nature portfolio

Peer Review File

Single-exposure holographic lithography of ultra-high-aspect-ratio microstructures

Corresponding Author: Professor Rajesh Menon

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

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

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The manuscript has several flaws that need to be fixed. As a result, the submitted manuscript cannot be accepted for publication in current format; however, it may be accepted following a significant change. Here are my comments and recommendations:

1. The abstract provides details on the primary aspect of the conducted study; however, a few phrases regarding the study's findings should be included.
2. The necessity of the conducted research must be explained by the authors. The study's objectives must be stated explicitly in the last part of the introduction.
3- The literature review needs to be improved. The following recent, pertinent studies must be studied, referred, and consulted by authors in this regard: (a) DOI: 10.1016/j.euromechsol.2021.104304 and (b) DOI:10.1016/j.jmrt.2024.11.052 and other related research works.
4- The introduction needs to be focused and must be effective. Discussion needs literature comparisons. Introduction should be clear and target the material in question. In this context, authors can search on Sciencedirect.com using “fracture in 3D printed parts” as key terms to find, read, refer, and use relevant published scientific papers.
5- Authors must explain why this approach is considered in this study. Limitations, challenges, and difficulties of conducting this research must be explained.
6- It is mentioned that “The structure exhibits a well-defined linear elastic regime at small strains”. It must be explained more and enriched with the relevant figures.
7- Standard deviation is the presented curves that must be discussed. In addition, errors in calculation must be considered and discussed. All curves must be illustrated in a more scientific way.
8- There are sentences which must be rewritten.
9- The conclusion needs to be more than a synopsis of the work. The list of references needs to be revised considering the suggested papers. In the updated version, please indicate all changes in red.

Reviewer #2

(Remarks to the Author)
The manuscript reports a single-exposure volumetric photolithography approach based on inverse-designed phase masks to fabricate ultra-high-aspect-ratio microstructures in SU-8. The authors demonstrate lattices with ~6 µm feature size and aspect ratios exceeding 120:1, fabricated within ~20 s exposure. The approach reconstructs a volumetric intensity distribution using an optimized phase mask to maintain optical contrast over large propagation distances.
The concept of using inverse-designed holographic phase masks for extended-depth lithography is interesting and potentially valuable for high-throughput fabrication of 3D microstructures. However, several important aspects regarding generality, resolution limits, material compatibility remain insufficiently addressed.
Therefore, I recommend major revision before the manuscript can be considered for publication.
1 SU-8 photoresist is used in this study. Can other liquid photoresists, such as acrylate-based UV-sensitive photoresists,

also be printed using this method? If the refractive indices of different photoresists vary, does the photomask need to be redesigned to match the refractive index of a specific photoresist? Since the optical field reconstruction depends strongly on propagation in the resist medium, this issue is critical for evaluating the general applicability of the method.
2 Other high-aspect-ratio structures, such as micropillars (e.g., Kamranikia, Keynaz, et al., Very high-aspect-ratio polymeric micropillars made by two-photon polymerization, Micromachines, 14(8), 1602, 2023), should be printed to demonstrate the universality of the proposed method.
3 What factors limit the resolution (6 µm) achieved in this work? Whether submicron resolution could be achieved using shorter lithography wavelengths, higher-NA optics, or smaller phase-mask pixels?
4 In Fig. 3h, some rectangular shapes appear distorted. Does this distortion arise from the optical system or from deformation during resin development?
5 The capillary experiment indicates that residual resist remains near the base of the structures, partially blocking the channels. The authors should discuss whether this issue becomes more severe for deeper or smaller features.

Reviewer #3

(Remarks to the Author)

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The comments are addressed and the manuscript has been improved. I think this manuscript can be considered for publication.

Reviewer #2

(Remarks to the Author)
The authors have addressed my concerns clearly, and I now recommend the manuscript for publication.

Reviewer #3

(Remarks to the Author)
I am happy with the revision that has addressed my previous concerns.

RESPONSES TO: REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The manuscript has several flaws that need to be fixed. As a result, the submitted manuscript cannot be accepted for publication in current format; however, it may be accepted following a significant change. Here are my comments and recommendations:

1. The abstract provides details on the primary aspect of the conducted study; however, a few phrases regarding the study's findings should be included.

Response R1.1:
We thank the reviewer for this suggestion. We agree that the abstract should more explicitly state the principal findings of the study. In the revised manuscript, we made minor edits to the abstract to highlight the key experimental results, including the achieved ~6 μm minimum feature size, fabrication of structures up to 800 × 800 × 720 μm^3 with aspect ratios exceeding 120:1, ~20 s exposure time, controlled capillary transport, and mechanically robust behavior revealed by micro-compression measurements. No major changes were required, but these additions improve clarity and better emphasize the main outcomes.

2. The necessity of the conducted research must be explained by the authors. The study's objectives must be stated explicitly in the last part of the introduction.

Response R1.2:
We thank the reviewer for this helpful comment. In the revised manuscript, we strengthened the Introduction to better explain the necessity of this research by emphasizing the limitations of existing 3D microfabrication approaches, including conventional photolithography, multilayer/grayscale lithography, two-photon polymerization, and current volumetric additive manufacturing methods. We now state more explicitly that there remains a significant unmet need for a fabrication method that can simultaneously provide micrometer-scale resolution, ultra-high aspect ratio, and high throughput. In addition, we revised the final paragraph of the Introduction to state the study objective explicitly. Specifically, we clarify that the objective of this work is to develop a single-exposure volumetric photolithography approach based on an inverse-designed phase mask that reconstructs a prescribed 3D intensity distribution in thick photoresist, suppresses diffraction-induced contrast loss over extended depths, and enables rapid fabrication of ultra-high-aspect-ratio microstructures with micrometer-scale features.

3- The **literature review** needs to be improved. The following recent, pertinent studies must be studied, referred, and consulted by authors in this regard: (a) DOI:

10.1016/j.euromechsol.2021.104304 and (b) DOI:10.1016/j.jmrt.2024.11.052 and other related research works.

Response R1.3:
We thank the reviewer for this valuable suggestion. We carefully reviewed the recommended papers and revised the manuscript accordingly. We now cite these studies to provide broader context on architected and mechanically functional photopolymer structures produced by additive manufacturing. In particular, we reference the work on 3D-printed multilayer perforated sound absorbers as an example of functional architected photopolymer structures fabricated by vat photopolymerization, and we cite the recent work on brittle fracture of stereolithography-fabricated notched components in the context of mechanical characterization and failure behavior of printed polymer structures. We further clarify in the manuscript that, although these studies do not address single-exposure volumetric photolithography or inverse-designed optical-field synthesis directly, they help position the present work within the broader literature on additively manufactured polymer architectures and their functional and mechanical performance.

4- The introduction needs to be focused and must be effective. Discussion needs literature comparisons. Introduction should be clear and target the material in question. In this context, authors can search on Sciencedirect.com using "fracture in 3D printed parts" as key terms to find, read, refer, and use relevant published scientific papers.

Response R1.4:
We thank the reviewer for this helpful suggestion. In the revised manuscript, we further focused the Introduction to better target the specific material and fabrication context of this study, namely thick SU-8 photoresist patterned by single-exposure volumetric photolithography using an inverse-designed phase mask. We also expanded the Discussion to include clearer comparison with relevant literature on the mechanical and fracture behavior of 3D-printed photopolymer parts, particularly stereolithography-fabricated polymer structures. These additions improve the focus of the Introduction and help position the present work relative to the broader mechanics literature on additively manufactured polymer components, while also clarifying the distinctions between our single-exposure volumetric process and conventional layer-wise additive-manufacturing approaches.

5- Authors must explain why this approach is considered in this study. Limitations, challenges, and difficulties of conducting this research must be explained.

Response R1.5:
We thank the reviewer for this comment. In the revised manuscript, we now explain more explicitly why this approach was considered. Specifically, we clarify that the single-exposure volumetric photolithography strategy was chosen to address a key unmet need in 3D microfabrication:

achieving micrometer-scale resolution, ultra-high aspect ratio, and high throughput simultaneously. We contrast this motivation with the limitations of conventional photolithography, serial point-writing methods such as two-photon polymerization, and current volumetric additive manufacturing approaches. We have also expanded the manuscript to discuss the main limitations and challenges of the present method. These include the fundamental trade-offs among structure depth, lateral feature size, and optical contrast; the increasing difficulty of printing deeper or denser patterns; and practical fabrication challenges such as alignment, incomplete development of deep features, adhesion-related issues, and the limited printability of unsupported standalone geometries. We further note possible mitigation strategies, including improved substrate preparation, adhesion-promoting layers, and optimized development protocols. These additions clarify both the motivation for the method and the present constraints on its implementation.

6- It is mentioned that "The structure exhibits a well-defined linear elastic regime at small strains". It must be explained more and enriched with the relevant figures.

Response R1.6:
We thank the reviewer for this helpful comment. In the revised manuscript, we expanded the explanation of the linear elastic regime to clarify that the initial approximately linear portion of the stress-strain curve at small compressive strain corresponds to predominantly elastic, reversible bending of the lattice walls before the onset of localized buckling and collapse. We also revised the main-text discussion to refer more explicitly to Figs. 6(e, f), where the in situ SEM images and stress-strain curve together illustrate the initial linear response and the subsequent transition to nonlinear deformation. Additional detail is provided in the Supplementary Information (Note S15).

7- Standard deviation is the presented curves that must be discussed. In addition, errors in calculation must be considered and discussed. All curves must be illustrated in a more scientific way.

Response R1.7:
We thank the reviewer for this important comment. In the revised manuscript, we improved the presentation of the plotted curves to make them more scientifically rigorous and easier to interpret. We revised the figure captions and associated discussion to clarify whether each curve represents averaged data or a representative individual measurement. Where fitted parameters are reported, we now describe the fitting procedure more explicitly and discuss the corresponding sources of uncertainty and calculation error. We also improved the graphical presentation of the curves through clearer axis labels, units, and fit annotations. In addition, the relevant manuscript text and supplementary plots, including Fig. 6(a) measured penetration of water, and Fig. S10(a) cross-sectional intensity of the designed hexagonal-cell lattice, were updated accordingly.

8- There are sentences which must be rewritten.

Response R1.8:

We thank the reviewer for this comment. In response, we carefully revised the manuscript throughout to improve sentence structure, clarity, grammar, and overall readability. A number of sentences were rewritten for greater precision and smoother flow, while preserving the original scientific content.

9- The conclusion needs to be more than a synopsis of the work. The list of references needs to be revised considering the suggested papers. In the updated version, please indicate all changes in red.

Response R1.9:

We thank the reviewer for this helpful suggestion. In the revised manuscript, we expanded the Conclusion so that it goes beyond a synopsis of the work. It now more clearly emphasizes the broader significance of the method, the key advances demonstrated here, the present limitations of the approach, and several promising future directions. We also revised the reference list to incorporate the suggested papers and to improve the completeness of the literature discussion. Finally, all changes in the revised manuscript have been highlighted, as requested.

Reviewer #2 (Remarks to the Author):

The manuscript reports a single-exposure volumetric photolithography approach based on inverse-designed phase masks to fabricate ultra-high-aspect-ratio microstructures in SU-8. The authors demonstrate lattices with ~6 µm feature size and aspect ratios exceeding 120:1, fabricated within ~20 s exposure. The approach reconstructs a volumetric intensity distribution using an optimized phase mask to maintain optical contrast over large propagation distances. The concept of using inverse-designed holographic phase masks for extended-depth lithography is interesting and potentially valuable for high-throughput fabrication of 3D microstructures. However, several important aspects regarding generality, resolution limits, material compatibility remain insufficiently addressed.
Therefore, I recommend major revision before the manuscript can be considered for publication.

1 SU-8 photoresist is used in this study. Can other liquid photoresists, such as acrylate-based UV-sensitive photoresists, also be printed using this method? If the refractive indices of different photoresists vary, does the photomask need to be redesigned to match the refractive index of a specific photoresist? Since the optical field reconstruction depends strongly on propagation in the resist medium, this issue is critical for evaluating the general applicability of the method.

Response R2.1:
We thank the reviewer for this important comment. In principle, this method is compatible with other photosensitive materials, including acrylate-based UV-sensitive resins, provided that their optical and photochemical properties are incorporated into the design and exposure process. Our prior single-exposure 3D fabrication work used an acrylate-based UV-sensitive resin formulation, supporting the broader applicability of the method beyond SU-8. In the present study, we selected SU-8 because it offers more favorable thresholding behavior, reduced oxygen sensitivity, better structural stability during processing, higher-resolution capability, and compatibility with MEMS-oriented fabrication workflows. The reviewer is also correct that the reconstructed optical field depends strongly on propagation in the resist medium. As a result, if the refractive index of the photoresist changes, the phase mask should generally be redesigned or re-optimized for that material. In our system, a target extent of 450 µm in air corresponds to approximately 720 µm in SU-8 with refractive index 1.613 at 405 nm. More generally, both the refractive index and the absorption/extinction coefficient of the resist should be included in the forward model when transferring the method to a different material system. We have clarified these points in the revised manuscript.

2 Other high-aspect-ratio structures, such as micropillars (e.g., Kamranikia, Keynaz, et al., Very high-aspect-ratio polymeric micropillars made by two-photon polymerization, Micromachines, 14(8), 1602, 2023), should be printed to demonstrate the universality of the proposed method.

Response R2.2:

We thank the reviewer for this helpful suggestion and for pointing us to the work by Kamranikia *et al.* To further assess the generality of the proposed method, we performed additional experiments on standalone cylindrical micropillar structures with varying aspect ratios, and these results have now been added to the Supplementary Information (Note S17). We found that isolated high-aspect-ratio micropillars are substantially more prone to collapse during processing than the connected, self-supported lattice architectures emphasized in the main text. We believe this comparison is informative because it helps clarify the present operating regime of the method: it is well suited for self-supporting ultra-high-aspect-ratio architectures, whereas unsupported standalone pillars remain more challenging. We have revised the manuscript to clarify this point.

3 What factors limit the resolution (6 µm) achieved in this work? Whether submicron resolution could be achieved using shorter lithography wavelengths, higher-NA optics, or smaller phase-mask pixels?

Response R2.3:

We thank the reviewer for this important question. As in other optical lithography techniques, the achievable resolution is fundamentally limited by diffraction, scaling approximately as λ/NA in the transverse direction and λ/NA^2 in the axial direction, with additional constraints imposed by the photoresist response, mechanical stability, and the exposure-development process. In the present system, the achieved printing resolution is primarily limited by the effective numerical aperture of the holographic reconstruction geometry. Specifically, the 2 mm phase-mask size and approximately 20 mm reconstruction distance correspond to an effective NA of about 0.05, which is consistent with the experimentally observed ~ 6 µm wall width and the simulated ~ 4 µm lateral resolution reported in the manuscript. We agree with the reviewer that shorter lithography wavelengths, higher-NA configurations, and smaller phase-mask pixels would all improve the achievable resolution. Accordingly, submicron resolution is possible in principle, and it would require a higher-NA implementation together with finer phase-mask fabrication and careful control of resist processing. We have added a brief discussion of these resolution limits and scaling considerations in the revised Supplementary Information (Note S16).

4 In Fig. 3h, some rectangular shapes appear distorted. Does this distortion arise from the optical system or from deformation during resin development?

Response R2.4:

We thank the reviewer for this careful observation. Based on our measurements and process understanding, the distortion in Fig. 3h arises primarily from post-exposure processing and mechanical deformation of the printed structure, rather than from a major limitation of the reconstructed optical field itself. The experimentally measured 3D intensity distribution agrees well with the design, indicating that the optical reconstruction is not the dominant source of the observed distortion. We attribute the deformation mainly to SU-8 reflow during post-exposure bake and to bending or collapse of thin unsupported walls during development. We have clarified this point in the revised manuscript and noted that improved resist formulations or mechanically stronger materials could help mitigate this effect.

5 The capillary experiment indicates that residual resist remains near the base of the structures, partially blocking the channels. The authors should discuss whether this issue becomes more severe for deeper or smaller features.

Response R2.5:

We thank the reviewer for this important comment. Yes, in general this issue is expected to become more severe for deeper structures and for smaller or denser features, because developer access becomes increasingly restricted and the removal of unpolymerized resist becomes more difficult over long, narrow channels. In the present work, the residual resist observed near the base of the capillary structures indicates incomplete development under these challenging high-aspect-ratio conditions. A practical limitation in our experiments was that the development time could not simply be extended indefinitely, because longer development led to partial delamination of the SU-8 structures from the soda-lime glass substrate. We therefore attribute the incomplete clearing near the base to a combination of hindered developer transport in deep features and limited substrate adhesion during prolonged processing. We have clarified this point in the revised manuscript and now note possible mitigation strategies, including improved substrate preparation, use of adhesion-promoting interlayers such as OmniCoat, alternative substrates, and more aggressive development protocols such as ultrasonic agitation.

Reviewer #3 (Remarks to the Author):

This manuscript reports an interesting direct-laser fabrication technique, single-exposure holographic lithography of 3D high-aspect-ratio microstructures. This approach uses a binary mask together with a holographic phase pattern to achieve a diffraction-free-like extended depth-of-field intensity distribution over a long propagation distance. The authors demonstrate lateral features of 6 µm and a structure height of 720 µm, achieving aspect ratios greater than 120. The fabricated microstructures also exhibit smooth sidewalls and good mechanical performance, including an effective Young's modulus of 5.7 GPa. Overall, this is a strong application of holographic phase masks for extended depth-of-focus exposure with non-diffraction-like characteristics. The manuscript is well written with comprehensive technical details and experimental data. I recommend publication after the authors address the following concerns:

1. The state-of-the-art discussion of 3D direct laser writing is incomplete. In the first sentence, the micro-optical systems section should cite at least:
https://www.science.org/doi/10.1126/science.1177031
https://www.nature.com/articles/ncomms11763
https://www.nature.com/articles/nphoton.2016.121.
The metasurfaces section should cite:
https://www.nature.com/articles/s41377-021-00491-z
https://www.nature.com/articles/s41565-020-0768-4
https://www.nature.com/articles/s41467-022-31902-3
https://www.science.org/doi/full/10.1126/sciadv.add4816
https://www.nature.com/articles/s41467-023-43068-7.

Response R3.1:

We thank the reviewer for this helpful suggestion. Following the reviewer's recommendation, we revised the Introduction to better position our work within the state of the art in 3D direct laser writing. In particular, we added the suggested references in the discussion of micro-optical systems and metasurface-related structures, and we revised the surrounding text to integrate these works more clearly into the narrative. The added references highlight prior advances in free-form micro-optics, ultracompact multi-lens objectives, and metasurface/meta-fiber platforms realized by 3D direct laser writing and related nanoprinting approaches. These additions improve the completeness of the Introduction and better situate the present work relative to prior developments in the field.

2. The sentence "Analytical structured-light approaches have demonstrated single-shot fabrication of simple 3D geometries, including helices and tubular structures, but these methods are generally restricted to certain geometries and thus far have not demonstrated extremely large aspect ratios" requires a supporting reference.

Response R3.2:
We thank the reviewer for pointing this out. In the revised manuscript, we added supporting references for analytical structured-light approaches that enable single-shot or scan-free fabrication of simple 3D geometries such as helices and tubular structures. We also slightly revised the sentence to clarify that such methods are typically restricted to analytically prescribed geometries, in contrast to the more general inverse-designed volumetric patterns pursued here.

3. The authors should discuss the limitations of their fabrication approach. In particular, can this method produce fully 3D standalone structures, or must all features remain interconnected for mechanical stability during development?

Response R3.3:
We thank the reviewer for this important comment. In principle, the method is not fundamentally restricted to interconnected geometries, and fully 3D standalone structures can be designed and optically reconstructed. In the present implementation, however, unsupported ultra-high-aspect-ratio features are significantly more difficult to preserve during post-exposure processing because they are more susceptible to bending, peeling, and collapse during post-exposure bake, development, and drying. Interconnected lattice geometries are therefore more robust in the current process window because they provide mutual mechanical support. We now clarify this limitation more explicitly in the revised manuscript. To further illustrate this point, we added supplementary results on standalone cylindrical micropillar structures in Supplementary Note S17, which show that isolated high-aspect-ratio features are considerably more prone to collapse than the connected architectures emphasized in the main text.

4. The authors should comment on how the fabrication resolution could be further improved. It would be valuable to discuss whether this platform could eventually reach the resolution needed for photonic crystal or metasurface structures, enabling photonic applications beyond the demonstrated mechanical properties.

Response R3.4:
We thank the reviewer for this important comment. In the present implementation, the achieved feature size is primarily limited by the effective numerical aperture of the holographic reconstruction geometry. The 2 mm phase-mask size and approximately 20 mm reconstruction distance correspond to an effective NA of about 0.05, consistent with the observed ~ 6 μm feature size and the simulated ~ 4 μm lateral resolution. More generally, as in other optical lithography techniques, the achievable resolution is fundamentally limited by diffraction, scaling approximately as λ/NA in the transverse direction and λ/NA^2 in the axial direction, with additional constraints from phase-mask pixelation, photoresist response, and the exposure-development process. Higher resolution could therefore be pursued through shorter wavelengths, higher effective NA, reduced reconstruction distance, and finer phase-mask patterning. Submicron

resolution appears plausible in principle under such conditions. This could make the platform relevant for selected photonic crystal and metasurface structures, especially at longer operating wavelengths where the required feature sizes are larger. We have added a brief discussion of these resolution limits and future photonic opportunities in the revised manuscript and Supplementary Information (Note S16).

5. Although the physical concepts behind generating non-diffraction beams have been introduced by the group before, the manuscript should still present the designed phase patterns and clarify how they were implemented experimentally.

Response R3.5:

We thank the reviewer for this valuable comment. We agree that the present manuscript should explicitly present the designed phase patterns and clarify how they were implemented experimentally, independent of our prior related work. In the revised manuscript and Supplementary Information, we now provide a clearer and more self-contained description of the designed phase profiles, the corresponding fabricated holographic phase masks, and their experimental implementation. Specifically, the phase-mask design and fabrication details are clarified in Supplementary Note S5, the optical testing procedure is described more clearly in Supplementary Note S6, and the alignment and mask–photoresist exposure process are detailed in Supplementary Note S7. We also strengthened the main-text discussion around Fig. 2 so that the manuscript more clearly connects the designed phase profile, fabricated mask, and experimentally reconstructed intensity distributions.

This manuscript reports an interesting direct-laser fabrication technique, single-exposure holographic lithography of 3D high-aspect-ratio microstructures. This approach uses a binary mask together with a holographic phase pattern to achieve a diffraction-free-like extended depth-of-field intensity distribution over a long propagation distance. The authors demonstrate lateral features of 6 µm and a structure height of 720 µm, achieving aspect ratios greater than 120. The fabricated microstructures also exhibit smooth sidewalls and good mechanical performance, including an effective Young's modulus of 5.7 GPa. Overall, this is a strong application of holographic phase masks for extended depth-of-focus exposure with non-diffraction-like characteristics. The manuscript is well written with comprehensive technical details and experimental data. I recommend publication after the authors address the following concerns:

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   The metasurfaces section should cite:
   https://www.nature.com/articles/s41377-021-00491-z
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2. The sentence "Analytical structured-light approaches have demonstrated single-shot fabrication of simple 3D geometries, including helices and tubular structures, but these methods are generally restricted to certain geometries and thus far have not demonstrated extremely large aspect ratios" requires a supporting reference.
3. The authors should discuss the limitations of their fabrication approach. In particular, can this method produce fully 3D standalone structures, or must all features remain interconnected for mechanical stability during development?
4. The authors should comment on how the fabrication resolution could be further improved. It would be valuable to discuss whether this platform could eventually reach the resolution needed for photonic crystal or metasurface structures, enabling photonic applications beyond the demonstrated mechanical properties.
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