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REVIEWER COMMENTS

Reviewer #2 (Remarks to the Author):

The manuscript by Ding and co-workers reports on a highly important result in the field of diamond nanophotonics with impact on the wider fields of quantum optics and quantum technology. Diamond is host to the most advanced optically active spin qubits; state-of-the-art experiments in quantum computing and quantum networking having been performed with diamond NV centers and SiV centers. To move beyond the state of the art, a high efficiency of the spin-photon interface is critical, as well as methods to perform reliable, high-yield and large-scale fabrication and integration of devices. In the absence of high-quality diamond device layers on insulator, groups have worked around this by using bulk diamonds and creating free-standing structures using complex etching techniques. Recently, several groups (including notably the group of Alex High, co-PI on this work) have made significant progress in using smart-cut-like processing to obtain thin, flat diamond membranes on insulator with almost no wedge and very low roughness. This breakthrough is anticipated to yield quantum-optical devices with significantly better quality and yield thanks to the smooth surfaces, a well-defined starting material and straightforward processing.

In the current manuscript, the authors demonstrate exactly this: record-high quality factor photonic crystal cavities, very high yield, and the ability to create 2D structures, all of which significantly surpass the state of the art. These results confirm that the new fabrication methods open exciting new possibilities for (diamond-based) quantum photonics. I therefore strongly recommend publication.

The manuscript is clear and well written. I have a few very minor questions and suggestions:

- On line 243, it is mentioned that a single-exponential fit is used. It appears that the fit function also contains a constant offset. It would be good to mention that (e.g. in the caption) to avoid confusion, and to provide the fit values (e.g. in supplementary)
- In the paragraph starting on line 390, the quantum efficiency of the SiV does not seem to be taken into account (or is it assumed to be part of the 19.3%)? The quantum efficiency (or stated differently: the presence of non-radiative channels) does affect how the Purcell enhancement translates into lifetime reduction.
- The expected cooperativity (line 400 and beyond) assumes the natural SiV linewidth. However, for quantum protocols it is the coherent cooperativity that matters, for which the full linewidth should be taken into account (in the current work this linewidth is a factor of 5 larger than this natural linewidth, leading to a factor of 5 lower coherent cooperativity). For this reason, for full disclosure, my advice is to also provide the expected coherent cooperativity based on actually measured values in this work alongside the best possible value.

Reviewer #3 (Remarks to the Author):

Ding et al. have presented an experimental demonstration of high-Q optical cavities in thin-film diamond, achieving quality factors that exceed 10^5 . This value sets a new record for photonic crystal cavities at this wavelength. The study successfully demonstrates high-quality factors in both two-

dimensional photonic crystals and one-dimensional nanobeam photonic crystals. They also demonstrate direct adiabatic coupling to a tapered fiber, which results in high collection efficiency. These advancements have the potential to significantly enhance the performance of diamond-based quantum photonic devices. I recommend this paper for publication once the authors address the following technical issues.

- 1) The authors employ ion slicing to fabricate thin film diamond, a method known to potentially adversely affect spin coherence times. Do the authors anticipate any negative impacts on spin coherence times due to this process?
- 2) The authors have reported a Purcell factor of 13, which is notably lower than anticipated given the exceptional quality factors presented. Could the authors specify the number of emitters characterized? Additionally, is the observed Purcell factor consistent across different emitters, or are there significant fluctuations as would be expected from the random emitter positions?
- 3) The use of gold and chrome deposition is unclear. The authors claim it is used to make a frame for the structure. Is this to prevent delamination or some other degradation from subsequent processing steps? It would help to have some more detail for better clarity.
- 4) In figure 3 the authors claim that their cavities achieve wavelengths very close to those predicted by numerical simulation. But the figure is plotted on a very large scale and in a way that is difficult to understand. The points are all highly clustered and it looks like the deviation in spectral wavelength can be as high as 100 nm for some target wavelengths (e.g. 680 nm target wavelength). It would be helpful if the authors provided a clearer representation of the deviation and how close they actually get to the predicted value. In particular, it would help to know what the variation in wavelength is for identically fabricated cavities. This is important because for scalability all cavities must have similar wavelengths, and although they can be tuned through methods like gas deposition, the tuning range is limited. Some additional quantitative discussion and how repeatable the wavelength is would be helpful to judge the potential for scalability.
- 5) For the measurements on the siv, the authors do not provide any count rates, or at least I could not find them. Based on the Purcell factor of 13 and the exceptional coupling efficiency to fiber attained through adiabatic tapering, I would expect extremely impressive count rates of hundreds of MHz. Do the authors observe such count rates? The authors should report their count rates and perform a photon budget to determine whether they are consistent with the observed efficiency of the nanobeam and radiative enhancement of the cavity. I think a discussion like this will significantly improve the manuscript.

Reviewers' comments

Author's response

Manuscript modified text

Original manuscript text

Reviewer 2

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In the current manuscript, the authors demonstrate exactly this: record-high quality factor photonic crystal cavities, very high yield, and the ability to create 2D structures, all of which significantly surpass the state of the art. These results confirm that the new fabrication methods open exciting new possibilities for (diamond-based) quantum photonics. I therefore strongly recommend publication.

The manuscript is clear and well written. I have a few very minor questions and suggestions:

Thank you for the accurate summary of the work. We also appreciate the positive reception of the work and the assessment that it is suitable for Nature Communications.

1. On line 243, it is mentioned that a single-exponential fit is used. It appears that the fit function also contains a constant offset. It would be good to mention that (e.g. in the caption) to avoid confusion, and to provide the fit values (e.g. in supplementary)

We thank the reviewer for their careful reading of our manuscript and this question. The fit function indeed contains an offset. The main text and the Method sections are modified to reflect this.

*By fitting the curves with a single exponential function **with a constant offset**, we obtain the lifetime of the SiV on resonance (τ_{on}) to be 0.47 ± 0.006 ns, which is reduced approximately by a factor of 3 from the off-resonance value (τ_{off}) of 1.3 ± 0.01 ns.*

The Purcell factor of the investigated zero-phonon line (ZPL) F_{ZPL} , is estimated using the following equation⁹: $F_{ZPL} = (\tau_{off}/\tau_{on} - 1)/\epsilon_{ZPL}$ ⁹, where ϵ_{ZPL} is defined by the fraction of the total emission into the ZPL visible at 4K for an SiV, which is estimated by a product of the Debye–Waller factor of 70%⁵⁰ and the

branching ratio of 19.3% into D line at 4K⁹. We use an exponential with a constant offset to fit the lifetimes in the main text (τ_{on} and τ_{off}). The constants are fitted to be 51 ± 2 and 38 ± 2 , which are contributed to the dark counts of the system.

2. In the paragraph starting on line 390, the quantum efficiency of the SiV does not seem to be taken into account (or is it assumed to be part of the 19.3%)? The quantum efficiency (or stated differently: the presence of non-radiative channels) does affect how the Purcell enhancement translates into lifetime reduction.

We thank the reviewer for the question. Indeed, quantum efficiency affects how the Purcell enhancement is related to lifetime reduction. We included the branching ratio in the expression we used to calculate the Purcell factor from the measurements, as in $F_{ZPL} = (\tau_{off}/\tau_{on} - 1)/\epsilon_{ZPL}$. However, Purcell factor in this expression: $F \equiv \frac{3}{4\pi^2} \frac{\lambda^3}{n^3} \frac{Q}{V}$, is calculated purely from cavity parameters, independent of the branching ratio. In other words, depending on the branching ratio, the same Purcell factor can correspond to different amount of lifetime reduction. I have added text to the Method section to clarify.

The theoretical Purcell factor is obtained from the following equation: $F \equiv \frac{3}{4\pi^2} \frac{\lambda^3}{n^3} \frac{Q}{V}$. The theoretical value is dictated entirely by the cavity parameters. We use the measured $Q \sim 1.2 \times 10^5$, simulated $V \sim 0.5(\lambda/n)^3$, and $B=19.3\%$ of the D line, and estimated F to be 1.8×10^4 in the most ideal case.

3. The expected cooperativity (line 400 and beyond) assumes the natural SiV linewidth. However, for quantum protocols it is the coherent cooperativity that matters, for which the full linewidth should be taken into account (in the current work this linewidth is a factor of 5 larger than this natural linewidth, leading to a factor of 5 lower coherent cooperativity). For this reason, for full disclosure, my advice is to also provide the expected coherent cooperativity based on actually measured values in this work alongside the best possible value.

We thank the reviewer for the comment. Our measurement of the linewidth is conducted at ~ 4 K, and the quoted value of nature linewidth is from a ~ 100 mK measurement, which is the temperature at which state-of-the-art SiV experiments are conducted. The linewidth at 4 K is usually wider. I have updated the related numbers and section in the main text, to include the clarification, and also the values calculated from the linewidth observed in this work.

If the SiV is placed at the cavity field maximum, the estimated g is 15.2 GHz, and experimentally, it has been observed to be ~ 8 GHz⁶. To be realistic, we use $g \sim 8$ GHz, the natural SiV linewidth ($\gamma \sim 0.12$ GHz)⁶. The SiV linewidth is $\gamma \sim 0.12$ GHz at ~ 100 mK⁶ or $\gamma \sim 0.61$ GHz as measured in this work at ~ 4 K, and the measured highest Q is $\sim 1.8 \times 10^5$ ($\kappa \sim 2.2$ GHz) or $Q \sim 8.4 \times 10^4$ ($\kappa \sim 4.8$ GHz) in the critically-coupled case. Using the ideal numbers, a C as large as 3500 at 100 mK can be obtained. Using the observed $g \sim 8$ GHz, the obtained values are $C \sim 970$ or $C \sim 440$ at 100 mK. Using the observed g and

linewidth of SiV at 4K from this work, the obtained values are $C \sim 190$ or $C \sim 87$ at 4K. ~~The obtained values are $C \sim 970$ or $C \sim 440$. If we use the theoretical $g \sim 15.2$ GHz, the C could be as large as 3500.~~

Using masked implantation, better overlap between SiV and optical mode can be achieved, resulting in cooperativities > 440 at ~ 100 mK (see Method).

Reviewer 2

General Comments:

Ding et al. have presented an experimental demonstration of high-Q optical cavities in thin-film diamond, achieving quality factors that exceed 10^5 . This value sets a new record for photonic crystal cavities at this wavelength. The study successfully demonstrates high-quality factors in both two-dimensional photonic crystals and one-dimensional nanobeam photonic crystals. They also demonstrate direct adiabatic coupling to a tapered fiber, which results in high collection efficiency. These advancements have the potential to significantly enhance the performance of diamond-based quantum photonic devices. I recommend this paper for publication once the authors address the following technical issues.

Thank you for the accurate summary of the work. We also appreciate the positive reception of the work and the assessment that it is suitable for Nature Communications.

1. The authors employ ion slicing to fabricate thin film diamond, a method known to potentially adversely affect spin coherence times. Do the authors anticipate any negative impacts on spin coherence times due to this process?

We thank the reviewer for the question. The thin film is produced through ion slicing and overgrowth, which should recover the high-quality of the diamond lattice for the spin. The production detail of the diamond sample is illustrated in a previous work¹, where people have observed coherent properties of nitrogen vacancies in the film (NVs) which are sensitive to defects. In this work, we did not characterize the coherent properties of the SiVs due to set up constraints, but the relatively narrow and stable linewidth is a positive piece of evidence that the lattice is not damaged. We did not observe any abnormality in optical signals (e.g. instability, or prominent background fluorescence). Combined with the observed long NV coherence time, we feel optimistic about the SiV spin properties. A clarification is added to the main text to address this concern.

The SiVs are generated through implantation across the membrane before the transfer, resulting in randomly distributed SiVs. This thin-film diamond has been shown to host emitters with promising optical and spin properties³², satisfying one of the pre-requisites of a promising platform for spin-photon interface.

2. The authors have reported a Purcell factor of 13, which is notably lower than anticipated given the exceptional quality factors presented. Could the authors specify the number of emitters characterized? Additionally, is the observed Purcell factor consistent across different emitters, or are there significant fluctuations as would be expected from the random emitter positions?

We thank the reviewer for the question. There were two SiVs that are placed physically close to a cavity that is <1 nm from 737 nm, so we characterized both of them. This is both limited by the sparse density of the SiV and the tunability of the available measurement set up: there are only a few SiVs (around 3, 5) per nanobeam, so only a small number of them can be close to the cavity (e.g. $< \lambda/2$ away); the set up also could only tune < 1 nm. The other characterized SiV has a relatively narrow linewidth as well, but did not show lifetime reduction, therefore not coupled to the cavity. In ideal situations, 50% of the SiVs would not couple to the cavity mode due to the zero overlap between the dipole moment and the optical mode of the cavity. Therefore, we think the other SiV is not coupled to the orthogonal spin orientation. We have taken confocal scan images of the SiVs implanted across the device, as shown in Fig. r1. The position is indeed random. The density is ~ 1 SiV/ $1 \mu\text{m}^2$ in the unpatterned area.

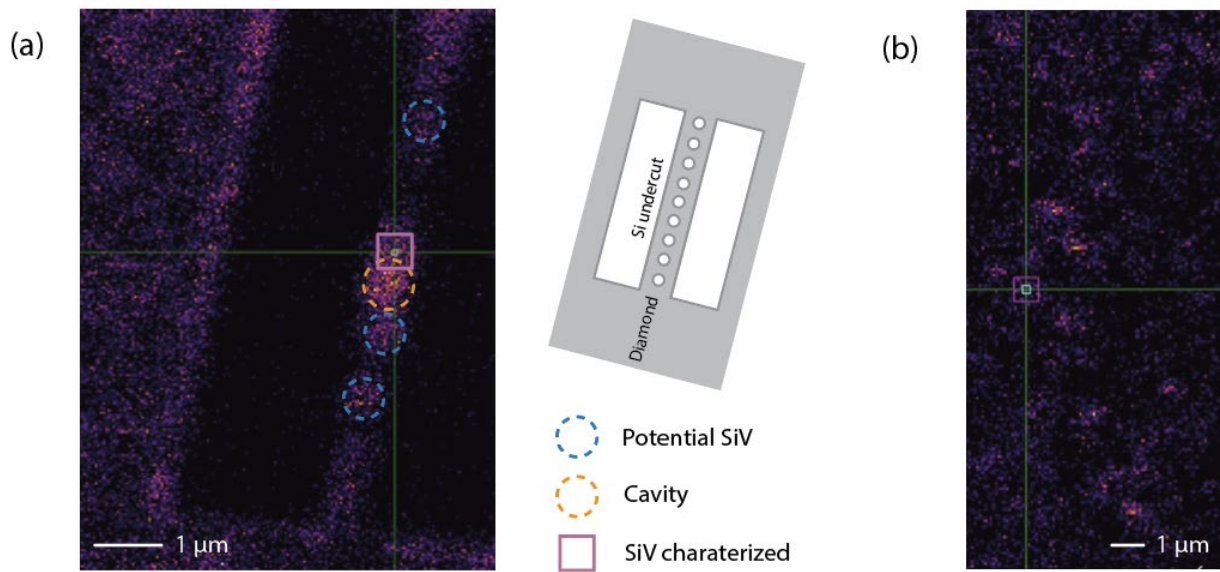


Fig. r1. Confocal scan photoluminescence (PL) images over the device and unpatterned area of the diamond. (a) PL image over the device. The brightness of the pixels corresponds to photon counts with arbitrary normalization, only for visual purposes. The cursor is parked at the SiV being characterized, that is close to the cavity. The cavity is bright located at the center of the nanobeam. The other bright spots are SiVs, that can be identified using the spectra. (b) PL image over the unpatterned area. The bright spots are potential SiVs (They could also be fluorescent defects or particles, and sometimes be multiple SiVs. This image only gives a rough estimate of the SiV location and density.)

3. The use of gold and chrome deposition is unclear. The authors claim it is used to make a frame for the structure. Is this to prevent delamination or some other degradation from subsequent processing steps? It would help to have some more detail for better clarity.

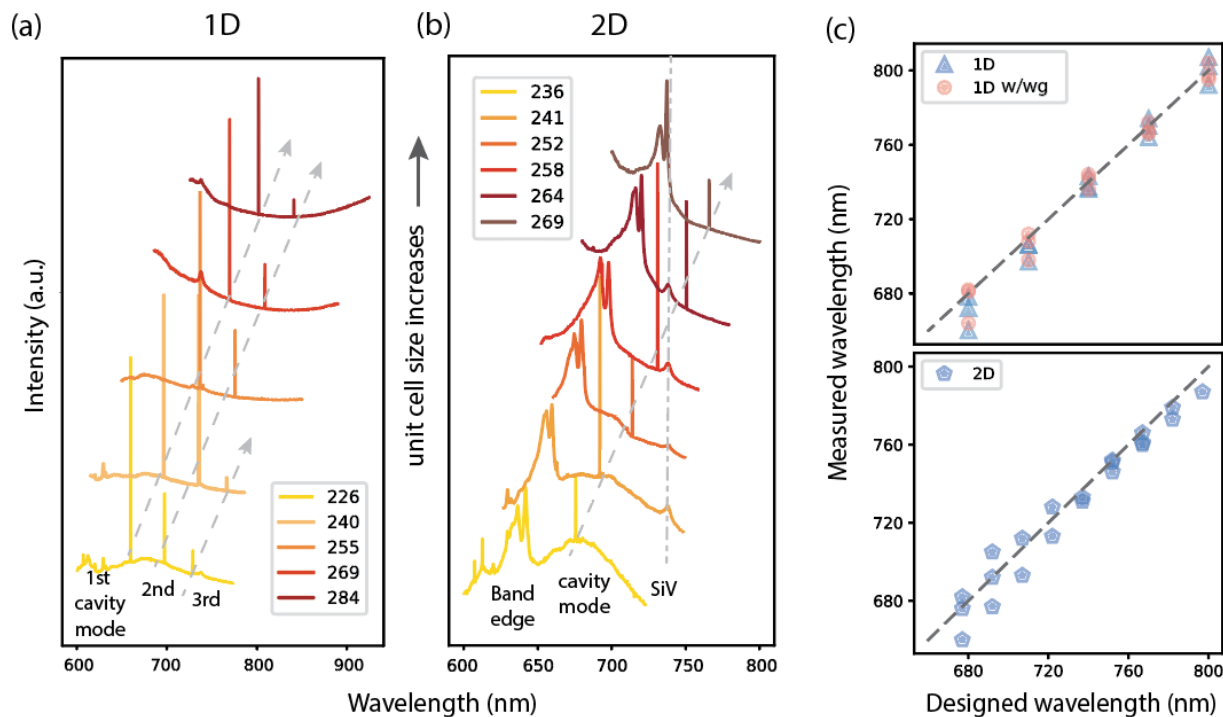
We thank the reviewer for the question. It is used to prevent delamination, since Cr and Au are not etched by HF or affected by the following processes which could affect the adhesion of the thin film to the substrate. We have added more texts to explain such a step.

(2) A liftoff of Cr/Au metal stack is performed to define a “frame” around the film in order to secure it on the carrier chip and to prevent delamination.

4. In figure 3 the authors claim that their cavities achieve wavelengths very close to those predicted by numerical simulation. But the figure is plotted on a very large scale and in a way that is difficult to understand. The points are all highly clustered and it looks like the deviation in spectral wavelength can be as high as 100 nm for some target wavelengths (e.g. 680 nm target wavelength). It would be helpful if the authors provided a clearer representation of the deviation and how close they actually get to the predicted value. In particular, it would help to know what the variation in wavelength is for identically fabricated cavities. This is important because for scalability all cavities must have similar wavelengths, and although they can be tuned through methods like gas deposition, the tuning range is limited. Some additional quantitative discussion and how repeatable the wavelength is would be helpful to judge the potential for scalability.

We thank the reviewer for the question. The points are at $x = 680$ (nm) correspond to $y = 682, 681, 664, 660, 678, 672$ (nm) for 1D, and $x = 677, y = 676, 682, 660$ for 2D, so much closer than 100 nm. It could have been the top and bottom panels of Fig. 3(c) are connected, so the y axis is not clear. I have adjusted the spacing between the two to avoid such confusion.

(modified Fig. 3)



The dense distribution of points around the dashed line in Fig. 3 (c), which represents the ideal condition, is a result of the small deviation of the fabricated device resonances from the designed ones. The standard deviation for 1D and 2D devices are 7.6 nm and 8.2 nm. Using the central wavelength of ~ 740 nm, the characteristic deviations are 1.0% and 1.1% in percentage respectively. I have added more explanation and some corrections to the main text.

The tuning range is conventionally ~ 10 nm with nitrogen gas at 4 K or 100 mK environments without sacrificing device performances^{2,3}. More context is added to the main text for clarifications.

This fabrication method exhibits high uniformity (resonances close to simulation) and high-yield (high-Q modes) for both 1D and 2D PhC cavities: from Fig. 3 (c), we estimate the standard deviation of the measured wavelengths from the simulation to be within 2.9% and 2.5% 1.0% and 1.1% (7.6 nm and 8.2 nm) for 1D and 2D cavities, respectively. Typically, the tuning range is ~ 10 nm with nitrogen gas deposition at 4 K or 100 mK environments without sacrificing device performances, so these devices are well within the tuning range.

- For the measurements on the SiV, the authors do not provide any count rates, or at least I could not find them. Based on the Purcell factor of 13 and the exceptional coupling efficiency to fiber attained through adiabatic tapering, I would expect extremely impressive count rates of hundreds of MHz. Do the authors observe such count rates? The authors should report their count rates and perform a photon budget to determine whether they are consistent with the observed efficiency of

the nanobeam and radiative enhancement of the cavity. I think a discussion like this will significantly improve the manuscript.

We thank the reviewer for the question. The SiV are measured confocally/through free space not through the tapered fiber coupling, because we need to check the position of the SiV through a confocal set up, and the 4 K cryostat currently does not have fiber-coupling capabilities. As a result, we did not see a very high photon rate, besides the enhancement from having suspended structures compared to bulk. We hope to set up a fiber coupling port for the cryostats for future demonstrations, which should significantly improve the count rates³.

However, we did notice faster rates when the cavity is tuned into resonance with the SiV. as shown in Fig. 5(e), the photon counts become enhanced on the D line by ~ 20 times relative to the other lines. This is consistent with the extracted Purcell factor of 13. Some clarification is added to the Method section to connect the photon rate difference and measured Purcell factor.

For this estimation, we also assume that the lifetime of a SiV measured in unpatterned area τ_{bulk} (~ 1.2 ns) is equal to the off-resonance value⁹. Notice, the extracted Purcell factor of 13 is consistent with the ~ 20 -fold relative increase in photon rate at the D line when the cavity is tuned into resonance with it. The error could come from the limited resolution of the spectrometer (1800 gr/mm).

[1] Guo, X. *et al.* Direct-bonded diamond membranes for heterogeneous quantum and electronic technologies. *arXiv [physics.app-ph]* (2023).

[2] Kuruma, K. *et al.* Coupling of a Single Tin-vacancy Center to a Photonic Crystal Cavity in Diamond. *Appl. Phys. Lett.* **118**, (2021).

[3] Bhaskar, M. K. *et al.* Experimental demonstration of memory-enhanced quantum communication. *Nature* **580**, 60–64 (2020).

REVIEWERS' COMMENTS

Reviewer #2 (Remarks to the Author):

The authors have made the requested adaptations and improvements based on the referees' comments and in my view the manuscript is now ready for publication.

Reviewer #3 (Remarks to the Author):

The authors have addressed all of my comments. I recommend publication.