nature portfolio

Peer Review File

Three-Dimensional Imaging of Topologically Protected Strings in a Multiferroic Nanocrystal

Corresponding Author: Dr Marcus Newton

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

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Dear Dr Newton,

Thank you for submitting your manuscript, "Three-Dimensional Imaging of Topologically Protected Strings in a Multiferroic Nanocrystal", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of potential interest, they have raised substantial concerns that must be addressed.

In particular, the reviewers are highlighting omissions of key references in the field, misleading statements and discussions regarding the Kibble-Zurek mechanism, and the need of a more comprehensive and accurate analysis. In light of these comments, we cannot accept the manuscript for publication, but are interested in considering a revised version that addresses these serious concerns.

We hope you will find the referees' comments useful as you decide how to proceed. Should further experimental analysis and discussion allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. However, please bear in mind that we will be reluctant to approach the referees again in the absence of major revisions. If the revision process takes significantly longer than three months, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type ‘Revised Manuscript - Marked Up’ when uploading the manuscript file to our online system.

-A clean version of the manuscript. Please select the file type ‘Article File’.

-An updated Editorial Policy checklist, uploaded as a ‘Related Manuscript File’ type. This checklist is to ensure your paper complies with all relevant editorial policies. If needed, please revise your manuscript in response to these points. Please note that this form is a dynamic ‘smart pdf’ and must therefore be downloaded and completed in Adobe Reader. Clicking this link will download a zip file containing the pdf.

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** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

Please do not hesitate to contact me if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Andreja Bencan Golob, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-2116-3779

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The major claim of the paper is that it describes the first three-dimensional, in-situ imaging of 1D topological defects in a multiferroic nanocrystal, which is achieved via Bragg CDI (i.e. x-ray microscopy). I question the broader importance of this, since very few multiferroics are known to contain 1D topological defects (R-MnO3 as far as I am aware), however the distribution and local structure of these defects in R-MnO3 is certainly an important issue to those studying these systems, so there is no doubt this is a valuable result.

The data presented in the paper is unquestionably beautiful and a credit to the authors. However, I have several major reservations about the paper itself:
1. This is not the first 3D imaging of the domain structure in R-MnO3 (see e.g. https://doi.org/10.1002/adma.202202614, though there may be others before this). Although BCDI is a very elegant solution for this, the fact that the domain structures are so stable means that they naturally lend themselves to mapping via destructive methods e.g. FIB-SEM tomography, which is typically more accessible than BCDI.
2. It is repeatedly claimed that this is an in-situ measurement which, as far as I can tell, it isn't. True, BCDI is non-destructive, but this is only relevant if the crystal is manipulated during the experiment (e.g. heating/cooling or electrical/mechanical/magnetic loading). The potential to do such in-situ experiments is obviously valuable, but I would imagine this is not trivial (since it seldom is. even with macroscopic crystals!).
3. The discussion of the results is virtually non-existent, so it's difficult for anyone reading the paper to learn much from it, other than that these types of measurements are feasible.

Consequently, the paper reads as if it is unfinished and/or hastily prepared.

Should the authors wish to finish/improve the manuscript, I would suggest addressing the following points:
- Narrow the scope of the title to something that befits the novelty of the results.
- Address the use of the term "in-situ" by either including data collected in-situ, or by removing the term entirely
- Appropriately comparing the results to those achieved previously via e.g. FIB-SEM and other methods.
- Include the valences when referring to ions (e.g. line 27-30)
- Proper citations in key places e.g. L52 "[BCDI] is able to reveal the ferroelectric domain structure in three dimensions" - this is not trivial and as far as I know is not a commonplace result for BCDI
- Consider explaining in the "Bragg CDI Synopsis" that "phase" refers to the optical phase of the x-rays, as this may not be obvious to e.g. a reader from the multiferroics field.
- Fix the typo in the abstract (L20) "nanocorystal"
- Provide a reference to the instrumentation used (e.g. I have no idea what a quad Merlin detector is, though I guess it's something developed in-house at Diamond).
- Provide a proper discussion of the results. How do they compare to e.g. FIB-SEM? What are the limitations for BCDI in this context? How about the effect of dense elements (e.g. Dy) in producing dynamical scattering that may influence the reconstruction?
- Similarly, consider analyzing the strain data. Your BCDI approach is unique because you measure lattice strain (which SEM and AFM-based methods cannot). There is considerable speculation regarding the degree of strain and and ionic displacement associated with both the improper domain walls and the 1D defects at their meeting points, and I am quite certain you are the first in the world to directly measure this. I imagine you should be able to compare the strain fields you measured to the DFT-based models published by Spaldin et al (or better yet, ask her for calculations of lattice displacements along the Q-vectors you measured so that you can compare them directly). In my opinion, this would be considerably more significant than just the demonstration of the approach as described in this paper.

Reviewer #2 (Remarks to the Author):

In the paper "Three-Dimensional Imaging of Topologically Protected Strings in a Multiferroic Nanocrystal", Mansoor and co-authors performed Bragg coherent X-ray diffraction imaging experiments to study multiferroic strings in DyMnO3 crystals. Multiple Bragg peaks from the same crystal were used to obtain strain tensors. Predicted 1D topologically protected strings within DyMnO3 crystals were imaged by Bragg CDI. This paper presents a new approach to understanding topologically protected defects in materials. The manuscript is well written and well organized. Therefore, I recommend publishing this paper.
1. As shown in Figure 1, the paraelectric phase is caused by the displacement of oxygen ions within the unit cell. However, Bragg CDI usually does not have atomic resolution and can only see modulated crystal planes. Not only these paraelectric phases, but also crystal defects can lead to modulation of crystal planes. Therefore, the authors would be better to add some notes in the paper on how these strain images are related to the paraelectric phases.
2. For Bragg CDI, the method does not converge well and does not give reproducible results when the phase of the crystal is strong. Figure 2 shows how reproducible these reconstructed images are. For example, does the phase procedure give the same results when running the reconstruction twice?
3. Did the authors try it on the strain tensor when estimating the path distribution function analysis? It would be more reasonable to do it on the strain tensor instead of the (111) Bragg peak.
4. Since these strain images are related to the crystal orientation, I suggest that the authors add the corresponding Q vector direction or crystal orientation to the figures in the manuscript.

Reviewer #3 (Remarks to the Author):

I find this article intriguing for its experimental advancements in imaging vortex lines in 3D. However, I must express significant concerns regarding the discussion of the Kibble-Zurek mechanism (KZM), which is both misleading and incorrect. Given these issues, I cannot recommend the article for publication in its current form, but a substantial revision could potentially address the concerns raised.

The manuscript presents data on vortex line formation following a cooling process at a single fixed rate. This limitation makes any comparison with the KZM problematic. KZM cannot predict the non-equilibrium correlation length with precision, as a fudge factor is typically required for such estimates (KZM correctly captures the scaling with the quench time). This important limitation of KZM is not acknowledged in the current work, which instead claims that the measured correlation length is well accounted for by KZM. Such statements are particularly gratuitous given that the work explores PhDf, which has not been previously analyzed in connection to KZM. The statements made in lines 152-153 lack supporting evidence (e.g., what value was estimated?), and the assertion lacks a solid foundation (KZM does not accurately estimate such values). Similarly, the claims in lines 17-19 of the abstract are misleading, as the current manuscript does not provide an advance in that regard.

Moreover, the manuscript cites Eq. (1), which, in the context of vortex strings, is incorrect. The proper expression can be found in Eq. (10) of Int. J. Mod. Phys. A 29, 1430018 (2014).

Furthermore, the manuscript overlooks seminal works that examine the KZM in multiferroics, such as Nature Physics 10, 970 (2014), which predate references [11-13] in the current manuscript. These omissions assign undue priority to refs. [11-13]. More recent advances, like those in Nature Physics 19, 1495 (2023), which also focus on extended defects, are also missing. This latter work is particularly relevant as an example of imaging 1D defects (domain walls) in condensed matter systems, questioning the claim of novelty in lines 17-19 in the abstract.

Additionally, the manuscript could acknowledge prior research on KZM and U(1) vortex strings, such as the work published in Phys. Rev. Lett. 82, 2824 (1999). Similarly, the study of defect spatial statistics by spacing distributions, of which the PthDF is a generalization to vortex lines, has been advanced in recent references such as Phys. Rev. B 106, L140101 (2022), Phys. Rev. Res. 6, 033152 (2024).

There is a minor typographical error in the affiliations: the second and third affiliations are both labeled with the number "2," which should be corrected.

they must do so prior to acceptance.
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Version 1:

Decision Letter:

** Please ensure you delete the link to your author homepage in this e-mail if you wish to forward it to your coauthors **

Dear Dr Newton,

Your manuscript titled "Three-Dimensional Imaging of Topologically Protected Strings in a Multiferroic Nanocrystal" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials.

We therefore invite you to edit your manuscript to comply with our journal policies and formatting style in order to maximise the accessibility and therefore the impact of your work.

EDITORIAL REQUESTS
* Your manuscript should comply with our policies and format requirements, detailed in our style and formatting guide (https://www.nature.com/documents/commsj-phys-style-formatting-guide-accept.pdf).

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* The editorial requests table also includes a full list of the files that must be provided upon resubmission. Please upload your files according to this table.

* An updated editorial policy checklist that verifies compliance with all required editorial policies must be completed and uploaded with the revised manuscript. All points on the policy checklist must be addressed; if needed, please revise your manuscript in response to these points. Please note that this form is a dynamic 'smart pdf' and must therefore be downloaded and completed in Adobe Reader. Clicking this link will download a zip file containing the pdf.

Editorial policy checklist: https://www.nature.com/documents/nr-editorial-policy-checklist.zip

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** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to hear from you within two weeks; please let us know if the process may take longer.

Best regards,

Andreja Bencan Golob, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-2116-3779

REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

The authors have addressed all my comments. I recommend the publication of the paper.

Reviewer #3 (Remarks to the Author):

The authors have revised the manuscript, addressing all the concerns I raised in the first version. In particular, the new text focuses on the experimental advancements in imaging vortex lines in 3D without overstating the results. I thus recommend the publication of the manuscript in its current form.

Dear Editor,

Thank you for the opportunity to resubmit our manuscript after corrections. We would like to thank the reviewers for their positive and constructive feedback. All reviewer comments were addressed in the revised manuscript. Please find below a point-by-point response to each of the reviewer's comments. Specifically, Blue text indicates our responses to the reviewers' comments, while red text highlights the changes made in the manuscript.

Reviewer #1 (Remarks to the Author):

The major claim of the paper is that it describes the first three-dimensional, in-situ imaging of 1D topological defects in a multiferroic nanocrystal, which is achieved via Bragg CDI (i.e. x-ray microscopy). I question the broader importance of this, since very few multiferroics are known to contain 1D topological defects (R-MnO3 as far as I am aware), however the distribution and local structure of these defects in R-MnO3 is certainly an important issue to those studying these systems, so there is no doubt this is a valuable result.

The authors would like to thank the reviewer for noting the valuable results contained within our paper.

The data presented in the paper is unquestionably beautiful and a credit to the authors. However, I have several major reservations about the paper itself:

1. This is not the first 3D imaging of the domain structure in R-MnO3 (see e.g. https://doi.org/10.1002/adma.202202614, though there may be others before this). Although BCDI is a very elegant solution for this, the fact that the domain structures are so stable means that they naturally lend themselves to mapping via destructive methods e.g. FIB-SEM tomography, which is typically more accessible than BCDI.

Thank you for sharing a link to this beautiful study on 3D imaging of the domain structure in R-MnO3. We have now added this reference to the manuscript. While it may be possible to image domain structure via destructive means such as FIB-SEM based methods, non-destructive alternatives such as BCDI are complementary and advantageous when destructive means become impractical. For example, when high resolution down to a single nanometer is required, in all three spatial dimensions. This could become important when attempting to image topological strings with higher density, quenched at a more rapid rate. In addition, BCDI is able to measure strain which is generally not possible with destructive methods.

2. It is repeatedly claimed that this is an in-situ measurement which, as far as I can tell, it isn't. True, BCDI is non-destructive, but this is only relevant if the crystal is manipulated during the experiment (e.g. heating/cooling or electrical/mechanical/magnetic loading). The potential to do such in-situ experiments is obviously valuable, but I would imagine this is not trivial (since it seldom is. even with macroscopic crystals!).

Apologies for the poor choice of words. We have replaced "in-situ" with "direct[ly]" or "non-destructive" throughout the manuscript, where appropriate.

3. The discussion of the results is virtually non-existent, so it's difficult for anyone reading the paper to learn much from it, other than that these types of measurements are feasible.

Consequently, the paper reads as if it is unfinished and/or hastily prepared.

Extensive discussions were added to the manuscript, featuring comparison with earlier FIB-SEM based methods. See below. The main point however was to demonstrate the BCDI method and its application to the study of topological structure.

Should the authors wish to finish/improve the manuscript, I would suggest addressing the following points:
- Narrow the scope of the title to something that befits the novelty of the results.

Thank you for the suggestion to revise the title. As we have successfully imaged topologically protected strings in a multiferroic nanocrystal and in three-dimensions, the existing title seems appropriate to us. We however welcome further suggestions.

- Address the use of the term "in-situ" by either including data collected in-situ, or by removing the term entirely

We have opted to remove the term entirely and replace it with “direct[ly]” or “non-destructive”, where appropriate.

- Appropriately comparing the results to those achieved previously via e.g. FIB-SEM and other methods.

We have added discussions where the results are compared with FIB-SEM and other methods. This reads:
Earlier studies aimed at revealing three-dimensional domains, domain walls, and multiferroic strings have employed destructive means such as sequential Focussed Ion Beam (FIB) cutting of cross-sections of bulk multiferroic materials. Subsequent imaging of these slices was performed using either Scanning Electron Microscopy (SEM), piezoresponse force microscopy (PFM) or conductive atomic force microscopy (cAFM). The resulting images were used to successfully reveal domain and domain wall contrast. SEM, PFM and cAFM measurements however were unable to reveal the winding direction of the vortex structure, which is readily observed when using Bragg CDI. While the robust nature of topologically protected structure lends itself to destructive mapping and imaging techniques, such measurements have the disadvantage of lacking repeatability. Bragg CDI however offers non-destructive imaging where measurements can be repeated on the same sample provided the dimensions of the material are below the coherence volume of the coherent x-ray probe. When this isn’t the case, alternative and more challenging approaches such as Bragg ptychography can in principle provide equivalent information.
Hexagonal manganite multiferroics often contain dense elements such as dysprosium that can produce dynamical scattering effects. While in principle this may influence the BCDI phase reconstruction process, dynamical scattering effects are often found to be less significant than refraction and absorption effects and therefore generally do not warrant further consideration.
Bragg CDI also provides strain information and has revealed in this study that multiferroic strings, which remain in the paraelectric phase, induce tensile and compressive strain of upto 0.1 along commensurate paths, with magnitude that often exceeds that of strain within the broader domain regions. Future BCDI studies on multiferroic strings where the effect strain is studied dynamically could provide further insight into the role of strain in the formation of multiferroic strings.

- Include the valences when referring to ions (e.g. line 27-30)

Ion valencies are now added, as requested.

- Proper citations in key places e.g. L52 "[BCDI] is able to reveal the ferroelectric domain structure in three dimensions" - this is not trivial and as far as I know is not a commonplace result for BCDI

Citations for BCDI of ferroelectric domains added:
1) Karpov, D., Liu, Z., Rolo, T. et al. Three-dimensional imaging of vortex structure in a ferroelectric nanoparticle driven by an electric field. Nat Commun 8, 280 (2017).
2) Jiecheng Diao, Xiaowen Shi, Tadesse A. Assefa, et al., Phys. Rev. Materials 4, 106001 (2020).
3) Mokhtar, A.H., Serban, D., Porter, D.G. et al. Three-dimensional domain identification in a single hexagonal manganite nanocrystal. Nat Commun 15, 3587 (2024).

- Consider explaining in the "Bragg CDI Synopsis" that "phase" refers to the optical phase of the x-rays, as this may not be obvious to e.g. a reader from the multiferroics field.

The following addition text was appended to further clarify this point:
"… present within the x-rays that is acquired when coherently scattered from the nanocrystal"

- Fix the typo in the abstract (L20) "nanocorystal"

This typo is now fixed.

- Provide a reference to the instrumentation used (e.g. I have no idea what a quad Merlin detector is, though I guess it's something developed in-house at Diamond).

The following reference was added to the manuscript:
1) R Plackett et al., Journal of Instrumentation, Volume 8,
C01038 (2013)

- Provide a proper discussion of the results. How do they compare to e.g. FIB-SEM? What are the limitations for BCDI in this context? How about the effect of dense elements (e.g. Dy) in producing dynamical scattering that may influence the reconstruction?
Discussions on the results, how they compare to equivalent FIB-SEM studies, and other points raised above by the reviewer were added (see above).
[see above for discussion text]

Regarding dynamical scattering, such effects are generally found to be less significant than refraction and absorption effects, and therefore generally do not warrant further consideration. This is discussed, for example, in the following reference: Phys. Rev. B 106, 184111 (2022). The following text was added to the discussions to address this point:

Hexagonal manganite multiferroics often contain dense elements such as dysprosium that can produce dynamical scattering effects. While in principle this may influence the BCDI phase reconstruction process, dynamical scattering effects are often found to be less significant than refraction and absorption effects and therefore generally do not warrant further consideration.

- Similarly, consider analyzing the strain data. Your BCDI approach is unique because you measure lattice strain (which SEM and AFM-based methods cannot). There is considerable speculation regarding the degree of strain and and ionic displacement associated with both the improper domain walls and the 1D defects at their meeting points, and I am quite certain you are the first in the world to directly measure this. I imagine you should be able to compare the strain fields you measured to the DFT-based models published by Spaldin et al (or better yet, ask her for calculations of lattice displacements along the Q-vectors you measured so that you can compare them directly). In my opinion, this would be considerably more significant than just the demonstration of the approach as described in this paper.

Thank you for the suggestion to perform DFT calculations for strain comparison. Unfortunately it is currently not possible to perform a DFT calculation of a vortex defect due to limitations of the unit cell size.

Reviewer #2 (Remarks to the Author):

In the paper "Three-Dimensional Imaging of Topologically Protected Strings in a Multiferroic Nanocrystal", Mansoor and co-authors performed Bragg coherent X-ray diffraction imaging experiments to study multiferroic strings in $DyMnO_3$ crystals. Multiple Bragg peaks from the same crystal were used to obtain strain tensors. Predicted 1D topologically protected strings within $DyMnO_3$ crystals were imaged by Bragg CDI. This paper presents a new approach to understanding topologically protected defects in materials. The manuscript is well written and well organized. Therefore, I recommend publishing this paper.

1. As shown in Figure 1, the paraelectric phase is caused by the displacement of oxygen ions within the unit cell. However, Bragg CDI usually does not have atomic resolution and can only see modulated crystal planes. Not only these paraelectric phases, but also crystal defects can lead to modulation of crystal planes. Therefore, the authors would be better to add some notes in the paper on how these strain images are related to the paraelectric phases.

Thank you for your positive comments, view that the manuscript is well written and recommendation for publication. The h-DMO nanocrystals are in the ferroelectric phase at room temperature. We have added the following discussions to the manuscript on how the strain relates to the ferroelectric phase:

Bragg CDI also provides strain information and has revealed in this study that the strings, which are lines of the paraelectric phase trapped in the ferroelectric structure, induce tensile and compressive strain of upto 0.1 along commensurate paths, with magnitude exceeding that of strain within the broader domain regions. Future BCDI studies on multiferroic strings where the effect strain is studied dynamically, via progressive deformation, [Phys. Rev. Materials 3, 043803 (2019)] could provide further insight into the role of strain in the formation of multiferroic strings

2. For Bragg CDI, the method does not converge well and does not give reproducible results when the phase of the crystal is strong. Figure 2 shows how reproducible these reconstructed images are. For example, does the phase procedure give the same results when running the reconstruction twice?

Convergence of the algorithm can become an issue when phase retrieval is performed for a single dataset from a single Bragg reflection. In this case however, phase retrieval was carried out concurrently for multiple Bragg reflections from the same nanocrystal. In this case, convergence of the phase retrieval algorithm is much stronger and highly reproducible. Please see the following reference, and articles that cite it, for further details:
1) Newton, M. C. Concurrent phase retrieval for imaging strain in nanocrystals. Phys. Rev. B 102,

014104 (2020).

We have added the following text to the manuscript to clarify this point:

Concurrent phase retrieval was previously shown to provide stronger convergence and high reproducibility than independent phase retrieval due to inherent redundancy in the concurrent approach.

3. Did the authors try it on the strain tensor when estimating the path distribution function analysis? It would be more reasonable to do it on the strain tensor instead of the (111) Bragg peak.

Intuition would suggest that calculating the path distribution function (PthDF) would be easier on the strain tensor data. This was also our initial assumption. The strain tensor data has six components each of which provide strain information. To calculate the PthDF would require extraction of topological strings from each tensor component and subsequent recombination of string information from each into a single image. This is equivalent to simply extracting the strings from a single reflection where phase vortices are clearly visible (i.e. one with a non-zero hexagonal c-axis Miller index).

4. Since these strain images are related to the crystal orientation, I suggest that the authors add the corresponding Q vector direction or crystal orientation to the figures in the manuscript.

Crystal orientations are present in figures 2, 3 and 4. Strain information in figure 4 is shown at the same orientation as the information shown in figure 3. This is indicated in the caption of figure 4.

Reviewer #3 (Remarks to the Author):

I find this article intriguing for its experimental advancements in imaging vortex lines in 3D. However, I must express significant concerns regarding the discussion of the Kibble-Zurek mechanism (KZM), which is both misleading and incorrect. Given these issues, I cannot recommend the article for publication in its current form, but a substantial revision could potentially address the concerns raised.

The manuscript presents data on vortex line formation following a cooling process at a single fixed rate. This limitation makes any comparison with the KZM problematic. KZM cannot predict the non-equilibrium correlation length with precision, as a fudge factor is typically required for such estimates (KZM correctly captures the scaling with the quench time). This important limitation of KZM is not acknowledged in the current work, which instead claims that the measured correlation length is well accounted for by KZM. Such statements are particularly gratuitous given that the work explores PhDf, which has not been previously analyzed in connection to KZM. The statements made in lines 152-153 lack supporting evidence (e.g., what value was estimated?), and the assertion lacks a solid foundation (KZM does not accurately estimate such values). Similarly, the claims in lines 17-19 of the abstract are misleading, as the current manuscript does not provide an advance in that regard.

The authors thank the reviewer for the insightful comments. We have removed the statements made in lines 152-153 and replaced them with the following:

The corresponding area density of strings, (n), is estimated from the preferred string separation and found to agree with those previously reported for the same cooling rate in bulk samples. From this we can infer that there are likely no fundamental changes in domain structure between our nanoscale structure and bulk samples.

We have also revised the claims in lines 17-19 by removing “to demonstrate Kibble-Zurek equivalence and scaling” and have also removed similar statements on other locations.

Moreover, the manuscript cites Eq. (1), which, in the context of vortex strings, is incorrect. The proper expression can be found in Eq. (10) of Int. J. Mod. Phys. A 29, 1430018 (2014).

The authors thank the reviewer for identifying the correct expression. This now features in the revised manuscript.

Furthermore, the manuscript overlooks seminal works that examine the KZM in multiferroics, such as Nature Physics 10, 970 (2014), which predate references [11-13] in the current manuscript. These omissions assign undue priority to refs. [11-13]. More recent advances, like those in Nature Physics 19, 1495 (2023), which also focus on extended defects, are also missing. This latter work is particularly relevant as an example of imaging 1D defects (domain walls) in condensed matter systems, questioning the claim of novelty in lines 17-19 in the abstract.

We have now incorporated the suggested references in the manuscript. The claim to novelty in lines 17-19 was revised to clarify that we are utilising a direct imaging approach.

Additionally, the manuscript could acknowledge prior research on KZM and U(1) vortex strings, such as the work published in Phys. Rev. Lett. 82, 2824 (1999). Similarly, the study of defect spatial statistics by spacing distributions, of which the PthDF is a generalization to vortex lines, has been advanced in recent references such as Phys. Rev. B 106, L140101 (2022), Phys. Rev. Res. 6, 033152 (2024).

The authors thank the reviewer for pointing out additional prior research. We have incorporated the above suggested references and also included a reference to the pair distribution function (PDF).

There is a minor typographical error in the affiliations: the second and third affiliations are both labeled with the number "2," which should be corrected.

The typographic error in the affiliation index was corrected.

Dear Editor,

We are delighted to learn that you are happy, in principle, to publish a suitably revised version of our manuscript in Communications Materials.

We would like to thank the reviewers for their positive and constructive feedback. As the reviewers have suggested no further changes, we have prepared a revised manuscript with the required editorial changes only, as outlined in the Editorial Requests Table. Reviewer comments are shown below for clarity.

Reviewer #2 (Remarks to the Author):

The authors have addressed all my comments. I recommend the publication of the paper.

Reviewer #3 (Remarks to the Author):

The authors have revised the manuscript, addressing all the concerns I raised in the first version. In particular, the new text focuses on the experimental advancements in imaging vortex lines in 3D without overstating the results. I thus recommend the publication of the manuscript in its current form.