REFEREE REPORT

Resonant enhancement of the zero-phonon emission from a colour centre in a diamond cavity

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I review a recent article [*Nat. Photon.* **5**, 301 (2011)] reporting on the resonant enhancement of the zero-phonon emission from nitrogen-vacancy centers in a single-crystal diamond microring cavity. I first provide a summary of the article's main results followed by a discussion of the article's figures. Then I provide an extensive list of shortcomings and suggestions that should be addressed prior to publication. I also discuss the originality of the article's conclusions, look at the significance of its findings, and provide some suggestions for additional references. Finally, I recommend the manuscript for publication however it should be extensively revised and undergo a second round of peer-review.

I. Article summary

> *Start by providing a paragraph summarizing what you think are the main findings and claims of the manuscript.*

I provide a technical summary for the purposes of the final exam followed by a general audience summary for pedagogical purposes as I found it very useful and insightful to write out for myself.

A. Technical summary

The authors demonstrate coupling of the zero-phonon line (ZPL) of individual nitrogen-vacancy (NV) centers to the modes of a single-crystal diamond microring resonator. This coupling lead to the decrease of the photoluminescence (PL) lifetime of the NV centers (desirabel for faster NV center manipulation times) and to the main result of the article—the Purcell enhancement of the spontaneous emission ratio into the ZPL by a factor of ∼10 without enhancing emission into the phonon sidebands. Without any enhancement, only ∼3% of emission is into the ZPL but the authors demonstrate an emission ratio of ∼25%. This is a critical step towards the utilization of NV centers as spin qubits and nanoscale sensors in spintronics and quantum technologies as a high ZPL emission ratio >99.5% absolutely crucial for these applications. Furthermore, the authors use a new method for fabricating structures out of single-crystal diamond that allowed for the creation of the high Q-factor cavity employed.

B. General audience summary

Nitrogen-vacancy (NV) centers are point defects in the tetrahedral crystal structure of diamond consisting of a nearest-neighbour pair of a substitutional nitrogen atom and a lattice vacancy. The lattice vacancy forms because unlike the carbon atom it replaces which can form four covalent bonds, nitrogen can only form three, leaving a "stuck" electron in the vacancy that is highly isolated and whose spin may be exploited. Nitrogen is not the only possible defect—silicon is another and over 500 have been detected [1]—and these defects together are sometimes referred to as color centers (e.g. in the title) as they exhibit photoluminescence (PL) in the visible region. Neutral NV centers (NV^0) also exist however they are usually just studied to obtain insight into NV^- centers and only speculative arguments exist for their utility [2].

Not only is diamond host to these versatile color centers, it also exhibits a unique combination of properties that make it by far the most promising candidate for quantum photonic technologies [3]. It has the largest optical bandgap of any material allowing for easy optical manipulation of the NV centers across a wide range of wavelengths from the ultraviolet through to the far infrared. It has a high refractive index, high thermal conductivity, is biocompatible, and chemically robust, making it suitable for a wide range of applications. For these reasons, and due to their color centers, diamond cavities have been very popular and are employed in this work as well.

The NV center should not have access to any phonon transition (i.e. excite any phonons as it changes state) as they will lead to reduced spin coherence times and the leakage of information. Thus the 'zero-phonon line' (ZPL) at 1.945 eV (637 nm) of the NV^- center must be used which excites the electron without exciting any phonons at all. This is as opposed to exciting the phonon sidebands which will excite both the NV^- center and lattice phonons.

Unfortunately the optical properties of NV centers are not all ideal. Two problems are that NV centers do not couple strongly to the electromagnetic field and that

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only a tiny fraction (∼3%) of the radiation is emitted into the ZPL. One way to increase this fraction is by coupling the NV center to an optical microcavity where the spontaneous emission rate is enhanced due to the Purcell effect. However, only enhancement of the ZPL emission is desirable, which is a significant accomplishment in this work. The ultimate goal, for implementing quantum protocols at least, is to reach a ZPL emission ratio of >99.5%.

NV centers have been studied for decades however they attracted considerable interest in 1997 when individual NV^- centers were detected [4] and subsequently used to build stable single photon sources at room temperature [5] in 2000. Today NV centers are of vehement interest because they possess highly desirable properties making them very promising candidates for spin qubits and nanoscale multi-sensors [6] with wide applications in spintronics [7] and quantum photonic technologies [8].

II. Figure summary

Outline in your own words the findings presented in the manuscript's figures and how they relate to the manuscript's claims.

A. Figure 1

Fig. 1 shows a scanning electron microscope (SEM) image of the fabricated diamond microring resonator. Embedded in it are NV centers whose ZPL were successfully coupled to the resonator's modes. The diamond microring is 4.8 μm in diameter, 700 nm wide, 280 nm thick, and sits on a 280 nm thick SiO_2 pedestal due to the fabrication process. Some surface roughness is visible on the sides of the microring, also due to the fabrication process, and is the main contribution for reducing the resonator's Q-factor from a theoretical value of $\sim 10^6$ (calculated using MEEP) to 5,000. As expected, these Q-factors are higher than those for microresonators fabricated using nanocrystalline diamond.

The microring is made of type IIa diamond which contain almost no impurities (generally <1 ppm) causing them to appear colorless and have the highest thermal conductivity among diamond types. They are very transparent over the full visible range down to 230 nm in the ultraviolet below which absorption occurs due to the diamond's structure rather than impurities. While type IIa diamonds rarely occur in nature (1–2% of natural diamond) due to the higher pressures and longer times required, synthetic diamonds produced through chemical vapor deposition (CVD) typically belong to this category [9]. High-plasma-density CVD is the most common commercial method of diamond growth due to its simplicity, flexibility, and extreme control over impurities, as opposed to high-temperature, high-pressure techniques [3].

The microring was obtained by thinning a monolithic single-crystal diamond membrane. Such single-crystals are ideal for fabricating cavities as their color centre properties are superior to those of emitters in other forms of diamond such as thin films or individual nanodiamonds which are crystalline and introduce undesirable boundaries in the diamond structure [3].

Azimuthal cross-sections of the simulated field profiles of the $TM_{m=40}$ and $TE_{m=40}$ modes are shown for $p = 1, 2, 3, 4$ where m is the azimuthal quantum number and p is the radial quantum number. The shaded black regions presumably represent the diamond region and thus we see that the modes visibly leak, especially vertically below and above the diamond

B. Figure 2

Fig. 2 shows a schematic diagram of the sequence of steps used to fabricate the microring resonator. The method shown is new and allowed for the fabrication of the high-Q diamond microring from Fig. 1. Without its development, resonant enhancement would not have been possible.

The process begins with a purchased and already polished 5 μm thick type IIa single-crystal diamond membrane from Element Six. The membrane was mounted on a 2 μm thick SiO_2 layer then thinned to 280 nm using reactive ion etching (RIE), a method of dry etching whereby the diamond is bombarded by an oxygen plasma in this case. A 500 nm thick silicon nitride layer is then deposited on top of the diamond using plasma-enhanced chemical vapor depositions (PECVD). An electron-beam resist layer consisting of PMMA was then spun onto the silicon nitride before electron beam lithography was used to pattern the ring onto the PMMA layer. The ring pattern was then transferred onto the silicon nitride using SF_6/C_4F_8 chemistry dry etching. Then it was finally transferred to the diamond layer through dry etching in oxygen plasma which got rid of the electron-beam resist layer as well. Then to finish, SF_6/C_4F_8 chemistry is again used to dry etch away the silicon nitride layer which also digs into the SiO_2 layer a bit.

C. Figure 3

Fig. 3 showcases the main result—the coupling of an NV center to the cavity modes and significant subsequent enhancement of the NV center's ZPL without enhancement of the phonon sidebands.

The microring is characterized using PL spectroscopy whereby it is pumped from the top by a 2 mW continuous-wave green laser. A high-resolution scan near the ZPL (634-637.5 nm) shows the ZPL along with a couple of cavity modes. Cavity Q-factors ($Q = \omega_0/\Delta\omega$) are calculated by fitting the mode PL spectra to Lorentzians and relying on the FWHM $\Delta\omega$.

To couple the cavity modes with an NV center's ZPL, xenon gas is condensed on the resonator which increases its effective size resulting in the redshift of its modes. Xenon gas is continuously condensed until the cavity mode is in resonance with an NV center's ZPL at which point it exhibits enhanced PL as shown in Fig. 3(e). Continuing to deposit more gas shows another NV center coming into resonance with a cavity mode at a scan number of 160-190 followed by the second cavity mode coming into resonance with the first NV center's ZPL at a scan number of 215 although the ZPL enhancement here isn't as great, possibly due to the mode's slightly lower Q-factor.

D. Figure 4

Fig. 4 shows the PL decay of a specific NV center's (denoted NV1) ZPL under pulsed excitation from which the PL lifetime is obtained and thus the Purcell enhancement factor is estimated. A 4.75 MHz, 520 nm, 200 μW excitation source with a 28 nm bandwidth is used. Figs. 4(a-b) show a clear enhancement of the NV center's ZPL when in resonance with the cavity mode C_2.

Fig. 4(c) shows the PL decay of the NV center's ZPL on and off-resonance, fitting them to decaying exponentials plus a constant to obtain lifetime measurements on (τ_{C_2}) and off-resonance (τ_0). Reduced lifetimes are observed on-resonance which is desirable as it leads to faster NV center manipulation times. The Purcell enhancement factor is then given by $F = (\tau_0/\tau_{C_2} - 1)/\xi_{\text{ZPL}}$ where $\xi_{\text{ZPL}} = \tau_0/\tau_{\text{ZPL}} \approx 0.03$ is the branching ratio (or ZPL emission ratio). This results in a Purcell factor of $\sim$10 for NV1.

These lifetime measurements are repeated for various detunings from the cavity modes. The NV center PL lifetime is plotted as a function of detuning in Fig. 4(d) showing dips when the NV center is in resonance with either cavity modes. The dip is of similar magnitude for both cavity modes as they seem to be counter-propagating modes with the same p and m however one exhibits a slightly enhanced dip, possibly due to that cavity mode having a higher Q.

E. Figure 5

Fig. 5 repeats the PL lifetime measurements of Fig. 4 but for two other NV centers (denoted NV2 and NV3). While less data is available, the enhancement of the ZPL is still clearly seen. NV2 exhibits a slightly larger reduction in the PL lifetime while NV3 exhibits a mild reduction. It seems that for this experiment, NV2 couples to C_1 while NV3 couples to C_2.

III. Flaws and typos

Does the manuscript have flaws that should prevent its publication? If so, please provide details. Are there any typos that should be fixed before the manuscript goes to print?

The research presented is of high quality and not fundamentally flawed; however, the manuscript has a few shortcomings. I believe it is imperative they be addressed prior to publication. *Nature Photonics* articles are widely read by the scientific community and as per the peer-review policy [10] they should provide sufficient methodological details such that experiments may be reproduced. Furthermore, I have provided some further suggestions for revisions that should improve the manuscript's utility, readability, and quality. Many of these comments are pedagogical in nature and should be easy to address.

A. Shortcomings

1. The authors should provide uncertainties on reported values, especially Q-factors, lifetimes, and Purcell factors. This is especially important for the calculated Q-factors from the Lorentzian fits in Fig. 3(b) and for the Purcell factors calculated using the lifetimes from Fig. 4(c) fits, both of which could have significant associated uncertainties due to the signal-to-noise ratio present. Most software packages provide confidence intervals and uncertainties on parameter estimates from fits.
2. Eqs. (1) and (2) are used to estimate the Purcell factors but Eq. (2) is only valid for ideally positioned and oriented NV centers as the authors state. The NV centers used in this work were incorporated accidentally during diamond growth so there is no guarantee that Eq. (2) is valid. How is its use justified? The fit in Fig. 4(d) suggests that the discrepancy is not large but this point is not addressed at all and raises concerns regarding the estimated Q-factors and Purcell factors.
3. Fig. 3(b): I assume the Lorentzian fits are used to calculate the cavity modes' Q-factors. While the fit for C_2 looks reasonable, the fit for C_1 is less convincing. The right portion of the spectrum is clearly wider than a Lorentzian profile would allow. This suggests that the calculated Q-factor ($Q_1 = 4,300$) is an overestimate. Furthermore, the Q-factor is used to calculate the Purcell enhancement factor which now casts doubts on its estimated value as well.
4. Figs 4(a-b): The authors claim no enhancement of the phonon sidebands in this work. However, in Fig. 4(a) the phonon sideband exhibits a PL inten-

sity of ~400 counts for the uncoupled case, and a PL intensity of ~700 in the coupled case of Fig. 4(b). The authors should clarify whether enhancement of the phonon sidebands occurs here or not. Figs. 5(a-b) also seem to exhibit the same phenomena.

5. Fig. 5(b): The authors should clarify why data exists only for the right side of the profile. I suspect the cavity mode could not be shifted enough however this should be clarified in the text. Otherwise the result looks deceptive as if the authors are trying to hide a highly non-Lorentzian profile on the left side.

B. General suggestions

1. The authors state that the cavity supports four modes. It might be helpful to some readers for the authors to discuss the reason for this. Obviously the radial mode depends on the dimensions of the microring but why are $p > 4$ modes not supported?
2. Can the authors determine which cavity modes were optically excited? Which values of m and p do the observed cavity modes C_1 and C_2 correspond to?
3. I suggest the authors add a short paragraph discussing their reasons for choosing the microring dimensions they did. This is especially insightful in the context of the new fabrication method as other groups may wish to use it as well in which case such a discussion would increase the scholarly value of the manuscript. A particular question I would like to see addressed is why the diamond microring just slightly thinner than the SiO_2 pedestal?
4. The authors should introduce F_i and F_{cav} as Purcell factors when Eq. (1) is first introduced on page 1. Currently this is not done until the end of page 3.

C. Experimental details

1. What was the nitrogen concentration and face orientation of the purchased diamond? (They are provided on the supplier's (Element Six) website for various samples but the reader does not know which sample was used.)
2. The authors should list the models of each Oxford Instruments machine used in the fabrication process.
3. The authors should provide details on the confocal microscope setup beyond just the spot size.
4. What was the bandwidth of the continuous-wave green laser used to pump the microring?
5. The authors should provide details on the cryostat setup used.
6. What purity grade was the xenon gas and from where was it obtained? Probably Paraxair?
7. The authors should report the range of values for the xenon flow rate if possible.
8. The authors should clarify why a non-constant flow rate was used. Is a constant flow rate desirable or not? Did experimental conditions prevent the use of a constant flow rate?
9. What was the source of the supercontinuum employed?
10. The authors should provide details on the photon counter employed.

D. Figure 1

1. Fig. 1(b): The authors should clarify the purpose of the shaded black box. It appears to be the diamond microring azimuthal cross-section region however this is not mentioned.
2. Fig. 1(b): Some short explanation for the mode asymmetry and $\text{TM}_{m=40}$ boundary nodes should be included for audiences not familiar with field profiles in ring resonators or who may have forgotten about electromagnetic boundary conditions.

E. Figure 3

1. Fig. 3(a): The whitespace on the edges of the plot should be cut out: the spectrum should occupy the entire horizontal stretch of the plot.
2. Fig. 3(a): The authors claim that "multiple cavity modes can be observed in the phonon sidebands" however I only see the two labeled. They should clarify what is a cavity mode and what is a phonon sideband mode. Are they all mixed together across the entire spectrum?
3. Fig. 3(b): If C_1 and C_2 represent counter-propagating modes of the same m and p as the authors suggest then why do their profiles look different?
4. Fig. 3(c): The color scale should be included.
5. Fig. 3(c): There appears to be a third cavity mode appearing at higher scan numbers. Can the authors comment on this?
6. Fig. 3(c): NV1 appears to exhibit enhanced ZPL emission twice, presumably once for each cavity mode. The authors should clarify which mode is responsible for which peak.
7. Fig. 3(c): NV2 appears to exhibit enhanced emission at scan numbers near 160-190. Can the authors confirm this? If so, the authors should label the NV2 and NV3 lines here as well.

F. Figure 4

1. Can the authors comment on why coupling NV1 to C_2 results in a lower lifetime? I suspect it is due to C_2 actually having a larger Q-factor due to bad fitting to the C_1 cavity mode in Fig. 3(b) as discussed earlier.
2. Fig. 4(d): Using the uncertainties calculated from the lifetimes, error bars should be added to the data points.

G. Figure 5

1. Why is NV2 coupled to C_1 and NV3 to C_2? I suspect the cavity modes could not be tuned sufficiently far enough but the authors should clarify.
2. Figs. 5(c-d): Again, error bars should be added here using lifetime uncertainties.

H. Typos

1. Three instances of "microdisk" throughout the manuscript should be changed to "microring" for consistency.
2. Fig. 4(c) caption has missing units: "$\Delta\lambda_2 = 0$" should be "$\Delta\lambda_2 = 0\,\text{nm}$".

IV. Originality of conclusions

Are the conclusions original? If not, please provide relevant references to similar work. Which references are most closely related to the manuscript? How do the manuscript's results differ from the work reported in these references?

This NV center-cavity coupling has been demonstrated in previous works and so has the NV center ZPL resonant enhancement, so in this context the conclusions presented are nothing new. However, what is new and interesting is the fabrication method used and that the resonant enhancement has been observed for NV centers in single-crystal diamond, the most promising system for spintronics and quantum technologies that is also amenable to future improvements so this work can be constructively improved on for future benefits.

For an example of a previous demonstration the authors cite Wolters et al. [11] who report on the coupling of an NV center's ZPL in a nanodiamond to a photonic crystal cavity with an observed Purcell enhancement of 12.1 and a Q-factor of $Q = 603$ on resonance. However, NV centers in nanodiamond have poor spectral properties due to nanodiamond's crystalline structure which is why coupling to a single-crystal diamond cavity is desirable, setting the current work apart from this one before it.

An interesting note is that a year after this work, the authors Faraon et al. [12] are able to couple individual NV centers to photonic crystal cavities in monocrystalline diamond. Due to the smaller mode volume (and thus larger coupling rates) of photonic crystal cavities they observe a ZPL Purcell enhancement of $\sim$70 and claim the photonic crystal cavaties to be superior to the microring structure employed here.

Furthermore, there have been multiple attempts by previous groups to fabricate resonators from single-crystal diamond; however, due to the difficulty of accurately dry etching diamond [3], none of the resonators produced had high enough Q-factors for resonant enhancement of an NV center's ZPL. The authors cite an attempt to fabricate single-crystal diamond nanobeam cavities by Babinec et al. [13] whose fabrication methods were not accurate enough to produce a high-Q cavity using focused ion beams due to their Gaussian profile. They conclude by suggesting that lithography and reactive ion etching techniques may be more successful and indeed they were, as shown in the current work. More recent work by Hausmann et al. [14] make use of the fabrication method employed in this work to demonstrate the successful in coupling an NV center to a single-crystal diamond nanobeam cavity.

V. Significance of findings

In your opinion, what is the significance of the findings? Do you feel that the results presented are only of interest to specialists, or would they appeal to the optics community in general?

The immediate results and fabrication method may be of limited interest outside the quantum and diamond photonics communities due to their lack of widespread utility; however, subsequent developments that build upon the results presented in this work may result in a host of other applications that would be of considerable interest not only to the optics community at large, but also the quantum computing and life sciences communities to name a couple.

The microring resonator may be used to construct a cavity-optomechanical wavelength converter with applications in classical and quantum communication [15]. Further enhancement of the spontaneous emission ratio into the ZPL will allow for the generation of a large number of indistinguishable photons [16] which is essential towards the implementation of linear optics quantum computing [17]. It may also be used in magnetometry as a nanoscale sensor for detecting and imaging weak magnetic fields as the magnetic field sensitivity of these devices can be significantly improved by enhancement of the ZPL emission ratio [18].

An improvement of the diamond microring may be possible from recent studies of surface-induced noise of shallow NV centers in diamond [19] which could allow for the passivation of the microring structures enhancing their Q-factor and thus their Purcell factor as surface roughness was the largest contribution of Q-factor degradation.

Of course, there are a few drawbacks present in this work that are worth discussing as multiple promising solutions have surfaced in recent years.

One drawback is the bottom-up approach of diamond growth relying on random placements and orientations of NV centers which means that devices must be fabricated with some trial-and-error. For optimum coupling and maximum Purcell enhancements, the NV centers should be placed at the field maxima so it could take multiple fabrication attempts to obtain a suitable cavity. The authors do not report how many fabrication trials were necessary to obtain a suitable microring. Very recently in 2016 there has been some significant progress on the implantation of NV centers; patterned formation of highly coherent NV centers has been achieved [20] and so has implantation into the mode maximum of diamond photonic crystal nanocavities [21]. While these techniques are still in their infancy and rely on implantation during diamond growth and thus and do work with single-crystal diamond yet, they are a promising step in the right direction.

Another drawback is that tuning of the cavity mode to the ZPL resonance must be done by gas condensation in a cryostat which can only redshift the cavity mode and makes it hard to stabilize the cavity to the resonance. One promising avenue in this regard is recent work by Johnson et al. [22]. Instead of redshifting the cavity mode using gas condensation, they demonstrate *in situ* tuning of an open cavity's resonance using piezoelectric actuators while still observing the Purcell enhancement of the NV center's ZPL.

There is a question of scalability as well, especially when NV center production relies on "accidents" and are randomly positioned and oriented. A step towards more scalable quantum networks was made by Choy et al. in 2011. They demonstrated enhancement of the spontaneous emission rate of single NV centres in ordered arrays of plasmonic apertures and report Purcell factors of 6.4 and 3.2 in certain samples with radiative lifetimes as low as 2.4 ns in other samples. However, their main advantage is that they controllably embed NV centres into metallic nanostructures and thus have the ability to create arrays of NV centers which is more promising for building scalable quantum networks.

VI. Suitability of references

Does this manuscript reference previous literature appropriately? Would the article benefit from including additional references? If so, make suggestions.

In general the manuscript references previous literature appropriately however it could benefit from additional references in a few areas where no sources are cited or where references would be of interest to readers, especially non-specialists. A list of suggestions follows.

1. The authors cite Neilson and Chuang which is a several-hundred page text and so page numbers should be included in the citations.
2. I recommend citing the 2007 *Scientific American* article on spintronics by Awschalom et al. [7] in the introduction to motivate the use on NV centers. It is well-written and accessible to a wide audience wishing to learn more about why everyone is interested in NV centers.
3. I recommend citing the 2006 *Phys. Rev. A* article on the electronic structure and dynamics of NV centers [23] for readers wishing to know more about NV centers. The authors should mention that despite it being a popular system of study, there are still many unresolved issues concerning the NV center. (The authoritative review on NV centers by Doherty et al. [6] did not appear until 2013.)
4. A reference should be provided for readers interested in how Eqs. (1) and (2) were obtained or derived. Citing Purcell's original paper [24] out of tradition is fine but does not provide sufficient background for the reader to convince themselves that Eqs. (1) and (2) are correct. Santori could have cited his own book [25] here.
5. The authors should cite previous work by Fairchild et al. [26] in fabricating microrings from single-crystal diamond membranes. Comparison of fabrication techniques could be used to highlight the strengths of the authors' new technique.

VII. Final verdict

If you recommend publication, please outline, in a paragraph or so, what you consider to be the outstanding features of the paper. Include suggestions for any revisions that you believe the manuscript would benefit from. Alternatively, if you would not recommend publication in the journal your paper has already been published in, explain why, list the issues the authors should address, and/or suggest an alternative journal that would be more suitable.

The quality of the results presented is outstanding and should be of significant interest in the photonics community with possible future benefits in other fields as well. Of particular note is the Purcell enhancement of the spontaneous emission ratio into the ZPL by a factor of ∼10 without enhancing emission into the phonon sidebands. This is a critical step towards the utilization of NV centers as spin qubits and nanoscale sensors in spintronics and quantum technologies as a high ZPL emission ratio >99.5% absolutely crucial for these applications. Furthermore, the authors employ a new method for fabricating structures out of single-crystal diamond that allowed for the creation of the high-Q cavity employed which will be of interest to other groups working on fabricating cavities and other structures out of single-crystal diamond.

However, the manuscript contains a few shortcomings that I believe must be addressed to bring the manuscript's quality up to the standards of *Nature Photonics*. I recommend the manuscript for publication however it should be extensively revised to address the shortcomings and suggestions listed in sections II and VI, and undergo a second round of peer-review.

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