

Supplementary Information

Photonic nanojet steering for parallel super-resolution laser writing

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MIE SIMULATIONS

When electromagnetic waves encounter particles with dimensions comparable to the wavelength of the incident light, the scattering behavior becomes highly complex to be accurately described by the Rayleigh approximation. In these scenarios, Mie theory offers an exact analytical solution to Maxwell's equations for light scattering by perfect spherical particles^{1,2}. This solution incorporates key parameters such as the particle size, the wavelength of the incident light, and the refractive indices of both the particle and its surrounding medium.

This study considers the scattering of a monochromatic plane wave of wavelength λ by a spherical particle of radius a , immersed in a homogeneous medium, where both the particle and its environment are non-magnetic. ϵ_m and ϵ_p are the dielectric permittivities of the surrounding medium and particle, respectively. By solving Maxwell's equations with the appropriate boundary conditions at the particle's surface, the resulting scattered field can be expressed in spherical coordinates (r, θ, ϕ) as follows²,

$$E_r = \frac{\cos\phi}{(k_m r)^2} \sum_{l=1}^{\infty} l(l+1) {}^e B_l \zeta_l(k_m r) P_l^{(1)}(\cos\theta) \quad (1)$$

$$E_\theta = -\frac{\cos\phi}{k_m r} \sum_{l=1}^{\infty} \left[{}^e B_l \zeta_l'(k_m r) P_l^{(1)'}(\cos\theta) \sin\theta - i {}^m B_l \zeta_l(k_m r) \frac{P_l^{(1)}(\cos\theta)}{\sin\theta} \right] \quad (2)$$

$$E_\phi = -\frac{\sin\phi}{k_m r} \sum_{l=1}^{\infty} \left[{}^e B_l \zeta_l'(k_m r) \frac{P_l^{(1)}(\cos\theta)}{\sin\theta} - i {}^m B_l \zeta_l(k_m r) P_l^{(1)'}(\cos\theta) \sin\theta \right] \quad (3)$$

where $P_l^{(m)}(x)$ denote associated Legendre polynomials, while $\zeta_l(x)$ and $\zeta_l'(x)$ are Riccati-Hankel function and its derivative; $k_p = 2\pi\sqrt{\epsilon_p}/\lambda$ and $k_m = 2\pi\sqrt{\epsilon_m}/\lambda$ are wave vectors in sphere and surrounding medium. The coefficients ${}^e B_l$ and ${}^m B_l$ are given by explicit expressions,

$${}^e B_l = i^{l+1} \frac{2l+1}{l(l+1)} \frac{q_p \psi_l'(q_m) \psi_l(q_p) - q_m \psi_l(q_m) \psi_l'(q_p)}{q_p \zeta_l'(q_m) \psi_l(q_p) - q_m \psi_l'(q_p) \zeta_l(q_m)} \quad (4)$$

$${}^m B_l = i^{l+1} \frac{2l+1}{l(l+1)} \frac{q_p \psi_l'(q_p) \psi_l(q_m) - q_m \psi_l(q_p) \psi_l'(q_m)}{q_p \psi_l'(q_p) \zeta_l(q_m) - q_m \psi_l(q_p) \zeta_l'(q_m)} \quad (5)$$

where $q_p = k_p a$ and $q_m = k_m a$ are size parameters in the sphere and surrounding media, respectively. Moreover, $\psi_l(x)$ and $\psi_l'(x)$ denote Riccati-Bessel function and its derivative. The series expansion, used to represent the scattered field, converges efficiently when the summation is truncated at a maximum multipole order given by $l_{max} \approx (q + 4q^{1/3} + 1)$ with $q = 2\pi\sqrt{\epsilon_m}a/\lambda$, the corresponding size parameter³.

Finally, the total electric field outside the sphere is quantified by the summation of the incident and scattered electric fields, that is

$$E_{total} = E_{incident} + E_{scattered} \quad (6)$$

Here, we first employ the analytical Mie solution to investigate the formation of photonic nanojets (PNJs) by interaction with a dielectric microsphere. The general characteristics of PNJs are verified before using numerical Maxwell solvers (FDTD simulations) to study how these characteristics vary for different incidence angles in the presence of a substrate supporting the microsphere. A schematic illustration of nanojet formation under inclined irradiation conditions is shown in Fig. S1a.

Fig. S1b shows the simulated intensity distributions in the xz -plane for varying angles of incidence. As clearly seen from the figure, the nanojet progressively shifts away from the original focal spot according to a rigorous rotation of the produced intensity profiles. With increasing angle, the peak intensity on the reference plane (surface position in the presence of a substrate) gradually decreases as the separation between the sphere and the reference plane along the PNJ path increases.

To quantitatively visualize the effect of angular variation, line profiles along the x -axis at the center of the transverse intensity distribution on the reference plane are plotted in Fig. S1c for the corresponding incident angles. Each line depicts the peak intensity experienced by the surface. The monotonic decrease of the peak intensity while the spot size increases supports the light delocalization due to the divergence of the nanojet on the reference plane. The ellipticity variation of the PNJ spot size is derived from these simulations and presented in Fig. S1d. Interestingly, one can note an initial ellipticity along the polarization direction that decreases with PNJ propagation before flipping analogously to an astigmatic focusing situation.

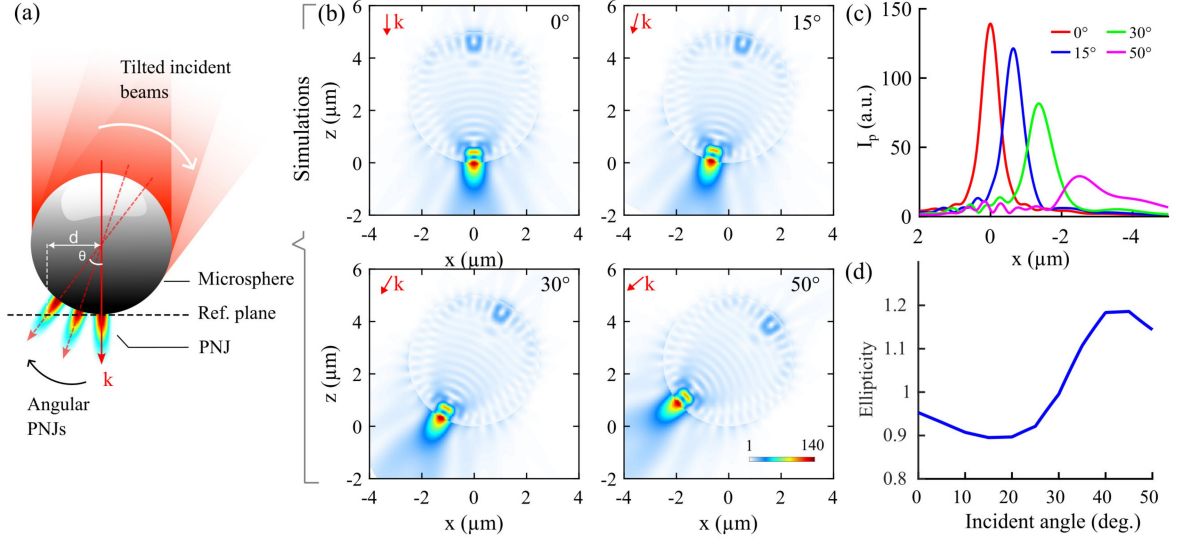


FIG. S1. (a) Schematic representation of PNJ formation through a dielectric microsphere under oblique illumination. The incident light wavevector (k) indicates the direction of propagation, and the white arrow denotes the direction of increasing angle of incidence (θ) relative to the reference plane (normal to propagation). d represents the expected lateral displacement of the peak intensity on the reference plane. (b) Cross-sectional view (xz -plane) illustrating the angular evolution of the PNJ intensity distributions, highlighting the angular dependence of nanojet orientation. (c) Transverse intensity profiles along x of the PNJ for different incident angles on the reference plane, showing a monotonic decrease of peak intensity relative to the normal incidence conditions. (d) Variation of the ellipticity of the PNJ on the reference plane with increasing angle of incidence.

FDTD SIMULATIONS

FDTD simulations using ANSYS Lumerical software are performed with Bloch periodic boundary conditions in the x and y directions. Perfectly matched layers (PMLs) are applied along the z direction to absorb the reflected and transmitted fields. The spatial grid resolution is set to 9 nm. Varying incidence illumination is systematically implemented using a plane-wave source in the simulations.

EFFECT OF OPTICAL IMAGING RESOLUTION ON NANOJET RECONSTRUCTION

The experimentally reconstructed profile shown in Fig. 1d (main text) confirms the formation of photonic nanojets. However, the finite optical resolution of the microscopy system broadens the reconstructed intensity profile, leading to an apparent PNJ FWHM of approximately $0.9 \mu\text{m}$. To account for this resolution effect, the FDTD-simulated intensity distribution (Fig. S2a) is convolved with a Gaussian point-spread function corresponding to the lateral resolution of the experimental imaging system. After applying this filtering, the simulated spot size becomes comparable to the experimentally reconstructed value, as shown by the intensity distribution and the line profiles in Fig. S2b-d, confirming a good correspondence between simulations and measurements.

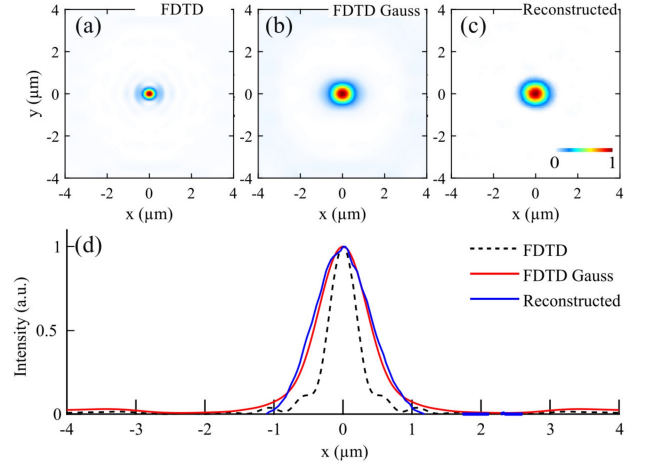


FIG. S2. Transverse intensity maps of the photonic nanojet formed in air through a $5 \mu\text{m}$ diameter microsphere; (a) FDTD simulation, (b) FDTD simulation after applying a Gaussian filter, and (c) experimental reconstruction; (d) Corresponding intensity line profiles extracted along the x -direction from panels (a-c).

RADIAL SHIFTS

The SEM images in Fig. S3 show how the PNJ-induced modification sites are systematically displaced from the microsphere centers under oblique illumination. Although the microspheres are removed prior to SEM imaging, their original center positions remain identifiable from weak residual

circular footprints at the sphere-substrate contact regions. The dashed yellow hexagon marks the close-packed lattice defined by the microsphere centers, serving as a reference for the geometrical arrangement of the monolayer. In contrast, the solid yellow hexagon is drawn through the experimentally observed modification sites, which correspond to the substrate-plane locations of the PNJ maxima. The offset between the dashed and solid hexagons therefore directly represents the radial shift of the PNJ maximum relative to the sphere center for each incidence angle. Notably, this displacement increases at larger incidence angles, indicating a progressively stronger deviation of the nanojet maximum from the sphere-centered position. We can also note different shift orientations despite an inclined orientation along the same direction. This is due to the systematic presentation of the same hexagonal pattern (orientation of the observation adjusted), while arrays have random orientations depending on the monolayer domains on the sample.

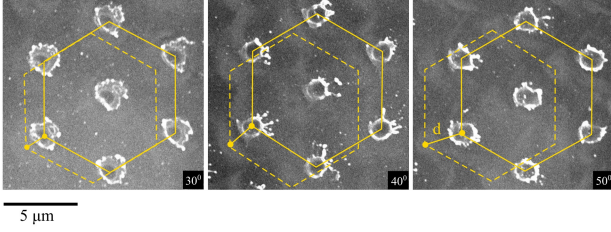


FIG. S3. SEM images showing the progressive lateral displacement (d) of the photonic-nanojet-induced irradiation site from the microsphere center (or sphere contact point with the substrate) using increasing incidence angles up to 50° .

EFFECT OF MONOLAYER OF MICROSPHERES ON PHOTONIC NANOJET FORMATION

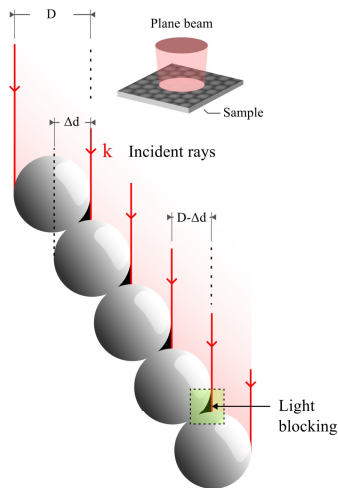


FIG. S4. Schematic illustration of the light blocking effect on a monolayer of microspheres at high-angle oblique incidence.

Under oblique illumination, the microsphere array can modify the effective entrance pupil of the individual microspheres through a light-blocking effect. As illustrated schematically in Fig. S4, part of the incident beam is shadowed by adjacent microspheres, reducing the effective illuminated aperture from D to $D - \Delta d$. This effect becomes increasingly significant as the incident angle increases. The resulting aperture reduction may alter the spatial field distribution entering the microsphere and influence the properties of the generated photonic nanojet.

RAMAN SPECTROSCOPY

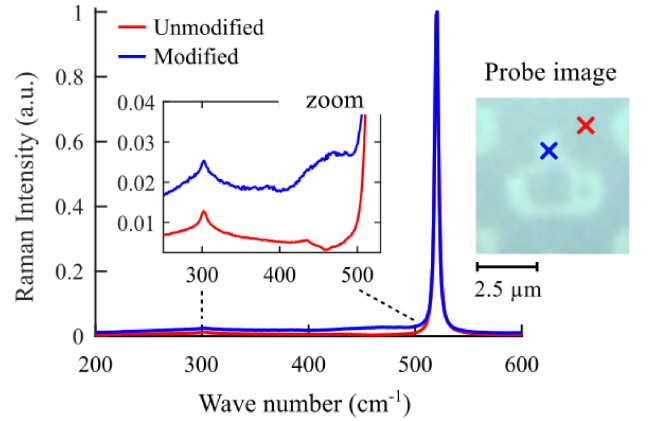


FIG. S5. Raman spectroscopy analysis comparing the unmodified and modified regions of the sample. The inset shows a magnified view of the selected Raman spectral region, while the probe image indicates the Raman measurement positions marked by colored crosses. The red and blue crosses correspond to the pristine Si surface and the processed patterns, respectively.

Raman spectroscopic measurements have been performed using a Renishaw inVia Raman microscope coupled to a Leica DM2500 M microscope in a confocal configuration. All measurements reported in this work are carried out by using a 532 nm diode-pumped solid-state laser (maximum output power up to 500 mW, class 3B). The laser beam focused onto the sample surface exhibits a lateral spot size of approximately 800 nm. The laser power at the sample surface is carefully controlled using neutral density filters to avoid local heating or sample damage. The spectrometer is calibrated prior to acquisition using the first-order Raman band of crystalline silicon at 520.7 cm^{-1} . Spectral acquisition and processing are performed using the Renishaw WiRE software.

SURFACE ROUGHNESS AND TRANSFORMATION MEASUREMENT

For structural characterization of the processed sample shown in Fig. 6c (main text), a lamella has been prepared

by focused ion beam milling (FIB) using HELIOS 5 UC FIB-SEM (Focused Ion Beam–Scanning Electron Microscope) system from ThermoFisher Scientific. The SEM image shown in Fig. S6a represents the top view of the processed sample, while the boxed region indicates the area selected for cross-sectional analysis. Prior to FIB milling, a protective Pt layer is deposited over the selected region to preserve the surface morphology and minimize milling-induced damage. A thin lamella containing the region of interest is then extracted and further thinned for cross-sectional inspection, as shown in Fig. S6b. The shadowgraphy image in Fig. S6c shows a cross-sectional view of the thinned FIB lamella, where the electron-transmission contrast allows the near-surface structure to be visualized. The bright top region corresponds to the protective Pt layer, below which there is a thin modified Si layer with a measured thickness of about 53 nm, followed by the bulk Si substrate. These results indicate that the processing mainly induces a shallow near-surface modification in the Si sample.

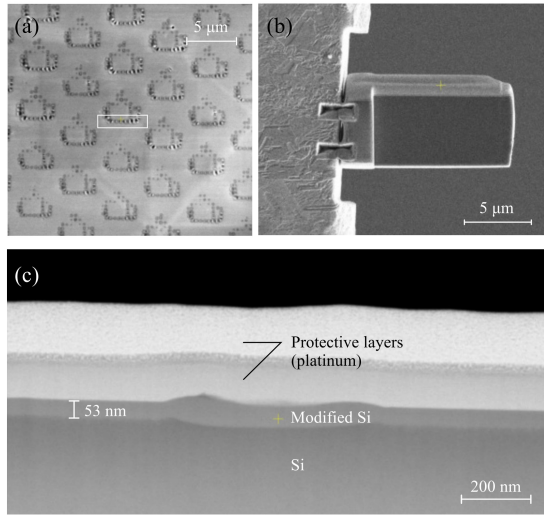


FIG. S6. Structural characterization of the laser-modified silicon surface. (a) Top-view SEM image of the processed Si sample. (b) FIB-prepared lamella extracted from the selected region. (c) FIB shadowgraphy image of the thinned lamella.

FEATURE SIZE MEASUREMENT

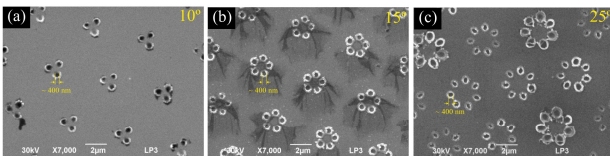


FIG. S7. SEM images of the laser-induced structures formed in the ablation regime at (a) 10°, (b) 15°, and (c) 25° angle of incidence.

To demonstrate the capability of the sphere-assisted laser

processing technique for dense surface structuring in the ablation regime, the sample is processed using different combinations of polar angle, θ , and azimuthal angle, ϕ , while keeping the pulse energy below the microsphere removal threshold. The SEM images show that this approach produces dense, flower-like arrangements of ablated features on the Si surface, enabling multiple closely spaced ablation sites to be generated under the same microsphere. It demonstrates that oblique sphere-assisted illumination can be used to fabricate dense surface patterns without removing the microspheres, even in the ablation regime.

SCATTERING EFFECTS FROM A CLOSE-PACKED MONOLAYER OF MICROSPHERES

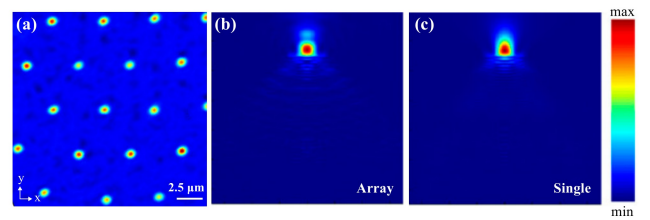


FIG. S8. (a) 2D array of photonic nanojets at the best focal plane; Optical field distributions from 7 μm dielectric microsphere for (b) infinite periodic array and (c) single microsphere. Reproduced from Sun *et al.*, *Nanomaterials* 2026, 16(2), 136. CC BY 4.0.

In contrast to some previous PNJ-based work studying isolated microspheres, our experiments are systematically carried out using a close-packed monolayer of microspheres assembled on a Si substrate. In particular, several reports have preferred to study sparsely distributed microspheres in order to minimize potential scattering or coupling effects from neighbouring particles⁴. However, we would like to mention here that we have not observed any effect causing real limitations with the conditions of our work. In our PNJ reconstructions, we did not observe any noticeable coupling effects affecting the scattering responses of microspheres, as shown in Fig. S8a. This observation is consistent with a recent numerical study by Sun *et al.*⁵, who have demonstrated that even for an infinite periodic array of microspheres, the main PNJ characteristics remain largely unchanged (Fig. S8b-c) and only weak periodic intensity modulations must be expected. In practice, we neglect these modulations in the compensation modeling approach in this work. The experiments support that these are sufficiently small to be ignored in the context of material processing applications.

LIMITATIONS AND ADVANTAGES OF SPHERE-ASSISTED LASER PROCESSING ON MANUFACTURING THROUGHPUT AND SCALABILITY

The key manufacturing metrics, including writing speed and areal processing rate, are strongly dependent on the com-

plexity of the target pattern. For instance, under normal-incidence illumination with 1-mJ pulses, an 8-inch wafer can be processed in approximately 1.8 s using a laser repetition rate of 10 kHz and an illumination area of 0.018 cm^2 to produce periodic single-spot modifications. For more complex patterns, such as the one shown in Fig. 6(c) (in the main manuscript), however, the processing time increases to approximately 1 minute (58 s) for the complete wafer, owing to the need to address multiple angular positions within each unit cell.

In the present implementation, the overall throughput is partly limited by the angular scanning speed of the rotational stages. This limitation is technical rather than fundamental. Replacing the rotational stages with a two-dimensional galvo scanner, while maintaining the same laser repetition rate and illumination area, would substantially reduce the angular addressing time and thereby further increase the processing throughput. Therefore, although the current configuration already enables parallel wafer-scale processing, the use of a faster beam-steering method provides a way toward high-throughput manufacturing.

In comparison with other parallel fabrication methods, SLM-based direct writing can provide high throughput and programmable patterning; however, these systems generally rely on conventional far-field optics and therefore remain diffraction-limited, whereas microsphere-assisted processing can go beyond the diffraction limit because of its near-field characteristics. Mask-based photolithography and nanoimprint lithography can offer very high throughput and excellent resolution. However, their pattern flexibility is limited because each new design typically requires a new mask. In contrast, the proposed microsphere-assisted method provides reconfigurable pattern generation by controlling the angular motions from one irradiation zone to another.

Regarding scalability, flexible patterning at the individual microsphere level has been demonstrated in approaches such as optically trapped microsphere lithography, where a single microsphere acts as a movable near-field lens for arbitrary direct writing. These methods confirm the feasibility of microsphere-level programmability, although they are serial in nature and limited in high-throughput. In the configuration studied in our work, all microspheres within the illuminated region experience the same incident beam condition at a given time. As a result, the proposed method is particularly suitable for high-throughput fabrication of periodic patterns over large areas, while large area processing with fully independent control of each microsphere remains unexplored, to the best of our knowledge.

¹Max Born and Emil Wolf. *Principles of optics: electromagnetic theory of propagation, interference and diffraction of light*. Elsevier, 2013.

²Boris Luk'yanchuk. *Laser cleaning: optical physics, applied physics and materials science*. World Scientific, 2002.

³Peter W Barber and Steven C Hill. *Light scattering by particles: computational methods*. World Scientific, 1990.

⁴Patrick Ferrand, Jérôme Wenger, Alexis Devilez, Martina Pianta, Brian Stout, Nicolas Bonod, Evgueni Popov, and Hervé Rigneault. Direct imaging of photonic nanojets. *Optics express*, 16(10):6930–6940, 2008.

⁵Zehua Sun, Shaobo Ge, Lujun Shen, Junyan Li, Shibo Xu, Jin Zhang, Yingxue Xi, and Weiguo Liu. Geometry-dependent photonic nanojet formation and arrays coupling. *Nanomaterials*, 16(2):136, 2026.