Peer Review File

Enhanced coherence of all-nitride superconducting qubits epitaxially grown on silicon substrate

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

7th May 21

Dear Dr Kim,

Thank you for submitting your manuscript, "Enhanced-coherence all-nitride superconducting qubit epitaxially grown on Si substrate", to Communications Materials. It has now been seen by 2 referees, whose comments are appended below. You will see that while they find your work of interest, some important points are raised. In particular, Reviewer #2 is asking to discuss the predicted/theoretical limit to the quality factor for the substrate material (MgO) used in previous reports, in order to strengthen the claim that the improvement of coherence time is mainly due the change in substrate. We believe this is a point that needs to be addressed in order to meet the level of impact that we are seeking in our journal. Naturally, we would expect all other requests and comments of the referees to be appropriately responded to, including their requests for further information and discussion.

We are 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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In particular, the Data availability statement should include:
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- Other unique identifiers (such as DOIs and hyperlinks for any other datasets)
- At a minimum, a statement confirming that all relevant data are available from the authors
- If applicable, a statement regarding data available with restrictions
- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

DATA SOURCES: We strongly encourage authors to deposit all new data associated with the paper in a persistent repository where they can be freely and enduringly accessed. We recommend submitting the data to discipline-specific, community-recognized repositories, where possible and a list of recommended repositories is provided at http://www.nature.com/sdata/policies/repositories.

If a community resource is unavailable, data can be submitted to generalist repositories such as figshare or Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

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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,

Dr Aldo Isidori
Associate Editor
Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Report about the manuscript COMMSMAT-21-0076 by Sunmi Kim et al, "Enhanced-coherence all-nitride superconducting qubit epitaxially grown on Si substrate"

At the present time the conventional Al technology with AlOx as atunnel burrier is widely used for fabrication of Al-based Josephson junctions of superconducting qubit devices with high coherence time. It is well known that the coherence time of superconducting quantum circuits made from Al-based Josephson junctions is limited by energy or phase relaxation due to microscopic two-level systems (TLS) in amorphous aluminium oxide tunnel barrier. In addition, the mechanical robustness of superconducting quantum devices fabricated from Al material is very low. Therefore the search of superconducting materials with high mechanical robustness and new tunnel burrier materials is a very important task for farther development of the large-scale superconducting quantum circuits with high coherence time.

The manuscript COMMSMAT-21-0076 reports the successful implementation of epitaxial NbN technology with AlN tunnel burrier for development of high energy relaxation time superconducting flux qubits. Indeed, the energy relaxation time of NbN/AlN/NbN C-shunt flux qubit presented in the manuscript COMMSMAT-21-0076, is of 16,3 mks is very near to the same parameter of the Al-based C-shunt flux qubit (55 kms). Replacing conventional MgO substrate by a Si substrate with a TiN buffer layer results in successful improvement in the energy relaxation time (NbN/AlN/NbN transmons on MgO substrate manifested the energy relaxation time of 500 ns, Nacamura Y. et al, Appl. Phys. Lett., Vol.99, 212502-1-3 (2011)). The manuscript COMMSMAT-21-0076 can be accepted to be published in the Communications Materials but after revision because some point have to be clarified.

Please include in the manuscript pi-pulse duration for all measurements reported in Fig. 3 (a), 4(a,b).

The realization of large scale superconducting quantum circuits require the technology which permits to fabricate large number of Josephson junctions with identical value of the Josephson critical currents, and hence, with small variation of the junction areas. Is the chemical mechanical polishing (CMP) is suitable for this purpose?

As was reported in paper Aaron D. O'Connell et al, Appl. Phys. Lett., Vol.92, 112903 (2008), the tangent loss of the MgO is of 0.5x10-4 – 0.8x10-4 while the tangent loss of the high resistance Si substrate is less than 5x10-6 – 12x10-6 . The Referee suggests to include this paper as an additional reference and use these values of the tangent loss in the discussion why the replacing of conventional MgO by a Si substrate with a TiN buffer layer leads to increase of the coherence time of superconducting qubits.

Reviewer #2 (Remarks to the Author, see also attached report):

The paper reports improved qubit coherence times for nitride-based epitaxial qubits and attributes this improvement to the low dielectric loss of the substrate material, in this case silicon, when compared to previous reports of nitride qubits fabricated using MgO substrate. The results are novel and interesting, and the significant improvement of the coherence time of nitride qubits based on substrate material is an important finding.
The report argues that the increase in qubit coherence times that they observe, when compared to previous reports of nitride qubits fabricated on MgO substrate, can be attributed to the lower dielectric loss of the silicon substrate. The evidence for their conclusion is based on the predicted qubit quality factor that would be expected with the assumption that the quality factor is limited only by the capacitive loss associated with the substrate.
To strengthen the conclusion that the substrate material is responsible for the improvement they demonstrate, they could discuss the predicted limit to the quality factor for the substrate material used in previous reports (MgO). This would strengthen the argument that the improved coherence time reported in this paper, in comparison to past reports, is due largely to changes in the substrate material.
The authors discuss several other mechanisms that could limit qubit coherence time, and argue that the Purcell effect is not responsible. They briefly mention the effect of two-level systems, but ultimately do not provide any analysis to indicate the degree to which two-level systems could be limiting the qubit coherence.
Overall, I feel the paper demonstrates a significant improvement, in comparison to past reports of nitride based superconducting qubits. As the authors readily point out, the coherence times of their qubits are well below the record value for Al-based superconducting qubits. Their report therefore demonstrates the potential for further improvements in alternative materials for superconducting qubits, and therefore may be somewhat influential in considering materials and strategies to develop qubits with increased coherence time.
The manuscript is easy to read and comprehend. There is at least one place where a symbol is used that is not defined (Qint). The manuscript is concise and direct. I feel the claims are completely in line with the evidence presented.

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Manuscript COMMSMAT-21-0076

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Response to Reviewer #1

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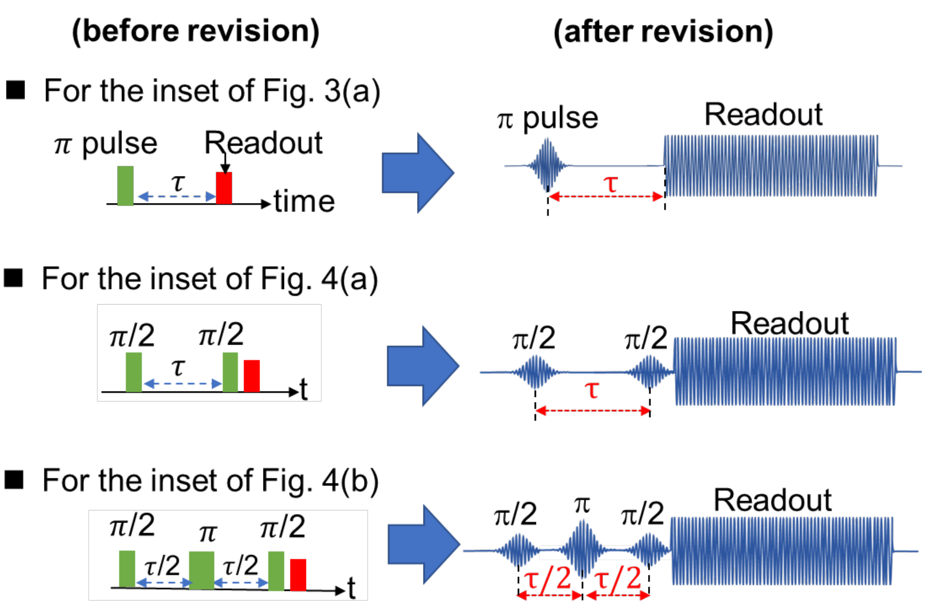
We would like to thank you for the careful reading of the manuscript and your valuable comments. Our point-by-point reply to your comments and revised points are described below. We sincerely hope that our reply satisfies you and that you now find our manuscript suitable for publication in Communications Materials.

1. Comment from Reviewer #1 mentioning "Please include in the manuscript pi-pulse duration for all measurements reported in Fig. 3 (a), 4(a,b)".
   Author response: Thank you for pointing this out. To explain the pulse sequence including pi-pulses for all measurements, we have added the following sentences (marked in red color in the manuscript) to the figure captions of Fig. 3(a), 4(a), and 4(b) as follows.

   (1) In the caption of Fig. 3 (a) on page 19:
   "**Figure 3 | Energy relaxation time $\boldsymbol{T_1}$ and its temporal variation.** (a) T_1 decay-profile with an exponential fit (solid line) with $T_1 = 18.25 \pm 0.91\ \mu$s. The inset shows the pulse sequence for T_1 measurement consisting of a π pulse (having a Gaussian envelope) with a 40 ns duration at ω_{01} and a readout pulse with a 400 ns duration at ω_r. (b) Multiple..."

   (2) In the caption of Fig. 4 on page 20:
   "**Figure 4 | Phase relaxation times: $\boldsymbol{T_2^*}$ from Ramsey measurement and $\boldsymbol{T_2}$ from spin-echo experiment.** Time-domain measurements showing (a) Ramsey fringe signal with $T_2^* = 3.33 \pm 0.30\ \mu\text{s}$ and (b) Hahn echo signal with $T_2 = 23.20 \pm 5.21\ \mu\text{s}$. The pulse sequences for the measurements are shown in the insets. The pulse sequences include a π pulse with a 40 ns duration, $\pi/2$ pulses with the same duration but half the amplitude of the π pulses, and readout pulses with 400 ns duration. Here the driving frequency ...."

   (3) Also, the corresponding insets in Fig. 3 (a) and Fig. 4(a and b) are modified as follows to express the pulse sequence properly.



2. Reviewer #1 asked whether the chemical mechanical polishing (CMP) is suitable for the fabrication of a large number of Josephson junctions with an identical value of the Josephson critical current and a small variation of the junction area.
   Author response: Thank you for your interest in our fabrication method. The uniformity of the Josephson junction properties depends on the uniformity of a thickness of AlN tunnel barrier and the patterning accuracy determined by lithography and RIE etching. The CMP process is used only to make the surface of the SiO_2 sacrificial layer flat, so there is no effect on the uniformity of the NbN/AlN/NbN junction. Therefore, we think that there is no problem in fabricating a large number of Josephson junctions with uniform junction properties and sizes with the CMP process.

3. Reviewer #1's suggestion: "As was reported in paper Aaron D. O'Connell et al, Appl. Phys. Lett., Vol.92, 112903 (2008), the tangent loss of the MgO is of 0.5×10^{-4} – 0.8×10^{-4} while the tangent loss of the high resistance Si substrate is less than 5×10^{-6} – 12×10^{-6} . The Referee suggests to include this paper as an additional reference and use these values of the tangent loss in the discussion why the replacing of conventional MgO by a Si substrate with a TiN buffer layer leads to increase of the coherence time of superconducting qubits."

Author response: Thank you for pointing this out. We have added the following sentence (marked in red) on page 4 and reference [28] on page 15 to explain why replacing the conventional MgO by a Si substrate with a TiN buffer layer leads to an increase in the coherence time of the superconducting qubit.

"To suppress the dominant dielectric loss from the MgO substrate, which has limited the energy relaxation time (T_1) in the previous work[25], we adopted a Si substrate with a TiN buffer layer for epitaxial growth of NbN/AlN/NbN junctions[27]. The loss tangent of the high-resistivity Si substrate is much lower than that of MgO, as reported in Ref. 28 ($5 \times 10^{-6} - 12 \times 10^{-6}$ for the former and $0.5 \times 10^{-4} - 0.8 \times 10^{-4}$ for the latter). Additionally, ..."

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Response to Reviewer #2

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We would like to thank you for the careful reading of the manuscript and your valuable comments. Our point-by-point reply to your comments and revised points are described below. We sincerely hope that our reply satisfies you and that you now find our manuscript suitable for publication in Communications Materials.

4. Reviewer #2 pointed out that "To strengthen the conclusion that the substrate material is responsible for the improvement they demonstrate, they could discuss the predicted limit to the quality factor for the substrate material used in previous reports (MgO). This would strengthen the argument that the improved coherence time reported in this paper, in comparison to past reports, is due largely to changes in the substrate material."

   Author response: Thank you for pointing this out. We have added the following sentences (marked in red) in the section *Main factors behind the enhanced coherence time* on page 8.

   "**Main factors behind the enhanced coherence time.** Compared with the NbN-based qubit epitaxially grown on a MgO substrate ($T_1 \approx T_2 \approx 0.5\ \mu s$)[25], the obtained coherence times for the qubit on a Si substrate ($\bar{T}_1 = 16.3\ \mu s$ and $\bar{T}_2 = 21.5\ \mu s$ as the mean values) represent a significant improvement,

namely a 32-fold increase in T_1 and a 43-fold increase in T_2. To support the argument that the substrate material is responsible for this improvement, we briefly discuss the predicted limit on the quality factor for the MgO substrate material. As reported in Ref. 25, the predicted limit for the MgO substrate clearly appears in the internal quality factor of the resonator $Q_{int} = \sim 2.35 \times 10^3$. This value of Q_{int} gives a higher value of the loss tangent for the MgO substrate $\tan\delta \sim 1/Q_{int} \sim 4.25 \times 10^{-4}$ compared to our result for the Si substrate, i.e., $\tan\delta = 3.68 \times 10^{-6}$ with $Q_{int} = 2.72 \times 10^5$. By considering the capacitive loss with the participation ratio of the capacitance across MgO among the total capacitance P_{MgO}~15% and ignoring other losses such as in the junctions as discussed in Ref. 25, the predicted limit of the quality factor of qubit (Q) on MgO substrate is estimated to be $Q \sim Q_{int}/P_{MgO} \sim 1.5 \times 10^4$ giving the expected relaxation time $T_1 = Q/(E_{01}/h)$~480 ns for E_{01}/h=5 GHz. This limit on T_1 set by the dielectric loss in the MgO substrate is indeed comparable to the experimentally observed value of $T_1 \sim 500$ ns.
We can use a similar calculation to estimate the expected quality factor of the qubit grown on a Si substrate. We first consider common sources limiting T_1, …"

5. Reviewer #2 pointed out that "They briefly mention the effect of two-level systems, but ultimately do not provide any analysis to indicate the degree to which two-level systems could be limiting the qubit coherence."
   Author response: This is indeed a good point. The contribution of two-level systems to the decoherence of the qubit cannot be ignored. As our response of this comment, we have added following sentences on page 7.
   "To reach a quantitative understanding of the degree to which two-level systems could be limiting the qubit coherence time, more experiments are needed. Such an investigation is beyond the scope of this work and could be the subject of future work."

6. Reviewer #2 pointed out that "There is at least one place where a symbol is used that is not defined (Q_{int})."
   Author response:
   (1) Since you mention "at least one place", we suspect that this might be a misunderstanding and we appreciate it if the reviewer can be more

specific. In terms of the symbol Q_{int} has been defined on page 5 in the manuscript.

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Summary of changes made

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Please refer the Marked-Up Manuscript (marked in red color) for this summary.

1. **[Figures]**
   (1) To clearly answer the comment of Reviewer #1(Comment 1), we added pi-pulse duration for all measurements in the caption of Fig. 3 and 4.
   (2) We also modified the insets in Fig. 3 (a) and Fig. 4(a and b) are modified for proper description to the applied pulse.
   (3) The fonts of T_1, T_2^*, and T_2 in of Fig. 3 and 4 are revised to be T_1, T_2^*, and T_2 (i.e., T becomes the italic character).

2. **[Reference]** In relation to Comment 3, new reference (Ref. 28) is added. Accordingly, the reference 28~37 is revised to 29~38.

3. **[Symbol]** The symbol of qubit quality factor is replaced with the broadly used 'Q' to avoid unconventional expression such as Q_1 on page 9.

4. Responding to Comment 3, additional explanations are added on page 4.

5. In relation to Comment 4, (1) new explanation is added on page 8 and 9 as described in author response 4.
   (2) We also removed the section of 'Qubit quality factors' on Page 8 to avoid overlap with the added sentences.
   (3) The term "To discuss the major factors contributing to the improvement in coherence time compared to the NbN-based transmon qubit epitaxially grown on MgO substrate[25]" (on page 9) is removed for smooth connection between new sentence and original sentence.

6. Regarding Comment 5, we have added an additional sentence to clarify the limitation of our analysis on page 7.

7. Considering Comment 6, the additional sentences are added on page 8.

8. We replace "63.14 μs" to "63 μs" in the first line on page 10.

9. The following modifications have been made.
   (1) The variable x of AlO_x on Page 1 and 3 is revised to be italic (i.e. AlO_{x}).
   (2) On page 6, the unit dimension of 38.6 A/cm^2 is corrected to be 38.6 A cm^{-2}.
   (3) On page 4, Results and discussions→ Results and Discussion
   (4) We removed unnecessary bar "|" in the figure captions from Figure 1 to 4.

20th Aug 21

Dear Dr Kim,

Your manuscript titled "Enhanced-coherence all-nitride superconducting qubit epitaxially grown on Si substrate" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to comply with our journal policies and formatting style in order to maximise the accessibility and therefore the impact of your work.

EDITORIAL REQUESTS
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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,

Dr Aldo Isidori
Associate Editor
Communications Materials

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The Referee is satisfied by the revision of the manuscript. The paper can be accepted for publication.

Reviewer #2 (Remarks to the Author):

I believe the changes that the authors have made all contribute to the improvement of the paper, and that as the paper stands it is a very interesting and valuable report. I fully recommend the paper for publication and have no additional questions or suggestions.