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- **Review synthesis:** summary of the reviewer reports provided by the editors.
- **Editorial recommendation:** personalised evaluation and recommendation from all 3 journals.
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- **Open research evaluation:** advice for adhering to best reproducibility practices.

About the editorial process

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- **Novelty** in comparison to prior publications;
- **Likely audience** of researchers in terms of broad fields of study and size;
- **Potential impact** of the study on the immediate or wider research field;
- **Evidence** for the claims and whether additional experiments or analyses could feasibly strengthen the evidence;
- **Methodological detail** and whether the manuscript is reproducible as written;
- Appropriateness of the literature review.

Editorial evaluation of reviews



Your editorial team discussed the potential suitability of your manuscript for each of the participating journals. They then discussed the revisions necessary in order for the work to be published, keeping each journal's specific editorial criteria in mind.

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If you have any questions about review portability, please contact our editorial office:

guidedoa@nature.com

Manuscript details

Tracking number		Submission date		Decision date	
GUIDEDOA-21-00074		18 March 2021		20 May 2021	
Title	Geometric entanglement of a photon and spin qubits in diamond	Corresponding author	Hideo Kosaka Affiliation: Yokohama National University		
Preprint information	No corresponding preprint identified.	Peer review type	Single-blind		

Editorial assessment team

Primary editor	Marco Bentivegna Home Journal: <i>Nature Communications</i> ORCID: orcid.org/0000-0002-3986-5760 Email: marco.bentivegna@nature.com
Editorial team members	Daniel Payne, <i>Communications Physics</i>, ORCID: 0000-0003-3368-2485 Richard Brierley, <i>Nature Physics</i>
About your primary editor	Marco completed his MS in particle physics and his PhD in quantum information and quantum optics in Sapienza University of Rome. For his doctoral research, he worked on quantum computation and simulation using integrated photonic circuits. His post-doctoral appointment involved performing experiments on quantum nonlocality and contextuality. He handles manuscripts in quantum information, computation and communication, fundamental aspects of quantum theory, information theory and high energy physics. Marco is based in the London office.

Editorial assessment and review synthesis

Editor's summary and assessment

The authors report entanglement generation between a geometric spin qubit in a nitrogen-vacancy center in diamond and a spontaneously emitted photon. The spin qubit is encoded in two degenerate energy levels, and can thus be addressed at zero magnetic field, which would ease its interfacing with other solid-state platforms. At the same time, the geometrical nature of the qubit is expected to improve robustness to noise. The experiment takes advantage of known techniques for manipulating this type of qubits using polarised light, and its main aim is confirming the entanglement of spontaneously emitted photons from such geometrically-encoded spin qubits. The work does not currently demonstrate prospective applications such as noise-resilient entanglement generation between remote spins. These considerations raise editorial reservations about potential suitability for publication in Nature Physics and Nature Communications.

Editorial synthesis of reviews

All the referees included comments about the similarities with previous works, such as the authors' earlier demonstrations of polarised microwave control, or previous examples of spin-photon entanglement generation at non-zero magnetic field. These statements confirmed the original editorial doubts for suitability of the manuscript for Nature Physics and Nature Communications. At the same time, the referees acknowledge the good fidelity reached in the experiment, which constitutes an indication of the potential advantages of this technique compared to non-zero field manipulations, and could constitute a motivation for consideration at Communications Physics.

Editorial recommendation

**nature
physics**

Revision not invited

Spin-photon entanglement, and magnetic-field-free manipulation of NV centre spin qubits have been demonstrated separately. As a result, this manuscript does not present the kind of conceptual advance we look for in a Nature Physics publication.

**nature
communications**

Major Revisions
with extension of
the study

The current version does not provide sufficient advance beyond previous demonstrations of polarised microwave control at zero magnetic field. In order to be reconsidered at Nature Communications, an experimental demonstration of the potential applications of this approach, e.g. noise-resilient entanglement generation between distant spins, or - as suggested by Reviewer #1 - generation of high-fidelity multiphoton entangled states, would be needed.

**communications
physics**

Minor Revisions

The reports indicate that your work offers a modest conceptual advance and appreciably-improved fidelity in comparison to the literature. The proposed scheme may eventually circumvent the limitations of alternative systems. As such, the manuscript is likely suitable for publication in Communications Physics pending minor revisions.

Next Steps

Recommendation Summary

- **Option 1 (recommended):** Revise for *Communications Physics*
- **Option 2:** Major Revision needed for further consideration at *Nature Communications*

See the previous page for details

Revision

If you would like to follow our recommendation, when you are ready you can upload the revised manuscript, along with your point-by-point response to the reviewer's reports and editorial advice [at this link](#)*.



Revision checklist

- Cover Letter, stating to which journal you are submitting
- Revised Manuscript
- Point-by-point response to reviews
- Updated Reporting Summary and Editorial Policy Checklist
- Supplementary materials (if applicable)



Submission elsewhere

To a journal outside of Nature Portfolio

We can share the reviews with another journal outside of the Nature Portfolio if requested. You will need to request that the receiving journal office contacts us at guidedOA@nature.com. We have included editorial guidance below in the reviewer reports and open research evaluation to aid in revising the manuscript for publication elsewhere.



Annotated Reviewer Reports

The editors have included some additional comments on specific points raised by the reviewers below, to clarify requirements for publication in the recommended journal(s). However, please note that all points should be addressed in a revision, even if an editor has not specifically commented on them.

Reviewer #1	
Reviewer #1	This reviewer has not chosen to waive anonymity. The reviewer's identity can only be shared with representatives of an established journal editorial office.
Reviewer #1 expertise Summarised by the editor	Spin qubits, quantum control, quantum communication, theorist
Editor's comments about this review	The reviewer acknowledges the potential advantages of this approach to spin manipulation, as well as the elements of novelty in this specific experiment, and suggests further potential applications. While agreeing with the reviewer on the good perspectives of applicability of geometrical spin qubit control, it is the editor's opinion that such potentialities were already clear from previous works, and that consideration for Nature Communications would require experimental demonstrations of novel capabilities rather than improved performances in known tasks.
Reviewer #1 comments	
Overview	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).

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

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:

Specific comments

#	Reviewer comment	Editorial comment
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?	In order to keep the claim of applicability to quantum state transfer, you would have to convince the referee that this technique can actually be used for that purpose. Otherwise, the claim would have to be removed.
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	This constitutes a possible application that you may want to explore with additional experiments in case you want to strengthen the paper for reconsideration at Nature Communications.

	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	
3	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

Reviewer #2	This reviewer has not chosen to waive anonymity. The reviewer’s identity can only be shared with representatives of an established journal editorial office.
Reviewer #2 expertise	solid-state spin systems for quantum technologies, NV centers in diamond, experimentalist

Summarised by the editor		
Editor's comments about this review	The reviewer points out the conceptual similarities with previous entanglement generation from NV centers at non-zero magnetic field, and with the authors' previous manipulations of NV spins at zero field. The main point of strength is found in the good reached fidelity. This is regarded by the editorial team as a commendable technical advance which may justify publication in Communications Physics.	
Reviewer #2 comments		
Overview	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.	
Specific comments		
#	Reviewer comment	Editorial comment
1	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.	
2	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.	
3	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?	
4	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.	The paper would clearly benefit from a much more complete and extensive description of the experimental apparatus and protocol (see also the same reviewer's comments below, or reviewer #3's third comment). These issues should be resolved in any subsequent resubmission.
5	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.	
6	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.	
7	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

Reviewer #3	This reviewer has not chosen to waive anonymity. The reviewer's identity can only be shared with representatives of an established journal editorial office.
Reviewer #3 expertise Summarised by the editor	solid-state spin systems for quantum technologies, NV centers in diamond, experimentalist
Editor's comments about this review	After highlighting the similarities with previous works, the reviewer expresses concerns on the advance constituted by merely confirming the spin-photon quantum correlations in this setting, as opposed to more complete demonstrations of two-qubit holonomic manipulation. At the same time, the reviewer acknowledges the superior fidelity compared to analogue demonstrations at non-zero field, which, in the editors' opinion, could justify consideration for Communications Physics

Reviewer #3 comments

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

Specific comments

#	Reviewer comment	Editorial comment
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).	The reviewer seems to challenge one of the main claims, or at least the fact that one could explicitly claim demonstration of geometric entanglement without performing holonomic two-qubit manipulation of the entangled qubits. We appreciate this may be seen as a matter of definitions, which nonetheless testifies the referee's opinion on the degree of advance this works represents over previous ones. The issue should be clarified in the answer to the reviewer, and, if needed, this should be reflected in modifications in the paper.
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.	Given that the high fidelity reached is one of the main strengths of the paper, it would be important, in our opinion, to be able to disentangle the effects on fidelity coming from geometric resilience from those due to zero-field operation. Whether and how this would be possible is something we cannot address at this stage. One possibility could be demonstrating some of the noise resilience capabilities that geometric encoding would have, such as insensitivity with respect to time, frequency, and space-mode matching. Another one would be estimating numerically the improvement on fidelity that

		comes from the zero-field. In case you are not able to convince the reviewer, you may have to tone down the claim of geometric entanglement observation.
3	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?	See related comments on Reviewer #2's report.

Open Research Evaluation

Data availability

Data Availability statement

Please add a Data Availability statement. Please ensure that your Data Availability statement includes accession details for deposited data, mentions where Source data can be found, and states that all other data are available from the corresponding author (or other sources, as applicable) on reasonable request. More information about our data availability policy can be found here: <https://www.nature.com/nature-portfolio/editorial-policies/reporting-standards#availability-of-data>

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The following figure panels should be accompanied by the underlying source data: **Figs 1e, 2b and 2d.**

Code availability and citation

Please ensure all the data collection/data analysis software/tools/algorithms/packages mentioned in the manuscript are also listed in the reporting summary (with version numbers). For example: GRAPE algorithm, etc.

See the Nature Portfolio policy on software availability here: <https://www.nature.com/nature-portfolio/editorial-policies/reporting-standards#availability-of-computer-code>

Ethics

Please provide a 'Competing interests' statement using one of the following standard sentences:

1. The authors declare the following competing interests: [specify competing interests]
2. The authors declare no competing interests.

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Statistics

Wherever statistics have been derived (e.g. error bars, box plots, statistical significance) the legend needs to provide and define the n number (i.e. the sample size used to derive statistics) as a precise value (not a range), using the wording “n=X biologically independent samples/animals/cells/independent experiments/n= X cells examined over Y independent experiments” etc. as applicable.

Please note that this information is missing in the legend of **Figure 1e**.

Please note that statistics such as error bars significance and p values cannot be derived from $n < 3$ and must be removed in all such cases.

We strongly discourage deriving statistics from technical replicates, unless there is a clear scientific justification for why providing this information is important. Conflating technical and biological variability, e.g., by pooling technically replicates samples across independent experiments is strongly discouraged. (For examples of expected description of statistics in figure legends, please see the following <https://www.nature.com/articles/s41467-019-11636-5> or <https://www.nature.com/articles/s41467-019-11510-4>).

All error bars need to be defined in the legends (e.g. SD, SEM) together with a measure of centre (e.g. mean, median). For example, the legends should state something along the lines of “Data are

presented as mean values $\pm$ SEM" as appropriate.

All box plots need to be defined in the legends in terms of minima, maxima, centre, bounds of box and whiskers and percentile.

Please note that the measure of centre for the error bars needs to be defined in the legends of **Figures 1e, 3b**.

The figure legends must indicate the statistical test used. Where appropriate, please indicate in the figure legends whether the statistical tests were one-sided or two-sided and whether adjustments were made for multiple comparisons.

For null hypothesis testing, please indicate the test statistic (e.g. F, t, r) with confidence intervals, effect sizes, degrees of freedom and P values noted.

Please provide the test results (e.g. P values) as exact values whenever possible and with confidence intervals noted.

Data presentation

Please ensure that data presented in a plot, chart or other visual representation format shows data distribution clearly (e.g. dot plots, box-and-whisker plots). When using bar charts, please overlay the corresponding data points (as dot plots) whenever possible and always for $n \leq 10$. (Please see the following editorial for the rationale behind this request and an example <https://www.nature.com/articles/s41551-017-0079>).

Please note that data presentation has to be revised to comply with our policy in **Figure 3b**.

Reproducibility

Please state in the legends how many times each experiment was repeated independently with similar results. This is needed for all experiments, but is particularly important wherever results from representative experiments (such as micrographs) are shown. If space in the legends is limiting, this information can be included in a section titled "Statistics and Reproducibility" in the methods section.