nature portfolio

Peer Review File

Monolithic 3D integration of tantalum pentoxide nonlinear photonics

Corresponding Author: Dr Grant Brodnik

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

Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)
In the manuscript "Monolithic 3D integration of tantalum pentoxide photonics on arbitrary substrates", G. M. Brodnik and co-authors present a multi-layer photonic integration platform based on a tantalum pentoxide film grown on top of a thin-film lithium niobate. The authors present the main technological step that are part of the platform and a large palette of results obtained with both passive devices and nonlinear ones, taking advantage of both χ(2) and χ(3) phenomena in both tantalum pentoxide and lithium niobate films.

The topic is of large interest in the community, which is very actively exploring different possible platforms to compensate the drawbacks of classical ones (especially silicon platforms), with the main goal of achieving full on chip integration of passive, active, electro-optic and nonlinear photonic devices. I believe the paper presents innovative results that could represent a significant step forward in the ongoing reflection within the community.

The manuscript is clear and very well organize, even though some additional information could be provided, as detailed below.

My main concern is about the general claim of the paper. The authors aims at demonstrating the integration of "tantalum pentoxide photonics on arbitrary substrates" but I believe the paper falls short at achieving this goal since it exclusively discusses the integration on top of lithium niobate. It is true that the authors mention that "Following CMP, the substrate resembles a standard, oxide-clad wafer from the perspective of tantala processing, allowing for fabrication to proceed with no modifications from previously developed process flows" (line 120) bur I do not find this justification convincing.

There are many constraints that could make the integration of a tantala layer challenging when different substrates are used, for example stresses generated by the bottom layers, different thermal expansion coefficients or diffusion problems to handle. I believe that showing a deposition at low temperature on top of a thin silica layer is a very promising result but does not demonstrate universal compatibility on arbitrary substrates. Either this universal compatibility is demonstrated or a solid argument should be done on why demonstrating deposition on lithium niobate is sufficient to demonstrate the deposition is possible on any possible substrate.

On my opinion the paper presents sufficiently novel and high quality results to re-focus its claim as the demonstration of an hybrid lithium niobate-tantala platform.

Some additional minor suggestions:

- Waveguide losses are only indirectly demonstrated via the Q factor of resonators. It would be more informative for the reader to have a value in dB/cm for both lithium niobate and tantala waveguides, possibly as a function of waveguide width.
- Edge couplers are not discussed. Are waveguide adapted/designed to optimize fiber-waveguide losses (eg., inverse tapers)?
- Figure1c, why tantala waveguides are shown with different thicknesses? Is this achieved on the same wafer with multiple

depositions? Is etching used? This is not discussed in the paper.
- I find Figure 1d unclear. I suppose the green and pink arrows indicate the transparency windows but there is also a vertical dotted green line whose function is unclear.
- Line 116, “After removing poling electrode metals”. How is this done?
- Line 180, the authors should discuss why they use a multimode fiber at the output facet.
- Polarization diversity is not discussed at all in the paper. I think it is important to at least briefly discuss if and how TM modes are guided in the presented platform.

Referee #2

(Remarks to the Author)
Monolithic 3D integration of tantalum pentoxide photonics on arbitrary substrates by Brodnick et al.

A.Summary of the key results

In this manuscript, the authors report the monolithic integration on the wafer scale level of a relatively recent material, Ta_2O_5 (tantalate), onto a lithium niobate photonic platform. Using this approach, they demonstrate a wide range of high quality tantalate linear devices (high Q resonators…), $LiNbO_3$ c(2) based devices (PPLN for SHG), nonlinear tantalate devices (OPO, FWM, microcombs), and combined functions by leveraging the c(2) and c(3) properties of the respective materials, thanks to appropriate and low loss (<0.5dB) vertical transition between the upper tantalate and bottom $LiNbO_3$ material layers.

B.Originality and significance

The novelty lies in the 3D monolithic integration of two photonic material platforms with complementary nonlinear properties for increased photonic chip functionality.
There have been separate demonstrations of the devices reported in this manuscript: PPLN LN devices for efficient SHG have been demonstrated already, and efficient nonlinear devices on Ta_2O_5 have been reported as well by the same group (frequency combs in Optica 2021 [6], high Q rings reaching a few millions, supercontinuum, OPO with so called photonic crystal rings in Optica 2022 [34]). The originality of the present work therefore lies in their integration using direct growth of Ta_2O_5 onto $LiNbO_3$ chips.

Heterogeneous integration of materials is critical to leverage the best of each material platform and increase the overall photonic functionality, since no material can have it all. The co-integration of other complementary materials such as SiN and $LiNbO_3$, for instance, have been demonstrated already and this is acknowledged in the introduction. First of all, the linear and nonlinear properties of Ta_2O_5 might be more attractive than those of SiN, but this should be clearly said from the beginning to motivate this work. Ta_2O_5 should be better benchmarked against traditional nonlinear photonic materials (Si, SiN, $LiNbO_3$, III-V…) to appeal to a broader audience, considering that this material is relatively recent for these applications.

Beside the potentially attractive properties of Ta_2O_5, the true significance of this work lies in the technological integration approach of the hybrid chips reported here. Indeed, SiN to $LiNbO_3$ integration has typically involved some micro-transfer printing technology rather than monolithic integration where one material can be directly grown on another material platform, which is the case for Ta_2O_5, as reported here. This is enabled by the relatively simple deposition process involved, which only require moderate temperature processes (deposition at room temperature and annealing at 500°C rather than 1000°C for SiN).

Overall, although I feel that there is a lot of potential related to this new integration approach, which is innovative and potentially involves a promising material (which should still be better motivated…), the rest of the manuscript makes me feel that there is not much novelty achieved with this hybrid platform since most of the manuscript reports devices already shown in the literature or reported by the same group on the stand-alone tantalate (or $LiNbO_3$) platform. At places, it seems that the main point is to check if the devices still work after the 3D integration. Besides, this comparison with the prior stand-alone device counterparts is poorly done, due to the absence of any true benchmark comparison or any quantitative assessment of the achieved device performance (see my comments in section E below). Finally, there is no new functionality or design leveraged to demonstrate the added value of this hybrid material platform apart from cascaded nonlinear devices that are interesting and significant but only qualitatively and rapidly shown for illustration towards the end of the manuscript.

C.Data & methodology: validity of approach, quality of data, quality of presentation

Some of the figures could be improved. I am not such a big fan of illustration figures like fig 1e which show many devices at once but they are so small that it is hard to fully understand the various range of devices intended or demonstrated.

Some details about the role of the silica layer that is left between $LiNbO_3$ and Ta_2O_5 after CMP should be clarified: how is it controlled? Is it uniform across the wafer? Are its thickness and uniformity critical? What is the tolerance for the hybrid devices to work, or the taper performance ensuring vertical coupling between the $LiNbO_3$ and Ta_2O_5. Considering the technological approach is the main point of the manuscript, additional information should be given.

D.Appropriate use of statistics and treatment of uncertainties

One added value of the 3 inch wafer scale process reported here lies in the possibility to manufacture many chips at once, with respect to recently developed micro-transfer printing for instance, where individual devices are typically transferred one by one. However, to showcase this advantage, one might expect some statistics on the device performance metrics or quantitative information about the measured deviations in device characteristics measured across multiple chips manufactured on the same wafer. Yet, there is no such reproducibility study anywhere in this manuscript (apart from having two different series of SHG results that have been measured possibly on two different chips across the same wafer, a point that should be also further clarified)

E.Conclusions: robustness, validity, reliability

Despite the use of qualitative terms like “ultra-efficient nonlinear optics” at the beginning of section 4, no comparison is drawn between the devices presented here and any nonlinear devices from the literature. I strongly advise the authors to add a comparison with prior nonlinear stand-alone Ta2O5 devices from their group, as well as with alternative material platforms for the demonstrated nonlinear effects. Some benchmark comparison is currently critically missing to help readers appreciate how the resulting devices compete with prior nonlinear demonstrators, including those already combining c(2) and c(3) materials (e.g. SiN/ LiNbO3 hybrid chips achieved with microtransfer printing).

Not only is there no direct comparison with the literature, but some essential performance metrics is missing for the reported nonlinear devices, like the conversion efficiency of several nonlinear c(3) based processes in tantalate, which is more novel that those related to SHG in LiNbO3 for instance.

The most quantitative performance description in this manuscript focuses on the demonstration of SHG in PPLN waveguides, which is discussed at length on page 8, although this represents the less novel aspect of the manuscript in my opinion, since there have been many related demonstrations. It seems to me that the main point of this section is to show that the LiNbO3 QPM SHG performance is not too much degraded by the integration of tantalate, although this material is not directly exploited for the SHG process. This is a good point to check but does not represent the main breakthrough of this manuscript.

F.Suggested improvements: experiments, data for possible revision

1. As raised above, several data and metrics are critically missing to appreciate the performance of the material and devices reported here, such as:

1a-the characteristics of the tantalate material (c(3) nonlinear properties, n2 of tantalate, gamma of the related waveguide, propagation loss…) which is less known than LiNbO3, should be explicitly stated and compared against more standard nonlinear materials.

1b-The intrinsic Q of tantalate ring resonators is given, but not the loaded Q which is the key metrics for microcomb generation. No indication is given regarding the coupling regime, and no comparison with the characteristics of Ta2O5 rings achieved before. Is it the same or worse?

1c-Similarly, the performance of these microcombs are not compared against prior microcombs on Ta2O5 or state of the art microcombs (conversion efficiency, frequency comb power threshold…)

1d-The so-called photonic crystal resonators in the abstract/ introduction are in fact corrugated rings. I would advise the authors to call them so to avoid any confusion, since some readers might expect actual photonic crystal resonators (e.g. holes etched in a high dielectric membrane) exhibiting some complete photonic band gaps thanks to a large index contrast. Most importantly, the Q factor for those structures are not given either, despite the “high Q” that is alluded to in the abstract and towards end of introduction. Eventually, the description of these devices is very concise and qualitative (a few lines only between page 8-9), so that we don’t know what to really grasp from it. There should be at least some comparison with the Q factors already obtained for similar stand-alone Ta2O5 devices demonstrated by the same group in the reference papers published between 2022-2025.

1e-Some information could be given regarding the short pass dichroic used for the SHG measurement on page 8: what are its spectral characteristics? By how much is the pump attenuated for the SH measurement, with respect to the SH signal? This directly impacts the SHG conversion efficiency.

1f-What is the coupling efficiency at input/ output of the chip, considering different fibers are used (single mode vs. multimode) at both ends. Also, is there any distinction for the pump and SH, as this would also impact the reported SHG efficiency.

1g-Why is the performance of the QPM SH efficiency worse with tantalate than without by a factor of 3? Showing a table of the three tested devices along with some literature landmark demonstration would have been clearer towards the end of p8. Why is there a difference between 787nm and 485nm tantalate- LiNbO3 device?

1h-What does limit the 0.5 dB loss related to the transition from the tantalate to the LiNbO3 layers? Is there some prospect to improve it?

1i-What is the upper limit of Ta2O5 thickness to be grown on the SiO2/LiNbO3 platform? Is it a limitation?

2. Section 2 starts by showing a resulting Ta_2O_5 waveguide integrated onto $LiNbO_3$ waveguide, but no details about how such an alignment is performed, or quantitative information about the achieved alignment precision, tolerance or length are given.

3. What is the purpose of the 500°C annealing step of Ta_2O_5 and why is it necessary? It is said that this is avoided for doped tantalate, but there is not much explanation for this…

4. What is the origin of the wavelength dependence of the vertical taper/ transition between Ta_2O_5 and $LiNbO_3$, as 0.2dB is measured at 1550nm and 0.5dB at 780nm. It would be useful to have error bars on these values, considering for instance multiple measurements across the various chips?

5. Regarding the design of the nonlinear c(3) devices (OPO, frequency combs…), is there any particular feature or intrinsic challenge due to the 3D integration of tantalate onto $LiNbO_3$? For all the reported tantalate devices, references to prior work from the group on stand-alone tantalate devices are cited, which seems to imply that everything is the same. Is it really true?

6. The last paragraph of section 4 is by far the most interesting because these demonstrations report the use of both $LiNbO_3$ and tantalate for cascaded device operation leveraging the c(2) and c(3) of the respective material. However, it remains very qualitative, showcasing the potential of the platform via illustrations: there are very few details about the resulting performance (pump power threshold, conversion efficiency etc…) so that it sometimes feels more like a bundle, rather than a scientific paper. I advise the authors to expand on these last results, so that we appreciate the novelty in a more quantitative way.

7. Several reference to “tantalate-LN devices” or “3D tantalate LN devices” and “interlayer functionality” are made in section 4, although these devices are always used separately and at best sequentially. This should be clarified as I found it misleading at first. The sequential use of the two platforms to demonstrate various nonlinear functions slightly decreases the appeal of the integration approach. Is there some prospect to truly leverage the full potential of this approach to realize fully hybrid devices?

G.References: appropriate credit to previous work?

The benchmark to the literature should be made more thoroughly (see my comments above).

Regarding the routing and tapers to connect the two material layers, no reference is given. I think that this has been used for SiN to Si interface or for hybrid III-V onto SiN chips already. What is really new here?

H.Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions

I advise the authors to temper down the claim on “arbitrary substrates” or “nearly universal compatibility in integration” in the title, abstract, introduction and conclusion (first sentence “we have presented monolithic 3D integration that combines tantalate photonics with arbitrary substrates”): only integration onto the $LiNbO_3$ platforms is demonstrated here, which, I acknowledge, is an important one. Even though the process flow involves steps that could be used for a wide range of material platforms, the present demonstration focuses onto $LiNbO_3$ exclusively, so it does not justify in my opinion the title/ abstract in its current form.

To reemphasize the previous point, no integration with active devices like lasers and modulators, nor active devices involving electrical control of the device operation, is shown. Instead, the group has recently demonstrated the heterogeneous integration of III-V lasers and Ta_2O_5 rings for OPO applications leveraging more traditional bonding technologies (ref [20] of the manuscript). Would there be any hurdle to integrate III-V lasers with Ta_2O_5 using the present approach?

Similarly, the use of low temperature several times in the introduction/ abstract is misleading since 500°C annealing is still performed here. While this is much less than what is used for SiN indeed, I would not qualify it as low temperature. For clarity, I would replace by moderate and state the number to avoid any confusion.

The claim that Ta_2O_5 enables “efficient mode matching between waveguides” might be true for $LiNbO_3$ but why would it be universally true for any other material platform? Efficient 3D routing is likely to work best for $LiNbO_3$. Could the authors comment on this, especially in the context of presenting this demonstration as a universal approach?

Version 1:

Reviewer comments:

Referee #1

(Remarks to the Author)
I have read the authors' rebuttal letter with interest and I think amendments significantly improve completeness and focus of the paper. Regarding the paper title, I do not find the phrase “functional substrate” immediately comprehensible. I would lean more toward something like “patterned substrate” but this is a personal feeling. I do not have further comments.

Referee #2

(Remarks to the Author)
I acknowledge the improvement brought by the authors to their manuscript to better highlight the main novelty/ claim of their findings, which are indeed centered on the demonstration of monolithic integration of tantalum pentoxide with LNOI for increased linear and nonlinear photonic functionalities.

The authors have made some efforts to better stress the advantages/ properties of this relatively recent material platform, and the results achieved are indeed convincing enough regarding the future prospects to integrate this material with other photonic platforms thanks to the less constraining processing method with respect to alternative approaches used in the literature.

I also appreciated the change in the title which I find more appropriate, as well as the clarification that is added early enough in the abstract/ introduction regarding the specific use of LNOI, as was chosen to illustrate the potential of this monolithic integration.

Finally, the addition of experimental nonlinear results (SCG + SHG) for f-2f self-referencing is relevant and appealing to illustrate the advantages of leveraging combined c(2) and c(3) nonlinear functions via exploiting this new hybrid Ta2O5/LN material platform.

I just have a few minor points to clarify the technical/ scientific content of the manuscript before fully endorsing the manuscript for publication:

-Regarding the use of edge couplers (which was also questioned by reviewer 1), the authors mention “For the designs presented in this work, the output waveguides are inverse tapered (increasing width approaching the edge of the chip) to a width of 2.5 um.” To my knowledge, inverse tapers refer to decreasing waveguide width close to the chip end-facet, which is an alternative strategy to increase fiber-chip coupling. Instead, if I understand correctly, here the authors use flared waveguides. I would advise them to change the terminology to be more accurate and avoid confusion.

-The authors mention the use of TE0 mode in LNOI (X cut) to leverage the best of the nonlinear waveguide. Could they also give the orientation of the LN waveguide axis? I don’t recall that it was specified. Is it along the Y axis?

-The authors now indicate the material properties of the Ta2O5 (in the additional table in fig 2b). Yet, could they specify the wavelength at which they give the n2 value? In the context of benchmarking it against SiN, although the respective fabrication processes are now better compared, they do not directly cite the n2 of SiN, but only mention in their response letter that it is larger for Ta2O5. For completeness, could they include the n2 value of SiN at 1550nm as well in the manuscript to stress how much larger is that of Ta2O5?

-Besides, regarding the use of c(3) Ta2O5 devices, could the authors also indicate the nonlinear gamma parameter (/W/m) at the different pump wavelengths, for both narrow and wide waveguides. This key metric would help the nonlinear photonics community to appreciate even further the potential of Ta2O5 nonlinear photonics and compare this nonlinear material platform against more traditional ones (like SiN or Si…)

-Fig 2d: could the authors indicate at which wavelength are the Qi values reported on the different points across the wafer? Is it 1550nm?

-I still do not support the misleading use of the terminology “photonic crystal resonator” in place of corrugated rings, but I don’t think we will agree on this…

-For the various nonlinear devices demonstrated on fig 4c, I strongly recommend the authors to indicate the pump power used for the different measurements and specify both the input (before the chip) and coupled pump power. Similarly, the total Q of the resonators should be specified in link with the related measurements.

-For the relevant SCG + SHG measurement added to the manuscript, I advise the authors to specify whether the quoted power is the input or coupled pump power, and to indicate the peak power (in addition to the average power), which is more relevant to SCG. They should also quote the nonlinear parameter gamma of their Ta2O5 waveguides here (see above).

Referee #1 (Remarks to the Author):

In the manuscript "Monolithic 3D integration of tantalum pentoxide photonics on arbitrary substrates", G. M. Brodnik and co-authors present a multi-layer photonic integration platform based on a tantalum pentoxide film grown on top of a thin-film lithium niobate. The authors present the main technological step that are part of the platform and a large palette of results obtained with both passive devices and nonlinear ones, taking advantage of both χ(2) and χ(3) phenomena in both tantalum pentoxide and lithium niobate films.

The topic is of large interest in the community, which is very actively exploring different possible platforms to compensate the drawbacks of classical ones (especially silicon platforms), with the main goal of achieving full on chip integration of passive, active, electro-optic and nonlinear photonic devices. I believe the paper presents innovative results that could represent a significant step forward in the ongoing reflection within the community.

The manuscript is clear and very well organize, even though some additional information could be provided, as detailed below.

My main concern is about the general claim of the paper. The authors aims at demonstrating the integration of "tantalum pentoxide photonics on arbitrary substrates" but I believe the paper falls short at achieving this goal since it exclusively discusses the integration on top of lithium niobate. It is true that the authors mention that "Following CMP, the substrate resembles a standard, oxide-clad wafer from the perspective of tantala processing, allowing for fabrication to proceed with no modifications from previously developed process flows" (line 120) but I do not find this justification convincing.

There are many constraints that could make the integration of a tantala layer challenging when different substrates are used, for example stresses generated by the bottom layers, different thermal expansion coefficients or diffusion problems to handle. I believe that showing a deposition at low temperature on top of a thin silica layer is a very promising result but does not demonstrate universal compatibility on arbitrary substrates. Either this universal compatibility is demonstrated or a solid argument should be done on why demonstrating deposition on lithium niobate is sufficient to demonstrate the deposition is possible on any possible substrate.

On my opinion the paper presents sufficiently novel and high quality results to re-focus its claim as the demonstration of an hybrid lithium niobate-tantala platform.

- We appreciate the Reviewer's insight and considerations discussed above.
- We agree with the Reviewer that our broadest claim of compatibility across 'arbitrary' substrates could be better explored or adapted to form a stronger manuscript.
- We also acknowledge the Reviewer's consideration that a re-focus on the integration with lithium niobate, rather than arbitrary substrates, could lend a more powerful and accurate scope

of our work. ***We are therefore open to changing the title to reflect this scope and are in support of suggestions for appropriate changes from the Reviewer.***

- We believe our revisions result in a stronger overall manuscript. We highlight our key claim of tantala and LN photonics operating in concert as a monolithic chi3/chi2 photonics platform, while also relating key information about tantala integration to facilitate broader adoption by the community for future fabrication with other novel platforms.
  - **INTRODUCTION PARAGRAPH 3**
  - In the space of nonlinear photonics, efforts have leveraged techniques like micro-transfer printing {vanackere_heterogeneous_2023} and direct bonding {chang_heterogeneous_2017,ghosh_wafer-scale_2023,churaev_heterogeneously_2023} to incorporate thin-film lithium niobate (LN) onto SiN platforms. The customized techniques used in these approaches arise from SiN material properties and processing conditions required to produce low-loss, thick-film photonic layers. Specifically, high residual stress (800 MPa) and high temperature deposition (700 ^oC) and annealing (1200 ^oC) necessitate independent SiN wafer processing and additional steps in integration such as incorporating stress-relief structures {luke_overcoming_2013,pfeiffer_photonic_2016}.}
  - **INTRODUCTION PARAGRAPH 4**
  - Monolithic 3D integration is made possible by tantala's room temperature, ion-beam-sputtering (IBS) deposition, low residual stress (38 MPa {jung_tantala_2021}), and moderate temperature (500 $C) anneals or anneal-free variations using metal oxide mixtures {carollo_amorphous_2025}. These advantages of tantala over, e.g. SiN, afford our monolithic 3D integration approach broad compatibility with underlying photonic materials and structures. We demonstrate seamless integration by realizing a tantala-LN platform…

Some additional minor suggestions:

1. Waveguide losses are only indirectly demonstrated via the Q factor of resonators. It would be more informative for the reader to have a value in dB/cm for both lithium niobate and tantala waveguides, possibly as a function of waveguide width.
   - We agree and address this item with manuscript revisions presented in red below.
   - We now report the associated waveguide loss for the corresponding intrinsic Q measurements of tantala microresonators.
     - Fig 2c Qi measurements
     - LINES 149-160
   - {We measure Qi and calculate waveguide loss, α, in microresonators with a RW of 4 µm optimized for normal dispersion 200 GHz dark pulse generation and microresonators with narrow RW designed for near-zero dispersion broadband OPO. In the 4 µm devices (Fig. 2c, top), we measure Qi to be ~ 4.7×10^6 (α = 0.12 dB/cm), ~ 5.5×10^6 (α = 0.09 dB/cm), and ~ 4.5×10^6 (α = 0.09 dB/cm) for wavelengths of 1064 nm, 1310 nm, and

1550 nm, respectively. In narrow RW devices (Fig. 2c, bottom), we measure Qi of ~ 7 × 10^5 for RW = 890 nm (blue), ~ 9 × 10^5 for RW = 1040 nm (green), and ~ 1 × 10^6 for RW = 1140 (red) for wavelengths spanning 780 nm to 1064 nm. The Qi values for the narrowest waveguide device (RW = 890 nm) correspond to waveguide losses of α = 1.2 dB/cm, α = 0.9 dB/cm, and α = 0.9 dB/cm for wavelengths of 780 nm, 980 nm, and 1064 nm respectively.}

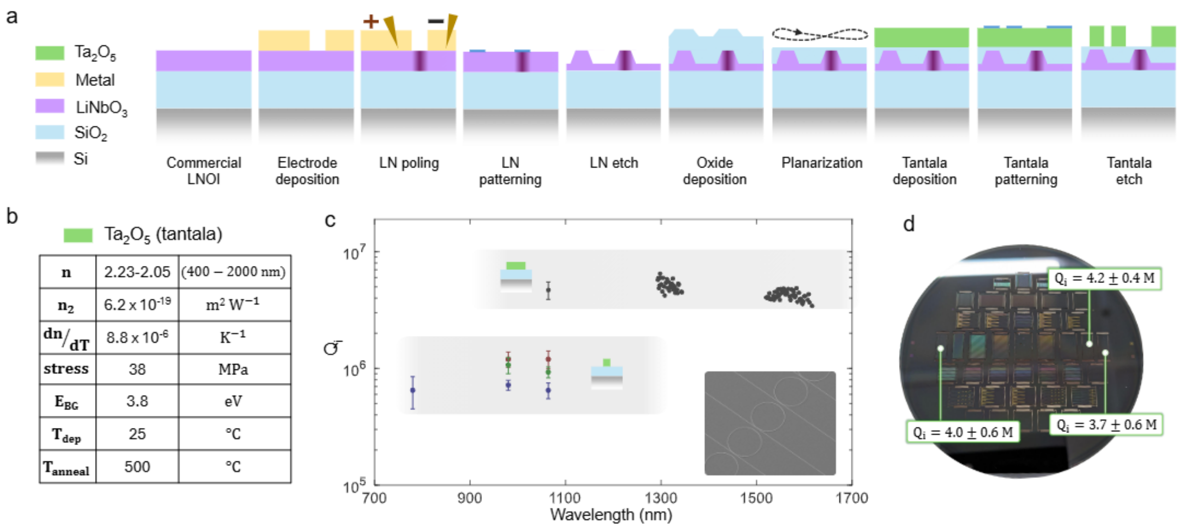
- We did not include loss test structures specifically for LN waveguides on the wafers for this work demonstrating nonlinear operation. We considered using SEM measurements of waveguide roughness to infer scattering loss, but didn't find this to be a suitable option. We thank the Reviewer for their suggestion and include this recommendation for future work.

2. Edge couplers are not discussed. Are waveguide adapted/designed to optimize fiber-waveguide losses (eg., inverse tapers)?
   - We address this item with manuscript revisions presented in red below.
   - For the designs presented in this work, the output waveguides are inverse tapered (increasing width approaching the edge of the chip) to a width of 2.5 um. This is done to improve mode matching between the air-clad waveguide and lensed fibers used in testing.
   - Beyond this, the designs have no specific optimization, for example, optimization to account for wavelength dependence or preference for SHG light coupling vs pump light
   - We revised the **Methods** section describing chip coupling to include the tantala waveguide facet coupling geometry and measurements
     - The tantala waveguides incorporate inverse tapers (increasing width) to a width of 2.5 um at the facets to improve mode matching with optical fiber.
   - We also now include a description of our insertion loss calibration in the **Methods** section **"SHG insertion loss measurements"**

3. Figure1c, why tantala waveguides are shown with different thicknesses? Is this achieved on the same wafer with multiple depositions? Is etching used? This is not discussed in the paper.
   - We agree this was confusing and address this item with manuscript revisions presented in red below.
   - Our goal was to present flexibility in waveguide thickness using control of deposited tantala thickness, but the figure was unclear.
   - In Figure 1, we've removed the varying thicknesses from the plot

4. I find Figure 1d unclear. I suppose the green and pink arrows indicate the transparency windows but there is also a vertical dotted green line whose function is unclear.
   - We agree and address this item with manuscript revisions presented in red below.
   - Figure 1d: Removed plot of index/transparency from the updated Figure 1.

- We now include a table listing critical material/fabrication parameters for tantala in Fig. 2b (notably, E_{BG} relates the material short wavelength transparency)



5. Line 116, "After removing poling electrode metals". How is this done?
   - We address this item with manuscript revisions presented in red below.
     - LINE 122
     - {After poling, electrode metals are removed through two selective wet chemical etches at room temperature. First, the gold layer is stripped in Transene TFA and rinsed in deionized (DI) water. The chromium layer is removed using CR-7 chromium etchant. The wafer is then rinsed again in DI water.}

6. Line 180, the authors should discuss why they use a multimode fiber at the output facet.
   - We agree and address this item with manuscript revisions referenced in red below.
   - We employ multimode fiber in our experimental setup for its broadband operation window / compatibility with testing device designs spanning a wide range of wavelengths
   - We now directly mention this in text in the Methods section, along with additional details on MMF with regards to coupling and insertion loss
     - Methods "SHG insertion loss measurements"
     - Calibrating chip coupling insertion loss is critical for characterizing SHG performance (Fig. 3f). For tantala-LN devices, light is coupled into upper layer tantala waveguides, routed down into LN for SHG, and routed back into tantala waveguides for output coupling. The tantala waveguides incorporate inverse tapers (increasing width) to a width of 2.5 μm at the facets to improve mode matching with optical fiber. Devices pumped at 980 nm and 1064 nm wavebands

employ HI1060 lensed fibre with a 2 µm spot size at the input. At the chip output, we edge couple a 50 µm core diameter multimode fibre, chosen for broadband compatibility. We independently measure insertion loss for each relevant wavelength (pump and all SHG wavelengths) for fibre-to-chip coupling. To do this, we characterize fibre coupling for symmetric tantala-LN interlayer-routed structures. Using test lasers operating at the experimental pump or SHG wavelength, we first couple light into and out-of the chip using matching lensed fibres designed for the test wavelength. We assume facet loss is symmetric and record per-facet insertion loss for lensed fibre coupling. We then replace the output fibre with the multimode fibre and measure the additional loss, thereby calibrating the per-wavelength, per-facet loss for either lensed fibre (pump light input) or multimode fibre (SHG light output) coupling configurations for the visible wavelength SHG measurements. We repeat these same calibrations for the LNOI device. For the 1550 nm tantala-LN device, we use lensed SMF-28 fibre on both the input and output, and measure SHG light on a visible detector insensitive to the pump (measured rejection >70 dB). The insertion losses for the tested devices are summarize in Table 1 below.

7. Polarization diversity is not discussed at all in the paper. I think it is important to at least briefly discuss if and how TM modes are guided in the presented platform.
    - We agree with the reviewer polarization/mode versatility is good to discuss. We address this item with the revisions presented in red below.
    - For the current work, we designed only TE0 devices. Our tantala platform supports operation using other polarization modes as well as higher order mode families.
    - We have addressed this comment by now mentioning other polarization and higher order mode operation for tantala. For the lower substrate, in this case X-cut TFLN, we make no claims about operating on other modes as we only consider TE0 operation in the current work to leverage the highest nonlinearity in X-cut TLFN.
        - LINE 149
        - The devices we characterize are designed for operation using the fundamental transverse electric mode (TE0), though designs that use other polarization or waveguide modes are possible {zhou_hybrid-mode-family_2022} are possible.

Referee #2 (Remarks to the Author):

Monolithic 3D integration of tantalum pentoxide photonics on arbitrary substrates by Brodnik et al.

A. Summary of the key results

In this manuscript, the authors report the monolithic integration on the wafer scale level of a relatively recent material, Ta2O5 (tantalate), onto a lithium niobate photonic platform. Using this approach, they demonstrate a wide range of high quality tantalate linear devices (high Q resonators…), LiNbO3 c(2) based devices (PPLN for SHG), nonlinear tantalate devices (OPO, FWM, microcombs), and combined functions by leveraging the c(2) and c(3) properties of the respective materials, thanks to appropriate and low loss (<0.5dB) vertical transition between the upper tantalate and bottom LiNbO3 material layers.

B. Originality and significance

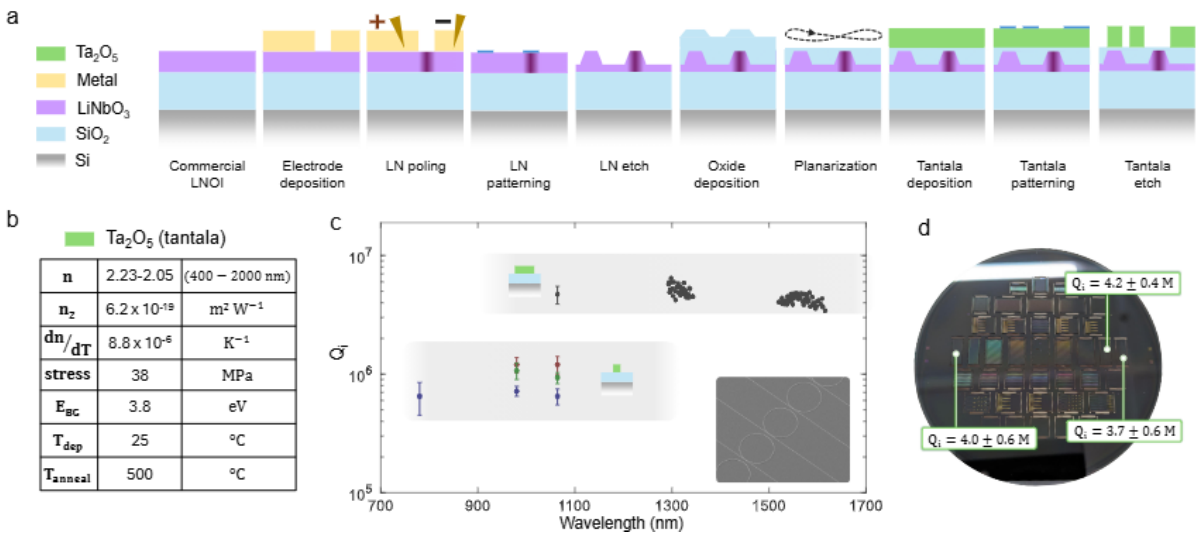
The novelty lies in the 3D monolithic integration of two photonic material platforms with complementary nonlinear properties for increased photonic chip functionality.
There have been separate demonstrations of the devices reported in this manuscript: PPLN LN devices for efficient SHG have been demonstrated already, and efficient nonlinear devices on Ta2O5 have been reported as well by the same group (frequency combs in Optica 2021 [6], high Q rings reaching a few millions, supercontinuum, OPO with so called photonic crystal rings in Optica 2022 [34]). The originality of the present work therefore lies in their integration using direct growth of Ta2O5 onto LiNbO3 chips.

- We agree with the reviewer, the originality of the paper is our monolithic integration approach enabled by the direct deposition of tantala.
- The reviewer relates a host of devices demonstrated in the literature on various platforms, as well as our own previous work in various aspects. We demonstrate broad functionality in terms of these device designs, but on monolithically integrated tantala on thin film lithium niobate for the first time – highlighting the key claim mentioned above.

Heterogeneous integration of materials is critical to leverage the best of each material platform and increase the overall photonic functionality, since no material can have it all. The co-integration of other complementary materials such as SiN and LiNbO3, for instance, have been demonstrated already and this is acknowledged in the introduction. First of all, the linear and nonlinear properties of Ta2O5 might be more attractive than those of SiN, but this should be clearly said from the beginning to motivate this work. Ta2O5 should be better benchmarked against traditional nonlinear photonic materials (Si, SiN, LiNbO3, III-V…) to appeal to a broader audience, considering that this material is relatively recent for these applications.

- We agree and address this item with manuscript revisions referenced in red below
- Tantala can be better benchmarked against comparable materials to lend stronger support for our claims and demonstrations of tantala as an efficient nonlinear photonics platform

- We now explicitly report relevant tantala material properties in Fig. 2b, along with references for metrics



- We have also revised the text about tantala and other materials, specifically SiN as the most direct comparison of nonlinear platform, directly comparing our deposited tantala platform in terms of nonlinear operation and fabrication compatibility.
  - **INTRODUCTION PARAGRAPH 3**
  - In the space of nonlinear photonics, efforts have leveraged techniques like micro-transfer printing {vanackere_heterogeneous_2023} and direct bonding {chang_heterogeneous_2017,ghosh_wafer-scale_2023,churaev_heterogeneously_2023} to incorporate thin-film lithium niobate (LN) onto SiN platforms. The customized techniques used in these approaches arise from SiN material properties and processing conditions required to produce low-loss, thick-film photonic layers. Specifically, high residual stress (800 MPa) and high temperature deposition (700 °C) and annealing (1200 °C) necessitate independent SiN wafer processing and additional steps in integration such as incorporating stress-relief structures {luke_overcoming_2013,pfeiffer_photonic_2016}.}
  - **INTRODUCTION PARAGRAPH 4**
  - Monolithic 3D integration is made possible by tantala's room temperature, ion-beam-sputtering (IBS) deposition, low residual stress (38 MPa {jung_tantala_2021}), and moderate temperature (500 °C) anneals or anneal-free variations using metal oxide mixtures {carollo_amorphous_2025}. These advantages of tantala over, e.g. SiN, afford our monolithic 3D integration approach broad compatibility with underlying photonic materials and structures. We demonstrate seamless integration by realizing a tantala-LN platform…

Beside the potentially attractive properties of Ta2O5, the true significance of this work lies in the technological integration approach of the hybrid chips reported here. Indeed, SiN to LiNbO3 integration has typically involved some micro-transfer printing technology rather than monolithic integration where one material can be directly grown on another material platform, which is the case for Ta2O5, as reported here. This is enabled by the relatively simple deposition process involved, which only require moderate temperature processes (deposition at room temperature and annealing at 500°C rather than 1000°C for SiN).

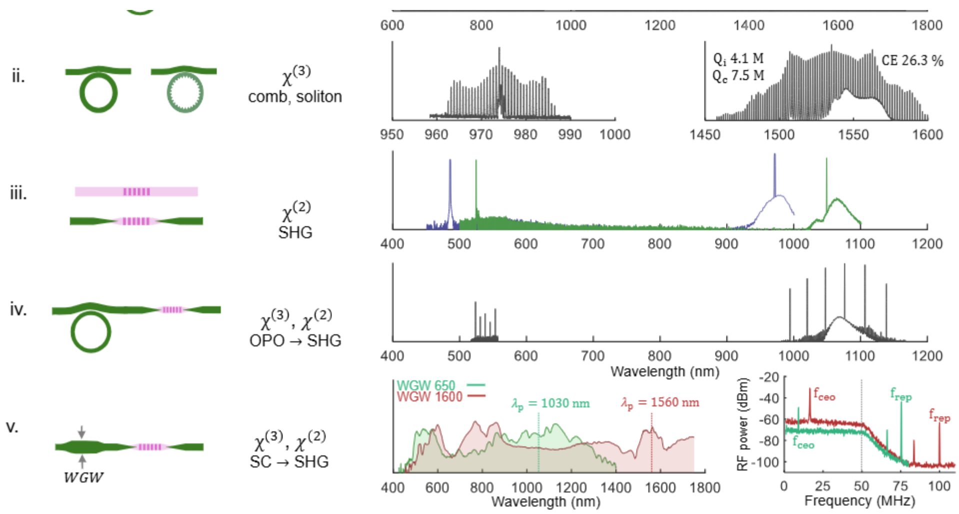
- We again agree with the identification of the true significance of the work being our technological integration approach, enabled by the direct deposition of tantala.
- The above items, along with the reviewer's previous related comments, strongly suggest that we could better compare our integration approach with others to more clearly highlight the impact of our work.
- We also now explicitly label the anneal temperature in Fig 2 and refer the anneal step as moderate temperature as requested by the reviewer, enabling direct comparison to comparable platforms.
- Our response to the item directly above addressing the Reviewer's similar comment provide additional discussion – We have revised the manuscript sections Introduction Paragraphs 3 and 4 (written out in the comment above.)

Overall, although I feel that there is a lot of potential related to this new integration approach, which is innovative and potentially involves a promising material (which should still be better motivated…), the rest of the manuscript makes me feel that there is not much novelty achieved with this hybrid platform since most of the manuscript reports devices already shown in the literature or reported by the same group on the stand-alone tantalate (or LiNbO3) platform. At places, it seems that the main point is to check if the devices still work after the 3D integration. Besides, this comparison with the prior stand-alone device counterparts is poorly done, due to the absence of any true benchmark comparison or any quantitative assessment of the achieved device performance (see my comments in section E below). Finally, there is no new functionality or design leveraged to demonstrate the added value of this hybrid material platform apart from cascaded nonlinear devices that are interesting and significant but only qualitatively and rapidly shown for illustration towards the end of the manuscript.

- A study on the metrics of individual devices is not the goal of this work – as the Reviewer previously describes, our key claim and novelty is the fabrication approach that affords the same palette of chi3/chi2 designs as other independent demonstrations, but in this case, for the first time, monolithically integrated directly at the wafer scale.
- We support the key claims by demonstrating an array of nonlinear devices – some in the tantala layer, some in lithium niobate, and some that leverage both – but we've made no effort to individually optimize designs at this stage. Certainly, these efforts are important for future work and broader adoption by the community.
- We report the critical platform metrics that *must be met* in order to support high-performance operation when optimizing designs in future efforts: the strong optical nonlinearity of tantala,

low-loss waveguides measured via intrinsic quality factor measurements, and low-loss interlayer routing that unlocks operation with underlying photonic substrates.

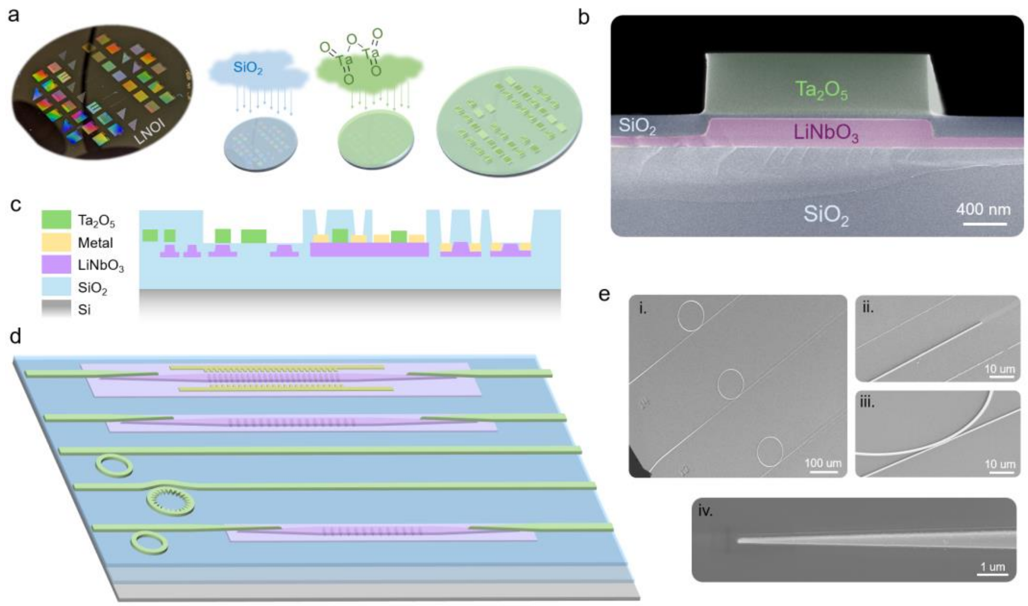
- We've also performed new experiments and measurements to provide quantified results via the conversion efficiency of the comb device, as well as a demonstration of a first-of-its kind supercontinuum (tantala) and SHG (LN) device for self referencing a comb in a fully monolithic design.
- FIG 4



C. Data & methodology: validity of approach, quality of data, quality of presentation

Some of the figures could be improved. I am not such a big fan of illustration figures like fig 1e which show many devices at once but they are so small that it is hard to fully understand the various range of devices intended or demonstrated.

- We've revised our Figures for clarity where noted, and/or to support specific considerations presented by the reviewers.
- In Figure 1, we removed the previous plot of index for tantala and LN
- This changes the scaling of the figure, to enlarge the schematic and address the reviewer's concern for the previously small device schematics of Fig 1d.



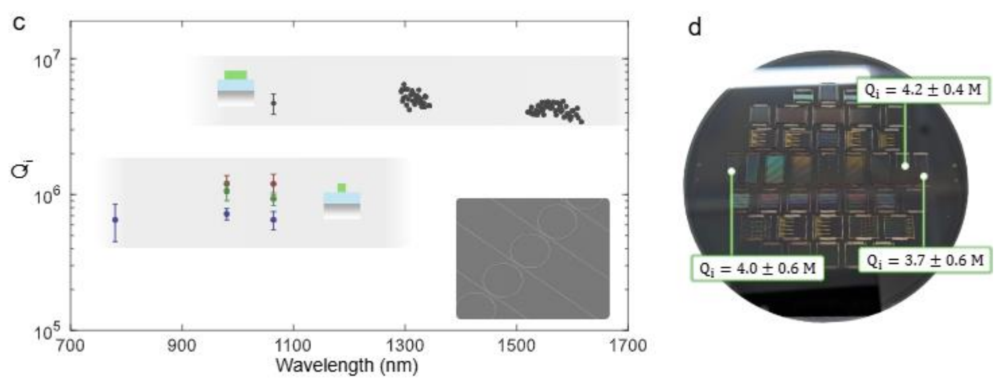
Some details about the role of the silica layer that is left between LiNbO3 and Ta2O5 after CMP should be clarified: how is it controlled? Is it uniform across the wafer? Are its thickness and uniformity critical? What is the tolerance for the hybrid devices to work, or the taper performance ensuring vertical coupling between the LiNbO3 and Ta2O5. Considering the technological approach is the main point of the manuscript, additional information should be given.

- We agree with the Reviewer on the importance of this discussion. We address these items with manuscript revisions using new numerical studies referenced in red below
  - LINES 172-181
  - We show a 3D schematic (exaggerated dimensions to show features) and a 2D slice labeling the silica interface thickness, dz, dependent on CMP planarization, overlaid with a sample finite-difference-time-domain (FDTD) simulation in Fig. 3a (left). Our E-beam lithography process is suited for high precision alignment of taper structures, with specified precision of ±20 nm. More critical in terms of processing is the CMP oxide planarization step, which controls dz between the waveguides and affects modal overlap of the taper structures. Further, modal overlap in general includes a wavelength dependence due to the stronger confinement of optical modes at shorter wavelengths {sacher_multilayer_2015}. In FDTD simulations with dz = 150 nm, we found the loss-per-transition to be <0.05 dB at 1550 nm and 0.3 dB at 780 nm. For dz = 200 nm, we simulate the loss-per-transition to be <0.05 dB at 1550 nm while falling to 1.5 dB at 780 nm.

D. Appropriate use of statistics and treatment of uncertainties

One added value of the 3 inch wafer scale process reported here lies in the possibility to manufacture many chips at once, with respect to recently developed micro-transfer printing for instance, where individual devices are typically transferred one by one. However, to showcase this advantage, one might expect some statistics on the device performance metrics or quantitative information about the measured deviations in device characteristics measured across multiple chips manufactured on the same wafer. Yet, there is no such reproducibility study anywhere in this manuscript (apart from having two different series of SHG results that have been measured possibly on two different chips across the same wafer, a point that should be also further clarified)

- We agree with the reviewer in highlighting the value of wafer scale processing, and address this item with manuscript revisions referenced in red below
- We performed additional measurements on chips from the same wafer, so that we can now include intrinsic Q data and statistics of tantala microresonators on chips sampled from opposite geographic locations on the 3 inch wafer to quantify yield/reliability in fabrication.
- The new data is presented in Fig. 2d



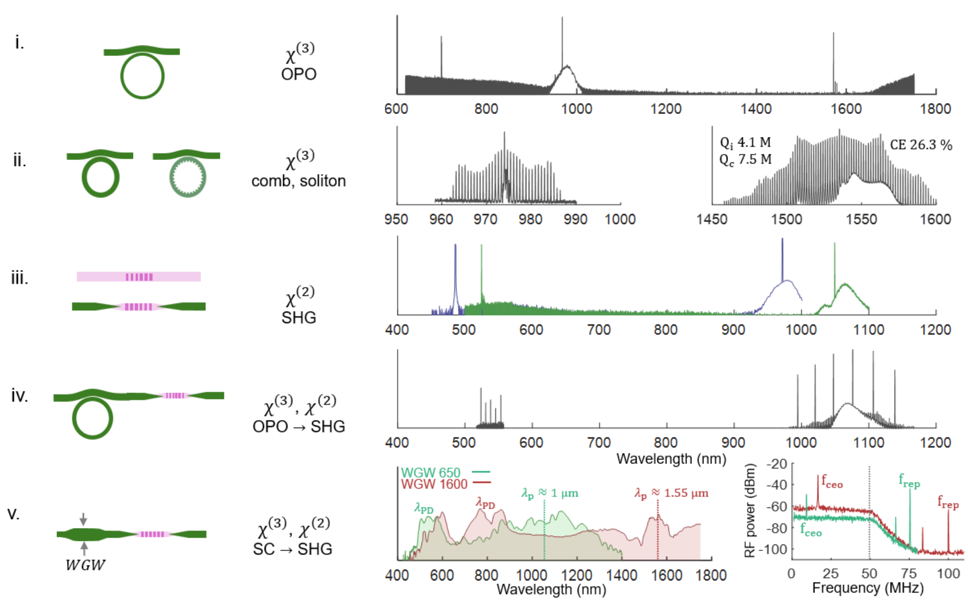
E. Conclusions: robustness, validity, reliability

Despite the use of qualitative terms like "ultra-efficient nonlinear optics" at the beginning of section 4, no comparison is drawn between the devices presented here and any nonlinear devices from the literature. I strongly advise the authors to add a comparison with prior nonlinear stand-alone Ta2O5 devices from their group, as well as with alternative material platforms for the demonstrated nonlinear effects. Some benchmark comparison is currently critically missing to help readers appreciate how the resulting devices compete with prior nonlinear demonstrators, including those already combining c(2) and c(3) materials (e.g. SiN/ LiNbO3 hybrid chips achieved with microtransfer printing).

Not only is there no direct comparison with the literature, but some essential performance metrics is missing for the reported nonlinear devices, like the conversion efficiency of several nonlinear c(3) based processes in tantalate, which is more novel that those related to SHG in LiNbO3 for instance.

The most quantitative performance description in this manuscript focuses on the demonstration of SHG in PPLN waveguides, which is discussed at length on page 8, although this represents the less novel aspect of the manuscript in my opinion, since there have been many related demonstrations. It seems to me that the main point of this section is to show that the LiNbO3 QPM SHG performance is not too much degraded by the integration of tantalate, although this material is not directly exploited for the SHG process. This is a good point to check but does not represent the main breakthrough of this manuscript.

- These items relate similar discussion as that given at the end of this Reviewer's Section B above
- As discussed there, our main claim, in agreement with the Reviewer's previous comments, is on the technological/integration approach rather than the specific optimization of any particular device (which is certainly important for future work). In context of 'ultra-efficient nonlinear performance,' we report the nonlinearity of tantala (higher than SiN) and our platform metrics (high intrinsic Q, low loss routing) that support our claim.
- We therefore respectfully disagree with the Reviewer's comments about specific device performance 'representing the main breakthrough of this manuscript', as the main breakthrough is the technological approach and integration, rather than device optimization.
- We are grateful for the Reviewer's considerations to strengthen our manuscript and find these suggestions provide motivation for interesting future work.
- We do find the Reviewer's comments present an opportunity to strengthen the main claim, while at the Reviewer's urging, we added entirely new device data and demonstration of a first of its kind device
    - We now include measurements of conversion efficiency for a microcomb device in Fig 4c (ii), enabling more direct comparison to similar devices.
    - We now include new device results – supercontinuum generation and SHG for fully integrated fceo detection, demonstrated in Fig 4c (v) at two different wavebands with plots of optical spectra, and RF spectra of fceo / frep



F. Suggested improvements: experiments, data for possible revision

1. As raised above, several data and metrics are critically missing to appreciate the performance of the material and devices reported here, such as:

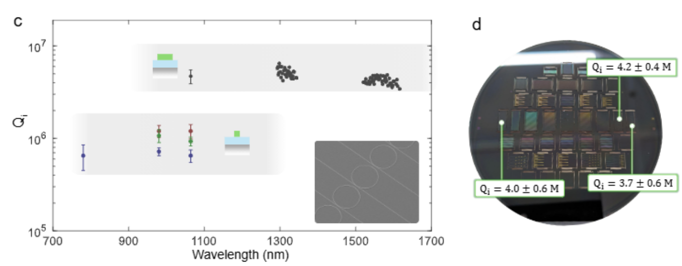
1a-the characteristics of the tantalate material (c(3) nonlinear properties, n2 of tantalate, gamma of the related waveguide, propagation loss…) which is less known than LiNbO3, should be explicitly stated and compared against more standard nonlinear materials.

- We agree with the reviewer in highlighting tantala characteristics and address this item with manuscript revisions referenced in red below
- In Fig. 2b we now report tantala material and fabrication parameters to enable direct comparison with other standard nonlinear materials e.g. SiN.
- We now report tantala waveguide loss in the text corresponding to the Q measurements in Fig. 2c.

b

Ta_2O_5 (tantala)

n	2.23-2.05	(400 – 2000 nm)
n_2	6.2 x 10^{-19}	$m^2 W^{-1}$
dn/dT	8.8 x 10^{-6}	K^{-1}
stress	38	MPa
E_{BG}	3.8	eV
T_{dep}	25	°C
T_{anneal}	500	°C



1b-The intrinsic Q of tantalate ring resonators is given, but not the loaded Q which is the key metrics for microcomb generation. No indication is given regarding the coupling regime, and no comparison with the characteristics of Ta2O5 rings achieved before. Is it the same or worse?

- We address the Reviewer's request for Loaded Q by providing the measurement on the tested microcomb device in Fig. 4
- We contend the intrinsic Q is the more relevant parameter when discussing the platform as a whole and its capabilities. It is determined by hard constraints of material properties and fabrication (e.g., material absorption, sidewall scattering due to etch quality). Loaded Q is indeed important for device optimization, but is a design choice controlled by the user e.g., ring-bus coupler gap for desired coupling rate (which, combined with Qi determines QL).
- We further include the following revised discussion to address the importance of Q
    - LINE 141
    - For microresonator-based FWM, high-Q is of key importance since threshold power scales inversely proportional to the square of loaded Q (Q_L) {yi_soliton_2015,lu_milliwatt-threshold_2019} and integrated devices aim for operation with practical on-chip pump powers. {Critically, to support high Q_L a photonics platform

must fundamentally support high Q_i since Q_L is tuned through control of coupling Q, (Q_C), via e.g. ring-bus coupling designs and through the relation $Q_L^{-1} = Q_c^{-1} + Q_i^{-1}$. We therefore characterize the (Qi) of the tantala-LN platform by performing microresonator spectroscopy of dispersion-engineered, wide and narrow ring width (RW) microresonators in Fig. 2c.}

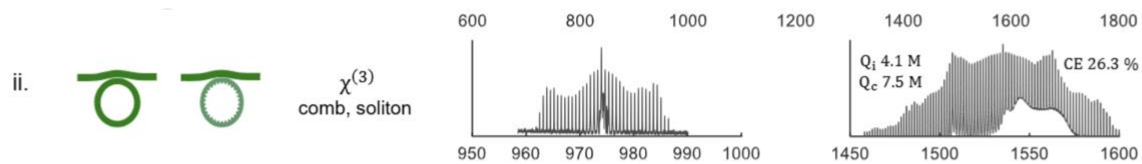
1c-Similarly, the performance of these microcombs are not compared against prior microcombs on Ta2O5 or state of the art microcombs (conversion efficiency, frequency comb power threshold…)

- At the Reviewer's request we now report the conversion efficiency of the comb device shown in Fig. 4c (ii) to provide a metric suited for comparison.
- Optimization efforts for device designs are outside the scope of our manuscript, but are important for future work. (please see item 'B' for similar discussion).
- We do maintain that the critical parameter that *must* be met for high efficiency operation on *any* platform is intrinsic Q. We already report this in Fig. 2, but now include additional Q data on the wafer map in Fig. 2 and also for the specific high efficiency device addressed here
- We also revise our discussion on why the reported metrics are of key importance.
    - LINE 141
    - For microresonator-based FWM, high-Q is of key importance since threshold power scales inversely proportional to the square of loaded Q (Q_L) {yi_soliton_2015,lu_milliwatt-threshold_2019} and integrated devices aim for operation with practical on-chip pump powers. {Critically, to support high Q_L a photonics platform must fundamentally support high Q_i since Q_L is tuned through control of coupling Q, (Q_C), via e.g. ring-bus coupling designs and through the relation $Q_L^{-1} = Q_c^{-1} + Q_i^{-1}$. We therefore characterize the (Q_i) of the tantala-LN platform by performing microresonator spectroscopy of dispersion-engineered, wide and narrow ring width (RW) microresonators in Fig. 2c.}

1d-The so-called photonic crystal resonators in the abstract/ introduction are in fact corrugated rings. I would advise the authors to call them so to avoid any confusion, since some readers might expect actual photonic crystal resonators (e.g. holes etched in a high dielectric membrane) exhibiting some complete photonic band gaps thanks to a large index contrast. Most importantly, the Q factor for those structures are not given either, despite the "high Q" that is alluded to in the abstract and towards end of introduction. Eventually, the description of these devices is very concise and qualitative (a few lines only between page 8-9), so that we don't know what to really grasp from it. There should be at least some comparison with the Q factors already obtained for similar stand-alone Ta2O5 devices demonstrated by the same group in the reference papers published between 2022-2025.

- We maintain the terminology of 'photonic crystal resonator' as that is the terminology used for similar designs in the referenced literature

- We agree with the reviewer on the importance of reporting Q for this device, and now include the intrinsic Q, coupling Q, and conversion efficiency for this device directly on the plotted data set for clarity in Fig 4c (ii).



1e-Some information could be given regarding the short pass dichroic used for the SHG measurement on page 8: what are its spectral characteristics? By how much is the pump attenuated for the SH measurement, with respect to the SH signal? This directly impacts the SHG conversion efficiency.

- We agree and now include these additional details.
- For our setup, we use 2 identical shortpass optical filters with a cut wavelength of 750 nm. Each is specified as having a rejection exceeding 50 dB. We verify suitable rejection of the pump laser by testing this– we sent only the pump light with a power of ~10 mW, and with the filters in place, measure below -55 dBm (meter limit) on the power meter.
- This information is in the Methods section relating the SHG experiments.
    - Methods
    - {We independently calibrate insertion loss for each relevant wavelength (pump and all SHG wavelengths) arising from fibre-to-chip coupling. To do this, we characterize fibre coupling for symmetric tantala-LN interlayer-routed structures. Using lasers operating at a given test wavelength, we couple light into and out-of the chip using lensed fibre designed for the given wavelength. We assume facet loss is symmetric and record per-facet insertion loss under these conditions. We then replace the output fibre with the multimode fibre, measure the additional loss, and thereby arrive at the per-wavelength, per-facet loss for either lensed fibre (pump light input) or multimode fiber (SHG light output) coupling. During high-power SHG measurements, we filter out the pump light using two shortpass optical filters, each cut at a wavelength of 750 nm and specified to have a rejection exceeding 50 dB. We confirm sufficient pump rejection by first measuring the pump laser directly, with and without the filters, observing a rejection exceeding 70 dB and limited by the noise floor of our power meter. These calibrations are used to report the on-chip pump and SHG powers in Fig. 3f as well as the associated uncertainties presented by the error bars.}

1f-What is the coupling efficiency at input/ output of the chip, considering different fibers are used (single mode vs. multimode) at both ends. Also, is there any distinction for the pump and SH, as this would also impact the reported SHG efficiency.

- We agree these are important and address this item with manuscript revisions presented in red below.
    - In the revised Methods section, we present a clearer description of our experimental approach and calibration of coupling loss, along with the measured values.
    - Calibrating chip coupling insertion loss is critical for characterizing SHG performance (Fig. 3f). For tantala-LN devices, light is coupled into upper layer tantala waveguides, routed down into LN for SHG, and routed back into tantala waveguides for output coupling. The tantala waveguides incorporate inverse tapers (increasing width) to a width of 2.5 um at the facets to improve mode matching with optical fiber. Devices pumped at 980 nm and 1064 nm wavebands employ HI1060 lensed fibre with a 2 um spot size at the input. At the chip output, we edge couple a 50 um core diameter multimode fibre, chosen for broadband compatibility. We independently measure insertion loss for each relevant wavelength (pump and all SHG wavelengths) for fibre-to-chip coupling. To do this, we characterize fibre coupling for symmetric tantala-LN interlayer-routed structures. Using test lasers operating at the experimental pump or SHG wavelength, we first couple light into and out-of the chip using matching lensed fibres designed for the test wavelength. We assume facet loss is symmetric and record per-facet insertion loss for lensed fibre coupling. We then replace the output fibre with the multimode fibre and measure the additional loss, thereby calibrating the per-wavelength, per-facet loss for either lensed fibre (pump light input) or multimode fibre (SHG light output) coupling configurations for the visible wavelength SHG measurements. We repeat these same calibrations for the LNOI device. For the 1550 nm tantala-LN device, we use lensed SMF-28 fibre with 2 um spot size on both the input and output, and measure SHG light on a visible detector insensitive to the pump wavelength (measured rejection >70 dB). The insertion losses for the tested devices are summarize in Table 1 below

Design	Pump IL (wavelength)	SHG IL (wavelength)
Tantala-LN	5.5±0.3 dB (970 nm)	7.5±0.3 dB (485 nm)
Tantala-LN	4.9±0.3 dB (1574 nm)	5.8±0.3 dB (787 nm)
LNOI	9.1±0.6 dB (1574 nm)	11.6±0.6 dB (1574 nm)

    - Table 1: Tantala-LN and LNOI SHG chip insertion loss measurements

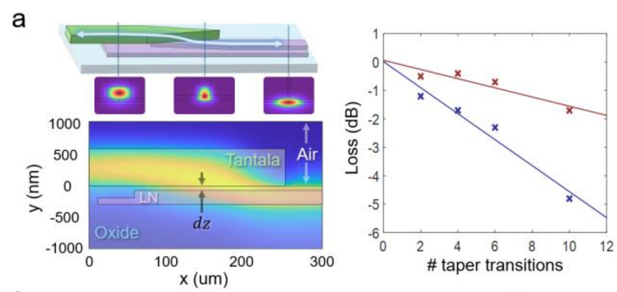
1g-Why is the performance of the QPM SH efficiency worse with tantalate than without by a factor of 3? Showing a table of the three tested devices along with some literature landmark demonstration would have been clearer towards the end of p8. Why is there a difference between 787nm and 485nm tantalate- LiNbO3 device?

- We note in the text that the SHG gain spectrum shifts on devices spaced across the wafer, and departs from ‘sinc^2’ theory suggesting deviations in phase matching even in single devices. This has been explored in the literature attributed to thickness variations in the underlying commercial LNOI wafer. With imperfect phase matching, variation in performance across single devices, chips and wafers is typical. Methods to improve upon this, for example using wafer level thickness measurements to bias phase matching designs, have also been explored in the literature. Because our processing employs ebeam lithography, we can directly realize high resolution and adaptable (wafer to wafer) QPM design biasing across the full wafer, which would be much more difficult or not possible using mask-based lithography. We aim to include such techniques in future work.

- We better clarify these points in the revised manuscript
  - LINE 219
  - We attribute variations in performance between the devices to deviations in phase matching based on our characterization of SHG gain presented above in Fig. 3d.

1h-What does limit the 0.5 dB loss related to the transition from the tantalate to the LiNbO3 layers? Is there some prospect to improve it?

- In this case, the thickness of the oxide interface layer is the likely limit of loss performance due to decreased modal overlap for thicker silica.
- We modified our Fig.3 schematic showing the interlayer taper to label the silica interface thickness, *dz*, to aid discussion



- We also performed FDTD studies to explore *dz* sensitivities and quantify performance / tolerances
  - LINE 175
  - Our E-beam lithography process is suited for high precision alignment of taper structures, with specified precision of +/-20 nm. More critical in terms of process control is the oxide planarization step using CMP, whereby its performance governs the silica interface layer thickness, *dz*, between the waveguides and affects modal overlap of the taper structures. In FDTD simulations with *dz* = 100 nm, we found the loss-per-transition at 1550 nm to be 0.05 dB and at 780 nm to be 0.4 dB, evidencing the wavelength dependence on performance.

1i-What is the upper limit of Ta2O5 thickness to be grown on the SiO2/LiNbO3 platform? Is it a limitation?

- We've fabricated >800nm thickness nonlinear devices and currently include this reference (H. et al. Tantala kerr nonlinear integrated photonics. Optica 8, 811–817 (2021))

- We don't find the tantala thickness to be a limiting factor, as the current thickness versatility is already suited to broad dispersion designs.

2. Section 2 starts by showing a resulting Ta2O5 waveguide integrated onto LiNbO3 waveguide, but no details about how such an alignment is performed, or quantitative information about the achieved alignment precision, tolerance or length are given.

- We now provide this information in the manuscript based on our E-beam lithography process precision.
    - LINE 175
    - Our E-beam lithography process is suited for high precision alignment of taper structures, with specified precision of +/-20 nm.

3. What is the purpose of the 500°C annealing step of Ta2O5 and why is it necessary? It is said that this is avoided for doped tantalate, but there is not much explanation for this…

- The text of the manuscript notes the ALD and anneal steps as mitigation of photorefractive effects, and includes references to literature that better explore these effects

4. What is the origin of the wavelength dependence of the vertical taper/ transition between Ta2O5 and LiNbO3, as 0.2dB is measured at 1550nm and 0.5dB at 780nm. It would be useful to have error bars on these values, considering for instance multiple measurements across the various chips?

- We agree that these considerations can be better explored.
- We attribute the wavelength dependence to the oxide interface layer thickness, as for shorter wavelengths the mode overlap is more strongly affected by waveguide separation. We also note that shorter wavelengths experience stronger sidewall scattering.
- We have a single chip with the test structures that lead to the taper loss plot in Fig 2a. However, we also have insertion loss measurements of actual nonlinear devices – we can directly compare straight waveguide tantala-only devices that are directly next to devices that route from tantala-LN-tantala which thereby provide quantitative data on the per taper loss.
- We have modified our manuscript to provide this detail and discussion.

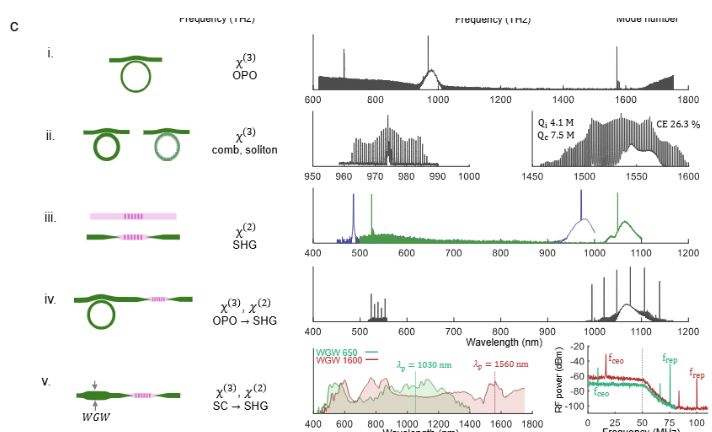
5. Regarding the design of the nonlinear c(3) devices (OPO, frequency combs…), is there any particular feature or intrinsic challenge due to the 3D integration of tantalate onto LiNbO3 ? For all the reported tantalate devices, references to prior work from the group on stand-alone tantalate devices are cited, which seems to imply that everything is the same. Is it really true?

- We did intend for this aspect to be presented as such – our process starting from the point of the oxidized wafer proceeds with no changes to our tantala-only processes that we reference

- The critical aspects to enable tantala with LN surround the interlayer transition for low loss routing between the platforms.
- We now include more discussion as well as new FDTD simulations to highlight the considerations in realizing low loss routing

6. The last paragraph of section 4 is by far the most interesting because these demonstrations report the use of both LiNbO3 and tantalate for cascaded device operation leveraging the c(2) and c(3) of the respective material. However, it remains very qualitative, showcasing the potential of the platform via illustrations: there are very few details about the resulting performance (pump power threshold, conversion efficiency etc…) so that it sometimes feels more like a bundle, rather than a scientific paper. I advise the authors to expand on these last results, so that we appreciate the novelty in a more quantitative way.

- We agree with the reviewer that devices that combine $\chi^{(3)}$ to $\chi^{(2)}$ dynamics are interesting. In the present work we demonstrate a broad array of the potential possibilities. Our current demonstrations aren't meant as an exhaustive exploration of designs but rather a foundational presentation of the technological integration and platform.
- We believe the current presentation of the already exceptional versatility in designs afforded by pairing tantala and LN, supported by our characterization of platform parameters that directly lend future designs, is highly novel and a suited to the current manuscript.
- To suit the reviewer's discussion and demonstrate even more $\chi^{(3)}$ to $\chi^{(2)}$ novelty, we performed additional experiments and include entirely new device designs that leverage $\chi^{(3)}$ to $\chi^{(2)}$ in a serially cascaded device (supercontinuum generation in tantala and SHG in LN for a monolithically integrated fceo detection device for f2f self referencing)
    - Fig. 4 c(v)
- We also added conversion efficiency as a quantitative metric to our comb results (discussed in previous comments similar this as well)
    - Fig. 4 c(iii)



7. Several reference to “tantalate-LN devices” or “3D tantalate LN devices” and “interlayer functionality” are made in section 4, although these devices are always used separately and at best sequentially. This should be clarified as I found it misleading at first. The sequential use of the two platforms to demonstrate various nonlinear functions slightly decreases the appeal of the integration approach. Is there some prospect to truly leverage the full potential of this approach to realize fully hybrid devices?

- We respectfully disagree. Fully integrated sequential $\chi^{(3)}$ to $\chi^{(2)}$ photonics unlocks a wealth of designs and makes a huge leap in integration compatibility. We demonstrate only a few potential possibilities, as our work isn’t meant as an exhaustive exploration of designs but a foundational presentation of the technological integration and platform.
- Devices that use $\chi^{(3)}$ / $\chi^{(2)}$ simultaneously are indeed interesting but beyond the scope of the current work.
- We thank the Reviewer for the comment and look forward to future work supporting these directions.

G. References: appropriate credit to previous work?

The benchmark to the literature should be made more thoroughly (see my comments above).

Regarding the routing and tapers to connect the two material layers, no reference is given. I think that this has been used for SiN to Si interface or for hybrid III-V onto SiN chips already. What is really new here?

- We previously included the reference **Sacher, W. D., Huang, Y., Lo, G.-Q. & Poon, J. K. S. Multilayer Silicon Nitride-on-Silicon Integrated Photonic Platforms and Devices. Journal of Lightwave Technology 33, 901–910 (2015)** which focuses in detail on the design, simulation, and test of multilayer taper structures, when we presented monolithic integration of multilayer photonics in the Introduction.
- To address the Reviewer’s comment, we now list this reference in closer proximity to the discussion on tapers to more clearly draw attention to it (in several locations where tapers are mentioned)
- Broadly, the approach of moving light between waveguides by tapering is not a new idea and indeed, the interlayer taper by itself is not the claim to novelty in this manuscript. Variations on taper design are common in literature and other platforms as the Reviewer notes. Their performance is an important consideration for our integration, but since their structure isn’t the claim to novelty we place limited focus on their design.
- We agree that more information on their design and performance could enable better benchmarking of performance, so we include new simulations and discussion:
    - We show a 3D schematic (exaggerated dimensions to show features) and a 2D slice labeling \rev{the silica interface thickness, dz, dependent on CMP planarization, overlaid

with a sample finite-difference-time-domain (FDTD) simulation in Fig. 3a (left). Our E-beam lithography process is suited for high precision alignment of taper structures, with specified precision of +\-20 nm. More critical in terms of processing is the CMP oxide planarization step, which controls dz between the waveguides and affects modal overlap of the taper structures. Further, modal overlap in general includes a wavelength dependence due to the stronger confinement of optical modes at shorter wavelengths **{sacher_multilayer_2015}.** In FDTD simulations with dz = 150 nm, we found the loss-per-transition to be <0.05 dB at 1550 nm and 0.3 dB at 780 nm. For dz = 200 nm, we simulate the loss-per-transition to be <0.05 dB at 1550 nm while falling to 1.5 dB at 780 nm, evidencing the wavelength dependence on modal overlap.}

H. Clarity and context: lucidity of abstract/summary, appropriateness of abstract, **introduction and conclusions**

I advise the authors to temper down the claim on "arbitrary substrates" or "nearly universal compatibility in integration" in the title, abstract, introduction and conclusion (first sentence "we have presented monolithic 3D integration that combines tantalate photonics with arbitrary substrates"): only integration onto the LiNbO3 platforms is demonstrated here, which, I acknowledge, is an important one. Even though the process flow involves steps that could be used for a wide range of material platforms, the present demonstration focuses onto LiNbO3 exclusively, so it does not justify in my opinion the title/ abstract in its current form.

- We agree with the reviewer and **are open to changing the title** since both Reviewers discuss this providing a more appropriate claim.
- We think the 'arbitrary' piece is not the most important, but that the most important piece is the monolithic integration and our technical integration approach/tantala's properties that make it broadly compatible with other platforms.
- We now include a more thorough description of tantala's material and processing parameters to better present our key claim, and lend future exploration using other platforms and integration.

To reemphasize the previous point, no integration with active devices like lasers and modulators, nor active devices involving electrical control of the device operation, is shown. Instead, the group has recently demonstrated the heterogeneous integration of III-V lasers and Ta2O5 rings for OPO applications leveraging more traditional bonding technologies (ref [20] of the manuscript). Would there be any hurdle to integrate III-V lasers with Ta2O5 using the present approach?

- Our current work is focused on exploring nonlinear designs, and we agree with the Reviewer that these are interesting directions for future work
- To address the Reviewer's comment, we support a title change that captures the novelty of our integration approach without the claim of arbitrary integration, as discussed above

Similarly, the use of low temperature several times in the introduction/ abstract is misleading since 500°C annealing is still performed here. While this is much less than what is used for SiN indeed, I would not qualify it as low temperature. For clarity, I would replace by moderate and state the number to avoid any confusion.

- We agree and address this by now using 500 C and/or 'moderate' wherever appearing to more clearly identify the anneal temperature where mentioned.
- We reference literature for 'anneal free' tantala photonics, whereby metal-oxide mixtures enable high Q performance with no final anneal step (ref Carollo et al.)

The claim that Ta_2O_5 enables "efficient mode matching between waveguides" might be true for $LiNbO_3$ but why would it be universally true for any other material platform? Efficient 3D routing is likely to work best for $LiNbO_3$. Could the authors comment on this, especially in the context of presenting this demonstration as a universal approach?

- Interlayer tapers are broadly effective for transitioning optical modes between materials even with higher index contrast, and have been demonstrated in the literature in many varieties e.g. between SiN and Si
- We previously included the reference **Sacher, W. D., Huang, Y., Lo, G.-Q. & Poon, J. K. S. Multilayer Silicon Nitride-on-Silicon Integrated Photonic Platforms and Devices. Journal of Lightwave Technology 33, 901–910 (2015)** which explores taper designs in great detail, including both transitions between SiN-SiN and SiN-Si, when we discussed multilayer monolithic integration
- We now explicitly include this reference in the text whenever we discuss taper structures as well

Reviewer Response Document: Manuscript ID: 2025-08-22671

Title: **"Monolithic 3D integration of tantalum pentoxide nonlinear photonics ~~on a functional substrate~~"**

Corresponding Author: Grant Brodnik; NIST Time and Frequency Division 688

Referees' comments:

Referee #1 (Remarks to the Author):

I have read the authors' rebuttal letter with interest and I think amendments significantly improve completeness and focus of the paper. Regarding the paper title, I do not find the phrase “functional substrate” immediately comprehensible. I would lean more toward something like “patterned substrate” but this is a personal feeling. I do not have further comments.

The authors graciously thank the Referee for their considerable efforts during our manuscript review, which contributed to a greatly strengthened final submission.

Referee #2 (Remarks to the Author):

I acknowledge the improvement brought by the authors to their manuscript to better highlight the main novelty/ claim of their findings, which are indeed centered on the demonstration of monolithic integration of tantalum pentoxide with LNOI for increased linear and nonlinear photonic functionalities.

The authors have made some efforts to better stress the advantages/ properties of this relatively recent material platform, and the results achieved are indeed convincing enough regarding the future prospects to integrate this material with other photonic platforms thanks to the less constraining processing method with respect to alternative approaches used in the literature.

I also appreciated the change in the title which I find more appropriate, as well as the clarification that is added early enough in the abstract/ introduction regarding the specific use of LNOI, as was chosen to illustrate the potential of this monolithic integration.

Finally, the addition of experimental nonlinear results (SCG + SHG) for f-2f self-referencing is relevant and appealing to illustrate the advantages of leveraging combined c(2) and c(3) nonlinear functions via exploiting this new hybrid Ta_2O_5/LN material platform.

We again the Referee for their detailed feedback and considerable efforts in review. By addressing these and previous items, we achieved a greatly strengthened final Manuscript.

I just have a few minor points to clarify the technical/ scientific content of the manuscript before fully endorsing the manuscript for publication:

-Regarding the use of edge couplers (which was also questioned by reviewer 1), the authors mention "For the designs presented in this work, the output waveguides are inverse tapered (increasing width approaching the edge of the chip) to a width of 2.5 um." To my knowledge, inverse tapers refer to decreasing waveguide width close to the chip end-facet, which is an alternative strategy to increase fiber-chip coupling. Instead, if I understand correctly, here the authors use flared waveguides. I would advise them to change the terminology to be more accurate and avoid confusion.

We agree this can be confusing terminology. To avoid confusion, we've removed 'inverse taper' and now just explicitly state the design of increasing width waveguides for the edge coupler.

> - '…incorporate waveguides with an increasing width to 2.5 um at the facets to improve mode matching with optical fibre.'

-The authors mention the use of TE0 mode in LNOI (X cut) to leverage the best of the nonlinear waveguide. Could they also give the orientation of the LN waveguide axis? I don't recall that it was specified. Is it along the Y axis?

We now include the waveguide orientation in the Methods section when describing our fabrication details.

- '…Waveguides in the LN layer are directed along the y-axis, affording TE polarized operation along in the z-axis.'

-The authors now indicate the material properties of the Ta_2O_5 (in the additional table in fig 2b). Yet, could they specify the wavelength at which they give the n2 value? In the context of benchmarking it against SiN, although the respective fabrication processes are now better compared, they do not directly cite the n2 of SiN, but only mention in their response letter that it is larger for Ta_2O_5. For completeness, could they include the n2 value of SiN at 1550nm as well in the manuscript to stress how much larger is that of Ta_2O_5?

We now include an additional reference for tantala n2 to provide literature values at 800 nm and 1550 nm (directly listed in the manuscript text), as well as the n2 value of SiN from referenced literature (also directly listing the wavelength for measurements in referenced literature).

- In terms of nonlinear performance, we note the strong nonlinear coefficient of tantala (6.2×10^{-19} m^2W^{-1} measured at 1550 nm [1] and 7.23×10^{-19} m^2W^{-1} measured at 800 nm [43]), in comparison to SiN (2.4×10^{-19} m^2W^{-1} measured at 1550 nm)

-Besides, regarding the use of c(3) Ta_2O_5 devices, could the authors also indicate the nonlinear gamma parameter (/W/m) at the different pump wavelengths, for both narrow and wide waveguides. This key metric would help the nonlinear photonics community to appreciate even further the potential of Ta_2O_5 nonlinear photonics and compare this nonlinear material platform against more traditional ones (like SiN or Si…)

We agree that directly reporting our calculated gamma is especially relevant for supercontinuum (SC) devices and performance. We therefore now report these parameters for both the narrow SC device (650 nm waveguide width, operated at 1030 nm) and for the wide SC device (1600 nm waveguide width, operated at 1560 nm) directly in the text accompanying results.

- We calculate the effective nonlinear parameter, $\gamma = 2\pi n2/\lambda Aeff$, where A_{eff} is the effective mode area, for these two designs to be 11.0 $W^{-1}m^{-1}$ (1030 nm pump, WGW = 650 nm) and 3.0 $W^{-1}m^{-1}$ (1560 nm pump, WGW = 1600 nm).

-Fig 2d: could the authors indicate at which wavelength are the Qi values reported on the different points across the wafer? Is it 1550nm?

We include now the wavelength for these measurements (1550 nm) in Fig. 2 caption where they are reported.

- '… tantala-LN wafer image identifying Qi, measured at 1550 nm, for microresonators of the same design from three separate chips at different locations on the wafer.'

-I still do not support the misleading use of the terminology "photonic crystal resonator" in place of corrugated rings, but I don't think we will agree on this…

-For the various nonlinear devices demonstrated on fig 4c, I strongly recommend the authors to indicate the pump power used for the different measurements and specify both the input (before the chip) and coupled pump power. Similarly, the total Q of the resonators should be specified in link with the related measurements.

We now include an additional section in the Methods ('Nonlinear testing conditions') to provide experimental details (on chip pump powers or off chip / insertion loss where applicable, wavelengths, resonator loaded/total Q as requested) supporting the data presented in Fig. 4.

> We provide here experimental conditions that accompany data presented in Fig. 4c of Main. Figure 4c(i) shows microresonator OPO for a device with a measured loaded Q of 0.43 M. The device was pumped at 968 nm with an on-chip pump power of 44 mW. Figure 4c(ii) presents dark pulse spectra of two normal dispersion PhCR devices. The 980 nm device (left) was measured to have loaded Q of 2.0 M and was operated with on-chip pump power of 89 mW. The 1550 nm device (right) was measured to have loaded Q of 2.65 M and was operated with on-chip pump power of 100 mW. Figure 4c(iv) presents microresonator OPO cascaded to LN for SHG. The microresonator was measured to have loaded Q of 0.62 M, and was operated with on-chip pump power of approximately 50 mW. Figure 4c(v) presents supercontinuum generation in tantala waveguides cascaded to LN SHG. For the 1030 nm device (blue), the chip insertion loss was measured to be 6 dB per facet and the laser source has pulse width 150 fs, average power 150 mW, and repetition rate of 100 MHz (peak power 10 kW). For the 1560 nm data (orange), the insertion loss was measured to be 5 dB per facet and the laser source has pulse width 150 fs, average power 58 mW, and repetition rate 75 MHz (peak power 5.2 kW).

-For the relevant SCG + SHG measurement added to the manuscript, I advise the authors to specify whether the quoted power is the input or coupled pump power, and to indicate the peak power (in addition to the average power), which is more relevant to SCG. They should also quote the nonlinear parameter gamma of their Ta_2O_5 waveguides here (see above).

- (see similar item above) – In the now-included Methods section 'Tantala nonlinear testing conditions' we fully detail the pump laser parameters for both CW resonator devices and for the SCG devices (average power, repetition rate, and pulse duration) which enables direct estimation of peak power.

- In the Main Manuscript accompanying our SCG results, we also now provide calculations for gamma as recommended to offer better comparison with literature.
    - We calculate the effective nonlinear parameter, $\gamma = 2\pi n2/\lambda Aeff$, for these two designs to be 11.0 $W^{-1}m^{-1}$ (1030 nm pump, WGW = 650 nm) and 3.0 $W^{-1}m^{-1}$ (1560 nm pump, WGW = 1600 nm).