypothetical and the interpretation of the role of AFM order-induced Zeeman splitting is not conclusive (although somewhat plausible). I think a clarification of this aspect will be particularly important."

Reply: We again largely agree with the referee on these points. In our unpublished ARPES data (shown above in Fig. B1), we do see a clear shift of the flat band and vHSs with temperature between T_{CDW} and room temperature. There is also DMFT calculation of the flat band in the paramagnetic and AFM states posted on arXiv after the submission of our work (arXiv: 2203.01930), showing a splitting of the kagome flat bands due to the ordered moment. Both of these results support our proposed scenario. Still, we do agree with the referee that the scenario is hypothetical. Since these are in the discussion section of the paper, we revised the paper to make absolutely clear that the scenario is only a possibility and is clearly subject to future experimental and theoretical verification.

"- Extended Data Figure 3. It would be very helpful if the authors could show the Hall resistivity data down to low temperature in panel c, where the AHE signal deviates significantly from zero. Additionally, I was expecting to see signatures of the change in slope of rho_xy vs B at the spin-flop field in panel e, but this is only visible for the large jump that takes place for the AFM phase. Could the authors show an additional panel that focuses on the AFM+CDW region, using a color map that makes the transition in rho_xy more apparent?"

Reply: In the revised Extended data Fig. 4, we clearly show the low temperature data. We appreciate very much these suggestions by the referee.

"In sum, I very much enjoyed reading this manuscript and I thought the experimental information is remarkably extensive, which has taught me a lot of interesting things about this new material. This kagome compound is clearly a very exciting, new playground to explore the interplay between correlation phenomena from a different angle, especially when it comes to the CDW/AFM interplay. At the same time, I cannot recommend this paper for publication in its current form for the reasons I outlined above, but I hope the authors can address my questions and, most importantly, provide new information in support of their proposed interpretation of the origin of the AHE."

Reply: We thank the referee for these comments. With new data provided in the reply, we hope to convince the referee about the validity of our results, particularly the interpretation of the data based on follow up ARPES and STM measurements shown here. The STM work is submitted for publication, and we are in the process of preparing ARPES work for publication.

Referee #3:

"In this manuscript, the authors report an intriguing set of experimental observations in their hexagonal FeGe single crystals. The central results are about a new phase below T ~ 100K that has pronounced signatures in physical properties (magnetic susceptibility, electric transport, and specific heat). Using single-crystal neutron diffraction, the authors attribute the phase to a charge-density-wave (CDW) order, whose propagating wave vectors are also seen with Fourier-transform scanning tunneling spectroscopy. A highlight is that the wave vectors are the same as the CDW wave vectors in the AV3Sb5 family, which shares the same layered kagome crystal structure as hexagonal FeGe and is currently under intense research. Based on this commonality, the authors argue that peculiarities of electronic structure and correlations in kagome metals may be at the origin of the intriguing physical properties of FeGe, in particular what they call an emergent anomalous Hall effect (AHE), which has also been claimed in the AV3Sb5 family. Finally, since the CDW wave vectors appear to connect van Hove singularities in FeGe that presumably follow from antiferromagnetic (AFM) order formation below 410K, it is suggested that the CDW is strongly coupled to the AFM order, which makes it unique among other CDW examples in correlated metals."

"To my knowledge, the 100K transition has not been reported in hexagonal FeGe. The manuscript is well-written with a broad vision. Conceptual connections (and contrasts) are made to the spin-charge stripes in high-Tc cuprate superconductors, and especially to a topical 'chiral flux phase' in AV3Sb5, which features time-reversal symmetry breaking due to inter-atomic circulating currents (similar physics has been proposed for explaining the pseudogap phase of high-Tc cuprates). A charge order with emergent orbital magnetization may indeed produce magnetoelectric transport consequences such as AHE, so the whole story sounds physically plausible, and can be anticipated to intrigue several major research communities. The collaboration also involves well-known research groups. With such a story, the manuscript will almost certainly make an impact.

These merits of the manuscript could warrant publication in Nature, given that the experiments and interpretation are technically sound. But this is where I see major issues, which occur at different levels. I try to indicate below how the issues might be overcome if the authors firmly believe in what they claim."

Reply: We are grateful to the referee for the detailed reading and summary of the merits of our manuscript and for acknowledging the impact our work would have on several major research communities. We are also thankful for the suggestions from the referee on improving the rigor in the interpretation of our results. Below is our detailed response to each of the suggestions.

"1. There is a stark discrepancy between the current study and a previous one in their experimental magnetometry data: between Fig.1f of the present manuscript, and Fig.2 of Beckman et al., Physica Scripta. Vol. 6, 151-151 (1972). The latter data were re-plotted in Ref.[24], yet upon citing this reference and discussing the result in Fig.1f, the authors did not mention the discrepancy. The issue cannot be overlooked. Hexagonal FeGe was first studied at low T and in B fields over half a century ago, quite carefully indeed by researchers in Sweden including M. Richardson, who first reported the growth method [48] used by the present study. Richardson was on the 1972 paper mentioned above, and the most important feature around T=100K (of χperp) in the present Fig.1f is completely absent in the previous data. Importantly, all the other features seem to correspond. So, the discrepancy concerning the 100K anomaly must be due to a difference in either the purity of samples, or the way how the measurements were done."

Replyy: We appreciate these insightful comments from the referee, particular the reference by Beckmann et al. In order to check the directional dependence of magnetic susceptibility, we have carried out additional measurements after carefully aligning the single crystal with a X-ray Laue machine. In the revised Fig. 1f, we include data at different in-plane orientations and show that there is basically no difference. While these data are clearly different from what was reported by Beckmann et al, we are not sure why that is the case. One possibility is that the earlier measurements have a rather large temperature steps (about 5 K), which may have missed this transition. However, we must confess that we are not sure whether this is the right interpretation.

"If it is related to sample quality, I am afraid that the present samples are problematic, e.g., contaminated by impurity phases that can inter-grow with hexagonal FeGe. Some candidates can be found in these recent papers: [doi:10.1016/j.jssc.2018.10.030], [doi:10.1039/d0dt03923c], and some of them even have a magnetic transition near 100K. I should say that I still do not consider impurity to be the likely explanation, since all other aspects of the present data set (especially specific heat) are in support of the 100K transition being genuine to hexagonal FeGe. But, in case there is a problem with the samples, essentially all conclusions of the present manuscript would be invalid."

Reply: We appreciate these additional references mentioned by the referees. In agreement with the referee, we do not believe impurity phase is the likely explanation of our data. As shown in the revised extended data Fig. 1, we have carried out careful crystallographic measurements of our sample, and have not seen any other Bragg peaks besides the hexagonal FeGe. These results are consistent with our single crystal neutron refinement at 440 K (Fig. 2 of main text) which again did not reveal any other impurity discussed in the above mentioned references. Any significant impurity phase should have been observed and the fact that specific heat (bulk measurement) shows signature of the CDW transition indicates that the transition is a true bulk phenomenon. We expanded discussion in the method section to make this clear and cited relevant papers.

"To show that the above discrepancy is not related to sample quality, the authors should try to reproduce the lack-of-100K-anomaly χperp data previously reported by Beckman et al. There is one possible way out: while the c-axis is unique, there are still two high-symmetry choices for the in-plane field direction that are mutually orthogonal. The authors should precisely define their in-plane field direction, and I am guessing that another in-plane direction would produce the result of Beckman et al., i.e., no anomaly at 100K. And if that turns out true, this statement in the manuscript is false: "the reduction of susceptibility is consistent with gapping density of states at the Fermi level by the CDW." It brings me to my next criticism, namely, if the transition at 100K leaves a highly anisotropic signature in the magnetic susceptibility, it is more likely to be magnetic than CDW."

Reply: We have carried out additional susceptibility measurements as suggested by the referee. As shown in the revised Fig. 1f, we find no angular dependence in χperp data for two angles we measured: in-plane magnetic field 0°, and 120° with respect to the a-axis. While this still leaves the question on how to reconcile our present results with that of Beckman et al, this leads us to the conclusion that the modern measurements are likely to be more accurate and correct, and that the CDW phase transition was somehow missed in the earlier work.

“2. I am unconvinced by the "CDW" interpretation of the 100K order even if it is genuine to FeGe. The key support for the CDW interpretation is this sentence: "Since these peaks persist to large Q-positions in higher zones where the Fe2+ magnetic form factor drops off dramatically, they must be nonmagnetic and associated with lattice distortions induced by a CDW or lower crystalline symmetry of unknown origin (Figs. 2f, 2k)."

I do believe that the H,K=integer, L=half-integer diffraction peaks (as shown in Fig.2e) are magnetic. However, those peaks are clearly still present in the "higher zones", which shows that the Fe2+ form factor alone is not enough to fully suppress magnetic scattering in those zones. Extended Data Fig.6g tells the same: neglecting the strong peaks around Q=3-4A^-1, it is not obvious at all that the magnetic intensity drops strongly up to Q=8A^-1. It means that due to the crystal structure, the form factor does not predominantly determine magnetic peak intensities, and that the structure factor also matters a lot. While the "CDW" peaks in Extended Data Fig.6h show an increasing trend towards larger Q, the overall intensities are weaker than the magnetic peaks in Extended Data Fig.6g. In other words, there is still 'room' for the new peaks below 100K to be magnetic.”

Reply: We appreciate very much these insightful statements from the referee and agree with them.

“The array of "CDW" peaks in Fig.2f actually have peculiar behaviors. As an example, I consider H=half-integer peaks in rows of the same integer L. At L=0 we essentially see no intensity, which implies that atoms move only in the c direction upon entering the "CDW" phase. The L=-1, -2, -3, and -4 rows of (H half-integer) peaks are common in that, in each row, the intensities monotonically *decrease* towards higher H. I cannot easily understand this by CDW, but it would be natural if the underlying order is actually magnetic. Both the form factor and the "S_perpQ" spin-polarization factor (by virtue of the magnetic scattering intensity being zero at L=0) would be able to explain the decrease in each row with increasing H.

The bottom line here is that the magnetic form factor argument is weak for proving a CDW origin. The STS data do not help much either, because when the system is already ordered magnetically for in-plane q=0 (below 410K), having an extra magnetic order at H or K=0.5 will lead to a second-order effect that looks like a (weak) charge order: the product of two time-reversal-odd order parameters is time-reversal-even (as a CDW should be). To make the CDW case stronger, the authors should consider improving their experimental evidence:”

Reply: We appreciate very much these insightful statements from the referee and agree with them completely. In the submitted draft of the paper, we only plotted data within H, and L below 4. Since our data covers much wider Q-range, we actually do have CDW peaks appearing at (6.5,0,-5.5), which corresponds to Q= 12.7 1/Angstrom and basically not possible to be magnetic (See extended data Fig. 7 for the magnetic form factor of Fe). In the revised draft of the paper, we included these new cuts in the revised Fig. 2 of the paper. In addition, if there is magnetic order at H or K=0.5 (L=0.5), we would expect spin waves stemming from these wave vectors. Instead, our inelastic neutron scattering experiments only reveal spin waves stemming from the wave vector of A-type AFM order at H,K= integer and L=0.5 at 70 K, a temperature below CDW ordering temperature.

Redacted Figure + Text

[redacted]

“1) A displacive model calculation can be done for the "CDW" neutron diffraction intensities (it does not have to be a full refinement of the distorted structure), to see how much of a (2x2) lattice distortion is needed to produce observed signal magnitudes. As explained above, I expect the atomic displacements to be along c, which should strongly affect tunneling currents in STM measurements. With that estimate, they would hopefully be able to find corresponding signatures with an STM tip (distance to top atoms of the same element).”

Reply: We have carried out a simple calculation assuming the lattice distortion is on Fe only and is only along the out-of-plane direction, with a wavevector same as the CDW wavevector (1/2 1/2 1/2). Under this assumption, we built a FeGe superlattice with $a^* = b^* = 2a = 9.98$Å and $c^* = 2c = 8.10$Å, extracted all CDW and integer Bragg peaks from 70K data (excluding the magnetic peaks) and multiply the *hkl* coordinate by a factor of 2. If the CDW phase has the symmetry shown in Fig. 1(d) in the main text, the space group of the superlattice is then reduced to P622 (#177), and the two Fe coordinates are (1/4 0 1/4+Δc_1) for Fe1 and (1/2 1/4 1/4+Δc_2) for Fe2. Fixing all Ge atom positions, we use JANA2006 to refine the two parameters Δc_1 and Δc_2. The result, as shown in extended data figure 10, gives Δc_1=0.0067 and Δc_2=0.0022, which corresponds to 0.054 Å and 0.018 Å, respectively. This estimation implies that the Fe atom will have a distortion within ~1% of the original lattice parameter in the CDW phase. These results are added in method section with Extended Data Figure 10.

“2) A non-resonant X-ray diffraction experiment should be ideal for revealing a CDW order. The authors seem to have in-house access to a variable-T X-ray diffractometer, and should attempt such measurements below 100K.”

Reply: We thank the referee for this suggestion. We have indeed carried out X-ray diffraction measurements at ORNL at temperatures above and below the CDW temperature. As shown in revised Extended Data Figs. 1c and 1d, we indeed observe superlattice peak at the expected positions below the CDW transition temperature and there are no peaks at 150 K. These results again confirm the structural nature of the superlattice peaks, consistent with our statement based on neutron data.

"3) A spin-polarized neutron diffraction measurement of several of the "CDW" peaks should show that they are all non-spin-flip and hence due to coherent nuclear scattering. Or, the authors can perform unpolarized neutron diffraction up to much higher Q, where no magnetic peaks are seen above 100K. If they are able to show that the peaks at half-integer H,K become visible below 100K at those high Q, it would be strong evidence for CDW."

Reply: Yes, we agree with the referee on this point, and have revised Fig. 2 to show exactly what the referee is requesting.

"3. I believe that the AHE interpretation is problematic. The AHE magnitude is extracted from the linear extrapolation of high-field \rho_{xy} data back to zero field for taking the intercept. In Fig.1k, the 180K data indeed look like a spin-flop transition, namely, the Hall coefficient changes abruptly across the transition, resulting in \rho_{xy} vs. B linear relationships on both sides of the transition. As both lines would go through the origin (under extrapolation to B=0), a jump must occur at the transition field.

However, the 10K data in Fig.1k show no jump, but only a change in slope around 7T. One might still argue that it is because the AHE's non-zero intercept cancels the spin-flop jump, but the argument is based on a coincidence. With curiosity I found additional data plotted in Extended Data Fig.3d-e (I am thankful that the authors are careful with showing raw data). To me, the jump disappears immediately below "T_CDW", which would mean that the coincidental cancellation holds true at every temperature below "T_CDW". That is very unlikely, and I think a more plausible explanation is that there is no AHE of the claimed type at all, and that the transition at 100K is of a magnetic origin. In particular, I suspect that the "spin-flop" transition becomes something else below "T_CDW" – I see a discontinuity in the magnetization data at "T_CDW" near 7.5T in Fig.1i. That would explain why no jump in \rho_{xy} is observed below ~100K. An investigation of the field-hysteresis behavior for temperatures both above and below 100K might be useful in this regard."

Reply: We appreciate these comments from the referee. As shown in revised extended Data Fig. 4, the jump \rho_{xy} vs. B is always there and it is not coincidentally "cancelled" below T_CDW. The size of the jump decreases continuously with decreasing temperature, and crosses zero around 60 K and becomes negative at lower temperatures. Regardless of the temperature evolution of the jump, the non-zero intercept of high field \rho_{xy} below T_CDW is robust. To our knowledge, such a non-zero intercept can only be understood as AHE. While we still don't have a complete understanding what is happening microscopically, we note that spin anisotropy gap at the AFM wavevector decreases with decreasing temperature. Although there is no appreciable change across CDW temperature, it vanishes in the canted magnetic phase below 60 K as shown below. These results may be related with the negative jump in \rho_{xy} vs. B. Nevertheless, we expect field-induced state in CDW, canted, and regular AFM phase to be similar, meaning the spin-flop phase. Only future neutron diffraction experiments under a magnetic field will sort this out. Finally, we did not observe hysteresis in \rho_{xy} either above or below T_CDW, as shown in revised extended Data Fig. 3.

[redacted]

Redacted Figure + Text

"4. I find several seemingly important statements which I cannot agree as they are phrased at present. They do not necessarily affect the validity of the whole paper but should still be revised.

(Page 7) "Therefore, while the CDW order clearly enhances the ordered moment of the collinear A-type AFM phase, we do not observe any effect of the CDW on the formation of the canted AFM structure."

The canted AFM structure develops at much lower temperatures than the CDW. In other words, there is no experimental comparison between canted AFM with/without CDW. How do the authors come to this conclusion?"

Reply: We apologize for confusing the referee with this sentence. What we meant to say is that when canted AFM structure occurs, there is no intensity change in the CDW ordering peak as seen in Figs. 3a and 3b. For comparison, the magnetic scattering commensurate wave vector (2,0,0.5) is clearly enhanced at the CDW temperature, but is then reduced at the canting temperature of 60 K. In the revised draft, we revised the sentence to make this very clear.

“(Page 9) "The energy splitting of the majority and minority bands increases with decreasing temperature and shifts the electronic bands such that the nesting of vHSs in Fig. 4a to form CDW becomes possible."

I think such nesting would be spin-flip nesting, and more likely to (if not can only) promote spin- rather than charge-density waves. It is another reason to suspect that the order below 100K is magnetic.”

Reply: We very much appreciate these comments. As extensively discussed above, we have no evidence of another magnetic structural transition below 110 K at (0.5,0,0.5), from inelastic spin waves, refinements of the neutron diffraction pattern, X-ray diffraction, and wave vector dependence of the scattering. From ARPES measurements, we know there is band structure shift below room temperature, our discussion provided a plausible interpretation of the CDW order. As emphasized in the draft, we recognize that this may not be the only interpretation of the data. Clearly, future theoretical work will shed more light on this problem.

“(Page 10) "Importantly, we observe no AHE in FeGe in the low field regime even from the circulating currents. This indicates that the circulating currents are locked to the ordered moments in the low field range, clearly different from AV3Sb5 where a small field already induces an AHE without magnetic order."

A net circulating-current magnetization at high fields could at most be an add-on response to the spin part of the magnetization, which could enhance the Hall signal, and when the spin magnetization goes to zero (extrapolation to zero field from the spin-flopped phase), the circulating-current magnetization also goes to zero. I hence do not think the authors have managed to explain the origin of AHE, even if I were to fully agree with their cartoon in the last panel of Fig.1.”

Reply: We fully agree with the referee and the editor that this scenario is somewhat speculative. Our original thoughts were that when spin-flop transition occurs, spin magnetization would basically go to zero and orbital part of the signal would give rise to the AHE since orbital current would not participate in the spin-flop transition as indicated in the last panel of Fig. 1. However, we do agree with the referee that this is speculative and we have no conclusive evidence for an orbital current in FeGe. In the revised draft, we have considerably tuned down this claim, and made absolutely clear that we actually do not completely understand the origin of the observed AHE in the spin flop phase.

“Calling such an effect "AHE" is potentially misleading, because AHE in the strict sense implies spontaneous breaking of time-reversal symmetry. Here, the time-reversal symmetry is in the first place broken by the applied c-axis field, i.e., the observed effect is a non-linear Hall effect rather than AHE. I understand that the terminology was (problematically) introduced first in some of the AV3Sb5 studies and the current authors are not the ones to blame, but if their work becomes published in a prestigious journal like Nature, this is a good opportunity to help correct misconceptions in some communities especially for the non-specialists.”

Reply: We agree with the referee that by the initial definition of the AHE is the spontaneous Hall effect at zero field. However, more recently the idea of AHE has been extended to antiferromagnetic materials and soft magnetic materials and study the Hall effect due to the field induced magnetization and non-colinear spin-structure. We would like to emphasize that the AHE we observed here is different from the non-linear Hall effect in AV3Sb5. The \rho_{xy} is linear both below and above the spin flop transition, yet the liner intercept of high field \rho_{xy} is non-zero below T_CDW. Similar observations were made and understood as AHE in several AFM materials, including MnBi2Te4, EuCd2As2, GdPtBi. For example, https://www.nature.com/articles/nphys3831 use the AHE terminology in describing their AHE from an antiferromagnetically order system. In the revised manuscript, we cited the above mentioned reference, and made absolutely clear of this point.

"5. (Minor) I was confused by some (likely) typos:

Fig.1k caption, \rho_xx should be \rho_xy

Text page 4, a reference to Fig. 2d should be Fig. 2c?

Text page 5, "... reminiscent of the out-of-plane magnetic susceptibility (Fig.1i) ..." I see nothing similar, the 2-4T part of Fig.1i is completely white in color. Perhaps they mean in-plane instead of out-of-plane, and Fig.1f instead of Fig.1i?

(There could be more as I did not note all of them.)"

Reply: Thank you very much, we meant to say there is a drop in in-plane susceptibility and modified text to make this clear. We did not plot field dependent susceptibility data below 4-T as there are no features there (for raw data please see Extended Data Fig. 2). In Fig. 1i, we want to emphasize the onset of spin-flop transition below Tcdw.

"In summary, the claims of the manuscript are novel and of broad interest. But I have concerns about the soundness of the experimental data and interpretation, and cannot recommend the work for publication. If, after further investigations, the authors conclude that the order below 100K is genuine to hexagonal FeGe, but it is something other than CDW, or that the AHE interpretation has to be altered, the finding could still be considered a nice discovery (a new order in a half-century old kagome metal is still exciting). Such result may warrant publication in a less selective journal."

Reply: We thank the referee for all the valuable suggestion. Based on our replies and additional data above, we believe that we have convincingly shown that the phase transition at 100 K is a CDW transition and that it is coupled with the magnetic order. We hope these pieces of evidence will convince the referee that the work is of sufficient interests for publication in Nature.

Referee #4

"In this paper, the authors report the coexistence of CDW and antiferromagnetic phase. The key point here is the COUPLING between CDW and magnetic order, which had never been reported in any material system. If this could be confirmed, the paper would definitely deserve to be published in Nature. However, based on the results in the paper, I can only confirm the coexistence of CDW and antiferromagnetic phase. The coupling between CDW and magnetic order cannot be demonstrated by the current experimental results. If the evidence of coupling could be provided, I would support the publication in Nature."

Reply: We thank the referee for the critical reading of our manuscript and for recognizing the importance of our work given that a coupling of the CDW and the magnetic order can be demonstrated. To this end, we believe that our results have indeed demonstrated the coupling between the magnetic order and the CDW order, as also positively commented by the previous referees. Specifically, as shown in Figs. 3a and 3b, the CDW order and associated superlattice peaks are both presence below 100 K. The magnetic ordered moment shows a clear increase at the onset temperature of the CDW order (Fig. 3f). Since we have shown that the magnetic structure of the system is unchanged below CDW, this leads us to the only plausible interpretation of the data that the magnetic order is enhanced by the CDW order, thereby demonstrating a coupling of the two orders.

Additional evidence for a strong CDW and magnetism coupling is through the observation of the AHE effect below CDW temperature as shown in Fig. 1K. Since AHE effect implies spontaneous breaking of time-reversal symmetry most likely associated with magnetism, its observation below CDW temperature strongly suggests a magnetism and CDW coupling.

“Figure 1 shows the transport and magnetic measurement results, which clearly demonstrates that something happens at T-CDW. Figure 2 and Figure 3 show the neutron scattering and STM data, which clearly demonstrate lattice distortion at T-CDW and the CDW gap. The ARPES data in Figure 4 clearly shows the vHSs in this material, which provides importance information for understanding the coexistence of CDW and AFM in this material. These experimental results convincingly confirm a CDW phase in the AFM system. However, to confirm the coupling, the authors need to show the CDW state can be tuned by manipulating the magnetic states, and vice versa.”

Reply: We thank the referee for pinpointing to the key experimental observations we have presented from neutron, STM and ARPES, which demonstrates the coexistence of CDW and magnetic order. The referee asked us to show that “the CDW state can be tuned by manipulating the magnetic states, and vice versa.” As discussed above, temperature dependence of the magnetic scattering and CDW order shows clear coupling. However, given the energy scale of the CDW order, application of a 14-Tesla magnetic field will likely not affect the CDW ordering temperature (see Figs. 1h and 1j). One possible way to manipulate CDW and magnetic order is through chemical doping, for example doping-Co into FeGe. As far as we know, there are no report of hexagonal Co-doped FeGe. Our group is in the process of carrying out these experiments, but we hope that the referee will agree that these future experiments are beyond the scope of the present paper.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

I understand the authors's responses to my comments. I would like to mention that the method for claiming the chiral CDW in Figs. A6 and A7 is not reliable. This method is strongly related the status of the STM tip, and two recent STM works have shown that there is no such chiral CDW in the AV3Sb5 kagome materials.
https://www.nature.com/articles/s41586-022-04493-8
https://www.nature.com/articles/s41567-021-01479-7

While I think the physics reported in this work needs further investigations, I agree that this work reports a new kagome antiferromagnet with CDW which is interesting. In this sense, I recommend the publication of this work.

Referee #2 (Remarks to the Author):

I have read the revised manuscript by Teng et al.. The authors have gone to great lengths in addressing my earlier remarks (and those of the other reviewers) and I very much appreciate their efforts.

First and foremost, I am happy with how the authors reformulated the discussion of the origin of the AHE signature, which was the major item in my initial assessment. I appreciate that the authors have taken a more agnostic approach to the interpretation of these experimental observations, which are clearly very intriguing and deserving of further investigation.

I am also satisfied with the authors' response to my other points. I appreciated the addition of the new ARPES panel in Fig. 4b and the provision of new temperature-dependent ARPES data on the flat band and van Hove singularity (Fig. B1 in the rebuttal letter). I have a couple of final questions and a request regarding these, which I hope the authors could address these in their next round revision.

The question is regarding the bands shown in Fig. B1 (left). If I understand correctly, the momentum cut is across the M point. Therefore, I take it that the measured van Hove singularity is the one labeled as "vHS1" in Fig. 4c. Looking at the band diagram in the same panel, I was expecting that the flat band would be located above the Fermi energy (for all the three main kagome bands identified here). How do the authors explain the energy position of the flat band in Fig. B1? If the shift is due to the internal exchange field acting on spin-polarized bands in the AFM, is it expected that only the flat band is affected, or should one expect a similar (downward) shift of the vHS?

In any case, the data of Fig. B1 are quite enlightening as they provide a rather direct demonstration of the relationship between magnetic order (via the ordered moment m^2) and the band structure. Given that the central result of this paper is the discovery of a novel type of interplay between AFM and CDW in a kagome system, I believe these data should be shown in the manuscript. I understand that the authors were planning to use these data for a subsequent publication, and I would not be suggesting their incorporation in the present manuscript if it was not for the fact that they represent key evidence in support of the very central message of this study. The inclusion of this final piece of experimental evidence would greatly add to the manuscript and to the other reported results.

Referee #3 (Remarks to the Author):

I appreciate the authors' revision and response to my comments. But after considering their whole package of results at present, I remain unconvinced of the authors' claim that they have uncovered CDW order in FeGe that is intrinsically related to the material's AFM order, or that the CDW causes a transport behavior that should be interpreted as an emergent anomalous Hall effect (AHE) or even arising from a chiral flux phase. I therefore do not find in the manuscript the level of novelty, importance, and rigor expected for a Nature paper.

The comparison to the previous result reported by Beckmann et al. remains an important unresolved issue. The authors have been unable to reproduce the previous results using their samples. The gross shape of the curves in Fig.1f looks the same as the previous result, yet the dip between 60K and 110K is only seen in the present data. I do not think a regular magnetometry measurement, even if done some 50 years ago, would entirely miss the dip (even if data points were taken in 5K steps as the authors guessed). I can only think of the difference as arising from some uncontrolled variations in the sample composition, such as Fe1.02Ge versus FeGe, and/or in structural imperfections, such as stacking faults. For instance, it is well-known that stacking faults in \alpha-RuCl3 matter a lot for the magnetic ordering as a bulk property (visible in specific heat; see, e.g., Cao PRB 93, 134423 (2016)), and Fe-Te compounds of slightly different compositions have the same structure at room temperature but behave very differently at low temperature (Bao PRL 102, 247001 (2009)). Maybe the present samples are better, or worse, in either/both of these regards compared to the samples of Beckmann et al., but it does not really matter. The bottom line here is that the AFM (intra-layer FM) order looks essentially identical between the two studies, yet the CDW transition is seen in only one type of samples, so there can be no causal relation between the AFM and the CDW orders. They are at best coexisting orders in the present samples.

I might agree with the authors that some version of Fe-Ge is a CDW material, yet there are already hundreds if not thousands of such, and finding one more is not really that novel.

Leaving the above issue aside for now, I appreciate the authors' inclusion of additional X-ray diffraction data in Extended Data Fig. 1, of new the neutron diffraction data up to a higher momentum range in Fig. 2, and display of inelastic neutron scattering data showing no spin waves stemming from (0.5,0,0.5) in their reply. However, I consider these to be at best circumstantial evidence, which might support the CDW interpretation, but which also certainly produces additional difficulties in the logic chain:

a) The authors claimed in the reply that at (6.5,0,-5.5) Q=12.7 1/Angstrom, neutron diffraction peaks are "basically not possible to be magnetic." I might agree with that, but I also see a few H=integer peaks not far from that Q, such as at (7,0,-4.5), which is supposed to be "AFM peak" according to the assignment in Fig.2k. Is the latter "AFM" or "CDW"? Either way, it would be a problem for the present story. In particular, if the "CDW" propagation vectors are found at q=(0.5,0,0), (0.5,0,0.5) AND (0,0,0.5), it becomes not only different from those in AV3Sb5, but also difficult to be related to the AFM-induced van Hove singularities - in other words the relation to the magnetism becomes highly questionable. Moreover, if the (0,0,0.5) *nuclear* diffraction intensity changes below 100K, the result in Fig.3f (Q_AFM) could be highly misleading, as the intensity increase at "QAFM" might not be magnetic at all!

This last issue is directly related to the abstract ("enhances the AFM ordered moment", it made me not raise an issue about the *coupling* in the previous round, but now I no longer believe it) and the authors' reply to Referee 4 concerning the coupling between the CDW and the AFM. The new diffraction data at very high Q actually weaken their argument for the coupling.

b) XRD in Extended Data Fig. 1 also shows weak diffraction peaks at both H,K=half-integer AND integer. The H,K=integer peaks are not really discussed by the authors. I should emphasize that for a system that is already magnetically ordered with q=(0,0,0.5) from the first place, having a secondary magnetic propagation vector of q=(0.5,0,0) [and (0,0.5,0)] would be able to produce many additional structural diffraction peaks, if one would suspect that the H,K=0.5 diffractions are partly magnetic. Alternatively, the system may simply undergo some complex structural distortion affecting all of q=(0.5,0,0), (0,0.5,0) and (0,0,0.5) and their combinations below 100K in the authors' sample of Fe-Ge. The bottom line here is that the case is not as simple as the authors are proposing, and the connection of the CDW to the AFM and AV3Sb5 is weak, if not absent.

c) Lack of spin waves emanating from a weak magnetic Bragg peak is actually not uncommon. In Fig.C1 of the authors' reply, even the spin waves stemming from H=1 is quite weak, whereas from H=0 is strong. I think the opposite is true for the H=integer magnetic Bragg peaks in Fig.2e, and it is not that simple. By a reversed argument, for a CDW Bragg reflection, one would expect to see Kohn-anomalous phonons at low but finite energies from the Bragg peaks (Hoesch et al., PRL 102, 086402 (2009)), even if the CDW is believed to be electronically driven (Le Tacon et al., Nat. Phys. 10, 52 (2014)). H=0.5 in the present case is very far from primary nuclear reflections, are the authors able to show such phonon INS data at high Q?

I see a discussion about this issue in the authors' reply to Referee 1, but the data in Fig.A3 are very noisy. The authors' purpose of showing no such phonons there also seems problematic to me -- seeing some excitations (either magnetic or structural) above the H=0.5 Bragg peaks would actually be a good thing for determining what the order below 100K is. Such excitations would have to be there regardless of the interpretation. The authors are offering simply no clear answer to this important point at present.

About AHE - It is clear from the above-mentioned points that the transition at 100K is rather complicated. It might not be present in all FeGe samples, and it causes many neutron and X-ray diffraction intensities to change, so there is no surprise that it also affects the transport property of the authors' samples. On top of that, at essentially all temperatures the system has a field-induced transition between 7T and 10T, and the so-called AHE signature is observed via (linear) extrapolation, which is by definition problematic in the present case, because such linear extrapolation relies on linear response, which is not guaranteed (otherwise, there should be no field-induced transition, and no change in size of the anisotropy gap as the authors have shown in their reply).

In summary, I believe that the authors have managed to show that in their FeGe sample, something complicated happens below about 100K. The precise nature of this something has not been determined, nor has its intrinsic relevance to the physics in FeGe (AFM order, in particular) or other kagome-structured systems been demonstrated well. The notion of AHE based on subtle field-dependent transport is not convincing. These being said, the volume of work is substantial. While I do not see them as making a Nature paper, the results might deserve open discussions by the community studying magnetic inter-metallic.

Referee #4 (Remarks to the Author):

After reading all the replies to the reviewers, I believe that the authors have successfully addressed my concerns and hence the manuscript is ready for the publication in Nature. I also expect the realization of mutual tunability between the CDW and magnetic states in the Co dope samples in their future work as mentioned in the reply.

Author Rebuttals to First Revision:

Replies to comments by the referees

Referee #1:

I understand the authors's responses to my comments. I would like to mention that the method for claiming the chiral CDW in Figs. A6 and A7 is not reliable. This method is strongly related the status of the STM tip, and two recent STM works have shown that there is no such chiral CDW in the AV3Sb5 kagome materials.
https://www.nature.com/articles/s41586-022-04493-8

https://www.nature.com/articles/s41567-021-01479-7

Reply: We appreciate very much these comments. Since the present paper did not claim chiral CDW and Figs. A6 and A7 will not be published as part of this manuscript, we will not address the comments by the referee on these points.

While I think the physics reported in this work needs further investigations, I agree that this work reports a new kagome antiferromagnet with CDW which is interesting. In this sense, I recommend the publication of this work.

Reply: We appreciate very much the support of the referee for the acceptance of our manuscript.

Referee #2

Referee #2 (Remarks to the Author):

I have read the revised manuscript by Teng et al.. The authors have gone to great lengths in addressing my earlier remarks (and those of the other reviewers) and I very much appreciate their efforts.

First and foremost, I am happy with how the authors reformulated the discussion of the origin of the AHE signature, which was the major item in my initial assessment. I appreciate that the authors have taken a more agnostic approach to the interpretation of these experimental observations, which are clearly very intriguing and deserving of further investigation.

I am also satisfied with the authors' response to my other points. I appreciated the addition of the new ARPES panel in Fig. 4b and the provision of new temperature-dependent ARPES data on the flat band and van Hove singularity (Fig. B1 in the rebuttal letter). I have a couple of final questions and a request regarding these, which I hope the authors could address these in their next round revision.

The question is regarding the bands shown in Fig. B1 (left). If I understand correctly, the momentum cut is across the M point. Therefore, I take it that the measured van Hove singularity is the one labeled as "vHS1" in Fig. 4c. Looking at the band diagram in the same panel, I was expecting that the flat band would be located above the Fermi energy (for all the three main kagome bands identified here). How do the authors explain the energy position of the flat band in Fig. B1? If the shift is due to the internal exchange field acting on spin-polarized bands in the AFM, is it expected that only the flat band is affected, or should one expect a similar (downward) shift of the vHS?

In any case, the data of Fig. B1 are quite enlightening as they provide a rather direct demonstration of the relationship between magnetic order (via the ordered moment m^2) and the band structure. Given that the central result of this paper is the discovery of a novel type of interplay between AFM and CDW in a kagome system, I believe these data should be shown in the manuscript. I understand that the authors were planning to use these data for a subsequent publication, and I would not be suggesting their incorporation in the present manuscript if it was not for the fact that they represent key evidence in support of the very central message of this study. The inclusion of this final piece of experimental evidence would greatly add to the manuscript and to the other reported results.

Reply: We are happy to hear that the referee appreciates our efforts. As the referee correctly interpreted, our picture of the exchange splitting is that the spin-degenerate bands of the paramagnetic phase splits into spin-majority and spin-minority bands for each ferromagnetic layer. Hence there is a set of spin-minority bands that shift up in energy while another set that shifts down as the moment orders. This picture is confirmed by the first-principle calculations shown in arXiv2203.01930 (reproduced below), where the vHSs near E_F in the AFM phase are those that shifted up as the spin-minority bands. The flat bands associated with these, as the referee correctly points out, must be above E_F, as shown to be within 0.5eV above E_F in the AFM calculations. The FB2 labeled in Fig. B1 cannot be the flat band associated with any of the vHSs near E_F. In our original understanding, this FB2 is a portion of the spin-majority bands that shifted down from E_F as the ordered moment developed. Since then, we have done a systematic study of the band structure on FeGe and identified two terminations which we ascribe to Ge termination and Fe kagome termination. While the vHSs are termination-independent and hence of kagome bulk nature, FB2 turns out to be termination dependent and hence we now ascribe it to a potential surface projection of a kagome flat band that appears on the Ge termination. Below in Fig. 2 we show a comparison of the cut along the Brillouin zone boundary taken on different terminations. As can be seen, the FB2 at ~-0.1eV appears strongly on the Ge termination but only weakly on the kagome termination. The shift of the electronic structure, and most importantly, the shift of the vHSs towards E_F as the moments order, is nevertheless the most important prerequisite for the formation of the CDW. We would be happy to include this information to the manuscript as suggested by the referee and editor. However, we have recently submitted this result as part of a separate manuscript to another journal (our preprint is enclosed here). We would be happy to include the shift of the vHS in comparison to the moment (Fig. 3) into Fig. 3 of this manuscript if it is deemed appropriate to do so by the referees and editors. Last by not least, as the referee pointed out that there should be flat bands associated with the vHS above E_F. We actually do see hints of this flat band crossing EF since this kagome flat band is dispersive and has a minimum energy point in the form of an electron-like band at the K point. This electron-like portion of the flat band can actually be observed at both the K and H points in Fig. 2 below, just barely crossing E_F with the band bottom below E_F.

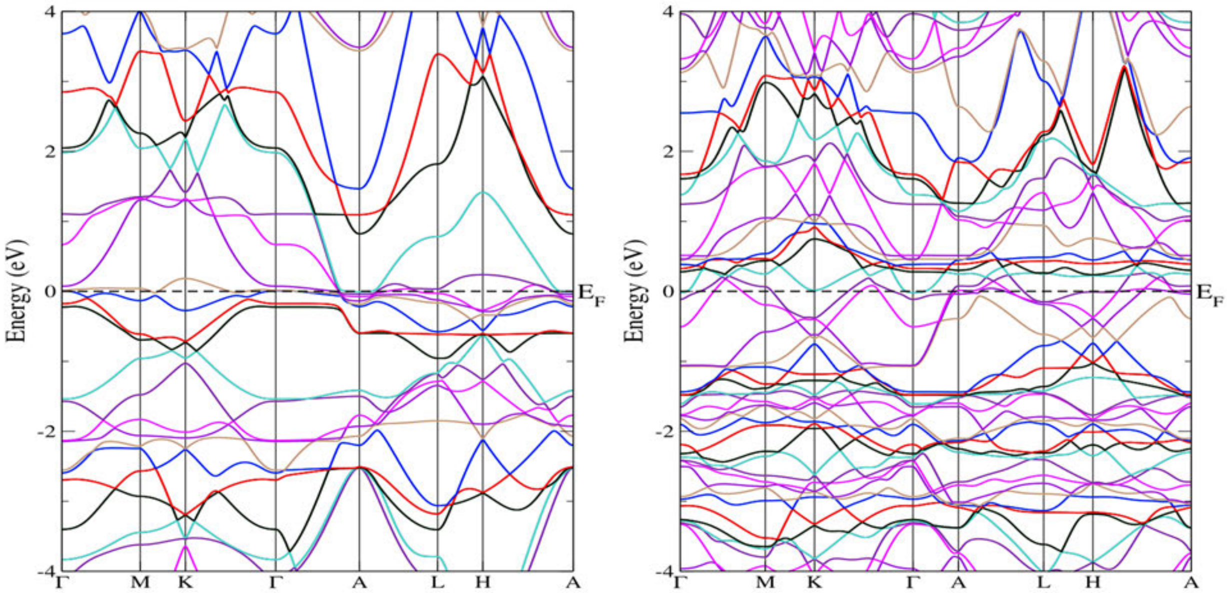


Fig. 1: Density Functional Theory calculations for (left) nonmagnetic and (right) magnetic phases of FeGe reproduce from arXiv: 2203.01930.

[redacted] Redacted Figure + Text [redacted]

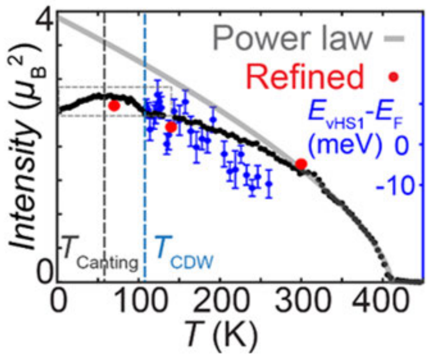


Fig. 3. Shift of the van Hove singularity (blue) towards the Fermi level as the ordered moment (black) forms.

Referee #3

I appreciate the authors' revision and response to my comments. But after considering their whole package of results at present, I remain unconvinced of the authors' claim that they have uncovered CDW order in FeGe that is intrinsically related to the material's AFM order, or that the CDW causes a transport behavior that should be interpreted as an emergent anomalous Hall effect (AHE) or even arising from a chiral flux phase. I therefore do not find in the manuscript the level of novelty, importance, and rigor expected for a Nature paper.

Reply: We appreciate very much the comments of the referee, but hope that our detailed replies below will be able to convince him/her about the validity and importance of our work.

The comparison to the previous result reported by Beckmann et al. remains an important unresolved issue. The authors have been unable to reproduce the previous results using their samples. The gross shape of the curves in Fig.1f looks the same as the previous result, yet the dip between 60K and 110K is only seen in the present data. I do not think a regular magnetometry measurement, even if done some 50 years ago, would entirely miss the dip (even if data points were taken in 5K steps as the authors guessed). I can only think of the difference as arising from some uncontrolled variations in the sample composition, such as Fe1.02Ge versus FeGe, and/or in structural imperfections, such as stacking faults. For instance, it is well-known that stacking faults in \alpha-RuCl3 matter a lot for the magnetic ordering as a bulk property (visible in specific heat; see, e.g., Cao PRB 93, 134423 (2016)), and Fe-Te compounds of slightly different compositions have the same structure at room temperature but behave very differently at low temperature (Bao PRL 102, 247001 (2009)). Maybe the present samples are better, or worse, in either/both of these regards compared to the samples of Beckmann et al., but it does not really matter. The bottom line here is that the AFM (intra-layer FM) order looks essentially identical between the two studies, yet the CDW transition is seen in only one type of samples, so there can be no causal relation between the AFM and the CDW orders. They are at best coexisting orders in the present samples.

Reply: We appreciate these comments and agree with most of them. Since we have carried out careful X-ray and neutron refinement of our single crystals (see extended data Fig. 1), we are quite confident about the quality and stoichiometry of our samples. We have also checked many single crystals grown in different batches with a magnetometer and the results are always the same. Since we don't have Beckmann et al's sample measured some 50 years ago, we really cannot comment on a possible origin of the data discrepancy. As earlier neutron diffraction work did not publish a careful order parameter measurement, it is unclear if there would be neutron scattering intensity change around the CDW temperature. So unfortunately we cannot compare our data directly with the earlier neutron work.

I might agree with the authors that some version of Fe-Ge is a CDW material, yet there are already hundreds if not thousands of such, and finding one more is not really that novel.

Reply: We hope our detailed discussion below will convince the referee about a coupling between CDW and AF order.

Leaving the above issue aside for now, I appreciate the authors' inclusion of additional X-ray diffraction data in Extended Data Fig. 1, of new the neutron diffraction data up to a higher momentum range in Fig. 2, and display of inelastic neutron scattering data showing no spin waves stemming from (0.5,0,0.5) in their reply. However, I consider these to be at best circumstantial evidence, which might support the CDW interpretation, but which also certainly produces additional difficulties in the logic chain:

a) The authors claimed in the reply that at (6.5,0,-5.5) Q=12.7 1/Angstrom, neutron diffraction peaks are "basically not possible to be magnetic." I might agree with that, but I also see a few H=integer peaks not far from that Q, such as at (7,0,-4.5), which is supposed to be "AFM peak" according to the assignment in Fig.2k. Is the latter "AFM" or "CDW"? Either way, it would be a problem for the present story. In particular, if the "CDW" propagation vectors are found at q=(0.5,0,0), (0.5,0,0.5) AND (0,0,0.5), it becomes not only different from those in AV_3Sb_5, but also difficult to be related to the AFM-induced van Hove singularities - in other words the relation to the magnetism becomes highly questionable. Moreover, if the (0,0,0.5) *nuclear* diffraction intensity changes below 100K, the result in Fig.3f (Q_AFM) could be highly misleading, as the intensity increase at "QAFM" might not be magnetic at all!

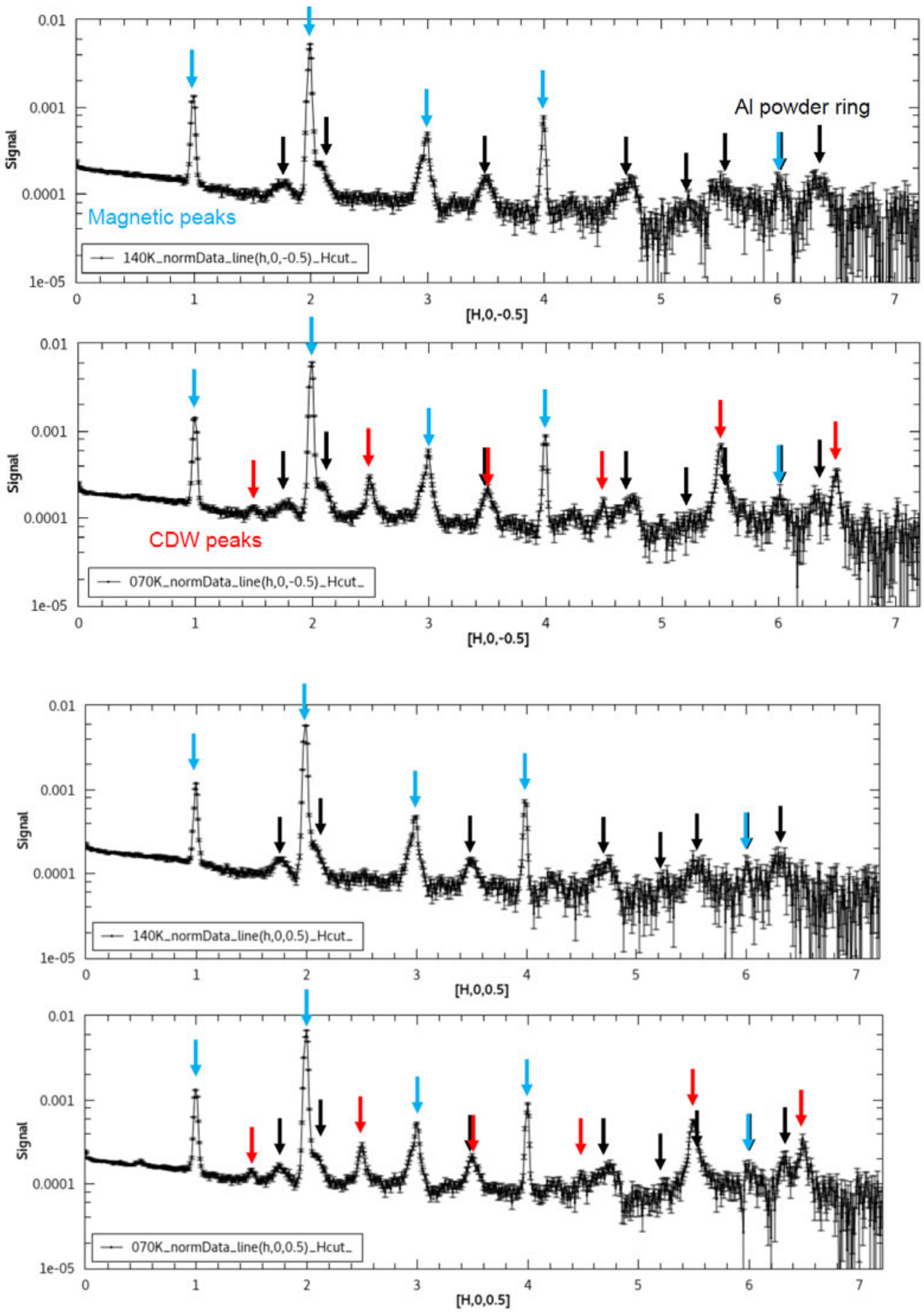
This last issue is directly related to the abstract ("enhances the AFM ordered moment", it made me not raise an issue about the *coupling* in the previous round, but now I no longer believe it) and the authors' reply to Referee 4 concerning the coupling between the CDW and the AFM. The new diffraction data at very high Q actually weaken their argument for the coupling.

Reply: We are grateful for these insightful comments from the referee, particularly for the referee's comment on the weak (7,0,4.5) peak which we honestly had overlooked previously. Because of these comments, we have completely redone the data analysis as previous data analysis did not include extremely high-Q data such as the weak peak at (7,0,4.5). We first consider data at 140 K, where there are no superlattice peaks and all peaks around (H,0,L/2) should be magnetic in principle. To obtain integrated intensity, we cut through all observable (H,0,L/2) peaks and subtract background (including powder ring) scattering. The obtained integrated intensity at different wavevectors are fitted with structural factor of A-type AFM order and also including magnetic form factor of Fe. If A-type magnetic structure can describe the data well, we expect that the Q-dependence of the scattering intensity should entirely obey the magnetic form factor of Fe. The new extended data Fig. 8a show our analysis at 140 K, which reveal that most data points agree with Fe as expected. However, there are two wave vectors that have higher intensity than expected. At Q=(1,0,1.5), the integrated intensity is higher and this is due to the fact that this peaks is sitting on top of an aluminum powder ring (See Fig. 2e, and Extended Data Fig. 5). At (1,0,3.5), we basically see very small scattering with huge uncertainty (see raw data cuts below), and cannot extract integrated intensity. This point is ignored in the data analysis of Extended Data Fig. 8a. Since the data agree well with magnetic form factor of Fe, we can safely assume that any additional large scattering intensity seen at large wave vectors at 70 K not seen in the data of 140 K would likely be nonmagnetic in origin, as the scattering intensity gain at (2,0,0.5) from 140 K to 70 K is only about 20% (see Extended Data Fig. 8c). Extended Data Fig. 8b shows the outcome of this analysis, showing four anomalous points at (green) (2,0,4.5), (3,0,4.5), (5,0,4.5), and finally (7,0,4.5) as noted by the referee. It is therefore clear that these four points are structural (or anomalous) in origin.

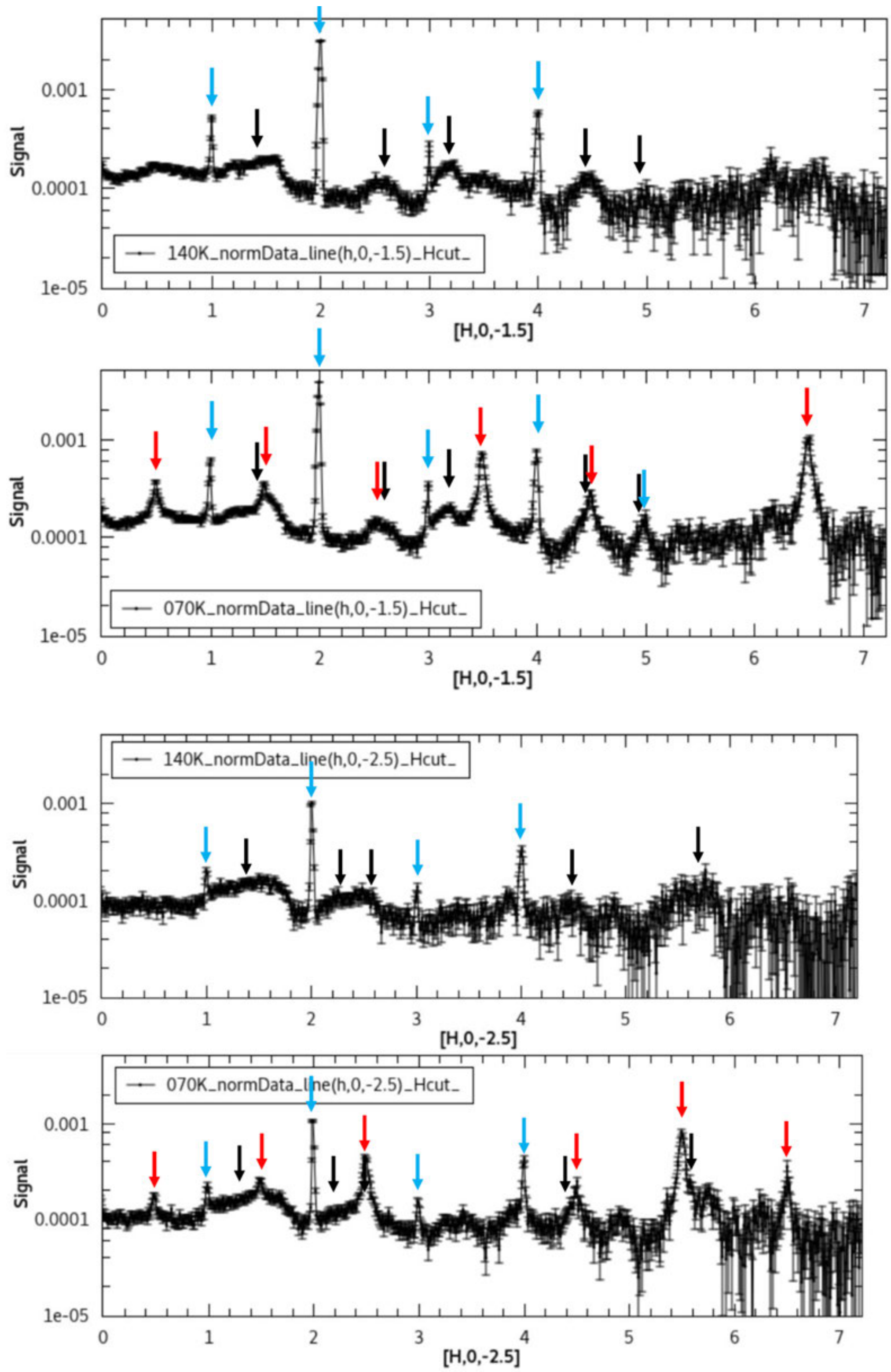
If we assume that these four peaks are indeed structural lattice distortion, we can estimate the structural lattice contribution to the magnetic scattering at (2,0,0.5), the wavevector where we see a clear intensity gain (about 20%) below CDW transition temperature (Figs. 3e and 3f of main text, Extended Data Fig. 8c). Extend Data Fig. 8c plots integrated intensity of all (H,0,L+0.5) and (H+0.5,0,L+0.5) peaks at 70 K and the integrated intensity of (2,0,0.5) at 140 K (solid black circle). As we can see from the figure, the scattering intensity at (7,0,4.5) is about 18 times smaller than that at (2,0,0.5), and is also 4 times smaller than the nearby superlattice peaks. Extended Data Fig. 8d shows the expanded version of Extended Data Fig. 8c, adding the magnetic peaks at 140 K, and marking the intensity gain at (2,0,0.5). Since intensity of superlattice decreases with decreasing momentum transfer as seen in red circles of (H+0.5,0,L+0.5) superlattice peaks, it is safe to assume that the scattering intensity of the lattice feature associated with (7,0,4.5) would decrease with decreasing Q. Assuming a simple linear decrease as shown in the green dashed line in Extended Data Fig. 8d, we conclude that the structural intensity, if present, cannot exceed 10% of the intensity gain at (2,0,0.5) position. Therefore, we are confident that the intensity gain from 140 K to 70 K at (2,0,0.5) is mostly due to magnetic scattering, and the central conclusion of the paper is correct.

To further test if the peaks at (2,0,4.5), (3,0,4.5), (5,0,4.5), and (7,0,4.5) are indeed superlattice peaks associated with lattice distortion, we performed systematic cuts along the [H,0,L+0.5] and [H+0.5,0,L+0.5] directions with L= +-1,2,3,4,5.

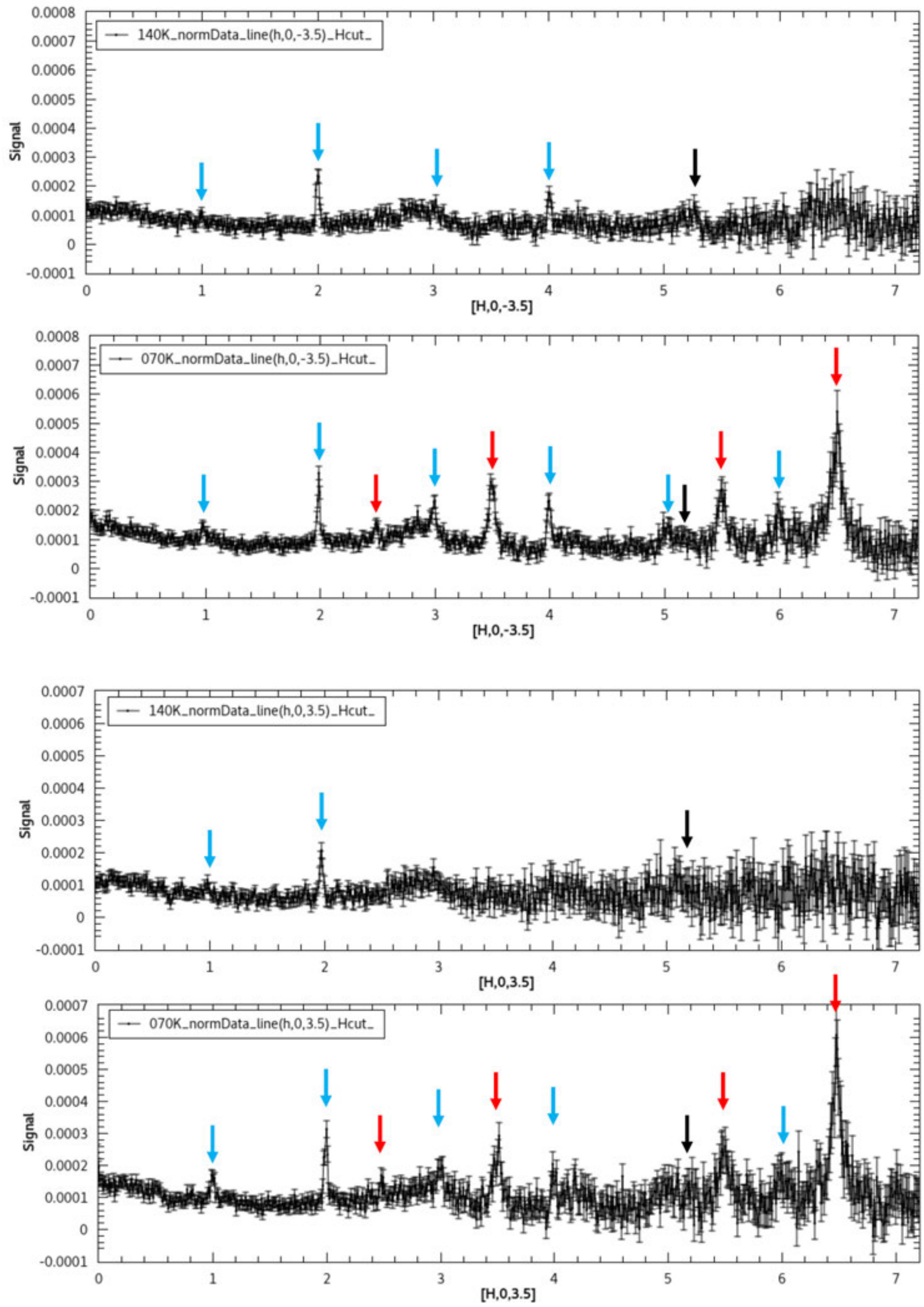
We first show all available data at L=+-0.5 at all H values. As seen from cuts below, plotted in log scale, one can see clear superlattice CDW peaks at (H+0.5,0,+-0.5) at all expected positions, and the intensity of superlattice peaks increase with increase Q, consistent of being structural peaks. On the other hand, we see NO evidence of any structural peaks at (H,0,+-0.5) at large H values (H=7), where there is no evidence of magnetism, suggesting there is NO or weak superlattice distortion peaks at L=0.5 for integer H positions.



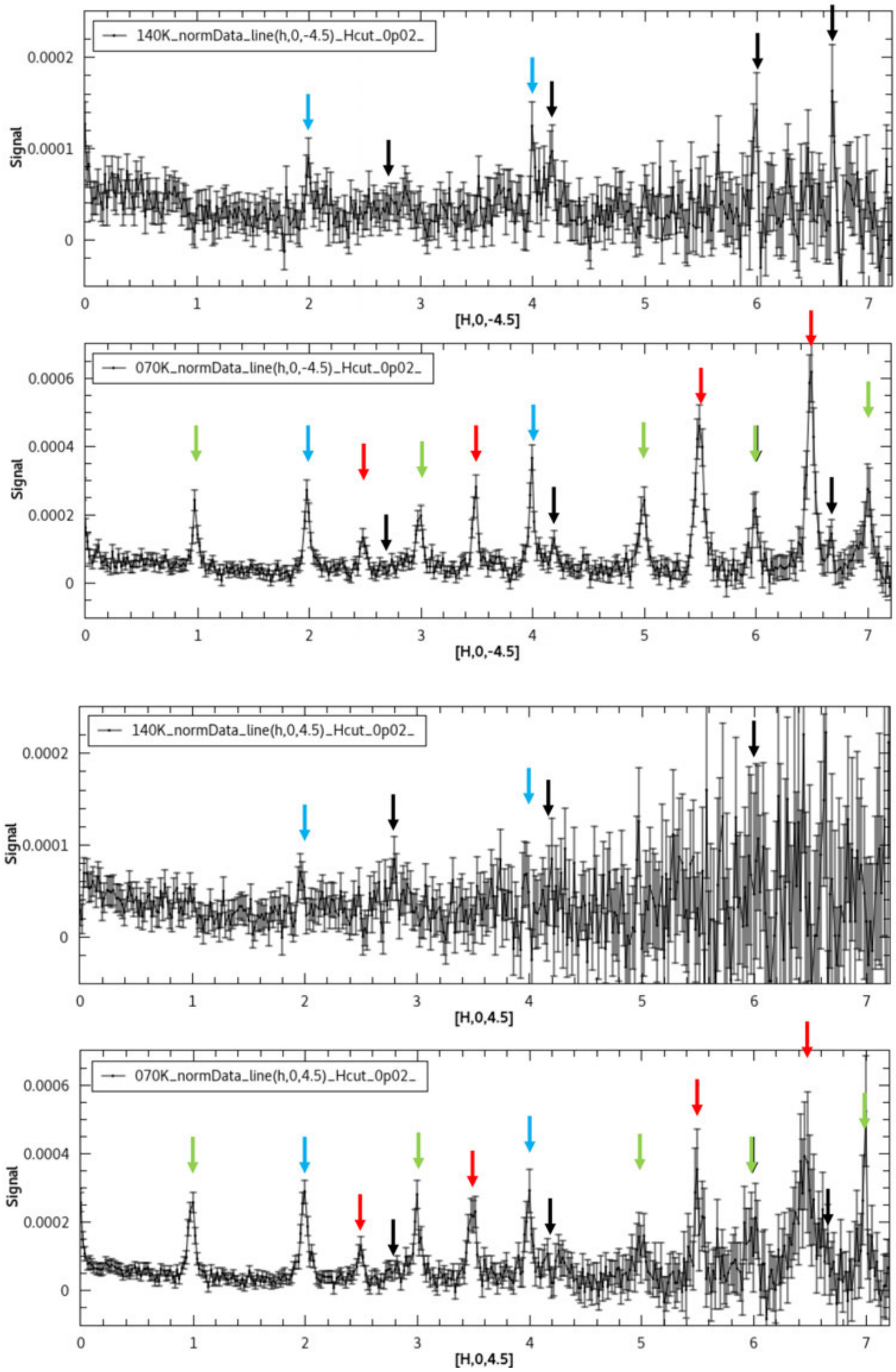
Next, we do the same data analysis at L=+-1.5,2.5 and all H values. In all cases, we only find evidence of superlattice peaks at (H+1/2,0,L+1/2) positions, and no evidence of superlattice peaks at (H,0,L+1/2).



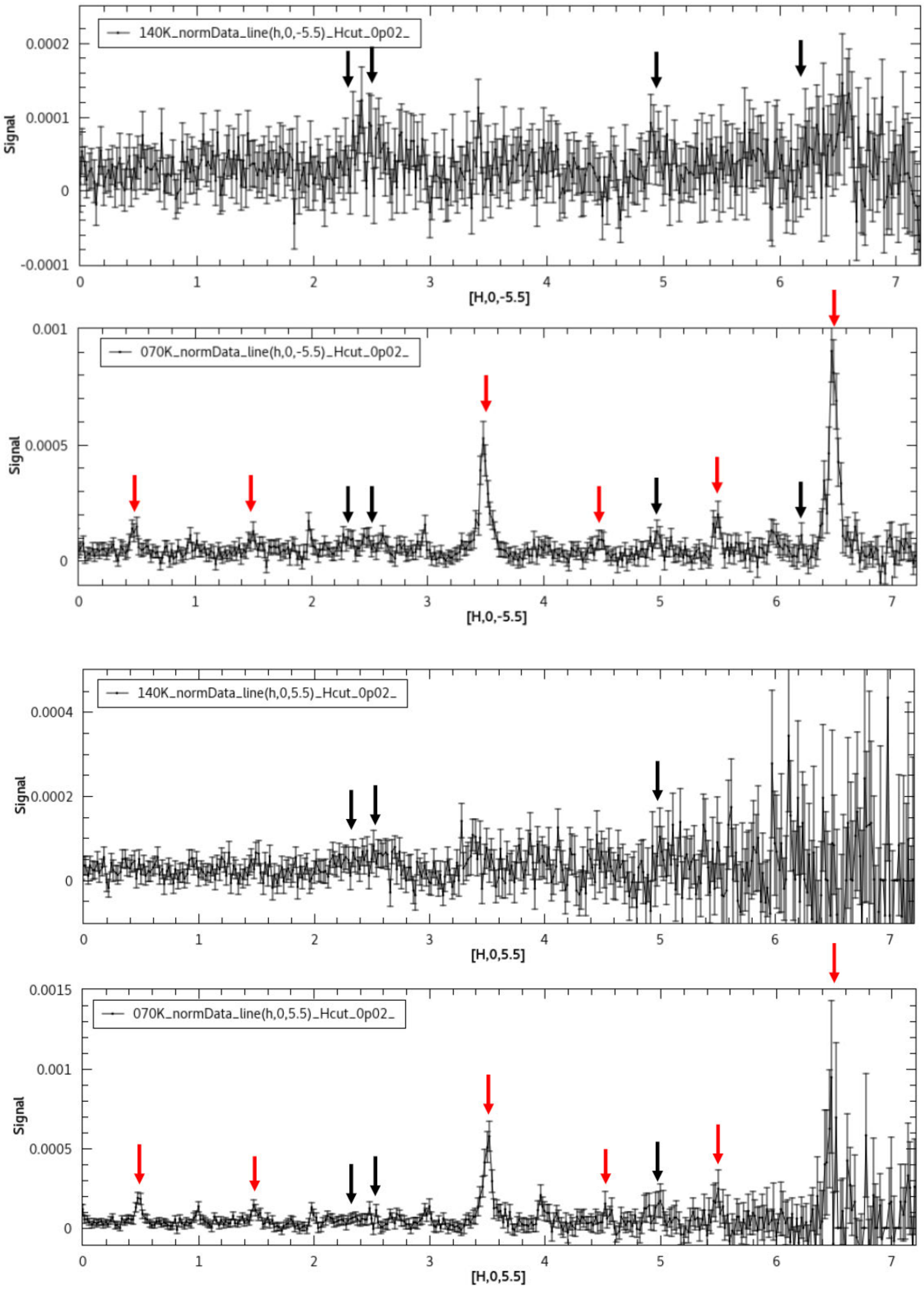
At L=+-3.5, we do the same cuts as shown below. We see again that there is also no evidence at all the superlattice peaks at (H,0,L+1/2) positions, especially we would expect that they are stronger with increasing Q. The small peak at (1,0,+-3.5) is magnetic in origin. This is the position where magnetic structural factor for A-type AFM order is weak. Since there is no evidence of peaks at large H, we conclude that there is no evidence of superlattice peaks at (H,0,+-3.5). At least their intensity will be much smaller than that of the superlattice peaks at (H+1/2,0,+-3.5) marked by the red arrows.



At L=+-4.5, where the referee first noted the anomalous peak at (7,0,-4.5). Cuts along H directions are shown below. At 140 K, we see weak magnetic peaks at (2,0,-4.5), and possibly (4,0,4.5). On cooling to 70 K, clear peak at (7,0,-4.5), as noted by the referee, and other H integer positions are seen. These results are consistent with integrated intensity measurements discussed earlier.



Finally, at L=+-5.5, we see no or very weak evidence of the superlatice peaks at large wave (H,0,L+1/2) positions. If these peaks are there at all, their intensity is much smaller than the superlattice peaks at (H+1/2,0,L+1/2) marked by the red arrows.



Having carried out all the possible cuts, we conclude that while there is nonmagnetic scattering at (H,0,+-4.5) positions. These peaks are not systematically present at other L positions, clearly different from the CDW peaks that occur at (H+1/2,0,L+1/2) positions. Therefore, we conclude that these peaks at L=+-4.5 are unlikely to be superlattice peaks associated with a CDW modulation along the c-axis. As discussed in the first part of our reply, even if we assume that the peaks at L=4.5 are superlattice peaks associated with lattice distortion, its intensity at (2,0,0.5) position is at least one order of magnitude smaller than the experimentally observed intensity gain below CDW temperature. Therefore, we are confident that the intensity gain reported in the paper is due to magnetic moment increase and cannot be due to structural lattice distortion peak intensity gain.

In agreement with the suggestion of the referee, neutron polarization analysis is the only way to conclusively determine this. Unfortunately, the only place this can be done currently is HFIR HB-1. However, HB-1 has beam shutter issues. We have applied beam time there to do this, but this will take at least 4 to 5 months from getting time to scheduling the experiment, assuming beam time is allocated. We hope that the referee will agree with our above data analysis and recommend publication of the work.

b) XRD in Extended Data Fig. 1 also shows weak diffraction peaks at both H,K=half-integer AND integer. The H,K=integer peaks are not really discussed by the authors. I should emphasize that for a system that is already magnetically ordered with q=(0,0,0.5) from the first place, having a secondary magnetic propagation vector of q=(0.5,0,0) [and (0,0.5,0)] would be able to produce many additional structural diffraction peaks, if one would suspect that the H,K=0.5 diffractions are partly magnetic. Alternatively, the system may simply undergo some complex structural distortion affecting all of q=(0.5,0,0), (0,0.5,0) and (0,0,0.5) and their combinations below 100K in the authors' sample of Fe-Ge. The bottom line here is that the case is not as simple as the authors are proposing, and the connection of the CDW to the AFM and AV3Sb5 is weak, if not absent.

Reply: We have carefully re-exam the Extended Data Fig. 1 again and discussed with Dr. Feng Ye, who carried out these measurements for us at ORNL. We conclude that XRD data in Extended Data Fig. 1 shows clear peaks at H,K= half integer but the situation at H,K= integer is unclear. Given that neutron data is bulk measurement looking over the entire volume of the sample, we trust the neutron data and our above analysis much better. In any case, we do not believe there is any magnetic component for peaks at (H+1/2,0,L+1/2) positions. We also do not believe that the scattering peaks at (H,0,+-4.5) positions are magnetic. Although the origin of these peaks deserve further investigation, it does not affect the central conclusion of the paper, meaning that there is clear magnetic intensity gain at the CDW transition temperature. We hope that the referee will agree with us on this. In terms of whether CDW in FeGe is similar to AV3Sb5, we would argue that current confirmed CDW peaks are similar. As we still do not understand the peaks at L=4.5, we added a sentence in the caption of the main text to make this absolutely clear to the reader. We will plan to carry out complete synchrotron X-ray diffraction to figure this out.

c) Lack of spin waves emanating from a weak magnetic Bragg peak is actually not uncommon. In Fig.C1 of the authors' reply, even the spin waves stemming from H=1 is quite weak, whereas from H=0 is strong. I think the opposite is true for the H=integer magnetic Bragg peaks in Fig.2e, and it is not that simple. By a reversed argument, for a CDW Bragg reflection, one would expect to see Kohn-anomalous phonons at low but finite energies from the Bragg peaks (Hoesch et al., PRL 102, 086402 (2009)), even if the CDW is believed to be electronically driven (Le Tacon et al., Nat. Phys. 10, 52 (2014)). H=0.5 in the present case is very far from primary nuclear reflections, are the authors able to show such phonon INS data at high Q?

Reply: We appreciate these comments from the referee. The structural factor of A-type AFM order is that magnetic scattering intensity at H=1, 3, is weak, while they are much stronger at H=2, 4, with L=0.5, as seen in cuts shown above for L=0.5 data. This is consistent with spin waves being very strong at H=even but rather weak at H=odd positions. Based on data at hand, we are confident that there are no acoustic phonons from the (H+1/2,0,L+1/2) positions. To address the question if there are phonons from high Q, we did collect data with Ei=100 meV, which cover higher Q, but we only have these data at 120 K as shown below. The data are consistent with earlier plot, show that phonon band top is around 40 meV.

[redacted]

Redacted Figure + Text

I see a discussion about this issue in the authors' reply to Referee 1, but the data in Fig.A3 are very noisy. The authors' purpose of showing no such phonons there also seems problematic to me -- seeing some excitations (either magnetic or structural) above the H=0.5 Bragg peaks would actually be a good thing for determining what the order below 100K is. Such excitations would have to be there regardless of the interpretation. The authors are offering simply no clear answer to this important point at present.

Reply: We appreciate these comments from the referee and agree with them that the data in Fig. A3 is kind of noisy. These data are the first attempt to measure phonons in the system. We also agree with the referee that there should be excitations stemming from the superlattice peaks. All we want to say at this point is that the current measurement is not sufficient to make any claims on phonon softening issue, this is similar to the case of AV3Sb5 situation. As the present paper is the first recent paper on this system, it should inspire future work in this direction.

About AHE - It is clear from the above-mentioned points that the transition at 100K is rather complicated. It might not be present in all FeGe samples, and it causes many neutron and X-ray diffraction intensities to change, so there is no surprise that it also affects the transport property of the authors' samples. On top of that, at essentially all temperatures the system has a field-induced transition between 7T and 10T, and the so-called AHE signature is observed via (linear) extrapolation, which is by definition problematic in the present case, because such linear extrapolation relies on linear response, which is not guaranteed (otherwise, there should be no field-induced transition, and no change in size of the anisotropy gap as the authors have shown in their reply).

Reply: We agree with the referee that the transition at 100 K is complicated and we don't completely understand it at present. At least such a transition is present in all of our samples, as seen by transport, neutron scattering, ARPES, and STM measurements. While we agree that our AHE signature is observed via linear extrapolation, we would argue, based on data at hand, that linear extrapolation is valid within the magnetic field regime probed in our experiments. We are currently performing measurements on these samples at higher field in high field magnet lab, and any deviation will be reported in the future. Regardless of the future, we hope that the referee would agree that the observation is interesting and should inspire future work in this direction.

In summary, I believe that the authors have managed to show that in their FeGe sample, something complicated happens below about 100K. The precise nature of this something has not been determined, nor has its intrinsic relevance to the physics in FeGe (AFM order, in particular) or other kagome-structured systems been demonstrated well. The notion of AHE based on subtle field-dependent transport is not convincing. These being said, the volume of work is substantial. While I do not see them as making a Nature paper, the results might deserve open discussions by the community studying magnetic inter-metallic.

Reply: We appreciate these comments and agree with the referee that we still do not completely understand the transition at 100 K. However, we hope that we are able to convince the referee with our additional data analysis that the intensity gain of (2,0,0.5) below 100 K is magnetic in origin regardless of the origin of the superlattice peak at (7,0,4.5). With these new analysis, and comments from other referees, we hope to convince the referee about suitability of this work for publication.

Referee #4 (Remarks to the Author):

After reading all the replies to the reviewers, I believe that the authors have successfully addressed my concerns and hence the manuscript is ready for the publication in Nature. I also expect the realization of mutual tunability between the CDW and magnetic states in the Co dope samples in their future work as mentioned in the reply.

Reply: We appreciate very much the support of the referee for the acceptance of our manuscript.

Reviewer Reports on the Second Revision:

Referees' comments:

Referee #2 (Remarks to the Author):

I have read the latest version of the manuscript by Teng et al. and the authors' response letter, and I believe all my remarks have been fully addressed. It was a pleasure to learn about this study and to help the authors in the review process. The manuscript has now my full endorsement for publication.

I would like to add one brief note in relation to the inclusion of the temperature dependent ARPES data. I appreciate the authors' transparency in disclosing their other submitted study which is more focused on the electronic band structure of FeGe. I'd like to further emphasize that, in my opinion, the ARPES data in the present manuscript, and those shown in the response letter, are crucial in demonstrating that the electronic band structure serves as a common denominator linking the CDW and AFM orders. Specifically, the ARPES data show that the system is potentially proximate to a nesting instability at the observed wavevector, while the very same bands of the kagome lattice also follow the temperature evolution of the magnetic order parameter, suggesting a connection to AFM order. Therefore, I believe that the inclusion of the data that putatively support the connection between the two phases is essential to this manuscript, whose core message is the discovery of coexisting AFM and CDW orders in a kagome compound.

Incidentally, this also appears to be one of the central takeaways of the other study by the same team, as is reflected in the abstract of their preprint. I hope the authors can sufficiently delineate the subjects, conclusions, and experimental datasets used in these two different manuscripts.

Referee #3 (Remarks to the Author):

I have studied the revised manuscript along with the rebuttal of Dai et al. The authors made an effort to address my reservations, but I must say that I still remain unconvinced. The key issues are really in my previous summary's first two sentences:

"In summary, I believe that the authors have managed to show that in their FeGe sample, something complicated happens below about 100K. The precise nature of this something has not been determined, nor has its intrinsic relevance to the physics in FeGe (AFM order, in particular) or other kagome-structured systems been demonstrated well."

One outstanding issue was why the present samples are so different from Beckmann et al.'s sample measured some 50 years ago, in the arguably simplest magnetometry measurement data concerning the 100K transition. While I still think this could be a major caveat, I did not turn critical solely for this reason. The discrepancy motivates me to scrutinize the claimed intimate relation between the charge-density-wave (CDW) and the intra-layer ferromagnetic (FM), inter-layer antiferromagnetic (AFM) order. I see a strong desire of the authors to defend this point by making a new Extended Data Figure 8 (EDFig.8), as well as detailed arguments and line-cut data plots in their rebuttal. However, I am troubled by some notable misleading statements and even counter-examples in the new content, which makes me remain doubtful. Specifically:

1) The authors admitted that they had overlooked several high-momentum H=integer peaks observed in the T<100K phase. They now call those peaks "anomalous", such as in the caption to main text Fig.2, on line 776. This is better than saying nothing, but I feel that because the unexplained nature of those peaks can actually affect (explained below) the important claim of enhanced "AFM ordered moment" by the CDW (abstract, line 31-32), calling them "anomalous" is effectively sweeping an important outlier under the carpet, and I strongly suggest the authors not do that.

2) Looking at the new EDFig.8, panel b, it is quite clear that the four new (green) reflections are too strong to be magnetic.

(But I do not quite understand, as written in the caption, how the authors "account for the A-type AFM structure factor" for those peaks. If the actual magnetic structure is different from nominal collinear A-type AFM, will it significantly change the placement of the green data points in the figure? I am not sure. For now, I choose to believe the authors and agree that the four green points are too high to be magnetic.)

However, I do not think that the authors' estimation of a related lattice contribution at (2,0,0.5) is adequately justified. From EDFig.8b, the four green data points obviously do not line up in a linear-with-Q trend. While displacive lattice distortions produce neutron scattering enveloped by an increasing function of Q, the unit cell's structure factor, as well as the Debye-Waller factor, could do something very different. I see from main text Fig.2d (440K data) that the nuclear Bragg reflections at H=5 and especially H=7 are weak, which probably reflects the role of the structure factor (and the DW factor, as T is high). The new reflections (green points in EDF.8b) intensities seem to be anti-correlated with the primary Bragg peaks. Then, when I look at H=2 and small L, my impression is that the primary nuclear Bragg peaks are not strong there, so the new lattice distortion might actually produce a strong signal at (2,0,0.5), contrary to the situation suggested by the green vertical bars in EDF.8c-d. The bottom line is that the structure factor of the new lattice distortion is unknown, and there are multiple ways to draw a line through two of the four green data points in EDFig.8d - for instance, drawing a line connecting the (2,0,4.5) and (3,0,4.5) data points and extrapolating to Q=3A^{-1}, one would almost obtain exactly the observed 70K-140K intensity difference at (2,0,0.5). I am not saying that I would prefer this extrapolation, but only to point out that it is hard to predict how much structural contribution should be present at (2,0,0.5).

3) In order to suggest that the H=2,3,5,7 and L=4.5 reflections are "anomalous", the authors presented line-cut data in their rebuttal and wrote that the integer-H superlattice peaks are absent for L=0.5, 1.5, 2.5, 3.5, and 5.5. But I disagree with this. There are some counter-examples: (5,0,-1.5) (I do not know why the authors put a blue arrow next to it, which implies a magnetic origin, in their corresponding rebuttal figure in the 70K panel), (7,0,-1.5), (6,0,-2.5) (clearly visible in Fig.2f), (6.0,-

3.5). Here, I am only considering high-Q data and primarily using my comparison of Fig.2e&f, but most of the examples are supported also by the line-cuts in the rebuttal.

The bottom line is that I cannot concur with the authors that "these peaks are not systematically present at other L positions" - I think they ARE, and are therefore real. Without understanding them (the structure factor as a function of \vec{Q}, in particular), it is in my opinion very difficult to disentangle between magnetic and nuclear contributions at locations where the two overlap, such as at (2,0,0.5).

4) The relative intensity increase from 140K to 70K at (2,0,0.5) depends on how the intensity is measured. The enhancement in the "integrated intensity" (EDFig.8c) is about 20%, but in Fig.3f where I presume the displayed quantity (black circles) is the peak intensity, the enhancement is only 10%. The authors did not comment on this factor of two difference, but the difference implies that the enhancement part has a broader momentum width than the magnetic peak already present at 140K. Because the authors have found that the CDW peaks are broader in momentum, this is another warning sign that the enhancement at (2,0,0.5) is not primarily magnetic.

I actually expect the authors to somewhat agree with my technical concerns above, as they have the expertise which enabled them to be fully aware of the previous problems once I pointed them out, and they agreed with most of my previous critical remarks. They seemed to be nevertheless confident for what they say, and even expressed their readiness to further verify their claim using spin-polarized neutrons on HB-1, which I consider a necessary step to provide a decisive proof for the *coupling* between the CDW and the magnetism. That being said, as the only referee in the second round who remained doubtful of the main conclusion of the manuscript, I will defer further decision to the Editor.

Author Rebuttals to Second Revision:

Reviewer Reports on the Third Revision:

Author Rebuttals to Third Revision:

Author Rebuttals to Second Revision:

Replies to referees:

Referee #2 (Remarks to the Author):

I have read the latest version of the manuscript by Teng et al. and the authors' response letter, and I believe all my remarks have been fully addressed. It was a pleasure to learn about this study and to help the authors in the review process. The manuscript has now my full endorsement for publication.

I would like to add one brief note in relation to the inclusion of the temperature dependent ARPES data. I appreciate the authors' transparency in disclosing their other submitted study which is more focused on the electronic band structure of FeGe. I'd like to further emphasize that, in my opinion, the ARPES data in the present manuscript, and those shown in the response letter, are crucial in demonstrating that the electronic band structure serves as a common denominator linking the CDW and AFM orders. Specifically, the ARPES data show that the system is potentially proximate to a nesting instability at the observed wavevector, while the very same bands of the kagome lattice also follow the temperature evolution of the magnetic order parameter, suggesting a connection to AFM order. Therefore, I believe that the inclusion of the data that putatively support the connection between the two phases is essential to this manuscript, whose core message is the discovery of coexisting AFM and CDW orders in a kagome compound.

Incidentally, this also appears to be one of the central takeaways of the other study by the same team, as is reflected in the abstract of their preprint. I hope the authors can sufficiently delineate the subjects, conclusions, and experimental datasets used in these two different manuscripts.

We appreciate very much these supportive comments of the referees. In agreement with the referee and the editor, we have now included the temperature dependent ARPES data in the final version of the paper.

Referee #3 (Remarks to the Author):

I have studied the revised manuscript along with the rebuttal of Dai et al. The authors made an effort to address my reservations, but I must say that I still remain unconvinced. The key issues are really in my previous summary's first two sentences:

"In summary, I believe that the authors have managed to show that in their FeGe sample, something complicated happens below about 100K. The precise nature of this something has not been determined, nor has its intrinsic relevance to the physics in FeGe (AFM order, in particular) or other kagome-structured systems been demonstrated well."

We appreciate very much the comments of the referees. Although we agree with the referee that we don't currently have a complete understanding of the charge order (we made that clear in the paper as well), we are confident that magnetic ordered moment increases below CDW transition temperature, and the charge density wave occur at the same wave vector as kagome lattice superconductor 135.

One outstanding issue was why the present samples are so different from Beckmann et al.'s sample measured some 50 years ago, in the arguably simplest magnetometry measurement data concerning the 100K transition. While I still think this could be a major caveat, I did not turn critical solely for this reason. The discrepancy motivates me to scrutinize the claimed intimate relation between the charge-density-wave (CDW) and the intra-layer ferromagnetic (FM), inter-layer antiferromagnetic (AFM) order. I see a strong desire of the authors to defend this point by making a new Extended Data Figure 8 (EDFig.8), as well as detailed arguments and line-cut data plots in their rebuttal. However, I am troubled by some notable misleading statements and even counter-examples in the new content, which makes me remain doubtful. Specifically:

We again appreciate very much these comments. We agree with the referee that we don't understand why our sample is so different from Beckmann's et al's sample (https://liu.se/en/employee/jonbe43). For this reason, we actually wrote to Prof. Jonte Berhard (https://liu.se/en/employee/jonbe43) to ask for their FeGe samples. He had wrote back to us to say that the original sample grower Marcus Richardson had passed away and worked on this system while he was a Ph. D student in 1987 and may have kept some of these crystals. He will look for them and let us know if he can find these samples. If we do get these crystals, we will carry out transport and neutron scattering experiments, and will report the results in a follow up paper.

In the following, we address detailed comments of the referee.

1)The authors admitted that they had overlooked several high-momentum H=integer peaks observed in the T<100K phase. They now call those peaks "anomalous", such as in the caption to main text Fig.2, on line 776. This is better than saying nothing, but I feel that because the unexplained nature of those peaks can actually affect (explained below) the important claim of enhanced "AFM ordered moment" by the CDW (abstract, line 31-32), calling them "anomalous" is effectively sweeping an important outlier under the carpet, and I strongly suggest the authors not do that. 1) The authors admitted that they had overlooked several high-momentum H=integer peaks observed in the T<100K phase. They now call those peaks "anomalous", such as in the caption to main text Fig.2, on line 776. This is better than saying nothing, but I feel that because the unexplained nature of those peaks can actually affect (explained below) the important claim of enhanced "AFM ordered moment" by the CDW (abstract, line 31-32), calling them "anomalous" is effectively sweeping an important outlier under the carpet, and I strongly suggest the authors not do that.

Thank you for these comments. In the revised draft, we replaced the word "anomalous" with "unexpected", and made clear that these deserve further investigation.

2) Looking at the new EDFig.8, panel b, it is quite clear that the four new (green) reflections are too strong to be magnetic.

We agree with the referee on this, and made this clearer in the caption.

(But I do not quite understand, as written in the caption, how the authors "account for the A-type AFM structure factor" for those peaks. If the actual magnetic structure is different from nominal collinear A-type AFM, will it significantly change the placement of the green data points in the figure? I am not sure. For now, I choose to believe the authors and agree that the four green points are too high to be magnetic.)

Again, we don't claim that these four green points are from A-type antiferromagnet. We made clear in the revised caption to say that these are unexpected nonmagnetic scattering we don't currently understand.

However, I do not think that the authors' estimation of a related lattice contribution at (2,0,0.5) is adequately justified. From EDFig.8b, the four green data points obviously do not line up in a linear-with-Q trend. While displacive lattice distortions produce neutron scattering enveloped by an increasing function of Q, the unit cell's structure factor, as well as the Debye-Waller factor, could do something very different. I see from main text Fig.2d (440K data) that the nuclear Bragg reflections at H=5 and especially H=7 are weak, which probably reflects the role of the structure factor (and the DW factor, as T is high). The new reflections (green points in EDF.8b) intensities seem to be anti-correlated with the primary Bragg peaks. Then, when I look at H=2 and small L, my impression is that the primary nuclear Bragg peaks are not strong there, so the new lattice distortion might actually produce a strong signal at (2,0,0.5), contrary to the situation suggested by the green vertical bars in EDF.8c-d. The bottom line is that the structure factor of the new lattice distortion is unknown, and there are multiple ways to draw a line through two of the four green data points in EDFig.8d - for instance, drawing a line connecting the (2,0,4.5) and (3,0,4.5) data points and extrapolating to Q=3A^{-1}, one would almost obtain exactly the observed 70K-140K intensity difference at (2,0,0.5). I am not saying that I would prefer this extrapolation, but only to point out that it is hard to predict how much structural contribution should be present at (2,0,0.5).

We agree with the referee that we don't know at present the structure factor of the unexpected superlattice peaks at H= integer and L=4.5. However, if we believe that these peaks are associated with charge density wave of some sort, then it is reasonable to assume that they should behave similarly to the regular charge density wave superlattic peaks seen at H=half integer, L=half integer peaks. Since all of these peaks decrease with decreasing Q (as explained by a simple model discussed in method section), we believe our estimation of the intensity at (2,0,0.5) is reasonable, particularly because we have no evidence of a superlattice peak at large H for this L position. We added a sentence in the revised caption of the extend data figure to make this clear.

3) In order to suggest that the H=2,3,5,7 and L=4.5 reflections are "anomalous", the authors presented line-cut data in their rebuttal and wrote that the integer-H superlattice peaks are absent for L=0.5, 1.5, 2.5, 3.5, and 5.5. But I disagree with this. There are some counter-examples: (5,0,-1.5) (I do not know why the authors put a blue arrow next to it, which implies a magnetic origin, in their corresponding rebuttal figure in the 70K panel), (7,0,-1.5), (6,0,-2.5) (clearly visible in Fig.2f), (6.0,-3.5). Here, I am only considering high-Q data and primarily using my comparison of Fig.2e&f, but most of the examples are supported also by the line-cuts in the rebuttal.

The bottom line is that I cannot concur with the authors that "these peaks are not systematically present at other L positions" - I think they ARE, and are therefore real. Without understanding them (the structure factor as a function of \vec{Q}, in particular), it is in my opinion very difficult to disentangle between magnetic and nuclear contributions at locations where the two overlap, such as at (2,0,0.5).

We appreciate these comments from the referee but respectfully disagree with some of them. Our key claim is that the structural peaks at L=4.5 are not systematically present at other L, certainly not as systematic as that of regular charge density wave superlattice peaks seen in the same sample This is clearly correct. In terms of some additional superlattice positions suggested by the referees, some of these examples, we believe, are magnetic and that is why we have blue arrows. One thing we do agree with the referee is that we do not completely understand these additional superlattice peaks. We made that very clear in the revised method section. However, we do believe our key conclusion, that is the magnetic intensity gain at charge order temperature, is correct regardless of these additional unknown peaks. We also note that the (5,0,-1.5) peak is almost overlapping with aluminum powder ring at H=4.98 and making the peak width appearing broader.

4) The relative intensity increase from 140K to 70K at (2,0,0.5) depends on how the intensity is measured. The enhancement in the "integrated intensity" (EDFig.8c) is about 20%, but in Fig.3f where I presume the displayed quantity (black circles) is the peak intensity, the enhancement is only 10%. The authors did not comment on this factor of two difference, but the difference implies that the enhancement part has a broader momentum width than the magnetic peak already present at 140K. Because the authors have found that the CDW peaks are broader in momentum, this is another warning sign that the enhancement at (2,0,0.5) is not primarily magnetic.

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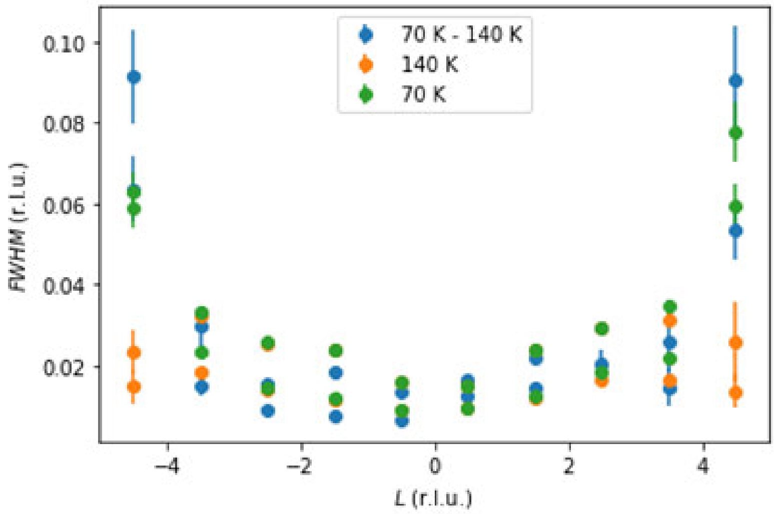
We appreciate these comments from the referee. We checked the integrated intensity calculation again, found a mistake in the program, it turns out that the integrated intensity gain below CDW transition temperature at (2,0,0.5) is about 12% obtained by averaging four equivalent positions within the (H,0,L) scattering plane, while the data shown in Fig. 1f is peak position raw data at (2,0,0.5) without background subtraction. So the intensity shown in vertical scale has both signal and background, and thus slightly underestimate but consistent with the integrated intensity. We also do not see scattering width change across charge ordering temperature as shown in the figure below. The peak width only broadens at L=+-4.5.



We agree with the referee that polarization analysis is the only unambiguous way to determine the magnetic component at (2,0,0.5) position. We are fully committed to do this, and will publish our results when such measurements are carried out. In any case, we are confident of our central conclusion concerning the magnetic intensity gain at charge ordering temperature.