The Dawn of an **Optical Revolution**

WHAT does the National Ignition Facility—the world's largest laser system—have in common with a handheld camera? Both rely on precisely calibrated optics—mirrors, lenses, and other components—to direct light toward a target. Industry standards often evolve as researchers strive for better performance and cost effectiveness. In support of the Laboratory's national security mission and a range of related applications, Lawrence Livermore scientists and engineers are continually advancing optical technology, including materials development and processing methods. (See *S&TR*, April/May 2017, pp. 17–20; July/August 2016, pp. 12–15; and September 2014, pp. 4–12.)

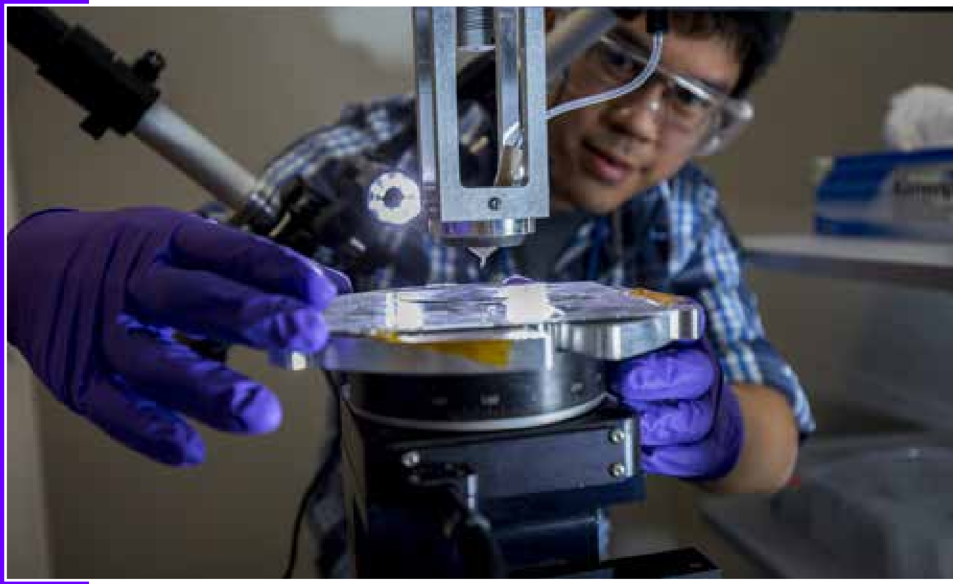
For an optical system of any size, innovations revolve around three variables: size, weight, and power. For example, a typical lens refracts light through its convex shape, which occupies valuable space when combined with other lenses in a series. Changing the compositional or structural properties of a lens can alter its shape, size, weight, performance, and production cost. Livermore chemical engineer Rebecca Dylla-Spears leads a team exploring new optical designs and fabrication methods. She explains, "With the right technology, we could potentially downsize an optical train to use fewer optics, creating a more compact system."

In a project funded by the Laboratory Directed Research and Development (LDRD) Program, Dylla-Spears's team is developing optical components with functionally graded material properties—that is, refractive and other properties resulting from changes to an optic's composition or structure, not its shape. Such advancement makes use of Livermore's capabilities in additive manufacturing (AM), also known as three-dimensional (3D) printing. In a breakthrough for AM technology, the team has "printed" optical-quality glass, lenses with a gradient refractive index (GRIN), and gradient-composition laser gain media.

The multidisciplinary team's goals lie at the intersection of the Laboratory's leadership in laser technology, optical engineering,

Livermore researchers are using additive manufacturing to print a new class of optics, such as this gradient-composition glass optic, shown gripped in a lens-holding tool. (Photo by Jason Laurea.)

Laboratory engineer Du Nguyen monitors the progress of ink deposition onto a substrate during direct ink writing. (Photo by Jason Laurea.)



advanced materials science, and AM technology. The project spans multiple classes of optics and different AM techniques, notably direct ink writing (DIW). "We're combining our skills to move rapidly," says Dylla-Spears. "Learning from other areas of the Laboratory and discovering different approaches to problem solving are important for moving forward."

Finding the Right Recipe

Besides transparency, next-generation optical glasses will have high-resolution architectures made without a mold. Conventional glass-processing techniques and many advanced manufacturing methods fall short of these goals. For instance, heat-based printing can generate forms by melting and fusing silica (silicon dioxide) powders, but the process leaves the silica filaments vulnerable to thermal stress and can result in nonuniform structures. The LDRD team saw potential in DIW, a mainstay of Livermore's AM capabilities that provides scientists with submillimeter precision for many material properties. (See *S&TR*, March 2012, pp. 14–20.) Dylla-Spears states, "We chose DIW because it gives us the most control over composition in three dimensions."

DIW-printed glass optics present significant challenges at all stages of fabrication. The ink needs to hold its shape, remain stable during printing, and flow at just the right rate to eliminate gaps between print lines. In addition, the printed structure must be mechanically strong to survive shrinkage during thermal treatment—drying, burnout, and sintering. Finally, to achieve the desired optical finish, the glass must withstand polishing without deformation or acquiring new defects. "These processes are dependent on pore and particle size," notes Dylla-Spears. "Altering ink composition affects the heat treatment process, as well. Cracking is a problem, as are bubbles, or voids, in the glass. If stresses develop during sintering, the piece is ruined."

Dylla-Spears and colleagues began by using a silica slurry to print glass. This series of experiments proved DIW's viability for producing silica optics, so the next step was determining how to control the glass's refractive properties. The team developed a custom ink by combining silica with titania (titanium dioxide), a common glass additive, to form a colloidal suspension that included precursors, binders, and solvents. In a second round of experiments, the inks were blended in a reservoir before extrusion through the nozzle. "By adding titania to silica and refining the process accordingly, we translate the optical prescription of the refractive index to a print prescription," explains Dylla-Spears.

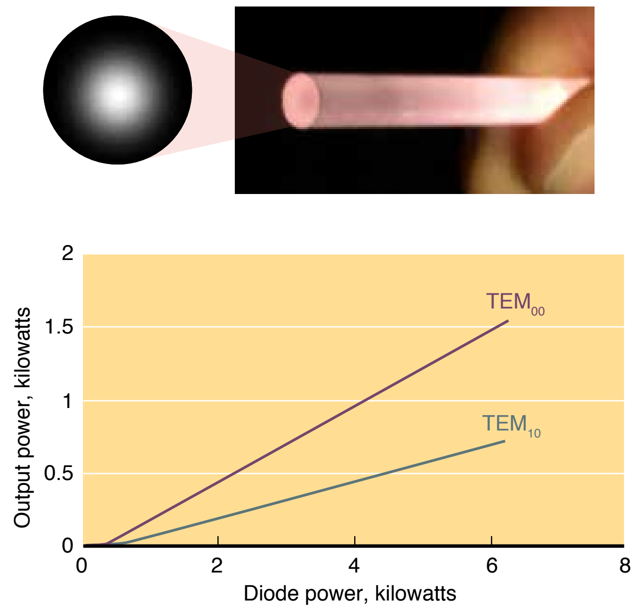
As the DIW printer deposits the ink on the substrate, the mixture is adjusted at different stages to create the structure's gradient composition in a radial pattern. The finished GRIN lens is a flat disc.

Making the Grade

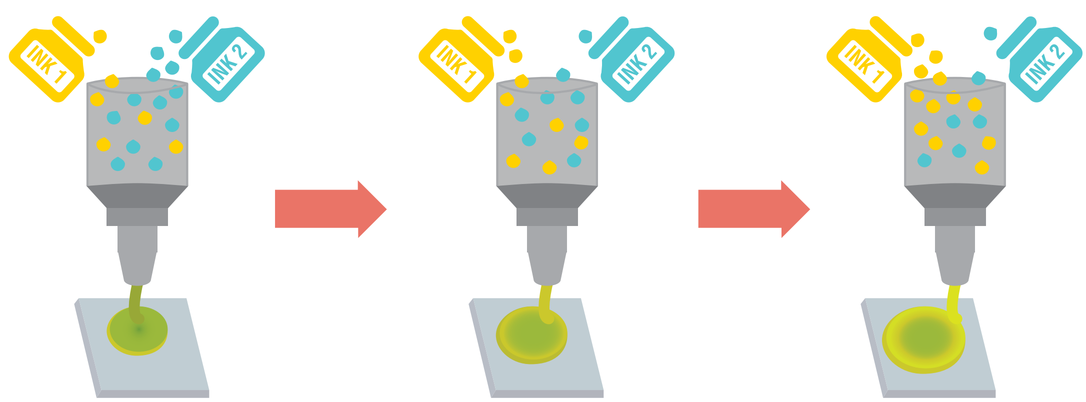
For decades, gain media—laser rods used to amplify laser energy—have been made from a single crystal of neodymium-doped yttrium–aluminum–garnet (Nd:YAG). However, uniform doping throughout the cylindrical structure leads to inefficiency as energy farther from the central axis of the rod is not extracted. A rod with tailored doping promises better laser efficiency and beam quality but cannot be grown as a single crystal, while manufacturing a rod from two or more parts poses fabrication challenges. As part of the LDRD project, Laboratory physicist Stephen Payne leads development of a cylinder of transparent Nd:YAG by DIW. Nd-doped and undoped YAG inks are extruded independently. In doped inks, Nd becomes concentrated in the cylinder's core so that the outer cladding remains Nd-free. According to Livermore scientist Nerine Cherepy, energy efficiency comes from this spatial change in the rod's structure.

Ink composition is key to creating a doped rod. The team combines high-surface-area nanoparticles with surfactants and polymers to produce unique shear-thinning characteristics, so that ink viscosity can change during printing. "The ink should flow freely through the nozzle, but afterwards it must gel on the substrate," explains Cherepy. "The particles within the ink are lubricated with surfactant to create a weak bond. Moving through the nozzle temporarily overcomes the bond between particles."

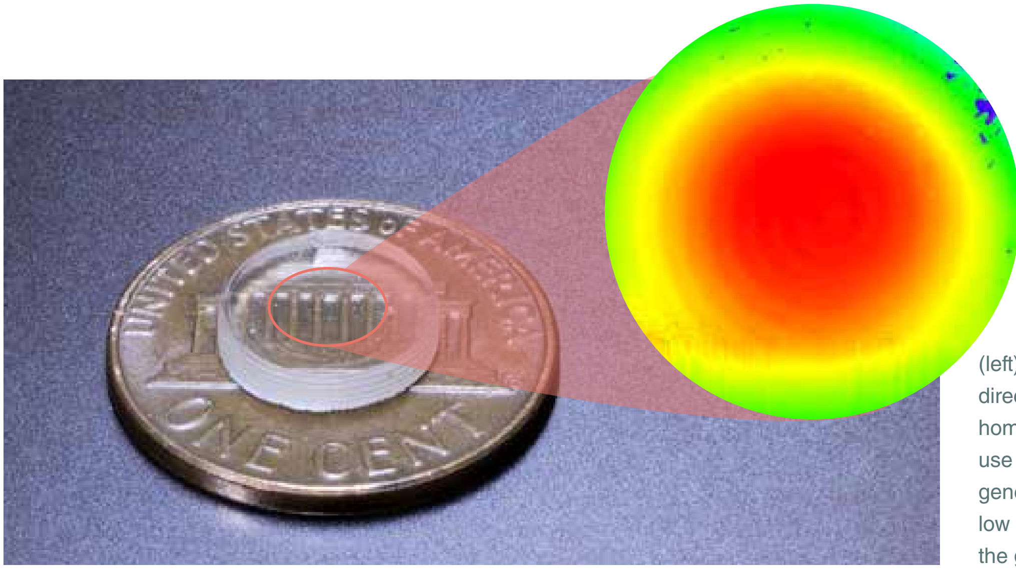
After printing by chemical engineer Tim Yee, the cylinder goes through ceramic processing, which had already proven successful and scalable for other transparent ceramic optics. (See *S&TR*, January/February 2017, pp. 12–13.) The piece undergoes multiple steps—including vacuum sintering and hot isostatic pressing—at different pressures and temperatures. Ceramist Zachary Seeley notes, "Our approach to manufacturing and processing should double the efficiency of gain media." Another goal is to create a cylinder with a high-aspect-ratio geometry, where the typical gain medium rod is many times longer than its diameter. Mechanical technologist Scott Fisher has developed ways to support the cylinder during DIW printing. Payne states, "As this technology matures, we will see many new possibilities. Optical components could be printed to design specifications."



(top) A rod of Livermore's tailored transparent ceramic gain medium contains a neodymium-doped core surrounded by undoped cladding. Optimized ink composition ensures uniform light propagation from the rod's center to edge. (bottom) Calculations of the Gaussian beam transverse electromagnetic (TEM) profiles shown illustrate how the graded rod amplifies power more effectively in the higher efficiency TEM_{00} mode than in the lower efficiency TEM_{10} mode.



Livermore researchers create gradient-composition optics with direct ink writing technology and tailored inks. Changing the proportions of the mixed inks while printing alters the refractive index, producing a refractive-index gradient indicated by the color gradient in the printed lens. (Subsequent lens-crafting steps omitted.)



(left) A gradient refractive index lens printed with direct ink writing is shown. (right) To determine the homogeneity of the lens's refractive index, researchers use phase-shifting diffraction interferometry to generate a map of the bulk material. The high (red) to low (blue) refractive indices of this piece correspond to the gradient pattern.

Quality Matters

Although gradients are visible in the team's 3D-printed structures, proof comes from refractive evaluation. The quality of a functionally graded lens is determined by homogeneity in its refractive index—that is, the consistency of light passing through. Using phase-shifting diffraction interferometry and correcting for any surface-shape effects, the LDRD team can quantify the lens's bulk material properties. Compared to commercial glass materials, Livermore's GRIN lens exhibits industry-standard density and approaches the chromatic dispersion and spectral absorption of conventionally manufactured silica–titania optics. Moreover, this technique has shown that printed glass can rival commercial silica glass in homogeneity of optical index. Dylla-Spears notes, "With control over material composition, the opportunities to modify other optical and material properties, such as absorption and thermal conductivity, are wide open."

Optical diagnostics confirm that the team's specially doped gain media can meet expectations. Scatterometry measures light traveling end-to-end through the rod, while interferometry measures refractive index. However, Nd doping introduces a slight change to the rod's refractive index. "To compensate for this change and obtain uniformity throughout the rod, we tune the composition of the outer cladding with lutetium doping," explains Seeley.

Just Getting Started

In another parallel effort, the LDRD team is using large-area projection microstereolithography to fabricate gradient-composition lattices for mirror components. The technique is ideal for manufacturing 3D structures with extremely fine features (see *S&TR*, January/February 2016, pp. 14–15). Optical mirrors are traditionally made from solid glass or metal, which is shaped before the surface is polished or coated for reflection. The larger the mirror, the more susceptible it is to problems associated with weight reduction, such as weakened support structures. By using AM in mirror fabrication to develop ultralight, ultrastiff microlattices, Livermore engineers can specify functional gradients to optimize the lattice supports. Postdoctoral researcher Nik Dudukovic explains, "We can control the global and local stiffness of the support structures by increasing the lattice density at points where high stresses are anticipated while also minimizing the lattice's overall density and weight." In addition to aerospace applications, Dudukovic says lightweight mirrors can potentially improve dynamic performance and reduce the power and size requirements for motor components in fast-scanning optical systems.

As Dylla-Spears's team advances the manufacturing possibilities for optical components, project goals include developing additional ink blends, scaling the 3D-printed pieces to larger sizes, and further refining multistep AM processes. "We're aiming for optics design and experimental efforts to go hand in hand," states Dylla-Spears. "The potential exists to revolutionize optical system design."

—Holly Auten

Key Words: additive manufacturing (AM), direct ink writing (DIW), functionally graded optical material, gain medium, gradient refractive index (GRIN), Laboratory Directed Research and Development (LDRD) Program, large-area projection microstereolithography, National Ignition Facility, neodymium-doped yttrium–aluminum–garnet (Nd:YAG), optics, phase-shifting diffraction interferometry, refraction, silica (silicon dioxide), three-dimensional (3D) printing, titania (titanium dioxide).

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