

This manuscript has been previously reviewed at another Nature Research journal. This document only contains reviewer comments and rebuttal letters for versions considered at Communications Physics.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The revised version of the manuscript has substantially improved with respect to the first version. The notation that was unclear has been changed and it is now well defined; the authors extended the description of the experimental procedures and of the data analysis and revised the text in order to avoid misleading sentences. I found now the description of the experiment substantially improved even if the paragraph regarding the occupation imbalance measurement can be further clarified as for example by adding the comment given as reply to Referee A criticism in (2) in the main text:

"Regarding the point #1 above (magnitude of occupation imbalance), one cannot obtain the absolute value of the occupation imbalance from XLD; one can only get a relative magnitude. When the temperature changes from normal state to the saturation temperature of orthorhombicity (~ 50 K), XLD develops with a magnitude of around 1 %. The increase in XLD when the temperature changes from 50 to 10 K is surely from the orbital contribution and it is larger than 2 %. This observation indicates that likely the XLD is dominated by the electronic (orbital) origin, not by the tiny structural distortion."

Concerning the previous literature. In my previous report I suggested the authors to refer and comment about their work in the context of previous works. While in the revised version the authors discussed more completely the results obtained in the context of previous experimental works I found that the discussion in connection with theoretical proposals is still poorly addressed. In that respect I would provide two suggestions:

(i) The authors could add the content of the reply to point (6) of Referee A to the discussion section of the main text

"However, we may offer some discussion on the issue. For the nematic band shift, it is not known why dxz and dyz bands get separated when the system goes into the nematic phase. There are theoretical suggestions such as anisotropic hopping of dxz/dyz orbitals, Pomeranchuk instability, orbital dependent correlation effect and orbital selective spin fluctuation*. However, there is no consensus on this issue yet. On the other hand, orbital selective hybridization in addition to the nematic band shift could be understood by relative orbital energy level. For the normal state of FeSe, the energy level of the dxy orbital is lower than that of the dxz/yz orbital [6]. When nematic band shift occurs in the nematic phase, the dxz level gets closer to the dxy level. Therefore, a strong hybridization between dxz and dxy orbitals is expected."

* Add proper citations e.g. Phys. Rev. B 95, 085108 (2017) (Pomeranchuk instability), Nature Materials 17 869–874(2018) (orbital dependent correlations), Phys. Rev. B 94, 155138 (2016) (orbital selective spin fluctuations)

(ii) Within the discussion of the absence of magnetism in FeSe, the weak-coupling point of view is not supported by any theoretical analysis. As far as I know the role of the orbital nesting in FeSe with respect to magnetism and nematicity has been explicitly addressed Phys. Rev. B 97, 121109 (2018). Please add this and any other relevant citations to support the orbital nesting argument.

In conclusion, having considered the reply to the previous Referees' comments together with the resubmitted manuscript I believe that most of the criticisms have been addressed and that the quality of the research and the presentation of the results have substantially improved. As I pointed out above, still there are few points that should be further considered. However I think that once properly addressed the manuscript should be accepted for publication in Communications Physics.

Reviewer #2 (Remarks to the Author):

I carefully reviewed this revised manuscript, and made my best effort to fairly evaluate it. Unfortunately, however, I could find almost no improvement in this version of the revised manuscript. In addition, the authors does not seem to have faithfully considered the referees' comments. Also, the replies to my previous comments are addressed in a confused way to me, to say the least. In the following, I briefly comment on the authors' replies point by point.

#1 The authors' reply is inconsistent with their description in Fig. 3d.

#2 Here, I must apologize for my typo. "dyz and dxy bands cross each other at the Y point below Ts" should be "dxz and dxy bands ...". Anyway, the authors' claim in Fig. 3d cannot be justified.

#3 The authors' claim is not based on the experimental data analysis. The amount of the nematic band shift of each orbital (dxy, dyz, and dxz) should be evaluated from the analysis of the temperature-dependent measurements.

#4 The authors' reply is unclear. The order parameter should be always temperature-dependent. The point is even if the experimentally determined orthorhombicity is proportional to the order parameter, the authors cannot claim that the structural contribution to the XLD area is also proportional to the order parameter. If the structural contribution to the XLD area was proportional to the square of the orthorhombicity, the temperature dependence of the XLD area could be explained only with the structural contribution.

#5 The authors' rebuttal whether the authors would like to emphasize the "origin" or "characteristics" also does not make sense.

#6 This reply is also nonsense. L_{2,3} edge of the absorption spectra reflects the transition strength from 2p_{1/2,3/2} to 3d orbitals. The calculated width of the Fe 3d band does not matter. The absorption spectra should be broadened more than the width due to the core-hole lifetime. Even if the satellite feature exists above 708 eV, the XLD signal should reflect the occupancy imbalance. The region may be contributed from the different multiplet configurations due to correlation effects, but should not be extrinsic.

#7 With the same reason described above, this reply is also inappropriate.

Unfortunately, the authors' replies are far from convincing, and therefore, I cannot recommend publication.

Lastly, I would like to give additional comments on this manuscript. Basically, the conclusion of this manuscript does not seem to be much different from that of the report from Y. Suzuki et al., [Ref. 12]. Even if the details are different, I think they cannot give any deep impact to most of readers. Since a new detwinning method of a piezo stack was employed in this study, the authors should try to report something new with the method. For example, if the authors investigate how the amount of detwinning depends on the voltage applied to the piezo device (and, if possible, on the applied strain), by implementing some analysis procedure to evaluate the amount, it will be a totally new result and valuable for many readers. It will be also good if the authors investigate the relation between the polarized ARPES spectra and the polarized microscope image depending on the applied voltage.

In conclusion, I cannot recommend this version of the manuscript for publication.

Reviewer #3 (Remarks to the Author):

The authors present a combined ARPES and XLD study of the nematicity in almost detwinned to fully detwinned FeSe. This is a relatively important issue that has been in heated debate in the iron-based superconductor community. The XLD portion on FeSe is indeed new, and presents a comparison to the authors' previous work on Ba122, showing a contrasting behavior between Ba122 and FeSe. The combination of the XLD results with ARPES gives a fresh view of the issue of nematicity in the iron-based superconductors, and in particular the comparison between the case of FeSe with the rest of the iron pnictides. I believe that the manuscript is timely, the results important, and has potential for making important contributions to the field. However, I hope that the manuscript could be written in a more approachable way for the general public who are not used to reading ARPES papers on iron-base superconductors. Below I point out some issues I found.

First paragraph under "results", "...elliptical pockets that are elongated along k_x - and k_y -directions with orbital characters of dx_y and dx_z (dy_z), respectively" I think you have them swapped, dx_z pocket is elongated along k_y .

In the description for "temperature evolution of the electronic structure", I think you have a typo regarding the direction of the dx_z (dy_z) hole band shift. At the $M_x(M_y)$ point, the dx_z hole band shifts DOWN as the temperature decreases. (First paragraph on page 8)

The hybridization between xy and xz/yz is not necessarily due to orbital selectivity. The crossing between xy and yz occurs along the high symmetry direction connecting Gamma and M, while the crossing between xy and xz occurs along the BZ boundary, which is 90deg rotated from the direction of the crossing between xy and yz . These two crossings do not have the same symmetry relations between the respective orbitals.

Last paragraph before the Discussion section, "An important point to note is that the orbital contribution of is positive" has a typo.

According to the data in Fig. 3, the dx_z electron band does not seem to disappear right at T_s , but some lower temperature, especially compared to the strain-induced elevated T_s judging from the merging of dx_z and dy_z bands at M. I think this point was not discussed at all in the manuscript, and is perhaps one of the questions one of the other referees raised about confusion of whether the pocket is there in the nematic phase. For this reason, it is important to state clearly the temperature at which each of the measurement shown in the figures was taken.

Regarding the XLD analysis, was the procedure used to extract the XLD for FeSe the same as that for Ba122 in the authors' previous publication? If so the two analyses may be comparable, and the comparison and therefore conclusion that the behavior is distinct between the two cases meaningful. I tried to search through the previous Ba122 work to identify the regions of integration for the XLD but could not find specific limits stated, such as what energy range was used for integration. It was only noted that area near an energy ω_0 was integrated. Could the authors state this more clearly?

I also want to point out the interesting observation of non-saturating increase of XLD below T_s may be consistent with the continuous shift of the electron band observed, which also does not happen sharply at T_s .

The fact that there exists hole band splitting does not exclude the possibility that there exists bond order in combination to some ferro-orbital order. This combination would still give you a finite splitting at Gamma and a splitting that is momentum-dependent.

The conclusion that dxy should be considered in nematicity on the top of page 11 is rather abrupt. The role in which dxy participates was not discussed in the results section, namely how dxy bands change in the nematic phase.

The claim in the discussion section regarding "the reduced dxy orbital weight due to strong orbital dependent hybridization should weaken the overall magnetic instability" needs some explanation. Do you mean that the hybridization between xy and xz/yz is enhanced in the nematic phase more than the normal phase and therefore the nematic phase has reduced localized dxy orbital weight? Is there evidence for this?

Also a technical inquiry: in doing the XLD measurement on detwinned crystal, do you rotate the sample or polarization? In both cases, how do you make sure that the signal you obtain under the two geometries are comparable for subtraction, that the beamspot stays on the same portion of the sample as well as the beamspot retains the same size and intensity?

Figure 1: what happened to the hole pockets at Gamma in d and e?

Editors Comments: please make sure to go thorough your text and have a thorough text revision for both spelling, notation mistakes and style. All referees have found that the paper can be written in a more fluent way and I agree with them.

We would like to thank the reviewers for their useful comments, which made us to reconsider some details of our manuscript and helped us improve it. We revised our manuscript to reflect the comments/suggestions as much as possible. Below are our point-by-point responses to reviewers' comments, followed by the list of changes made. We put the reviewers' comments in italic followed by our reply in regular fonts.

To reviewer A

1. *I found now the description of the experiment substantially improved even if the paragraph regarding the occupation imbalance measurement can be further clarified as for example by adding the comment given as reply to Referee A criticism in (2) in the main text:*

“Regarding the point #1 above (magnitude of occupation imbalance), one cannot obtain the absolute value of the occupation imbalance from XLD; one can only get a relative magnitude. When the temperature changes from normal state to the saturation temperature of orthorhombicity (~ 50 K), XLD develops with a magnitude of around 1 %. The increase in XLD when the temperature changes from 50 to 10 K is surely from the orbital contribution and it is larger than 2 %. This observation indicates that likely the XLD is dominated by the electronic (orbital) origin, not by the tiny structural distortion.”

The authors could add the content of the reply to point (6) of Referee A to the discussion section of the main text

“However, we may offer some discussion on the issue. For the nematic band shift, it is not known why dxz and dyz bands get separated when the system goes into the nematic phase. There are theoretical suggestions such as anisotropic hopping of dxz/dyz orbitals, Pomeranchuk instability, orbital dependent correlation effect and orbital selective spin fluctuation. However, there is no consensus on this issue yet. On the other hand, orbital selective hybridization in addition to the nematic band shift could be understood by relative orbital energy level. For the normal state of FeSe, the energy level of the dxy orbital is lower than that of the dxz/yz orbital [6]. When nematic band shift occurs in the nematic phase, the dxz level gets closer to the dxy level. Therefore, a strong hybridization between dxz and dxy orbitals is expected.”*

** Add proper citations e.g. Phys. Rev. B 95, 085108 (2017) (Pomeranchuk instability), Nature Materials 17 869–874(2018) (orbital dependent correlations), Phys. Rev. B 94, 155138 (2016) (orbital selective spin fluctuations)*

Within the discussion of the absence of magnetism in FeSe, the weak-coupling point of view is not supported by any theoretical analysis. As far as I know the role of the orbital nesting in FeSe with respect to magnetism and nematicity has been explicitly addressed Phys. Rev. B 97, 121109 (2018). Please add this and any other relevant citations to support the orbital nesting argument.

We thank the reviewer for the suggestion and information. We revised manuscript and added the references as suggested.

To reviewer B

1. *The authors' reply is inconsistent with their description in Fig. 3d.*

The reviewer mentions 'inconsistency'. However, he/she is not clear on which point of our reply is inconsistent with description in Fig 3d. If it is regarding the comment #2, please see below.

2. *Here, I must apologize for my typo. "dyz and dxy bands cross each other at the Y point below T_s " should be "dxz and dxy bands ...". Anyway, the authors' claim in Fig. 3d cannot be justified.*

The reviewer's initial comment on Fig. 3d then becomes "In addition, according to Fig. 3d, the authors claim that the dxz and dxy bands cross each other at the Y point below T_s . However, this also cannot be justified from Fig. 3b. The structure at lower energy of the EDCs at the Y point seems to shift downward below T_s . This should mean that the dxy band shifts downward below T_s ." We respectfully disagree with the reviewer on the criticism based on the data in Fig. 3b for the following reasons.

First, we would like to point out that the d_{xz} and d_{xy} band do not cross each other at the Y point below T_s as shown in Fig. 3d (right hand side). If d_{xz} hole band and d_{xy} electron band cross each other as the reviewer asserts, it should be seen in the M_x and M_y cut data. Corresponding cut data are shown in Figs. 2e and 2i, respectively. The data show that d_{xz} and d_{xy} bands do not cross each other as we claimed in Fig. 3d.

Second, we would like to point out that EDCs at the Y point in Fig. 3b (now it is M_x point in cut 1) are not expected to have contribution from the d_{xy} orbital hole band (see Supplementary Information). Therefore, we did not state that the data in Fig. 3b contain d_{xy} orbital contribution.

3. *The authors' claim is not based on the experimental data analysis. The amount of the nematic band shift of each orbital (dxy, dyz, and dxz) should be evaluated from the analysis of the temperature-dependent measurements.*

We would like to point out that, contrary to what the reviewer said, our claim is based on the experimental result. While the exact amount of the nematic band shift is difficult to evaluate because nematic band shift occurs with hybridization, we can evaluate amount of the shift that include hybridization. In our case, compared to normal state band dispersion of twinned sample [1], d_{xz} , d_{yz} and d_{xy} band shifts extracted from the fitting the 30 K data are about 45, 15 and 20 meV, respectively (Fig R1).

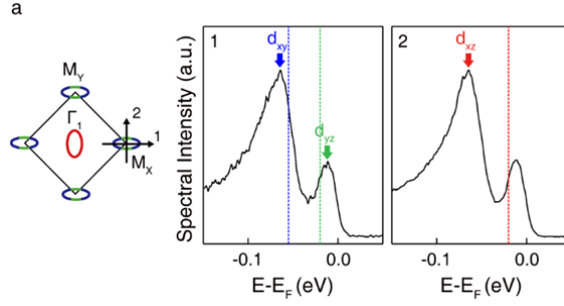


Fig R1. (a) Energy distribution curves (EDCs) at M_x point at 30 K. The schematic experimental geometry is illustrated in the left figure. The peak position of the d_{xz} , d_{yz} and d_{xy} are indicated by red, green and blue arrows, respectively. The peak position of the normal state [1] is indicated by the dashed line with same color representation.

4. *The authors' reply is unclear. The order parameter should be always temperature-dependent. The point is even if the experimental determined orthorhombicity is proportional to the order parameter, the authors cannot claim that the structural contribution to the XLD area is also proportional to the order parameter. If the structural contribution to the XLD area was proportional to the square of the orthorhombicity, the temperature dependence of the XLD area could be explained only with the structural contribution.*

As far as we understand, the reviewer is saying that there is still possibility that the XLD area can be from structural contribution only. The underlying assumption is that orthorhombicity (order parameter) is always temperature dependent, as the reviewer explicitly stated in the comment. However, orthorhombicity is almost temperature independent at low temperature as can be seen in Fig R2 (red squares for square of the orthorhombicity). We also plot XLD area and compare it with square of the orthorhombicity. Below the 40 K, square of the orthorhombicity saturates while XLD area continues to increase. This comparison clearly indicates that XLD area cannot be explained solely by the structure contribution.

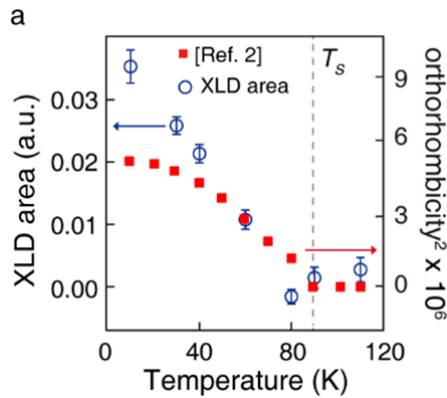


Fig R2. (a) Temperature dependence of XLD area (left, blue circle) and square of the orthorhombicity (right, red square).

5. *The authors' rebuttal whether the authors would like to emphasize the "origin" or "characteristics" also does not make sense.*

The reviewer is not specific on in what sense our rebuttal does not make sense. We therefore answer to the comment based on our best guess of the reviewer's comment.

In our rebuttal, we stated *"First, we would like to point out that we do not emphasize the different 'origin' of the nematic phase between FeSe and pnictides. Our results reveal different 'characteristics' of FeSe nematic phase, but within the same picture."* The word 'origin' meant the microscopic mechanism while 'characteristic' meant observed properties. The following is the message we wanted to convey in the previous rebuttal. Compared to pnictides, FeSe has different nematic properties such as reversed resistivity anisotropy and absence of the long-range magnetism. Through the study, we showed why the different properties ('characteristics') appear in FeSe in terms of the electronic structure. Furthermore, our results suggest that the same mechanism can explain nematic phases with different properties.

6. *This reply is also nonsense. L2,3 edge of the absorption spectra reflects the transition strength from $2p_{1/2,3/2}$ to 3d orbitals. The calculated width of the Fe 3d band does not matter. The absorption spectra should be broadened more than the width due to the core-hole lifetime. Even if the satellite feature exists above 708 eV, the XLD signal should reflect the occupancy imbalance. The region may be contributed from the different multiplet configurations due to correlation effects, but should not be extrinsic.*

We respectfully disagree with the reviewer's point *"The calculated width of the Fe 3d band does not matter. The absorption spectra should be broadened more than the width due to the core-hole lifetime."* Contrary to the reviewer's comment, the core-hole lifetime broadening is known to be about 180 meV which is much smaller than the calculated Fe 3d band width [3]. Even if XLD signal above 708 eV reflects occupancy imbalance, this region cannot give accurate information than 704 – 708 eV region due to the existence of satellite features.

7. *With the same reason described above, this reply is also inappropriate*

We believe the reviewer made the comment due to misunderstanding of what we stated in the original rebuttal. Therefore, we tried to answer reviewer's points in more detail to convey our message more clearly.

Lastly, I would like to give additional comments on this manuscript. Basically, the conclusion of this manuscript does not seem to be much different from that of the report from Y. Suzuki et al., [Ref. 12]. Even if the details are different, I think they cannot give any deep impact to most of readers. Since a new detwinning method of a piezo stack was employed in this study, the authors should try to report something new with the method. For example, if the authors investigate how the amount of detwinning depends on the voltage applied to the piezo device (and, if possible, on the applied strain), by implementing some analysis procedure to evaluate the amount, it will be a totally new result and valuable for many readers. It will be also good if the authors investigate the relation between the polarized ARPES spectra and the polarized microscope image depending on the applied voltage.

We respectfully disagree with the reviewer's subjective opinion about the impact of our work. Although the dispersion of the hole bands is the same as that reported in Y. Suzuki *et al.*, [5], our finding on the electron band provides new perspectives to the readers. An independent work that claims consistent result (lifted electron pocket) is very recently published in PRX [4], which also indicates sufficient impact on the field. Therefore, we believe that orbital occupancy imbalance and lifted electron pocket found through our work are important enough to be published in Communications Physics.

Reviewer also suggests strain dependent studies, such as investigating the amount of detwinning or the relation between ARPES spectra and polarized microscope image. From such studies, we may be able to obtain some information on continuous change from twinned domain to detwinned domain. However, such information, while interesting to some readers, is not helpful for the understanding of the electronic structure of nematic phases.

To reviewer C

Let us start this by thanking the reviewer for thorough reading of the manuscript and also for going extra distance in reviewing.

1. *First paragraph under “results”, “...elliptical pockets that are elongated along k_x - and k_y -directions with orbital characters of d_{xy} and d_{xz} (d_{yz}), respectively” I think you have them swapped, d_{xz} pocket is elongated along k_y .*

In the description for “temperature evolution of the electronic structure”, I think you have a typo regarding the direction of the d_{xz} (d_{yz}) hole band shift. At the $M_x(M_y)$ point, the d_{xz} hole band shifts DOWN as the temperature decreases. (First paragraph on page 8)

Last paragraph before the Discussion section, “An important point to note is that the orbital contribution of is positive” has a typo.

We would like to thank the reviewer for the comments. We have corrected all the mistakes the reviewer pointed out.

2. *The hybridization between xy and xz/yz is not necessarily due to orbital selectivity. The crossing between xy and yz occurs along the high symmetry direction connecting Gamma and M, while the crossing between xy and xz occurs along the BZ boundary, which is 90deg rotated from the direction of the crossing between xy and yz . These two crossings do not have the same symmetry relations between the respective orbitals.*

The crossing between d_{xy} and d_{yz} occurs along the 1st Gamma-X line (along k_x -direction) while the crossing between d_{xy} and d_{xz} occurs along the 2nd Gamma-Y line (still along k_x -direction) in 1Fe BZ scheme (please see Fig. 3d). Therefore, directions where crossing occur are parallel to each other. In other words, they have the same symmetry relations. Along these directions, d_{xz} and d_{xy} have the same parity that allows hybridization between them. Meanwhile, d_{yz} and d_{xy} have opposite parity that does not allow hybridization [4, 6].

3. *According to the data in Fig. 3, the d_{xz} electron band does not seem to disappear right at T_s , but some lower temperature, especially compared to the strain-induced elevated T_s judging from the merging of*

dxz and dyz bands at M. I think this point was not discussed at all in the manuscript, and is perhaps one of the questions one of the other referees raised about confusion of whether the pocket is there in the nematic phase. For this reason, it is important to state clearly the temperature at which each of the measurement shown in the figures was taken.

As the reviewer pointed out, the d_{xz} electron band does not disappear right at T_S . The spectral weight of d_{xz} electron band disappears between 80K and 50K as can be seen in the momentum distribution curve analysis (Fig. 3a). We note that there is a recent temperature dependent ARPES work [4] that claims that the d_{xz} electron band disappears about 30 K.

We realized that this point should be stated more clearly. Therefore, we revised our manuscript accordingly.

4. *Regarding the XLD analysis, was the procedure used to extract the XLD for FeSe the same as that for Ba122 in the authors' previous publication? If so the two analyses may be comparable, and the comparison and therefore conclusion that the behavior is distinct between the two cases meaningful. I tried to search through the previous Ba122 work to identify the regions of integration for the XLD but could not find specific limits stated, such as what energy range was used for integration. It was only noted that area near an energy ω_0 was integrated. Could the authors state this more clearly?*

Since FeSe and Ba122 are two different materials, corresponding features in XLD may appear at different energies. Therefore, for a meaningful comparison, it is important to use a regions where corresponding features exist. The energy range for the XLD integration was 702 – 707.5 eV in the previous Ba122 work (the region the dip appears in the XLD data). Similar to the case of Ba122, we analyzed the integrated XLD area in the 704 – 708 eV range for FeSe (the region the peak appears in XLD data). Therefore, we believe comparison of XLD data from Ba122 and FeSe is meaningful.

5. *I also want to point out the interesting observation of non-saturating increase of XLD below T_S may be consistent with the continuous shift of the electron band observed, which also does not happen sharply at T_S .*

We would like to thank the reviewer for pointing this out. We agree that continuous shift of d_{xz} electron band may be consistent with the non-saturating increase of XLD below T_S . On the other hand, we would like to point out that not only to the electron bands at the BZ corner but also hole bands at the BZ center should be considered to figure out the occupancy imbalance. Another careful temperature dependent APRES study for the BZ center is required to explain the non-saturating increase of XLD below T_S which we would like to leave it for a future study.

6. *The fact that there exists hole band splitting does not exclude the possibility that there exists bond order in combination to some ferro-orbital order. This combination would still give you a finite splitting at Gamma and a splitting that is momentum-dependent.*

We would like to thank the reviewer for raising the issue. As the reviewer correctly pointed out, combination of bond order and ferro-orbital order could create the momentum dependent hole band

splitting. However, the observed sign reversal hole band splitting (d_{xz} hole band located above d_{yz} at the Gamma point) may exclude the possibility the reviewer mentioned for the following reason. When ferro orbital order or bond order exists, d_{yz} hole band should be located above d_{xz} hole band [7]. However, d_{xz} hole band is located above d_{yz} hole band at the Gamma point, which cannot be explained by the combination of ferro orbital order and bond order.

7. *The conclusion that d_{xy} should be considered in nematicity on the top of page 11 is rather abrupt. The role in which d_{xy} participates was not discussed in the results section, namely how d_{xy} bands change in the nematic phase.*

We fully agree with the reviewer's point. Therefore, we added more details on the role of d_{xy} band in the nematic phase in the revised manuscript.

8. *The claim in the discussion section regarding “the reduced d_{xy} orbital weight due to strong orbital dependent hybridization should weaken the overall magnetic instability” needs some explanation. Do you mean that the hybridization between xy and xz/yz is enhanced in the nematic phase more than the normal phase and therefore the nematic phase has reduced localized d_{xy} orbital weight? Is there evidence for this?*

Evidence for the enhancement of hybridization between d_{xy} and d_{xz} in the nematic phase is the d_{xz} electron band shift. If hybridization occurs, it is expected that d_{xz} (d_{xy}) band shifts upward (downward). A previous study reports that d_{xz} electron band starts to shift upward near T_S due to hybridization [4]. Therefore, this observation indicates that hybridization between d_{xy} and d_{xz} is enhanced in the nematic phase.

We would like to point out that there is recent NMR result which supports our view of reduced d_{xy} orbital weight in the nematic phase [8].

9. *Also a technical inquiry: in doing the XLD measurement on detwinned crystal, do you rotate the sample or polarization? In both cases, how do you make sure that the signal you obtain under the two geometries are comparable for subtraction, that the beamspot stays on the same portion of the sample as well as the beamspot retains the same size and intensity?*

Reviewer's questions are related to reliability of the data. Here are details of experimental and data analysis procedures. First, we rotate the polarization in the experiment. This is to keep the beam spot on the same part of the sample as much as possible. Second, the intensity is checked before and after a scan cycle to ensure that beam stays on the same part of the sample. Third, because a beam chopper was used in front of the chamber, the beam size remained the same. Fourth, all spectra were normalized by the incident photon flux intensity measured from a gold mesh in front of the chamber. We found that these procedures allowed us to have reproducible data, with a reproducibility within the error bar. We believe the reproducibility ensures that we have reliable data.

10. *Figure 1: what happened to the hole pockets at Gamma in d and e ?*

For the simplicity, we showed schematic of the BZ corner. Considering the reviewer's question, we modified the figure and added the change in hole pockets at the BZ center in the revised manuscript.

References:

- [1] Watson, M.D. et al. Evidence for unidirectional nematic bond ordering in FeSe. *Phys. Rev. B* **94**, 201107 (2016).
- [2] Margadonna, S. et al. Crystal structure of the new FeSe_{1-x} superconductor. *Chem. Commun.* 2008, 5607-5609 (2008).
- [3] Rahn, M. C. et al. Paramagnon dispersion in β -FeSe observed by Fe *L*-edge resonant inelastic x-ray scattering *Phys. Rev. B* **99**, 014505 (2019).
- [4] Yi, M. et al. Nematic Energy Scale and the Missing Electron Pocket in FeSe. *Phys. Rev. X* **9**, 041049 (2019).
- [5] Suzuki, Y. et al. Momentum-dependent sign inversion of orbital order in superconducting FeSe. *Phys. Rev. B* **92**, 205117 (2015).
- [6] Brouet, V. et al. Impact of the two Fe unit cell on the electronic structure measured by ARPES in iron pnictides. *Phys. Rev. B* **86**, 075123 (2012).
- [7] Zhang, P. et al. Observation of two distinct d_{xz}/d_{yz} band splittings in FeSe. *Phys. Rev. B* **91**, 214503 (2016).
- [8] Li, J. et al. Spin-orbital-intertwined nematic state in FeSe. *arXiv:1903.05798* (2019).

Summary of changes made

1. The result and discussion part has been revised.
2. Fig. 1d and 1e are modified.

REVIEWERS' COMMENTS:

Reviewer #3 (Remarks to the Author):

I have read through the revised manuscript and the authors' response to all the reviewer comments. I believe that the manuscript is much improved and suitable for publication, after the authors correct the following remaining typos that I found.

Page 8, "Meanwhile, the dxz (dyz) hole band at MY (MX) shifts downward (upward)..." You got Mx and My flipped I think.

Page 8, "Next, we discuss how the electron pocket at Y in 1-Fe BZ is" delete "is".

We thank the reviewer for carefully reading our manuscript and recommending our manuscript for publication. Reviewer comments and our replies to the comments are as follows.

To reviewer C

Page 8, “Meanwhile, the dxz (d_{yz}) hole band at M_Y (M_X) shifts downward (upward)...” You got M_x and M_y flipped I think.

→ We would like to confirm that the d_{xz} hole band shifts downward at the M_Y point (on the cut along the k_x -direction) while the d_{yz} hole band shifts upward at the M_X point (on the cut along the k_y -direction). Therefore, what was originally described was correct. The confusion may have come from the fact that the M_Y point data was taken along the k_x -direction, instead of the k_y -direction.

Page 8, “Next, we discuss how the electron pocket at Y in 1-Fe BZ is” delete “is”.

→ We thank the reviewer for finding the typo. We corrected it in the revised manuscript.