

Chiral Diffraction from Aperiodic Monotile Structure

Corresponding Author: Dr Yuto Moritake

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

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors describe a diffraction experiment from an aperiodic point-set which they have designed based on the Hat tiling. It is defined by placing scatterers on the centroids of the T metatiles, it features perfect threefold rotational symmetry (unlike the Hat tiling) and lacks mirror symmetry (like the Hat tiling). The authors carried out diffraction experiments, showing well defined aperiodic Bragg peaks, pinwheel like diffraction patterns and circular polarization dependence. These experiments prove that the point-set has is aperiodic, as expected for quasicrystals, has C3 symmetry, and that it lacks mirror symmetry. The conclusions are supplemented by theoretical calculations that seem to reproduce the experiment.

I think the work is interesting and it is nice to see an experimental work based on the Hat tiling after several theoretical discussions. It is a timely work in my opinion. One of the main novelties is that "The observed circular polarisation dependence has never been observed in conventional quasicrystals". While this is not surprising given the absence of mirror symmetry, it is nice to see it working experimentally.

Here are a few comments I would like the authors to address before giving a recommendation.

1. I find the claims of a new monotile lattice a little overemphasised in the introduction, given the simplicity of the construction based on centroids.
2. I think the authors should discuss in detail the connection with the article by J. Socolar Phys. Rev. B 108, 224109 (2023). As far as I am aware, he was the first to compute the diffraction patterns of the related Key tiling and appeared at a similar time to Ref. 23 in the manuscript. It should be cited also in the main text. The pinwheel pattern in Fig. 2D,E is reminiscent of some figures in the PRB mentioned above, and I suspect their construction is related to that of Socolar. If this is so, it would be an experimental confirmation of Socolar's calculation.
3. The explanation of the "twisting" is a little scattered and a little misleading in some parts. I find the supplementary to be clearer. Specifically:
 - In line 71 the authors say that the twisting "introduces" the golden mean into the inflation factor, but this sentence is quite unclear. First, it is unclear what is being twisted with respect to what; it seems it is more rotated than twisted, which seems to be what they mean in my next comment.
 - in Line 120 the authors say: "the twist of the metatiles induces a global rotation of the entire structure, and this directionality manifest as chirality in the diffraction pattern". I do not understand how a rotation can manifest as a chirality (the absence of mirror). Please explain.
4. Many times in the main text they refer to "weak rotational symmetry" without explaining it, e.g. in line 88. In the Supplementary material they provide a succinct definition that should also be in the main text.
5. The two paragraphs of the first section of the Results have several redundant statements at different locations, and also overlap with some parts of the introduction. I would recommend the authors to group statements about symmetry together, statements about the chirality together, and so on.

6. It would be nice to see 1D cuts along different momentum lines of Fig. 2G and H to showcase better the agreement between theory and experiment. Namely, it would be more convincing to see plots of the intensity vs k for some representative cuts comparing theory and experiment. I think this will support further the claims related to these figures on the agreement between theory and experiment.

7. The findings are discussed for H-type metatiles. From the main text it seems the authors chose to work with the H_n metatiles because of the C_3 symmetry. How do their conclusions change for other types of metatiles?

In summary, I found the manuscript enjoyable and well presented. However, I was uncertain about what additional insight the experiment provides beyond what is already established theoretically. It would be helpful if the authors could argue further about the specific new understanding gained from performing this experiment. I would appreciate the opportunity to review their explanation before making a final recommendation.

Other comments:

- The a_1 and a_2 in Fig. 3A are not explained. From the supplementary information I gathered that they are the triangular lattice primitive vectors. This should be explicitly stated in the main text. Perhaps they could be already depicted in Fig. 1 B

- Please include in the caption what are the dashed and dotted lines in Fig. 3 E. This caption should explicitly mention that the dotted lines are not straight, and that they are meant to represent the "meandering" of the weak peaks.

- Line 36: However, An experimental... (misplaced capital letter)

- As the authors mention in line 74, calling a 2D lattice chiral is an abuse of notation, as chirality only exists in 3D. This should be mentioned already in the introduction.

A related comment: there are parts in the work where they mention chirality and the absence of mirror symmetry as if they were different properties (e.g. line 53). However, these are the same property.

- Line 82 I guess they mean "...rotated by $2\pi/3$ " not $2/3\pi$

Reviewer #2

(Remarks to the Author)

The focus of the manuscript is on the study of the diffraction properties of a particular aperiodic monotile structure. This structure is formed by the centroids of the unit tiles of the hat tiling. While the hat tiling has been studied in great detail by several groups, the present study of the chiral diffraction from this aperiodic monotile structure is novel and original. It certainly deserves publication after a minor revision:

(a) The term "lattice" is misused in the manuscript. According to the IUCr, a lattice is an ordered arrangement of points in space. A lattice in the vector space V_n is the set of all integral linear combinations $t = u_1 a_1 + u_2 a_2 + \dots + u_k a_k$ of a system ($a_1, a_2, \dots, a_k$) of linearly independent vectors in V_n . Thus, the term "structure" should be used instead.

(b) The authors emphasize the potential importance of some optical properties for applications. It would be helpful to explain the particular role of aperiodicity.

(c) The excellent paper on the "Diffraction of the Hat and Spectre tilings and some of their relatives" by Baake et al. (J. Math. Phys. 66, 092707 (2025)), which appeared in September this year, should be cited.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I appreciate the author's replies to my comments, which I find satisfactory.

The only minor comment I have left is that the newly added sentences:

"As shown by these dashed lines, the peak positions in k -space are in good agreement between the experiment and the calculation. It can also be seen that the relative magnitudes of the peak intensities agree well between the experiment and the calculation. These results demonstrate excellent agreement between experiment and calculation."

seem quite redundant. It seems to me this can be said in a single sentence.

Aside from this, I recommend publication.

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Response to reviewers

We sincerely thank you for your careful review and constructive suggestions. In this revision, we have carefully addressed all the points raised by the reviewers and updated the manuscript to further improve its clarity. Below is our point-by-point response to the reviewers' comments.

In addition to addressing the reviewer's comments, we have revised several sentences to clarify the logic and improve the overall presentation. Please refer to the revised manuscript in which all changes are marked in red.

We hope that our response meets your expectations.

Sincerely yours,

Yuto Moritake and Masaya Notomi

Dear Referee 1,

Comment:

I think the work is interesting and it is nice to see an experimental work based on the Hat tiling after several theoretical discussions. It is a timely work in my opinion. One of the main novelties is that “The observed circular polarisation dependence has never been observed in conventional quasicrystals”. While this is not surprising given the absence of mirror symmetry, it is nice to see it working experimentally.

Answer:

Thank you very much for reviewing our manuscript carefully and providing constructive feedback. We appreciate your positive assessment of our research.

Comment:

1. I find the claims of a new monotile lattice a little overemphasised in the introduction, given the simplicity of the construction based on centroids.

Answer:

Thank you for your comment. As you pointed out, our point set is generated by the centroids of the Smith hat tiling rather than a new lattice. Therefore, we have weakened the corresponding statements in the Introduction. In addition, in response to Referee 2’s comment (a), we decided not to use the term “lattice” and replaced it with “quasilattice” or “structure” throughout the manuscript. As we revised many parts of the manuscript, please refer to the marked-up version for the detailed changes.

Comment:

2. I think the authors should discuss in detail the connection with the article by J. Socolar Phys. Rev. B 108, 224109 (2023). As far as I am aware, he was the first to compute the diffraction patterns of the related Key tiling and appeared at a similar time to Ref. 23 in the manuscript. It should be cited also in the main text. The pinwheel pattern in Fig. 2D,E is reminiscent of some figures in the PRB mentioned above, and I suspect their construction is related to that of Socolar. If this is so, it would be an experimental confirmation of Socolar’s calculation.

Answer:

Thank you for your comment. We thank the reviewer for raising this important point. We agree that it should also have been cited in the main text. As discussed below, our diffraction results are

closely related to those reported in the PRB paper, but we do not regard the present work as a mere experimental verification of that study.

Socolar *et al.* theoretically analyzed diffraction from unit masses placed at the vertices of the related key tiling and established the quasicrystalline nature of the hat/key-tiling family through a dense set of Bragg peaks. Their diffraction patterns are organized around an overall hexagonal (C6-like) reciprocal-space skeleton. In particular, for the golden key tiling (Fig. 12 in the PRB paper), the diffraction pattern is essentially mirror-symmetric and consistent with complete hexagonal symmetry. For the generic key tiling (Fig. 9 in the PRB paper), the diffraction pattern is described as chiral. However, the chirality is mainly reflected in the intensity distribution of the peaks, while the overall arrangement of the peak positions retains a hexagonal character.

By contrast, in our monotile quasilattice, the observed pinwheel-like diffraction pattern originates from a distinct hat-derived structure with C_3 symmetry and broken mirror symmetry. In our case, the chiral character is manifested not only in the diffraction intensity but also in the geometric arrangement of the prominent diffraction features. In addition, our manuscript discusses aspects that are not treated in the PRB paper, including the polarization-dependent diffraction response and the relation between the pinwheel-like pattern and the rotational hierarchy of the metatiles.

Therefore, although the PRB paper is clearly related and provides important theoretical context, the present work is not a mere experimental reproduction of that calculation. Rather, we experimentally demonstrate a distinct chiral diffraction pattern extracted from a monotile-based structure and clarify its physical origin. We have added this reference to the main text and clarified its connection to the present study.

Added sentences

Line 125

A related theoretical study reported chiral diffraction in a hat-derived tiling system [31]. However, that work considered diffraction from a key tiling based on a nearly mirror-symmetric hexagonal framework, in which the chirality is manifested mainly in the intensity distribution of the diffraction peaks. By contrast, our quasilattice is characterized by C_3 symmetry without mirror symmetry, and the resulting diffraction exhibits a pinwheel-like arrangement.

Comment:

3. The explanation of the “twisting” is a little scattered and a little misleading in some parts. I find the supplementary to be clearer. Specifically:

- In line 71 the authors say that the twisting “introduces” the golden mean into the inflation factor, but this sentence is quite unclear. First, it is unclear what is being twisted with respect to what; it seems it is more rotated than twisted, which seems to be what they mean in my next comment.
- in Line 120 the authors say: "the twist of the metatile induces a global rotation of the entire structure, and this directionality manifest as chirality in the diffraction pattern". I do not understand how a rotation can manifest as a chirality (the absence of mirror). Please explain.

Answer:

Thank you for your comment. As you pointed out, the sentences in lines 71 and 120 were unclear. First, we replaced the expression “twist” with “rotation.” Regarding line 120, our intention was to indicate that the direction of the rotation determines the handedness of the structure, which is reflected in the shape of the diffraction pattern. However, because this wording was unclear, we removed the sentence.

Comment:

- 4. Many times in the main text they refer to “weak rotational symmetry” without explaining it, e.g. in line 88. In the Supplementary material they provide a succinct definition that should also be in the main text.

Answer:

Thank you for your comment. As you pointed out, we added the explanation from the Supplementary Information to the main text.

Added sentence

Line 89

Here, “weak” means that, when the aperiodic tiling is extended infinitely, the set of points that break the rotational symmetry has a scale smaller than the spatial dimension of the tiling.

Comment:

- 5. The two paragraphs of the first section of the Results have several redundant statements at different locations, and also overlap with some parts of the introduction. I would recommend the authors to group statements about symmetry together, statements about the chirality together, and so on.

Answer:

Thank you for your comment. Following your suggestion, we rewrote the first two paragraphs of the Results section. Please see the first three paragraphs of the Results section in the marked-up manuscript.

Comment:

6. It would be nice to see 1D cuts along different momentum lines of Fig. 2G and H to showcase better the agreement between theory and experiment. Namely, it would be more convincing to see plots of the intensity vs k for some representative cuts comparing theory and experiment. I think this will support further the claims related to these figures on the agreement between theory and experiment.

Answer:

Thank you for your constructive suggestion. Following your suggestion, we have added the diffraction intensities along selected lines in k -space to S3 in the Supplementary Information. Please see Section S3 of the Supplementary Information and Fig. S6. The comparison between the experiment and the calculation confirms that the peak positions and their relative intensities are in very good agreement.

Added sentence

S3 in Supplementary Information

Figure S6 compares the diffraction peaks obtained experimentally and numerically. Figures S6(C) and (D) show the experimental and calculated diffraction intensities, respectively, along the lines indicated in Figs. S6(A) and (B). Figures S6(C) and (D) are displayed on a logarithmic scale, and the positions of the corresponding peaks are indicated by red dashed lines. As shown by these dashed lines, the peak positions in k -space are in good agreement between the experiment and the calculation. It can also be seen that the relative magnitudes of the peak intensities agree well between the experiment and the calculation. These results demonstrate excellent agreement between experiment and calculation.

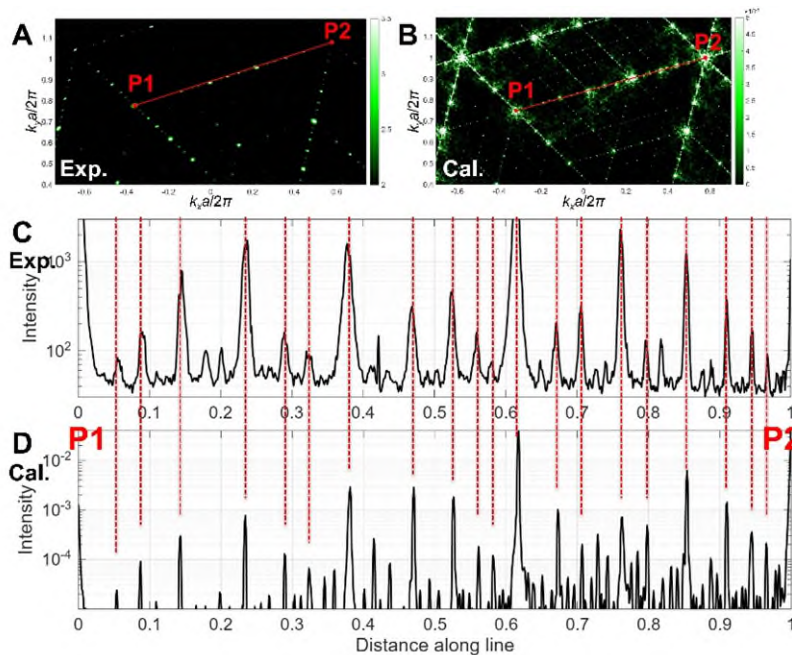


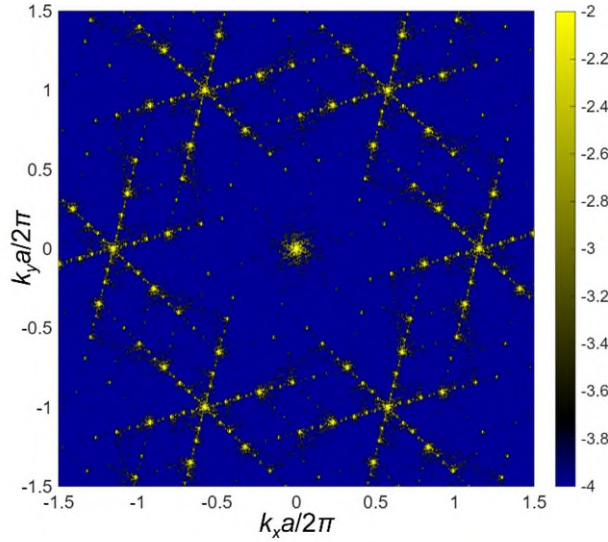
Fig. S6 Comparison of diffraction peaks between experiments and calculations. (A), (B) Lines from P1 to P2 used to extract the diffraction peaks from the experimental and calculated diffraction patterns, respectively. Panel (A) is shown on a logarithmic scale. The diffraction intensities along these lines are presented in panels (C) and (D), respectively. The red dashed lines indicate peaks that coincide between the experiment and the calculation.

Comment:

7. The findings are discussed for H-type metatiles. From the main text it seems the authors chose to work with the H_n metatiles because of the C_3 symmetry. How do their conclusions change for other types of metatiles?

Answer:

Thank you for your comment. We chose the H-metatiles because they most clearly exhibit the exact C_3 symmetry at the center of the monotile quasilattice. For other types of metatiles, we calculated the diffraction pattern for the F-metatile as an example. The diffraction pattern for the F-metatile shown in the figure below exhibits patterns similar to those of the H metatile. Even if we use a different metatile, the resulting diffraction pattern is basically the same, which is consistent with the independence of the illumination position (S2 in Supplementary Information). To explain this point, we added some sentences in the Discussion section.



Added sentence

Line 161

The essential properties of the hat-tiling structure do not depend on the specific choice of metatiles. Therefore, the chiral diffraction patterns and circular-polarization dependence observed in this study appear in hat tilings constructed from metatiles other than the H metatile.

Comment:

In summary, I found the manuscript enjoyable and well presented. However, I was uncertain about what additional insight the experiment provides beyond what is already established theoretically. It would be helpful if the authors could argue further about the specific new understanding gained from performing this experiment. I would appreciate the opportunity to review their explanation before making a final recommendation.

Answer:

Thank you for the important comment. We have added sentences to explain more explicitly what new understanding is gained from the experiment. We addressed this comment together with Referee 2's (b) comment, as they are closely related.

We found a clear helicity dependence of the diffraction pattern under circularly polarized excitation. To our knowledge, such a polarization-resolved optical response has not been established in previous theoretical studies of monotile structures. This result shows that the monotile structure is not only geometrically aperiodic and chiral, but also exhibits a nontrivial chiral optical response that becomes apparent only through experiment.

Importantly, the observed circular-polarization dependence clearly indicates that quasiperiodicity provides a new degree of freedom for breaking inversion symmetry. If delta functions are placed on a periodic lattice with C_6 symmetry, the symmetry of the system does not allow the circular-polarization dependence observed in our work. It is already well understood that such helicity dependence can emerge in valley photonic systems when inversion symmetry is broken at the level of the unit cell, reducing the symmetry from C_6 to C_3 . In contrast, in our structure, the symmetry reduction from C_6 to C_3 is realized not through a local unit cell, but through the hatching-based quasiperiodic structure applied to the honeycomb lattice as a whole. From this viewpoint, the observed circular-polarization dependence can be understood as a consequence of global symmetry reduction induced by quasiperiodicity. This demonstrates a degree of freedom unique to quasiperiodic systems for breaking inversion symmetry, and highlights a new functionality enabled by the monotile structure.

Added sentence

Line 222

If the structure has C_6 symmetry, such circular-polarization dependence does not appear unless chiral structures are placed at the lattice points. To induce circular-polarization dependence, it is necessary to break inversion symmetry and lower the symmetry of the unit cell from C_6 to C_3 , as in valleytronic systems. In contrast, in our quasicrystalline structure, we globally reduce the

overall symmetry of the structure to C_3 by introducing quasiperiodic defects into a honeycomb lattice, providing a new method for realizing circular polarization dependence.

Line 238

Such circular-polarization-dependent diffraction has not been identified in previous theoretical studies and is revealed here for the first time through our optical experiments.

Comment:

- The a_1 and a_2 in Fig. 3A are not explained. From the supplementary information I gathered that they are the triangular lattice primitive vectors. This should be explicitly stated in the main text. Perhaps they could be already depicted in Fig. 1 B
- Please include in the caption what are the dashed and dotted lines in Fig. 3 E. This caption should explicitly mention that the dotted lines are not straight, and that they are meant to represent the “meandering” of the weak peaks.
- Line 36: However, An experimental... (misplaced capital letter)
- As the authors mention in line 74, calling a 2D lattice chiral is an abuse of notation, as chirality only exists in 3D. This should be mentioned already in the introduction.
A related comment: there are parts in the work where they mention chirality and the absence of mirror symmetry as if they were different properties (e.g. line 53). However, these are the same property.
- Line 82 I guess they mean “...rotated by $2\pi/3$ ” not $2/3\pi$

Answer:

Thank you for your comment. We added the vectors a_1 and a_2 to Fig. 1B. We also added an explanation of the dashed and dotted lines in Fig. 3 to the figure caption. In addition, we revised the typos and moved the definition of chirality to the Introduction. Please see the marked-up version for the changes.

Dear Referee 2,

Comment:

The focus of the manuscript is on the study of the diffraction properties of a particular aperiodic monotile structure. This structure is formed by the centroids of the unit tiles of the hat tiling. While the hat tiling has been studied in great detail by several groups, the present study of the chiral diffraction from this aperiodic monotile structure is novel and original. It certainly deserves publication after a minor revision:

Answer:

Thank you very much for reviewing our manuscript carefully and providing constructive feedback. We appreciate your high evaluation of our research and recommendation for publication in Nature Communications.

Comment:

(a) The term "lattice" is misused in the manuscript. According to the IUCr, a lattice is an ordered arrangement of points in space. A lattice in the vector space V_n is the set of all integral linear combinations $t = u_1a_1 + u_2a_2 + \dots + u_ka_k$ of a system $(a_1, a_2, \dots, a_k)$ of linearly independent vectors in V_n . Thus, the term "structure" should be used instead.

Answer:

Thank you for your comment. Following your suggestion, we changed the title from "lattice" to "structure." We also replaced "lattice" with "quasilattice" because our point set shows quasiperiodic order. In our view, "quasilattice" is more appropriate than "structure" for describing our point set because an essential aspect of our system is that the point set is defined using the centroids of the hat tiling. To explain this point, we added the following sentences.

Added sentence

Line 42

In this work, we use the term quasilattice to denote a non-periodic point set with quasiperiodic order. We therefore refer to the present centroid-based point set as the monotile quasilattice because it exhibits long-range quasiperiodic order.

Comment:

(b) The authors emphasize the potential importance of some optical properties for applications. It would be helpful to explain the particular role of aperiodicity.

Answer:

Thank you for this helpful comment. We addressed this comment together with Referee 1's final comment, as they are closely related. We agree that the original manuscript did not clearly explain the role of aperiodicity in relation to the possible optical applications mentioned in the Conclusion. In response, we revised the relevant part to clarify that the significance of aperiodicity here lies not merely in the absence of translational periodicity, but in the additional structural degree of freedom it provides for generating rich reciprocal-space features and symmetry-dependent optical responses while maintaining deterministic long-range order.

For example, if delta functions are placed on a periodic point lattice with C_6 symmetry, the circular-polarization dependence observed in the present work does not appear. (Of course, if some chiral structure is placed at each lattice point instead of a delta function, circular-polarization dependence may arise.) In valleytronic systems, circular-polarization dependence can emerge when inversion symmetry is broken, typically by lowering the symmetry of the unit cell from C_6 to C_3 . In our structure, by contrast, the symmetry reduction from C_6 to C_3 can be regarded as being realized not at the level of the unit cell, but through the quasiperiodic structure as a whole. We believe that this is the origin of the observed circular-polarization dependence. In this sense, our system provides a symmetry-breaking mechanism that is clearly distinct from conventional periodic structures, and may be viewed as a new design degree of freedom enabled by quasiperiodicity.

Added sentence

Line 222

If the structure has C_6 symmetry, such circular polarization dependence does not appear unless chiral structures are placed at the lattice points. To induce circular polarization dependence, it is necessary to break inversion symmetry and lower the symmetry of the unit cell from C_6 to C_3 , as in valleytronic systems. In contrast, in our quasicrystalline structure, we globally reduce the overall symmetry of the structure to C_3 by introducing quasiperiodic defects into a honeycomb lattice, thereby providing a new method for realizing circular polarization dependence.

Line 245

In this context, the aperiodicity provides an additional structural degree of freedom, enabling rich reciprocal-space features and symmetry-dependent optical responses that are difficult to realize in conventional periodic lattices. Thus, the monotile point set may offer a new platform for exploring such optical phenomena in deterministic aperiodic structures.

Comment:

(c) The excellent paper on the "Diffraction of the Hat and Spectre tilings and some of their

relatives" by Baake et al. (J. Math. Phys. 66, 092707 (2025)), which appeared in September this year, should be cited.

Answer:

Thank you for information about an important paper. Following your suggestion, we have cited the paper.

Response to reviewers

Dear Referee 1,

We sincerely thank you for your careful review and constructive comments. According to your comment, we revised the sentences as follows:

Comment:

“As shown by these dashed lines, the peak positions in k-space are in good agreement between the experiment and the calculation. It can also be seen that the relative magnitudes of the peak intensities agree well between the experiment and the calculation. These results demonstrate excellent agreement between experiment and calculation.” seem quite redundant. It seems to me this can be said in a single sentence.

Revised sentence

Page 7 in Supplementary Information

As indicated by the dashed lines, the experimental and calculated k-space peak positions, as well as the relative magnitudes of the peak intensities, show excellent agreement.

We thank you again for reviewing our manuscript.

Sincerely yours,

Yuto Moritake and Masaya Notomi