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| 09 Nov 2018 |
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# Title: Tuning oxide properties by oxygen vacancy control during growth and annealing Authors: Dennis V Christensen, Felix Trier, Merlin von Soosten, Yulin Gan, Yu Zhang, Simone Sanna, Yunzhong Chen, and Nini Pryds

# Dear editor

Thank you for your recent communication regarding our submission JoVE58737. We greatly appreciate the overall positive comments received from the two reviewers, and we have modified the manuscript according to your as well as their suggestions. Point-by-point comments are found later in this document marked with blue.

Please do not hesitate to contact us for further clarification.

On behalf of all the co-authors,  
sincerely,

Dennis Christensen

**Editorial comments:**  
Changes to be made by the Author(s) regarding the written manuscript:  
1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.  
Author: We have checked the manuscript carefully for spelling and grammar issues.  
  
2. Please obtain explicit copyright permission to reuse any figures from a previous publication. Explicit permission can be expressed in the form of a letter from the editor or a link to the editorial policy that allows re-prints. Please upload this information as a .doc or .docx file to your Editorial Manager account. The Figure must be cited appropriately in the Figure Legend, i.e. “This figure has been modified from [citation].”  
Author: Copyright permissions are included.  
  
3. Figure 4: Please use °C for temperature unit. Please include a space between all numbers and their corresponding units (i.e., 20 °C, 37 °C, etc.).  
Author: Revised accordingly.  
  
4. Please provide figures with higher resolution if possible.  
Author: All figures are now provided in high quality.  
  
5. Please revise the title to be more concise.  
Author: The title has been shortened to “Tuning oxide properties by oxygen vacancy control during growth and annealing”  
  
6. Please provide an email address for each author.  
Author: Emails are added.  
  
7. JoVE cannot publish manuscripts containing commercial language. This includes trademark symbols (™), registered symbols (®), and company names before an instrument or reagent. Please remove all commercial language from your manuscript and use generic terms instead. All commercial products should be sufficiently referenced in the Table of Materials and Reagents. For example: Crystec, milli-Q, LEMO, etc.  
Author: Revised accordingly.  
  
8. Please revise the protocol text to avoid the use of any personal pronouns (e.g., "we", "you", "our" etc.).  
Author: Revised accordingly.  
  
9. Please revise the protocol to contain only action items that direct the reader to do something (e.g., “Do this,” “Ensure that,” etc.). The actions should be described in the imperative tense in complete sentences wherever possible. Avoid usage of phrases such as “could be,” “should be,” and “would be” throughout the Protocol. Any text that cannot be written in the imperative tense may be added as a “Note.” Please include all safety procedures and use of hoods, etc. However, notes should be used sparingly and actions should be described in the imperative tense wherever possible.  
Author: Revised accordingly.  
  
10. Lines 100-105: The Protocol should contain only action items that direct the reader to do something. Please move the introduction about the protocol to the end of Introduction section.  
Author: Revised accordingly.  
  
11. Please simplify the Protocol so that individual steps contain only 2-3 actions per step and a maximum of 4 sentences per step. Use sub-steps as necessary.  
Author: Revised accordingly.

12. Please add more details to your protocol steps. There should be enough detail in each step to supplement the actions seen in the video so that viewers can easily replicate the protocol. Please ensure you answer the “how” question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action. Some examples:  
Lines 119-123: Please specify the mass of substrates and volume of water/acidic solution added.  
Author: Example of acid volume is added. The mass of the substrate (less important) follows directly from its size (more important), which is stated above.   
  
Lines 125-126: Please specify again the volume of water used to wash.  
Author: Revised accordingly  
  
1.1.3: What type of device is used for thermal treatment?  
Author: Revised accordingly  
  
2.1: Please describe how this is done.  
Author: Revised accordingly  
  
13. Please include single-line spaces between all paragraphs, headings, steps, etc.  
Author: Revised accordingly  
  
14. After you have made all the recommended changes to your protocol (listed above), please highlight 2.75 pages or less of the Protocol (including headings and spacing) that identifies the essential steps of the protocol for the video, i.e., the steps that should be visualized to tell the most cohesive story of the Protocol.   
Author: Highlighted in yellow   
  
15. Please highlight complete sentences (not parts of sentences). Please ensure that the highlighted part of the step includes at least one action that is written in imperative tense.   
Author: Highlighted accordingly  
  
16. Please include all relevant details that are required to perform the step in the highlighting. For example: If step 2.5 is highlighted for filming and the details of how to perform the step are given in steps 2.5.1 and 2.5.2, then the sub-steps where the details are provided must be highlighted.   
Author: Highlighted accordingly  
  
17. Discussion: Please also discuss critical steps within the protocol.  
Author: The most critical parts of the protocol are to get the temperature profiles and oxygen partial pressures right. This has been merged into the discussion.  
  
18. References: Please do not abbreviate journal titles.

Author: Revised accordingly.

**Reviewers' comments:**  
  
**Reviewer #1:**  
Manuscript Summary:  
The manuscript "Tuning oxide properties by oxygen vacancy control during growth and annealing - the case of SrTiO3-based heterostructures" by Christensen et al. describes experimental means to control and characterize the oxygen vacancy concentration and profile in SrTiO3-based 2DEG electron gases. The contribution is very interesting and suitable to be illustrated by video. The accompanying manuscript reads well and explains all experimental procedure clearly and in sufficient detail. Therefore, I am happy to recommend its publication.  
Author: We thank the reviewer for the positive evaluation of the manuscript.

Minor Concerns:  
While reading, I had a few minor issues that may be addressed by the authors:  
  
End of first paragraph (lines 56, 57): I think it may be useful to generalize the first section also to defects beyond oxygen vacancies. They are of course the most prominent species, also the most relevant to this contribution, however, other defects such as cation vacancies, anti-site, extended defects, etc. can all cause strain fields, break symmetry and modify transport. Therefore, I think it is important to note that oxygen vacancy defects are not the only engineering tool available in oxides.  
Author: The reviewer is certainly correct that other defects than oxygen vacancies may also play a very important role in determining the properties of oxides. Controlling these defects can therefore also be used to tune the properties. We have extended the sentences in question in the introduction to cover both oxygen vacancies as well as other defects. In addition, we have added the following sentence to the discussion: “It should, however, be noted that in some cases cation diffusion and variations in the defect configuration induced by the deposition or post-annealing can also be a desirable way to tune the oxide properties.”  
  
Line 67: Reference for opaqueness of SrTIO3 after reduction: Ref.2 refers to a 2017-paper - however, this phenomenon was of course observed already much earlier. I feel a proper reference to original literature should be added here.  
Author: The change in color when introducing oxygen vacancies in SrTiO3 has been known for more than half a century, however, we have not been able to find the original reference. We therefore simply use one of the references already used in the manuscript, which also happens to show this change in the optical properties very clearly.   
  
Protocol step 1.1.3.: Is it really to relax surface tension or to allow formation of smooth step terraces? Or is this what the authors mean?  
Author: The annealing process is to bring the substrate surface into a low energy state. We have rephrased the sentence to ”(..) to relax the substrate surface into a state with low energy”  
  
Line 232-234: Do I understand correctly that if side-diffusion is determining the conduction state, this implies that the lateral distribution of defect may vary over the sample? Hence, while the edges will be more oxidized than the center of the sample?  
Author: The oxidation in the case of GAO/STO can occur from the bottom and from the sides. This will depend on the boundary conditions of the sample, i.e. whether is it exposed air from the sides and bottom or are there some parts covered such as when mounted on a chip carrier. Therefore, the relative contributions of oxidation from the sides and the bottom may vary, and a systematic study on this under various conditions is missing. We have observed earlier some gradient in the sheet resistance along the interface pointing towards the outer parts are being somewhat more oxidized as the reviewer correctly mention.  
  
In the same section, I would also recommend to add a clearer statement that the actual state controlled via annealing depend on three parameters, i.e. temperature, annealing time and atmosphere (I assume).  
Author: The reviewer is correct, and we have now added the following sentence to the manuscript: “Here, the final state after annealing is determined by three parameters: annealing time, temperature and atmosphere”.

Figures: Please check if GAO (used in the figures instead of g-Al2O3) is properly defined in the main text (figure captions).  
Author: Excellent point. We have added the abbreviation GAO in the caption for figure 5.  
  
  
  
  
**Reviewer #2:**  
Manuscript Summary:  
The authors deal with the problem of oxygen content fine tuning in strontium titanate based heterostructure, proposing two different (but not alternative) approaches based on controlling the growth conditions and on oxygen thermal annealing respectively. The paper, mainly based on already published results (suitably cited), is clear and well-written and of some interest for the community. For such reason, I recommend it for publication in JOVE.  
Author: We thank the reviewer for the positive evaluation of the manuscript.