

Supplementary Information for

Mosquito-eye inspired hydrophobic metasurface enabled by NIL

with sub-100 nm resolution

Dingyu Xu¹, Qianqian Zhang¹, Zhiwei Liu¹, Jingcheng Zhang⁴, Qisong Li^{1,2}, Junxiao Zhou^{3,*} and Xiewen Wen^{1*}

¹Department of Industrial and Systems Engineering, The Hong Kong Polytechnic University, Kowloon, Hong Kong, SAR, China.

²School of Optical-Electrical and Computer Engineering, University of Shanghai for Science and Technology, Shang-hai, 200093, China.

³National Laboratory of Solid State Microstructures, Key Laboratory of Intelligent Optical Sensing and Manipulations, Jiangsu Key Laboratory of Artificial Functional Materials, Collaborative innovation center of advanced microstructures, College of Engineering and Applied Sciences, Nanjing University, Nanjing, China.

⁴School of Electronics and Information, Northwestern Polytechnical University, Xi'an, China.

[#]Corresponding author. Email: junxiaozhou@nju.edu.cn; xw.wen@polyu.edu.hk

Supplementary Note 1: XPS of hydrophobic resin

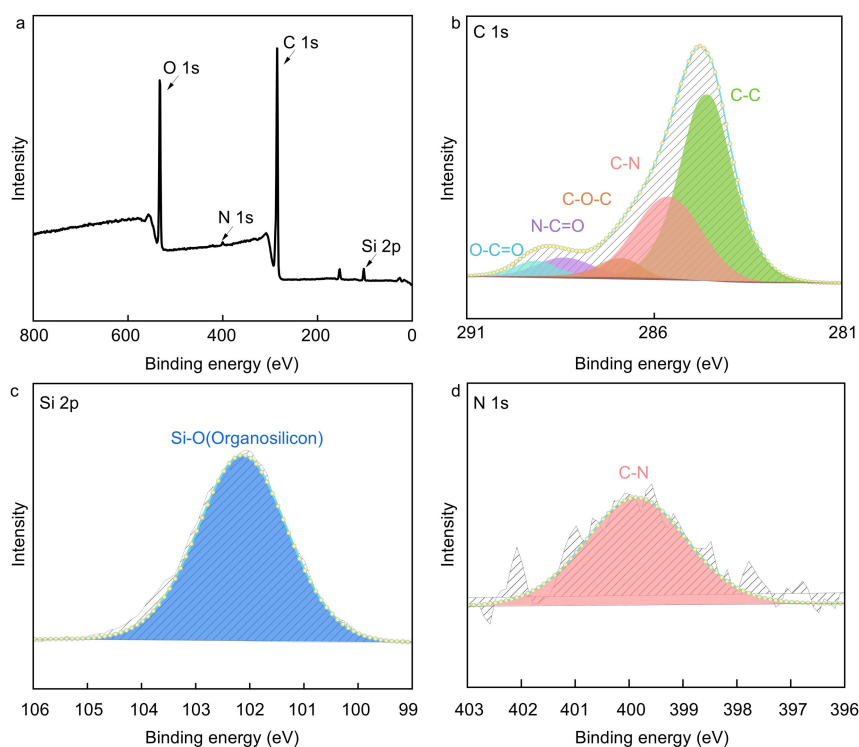


Fig. S1 X-ray photoelectron spectroscopy (XPS) analysis of the hydrophobic resin surface. (a) The full XPS spectrum of the sample demonstrates that the surface is mainly composed of four elements: C, O, N, and Si. (b) The deconvolution result of the C 1s high-resolution spectrum. (c) The Si 2p high-resolution spectrum. (d) The N 1s high-resolution spectrum.

Fig. S1 provides the X-ray photoelectron spectroscopy (XPS) survey scan and high-resolution spectra for the carbon, nitrogen, and silicon elements of the hydrophobic resin surface. The survey spectrum (Fig. S1a) reveals that the surface is composed of carbon (C 1s), nitrogen (N 1s), oxygen (O 1s), and silicon (Si 2p), with no other significant impurity elements detected. High-resolution spectra for the C 1s, Si 2p, and N 1s core levels were subsequently acquired to probe the chemical bonding environment. The deconvoluted C 1s spectrum (Fig. S1b) distinguishes several chemical states of carbon: the main peak at 284.8 eV is assigned to C-C bonds in alkyl chains, whereas the peaks at approximately 285.63 eV, 286.84 eV, 288.40 eV and 289.15 eV are characteristic of C-N, C-O-C, N-C=O and O-C=O groups, respectively. The Si 2p spectrum (Fig. S1c) exhibits a single, symmetric peak centered at 102.13 eV. This binding energy is characteristic of Si-O bonds, a finding that strongly supports the conclusion of the presence of a polysiloxane backbone and is consistent with the analysis of the Si-O-Si component from the O 1s spectrum. The N 1s spectrum (Fig. S1d) further corroborates the presence of nitrogen-containing functional groups. Its signal peak at 399.87 eV is definitively assigned to the C-N chemical environment. This observation is mutually supportive of the urethane (N-C=O) functional group identified in the O 1s

spectrum.

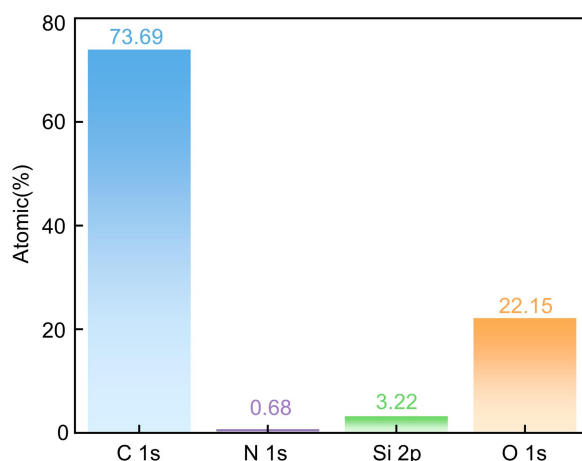


Fig. S2 XPS quantitative elemental analysis of the hydrophobic resin surface. The atomic percentages of the four primary elements on the hydrophobic resin surface were determined using X-ray photoelectron spectroscopy (XPS) to be C (73.69%), O (22.15%), Si (3.22%), and N (0.68%). These quantitative results were obtained by calculating the peak areas from the XPS survey spectrum.

To quantitatively determine the surface elemental composition of the hydrophobic resin (Si-PUA), X-ray photoelectron spectroscopy (XPS) analysis was performed. Fig. S2 presents the atomic percentages of the four primary elements identified on the resin surface. These quantitative results were derived by calculating the integrated peak areas from the XPS survey spectrum. The analysis confirms that the surface is predominantly composed of carbon (C1s) at 73.69% and oxygen (O1s) at 22.15%. Critically, the presence of silicon (Si2p) was detected at 3.22%, along with nitrogen (N1s) at 0.68%. This elemental composition, particularly the detection of C, O, N, and Si, is fully consistent with the chemical structure of the silicone-modified polyurethane acrylate material used in this study. The significant presence of silicon on the surface is a key factor contributing to the material's observed hydrophobic properties.

Supplementary Note 2: DSC of hydrophobic resin

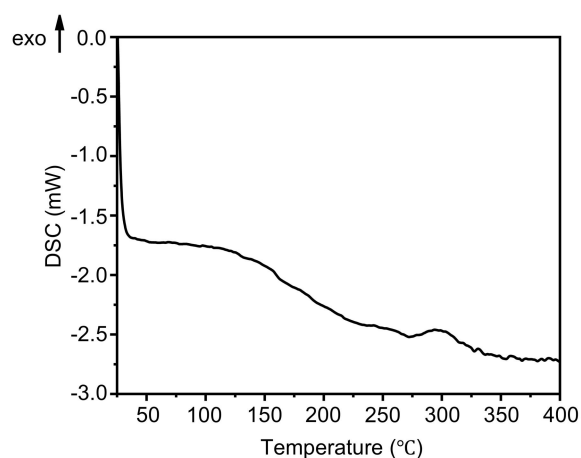


Fig. S3 Differential Scanning Calorimetry analysis of the hydrophobic resin. The DSC curve illustrating the heat flow as a function of temperature. The plot features a broad, step-like decrease in heat flow, which is indicative of a wide glass transition temperature range.

The thermal stability of the resin, whose chemical structure was previously confirmed by XPS, was evaluated using Differential Scanning Calorimetry (DSC) Analysis (Fig. S3). The X-axis represents temperature (°C), while the Y-axis represents heat flow. The heat flow curve in the figure exhibits a wide step-like change in the temperature range of approximately 30°C to 170°C, which manifests as a broad glass transition temperature range for the material, a typical characteristic of the glass transition in amorphous polymers. This broad range suggests sluggish chain segment motion.

Supplementary Note 3: Nanoimprint of high-resolution patterns.

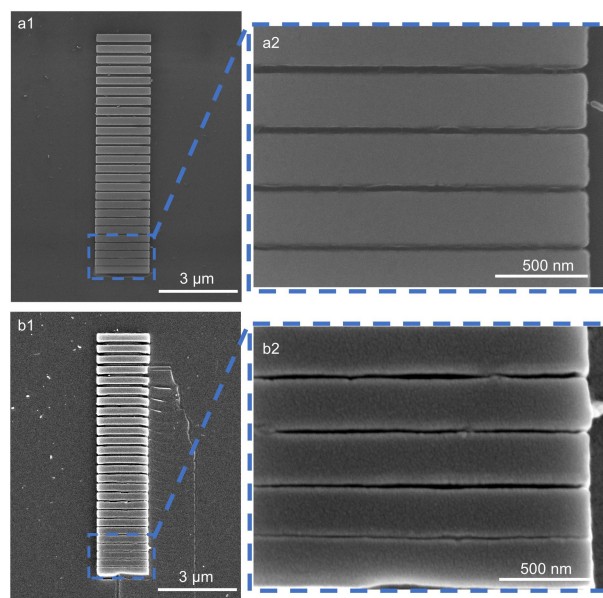


Fig. S4 SEM images of the large-area nanoimprint structures. (a)-(b) SEM images of nanostructures within different rectangular regions of the master stamp and the nanoimprinted sample, respectively.

To evaluate the reliability and structural replication accuracy of NIL technology, SEM characterization was performed on the fabricated regular different rectangular nanopillar arrays. As shown in Fig. S4, the nanostructures of the nanoimprinted samples exhibit distinct rectangular edge profiles, regular columnar arrangements, and an absence of significant collapse when compared to the master stamp used for nanoimprint. Statistical analysis across various sampling regions indicates that deviations in the critical dimensions of the nanopillars remain at a consistently low level. This high-fidelity fabrication capability at the nanoscale ensures excellent structural control and process robustness when processing large-area, high-resolution regular patterns, providing support for subsequent large-scale, high-quality nanostructure integration fabrication.

Besides precise control of planar dimensions, the consistency of structural height is crucial for phase accumulation in metalens. Fig. S5 provides SEM images of the nanopillar array from a tilted perspective. Observation of different regions reveals that the nanopillars not only exhibit verticality but also height consistency. These results effectively demonstrate that this process possesses stable structural control capabilities when handling large-area functional devices.

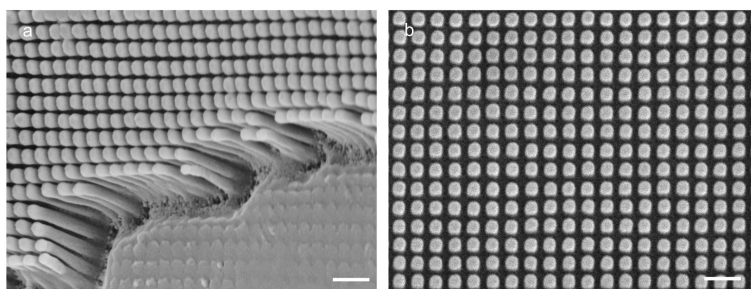


Fig. S5 SEM images of the nanocylinder array. (a)-(b) The height consistency of the nanocylinders in different sampling areas.
Scale bar, 500 nm.

Supplementary Note 4: Repeatability test of stamp.

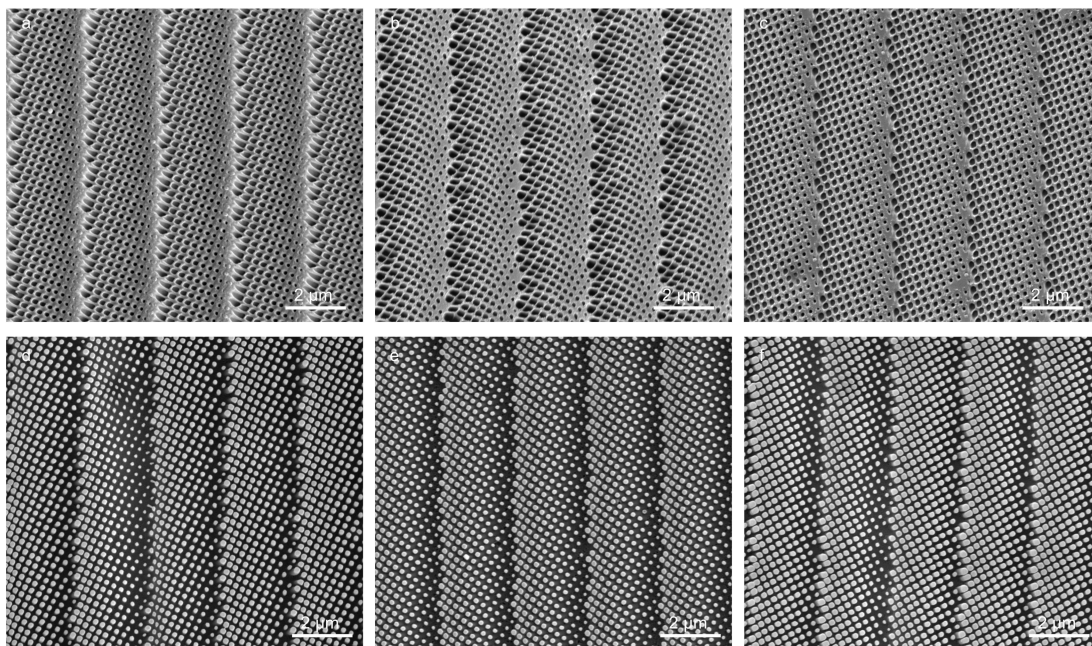


Fig. S6 Repeatability test of stamp. (a)-(c) SEM images of secondary templates replicated from a master template after 10, 15, and 20 imprinting times, respectively. (d)-(f) The final resin structures obtained after the corresponding number of imprinting times (10, 15, and 20), respectively.

The lifetime of the master template and the reliability of the fabrication process were evaluated by repeatedly replicating identical structures [1]. Specifically, we tested the secondary stamps and samples produced during the imprinting process. To create the secondary stamp, the master stamp pattern was transferred to TOPAS at 100°C under 5 bar of pressure, and the final nanostructures were fabricated using a UV-curing process at 365 nm (4 W) for 1 minute under 2 bar of pressure. Figs. S6a-S6c depict the surface morphologies of the secondary molds obtained by replication after the master template had completed 10, 15, and 20 imprinting cycles, respectively. Figs. S6d-S6f correspondingly display the final resin nanostructures fabricated using these molds. Through a comparative analysis of the SEM images, we demonstrated that the master template can be used over 20 times, thereby also validating the reliability of this process for the large-area, continuous production of metasurface devices.

Supplementary Note 5: Measured complex refractive index of hydrophobic resin

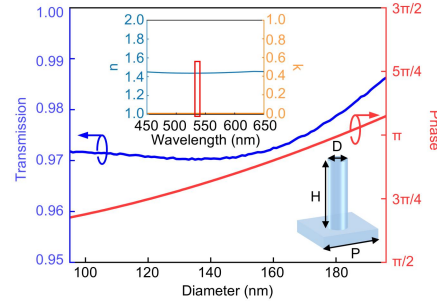


Fig. S7 Simulation of the optical properties of meta-atom. The graph displays the simulation results for the transmittance (blue curve) and phase (red curve) with different nanopillar diameters. The upper inset shows the refractive indices at different wavelengths. The lower right inset is a schematic of the meta-atom structure.

Ellipsometry was utilized to analyze the optical properties of the hydrophobic material, in the visible wavelength range of 450 nm to 650 nm, the real part of resin's refractive index of 1.43 was recorded, and the extinction coefficient k approached 0, as shown in the upper inset of Fig. S7. It exhibiting low optical absorption and high optical transmission characteristics, which is consistent with the results of the highly transparent metalens presented in Fig. 4a. The designed meta-atoms depicted in the inset of Fig. S7 have diameters (D) ranging from 90 nm to 196 nm, a period (P) of 260 nm, and a height (H) of 730 nm. The simulation results about the optical properties of the nanostructures composed on metalens are summarized in Fig. S7. The transmission curve (blue curve) shows that this hydrophobic resin exhibits a high optical transmittance exceeding 95%, which proves it is suitable for use as optical elements. Furthermore, COMSOL simulations (red curve) indicate that by precisely controlling the nanopillar diameter, it is possible to modulate the phase of the transmitted light.

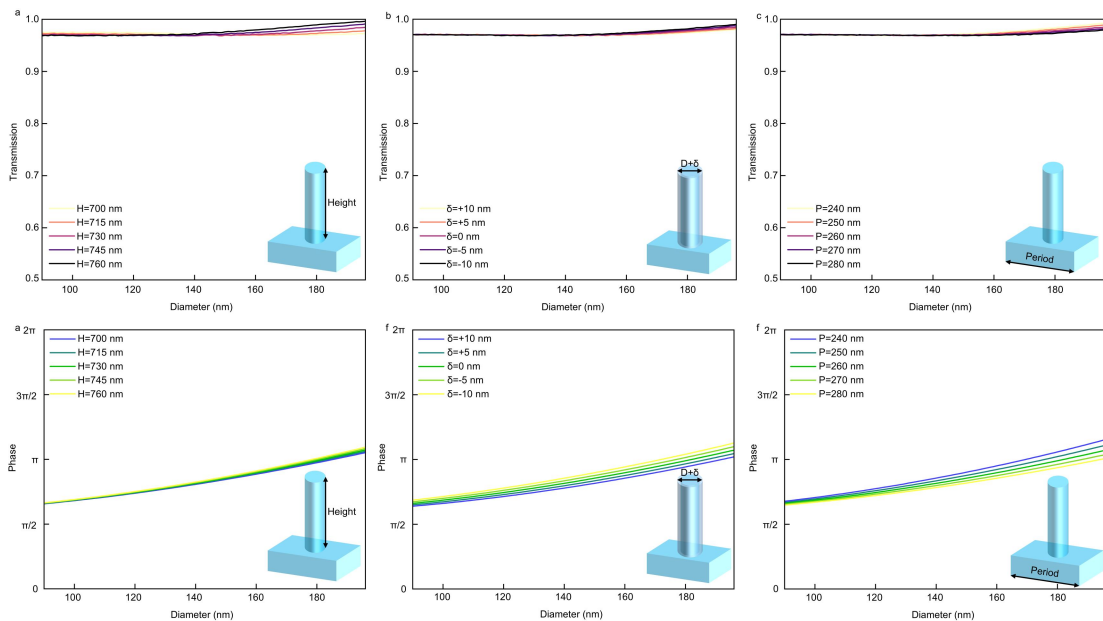


Fig. S8 Simulation analysis of the influence of nanopillar manufacturing errors on transmittance and phase distribution. (a)-(c) The effects of height deviation H , diameter deviation δ , and period deviation P on the device's transmission, respectively. (d)-(f) The effects of the above three parameter deviations on the device's phase profile, respectively.

To assess the specific impact of potential manufacturing deviations during large-area NIL on device performance, a physical model was established using COMSOL, and systematic parameter scanning simulations were performed on key geometric parameters. Considering the minute morphological fluctuations observed in the experiments, a more stringent simulation range was adopted, expanding the maximum deviation range of height, diameter, and period to $\pm 30\text{ nm}$ to simulate device performance under extreme manufacturing conditions. As shown in Figs. S8a-S8c, the transmittance curves of the device exhibit small amplitude fluctuations with parameter variations. Regarding phase modulation characteristics (Figs. S8d-S8f), although the shift in geometric parameters caused a slight drift in the phase, the trend of phase variation with nanopillar diameter remained unchanged. Simulation results indicate that even with slight manufacturing deviations, the optical response of the device still meets expectations.

Supplementary Note 6: Nanoimprint transfer stamp and samples.

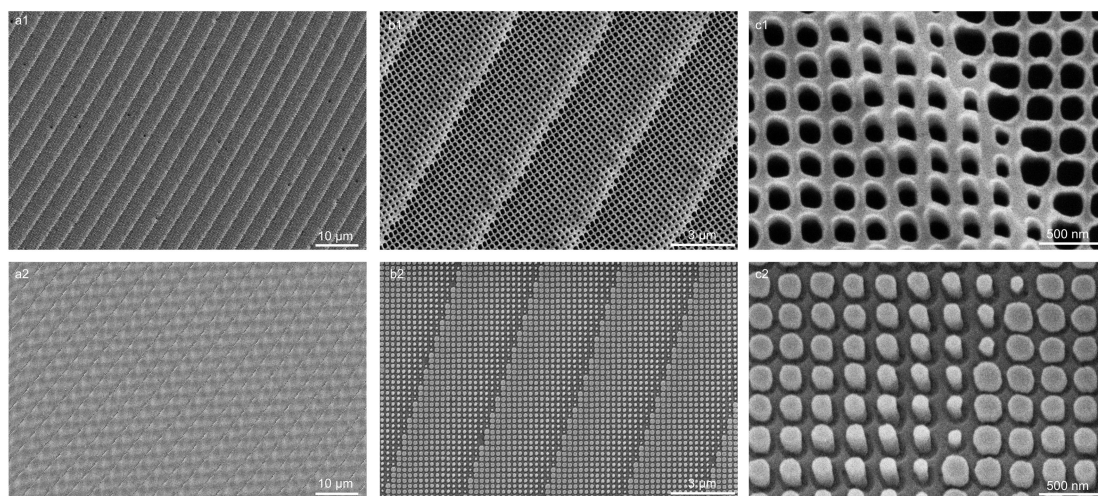


Fig. S9 SEM characterization of the metalens fabrication process. (a1)-(c1) SEM images of the TOPAS secondary stamp at different magnifications, showing an array of circular nano-holes. (a2)-(c2) Corresponding SEM images of the final hydrophobic resin sample, showing the replicated array of circular nano-pillars.

Fig. S9 provides SEM micrographs that demonstrate the pattern transfer fidelity of the NIL process used for metalens fabrication. The top row (Figs. S9a1-S9c1) shows the morphology of the intermediate TOPAS secondary stamp, which was prepared by hot embossing. Figures S8a1 and S8b1 are low- and medium-magnification views, respectively, showing the large-area layout of the patterned arrays. Fig. S9c1 is a high-magnification view that shows the circular nano-hole structures on the TOPAS. The bottom row (Figs. S9a2-S9c2) correspondingly shows the SEM images of the final metalens sample replicated from the TOPAS stamp. These results demonstrate that this fabrication process possesses excellent robustness and high resolution.

Supplementary Note 7: Analysis of hydrophilic and hydrophobic metalenses.

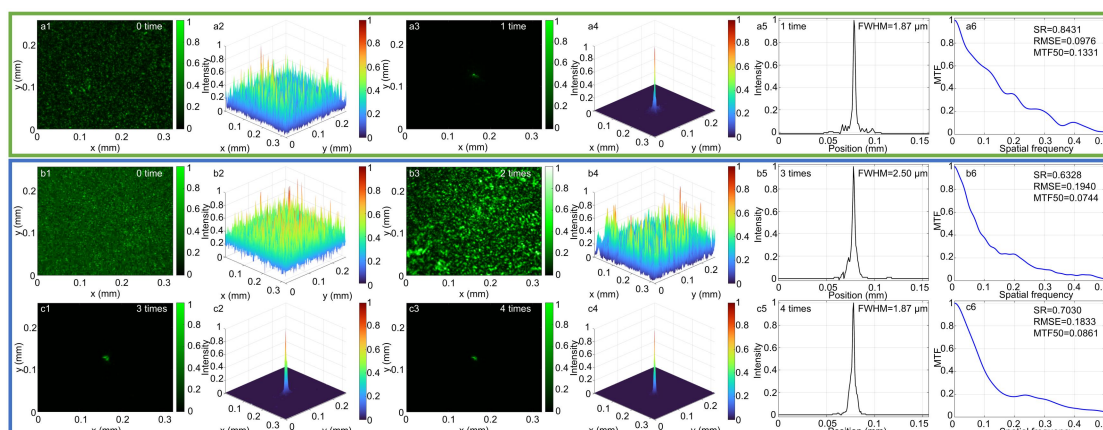


Fig. S10 A comparison of the optical performance of hydrophobic and hydrophilic metalenses in contamination and cleaning tests. (a) Focal plane intensity distributions and optical parameters (a5, a6) of the hydrophobic metalens in the contaminated state (a1, a2) and after 1 cleaning cycle (a3, a4). (b) Performance of the hydrophilic metalens in the contaminated state (b1, b2) and after 2 cleaning cycles (b3, b4). (c) Focal plane intensity of the hydrophilic metalens after 3 (c1, c2) and 4 (c3, c4) cleaning cycles, along with the corresponding optical parameters (b5, b6, c5, c6).

To verify the enhancement of anti-fouling and self-cleaning capabilities provided by the hydrophobic design, the performance recovery processes of hydrophobic and hydrophilic metalenses were compared under identical simulated contamination conditions. Experimental results (Fig. S10) show that the hydrophobic metalens achieves effective restoration of its focusing performance after only one cleaning cycle following contaminant adhesion (Fig. S10a). The central intensity of the focal spot recovers significantly, and the quantitative evaluation metrics remain stable, demonstrating low adhesion between the contaminants and the hydrophobic nanostructures. In contrast, the hydrophilic metalens exhibits notable difficulty in cleaning under the same contamination levels (Figs. S10b-S10c). After two consecutive cleaning cycles, the hydrophilic metalens remains unable to form a clear focal center due to scattering interference from residual surface contaminants (Figs. S10b3-S10b4). Its focusing capability gradually recovers only as the number of cleaning cycles increases to three and four (Figs. S10c1-S10c4). Quantitative analysis indicates that parameters such as the Strehl ratio (SR) and full width at half maximum (FWHM) of the hydrophilic metalens return to detectable levels only after four cleaning cycles. This comparative experiment directly reflects the critical role of the hydrophobic characteristics of the metalens in reducing cleaning frequency and maintaining the optical lifespan of the device. The hydrophobic design not only mitigates the risk of contaminant coverage on the nanostructures but also ensures that the device maintains optical fidelity through simpler maintenance procedures in complex environmental applications.

Supplementary Note 8: Long-term reliability test of hydrophobic metalens.

To evaluate the stability of the hydrophobic metalens in practical application environments, cyclic tests were conducted under four stress conditions, including ultraviolet (UV) exposure, thermal cycling, chemical solvent immersion, and contamination-cleaning. In the characterization of optical focusing performance, a comparison of the three-dimensional light field distributions before and after cycling reveals the physical stability of the device. The longitudinal light field evolution curves along the propagation direction (Figs. S11-S14) show that after 50 cycles of UV irradiation (Fig. S11) or thermal cycling (Fig. S12), the spatial coordinates of the focal spot remain constant, with no focal shift induced by UV aging or thermal stress observed. The two-dimensional intensity maps (x2 series) and corresponding 3D intensity profiles (x3 series) at the focal plane confirm that the energy distribution remains concentrated. In the imaging verification (x2 insets), the resolution quality of the target characters shows no significant change after testing, maintaining clear imaging performance.

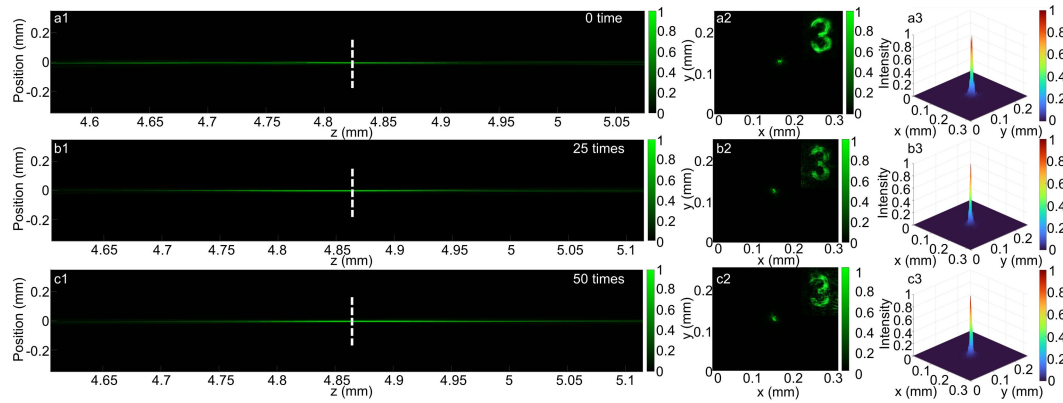


Fig. S11 Optical performance of hydrophobic metalens after UV exposure test. (a1-a3) Performance of the original device. (b1)-(b3) and (c1)-(c3) Performance after 25 and 50 UV exposure cycles, respectively. UV exposure test: 50 cycles of aging, 10 minutes of irradiation time.

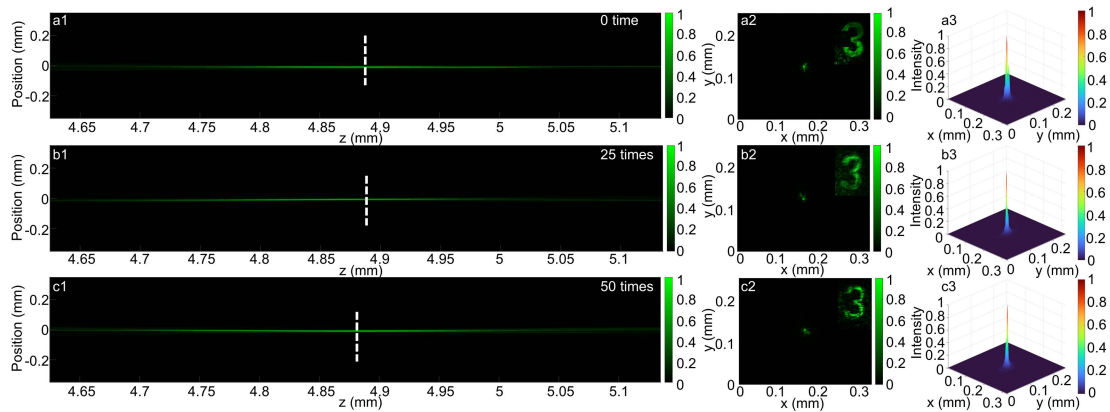


Fig. S12 Optical performance of hydrophobic metalens after multiple thermal cycling treatments. (a1-a3) Performance of the original device. (b1-b3) and (c1-c3) Performance after 25 and 50 thermal cycles, respectively. Thermal cycling experiment: 50

cycles, each cycle held at 80°C for 10 minutes.

The characterization results for chemical and stability similarly record the device's performance. Following immersion in ethanol (Fig. S13), the nanostructures exhibit no swelling or detachment, and the focal spot distribution shows slight fluctuations compared to the initial state. In the contamination-cleaning cycles simulating practical usage (Fig. S14), the device maintains a regular focal spot morphology after 50 repetitions of contamination and cleaning, with no significant degradation in imaging contrast.

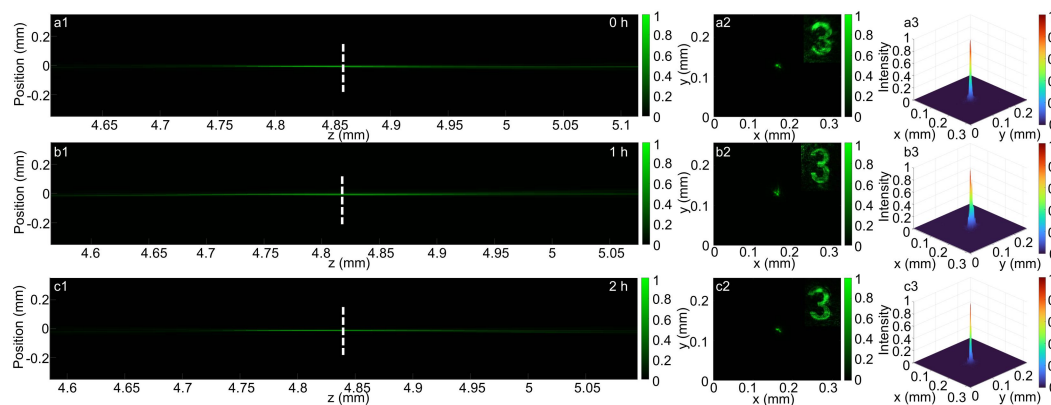


Fig. S13 Optical performance of hydrophobic metalens after immersion in organic solvents. (a1-a3) Performance of the original device. (b1-b3) and (c1-c3) Performance after immersion in organic solvents for 1 h and 2 h, respectively.

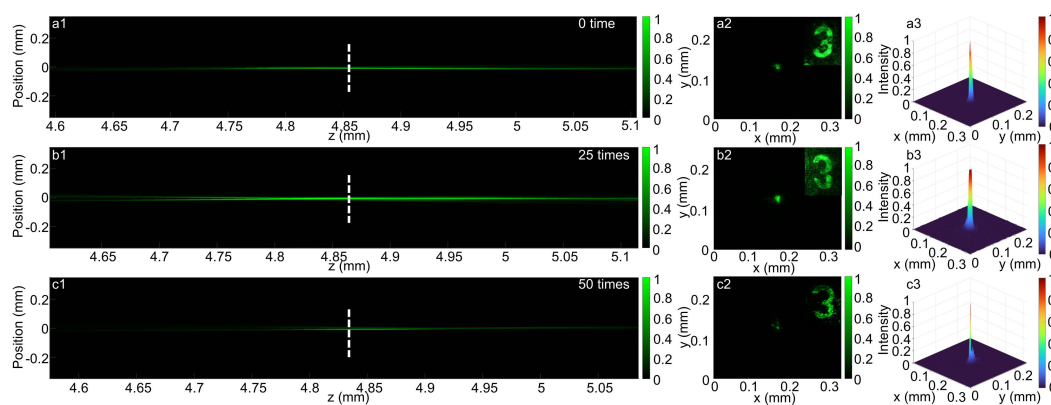


Fig. S14 Optical performance of hydrophobic metalens after multiple contamination-cleaning processes. (a1-a3) Performance of the original device. (b1-b3) and (c1-c3) Performance after 25 and 50 contamination-cleaning processes, respectively.

Analysis of the measured one-dimensional intensity distribution curves indicates that the full width at half maximum (FWHM) fluctuations across the four cyclic tests remain small (Figs. S15a, S15c, S15e, S15g), consistent with design expectations. Furthermore, the modulation transfer function (MTF) curves trend closely with those of the original device (Figs. S15b, S15d, S15f, S15h). Key evaluation metrics, including the Strehl ratio (SR), RMSE, and MTF50, had relatively minor changes before and after the 50 cycles, suggesting that environmental damage has a slight impact on optical transfer efficiency.

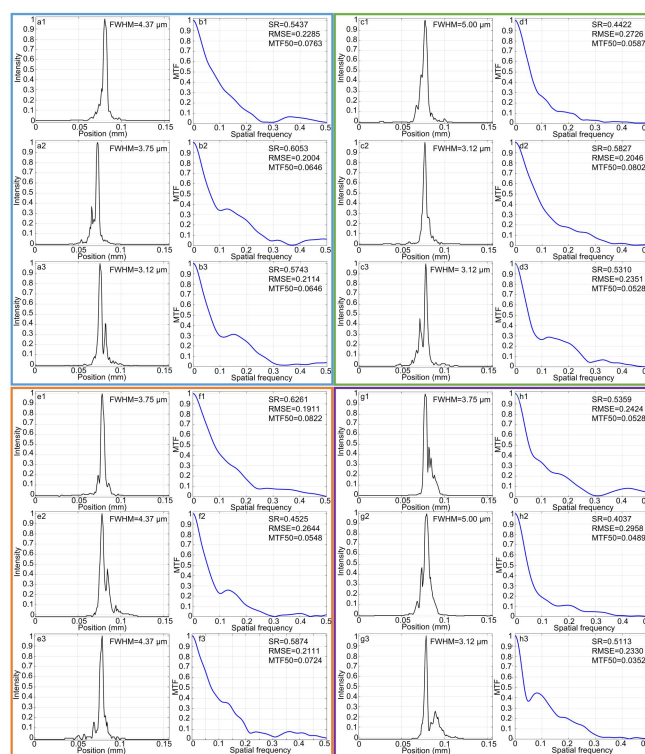


Fig. S15 Parameters of the hydrophobic metalens under different tests. The left column shows the one-dimensional intensity distribution curve of the focal spot, including FWHM, and the right column shows the corresponding modulation transfer function curve and related indicators (SR, RMSE, MTF50). (a)-(b) Blue boxes: UV exposure test. (c)-(d) Green boxes: Thermal cycling test. (e)-(f) Orange boxes: Solvent immersion test. (g)-(h) Purple boxes: Contamination-cleaning test.

Additionally, the hydrophobic characteristics of the device surface were verified through static water contact angle measurements (Fig. S16). Experimental data show that after undergoing UV aging, thermal cycling, solvent immersion, and repeated contamination-cleaning, the values of the samples remain above 100° (Figs. S16a-S16d). These results indicate that the intrinsic hydrophobic properties of the device do not undergo significant degradation when subjected to environmental change. The experimental evidence demonstrates that the hydrophobic metalens maintains its key optical indicators and surface wetting characteristics under environmental interference, providing support for its application in fields such as outdoor monitoring and industrial sensing.

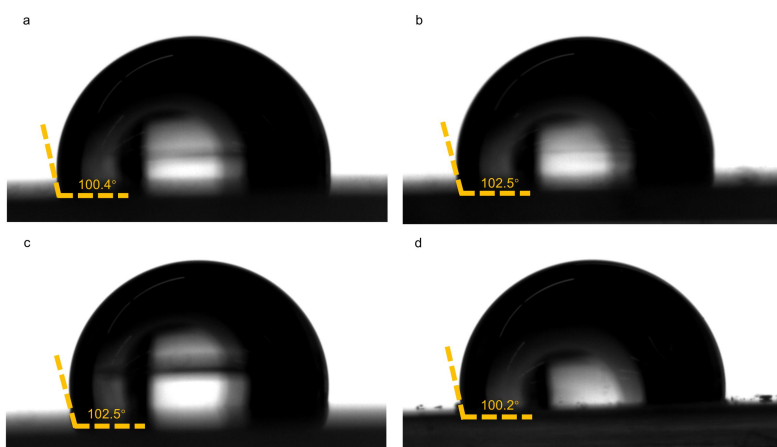


Fig. S16 The static water contact angle measurements of the hydrophobic metalens under different tests. (a) UV exposure test. (b) Thermal cycling test. (c) Solvent immersion test. (d) Contamination-cleaning test.

Reference

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