nature portfolio

Peer Review File

300 units per second roll-to-roll manufacturing of visible metalenses

Corresponding Author: Professor Junsuk Rho

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

Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)
This well-written manuscript describes the fabrication of 300 metalenses per second using roll-to-roll nanoimprint lithography (R2R NIL), demonstrating significant promise for high-throughput metalens manufacturing. While the work clearly merits publication in a high-quality journal, I am not convinced that its impact is sufficient to warrant publication in Nature.

Although there is limited published information on R2R NIL for metalens fabrication, several companies including NILT, Toppan Photomask (in collaboration with EVG), and Myrias Optics claim to have high-throughput metalens manufacturing capabilities. To my knowledge, however, none of these companies publicly report throughputs as high as 300 metalenses per second or demonstrate true R2R fabrication, as described here. This positions the manuscript as a valuable and exciting contribution. The authors also appropriately cite work from some of these companies. One notable omission is the following recent publication:

Full Wafer Scale Manufacturing of Directly Printed TiO_2 Metalenses at Visible Wavelengths with Outstanding Focusing Efficiencies, Advanced Materials, 2025 (DOI: 10.1002/adma.202500327). This article reports the fabrication of 1,000 efficient metalenses in under 5 minutes via NIL of a TiO_2 ink on an 8-inch wafer. Although this throughput is significantly lower than the 300 metalenses/second presented in the current manuscript, it is relevant work that should be cited.

I would also appreciate a more detailed discussion of the durability of the polymer replica mold. The authors note the use of an anti-stick self-assembled monolayer (SAM) on the mold. While SAMs can help prevent adhesion between the mold and the cured polymer, several R2R manufacturing companies I have worked with avoid using SAMs due to degradation over time, which can lead to variation in the imprinted features over the course of a run. Although 20 meters is insufficient to fully assess long-term mold durability or SAM stability, it would still be valuable to include metrology data at both the beginning and end of the 20-meter roll. Specifically, I suggest:

1. Showing SEM images of the polymer replica mold before imprinting begins and after completion of the 20-meter run.
2. Comparing the same metalens at different points along the roll. This would be similar to Figure 4d, but would track a single lens across the entire 20 meters.

Finally, the authors assert that their atomic layer deposition (ALD) process is R2R-compatible. While this may be theoretically feasible, the four-step ALD process described in the Methods section seems difficult to implement in a true R2R system at a practical throughput and cost. Can the authors provide further justification or references supporting the claim that this ALD process is realistically scalable to R2R? Additionally, is there a specific reason why ALD of TiO_2 on top of the cured resin is preferred over alternatives such as TiO_2-infused resins or printable TiO_2 inks?

Referee #2

(Remarks to the Author)

The authors describe roll-to-roll (R2R) nanoimprint of metalenses in the visible spectrum on flexible PET substrate with high throughput. The proposed process utilizes UV-curable imprint to replicate the metalens geometry from a 12 in mold patterned using 193 nm immersion lithography. A conformal layer of TiO_2 is then deposited using a subsequent atomic layer deposition (ALD) process to increase the effective index of the imprinted polymer and thus the efficiency of the printed device. This work is among the first demonstration of scalable manufacturing of hybrid metalens that would be suitable for commercialization and would find broad interests. However, I think there are a number of critical points that require further clarification. These points are outlined in the following questions/comments:
1. There has been a number of recent publications over the last few month on similar topic of industry-scale manufacturing of metalens (For example: https://doi.org/10.1002/adma.202500327, which focuses on direct printing of inorganic TiO_2 composite but also includes a backfill ALD step to increase index). Can the authors comment on the novelty of the current manuscript in comparison to what has recently been published? It would be helpful to quantitatively compare the throughput (linespeed in terms of mm/s), focusing efficiency, yield, etc..
2. The measured focusing efficiency of the metalenses are around 49.5%, which is much lower than the predicted efficiency of up to 90%. Can the authors comment on the discrepancy between model and fabrication? The low efficiency of the lens is a concern since other R2R manufactured metalenses have efficiency of over 80% (cited above).
3. Figure 4b and 4c plot the point spread functon(PSF) and MTF of the metalenses, respectively, which shows a sizable error between the measured vs model. Can the authors comment on the sources of error and why the focus is more broad than expected? Also it appears that the error in the calculated MTF is worse compared with the PSF. Given that the MTF is a normalized Fourier transform of the PSF, it is not clearly to me why the MTF looks so much worse. The scaling property of Fourier transform indicate that if the experimental MTF is half of the bandwidth in the spatial frequency domain (which appears to be the case), the PSF should be twice as wide in the spatial domain? Are the numerical aperture impose by the size of lens/illumination area the same for both cases?
4. The authors state “The full width at half maximum of the focal spots approached the diffraction limit, indicating near-ideal focusing behaviour.” It would be helpful to quantitatively state the FWHM beam width for Figure 4 and compare that to the theoretical calculated width based on the numerical aperture of the lens.
5. Can the authors comment on the defect density of the samples fabricated? This point is critical given the manufacturing focus of this paper.
6. One of the key limitations in the throughput of the R2R system is the need of ALD to increase the diffraction efficiency. One approach might be spatial ALD (https://doi.org/10.1116/1.3670745), which can greatly increase throughput and is compatible with R2R. Can the authors comment on potential use of spatial ALD and limitations on throughput?

Referee #3

(Remarks to the Author)
The manuscript reports a big step towards continuous and high-throughput manufacturing of large area meta-lens by roll-to-roll (R2R) nanoimprint (NIL) technology. It's an impressive work through the collaboration of three research institutes. This contribution is somewhat different from a typical Nature submission, because the presented work and results represent a combined expertise of multiple PI's labs, which individually have been developed and demonstrated before, e.g. meta-surface fabrication by using NIL and coating with ALD high refractive index layer to significantly improve the index contrast; R2R NIL process, and the use of polymer templates from a Master mold for cost-effective replication of the sub-wavelength structures and for managing defects by finite use of the polymer template. Since the focus is on a scalable manufacturing process, the authors would want to provide some more detailed comparison and analysis on the uniformity and repeatability of the process and the fabricated metalenses. Specifically I'd recommend adding these analysis:

1. The residual layer thickness control is important for this application. It is mentioned "We optimized the imprint pressure at 45 kgf/cm2 to minimize the residual layer thickness..." please give a range of the residual thickness obtained, and the thickness distribution (uniformity) across the samples and multiple imprints. What are impacts of the thickness variation of the residual layer on the behavior of the metalenses?

2. Report or suggest a way to quickly characterize or sample the optical performance of the fabricated metalenses, across multiple locations distributed on the imprinted 300 metalenses per unit time. Especially are there significant optical efficiency variations, and if there is, what's the main structural parameter that influences the optical performance. Supply some representative SEM showing such variations and estimate the percentage of samples of out-of-range variations.

3. What are the major defects identified in characterizing a large number of metalenses? what would you recommend/suggest to minimizing such defects?

Referee #4

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Version 1:

Reviewer comments:

Referee #1

(Remarks to the Author)
The revision is a substantially improved version of an already impressive manuscript. I appreciate the additional discussions on mold durability, optical uniformity, and the scalability of the ALD process.

However, I cannot wholeheartedly recommend publication in Nature. Rather, I recommend publication of this work in a journal such as Nature Materials, Nature Photonics, Nature Communications, or Advanced Materials. While the engineering achievements and manufacturing scalability demonstrated here are excellent and will be valuable to the field, the manuscript focuses on advancing fabrication throughput rather than introducing new scientific phenomena. As such, it does not align with Nature's typical editorial criteria for conceptual scientific novelty.

Referee #2

(Remarks to the Author)
The authors have adequately address all of my concerns and questions and I think the article is suitable for publication.

Referee #3

(Remarks to the Author)
I am satisfied with the authors responses and revised manuscript.

Referee #4

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Reviewer #1 (Comments to the Author):

Comment 1

This well-written manuscript describes the fabrication of 300 metalenses per second using roll-to-roll nanoimprint lithography (R2R NIL), demonstrating significant promise for high-throughput metalens manufacturing. While the work clearly merits publication in a high-quality journal, I am not convinced that its impact is sufficient to warrant publication in Nature.

Response 1

We sincerely appreciate the reviewer's constructive and insightful comments, which have greatly helped us improve our manuscript. We are especially grateful for the reviewer's description of our work as demonstrating "**significant promise for high throughput metalens manufacturing**" and acknowledging that it "**clearly merits publication in a high-quality journal**".

The novelty and impact of our work lie in the first practical realization of an industrially scalable platform for large-area metalens fabrication. This achievement transforms metalenses from an academic demonstration into a manufacturable optical technology, bridging the gap between fundamental nanophotonics and real-world applications. In particular, the realization of a highly continuous roll-to-roll (R2R) process for metalens fabrication marks the first demonstration of a truly scalable route toward practical, large-area optical devices.

To further address the reviewer's concern and strengthen the manuscript, we have incorporated additional clarifications and investigations. Specifically, we have revised the manuscript by (1) incorporating comparisons with key recent studies to better highlight the novelty of our work, (2) adding results of durability tests for the polymer replica mold and stability tests for the SAM coatings to demonstrate their robustness under R2R conditions, and (3) providing a discussion on replacing the conventional ALD process with a more industrially viable alternative, spatial ALD. Overall, we have further **detailed the R2R framework to make the manuscript more informative and accessible to a broad readership**. We hope the reviewer finds our revision and responses satisfactory.

Comment 1.1

Although there is limited published information on R2R NIL for metalens fabrication, several companies including NILT, Toppan Photomask (in collaboration with EVG), and Myrias Optics claim to have high-throughput metalens manufacturing capabilities. To my knowledge, however, none of these companies publicly report throughputs as high as 300 metalenses per second or demonstrate true R2R fabrication, as described here. This positions the manuscript as a valuable and exciting contribution. The authors also appropriately cite work from some of these companies. One notable omission is the following recent publication:

Full Wafer Scale Manufacturing of Directly Printed TiO_2 Metalenses at Visible Wavelengths with Outstanding Focusing Efficiencies, Advanced Materials, 2025 (DOI: 10.1002/adma.202500327). This article reports the fabrication of 1,000 efficient metalenses in under 5 minutes via NIL of a TiO_2 ink on an 8-inch wafer. Although this throughput is significantly lower than the 300 metalenses/second presented in the current manuscript, it is relevant work that should be cited.

Response 1.1

We sincerely appreciate the reviewer's thoughtful and constructive comments. We are particularly grateful that the reviewer clearly recognizes the main novelty of our work. We also thank the reviewer for recommending valuable work. The cited study indeed makes a meaningful contribution by demonstrating high optical efficiency along with excellent throughput. In response to the reviewer's suggestion, we have included this reference in the revised manuscript as follows:

On page 3,

At the other end of the spectrum, deep-ultraviolet photolithography provides wafer-scale high-fidelity metasurface patterning, but its reliance on expensive semiconductor processes makes it economically infeasible for many optical applications18-20. To address this gap, nanoimprint lithography (NIL) has emerged as a promising alternative21-29.

In References,

29 Jung, D. E. et al. Full Wafer Scale Manufacturing of Directly Printed TiO2 Metalenses at Visible Wavelengths with Outstanding Focusing Efficiencies. Advanced Materials 37, 2500327 (2025).

In recent years, the commercialization of metalenses has become increasingly important, leading to significant efforts by major companies as well as academic groups, to develop scalable manufacturing processes. A notable example of these recent advances includes the use of hybrid materials [***Nat. Mater.*** **22**, 474-481 (2023)] and nanoparticle-infused resins [***Adv. Mater.*** **37,** 2500327 (2025)]. These two studies represent the most advanced demonstrations of scalable metalens manufacturing to date: the hybrid material approach achieves printing over a 12-inch area in approximately 15 minutes, while the nanoparticle-infused resin method enables printing over an 8-inch area in about 5 minutes. By contrast, as the reviewer noted, our roll-to-roll process is much faster than these state-of-the-art techniques, enabling the printing of a **12-inch area within 1.5 seconds**. This corresponds to an area throughput more than **90 times faster** than the other works, and to emphasize this novelty, we have revised manuscript as follows.

In Discussion section,

*To our knowledge, no previous study has reported a throughput as high as 300 lenses per second (**Supplementary Note 9**).*

In Supplementary Information,

Supplementary Note 9. Comparison table of this work and related works.

	Our work	***[2]***	***[3]***	***[4]***	***[5]***	***[6]***
Material	*Resin+ALD*	*Resin+ALD*	*Resin+NPs*	*Resin*	*Resin+NPs*	*Resin+NPs*
Imprint type	*R2R*	*P2P*	*P2P*	*R2P*	*P2P*	*P2P*
Diameter (mm)	*10*	*10*	*4*	*10*	*0.45*	*0.4*
Speed (lens/min)	*18,000*	*45*	*200*	*16*	*0.2*	*0.5*

Supplementary Table 1. Comparison of previously reported nanoimprinted metalenses and this work. *Summary of representative metalens fabricated by nanoimprint lithography, comparing the material, imprint type, lens diameter, and imprinting speed with the R2R-imprinted metalenses demonstrated in this work.*

In References,

2. Kim, J. et al. Scalable manufacturing of high-index atomic layer–polymer hybrid metasurfaces for metaphotonics in the visible. Nat. Mater. ***22****, 474-481 (2023).*

3. Jung, D. E. et al. Full wafer scale manufacturing of directly printed TiO_2 metalenses at visible wavelengths with outstanding focusing efficiencies. Adv. Mater. ***37****, 2500327 (2025).*

4. Choi, M. et al. Roll-to-plate printable RGB achromatic metalens for wide-field-of-view holographic near-eye displays. Nat. Mater. ***24****, 535-543 (2025).*

5. Yoon, G. et al. Single-step manufacturing of hierarchical dielectric metalens in the visible. Nat. Commun. ***11****, 2268 (2020).*

*6. Einch, V. J. et al. Scalable nanoimprint lithography process for manufacturing visible metasurfaces composed of high aspect ratio TiO_2 meta-atoms. ACS Photonics **8**, 2400-2409 (2021).*

While throughput is an important metric, the continuity of the manufacturing process is equally critical for industrial scalability. Recent advances have transitioned from plate-to-plate (P2P) to roll-to-plate (R2P) processing, and ultimately, achieving true roll-to-roll (R2R) processing is regarded as the next milestone for metalens production. Although R2P nanoimprint method has recently been demonstrated [***Nat. Mater.*** **24**, 535-543 (2025)], as the reviewer noted**, no metalens has yet been manufactured using an R2R process**. R2R processing has been reported for film types of metamaterials; for example, [***Science*** **355**, 1062-1066 (2017)] demonstrated continuous production of 30 cm-wide metamaterials at 5 m/min, showcasing the speed and automation potential of R2R processing. However, the prior demonstrations are limited to film-type structures composed of randomly dispersed nanoparticles, which lack the precise pattern control required for metalens. In contrast, our work demonstrates, for the first time, true patterned R2R processing for visible metalenses, and achieves throughput exceeding that of film-type R2R approaches. This result represents a **breakthrough in metasurface manufacturing, proving that R2R processing can meet the strict precision and scalability demands for industrial deployment of large-area, high-performance metalenses**.

Comment 1.2

I would also appreciate a more detailed discussion of the durability of the polymer replica mold. The authors note the use of an anti-stick self-assembled monolayer (SAM) on the mold. While SAMs can help prevent adhesion between the mold and the cured polymer, several R2R manufacturing companies I have worked with avoid using SAMs due to degradation over time, which can lead to variation in the imprinted features over the course of a run. Although 20 meters is insufficient to fully assess long-term mold durability or SAM stability, it would still be valuable to include metrology data at both the beginning and end of the 20-meter roll. Specifically, I suggest: 1. Showing SEM images of the polymer replica mold before imprinting begins and after completion of the 20-meter run. 2. Comparing the same metalens at different points along the roll. This would be similar to Figure 4d, but would track a single lens across the entire 20 meters.

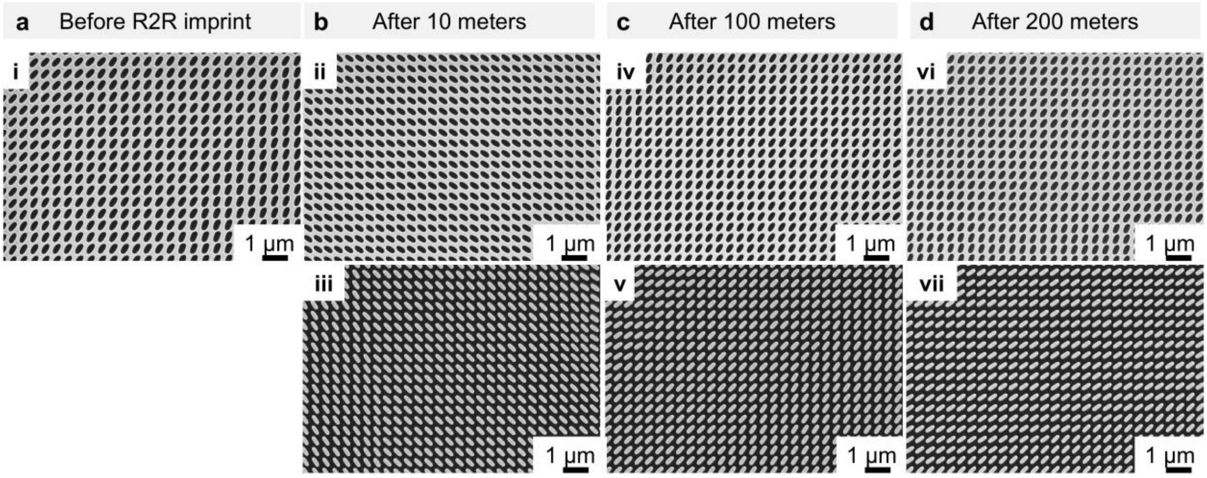
Response 1.2

We sincerely appreciate the constructive comment regarding the durability of the replica mould. The durability of the SAM coating is critical for consistent fabrication of metalenses. To evaluate the durability of SAM coating method, we have continuously imprinted metalenses over 200 m length using the same replica mould. As a result, we captured scanning electron microscopy (SEM) images of both the replica mould and the imprinted metalenses before imprinting, after 10, 100, and 200 meters of R2R imprinting. The results demonstrate that, even after 200 meters of continuous imprinting, the replica mould maintains its fidelity, and the imprinted metalens features remain well-preserved without noticeable structural changes. The stable coating property was further verified by atomic force microscopy (AFM) height profiles, showing that the heights of both the replica mould and the imprinted metalenses were well maintained, as shown below:

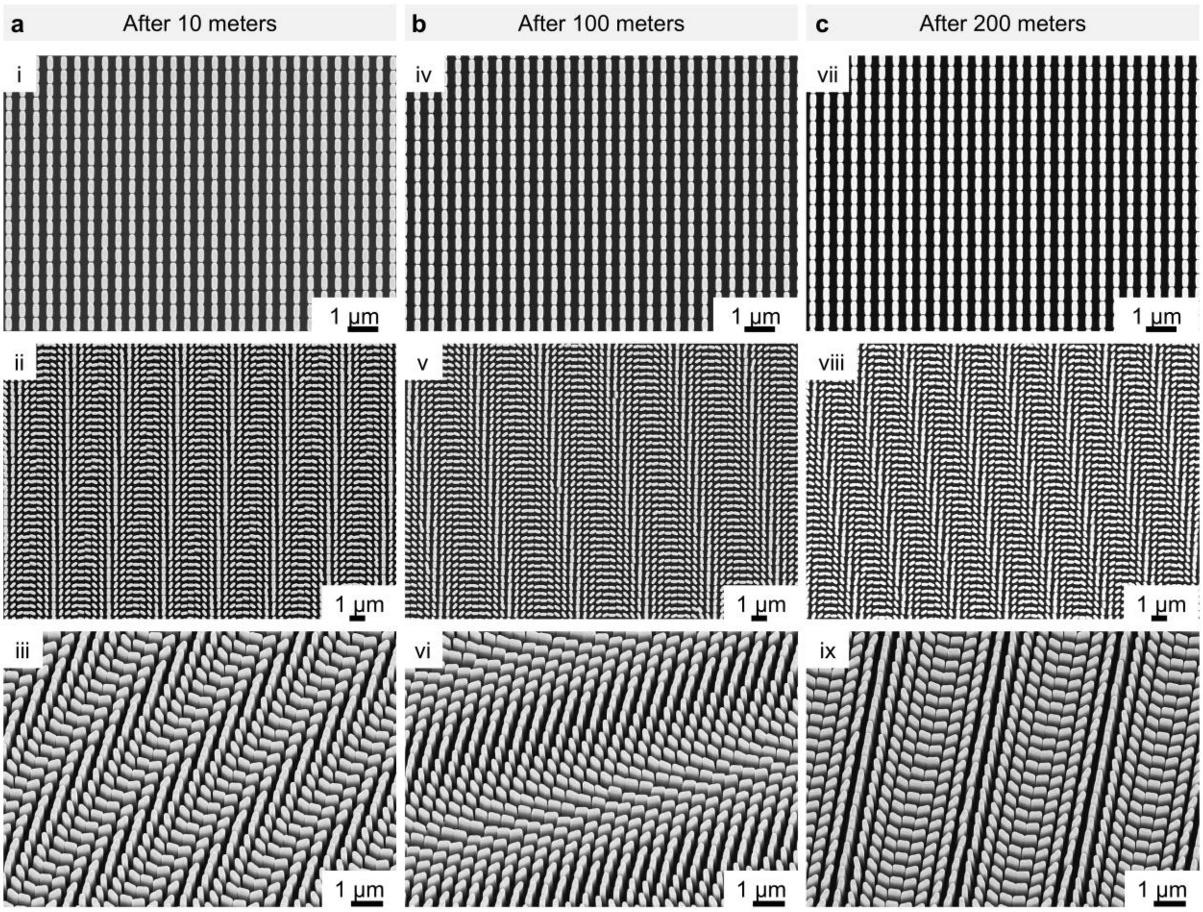
On Page 6,

*Subsequently, the replica mould is treated with a self-assembled monolayer (SAM) anti-stick coating to enhance release performance. To assess its durability during continuous imprinting, SEM images and atomic force microscopy (AFM) height profiles of the replica mould and imprinted structures have been taken before imprinting and after 200 meters of use (**Extended Data Fig. 2, 3 and Supplementary Note 5**).*

In Extended Data Figure,



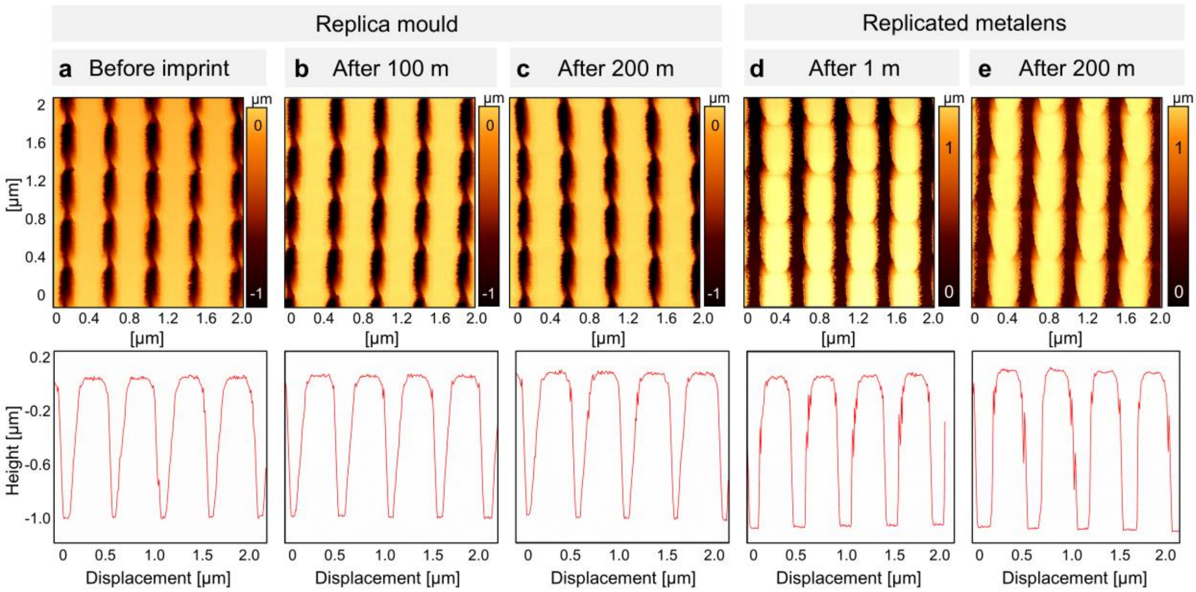
***Extended Data Figure 2. Durability test of the polymer replica mould. (a.i–d.vi)** SEM images of the replica mould: **(a.i)** before R2R imprinting, **(b.ii)** after 10 m, **(c.iv)** after 100 m, and **(d.vi)** after 200 m of R2R imprinting. **(b.iii–d.vii)** Corresponding SEM images of the imprinted resin structures: **(b.iii)** after 10 m, **(c.v)** after 100 m, and **(d.vii)** after 200 m of R2R imprinting.*



Extended Data Figure 3. R2R imprinted metalens structures at different positions along 200 m-long PET substrate. (a-e) SEM images (top and tilted) at different positions of R2R manufactured metalenses taken at (a. i-iii) 10 m, (b. iv-vi) 100 m, and (c. vii-ix) 200 m.

In Supplementary Information,

Supplementary Note 5. AFM surface profiles of the replica mold and R2R manufactured metalens.



Supplementary Figure 5. Atomic Force Microscopy (AFM) height profiles of polymer replica mould and R2R manufactured metalens. (a-e) *AFM images and corresponding height profiles of the* ***(a)*** *original mould before R2R imprinting,* ***(b)*** *the mould after 100 m,* ***(c)*** *the mould after 200 m of R2R imprinting,* ***(d)*** *the R2R replicated metalens structure at 1 m, and* ***(e)*** *the R2R replicated metalens structure at 200 m.*

In this section, we perform AFM analysis to further verify the structural integrity of the flexible polymer replica mould before and after 200 m of R2R imprinting, as well as to evaluate the repeatability and uniformity of the R2R manufactured metalens throughout the entire process.

In addition, as suggested by the reviewer, we have compared the focusing efficiencies of metalenses measured at different imprint lengths along the roll, as shown below.

On page 8,

*Beyond a single wafer, reproducibility is examined by measuring four central lenses at different imprint lengths (50, 100, 150, and 200 m) along a 200 m roll, yielding consistent efficiencies of 51.5%, 47.5%, 48.3%, and 47.0%, respectively (**Extended Data Table 1**).*

In Extended Data,

Efficiency (%)	*R1*		*R2*	
Imprint length	*C1*	*C2*	*C1*	*C2*
50 m	*53.3*	*48.1*	*54.5*	*50.2*
100 m	*50.4*	*40.5*	*52.0*	*46.9*
150 m	*48.1*	*44.0*	*51.2*	*49.9*
200 m	*46.6*	*40.9*	*50.6*	*49.7*

Extended Data Table 1. Measured focusing efficiencies of R2R-imprinted metalenses at different imprint lengths from a single replica mould. *Focusing efficiencies of four representative metalenses (2 rows × 2 columns) are measured at imprint distances of 50 m, 100 m, 150 m, and 200 m along the 200-m roll fabricated using a single replica mold.*

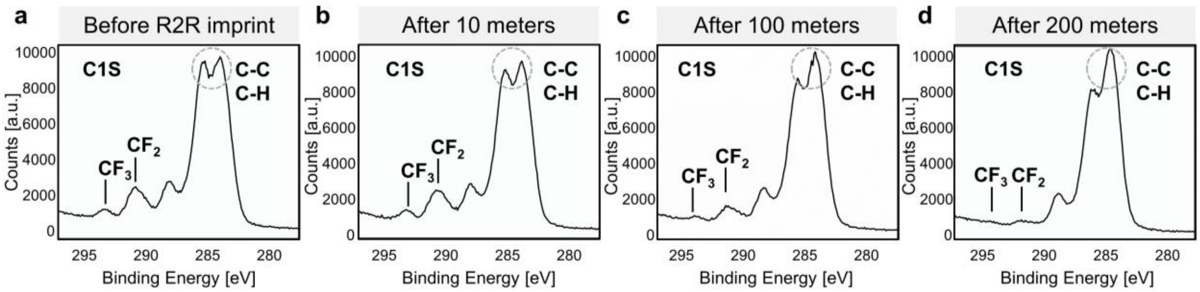
To further verify the durability of the SAM coating, we have analyzed the surface of the polymer replica mould using X-ray photoelectron spectroscopy (XPS) and contact angle measurements, as shown below:

On page 6,

*Furthermore, the x-ray photoelectron spectroscopy (XPS) and contact angle analyses (**Supplementary Note 6**) have been conducted to confirm the chemical and physical stability of the SAM coating throughout the process.*

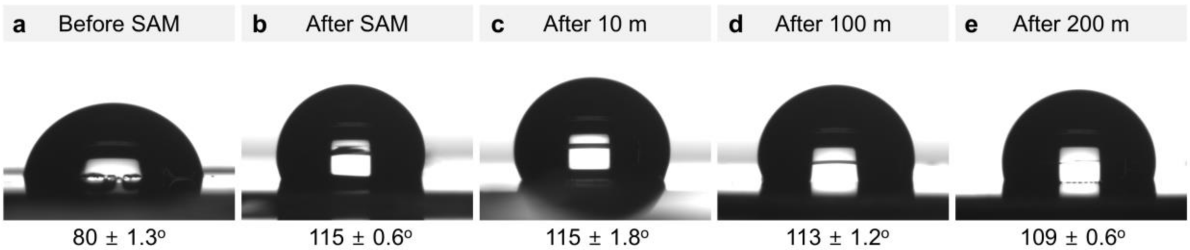
In Supplementary Information,

Supplementary Note 6. Durability test of the replica mold after 200 m of printing.



Supplementary Figure 6. Durability test of the SAM coating layer on the replica mould. (a-d) *X-ray Photoelectron Spectroscopy (XPS) C1s spectra of a deposited F13-TCS film on our flexible polymer shim* ***(a)*** *before R2R imprint process,* ***(b)*** *after 10 m of imprinting****, (c)*** *after 100 m of imprinting, and* ***(d)*** *after 200 m of imprinting.*

In this section, we confirm the durability of the SAM coating on the polymer replica mould by providing more detailed insight into the chemical composition of the coating layer using XPS surface analysis method. The figure shows C1s spectra of an absorbed F13-TCS film on the polymer mould. The two strong peaks at 293.8, 291.4 eV and the broad spectrum that has two peaks around 285 eV can be assigned to the CF3, the CF2 groups and alkane groups of the F13-TCS molecule[1], respectively, which shows the good durability of the SAM coating layer for our mass production process.



Supplementary Figure 7. Measured contact angles of DI water on the replica mould. (a-e) *Water contact angles measured on the replica mould* ***(a)*** *before SAM coating,* ***(b)*** *after SAM coating,* ***(c)*** *after 10 m,* ***(d)*** *after 100 m, and* ***(e)*** *after 200 m of R2R imprinting.*

In References,

1. Beck, M. et al. Improving stamps for 10 nm level wafer scale nanoimprint lithography. Microelectron. Eng. 61, 441–448 (2002).

Comment 1.3

Finally, the authors assert that their atomic layer deposition (ALD) process is R2R-compatible. While this may be theoretically feasible, the four-step ALD process described in the Methods section seems difficult to implement in a true R2R system at a practical throughput and cost. Can the authors provide further justification or references supporting the claim that this ALD process is realistically scalable to R2R? Additionally, is there a specific reason why ALD of TiO_2 on top of the cured resin is preferred over alternatives such as TiO_2-infused resins or printable TiO_2 inks?

Response 1.3

We appreciate the reviewer's insightful comments regarding the ALD process. As the reviewer correctly noted, the ALD employed in our work is a time-divided process, which can limit throughput and increase costs. Because the primary focus of this work is on demonstrating R2R nanoimprint lithography, we have adopted a commercially available time-divided ALD system widely used in academic level. However, in industrial settings, spatial-divided ALD has emerged as a practical alternative that offers significantly higher throughput and lower cost while maintaining the material quality typical of ALD. Notably, Spatial ALD is already being adopted in industry, including in R2R configurations, and its integration would enable true R2R systems with minimal compromise in throughput or cost. For clarity, we have added a discussion in the revised manuscript highlighting this distinction and summarizing several companies actively developing such scalable spatial ALD systems.

Company	Method	Deposition speed	Process condition
Holst Centre	Spatial ALD	1 nm/s	Atmospheric process
SLAD	Spatial ALD	60 nm/ min	Atmospheric process
Beneq	Spatial ALD	10 nm*m/min	Vacuum process

Table R1. Representative examples of spatial ALD implementations by industry.

In Discussion section,

Moreover, improving productivity in the deposition step remains another major challenge. In this work, a time-divided ALD process has been used to deposit the TiO_2 film; however, employing a spatial-divided ALD system could further enhance the overall throughput of the R2R system[41-43].

In References,

41. Poodt, P. et al. Spatial atomic layer deposition: A route towards further *industrialization of atomic layer deposition. Journal of Vacuum Science & Technology A* ***30****, 010802 (2011).*

42 Dickey, E. & Barrow, W. A. High rate roll to roll atomic layer deposition, and its application to moisture barriers on polymer films. Journal of Vacuum Science & Technology A ***30****, 021502 (2012).*

43 Chao, C.-H. et al. Roll-to-roll atomic layer deposition of titania coating on polymeric separators for lithium ion batteries. Journal of Power Sources ***482****, 228896 (2021).*

As the reviewer pointed out, **particle-infused resin** is highly advantageous for one-step fabrication processes. However, the process inherently leaves behind a **high-index residual layer** composed of nanoscale particles, leading to optical losses due to the unwanted scattering and reduced transmittance [***Adv. Sci.*** **12**, 2470317 (2025)]. The residual layer, which consists of nanoparticles and matrix resin, cannot be removed by plasma etching because of the different etching rates between the two materials. In addition, due to the nanoparticle size of approximately 30 nm, it is difficult to maintain a consistent thickness of the residual layer, which could lead to **variations in the metasurface efficiency**. Furthermore, dispensing of nanoparticle-based inks presents a significant challenge in high-speed roll-to-roll (R2R) nanoimprinting processes, owing to the increased risk of nozzle clogging, unstable flow dynamics, and particle aggregation under continuous high-throughput operation [***Langmuir*** **35** (16), 5517-5524 (2019)]. Clogging typically arises from particle agglomeration, solvent evaporation, and sedimentation within the printhead, which obstruct the nozzle orifice and disrupt droplet ejection. In UV-curable formulations, premature photopolymerization or partial crosslinking near the nozzle tip—induced by stray UV exposure or thermal effects—can further exacerbate this problem, leading to irreversible solidification of the ink. Under high-frequency jetting conditions, enhanced shear stress and localized drying promote nanoparticle aggregation and resin crust formation, narrowing the effective flow path. Therefore, while particle infused resin is certainly a promising alternative for metasurface commercialization, we believe that its integration into R2R processes still requires further investigation.

In contrast, our approach produces a residual layer **composed solely of pure resin**, whose refractive index closely matches that of the substrate. As a result, the resin forms a uniform residual layer that does not introduce optical scattering or thin-film interference, thereby ensuring consistent optical performance at an industrial scale. To further verify that the optical efficiency remains consistent under variations in residual layer thickness, we performed imprinting under different pressures of 5, 15, 30, and 45 kg/cm^2 and measured the corresponding optical efficiency. The results confirm that the average efficiency remains nearly constant regardless of the applied pressure, as shown below:

On page 8,

*Finally, focusing efficiencies measured under various imprint pressures (5, 15, 30, and 45 kgf/cm2) average 51.5%, 52.8%, 51.6%, and 46.7%, confirming that imprint pressure has negligible influence on optical performance (**Extended Data Table 2**).*

In Extended Data,

Efficiency (%)	*R1*			*R2*			*R3*		
Imprint pressure	*C1*	*C2*	*C3*	*C1*	*C2*	*C3*	*C1*	*C2*	*C3*
5 kgf/cm²	*49.8*	*52.5*	*53.4*	*41.4*	*55.8*	*51.4*	*52.3*	*50.7*	*52.3*
15 kgf/cm²	*50.4*	*55.6*	*61.4*	*52.0*	*54.1*	*58.1*	*50.4*	*38.1*	*55.4*
30 kgf/cm²	*46.3*	*50.5*	*52.6*	*51.4*	*51.5*	*52.8*	*48.7*	*57.0*	*54.0*
45 kgf/cm²	*49.8*	*44.7*	*48.7*	*45.3*	*48.8*	*45.7*	*45.6*	*46.9*	*44.8*

Extended Data Table 2. Measured focusing efficiencies of R2R-imprinted metalenses under different imprint pressures. *Focusing efficiencies of nine representative metalenses (3 rows × 3 columns) are measured at imprint pressure of 5, 15, 30, and 45 kgf/cm2 to evaluate the effect of imprint pressure on device performance.*

Reviewer #2 (Comments to the Author):

Comment 2

The authors describe roll-to-roll (R2R) nanoimprint of metalenses in the visible spectrum on flexible PET substrate with high throughput. The proposed process utilizes UV-curable imprint to replicate the metalens geometry from a 12 in mold patterned using 193 nm immersion lithography. A conformal layer of TiO_2 is then deposited using a subsequent atomic layer deposition (ALD) process to increase the effective index of the imprinted polymer and thus the efficiency of the printed device. This work is among the first demonstration of scalable manufacturing of hybrid metalens that would be suitable for commercialization and would find broad interests. However, I think there are a number of critical points that require further clarification. These points are outlined in the following questions/comments:

Response 2

We sincerely appreciate the reviewer's careful reading and thoughtful feedback. We also appreciate the reviewer's recognition of the significance of our work. In response to the reviewer's comments and questions, we have carefully addressed each point in a point-by-point manner below. We hope our responses meet the reviewer's expectations and satisfactorily address the concerns.

Comment 2.1

There has been a number of recent publications over the last few month on similar topic of industry-scale manufacturing of metalens (For example: https://doi.org/10.1002/adma.202500327, which focuses on direct printing of inorganic TiO2 composite but also includes a backfill ALD step to increase index). Can the authors comment on the novelty of the current manuscript in comparison to what has recently been published? It would be helpful to quantitatively compare the throughput (linespeed in terms of mm/s), focusing efficiency, yield, etc..

Response 2.1

We sincerely appreciate the reviewer's insightful comment and for pointing out the recent publications on industry-scale metalens manufacturing, including the study by Jung *et al*. We agree that this work represents a meaningful step toward scalable manufacturing of metalenses. However, our manuscript distinguishes itself from these recent studies in several key aspects.

First, the approach by Jung *et al.* [***Adv. Mater.*** **37**, 2500327 (2025)] and other related works primarily rely on plate-to-plate (P2P) NIL, with one recent demonstration of roll-to-plate (R2P) NIL [***Nat. Mater.*** **24**, 535-543 (2025)] as an intermediate step toward continuous processing. In contrast, roll-to-roll (R2R) NIL has enabled fully continuous processing suitable for industrial-scale production, as demonstrated in [***Science*** **355**, 1062-1066 (2017)], where 30 cm-wide metamaterials were fabricated at a speed of 5 m/min. However, since the demonstrated metamaterials were based on films with randomly dispersed nanoparticles, R2R NIL has not yet been applied to metalens fabrication, which requires precise nanoscale patterning. Here, we demonstrate for the first time that **R2R NIL can achieve fully continuous patterning of high-resolution metalenses** over arbitrarily long substrates, distinguishing our work from previous studies. Moreover, we present the first large-scale realization of a 200 m-long, 12-inch metalens array fabricated through continuous R2R manufacturing using the same replica mold, further underscoring the exceptional scalability of our approach.

Second, the throughput of our process is unprecedented: our R2R system achieves a new record of 300 metalenses per second during this revision, corresponding to 1.5 seconds per 12-inch wafer, which is more than **90 times faster than the highest-throughput method** previously reported [***Adv. Mater.*** **37**, 2500327 (2025)]. Overall, our work establishes a new manufacturing paradigm where nanoscale optical precision and industrial-scale throughput coexist, bridging the long-standing gap between laboratory nanofabrication and scalable production of metasurfaces.

To address the reviewer's suggestion, we have included a quantitative comparison table in the **Supplementary Information**, summarizing materials, imprint types, lens diameters, and imprinting speeds from representative NIL-based studies alongside our work. This clearly highlights the novelty of our approach in achieving both the highest throughput and scalable patterned R2R fabrication, which has not been previously demonstrated for metalenses.

In Discussion section,

*To our knowledge, no previous study has reported a throughput as high as 300 lenses per second (**Supplementary Note 9**).*

In Supplementary Information,

Supplementary Note 9. Comparison table of this work and related works.

	Our work	***[2]***	***[3]***	***[4]***	***[5]***	***[6]***
Material	*Resin+ALD*	*Resin+ALD*	*Resin+NPs*	*Resin*	*Resin+NPs*	*Resin+NPs*
Imprint type	*R2R*	*P2P*	*P2P*	*R2P*	*P2P*	*P2P*
Diameter (mm)	*10*	*10*	*4*	*10*	*0.45*	*0.4*
Speed (lens/min)	*18,000*	*45*	*200*	*16*	*0.2*	*0.5*

Supplementary Table 1. Comparison of previously reported nanoimprinted metalenses and this work. *Summary of representative metalens fabricated by nanoimprint lithography, comparing the material, imprint type, lens diameter, and imprinting speed with the R2R-imprinted metalenses demonstrated in this work.*

In References,

*2. Kim, J. et al. Scalable manufacturing of high-index atomic layer–polymer hybrid metasurfaces for metaphotonics in the visible. Nat. Mater. **22**, 474-481 (2023).*

*3. Jung, D. E. et al. Full wafer scale manufacturing of directly printed TiO_2 metalenses at visible wavelengths with outstanding focusing efficiencies. Adv. Mater. **37**, 2500327 (2025).*

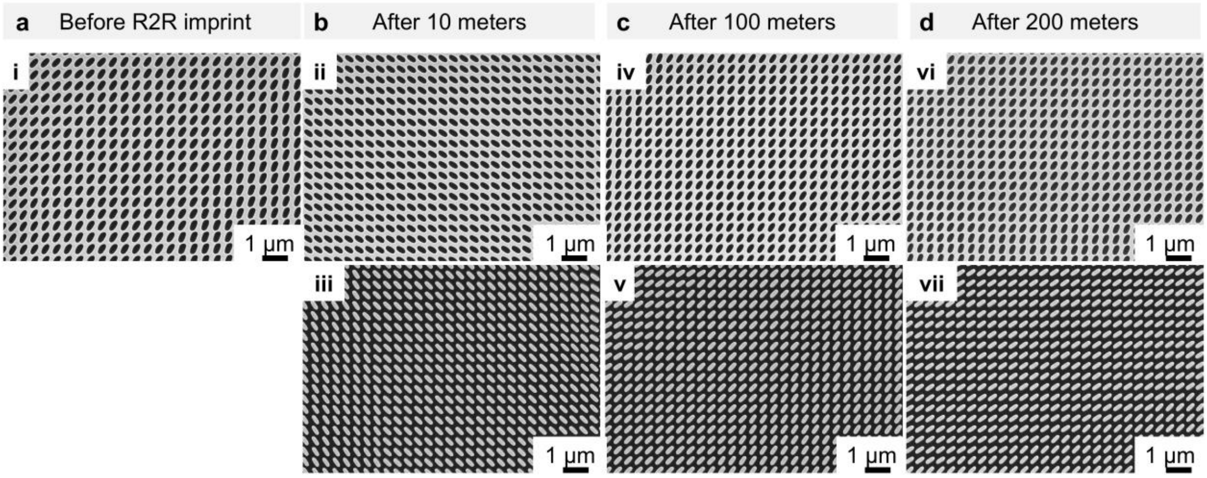
*4. Choi, M. et al. Roll-to-plate printable RGB achromatic metalens for wide-field-of-view holographic near-eye displays. Nat. Mater. **24**, 535-543 (2025).*

*5. Yoon, G. et al. Single-step manufacturing of hierarchical dielectric metalens in the visible. Nat. Commun. **11**, 2268 (2020).*

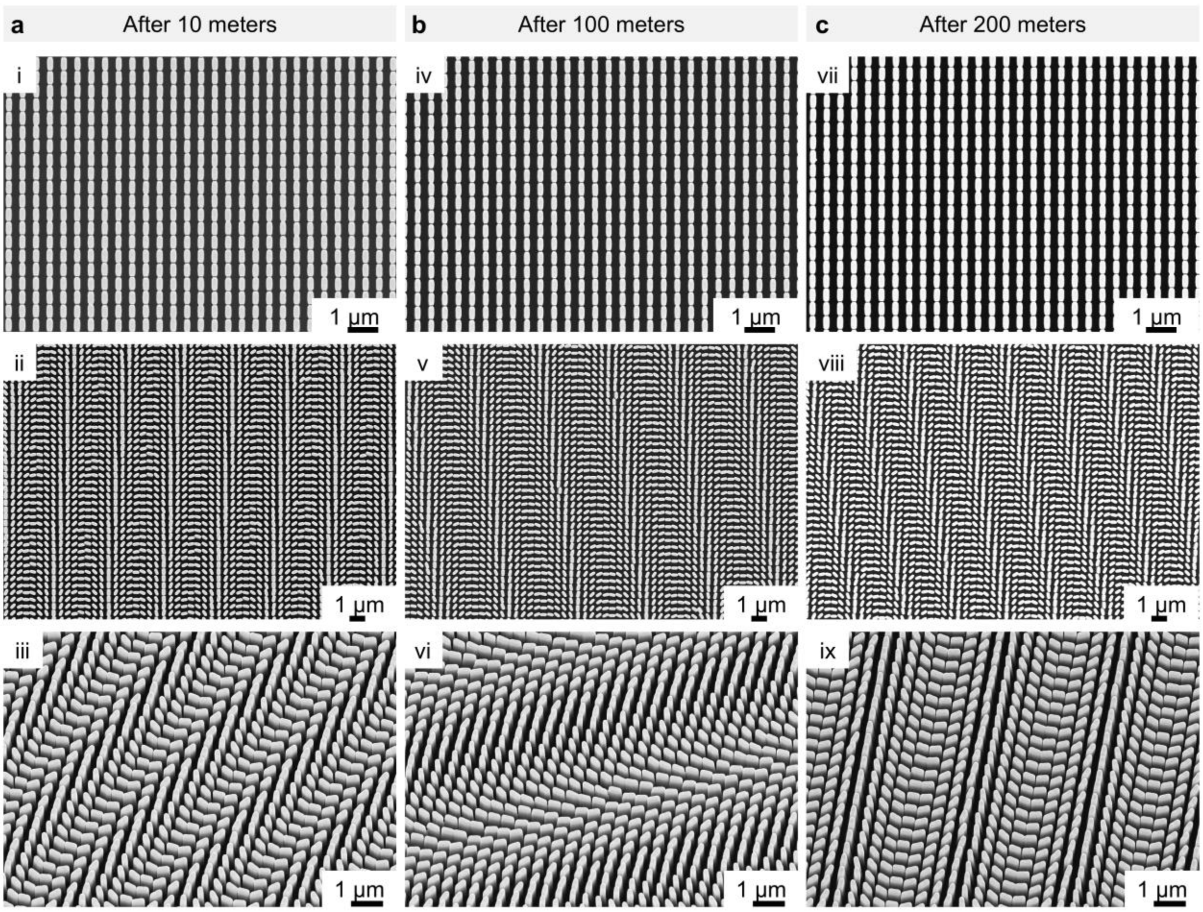
*6. Einch, V. J. et al. Scalable nanoimprint lithography process for manufacturing visible metasurfaces composed of high aspect ratio TiO_2 meta-atoms. ACS Photonics **8**, 2400-2409 (2021).*

In addition, we have included the SEM images and optical efficiency results of 200-m-long continuous R2R imprinting using the same replica mold, as shown below:

In Extended Data Figure,

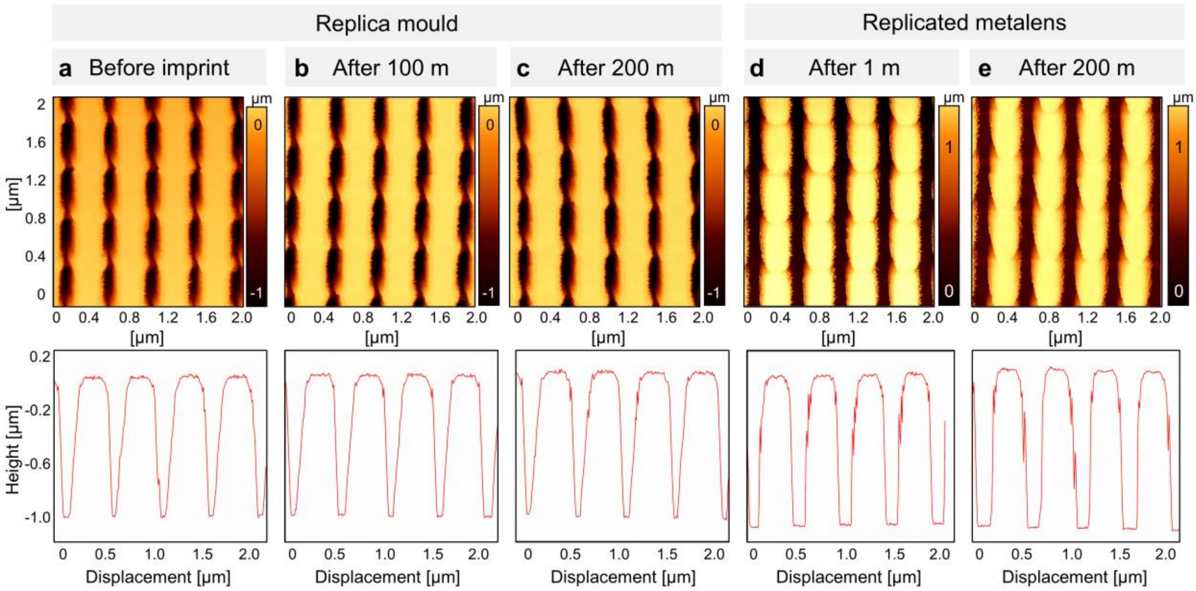


***Extended Data Figure 2. Durability test of the polymer replica mould. (a.i–d.vi)** SEM images of the replica mould: **(a.i)** before R2R imprinting, **(b.ii)** after 10 m, **(c.iv)** after 100 m, and **(d.vi)** after 200 m of R2R imprinting. **(b.iii–d.vii)** Corresponding SEM images of the imprinted resin structures: **(b.iii)** after 10 m, **(c.v)** after 100 m, and **(d.vii)** after 200 m of R2R imprinting.*



In Supplementary Information,

Supplementary Note 5. AFM surface profiles of the replica mold and R2R manufactured metalens.



***Supplementary Figure 5**. **Atomic Force Microscopy (AFM) height profiles of polymer replica mould and R2R manufactured metalens. (a-e)** AFM images and corresponding height profiles of the **(a)** original mould before R2R imprinting, **(b)** the mould after 100 m, **(c)** the mould after 200 m of R2R imprinting, **(d)** the R2R replicated metalens structure at 1 m, and **(e)** the R2R replicated metalens structure at 200 m.*

In this section, we perform AFM analysis to further verify the structural integrity of the flexible polymer replica mould before and after 200 m of R2R imprinting, as well as to evaluate the repeatability and uniformity of the R2R manufactured metalens throughout the entire process.

On page 8,

*Beyond a single wafer, reproducibility is examined by measuring four central lenses at different imprint lengths (50, 100, 150, and 200 m) along a 200 m roll, yielding consistent efficiencies of 51.5%, 47.5%, 48.3%, and 47.0%, respectively (**Extended Data Table 1**).*

In Extended Data,

Efficiency (%)	*R1*		*R2*	
Imprint length	*C1*	*C2*	*C1*	*C2*
50 m	*53.3*	*48.1*	*54.5*	*50.2*
100 m	*50.4*	*40.5*	*52.0*	*46.9*
150 m	*48.1*	*44.0*	*51.2*	*49.9*
200 m	*46.6*	*40.9*	*50.6*	*49.7*

Extended Data Table 1. Measured focusing efficiencies of R2R-imprinted metalenses at different imprint lengths from a single replica mould. *Focusing efficiencies of four representative metalenses (2 rows × 2 columns) are measured at imprint distances of 50 m, 100 m, 150 m, and 200 m along the 200-m roll fabricated using a single replica mold.*

Comment 2.2

The measured focusing efficiency of the metalenses are around 49.5%, which is much lower than the predicted efficiency of up to 90%. Can the authors comment on the discrepancy between model and fabrication? The low efficiency of the lens is a concern since other R2R manufactured metalenses have efficiency of over 80% (cited above).

Response 2.2

We sincerely thank the reviewer for raising this important point regarding focusing efficiency. First, we would like to clarify that the reference cited above, which reports efficiencies over 80%, employs a **P2P NIL** process, rather than a **R2R NIL** process. As described in our **response 2.1**, our manuscript represents the first demonstration of patterned R2R NIL for visible metalenses, enabling fully continuous fabrication over arbitrarily long substrates.

Regarding the discrepancy between simulated and measured focusing efficiencies, it is important to note that conversion efficiency (simulated efficiency) and focusing efficiency (measured efficiency) are fundamentally different metrics. Conversion efficiency refers to the fraction of the incident light in the conjugate polarization (RCP) that acquires the desired phase profile, whereas focusing efficiency quantifies the fraction of the total incident light that is concentrated at the focal spot. As such focusing efficiency is a stricter measure of device performance and is generally lower than conversion efficiency.

For instance, even if the conversion efficiency is 100%, the focusing efficiency can still be reduced by several factors: (i) unwanted multiple diffraction orders (for example, aliasing effects originating from lattice sampling [***Nat. Commun.*** **16**, 411 (2025)]), (ii) rotation of meta-atoms, which introduces weak coupling between neighboring elements that is not captured in periodic conversion efficiency calculations, and (iii) dimensional deviations between the designed and fabricated meta-atoms. A more detailed discussion of how the mixing of focusing and conversion efficiency definitions can lead to variations in reported efficiencies across studies, as well as how unwanted multiple diffraction orders can reduce focusing efficiency, can be found in the paper [***Nat. Photonics*** **17**, 923-924 (2023)].

These factors can be mitigated through further optimization of the phase map, meta-atom design, and fabrication processes. We anticipate that such refinements will enable closer agreement between simulated and experimental efficiencies in future iterations. We have added this clarification to the revised manuscript for completeness.

On page 8,

Although differences between conversion and focusing efficiencies are expected, additional discrepancies between simulated and measured results can arise from aliasing at the metalens level, phase-dependent meta-atom rotation causing weak coupling, and fabrication-induced dimensional variations. These effects can be mitigated through optimized design and fabrication, enabling closer agreement between calculation and experiment.

Comment 2.3

Figure 4b and 4c plot the point spread functon(PSF) and MTF of the metalenses, respectively, which shows a sizable error between the measured vs model. Can the authors comment on the sources of error and why the focus is more broad than expected? Also it appears that the error in the calculated MTF is worse compared with the PSF. Given that the MTF is a normalized Fourier transform of the PSF, it is not clearly to me why the MTF looks so much worse. The scaling property of Fourier transform indicate that if the experimental MTF is half of the bandwidth in the spatial frequency domain (which appears to be the case), the PSF should be twice as wide in the spatial domain? Are the numerical aperture impose by the size of lens/illumination area the same for both cases?

The authors state "The full width at half maximum of the focal spots approached the diffraction limit, indicating near-ideal focusing behaviour." It would be helpful to quantitatively state the FWHM beam width for Figure 4 and compare that to the theoretical calculated width based on the numerical aperture of the lens.

Response 2.3

We thank the reviewer for this insightful comment regarding the discrepancies observed between the measured and modeled PSF and MTF data. We agree that the initial results show noticeable deviations from the ideal predictions and have investigated the sources of error as follows.

First, both PSF and MTF measurements involve focusing a 1-cm-diameter beam to a diffraction-limited focal spot on the order of micrometers. In such conditions, even minor curvature or non-uniformity of the flexible substrate onto which the metalenses are transferred can affect the measurements. We believe this substrate-induced curvature is a major factor contributing to the deviations in the original data. To address this, we perform new measurements after carefully flattening the sample, which resulted in PSF and MTF data that are significantly closer to the ideal values.

Second, regarding the reviewer's observation that the MTF appeared worse than the PSF, we found that this was due to the incident beam size being smaller than the metalens aperture in the initial measurement, effectively reducing the numerical aperture (NA) realized in the experiment. In the new measurements, we employ a beam expander to overfill the lens aperture, ensuring that the measured MTF accurately reflected the designed NA.

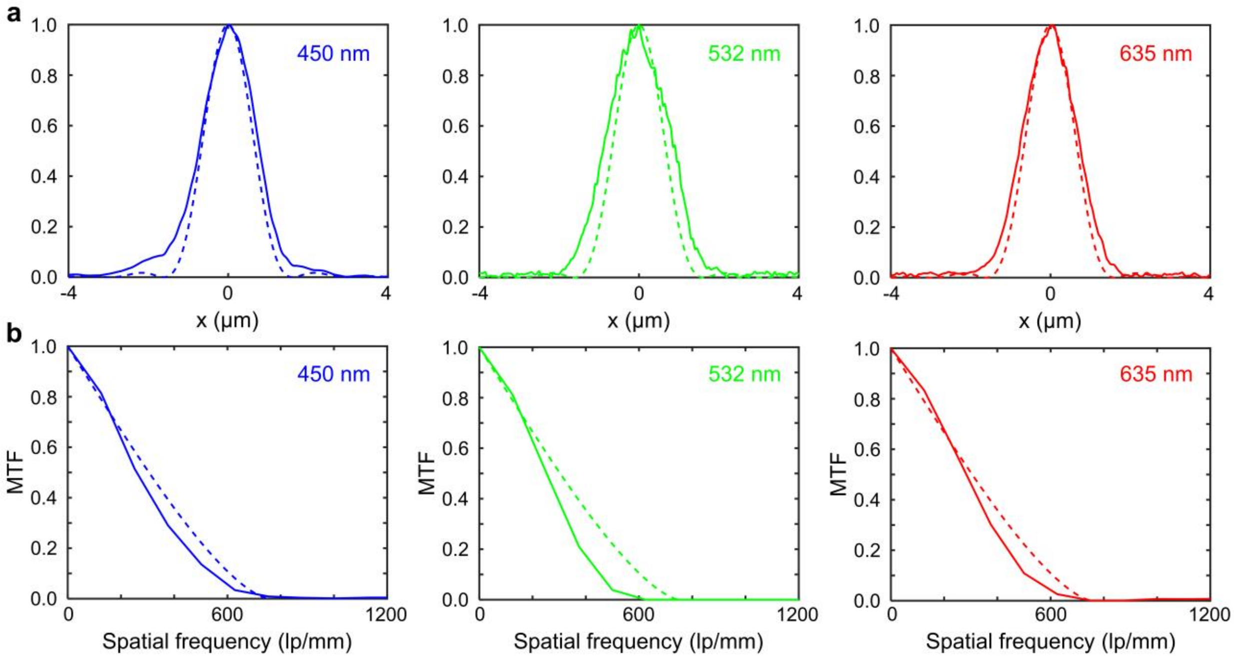
We have also included a quantitative analysis of the full width at half maximum (FWHM) beam widths: the measured FWHMs differ from the theoretical diffraction-limited values by approximately 17% at 450 nm, 28% at 532 nm, and 13% at 635 nm. Given that the theoretical values themselves represent the diffraction limit, these results confirm that our fabricated metalenses operate near the ideal focusing performance.

Finally, to further validate the focusing capability, we perform new imaging experiments using a 1951 USAF resolution target, successfully resolving the smallest elements in the groups 6 and 7 under all three wavelengths. We have revised the characterization section of the manuscript to incorporate these new measurements and quantitative comparisons.

On page 8,

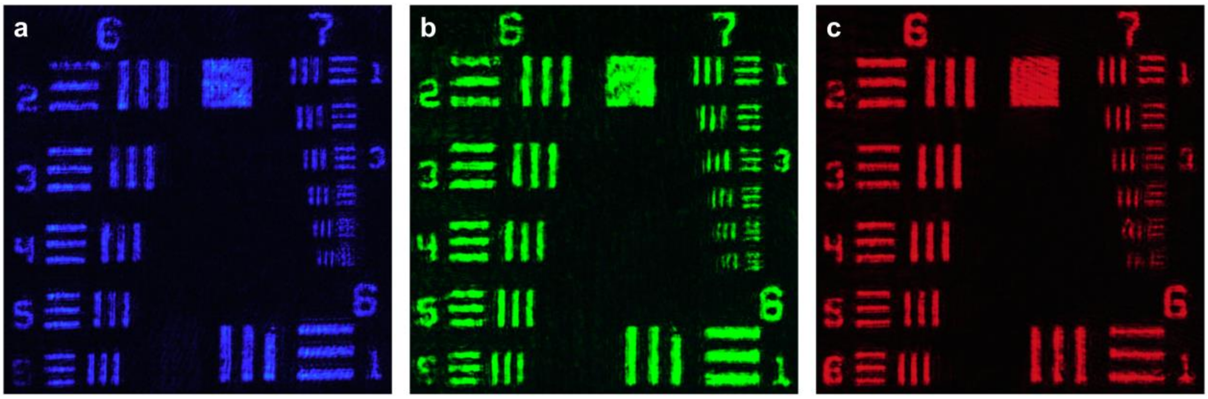
*To experimentally validate the imaging performance of the R2R-imprinted metalens, we image a negative 1951 USAF resolution target, covering all elements from groups 6 and 7, under 450, 532, and 635 nm illumination (**Extended Data Fig. 5**).*

In Extended Data,



Extended Data Figure 4. Intensity profiles and MRF analysis of the fabricated metalens. (a) *Cross-sectional intensity profiles along the x-axis at the focal plane (solid lines) compared with simulated results (dashed lines) for each wavelength.* ***(b)*** *Modulation transfer function (MTF) analysis showing the spatial frequency response of the fabricated metalens (solid lines) and theoretical design (dashed lines) at different wavelengths.*

In Extended Data,



Extended Data Figure 5. Imaging results of the fabricated metalens using a USAF resolution target under RGB illumination. *Imaging results of the fabricated metalens using a negative 1951 USAF resolution target under* ***(a)*** *450 nm,* ***(b)*** *532 nm, and* ***(c)*** *635 nm illumination, where all elements from groups 6 and 7 are imaged.*

Comment 2.4

Can the authors comment on the defect density of the samples fabricated? This point is critical given the manufacturing focus of this paper.

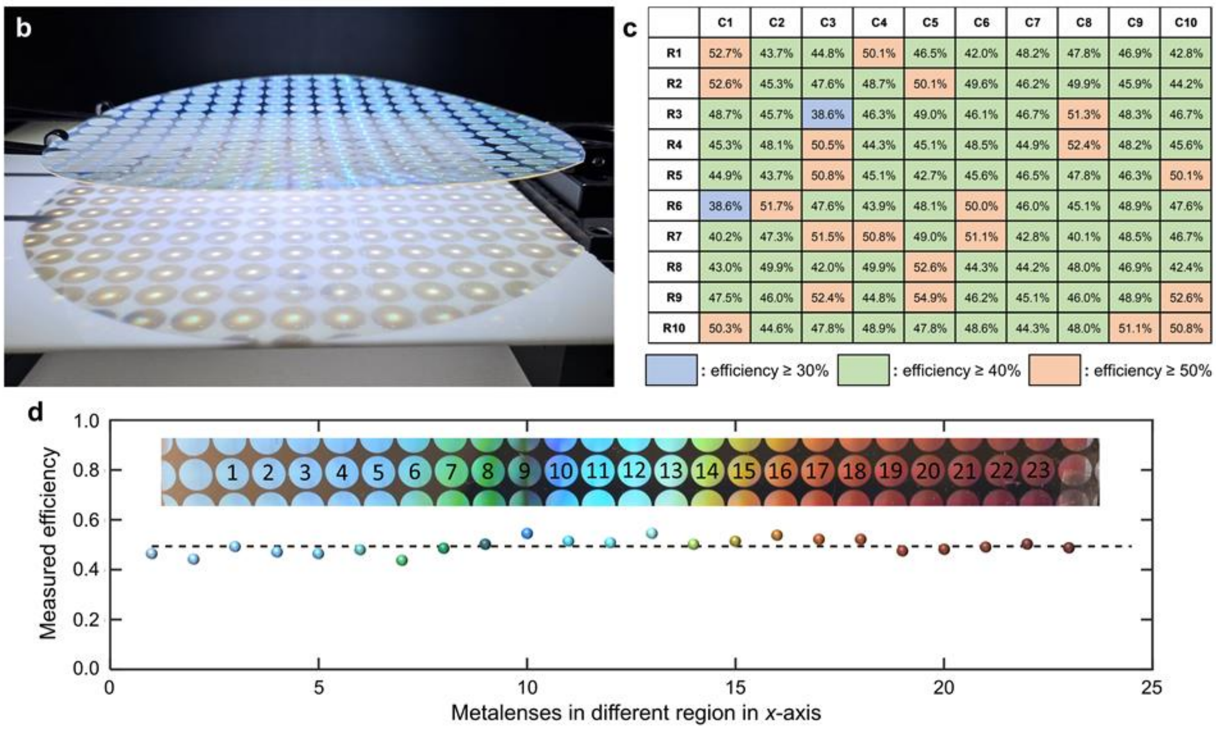
Response 2.4

We thank the reviewer for highlighting the importance of defect density in the context of large-scale manufacturing. In this work, we indirectly assessed defect density by verifying the optical functionality of the fabricated metalenses across the full 8-inch area. A qualitative assessment is first performed by illuminating the entire 8-inch R2R-imprinted sample with white light, which confirms that all metalenses across the wafer produced clear focusing spots. For a more quantitative evaluation, we measure the focusing efficiencies of a 10 × 10 array (100 metalenses) located in the central region of the 8-inch wafer, as below. All measured metalenses exhibit proper focusing behavior with average efficiencies of 47.2% and a standard deviation of ~3.2%, with no metalenses showing catastrophic failure. These results indicate a very low defect density within the fabricated sample and demonstrate the reliability of our R2R nanoimprint process for large-area metalens production.

On page 8,

*Given the large number of metalenses mass-produced via the R2R NIL process, it is essential to ensure consistent optical performance and uniformity. Qualitative inspection under white-light illumination of randomly selected 8-inch sections confirms that all metalenses act as focusing elements (**Fig. 4b**). For quantitative evaluation, a 10 × 10 array in the central region of a 12-inch wafer exhibits an average focusing efficiency of 47.2% with a standard deviation of 3.2%, where only two out of 100 lenses show efficiencies below 40% (**Fig. 4c**). To assess large-area uniformity, focusing efficiencies measured along a single line across the entire 12-inch sample show an average of 49.4% with a deviation of 2.9%, confirming uniform performance across the wafer scale (**Fig. 4d**). Beyond a single wafer, reproducibility is examined by measuring four central lenses at different imprint lengths (50, 100, 150, and 200 m) along a 200 m roll, yielding consistent efficiencies of 51.5%, 47.5%, 48.3%, and 47.0%, respectively (**Extended Data Table 1**). Finally, focusing efficiencies measured under various imprint pressures (5, 15, 30, and 45 kgf/cm2) average 51.5%, 52.8%, 51.6%, and 46.7%, confirming that imprint pressure has negligible influence on optical performance (**Extended Data Table 2**).*

In Figure 4,



	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10
R1	52.7%	43.7%	44.8%	50.1%	46.5%	42.0%	48.2%	47.8%	46.9%	42.8%
R2	52.6%	45.3%	47.6%	48.7%	50.1%	49.6%	46.2%	49.9%	45.9%	44.2%
R3	48.7%	45.7%	38.6%	46.3%	49.0%	46.1%	46.7%	51.3%	48.3%	46.7%
R4	45.3%	48.1%	50.5%	44.3%	45.1%	48.5%	44.9%	52.4%	48.2%	45.6%
R5	44.9%	43.7%	50.8%	45.1%	42.7%	45.6%	46.5%	47.8%	46.3%	50.1%
R6	38.6%	51.7%	47.6%	43.9%	48.1%	50.0%	46.0%	45.1%	48.9%	47.6%
R7	40.2%	47.3%	51.5%	50.8%	49.0%	51.1%	42.8%	40.1%	48.5%	46.7%
R8	43.0%	49.9%	42.0%	49.9%	52.6%	44.3%	44.2%	48.0%	46.9%	42.4%
R9	47.5%	46.0%	52.4%	44.8%	54.9%	46.2%	45.1%	46.0%	48.9%	52.6%
R10	50.3%	44.6%	47.8%	48.9%	47.8%	48.6%	44.3%	48.0%	51.1%	50.8%

Fig. 4. Optical characterization of the R2R-imprinted metalens. (a) *Two-dimensional intensity distributions at the focal plane for 450, 532, and 635 nm wavelengths.* ***(b)*** *Photograph of R2R-imprinted metalenses with an 8-inch-wide section cut from the roll to demonstrate white-light focusing onto a sheet of paper.* ***(c)*** *Measured focusing efficiencies of the central 10 × 10 array (100 metalenses) within the R2R-imprinted sample shown in Fig. 4b, demonstrating uniform performance across the central region.* ***(d)*** *Measured focusing efficiencies of metalenses across different regions along the x-axis of the 12-inch R2R-imprinted sample, highlighting the uniformity of performance over the full area.*

Comment 2.5

One of the key limitations in the throughput of the R2R system is the need of ALD to increase the diffraction efficiency. One approach might be spatial ALD (https://doi.org/10.1116/1.3670745), which can greatly increase throughput and is compatible with R2R. Can the authors comment on potential use of spatial ALD and limitations on throughput?

Response 2.5

We thank the reviewer for this constructive comment on the throughput limitation associated with the ALD steps. We note that in this work our focus is on validating the R2R NIL process, and therefore we adopt a commercially available time-divided ALD system that is widely used in academic settings. While this approach allows us to demonstrate the concept, the four-step time-divided ALD process indeed represents the primary bottleneck in achieving higher overall throughput.

We concur with the reviewer that spatial ALD offers a promising solution to this limitation. Spatial ALD enables continuous material deposition by separating precursor zone in space rather than in time, which significantly increases deposition rates while maintain the material quality typical of conventional ALD. This approach has already been demonstrated in industrial settings and is inherently compatible with R2R processing. Integration of spatial ALD with our R2R NIL platform would thus allow high-index coatings to be deposited at practical throughput and cost, overcoming the primary limitation identified by the reviewer. Representative examples of industrial implementations of spatial ALD are summarized in **Table R1**, which highlights the achievable deposition speeds and process conditions demonstrated by leading research centers and companies. These examples further support the feasibility of integrating spatial ALD with our R2R NIL platform for high-throughput production of high-index coatings.

Company	Method	Deposition speed	Process condition
Holst Centre	Spatial ALD	1 nm/s	Atmospheric process
SLAD	Spatial ALD	60 nm/ min	Atmospheric process
Beneq	Spatial ALD	10 nm*m/min	Vacuum process

Table R1. Representative examples of spatial ALD implementations by industry.

To address this point, we have revised the manuscript to include a discussion on spatial ALD as a scalable alternative to the time-divided ALD used in this study, and to cite relevant industrial and academic developments in this field.

In Discussion section,

Moreover, improving productivity in the deposition step remains another major challenge. In this work, a time-divided ALD process has been used to deposit the TiO2 film; however, employing a spatial-divided ALD system could further enhance the overall throughput of the R2R system[41-43].

In References,

41. Poodt, P. et al. Spatial atomic layer deposition: A route towards further industrialization of atomic layer deposition. Journal of Vacuum Science & Technology A ***30****, 010802 (2011).*

42 Dickey, E. & Barrow, W. A. High rate roll to roll atomic layer deposition, and its application to moisture barriers on polymer films. Journal of Vacuum Science & Technology A ***30****, 021502 (2012).*

43 Chao, C.-H. et al. Roll-to-roll atomic layer deposition of titania coating on polymeric separators for lithium ion batteries. Journal of Power Sources ***482****, 228896 (2021).*

Reviewer #3 (Comments to the Author):

Comment 3

The manuscript reports a big step towards continuous and high-throughput manufacturing of large area meta-lens by roll-to-roll (R2R) nanoimprint (NIL) technology. It's an impressive work through the collaboration of three research institutes. This contribution is somewhat different from a typical Nature submission, because the presented work and results represent a combined expertise of multiple PI's labs, which individually have been developed and demonstrated before, e.g. meta-surface fabrication by using NIL and coating with ALD high refractive index layer to significantly improve the index contrast; R2R NIL process, and the use of polymer templates from a Master mold for cost-effective replication of the sub-wavelength structures and for managing defects by finite use of the polymer template. Since the focus is on a scalable manufacturing process, the authors would want to provide some more detailed comparison and analysis on the uniformity and repeatability of the process and the fabricated metalenses. Specifically I'd recommend adding these analysis:

Response 3

We sincerely appreciate the reviewer's constructive and insightful comments. In particular, we are grateful for the recognition of our work as a "**big step towards continuous and high-throughput manufacturing of metalens**". We fully agree that a thorough comparison and analysis of our approach is essential. To address the reviewer's comments comprehensively, we have provided detailed and point-by-point responses below. We hope that our revisions and explanations sufficiently address the concerns raised.

Comment 3.1

The residual layer thickness control is important for this application. It is mentioned "We optimized the imprint pressure at 45 kgf/cm2 to minimize the residual layer thickness..." please give a range of the residual thickness obtained, and the thickness distribution (uniformity) across the samples and multiple imprints. What are impacts of the thickness variation of the residual layer on the behavior of the metalenses?

Response 3.1

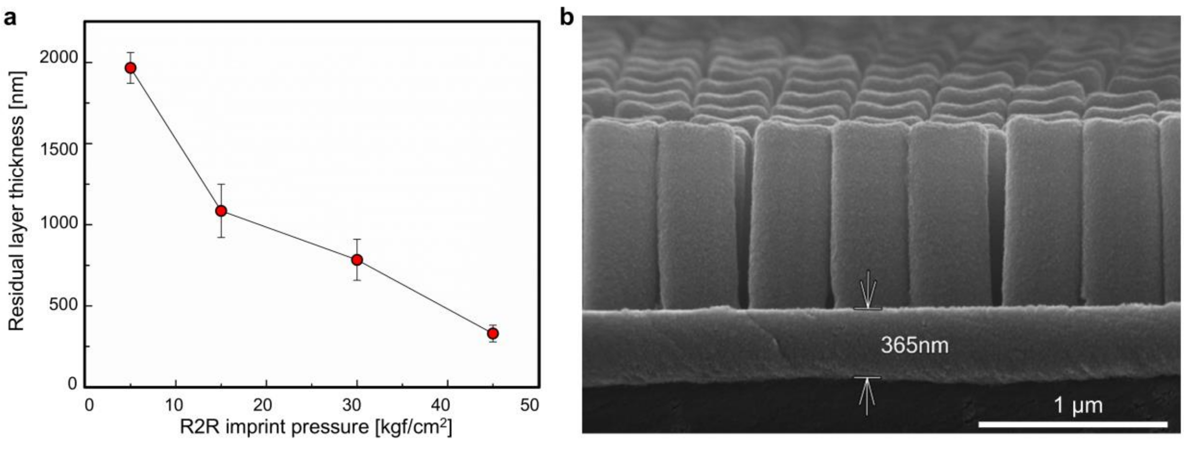
We appreciate the reviewer's insightful comment regarding the residual layer thickness and its potential impact on metalens performance. In our design, the residual layer and the PET substrate share nearly identical refractive index (~1.5). Therefore, variations in its absolute thickness do not introduce significant optical phase errors or affect the focusing behavior of the metalenses. This is one of the key advantages of our work, as maintaining consistent efficiency across mass-produced metalenses is particularly critical in industrial applications. To experimentally validate this, we first applied different imprint pressures (5, 15, 30, and 45 kgf/cm^2) to obtain various residual layer thicknesses, as shown in **Supplementary Note 8**. Among them, the highest imprint pressure of 45kgf/ cm^2 yielded thinnest residual layer of 329.47 nm, with a standard deviation of 51.16 nm. 10 batches were manufactured for each imprint pressure, and each measurement was conducted on different samples in multiple imprint batches.

On page 7,

*In our system, the imprinting pressure can be applied uniformly over a large area by precisely controlling the counter roller and sensor system. We optimize the imprint pressure at 45 kgf/cm2 to minimize the residual layer thickness (329.67 ± 51.16 nm) and enable faithful replication of high aspect ratio features at a high speed (**Supplementary Note 8**).*

In Supplementary Information,

Supplementary Note 8. Optimization of imprint pressure for lower residual layer.



*Supplementary Figure 9. **Optimization of imprint pressure for residual layer thickness.** (a) Plot of residual layer thickness as a function of imprint pressure. **(b)** Cross-sectional SEM images of R2R manufactured metalens.*

We then measured the focusing efficiencies of nine representative metalenses for each condition and observed negligible differences in performance (average efficiencies ranging from ~46% to ~53%). These results confirm that residual layer thickness variations have minimal impact on optical performance. We have added this explanation and the corresponding experimental data to the revised manuscript to clarify the influence of residual layer thickness and its uniformity across different imprint pressures.

On page 8,

*Finally, focusing efficiencies measured under various imprint pressures (5, 15, 30, and 45 kgf/cm2) average 51.5%, 52.8%, 51.6%, and 46.7%, confirming that imprint pressure has negligible influence on optical performance (**Extended Data Table 2**).*

In Extended Data,

Efficiency (%) / Imprint pressure	R1			R2			R3		
	C1	C2	C3	C1	C2	C3	C1	C2	C3
5 kgf/cm^2	49.8	52.5	53.4	41.4	55.8	51.4	52.3	50.7	52.3
15 kgf/cm^2	50.4	55.6	61.4	52.0	54.1	58.1	50.4	38.1	55.4
30 kgf/cm^2	46.3	50.5	52.6	51.4	51.5	52.8	48.7	57.0	54.0
45 kgf/cm^2	49.8	44.7	48.7	45.3	48.8	45.7	45.6	46.9	44.8

Extended Data Table 2. Measured focusing efficiencies of R2R-imprinted metalenses under different imprint pressures. *Focusing efficiencies of nine representative metalenses (3 rows × 3 columns) are measured at imprint pressure of 5, 15, 30, and 45 kgf/cm^2 to evaluate the effect of imprint pressure on device performance.*

Comment 3.2

Report or suggest a way to quickly characterize or sample the optical performance of the fabricated metalenses, across multiple locations distributed on the imprinted 300 metalenses per unit time. Especially are there significant optical efficiency variations, and if there is, what's the main structural parameter that influences the optical performance. Supply some representative SEM showing such variations and estimate the percentage of samples of out-of-range variations.

Response 3.2

We appreciate the reviewer's valuable suggestion to discuss rapid characterization and optical sampling strategies across the mass-produced metalenses. For industrial adoption, in-situ characterization is critical to ensure real-time process control. To quickly characterize the optical performances of 300 metalenses, we performed a qualitative white-light illumination test on entire 8-inch sections cut from the roll, which enables rapid verification that all metalenses across the wafer produced focusing spots (**Fig. 4b**). This method provides a straightforward way to screen multiple locations simultaneously and identify catastrophic defects at a glance. In addition, such rapid optical assessment could be further enhanced by integrating a CCD or high-speed camera at the focal plane to acquire PSF images in real time for quantitative evaluation.

In regards to the reviewer's comment on efficiency variations, we carried out quantitative focusing efficiency measurements on representative subsets of the metalenses. Specifically, we measured 100 metalenses arranged in a 10 x 10 array at the wafer center (**Fig. 4c**) and observed an **average efficiency of 47.2%** with a standard deviation of 3.2%. **Only two metalenses exhibited efficiencies below 40%**, corresponding to 2% of the total, indicating minimal out-of-range variations. To further check for large-area variations, we measure efficiencies along a single line across the entire 12-inch sample (**Fig. 4d**) and found an **average efficiency of 49.4%** with a standard deviation of 2.9%, comparable to the central region.

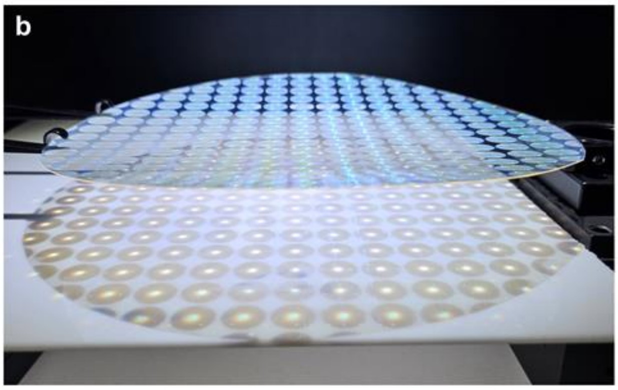
These results indicate that both optical efficiency variations and the percentage of out-of-spec samples are minimal. Therefore, the defects that affect optical efficiency are of very low density across the continuously imprinted samples. The details regarding the main defects and the possible solutions are provided in response to **Comment 3.3**, and the manuscript has been revised accordingly to include these additional measurements.

On page 8,

*Given the large number of metalenses mass-produced via the R2R NIL process, it is essential to ensure consistent optical performance and uniformity. Qualitative inspection under white-light illumination of randomly selected 8-inch sections confirms that all metalenses act as focusing elements (**Fig. 4b**). For quantitative evaluation, a 10 × 10 array in the central region of a 12-inch wafer exhibits an average focusing efficiency of 47.2% with a standard deviation of 3.2%, where only two out of 100 lenses show efficiencies below 40% (**Fig. 4c**). To assess large-area uniformity, focusing efficiencies measured along a single line across the entire 12-*

*inch sample show an average of 49.4% with a deviation of 2.9%, confirming uniform performance across the wafer scale (**Fig. 4d**). Beyond a single wafer, reproducibility is examined by measuring four central lenses at different imprint lengths (50, 100, 150, and 200 m) along a 200 m roll, yielding consistent efficiencies of 51.5%, 47.5%, 48.3%, and 47.0%, respectively (**Extended Data Table 1**).*

In Figure 4b-d,



c

	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10
R1	52.7%	43.7%	44.8%	50.1%	46.5%	42.0%	48.2%	47.8%	46.9%	42.8%
R2	52.6%	45.3%	47.6%	48.7%	50.1%	49.6%	46.2%	49.9%	45.9%	44.2%
R3	48.7%	45.7%	38.6%	46.3%	49.0%	46.1%	46.7%	51.3%	48.3%	46.7%
R4	45.3%	48.1%	50.5%	44.3%	45.1%	48.5%	44.9%	52.4%	48.2%	45.6%
R5	44.9%	43.7%	50.8%	45.1%	42.7%	45.6%	46.5%	47.8%	46.3%	50.1%
R6	38.6%	51.7%	47.6%	43.9%	48.1%	50.0%	46.0%	45.1%	48.9%	47.6%
R7	40.2%	47.3%	51.5%	50.8%	49.0%	51.1%	42.8%	40.1%	48.5%	46.7%
R8	43.0%	49.9%	42.0%	49.9%	52.6%	44.3%	44.2%	48.0%	46.9%	42.4%
R9	47.5%	46.0%	52.4%	44.8%	54.9%	46.2%	45.1%	46.0%	48.9%	52.6%
R10	50.3%	44.6%	47.8%	48.9%	47.8%	48.6%	44.3%	48.0%	51.1%	50.8%

(blue): efficiency ≥ 30% (green): efficiency ≥ 40% (orange): efficiency ≥ 50%

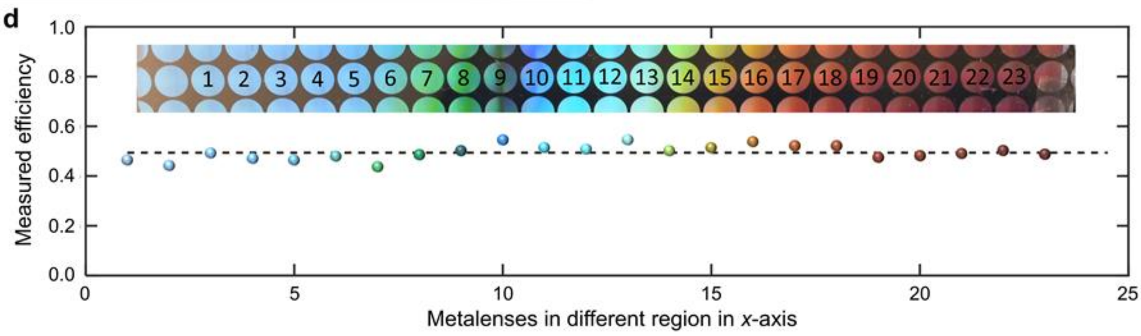


***Fig. 4. Optical characterization of the R2R-imprinted metalens. (a)** Two-dimensional intensity distributions at the focal plane for 450, 532, and 635 nm wavelengths. **(b)** Photograph of R2R-imprinted metalenses with an 8-inch-wide section cut from the roll to demonstrate white-light focusing onto a sheet of paper. **(c)** Measured focusing efficiencies of the central 10 × 10 array (100 metalenses) within the R2R-imprinted sample shown in Fig. 4b, demonstrating uniform performance across the central region. **(d)** Measured focusing efficiencies of metalenses across different regions along the x-axis of the 12-inch R2R-imprinted sample, highlighting the uniformity of performance over the full area.*

Comment 3.3

What are the major defects identified in characterizing a large number of metalenses? what would you recommend/suggest to minimizing such defects?

Response 3.3

We appreciate the reviewer's constructive comments regarding the major defects in our manufactured metalenses. The major defects identified in our manufactured metalenses are primarily associated with:

- Incomplete pattern replication: this defect occasionally occurred in some of our manufactured metalens due to **demolding defects** or **incomplete resin filling** in our high-aspect-ratio mold feature as shown in **Figure R1 a** and **b**.
- Local surface contamination defects: this defect mainly caused by particle contamination on PET substrate during the delivery process as well as transferring from many rollers surface during open-air and continuous R2R operation which leads to a big void in our fabricated devices as shown in **Figure R1 c** and **d**.

To minimize these defects, several process optimizations are recommended:

- Further optimization of roller geometry for uniform nip pressure over a large area: implementation of multi-backup-roller configurations or compliant sub-rollers can help to minimize roller bending and ensure homogeneous contact pressure.
- Environmental and dust control: lengthy & continuous imprinting process increases the risk of contamination. Automated cleaning systems for molds and rollers should be installed to remove the dust particles before imprinting which can further improve production yield.
- In-line metrology and data feedback: Integrating optical inspection, and defect-detection imaging enables precise identification of systematic process deviations. When combined with machine-learning-based feedback control and a digital twin, these in-line diagnostics enable automatic process optimization in next-generation pilot R2R manufacturing systems, paving the way toward the commercialization of large-area metalenses.

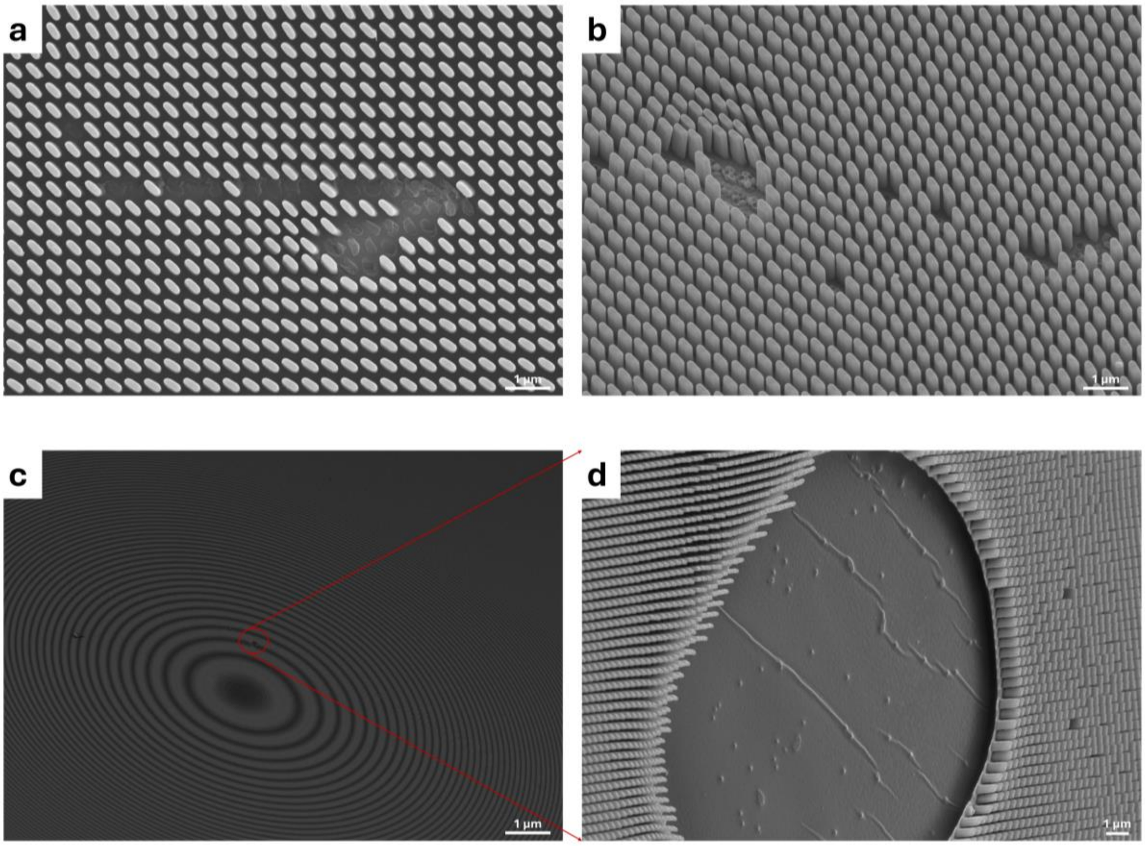


***Figure R1. Defects in R2R manufactured metalens**. **(a-b)** SEM images of defects caused by incomplete pattern replication, **(c-d)** SEM images of local surface contamination defects.*

In addition, we have discussed this aspect in the manuscript, as shown below:

In Discussion section,

Despite these advances, in-situ metrology and quality control remain crucial for the mass production of metalenses, and further investigation into real-time monitoring of structural uniformity and optical efficiency are required[40].

Reviewer #4 (Remarks to the Author):

Comments 4

I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Response 4

We sincerely appreciate the reviewer for the time and effort in co-reviewing our manuscript. We truly appreciate the collaborative evaluation and the constructive feedback provided through this process, and we hope that our responses satisfactorily address all of the concerns raised.

Response Letter

Reviewer #1 (Remarks to the Author):

Comment 1

The revision is a substantially improved version of an already impressive manuscript. I appreciate the additional discussions on mold durability, optical uniformity, and the scalability of the ALD process.

However, I cannot wholeheartedly recommend publication in Nature. Rather, I recommend publication of this work in a journal such as Nature Materials, Nature Photonics, Nature Communications, or Advanced Materials. While the engineering achievements and manufacturing scalability demonstrated here are excellent and will be valuable to the field, the manuscript focuses on advancing fabrication throughput rather than introducing new scientific phenomena. As such, it does not align with Nature's typical editorial criteria for conceptual scientific novelty.

Response 1

We appreciate the reviewer for the thoughtful evaluation and for recognizing the substantial improvements in our revised manuscript. We are particularly grateful for the acknowledgement of the impact of our engineering achievements and demonstrated manufacturing scalability on the community which can reach a conclusion of interest to an interdisciplinary readership. We respectfully believe that our work aligns with Nature's stated editorial criteria in several key respects. First, Nature considers technical advances that "are of outstanding scientific importance" and "have significant impacts on communities of fellow researchers". To the best of our knowledge, this work is the first demonstration of large-scale continuous roll-to-roll manufacturing of visible metalenses enabling high-volume production of metasurface optics, addressing a long-standing translational barrier in nanophotonics. Second, our work also "reach a conclusion of interest to an interdisciplinary readership" by bridging nanophotonics & optical physics, materials science and industrial-scale precision manufacturing, enabling manufacturing paradigm of metasurface optics for real-world systems.

Reviewer #2 (Remarks to the Author):

Comments 2

The authors have adequately address all of my concerns and questions and I think the article is suitable for publication.

Response 2

We sincerely thank the reviewer for the positive assessment of our revised manuscript and the constructive feedback provided during the review process, which has significantly improved our work.

Reviewer #3 (Remarks to the Author):

Comments 3

I am satisfied with the authors responses and revised manuscript.

Response 3

We are grateful for the reviewer's positive evaluation of our revised manuscript and confirmation that the revisions have satisfactorily addressed all concerns.

Reviewer #4 (Remarks to the Author):

Comment 4

The author has addressed most of my previous questions. However, after carefully reviewing the revised manuscript again, I still have several remaining concerns and questions.

Response 4

We sincerely thank the reviewer for the constructive follow-up comments. We have carefully considered the remaining concerns and provide detailed responses below.

Comment 4.1: Scope and performance relevance

Given the broad readership of Nature, I believe it is important for the manuscript to clearly address two fundamental questions at the outset:

Performance: Is the demonstrated metalens performance sufficient for any meaningful or practical applications?

Manufacturability: Can this technology consistently produce devices with acceptable yield, throughput, and cost?

After reading the manuscript, I find that the authors have addressed the second question (manufacturability) very well. However, the first question regarding performance and application relevance remains insufficiently clarified.

In particular, it is still unclear whether the work primarily demonstrates a scalable fabrication method for metalenses with relatively low performance, or whether the demonstrated devices meet the requirements of any realistic applications. Without this context, it is difficult to assess the overall impact of the work.

Response 4.1

We sincerely appreciate the reviewer's constructive feedback on the practical impact and performance relevance of our work. Following the suggestions, we have thoroughly revised the manuscript to define application-specific metrics, benchmark our results against exiting fabrication methods, and justify the trade-offs between performance and scalability. We have further addressed the specific points regarding application-relevant thresholds and benchmarking in the detailed responses below.

Comment 4.2: Suggestions to address the above concerns

To strengthen the manuscript and clarify its significance, I suggest the authors consider the following:

a) Define application-relevant performance metrics

   The authors should clearly identify which metrics are most meaningful for the intended applications (e.g., conversion efficiency, focusing efficiency, Strehl ratio, imaging contrast, etc.), and provide reference values or thresholds that are generally considered acceptable for those applications.

Response 4.2a

We would like to express our sincere gratitude to the reviewer for this constructive suggestion. We agree that providing a standardized performance metric is essential for assessing the practical viability of our metalenses. In response, we have now incorporated the Strehl ratio (SR) as a primary metric to evaluate the imaging performance and contrast of our devices.

The Strehl ratio, which compares the peak intensity of the actual focal spot to that of an ideal diffraction-limited system, is a widely accepted benchmark in optical engineering. Specifically, according to the **Maréchal criterion**, an **SR value of 0.8 or higher indicates that a system has achieved diffraction-limited performance**, making it suitable for high-end imaging applications. While our previous manuscript focused on fabrication metrics, we have now newly measured the Strehl ratio across the visible spectrum to provide the clarity the reviewer requested.

The measured SR values are 0.9027, 0.8096, and 0.9179 for wavelengths of 450 nm, 532 nm, and 635 nm, respectively. Since all values consistently exceed the 0.8 threshold, these results quantitatively demonstrate that our scalable fabrication method delivers performance that meets the rigorous standards required for practical optical systems. We believe this additional analysis, prompted by the reviewer's insight, significantly strengthens the manuscript by validating the real-world applicability of our work.

On page 7-8,

Furthermore, the Strehl ratio (SR) is calculated to further evaluate the focal spot quality. The obtained SR values are 0.9027, 0.8096, and 0.9179 for 450, 532, and 635 nm, respectively. These values, being above the Marechal criterion (SR > 0.8), confirm that our R2R-imprinted metalens operates as a diffraction-limited imaging system across the visible spectrum.

Comment 4.2b

b) Benchmark against existing fabrication approaches

A quantitative comparison of device performance with metalenses fabricated using other methods (e.g., e-beam lithography, DUV lithography, or alternative NIL approaches) would greatly help place this work in context.

Response 4.2b

We fully agree that a quantitative benchmark against established fabrication methods is crucial for contextualizing the impact of our work. In response to the reviewer’s request, we have performed a comparative analysis of the Strehl ratio (SR) among metalenses fabricated via E-beam lithography (EBL) [Ref 1: ***Science*** **352**, 1190-1194 (2016)], Deep-UV (DUV) lithography [Ref 2: ***Laser Photonics Rev.*** **19**, 2401425 (2025)], and Plate-to-Plate (P2P) nanoimprint lithography (NIL) [Ref 3: ***Nat. Mater.*** **22**, 474-481 (2023)], as summarized in the table below.

Strehl Ratio	Blue	Green	Red
This work	0.90	0.81	0.92
Ref 1	0.76	0.78	0.77
Ref 2	0.94	0.96	0.99
Ref 3	0.81	0.90	0.86

As shown in the table, all the benchmarked metalenses, including ours, achieve SR values around or above the Maréchal criterion (0.8), indicating successful fabrication. Notably, the metalenses produced by our method exhibit higher SR values compared to those fabricated via EBL and P2P NIL. While DUV lithography remains a high-performance standard, our results demonstrate that our scalable manufacturing approach delivers optical quality that is not only comparable to DUV but also superior to the widely used EBL and conventional NIL techniques.

This benchmark provides clear evidence that our technology transcends the typical trade-off between throughput and precision. By achieving near-ideal SR values consistently across the visible spectrum, we confirm that our work enables the **mass production of metasurfaces without sacrificing the high-fidelity optical performance** required for advanced applications.

Comment 4.2c

c) Justify performance trade-offs

If the proposed method does not outperform existing techniques in terms of optical performance, the authors should clearly argue why the demonstrated performance is nevertheless sufficient for practical use and why it represents a compelling trade-off for large-scale manufacturing.

Response 4.2c

We appreciate the reviewer's request to clarify the positioning of our work regarding performance trade-offs. While our method may not drastically outperform the absolute highest values reported by individual, labor-intensive EBL-fabricated samples, it delivers optical performance that is highly comparable to established benchmarks.

We acknowledge that printable materials typically possess an effective refractive index in the range of 2.0 to 2.2. Due to this moderate index, a little larger sampling period that is close to the wavelength is required, which can lead to a trade-off in efficiency in high-numerical aperture (NA) designs requiring steep light bending. However, this is a fundamental challenge shared by all metasurfaces that do not utilize extremely high-index, non-printable inorganic materials (TiO_2, SiN, GaN). It is not a limitation unique to our proposed fabrication process.

The true significance of our work, and why we believe it holds high impact for the readership of Nature, is that it effectively overcomes the long-standing trade-off between scalability and performance. Historically, the field has been polarized between high-performance but unscalable methods (EBL) and scalable but low-precision techniques. Our work demonstrates, for the first time, that high-fidelity metasurfaces can be manufactured at a massive scale without the typical "performance degradation". By bridging this gap, we provide a compelling solution for the practical deployment of nanophotonics, representing a major leap forward from laboratory-scale demonstrations to real-world industrial application.