nature portfolio

Peer Review File

Spectral Properties of Two Superconducting Artificial Atoms Coupled to a Resonator in the Ultrastrong Coupling Regime

Corresponding Author: Dr Akiyoshi Tomonaga

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

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
In this paper, the authors report on an ultrastrongly-coupled circuit QED setup, where two flux qubits are coupled to a common LC resonator in a regime, where the coupling strengths are comparable to the qubit energies and the resonator frequencies. The system is modeled in terms of an effective cavity QED Hamiltonian with parameters extracted from the measured excitation spectra. The paper then focuses on a higher-order avoided crossing between a single photon state and a state with both of the qubits being excited. This avoided crossing indicates a strong coupling between these two states, such that a single photon can in principle excite two (artificial) atoms. The observation of this feature is the main selling point of this work.

Overall, I find the paper very interesting and suited for publication in Nature Communications. The experimental results are clearly presented and supported by theoretical models. The experiment with two atoms represents a significant advancement in the field of ultrastrong coupling QED, which should be of interest for a broad readership. My only "complaint" about this work is its selling point. The fact that two states that are in resonance exhibit an avoided crossing is not specific to ultrastrongly coupled systems and occurs, essentially, in any non-linear circuit QED system due some higher-order processes. Although the strong coupling helps to observe such an avoided crossing, similar processes are frequently used in driven circuit QED systems to engineer various two- and multi-excitation terms. In addition, in contrast to what is suggested by the title, only the avoided crossing and not the excitation process itself has been experimentally observed.

On the other side, (as far as I am aware) this is the first clean experimental demonstration of an ultrastrongly-coupled cavity QED system with more than one atom. Several years ago, there were many contradictory theoretical prediction about the correct modelling of such systems, including, e.g., the role of the A^2 term or the appearance of a superradiant phase transition [P. Nataf, C. Ciuti, Nat. Commun. 1, 72 (2010), O. Viehmann, J. von Delft, and F. Marquardt, Phys. Rev. Lett. 107, 113602 (2011), M. Bamba, K. Inomata, and Y. Nakamura, Phys. Rev. Lett. 117, 173601 (2016), Ref. [44], etc.] . It seems that the current paper resolves at least .some of these issues at the experimental level. For example, the authors point out the role of the qubit-qubit interaction predicted in Ref. [44], which is necessary to explain the data, but is not included in standard QED Hamiltonians. In my opinion, the experimental clarification of the correct model Hamiltonian for such a system is more significant than the avoided crossing that is then predicted by this model. Therefore, I encourage the authors to place a stronger emphasis on this point, which I believe is most interesting for the community.

Minor detail:
For the derivation of Eq. (5) and (6), the authors cite Ref. [40] and [41]. Neither of those considers the multi-qubit case and the appearance of the qubit-qubit coupling term is not immediately obvious. In particular, the Hamiltonian in Ref. [41], which includes the A^2 term is very different from Eq. (6). Please provide more appropriate references.

Reviewer #2

(Remarks to the Author)
In the current manuscript, the authors experimentally demonstrate the concurrent excitation of two atoms using a single

photon within a straightforward architecture of circuit quantum electrodynamics. Experimental confirmation of theoretical predictions from Ref. 30 regarding the simultaneous excitation of two atoms by a single photon is achieved through the measurement of the circuit spectrum, simulating two atoms ultra-strongly coupled to an LC resonator.

I believe the article is relevant as it effectively translates a theoretically interesting result into the laboratory setting. Importantly, the implementation appears to be reproducible and paves the way for future implementations of multi-qubit gates, photon conversion, and quantum information studies in relation to numerous atoms within circuit quantum electrodynamics (cQED) setups.

Some minor comments could be beneficial for a more thorough understanding and utilization of the paper. For instance, there is no discussion regarding why the system is so robust concerning dissipation or possible sources of noise (such as potential spontaneous decay of qubits or artificial atoms, dissipation processes, and/or dephasing). Specifically, is the readout for obtaining the spectrum done through a transmission line? Why doesn't it induce any form of dissipation? Why are there no references to errors in the measurements of the spectra? I believe these questions should be addressed and clarified in the manuscript, providing relevant orders of magnitude for each process.

Why flux qubits? Why not transmons?

If one were to use atoms with non-anharmonicities (Kerr-type), what is expected to be found? Could one consider exciting higher levels of an atom or a pair of atoms?

A couple of grammar details, for instance, in the introduction, there are a couple of sentences that should have been written more carefully. For example, where it says: 'when is broken the parity symmetry is broken in an atom-light', or also: 'one of the most fascinating theoretical predictions is, when the parity symmetry is broken [27–29], one of the most fascinating is that one photon can simultaneously excite two atoms'.

In the second paragraph of page 4, there is a mention of a 'white arrow' in Fig 3(a) that I couldn't locate.

Reviewer #3

(Remarks to the Author)
This is a review of a manuscript by A. Tomonaga et al. "One photon simultaneously excited two atoms ...". In this work the authors created ultra-strong coupling between two flux qubits and a harmonic oscillator and performed spectroscopy of the resulting 3-degree of freedom "molecule" system as a function of flux in one of the qubit's loop. The resulting spectrum shows a number of anti crossings, all of which are captured by a simple model. The authors interpret one of the anti crossings as a process of "one photons simultaneously excites two qubits". In my opinion this experiment is similar to Ref. 7 from almost 15 years ago where similar splittings have been observed and modelled in a system of 2 oscillators ultra-strongly coupled to 1 flux qubit. I therefore do not see a conceptual advance in the present work.

I believe some of the key statements by the authors are confusing to an average reader. Listing some of them below.

The authors interpret their splitting as a "reverse" process to 2-photon spectroscopy which is widely used in imaging and material characterisation. I believe this is hardly the case. Imaging does not involve any ultra-strong coupling between radiation and the system in question. By contrast the authors coupled flux qubits to an oscillator so strongly that assigning states |gg1> and |ee0> is perhaps no longer meaningful. Just for the fun of it, I superimposed their calculations for g/w << 1 and for g/w ~ 0.5 on top of each other. The image file is entitled Illustration.jpeg and should be attached to this review. In the weak coupling case (semi-transparent plot) one can indeed clearly identify the state |gg1>, which is basically one photon in the resonator and the qubits are in their ground states, this is the flux-independent line. This line crosses another line which has a linear flux dependence in the vicinity of the crossing, this is indeed the state |ee0>, where the flux dependence is predominantly due to one of the qubits. In this case we can indeed see which states are to interact, but they don't interact significantly, because g/w << 1. In case g/w ~0.5 (opaque plot), we see that indeed the blue and red lines are split, but they also are quite different from the uncoupled red and blue lines. The "resonator" excitation, "gg1" states, clearly acquired flux dependence, which means it's not a photon anymore. The qubit frequencies also have shifted, which means that they are not just qubits anymore. So the interpretation put into the title of the paper seems meaningless to me.

Another issue with the presentation is that the authors seem to suggest that the splitting is due to ultra-strong coupling and non-linearity of the qubits. However, in reality, the most important effect is that the qubits are flux-frustrated: should the qubits be biased at their flux sweet spot where their coherence time is maximal, the excitation parity number would need to be conserved and the reported interaction is forbidden. However, the decoherence time of flux qubits away from the flux =0 is very short (the authors do not quote it, but it's probably as bad as 10-100 ns or so), which makes their spectral lines so wide. Because the reported interaction exists only when the qubits have a very low coherence time it will unlikely to lead to further interesting quantum experiments.

Suggestions to improve the paper:

1) Please plot the uncoupled spectrum directly in Fig. 2 and Fig. 3 and label the states of interests. This way the readers will be able to decide themselves whether the interpretation of "one photon excites 2 qubits" has much meaning. In my analysis it does not, because ultra-strong coupling simply turns the system into a "molecule" and flux bias eliminates selection rules,

so all crossing lines in the spectrum would be split to some degree.

2) Please precise the statements of interest. For example, "one of the most fascinating theoretical predictions is, when the parity symmetry is broken [27–29], one of the most fascinating is that one photon can simultaneously excite two atoms [30, 31]."

This seems subjective. And there are many typos.

3) "The reverse phenomenon, i.e., two photon excitation of an atom or molecule, has been adopted for specific spectroscopic instruments [32, 33]."

As explained above, I find this statement misleading, if not wrong, so my suggestion is to remove references to the 2-photon spectroscopy.

4) In the discussion of the spectrum asymmetry with flux, one gets an impression that it is some kind of a fundamental phenomenon. In reality it's probably just a classical flux cross-talk from one qubit to the other + different flux offsets in the two qubits. I would suggest that the authors explain this point more clearly, ideally by writing an explicit Hamiltonian function

5) In relation to 4) please list all the fit parameters.

6) The spectrum is shown in a rather narrow range of flux and frequency. Does their model fit the rest of the spectrum? How can we be sure that the proposed model of Eq. 6 does not overfit the data?

7) The meaning of the bottom-right image in Fig. 3 is unclear to the Referee. If anything, it shows that the photon is no longer just a photon!

8) The authors make a statement that "However, the spin-spin interaction, that is not considered in Ref. 30, reduces the dressing." It is hard to see how this statement can be related to the data. If anything I see the qubit-qubit anti crossing as an anti crossing (once again noting that each qubit is partly a photon...)

9) The author mentioned in the outlook that their results can be extended to "explore, for example, photon down- and up-conversion [29]"

Recently coherent photon down-conversion was reported in N. Mehta et al. (Nature 2023) in a system which seems not too different. It would be good to see a more extended discussion of what kind of novel observations can be reasonably expected from the authors specific demonstration.

Version 2:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
In the revised version of the manuscript, the authors have addressed all of the minor points, which I mentioned in my previous report. They disagreed with my suggestion to focus more on the fundamental aspects of their ultrastrongly coupled two-qubit system, which I personally found most relevant. This is, however, up to personal taste and in the end it should be left up to the authors, how to present and sell their findings. I am happy with the arguments in their reply and the additional clarifications added in the paper.

Reviewer #2

(Remarks to the Author)
The authors have satisfactorily addressed all my questions and comments from the previous review. I have also read the other reports and their corresponding responses, and I am completely satisfied with the new version of the manuscript. I consider it suitable for publication.

Reviewer #3

(Remarks to the Author)
I welcome the revisions of the authors, including the removal of ambiguous statements and supplementary theory plots. Yet, I maintain my opinion that the present experiment is a variation on the one reported in Ref. 7 and we learn nothing new on electrodynamics of quantum superconducting circuits from this one. In Ref 7 it was 1 flux qubit and 2 resonator modes ultra-strongly coupled, here we have 2 flux qubits and 1 resonator mode ultra-strongly coupled. Since superconducting circuits are not real atoms (not fermions), one can think of a coupled system as just 3 non-linear bosonic modes and the experiment is just showing a 3 wave mixing process. The authors are heavily citing the theory proposal by Garziano et al. published in PRL to justify the novelty of their experiment, but this is not a strong argument, in my opinion. A strong argument is a comparison of the present experiment to Ref. 7 NIemczyk et al. Nature Physics 2010 (single photon conversion in a 3-mode

system) and Ref. 42 N. Mahta et al. Nature 2023 (single photon conversion in a massively multi-mode system). I of course welcome any publication decision by the Editors; however, the connection to these two previous works, the description of similarities and differences, should be very clearly spelled out in the introduction to this paper.

Reply to Reviewer #1:

Reviewer

In this paper, the authors report on an ultrastrongly-coupled circuit QED setup, where two flux qubits are coupled to a common LC resonator in a regime, where the coupling strengths are comparable to the qubit energies and the resonator frequencies. The system is modeled in terms of an effective cavity QED Hamiltonian with parameters extracted from the measured excitation spectra. The paper then focuses on a higher-order avoided crossing between a single photon state and a state with both of the qubits being excited. This avoided crossing indicates a strong coupling between these two states, such that a single photon can in principle excite two (artificial) atoms. The observation of this feature is the main selling point of this work.

Overall, I find the paper very interesting and suited for publication in Nature Communications. The experimental results are clearly presented and supported by theoretical models. The experiment with two atoms represents a significant advancement in the field of ultrastrong coupling QED, which should be of interest for a broad readership.

Authors

We thank the Referee for the positive evaluation of our manuscript and the useful comments that helped us to improve our manuscript.

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Reviewer

My only "complaint" about this work is its selling point. The fact that two states that are in resonance exhibit an avoided crossing is not specific to ultrastrongly coupled systems and occurs, essentially, in any non-linear circuit QED system due some higher-order processes. Although the strong coupling helps to observe such an avoided crossing, similar processes are frequently used in driven circuit QED systems to engineer various two- and multi-excitation terms. In addition, in contrast to what is suggested by the title, only the avoided crossing and not the excitation process itself has been experimentally observed.

Authors

Avoided-level crossings are a ubiquitous phenomenon that indicates interaction among different energy levels. The “one photon simultaneously excites two atoms" effect was first predicted in 2016 (Garziano et al, PRL 2016; ref [28] in the revised manuscript), and until now, no experimental evidence had been found of it. This article and the process it predicts gained significant attention, being selected as an "Editors' Suggestion" in Phys. Rev. Lett., with a Focus article dedicated to it [i.e., https://physics.aps.org/articles/v9/83]. Even though the excitation process itself has not been directly observed, the avoided-level crossing provides strong evidence of the existence of this phenomenon. Unfortunately, our sample was not designed for the Rabi oscillation experiment, but this could be achieved in future work.

The manuscript now writes this:

"Clearly, only the avoided-level crossing and not the excitation process itself has been experimentally observed, because our sample was not designed for the Rabi oscillation experiment. However, our observations prove the existence of this excitation."

Reviewer

On the other side, (as far as I am aware) this is the first clean experimental demonstration of an ultrastrongly-coupled cavity QED system with more than one atom.

Several years ago, there were many contradictory theoretical prediction about the correct modelling of such systems, including, e.g., the role of the A^2 term or the appearance of a superradiant phase transition [P. Nataf, C. Ciuti, Nat. Commun. 1, 72 (2010), O. Viehmann, J. von Delft, and F. Marquardt, Phys. Rev. Lett. 107, 113602 (2011), M. Bamba, K. Inomata, and Y. Nakamura, Phys. Rev. Lett. 117, 173601 (2016), Ref. [44], etc.] . It seems that the current paper resolves at least some of these issues at the experimental level. For example, the authors point out the role of the qubit-qubit interaction predicted in Ref. [44], which is necessary to explain the data, but is not included in standard QED Hamiltonians. In my opinion, the experimental clarification of the correct model Hamiltonian for such a system is more significant than the avoided crossing that is then predicted by this model. Therefore, I encourage the authors to place a stronger emphasis on this point, which I believe is most interesting for the community.

Authors

We sincerely thank Reviewer 1 for underlining the importance of other results we obtained. However, in this work, we would rather not enter on the debate about the A^2 term. This is an issue that was sufficiently discussed elsewhere in [Yoshihara *et al*., https://www.nature.com/articles/nphys3906], as well as about the superradiant phase transition, which is also involves a separate debate in the community.

In the current submission, we would like to make this point: the simultaneous excitation of two atoms by a single photon, which is a fundamentally important and novel topic in quantum mechanics, has never been experimentally confirmed in any system before. We present the first experimental spectroscopic evidence of it, which is a very novel result. This first step will motivate others to look for Rabi oscillations.

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Reviewer

Minor detail:

For the derivation of Eq. (5) and (6), the authors cite Ref. [40] and [41]. Neither of those considers the multi-qubit case and the appearance of the qubit-qubit coupling term is not immediately obvious. In particular, the Hamiltonian in Ref. [41], which includes the A^2 term is very different from Eq. (6). Please provide more appropriate references.

Authors

Following the Referee suggestion, we removed Ref. [41] (in the original submission). When discussing the multi-qubit case around Eq. (6), in the first column of page 3, we added a new reference [T. Jaako *et al*., Ultrastrong-coupling phenomena beyond the Dicke model, Physical Review A 94, 033850 (2016)]. We have retained Ref. [40] (in the original submission), because we have found it useful for deriving Eqs. (1) to (5).

Reviewer #2:

Reviewer

In the current manuscript, the authors experimentally demonstrate the concurrent excitation of two atoms using a single photon within a straightforward architecture of circuit quantum electrodynamics. Experimental confirmation of theoretical predictions from Ref. 30 regarding the simultaneous excitation of two atoms by a single photon is achieved through the measurement of the circuit spectrum, simulating two atoms ultra-strongly coupled to an LC resonator.

I believe the article is relevant as it effectively translates a theoretically interesting result into the laboratory setting. Importantly, the implementation appears to be reproducible and paves the way for future implementations of multi-qubit gates, photon conversion, and quantum information studies in relation to numerous atoms within circuit quantum electrodynamics (cQED) setups.

Authors

We thank the Referee for the favorable evaluation of our work and the constructive feedback.

——

Reviewer

Some minor comments could be beneficial for a more thorough understanding and utilization of the paper. For instance, there is no discussion regarding why the system is so robust concerning dissipation or possible sources of noise (such as potential spontaneous decay of qubits or artificial atoms, dissipation processes, and/or dephasing). Specifically, is the readout for obtaining the spectrum done through a transmission line? Why doesn't it induce any form of dissipation? Why are there no references to errors in the measurements of the spectra? I believe these questions should be addressed and clarified in the manuscript, providing relevant orders of magnitude for each process.

Authors

As indicated in the caption of Fig. 1: "The spectrum is measured using a vector network analyzer (VNA) for probing and reading from the transmission line shown below the circuit". Due to the characteristics of superconducting circuits, with the circuit being two-dimensional and the transmission line being considered one-dimensional, we can obtain relatively good signal-to-noise ratio (S/N) spectrum.

The spectra of superconducting circuits have been obtained with a sufficient S/N ratio using similar methods for over 20 years, and our circuit is no exception. The spectrum appears as the absorption of transition energy (frequency) between eigenstates. And the frequency shift in time during spectrum measurement due to noises is usually much smaller than the half width of the Lorentzian absorption. It does not affect the spectrum analysis.

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Reviewer

Why flux qubits? Why not transmons?

Authors

In case of transmons, achieving a coupling ratio over unity is not easy even though a large coupling capacitance is used. For example, in the article by Sal J. Bosman et al. [npj Quantum Inf 3, 46 (2017)] is reached a coupling ratio of g/ω_c = 0.19, while in our case we have g/ω_c = 0.65. Moreover, transmon qubits have low anharmonicity and cannot reproduce the Rabi model.

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Reviewer

If one were to use atoms with non-anharmonicities (Kerr-type), what is expected to be found? Could one consider exciting higher levels of an atom or a pair of atoms?

Authors

If we use a weak anharmonicity system like transmons or Kerr qubits, it might lead to new physics by utilizing higher levels. However, as we mentioned, using a weak anharmonicity system, the coupling constant is not expected to be as high as that of flux qubits. The multilevel ultrastrong coupling system is indeed interesting, but these deviates from the current topic.

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Reviewer

A couple of grammar details, for instance, in the introduction, there are a couple of sentences that should have been written more carefully. For example, where it says: 'when is broken the parity symmetry is broken in an atom-light', or also:

'one of the most fascinating theoretical predictions is, when the parity symmetry is broken [27–29], one of the most fascinating is that one photon can simultaneously excite two atoms'.

Authors

We revise as follows:

"In 2016 a theoretical work claimed that one photon can simultaneously excite two atoms. This effect is observable if the atoms are ultrastrongly coupled with the same cavity mode, and the parity symmetry of the atoms is broken".

——

Reviewer

In the second paragraph of page 4, there is a mention of a 'white arrow' in Fig 3(a) that I couldn't locate.

Authors

We apology for this, the arrows had disappeared. We draw again a black arrow in Figure 3(a).

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Reviewer #3:

Reviewer

This is a review of a manuscript by A. Tomonaga et al. "One photon simultaneously excited two atoms ...". In this work the authors created ultra-strong coupling between two flux qubits and a harmonic oscillator and performed spectroscopy of the resulting 3-degree of freedom "molecule" system as a function of flux in one of the qubit's loop. The resulting spectrum shows a number of anti crossings, all of which are captured by a simple model. The authors interpret one of the anti crossings as a process of "one photons simultaneously excites two qubits".

In my opinion this experiment is similar to Ref. 7 from almost 15 years ago where similar splittings have been observed and modelled in a system of 2 oscillators *ultra-strongly coupled to 1 flux qubit. I therefore do not see a conceptual advance in the present work.*

Authors

Reference 7 (in the original submission) is a Nature Physics article from 2010 that reports the first superconducting circuit in the ultrastrong coupling regime. This article demonstrates an avoided level crossing that indicates excitation exchange between two cavity modes and a flux qubit.

In sharp contrast to this, L. Garziano et al. (PRL, 2016) predicted something different, never studied before 2016: the simultaneous excitation of two atoms by a single photon. Although, superficially, these phenomena may naively appear somewhat similar, they represent fundamentally different processes.

The phenomenon we observed was predicted in 2016, not 2010: six years after the publication of Reference 7. The article by Garziano et al. (PRL, 2016) and the process it predicted received significant attention. It was selected as an “Editors' Suggestion” in Physical Review Letters, was the subject of a Physics magazine Focus article (see: https://physics.aps.org/articles/v9/83) and was covered by various online scientific outlets. We believe that our findings are equally or even more significant than those reported in Reference 7, as we have demonstrated that the process predicted by Garziano et al. (PRL, 2016) is indeed possible and that the theoretical framework used to predict it is accurate.

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Reviewer

I believe some of the key statements by the authors are confusing to an average reader. Listing some of them below.

The authors interpret their splitting as a "reverse" process to 2-photon spectroscopy which is widely used in imaging and material characterization. I believe this is hardly the case. Imaging does not involve any ultra-strong coupling between radiation and the system in question.

Authors

We agree on this point, but we do not consider that our process is, strictly speaking, a reverse process to 2-photon spectroscopy. We just mention the importance of this process (2-photon spectroscopy) and we believe that our result could have a similar impact in the future. Therefore, to avoid possible misunderstandings, we eliminated this sentence.

Please note that the 2016 PRL of Garziano et al. also mentioned it in passing, without any emphasis on this, and no one ever objected to this. Anyhow, to avoid peripheral distractions, that sentence has been removed. It has no effect on the rest of the manuscript.

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Reviewer

By contrast the authors coupled flux qubits to an oscillator so strongly that assigning states |gg1> and |ee0> is perhaps no longer meaningful. Just for the fun of it, I superimposed their calculations for g/w << 1 and for g/w ~ 0.5 on top of each other. The image file is entitled Illustration.jpeg and should be attached to this review. In the weak coupling case (semi-transparent plot) one can indeed clearly identify the state |gg1>, which is basically one photon in the resonator and the qubits are in their ground states, this is the flux-independent line. This line crosses another line which has a linear flux dependence in the vicinity of the crossing, this is indeed the state |ee0>, where the flux dependence is predominantly due to one of the qubits. In this case we can indeed see which states are to interact, but they don't interact significantly, because g/w << 1. In case g/w ~0.5 (opaque plot), we see that indeed the blue and red lines are split, but they also are quite different from the uncoupled red and blue lines. The "resonator" excitation, "gg1" states, clearly acquired flux dependence, which means it's not a photon anymore. The qubit frequencies also have shifted, which means that they are not just qubits anymore. So the interpretation put into the title of the paper seems meaningless to me.

Authors

We understand the point of view of the reviewer; however, Fig.3c shows that before and after the anti-crossing, the states |gg1> and |ee0> have only a small dressing producing a shift in frequencies. Shifts due to dressing are not uncommon in nature (i.e., Lamb shift) and they do not change the physics of the system.

As the reviewer suggested, in Figs. 3b, A1, B2, we overlapped the eigenvalues of the generalized Dicke Hamiltonian with and without interactions considering the shift in frequency of the bare atoms and cavity. As it is possible to see, these coincide, showing that our system can still be considered composed of two independent atoms and one cavity with shifted bare frequencies. This behavior can be justified by the fact that the spin-spin and the spin-light couplings are competing interactions that reduce the dressing.

Moreover, the longitudinal interaction, active when epsilon is not zero, decouples for specific atomic states as a function of the sign of epsilon1 (see Appendix B). However, there is still a residual transverse coupling that dresses our system, as shown in fig.3c.

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Reviewer

Another issue with the presentation is that the authors seem to suggest that the splitting is due to ultra-strong coupling and non-linearity of the qubits. However, in reality, the most important effect is that the qubits are flux-frustrated: should the qubits be biased at their flux sweet spot where their coherence time is maximal, the excitation parity number would need to be conserved and the reported interaction is forbidden. However, the decoherence time of flux qubits away from the flux =0 is very short (the authors do not quote it, but it's probably as bad as 10-100 ns or so), which makes their spectral lines so wide. Because the reported interaction exists only when the qubits have a very low coherence time, it will unlikely to lead to further interesting quantum experiments.

Authors

Indeed, to observe the effect, one photon exciting two qubits, the flux qubit must be slightly away from the optimal point, where the coherence time is shorter. However, in recent years, flux qubit coherence times have increased due to optimization of fabrication processes and designs. For example, the coherence time of a C-shunt flux qubit [F. Yan et al. Nat Commun 7, 12964 (2016)] exceeds 10 μs over a wide frequency, and these are results from about nine or so years ago.

In addition, the bandwidth of the spectrum (absorption lines) is mainly determined by the strength of the coupling between the resonator and the feedline, and it does not directly reflect the coherence time of this system.

For these reasons, we believe that coherence time is not a limiting factor. By selecting appropriate parameters, time-resolved experiments can further validate the existence of this phenomenon.

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Reviewer

Suggestions to improve the paper:

1)Please plot the uncoupled spectrum directly in Fig. 2 and Fig. 3 and label the states of interests. This way the readers will be able to decide themselves whether the interpretation of "one photon excites 2 qubits" has much meaning. In my analysis it does not, because ultra-strong coupling simply turns the system into a "molecule" and flux bias eliminates selection rules, so all crossing lines in the spectrum would be split to some degree.

Authors

We overlapped the non-interacting Hamiltonian's spectrum and the interacting one in Fig.3b, and Figs. A1 A2. In Appendix B we describe the fact that the atoms and the cavity are still independent dressed physical objects.

2) Please precise the statements of interest. For example, "one of the most fascinating theoretical predictions is, when the parity symmetry is broken [27–29], one of the most fascinating is that one photon can simultaneously excite two atoms [30, 31]."

This seems subjective. And there are many typos.

Authors

Thanks for noticing that unfortunate wording. We removed the mentioned sentence, and we write now: "*In 2016, a theoretical work showed that one* *photon can simultaneously excite two atoms [28,29]. This effect is observable if the atoms are ultrastrongly coupled with the same cavity mode, and the parity symmetry of the atoms is broken*".

Also, we carefully checked the manuscript and used a proof-reading service to improve the text.

3) "The reverse phenomenon, i.e., two photon excitation of an atom or molecule, has been adopted for specific spectroscopic instruments [32, 33]."

As explained above, I find this statement misleading, if not wrong, so my suggestion is to remove references to the 2-photon spectroscopy.

Authors
Those peripheral and marginal sentences are not essential at all, and have been removed.

4) In the discussion of the spectrum asymmetry with flux, one gets an impression that it is some kind of a fundamental phenomenon. In reality it's probably just a classical flux crosstalk from one qubit to the other + different flux offsets in the two qubits. I would suggest that the authors explain this point more clearly, ideally by writing an explicit Hamiltonian function

Authors
This asymmetry is a fundamental phenomenon which comes directly from studying the generalized Dicke model. This asymmetry originates from the longitudinal interaction, as explained in Appendix B. The crosstalk between the qubits is already considered in Eq. (6), here the energy bias of qubit 2 is modified by

epsilon2 —> epsilon2 + A × epsilon1, where A = −9.43 × 10e−3.

This is explained in the section "Energy spectrum", on page 3.

5) In relation to 4) please list all the fit parameters.

Authors

All the eleven fitting parameters are listed in page 4 in the main text of the new version. The new set of parameters provides a better fit.

6) The spectrum is shown in a rather narrow range of flux and frequency. Does their model fit the rest of the spectrum? How can we be sure that the proposed model of Eq. 6 does not overfit the data?

Authors

We now explain in more detail that the fit is reasonable and physically correct in the section "Energy spectrum" and the caption for Table 1. In the caption of table 1, we write: "We use the first eight listed parameters for an initial fit. Afterwards, we add the last three parameters to slightly modify the fitting. The initial parameters $\omega_r, \tilde{\varepsilon}_0, \text{and } I_{b0}$ can be obtained from the spectrum. Also, the qubit parameters *g* and Δ can be estimated from the design parameters." As we write in the main text, the design of the two qubits is nominally identical and the fitting parameters for two qubits have similar values.

Additionally, the energy calculations for the flux qubit are based on an approximation that is most accurate around a flux value of 0.5. This is a general characteristic of circuit Hamiltonian models for flux qubits, and when the flux deviates significantly from 0.5, a different model would be required. Since our study focuses on the physics near a flux value of 0.5, we believe our model is appropriate and valid in this context.

7) The meaning of the bottom-right image in Fig. 3 is unclear to the Referee. If anything, it shows that the photon is no longer just a photon!

Authors

In Figure 3(c) the projection of the third and fourth eigenstates to the bare states gg1 and ee0 is plotted in correspondence of the avoided-level crossing where these states hybridize. The plot shows that the large couplings do not significantly alter their nature, even if they generate a small dressing.

8) The authors make a statement that "However, the spin-spin interaction, that is not considered in Ref. 30, reduces *the dressing." It is hard to see how this statement can be related to the data. If anything I see the qubit-qubit anti crossing as an anti-crossing (once again noting that each qubit is partly a photon...)*

Authors

We deleted this specific sentence, because its wording was ambiguous. In the section "One photon simultaneously excites two atoms" now we add this text:

> *"On the contrary, Fig. 3c shows that the dressing is low for those states, and, as shown in Appendix B, our system can still be considered formed by separated two two-level atoms and one cavity mode with shifted eigenfrequencies. This behavior is heuristically justified by the fact that spin–spin and spins–light couplings are competing interactions"*

We do not give a demonstration in this manuscript; we found this suppression heuristically noting that the dressing is higher if only one of the two couplings is active. In the section "Discussion" we add:

> *"Theoretically, the reduction of dressing and the asymmetry in the spectrum could be further investigated. "*

9) The author mentioned in the outlook that their results can be extended to "explore, for example, photon down- and up-conversion [29]"

Recently coherent photon down-conversion was reported in N. Mehta et al. (Nature 2023) in a system which seems not too different. It would be good to see a more extended discussion of what kind of novel observations can be reasonably expected from the authors specific demonstration.

Authors

Thank you for this advice. We added that reference, but they use a very different type of qubit, a fluxonium. We also add another reference "K. Inomata et al., Microwave Down-Conversion with an Impedance-Matched Λ System in Driven Circuit QED, PRL (2014)". We also expect similar photon down- and up-conversion experiments in our circuit, in the future.

Reviewer #1 (Remarks to the Author):

In the revised version of the manuscript, the authors have addressed all of the minor points, which I mentioned in my previous report. They disagreed with my suggestion to focus more on the fundamental aspects of their ultrastrongly coupled two-qubit system, which I personally found most relevant. This is, however, up to personal taste and in the end it should be left up to the authors, how to present and sell their findings. I am happy with the arguments in their reply and the additional clarifications added in the paper.

Answer
Thank you for your comment. Regarding your suggestion to focus more on the fundamental aspects of the two-qubit system, we have reconsidered it. As a result, we have revised the title, abstract, and introduction to better highlight these fundamental aspects of the study. We hope that these revisions improve the overall balance of the manuscript and make it more accessible to a broader audience.

Reviewer #2 (Remarks to the Author):

The authors have satisfactorily addressed all my questions and comments from the previous review. I have also read the other reports and their corresponding responses, and I am completely satisfied with the new version of the manuscript. I consider it suitable for publication.

Answer
Thank you for your positive feedback. We are pleased to hear that you are satisfied with the revised version of the manuscript. Your support and careful consideration of the revisions are greatly appreciated.

Reviewer #3 (Remarks to the Author):

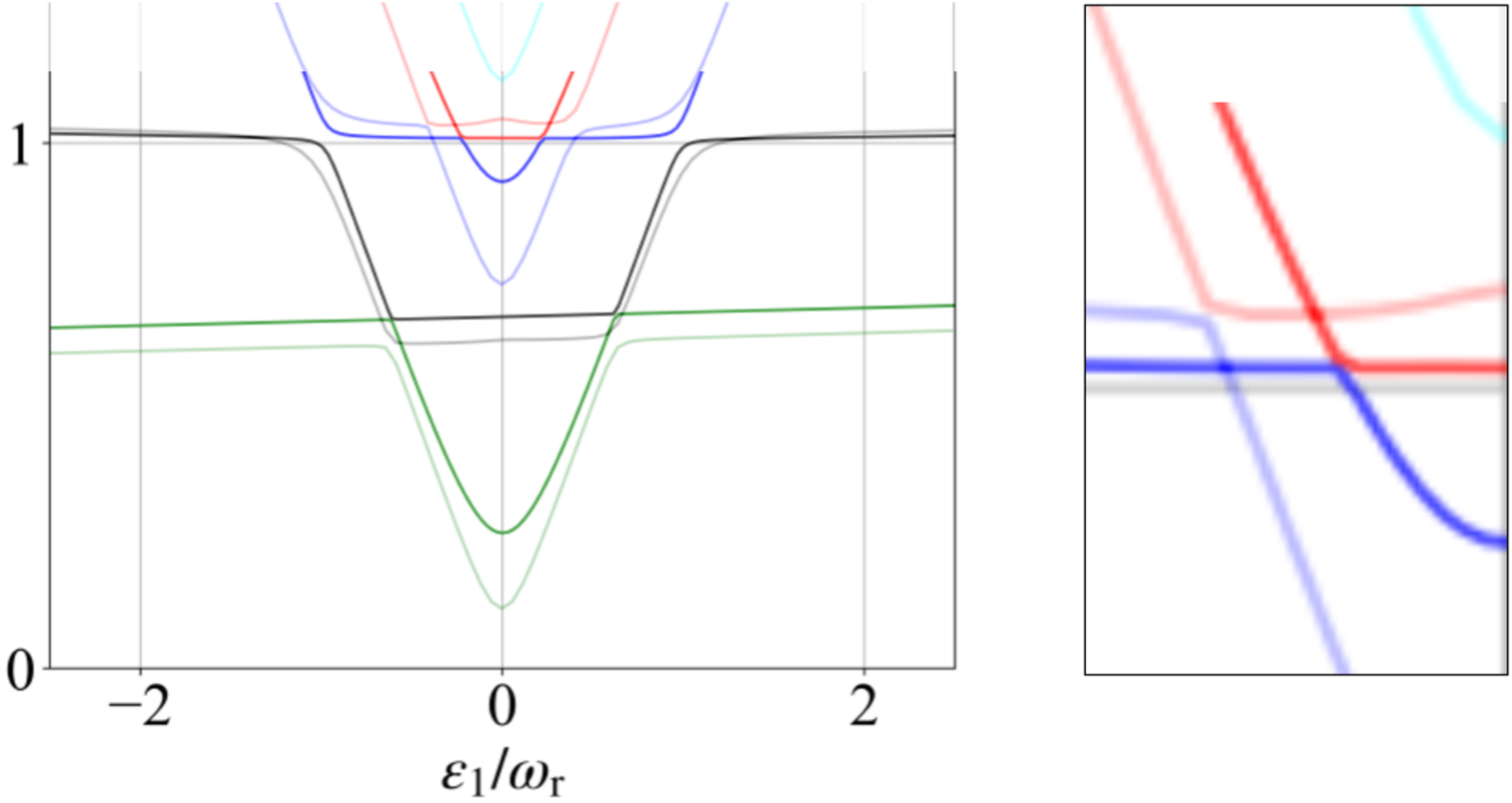
I welcome the revisions of the authors, including the removal of ambiguous statements and supplementary theory plots. Yet, I maintain my opinion that the present experiment is a variation on the one reported in Ref. 7 and we learn nothing new on electrodynamics of quantum superconducting circuits from this one. In Ref 7 it was 1 flux qubit and 2 resonator modes ultra-strongly coupled, here we have 2 flux qubits and 1 resonator mode ultra-strongly coupled. Since superconducting circuits are not real atoms (not fermions), one can think of a coupled system as just 3 non-linear bosonic modes and the experiment is just showing a 3 wave mixing process. The authors are heavily citing the theory proposal by Garziano et al. published in PRL to justify the novelty of their experiment, but this is not a strong argument, in my opinion. A strong argument is a comparison of the present experiment to Ref. 7 NIemczyk et al. Nature Physics 2010 (single photon conversion in a 3-mode system) and Ref. 42 N. Mahta et al. Nature 2023 (single photon conversion in a massively multi-mode system). I of course welcome any publication decision by the Editors; however, the connection to these two previous works, the description of similarities and differences, should be very clearly spelled out in the introduction to this paper.

Answer

We have included both references in the introduction.

Our work and that of T. Niemczyk et al. belong to the same class of phenomena that are only observable in the ultrastrong coupling regime, but both are important for different reasons. While a multimode cavity coupled to a single atom, as in Niemczyk et al., allows frequency-selective interactions, its photon conversion capability is limited by mode spacing. In contrast, multiple atoms coupled to a single cavity (as in our work) can facilitate the conversion of a single photon into multiple lower-frequency excitations. Despite superficial similarities, these two circuits are different in configuration and serve distinct roles.

On the other hand, a recent experiment by the group of Manachuryan, based on many-body localization (MBL), not studied in our work, shows the suppression of the photon down-conversion, due to MBL. This is in stark contrast to our system, which is designed to actively promote both down- and up-conversion processes. Moreover, that work by the group of Manachuryan operates in what they call superstrong coupling regime, which arises when the atomic linewidth exceeds the cavity mode spacing. The resulting three-wave mixing interaction induces multiphoton processes across many modes, which is not our case. Furthermore, ultrastrong coupling, as realized in our work, occurs in a different regime: when the light–matter coupling strength exceeds 10% of the cavity photon energy. This system is described by a Generalized Dicke Hamiltonian including counter-rotating terms, leading to strong hybridization of the qubits with a cavity mode.



1
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−2
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2
$\varepsilon_1/\omega_\mathrm{r}$