
Supplementary information

A quantum enhanced search for dark matter axions

In the format provided by the
authors and unedited

Peer Review File

Manuscript Title: A Quantum-Enhanced Search for Dark Matter Axions

Editorial Notes:**Redactions – Mention of other journals**

This document only contains reviewer comments, rebuttal and decision letters for versions considered at *Nature*. Mentions of the other journal have been redacted.

Reviewer Comments & Author Rebuttals**Reviewer Reports on the Initial Version:**

Referee #1 (Remarks to the Author):

The work presented in this article has been carried out as part of the HAYSTAC experiment to look for dark matter (DM) axions. The result presented includes a new experimental run in a particular axion mass range (4.1 to 4.2 microelectron-volts), producing a new excluded region that improves previous bounds in this range. The significance of the work resides not so much in this result, but in the fact that it is the first time a squeezed quantum vacuum state is used to reduce the experimental noise and improve the effective scan rate. It is known that conventional axion haloscope searches suffer from a limit to the potential noise reduction imposed by the uncertainty principle, and that leading experiments (like that of the authors) have been getting close to such “quantum limit”. The preparation of the cavity vacuum as a squeezed photon state could reduce (some component of) the experiment noise, effectively going beyond this limit. The actual improvement achieved in this paper is equivalent to a factor of 2 in scanning rate (that is, the same sensitivity would be achieved by a run without squeezing but twice longer in exposure). Although this does not seem like a quantitative breakthrough, the relevance of this result is qualitatively very high, as it opens the door to break the quantum limit and obtain further improvements in the future.

The work is original, as it is the first time the concept of squeezing is experimentally applied in a running axion experiment. I consider this work will become a reference for axion experimenters. Although the impact is mostly circumscribed inside the field of axion and dark matter searches, I anticipate some interest beyond it, as the result can be considered an application of quantum information technology to axion searches. Whether this is enough to warrant a publication in *Nature*, in contrast to, e.g. in [REDACTED], I leave it to the consideration of the editor in view of the other referee reports. The methodology is adequate and the authors present it clearly. The text is well written and, although the descriptions are succinct, the authors count with a few excellent references of previous works of them that include exhaustive descriptions of the experimental setup or the analysis procedure.

In summary, I consider this is first-level article and totally recommend its publication. I want to congratulate the authors for the excellent and pioneering work, that is guiding and enlightening that of the whole community in search for axions.

I have two small comments / suggestions to the text that I would invite the authors to consider, but they are of small importance and they do not condition my recommendation for publication:

1) In figure 2a and in the related discussion in the text it is not clear the units that the author chose to represent the noise components. From the plot label "PSD/vacuum" and the values of the plot, I would guess that what is shown is the fraction of the noise PSD that comes from the reflected or the internal components respectively. But this is not clearly stated nowhere (I think) in the text or caption.

2) The last sentence of the paper mentions the prospect of getting better scan rate increases in future developments. Given the importance of this statement (in fact the impact of the work depends on the feasibility of going to improvement factors potentially much larger than the one obtained in this first work), I would appreciate if the authors devote a few sentences with a discussion of the feasibility of such possibility. For example, the authors mention greater-than-tenfold factors. How feasible is this and what would that entail?

Referee #2 (Remarks to the Author):

Summary of key results:

Dark matter detection using axion haloscopes require the precise measurement of low-signal electromagnetic field emitted from a high-Q cavity with large magnetic fields at cryogenic temperatures. Previous experiments have been constrained by quantum (Nyquist) noise and the bandwidth of the high-Q detector cavity, therefore limiting the scan-rate of a detector and the range of detection within a given time.

In this report, the authors demonstrate the utility of quantum squeezing to improve the dark matter axion search. In particular, the authors show a x1.9 enhancement in scan rate beyond what is achievable by the standard quantum limit and are able to exclude a region of mass between 16.92 -17.12 and 17.14-17.28 $\mu\text{eV}/c^2$.

The key result here is the integration of delicate quantum computing technology (namely two flux-pumped Josephson parametric amplifiers, JPAs) with equipment required for axion detection (e.g. 8T magnetic field, long-time stability). The authors are able to show equivalent squeezing and amplification performance [Ref 9, Malnou et al] in their detection setup [Ref 3, Brubaker et al] and then use this setup to perform a 'sub-quantum-limited' search of a region of masses. This results in a nearly two-fold improvement in scan-rate compared to an 'un-squeezed' measurement protocol.

Originality and significance:

The authors demonstrate the successful integration of disparate technologies to improve dark-matter axion searches. While the squeezing/amplification setup has been demonstrated [Ref 9, Malnou et al] and characterised, the rig had not been integrated in an axion-detector setup (namely with magnetic fields up to 8T) requiring long-time stability over month-long collection periods [Ref 3, Brubaker et al].

The significance of this manuscript is that these technologies are working in tandem and that squeezing using a JPA is a useful tool for enhanced axion detection which speeds up the detection scan rate. Future axion detection setups will likely use this technique for faster data collection.

Data & Methodology:

Much of the squeezing/amplification theory and characterisation methods have already been

introduced in more detail in [Ref 9, Malnou et al] and the axion detection methods in [Ref 3, Brubaker et al]. The authors use these same methods to characterise squeezing and in turn compare the amount of noise which can be attributed to the amplifier/squeezer added noise (N_a) vs noise added by Johnson-Nyquist noise due to cavity internal loss (N_c).

The analysis of the squeezing/amplifier setup appears sound to me. The authors use two methods to determine noise added in their measurement chain (using a 50ohm load at different temperatures).

The authors find the cavity resonator somewhat 'hotter' than expected which is typical in many cryogenic setups. I appreciate this careful measurement and their reported action to ameliorate its effects.

I do have some confusion on their reported 'squeezing' versus their reported theoretical maximum (see clarification section).

My area of expertise lies outside axion searches and can therefore not speak towards their data analysis for determining mass exclusions. I do however appreciate that analysis was performed by two separate teams with 1% agreement.

Conclusions:

This work strongly motivates the integration of squeezing for quantum-enhanced axion searches. The near two-fold improvement in scan-rate demonstrated here has immediate utility and has a good outlook for further improvements in detector efficiency.

This work demonstrates that quantum computing technology, namely microwave quantum squeezers and amplifiers can be used in tandem with detection hardware to generate an enhanced axion search-rate. Quantum-limited amplifiers have been used in previous works so the ability to add of squeezers may seem obvious. However, the stability and operation of all this equipment in unison is a useful result.

Overall, I find the design, implementation, and results of this experiment sound. I believe the results will have a positive contribution to the field of axion detection which may dramatically change the way detectors are built in the future. However in my opinion I find the novelty of the result diminished as the methods have already been demonstrated in better detail elsewhere [Ref 9, Malnou et al]. I find it disappointing that no new outlooks have been drawn compared to this previous work.

Additional clarification:

Why is the measured squeezing ($S=0.40$) larger than the expected maximum ($S=0.37$)? Is this due to the approximate value for efficiency ($\eta = 0.63$) being overestimated?

How does amplifier/squeezer bandwidth affect future experiments? I cannot find an amplifier bandwidth reported here so I presume in these devices, bandwidth is far from becoming a limiting factor.

What particular challenges were there in this experiment as opposed to the works in [Ref 9, Malnou et al], [Ref 3, Brubaker et al]? Was stability more of an issue due to twice the number of Josephson devices (which both need to be phase-locked)?

Referee #3 (Remarks to the Author):

* Key results

The paper presents a search for dark matter axions at relatively high frequency compared to other microwave cavity searches, and reaching sensitivity close to the benchmark QCD axion. The nature of such searches is that the given frequency range, while narrow, has not been probed before. The key point of the paper, however, is that this sensitivity has been achieved exploiting a novel measurement procedure. The signal is ``squeezed' in phase space, overcoming the standard quantum limit for an unsqueezed distribution. The squeezing procedure widens the bandwidth of the experiment, and allows a wider range of frequencies to be searched in a shorter measurement time.

* Validity

I can see no immediate problems with the validity, however see my expertise below.

* Significance

Searches for the QCD axion are appearing more and more around the world. This is a hot topic at the moment. There are many technologies being developed. The present work uses the tried-and-tested microwave cavity, but enhanced with the squeezing technique to extend its usefulness to higher frequencies, clearly with an eye to future campaigns.

The frequency probed is novel, and the sensitivity is close to the QCD axion range (although not yet reaching KSVZ or DFSZ). This method will surely be useful to other axion searches operating at high frequency in the future, a significant technological step forwards for the field.

* Data and methodology/Analytical approach

The methodology appears extremely sound is based on well documented and clear methods previously published by the HAYSTAC collaboration. That being said, I comment below on places where the presentation could be made more clear. Many symbols are undefined, and I think a short review of the relevant papers in the Methods section would be helpful to the reader.

* Suggested improvements

1) Recent theoretical predictions of the axion mass, discussion of refs. [10,11].

a) The discussion makes out like these predictions are completely model independent, and uncontroversial, but, as I am sure the authors know, many caveats apply.

The authors should refer to Gorghetto et al's two papers on this subject, which I believe to be superior. After including many sources of uncertainty in the extrapolations taking simulations to the physical coupling values, they conclude that only a lower limit on the axion mass can be placed, and it is of order 1 meV (this in itself is not uncontroversial either).

Furthermore, all of these theoretical predictions depend on cosmology, and the assumption that there is only a single DM particle. The cosmology dependence assumes that the Universe had a maximum temperature higher than the PQ transition, and that all physics below that temperature is known in the Standard Model. Both of these facts are clearly very large assumptions.

In low scale inflation (pre-inflation symmetry breaking), as is well known, a very wide axion mass range is allowed and the theoretical predictions cited do not apply. Adopting a uniform prior on the misalignment angle and demanding that axions are all the DM in this case leads to the 95% C.L.

posterior on the mass $0.53 \mu\text{eV} \leq m \leq 0.13 \text{ meV}$ (GAMBIT axion global fits, <https://arxiv.org/abs/1810.07192>). The present work is square in the middle of this range.

If more DM candidates are allowed (e.g. axions+WIMPs), then the mass predictions of [10,11] become lower bounds.

Finally, even taking the statement in the abstract completely with these caveats, the predictions only apply to $N=1$. For $N > 1$ (e.g. DFSZ), the prediction was made by the Kawasaki group for m . With the least amount of fine-tuning to solve the strong CP problem, the prediction for $N > 1$ saturates at the SN1987A bound, $m \sim 5\text{--}10 \text{ meV}$.

The above considerations should be made in brief, and Refs. [10,11] should not be give such prominence in setting an expected mass range. I would agree with something along the lines of "the range $m > 10 \mu\text{eV}$ favoured when the Peccei-Quinn symmetry is broken during the hot big bang, and not during inflation" and then an expanded list of references. Maybe add something about ``about half of the posterior volume in $\log(m)$ in the alternative scenario for symmetry breaking assuming a uniform prior on the initial axion field value'.

b) Later, Refs. [10,11] are fundamentally misunderstood. These are NOT Lattice QCD. They are lattice classical field theory for the Peccei-Quinn field and include no QCD whatsoever. This statement must be corrected.

Recent lattice QCD simulations were carried out by Borsanyi et al to determine the temperature dependence of the axion mass, but they do NOT simulate the lattice classical theory of string decay. Moore's results did not use the Borsanyi et al QCD-calculated $m(T)$. Foster et al use a parameterization of $m(T)$ and discuss the uncertainties from it coming from lattice QCD, which are around 27% on the mass prediction.

2) Why is Ref. [33] used for the axion model band? This seems limited, and out of date. A more complete and recent study is by di Luzio et al, which gives the maximum of the band in terms of the ratio E/N in self-consistent models. This was adopted in the GAMBIT global fit of axion models cited earlier. The model band is reviewed in <https://arxiv.org/abs/2003.01100> I suggest to either include a more detailed view of the band following di Luzio and the GAMBIT analysis, or to simply show KSVZ and DFSZ in Fig. 3.

In Fig. 3 the red band at just slightly higher frequency than the present analysis is not labelled. Is it the ORGAN result, or the QUAX (photon coupling) result?

3) Why does the axion mass window scanned have a gap from 17.12 to 17.14 micro eV? I presume this is some background noise frequency or troublesome resonance of the apparatus. Please explain.

4) Presentation/discussion of squeezing

I struggle to fundamentally understand what happens when ``the axion displaces the squeezed state from the origin". Clearly, this must be done by the inverse Primakoff process where the axion creates a photon in the presence of a magnetic field. However this is not actually stated anywhere in the manuscript. The authors should explain a bit more how this interaction relates to the Hamiltonian (1).

How is the power generated by the axion related to the shift in the operator X ? The authors should improve the description of squeezing so that the link to the particle physics model is clearer. I think this would improve the wider readability of the manuscript.

I understand that squeezing increases the scan rate. But I do not understand what ``squeezing

N_r means. The integral on $\delta\nu$ is a one dimensional integral, and N_r is written as a function of a single variable, not two, one of which could be squeezed.

Can the authors give slightly more explanation of how Fig. 2a is arrived at, perhaps with reference formulae given in a supplemental? The role of the parameter S for the deliverable squeezing is not clear. How does it enter in Eq. 2?

S is minimized with the parameter η of the experiment. Could this in principle be improved, or is it a fundamental limit to this technology?

I feel like the description of squeezing came a bit backwards. I was always asking questions which were answered about a half page later. To me it seems most logical to define S , the measure of squeezing, and the noise quantities, then give R , then α . $S_{\rm ax}$ is then clearly the power given by the familiar formulae for a cavity. Then finally you could say "this is understood in quantum mechanics by thinking of Eq. 1 ...". I find starting with the quantum picture confusing.

5) It would be nice to see somewhere in the conclusions a discussion about the future of HAYSTAC. Will this technology be used to go "wider" or "deeper"? Plans for DFSZ? Plans for other frequencies?

What about other detection methods? How does the present method using squeezing+heterodyne stack up against a (hypothetical) microwave single photon counter?

* Clarity and context

There are many undefined symbols used.

The axion-photon coupling is not defined. The cavity form factor is not defined. The cavity power is not given. What model was used for the axion linewidth/lineshape? How do uncertainties in the halo model impact the exclusions? What is the "axion distribution $\mathcal{A}_i$ "?

A slightly longer description of Ref. [30] in the Methods section is required.

* Your expertise

I am not an experimentalist, so the details of the actual procedure of measurement, and some areas of the statistical analysis are outside of my expertise.

Comments from a colleague on these points:

My brief and incomplete impression is that this is an interesting entry in HAYSTAC's list of publications that seek to improve haloscope sensitivity via improvements in software or quantum-limited hardware. It is an intriguing proposal to use a squeezed thermal state prior to the cavity to boost SNR, though it is not clear to me how this would favorably impact the sum of the squeezed resistor + cavity noise. I would be interested to know how they confirmed their sensitivity increase, as I do not recall seeing discussion of hardware signal injections or other common approaches. But again, this was only after a brief reading.

Author Rebuttals to Initial Comments:

Reviewer 1's Report:

The work presented in this article has been carried out as part of the HAYSTAC experiment to look for dark matter (DM) axions. The result presented includes a new experimental run in a particular axion mass range (4.1 to 4.2 microelectron-volts), producing a new excluded region that improves previous bounds in this range. The significance of the work resides not so much in this result, but in the fact that it is the first time a squeezed quantum vacuum state is used to reduce the experimental noise and improve the effective scan rate. It is known that conventional axion haloscope searches suffer from a limit to the potential noise reduction imposed by the uncertainty principle, and that leading experiments (like that of the authors) have been getting close to such “quantum limit”. The preparation of the cavity vacuum as a squeezed photon state could reduce (some component of) the experiment noise, effectively going beyond this limit. The actual improvement achieved in this paper is equivalent to a factor of 2 in scanning rate (that is, the same sensitivity would be achieved by a run without squeezing but twice longer in exposure). Although this does not seem like a quantitative breakthrough, the relevance of this result is qualitatively very high, as it opens the door to break the quantum limit and obtain further improvements in the future.

The work is original, as it is the first time the concept of squeezing is experimentally applied in a running axion experiment. I consider this work will become a reference for axion experimenters. Although the impact is mostly circumscribed inside the field of axion and dark matter searches, I anticipate some interest beyond it, as the result can be considered an application of quantum information technology to axion searches. Whether this is enough to warrant a publication in Nature, in contrast to, e.g. in [REDACTED], I leave it to the consideration of the editor in view of the other referee reports. The methodology is adequate and the authors present it clearly. The text is well written and, although the descriptions are succinct, the authors count with a few excellent references of previous works of them that include exhaustive descriptions of the experimental setup or the analysis procedure.

In summary, I consider this is first-level article and totally recommend its publication. I want to congratulate the authors for the excellent and pioneering work, that is guiding and enlightening that of the whole community in search for axions.

I have two small comments / suggestions to the text that I would invite the authors to consider, but they are of small importance and they do not condition my recommendation for publication:

General response: We are happy that reviewer #1 found our work original and worthy of publication. We have responded to their two comments below. Both comments were useful in helping us clarify our work.

- In figure 2a and in the related discussion in the text it is not clear the units that the author chose to represent the noise components. From the plot label “PSD/vacuum” and the values of the plot, I would guess that what is shown is the fraction of the noise PSD that comes from the reflected or the internal components respectively. But this is not clearly stated nowhere (I think) in the text or caption.

Response: The “vacuum” in “PSD/vacuum” is a quantity with dimensions of power spectral density (PSD). Therefore “PSD/vacuum” refers to the noise PSD normalized to the single- quadrature vacuum value of $h\nu/4$, and not quite to the noise fraction from each source (although the two are equal in the unsqueezed case). We have added “PSDs are normalized to their single quadrature vacuum value of $h\nu/4$ ” to the figure caption to clarify this.

- The last sentence of the paper mentions the prospect of getting better scan rate increases in future developments. Given the importance of this statement (in fact the impact of the work depends on the feasibility of going to improvement factors potentially much larger than the one obtained in this first work), I would appreciate if the authors devote a few sentences with a discussion of the feasibility of such possibility. For example, the authors mention greater-than-tenfold factors. How feasible is this and what would that entail?

Response: We have added language to the final sentence of the paper alluding to one way that scan rates might be significantly improved: in particular, through a parametric swapping interaction between the axion cavity and the JPA circuit, which would allow for the elimination of lossy, nonreciprocal elements. We note for context that this technique is currently a field of active research by our colleagues at JILA/CU Boulder, where there are plans to feature it in its own publication. It represents the most exciting and potentially transformative path forward that we know of for advancing the technology presented in our paper. We therefore chose to highlight it over more mundane avenues for improvement such as through microwave technologies becoming steadily less lossy over time. We consider the feasibility of improvements along these or other avenues to be high, but it is beyond the scope of this work to explore the technical considerations that lead us to making that judgement.

Reviewer 2's Report:

Summary of key results:

Dark matter detection using axion haloscopes require the precise measurement of low-signal electromagnetic field emitted from a high-Q cavity with large magnetic fields at cryogenic temperatures. Previous experiments have been constrained by quantum (Nyquist) noise and the bandwidth of the high-Q detector cavity, therefore limiting the scan-rate of a detector and the range of detection within a given time.

In this report, the authors demonstrate the utility of quantum squeezing to improve the dark matter axion search. In particular, the authors show a $\times 1.9$ enhancement in scan rate beyond what is achievable by the standard quantum limit and are able to exclude a region of mass between 16.92 – 17.12 and 17.14 – $17.28 \mu\text{eV}/c^2$.

The key result here is the integration of delicate quantum computing technology (namely two flux-pumped Josephson parametric amplifiers, JPAs) with equipment required for axion detection (e.g. 8T magnetic field, long-time stability). The authors are able to show equivalent squeezing and amplification performance [Ref. 9, Malnou et al] in their detection setup [Ref. 3, Brubaker et al] and then use this setup to perform a ‘sub-quantum-limited’ search of a region of masses. This results in a nearly two-fold improvement in scan-rate compared to an ‘un-squeezed’ measurement protocol.

Originality and significance:

The authors demonstrate the successful integration of disparate technologies to improve dark-matter axion searches. While the squeezing/amplification setup has been demonstrated [Ref. 9, Malnou et al] and characterised, the rig had not been integrated in an axion-detector setup (namely with magnetic fields up to 8T) requiring long-time stability over month-long collection periods [Ref. 3, Brubaker et al]. The significance of this manuscript is that these technologies are working in tandem and that squeezing using a JPA is a useful tool for

enhanced axion detection which speeds up the detection scan rate. Future axion detection setups will likely use this technique for faster data collection.

Data & Methodology:

Much of the squeezing/amplification theory and characterisation methods have already been introduced in more detail in [Ref. 9, Malnou et al] and the axion detection methods in [Ref. 3, Brubaker et al]. The authors use these same methods to characterise squeezing and in turn compare the amount of noise which can be attributed to the amplifier/squeezer added noise (N_a) vs noise added by Johnson-Nyquist noise due to cavity internal loss (N_c). The analysis of the squeezing/amplifier setup appears sound to me. The authors use two methods to determine noise added in their measurement chain (using a 50ohm load at different temperatures). The authors find the cavity resonator somewhat 'hotter' than expected which is typical in many cryogenic setups. I appreciate this careful measurement and their reported action to ameliorate its effects. I do have some confusion on their reported 'squeezing' versus their reported theoretical maximum (see clarification section). My area of expertise lies outside axion searches and can therefore not speak towards their data analysis for determining mass exclusions. I do however appreciate that analysis was performed by two separate teams with 1% agreement.

Conclusions:

This work strongly motivates the integration of squeezing for quantum-enhanced axion searches. The near two-fold improvement in scan-rate demonstrated here has immediate utility and has a good outlook for further improvements in detector efficiency.

This work demonstrates that quantum computing technology, namely microwave quantum squeezers and amplifiers can be used in tandem with detection hardware to generate an enhanced axion search-rate. Quantum-limited amplifiers have been used in previous works so the ability to add of squeezers may seem obvious. However, the stability and operation of all this equipment in unison is a useful result.

Overall, I find the design, implementation, and results of this experiment sound. I believe the results will have a positive contribution to the field of axion detection which may dramatically change the way detectors are built in the future. However in my opinion I find the novelty of the result diminished as the methods have already been demonstrated in better detail elsewhere [Ref. 9, Malnou et al]. I find it disappointing that no new outlooks have been drawn compared to this previous work.

General response: We thank reviewer #2 for their thoughtful comments. They have helped us clarify several aspects of the discussion of the squeezed state receiver.

Additional clarification:

- Why is the measured squeezing ($S=0.40$) larger than the expected maximum ($S=0.37$)? Is this due to the approximate value for efficiency ($\eta = 0.63$) being overestimated?

Response: We realize that our phrasing was confusing. Squeezing is colloquially discussed as a desirable thing which it is good to maximize. Mathematically, however, we define squeezing in power units as a ratio of variances ranging from 0 to 1, with lower quantities indicating stronger squeezing. Therefore, our measured value of $S = 0.40$ indeed fails to realize the expected best value (what we misleadingly called a "maximum") of $S = 0.37$. To avoid such confusion, we switched "maximum" to "optimum."

- How does amplifier/squeezer bandwidth affect future experiments? I cannot find an

amplifier bandwidth reported here so I presume in these devices, bandwidth is far from becoming a limiting factor.

Response: The reviewer is correct. We have added the approximate average bandwidths for both the squeezer (~ 26 MHz) and amplifier (~ 3 MHz) JPAs to the Methods section and noted that they are far from limiting.

- What particular challenges were there in this experiment as opposed to the works in [Ref. 9, Malnou et al], [Ref. 3, Brubaker et al]? Was stability more of an issue due to twice the number of Josephson devices (which both need to be phase-locked)?

Response: The main challenges present in this work vs [Ref. 3, Brubaker et al] is the need to tune a second JPA in tandem with the first and optimize squeezing, as well as additional calibrations needed to fully characterize the squeezed state receiver. The simultaneous tuning in particular opened up a host of new technical challenges, including frequency- and phase- locking the pumps of the two JPA and “orthogonalizing” their tuning coils in software, which due to spatial constraints imposed by the magnetic field profile had to be mutually adjacent, allowing each JPA’s coil to inadvertently bias the other. Finally, because this work deals with quantum states of the microwave fields, intra-receiver losses had to be at an absolute minimum in our present work, whereas our former, classical measurement was much more forgiving. Reference numbers correspond to the revised manuscript.

Compared with [Ref. 9, Malnou et al], new challenges include: spectrally tuning the squeezed state receiver (including both JPAs as well as the TM_{010} -like mode of the cavity), coupling to a many-times larger cavity, and operation of superconducting circuitry in the vicinity of an 8 T magnet. We also had to introduce additional calibrations not needed in Ref. 9, and pay sharper attention to live-time considerations. We have added some of these comparisons in the methods section of the paper to highlight the key challenges overcome in this work which were not present in our non-quantum-enhanced and/or prototype work. We thank the reviewer for this comment. It has helped strengthen the discussion of this new work.

Reviewer 3’s Report

Key results

The paper presents a search for dark matter axions at relatively high frequency compared to other microwave cavity searches, and reaching sensitivity close to the benchmark QCD axion. The nature of such searches is that the given frequency range, while narrow, has not been probed before. The key point of the paper, however, is that this sensitivity has been achieved exploiting a novel measurement procedure. The signal is “squeezed” in phase space, overcoming the standard quantum limit for an unsqueezed distribution. The squeezing procedure widens the bandwidth of the experiment, and allows a wider range of frequencies to be searched in a shorter measurement time.

Validity

I can see no immediate problems with the validity, however see my expertise below.

Significance

Searches for the QCD axion are appearing more and more around the world. This is a hot topic at the moment. There are many technologies being developed. The present work

uses the tried- and-tested microwave cavity, but enhanced with the squeezing technique to extend its usefulness to higher frequencies, clearly with an eye to future campaigns.

The frequency probed is novel, and the sensitivity is close to the QCD axion range (although not yet reaching KSVZ or DFSZ). This method will surely be useful to other axion searches operating at high frequency in the future, a significant technological step forwards for the field.

Data and methodology/Analytical approach

The methodology appears extremely sound is based on well documented and clear methods previously published by the HAYSTAC collaboration. That being said, I comment below on places where the presentation could be made more clear. Many symbols are undefined, and I think a short review of the relevant papers in the Methods section would be helpful to the reader.

general response: We are fortunate to have had a theorist among the three Referees (all of the authors are experimentalists), and in addition to the detailed comments and corrections, we interpret the larger sense of this referee's comments to be that it is unnecessary to pin the results of this paper so tightly to theoretical predictions at all. We concur with Referee #3 in this regard, and in addition to amending and correcting the detailed discussion as pointed out below, we have made the theory discussion overall more appropriately tentative.

Suggested improvements:

- Recent theoretical predictions of the axion mass, discussion of refs. [10,11].
 - The discussion makes out like these predictions are completely model independent, and uncontroversial, but, as I am sure the authors know, many caveats apply.

The authors should refer to Gorghetto et al's two papers on this subject, which I believe to be superior. After including many sources of uncertainty in the extrapolations taking simulations to the physical coupling values, they conclude that only a lower limit on the axion mass can be placed, and it is of order 1 meV (this in itself is not uncontroversial either).

response: We thank Referee #3 for pointing out the Gorghetto et al references to us, and have now cited the paper with their lower limit on the axion mass as a caveat to the predictions of refs. [10,11].

Furthermore, all of these theoretical predictions depend on cosmology, and the assumption that there is only a single DM particle. The cosmology dependence assumes that the Universe had a maximum temperature higher than the PQ transition, and that all physics below that temperature is known in the Standard Model. Both of these facts are clearly very large assumptions.

response: Essentially all dark matter experiments are designed to achieve a target sensitivity assuming that the candidate particle (WIMP, axion, ALP, etc.) saturates the local dark matter density, and report their sensitivity under that assumption as well; we are no exception. But to make that absolutely clear, we have inserted that qualification into the text.

In low scale inflation (pre-inflation symmetry breaking), as is well known, a very wide axion mass range is allowed and the theoretical predictions cited do not apply. Adopting a uniform prior on the misalignment angle and demanding that axions are all the DM in this case leads to the 95% C.L.

posterior on the mass $0.53 \mu\text{eV} \leq m \leq 0.13 \text{ meV}$ (GAMBIT axion global fits, <https://arxiv.org/abs/1810.07192>). The present work is square in the middle of this range.

response: We agree with the Referee. If more DM candidates are allowed (e.g. axions+WIMPs), then the mass predictions of [10,11] become lower bounds. This is certainly true, but it should be noted that having two dark matter particles whose relic densities would be within an order of magnitude of one another would be very surprising, absent a causal connection in their production.

Finally, even taking the statement in the abstract completely with these caveats, the predictions only apply to $N = 1$. For $N > 1$ (e.g. DFSZ), the prediction was made by the Kawasaki group for m . With the least amount of fine-tuning to solve the strong CP problem, the prediction for $N > 1$ saturates at the SN1987A bound, $m \sim 5 - 10 \text{ meV}$. The above considerations should be made in brief, and Refs. [10,11] should not be give such prominence in setting an expected mass range. I would agree with something along the lines of “the range $m > 10 \mu\text{eV}$ favoured when the Peccei-Quinn symmetry is broken during the hot big bang, and not during inflation” and then an expanded list of references. Maybe add something about “about half of the posterior volume in $\log(m)$ in the alternative scenario for symmetry breaking assuming a uniform prior on the initial axion field value’.

response: As this is an experimental paper, and further whose primary emphasis is on the application of quantum measurement to a dark matter search, we would prefer to keep the theory discussion tight, and resist the temptation to let it expand into an expert-level discussion. Thus we propose that we include the important qualifications the Referee has suggested above, including adding the Gorghetto et al reference, but not go much further here.

- Later, Refs. [10,11] are fundamentally misunderstood. These are NOT Lattice QCD. They are lattice classical field theory for the Peccei-Quinn field and include no QCD whatsoever. This statement must be corrected. Recent lattice QCD simulations were carried out by Borsanyi et al to determine the temperature dependence of the axion mass, but they do NOT simulate the lattice classical theory of string decay. Moore’s results did not use the Borsanyi et al QCD-calculated $m(T)$. Foster et al use a parameterization of $m(T)$ and discuss the uncertainties from it coming from lattice QCD, which are around 27% on the mass prediction.

response: We are thankful to the Referee for pointing this out, and have made the correction.

- Why is Ref. [33] used for the axion model band? This seems limited, and out of date. A more complete and recent study is by di Luzio et al, which gives the maximum of the band in terms of the ratio E/N in self-consistent models. This was adopted in the GAMBIT global fit of axion models cited earlier. The model band is reviewed in <https://arxiv.org/abs/2003.01100> I suggest to either include a more detailed view of the band following di Luzio and the GAMBIT analysis, or to simply show KSVZ and DFSZ in Fig. 3.

response: The suggestion to cite di Luzio is a welcome one, as it makes the case that even while it is reasonably bounded (unlike WIMP parameter space), the axion-photon coupling is still plausibly broad. This change brings us back in line

with rest of the field, which has been using this model band for a few years now. We will adopt this reference and modify the figure appropriately.

- In Fig. 3 the red band at just slightly higher frequency than the present analysis is not labelled. Is it the ORGAN result, or the QUAX (photon coupling) result?

Response: In figure 3, the labeling is performed by experiment, not individual bands. The red band pictured is a secondary result published by the ADMX collaboration (from a small cavity called ‘Side Car’), which is labeled at the left of the plot. Due to space constraints, results from each collaboration are only labeled once.

- Why does the axion mass window scanned have a gap from 17.12 to 17.14 micro eV? I presume this is some background noise frequency or troublesome resonance of the apparatus. Please explain.

Response: In the third-to-last paragraph of the main text, we already include the explanation for this “skipping a TE mode at 4.140–4.145 GHz which does not couple to the axion.” We have altered TE to “transverse electric” since the shorthand is not used elsewhere.

- Presentation/discussion of squeezing

- I struggle to fundamentally understand what happens when “the axion displaces the squeezed state from the origin”. Clearly, this must be done by the inverse Primakoff process where the axion creates a photon in the presence of a magnetic field. However this is not actually stated anywhere in the manuscript. The authors should explain a bit more how this interaction relates to the Hamiltonian (1). How is the power generated by the axion related to the shift in the operator X ? The authors should improve the description of squeezing so that the link to the particle physics model is clearer. I think this would improve the wider readability of the manuscript.

Response: We appreciate this comment and agree that inclusion of the Primakoff effect in our explanation would make this section more widely readable. We have changed the

text to clarify that it is due to the inverse Primakoff effect that the axion is able to displace the squeezed state. We address the reviewer’s concerns about our description of squeezing below.

- I understand that squeezing increases the scan rate. But I do not understand what “squeezing N_r ” means. The integral on δ_ν is a one dimensional integral, and N_r is written as a function of a single variable, not two, one of which could be squeezed.

response: We apologize for this confusion: the referee is correct that one variable may only be squeezed at the expense of amplifying another. Our equation accounts for this, but we acknowledge that our definitions may lead to confusion. Accordingly, we include within the manuscript: “where all spectral densities and hence α are understood to refer to a single quadrature,” after introducing N_r and the other noise sources. Then, when we perform the integral in Eq. (3), we add “taking α to be along the squeezed quadrature.” Thus we are implicitly ignoring the noise along the Y quadrature, which, as the referee suggests, is also present, and increases as a result of our measurement protocol. However, we never measure the Y-quadrature noise, and as referenced in the same paragraph, this does not slow down the scan rate at all relative to a quantum-limited two-quadrature measurement. The integral on δ_ν is valid

subject to the language we have inserted above.

- Can the authors give slightly more explanation of how Fig. 2a is arrived at, perhaps with reference formulae given in a supplemental? The role of the parameter S for the deliverable squeezing is not clear. How does it enter in Eq. 2?

Response: We have added language to the figure caption to clarify that each of the lines in figure 2 is a Lorentzian cavity response profile obtained by optimizing the scan rate subject to squeezed and unsqueezed operation. This was a welcome suggestion and we think that it will benefit the clarity of the paper as a whole.

- S is minimized with the parameter η of the experiment. Could this in principle be improved, or is it a fundamental limit to this technology?

Response: The mathematical dependence on η is fundamental to this configuration. However, in principle, η itself can be improved. In the current text of the paper we include forward looking language which addresses this in the conclusion: “As intense interest in quantum information processing technology continues to drive transmission losses downward, quantum-enhanced measurement will deliver transformative benefits to searches for new physics.”

- I feel like the description of squeezing came a bit backwards. I was always asking questions which were answered about a half page later. To me it seems most logical to define S , the measure of squeezing, and the noise quantities, then give R , then α . S_{ax} is then clearly the power given by the familiar formulae for a cavity. Then finally you could say “this is understood in quantum mechanics by thinking of Eq. 1 ...”. I find starting with the quantum picture confusing.

Response: We respectfully disagree with the reviewer here. The current organization of the description of squeezing allows us to start with the big picture and gradually move into more detailed experimental explanations. We hope that the other clarifications that we made based off of the other comments from the reviewer help improve their understanding of the description of squeezing and the order that we chose.

- It would be nice to see somewhere in the conclusions a discussion about the future of HAYSTAC. Will this technology be used to go “wider” or “deeper”? Plans for DFSZ? Plans for other frequencies?

response: In regard to the Referee’s questions about our intentions for future HAYSTAC operations, these are collaboration strategic issues that dynamically evolve according to our understanding of the physics landscape and our own technological progress. Thus we think it wouldn’t be helpful to the reader to try to be too precise about our projected coverage in mass and sensitivity. What should be clear from the text already is that the current mass range of HAYSTAC operation is a very interesting one. Also as we have tried to clarify in the text, DFSZ and KSVZ are in fact both manifolds of models, spanning a broad range in axion-photon coupling; HAYSTAC is already squarely in the middle of the QCD axion band.

- What about other detection methods? How does the present method using squeezing+heterodyne stack up against a (hypothetical) microwave single photon counter?

Response: We are also excited about the potential of single-photon counting in axion experiments. It is beyond the scope of this paper to comment on single-photon counting and other technologies, but we wish to supply some of that context here, and

consequently show that photon counting would have been a comparably poor readout technique for our haloscope in the frequency range of interest. In particular, the efficacy of photon counting depends on the dark count rate, set in part by the thermal occupancy n of the cavity. Taking the axion to have a fractional linewidth of 10^{-6} , photon counting could theoretically surpass coherent readout when

$$n \lesssim Q_0/10^6. \quad (1)$$

In our haloscope the thermal occupancy of the cavity was $n \sim 0.3$ photons, whereas our $Q_0 = 47000$ implies that our thermal occupancy is still roughly two orders of magnitude too high, *before* we account for squeezing. A more in depth discussion of this can be found in this article, written by members of our collaboration: Analysis of single-photon, squeezed- state and linear amplifier detectors for microwave cavity dark matter axion searches, S.K. Lamoreaux, K.A. van Bibber, K. Lehnert, G. Carosi, Phys. Rev. D 88, 035020 (2013)

- There are many undefined symbols used.

response: We thank the reviewer for this comment. We have gone through the text and defined a number of symbols that were lacking a definition before: TM_{0n0} , TE, g_Y^{KSVZ} , σ_{on} , and σ_{off} .

The axion-photon coupling is not defined. The cavity form factor is not defined. The cavity power is not given. What model was used for the axion linewidth/lineshape?

Response: In an effort to ensure that the paper remains as broadly readable as possible, we have kept it equation-light. In addition to being readily available in previous work, the expressions for these quantities are somewhat cumbersome and contain a lot of terms not necessary for understanding the main claims of this paper. Each of the quantities listed above is fully written out in equation form in our previous publications. As such, we have added a citation to the text for [Ref. 30, Kenany et al] (reference number corresponds to the revised manuscript) where the expression for axion converted power can be found, and [Ref. 33, Brubaker et al] for the axion lineshape.

- How do uncertainties in the halo model impact the exclusions? What is the “axion distribution A_i ”?

Response: To clarify for the reviewer, our use of the term “axion distribution” refers to the probability density function of grand spectrum power excesses measured at a frequency containing the axion, and not the velocity or energy distribution of the axions in the galactic halo. Specifically, we include: “axion A_i distributions, well approximated as Gaussian with $\sigma = 1$ and $\mu_0 = 0$ (no-axion) or $\mu_a = g_{Y,i}^2 n_i$ (axion), evaluated at x_i . For clarity, we had inserted “probability” between “ A_i ” and “distributions.”

- A slightly longer description of Ref. [30] in the Methods section is required.

Response: We have added several new sentences and clarifications as well as a useful citation, Ref. [39], which had not yet been published at the time of our original submission. We believe these fill in key details and complete the picture of the processing and accompanying analysis for the reader at an appropriate level of detail.

Summary of Changes

1. In the abstract, we added “some” to the sentence “we double the search rate for axions over a mass range favored by some recent theoretical projections.”
2. In the second-to-last sentence of the abstract, we added the excluded mass range with the language “within the 16.96–17.12 and 17.14–17.28 $\mu\text{eV}/c^2$ axion mass windows.”
3. In the second paragraph of the main text, we added “transverse magnetic” in front of TM_{0n0} -like mode. A citation for [22] was added as well.
4. In the third paragraph of the main text, we added “ g_Y^{KSVZ} ” after “the benchmark KSVZ coupling” in order to introduce the symbol.
5. In the fourth paragraph of the main text, we changed “favored by large-scale lattice QCD calculations of post-inflationary Peccei-Quinn symmetry breaking scenarios [10,11].” to read “favored by recent calculations based on lattice classical field theory for the Peccei-Quinn field in post-inflationary Peccei-Quinn symmetry breaking scenarios, and assuming that axions saturate the cosmological dark matter density [10,11] (alternative views should be noted however; see e.g. Ref. [27]).”
6. In the fifth paragraph, we changed “An axion field, if present, displaces the squeezed state from the origin in the cavity field quadrature phase space, proportional to the Lorentzian transmission profile of the cavity at ν_a .” to read “An axion field, if present, displaces the squeezed state along a random direction from the origin in the cavity field quadrature phase space via the inverse Primakoff effect [29]. The displacement is proportional to the Lorentzian transmission profile of the cavity at ν_a and the square root of the axion-converted power [30].”
7. After Eq. (2), we added the language “where all spectral densities and hence α are understood to refer to a single quadrature.”
8. After Eq. (3) we changed “ N_r ” to “the reflected noise and taking α to be along the squeezed quadrature.”
9. At the beginning of the next paragraph, we added “in Eq. (2)” after N_r .
10. In the same paragraph, we changed “defined as the variance reduction at the system output” to “defined as the SQ on/off-variance reduction at the system output.”
11. In the Fig. 2 caption, we added “PSDs are normalized to the single-quadrature vacuum value of $\hbar\nu/4$. The solid and dashed lines are Lorentzian cavity response profiles obtained by optimizing the scan rate subject to squeezed and unsqueezed operation.”
12. In the fourth-from-last paragraph, we changed “we measure a delivered squeezing” to “we measure a deliverable squeezing.”
13. In the fourth-from-last paragraph, we changed “maximum” to “optimal.”
14. In the third-from-last paragraph, we changed “TE” to “transverse electric.”
15. In the Fig. 3 caption, we changed “The QCD axion model band [32] is shown in yellow” to read “The QCD axion model band representing the most natural KSVZ and DFSZ models from a recent analysis [35] is shown in yellow.”

16. The model band in Fig. 3 was changed.
17. We changed the last sentence to read “In particular, the prospect of removing nonreciprocal signal routing, as could be accomplished by creating a parametric swap interaction [38] between the axion cavity and the JPA circuit, would boost transmission efficiencies above 90%, yielding a greater-than-tenfold scan rate increase beyond the quantum limit [9].”
18. We added “(the constraints of operation near an 8 T region were not present in the previous work described in Ref. [9])” to the first paragraph of the Experimental Operations section in the Methods.
19. In the second paragraph of that section, we changed “Then,” to “This corresponds to an
AMP (bandwidth of ~ 3 MHz). Both SQ (bandwidth ~ 26 MHz) and AMP therefore have bandwidths much greater than that of the cavity (bandwidth $\kappa_m + \kappa_l \lesssim 1$ MHz). With the AMP gain set...”
20. We added “Simultaneously tuning the two JPAs and optimizing the squeezing at each frequency step posed new challenges not present in either previous HAYSTAC runs [3,19] where only one JPA was used, or in the work of Ref. [9], where no tuning was performed” to the same paragraph.
21. In the next paragraph, we changed “delivered” to “deliverable.”
22. We added “The shared structure in the IF is a result of the shape of the receiver chain’s spectral response and noise. An axion signal would line up in the RF and wash out in the IF, making it safer to remove power excesses in the IF” to the second paragraph of the Data Analysis section in the Methods.
23. In the same paragraph, we changed “We then coadd groups of adjacent frequencies” to “Again using maximum likelihood weights, we then coadd groups of adjacent frequencies.”
24. We added “[33] in order” to the last sentence of that paragraph.
25. We added a sentence before the second sentence of the last paragraph of the section and changed the lead in to the next sentence: “In the limit of low but nonzero prior probability of the axion residing anywhere in the scanned window, the prior probability itself drops out of the expression for the update. The update then...”
26. In the next sentence, we added “probability” before “distribution.”
27. We fixed a typo that was found in the second to last equation in the paper. It now correctly reads:

$$u_i = \frac{P(x_i|\mathcal{A}_i)}{P(x_i|\mathcal{N}_i)} = \exp \left[-\frac{\mu_{a,i}^2}{2} + \mu_{a,i}x_i \right].$$

28. We added “PHY-2011357” to the acknowledgements.
29. We added four citations. Their locations in the text are in the changes above.

- [22] Rapidis, N. M., Lewis, S. M. & van Bibber, K. Characterization of the HAYSTAC axion dark matter search cavity using microwave measurement and simulation techniques. *Rev. Sci. Instrum.* **90** (2019).
- [27] Gorghetto, M., Hardy, E. & Villadoro, G. Axions from strings: the attractive solution. *Journal of High Energy Physics* **2018**, 151 (2018).
- [35] Di Luzio, L., Giannotti, M., Nardi, E. & Visinelli, L. The landscape of QCD axion models. *Physics Reports* **870**, 1–117 (2020).
- [39] Palken, D. A. *Enhancing the scan rate for axion dark matter: Quantum noise evasion and maximally informative analysis*. Ph.D. thesis, University of Colorado Boulder (2020).

30. We removed one citation.

- [32] Cheng, S. L., Geng, C. Q. & Ni, W.-T. Axion-photon couplings in invisible axion models. *Phys. Rev. D* **52**, 3132–3135 (1995).