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Peer Review File

Holographic Direct Sound Printing

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors present the application of acoustic holography to sound-based printing, with the ability to create polymerized PDMS components based on acoustic fields defined at a given plane. This appears to be an extension of the authors' previous work, in which they previously applied the acoustic printing principle using a single focused point, where this work produces planes of polymerized images, rather than doing this point by point. The authors compare this work to the difference between SLA vs DLP in optical-based printing, although there is an important caveat. Whereas in the author's previous work designed fully 3D geometries could be produced, with the part being built up point-by-point, in this work the produced parts are limited to ~2.5D geometries, where an identical cross-section is extruded from a PDMS bath. This is somewhat mitigated by the integration of an articulated arm that permits the extrusion to be bent/rotated as it is extruded. The authors could accordingly do more to articulate the limitations and relative merits of the previous work compared to the current one, especially in terms of print speed and highlighting particular applications where the dimensional limitations are not overly deleterious. The authors are advised to comprehensively respond to this in their revisions, as well as to the comments below.

1. Is there a threshold acoustic power input that's required for cavitation/polymerization? For instance, if the power were decreased to 1W from 5W for a given image, would the polymerization take 5x as long? Or would it not occur at all (or much much slower). Namely, what is the significance of the 2MPa value? Similarly, is there a maximum acoustic pressure beyond which printing speed is not enhanced and/or part quality is negatively impacted? Can the polymerization rate as a function of acoustic power or pressure be explicitly quantified?

2. Can the authors expand on the mechanism/physical basis by which the printed object appears to polymerize on the platform first/preferentially, rather than in the bulk of the material (where the acoustic field is also present)?

3. Can the authors better characterize/model the improvement in print speed that is feasible using acoustic holograms vs. a single focussed point to print objects?

4. Are there limits to the dimensions/resolution that can be printed using this approach? I.e. the pattern widths in DOI 10.1002/adma.202208002, doi.org/10.1021/acsnano.0c03754 and DOI 10.1002/adma.201904181 appear to be well below 1 mm, though are >=3mm here. Would this approach be scalable to higher frequencies that might permit greater resolution?

5. When comparing HDSP vs. DSP times, there should be greater clarity on what basis the comparison is made. Is the total energy deposition identical in both cases, and/or are there limits to the total energy density that are appropriate for DSP? Or is this limited by the translation speed of the DSP method?

6. The examination of printing at multiple heights (as in Fig. 4b) in the presented application would appear to have limited utility. What applications would benefit from such multi-object/location printing? Moreover, the authors may wish to discuss potential future implementations based on modifiable acoustic holograms in which the entire acoustic hologram changes at a given plane, which may give this approach additional functionality (e.g. doi.org/10.1038/s41467-020-18347-2, doi/10.1002/advs.202301489)

7. “accuracy of λw/8” – does this exactly correlate to a pixel dimension (80μm in the methods)?

8. “Therefore, there is an optimum value for f0 based on the printing setup and the printing material used” Can the authors estimate what this might be for the printing setup used in this work?

9. There are a range of powers and DC (10-100%) values used in the various prints in this work. Can the authors provide a better understanding of how/why a given power/DC values were chosen for these different prints? Is it the case that a more opaque part was desired in the higher DC% cases? It’s notable that “DC 100% results in fully non-transparent and porous structurers [sic]”, but no DC 100% data is presented.

10. “similar to SLA when DLP was utilized” – the authors could be more specific in noting the increase in dimensionality between these methods (1D vs 2D).

11. Please provide additional details on the “step-down matching unit” that was used (i.e. manufacturer/model or circuit makeup).

12. What are the transducer thicknesses?

13. Please provide more detail on the luminol solution preparation (concentrations, amounts, etc.)

14. What other materials might this method be applicable to besides PDMS?

Language. This work could do with a comprehensive review of the wording/writing to pick up gramaticall errors and issues. A few are given below.

1. P1. “acoustic field could reach”, could be “acoustic field can reach”

2. “so that it could break”, could be “to break”

3. “DSP tames the cavitation phenomena for creation rather than destruction”, this seems a bit more poetic than typical academic discourse, especially when the “destruction” that cavitation produces often “creates” beneficial ends, for example enhancing blood brain barrier permeability.

4. “in which difficult to”, should be “in which it is difficult to”

5. P3. “Hologram coordinate system”, “The hologram coordinate system”

6. “single chemically active region”, “a single chemically active region”

7. “need to be calculated”, “needs to be calculated”,

8. P5. “Inside body”, “inside the body”

9. There are several acronyms that are introduced throughout the manuscript that are seldom used (only once or twice, or even not at all, e.g. ECS, HCS, UAMR). These do not enhance clarity and should be removed.

Reviewer #2 (Remarks to the Author):

This paper presents a layer-based additive manufacturing technique based on the process of holographic acoustic assisted 3D printing, called as HDSP. This research builds on previous work of the authors (Habibi, M., Foroughi, S., Karamzadeh, V. & Packirisamy, M. Direct sound printing. Nat. Commun. 13, 1–11, 2022) and aims at extending the previous point-by-point printing process to a full cross-section of the segmented object, therby speeding up the printing process. The authors describe the various aspects of HDSP, including the printing process, material characterization, image quality analysis, and process characterization. They also explore different applications of HDSP, such as printing through optically opaque obstacles, overprinting, multi-material printing, and robot-assisted printing. The paper highlights the importance of accurate hologram manufacturing for achieving precise printing results. Overall, this is a comprehensive study that aims at tackling an interesting problem for a clear purpose: Making acoustic assisted 3D printing fast! However, I believe, while this work presents a proof-of-concept and demonstrated some preliminary prototypes, the presented results are mostly not very impressive in terms of resolution, sharpness of the edges, uniformity of the material, and overall quality of printing. There are many issues that are not resolved yet which makes the presentation of this work immature and hence not at the level of the expectations of the readers of Nature Communications. Authors acknowledged several issues listed below that should be overcome to improve the quality of the prints, however this work does go beyond some early characterizations and the depth of issues makes the feasibility of the proposed printing technique for real applications highly questionable:

1. Manufacturing process of holograms is a complex task: The accuracy of the printed parts depends heavily on the accuracy of the manufactured holograms. The hologram manufacturing process can introduce errors and distortions, which can affect the quality and accuracy of the printed objects. It is questionable that if there is a universal hologram that can handle 3d printing of objects with different complexity and geometrical parameters.

2. Uniformity of pressure patterns is challenging: The uniformity of the pressure patterns at the target planes is crucial for the accuracy of the printed parts. If the pressure values are not uniform, it can result in non-uniform printed patterns and image distortion.

3. Selection of ultrasound center frequency and transducer size: The choice of ultrasound center frequency (f0) and transducer active diameter (OD) affects the image quality and accuracy of the printed parts. However, there is a limitation on how much f0 can be increased, as higher frequencies can lead to increased acoustic attenuation and loss of energy reaching the platform.

4. Optimal image plane location: The position of the image plane (Z target) relative to the source can affect the signal-to-noise ratio (SNR) and peak amplitude of the reconstructed image. Selecting

the optimal distance between the object plane and the source is important for achieving better SNR and image quality.

5. Cavitation bubble cloud formation: The interval of the input signal can also affect the formation of cavitation bubble clouds inside the printing part. Altering the interval of the input signal can be another parameter to control the formation of bubble clouds and improve the transparency of the printed objects.

Still, the paper is very well written and the conducted research follows a comprehendible logic that addresses many issues that are facing hologram acoustic printing which is not necessary a weakness, and can be published in more specialized journals focused on additive manufacturing and 3D printing techniques.

Reviewer #3 (Remarks to the Author):

This manuscript presents a novel idea of a holography-based DSP 23 process. The information for printing is stored in cross sections as acoustic holograms. The cavitation caused by sonication polymerizes the printing material at the location instantly. This is an excellent technique for 3D printing of complex structures. One of the advantages of the described technique is its ability for printing at multiple locations simultaneously. Using holographic approach enables storing information content of multiple images in a single hologram. This opens up enormous possibilities for printing multiple objects at different locations.

These authors originated the original concept. The holographic idea using phased array of multiple nozzles is breakthrough innovation.

Overall, this is an excellent manuscript. The concept is very novel and has the potential to revolutionize the area.

The manuscript is well written. The images provided are of very high quality. The authors have done an excellent job in making the material interesting to general readers.

I have couple of questions and comments.

1) What is the defect density in the printed material?

2) Does cavitation process result in any reduction in the material (polymer) properties?

3) What is the spatial separation that can be achieved when printing multiple objects using hologram approach in the printing space?

4) What is the effect of viscosity in the printing process? Does the polymer solution need to be homogeneous?

5) What about the presence of air bubbles in the polymer?

6) It would be helpful for general readers if the authors could add some physics of cavitation process.

Response to Reviewers:

We appreciate very much the constructive and valuable suggestions feedback from all the Reviewers, which have enhanced the quality of our manuscript. In response to their suggestions and comments, we have performed multiple investigation and have revised the manuscript meticulously and thoroughly. The explanations and corrections to all comments were added in the revised manuscript.

To Reviewer 1:

The authors present the application of acoustic holography to sound-based printing, with the ability to create polymerized PDMS components based on acoustic fields defined at a given plane. This appears to be an extension of the authors' previous work, in which they previously applied the acoustic printing principle using a single focused point, where this work produces planes of polymerized images, rather than doing this point by point. The authors compare this work to the difference between SLA vs DLP in optical-based printing, although there is an important caveat. Whereas in the author's previous work designed fully 3D geometries could be produced, with the part being built up point-by-point, in this work the produced parts are limited to ~2.5D geometries, where an identical cross-section is extruded from a PDMS bath. This is somewhat mitigated by the integration of an articulated arm that permits the extrusion to be bent/rotated as it is extruded

RE1: We truly thank you for the detail insights and surgical comments about our paper and we really appreciate your time. Your comments helped us to modify the paper accordingly which elevated the quality of the work. We agree with the reviewer's assessment completely about ~2.5D parts being printed in the present paper. This is the limitation due to the utilization of passive holograms as the stored images on the hologram is permanent/fixed. In order to overcome this limitation of HDSP, active or dynamic holograms are needed to continuously update projected acoustic pressure image.

The goal of the present paper is to introduce the application of acoustic holography in the field of printing with ultrasound. For this cause, we utilized a relatively well-developed passive hologram design workflow. We hope our work could ignite other researchers to not only pursue passive hologram but also develop further in the field of active holograms which is relatively less matured. The future ultimate aim is to develop a dynamic acoustic hologram, functioning analogously to a Digital Micromirror Device (DMD) in light based methods, capable of facilitating fully sound-based 3D printing. This manuscript presents the pioneering effort to incorporate a passive acoustic hologram in Direct Sound Printing (DSP), laying the groundwork for future advancements in the sound-based 3D printing.

We added a descriptive paragraph the Conclusion section to address this comment as:

"Furthermore, in the pursuit of advancing HDSP technology, the integration of active/dynamic acoustic holograms presents a substantial opportunity for enhancement. Programmable acoustic holograms, capable of actively generating desired acoustic fields for various geometries in real-time, can be a substantial development [1]–[3]. Analogous to Digital Micromirror Devices (DMD) used in optics, the Spatial Ultrasound Modulator (SUM) exemplifies this innovation [2], [3]. Utilizing active acoustic holograms such as SUM significantly enhances HDSP's capability, enabling the creation of complex, fully three-dimensional printed objects with intricate details."

The authors could accordingly do more to articulate the limitations and relative merits of the previous work compared to the current one, especially in terms of print speed and highlighting particular applications where the dimensional limitations are not overly deleterious

RE2: We agree with you about the dimensional limitations of the current work, as discussed in RE1, due to the utilization of passive holograms. To address the "particular applications where the dimensional limitations are not overly deleterious" of this comment, the current paper can have a direct application with its current form (using passive hologram) for the application of bioprinting inside the body as the required pattern is relatively simple and 3D geometry in most application (like bone restoration) is not complicated and they are mostly 2D. Recently (after submission of the present HDSP paper) 4 other papers from other research groups were published based on the idea of DSP. The idea of non-invasive deep printing inside the body and its ex-vivo proof of concept was introduced in the early DSP demonstration [4]. A paper was published in *Science* [5] in Dec. 2023 and replicated our work with different printing material and redemonstrated the concept of printing deep inside body. Another paper in *Small Methods*[6] in Jan. 2024 investigated the cell viability and drug delivery. Both works printed extremely simple 1D or 2D objects with single focused ultrasound for the purpose of bioprinting with DSP. Therefore, the current HDPS paper could have a direct application in this exciting field of non-invasive bioprinting introduced by our research group. We have mentioned this in the Special Applications section in the paper and elaborated further in the present modified manuscript in the same section. Two other papers, which are not bioprinting oriented, were published in *Advanced Functional Materials* [7] and *IEEE Ultrasound* [8] respectively, and printed simple objects and filaments in epoxies and elastomers with the DSP concept in late 2023 as well using single focused ultrasound.

We added a descriptive paragraph in the Introduction section to cover the recent published papers as:

"Our pioneering works on DSP and RDP [4], [9]–[11] have garnered significant interest, igniting new research into ultrasound-driven 3D printing. We introduced the paradigm of printing through physical barriers, such as directly inside the human body[4]. The concept of non-invasive deep printing inside the body was initially demonstrated ex-vivo[4]. The chemical reactivity induced by ultrasound sources arises from a mix of thermochemistry, caused by heat generation through acoustic attenuation, and sonochemistry, resulting from acoustic cavitation bubbles. Notably, the temperature rise at the macroscopic scale of the focal region due to acoustic attenuation was insufficient to facilitate on-demand curing in materials like silicone elastomers or epoxies through thermochemistry alone; hence, sonochemical reactions within the cavitation bubbles have been proposed to enable on-demand curing for such materials. Later, hydrogels have been successfully printed using DSP to reaffirm the concept of in-body printing[5], [6]. Recently, an ultrasonic horn with a concave front surface has been used as the primary ultrasound source to create an acoustic focal region and print filaments of thermoset materials[7]. Additionally, a cost-effective sound-based 3D printer has been developed, capable of solidifying silicone elastomer and egg white[8]. In all of the above mentioned works on DSP, single focal ultrasound sources have been utilized."

We also added a descriptive paragraph in the Conclusion as:

"However, the current form of HDSP utilizing passive hologram in this paper could have direct applications in printing the body as the complexity of the desired objects are limited."

1.1. Is there a threshold acoustic power input that's required for cavitation/polymerization? For instance, if the power were decreased to 1W from 5W for a given image, would the polymerization take 5x as long? Or would it not occur at all (or much much slower).

RE3: We sincerely thank the reviewer for the in-depth feedback. Yes, the activation of sonochemical reactions for polymerization indeed requires a certain threshold of acoustic power. The holographic field we employ generates a distributed pattern across the target printing plane, which consists of a grid of voxels. These voxels, when illuminated by the holographic field at higher intensities, must receive sufficient power to initiate polymerization. The power distribution is affected by the geometry of the target object; specifically, it is influenced by the area of the object being printed.

For instance, consider a simple line geometry compared to a maple Leaf target object with an intricate contour. The total power required for printing and solidifying all the desired voxels in the case of the line geometry is lower than that needed for the maple Leaf object, which has a more complex geometry and a greater cumulative number of voxels. Thus, the required power input is directly related to the total area and complexity of the targeted geometry, as the acoustic energy is distributed among the voxels within this perimeter.

As per the reviewer's example of 5W down to 1W and to provide more clarity to the scenario, the relationship between power reduction and polymerization time does follow a somewhat proportional pattern (though not linearly as discussed in Question 1.4 (RE6)), but only above a certain threshold necessary for polymerization to occur. For example, if the geometry used in our manuscript, such as a line, initially requires a minimum of 6W to start the polymerization process, reducing the input power from, for example 15W to 6W would indeed result in an approximately increase in polymerization time by a factor of 2.5^2, proportional to the 2.5-fold decrease in power. This proportionality, however, is predicated on the condition that the power remains above the minimum threshold required to initiate polymerization and sonochemistry.

Below this threshold, the polymerization process would be significantly slowed down or might not occur at all. Thus, while there is a proportional relationship between power input and polymerization time within certain bounds, this proportionality is contingent upon maintaining power levels above the critical threshold required for initiating polymerization. This highlights the importance of optimizing power input based on the complexity of the target geometry and ensuring it exceeds the minimum requirement for effective polymerization.

This comment was addressed in the manuscript as stated in RE6.

1.2. Namely, what is the significance of the 2MPa value?

RE4: The 2 MPa value mentioned in the manuscript represents the estimated minimum pressure required for printing in PDMS obtained in our previous work [4] and also the present work. Therefore, enough power should be utilized to reach or exceed this pressure level. This pressure threshold is critical for initiating the sonochemical reactions required for the polymerization process in PDMS. Due to the limitations of our needle hydrophone, which was not calibrated for immersion in a resin bath, we relied on simulated experiments to determine this threshold. We employed COMSOL simulations (which were validated by experiments in water in our previous work [4]) with input parameters matching those of our actual experimental setup (e.g., the input printing power) to accurately model and determine the input power to create the pressure at the target location necessary to activate these sonochemical reactions.

It is important to note that this pressure threshold may vary when using materials other than PDMS, due to differences in material properties such as attenuation and bulk modulus. These material-specific factors can significantly influence the required pressure threshold for successful polymerization.

This comment is addressed in the Introduction section as:

"this pressure threshold is critical for initiating the sonochemical reactions required for the polymerization process in PDMS"

1.3. Similarly, is there a maximum acoustic pressure beyond which printing speed is not enhanced and/or part quality is negatively impacted?

RE5: Indeed, there is a maximum acoustic pressure threshold beyond which we do not observe further enhancements in printing speed, and part quality may begin to suffer. With the moderately viscous resin used in our experiments, such as PDMS, which is a common and conventional material found in laboratories, we have found that increasing the emitted power or acoustic pressure beyond this threshold disturbs the pattern uniformity. This disturbance occurs because the excessive acoustic force generates streaming within the resin, negatively impacting the print quality. Furthermore, in applications involving extrusion with a feed rate, an increase in printing power that is not proportionately matched with the extrusion feed rate can deteriorate the quality of the print, resulting in shapes that do not closely match the intended design.

Other than increasing the input power, based on our experiments, we observed that Duty Cycle (DC) plays a vital role in printing in HDSP. Although the higher DC will contribute to the higher delivered power, but cavitation generated bubble clouds will form in the final part which will affect the type of the structure of the part (being porous or solid).

Addressing the potential for future improvements, if we were to synthesize and develop sound-sensitive materials with modified characteristics, it could be possible to adjust the threshold of acoustic pressure that the material can withstand before negative effects begin to manifest. This would allow for a broader range of printing conditions and potentially higher quality prints under increased power settings, provided that the material properties are tailored to mitigate the adverse effects of acoustic streaming and other phenomena associated with high acoustic pressures.

We addressed this comment in the Introduction section as:

"we have found that increasing the emitted power or acoustic pressure beyond this threshold disturbs the pattern uniformity. This disturbance occurs because the excessive acoustic force generates streaming within the resin, negatively impacting the print quality."

1.4. Can the polymerization rate as a function of acoustic power or pressure be explicitly quantified?

RE6: The polymerization rate within our HDSP system can indeed be correlated with acoustic power and pressure, although with certain complexities inherent to the process. Importantly, in

HDSP, the polymerization rate is influenced not only by the delivered acoustic power but also by the duty cycle (*DC*) of the transmitted acoustic power applied to the resin.

To quantify this relationship in our system, we conducted a series of experiments in which we systematically varied both the acoustic power and the *DC* delivered to the resin. We have documented these variations and their impact on the printing time required to generate a solid wall by extruding a holographically generated line. This is detailed in Extended Data Fig. 3, which illustrates the combination of power and *DC* with the printing time.

Responding to reviewer's insightful comment, we have conducted a more detailed analysis and included additional data in Fig. R1, now referenced in the manuscript and added to the supplementary materials section. This figure illustrates our parametric study on printing a wall with dimensions of 15×1×20 mm^3, as depicted in Fig. R1d. This schematic represents the line cross-section extrusion generated through holography, originally presented in Extended Data Fig. 3c.

We have expanded our analysis to include a new representation that specifically illustrates the relationship between printing power and printing time when the *DC* is varied, as shown in Fig. R1a. We observed that increasing the *DC* results in extended printing times. Additionally, we introduce the concept of Interaction Strength, σ (W), calculated by multiplying the maximum power (W) and the *DC* (%). Fig. R1b demonstrates how this interaction influences the printing time, allowing us to derive corresponding polymerization rates under these conditions.

This analysis enables us to establish a quantifiable correlation between acoustic power, *DC*, and the polymerization rate, in terms of a Volumetric Deposition Rate (VDR) function, given in Eq. R1. Our findings indicate that the total power required to print geometries of varying circumferential lengths differs, even though the intensity needed for printing each voxel remains constant. Consequently, to enhance the versatility of the VDR function presentation, we incorporated the intensity used in the parametric study, calculated as power divided by the circumferential area of the voxels targeted by acoustic holography for solidification. Fig. R1c shows the plot of this numerically derived VDR function, modeled using a second-order polynomial (with R^2=0.9564):

$$\begin{aligned} &VDR(I,DC) = a_0 + a_{10}I + a_{01}DC + a_{20}I^2 + a_{11}I.DC + a_{02}DC^2 \\ &\text{where: } \{a_0 = 1.24\times10^4,\ a_{10} = -2.88\times10^4,\ a_{01} = -725.89, \\ &a_{20} = 1.96\times10^4, a_{11} = 982.74, a_{02} = 11.53\} \end{aligned} \qquad \text{Eq. R1}$$

where $I = \dfrac{P}{A_{ROI}}$ is the acoustic intensity on the image area, A_{ROI}, and P is delivered acoustic power.

The VDR function is based on two key input parameters, Intensity of the printing, I (W/mm^2) and *DC*. The results are clearly depicted in Fig. R1 c, which shows nonlinear trait. The quadratic nature of the numerically obtained VDR function, is consistent with the quadratic-like trends observed in Fig. R1a and b.

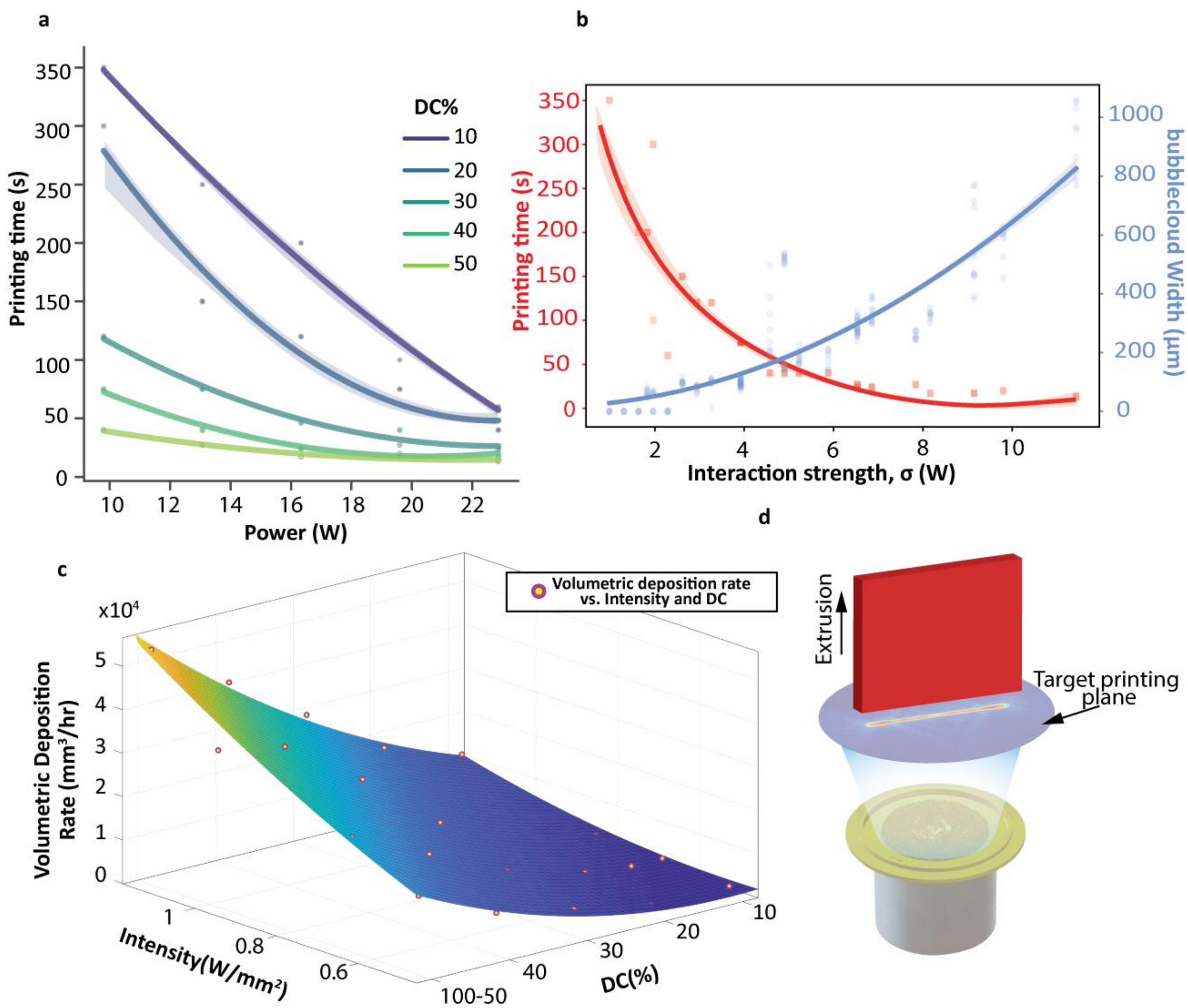


Fig. R1. a. The relation between the power and the printing time for the wall printing with the schematics of the printing process shown in **d.** The relation between the effective power parameter Interaction strength, σ(W) vs. Printing time (s) is shown by red line in **b**, while the bubble cloud formation width is shown in blue along with data with respect to the interaction strength.

We summarized this RE in a section titled "Volume Deposition Rate in HDSP vs. DSP" in the extended data and added Extended Data Fig. 4.

2. Can the authors expand on the mechanism/physical basis by which the printed object appears to polymerize on the platform first/preferentially, rather than in the bulk of the material (where the acoustic field is also present)?

RE7: We thank the reviewer for this question. The platform is placed at the image plane where the acoustic waves form the image and also to meet the threshold of acoustic pressure suitable for printing. Other regions in the bulk of printing medium does not reach this threshold pressure as described in the previous response. Moreover, in low viscosity medium, due to acoustic streaming induced by the acoustic waves, a platform is needed to hold the already solidified part.

We have added a description of this comment in the Introduction as:

> "The platform is placed at the target image plane where the acoustic pressure more than the threshold for printing is reached. Moreover, due to the low viscosity medium, the platform is needed to adhere the already solidified part."

3. Can the authors better characterize/model the improvement in print speed that is feasible using acoustic holograms vs. a single focused point to print objects?

RE8: We thank the reviewer for their insightful query. To address the comparison between the print speed capabilities of HDSP and point-based DSP, we have detailed the mechanism behind the HDSP's enhanced efficiency. Assuming the target printing plane is segmented into a grid of voxels, where each voxel is the minimum resolution that can be solidified with the current experimental setup. The voxels designated by the target image are marked for printing. Fig. R2 a and c, illustrates the targeting operation of HDSP vs. DSP, respectively, for the better visually comparison of the processes. Fig R2 b and d refer to the intensity pattern generated by HDSP's field and DSP's focal point, respectively.

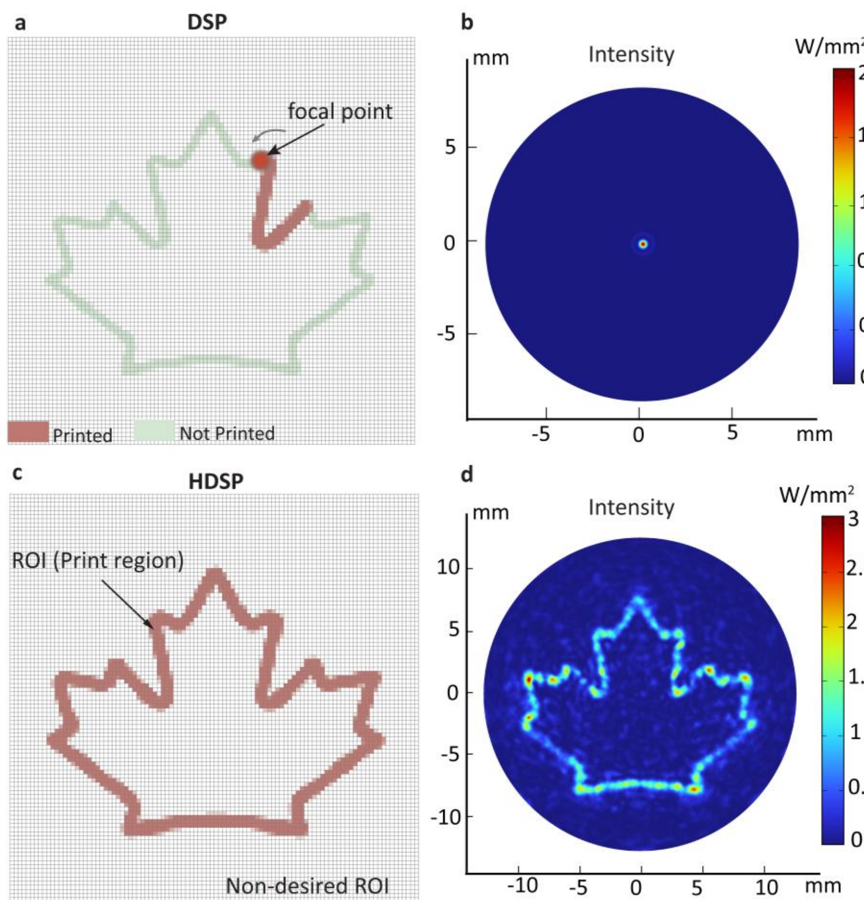
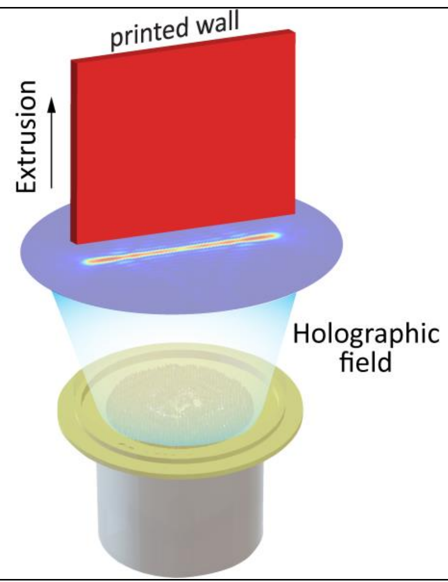
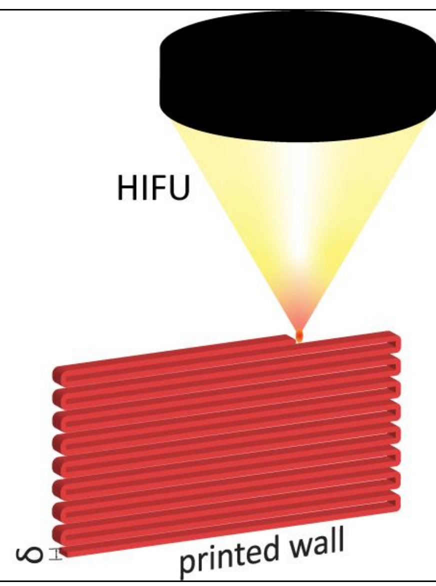


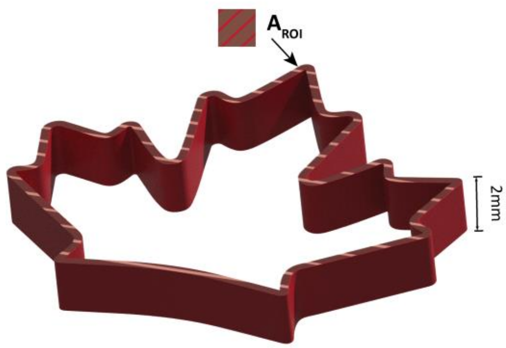
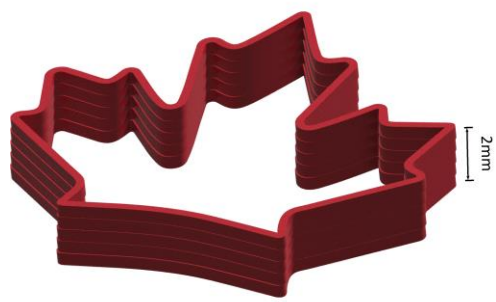
Fig. R2. a and **c** Comparison of the HDSP process vs. DSP in terms of their unit operation. HDSP process is targeting the desired region simultaneously at once, while DSP process is voxel-by-voxel process. **b** and **d** show the minimum intensity pattern required for activating the printing for the DSP and HDSP, respectively.

The time saved by HDSP due to stationary source, and not moving the focal point as in DSP, translates directly into faster overall printing speeds. The ability to solidify a large area at once, rather than point-by-point, is a clear advantage in scenarios where the production speed is a critical factor. We have experimentally compared the printing of a wall of 15×1×20mm^3 obtained by the 2 methods. Since we know the total volume of the printed part, and by selecting a modest power/feed rate setting for both methods of DSP and HDSP, the time takes for complete printing can be measured. The printing time for HDSP using 30W and DC50%was 30s while for DSP with moderate feed rate (240 mm/min), the printing took 12.56 min. Comparing the printing time for HDSP vs. DSP, we can conclude that since the HDSP has more voxels to simultaneously print, it requires much higher energy compared to the DSP, and that's why it is faster. The printing time of HDSP is calculated using the volumetric deposition rate (VDR) calculated in RE6 and the printing time for DSP is path length divided by the feed rate of the source. Therefore, printing time can be experimentally obtained for the HDSP vs. DSP as given in Table R1.

Table R1 Printing time predication for HDSP vs DSP.

<table>
<tr><th>Printing mechanism</th><th>HDSP</th><th>DSP</th></tr>
<tr><td>Schematics of printing

Case study: Wall</td><td>
</td><td>
</td></tr>
<tr><td>Printing time formulation</td><td>$time_{HDSP} = \frac{Volume}{VDR(I, DC(\%))}$
The calculation of Intensity and VDR function is presented in Eq. R1, in RE6</td><td>$time_{DSP} = \frac{length\ of\ path}{feed\ rate}$</td></tr>
<tr><td>Printing time for a wall of 15×1×20mm^3</td><td>$time_{HDSP}$: 0.25 – 5min
Printing wall with (23 W, DC50%) setting:
$time_{HDSP} = \mathbf{0.5\ min}$</td><td>• Length of path/layer = 15mm
• Wall height=20mm
• Layer thickness≈0.3mm</td></tr>
</table>

(from actual experiment)		• Number of layers (transversal)=20mm/0.3mm=67 • Number of layers (lateral)=1mm/0.3mm≈3 • Total length = 15×3×67(mm) • Feed rate (mm/min) = 240 $time_{DSP} = \frac{Total\ length}{Feed\ rate} = \mathbf{12.56min}$
Another example of the maple leaf		
Example Prediction of the time to print the maple leaf object with 2mm thickness	$time_{\mathrm{HDSP}} = \frac{(61.0 \times 2\mathrm{mm}^3)}{\mathrm{VDR}\left(\frac{45\mathrm{W}}{61\mathrm{mm}^2}, 50\right)}$ $= \mathbf{14.35s}$	• length of path/layer (mm) = 61.0 • Part thickness=2mm • Each layer thickness≈0.3mm • Number of layers=7 • Total path length = 61.0×7(mm) • Feed rate (mm/min) = 240 $time_{\mathrm{DSP}} = \frac{Total\ length}{Feed\ rate} = \mathbf{1.77min}$

We addressed this comment in the extended data under "Volume Deposition Rate in HDSP vs DSP" and added Table R1 as a new analysis in the Extended Data Table 1.

4. Are there limits to the dimensions/resolution that can be printed using this approach? I.e. the pattern widths in DOI10.1002/adma.202208002, doi.org/10.1021/acsnano.0c03754 and DOI10.1002/adma.201904181 appear to be well below 1 mm, though are >=3mm here. Would this approach be scalable to higher frequencies that might permit greater resolution?

RE9: We thank the reviewer for referring and highlighting the resolution limitation in HDSP, as this is crucial in the printing process. Yes, increasing the frequency would lead to better resolution in HDSP. The resolution in HDSP is primarily governed by the wave interference principles of acoustic holography and the wavelength, λ, of the acoustic waves used. Theoretically and ideally, the acoustic hologram can achieve a diffraction-limited resolution of approximately λ/2, in areas of higher-pressure. However, in practice, the actual reconstructed image thickness is observed to be around λ, as demonstrated in Fig. R3. This discrepancy happens due to the diffraction effects and interference that leads to a softening of sharp edges or blending in the reconstructed image. This can be seen from the inset of Fig. R3 **a-c** where the total thickness of the reconstructed image including the high-pressure zones and the neighboring blending area is technically around ~λ. The resolution of only region of the high pressure is of ~λ/2. On the other hand, the uniformity of the high-pressure zones from the reconstruction with the Iterative Angular Spectrum Method (IASA) is needs to be improved. This is the topic of our ongoing study, which we believe will significantly advance the fidelity of acoustic holography used for HDSP.

Depending on the thickness of the desired target image in terms of λ, minimum achievable feature thickness of the reconstructed image out of acoustic hologram is almost one wavelength. To explain in detail, assume the grid of the pixels in the holographic plane as well as the target plane of N×N dimension with each pixel size, δ, is λ/5 and the target image with the same dimension. Theoretically, the reconstructed image's minimum feature thickness is approximately one λ, or nearly five pixels. This means that even if the target image has a thickness of 1 pixel, the reconstructed image will have a thickness of about λ, equating to roughly five pixels. For thicknesses below one λ (e.g., 1px, 2px, or 4px), the reconstructed feature will still span one wavelength. If the target image's thickness lies between λ and 2λ, the reconstructed feature will span 2λ, and so forth.

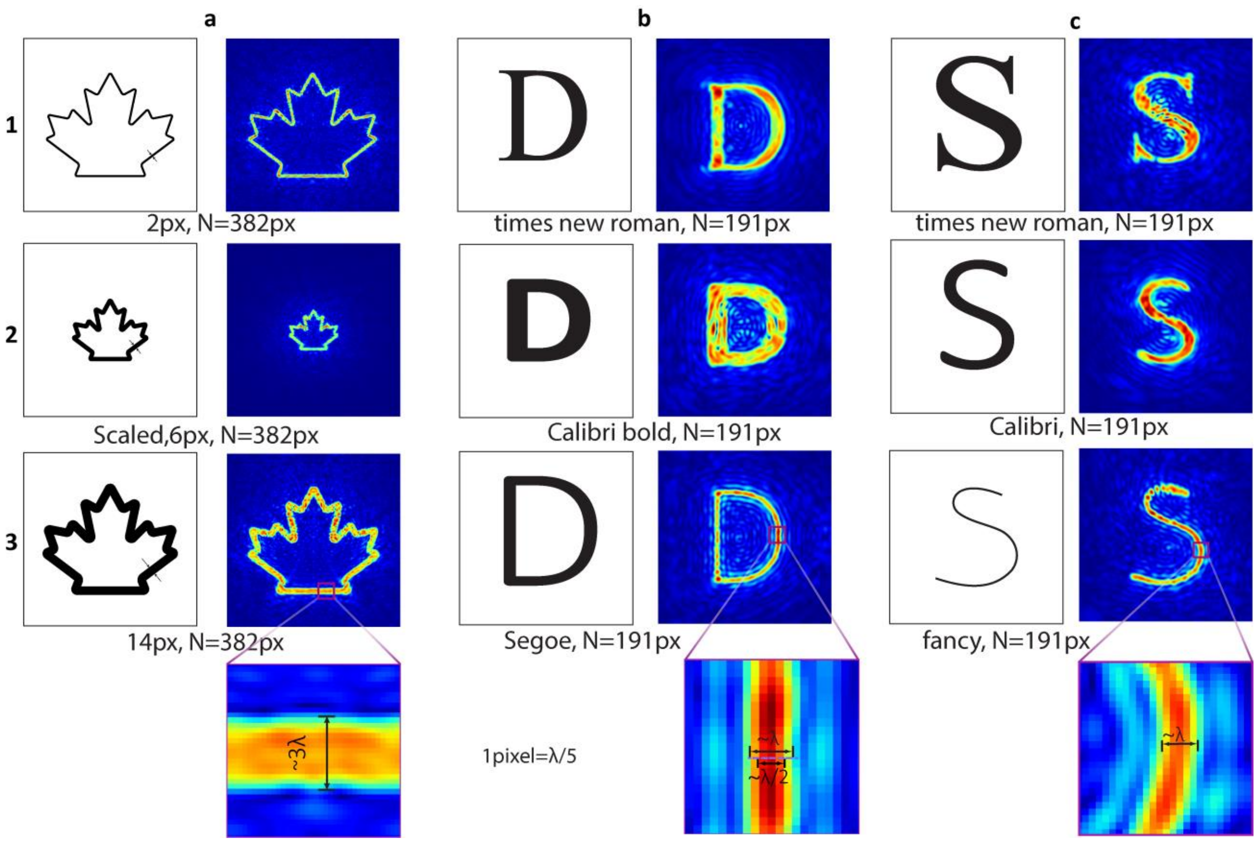


Fig R3. Reconstruction of the various target images. **a** 1-3 shows the effect of geometry variation such as scaling and thickening on the reconstruction. **b** and **c** illustrate the text geometry and the variations such as various font on the font size on the reconstruction

To explain the feature resolution appears in our printed parts, firstly, we used thicker images to ensure uniform pressure projection across the target plane within the resin. If we were to use the smaller thickness of the target image, let’s say 1px, although theoretically we get the better resolution, however, the reconstructed pattern does not have satisfactory uniformity, results in partial solidification of the part. It can be seen in the reconstructed image from the suggested article by the reviewer [12]–[14]. Although the reconstructed thickness is approximately one λ, which in the frequency range of 1.5-2.5MHz, will be around 0.7-0.9mm.

In the reference [12] it is noted that they experimentally found that the resolution is below ~λ/2 which corresponds to only higher-pressure zones which are also disrupted and not uniform generating acoustic force for cell/particle trapping. In our case, along with those diffraction limited resolution from high pressure region, the blending area is also might contribute to the printing and inducing heat, making the pattern thicker. By further improving the pattern uniformity and improving the IASA acoustic holography technique and tuning the power threshold for printing so that only the high-pressure zone reaches the printing and not the lower pressures.

The second reason is that acoustic holography allows us to freely manipulate various geometric features, such as thickening, scaling, warping, and even the type of fonts for text shapes. The printed parts presented in Fig. 1 of our manuscript are deliberately selected with various types rather than a line/spline shape with default 1px thickness. To provide a detailed example, Fig. R3 illustrates reconstructions for various target objects. Fig R3. a-1 displays the target image for a shape geometry with 2px thickness and 382×382 pixels image on the left, alongside the corresponding reconstructed image on the right. The effect of scaling and the thickening the pattern is evident in Fig. R3 a-2 and a-3. Demonstrating how the scaling and thickness of the target image affect on the reconstruction. Moreover, regarding the text geometry, Fig. R3 b and c show the letter “D” and “S” in various fonts in images 1-3, along with their corresponding reconstructed images. These results clearly show how variation to the geometry of the target image affect on the reconstruction process. Specifically, the letters “D” and “S” in our manuscript, with a thickness greater than λ, results in printed thickness of approximately ~2λ in final product.

The third reason, is that since the platform was submerged into the resin, during the experiments we couldn’t visually monitor the ongoing printing in real-time, making it challenging to halt the printing process in time. To compensate, we extend the duration of the printing process slightly for a few seconds more, even beyond what was strictly necessary, to ensure all the regions are solidified completely. However, keeping the acoustic source active in the resin creates more polymerized material in previously solidified areas. These spots promote further solidification in adjacent, yet-to-be-solidified resin, leading to thicker final parts than initially intended. Therefore,

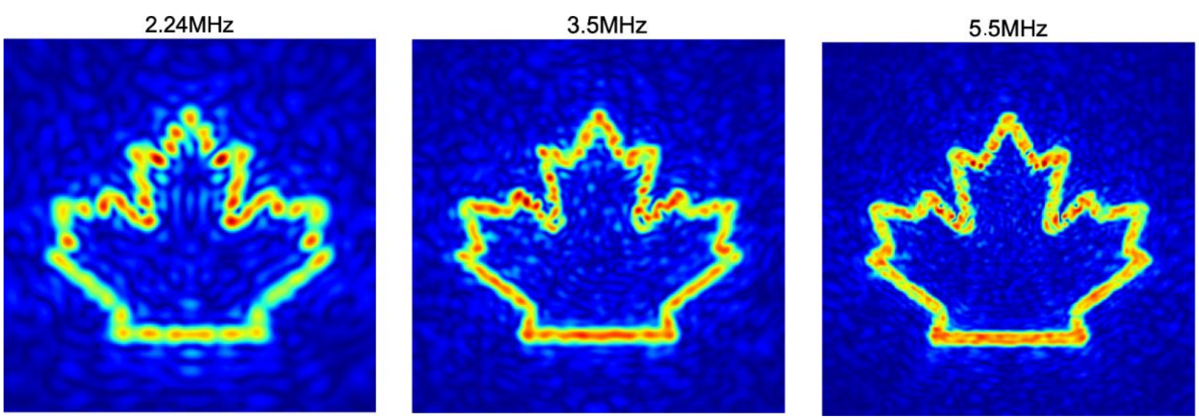


Fig. R4. Demonstration of resolution improvement by higher frequencies. The images are reconstructed with transducer OD25mm and target plane of Z=30mm.

there is a need for careful timing and control of the printing process, especially in setups where direct visual monitoring is not feasible.

Finally, upon reviewing the experimental observations from both DSP and HDSP, we realized that the printing resolution, while directly related to the image resolution from the patterned acoustic waves, is not the only factor affecting the resolution of the final printed part. The parameter *DC* (which mainly corresponds to the transparency and porosity) in HDSP plays a crucial role. We observed that using a higher *DC* leads to increased resolution, while decreasing the *DC* results in lower resolution.

In response to the reviewer's question regarding "*The possibility of using higher frequencies for the improved resolution*", theoretically the higher frequency such as 3.5MHz can have much pattern uniformity and better Sound to Noise Ratio (SNR), due to the fact that the spatial resolution of the hologram increases, enabling the hologram to reconstruct with much richer details. Moreover, the reconstruction with higher frequency, results in inherently more Uniformity of the pattern and significantly improved quality. Specially the images with finer details such as sharp edges and corners such as the reconstructed "Mandala" design shown in the Fig. 6 in the manuscript. Additionally, here Fig. R4 illustrates the reconstruction of the similar geometries for higher frequencies for the same transducer size of OD 25mm, targeting the image plane of Z=30mm. However, upon increasing the frequency, the acoustic penetration depth diminishes due to increased material attenuation, which can limit the effective range of the sound waves within the material.

Managing the adaptability of HDSP to use higher frequencies while maintaining the trade-off between the penetration depth and attenuation is a critical area of our ongoing research. Our future efforts aim to control more on balancing the resolution improvement with practical limitation with the material's inherent properties.

We have addressed the physical resolution limitation in HDSP in the revised manuscript as below:

> "In exploring the resolution capabilities of HDSP, it is crucial to understand the physical limitations imposed by the principles of acoustic holography. The resolution of HDSP is primarily governed by the wave interference patterns and the wavelength, λ, of the acoustic waves utilized. Theoretically, the acoustic hologram is capable of achieving a diffraction-limited resolution of approximately $\lambda/2$ in areas of higher pressure. However, in practical implementations, the actual reconstructed image often exhibits a thickness around λ. This discrepancy arises due to diffraction effects and interference that soften sharp edges or cause blending in the reconstructed image, as can be seen in the inset of Fig. 6b.
>
> Moreover, beyond the acoustic parameters, the *DC* (which mainly corresponds to the transparency and porosity) in HDSP significantly influences the final print resolution. Experimental observations indicate that higher *DC* settings enhance resolution by increasing the precision in energy delivery to the target regions. Conversely, lowering the *DC* leads to reduced resolution."

5. When comparing HDSP vs. DSP times, there should be greater clarity on what basis the comparison is made. Is the total energy deposition identical in both cases, and/or are there limits to the total energy density that are appropriate for DSP? Or is this limited by the translation speed of the DSP method?

RE10: We thank the reviewer for their insightful comment to have better comparison between the HDSP vs. DSP in terms of the total energy and the timing. We have investigated deeply into our simulation models as well as new experimental observation and tryouts, in order to comprehensively and quantitatively compare the HDSP vs DSP in terms of total energy deposition.

We aimed to provide clarity on the basis of comparison, particularly concerning the total energy deposition and the energy density appropriate for each method. Considering the case of printing the same object "Wall", previously mentioned in response to question 3 of the reviewer. The simulations, with simplified environmental parameter neglecting the heat dissipation etc., assume an equal pressure of 2MPa, both at the focal point in DSP and across the target image in HDSP. We have obtained the outcomes such as the intensity pattern as well as the overall generated power on the target plane, as can be seen listed in Table R2. We have compared HDSP and DSP in terms of total energy deposition and total energy density, two critical metrics for assessing the efficiency and suitability of each method.

Total Energy Deposition (J):

Our analysis reveals distinct differences in total energy deposition between HDSP and DSP. Specifically:

- DSP, conversely, operates at a lower power (1.18W) but over a longer period (12.56 minutes), leading to a total energy deposition of approximately 889.25 Joules.
- HDSP uses a higher power (23.18W) over a shorter duration (~0.5 minutes), resulting in a total energy deposition of approximately 695.4 Joules.

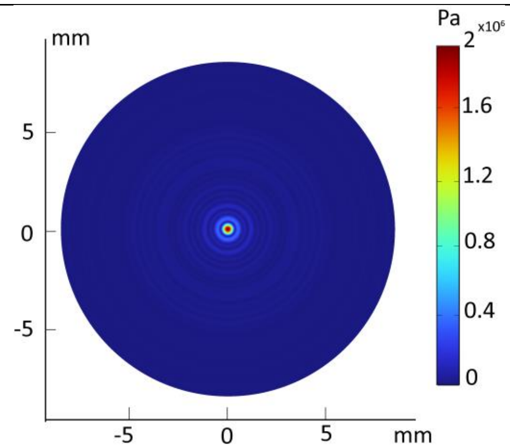
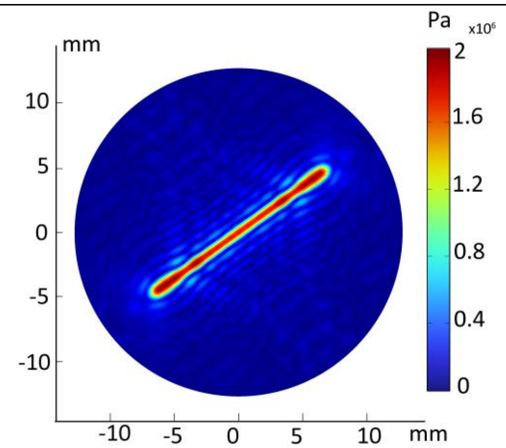
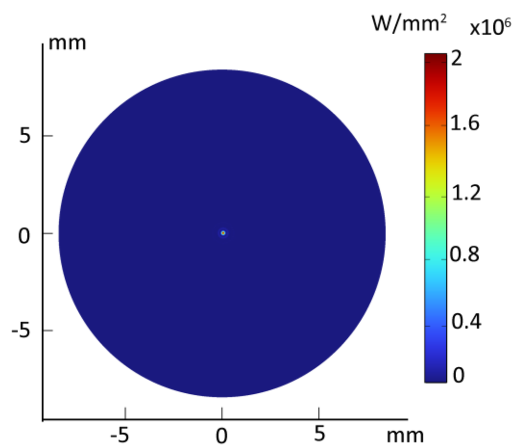
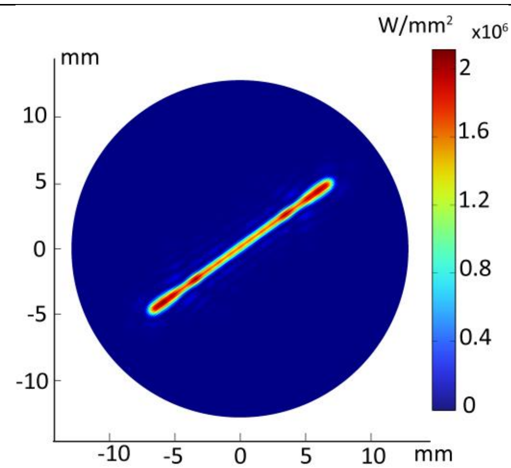
Total Energy Density (J/m^3):

The total energy density, which considers the energy used per cubic meter of the printed object, provides a metric that more accurately represents the efficiency of each printing method:

- **DSP** Utilizes a total energy density of approximately 2.964 J/mm^3. This higher energy density indicates a concentrated use of energy per unit volume, which is inherent to DSP's method of focusing energy tightly to a single point or small area.
- **HDSP** Shows a total energy density of approximately 2.318 J/mm^3. While this is slightly lower than DSP, it reflects HDSP's ability to efficiently distribute energy across a larger area, thus completing the printing process more quickly and using energy over a broader area effectively.

From the calculations, despite using lower power, DSP consumes more total energy due to the significantly longer time required to complete the printing. This is primarily because DSP has to mechanically trace the path layer by layer, which is inherently more time-consuming compared to HDSP, which can create complex layers in one go.

Table R2. Comparison of the HDSP vs DSP in terms of deposition power, total deposition energy and energy density

Parameter	DSP	HDSP
Frequency (MHz)	2.15	2.24
Geometry and area of the target image	Focal point, 0.3848 (mm^2)	Line, 6.63 (mm^2)
Pressure pattern		
Intensity pattern(W/mm²)		
Deposition power at the image plane (W)	1.18	23.18
Total Printing Time (min)	~12.56	~0.5
Total Energy Deposition (J): power × time	**Simulation:** 1.18W×12.56×60=889.25	**Simulation:** 23.18W×0.5×60=695.4
Total Energy Input to the system (J)- (from experiment)	**Experiment:** 10W×12.56×60=7536J	**Experiment:** 30W×0.5×60=2790

Moreover, in actual experimental conditions, factors such as scattering, temperature variations, setup accuracy, and transducer efficiency in converting electrical to acoustic power necessitated the use of higher power than that suggested by the simulation. Despite the increased power supplied in the experiments, the total energy deposition for DSP remained higher than that for HDSP.

In summary, the total energy consumption depends on how efficiently each method uses power over time. In this example, although HDSP uses more power at any given moment, it completes the task much faster than DSP, leading to a lower total energy consumption when calculated over the entire duration of printing. Thus, HDSP could be considered more energy-efficient for the given scenario, despite its higher instantaneous power usage. The results finding from simulation insights and real-world experimental data provides a comprehensive understanding of the relative efficiencies of DSP and HDSP. Looking at a broader framework, these analyses suggests that the choice between HDSP and DSP can be tailored to specific application needs. If the focus is on maximizing energy concentration, DSP is suitable; for enhancing production speed with efficient energy distribution, HDSP proves more advantageous.

Regarding the reviewer's comment about energy density of DSP: "Or is this limited by the translation speed of the DSP method?" they have referred to a very useful findings in terms of energy and energy density:

Based on our findings, the translation speed of the DSP method inherently limits its operational efficiency. DSP's need to mechanically trace each voxel of the print path slows down the process, thereby increasing the total print time and energy expenditure. HDSP, with its capability to impact a larger area simultaneously through holographic techniques, markedly reduces the print time, albeit at the expense of requiring higher power to spread the energy over a larger area.

We have added these energy analyses discussion to the revised manuscript, and added the Table R2 as the Extended Data Table 2. We have added the following text in the revised manuscript:

> "In this study, we have conducted detailed analyses of the printing processes for HDSP and DSP, focusing on power consumption and total energy in deposition. Both simulation and experimental data were analyzed for printing a wall with identical dimensions using both methods. Our findings reveal that while DSP requires less power per unit of time, it consumes more total energy due to its longer operational time compared to HDSP. Furthermore, although DSP exhibits a higher energy density, suggesting intense energy use per unit volume, HDSP compensates for this with substantially faster printing times, making it highly advantageous for applications that require quick production without significantly compromising on energy efficiency. These insights are further elaborated in Extended Data Table 2, which details the comparative analyses"

6.1. The examination of printing at multiple heights (as in Fig. 4b) in the presented application would appear to have limited utility. What applications would benefit from such multi-object/location printing?

RE11: We thank the reviewer for providing their thoughtful comment on this application of the HDSP, which encourage us to delve deeper into the potential applications. When conceptualizing this application of the multi-object printing using the acoustic hologram, which capable of encoding the information for multiple target planes, we were primarily considering the possible application of in-body printing, under the group of Remote Distance Printing (RDP) application. Our findings show promising approach as listed below:

- Tissue Structures: Knowing the tissue structures that comprises various layers, it is possible to print multi-layered tissue structures for example creating skin grafts/patch with distinct layers (dermis, epidermis). This requires development and synthesize of the suitable sound-printable bio materials.
- Localized Drug and Cell Delivery Systems: The ability to print drug-loaded structures at multiple locations within a gel matrix that could be implanted in the body, for the release of different therapeutic agent at targeted sites for combination therapies.
- Tissue Engineering: Printing multi-layered tissue patches for repairing an organ, where different growth factors are deposited at specific depths could significantly enhance regeneration and integration with host tissue.

It is important to note that realizing the full potential of this technology needs further collaboration and more interdisciplinary approach to enhance this research.

We have added these applications for the multi-height/object printing in the revised manuscript as follows:

> "The HDSP's ability to encode information content for multiple objects thanks to the precise control on acoustic holography, allows for innovative applications such as complex tissue structures, localized drug and cell delivery systems, and advanced tissue engineering. For instance, it enables the printing of multi-layered tissue structures like skin grafts with distinct layers (dermis and epidermis), each tailored with different cells and growth factors for enhanced healing. Additionally, HDSP can be used to create drug-loaded structures within a gel matrix at targeted locations, crucial for combination therapies where different therapeutic agents are released at specific sites. Furthermore, the technology supports the creation of tissue engineering scaffolds with gradient distributions of growth factors or cells, promoting organ repair and regeneration. These capabilities demonstrate HDSP's potential to address complex biomedical challenges, though further development and interdisciplinary collaboration are essential for optimizing material properties and printing parameters for clinical applications"

6.2. Moreover, the authors may wish to discuss potential future implementations based on modifiable acoustic holograms in which the entire acoustic hologram changes at a given plane, which may give this approach additional functionality (e.g. doi.org/10.1038/s41467-020-18347-2, doi/10.1002/advs.202301489)

RE12: We thank the reviewer for their encouraging us to enhance our manuscript further by discussing the potential of modifiable acoustic holograms. The integration of active, reconfigurable acoustic holography with HDSP, rather than using passive acoustic holograms, is indeed represents a significant improvement.

The studies referenced by the reviewer, emphasize the feasibility and added functionalities of employing reconfigurable acoustic holography. Such advancements would allow for the precise fabrication of complex tissue structures with varying layers and geometries, significantly expanding the applicability of HDSP in creating intricate and multi-layered objects.

A notable challenge that we anticipate in multi-plane object printing in HDSP is that each target image planes should have a sufficient separation to achieve a structural fidelity to the desired target image and maintain high SNR, without introduction of noise. The use of reconfigurable acoustic hologram can mitigate this issue, by allowing on the fly adjustment of the holographic patterns. This ensures that each generated image closely matches the target design with high structural similarity and SNR, thereby enhancing the quality and versatility of the printing process.

We have added included the advancement of HDSP utilizing the active hologram in the revised manuscript in the discussion section, and cited the suggested articles. The added text is as follows:

> "Furthermore, in the pursuit of advancing our HDSP technology, the integration of active/dynamic acoustic holograms presents a substantial opportunity for enhancement. Programmable acoustic holograms, capable of actively generating desired acoustic fields for various geometries in real-time, can be a substantial development [1]–[3]. Analogous to Digital Micromirror Devices (DMD) used in optics, the Spatial Ultrasound Modulator (SUM) exemplifies this innovation [2], [3]. Utilizing active acoustic holograms such as SUM significantly enhances HDSP's capability, enabling the creation of complex, fully three-dimensional printed objects with intricate details."

7. "accuracy of λw/8" – does this exactly correlate to a pixel dimension (80µm in the methods)?

RE13: Yes indeed. We wanted to mention the pixel dimension and data acquisition using the confocal microscopy, in terms of λ_w. Since one wavelength in water medium in that frequency is 660µm, then 80µm will be ~$\lambda_w/8$.

8. "Therefore, there is an optimum value for f0 based on the printing setup and the printing material used" Can the authors estimate what this might be for the printing setup used in this work?

RE14: Through our extensive experimentations within our printing framework, we have observed that higher frequencies, particularly above 2.5MHz yield sharper pattern due to finer resolution associated with the shorter acoustic wavelength. However, higher acoustic power is required for the printing with our current material choice, PDMS. This is due to the fact that higher frequency waves tend to be absorbed as the attenuation increase with the frequency of the incoming wave.

Similarly, we have also observed that lower frequencies, such as 1.85MHz is also not suitable since they cannot generate high fidelity image. Although they require less power, these frequencies do not provide the high fidelity in the printed images that we aim for, likely due to the coarser resolution afforded by the longer wavelengths.

Given these considerations, we have identified an optimum frequency range that balances pattern sharpness with energy efficiency and material compatibility. For the PDMS material used in both HDSP and DSP methods in our work, frequencies around 2.15MHz to 2.3MHz represent a practical compromise, offering a balance between resolution and the acoustic power required for effective printing.

It is noteworthy that this finding is specific to the materials currently employed in our HDSP and DSP setups, mainly PDMS. Upon the development of the more specialized sound sensitive resins, which might be functionalized with higher frequencies, we anticipate that the optimum frequency for achieving high-resolution prints with efficient energy use could shift. Such materials could potentially reduce the attenuation challenges at higher frequencies, allowing for sharper and more precise patterning without the need for proportionally increased acoustic power.

9. There are a range of powers and DC (10-100%) values used in the various prints in this work. Can the authors provide a better understanding of how/why a given power/DC values were chosen for these different prints? Is it the case that a more opaque part was desired in the higher DC% cases? It's notable that "DC 100% results in fully non-transparent and porous structurers [sic]", but no DC 100% data is presented.

RE15: The power we present in the Extended data Fig. 3 is actually the calculated delivered acoustic power in the printing location. We obtained the values from the following equation, Eq. R2:

$$P_{delivered} = P_{\text{electrical input}} \times \eta_{\text{transducer}} \times transmission\ loss \quad \text{Eq. R2}$$

where:

- $P_{delivered}$ is the acoustic power effectively delivered to the printing location.
- $P_{\text{electrical input}}$ is the input electrical power supplied to the transducer.
- $\eta_{\text{transducer}}$ represents the efficiency of the transducer in converting electrical power into acoustic power. According to the manufacturer, it was 49%.
- transmission loss is obtained from COMSOL simulations, representing the ratio of power at the target plane to the power at the source plane, accounting for the decrease due to the propagation through multiple materials. For the transducer with the OD25mm, the calculated $transmission\ loss$ is 0.67 times.

This calculation approach enables us to accurately quantify the acoustic power that reaches the printing location, rather than the mere input power to the transducer. By doing so, we account for both the inherent efficiency of the transducer in converting electrical to acoustic power and the attenuation of power as the acoustic waves propagate through various materials.

Regarding the *DC* we observed that it has an important impact on the bubble cloud formation within the resin, that is critical for printing quality. Bubble clouds tend to form in areas experiencing the maximum negative pressure, and their formation is significantly influenced by the *DC*. To systematically assess and control this phenomenon, we conducted a parametric sweep of the DC values, ranging from 100% down to levels where no bubble cloud formation was observable.

In the current development of HDSP, we realized that the bubble cloud formation occurred in an arbitrary and inconsistent manner, predominantly at locations of highest negative pressure. This is in contrast to the consistent porous structures we printed with DSP. Our results indicate that from 100% *DC* down to 50%, the print quality was almost similar. However, upon further decreasing the *DC* in steps of 10% down to 10%, we meticulously identified the specific *DC* level at which bubble clouds ceased to form. Through detailed inspection with smaller steps of 1% *DC*, we determined the *DC* threshold where no bubbles were observed was 14%.

We should note that, the presented result in Extended Data Fig.3 is applicable for the line geometry, which is an example specific to it. This means that the exact *DC* values observed cannot universally applied to print different geometries. However, the observed trend on influence of the *DC*% to the bubble cloud formation is similar for various shapes. As mentioned in response to question 5 (RE10), discussing about the energy density and the total deposition energy, for more complex target images, higher total energy must be used, to ensure that all the voxels will receive the solidification energy. Introducing *DC*% influences the average power delivered to the printing location, thereby affect the bubble cloud formation in geometry-dependent manner. We anticipate for the geometries with the same circumferential length, bubble could formation with specific *DC*% values, potentially exhibit similar patterns.

We have modified the section explaining the role of *DC* on the porosity in the revised manuscript and enhanced the clarity of the text as can be read below:

"In this parametric study, we observed that from 100% *DC* down to 50%, the porous width was almost similar. However, upon further decreasing the *DC* in steps of 10% down to 10%, we meticulously identified the specific *DC* level at which bubble clouds ceased to form. Through detailed inspection with smaller steps of 1% *DC*, we determined the *DC* threshold where no bubbles were observed was ~14%."

10. "similar to SLA when DLP was utilized" – the authors could be more specific in noting the increase in dimensionality between these methods (1D vs 2D).

RE16: We thank the reviewer for their constructive feedback. We have enriched this sentence according to their suggestion. Here is the updated version in the revised manuscript:

"The present work introduces a significant advancement in the dimensionality of the sound-based printing techniques, transitioning from the point-based approach of DSP, to employing acoustic holography. This method allows for the manipulation of acoustic fields to simultaneously create an image of the entire layer. This paradigm shift is similar in the evolution observed in the photopolymerization printing processes which initially originated from the laser point-based SLA. SLA utilized a methodical approach, employing a laser to solidify resin selectively, tracing specific areas one at a time in a one-dimensional (1D) manner. Subsequently, the introduction of DLP represented a significant evolution in printing speed and efficiency, shifting from SLA's sequential, point-by-point curing method, to a more efficient, 2D strategy. By projecting entire cross-sectional images onto the printing platform, DLP facilitates the simultaneous curing of whole layers, significantly enhancing both speed and efficiency in the photopolymerization printing process."

11. Please provide additional details on the "step-down matching unit" that was used (i.e. manufacturer/model or circuit makeup).

RE17: The stepdown electrical matching transformer we used is a commercial matching unit from the same company of the transducer manufacturer, American Piezo Company (APC). The model is #90-4496 and it has the following specifications:

- Freq 500kHz to 5MHZ
- Power 200W Max.
- Input = 50 Ohm
- Output ={9, 16, 25, 38} Ohm

The details of the model can be found in the following link:
https://www.americanpiezo.com/images/stories/content_images/pdf/amplifers_pdf.pdf

We have added further details for the electrical matching transformer in the revised manuscript:

> "A step-down electrical matching unit (model #90-4496, American Piezo Co, USA) to connect the transducers with 20Ω impedance to the power generator with 50Ω was employed to ensure the best electrical matching between transducer and the power generator"

12. What are the transducer thicknesses?

RE18: The transducers that we used in this study is commercial from American Piezo company (APC) with the piezo disk potted with the aluminum housing. All the transducers, with both piezo disks having the same thickness of ~0.9mm for the transducers with 2.28MHz and 2.24MHz center frequency and ∼1.1mm for the transducer with 1.86MHz center frequency. We have added the detailed information of these transducers to the revised manuscript as follows:

> "Three single element flat transducers of various dimensions and frequencies were used in this study. These commercially available flat transducers (American Piezo Co, USA) have active elements of ~0.9-1.1mm thicknesses, with *OD* = 50mm, 35mm and 25 mm with center frequency, f_0, of 2.28 MHz, 1.86MHz and 2.24 MHz, respectively. Each transducer was encapsulated with aluminum housing, as provided by the manufacturer"

13. Please provide more detail on the luminol solution preparation (concentrations, amounts, etc.)

RE19: The luminol solution for the sonochemonoluminescene experiments, we used the exact solution from DSP [4]. We have added the details of the solution in the revised manuscript as follows:

> "A 1 mM solution of luminol (3-aminophthalhydrazide, Sigma-Aldrich, Canada) is prepared, and the pH is adjusted to 12 using NaOH (Sigma-Aldrich, Canada). The pH is continuously monitored in real time using a pH 315i meter (Wissenschaftlich-Technische Werkstätten, WTW, GmbH, Germany). Subsequently, 0.5 M sodium carbonate (Na2CO3) is added to the solution. The sodium carbonate is produced by heating sodium bicarbonate (NaHCO3, commonly known as baking soda) to 100°C."

14. What other materials might this method be applicable to besides PDMS?

RE20: We appreciate the reviewer's question. We have predicted in DSP paper [4] that any heat curing polymer could be a candidate for DSP and DSP related methods. We have tested on different types of silicone elastomers and epoxies as we reported in DSP paper. Recently (after submission of this paper), 4 research groups independently could print in acrylates [7], epoxies [7], silicone elastomers[7], [15], egg white [15] and hydrogels [5], [6] such as poly(ethylene glycol) diacrylate (PEGDA) and polyvinyl alcohol methacrylate (PVA-MA). We are extremely excited to see that other research groups worldwide started to explore different possibilities in DSP and related technologies. It should be noted that the material is better to have a low exothermic behavior in order to maintain the desired resolution and avoid unwanted macroscopic heat induced polymerization.

Language. This work could do with a comprehensive review of the wording/writing to pick up gramaticall errors and issues. A few are given below.
RE21: We appreciate the reviewer's detailed feedback that helped us to enhance our manuscript. We have implemented all the mentioned grammatical corrections in our revised manuscript, as highlighted by yellow in the manuscript.

1. P1. "acoustic field could reach", could be "acoustic field can reach"
2. "so that it could break", could be "to break"
3. "DSP tames the cavitation phenomena for creation rather than destruction", this seems a bit more poetic than typical academic discourse, especially when the "destruction" that cavitation produces often "creates" beneficial ends, for example enhancing blood brain barrier permeability.

Indeed, the cavitation has useful advantages that are being discovered and implemented such as their application in enhancing the permeability of blood brain barrier, as mentioned. We have modified the sentence as follows:

> "Cavitation, once predominantly viewed as a destructive force[16], has recently become the focus of exploration for beneficial and innovative outcomes. This includes applications in medical and biomedical[17], environmental management[18] and industrial processing[19]. DSP also leverages the cavitation phenomena for creation. DSP is a unique AM method enabling the direct printing of materials, such as heat curing thermosets, which are difficult to process with light or heat."

4. "in which difficult to", should be "in which it is difficult to"
5. P3. "Hologram coordinate system", "The hologram coordinate system"
6. "single chemically active region", "a single chemically active region"
7. "need to be calculated", "needs to be calculated",
8. P5. "Inside body", "inside the body"
9. There are several acronyms that are introduced throughout the manuscript that are seldom used (only once or twice, or even not at all, e.g. ECS, HCS, UAMR). These do not enhance clarity and should be removed.

Reviewer #2 (Remarks to the Author):

This paper presents a layer-based additive manufacturing technique based on the process of holographic acoustic assisted 3D printing, called as HDSP. This research builds on previous work of the authors (Habibi, M., Foroughi, S., Karamzadeh, V. & Packirisamy, M. Direct sound printing. Nat. Commun. 13, 1–11, 2022) and aims at extending the previous point-by-point printing process to a full cross-section of the segmented object, therby speeding up the printing process. The authors describe the various aspects of HDSP, including the printing process, material characterization, image quality analysis, and process characterization. They also explore different applications of HDSP, such as printing through optically opaque obstacles, overprinting, multi-material printing, and robot-assisted printing. The paper highlights the importance of accurate hologram manufacturing for achieving precise printing results. Overall, this is a comprehensive study that aims at tackling an interesting problem for a clear purpose: Making acoustic assisted 3D printing fast! However, I believe, while this work presents a proof-of-concept and demonstrated some preliminary prototypes, the presented results are mostly not very impressive in terms of resolution, sharpness of the edges, uniformity of the material, and overall quality of printing. There are many issues that are not resolved yet which makes the presentation of this work immature and hence not at the level of the expectations of the readers of Nature Communications.
Authors acknowledged several issues listed below that should be overcome to improve the quality of the prints, however this work does go beyond some early characterizations and the depth of issues makes the feasibility of the proposed printing technique for real applications highly questionable:

RE22: We appreciate the reviewer's opinion and thank you deeply for your comments. We understand the point view of the reviewer. However, we respectfully disagree with this assessment. We would like to highlight the foundational nature of this work as an initial exploration into the realm of patterned sound waves in ultrasound based 3D printing. *Nature Communications* is a journal covering pioneering papers with multidisciplinary audience. This technique initiates a new paradigm in Additive Manufacturing (AM) by integrating various research fields including, to name a few, acoustics, holography, material science, polymerization, bio and tissue engineering and computer science, additive manufacturing, mechanical engineering, material and chemical engineering and in-body printing applications.

To emphasis the *Nature Communications'* role in advancing science by covering pioneering works, we can mention the publication of our previous work (Direct Sound Printing) in Nature Communications back in 2022 which opened up exciting and uncharted territories for future research. Different researchers worldwide started working on this concept and published their works recently. A paper published in *Science* in Dec. 2023 [5] and replicating our work even with the same equipment for the purpose of inside body printing. Another paper in *Advanced Functional Materials* [7] used an acoustic horn to print filaments. A different work published in *Small Methods* [6] used an ultrasound imaging head to print hydrogels for the purpose of in-body printing. And a desktop DSP printer was also observed recently by another research group [15]. *Science* recently covered the topic of printing with ultrasound in a perspective piece "Using ultrasound to 3D-print materials) [20] and *Nature Reviews Materials* also covered an opinion piece "3D printing through tissues" [21].

We are proud that our initial work opened up new directions for researchers in different disciplines in different parts of the world to build on what we have started and publish in well-respected avenues (rest alone the media outreach of the work [22]). One of the main reasons of such a worldwide reaction is the reach of *Nature Communications* to diverse audience. The current work on HDSP has the same value and one of the best outlets for such multidisciplinary work is *Nature Communications*. Comparing DSP/HDSP with SLA print quality is wrong because SLA can not print in the materials that DSP/HDSP can print in. SLA cannot print remotely as DSP/HDSP is able to print deep into opaque materials and passing through physical obstacles. The print quality of none of the other works (including us and other research groups) based on DSP are impressive as SLA parts however, this comparison is flawed since SLA (nor any other 3D printing methods) can do what DSP/HDSP is capable of.

1. **Manufacturing process of holograms is a complex task**: The accuracy of the printed parts depends heavily on the accuracy of the manufactured holograms. The hologram manufacturing process can introduce errors and distortions, which can affect the quality and accuracy of the printed objects. It is questionable that if there is a universal hologram that can handle 3d printing of objects with different complexity and geometrical parameters.

RE23: We thank the reviewer for highlighting significance of hologram manufacturing accuracy and its potential effects on printed parts. Yes, acoustic holography is a complex field of study but we expect that the collective efforts of researchers push the boundaries of this discipline and make the acoustic holography widespread, similar to Digital Micromirror Device (DMD) that can be found in most optical projectors and many light-based 3D printers. DMDs were invented by Texas Instrument back in 80s and at that time this belief that DMDs manufacturing is a complex task was wide spread too. But nothing earned easy in the scientific and engineering fields. DMDs have gone through decades of development and now we are fortunate to access such articulated technology extremely easy and inexpensively. We expect to see the same technological development history for acoustic holography too.

We elaborated in our response RE12, where we discussed the advancement toward a programmable acoustic hologram is a pivotal development. Such holograms can actively generate the desired acoustic field for various geometries in real-time [1]. A significant breakthrough in this direction is the introduction of the Spatial Ultrasound Modulator (SUM), analogous to the Digital Micromirror Device (DMD) in optics, recently introduced in this study [2]. This technology enables dynamically reconfiguration of the acoustic field through digitally controlled patterns of microbubbles, generated by a complementary metal–oxide–semiconductor (CMOS) chip. The generated pattern of microbubbles, act as a binary hologram that shapes the incoming acoustic wavefront. Following this study, the more sophisticated and robust enhancement was introduced which shows faster response times, where the microbubble generation is enabled by directing light patterns from a Digital Light Processing (DLP) onto a photoconductive substrate [3]. These studies introduce dynamic acoustic holograms that show a feasibility of dynamic acoustic holography for HDSP, enabling a full 3D printed object, which will significantly expand the applicability of HDSP in creating intricate objects.

Employing dynamic acoustic holography, opens up a new avenue for enhancing HDSP capabilities from Sound-based 3D printing, comparable with the DLP from photopolymerization process. This approach significantly broadens the spectrum of achievable designs, indicating that while a 'universal' hologram may not yet exist today, the technology is moving rapidly towards flexible, adaptable solutions that could accommodate a vast range of object complexities.

In addition, we have mentioned this concern in the paper as well and provided an FEA simulation and exact measurement of the fabricated hologram with the theoretical 3D model that first might seems to be contingent only upon using high accuracy 3D printed hologram. The defects in the

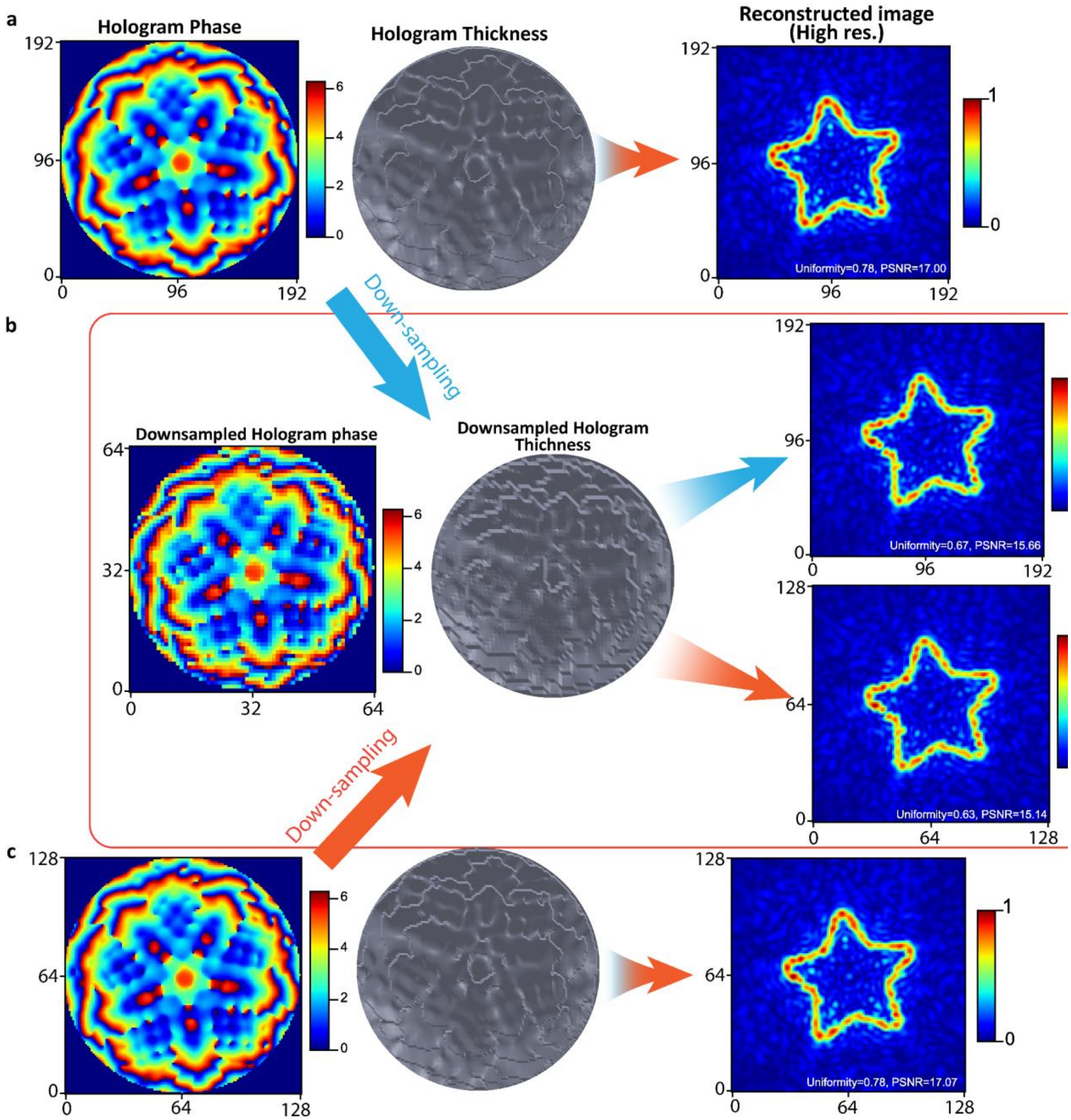


Fig. R1. Compression of high-resolution hologram into a lower resolution hologram without the reduction in accuracy and reconstruction fidelity. a and c correspond to the 192×192 and 128×128 pixels holograms with their corresponding thickness pattern and their reconstructed image. Downsampling hologram a and c with kernel size of 3 and 2, respectively, the compressed hologram in b with 64×64 pixel can be obtained.

manufacturing of the hologram, such as blobs and flaps, indeed influence the pattern formation and, therefore the object printing quality. However, the sensitivity of the employed acoustic hologram of the fabrication defects can be diminished in multiple ways. There are ways to reduce the effects of manufacturing errors in the holograms. In the following, we discuss a solution.

A straightforward method to reduce manufacturing defects involves downsampling a high-resolution hologram to a lower resolution, without inherently compromise the resolution of the reconstructed image. To illustrate this process, consider the space bandwidths for both the hologram plane and the target plane, where the image is generated. Typically, these bandwidths are similar, with the target image matching the size of the hologram, as seen in conventional optical holography. However, if we follow the same principle to get a defect-free, low-resolution hologram which ensures manufacturing fidelity, might initially suggest a reconstructed image with low resolution. Yet, by employing a high-resolution target image and creating a corresponding high-resolution hologram with a large number of pixels, then we can downsample it using average pooling. This reduces the number of elements, followed by an upsampling with nearest neighborhood method to match with the high-resolution target image size. This approach has proven to be highly effective.

Fig. R1 a and c, shows the high-resolution holograms of 192×192 and 128×128 elements respectively, along with their depicted reconstructed image, designed for a transducer with outer diameter (OD) of 25mm. These are then downsampled to a low resolution of 64×64 element hologram, as depicted in Fig R1 b, utilizing kernel sizes of 3 and 2 for average pooling, respectively. Despite the reduction in resolution of these holograms, the quality of the reconstructed images remains comparably high, as it is clear from their corresponding reconstructed images.

Quantitatively, when comparing the image quality measures such as Peak Sound to Noise Ratio (PSNR), as well as Uniformity in all cases, we can see modest drop in image quality. For instance, while the reconstructed image for the high resolution 192×192px hologram achieves a Uniformity of 0.78 and PSNR of 17.00, the image reconstructed from the downsampled hologram exhibits a Uniformity of 0.67 and a PSNR of 15.66, a decreased of only 14.1% and 7.88%, respectively.

This method effectively reduces the risk of manufacturing defects while maintaining the quality of the reconstructed image, demonstrating our ability to optimize hologram production for improved acoustic holography-based 3D printing. The methodology discussed here is the topic of our ongoing research, which aims to address how the print quality is influenced by the manufacturing quality of the acoustic holograms used in HDSP. Details of these studies will be elaborated in subsequent publications.

As suggested by Reviewer 1 and 2, we have highlighted the advancement of HDSP utilizing the active hologram in the revised manuscript in the Discussion section, that can be read as follows:

> "Furthermore, in the pursuit of advancing our HDSP technology, the integration of active/dynamic acoustic holograms presents a substantial opportunity for enhancement. Programmable acoustic holograms, capable of actively generating desired acoustic fields for various geometries in real-time, can be a substantial development [1]–[3]. Analogous to Digital Micromirror Devices (DMD) used in optics, the Spatial Ultrasound Modulator (SUM) exemplifies this innovation [2], [3]. Utilizing active acoustic holograms such as

SUM significantly enhances HDSP's capability, enabling the creation of complex, fully three-dimensional printed objects with intricate details."

2. **Uniformity of pressure patterns is challenging**: The uniformity of the pressure patterns at the target planes is crucial for the accuracy of the printed parts. If the pressure values are not uniform, it can result in non-uniform printed patterns and image distortion.

RE24: We appreciate the reviewer's helpful insight. Similar to the significance of energy source beam shaping in other additive manufacturing (AM) methods, such as Selective Laser Sintering (SLS), volumetric AM, and Digital Light Processing (DLP) and their ilk, uniformity in the generated acoustic pattern plays a crucial role in our developed HDSP process as well.

Recognizing the challenges associated with pattern uniformity and potential image distortion, we are actively refining our methodologies to refine these aspects. Currently, we are exploring a range of optimization techniques aimed specifically at enhancing the Uniformity of the reconstructed images and minimizing noise, and correct errors inherent in the Iterative Angular Spectrum Approach (IASA).

In our ongoing research, which will be detailed in subsequent publications, we have made promising advancements using both iterative techniques (such as iterative penalization) and a pioneering Deep Learning-based model tailored for HDSP pattern generation. Preliminary theoretical results indicate significant improvements in pattern uniformity, which is critical for enhancing the quality of the printed parts.

Currently, while these developments are at the forefront of our next steps in research and are in the theoretical phase, they are part of an evolutionary process that builds upon the foundational work discussed in this manuscript. As such, they are not yet ready for integration into the current experimental framework but signal the direction of our future efforts. By Focusing on these future directions, we are committed to address the inherent challenges within HDSP. This strategic approach ensures that once fully developed and validated, these advanced techniques can be applied to further refine HDSP technology effectively and efficiently.

We have modified the section discussing the future directions of HDSP, and appended the text below in the revised manuscript:

> "To address these challenges, advancing our reconstruction algorithms to better manage the uniformity of the pressure patterns is essential. Implementing sophisticated computational techniques that can dynamically adjust and optimize pressure distributions will be a crucial focus of our future research efforts. This approach will enhance the overall quality and consistency of the printed objects, reducing anomalies and uniformity in the final products."

3. **Selection of ultrasound center frequency and transducer size**: The choice of ultrasound center frequency (f0) and transducer active diameter (OD) affects the image quality and accuracy of the printed parts. However, there is a limitation on how much f0 can be increased, as higher frequencies can lead to increased acoustic attenuation and loss of energy reaching the platform.

RE25: We thank the reviewer for their helpful feedback. The critical role of the ultrasound center frequency and the transducer's diameter in determining the quality and accuracy, particularly referring to our characterization graph Fig. 6 d and e in manuscript. The reviewer correctly noted the trade-off between increasing frequency for improved resolution and the consequent risk of enhanced acoustic attenuation, which can reduce the energy reaching the printing platform. However, the sonochemical reaction is also might depend on the specific frequency as well as the rate of solidification.

With the PDMS resin currently used in our study, we found an optimal frequency range between 2-2.5 MHz that balances resolution enhancement with the sonochemical reaction's efficiency for solidification. This observation aligns with PDMS's acoustic properties and its responsiveness within this frequency range.

However, looking forward, we are exploring the synthesis of novel sound-sensitive, sound-printable resin that can functionalize to respond positively with higher frequencies. Developing such materials, allow us to potentially to expand the operation of HDSP with higher frequencies than 3MHz and finer printed objects.

Moreover, to utilize higher frequency in HDSP, we are not limited to keep the target printing plane fixed, and can dynamically be adjusted. By fine-tuning the target printing plane's distance, we can mitigate some of the challenges associated with higher frequency use, such as increased attenuation, ensuring that the reconstructed image maintains high-quality metrics.

These approaches collectively suggest a multifaceted strategy to navigate the complexities of using higher frequencies in HDSP. By integrating novel resin chemistries, adaptive printing plane adjustments, we aim to enhance both the resolution and overall quality of HDSP-produced objects, pushing the boundaries of what's achievable within the constraints of acoustic attenuation.

4. **Optimal image plane location**: The position of the image plane (Z target) relative to the source can affect the signal-to-noise ratio (SNR) and peak amplitude of the reconstructed image. Selecting the optimal distance between the object plane and the source is important for achieving better SNR and image quality.

RE26: Yes indeed. The target printing plane for relative the source does affect on the SNR, PSNR. Also, the proximity to the source will effect on the complexity of the retrieved phases, as the generated holograms targeting closer distance compared to those aimed at further distances. However, we view this not as a limitation, but as an opportunity to fine-tune our systems to achieve optimal resolution and image quality.

Through development and simulation-based investigations, we conduct parametric sweeps to identify the optimal set of printing parameters, including the workable frequency, the outer diameter (OD) of the transducer, and the target printing distance, before actual printing. This is a crucial step for tailoring the system, as demonstrated in the graph presented in Fig. 6 of our manuscript.

Regarding the selection of the target printing plane, the significant advantage of using an acoustic hologram over, High Intensity Focused Ultrasound (HIFU) transducers is that they can generate acoustic field for arbitrary plane at distance, with more versatility. By analyzing the hologram characterization graphs, as detailed in Fig. 6 in the manuscript, users can determine the optimal frequency, transducer OD, and target printing distance for their specific application. This methodology ensures that we can achieve the best possible SNR and image quality, which demonstrates the robustness and adaptability of our approach to holography-based sound 3D printing.

5.**Cavitation bubble cloud formation**: The interval of the input signal can also affect the formation of cavitation bubble clouds inside the printing part. Altering the interval of the input signal can be another parameter to control the formation of bubble clouds and improve the transparency of the printed objects.

RE27: We appreciate the reviewer's attention to the issue of bubble cloud formation, a challenge we are indeed keen to address. As discussed in the manuscript, we employ a strategy of adjusting the Duty Cycle (DC) by modulating the signal burst length and interval. This approach enables us to control not just the transparency of the printed parts but also their porosity. In our initial study on Direct Sound Printing (DSP), we demonstrated the ability to print structures ranging from porous to fully transparent. Unlike DSP, where bubble cloud formation is uniformly distributed throughout the part, in the current HDSP methodology, bubble clouds predominantly form in regions of high negative pressure, resulting in a more localized distribution.

By optimizing our holograms and employing the proposed methods discussed in response RE24, we aim to achieve more uniform bubble cloud formation across the printed parts. Additionally, it is important to note that using the formation of bubble cloud is not a necessarily unfavorable and detrimental; rather it presents a unique opportunity for creating porous structures intentionally. Such porosity has significant potential in biomedical applications, including drug delivery systems. For instance, by infusing the resin with therapeutic agents, we can encapsulate drugs within the bubble clouds during printing, offering a novel method for fabricating drug-loaded implants or scaffolds. With this approach, underlines our dual focus on improving the clarity of printed parts while also exploring the beneficial aspects of bubble cloud formation for the development of functional porous materials.

Still, the paper is very well written and the conducted research follows a comprehendible logic that addresses many issues that are facing hologram acoustic printing which is not necessary a weakness, and can be published in more specialized journals focused on additive manufacturing and 3D printing techniques.

RE28: We really appreciate the time and insightful comments of the reviewer. We respectfully disagree with the reviewer as elaborated in RE22 to the first comment of the reviewer. Our work in the present paper has a multidisciplinary nature beneficial for researchers in many disciplines such as physics, acoustics, chemistry, manufacturing, bioprinting, tissue engineering and sonochemistry. Therefore, specialized journals would not provide the proper platform for diverse audience that this work deserves to have. Nature Communications has a wide reach with multidisciplinary audience; therefore, we believe this would be a proper venue for our current work.

Reviewer #3 (Remarks to the Author):

This manuscript presents a novel idea of a holography-based DSP process. The information for printing is stored in cross sections as acoustic holograms. The cavitation caused by sonication polymerizes the printing material at the location instantly. This is an excellent technique for 3D printing of complex structures. One of the advantages of the described technique is its ability for printing at multiple locations simultaneously. Using holographic approach enables storing information content of multiple images in a single hologram. This opens up enormous possibilities for printing multiple objects at different locations.

These authors originated the original concept. The holographic idea using phased array of multiple nozzles is breakthrough innovation.

Overall, this is an excellent manuscript. The concept is very novel and has the potential to revolutionize the area.

The manuscript is well written. The images provided are of very high quality. The authors have done an excellent job in making the material interesting to general readers.

RE29: We are deeply grateful for the reviewer's appreciation for our hard research work. These supportive words have energized our team, and we are thrilled to see such recognition of our efforts. The acknowledgment of the novelty and potential impact of our work is immensely encouraging. We sincerely thank the reviewer for their kind support and for recognizing the contributions our research aims to make.

I have couple of questions and comments.
1) What is the defect density in the printed material?

RE30: Thank you for your inquiry. We acknowledge the importance of understanding defect characteristics to evaluate the quality of the printed parts.

In our study, we have not quantified defect (porosity) density in traditional terms such as defects per unit volume directly. However, we have conducted comprehensive assessments of the structural integrity and uniformity of the printed objects. These assessments include visual inspections, mechanical testing, and microscopic examinations with Confocal Microscopy to identify any irregularities such as voids, unreacted or uneven areas. As an example, Extended Data Fig. 3 in the manuscript shows the measurement of formed porosity width with relation to the input power and the *DC*, the two important input parameters in HDSP. Moreover, we have added a new analysis as can be seen in Fig. R1 to have better understanding on the porosity with the interaction strength parameter σ (W) which is the multiplication of power and *DC*. It shows although the printing time decrease with increasing the σ, however, the width of the porosity region increases. We have appended this figure to the Extended Data Fig. 4b in the revised manuscript.

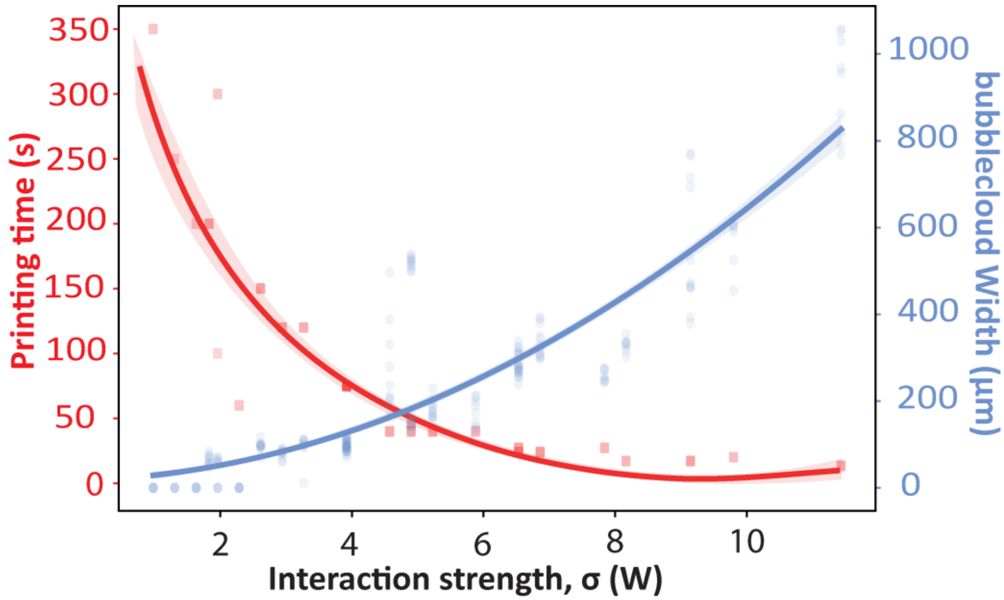


Fig. R1. Relation between the Interaction strength, porous region width and printing time.

Our observations indicate that inaccuracies in our printing setup are primarily caused by several factors including the setup misalignment and efficiency, temperature of the medium and resin, the stability of the acoustic field, and the precision of the holographic patterning. For instance, areas where the generated patterned pressure is inconsistent or where the platform is not completely aligned horizontally, parallel to the holographic field, can lead to increased incidences of weak spots in the printed structure.

Furthermore, we plan to implement more rigorous quantitative methods in future studies to accurately measure defect and pore density and correlate these findings with the variations in printing parameters and material properties.

2) Does cavitation process result in any reduction in the material (polymer) properties?

RE31: Thank you for your comment. Following the DSP, we have investigated the properties of the parts, post printing, from material composition as well as the mechanical properties. The FTIR analysis of the printed part, whether transparent or porous, shows no deviation in the material composition from that of resin typically cured in an oven. Therefore, the cavitation doesn't alter the composition of the material.

However, mechanical tensile testing revealed difference in mechanical properties. Following the DSP, the printed parts revealed consistent elastic modulus compared with the normally cured resin, although the tensile strength is different. Specifically, parts with a porous structure exhibit reduced tensile strength compared to the transparent parts. This indicates that the cavitation does not affect the material's elastic modulus, it does influence tensile strength, particularly in the printed structures with porosity.

We addressed this comment in the section of "HDSP process characterization" as:

> "Mechanical tensile tests revealed difference in mechanical properties[1]. Following the DSP, the printed parts revealed consistent elastic modulus compared with the normally

cured resin, although the tensile strength is different. Specifically, parts with a porous structure exhibit reduced tensile strength compared to the transparent parts. This indicates that the cavitation does not affect the material's elastic modulus, it does influence tensile strength, particularly in the printed structures with porosity."

3) What is the spatial separation that can be achieved when printing multiple objects using hologram approach in the printing space?

RE32: We thank the reviewer for highlighting the spatial separation between the multiple objects in printing space. The achievable separation depends several factors, including the spatial bandwidth and complexity of the target images, as well as critical input parameters such as the transducer's OD, the frequency used, and the number of elements in the hologram. These factors collectively determine the image quality and the distinctness of each reconstructed image within a multi-plane printing setup.

As an example, utilizing a transducer with an OD of 25mm and operating at a frequency of 2.24 MHz, we can achieve spatial separations ranging from approximately 2 to 6mm, depending on the

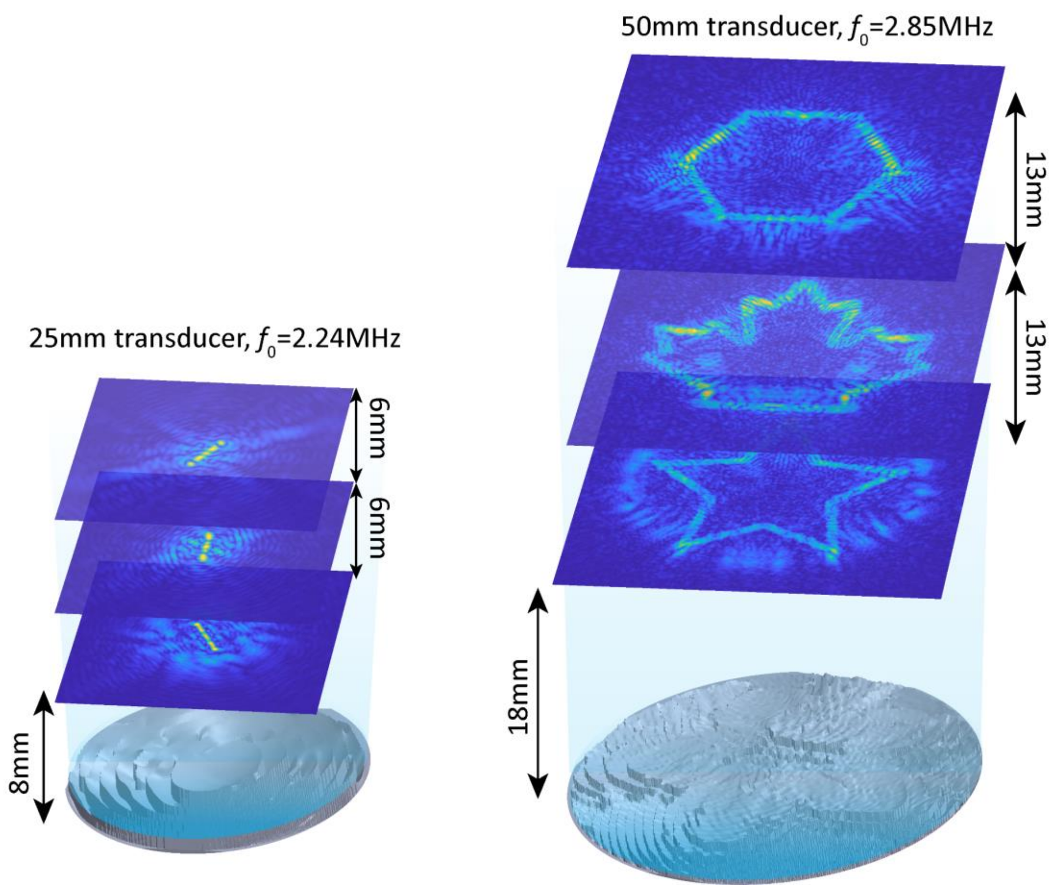


Fig. R2. Reconstruction for Multi-plane object printing capability with HDSP. Input parameter of transducer OD25mm and 2.24MHz frequency with 191×191 element hologram generating various line images (Left) and transducer OD50mm and 2.85MHz frequency with 386×386 element hologram (Right) complexity of the images and the desired clarity of the reconstructed images. In our demonstrations within the manuscript, multi-plane printing of various point images placed at different locations on the plane required a minimum separation of about 2mm. Conversely, for line geometries positioned at various angles, as depicted on the left side of Fig. R2, achieving high-quality reconstructions required separations of roughly 6mm.

Furthermore, if we increase the spatial bandwidth of the hologram as well as the OD of the transducer source, and utilizing higher frequencies, allows for reconstruction of more complex images with greater fidelity. As shown on the right side of Fig. R2, using a transducer with an OD of 50mm and a frequency of 2.85MHz, facilitated the high-fidelity reconstruction of more intricate images such as a star, maple leaf, and hexagon, separated by gaps of approximately 13mm. Using lower separation distances between the image planes, reduces the fidelity and compromise the quality of the images, make it less favorable for HDSP.

To optimize the practicality of the multi-plane object printing capability of HDSP, we strongly recommend prior simulations with varied parameters. This approach enables the determination of optimal settings to achieve the highest image quality and the most accurate final printed parts.

We have included Fig. R2 as the Extended Data Fig. 6 as well as the following explanations into the revised manuscript:

> "The effectiveness of HDSP in generating multiple objects with distinguishable and high-fidelity results depends on maintaining specific separation distances. This separation correlates directly with the spatial bandwidth of the acoustic hologram and the diameter of the transducer. Smaller transducer diameters and limited hologram bandwidth allow for the creation of simpler geometries, such as various lines and points. Conversely, utilizing larger acoustic field areas enables the incorporation of more complex shapes. Furthermore, the reconstruction of more complex images using HDSP requires increased separation distances between objects to ensure high-fidelity outcomes. The details of the reconstructions with various transducer diameters are depicted in Extended Data Fig. 6."

4) What is the effect of viscosity in the printing process? Does the polymer solution need to be homogeneous?

RE33: Thank you for your feedback. The viscosity of the printing material is crucial, especially in the sound-based printing process. High-viscosity materials are beneficial because they help control the streaming caused by acoustic waves pushing through the resin. However, materials with high viscosity, such as silicone-based Sylgard 186, often exhibit high attenuation, which can significantly hinder proper wave propagation within the resin. Thus, an optimal resin for sound-based 3D printing would possess high viscosity to mitigate streaming while also maintaining low attenuation to facilitate effective wave transmission.

Regarding the homogeneity of the resin, it is indeed crucial for ensuring consistent acoustic properties throughout the material, such as speed of sound and attenuation. Inhomogeneities in the resin can lead to disturbances in the desired acoustic patterning, adversely affecting the print quality. We could also print composite polymers (polymer mixed micro/nano solid particles) as shown in [1] however the mixture should be homogenous too to avoid uneven scattering from the added particles.

To address these challenges, our future work will focus on synthesizing specialized resins tailored for sound-based 3D printing. These materials will aim to balance viscosity and attenuation properties optimally, enhancing the efficacy and reliability of the printing process.

We addressed this comment in the section of HDSP Process Characterization as

> "It is indeed crucial to ensure consistent acoustic properties throughout the material, such as speed of sound and attenuation. Inhomogeneities in the resin can lead to disturbances in the desired acoustic patterning, adversely affecting the print quality. We could also print composite polymers (polymer mixed micro/nano solid particles)[1] however the mixture should be homogenous too to avoid uneven scattering from the added particles."

5) What about the presence of air bubbles in the polymer?

RE34: Thank you for raising this important issue. The presence of air bubbles inside the resin should always be prevented prior to the printing, as bubbles can significantly disrupt the propagation of sound waves due to the marked difference in sound speed between air and resin. To mitigate this, we typically degas the resin in a vacuum chamber to remove large air bubbles and achieve a homogeneous material composition. Additionally, careful handling of the resin after degassing is essential to avoid reintroducing air bubbles into the resin. However, it's important to note that a small percentage of microbubbles may still remain dissolved in the resin after the degassing process. These microbubbles are generally smaller than the wavelength of the sound waves used and do not significantly interfere with wave transmission. Instead, they can play a beneficial role in the cavitation process as the weak points in the medium to initiate bubble generation, contributing positively to the overall printing mechanism.

We addressed this comment in the HDSP Process Characterization as:

"The presence of air bubbles inside the resin should always be prevented prior to the printing, as bubbles can significantly disrupt the propagation of sound waves due to the marked difference in sound speed between air and resin. To mitigate this, we typically degas the resin in a vacuum chamber to remove large air bubbles and achieve a homogeneous material composition. Additionally, careful handling of the resin after degassing is essential to avoid reintroducing air bubbles into the resin. However, it's important to note that a small percentage of microbubbles may still remain dissolved in the resin after the degassing process. These microbubbles are generally smaller than the wavelength of the sound waves used and do not significantly interfere with wave transmission. Instead, they can play a beneficial role in the cavitation process as the weak points in the medium to initiate bubble generation, contributing positively to the overall printing mechanism."

6) It would be helpful for general readers if the authors could add some physics of cavitation process.

RE35: Thank you for your suggestion. We had provided correlation between the bubble dynamics and the pores' sizes in the printed objects in our initial work on DSP in the supplementary data [23]. We also elaborated on the physics of the cavitation and added in the revised manuscript, to highlight the fundamental cavitation process happening in DSP and HDSP. The revised text is highlighted in the manuscript and can be find as below:

"However, DSP employs sonochemistry, which utilizes the dynamic behavior of cavitation bubbles within an acoustic field. During the sonochemical process, these cavitation bubbles undergo rapid oscillations, expanding during periods of low pressure and collapsing violently under high pressure. The collapse of these bubbles generates localized hot spots where temperatures and pressures can reach extreme levels[8-10] [24]–[26][23]–[25] sufficient momentarily to break and form chemical bonds or also create ones. This intense environment within the bubbles enables various chemical reactions, crucial for the DSP process, allowing for precise manipulation of the material's polymerization at microscopic scales"

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REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have sufficiently addressed the reviewer comments. The revised work is improved in terms of its clarity and comprehensiveness, with a substantially improved discussion.

Reviewer #2 (Remarks to the Author):

The authors have clarified many issues raised by reviewers including myself. I have no further comments and I am convinced that the revised version of this manuscript is suitable for publication in Nature Communications.

Reviewer #3 (Remarks to the Author):

The authors have addressed all my concerns, comments, and questions. I recommend accepting the revised version of the manuscript.

Response to the Reviewers

Reviewer #1:

The authors have sufficiently addressed the reviewer comments. The revised work is improved in terms of its clarity and comprehensiveness, with a substantially improved discussion.

RE #1: We sincerely appreciate the reviewer's insightful comments during the review process, which significantly enriched our work. We are glad that the reviewer found the changes to the manuscript as well as the response to the concerns in the review process satisfactory.

Reviewer #2:

The authors have clarified many issues raised by reviewers including myself. I have no further comments and I am convinced that the revised version of this manuscript is suitable for publication in Nature Communications.

RE #2: We are pleased that the reviewer found our revised manuscript suitable for publication in Nature Communications. We appreciate their thoughtful feedback during the review process and for recognizing the value of our work.

Reviewer #3:

The authors have addressed all my concerns, comments, and questions. I recommend accepting the revised version of the manuscript.

RE #3: We are delighted that the reviewer recommended our manuscript for publication. We are grateful for the reviewer's invaluable comments in strengthening our manuscript.