A New Dimension of GLASS

FUSED silica is the go-to material for fashioning laser optics. Composed simply of one atom of silicon for every two oxygen atoms, the glass is mechanically robust, thermally resistant, and can efficiently transmit a broader spectrum of light than most materials.

Still, no material is perfect. High-energy laser systems regularly encroach upon the glass's physical tolerances, subjecting it to extreme experimental regimes that cause irreparable damage. Yet, superficial changes to fused silica could hold the keys to optical optimization and open a new dimension of design freedom.

When light passes between different media, portions of it can be reflected. Air and glass, for example, have different refractive indices—light propagates at different speeds through each material, with about 4 percent of the incident light reflected when it strikes a lens surface. This phenomenon hinders laser systems designed to maximize the energy throughput per unit area (fluence) of their optical components. Scientists often tweak optics by applying coatings made from multilayered dielectrics or sol-gel. The coatings help reduce reflectance and accept off-angle light by easing the transition between the refractive index of each medium. However, coatings unleash a cascade of new challenges. For example, the added layers can compromise the glass's durability.

Glass-only metasurfaces can tailor optical performance without added coatings. Precise patterns carved into the glass instill emergent material properties inaccessible with silica alone, such as altering the refractive index of a material by changing the volumetric proportion of glass to air on its surface. The potential uses for metasurfaces are manifold—provided they can be built at scale. "A major challenge of creating a metasurface is that its engineered features need to be substantially smaller than the targeted wavelengths of light," says Livermore scientist Eyal Feigenbaum.

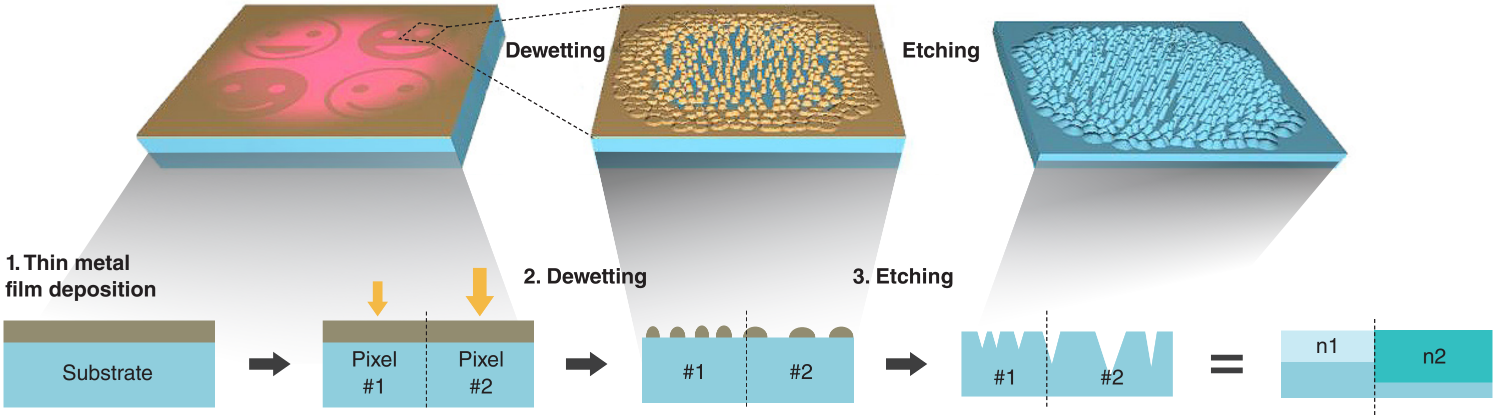
While some metasurfaces have been achieved on very small scales, their fabrication techniques trace out features one by one—impractical for scaling to large optics such as those installed at the National Ignition Facility (NIF). Funded by the Laboratory Directed Research and Development Program, Feigenbaum led a team of scientists and engineers from NIF and Photon Science and Physical and Life Sciences principal directorates in an effort to scale production of all-glass metasurfaces. Having reimagined the fabrication technique, the team developed a process for generating trillions of nanoscale structures on meter-scale glass surfaces.

Behind the Mask

The Livermore metasurface fabrication process starts by temporarily laying down a 5- to 15-nanometer-thick film of metal (usually gold or platinum) onto the glass surface using electron beam vapor deposition. The film is then heated, initiating a dewetting process in which the metal self-organizes into tiny islands of metal nanoparticles whose size and spacing directly vary with the induced temperature. Feigenbaum explains, "Whether we use a furnace for uniformity on the entire surface or raster-scan laser exposure to get local variation, heat application is a lever for defining the shape and period of the organization pattern, avoiding the need to trace each resulting meta-element individually." This thin mask of metal nanoparticles now acts akin to a photographic negative, shielding the underlying glass from the etching process that follows. During etching, bursts of plasma-phase particles buffet the film, eroding it and digging into the glass underneath. Glass protected by mask regions, however, remains untouched. Once the mask is depleted (or any traces chemically removed), all that remains is the glass surface that exhibits countless nanoscale structures with precisely controlled shapes.

Staff scientist Nathan Ray holds a fused silica optic whose engineered surface features instill antireflective properties. The optic's all-glass metasurface is composed of millions of nanoscopic structures.

"Getting to the relevant length scales for these surface features was no easy task," says staff scientist Nathan Ray. "We

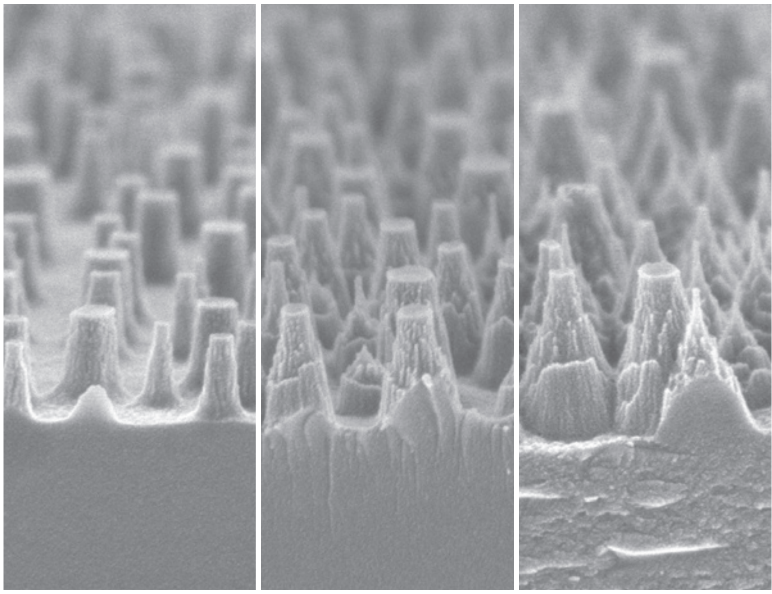


The metasurface fabrication process begins when a thin metal film is deposited atop the substrate (often fused silica). Heat application by furnace or laser (yellow arrows) initiates dewetting, during which the film self-assembles into nanoparticles forming a patterned, protective mask (brown layer). Ion etching then erodes the surface. Heat, mask height, and etching time and intensity can all be modified to customize the metasurface. Once the mask is removed, the different specific geometry and spacing of nanostructures dictate surface properties (represented as n1 and n2), which may include antireflection and altered refractive index.

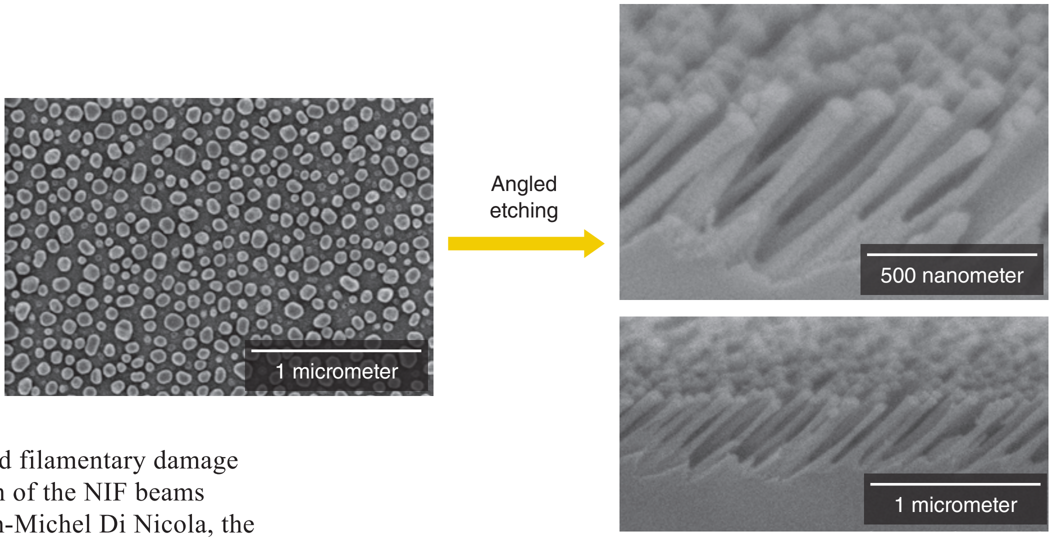
devoted much effort to be able to etch deep into the substrate and create structures tall enough to function as intended.” To achieve high-aspect-ratio features whose height is 10 to 15 times their width necessitated a longer etching process to dig deeper into the glass substrate without completely eroding the mask material. The mask nanoparticles could be made thicker to withstand more etching. However, simply thickening the initial metal film would produce nanoparticle spacing through the standard dewetting process exceeding the sub-wavelength period benchmark. Livermore’s novel seeded dewetting technique instead swells the initial metal nanoparticles in position over multiple depositions, ensuring high mask fill factor and high aspect ratio of etched features.

This method is not only scalable, it also generates previously unthinkable surface structures with unique optical properties. Ray explains, “Different applications require different surface geometries. With this process, we can precisely define the shape of nanoscale structures. That is, fittingly, a really big deal.”

During etching, the mask’s nanoparticles erode and shrink, and nanostructures honed from the glass surface gradually transition from pillars to cones. Pillars function similarly to sol-gel, producing thin layers of uniform index. Cones, on the other hand, tamp down broadband reflection and widen acceptable angles of incidence thanks to the gradient of refractive indices between their tips and bases. The team can therefore determine the optimal etch time to obtain any structure type on this cylinder-to-cone continuum. Other structures, such as truncated pyramids or needlelike features that were previously impossible to create, can instill new physical characteristics in glass ranging from increased mechanical robustness to modified particle adhesion. An expanded repertoire of metasurface structures is indispensable for meeting the rising performance demands of some of the world’s most challenging optical designs.



As etching time increases, a mask’s metal nanoparticles shrink, and nanostructures honed from the glass surface transition from pillars to cones as pictured above in images taken from metasurface sections subjected to different etching times.



Angled etching produces tilted pillar nanostructures used to induce birefringence in silica glass. Birefringence causes light to refract differently depending on its polarization, making it a key feature of waveplate optics that convert between light's polarization states. For high-power experiments at the National Ignition Facility, researchers found that converting linearly polarized light to circular polarization decreased filamentation damage.

A Spectrum of Applications

Although it may seem fitting for an enormous laser facility such as NIF to boast similarly enormous optics, scientists prefer to slim them down. Optical damage can occur when a high-irradiance (power per unit area) laser beam has ample time to interact with the bulk material—that is, if the glass is thick. This is the case inside some of NIF's ultraviolet-fused silica lenses where scientists have documented filamentary damage called "angel's hair" for a small fraction of the NIF beams when operated at maximum power. Jean-Michel Di Nicola, the chief laser systems engineer at NIF and co-program director for Laser Science and System Engineering, describes the physical phenomenon fueling filamentation: "Laser light effectively modifies its own propagation path through the glass. The laser's strong electric field can alter the glass's index of refraction, creating micro-lenslets that intensify the laser hotspots. This runaway self-focusing process can form micrometer-wide voids or bubbles in the optic—precursors to cracks."

Motivated to protect these optics, Di Nicola and colleagues calculated that switching from NIF's linearly polarized light to circular polarization would unlock what he calls a "new degree of freedom" untapped by most laser systems, making the optics more resilient to filamentation. The metric defined as the laser irradiance times length of the optics traversed (the IxL product) is effectively increased by 1.5 times when the laser is circularly polarized instead of linearly. Consequently, if circularly polarized light could be used, NIF engineers could opt for up to 50 percent greater power at the same filamentation probability to support higher-energy NIF experiments or gain more margins against filamentation at current regimes. Polarization rotation could also help generate magnetic fields or mitigate backscatter caused by laser–plasma interactions. However, switching polarization requires a specialized waveplate element to reorient the electromagnetic field components of transmitted light. "We can't simply buy this optic from a catalog. No waveplate exists that could meet the size and fluence resilience demands of NIF lasers," says Di Nicola.

Metasurfaces are uniquely suited for modifying the properties of fused silica to produce durable waveplates. Unlike crystals, which have defined, repeating arrangements of atoms, silica glass is amorphous; it, therefore, lacks birefringence, the property that allows a material to exhibit different refractive indices depending on the polarization of incoming light. Using computational tools such as finite-difference time-domain solvers of Maxwell's equations, Feigenbaum determined the dimensions of tilted features that optimally induce birefringence in fused silica to convert light from linear to circular polarization.

Beyond waveplates, the metasurface manufacturing process offers promise for fabricating the optics implicated in diverse technologies. Staff scientist Jae-Hyuck Yoo refined the laser dewetting step to obtain metalenses that are more compact, durable, and customizable than conventional lenses. In addition to their antireflective potential for optical systems, surface patterning that increases broadband acceptance can augment photovoltaic windows performance by permitting more wavelengths of solar radiation. Scientists can also construct metasurfaces on which fluid flows preferentially in one direction, an indispensable tool for designing microfluidic devices. The Livermore team continues to explore new surface structures and further applications.

—Elliot Jaffe

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