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## Supporting Information

for

# Influence of laser beam profile on morphology and optical properties of silicon nanoparticles formed by laser ablation in liquid

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## Additional experimental results and analysis of the produced Si NPs

## Transmission electron microscopy

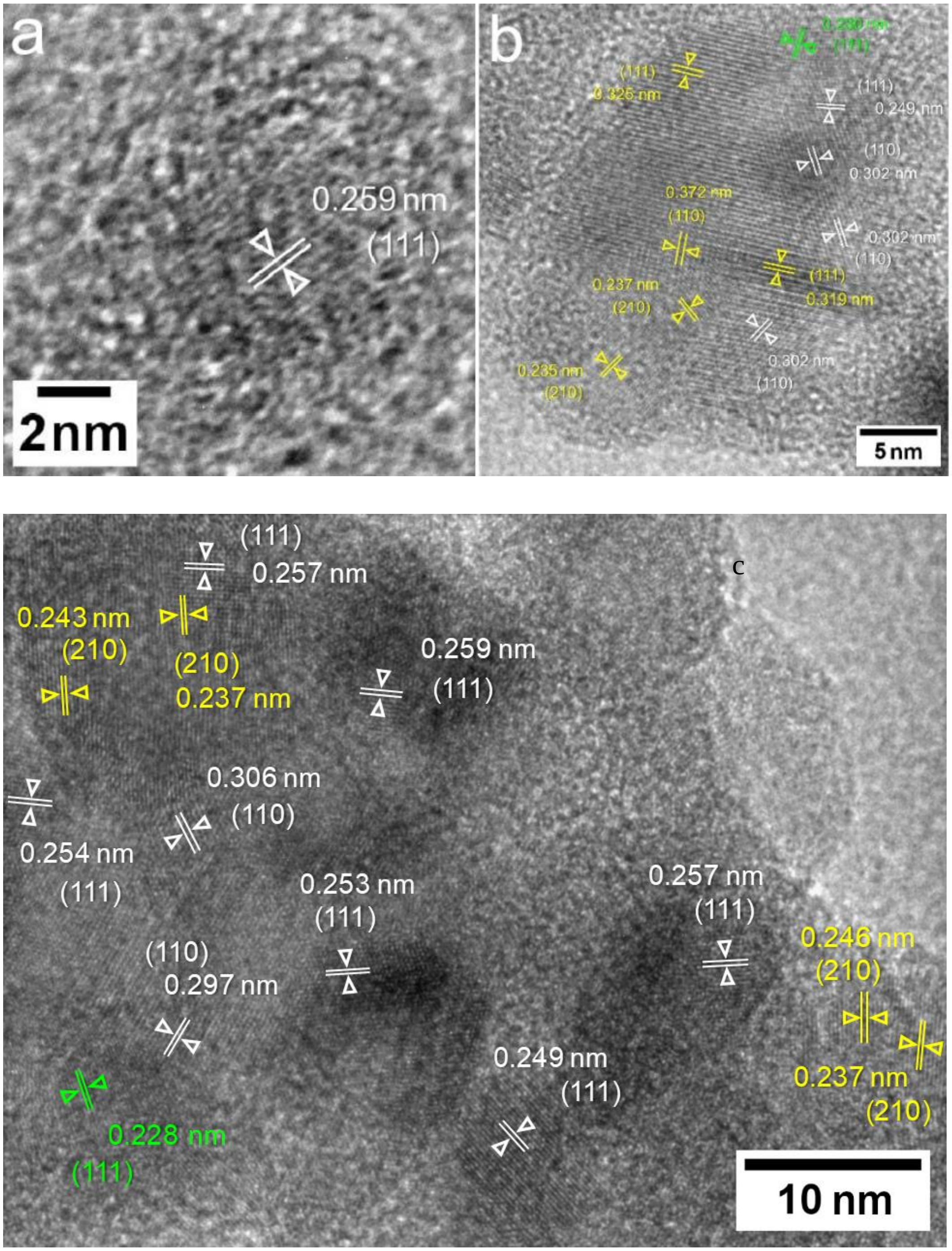


**Figure S1:** HRTEM images of separate particles prepared by laser ablation of Si wafer with the Bessel (a) and annular (b,c) laser beams indicating the interplanar spacings**,** which can be attributed to the planes of cubic Si, cubic SiC**,** and silicon oxide phases.

**Table S1:** Determination of the interplanar spacings.

| Color highlighting | Material | Crystal structure and space group | $d_{measured}$, nm | $d_{calc}$, nm | Miller indices (hkl) | Δ*d*, nm |
|---|---|---|---|---|---|---|
| yellow | Si | cubic *Fd*−3*m* | 0.237 | 0.243 | (210) | ±0.006 |
| | | | 0.246 | 0.243 | (210) | ±0.003 |
| | | | 0.243 | 0.243 | (210) | ±0.000 |
| | | | 0.235 | 0.243 | (210) | ±0.008 |
| | | | 0.319 | 0.314 | (111) | ±0.005 |
| | | | 0.325 | 0.314 | (111) | ±0.011 |
| | | | 0.372 | 0.385 | (110) | ±0.013 |
| green | $SiO_2$ | hexagonal $P3_221$ | 0.228 | 0.224 | (111) | ±0.004 |
| | | | 0.230 | 0.224 | (111) | ±0.006 |
| white | SiC | cubic *F*−43*m* | 0.297 | 0.307 | (110) | ±0.010 |
| | | | 0.302 | 0.307 | (110) | ±0.005 |
| | | | 0.249 | 0.251 | (111) | ±0.002 |
| | | | 0.253 | 0.251 | (111) | ±0.002 |
| | | | 0.254 | 0.251 | (111) | ±0.003 |
| | | | 0.306 | 0.307 | (110) | ±0.001 |
| | | | 0.257 | 0.251 | (111) | ±0.006 |
| | | | 0.259 | 0.251 | (111) | ±0.008 |

**Table S2:** Phase composition of the NPs prepared using different beam profiles.

| Phase composition, Miller indices (hkl) | Beam profile | |
|---|---|---|
| | Bessel | annular |
| Si | (111) | (111), (110), (210) |
| SiC | (111) | (111), (110) |
| $SiO_2$ | — | (111) |

## Size distributions of the NPs

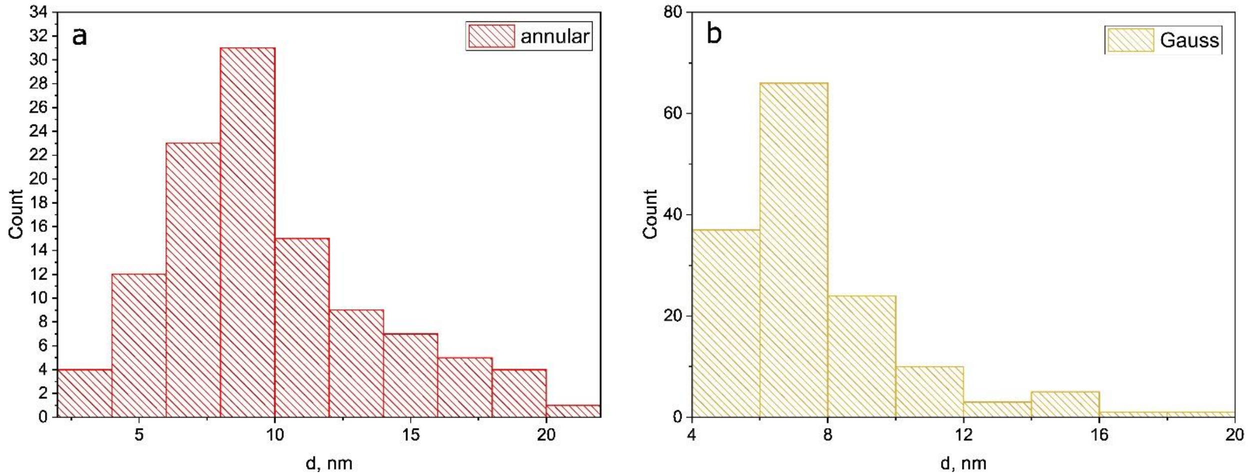


**Figure S2:** Size distribution of the NPs prepared by laser ablation of Si target in ethanol using (a) annular and (b) Gaussian beam profiles.

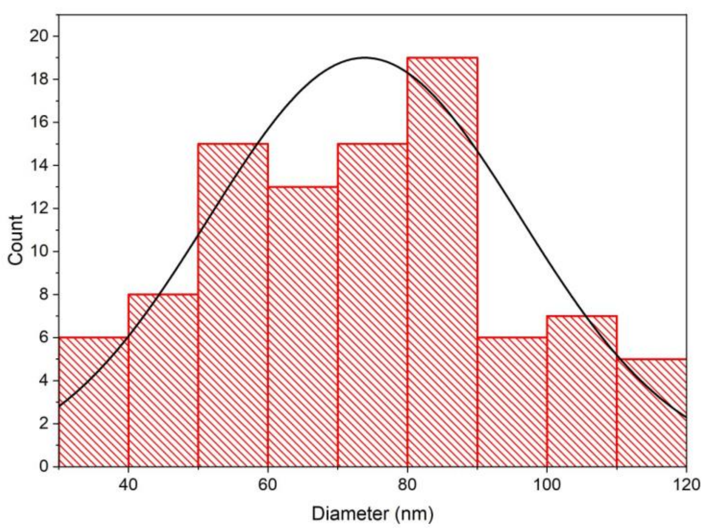


**Figure S3:** Histogram of diameter distribution of nanofilaments prepared by laser ablation of a Si target in ethanol using a Bessel beam profile. The size distribution was determined with the aid of ImageJ software. The distribution was nearly normal (Gaussian) type.

## Scanning electron microscopy of the ablated Si target

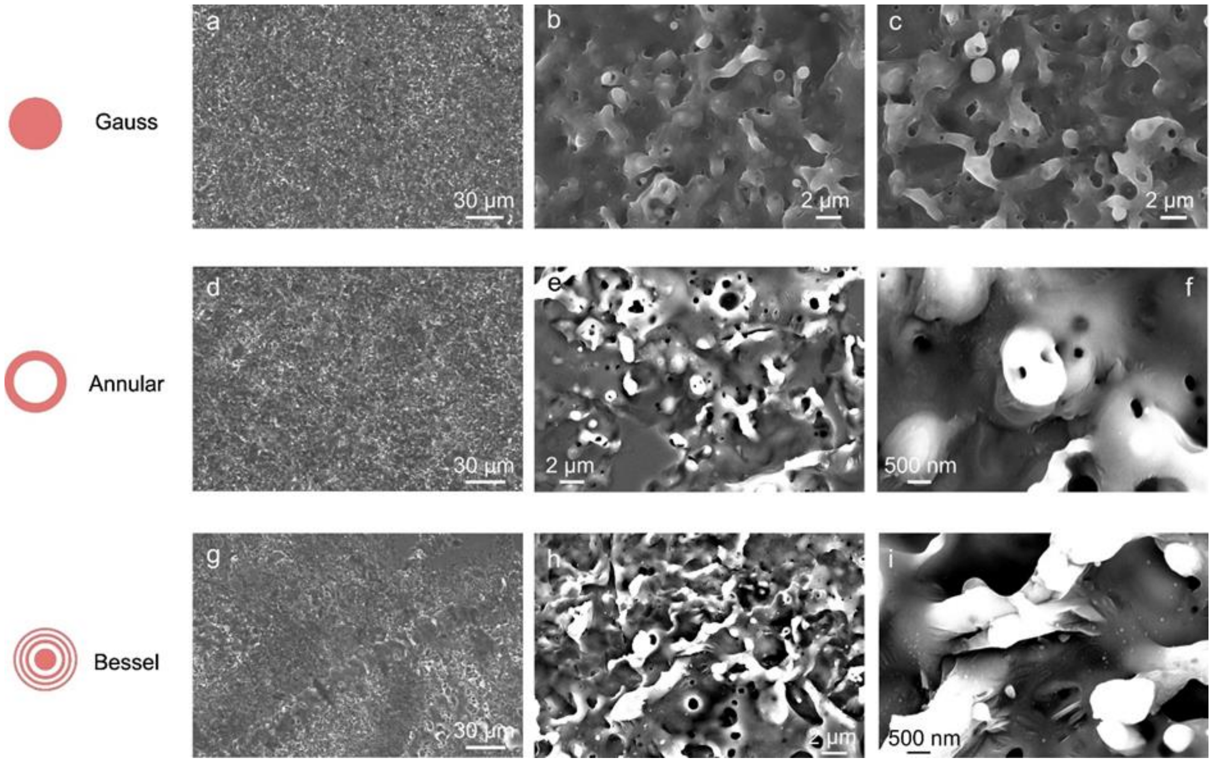


**Figure S4:** SEM characterization of Si targets after laser ablation using (a–c) Gauss, (d–f) annular, and (g–i) Bessel beam profiles. The SEM images are presented with different magnifications.

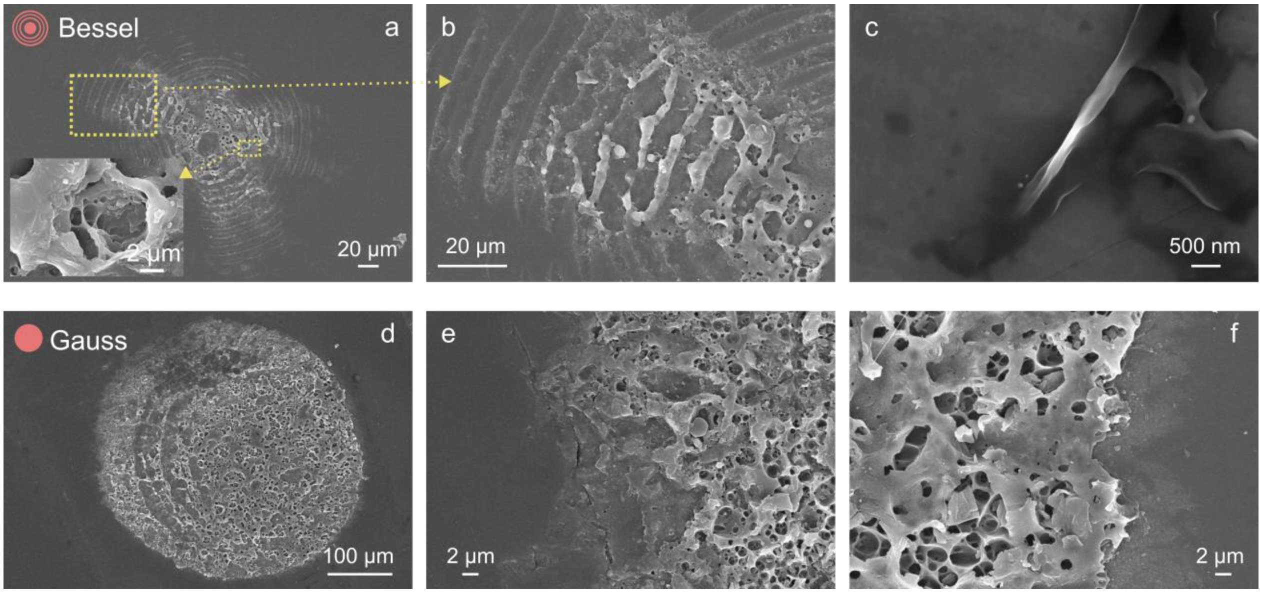


**Figure S5:** SEM images of Si targets exposed to a single laser pulse of (a–c) Bessel and (d–f) Gaussian beams. Panel c shows individual twisted filaments forming on the surface in the area of outer-ring irradiation.

## Raman spectra of Si NPs

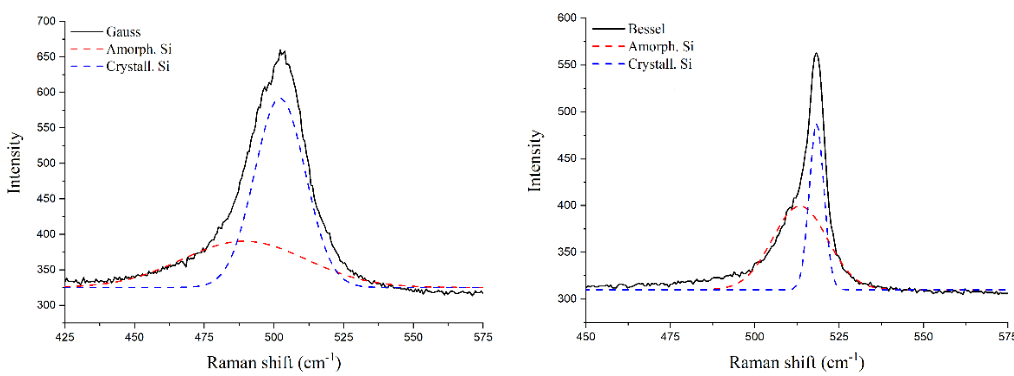


**Figure S6:** Raman spectra deconvolution into Gaussian (crystalline, blue line) and asymmetric (amorphous, red line) components for Si NPs prepared by laser ablation of Si in ethanol using Gauss and Bessel laser beams.

Deconvolution of the Raman spectra in Figure S6 in the range from 400 to 540 $cm^{-1}$ into Gaussian and asymmetric components based on the method described in [1] enables the estimation of the partial volumes of crystalline and amorphous phases in the prepared Si samples. In all cases, the best fit results were obtained using two components. The relative content of the crystalline phases can be estimated as 0.77 and 0.67 for samples prepared with Bessel and Gauss laser beams, respectively.

## References


1. Neimash, V.; Poroshin, V.; Shepeliavyi, P.; Yukhymchuk, V.; Melnyk, V.; Kuzmich, A.; Makara, V.; Goushcha, A. O. *J. Appl. Phys.* **2013,** *114*, 213104. doi:10.1063/1.4837661