nature portfolio

Peer Review File

Self-organization of ferroelectric domains induced by water and reinforced via ultrasonic vibration

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality

3rd Feb 23

Dear Professor Lu,

Thank you for submitting your manuscript, "Self-organization of ferroelectric domains induced by water and reinforced via ultrasonic vibration", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that they give mixed recommendations, and raise a number of technical concerns. In light of these comments, we cannot accept the manuscript for publication. It is not clear to us whether you will be able to convince the referees, but we may be willing to send a substantially revised manuscript back to the referees for further comment.

You will see in particular that Reviewer 2 and 3 both raise issues with the claimed underpinning mechanism, mentioning several aspects that they feel are not sufficiently convincing, and request further data to understand this. Furthermore, Reviewer 1 was unable to replicate these findings in their own experiments. Whether or not you can convincingly address these comments is not something we can determine at the moment. Indeed, it might be in your best interest to submit the paper to another journal and not continue the submission process with us.

However, should further experimental data allow you to address these criticisms we may be willing to look at a substantially revised manuscript, but 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.

Please use the following link to submit your revised manuscript files:

[link redacted]

** 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 understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

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):

Dear authors,
I do not recommend this paper for publication in Communications Materials. Please see my comments below:
1) Self-organization is not an appropriate term for the results described. Therefore, the introduction is not properly written because it describes self-organization in nature in general, which is not the subject of this paper.
2) The work is not new; a similar topic has already been described by authors about the BiFeO3. The BiFeO3 is much more sensitive to moisture compared to the Pb(Mg1/3Nb2/3)O3-PbTiO3, so the influence of water on the change of the domain pattern is more credible.
3) My biggest concern with this work is that after I received this work for review, we repeated the exact same experiment with Pb(Mg1/3Nb2/3)O3-PbTiO3 bulk material with composition near the morphotropic phase boundary and did not get the same result. In fact, we immersed the sample in the water for 1 hour and 25 hours, and then we performed the PFM study of the domain patterns. No changes in the domain structure were observed. Therefore, I doubt that this experiment is repeatable.

4) Is the same area always seen in figures 1 and 3? The information is limited because only the PFM amplitude image is shown. What about the topography, from which we can judge if the area is really the same? Also, the PFM phase images should be added. A similar comment applies to figures S1-S5.

Reviewer #2 (Remarks to the Author):

The paper reports a self-organization process of domain structures in the PMN-0.28PT system when subjected to water, which can be further accelerated by the ultrasonic vibration. The interaction between the screening charges in water, i.e., H+ and OH-, and the bound charges is believed to be the origin of the observed switching and coarsening phenomenon. A transition point is shown in the percolation model, and this indicates a non-conservative nature of the system. This paper for the first time shows that in addition to low dimensional ferroelectrics such as epitaxial thin films and 2D van der Waals ferroelectrics, the direction of the polarization in bulk single crystals, can also be modulated/reoriented by water. It will elicit broad interest in the ferroelectric community and the ultrasonication part is quite likely of interest of the emerging topic of 'piezocatalysis' which the authors can have a brief discussion about in the introduction. However, there are a few pending question marks on the mechanism of polarization switching, which need to be further explained before publication.

1. The authors think that when the polarization is pointing up, the positive H+ is absorbed onto the upper surface with positive bound charges. This attraction between two positive charge species is quite counterintuitive. On top of that, both PbO and TiO2 terminations are charge neutral, which also makes it unlikely to attract positive ions. Can authors further justify this?
2. In both references the authors cite (Nat. Commun. 12, 655 (2021); Nat. Commun. 9.1 (2018): 1-8.), only one direction of polarization can be switched by the ionic liquid, which is easy to understand. Why both directions of the polarization can be switched at the same time?
3. It would be great if the authors can add extra experiments to show how the polarization will be modulated when exposed to an alkaline/acidic aqueous solution. In the current model, both positive H+ and negative OH- ions play a role in the switching.
4. What role will flexoelectricity and the cavitation pressure of the water bubble play in this polarization switching? The collapse of the water bubbles will release small shock waves and very intense local heating, which is not unlikely when an ultrasonication condition of 40 kHz 50 W is used. The power and the frequency are quite large.
5. Will there be a phase transition for the PMN-PT sample when exposed to the aqueous liquid given the dramatic change happening in the domain structure? PMN-28PT should have a rhombohedral phase and will it be transformed into a T phase? Any evidence such as the XRD to show the structure before and after the treatment?
6. Can the authors also provide the topography images for Figure 1b in the Supplementary Information to show it is in the same area? Also, for figure 3b, can authors provide the amplitude images that can show how the piezoelectric response is evolving over time?

7. There are a few minor things that can be easily fixed:
7.1 Abstract, “Combined with experimental observations and theoretical analysis”
Should it be changed to “By combining experimental xxx”
7.2 Page 3, “free from the external instructive field and low energy consumption”
Should it be changed to “free from the external instructive field but requires low energy consumption”.
7.3 Page 11: “such as domains above 20 μm²”
“over” probably is better than “above”
7.4 Page 5: “That is to say, domain formation and surface screening are competitive for obtaining low-energy states”
Changing this to “That is to say, domain formation and surface screening are competing in order to reach obtaining low-energy states” is probably better.
7.5 Fig. 3 caption: the white squares and green and blue circles need to be specified.
7.6 To improve the overall clarity and coherence of the manuscript, I suggest that the authors pay particular attention to the grammar and language usage throughout the paper.

Overall, it is a very good paper and up to the standard of Communications Materials. I would suggest the acceptance of the paper after addressing those questions.

Reviewer #3 (Remarks to the Author):

The present manuscript deals with the influence of water on the domain structure and functional properties of PMN-PT single crystals. It is well structured and presents the results in an organized and easy to follow manner. The images are of good quality. The findings are very interesting as they seem to imply that the domain structures of bulk single crystals can be manipulated by the presence of water in an easy manner.
Some aspects of the manuscript could be improved and some questions remained as summarized below:

technical aspects:
the description of the methods should be expanded by a detailed description of the water and ultrasound treatment, e.g. what kind of water was used (tap, DI, ultrapure,...), what was the temperature,....; for the PFM measurement it should be noted if it was checked that applied measurement signal did not induce changes in the domain structures. Furthermore, it should be noted which mode is depicted in Figure 1 and 3. And it would be of interest to know how the samples were treated after the soaking and before the PFM/before applying electrodes for polarization measurements (like, were they dried and in which way? were they washed additionally). For the ultrasound experiment, was there a control experiment done without water? In Figure 4, it is difficult to follow the self organization process based on the colours given, as seemingly disconnected areas switch from one to another colour (e.g. the white region from 40 to 50 to 60 min).

general comments and questions:
the work shows a quite interesting change in the domain structure on the surface without a change in the overall poling state. This is in contrast to the literature on films, where there is the development of a dead layer or an imprint reported due to the preferred alignment of

the surface dipoles with the adsorbates (e.g. in ref 31). It does not become quite clear from the manuscript why the adsorbates in the present case would change the domain structure, but would not lead to a preferred orientation of the surface domain pattern. Furthermore, it does not become clear if this domain pattern change is only a surface effect or if it can be expected to reach through the bulk.

On page 8 the authors mentioned that they broke the time resolution limit of PFM. It is not quite clear what is meant by that.

The P-E loops shown in the appendix indicate that after soaking in water the switching dynamics are stretched out over a longer time scale and the authors argue that the coercive fields vary more for large domains than for small domains. It does not become clear why there should be a change in the coercive field distribution just based on the domain structures. The authors should elaborate more on that.

Related to this point: the argumentation in the paper does not include the possibility that the material surface of the samples has changed chemically due to the soaking procedure apart from the presence of adsorbates. It is known that perovskite systems easily loose preferably A-site cations, but as well B-site cations when in contact with water. Maybe this could be a reason for the changes observed in the polarization loops? And as well influence the domain pattern? If the pH of the water monitored during soaking, this could give indications as well.

Point-by-point Response to Reviewers' Comments

Manuscript number: COMMSMAT-22-0314-T

Title: Self-organization of ferroelectric domains induced by water and reinforced via ultrasonic vibration

Authors: Shuo Yan, Xueli Hu, Xiaomei Lu, Junting Zhang, Xiaofan Shen, and Fengzhen Huang

We thank the reviewers for their critical evaluation and constructive suggestions. Below we address the itemized comments and provide our response with the updated text, figures, and relevant literature references.

The reviewers' comments are given in italics, our responses in normal font, and the changes in the main text are given in blue font.

■ Reviewer #1:

I do not recommend this paper for publication in Communications Materials. Please see my comments below:

■ Response

Thanks for your comments and suggestions on our work. We revised our manuscript with the main points described in the following response.

■ Comment 1

Self-organization is not an appropriate term for the results described. Therefore, the introduction is not properly written because it describes self-organization in nature in general, which is not the subject of this paper.

■ Response

We thank the reviewer for this comment. Self-organization is considered appropriate for the following reasons:

(1) Self-organization is a widespread phenomenon, both in nature and in laboratories. Thus, as an introduction of this work, we simply listed several relevant phenomena in nature in the first paragraph. And then beginning from the second paragraph, all the discussions focused on ferroelectrics. Considering that the first

paragraph may mislead the reviewer, we have further condensed related descriptions.

(2) Self-organization is the spontaneous emergence of ordered collective patterns driven by the interactions among abundant individual components [1-2]. Here in our case, the domain evolution is driven by the interactions among polarizations (dipoles) in the $Pb(Mg_{1/3}Nb_{2/3})O_3$-$28PbTiO_3$ (PMN-28PT) crystals, with water as the non-instructive environment, which is in line with the definition of self-organization.

(3) A large portion of the fractals in nature may be created by a self-organizing process, and the main feature of fractal structures is self-similarity [3-4]. In fact, it is further revealed that here the domain evolution does exhibit self-similar characteristics as shown in Fig. R1-1(a-d). On the one hand, if there is no scale bar, the domain configurations after 30 min (b, 20 μm × 20 μm) and that after a longer time and of a larger area (d, 120 min, 40 μm × 40 μm) look quite similar, showing visual similarity. On the other hand, as shown in Fig. R1-1(e, f), the fractal dimension of domain walls in Fig. R1-1(b) and Fig. R1-1(d) are almost equal, representing the self-similarity in a statistical sense. The self-similarity in the domain evolution process confirms the self-organization nature.

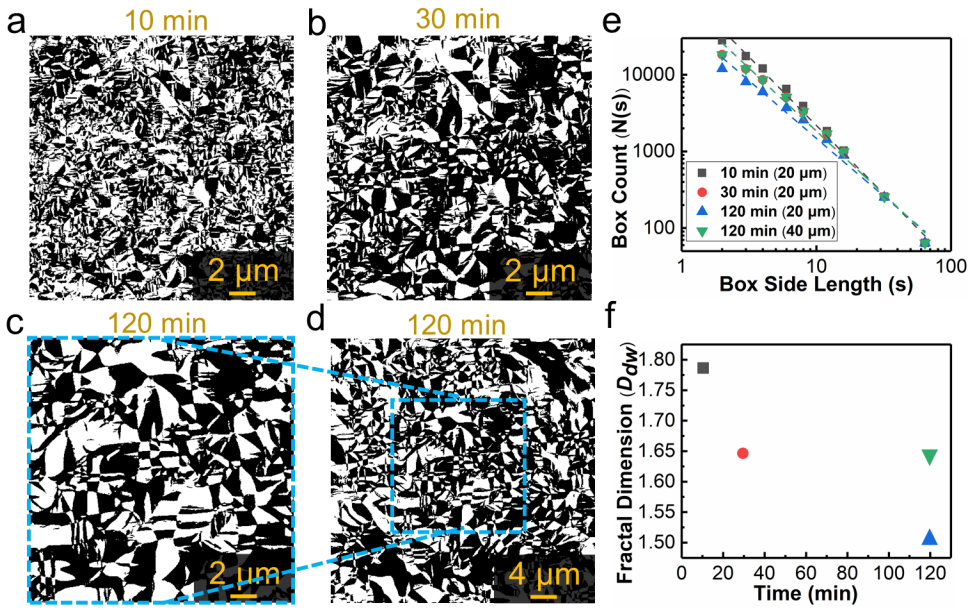

Fig. R1-1 (a-d) Binarized PFM images of the PMN-28PT crystal after different time in water with ultrasound, where white and black represent upward and downward polarization, respectively. (a-c) are measured in a same area, while (d) in a larger area including that of (c). e) Box count vs box side length in the box-counting method [5] for the fitting of fractal dimension, which corresponds to the absolute value of the slope. f) Plot of fractal dimensions over time.

Thanks for your comment, accordingly, we revised the first paragraph of the introduction **(Page 3, line 2-7)** and added a discussion of self-similarity **(Page 10 line 20 to Page 11 line 11)**. We also provide Fig. R1-1 as **Fig. 4 (Page 29)**.

The above discussions are added in the revised manuscript as:

Page 3, line 2-7: The self-organization is a process with only energy input but not external commands. In this process, the intricate and orderly patterns are generated through simple interactions among a large number of components in a system [1, 2]. Self-organization is ubiquitous in biology, material science, and chemistry, ranging from molecular to centimeter scales, and exhibiting rich and diverse phenomena, such as material crystallization events in physicochemical systems [3-6].

Page 10 line 20 to Page 11 line 11: Self-organized growth tends to generate fractal structures, and the core feature of fractals is self-similarity [39, 40]. In fact, the self-organized growth of the ferroelectric domains exhibits a self-similarity over time. Fig. 4a to c are the evolution of binarized PFM images after different time in water with ultrasound. Fig. 4d gives the image taken also for the 120 min-treated sample but with a larger area than that of Fig. 4c. Comparing Fig. 4b with Fig. 4d, the similar domain morphologies and distributions were observed. Without a scale bar, it is challenging to differentiate the actual scan size.

The self-similarity behavior can be quantified by the fractal dimension [41]. In this study, we compute the fractal dimension of domain walls (D_{dw}) using the box-counting method (Fig. 4e). Our findings (Fig. 4f) demonstrate that for the same observation area (Fig. 4a to c), the D_{dw} decreases over time, while the D_{dw} of Fig. 4d (120 min, scan size 40 μm) is approximately equal to that of Fig. 4b (30 min, scan size 20 μm), indicating the self-similarity in a statistical sense. The self-similarity further confirms the self-organization nature of the domain evolution process.

■ Comment 2

The work is not new; a similar topic has already been described by authors about the $BiFeO_3$. The $BiFeO_3$ is much more sensitive to moisture compared to the

$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$, so the influence of water on the change of the domain pattern is more credible.

■ **Response**

We thank the reviewer for this comment. We would like to explain our opinion from the following three aspects.

(1) New information in this work

Compared with the existing reports [6-7], our work is obviously different in material system, working methods, internal mechanism, and experimental results.

A. Material system. As mentioned in the introduction of our manuscript, the existing reports involve water-induced polarization switching in $BiFeO_3$ (BFO) films [6], and ionic liquid-induced polarization switching in a van der Waals layered ferroelectric $CuInP_2S_6$ (CIPS) [7]. Both BFO and CIPS are low-dimensional materials, while we focus on PMN-PT bulk crystals. It is evident that the difference between low-dimensional materials and bulk crystals can be huge.

B. Working methods. For low-dimensional BFO and CIPS, ion-adsorption on one side of the sample could induce dramatic changes in the polarization distribution. Nevertheless, for the PMN-PT crystal, the domain evolution is found to be quite slow even if we immerse it wholly in water. To address this issue, we innovatively applied ultrasonic vibration, which greatly facilitate the domain growth.

C. Internal mechanism. In low-dimensional BFO and CIPS, what happened is the polarization reversal caused by the difference in the adsorption of negative and positive charges. While in the PMN-28PT crystal we investigated, it is the self-organized growth of the domain structure induced by equally adsorbable negative and positive charges.

D. Experimental results. For low-dimensional BFO and CIPS, the results of the polarization reversal are the preferential polarization direction. While for our PMN-28PT crystal, the self-organization mainly results in the domain coarsening, with the area ratio of the upward to downward domains remain around 1:1.

(2) The sensitivity of BFO and PMN-PT

As mentioned above, the existing work is about BFO films, while we focus on PMN-PT bulk crystals. To our understanding, it is difficult to make a judgement on two materials (BFO and PMN-PT) by comparing data between films and crystals.

The current literatures on ion-adsorption-induced polarization reversal primarily focus on low-dimensional materials. For crystals, it is reported that humidity would affect the shape and size of the switching domains induced by a biased scanning tip [8-9]. Thus, we agree if it is the reviewer's opinion that, the crystals are less sensitive to moisture compared with the films.

As we also mentioned in the manuscript "while for ferroelectric bulk crystals, the solid-liquid interface is rarely considered to affect the polarization state and the domain configuration" (Page 4 line 8-10). Because of this, we think our work on crystals is instructive. Through careful research, we found that the domain structure of PMN-28PT crystal changes slowly after exposure to water, and the domain coarsening process can be greatly accelerated by ultrasonic vibration.

(3) Our results are credible

The reliability of this research can be confirmed by the repeatable data provided in the manuscript and in the supplementary material. In fact, we mentioned the experimental reproducibility in our submitted manuscript, and we can provide additional information upon requests. For more details, please see the response to the next comment (comment 3).

With all these issues in mind, we have revised part of the introduction **(Page 4, line 10-16)** and added a comparison with low-dimensional materials **(Page 15 line 9-21)**. The above discussions are added in the revised manuscript as:

Page 4, line 10-16: Some recent studies have demonstrated that humidity would affect the shape and size of the domains formed by a scanning tip in $LiNbO_3$ crystals [26, 27]. This phenomenon was attributed to the interplay between polarization switching and screening charge dynamics, and suggested that the presence of water in the environment could influence polarization switching under an electric field. While water did not

directly modulate crystal polarization, the findings raised intriguing possibilities for further exploration.

Page 15 line 9-21: In the end, we perform a discussion of our work and that of others on low-dimensional materials. For low-dimensional material [22, 23, 29], an initial polarization direction is present, either upward or downward. Positive and negative charges preferentially adsorb on surfaces with initial upward and downward polarizations, respectively, resulting in a polarization reversal from upward to downward or vice versa. However, in the PMN-28PT crystal we investigated, there is no specific initial polarization direction in the crystal. The area ratio of the upward to downward domains is around 1:1. When the crystal is immersed in water, a non-instructive environment with comparable numbers of positive and negative ions, polarization switching occurs with equal probabilities in the upward and downward directions, leading to the self-organized growth of domains driven by the system's total energy. Throughout the evolution process, the proportion of upward and downward polarization areas remains roughly equal.

■ **Comment 3**

My biggest concern with this work is that after I received this work for review, we repeated the exact same experiment with $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ *bulk material with composition near the morphotropic phase boundary and did not get the same result. In fact, we immersed the sample in the water for 1 hour and 25 hours, and then we performed the PFM study of the domain patterns. No changes in the domain structure were observed. Therefore, I doubt that this experiment is repeatable.*

■ **Response**

We thank the reviewer for the interests in repeating our experiments. We would like to emphasize that we have enough confidence in the repeatability of our experiments.

In fact, this research was started in 2020. Since then, we have repeated the experiments using different batches of crystals to adjust the experimental conditions

and investigate the stability of the domains. In response to the reviewer's comments, we have conducted another experiment using a new batch of PMN-28PT crystals. As shown in Fig. R1-2, after the crystal was exposed to water, the domain configuration did change gradually. In addition, we provided a control experiment using ethanol instead of water, and found that ethanol caused little change in domain structure even with ultrasound (Fig. S9b).

We can also understand the concern of the reviewer. We apologize the experimental procedure was not sufficiently detailed for repeating the experiment in a short time. Therefore, we would like to discuss some key points of the experiments in the following:

A. Sample. The sample we used is the commercial [001]-oriented PMN-28PT crystal. We cannot guarantee similar results for ceramics.

B. Pre-annealing of the samples. To minimize the influence of internal stress and surface defects caused by polishing, the crystals were pre-annealed at 300 °C for 2 hours. Thus, the initial samples for our experiments are composed of irregularly-shaped condensed small domains. We cannot guarantee similar results for samples with initial large domains.

C. Observation position. The PFM images before and after the water treatment were obtained at the same position of the crystal, determined by topography as described in comment 4. Otherwise, it would be difficult to distinguish the domain changes caused only by water. While, for crystals exposed to water with ultrasound, the domain changes can be observed easily.

D. PFM measurement. The probe type and the scanning parameters should be carefully chosen to ensure that the PFM measurement would not affect the domain structure.

In addition to the points mentioned above, we would be glad to answer any further questions regarding this experiment.

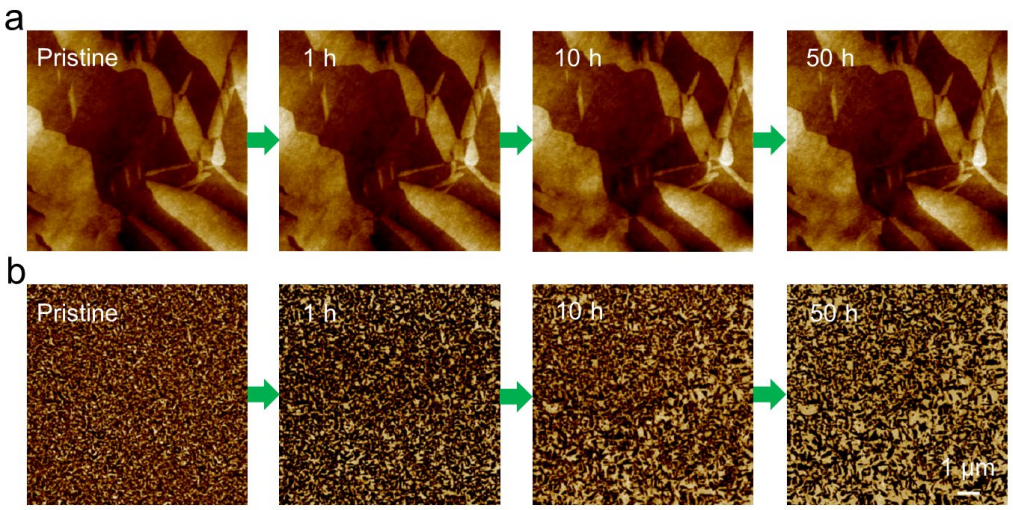

Fig. R1-2 (a) Topography, and (b) PFM phase images at the same position of the crystal after being exposed to water for different time.

Thanks for your comment, accordingly, we have added the picture of this repeated experiment to the supplementary information (**Fig. S3**) and improved the description of the experimental details in Methods as:

Page 16 line 20 to Page 17 line 4: For the water treatment: The PMN-28PT crystals were treated with deionized water at room temperature in a 25 mL beaker. The inner diameter and the height of the beaker were 3.2 cm and 5.5 cm, and the height of deionized water was 3 cm. The beaker was sealed and placed on a stationary desktop during the water treatment. For the water treatment with ultrasonic vibration: The above beaker containing a crystal was suspended within the ultrasonic bath and sonicated (40 kHz, 50 W). The height of water in the ultrasonic bath was about 7 cm.

■ **Comment 4**

Is the same area always seen in figures 1 and 3? The information is limited because only the PFM amplitude image is shown. What about the topography, from which we can judge if the area is really the same? Also, the PFM phase images should be added. A similar comment applies to figures S1-S5.

■ **Response**

We thank the reviewer for raising this important question. As we described in the manuscript, all the PFM images in Fig. 1b and Fig. 3b were measured at the same

position and determined by the topography. We apologize for providing only PFM phase images in the previous manuscript. Here the topography images corresponding to Fig. 1b and Fig. 3b are shown in Fig. R1-3 and Fig. R1-4, respectively. It is noticeable that there is little variation in the topography before and after water treatment.

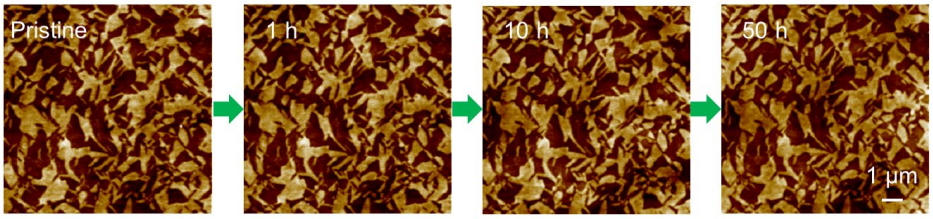

Fig. R1-3 Topography images of the PMN-28PT crystal with different time in water, in corresponding to the PFM phase images in Fig. 1b. The measurements were performed at the same location of the crystal.

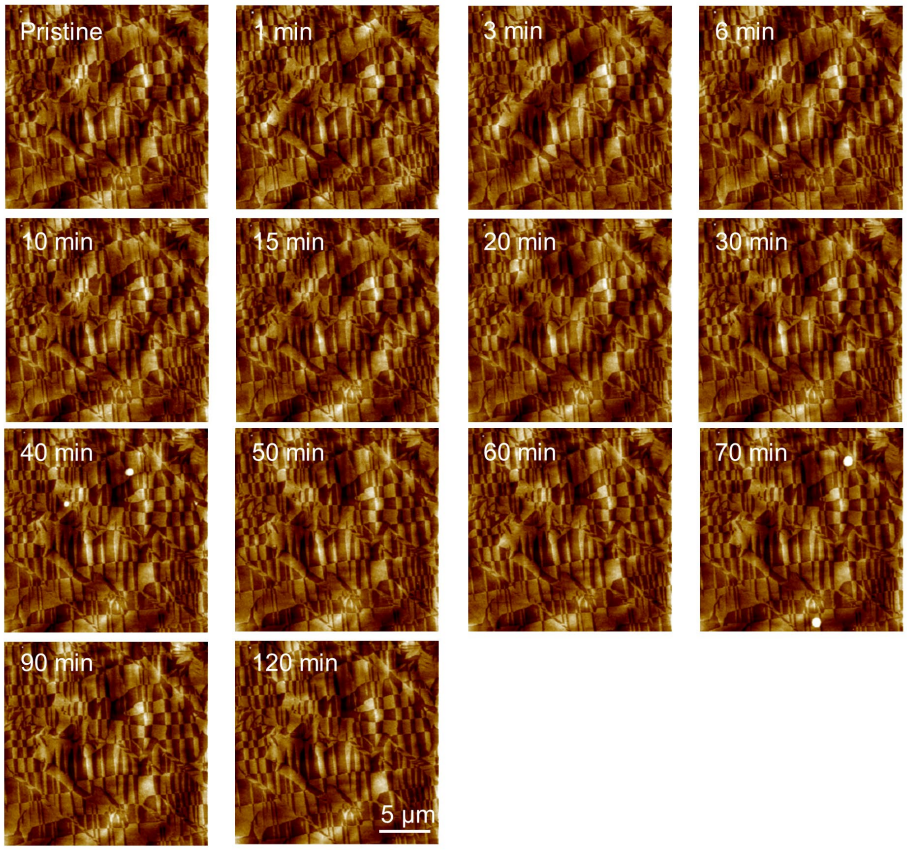

Fig. R1-4 Topography images of the PMN-28PT crystal with different time in water under ultrasonic vibration, in corresponding to the PFM phase images in Fig. 3b. The measurements were performed at the same location of the crystal.

Thanks for your comment, accordingly, we have pointed out in the methods **(Page 17, line 7-9)** that all the PFM images in the manuscript are phase images, provided the

topography and amplitude images corresponding to Fig. 1b and Fig. 3b in the supplementary information as **Fig. S1**, **Fig. S4** and **Fig. S5**, and added related discussions **(Page 5, line 11-13; Page 8, line 18-21)** in the revised manuscript.

Page 17, line 7-9: The ferroelectric domains were characterized under the Vertical PFM mode, and all the PFM images given in Fig. 1 and Fig. 3 are out-of-plane phase images.

Page 5, line 11-13: Note that the Piezoresponse Force Microscopy (PFM) images in Fig. 1b were measured at the same position determined by the topography (Fig. S1).

Page 8, line 18-21: All PFM images in Fig. 3b were acquired at the same location and determined by the topography (Fig. S4). We also provide the corresponding PFM amplitude images in Fig. S5. With the time (average domain size) increasing, the average amplitude over the measured area also increases.

■ Reference

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7. Xu, D. D. *et al.* Ion adsorption-induced reversible polarization switching of a van der Waals layered ferroelectric. *Nat. Commun.* **12**, 655 (2021).
8. Ievlev, A. V. et al. Intermittency, quasiperiodicity and chaos in probe-induced ferroelectric domain switching. *Nat. Phys.* **10**, 59 (2013).
9. Ievlev, A. V., Morozovska, A. N., Eliseev, E. A., Shur, V. Y. & Kalinin, S. V. Ionic field effect and memristive phenomena in single-point ferroelectric domain switching. *Nat. Commun.* 5, 1 (2014).

■ **Reviewer #2:**

The paper reports a self-organization process of domain structures in the PMN-0.28PT system when subjected to water, which can be further accelerated by the ultrasonic vibration. The interaction between the screening charges in water, i.e., H+ and OH-, and the bound charges is believed to be the origin of the observed switching and coarsening phenomenon. A transition point is shown in the percolation model, and this indicates a non-conservative nature of the system. This paper for the first time shows that in addition to low dimensional ferroelectrics such as epitaxial thin films and 2D van der Waals ferroelectrics, the direction of the polarization in bulk single crystals, can also be modulated/reoriented by water. It will elicit broad interest in the ferroelectric community and the ultrasonication part is quite likely of interest of the emerging topic of 'piezocatalysis' which the authors can have a brief discussion about in the introduction. However, there are a few pending question marks on the mechanism of polarization switching, which need to be further explained before publication.

■ **Response**

Thank you very much for your positive evaluation and suggestions on our work. We have carefully revised our manuscript according to your valuable comments and detailed advices, which greatly contributed to the improvement of our paper.

As our work emphasizes ion-adsorption induced polarization switching in ferroelectrics, we would like to incorporate discussions about piezocatalysis in the main text, as described in our response to comment 4. The details of our revision are described as follows.

■ **Comment 1**

The authors think that when the polarization is pointing up, the positive H^+ is absorbed onto the upper surface with positive bound charges. This attraction between two positive charge species is quite counterintuitive. On top of that, both PbO and TiO_2 terminations are charge neutral, which also makes it unlikely to attract positive ions. Can authors further justify this?

■ **Response**

Thanks for the valuable comment. This question can be approached from the following aspects:

(1) When polar ferroelectric crystals are immersed in water, despite the presence of bound charges on the surface, the random thermal moving H^+ and OH^- ions still have probability of moving to the surface and being adsorbed.

(2) This adsorption is different from the long-range attraction between opposite charges, but bonding between atoms close to each other. Although PbO and TiO_2 terminations are charge neutral, Pb and O (Ti and O) are not in the same horizontal position on the surface of crystals. As a result, H^+ and OH^- ions may potentially be adsorbed near any atom on the terminal through bonding, and the final adsorption position can be determined by adsorption energy.

(3) We calculated the adsorption energies of H^+ and OH^- ions at different locations on the crystal surface. It is found that H^+ ions tend to be adsorbed near O ions, while OH^- ions near Pb (Ti) ions, resulting in changes of the local polarization state. Nevertheless, in the case of upward (downward) polarization, when the H^+ (OH^-) ions bonds with O (Ti) ions on the upper surface while OH^- (H^+) ions bond with Pb (O) ions on the lower surface, the polarization switching could occur in a large range.

(4) Similar phenomena have been reported by other researchers [1]. As shown in Fig. R2-1(Nat. Commun. 9, 3809 (2018); Figure 4b), the negative OH^- ions can be adsorbed by the negative surface with negative bond charges. The OH^- ions bond with Fe ions on negative surface, inducing a polarization reversal from downward to upward.

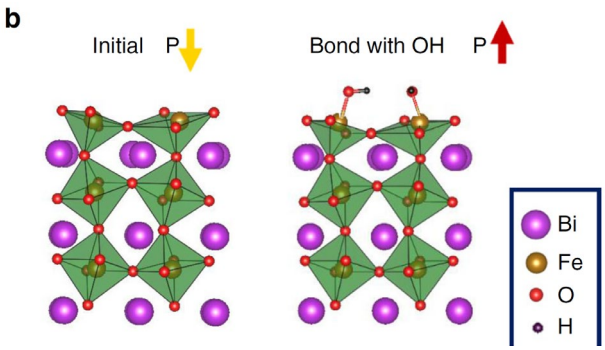

Fig. R2-1 BFO polarizations are switched from downward to upward with adsorbed two OH ions. [Picture comes from Nat. Commun. 9, 3809 (2018)]

Thanks for your comment, accordingly, the above discussions are added in the revised manuscript as:

Page 18 line 12-18: The adsorption energies of H^+ and OH^- ions at different locations on the crystal surface were calculated. The H^+ ions tend to be adsorbed near O ions, while OH^- ions near Pb (Ti) ions, resulting in changes of the local polarization state. Nevertheless, in the case of upward (downward) polarization, when the H^+ (OH^-) ions bonds with O (Ti) ions on the upper surface while OH^- (H^+) ions bond with Pb (O) ions on the lower surface, the polarization switching could occur in a large range.

■ **Comment 2**

In both references the authors cite (Nat. Commun. 12, 655 (2021); Nat. Commun. 9.1 (2018): 1-8.), only one direction of polarization can be switched by the ionic liquid, which is easy to understand. Why both directions of the polarization can be switched at the same time?

■ **Response**

We thank the reviewer for the excellent comment. Below, we would like to answer your question by comparing our work with that in the references [1-2].

(1) In the references

The samples in the references are thin films $BiFeO_3$ (BFO) and two-dimensional materials $CuInP_2S_6$ (CIPS). There is an initial polarization direction in these low-dimensional materials, either upward or downward, and the positive and negative charges (cations and anions) are preferred to be adsorbed by surface with initial upward (positive bound charges) and downward (negative bound charges) polarizations, respectively. Researchers have suggested that "The adsorbed like-charges provide an additional electric field with the same direction of depolarization field" [2], driving the polarization reversal in the sample.

(2) Our work

The samples in our work are PMN-28PT bulk crystals. On the one hand, there is no specific initial polarization direction in the crystals, meaning, the area ratio of the

upward to downward domains is around 1:1, showing no priority. On the other hand, the crystals are immersed in water with comparable numbers of positive and negative ions, and the adsorption probability of them on the crystal surface are also comparable. In this case, the environment of the crystal is non-instructive, and there is no directional field similar to the aforementioned low-dimensional materials.

What happens in the crystals is the polarization switching with equal opportunities in the up and down directions. From another point of view, it is the self-organization process driven by the total energy of the system, manifested mainly as the coarsening of the domains with the area ratio of the upward to downward domains remain around 1:1.

Thanks for your comment, accordingly, the above discussions are added in the revised manuscript as:

Page 15 line 9-21: In the end, we perform a discussion of our work and that of others on low-dimensional materials. For low-dimensional material [22, 23, 29], an initial polarization direction is present, either upward or downward. Positive and negative charges preferentially adsorb on surfaces with initial upward and downward polarizations, respectively, resulting in a polarization reversal from upward to downward or vice versa. However, in the PMN-28PT crystal we investigated, there is no specific initial polarization direction in the crystal. The area ratio of the upward to downward domains is around 1:1. When the crystal is immersed in water, a non-instructive environment with comparable numbers of positive and negative ions, polarization switching occurs with equal probabilities in the upward and downward directions, leading to the self-organized growth of domains driven by the system's total energy. Throughout the evolution process, the proportion of upward and downward polarization areas remains roughly equal.

■ **Comment 3**

It would be great if the authors can add extra experiments to show how the polarization will be modulated when exposed to an alkaline/acidic aqueous solution.

In the current model, both positive H^+ and negative OH^- ions play a role in the switching.

■ **Response**

We thank the reviewer for this insightful comment. Based on your comments, we have treated the crystals using acidic (pH~2) and alkaline (pH~12) aqueous solutions, respectively. As shown in Figure R2-2, the domain evolution in either acidic or alkaline aqueous solution is significantly suppressed compared to that in water.

As supported by first-principles calculations and discussed in comment 1, the polarization switching can be stimulated when positive and negative charges are adsorbed simultaneously by the crystal. In addition, as shown by the Monte Carlo simulations, equally sufficient positive and negative screening charges are beneficial for the domain coarsening.

As we know that, positive H^+ (negative OH^-) ions are dominant in acidic (alkaline) aqueous solutions. Thus, in acidic (alkaline) aqueous solutions, the probability of negative OH^- (positive H^+) adsorption on the crystal surface is very low, which largely reduces the probability of adsorbing the same amount of positive and negative ions, and significantly delay the process of polarization switching and domain coarsening.

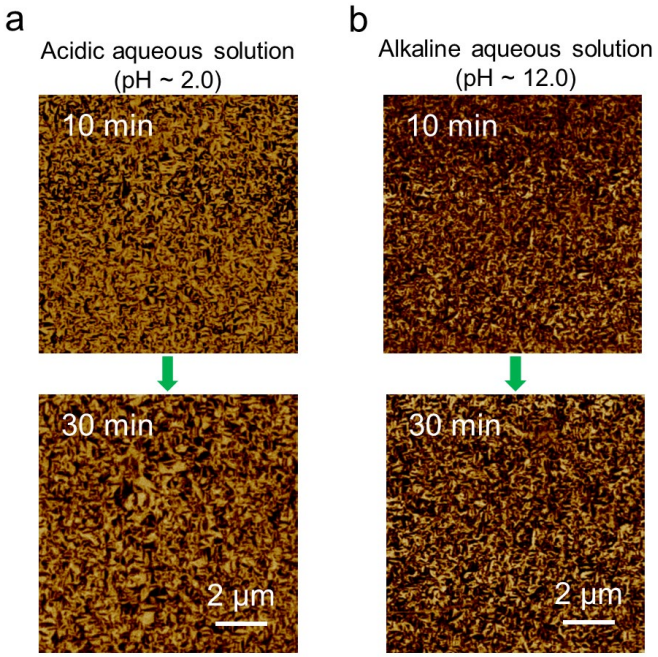

Fig. R2-2 PFM phase images of the domains in (a) acidic and (b) alkaline aqueous solution with ultrasound (40 kHz, 50 W).

Thanks for your comment, Fig. R2-2 is added as **Fig. S9(c, d)**. The above discussions are added in the revised manuscript as:

Page 10 line 1-9: Further, with ultrasound (40 kHz, 50 W), we also exposed the samples to ethanol, acidic, and alkaline aqueous solution, respectively. As shown in Fig. S9b to d, there is no observable change in domain configuration after 30 mins, which means the self-organization process of ferroelectric domains in these solutions are greatly suppressed compared with that in water (Fig. 3b). Ethanol is a non-electrolyte with very few ions, and H^+(OH^-) is dominant in acid (alkaline) solution. The different effects of these solutions and water indicate that abundant and comparable amount of positive and negative ions in water are the cause of domain changes, and ultrasonic vibration plays a role in promoting the self-organization process of domains.

■ **Comment 4**

What role will flexoelectricity and the cavitation pressure of the water bubble play in this polarization switching? The collapse of the water bubbles will release small shock waves and very intense local heating, which is not unlikely when an ultrasonication condition of 40 kHz 50 W is used. The power and the frequency are quite large.

■ **Response**

We would like to express our appreciation to the reviewer for the constructive comments. As pointed out by the reviewer, the ultrasonic cavitation bubbles that collapse instantaneously will release local electric, mechanical, and thermal fields, inducing changes in the polarization states of the particles [3-5].

However, as discussed in the previous manuscript and comment 3, in an ultrasound environment, it was observed that the domain growth in ethanol, acidic, and alkaline aqueous solutions were suppressed. These comparative experiments suggest that abundant and comparable amount of positive and negative ions in water are the cause of domain changes, and ultrasonic cavitation could also promote polarization switching.

Thanks for your comment, we have included a discussion on the topic of

piezocatalysis in the revised manuscript as:

Page 10, line 15-19: Moreover, in the emerging field of piezocatalysis, studies have indicated that ultrasonic waves could promote the formation and collapse of bubbles on the particle surface. The ultrasonic cavitation bubbles that collapse instantaneously will release local electric, mechanical, and thermal fields, which also promote polarization switching. [36-38].

■ **Comment 5**

Will there be a phase transition for the PMN-PT sample when exposed to the aqueous liquid given the dramatic change happening in the domain structure? PMN-28PT should have a rhombohedral phase and will it be transformed into a T phase? Any evidence such as the XRD to show the structure before and after the treatment?

■ **Response**

We thank the reviewer for this valuable comment. Based on your comment, X-ray diffraction (XRD, Bruker D8 Advance, Cu Kα rays) measurements were performed on PMN-28PT crystals before and after treatment (exposed to water with ultrasound for 120 min). As shown in Fig. R2-3, the domain changes are not accompanied by a phase transition, and the crystal remains in the rhombohedral phase.

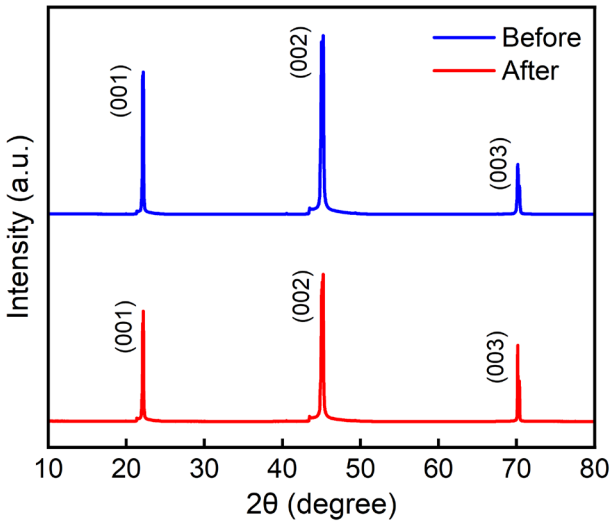

Fig. R2-3 XRD patterns of the [001]-oriented PMN-28PT crystal before and after treatment (in water with ultrasound for 120 min).

Thanks for your comment. The XRD patterns are given as **Fig. S6**, and the above

discussions are added in the revised manuscript as:

Page 8 line 21 to Page 9 line 3: In addition, X-ray diffraction (XRD, Bruker D8 Advance, Cu Kα rays) measurements were performed on PMN-28PT crystals before and after treatment (exposed to water with ultrasound for 120 min). As shown in Fig. S6, the domain changes are not accompanied by a phase transition, and the crystal remains in the rhombohedral phase.

■ **Comment 6**

Can the authors also provide the topography images for Figure 1b in the Supplementary Information to show it is in the same area? Also, for figure 3b, can authors provide the amplitude images that can show how the piezoelectric response is evolving over time?

■ **Response**

We thank the reviewer for raising this question. As we described in the main text, all the PFM images were measured in the same area, determined by the topography. To address your concern, we provide the topography images for Fig. 1b and Fig. 3b as **Fig. S1** and **Fig. S4**, respectively (Here Fig. R2-4 and Fig. R2-5). It is noticeable that there is little variation in the topography before and after treatment.

Additionally, we provide the PFM amplitude images (**Fig. S5**) corresponding to Fig. 3b, as shown in Fig. R2-6. With the time (average domain size) increasing, the average amplitude over the measured area also increases.

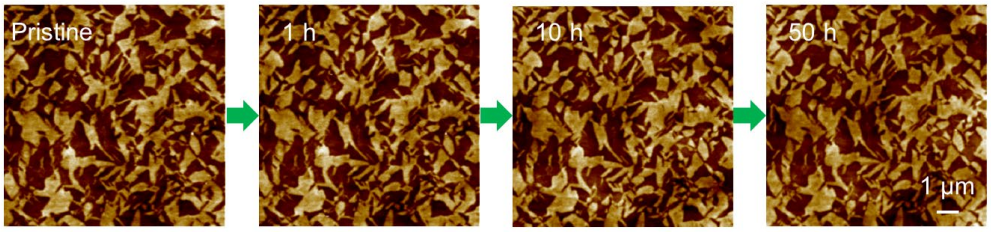

Fig. R2-4 Topography images of the PMN-28PT crystal with different time in water, in corresponding to the PFM phase images in Fig. 1b. The measurements were performed at the same location of the crystal.

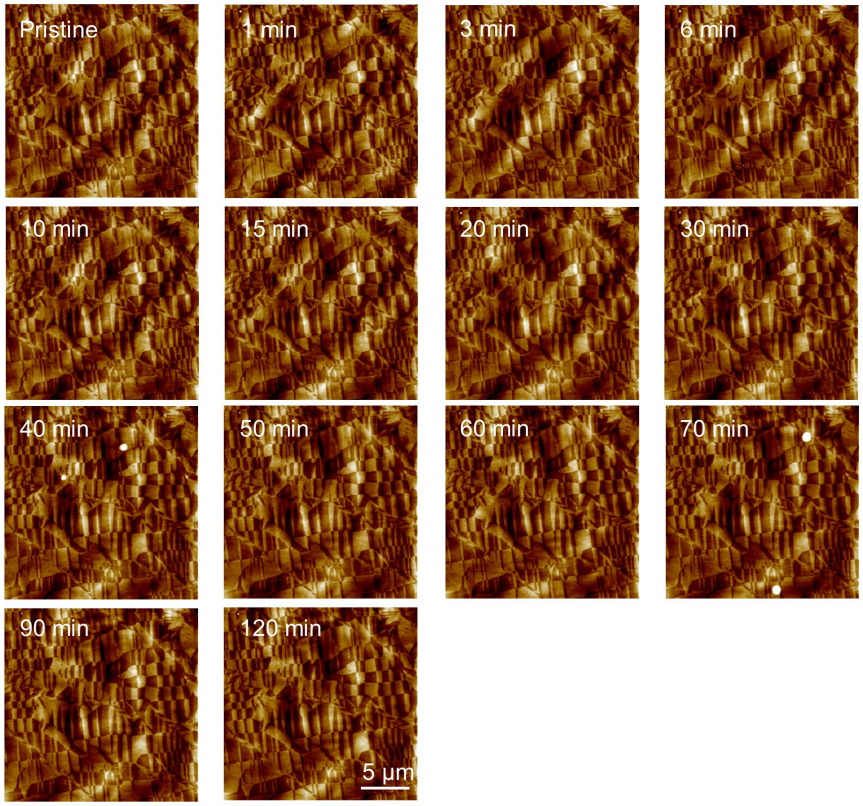

Fig. R2-5 Topography images of the PMN-28PT crystal with different time in water under ultrasonic vibration, in corresponding to the PFM phase images in Fig. 3b.

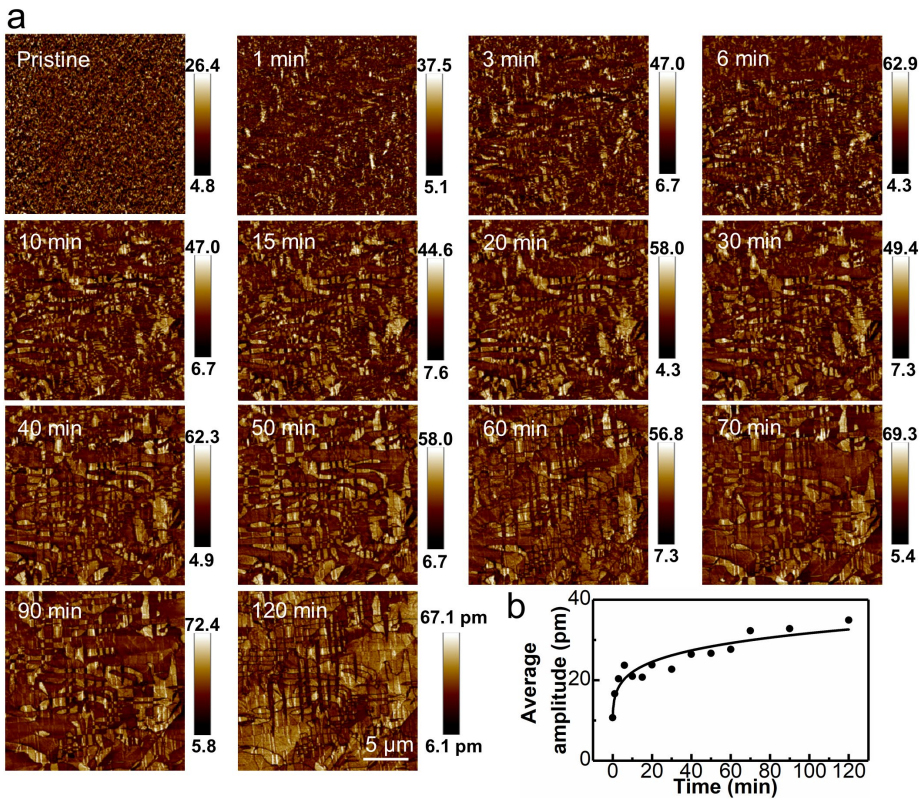

Fig. R2-6 a) PFM amplitude images of PMN-28PT corresponding to the phase images in Fig. 3b. b) Change of average PFM amplitude with time.

Thanks for your comment, accordingly, the related descriptions have been added in the revised manuscript as:

Page 5, line 11-13: Note that the Piezoresponse Force Microscopy (PFM) images in Fig. 1b were measured at the same position determined by the topography (Fig. S1)

Page 8, line 18-21: All PFM images in Fig. 3b were acquired at the same location and determined by the topography (Fig. S4). We also provide the corresponding PFM amplitude images in Fig. S5. With the time (average domain size) increasing, the average amplitude over the measured area also increases.

■ **Comment 7**

There are a few minor things that can be easily fixed:

7.1 Abstract, "Combined with experimental observations and theoretical analysis"

Should it be changed to "By combining experimental xxx"

7.2 Page 3, "free from the external instructive field and low energy consumption"

Should it be changed to "free from the external instructive field but requires low energy consumption".

7.3 Page 11: "such as domains above 20 μm²"

"over" probably is better than "above"

7.4 Page 5: "That is to say, domain formation and surface screening are competitive for obtaining low-energy states"

Changing this to "That is to say, domain formation and surface screening are competing in order to reach obtaining low-energy states" is probably better.

7.5 Fig. 3 caption: the white squares and green and blue circles need to be specified.

7.6 To improve the overall clarity and coherence of the manuscript, I suggest that the authors pay particular attention to the grammar and language usage throughout the paper.

■ **Response**

It's so kind for the reviewer to provide such a detailed correction list. We have made the revisions in the revised manuscript and have clearly marked them.

■ **Comment 8**

Overall, it is a very good paper and up to the standard of Communications Materials. I would suggest the acceptance of the paper after addressing those questions.

■ **Response**

Many thanks for the reviewer's high affirmation of our work. We have carefully revised our manuscript according to your suggestions. We sincerely hope that our revised manuscript meets the standards for publication and can be accepted.

■ **Reference**

1. Tian, Y. *et al.* Water printing of ferroelectric polarization. *Nat. Commun.* **9**, 3809 (2018).
2. Xu, D. D. *et al.* Ion adsorption-induced reversible polarization switching of a van der Waals layered ferroelectric. *Nat. Commun.* **12**, 655 (2021).
3. Su, R. *et al.* Nano-Ferroelectric for High Efficiency Overall Water Splitting under Ultrasonic Vibration. *Angew. Chemie - Int. Ed.* **58**, 15076 (2019).
4. Wang, Y. *et al.* Piezo-catalysis for nondestructive tooth whitening. *Nat. Commun.* **11**, 1328 (2020).
5. Wang, Y. *et al.* Ultrasonic activation of inert poly(tetrafluoroethylene) enables piezocatalytic generation of reactive oxygen species. *Nat. Commun.* **12**, 3508 (2021).

■ **Reviewer #3:**

The present manuscript deals with the influence of water on the domain structure and functional properties of PMN-PT single crystals. It is well structured and presents the results in an organized and easy to follow manner. The images are of good quality. The findings are very interesting as they seem to imply that the domain structures of bulk single crystals can be manipulated by the presence of water in an easy manner.

Some aspects of the manuscript could be improved and some questions remained as summarized below:

■ **Response**

Thank you very much for the affirmation of our work. Your suggestions are very helpful, and we revised our manuscript accordingly. The details are highlighted in a different color in the text and described as follows.

■ **Comment 1**

the description of the methods should be expanded by a detailed description of the water and ultrasound treatment, e.g. what kind of water was used (tap, DI, ultrapure,...), what was the temperature,....;

■ **Response**

We thank the reviewer for this advice.

For the water treatment: The PMN-28PT crystals were treated with deionized water at room temperature in a 25 mL beaker. The inner diameter and the height of the beaker were 3.2 cm and 5.5 cm respectively, and the height of deionized water was 3 cm. The beaker was sealed and placed on a stationary desktop during the water treatment.

For the water treatment with ultrasonic vibration: The above beaker containing a crystal was suspended within the ultrasonic bath and sonicated (40 kHz, 50 W). The height of water in the ultrasonic bath is about 7 cm.

Thank you for your comment. We have added the above discussions to the revised manuscript in the Methods section as:

Page 16 line 20 to Page 17 line 4: For the water treatment: The PMN-28PT crystals

were treated with deionized water at room temperature in a 25 mL beaker. The inner diameter and the height of the beaker were 3.2 cm and 5.5 cm, and the height of deionized water was 3 cm. The beaker was sealed and placed on a stationary desktop during the water treatment. For the water treatment with ultrasonic vibration: The above beaker containing a crystal was suspended within the ultrasonic bath and sonicated (40 kHz, 50 W). The height of water in the ultrasonic bath was about 7 cm.

■ **Comment 2**

For the PFM measurement it should be noted if it was checked that applied measurement signal did not induce changes in the domain structures.

■ **Response**

We thank the reviewer for this constructive comment, and this is very important. The driving amplitude for PFM measurement is 200 mV, which is a small signal and less possible to cause any change in the domain structure.

To further prove this, we conducted repeated PFM measurements in the same area of an untreated PMN-28PT crystal. As shown in Fig. R3-1, even for samples with nanometer-sized domains, no changes can be observed after repeated PFM measurements.

Fig. R3-1 PFM images of the repeated scan in the same area of an untreated PMN-28PT crystal. The untreated initial sample is composed of irregularly-shaped condensed domains with a 100 nm average size.

Thanks for your comment, accordingly, we have provided this figure in the supplementary information as **Fig. S2**, and added a related discussion in the revised manuscript as:

Page 5, line 13-14: and the PFM measurements did not induce any change in the domain structure (Fig. S2).

■ **Comment 3**

Furthermore, it should be noted which mode is depicted in Figure 1 and 3. And it would be of interest to know how the samples were treated after the soaking and before the PFM/before applying electrodes for polarization measurements (like, were they dried and in which way? were they washed additionally).

■ **Response**

We thank the reviewer for this comment.

(1) The ferroelectric domains were characterized under the Vertical PFM mode, and all the PFM images given in Fig. 1 and Fig. 3 are out-of-plane phase images.

(2) After the soaking and before the PFM measurements, the crystals were dried with an N_2 gun for about 5 s at room temperature.

Thanks for your comment, accordingly, we have incorporated the above descriptions into the Methods section of the revised manuscript.

Page 17 line 7-11: The ferroelectric domains were characterized under the Vertical PFM mode, and all the PFM images given in Fig. 1 and Fig. 3 are out-of-plane phase images. After the water treatment and before the PFM characterization, the crystals were dried with a N_2 gun for about 5 s at room temperature.

■ **Comment 4**

For the ultrasound experiment, was there a control experiment done without water?

■ **Response**

Thanks for your comment. In our manuscript, we provided a control experiment using ethanol instead of water to confirm the role of water. Following your suggestion, we also conducted an ultrasound experiment without water as described below:

A PMN-28PT crystal was placed in an empty 25 mL beaker (without any liquid including water), which was then fixed in the ultrasonic bath (with a water height of

about 7 cm) through the beaker clamp and sonicated (40 kHz, 50 W) for 20 min. Note that the beaker was sealed to prevent water from splashing in.

Fig. R3-2 shows the corresponding domain patterns before and after ultrasonic treatment for 20 min. Compared with water, there is no observable change in domain configuration for the crystal in air. This control experiment indicates that, water serves as the primary factor responsible for domain changes, whereas ultrasonic vibration plays a role in promoting the self-organization process of domains.

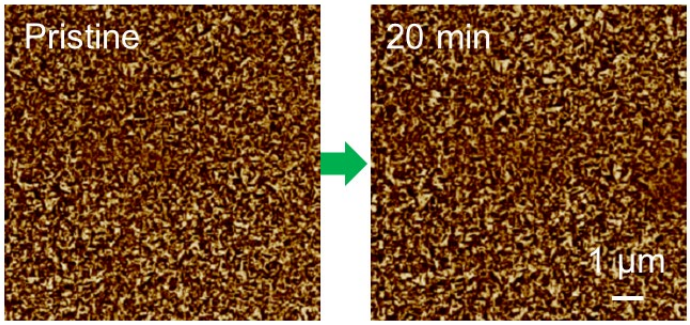

Fig. R3-2 PFM image of PMN-28PT crystals before and after ultrasonic treatment. The samples were sonicated for 20 min in a beaker without water.

Thanks for your comment. Figure R3-2 is added as **Fig. S9a.** The above discussions are added in the revised manuscript as:

Page 9 line 21 to Page 10 line 1: As a comparison to the experiment in Fig. 3, we performed a controlled experiment without water (Fig. S9a), and found that ultrasound alone has a limited impact on the domains.

■ **Comment 5**

In Figure 4, it is difficult to follow the self organization process based on the colours given, as seemingly disconnected areas switch from one to another colour (e.g. the white region from 40 to 50 to 60 min).

■ **Response**

We thank the reviewer for this comment. The coloring rule is that, different colors represent different connected regions (clusters), with the largest cluster marked in white.

With time increasing, the number, the size, and the shape of the clusters undergo changes, making it difficult to track and assign the same color to each region. In our

study, MATLAB was utilized to color the clusters automatically, with the largest cluster specified in white. Since the largest cluster is most concerned in percolation model, fixing this color is useful and common [1].

It can be seen from Fig. 4b (revised manuscript Fig. 5b), before percolation transition point, the position of the largest cluster may change due to the evolution of the domains (40 min to 60 min). While after the transition point, the dominant connected region continues growing (70 min to 120 min) around a certain location.

Thanks for your comment, accordingly, the above discussions are added in the revised manuscript as:

Page 19, line 7-10: With time increasing, the number, the size, and the shape of the clusters undergo changes, making it difficult to track and assign the same color to each region. Hence, MATLAB was utilized to color the clusters automatically, with the largest cluster specified in white.

■ **Comment 6**

The work shows a quite interesting change in the domain structure on the surface without a change in the overall poling state. This is in contrast to the literature on films, where there is the development of a dead layer or an imprint reported due to the preferred alignment of the surface dipoles with the adsorbates (e.g. in ref 31). It does not become quite clear from the manuscript why the adsorbates in the present case would change the domain structure, but would not lead to a preferred orientation of the surface domain pattern.

■ **Response**

We thank the reviewer for the excellent comment. Below, we try to compare our work with that in the references.

(1) In the references

Concerning the polarization changes, there are two cases in the references. One is the polarization reversal in the surface layer [2], and the other is that throughout the whole sample [3, 4].

The samples in the references are thin films and two-dimensional materials. There is an initial polarization direction in these low-dimensional materials, either upward or downward, and the positive and negative charges (cations and anions) are preferred to be adsorbed by surface with initial upward (positive bound charge) and downward (negative bound charge) polarizations, respectively. The chemisorption, with bond between adsorbed ions and sample atoms, somewhat likes an additional electric field opposite to the initial polarization direction, driving the polarization reversal in the surface layer (in case of considerable surface charged defects) [2] and even in the whole sample [3, 4].

(2) Our work

The samples in our work are PMN-28PT bulk crystals. On the one hand, there is no specific initial polarization direction in the crystals, meaning, the area ratio of the upward to downward domains is around 1:1, showing no priority. On the other hand, the crystals are immersed in water with comparable numbers of positive and negative ions, and the adsorption probability of them on the crystal surface are also comparable. In this case, the environment of the crystal is non-instructive, and there is no directional field similar to that of the aforementioned low-dimensional materials. What happens in the crystals is the polarization switching with equal opportunities in the up and down directions. From another point of view, it is the self-organization driven by the total energy of the system, manifested mainly as the coarsening of the domains with the area ratio of the upward to downward domains remains around 1:1.

Thanks for your comment, accordingly, the above discussions are added in the revised manuscript as:

Page 15 line 9-21: In the end, we perform a discussion of our work and that of others on low-dimensional materials. For low-dimensional material [22, 23, 29], an initial polarization direction is present, either upward or downward. Positive and negative charges preferentially adsorb on surfaces with initial upward and downward polarizations, respectively, resulting in a polarization reversal from upward to downward or vice versa. However, in the PMN-28PT crystal we investigated, there is

no specific initial polarization direction in the crystal. The area ratio of the upward to downward domains is around 1:1. When the crystal is immersed in water, a non-instructive environment with comparable numbers of positive and negative ions, polarization switching occurs with equal probabilities in the upward and downward directions, leading to the self-organized growth of domains driven by the system's total energy. Throughout the evolution process, the proportion of upward and downward polarization areas remains roughly equal.

■ **Comment 7**

Furthermore, it does not become clear if this domain pattern change is only a surface effect or if it can be expected to reach through the bulk.

■ **Response**

We thank the reviewer for this insightful comment. The evolution of domain patterns is considered to reach into the bulk, not only a surface effect. According to your comment, we performed additional experiments to further clarify this point.

It is known for ferroelectrics that the upward polarizations lead to faster etching rates than downward in acid solutions [5-6]. Here, we use the buffered oxide etchant (BOE 6:1, Average etching rate: about 5 nm/s) to etch a treated crystal (in water with ultrasound for 120 min). Fig. R3-3 shows the 3D topography and PFM images after etching. We can see that after etching, the boundaries in the topography images coincide with that in the corresponding PFM images. Moreover, as the etching time increases, the height difference between the upward and downward polar regions enlarges, and the domain structure shown by PFM does not change.

These phenomena suggest that the domain configuration on the surface extends to the interior of the crystal.

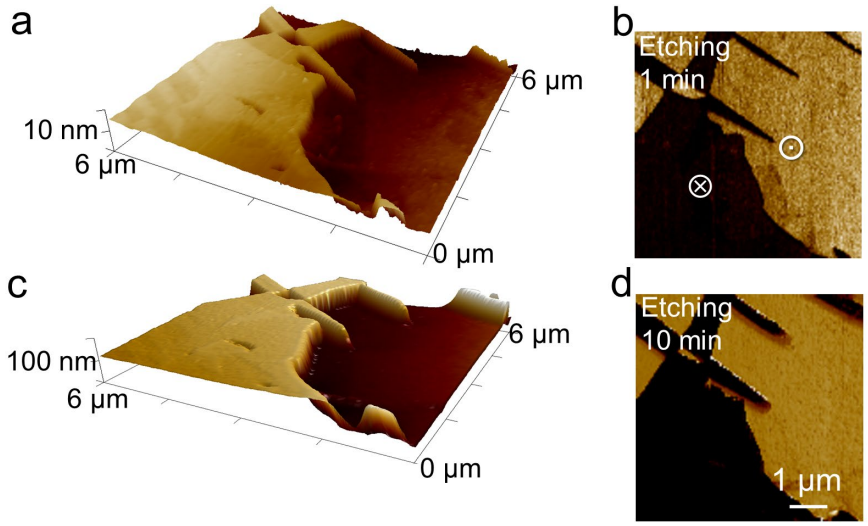

Fig. R3-3 3D morphology and PFM images of a treated PMN-28PT crystal (in water with 120 min ultrasound) after etching for (a, b) 1 min, and (c, d)10 min. The measurements were performed at the same location of the crystal.

Thanks for your comment, Figure R3-3 is added as **Fig. S8**. The above discussions are added in the revised manuscript as:

Page 9, line 14-20: The stable domain structures are expected to extend to the interior of the crystal. To confirm this, we used the buffered oxide etchant to etch a treated crystal, as we know that the upward polarizations lead to faster etching rates than downward [5-6]. Based on the 3D topography and PFM images in Fig. S8 after etching, the boundaries in the topography images coincide with that in the corresponding PFM images. Moreover, as the etching time increased, the height difference between the upward and downward polar regions enlarged, and the domain structure shown by PFM did not change.

■ **Comment 8**

On page 8 the authors mentioned that they broke the time resolution limit of PFM. It is not quite clear what is meant by that.

■ **Response**

As a technique using scanning tip, the time resolution limit of PFM refers to the time interval between two successive images, which is typically no less than 17 min (scan rate 1 Hz, lines 512). Consequently, it is challenging using PFM to capture the

rapid and continuous evolution of domain patterns in water with ultrasound. However, the domain patterns stop changing (frozen) once the crystal exits the water (and the ultrasound), allowing for a recording of the domain growth process.

■ **Comment 9**

The P-E loops shown in the appendix indicate that after soaking in water the switching dynamics are stretched out over a longer time scale and the authors argue that the coercive fields vary more for large domains than for small domains. It does not become clear why there should be a change in the coercive field distribution just based on the domain structures. The authors should elaborate more on that.

■ **Response**

We appreciate the reviewer's comment.

The macroscopic P-E loop originates mainly from microscopic domain switching [7]. The change of the hysteresis loops can be explained by the variant threshold voltage of the domains with different size. In the samples with smaller average domain size, the size and threshold voltage of different domains are close. Therefore, most domains reverse nearly simultaneously as the applied voltage increases to a certain value, leading to a rapid increase in the macroscopic polarization. Conversely, in the samples with larger average domain size, the size and threshold voltage of different domains vary significantly. As the applied voltage increases, the domains with different size reverse one after another, resulting in a slow increase in the macroscopic polarization.

Thank you for your comment, and we have incorporated the above discussions in the revised manuscript as:

Page 14 line 22 to Page 15 line 8: The change of the hysteresis loops can be explained by the variant threshold voltage of the domains with different size [52]. In the samples with smaller average domain size, the size and threshold voltage of different domains are close. Therefore, most domains reverse nearly simultaneously as the applied voltage increases to a certain value, resulting in a rapid increase in macroscopic polarization. Conversely, in the samples with larger average domain size, the size and threshold

voltage of different domains vary significantly. Then as the applied voltage increases, the domains with different sizes reverse one after another, leading to a slow increase in the macroscopic polarization.

■ **Comment 10**

Related to this point: the argumentation in the paper does not include the possibility that the material surface of the samples has changed chemically due to the soaking procedure apart from the presence of adsorbates. It is known that perovskite systems easily loose preferably A-site cations, but as well B-site cations when in contact with water. Maybe this could be a reason for the changes observed in the polarization loops? And as well influence the domain pattern? If the pH of the water monitored during soaking, this could give indications as well.

■ **Response**

We express our gratitude to the reviewer for providing us with a constructive comment. After careful consideration, we think that the chemical change in the surface is unlikely to be the primary cause of the changes in domain patterns and the loops. Our judgment is based on the following observed phenomena:

(1) According to your suggestion, we monitored the pH of the water during soaking. We placed an untreated PMN-28PT crystal in a 25 ml beaker containing 20 ml of water. Over a 5-hour soaking period, we did not observe any change in the pH beyond the error range (less than 0.05) of the pH meter.

(2) Fig. R3-4 gives the topography and the PFM phase images before and after 5-hour water treatment. After soaking, the domain structure changed, while the surface topography did not show any change, indicating that the water treatment did not lead to the degradation of the crystal surface [2].

(3) The change in the domain pattern can be well explained by the ion adsorption-induced polarization switching, which was supported by the first principles calculations and Monte Carlo simulation. Meanwhile, as discussed in Comment 9, the change in the hysteresis loops is closely linked with the ferroelectric domain structure.

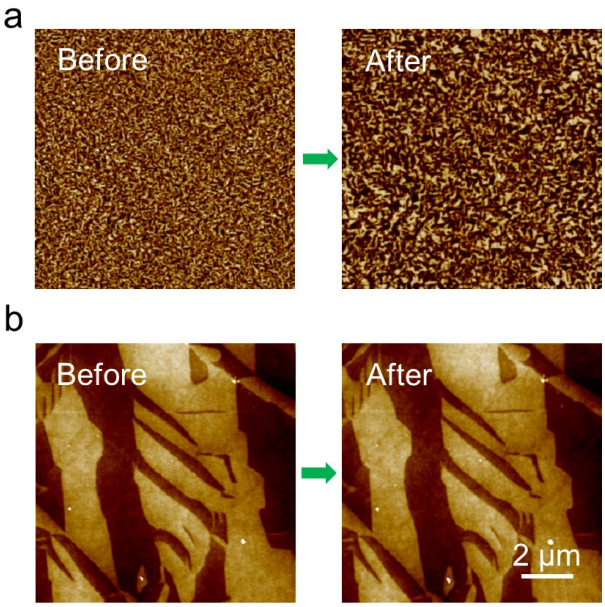

Fig. R3-4 (a) topography and (b) PFM images of the crystals before and after water treatment. The measurements were performed in the same location of the crystal.

According to your comment, the above discussions are added as:

Page 5 line 14-16: What's more, the topography did not show any change (Fig. S1), indicating that the water treatment did not lead to any degradation of the sample surface [29].

■ Reference

1. Falsi, L. *et al.* Direct Observation of Fractal-Dimensional Percolation in the 3D Cluster Dynamics of a Ferroelectric Supercrystal. *Phys. Rev. Lett.* **126**, 037601 (2021).
2. Lee, H. *et al.* Imprint Control of $BaTiO_3$ Thin Films via Chemically Induced Surface Polarization Pinning. *Nano Lett.* **16**, 2400–2406 (2016).
3. Tian, Y. *et al.* Water printing of ferroelectric polarization. *Nat. Commun.* **9**, 3809 (2018).
4. Xu, D. D. *et al.* Ion adsorption-induced reversible polarization switching of a van der Waals layered ferroelectric. *Nat. Commun.* **12**, 655 (2021).
5. Hooton, J. A. & Merz, W. J. Etch Patterns and Ferroelectric Domains in $BaTiO_3$ Single Crystals. *Phys. Rev.* **98**, 409 (1955).
6. Peng, J., Chao, C., Dai, J., Chan, H. L. W. & Luo, H. Micro-patterning of $0.70Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.30PbTiO_3$ single crystals by ultrasonic wet chemical etching. *Mater. Lett.* **62**, 3127 (2008).
7. Jin, L., Li, F. & Zhang, S. Decoding the fingerprint of ferroelectric loops: Comprehension of the material properties and structures. *J. Am. Ceram. Soc.* **97**, 1–27 (2014).

4th Apr 23

Dear Professor Lu,

Thank you for submitting your revised manuscript, "Self-organization of ferroelectric domains induced by water and reinforced via ultrasonic vibration", to Communications Materials. It has now been seen again by the 3 referees, whose comments are appended below. You will see that Reviewer 1 and 2 support publication, but Reviewer 3 raises important points that they feel have not been addressed from their first report. We remain interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

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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- Other unique identifiers (such as DOIs and hyperlinks for any other datasets)
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We hope to receive your revised paper within three months; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still 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 understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and 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 paper is accepted.

Reviewer #2 (Remarks to the Author):

The authors have performed more experiments to address raised questions and I am convinced by their results and response. Therefore, I think the manuscript is now up to the standard for publication.

Reviewer #3 (Remarks to the Author):

Most comments were addressed satisfactorily, but a few open questions remain:

- regarding the question if the observed effect on the domain structure is a surface or a bulk effect, the authors etched the surface to reveal the depth of the domain structure. However, the etching procedure could only reveal the subsurface (from the images given about 8 - 55 nm down). Domains are usually 3D structures, so it is expected that the subsurface shows a similar domain structure than the surface, as the same domains are imaged as the ones seen on the surface. That does not clarify the question if the effect influences the whole bulk of the sample.

- regarding the question about the broken time-resolution, the authors state that the domain patterns freeze in after they are taken out of the water-ultrasound bath, which then makes it possible to image the evolution of the water induced changes. I would think there is no proof for this. There is certainly a significant time gap between the end of the treatment and beginning of the PFM measurement, where there exists no information about the potential relaxation of the domain pattern.

- regarding the changes in polarization switching dynamics after soaking, the authors point out that in a sample with small domains the switching field distribution is narrow. They point towards the paper of Jin, Li and Zhang to support their statement. When I re-read the paper, I could not find supporting arguments in it. The only point, where switching dynamics are discussed is on page 8.

However, there they compare the dynamics of polar nanoregions (PNRs) with those of ferroelectric domains. The statements made there do not hold for the situation present in the current manuscript, as PNRs show non-correlated switching behavior, whereas ferroelectric domain behave in a correlated manner. There is more that influences the switching dynamics in the paper of Zhang than just the size.
furthermore, this paper points out multiple times that domain switching and with this the polarization response is strongly influenced by all kinds of defects present in the system. Point defects as introduced during processing (deliberately or not) pin domain walls and introduce all kind of changes to the P-loops depending on their kind, location, number, mobility,.... . Time dependent effects like aging are discussed. As pointed out below, the creation of surface defects is quite likely to happen during the soaking procedure, which would alter the way domain walls are pinned on the surfaces (as well as source of pinning mentioned in the Zhang paper). I think this point can not be completely neglected.

- regarding the possibility of surface dissolution, the authors added an experiment measuring the pH during soaking, where they did not observe any change over a 5 hour period. Did you as well calculate how many ions would need to dissolve to observe a change in pH? As a single crystal was used, the surface area from which dissolution occurs is rather limited and depending on the ratio of surface area to liquid volume it could be that no change is observed in the pH even if the whole outer layer of the single crystal was dissolved.

- in figure R3-4 the authors give additional topography and domain structure images and state that the topography does not show changes, whereas the domain structure does. Looking at r3-4 I would say it is exactly the opposite: in a) there is a clear coarsening of the topography, whereas in b) the images look quite identical. Please comment.
Based on my last three comments, I think that the influence of surface dissolution on the topography, domain patterns and functional properties is still open for debate.

Point-by-point Response to Reviewers' Comments

Manuscript number: COMMSMAT-22-0314A

Title: Self-organization of ferroelectric domains induced by water and reinforced via ultrasonic vibration

Authors: Shuo Yan, Xueli Hu, Xiaomei Lu, Junting Zhang, Xiaofan Shen, and Fengzhen Huang

We express our gratitude to the reviewers for their insightful evaluation and constructive suggestions. Below we address the itemized comments and provide our response with the updated text, figures, and relevant literature references.

The reviewers' comments are given in italics, our responses in normal font, and the changes in the main text are given in blue font.

- **Reviewer #1:**

The paper is accepted.

- **Response**

We thank the reviewer again for the valuable suggestions and affirmation of our revised manuscript.

- **Reviewer #2:**

The authors have performed more experiments to address raised questions and I am convinced by their results and response. Therefore, I think the manuscript is now up to the standard for publication.

- **Response**

We thank the reviewer again for the valuable suggestions and affirmation of our revised manuscript.

- **Reviewer #3:**

Most comments were addressed satisfactorily, but a few open questions remain:

- **Response**

Thanks for your positive feedback on our manuscript. We greatly appreciate your

time and effort in reviewing our work. In response to the new comments, we have carefully revised the manuscript again.

Before addressing your comments, we would like to express our sincere apologies for any confusion caused by Fig. R3-4 in our previous response letter. We have identified an error in the figure captions, that is, the captions of Fig. R3-4a and Fig. R3-4b were exchanged in mistake. And we would like to clarify that Fig. R3-4a is the PFM images, while Fig. R3-4b is the topography images. Therefore, we confirm that the surface morphology did not change before and after water treatment.

Once again, we appreciate your valuable feedback and hope that our revised manuscript meets your expectations.

■ **Comment 1**

- regarding the question if the observed effect on the domain structure is a surface or a bulk effect, the authors etched the surface to reveal the depth of the domain structure. However, the etching procedure could only reveal the subsurface (from the images given about 8 - 55 nm down). Domains are usually 3D structures, so it is expected that the subsurface shows a similar domain structure than the surface, as the same domains are imaged as the ones seen on the surface. That does not clarify the question if the effect influences the whole bulk of the sample.

■ **Response**

Thank you for your question, and we apologize for the confusion caused by our previous statement. We would like to clarify that both the upward and downward surfaces of the crystal can be etched by buffered oxide etchant (BOE) with an average etching rate of about 5 nm/s, although the etching rate of the upward surface is slightly faster than that of the downward surface [1, 2]. In our experiment, we etched the crystal for 10 minutes, resulting in a total etching depth of about 3 μm and a height difference of approximately 100 nm between the upward and downward surface.

To further investigate the domain structure, we utilized confocal second-harmonic microscopy, which is an effective imaging technique for analyzing domain patterns within crystals [3]. **Figure R1** presents the 3D domain image of the treated (in water

with 120 min ultrasound) PMN-28PT crystal to a depth of 20 μm beneath the crystal surface, which suggests that the domain patterns are not limited to the surface but extend deep into the bulk.

To our understanding, the observed change in domain structure is a bulk effect. However, it does not mean that the domains observed on the surface penetrate the entire 500 μm thickness of the crystal, a point for which we cannot provide conclusive evidence at present.

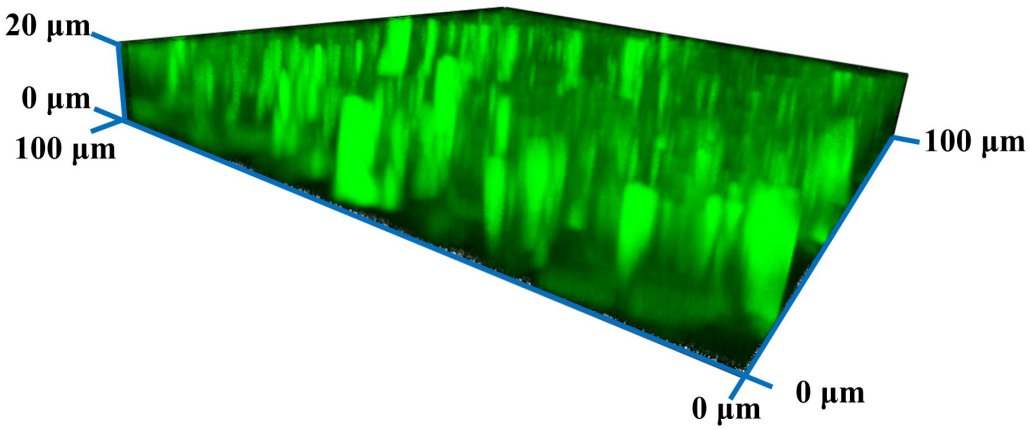

Figure R1 3D domain image recorded using confocal second-harmonic microscopic system.

Thanks for your comment. **Figure R1** is added as Fig. S9 in the supplementary material. The above discussions are added in the revised manuscript as:

Page 9 line 11-19: To investigate the domain structure inside the bulk of the crystal, we used the buffered oxide etchant to etch a treated crystal [35]. The results presented in Fig. S8 demonstrate that the domain structure remains unchanged up to an etching depth of approximately 3 μm. Furthermore, confocal second-harmonic microscopy [36] reveals that the domain evolution extends to a depth of at least 20 μm beneath the surface (Fig. S9). The water-induced domain evolution is expected to extend to the interior of the crystal. However, it does not necessary mean that the domains observed on the surface penetrate the entire 500 μm thickness of the crystal, as conclusive evidence for this point is not currently available.

■ **Comment 2**

- regarding the question about the broken time-resolution, the authors state that the domain patterns freeze in after they are taken out of the water-ultrasound bath, which then makes it possible to image the evolution of the water induced changes. I would think there is no proof for this. There is certainly a significant time gap between the end of the treatment and beginning of the PFM measurement, where there exists no information about the potential relaxation of the domain pattern.

■ **Response**

We would like to express our appreciation to the reviewer for the constructive comments.

We believe that the domains are stable after water treatment for the following reasons. Firstly, our analysis in Fig. 2 suggests that the sufficient surface screening charges from water assist in stabilizing the domain structures. Secondly, as mentioned in Comment 1, the domain evolution extends to the interior of the crystal, which may also contribute to stability. Thirdly, after water treatment, the samples were dried quickly by N_2 gun, and our experimental observations (Fig. S7) indicate that the domain pattern remains unchanged after being kept in air for 1 and 48 hours. Lastly, our experimental observations in Fig. 3b show that, as the water treatment time increases, the evolution of the domain pattern is continuous. If the domains relax in a short time, it is hard to imagine the current results.

However, with regard to the reviewer's comment on the time gap between the end of treatment and beginning of PFM measurement, we recognize that we do not have direct evidence of domain relaxation during this period. Therefore, we removed the terms "frozen" and "break the time resolution of PFM", and revised the related descriptions as:

Page 9 line 4-10: The water-induced self-organized domain structure was quite stable, and no change was observed after the sample being kept in atmosphere for a long time (Fig. S7). This is also the key point of the self-organization process, that is, although the formation of ordered domain patterns is driven by the interaction among dipoles, energy input is required from the outside. Once the energy input stops, the self-

organization process stops. Based on this special mechanism of domain growth, we were able to record the growth and merge process of ferroelectric domains.

■ **Comment 3**

- regarding the changes in polarization switching dynamics after soaking, the authors point out that in a sample with small domains the switching field distribution is narrow. They point towards the paper of Jin, Li and Zhang to support their statement. When I re-read the paper, I could not find supporting arguments in it. The only point, where switching dynamics are discussed is on page 8. However, there they compare the dynamics of polar nanoregions (PNRs) with those of ferroelectric domains. The statements made there do not hold for the situation present in the current manuscript, as PNRs show non-correlated switching behavior, whereas ferroelectric domain behave in a correlated manner. There is more that influences the switching dynamics in the paper of Zhang than just the size.

furthermore, this paper points out multiple times that domain switching and with this the polarization response is strongly influenced by all kinds of defects present in the system. Point defects as introduced during processing (deliberately or not) pin domain walls and introduce all kind of changes to the P-loops depending on their kind, location, number, mobility,.... . Time dependent effects like aging are discussed. As pointed out below, the creation of surface defects is quite likely to happen during the soaking procedure, which would alter the way domain walls are pinned on the surfaces (as well as source of pinning mentioned in the Zhang paper). I think this point can not be completely neglected.

■ **Response**

We thank the reviewer for this valuable comment.

1. Switching field

We agree with the reviewer that there is a distinction between PNRs and domains. The reason why we cite Zhang's article is that the article mentions both microdomains and PNRs. There is such a sentence on page 8 "Due to their much smaller characteristic size, microdomains response to the external field much faster than macroscopic

domain....” [4]. In addition, as shown in **Figure R2** (Nature, 2016, 534(7607): 360-363, Fig. 3a), some other researchers believe that the coercive field of small domain is smaller than that of large ones [5], which can be obvious at the measuring frequency of 100 Hz in our experiment.

Moreover, we have observed that the distribution of domain sizes varies among different samples. In sample S (treated for 5 min, Fig. S12), the size of different domains is similar, whereas in sample L (treated for 120 min, Fig. S12), the domain size varies widely. We propose that the distribution of domain size contributes much to the observed loop shapes.

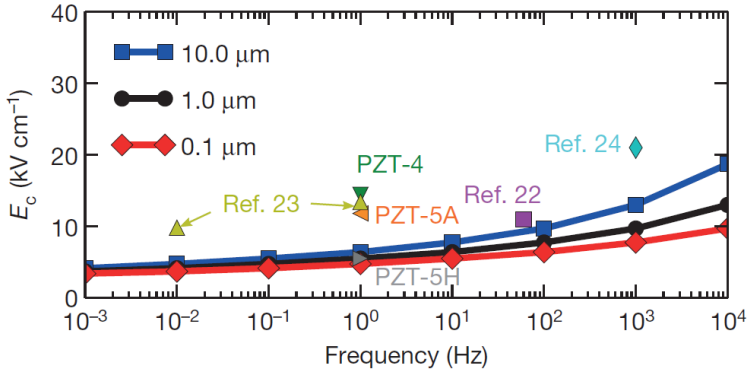

Figure R2 Simulated frequency dependence of coercive fields Ec for PZT ceramics for various domain sizes (see legend) at 300 K. Theoretical values are comparable to various experimental values. (Nature, 2016, 534(7607): 360-363, Fig. 3a)

2. Defects

We apologize for the confusion caused by Fig. R3-4 in the previous response letter. As described in the beginning of the response and detailed later in Comment 5, the captions of the former Fig. R3-4a and Fig. R3-4b were exchanged in mistake. We confirm that the surface topography did not change before and after 5 h water treatment.

In addition, we provided the corresponding topography images for Fig. 3b in the supplementary material as Fig. S4. As shown here in **Figure R3**, the topography of the crystals remains unchanged, despite the significant domain changes observed in Fig. 3b, indicating the absence of observable surface defects. However, as the reviewer suggested, we cannot completely rule out the possibility that defects may form during the soaking procedure, which could potentially affect the ferroelectric loop by pinning.

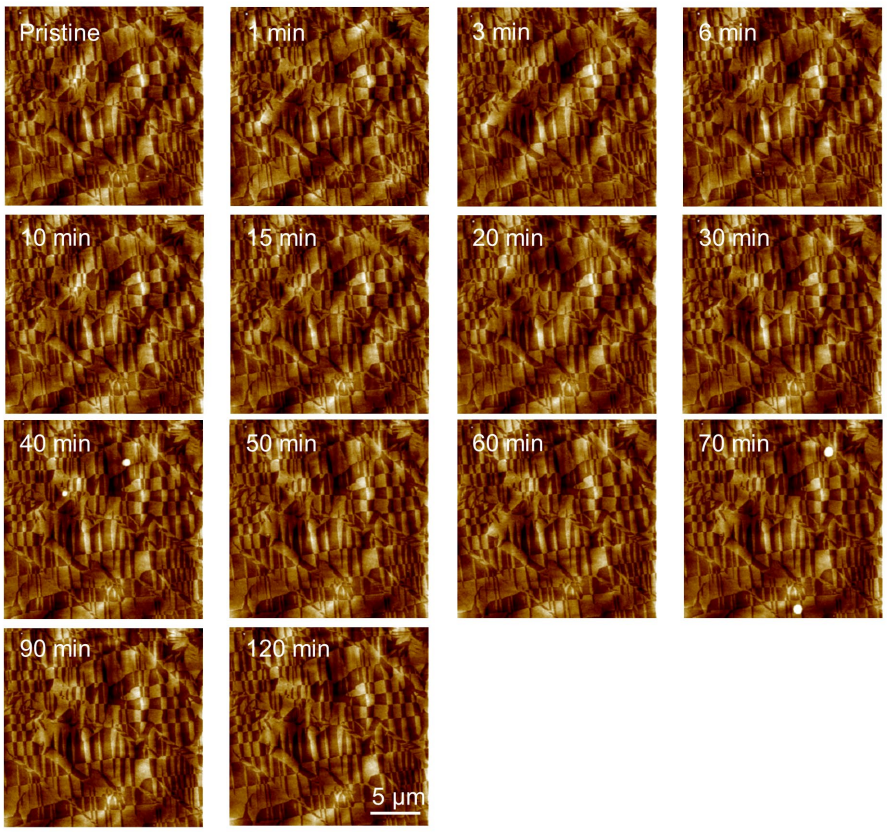

Figure R3 Topography images of the PMN-28PT crystal with different time in water under ultrasonic vibration, in corresponding to the PFM phase images in Fig. 3b. The measurements were performed at the same location.

In summary, we agree that ferroelectric switching can be influenced by many factors. Although domain structure can be considered a dominant factor in many cases, we cannot completely ignore other possibilities. According to the above discussion, we have made revisions to our manuscript, improving the discussion on the influence of domain size distribution on ferroelectric loops and adding a description on the potential impact of defects.

Page 14 line 17 to Page 15 line 7: We try to understand the shape of the hysteresis loops based on the variant threshold voltage of each domain related to the domain size, which is expected to be larger for larger domains [53, 54]. In sample S (treated for 5 min), the size and the threshold voltage of different domains are similar. While in sample L (treated for 120 min), the size and the threshold voltage of different domains exhibit a significant range of variation, resulting in a distribution in the switching

threshold of different domains. As the applied voltage increases, the domains in sample L might reverse gradually starting from smaller ones, leading to a slow increase in the macroscopic polarization. Although the domain structure is considered dominating the ferroelectric hysteresis behavior, it should be noted that ferroelectric switching can be affected by many factors. Here we cannot discount the possibility that defects may form during the water treatment process, which may also contribute to the changes in hysteresis loops through pinning.

■ **Comment 4**

- regarding the possibility of surface dissolution, the authors added an experiment measuring the pH during soaking, where they did not observe any change over a 5 hour period. Did you as well calculate how many ions would need to dissolve to observe a change in pH? As a single crystal was used, the surface area from which dissolution occurs is rather limited and depending on the ratio of surface area to liquid volume it could be that no change is observed in the pH even if the whole outer layer of the single crystal was dissolved.

■ **Response**

We thank the reviewer for this advice.

We concur with your assessment that detecting pH changes would be challenging, even if the entire outer layer of the crystal was dissolved. Nonetheless, as we discussed in Comment 3 above and Comment 5 below, we observed changes in the domain structure after water treatment, while the surface topography remained unchanged. This finding implies that the water treatment did not lead to any observable degradation of the crystal surface.

■ **Comment 5**

- in figure R3-4 the authors give additional topography and domain structure images and state that the topography does not show changes, whereas the domain structure does. Looking at r3-4 I would say it is exactly the opposite: in a) there is a clear coarsening of the topography, whereas in b) the images look quite identical.

Please comment.

■ **Response**

We thank the reviewer for bringing up this question. We sincerely apologize for the error in the labeling of the images and any confusion it may have caused. In the last response letter, the captions of Fig. R3-4a and Fig. R3-4b were exchanged in mistake, and the corrections is shown here as **Figure R4**. The images in the former Fig. R3-4b are actually topography images, which are quite identical as you mentioned. We confirm that the surface topography remained unchanged after water treatment, while the domain structure did change.

This result is consistent with that provided as the supplementary material (Fig. S1a and Fig. S4) in the last round of review. Fig. S1a and Fig. S4 give the corresponding topography images of Fig. 1b and Fig. 3b, which shows that, after water treatment, no matter with ultrasound or not, the topography of the sample remains unchanged.

We have carefully reviewed the manuscript to ensure that there are no further errors.

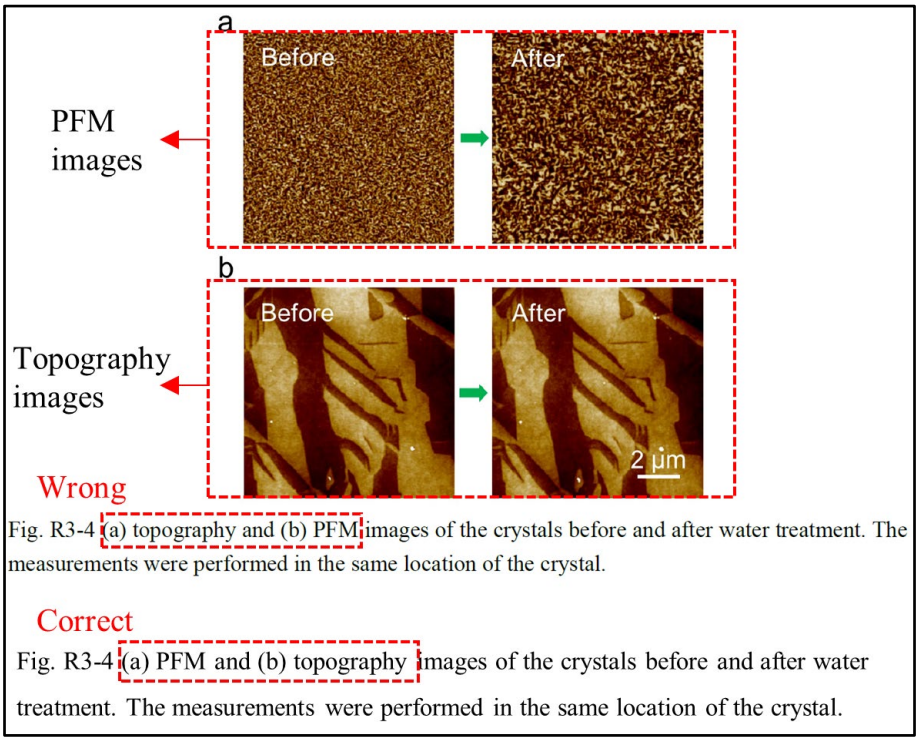

Figure R4 Corrections to the former Fig. R3-4.

■ **Comment 6**

Based on my last three comments, I think that the influence of surface dissolution

on the topography, domain patterns and functional properties is still open for debate.

■ **Response**

Thanks for your comment.

We apologize again for the confusion caused by the mistake in the captions of Fig. R3-4a and Fig. R3-4b. Based on our responses in Comments 3, 4, and 5, it appears that the surface topography remains unchanged while the domain structure changes, indicating that the water treatment did not cause any observable surface dissolution. However, we also agree that, due to the limitations of current research methods, we cannot completely rule out the possibility of defects and their potential impacts on the ferroelectric behavior. According to your comments, we have revised our manuscript carefully as described above.

■ **Reference**

[1] Hooton, J. A. & Merz, W. J. Etch Patterns and Ferroelectric Domains in $BaTiO_3$ Single Crystals. Phys. Rev. 98, 409 (1955).

[2] Peng, J., Chao, C., Dai, J., Chan, H. L. W. & Luo, H. Micro-patterning of $0.70Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.30PbTiO_3$ single crystals by ultrasonic wet chemical etching. Mater. Lett. 62, 3127 (2008).

[3] Wei, D. et al. Efficient nonlinear beam shaping in three-dimensional lithium niobate nonlinear photonic crystals. Nat. Commun. 10, 4193 (2019).

[4] Jin, L., Li, F. & Zhang, S. Decoding the fingerprint of ferroelectric loops: Comprehension of the material properties and structures. J. Am. Ceram. Soc. 97, 1–27 (2014).

[5] Liu, S., Grinberg, I. & Rappe, A. M. Intrinsic ferroelectric switching from first principles. Nature 534, 360–363 (2016).

2nd May 23

Dear Professor Lu,

Your manuscript titled "Self-organization of ferroelectric domains induced by water and reinforced via ultrasonic vibration" has now been seen again by Reviewer 3, 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 under the open access CC BY license (Creative Commons Attribution v4.0 International License).

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.

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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 #3 (Remarks to the Author):

The authors have addressed all remaining questions in a concise and constructive way. It was a pleasure to review and discuss this manuscript.