nature portfolio

Peer Review File

Direct-bonded diamond membranes for heterogeneous quantum and electronic technologies

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This manuscript has been previously reviewed at another journal. This document only contains reviewer comments, rebuttal and decision letters for versions considered at Nature Communications.

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

Version 0:

Reviewer comments:

Reviewer #2

(Remarks to the Author)
The authors have made careful and detailed revisions and have adequately addressed my questions. I recommend the publication of the manuscript in Nature Comm.

Reviewer #3

(Remarks to the Author)
To what extent have the authors responded to my original points and what further changes are needed?

My original point "A weakness of the paper is that the potential of the fabrication process in a variety of contexts is explained, but no one application is pushed to a firm result."
The authors respond "We hope the Referee can support our preference to organize the manuscript in such a fashion." Yes, I can support this preference of the authors. My concern remains however, and is amplified a little by the authors' comment "In follow-up works, we will revisit these applications and conduct more in-depth studies" - a lot is pushed into the future.

My original point "A specific weakness in my opinion is the lack of information on charge noise."
Unfortunately, the authors are unable to make a clear statement on the charge noise in their diamond membranes. To my mind, this remains a weakness of the paper. The authors agree that probing the optical linewidth of NV centers is a good experiment to carry out in order to determine the charge noise but are presently unable to carry it out. I note that Referee 2 raises the issue of "optical and spin properties of NV centers" in the membranes - on optical properties, this is the same point. The authors respond by discussing mostly the spin properties. And the statement "we anticipate that ultra-shallow NV centers will have a similar environment as shallow NVs in bulk diamond and thus acquire comparable coherence values" is unconvincing to my mind given the complexity of the diamond surface and its potential to destroy optical and spin coherence.

My point 1 on adding the membrane thickness: all fine.

My point 2 on the 10nm-thick membrane thickness: this remains slightly ambiguous. The authors state that the 10-nm membrane was created from a 309-nm membrane. My question is this - is the membrane thickness 10 nm throughout the entire 200 micron x 200 micron area? The authors should change the wording so that there is no ambiguity.

My point 3 on spin coherence: the response in the rebuttal is fine (in fact very impressive spin coherence times) but the criticism of Referee 2 remains, namely that data is only presented for relatively thick membranes. The abstract claims in one sentence that bonded membranes as thin as 10 nm can be created, and in the next sentence that 623 micro-s NV spin coherence times in bonded membranes are measured. It is NOT clear from this text that it is presently unknown if 10-nm thick membranes support NVs with such long coherence times. This ambiguity must be removed.

Once these ambiguities have been resolved, the remaining weakness in my opinion is the lack of information on charge noise. I have two recommendations.

A the authors state on the subject of GeV linewidths: "In fact, as pointed out in section 3.2 of SI, the discrepancy between the single scan and multiple scan average is primarily due to the resolution and stability of our wavelength meter stabilizing the scanning laser, which was used for resonant excitation measurements." I would point out that the deficiencies of the authors' wave meter are not of interest to the community at large and hence that it would make sense to remove the statements and accompanying data on the linewidths measured with multiple scans - this data tells us nothing about the GeV centers.

B the authors should make a clear statement in the main text on charge noise, for instance "The level of charge noise in the fabricated membranes is presently unknown."

The above focusses on the remaining (and unacceptable) ambiguities and on a negative point, the lack of information on charge noise. I re-state the strengths of the paper as I see them, quoting from my original report: "This paper describes a full process flow to create diamond heterostructures by bonding thin diamond membranes to a variety of materials. The paper reports the details of the process and
a comprehensive study of the materials aspects - these are the paper's strengths." and "The results are impressive: I found the results in Figure 2 striking - ultra-smooth interfaces, constant-thickness membranes, good bonding at the diamond-substrate interface." Once the ambiguities have been removed and the issue of charge noise addressed, the paper is suitable for Nature Communications.

We thank the editor for accepting our manuscript and the reviewers for their time and effort. We are grateful for reviewer #2's satisfaction with our revised manuscript and reviewer #3's positive appraisal of the significance, novelty, and importance of our work. Additional comments provided by reviewer #3 were systematically considered, and revisions were made accordingly, as detailed below. These comments have improved the quality and clarity of our manuscript.

Reviewer #2:

1. *The authors have made careful and detailed revisions and have adequately addressed my questions. I recommend the publication of the manuscript in Nature Comm.*

We are grateful for the reviewer's satisfaction of our revised manuscript and support in the publication of it.

Reviewer #3:

2. *My original point "A weakness of the paper is that the potential of the fabrication process in a variety of contexts is explained, but no one application is pushed to a firm result." The authors respond "We hope the Referee can support our preference to organize the manuscript in such a fashion." Yes, I can support this preference of the authors. My concern remains however, and is amplified a little by the authors' comment "In follow-up works, we will revisit these applications and conduct more in-depth studies" - a lot is pushed into the future.*

We thank the reviewer for supporting our decision to demonstrate the direct-bonding diamond membrane platform's great potential and a wide variety of applications. This study is primarily focused on material characterization and nanofabrication innovation while shedding light on a few promising applications that require additional in-depth characterizations. More detailed studies demonstrating the potential of each application are underway. For instance, we have recently published a follow-up work of direct-bonded diamond membranes for nanophotonics and quantum communication [S. Ding at al., High-Q cavity interface for color centers in thin film diamond. *Nature Communications*, **15**, 6358 (2024)]. Additionally, we are collaborating with two other groups to study the potential of direct-bonding diamond membranes in magnetic material quantum sensing and heat dissipation of electronic devices. They have shown promising results, and we hope to share them soon.

3. *My original point "A specific weakness in my opinion is the lack of information on charge noise." Unfortunately, the authors are unable to make a clear statement on the charge noise in their diamond membranes. To my mind, this remains a weakness of the paper. The authors agree that probing the optical linewidth of NV centers is a good experiment to carry out in order to determine the charge noise but are presently unable to carry it out. I note that Referee 2 raises the issue of "optical and spin properties of NV centers" in the membranes - on optical properties, this is the same point. The authors respond by discussing mostly the spin properties. And the statement "we anticipate that ultra-shallow NV centers will have a similar environment as shallow NVs in bulk diamond and thus acquire comparable coherence values" is unconvincing*

to my mind given the complexity of the diamond surface and its potential to destroy optical and spin coherence.

We thank the reviewer for highlighting this point. As we mentioned in the previous response, we agree that both NV optical and spin properties are important factors in determining the charge and magnetic noise of the color center. However, due to the equipment limitation, we were only able to conduct the spin coherence measurement at this moment. The optical coherence will be performed in follow-up experiments once adequate equipment is available.

Regarding the coherence estimation of shallow NV centers in the diamond membrane, we are considering noise from (1) the carrier substrate, (2) the direct bonding process, (3) the crystal quality of the diamond membrane, and (4) the membrane surface. By measuring the NV spin coherence inside the diamond membrane (Figure 2g), we show that the spin coherence is the same as NV centers in bulk diamond with a similar depth. Therefore, we showed that the carrier substrate, the bonding process, and the diamond membrane crystal quality will not affect the spin coherence of near-surface NV centers. In addition, the diamond membrane surface has atomically smooth surfaces (Figure 2a), an ideal surface morphology for realizing low noise levels (See Ref. 5 from Supplementary Information). With proper oxygen termination which is compatible with the membrane fabrication process, the surface noise of the diamond membrane should be the same as that of a properly treated bulk diamond, and near-surface NV centers should have a similar performance.

4. *My point 1 on adding the membrane thickness: all fine.*

We thank the reviewer for acknowledging our clarification on the membrane thickness.

5. *My point 2 on the 10nm-thick membrane thickness: this remains slightly ambiguous. The authors state that the 10-nm membrane was created from a 309-nm membrane. My question is this - is the membrane thickness 10 nm throughout the entire 200 micron x 200 micron area? The authors should change the wording so that there is no ambiguity.*

We confirm that the membrane thickness is 10 nm throughout the entire area, as no etching mask was applied during the etching. The thickness uniformity is confirmed by high-resolution TEM measurements at different places (almost identical images as Figure 2(d), not shown in the manuscript due to repeated information). To further clarify this, we have added "uniformly" in the main text:

Main text, Material Chacterization:

Fig.2 (d)-(e) shows a uniformly ICP-thinned (from $\approx 309\,\mathrm{nm}$ to $10 \pm 0.3\ \mathrm{nm}$ while the lateral dimension remains to be $200\,\mu\mathrm{m} \times 200\,\mu\mathrm{m}$) diamond membrane bonded to a sapphire substrate.

6. *My point 3 on spin coherence: the response in the rebuttal is fine (in fact very impressive spin coherence times) but the criticism of Referee 2 remains, namely that data is only presented for relatively thick membranes. The abstract claims in one sentence that bonded membranes as*

thin as 10 nm can be created, and in the next sentence that 623 micro-s NV spin coherence times in bonded membranes are measured. It is NOT clear from this text that it is presently unknown if 10-nm thick membranes support NVs with such long coherence times. This ambiguity must be removed.

We thank the reviewer for pointing out this ambiguity. We have added the thickness information in the abstract to elimination the confusion.

Main text, Abstract

We measure spin coherence times T_2 for nitrogen vacancy centers in 150 nm-thick bonded membranes of up to $623(21)$ µs, suitable for advanced quantum applications.

7. *Once these ambiguities have been resolved, the remaining weakness in my opinion is the lack of information on charge noise. I have two recommendations.*

A the authors state on the subject of GeV linewidths: "In fact, as pointed out in section 3.2 of SI, the discrepancy between the single scan and multiple scan average is primarily due to the resolution and stability of our wavelength meter stabilizing the scanning laser, which was used for resonant excitation measurements." I would point out that the deficiencies of the authors' wave meter are not of interest to the community at large and hence that it would make sense to remove the statements and accompanying data on the linewidths measured with multiple scans - this data tells us nothing about the GeV centers.

B the authors should make a clear statement in the main text on charge noise, for instance "The level of charge noise in the fabricated membranes is presently unknown."

The above focusses on the remaining (and unacceptable) ambiguities and on a negative point, the lack of information on charge noise.

We thank the reviewer for pointing out that data produced by defective equipment has little scientific value. We agree with recommendation A provided by the reviewer and remove the average scan date from Figure S9. We have also revised the text accordingly. However, for statement B, we disagree with the reviewer that the charge noise of the direct-bonding diamond membrane is unknown. As we mentioned in the supplementary material section 3.2, the same batch of diamond membrane containing SnV (similar performance to GeV) has been measured from a different lab, which has shown transform-limited linewidths, indicating very low charge noise. We have also demonstrated that plasma treatment and direct bonding do not affect GeV linewidth compared to our previous result [Ref. 19 in the main text], indicating that the direct bonding method introduces limited charge noise.

Supplementary Information, 3.2 Effect of plasma treatments on the optical coherence of GeV– centers:

...Figure S9 (c) shows the single ~~(2.5 min average)~~ scan ZPL linewidths with resonant excitation. We observed no statistical difference of the linewidth distribution, with mean single scan linewidth of $97\,\mathrm{MHz}$ ($85\,\mathrm{MHz}$) ~~and mean average scan linewidth of 212 MHz (196 MHz)~~ for membrane 1 (2). ~~The measured linewidths are broader than the real value due to the resolution limit of the wavelength meter (High Finesse WS6-600, 20 MHz measurement resolution, 500 MHz wavelength accuracy).~~ A separate characterization of tin vacancy centers in our membranes ~~using a higher resolution wavelength meter~~ using higher resolution equipment reported a much narrower linewidth profile, with the average linewidth only $\approx 50\%$ higher than the transform-limited value [Guo2023], indicating a low charge noise environment.

Supplementary Information, Figure S9 caption:

...(c) ZPL wavelength and single ~~/2.5 min average~~ scan linewidth distribution of GeV^- centers. Centers from native (plasma treated) membranes are labelled in blue (orange).

8. *I re-state the strengths of the paper as I see them, quoting from my original report: "This paper describes a full process flow to create diamond heterostructures by bonding thin diamond membranes to a variety of materials. The paper reports the details of the process and a comprehensive study of the materials aspects - these are the paper's strengths." and "The results are impressive: I found the results in Figure 2 striking - ultra-smooth interfaces, constant-thickness membranes, good bonding at the diamond-substrate interface." Once the ambiguities have been removed and the issue of charge noise addressed, the paper is suitable for Nature Communications.*

We thank the reviewer for supporting the publication of the manuscript. We hope the revised manuscript has removed the ambiguity and addressed the charge noise issue.