

Peer Review Information

Journal: Communications Physics

Manuscript title: Geometric entanglement of a photon and spin qubits in diamond

Corresponding author name(s): Hideo Kosaka

Reviewer comments & decisions:

Reviewer comments, first version:
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Reviewer #1 (Remarks to the Author: Overall significance):

The authors report the experimental demonstration of spin-photon entanglement between the spin of an NV center in diamond and a spontaneously emitted optical photon. The key feature of this experiment compared to prior works that demonstrated spin-photon entanglement in this system is that the present work uses polarized microwaves to manipulate the spin. This capability lifts the need for an applied magnetic field, which opens up the possibility of transduction between the NV spin and other types of microwave-domain qubits that react negatively to strong magnetic fields, such as superconducting qubits. It also enhances the controllability of the spin, enabling the implementation of holonomic gates that are robust to certain types of control errors. Finally, it allows the logical spin states to be degenerate, which in turn eliminates dynamical phases in the resulting entangled spin-photon state. This helps to avoid complications that otherwise arise when confirming the existence of entanglement as well as when using this entanglement for quantum information applications such as quantum communication.

The work is interesting and timely. Achieving high-precision control over the spin in a spin-photon interface has been a major challenge in using these systems for various applications in quantum information technologies. The present work constitutes an important step toward overcoming this challenge. While polarized microwave control has been demonstrated in previous works (including by the authors), it is by no means a standard technique, and to my knowledge it has not been used in demonstrating spin-photon entanglement before. In addition to microwave-to-optical transduction, the techniques introduced in this work could also have important repercussions for the deterministic

generation of multiphoton entangled resource states from quantum emitters (see below).

Reviewer #1 (Remarks to the Author: Impact):

I think the scope and importance of the work makes it appropriate for publication in Nature Communications.

Reviewer #1 (Remarks to the Author: Strength of the claims):

The results presented in this work are convincing, and I do not see a need for further experimental work. However, the impact of the work would benefit from further elaboration, specifically on the following points:

1. On lines 168-172, it is suggested that the techniques demonstrated in this work could be used to facilitate quantum state transfer from the microwave to the optical domain. In order for transduction to be useful, it is necessary to be able to transfer unknown quantum states from one domain to the other. It is not clear to me how an unknown state stored on a microwave photon can be mapped onto an optical photon using the methods described in this paper. Can the authors elaborate on this point?
2. In addition to microwave to optical transduction, another important application of this work could be in the generation of multiphoton entangled resource states for quantum networks and computation. In Ref. [Phys. Rev. Lett. 103, 113602 (2009)], it was shown that linear cluster states can be produced deterministically from a single quantum emitter provided one has the ability to perform fast spin rotations on the emitter. While the original proposal focused on self-assembled quantum dots, there are also more recent proposals that focus on color centers also [Phys. Rev. X 7, 041023 (2017); PNAS 114, 11362 (2017); Phys. Rev. B 98, 085303 (2018)]. In this case, the lack of high-fidelity spin control has been a limitation that the present work could potentially circumvent.

Reviewer #1 (Remarks to the Author: Reproducibility):

The paper is well written overall. However, there are a few points that would benefit from further clarification:

1. The microwave GRAPE pulses that were used to generate the results in Fig. 2 should themselves be shown.
2. On lines 99-102, it is mentioned that an electro-optic modulator is used to suppress the reflected light, but that this is not sufficient, and so the polarization of the optical pump is also chosen orthogonal

to the measurement basis. How much reflected light is suppressed by the modulator? How much reflection remains after the polarizations of excitation and measurement are made orthogonal to each other?

3. On line 124-125, it states that the spin measurement error can be suppressed by “high-fidelity manipulation with a stronger microwave”. It is not clear what is meant here. Are the authors proposing to use additional pulse-shaping techniques beyond GRAPE?

4. I also noticed a few typos that could create confusion:

Line 80: “projects the arbitrary state” should read “rotates the arbitrary state” since this is a unitary process.

Line: 109: “Figure 1c shows” should read “Figure 1e shows”.

Line 111: “detection merely occurs” should read “detection rarely occurs”.

Reviewer #2 (Remarks to the Author: Overall significance):

The manuscript by Sekiguchi and co-authors describes the achievement of spin-photon entanglement in the nitrogen vacancy center. The particular form of entanglement (circularly polarized photon emitted to the final ground state spin after emission) was previously achieved by Togan et al. (Nature 466 2010). However, Togan's experiments were performed at non-zero magnetic field, where the ground state spin states are separated in energy, while the present manuscript achieves entanglement at zero magnetic field (degenerate lambda system). Under a degenerate lambda system, there are certain geometric aspects related to the spin, which the authors previously demonstrated (Ref. 1, 2). Overall, I think the novelty of the result is basically just the demonstration in Figure 3b of photon correlations.

I should note that Fig. 4 is very similar to:

<https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.119.063601>
which is not cited and should be cited.

So the question is how important is the degenerate aspect of the ground state for the final spin:

"The magnetic field lifts the degeneracy to make the light polarization time-dependent although quantum eraser technique erases the frequency information, while the degeneracy remains under a

zero magnetic field in our case to keep the light polarization time-independent as well as the geometric spin qubit state."

Could the author explain more clearly why the polarization of the "light" (emitted photon) changes with time? "as well as the geometric spin qubit state" is tacked on, can the authors explicitly explain the significance? At the end of the day, I think the distinction from Togan is not very convincing.

Other comments:

1) For Fig. 4, could the authors more clearly explain where the attenuation coefficient comes from? It seems to be cosine(53 degrees) for the horizontal component of the E-field (assuming horizontal NV axis). The vertical component, however, would not be attenuated if I understand correctly. Does this rotate the polarization then? The section is not well explained and a figure in the SI would help.

2) The authors should define "A2 state fidelity" in Fig. 3c. Does the angle of the strain affect different cases of the photon polarization differently?

Reviewer #2 (Remarks to the Author: Impact):

I think as currently written the paper is most suitable for Communications Physics. Without a supplementary information and very cursory explanation of the experiment, I was basically relying on Togan et al. to even understand this paper.

For example, the most important Fig. 3b is introduced as:

"The obtained results are in good agreement with the entangled state written as..."
 $|+1\rangle_p$ and $|+\rangle_p$, etc. for the photon are not defined (it's drawn in Fig. 1b).

I'm doubtful the paper will influence thinking on the field. However, I should say that fidelity of entanglement >90% is indeed rather good and thus is a strong point of the paper.

Reviewer #2 (Remarks to the Author: Strength of the claims):

1. The estimated fidelity of 91.2% - the main selling point of this paper - should be unpacked in a Supplementary Information. The authors should give the raw photon counts (hi/low) for each of the cases (for the 117, 130, etc. events) and then detail step by step the background corrections they made to find an estimated fidelity of 91.2%. There's a comment about the error due to misalignment (7%), spin phase rotation (2%), etc. This needs to be unpacked in the supplementary information.

Reviewer #2 (Remarks to the Author: Reproducibility):

There is fairly minimal detail provided on the experimental methods or data analysis without a supplementary information. Usually papers in this field contain detailed experimental methods where implementation of key technical aspects (such as the spin readout conditioned on photon detection) is elaborated on to help reproducibility.

Reviewer #3 (Remarks to the Author: Overall significance):

In this paper, Sekiguchi et al report on entanglement demonstration between spin and photon polarization degrees of freedom of an NV center qubit in diamond. They claim to observe entangled spin-photon emission with >90% fidelity due to geometric resilience of the qubit degrees of freedom to noise.

As such, earlier work has shown spin-photon entanglement (Ref. 26) and remote spin entanglement (Ref. 27) with the same qubit system, thus the main novel result in this paper is the claim of geometric entanglement. Ref. 26 is probably closest to this paper as the same states and nearly similar experimental protocol is used by the authors.

Reviewer #3 (Remarks to the Author: Impact):

As noted below, I find the paper not recommended for publication in Nature journals.

Reviewer #3 (Remarks to the Author: Strength of the claims):

1. Firstly, we can ask: what do the authors mean by geometric entanglement? After all, the Bell state of two qubits remains invariant under basis change, therefore it is not surprising that if the spin/photon qubits is measured in a subspace spanned by the basis states $\{+1, -1\}$ that entanglement would be preserved. So the mere demonstration of correlations, even quantum correlations, is not surprising or unexpected. Do the authors mean that the entanglement is geometric because they carry out manipulations in the two-qubit subspace of all possible spin-photon states in a holonomic fashion? I think not, because they only manipulate the spin in geometric fashion and measure its correlation with certain photon states, after post-selection. They do not carry out any quantum gates between the two qubits (here the spin and photon).

2. Furthermore, while the fidelity of the spin-photon state reported appears fairly high, this could be entirely because they operate at zero magnetic field, where the frequency and other degrees of

freedom of the spontaneous emission photon wavepacket from A2 to the two ground states $\{+1, -1\}$ are well overlapped. As noted by the authors, the fidelity in the Togan et al paper was increased by a quantum eraser technique, and operating at zero magnetic field is similar to that approach. So I am not convinced at all that the fidelity is increased due to resilience of the geometric phase, as claimed by the authors.

3. Another issue is that when the authors prepare coherent superpositions of the basis states, then due to small magnetic fields and the nitrogen hyperfine interactions I would expect the state should evolve in phase between preparation and photon measurement, and certainly between one run of the experiment and the next. The resulting photon measurement should take this into account, either with a geometric spin-echo to zero out the phase evolution or clocking the phase evolution with high time resolution. The authors don't seem to discuss this step at all.

4. I also find this statement in page 8 troubling: "The scheme does not require any of the mode matching in time, frequency, and space required in the previous scheme for photon indistinguishability with the Bell measurement of photons using a beam splitter in the middle[51–55]". I think this statement is completely incorrect as photon indistinguishability is a requirement for Bell measurement, and none of the references given by the authors indicates otherwise.

Given all these issues and other ones noted below, I therefore find the paper not recommended for publication in Nature journals.

Reviewer #3 (Remarks to the Author: Reproducibility):

I found the paper somewhat short on important experimental details necessary for others to reproduce the work: for example,

1. how the fidelity is defined for state preparation and the actual spin-photon entanglement and what are the error bars?
2. How is the data in Fig. 2 taken, with preparation and measurement steps?
3. Are only post-selected events where the NV is in the correct charge and spin state kept or are the events that are shown not post-selected?
4. The data in Fig. 3 shows the parity measurements between states prepared in different axes, e.g. $|+1s, +1p\rangle$, $|+s, +p\rangle$, $|+is, +ip\rangle$ etc. But they don't measure in other bases to the preparation axes, e.g. $|+1s, +ip\rangle$ or $|+s, -1p\rangle$. This would be useful to know the mixedness of the final state that is prepared.
5. Is spin and photon state measurement carried out in quantum projective fashion, and if so what are the measurement fidelities?

Author rebuttal, first version:
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**Response to referee comments on “Geometric entanglement of a photon and spin qubits in diamond”
(GUIDEDOA-21-00074)**

Reviewer #1 (Remarks to the Author: Overall significance):

The authors report the experimental demonstration of spin-photon entanglement between the spin of an NV center in diamond and a spontaneously emitted optical photon. The key feature of this experiment compared to prior works that demonstrated spin-photon entanglement in this system is that the present work uses polarized microwaves to manipulate the spin. This capability lifts the need for an applied magnetic field, which opens up the possibility of transduction between the NV spin and other types of microwave-domain qubits that react negatively to strong magnetic fields, such as superconducting qubits. It also enhances the controllability of the spin, enabling the implementation of holonomic gates that are robust to certain types of control errors. Finally, it allows the logical spin states to be degenerate, which in turn eliminates dynamical phases in the resulting entangled spin-photon state. This helps to avoid complications that otherwise arise when confirming the existence of entanglement as well as when using this entanglement for quantum information applications such as quantum communication.

The work is interesting and timely. Achieving high-precision control over the spin in a spin-photon interface has been a major challenge in using these systems for various applications in quantum information technologies. The present work constitutes an important step toward overcoming this challenge. While polarized microwave control has been demonstrated in previous works (including by the authors), it is by no means a standard technique, and to my knowledge it has not been used in demonstrating spin-photon entanglement before. In addition to microwave-to-optical transduction, the techniques introduced in this work could also have important repercussions for the deterministic generation of multiphoton entangled resource states from quantum emitters (see below).

Reviewer #1 (Remarks to the Author: Impact):

I think the scope and importance of the work makes it appropriate for publication in Nature Communications.

Reviewer #1 (Remarks to the Author: Strength of the claims):

The results presented in this work are convincing, and I do not see a need for further experimental work. However, the impact of the work would benefit from further elaboration, specifically on the following points:

Response:

We appreciate the reviewer's understanding of the significance of using microwave polarization to show the possibility of microwave-to-optical transduction with superconducting qubits and generating the ideal entanglement without the dynamic phase.

1. On lines 168-172, it is suggested that the techniques demonstrated in this work could be used to facilitate quantum state transfer from the microwave to the optical domain. In order for transduction to be useful, it is necessary to be able to transfer unknown quantum states from one domain to the other. It is not clear to me how an unknown state stored on a microwave photon can be mapped onto an optical photon using the methods described in this paper. Can the authors elaborate on this point?

Response:

Since the quantum state transfer from the microwave to optical domain is one of the important applications of our work, we added it to the supplementary information.

2. In addition to microwave to optical transduction, another important application of this work could be in the generation of multiphoton entangled resource states for quantum networks and computation. In Ref. [Phys. Rev. Lett. 103, 113602 (2009)], it was shown that linear cluster states can be produced deterministically from a single quantum emitter provided one has the ability to perform fast spin rotations on the emitter. While the original proposal focused on self-assembled quantum dots, there are also more recent proposals that focus on color centers also [Phys. Rev. X 7, 041023 (2017); PNAS 114, 11362 (2017); Phys. Rev. B 98, 085303 (2018)]. In this case, the lack of high-fidelity spin control has been a limitation that the present work could potentially circumvent.

Response:

We thank the reviewer for introducing us to the application as a multi-photon entanglement source. We have cited it in the main text although we have not initially envisioned the cluster state, which is not covered in this paper because it is difficult to perform immediately.

Reviewer #1 (Remarks to the Author: Reproducibility):

The paper is well written overall. However, there are a few points that would benefit from further clarification:

1. The microwave GRAPE pulses that were used to generate the results in Fig. 2 should themselves be shown.

Response:

Since we thought that the specific GRAPE pulses used in our experiment should be shown, we added them in the supplementary information.

2. On lines 99-102, it is mentioned that an electro-optic modulator is used to suppress the reflected light, but that this is not sufficient, and so the polarization of the optical pump is also chosen orthogonal to

the measurement basis. How much reflected light is suppressed by the modulator? How much reflection remains after the polarizations of excitation and measurement are made orthogonal to each other?

Response:

The extinction ratio of two consecutive EOM is $1e6$. In addition to this, the polarization of the excitation light and the polarization of the entangled photon are orthogonal, which further increases the extinction ratio by a factor of $1e3$. However, due to the finite bandwidth of the EOM, about 10% of the detected photons originate in the tail of the excitation pulse, as shown in Figure 2e. This could be reduced by applying an AR coating to the diamond or by increasing the excitation and collection efficiency of the optical system.

3. On line 124-125, it states that the spin measurement error can be suppressed by “high-fidelity manipulation with a stronger microwave”. It is not clear what is meant here. Are the authors proposing to use additional pulse-shaping techniques beyond GRAPE?

Response:

Our GARPE algorithm searches for optimized pulses under the condition that the maximum Rabi frequency of the pulse is smaller than the experimentally achievable magnitude. Therefore, if experimentally larger Rabi frequencies can be achieved by improving the microwave antenna, the search range would be extended, and thus higher-fidelity pulses would be found.

4. I also noticed a few typos that could create confusion:

Line 80: “projects the arbitrary state” should read “rotates the arbitrary state” since this is a unitary process.

Response:

We thank the reviewer for pointing out the wrong choice of word. We have corrected “projects” to “rotates”.

Line 109: “Figure 1c shows” should read “Figure 1e shows”.

Response:

We thank the reviewer for pointing out the typo. We have corrected the figure number.

Line 111: “detection merely occurs” should read “detection rarely occurs”.

Response:

We thank the reviewer for pointing out the typo. We have corrected “merely” to “rarely”.

Reviewer #2 (Remarks to the Author: Overall significance):

The manuscript by Sekiguchi and co-authors describes the achievement of spin-photon entanglement in the nitrogen vacancy center. The particular form of entanglement (circularly polarized photon emitted to the final ground state spin after emission) was previously achieved by Togan et al. (Nature 466 2010). However, Togan's experiments were performed at non-zero magnetic field, where the ground state spin states are separated in energy, while the present manuscript achieves entanglement at zero magnetic field (degenerate lambda system). Under a degenerate lambda system, there are certain geometric aspects related to the spin, which the authors previously demonstrated (Ref. 1, 2). Overall, I think the novelty of the result is basically just the demonstration in Figure 3b of photon correlations.

Response:

We thank the reviewer's understanding of the novelty of the correlation measurements between the degenerate spin and photon under a zero magnetic field.

I should note that Fig. 4 is very similar to:

<https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.119.063601>

which is not cited and should be cited.

Response:

We thank the reviewer for informing the related paper. We have cited it in the main text because they are similar in the way that they show phase exchange between microwave and light, although they do not use polarization as our work.

So the question is how important is the degenerate aspect of the ground state for the final spin:

"The magnetic field lifts the degeneracy to make the light polarization time-dependent although quantum eraser technique erases the frequency information, while the degeneracy remains under a zero magnetic field in our case to keep the light polarization time-independent as well as the geometric spin qubit state."

Could the author explain more clearly why the polarization of the "light" (emitted photon) changes with time? "as well as the geometric spin qubit state" is tacked on, can the authors explicitly explain the significance? At the end of the day, I think the distinction from Togan is not very convincing.

Response:

Since the relaxation time of the electron orbital in the NV center is about 12 ns, the phase rotation due to the ground state spin splitting at 122 MHz, as in Togan's experiment, is non-negligible. This is because the ground state spin begins to rotate as soon as the orbital emits a photon. This means that the phase of the entangled state depends on the timing of the emission even under the rotating frame at 122 MHz. Therefore, when we consider the spin phase to be fixed, the wave function of the emitted photon has a time-dependent polarization with a spread of the relaxation time.

If we try to generate remote spin entanglement through a two-photon interference, the two photons must have exactly the same time-dependence. In addition, the detection time of the photon after the interference must be

known precisely, even at long-distant location. Furthermore, the scheme of transferring a spin-entangled photon to a distant spin (Ref. 5, 6) does not apply since the scheme does not allow fast detection of success event required for quantum erasure technique. Therefore, the non-degenerate setup makes the experiment unrealistic.

Other comments:

1) For Fig. 4, could the authors more clearly explain where the attenuation coefficient comes from? It seems to be $\cos(53 \text{ degrees})$ for the horizontal component of the E-field (assuming horizontal NV axis). The vertical component, however, would not be attenuated if I understand correctly. Does this rotate the polarization then? The section is not well explained and a figure in the SI would help.

Response:

Since the explanation of the polarization distortion in Fig. 4 was not sufficient, we have added it to the Supplemental Information including the figure.

2) The authors should define "A₂ state fidelity" in Fig. 3c. Does the angle of the strain affect different cases of the photon polarization differently?

Response:

Based on reviewer's recommendation, we have added the definition of A₂ state fidelity. The elements of the A₂ state change depending on the angle of the non-axial strain, but the fidelity of the maximally entangled state itself does not change.

Reviewer #2 (Remarks to the Author: Impact):

I think as currently written the paper is most suitable for Communications Physics. Without a supplementary information and very cursory explanation of the experiment, I was basically relying on Togan et al. to even understand this paper.

Response:

Since the experimental description was not sufficient, we have added the methods.

For example, the most important Fig. 3b is introduced as: "The obtained results are in good agreement with the entangled state written as..." $|+1\rangle_p$ and $|+\rangle_p$, etc. for the photon are not defined (it's drawn in Fig. 1b).

Response:

Since the definition of the photon state was not written, we have added it to the main text.

I'm doubtful the paper will influence thinking on the field. However, I should say that fidelity of entanglement $>90\%$ is indeed rather good and thus is a strong point of the paper.

Reviewer #2 (Remarks to the Author: Strength of the claims):

1. The estimated fidelity of 91.2% - the main selling point of this paper - should be unpacked in a Supplementary Information. The authors should give the raw photon counts (hi/low) for each of the cases (for the 117, 130, etc. events) and then detail step by step the background corrections they made to find an estimated fidelity of 91.2%. There's a comment about the error due to misalignment (7%), spin phase rotation (2%), etc. This needs to be unpacked in the supplementary information.

Response:

Although the fidelity in the previous version showed the fidelity in the correlations measured on the x, y, z axis, we changed it to match the fidelity of Togan et al. We have also written a description of how to calculate the fidelity, how to handle background, and the comments on the main errors in the supplementary information.

Reviewer #2 (Remarks to the Author: Reproducibility):

There is fairly minimal detail provided on the experimental methods or data analysis without a supplementary information. Usually papers in this field contain detailed experimental methods where implementation of key technical aspects (such as the spin readout conditioned on photon detection) is elaborated on to help reproducibility.

Response:

Since the technical aspects and data analysis were not sufficient, we have added them in the Supplemental Information.

Reviewer #3 (Remarks to the Author: Overall significance):

In this paper, Sekiguchi et al report on entanglement demonstration between spin and photon polarization degrees of freedom of an NV center qubit in diamond. They claim to observe entangled spin-photon emission with >90% fidelity due to geometric resilience of the qubit degrees of freedom to noise.

As such, earlier work has shown spin-photon entanglement (Ref. 26) and remote spin entanglement (Ref. 27) with the same qubit system, thus the main novel result in this paper is the claim of geometric entanglement. Ref. 26 is probably closest to this paper as the same states and nearly similar experimental protocol is used by the authors.

Response:

We are sorry to have confused you. We are not claiming that the geometric nature of geometric entanglement makes the spin-photon entangled high fidelity. What we mean by “geometric” is that the qubit system has a geometric degree of freedom, likewise the polarization of light, without the intentional inclusion of dynamic phase.

Reviewer #3 (Remarks to the Author: Impact):

As noted below, I find the paper not recommended for publication in Nature journals.

Reviewer #3 (Remarks to the Author: Strength of the claims):

1. Firstly, we can ask: what do the authors mean by geometric entanglement? After all, the Bell state of two qubits remains invariant under basis change, therefore it is not surprising that if the spin/photon qubits is measured in a subspace spanned by the basis states $\{+1, -1\}$ that entanglement would be preserved. So the mere demonstration of correlations, even quantum correlations, is not surprising or unexpected. Do the authors mean that the entanglement is geometric because they carry out manipulations in the two-qubit subspace of all possible spin-

photon states in a holonomic fashion? I think not, because they only manipulate the spin in geometric fashion and measure its correlation with certain photon states, after post-selection. They do not carry out any quantum gates between the two qubits (here the spin and photon).

Response:

We are sorry that we have confused the reviewer by the terminology “geometric”. “Geometric entanglement” refers here to entanglement due to degrees of freedom of a geometric nature where no dynamic phase is introduced. Although a complete energy degeneracy cannot be experimentally achieved, our demonstration is completely different from the Ref. 26, which intentionally lift the energy degeneracy to incorporate dynamic phases. With energy splitting, the entangled state becomes time-dependent and precise time synchronization is required remotely, which is a major difference from our work. Although we also use geometric manipulations to generate and measure geometric entanglement, this is not the essence of our achievements. The essence of this paper is that we can reveal the geometric nature (freedom) of the photon at a completely zero magnetic field. Additional important point is that we can avoid the application of a magnetic field, which negatively acts on microwave-domain qubits such as superconducting qubits. This enables transfer of unknown quantum states from microwave-domain qubits to optical-domain qubits or vice versa.

2. Furthermore, while the fidelity of the spin-photon state reported appears fairly high, this could be entirely because they operate at zero magnetic field, where the frequency and other degrees of freedom of the spontaneous emission photon wavepacket from A2 to the two ground states $\{+1, -1\}$ are well overlapped. As noted by the authors, the fidelity in the Togan et al paper was increased by a quantum eraser technique, and operating at zero magnetic field is similar to that approach. So I am not convinced at all that the fidelity is increased due to resilience of the geometric phase, as claimed by the authors.

Response:

Again, we are not claiming that the geometric phase increases the fidelity of the entanglement. On the other hand, it is correct that experimentally it is important that the system degenerates under a zero magnetic field. It should be noted, however, that the zero magnetic field intentionally eliminates the dynamic phase, which in principle creates a time-independent entanglement. Hence, there is no need for precise time synchronization between remote locations.

3. Another issue is that when the authors prepare coherent superpositions of the basis states, then due to small magnetic fields and the nitrogen hyperfine interactions I would expect the state should evolve in phase between preparation and photon measurement, and certainly between one run of the experiment and the next. The resulting photon measurement should take this into account, either with a geometric spin-echo to zero out the phase evolution or clocking the phase evolution with high time resolution. The authors don't seem to discuss this step at all.

Response:

As the reviewer pointed out, the phase rotation starts from the timing of relaxation due to the residual external magnetic field or the nitrogen hyperfine interaction. However, the relaxation time of about 12ns is relatively shorter than the phase rotation, so even without measuring the timing of the photon emission, the fidelity immediately after entanglement generation is relatively high. When transmitting entangled photons over long distances, it takes time for the photons to be measured or interact with other qubits, but as long as the photon state is not destroyed by the transmission, the entangled state can be maintained by local dynamical decoupling of the spins.

4. I also find this statement in page 8 troubling: "The scheme does not require any of the mode matching in time, frequency, and space required in the previous scheme for photon indistinguishability with the Bell measurement of photons using a beam splitter in the middle[51–55]". I think this statement is completely incorrect as photon indistinguishability is a requirement for Bell measurement, and none of the references given by the authors indicates otherwise.

Response:

We are sorry for misleading you. We claim that our method is different from Refs. 51-55. The scheme we are proposing is a quantum state transfer of the emitted photon to the spin of the next node. That quantum state transfer does not depend on mode matching, as it reduces efficiency instead of fidelity with respect to spatial, timing, and frequency deviations. Of course, if the deviation is too large, the efficiency will decrease but it is not realistic. Since we have maybe overstated the mode matching issue, we rewrote it conservatively.

Given all these issues and other ones noted below, I therefore find the paper not recommended for publication in Nature journals.

Reviewer #3 (Remarks to the Author: Reproducibility):

I found the paper somewhat short on important experimental details necessary for others to reproduce the work: for example,

1. how the fidelity is defined for state preparation and the actual spin-photon entanglement and what are the error bars?

Response:

Since the definition of fidelity was not written in detail, we have added it to the supplementary information.

2. How is the data in Fig. 2 taken, with preparation and measurement steps?

Response:

Since there was not much explanation of the experiment on Fig. 2, we have added it to the supplementary information.

3. Are only post-selected events where the NV is in the correct charge and spin state kept or are the events that are shown not post-selected?

Response:

The charge state of the NV center is confirmed to be negative by the detection of the emitted ZPL photon, so the negative state is post-selected. The spin measurement result is conditioned on the ZPL photon detection and subsequently post-selected upon probabilistic PSB photon detection.

4. The data in Fig. 3 shows the parity measurements between states prepared in different axes, e.g. $|+1s, +1p\rangle$, $|+s, +p\rangle$, $|+is, +ip\rangle$ etc. But they don't measure in other bases to the preparation axes, e.g.

$|+1s, +ip\rangle$ or $|+s, -1p\rangle$. This would be useful to know the mixedness of the final state that is prepared.

Response:

Reviewer-recommended measurements such as $|+1s, +ip\rangle$ or $|+s, -1p\rangle$ were omitted to save experimental time.

5. Is spin and photon state measurement carried out in quantum projective fashion, and if so what are the measurement fidelities?

Response:

“Projection” is a writing error, and “Transfer” is correct. Hence the reviewer's question is not applicable. We have fixed the writing error.

Reviewer comments, second version:

Reviewer #1

(Remarks to the Author: Overall significance):

The authors have thoroughly addressed all my comments and questions. I recommend publication of the manuscript in its current form.

Reviewer #2

(Remarks to the Author: Overall significance):

In the referee response, the authors have done a better job in explaining the ramifications of using degenerate spin states in spin-photon entanglement protocol. The main advantage is that one would not have to keep track of the random arrival time of the photon due to the 12 ns excited state lifetime of the NV center, which would induce a dynamical phase in the spin state used in subsequent entanglement operations. This is an important, but incremental, advance toward demonstrating the full protocol sketched in Fig. 1a. However, significant gaps in efficiency in each of the steps seem render the vision of Fig. 1a practically out-of-reach in my opinion.

(Remarks to the Author: Impact):

I still standby my initial assessment that the manuscript is most appropriate for Communication Physics. I would argue that outside of Fig. 3 (which is novel), the other experimental figures are basic experimental checks. Further, I think the present paper does not make significant progress towards quantum transduction of microwave to optical polarization, since the experiment is far from using single microwave photons. Fig. 4 is mainly verifying bright state/dark state selection rules which has been previously demonstrated. The distinction between two microwave polarizations generated from two crossed wires versus the two microwave frequencies in Lekavicius et al. is very little until you get to the single photon level.

(Remarks to the Author: Strength of the claims):

The claim that spin-photon entanglement has been generated is convincing (and previously seen in

Togan et al. on which this experiment is based). The paper does not make any other claims.

(Remarks to the Author: Reproducibility):

The authors have now done a better job in explaining the experiment and data analysis in the supplementary info and methods. The description of how the fidelity of the entangled state is calculated is appreciated.