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27th Feb 24

Dear Dr Kaleris,

Thank you for submitting your manuscript, "Acoustic metamaterials characterization via laser-plasma sound sources", to Communications Materials. It has now been seen by 2 referees, whose comments are appended below. You will see that while they find your work of interest, some important points are raised. We are interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

-A clean version of the manuscript. Please select the file type 'Article File'.

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- If applicable, a statement regarding data available with restrictions
- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

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We hope to receive your revised paper within three months; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Aldo

Dr Aldo Isidori

Senior Editor

Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors propose to create sound in acoustic experiment by using the laser-plasma breakdown. Compared to previous methods by using speakers, a main advantage of the current one is to induce sound inside materials. Measurements on several metamaterials are conducted to verify the method and the results are compared with numerical simulations and results from measurements in impedance tube.

The method looks interesting to me, however, the presentation is not quite satisfying and needs improvement. Given that laser-plasma sound source might be interesting to the metamaterial community, I recommend the authors to address below questions adequately:

The authors should be more modest about their method in the abstract and in the introduction. There are different approaches to characterize acoustic properties of materials, either by using waveguides, anechoic chambers, or impedance tubes. Properties along different directions as well as different frequency ranges can be obtained. What the authors present here is essentially an alternative way to generate sound. Therefore, I would refrain to state “However, although their design and modeling are based on well-established methods, experimental evaluation of their acoustic performance is currently hindered by major limitations.”

As the main novelty lies in the sound generation method, I prefer to see more elaborations on the mechanism and also more characterizations of the generated sound. For examples, what is needed to allow for plasmon in an acoustic material, is air alone in the tubes enough? How do the time-domain signal looks like in the actual experiment? And how is the time-domain signal after propagating through the sample?

The authors first present the experiment and numerical results on transmission in Fig. 4 and then show the band structure in Fig. 5. The order of the two figures is not optimal. Since both figures explain the bandgap, I recommend that the authors somehow prepare the band structures and the transmission curves side-by-side. In addition, the authors may consider swapping the vertical axis and the horizontal axis of the transmission curves. This enables comparing the bandgaps from the calculation and the measurement more easily.

Figure 6 presents the eigenmodes for the model with defects. I understood this figure as a numerical support to the results in Fig. 4. If so, the authors should immediately refer to the results while discussing Fig. 4 in the main text instead of coming back to the results after discussing something completely different in Fig. 5.

In Fig. 5b, the authors model the open peripheral holes as soft boundaries, which no surprisingly leads to suppression of transmission at zero frequency. The authors may consider modeling the open holes with alternative boundary conditions that are more close to reality, e.g., impedance boundaries. The authors could discuss the difference between the two modeling methods in the supplementary information.

The defected model in Fig. 6(d) is already presented in Fig. 2(b) and should not be depicted again here if the figure is meant to be in the main paper. If the figure is for supplementary information, then it is fine.

Some general remarks:

How strong the laser induced sound pressure is in the experiment? Is it necessary to consider acoustic nonlinear effects? The author might comment on this.

How long does it take to measure one sample?

In the numerical calculation of transmission spectra, the transmitted and incident pressure filed are extracted from the simulation. In the experiment, how is the incident acoustic power determined? This should be specified in the main text or in the method section.

Reviewer #2 (Remarks to the Author):

This is a concise study demonstrating the use of laser-induced plasma shock/acoustic waves as a consistent, repeatable, and robust acoustic source to probe PC and AMM structures with. I recommend publishing with minor revisions:

Laser induced shockwaves decay significantly from the origin. Have the authors explored laser energy/unit cell ratios in order to determine if their results hold at various scales of PC structure and energy pairings?

How much is the duration of the N-wave produced by the laser a function of laser-energy coupling and inherent acoustic wave properties and how much is it dependent on microphone response properties e.g. minimum response or rise times of the mic?

How did the authors ensure the laser-induced breakdown was exactly in the center of the PC structure sphere centers? They should include some sort of visualization of the laser-induced breakdown wave (shadowgraphy, schlieren, etc) to confirm it is spherical or cylindrical, which will depend on the laser properties as they note.

Fig 3: please label the laser configuration/ conditions on each figure for posterity

Several references discuss in more details the laser-induced shockwave/acoustic properties from a propagation standpoint. The authors may want to consider reviewing the following resources:

a) <https://doi.org/10.1016/j.apacoust.2003.11.003>

b) <https://doi.org/10.1088/1004-423x/7/2/007>

c) <https://doi.org/10.1364/AO.56.006902>

Response to reviewers

Acoustic metamaterials characterization via laser-plasma sound sources

We thank the reviewers for their considerate evaluation of our work and their constructive comments, which we believe have helped us improve the manuscript. Answers to all their comments are provided below. Changes related to the reviewer comments are marked with yellow in the revised manuscript. Minor phrasing changes are marked with green.

pg. = page, par.= paragraph

Reviewer #1:

The authors propose to create sound in acoustic experiment by using the laser-plasma breakdown. Compared to previous methods by using speakers, a main advantage of the current one is to induce sound inside materials. Measurements on several metamaterials are conducted to verify the method and the results are compared with numerical simulations and results from measurements in impedance tube.

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Comment 1: The authors should be more modest about their method in the abstract and in the introduction. There are different approaches to characterize acoustic properties of materials, either by using waveguides, anechoic chambers, or impedance tubes. Properties along different directions as well as different frequency ranges can be obtained. What the authors present here is essentially an alternative way to generate sound. Therefore, I would be refrain to state “However, although their design and modeling are based on well-established methods, experimental evaluation of their acoustic performance is currently hindered by major limitations.”

Response: Following the reviewer’s suggestion, we have mitigated the statements regarding the drawbacks of existing AMM evaluation methods in the revised manuscript. We have removed the abovementioned phrase and have made relevant changes in the Abstract (pg. 1, par. 1), Introduction (pg. 2, par. 2), Discussion (pg. 11, par. 2 and pg. 12, par. 1) and Outlook (pg. 12, par. 1), which we hope are in the suggested direction towards the existing approaches.

Comment 2: As the main novelty lies in the sound generation method, I prefer to see more elaborations on the mechanism and also more characterizations of the generated sound.

- a) For examples, what is needed to allow for plasmon in an acoustic material, is air alone in the tubes enough?
- b) How do the time-domain signal looks like in the actual experiment? And how is the time-domain signal after propagating through the sample?

Response: Following the reviewer's request, we have added details on sound generation via laser-induced breakdown of ambient air in the Introduction section (pg. 3, par. 1). Moreover, since both reviewers asked for more details on the acoustic sources (see also Comment 3 of Reviewer 2), we have added information regarding the source characterization via acoustic directivity measurements in the subsection "LPSS characterization" of the Methods section of the revised manuscript.

a) We are not sure if we have correctly understood the question of the reviewer regarding "plasmon in an acoustic material" and the relation between acoustic plasmons and laser-plasma sound generation in the air. Generally acoustic plasmons entail the interaction of acoustic waves with electromagnetic waves on a surface or at the interface of solid materials. If the reviewer refers to the collective motion of plasma ions (ion acoustic wave) in the presence of the EM-Field of the laser, these are in frequencies far above the audible range, while the intensity of the used nanosecond laser is very low to drive such phenomena. In our understanding, acoustic plasmons are not relevant to this work since LPSSs generate classical pressure waves.

In case the reviewer refers to "plasma" (instead of "plasmon"), air alone is sufficient for plasma and consequent acoustic N-pulse generation inside the tubes. High concentration of optical power within a small air volume ($> 10^{12} \text{ W/cm}^2$) leads to multiphoton ionization and the generation of an initial small free electron density. These free electrons initiate a cascade ionization process which leads to a larger density of hot free electrons that is capable to thermalize the heavy particles in the interaction volume. Please refer to the information added in the revised manuscript (pg. 3, par. 1) for more details.

b) In the manuscript, Fig. 1a) shows an actual N-pulse pressure signal (and its frequency spectrum in Fig. 1b)) from the experiments. Please note that time-domain signals are not particularly helpful for observing delicate differences in the behavior of the materials, but they clearly reflect the filtering effect of the meta-structure, which manifests as a broadening of the acoustic transmission signal with multiple oscillations before the pressure fades to zero. Following the reviewer's suggestion, we have added a new figure (Fig. 3 of the revised manuscript) with a typical time-domain signal measured at the output hole of the normal PC with closed holes and source outside. The description of the figure is in the respective section (Results, cylinder evaluation, pg. 4, par. 1)

Comment 3: The authors first present the experiment and numerical results on transmission in Fig. 4 and then show the band structure in Fig. 5. The order of the two figures is not optimal. Since both figures explain the bandgap, I recommend that the authors somehow prepare the band structures and the transmission curves side-by-side. In addition, the authors may consider swapping the vertical axis and the horizontal axis of the transmission curves. This enables comparing the bandgaps from the calculation and the measurement more easily.

Response: please see Response to the next comment.

Comment 4: Figure 6 presents the eigenmodes for the model with defects. I understood this figure as a numerical support to the results in Fig. 4. If so, the authors should immediately refer to the results while discussing Fig. 4 in the main text instead of coming back to the results after discussing something completely different in Fig. 5.

Response: Comments 3 and 4 of the reviewer address the fact that in the original manuscript the computational results were presented separately from the experimental results, in the form of discussion, instead of being presented in the Results sections side-by-side to the experimental measurements. This was initially done to highlight the novelty and main contribution of the work, which lie in the experimental evaluations. However, we appreciate the reviewer's concern that this "back and forth" could confuse readers and reduce clarity. For this reason, in the revised manuscript we have moved the computational results to the Results section. In this way, the computational findings are presented and discussed side-by-side with the experimental results. We believe that this change suggested by the reviewer enhances the readability of the manuscript and enables direct comparison between experimental and computational results. To keep the experimental results concise, they are again presented in one figure for the cylinder and one for the cube. This helps the reader to easily identify differences in the acoustic behavior of the different structures or propagation axes, respectively. The computational results are included in separate figures but are referenced in the text directly in the results section, comparatively to the experimental results. We hope that the reviewer finds this presentation more adequate compared to the original one.

The reviewer has also suggested flipping the axes of the transmission curves in order for the experimental findings to be in the same format as the band structure. Although we understand that this would facilitate comparison between the two, we ~~believe~~ have observed that flipping the axes of the transmission curves would hinder their readability. Frequency spectra, such as sound transmission or transmission loss, are very typically presented with the frequency in the horizontal axis and the spectral magnitude in the vertical axis, both in the acoustics and the materials communities. Of course, in the materials community sometimes the axes are flipped, however a quick search in the present journal has shown that the majority of publications use the transmission vs frequency format. On the other hand, band structures are almost always presented with flipped axes, a convention that stems from the electronic band structure curves. Hence, although flipping the axes of the transmission curves would facilitate comparison to the band structure, we believe that it would significantly hinder the readability and immediacy of the acoustic spectra. Since these transmission curves constitute the main results of our work, and since the reviewer was not absolute on the matter, we have decided to keep the axes as they were. However, if the reviewer insists, we are willing to make changes in a next revision round.

Comment 5: In Fig. 5b, the authors model the open peripheral holes as soft boundaries, which no surprisingly leads to suppression of transmission at zero frequency. The authors may consider modeling the open holes with alternative boundary conditions that are more close to reality, e.g., impedance boundaries. The authors could discuss the difference between the two modeling methods in the supplementary information.

Response: We thank the reviewer for this comment which directed us to further investigate the different choices for modeling the boundaries for the open holes. Sound soft boundaries impose zero pressure at the position of the holes, which is known to accurately describe the physics of acoustic wave propagation in such structures, especially in the low frequency range. Direct connection of the structure's air conduits with the ambient air leads to a significant suppression of the acoustic pressure at the position of the holes, which tends to zero. This pressure drop leads to a strong reflection of the propagating wave in the low frequencies, resulting to the formation of the observed low frequency bandgaps. Such a condition is well described by the sound soft boundaries, as also shown in previous works [50,51].

Nevertheless, following the reviewer's suggestion, we carried out test simulations using impedance boundaries, which resulted to the emergence of a passband in the low frequencies (from zero up to 2.0 kHz), as shown in the calculated band structure of Figure c) below. This behavior is similar to the sound hard boundary (Figure b), while it contradicts both the relevant measurements of Figs 4c)-e) and the band structure calculated via sound soft boundaries shown in Figure a). As previously mentioned, this is because the phenomenon creating the low-frequency stopband emerges from the direct coupling of the air conduits with the ambient air. The coupling creates open tube conditions which result in vanishing pressure at the holes. In the transmission calculations, such a direct connection was realistically simulated by the use of an air domain surrounding the structure, so that no boundaries were needed at the peripheral holes. This approach showed zero pressure in the holes, while it accurately predicted (Figure a) the measured transmission spectra of Figs 4c)-4e). In the band structure calculation, where a single cell with boundary conditions and no surrounding air domain is used, such a pressure condition is best modeled via sound soft boundaries [50,51]. Justification of the selection of the boundary conditions has been added in the Simulations subsection of the revised manuscript (pg. 14, par. 1)

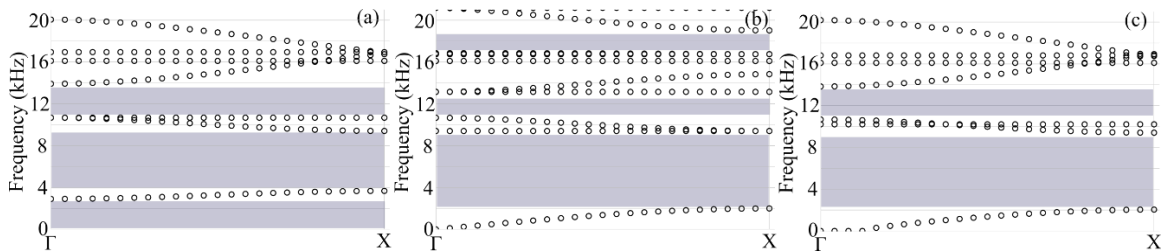


Figure: Calculated band structure for the cylinder with open peripheral holes and periodicity along the ΓX direction considering different boundary conditions at the peripheral holes, (a) Sound soft boundaries, (b) Sound hard boundaries, and (c) Impedance boundaries. The grey zones denote the phononic band gaps.

Comment 6: The defected model in Fig. 6(d) is already presented in Fig. 2(b) and should not be depicted again here if the figure is meant to be in the main paper. If the figure is for supplementary information, then it is fine.

We confirm that Fig. 6(d) is placed only as supplementary information and does not differ from the one presented in Fig. 2(b) in any way. We believe that it is helpful for the reader to have a

direct impression of the defective cylinder's structure, next to the FEM simulation results and for this reason we have kept it in the revised manuscript. We also explicitly state that the schematic is the same as that of Fig. 2b).

Comment 7: Some general remarks:

a) How strong the laser induced sound pressure is in the experiment?

We thank the reviewer for his notice. The sound pressure level is about 97 dB at a distance of one meter from the plasma for a measurement time window of 35 ms, which corresponds to a peak pressure of ~ 130 dB. A relevant mention was included in the newly added subsection "LPSS characterization" of the Methods section (pg. 14, par. 1).

Is it necessary to consider acoustic nonlinear effects? The author might comment on this.

Please refer to the response to Comment 1 of Reviewer #2.

b) How long does it take to measure one sample?

A measurement can be carried out with a single pulse or with a train of pulses. For a single pulse, the measurement is instantaneous (less than 1 sec), while for pulse trains it depends on the number of pulses. Here, pulse trains with ~ 300 pulses were used at a repetition rate of 5 Hz, so each sample was measured for 1 min. This was done in order to have a sufficient number of responses to average out environmental noise. Details can be found in the subsection "Signal analysis" of the Methodology section (pg. 15).

c) In the numerical calculation of transmission spectra, the transmitted and incident pressure filed are extracted from the simulation. In the experiment, how is the incident acoustic power determined? This should be specified in the main text or in the method section.

It is well known that the pressure profile of the LPSSs within the audible range is 1st order high pass, as shown in Fig. 1b but also from works, such as [31,36]. This is a global attribute of LPSSs and hence the spectrum of the incident acoustic N-pulse is taken as in Fig. 1b. To account for the high-pass profile of the source and equalize the measurement results and derive the objective transmission spectrum of the structures, we have used post-processing which is described in the subsection "Signal analysis" (pg. 15) of the Methods section. The absolute value of the incident acoustic power can be calculated from the measured acoustic pressure level; however, it is not of interest for this work since all the transmission spectra are presented in a normalized logarithmic scale.

Reviewer #2

This is a concise study demonstrating the use of laser-induced plasma shock/acoustic waves as a consistent, repeatable, and robust acoustic source to probe PC and AMM structures with. I recommend publishing with minor revisions:

Comment 1: Laser induced shockwaves decay significantly from the origin. Have the authors explored laser energy/unit cell ratios in order to determine if their results hold at various scales of PC structure and energy pairings?

The reviewer is right in that laser-induced shock waves (LISW) decay fast as they propagate away from the plasma source. However, under the specific experimental conditions (6 ns, 532 nm, 120 mJ laser pulses focused via a 10 cm lens) and due to expansion and energy dispersion, LISWs quickly transform into linear sound waves, within less than a centimeter. This has been evaluated in the past by measurements of the LISW propagation speed via laser beam deflection at various distances from source. It also becomes apparent from reference [38] of the new manuscript, where the times of arrival of N-pulses generated under identical laser radiation conditions are measured for propagation of 8 cm from the source. Within this distance, the average N-pulse propagation speed is about 360 m/s, which shows that the LISW quickly converts into a linear soundwave. From that point on, the laser-induced N-pulses decay proportionally to the propagation distance. Given that the sound pressure level of the N-pulse was measured ~ 97 dB at 1 m distance for an observation window of 35 ms, it becomes apparent that there would be no obstacles in measuring PC structures of significantly larger scales than the ones used here. Of course, this also depends on the noise conditions in the measurement environment. A relevant mention was included in the newly added subsection “LPSS characterization” of the Methods section.

Comment 2: How much is the duration of the N-wave produced by the laser a function of laser-energy coupling and inherent acoustic wave properties and how much is it dependent on microphone response properties e.g. minimum response or rise times of the mic?

In this work we have used a sound capturing system consisting of a microphone (G.R.A.S 46BE and power module 12AK) and a sound card (RME Fireface 802), with a combined frequency response of > 90 kHz. As shown in Fig. 1 of the manuscript, also well-known from other works [31,32,34, 37], the main part of the LPSS spectral content for such laser radiation conditions lies in the 10 - 30 kHz frequency range and fades with increasing frequency. For this reason, it is safe to consider that the captured waveforms are not compromised by the spectral bandwidth of the acoustic measurement system – and correspondingly by the microphone’s transient response. Thus, we can be confident that the observed acoustic features are inherent properties of the structure or the source, respectively.

Comment 3: How did the authors ensure the laser-induced breakdown was exactly in the center of the PC structure sphere centers? They should include some sort of visualization of the laser-

induced breakdown wave (shadowgraphy, schlieren, etc) to confirm it is spherical or cylindrical, which will depend on the laser properties as they note.

We thank the reviewer for giving us the opportunity to clarify this issue. To place the LPSSs at the center of the spheres, a camera was used to monitor the plasma position (see Figure below). We have added a relevant mention in the subsection “Experiments” of the Methods section of the revised manuscript (pg. 15, par. 1).

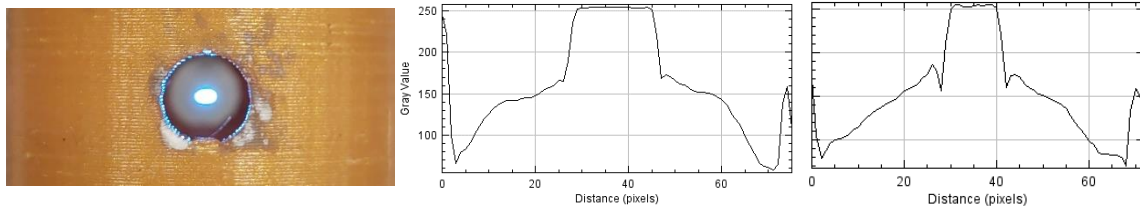


Figure: a) image of the LPSS placement inside the cylindrical phononic crystal and b) horizontal, c) vertical lineouts of the image, showing the symmetrical placement of the source.

For the source characterization, we have carried out acoustic measurements of LPSSs generated by 532 nm, 6 ns, 120 mJ pulses focused by a 10 cm lens, showing that they exhibit an almost perfectly omnidirectional acoustic emission. Particularly, at 12 cm distance from the plasma, a maximum deviation of 4 dB was observed between the sound pressure levels measured at 0 and 90 degrees with respect to the laser pulse propagation direction, while at 60 cm distance, no deviation was observed. The measurements verify that the LPSS is almost point-like and omnidirectional. The experimental procedure and the directivity polar plots are included in the revised manuscript in a newly added subsection “LPSS characterization” of the Methods section.

Finally, it is known from previous works that strongly focused few nanosecond pulses produce spherical acoustic waves. We have included references to relevant articles showing shadowgrams of shock waves generated under laser radiation conditions that are similar to the ones used here, where the roundness and omnidirectionality of the LPSS is clearly seen (refs. [40,41] in the revised manuscript). Since the interest of the present work is focused on the acoustic emission of the source, and since detailed works dedicated on the imaging of the laser-plasma shock wave generation exist, we believe it is more appropriate to restrict ourselves to the acoustic measurements.

Comment 4: Fig 3: please label the laser configuration/ conditions on each figure for posterity
Following the reviewer’s suggestion, we have added the information of the laser configuration and conditions in all measurement related figures, particularly Figs 1,3,4 and 5 of the revised manuscript.

Comment 5: Several references discuss in more details the laser-induced shockwave/acoustic properties from a propagation standpoint. The authors may want to consider reviewing the following resources:

- a) <https://doi.org/10.1016/j.apacoust.2003.11.003>
- b) <https://doi.org/10.1088/1004-423x/7/2/007>
- c) <https://doi.org/10.1364/AO.56.006902>

The suggested articles are relevant to our work and we have added them in the introduction of the revised manuscript (references [35, 36, 39]).

2nd May 24

Dear Dr Kaleris,

Your manuscript titled "Acoustic metamaterials characterization via laser-plasma sound sources" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials under the open access CC BY license (Creative Commons Attribution v4.0 International License).

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We hope to hear from you within two weeks; please let us know if the process may take longer.

Best regards,

Aldo

Dr Aldo Isidori

Senior Editor

Communications Materials

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have addressed my questions. Now, I support its publication.

Reviewer #2 (Remarks to the Author):

The authors have responded adequately to the reviewer comments. Accept as is.