nature portfolio

Peer Review File

Strong coupling between a single-photon and a two-photon Fock state

Corresponding Author: Dr Shuai-Peng Wang

This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Communications.

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

Version 0:
Reviewer comments:
Reviewer #2
(Remarks to the Author)

While in general the authors have advanced in addressing the points raised in my first report, there is a lack of depth in the answers. In order for this work to be publishable in Nature Communications, more quantitative arguments must be provided, as otherwise it is not clear that the work describes the actual physics taking place in this experiment.

-While the theoretical tools defined are the appropriate ones to describe a system in the ultrastrong coupling regime, the analysis is confined to the 2 lowest modes of the waveguide. If one considers the actual physics of the system there will be nonnegligible interactions to the higher-energy modes, particularly the one lying around 15 GHz which is quite close to the qubit gap of 12 GHz. The theoretical plots do look like they describe the observations, however there are enough free parameters in the system (qubit frequency and current, mode frequencies, coupling strengths, etc.) that the fitted parameters may effectively hide the effects from higher-energy modes. The authors should demonstrate that this is not the case, otherwise the results are under question.

Reply

The flux qubit is placed in the position corresponding to the node of the third mode. This results in a negligible coupling to that mode.

Counter-reply:

While it is indeed the case that the third mode will be very weakly coupled to the qubit directly, there are additional modes higher up in frequency. The 4th mode, which has non-negligible amplitude at the qubit position, resonates near 25GHz. The qubit at the biasing of 45mPhi0 has a frequency of about 22.5GHz, using the parameters from the manuscript. One cannot thus simply ignore such higher-frequency modes. I request the authors to validate their theoretical model with the addition of at least the 4th mode of the resonator and provide quantitative proof that such additional mode plays no role in the process described in this work.

-Another limitation in the theoretical analysis is the use of a limited quantum optical model (Eq. 1), while what is actually needed is a circuit quantization analysis, especially given the galvanic connection of the qubit to a resonator. For instance, a proper analysis should follow the procedure described in the appendix of this reference: B. Peropadre, D. Zueco, D. Porras, and J. J. García-Ripoll, Phys. Rev. Lett. 111, 243602 (2013). Failure to perform such an analysis may result in an effective coupling strength which is not the actual circuit-induced coupling strength but the one of an effective

quantum optical model that misses the actual physics lying underneath.

Reply:

We have added a circuit quantization analysis in the supplementary information. The resulting Hamiltonian confirms the validity of the model used in the previous version of the manuscript.

Counter-reply:

The authors have indeed begun producing a circuit QED model of their experiment. However, the model is incomplete. Eq. 3 is not strictly speaking correct. Inside the first term, the inductive sum of elements contains terms that involve the qubit phases and therefore have to be taken into account when modelling the qubit, as these terms will renormalize the qubit frequency as well as the coupling to each mode.

Similarly, one would expect capacitive terms involving phase differences with the qubit islands and the resonator islands.

Eventually, with the form of Eq. 3, one can estimate the value of coupling strength g_n following the methods from Peropadre, et al. (Eq. 33 of their supplementary) for each resonator mode n, and compare them to the values obtained in the experiment. This would be useful to the readers of this work as a convincing evidence of the device being in the ultrastrong coupling regime.

-Several important technical details are omitted both from the main text and the supplementary and are rather critical to make several claims. One of the main points is the discussion on how the effective photon number is calibrated to claim that the effects observed are below average single photon 1. For instance, how is the x-axis of Fig. 3c calibrated? what is the error?

Reply:

The average photon number in Fig. 3c is determined by contrasting the experimental data—plotting the maximum second-harmonic generation (SHG) amplitude against input power—with the theoretical simulation outlined in Eq. 21 of the supplementary information, as depicted in Fig. 3c. During the calibration of the photon number, the obtained fitted parameters allowed us to reproduce in very good agreement not only the data in Fig. 3c but also the spectra in Fig. 2 and the interference fringes in Fig. 4. Hence, although there might be a slight deviation from the true photon number, we expect it to be less than 10% of our fitted values.

Counter-reply:

This information cannot be found on the revised text. Please, add it either to the main text and/or at the caption of Fig. 3.

-Other less relevant details but important for the information they omit is a technical description of how each measurement is performed. For instance, the data on Fig. 3, is it a coherent signal measured with a VNA, or is it incoherent signal acquired in a spectrum analyzer? This applies similarly to Figs. 2 and 4. And Fig. 4, what is the y-axis representing? this variable is not stated in the text.

Reply:

The data presented in Figs. 2, 3, and 4 were all acquired as coherent signals using a vector network analyzer (VNA). For Fig. 2, the VNA was employed to measure the S21 parameter of the transmitted signal. In the case of Fig. 3, the VNA detected the SHG signal emanating from the device, which was excited by an external signal generator. The VNA's output port was terminated, and it functioned solely to capture the SHG signal. For Fig. 4, the VNA measured the S21 parameter of the signal at the frequency $\omega/2\omega$, while a simultaneous signal at $2\omega/\omega$ frequency was applied from a separate signal generator. We have added this clarification in the supplementary information.

Counter-reply:

Still, I do not see the definition of gain neither in the figure caption (where it should be stated) nor in the supplementary information.

-Towards the end of the manuscript (line 307) the authors state that "without exciting atomic transitions". Given the ultrastrong coupling from the qubit to the two modes analyzed in this work, I believe the degree of hybridization of qubit and photon of each energy level excited is rather high. Therefore, the claim that no atomic transitions are excited is arguable. In fact, it would be convenient to discuss the effect of losses in the qubit how they translate into losses of the photon-like states involved in the system.

Reply:

Indeed, even with the ultrastrong coupling present, the relatively high loss rates of the flux qubit in our device do not substantially degrade the coherence of the interacting photons. This is confirmed by the theoretical simulations presented in Supplementary Fig. 3, and in the new dedicated Appendix. The resilience of coherence can be ascribed to the significant detuning between the qubit transition frequency, which is 22.5 GHz at the anticrossing flux bias point, and the resonator frequencies of 4.9 GHz and 9.8 GHz. The detuning gaps are 17.6 GHz and 12.7 GHz, respectively, far surpassing the coupling strength of 2.18 GHz. Operating in this dispersive regime ensures that the excitation of atomic transitions is effectively negligible.

Counter-reply:

I respectfully disagree with the statement that a detuning of 17.6GHz "far surpasses" a coupling strength of 2.18GHz. In order for the qubit-resonator hybridization of the levels to be merely dispersive up to second order in g/detuning, the ratio g/detuning should be far smaller than 1. Taking the actual parameters, this is 16% for mode 1 and 17% for mode 2, and it may likely be higher for mode 4 which is just 2.5GHz away in frequency. Therefore, all of them are far from dispersive. The authors should quantify the potential amount of atomic excitation in numbers, not just with qualitatively statements such as "confirmed by the theoretical simulations" which, as already stated in my previous report, contain numerous fitting parameters and may mask the actual physics going on.

-The authors state that their transmission line geometry is inhomogeneous and refer to Fig 1b, which only shows a very small fraction of the waveguide where the qubit is placed. Could the authors elaborate more (why is it inhomogeneous?) and detail the consequences of this inhomogeneity in the resonances?

Reply:

Despite the geometric inhomogeneity inherent in the transmission line, an even more pronounced form of inhomogeneity arises from the inductance introduced by the Josephson junctions (JJs, Josephson inductance ~0.6 nH) embedded within the transmission line (geometry inductance ~5 nH). A key consequence of this inhomogeneity is that the second mode frequency is not precisely double that of the first mode frequency, as would be expected in a standard transmission-line resonator.

Counter-reply:

Could the authors state in numbers such an inhomogeneity?

-Reference 39 is now published as https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.134.013804

(Remarks on code availability)

Reviewer #3

(Remarks to the Author)

I checked the revised manuscript and their reply to 3 referees. My overall impression is that the authors do not respond to the referee comments faithfully and the revisions are insufficient. I cannot recommend acceptance of this manuscript in Nature Communications in its current form.

Comments

Regarding comment (1) of Referee 3: from the revised manuscript and the authors' response, it turned out that the conversion efficiency only reaches 1%(=\eta^2). Is this value really an unprecedented efficiency as the authors argue in their manuscript?

Regarding comment (2) of Referee 3: from the authors' response, it turned out that the authors have no clear experimental evidence on downconversion. Within the theory, near deterministic conversion efficiency has already been predicted, so "unprecedented" should be removed.

Regarding comment (4) of Referee 3: the authors state in their reply that "We have updated the manuscript to avoid this misleading". However, no revisions are made on this point. For example, "photon pair" is still used in the title. As I wrote in my previous comment, "two-photon Fock state" and "photon pair" are quite different concept. If the author sticks to the use of

"photon pair", they should explain its reason.

Regarding comment (5) of Referee 3: in my previous report, I made a question that "Regarding the external coupling rates, why the authors set \kappa_{out}=3\kappa_{in}?". No answer is given in the revised manuscript nor their reply.

(Remarks on code availability)

Version 1:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The authors have addressed all my points in a satisfactory manner, and I consider the manuscript to be suitable for publication after some minor revision of the last modifications by the authors.

There seems to be no mention added to the main text about the conclusions of the checks I required the authors to run. This makes the analysis buried in the long supplementary. Therefore, I encourage the authors to properly state the conclusions of the new sections in the supplementary information (role of the 4th mode, estimate of coupling from circuit quantization, qubit hybridization) to the main text, and reference each result to the appropariate section of the supplementary.

We sincerely thank the reviewer for the insightful comments and detailed arguments. We believe that, thanks to these valuable suggestions, the revised manuscript now presents the underlying physics with the quantitative depth expected, and we hope it meets the referee's high standards. Below we provide a point-by-point response to all concerns raised. The fourth mode of the resonator is at approximately 18 GHz. We agree that this mode interacts with the qubit when the bias is around 45 mPhi0. However, this interaction

happens at higher energies with respect to those studied in our work. Due to the small pump amplitude (less than one photon in the resonator) and the small drive frequency, the fourth mode has no photons. In our work we focus on the low-energy region, ensuring that the higher-energy modes are not excited.

To quantitatively demonstrate that the fourth mode has no influence on the effect, we have added a section in the Supplemental Materials, showing both the eigenvalues of the total Hamiltonian and the transmission spectrum (Supplementary Fig. 2). The eigenvalues in the interested region remained the same, and the transmission spectrum is identical to Fig. 2 of the main text.

We hope that this clarifies that the higher-energy modes don't have any influence on the effect we study in this work.

The analysis of the fourth mode is sound. However, I would expect to see a comment regarding it on the main text and a reference to the supplementary.

We would like to point out first that Supplementary Eq. 3 of the previous version was exactly equivalent to the total Lagrangian present in other works like in Peropadre, Physical review letters, 111(24), 243602 (2013) or Bourassa, Physical Review A, 86(1), 013814.402 (2012). Our previous Supplementary Eq. 3 has exactly the same form of, e.g., Supplementary Eq. 14 in the Peropadre work. Perhaps, the Referee didn't notice that the sum is excluding the m-th index, and so there is no qubit phase contribution in the sum of the inductive terms of the waveguide.

To make everything more explicit, we have performed a more complete quantization of the system. The derivation generalizes the one- and two-junction treatment of Bourassa, Physical Review A, 86(1), 013814.402 (2012) to our three-junction device. This method takes into account the renormalization of the modes frequencies with respect to the external flux (see, e.g., Fig. 12 of the Bourassa work), giving the V-shape behavior observed in our data and also found in, e.g., Yoshihara, Nature Physics, 13(1), 44-47 (2017). And, we have numerically calculated the coupling strength g_n (Supplementary Eq. 19) using the new circuit quantization approach, the results qualitatively agree with the fitted values in the main text (see the updated Supplementary Information).

We hope that this expanded analysis addresses the referee's concern. Moreover, we would like to point out the excellent agreement between the Hamiltonian in Eq. 1 and all the presented data further support the applicability of Eq. 1 as a proper model Hamiltonian for the investigated device.

The authors have made a significant effort to properly describe their circuit using a quantization model known in the literature, equivalent to my original suggestion (which was incomplete). There are some minor corrections regarding their final expressions (Eqs. 17-19 of the supplementary):

- -The approximation that sin(Phi_ext/Phi_0) is negligible is arguable, as this is only valid near Phi_ext = 0. However, in the experiment Phi_ext is always near Phi_0/2 where that sine term is maximum. Therefore, I would suggest the authors to revise that statement and provide the correct full expression of the system Hamiltonian in Eq. 17.
- -In the final expression for the coupling g_n, it seems the flux dependency on Phi_ext one can see in the last term of Eq. 17 is gone. Could the authors verify the expression in Eq. 19?
- -In the numerical evaluation of the coupling below Eq. 19, the correct units should be g/2pi as the authors use in the main text.

We think that a ratio g/detuning of 0.12 still qualifies as dispersive regime. Such values are also typical in the dispersive readout of qubits (see, e.g., Gautier, Phys. Rev. Lett. 134, 070802 (2025)). In any case, we have computed the dressed qubit

population as a function of the flux bias, showing that it has a neglible participation in the region of the modes hybridization. We now show a figure (Supplementary Fig. 4) demonstrating this for the first three eigenstates. To dispel any remaining ambiguity, the calculation explicitly includes the fourth resonator mode, ensuring that the influence of higher harmonics is fully captured.

We hope that this Figure, together with Fig. 5 in the Supplemental Materials, should clarify this fact.

The analysis is sound. Again, please make sure it is mentioned and properly referenced on the main text.

The geometric inductance of the CPW is approximately 0.0004 nH/ μ m along the transmission line, whereas the average inductance across the Josephson junction (JJ) section (aluminum strips and the qubit loop) reaches 0.0075 nH/ μ m. This results in a local inhomogeneity of ~ $(0.0075/0.0004)\times100\% \approx 1875\%$.

My comment was referring to whether the authors considered any other inhomogeinities such as impedance engineering as typically used in parametric donwconversion experiments to avoid degeneracies between modes. Just to make this point clear, I suggest the authors to modify the statement under Eq. 1 of the main text as:

"Owing to the inhomogeneous transmission line geometry due to the qubit presence..."

(Remarks on code availability)

Reviewer #3

(Remarks to the Author)

I checked the revised manuscript and their reply to Referee 2 and 3. Again, my overall impression is that the authors do not respond to the referee comments faithfully and the revisions are insufficient. I cannot recommend publication of this manuscript in Nature Communications.

Comments

Regarding comment (1) of Referee 3: By reading authors' response, I could understand that the 1% conversion efficiency is higher than the previous works on SHG using weak input fields. However, I feel that SHG with 1% efficiency is not very impressive in the context of quantum nonlinear optics, since near deterministic down-conversion has been demonstrated already. I recommend the authors to include these comments in the main text so that the readers can recognize the significance of these results.

Regarding comment (2) of Referee 3: I could understand the referees' statement that spontaneously down converted photons are incoherent and cannot be measured through amplitude measurements. The authors' comments are instructive to the readers so should be included in the main text. I also hope to see the authors' explanation on the process in which the incoherent signal (DC photons) can generate coherent signal (Fig.4a).

Regarding comment (4) of Referee 3: The authors state (in previous response) that "We have updated the manuscript to avoid this misleading" and (in this response) that "In the last reply, we have already changed many instances of "photon pair" to "two photons"". However, in the previous response, they did not revise the title, which is the most important part of the paper. I wonder that the authors correctly recognize the difference between "two-photon Fock state" and "photon pair".

Regarding comment (5) of Referee 3: The authors statement "We used different values for \kappa{in} and \kappa{out}" is obvious. My question was "why the authors set \kappa_{out}=3\kappa_{in}?" and I feel that the authors do not respond to the question

kappa_{in} and kappa_{out} are the cavity decay rates at the input/output frequencies, which should be determined experimentally in principle and are not the quantities the authors can arbitrarily choose. Since this is an experimental paper, the authors should measure them and assign more reliable parameter values.

(Remarks on code availability)	on code availabilit	v)
--------------------------------	---------------------	----

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The authors have addressed all my comments. The manuscript thus can be published. I only found that in Eq. 19 of the supplementary should be $\beta_n = ...$

(Remarks on code availability)

Reviewer #3 (Remarks to the Author) In this revision, the authors have revised the manuscript adequately in response to the comments of both reviewers. I now recommend publication of this work in Nature Communications.

(Remarks on code availability)

Open Access This Peer Review File is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

In cases where reviewers are anonymous, credit should be given to 'Anonymous Referee' and the source. The images or other third party material in this Peer Review File are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

To view a copy of this license, visit https://creativecommons.org/licenses/by/4.0/

Referees' comments:

Reviewer #2 (Remarks to the Author):

While in general the authors have advanced in addressing the points raised in my first report, there is a lack of depth in the answers. In order for this work to be publishable in Nature Communications, more quantitative arguments must be provided, as otherwise it is not clear that the work describes the actual physics taking place in this experiment.

We sincerely thank the reviewer for the insightful comments and detailed arguments. We believe that, thanks to these valuable suggestions, the revised manuscript now presents the underlying physics with the quantitative depth expected, and we hope it meets the referee's high standards. Below we provide a point-by-point response to all concerns raised.

-While the theoretical tools defined are the appropriate ones to describe a system in the ultrastrong coupling regime, the analysis is confined to the 2 lowest modes of the waveguide. If one considers the actual physics of the system there will be nonnegligible interactions to the higher-energy modes, particularly the one lying around 15 GHz which is quite close to the qubit gap of 12 GHz. The theoretical plots do look like they describe the observations, however there are enough free parameters in the system (qubit frequency and current, mode frequencies, coupling strengths, etc.) that the fitted parameters may effectively hide the effects from higher-energy modes. The authors should demonstrate that this is not the case, otherwise the results are under question.

Reply:

The flux qubit is placed in the position corresponding to the node of the third mode. This results in a negligible coupling to that mode.

Counter-reply:

While it is indeed the case that the third mode will be very weakly coupled to the qubit directly, there are additional modes higher up in frequency. The 4th mode, which has non-negligible amplitude at the qubit position, resonates near 25GHz. The qubit at the biasing of 45mPhi0 has a frequency of about 22.5GHz, using the parameters from the manuscript. One cannot thus simply ignore such higher-frequency modes. I request the authors to validate their theoretical model with the addition of at least the 4th mode of the resonator and provide quantitative proof that such additional mode plays no role in the process described in this work.

The fourth mode of the resonator is at approximately 18 GHz. We agree that this mode interacts with the qubit when the bias is around 45 mPhi0. However, this interaction

happens at higher energies with respect to those studied in our work. Due to the small pump amplitude (less than one photon in the resonator) and the small drive frequency, the fourth mode has no photons. In our work we focus on the low-energy region, ensuring that the higher-energy modes are not excited.

To quantitatively demonstrate that the fourth mode has no influence on the effect, we have added a section in the Supplemental Materials, showing both the eigenvalues of the total Hamiltonian and the transmission spectrum (Supplementary Fig. 2). The eigenvalues in the interested region remained the same, and the transmission spectrum is identical to Fig. 2 of the main text.

We hope that this clarifies that the higher-energy modes don't have any influence on the effect we study in this work.

-Another limitation in the theoretical analysis is the use of a limited quantum optical model (Eq. 1), while what is actually needed is a circuit quantization analysis, especially given the galvanic connection of the qubit to a resonator. For instance, a proper analysis should follow the procedure described in the appendix of this reference: B.

Peropadre, D. Zueco, D. Porras, and J. J. García-Ripoll, Phys. Rev. Lett. 111, 243602

(2013). Failure to perform such an analysis may result in an effective coupling strength which is not the actual circuit-induced coupling strength but the one of an effective quantum optical model that misses the actual physics lying underneath.

Reply:

We have added a circuit quantization analysis in the supplementary information. The resulting Hamiltonian confirms the validity of the model used in the previous version of the manuscript.

Counter-reply:

The authors have indeed begun producing a circuit QED model of their experiment. However, the model is incomplete. Eq. 3 is not strictly speaking correct. Inside the first term, the inductive sum of elements contains terms that involve the qubit phases and therefore have to be taken into account when modelling the qubit, as these terms will renormalize the qubit frequency as well as the coupling to each mode.

Similarly, one would expect capacitive terms involving phase differences with the qubit islands and the resonator islands.

Eventually, with the form of Eq. 3, one can estimate the value of coupling strength g_n following the methods from Peropadre, et al. (Eq. 33 of their supplementary) for each resonator mode n, and compare them to the values obtained in the experiment. This would be useful to the readers of this work as a convincing evidence of the device being in the ultrastrong coupling regime.

We would like to point out first that Supplementary Eq. 3 of the previous version was exactly equivalent to the total Lagrangian present in other works like in *Peropadre*, *Physical review letters*, 111(24), 243602 (2013) or *Bourassa*, *Physical Review A*, 86(1), 013814.402 (2012). Our previous Supplementary Eq. 3 has exactly the same form of, e.g., Supplementary Eq. 14 in the *Peropadre* work. Perhaps, the Referee didn't notice that the sum is excluding the m-th index, and so there is no qubit phase contribution in the sum of the inductive terms of the waveguide.

To make everything more explicit, we have performed a more complete quantization of the system. The derivation generalizes the one- and two-junction treatment of *Bourassa, Physical Review A, 86(1), 013814.402 (2012)* to our three-junction device. This method takes into account the renormalization of the modes frequencies with respect to the external flux (see, e.g., Fig. 12 of the *Bourassa* work), giving the V-shape behavior observed in our data and also found in, e.g., *Yoshihara, Nature Physics, 13(1), 44-47 (2017)*. And, we have numerically calculated the coupling strength g_n (Supplementary Eq. 19) using the new circuit quantization approach, the results qualitatively agree with the fitted values in the main text (see the updated Supplementary Information).

We hope that this expanded analysis addresses the referee's concern. Moreover, we would like to point out the excellent agreement between the Hamiltonian in Eq. 1 and all the presented data further support the applicability of Eq. 1 as a proper model Hamiltonian for the investigated device.

-Several important technical details are omitted both from the main text and the supplementary and are rather critical to make several claims. One of the main points is the discussion on how the effective photon number is calibrated to claim that the effects observed are below average single photon 1. For instance, how is the x-axis of Fig. 3c calibrated? what is the error?

Reply:

The average photon number in Fig. 3c is determined by contrasting the experimental data—plotting the maximum second-harmonic generation (SHG) amplitude against

input power—with the theoretical simulation outlined in Eq. 21 of the supplementary

information, as depicted in Fig. 3c. During the calibration of the photon number, the obtained fitted parameters allowed us to reproduce in very good agreement not only the data in Fig. 3c but also the spectra in Fig. 2 and the interference fringes in Fig. 4. Hence, although there might be a slight deviation from the true photon number, we expect it to be less than 10% of our fitted values.

Counter-reply:

This information cannot be found on the revised text. Please, add it either to the main

text and/or at the caption of Fig. 3.

Sorry for this oversight. We have added this information in the updated manuscript.

-Other less relevant details but important for the information they omit is a technical description of how each measurement is performed. For instance, the data on Fig. 3, is it a coherent signal measured with a VNA, or is it incoherent signal acquired in a spectrum analyzer? This applies similarly to Figs. 2 and 4. And Fig. 4, what is the y-axis representing? This variable is not stated in the text.

Reply:

The data presented in Figs. 2, 3, and 4 were all acquired as coherent signals using a vector network analyzer (VNA). For Fig. 2, the VNA was employed to measure the S21 parameter of the transmitted signal. In the case of Fig. 3, the VNA detected the SHG signal emanating from the device, which was excited by an external signal generator. The VNA's output port was terminated, and it functioned solely to capture the SHG signal. For Fig. 4, the VNA measured the S21 parameter of the signal at the frequency $\omega/2\omega$, while a simultaneous signal at $2\omega/\omega$ frequency was applied from a separate signal generator. We have added this clarification in the supplementary information.

Counter-reply:

Still, I do not see the definition of gain either in the figure caption (where it should be stated) nor in the supplementary information.

The gain is the amplitude of the normalized S21 parameter measured at frequency ω /2 ω . This is defined as the ratio between the output signal amplitude at ω /2 ω when a secondary signal is applied at 2 ω / ω , and the output amplitude of the same signal at ω /2 ω in the absence of the secondary signal applied at 2 ω / ω . We have added this clarification in the updated manuscript.

-Towards the end of the manuscript (line 307) the authors state that "without exciting atomic transitions". Given the ultrastrong coupling from the qubit to the two modes analyzed in this work, I believe the degree of hybridization of qubit and photon of each energy level excited is rather high. Therefore, the claim that no atomic transitions are excited is arguable. In fact, it would be convenient to discuss the effect of losses in the qubit how they translate into losses of the photon-like states involved in the system.

Reply:

Indeed, even with the ultrastrong coupling present, the relatively high loss rates of the flux qubit in our device do not substantially degrade the coherence of the interacting photons. This is confirmed by the theoretical simulations presented in Supplementary Fig. 3, and in the new dedicated Appendix. The resilience of coherence can be ascribed to the significant detuning between the qubit transition frequency, which is 22.5 GHz at the anticrossing flux bias point, and the resonator frequencies of 4.9 GHz and 9.8 GHz. The detuning gaps are 17.6 GHz and 12.7 GHz, respectively, far surpassing the coupling strength of 2.18 GHz. Operating in this dispersive regime ensures that the excitation of atomic transitions is effectively negligible.

Counter-reply:

I respectfully disagree with the statement that a detuning of 17.6GHz "far surpasses" a coupling strength of 2.18GHz. In order for the qubit-resonator hybridization of the levels to be merely dispersive up to second order in g/detuning, the ratio g/detuning should be far smaller than 1. Taking the actual parameters, this is 16% for mode 1 and 17% for mode 2, and it may likely be higher for mode 4 which is just 2.5GHz away in frequency. Therefore, all of them are far from dispersive. The authors should quantify the potential amount of atomic excitation in numbers, not just with qualitatively statements such as "confirmed by the theoretical simulations" which, as already stated in my previous report, contain numerous fitting parameters and may mask the actual physics going on.

We think that a ratio g/detuning of 0.12 still qualifies as dispersive regime. Such values are also typical in the dispersive readout of qubits (see, e.g., *Gautier, Phys. Rev. Lett.* 134, 070802 (2025)). In any case, we have computed the dressed qubit population as a function of the flux bias, showing that it has a neglible participation in the region of the modes hybridization. We now show a figure (Supplementary Fig. 4) demonstrating this for the first three eigenstates. To dispel any remaining ambiguity, the calculation explicitly includes the fourth resonator mode, ensuring that the influence of higher harmonics is fully captured.

We hope that this Figure, together with Fig. 5 in the Supplemental Materials, should clarify this fact.

-The authors state that their transmission line geometry is inhomogeneous and refer to Fig 1b, which only shows a very small fraction of the waveguide where the qubit is placed. Could the authors elaborate more (why is it inhomogeneous?) and detail the consequences of this inhomogeneity in the resonances?

Reply:

Despite the geometric inhomogeneity inherent in the transmission line, an even more pronounced form of inhomogeneity arises from the inductance introduced by the

Josephson junctions (JJs, Josephson inductance ~0.6 nH) embedded within the transmission line (geometry inductance ~5 nH). A key consequence of this inhomogeneity is that the second mode frequency is not precisely double that of the first mode frequency, as would be expected in a standard transmission-line resonator.

Counter-reply:

Could the authors state in numbers such an inhomogeneity?

The geometric inductance of the CPW is approximately 0.0004 nH/ μ m along the transmission line, whereas the average inductance across the Josephson junction (JJ) section (aluminum strips and the qubit loop) reaches 0.0075 nH/ μ m. This results in a local inhomogeneity of ~(0.0075/0.0004)×100% ≈ 1875%.

-Reference 39 is now published as https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.134.013804

We thank the reviewer for the information. We have updated reference 39 in the main text.

Reviewer #3 (Remarks to the Author):

I checked the revised manuscript and their reply to 3 referees. My overall impression is that the authors do not respond to the referee comments faithfully and the revisions are insufficient. I cannot recommend acceptance of this manuscript in Nature Communications in its current form.

We apologize for not adequately addressing the reviewer's concerns in our previous response. We have clarified the points raised below and hope these explanations resolve the reviewer's concerns.

Comments

Regarding comment (1) of Referee 3: from the revised manuscript and the authors' response, it turned out that the conversion efficiency only reaches 1%(=\eta^2). Is this value really an unprecedented efficiency as the authors argue in their manuscript?

We should emphasize that the 1% second-harmonic generation (SHG) efficiency is achieved with only ~0.25 photons input into the resonator—operating in the quantum limit. When normalized per input photon, this conversion efficiency is exceptionally high. For comparison, classical SHG systems exhibit efficiency proportional to the photon number in the cavity. Achieving 1% efficiency in such regimes typically requires thousands or more input photons (see, e.g., *Wu, Laser Photonics Rev. 18(7), 2300951*

(2024)).

To the best of our knowledge, SHG in the quantum limit has never been experimentally observed until now. Previous devices reported near-zero SHG efficiency under quantum-limited conditions (sub-photon input levels). Thus, we think that the results presented here are unprecedented.

Regarding comment (2) of Referee 3: from the authors' response, it turned out that the authors have no clear experimental evidence on downconversion. Within the theory, near deterministic conversion efficiency has already been predicted, so "unprecedented" should be removed.

This work does not provide any experimental evidence of spontaneous down-conversion. This is a direct consequence of the fact that our setup selects only coherent signals, with the phase determined by the input signal. This technique allows to filter thermal noise and to allow the detection of very small signals. However, spontaneous down conversion is not a coherent signal and, it is thus filtered out. Nonetheless, Fig. 4a provides clear evidence of stimulated down-conversion, which gives rise to a coherent signal.

To avoid misunderstanding, we change the sentence to "SHG and stimulated downconversion can occur efficiently at sub-photon input levels".

Regarding comment (4) of Referee 3: the authors state in their reply that "We have updated the manuscript to avoid this misleading". However, no revisions are made on this point. For example, "photon pair" is still used in the title. As I wrote in my previous comment, "two-photon Fock state" and "photon pair" are quite different concept. If the author sticks to the use of "photon pair", they should explain its reason.

We have updated the title and the related sentences. In the last reply, we have already changed many instances of "photon pair" to "two photons" (red font).

Regarding comment (5) of Referee 3: in my previous report, I made a question that "Regarding the external coupling rates, why the authors set \kappa_{out}=3\kappa_{in}?". No answer is given in the revised manuscript nor their reply.

In the last reply, we stated that "We used different values for \kappa{in} and \kappa{out} for a more efficient extraction of the signal". This setting is just to enchance the transmitted amplitude from the output port.

Reviewer #2 (Remarks to the Author):

The authors have addressed all my points in a satisfactory manner, and I consider the manuscript to be suitable for publication after some minor revision of the last modifications by the authors.

There seems to be no mention added to the main text about the conclusions of the checks I required the authors to run. This makes the analysis buried in the long supplementary. Therefore, I encourage the authors to properly state the conclusions of the new sections in the supplementary information (role of the 4th mode, estimate of coupling from circuit quantization, qubit hybridization) to the main text, and reference each result to the appropriate section of the supplementary.

Reply:

We thank the reviewer for the positive comments and the recommendation for publication! We have addressed all the remaining concerns in the following.

We sincerely thank the reviewer for the insightful comments and detailed arguments. We believe that, thanks to these valuable suggestions, the revised manuscript now presents the underlying physics with the quantitative depth expected, and we hope it meets the referee's high standards. Below we provide a point-by-point response to all concerns raised.

The fourth mode of the resonator is at approximately 18 GHz. We agree that this mode interacts with the qubit when the bias is around 45 mPhi0. However, this interaction happens at higher energies with respect to those studied in our work. Due to the small pump amplitude (less than one photon in the resonator) and the small drive frequency, the fourth mode has no photons. In our work we focus on the low-energy region, ensuring that the higher-energy modes are not excited.

To quantitatively demonstrate that the fourth mode has no influence on the effect, we have added a section in the Supplemental Materials, showing both the eigenvalues of the total Hamiltonian and the transmission spectrum (Supplementary Fig. 2). The eigenvalues in the interested region remained the same, and the transmission spectrum is identical to Fig. 2 of the main text.

We hope that this clarifies that the higher-energy modes don't have any influence on the effect we study in this work.

The analysis of the fourth mode is sound. However, I would expect to see a comment regarding it on the main text and a reference to the supplementary.

Reply:

We thank the reviewer for the positive comment and have updated the manuscript accordingly.

We would like to point out first that Supplementary Eq. 3 of the previous version was exactly equivalent to the total Lagrangian present in other works like in Peropadre,

Physical review letters, 111(24), 243602 (2013) or Bourassa, Physical Review A, 86(1), 013814.402 (2012). Our previous Supplementary Eq. 3 has exactly the same form of, e.g., Supplementary Eq. 14 in the Peropadre work. Perhaps, the Referee didn't notice that the sum is excluding the m-th index, and so there is no qubit phase contribution in the sum of the inductive terms of the waveguide.

To make everything more explicit, we have performed a more complete quantization of the system. The derivation generalizes the one- and two-junction treatment of Bourassa, Physical Review A, 86(1), 013814.402 (2012) to our three-junction device. This method takes into account the renormalization of the modes frequencies with respect to the external flux (see, e.g., Fig. 12 of the Bourassa work), giving the V-shape behavior observed in our data and also found in, e.g., Yoshihara, Nature Physics, 13(1), 44-47 (2017). And, we have numerically calculated the coupling strength g_n (Supplementary Eq. 19) using the new circuit quantization approach, the results qualitatively agree with the fitted values in the main text (see the updated Supplementary Information).

We hope that this expanded analysis addresses the referee's concern. Moreover, we would like to point out the excellent agreement between the Hamiltonian in Eq. 1 and all the presented data further support the applicability of Eq. 1 as a proper model Hamiltonian for the investigated device.

The authors have made a significant effort to properly describe their circuit using a quantization model known in the literature, equivalent to my original suggestion (which was incomplete). There are some minor corrections regarding their final expressions (Eqs. 17-19 of the supplementary):

-The approximation that sin(Phi_ext/Phi_0) is negligible is arguable, as this is only valid near Phi_ext = 0. However, in the experiment Phi_ext is always near Phi_0/2 where that sine term is maximum. Therefore, I would suggest the authors to revise that statement and provide the correct full expression of the system Hamiltonian in Eq. 17. -In the final expression for the coupling g_n, it seems the flux dependency on Phi_ext one can see in the last term of Eq. 17 is gone. Could the authors verify the expression in Eq. 19?

-In the numerical evaluation of the coupling below Eq. 19, the correct units should be g/2pi as the authors use in the main text.

Reply:

These issues come from the fact that we used a wrong symbol (Φ_0) for the reduced flux quantum in the Supplementary Information, which is the same as we used in the main text for the standard flux quantum (Φ_0) . The reduced flux quantum should be $\phi_0=\Phi_0/2\pi$, then $sin(\Phi_{ext}/\phi_0)\simeq sin(\pi)=0$ near the optimal point $\Phi_{ext}=\Phi_0/2$. And, the Φ_{ext} -dependency in Supplementary Eq. 19 was omitted due to $|cos(\Phi_{ext}/\phi_0)|\simeq 1$ near the optimal point $\Phi_{ext}=\Phi_0/2$.

We thank the reviewer very much for pointing out these typos and have updated the manuscript accordingly.

We think that a ratio g/detuning of 0.12 still qualifies as dispersive regime. Such values are also typical in the dispersive readout of qubits (see, e.g., Gautier, Phys. Rev. Lett. 134, 070802 (2025)). In any case, we have computed the dressed qubit population as a function of the flux bias, showing that it has a negligible participation in the region of the modes hybridization. We now show a figure (Supplementary Fig. 4) demonstrating this for the first three eigenstates. To dispel any remaining ambiguity, the calculation explicitly includes the fourth resonator mode, ensuring that the influence of higher harmonics is fully captured.

We hope that this Figure, together with Fig. 5 in the Supplemental Materials, should clarify this fact.

The analysis is sound. Again, please make sure it is mentioned and properly referenced on the main text.

Reply:

We have updated the manuscript accordingly.

The geometric inductance of the CPW is approximately 0.0004 nH/um along the transmission line, whereas the average inductance across the Josephson junction (JJ) section (aluminum strips and the qubit loop) reaches 0.0075 nH/um. This results in a local inhomogeneity of \sim (0.0075/0.0004) \times 100% \approx 1875%.

My comment was referring to whether the authors considered any other inhomogeneities such as impedance engineering as typically used in parametric donwconversion experiments to avoid degeneracies between modes. Just to make this point clear, I suggest the authors to modify the statement under Eq. 1 of the main text as:

"Owing to the inhomogeneous transmission line geometry due to the qubit presence..."

Reply:

We thank the reviewer for the suggestion and have updated the manuscript accordingly.

Reviewer #3 (Remarks to the Author):

I checked the revised manuscript and their reply to Referee 2 and 3. Again, my overall impression is that the authors do not respond to the referee comments faithfully and the revisions are insufficient. I cannot recommend publication of this manuscript in Nature Communications.

Reply:

We thank the reviewer for the valuable comments. We have addressed each of the

points raised and provide our detailed responses below.

Comments

Regarding comment (1) of Referee 3: By reading authors' response, I could understand that the 1% conversion efficiency is higher than the previous works on SHG using weak input fields. However, I feel that SHG with 1% efficiency is not very impressive in the context of quantum nonlinear optics, since near deterministic down-conversion has been demonstrated already. I recommend the authors to include these comments in the main text so that the readers can recognize the significance of these results.

Reply:

The efficiency was provided in the caption of Fig. 3 in the previous version of the manuscript. We now added a sentence in the main text reporting the estimated efficiency as well as additional comments and a reference to Supplementary Fig. 3. We note that our setup can be directly optimized to approach deterministic operation in the propagation mode (see Ref. 35). To the best of our knowledge, near-deterministic photon down-conversion has so far been achieved predominantly using atomic level transitions [see Ref. 48]. Crucially, our approach has the potentiality to achieve near-deterministic pure photon up/down conversion without involving atomic transitions, thereby avoiding the associated losses.

Regarding comment (2) of Referee 3: I could understand the referees' statement that spontaneously down converted photons are incoherent and cannot be measured through amplitude measurements. The authors' comments are instructive to the readers so should be included in the main text. I also hope to see the authors' explanation on the process in which the incoherent signal (DC photons) can generate coherent signal (Fig.4a).

Reply:

We thanks Reviewer #3 for considering instructive our comments. We have now included them in the main text. Concerning the last sentence of the above comment, the quantum nonlinear process investigated in our work corresponds to degenerate parametric amplification (with modified photonic matrix elements and in the weak amplification limit). Degenerate amplifiers are known to be phase sensitive amplifiers. Already in the previous versions of the manuscript we cited two papers demonstrating phase-sensitive amplification of such amplifiers. We have now added a few lines in the Supplementary Information file to show the origin of phase sensitive amplification of the signal at ω in the presence of a tone at 2ω . The specific full theoretical analysis for the present case, however is based on numerical calculations (see Supplementary Eq. 36) and leads to Fig. 4(b,d).

Regarding comment (4) of Referee 3: The authors state (in previous response) that "We have updated the manuscript to avoid this misleading" and (in this response) that "In

the last reply, we have already changed many instances of "photon pair" to "two photons"". However, in the previous response, they did not revise the title, which is the most important part of the paper. I wonder that the authors correctly recognize the difference between "two-photon Fock state" and "photon pair".

Reply:

To the best of our knowledge, a photon pair refers to the simultaneous or cascade generation of two photons. They do not necessarily constitute a two-photon Fock state. In the present case, the system in the studied configuration is similar to a degenerate parametric amplifier in the very low excitation regime, hence we think that it is not wrong to speak about photon pairs. However, since these belongs to the same mode, it is more precise, to use "two-photon Fock states", as suggested by the Reviewer.

Regarding comment (5) of Referee 3: The authors statement "We used different values for \kappa{in} and \kappa{out}" is obvious. My question was "why the authors set \kappa_{out}=3\kappa_{in}?" and I feel that the authors do not respond to the question.

kappa_{in} and kappa_{out} are the cavity decay rates at the input/output frequencies, which should be determined experimentally in principle and are not the quantities the authors can arbitrarily choose. Since this is an experimental paper, the authors should measure them and assign more reliable parameter values.

Reply:

We are sorry for having missed to answer to the Reviewer's question. We added a paragraph in the Methods sections providing all the answers to the above comment.