

SUPPLEMENTARY MATERIAL

Experimental details

Nanostructured Substrate Fabrication: The process of fabrication of the nanopillars and nanopores is schematically shown in Figure S1. The front surface of each substrate is cleaned using O₂ plasma exposure and then coated by a large-area closed-packed polystyrene (PS) latex microsphere (1 μm or 0.5 μm in diameter) monolayer formed at a water-air interface, as shown in Figure S2. The diameter of the PS spheres is reduced to different sizes by reactive-ion etching using O₂ plasma exposure at different times. For nanopillar fabrication, the PS spheres are used as the mask. The InP or Si wafer to be processed is etched by inductively coupled plasma (ICP) etching. After ICP etching, the PS spheres are removed via exposure in O₂ plasma for 4 min. For nanopore fabrication, a layer of Cr is deposited via thermal evaporation for use as the hard mask. Subsequently, the PS spheres coated with Cr are removed before performing ICP etching of the wafer.

Epitaxial InP deposition: Using a proprietary metal-organic chemical vapor deposition (MOCVD) process of SMART Photonics, the InP film was deposited onto all of the substrates in the same deposition run. In addition to nanostructured and flat Si substrates, reference InP substrates were also used. The InP and Si substrates were cleaned to remove organics and particles prior to deposition. To remove the native oxide, the Si substrates were also dipped in HF solution, rinsed with water, and then dried by blowing with N₂ prior to placement into the MOCVD chamber for InP deposition. The growth rate was approximately 1 micron per hour. According to capacitance-voltage measurements performed by SMART Photonics on a reference sample with the p-type InP film, the doping level was $8.0 \times 10^{17} \text{ cm}^{-3} \pm 0.5 \times 10^{17} \text{ cm}^{-3}$.

X-Ray Diffraction: The XRD measurements were performed using a high-resolution diffractometer equipped with a Cu-K_α x-ray source at the Eyring Materials Center at Arizona State University.

Photoluminescence Spectroscopy: The photoluminescence from an InP film sample is obtained by illuminating the sample with infrared light (810 nm) from a Ti:sapphire laser (16 μW average power) incident normal to the surface of the sample and focused onto the sample (ca. 3 μm diameter spot size) and then collecting the photoluminescence using a custom near-infrared microscope setup coupled to a spectrometer with a LN₂-cooled InGaAs linear array detector. Note that the InGaAs array detector has a long wavelength cutoff at approximately 0.77 eV and that the broad spectral dip at ca. 0.9 eV in the broad sub-bandgap PL peak is caused by atmospheric absorption. See Nano Lett. 2012, 12, 7, 3378-3384, <https://doi.org/10.1021/nl300015w> for information of the relationship between surface states and sub-bandgap PL.

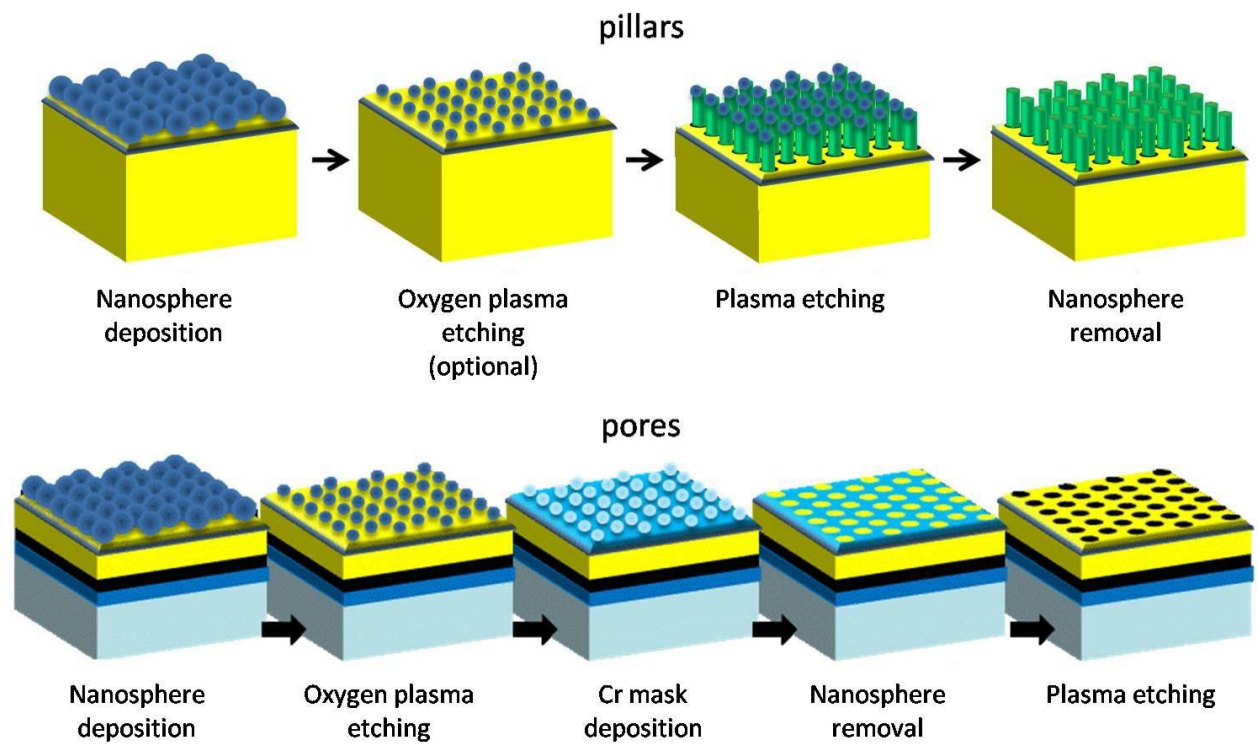


Figure S1. Schematic of the nanosphere lithography and etching process to produce nanopillars and nanopores.

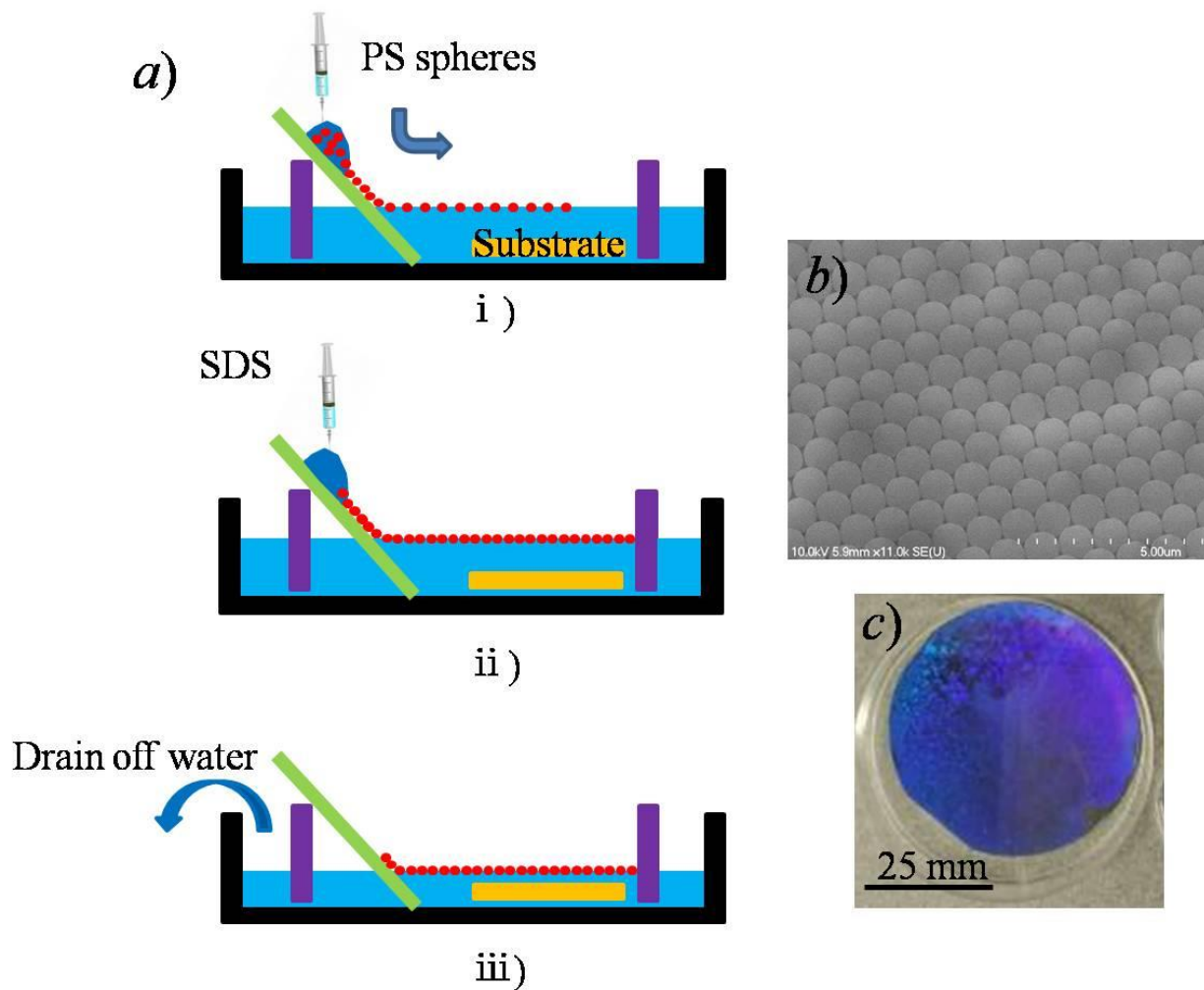
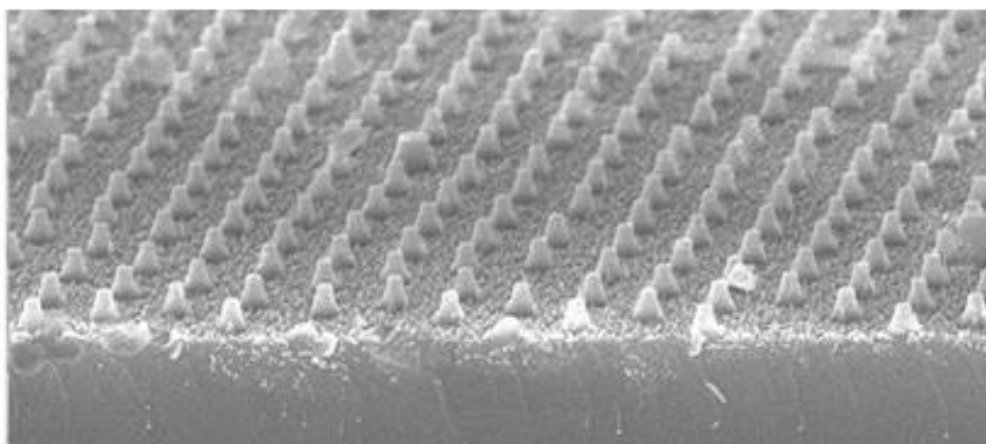


Figure S2. a) Schematic of the procedure of deposition of a monolayer of close-packed PS spheres at the water-air interface; b) scanning electron microscope images of a monolayer of PS spheres of 1 μm in diameter on a silicon substrate; c) photograph of a monolayer of PS spheres on a 2" silicon wafer.



250 nm diameter pillars
1 μm spacing

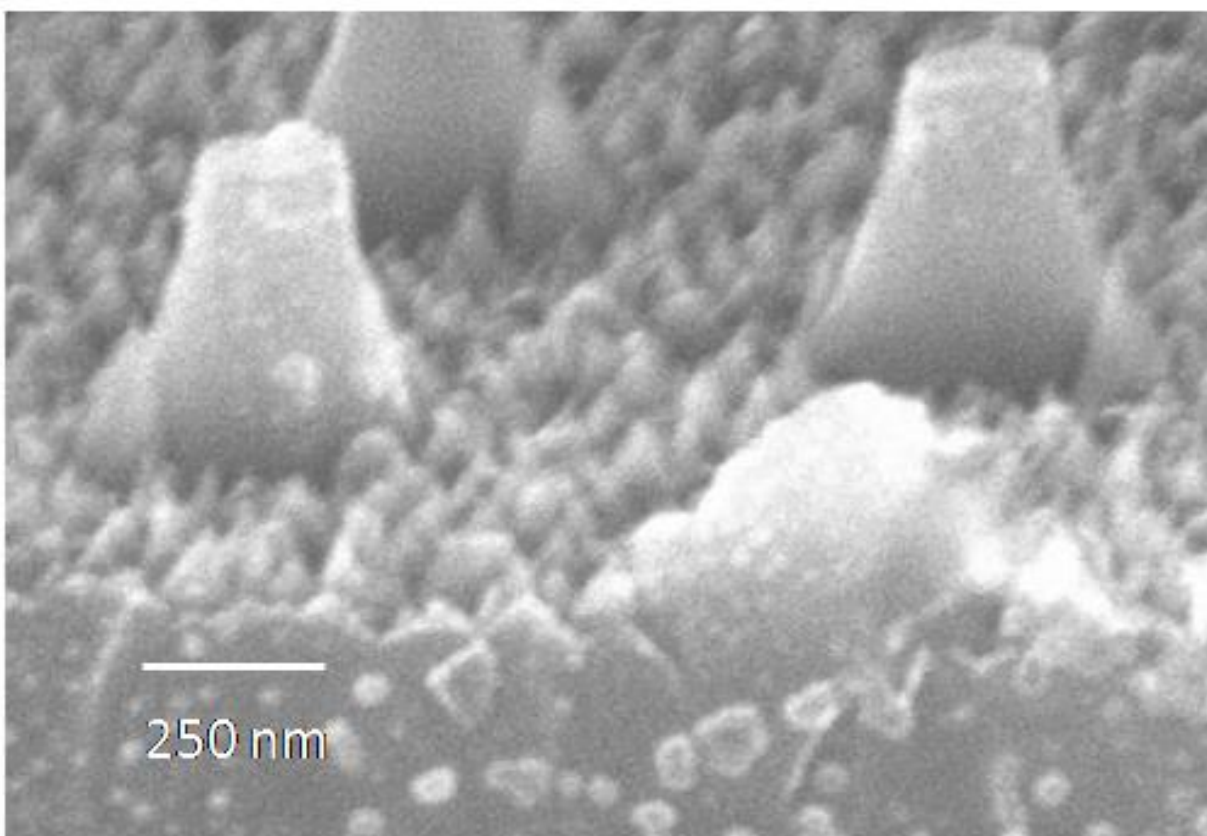


Figure S3. SEM images (30° tilt view) of etched Si pillars defined by nanosphere lithography.

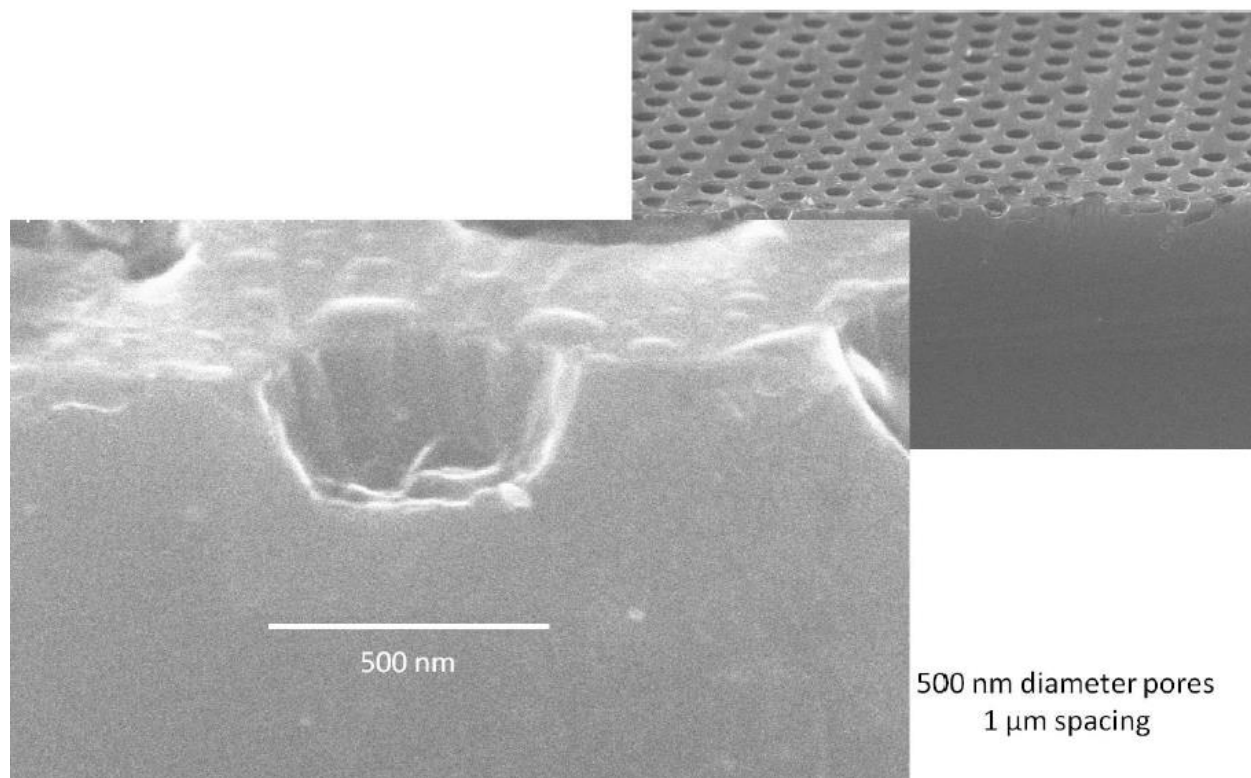


Figure S4. SEM images (30° tilt view) of etched nanopores in Si(100) using nanosphere lithography to define a Cr etch mask.

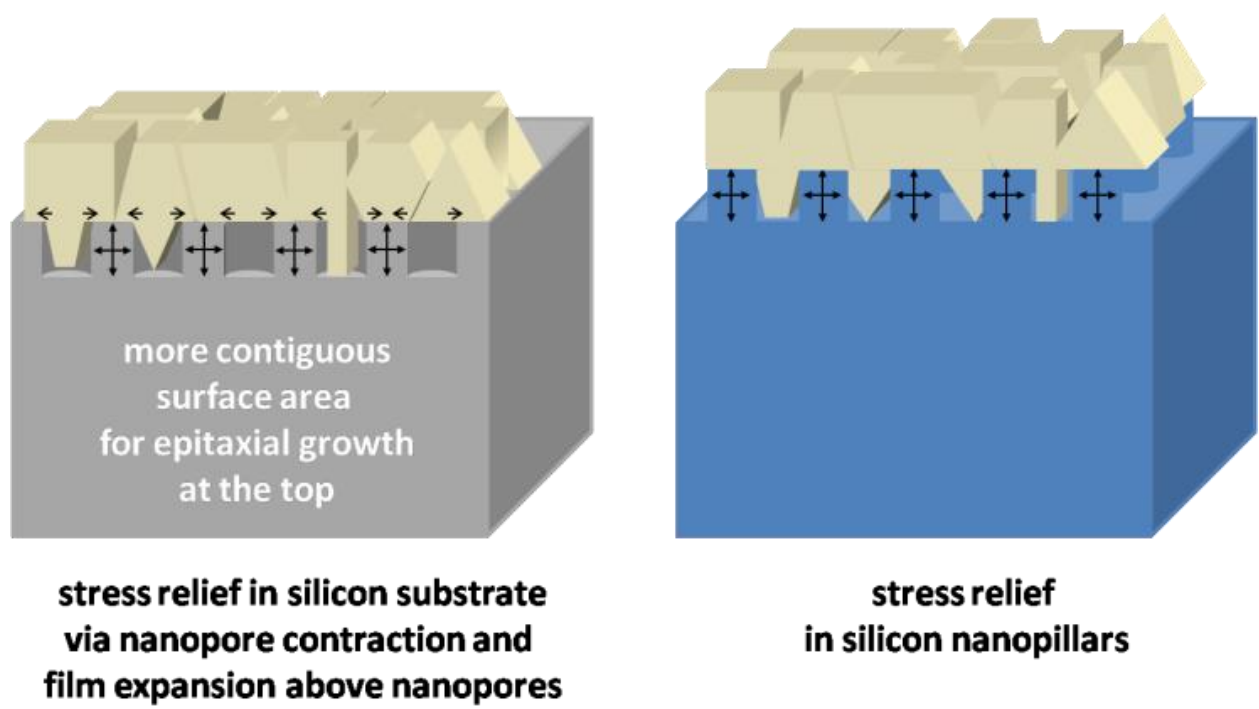


Figure S5. Comparison of the details of the stress relief at the hetero-interface between the InP film and the nanostructured Si substrate for nanopore and nanopillar morphologies.