Simultaneous Printing and Deformation of Microsystems via Two-Photon Lithography and Holographic Optical Tweezers

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The purpose of this work is to enable the simultaneous printing and deformation of polymer microsystems using an integrated two-photon lithography (TPL) and holographic optical tweezers (HOT) approach. This approach is the first of its kind to enable the fabrication of advanced metamaterials, micromechanisms, soft microrobots, and sensors that require embedded strain energy in their constituent compliant elements to achieve their intended behaviors. We introduce a custom-developed photopolymer chemistry that is suitable for near-infrared (NIR) TPL fabrication but remains unreactive in the visible-light regime for HOT-based handling. We facilitated the optimal HOT-based actuation of TPL-fabricated microsystems by advanceing a ray-optics-based optical-force simulation tool to work with microbodies of any arbitrary shape. We demonstrate the utility of this integrated system via fabrication of three unique case studies, which could not be achieved using any alternative technologies.

Introduction

New technologies have been evolving rapidly to enable the fabrication of microsystems, which are currently not feasible to make using conventional methods. Of particular interest are microsystems that require the storage of strain energy in their compliant constituent elements to facilitate new kinds of metamaterials, micro-mechanisms, soft micro-robots, and other compliant microarchitectures. General systems that possess embedded strain energy often exhibit unique non-linear characteristics such as negative and/or tunable stiffness[1] and show behaviors such as buckling, snapping, and wrinkling. Although such behaviors have typically been avoided when designing traditional systems due to their nonlinear complexities, these behaviors have more recently been exploited for enabling more advanced applications[2]. Sensor, actuator, and switch applications have, for instance, been achieved by leveraging the configuration changes that dramatically occur when some multi-stable strain-energy-storing systems are subjected to small perterbations[3]. Much research has been conducted toward enabling such embedded-strain-energy applications on the macroscale, however, limitations in fabrication technologies have prevented the

Conceptual insights

Systems with embedded strain energy have become an important area of research due to their ability to achieve extraordinary capabilities as a result of how their constituent elements are deformed within equilibrium states. Such systems can store and release energy in controlled ways in response to external stimuli and can be engineered to exhibit large deformations with minimal actuation forces. However, fabrication of these systems on the microscale requires the creation of an advanced technique for deforming the systems' constituent elements in any way desired. Here we demonstrate a new technique that combines two-photon lithography (TPL) with holographic optical tweezers (HOT) for enabling polymer microsystems to be printed with embedded strain energy. Our system uses light as the sole agent to simultaneously print and deform polymer microsystems. Furthermore, we have developed a photopolymer chemistry that is specially designed for the different working laser wavelengths of our system's hybrid TPL and HOT processes. A geometric optics simulation tool is also used to calculate optical force profiles on arbitrarily shaped bodies to enable the optimal manipulation of printed structures for general scenarios. This work facilitates the microfabrication of new metamaterials, energy harvesters, soft robots, shape-morphing structures, sensors, and actuators.

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† Electronic supplementary information (ESI) available: MATLAB-based optical-force simulation tool for arbitrary shapes, integrated TPL and HOT system component details, photopolymer absorption spectrum data, and experimental videos of the fabricated microsystems. See DOI: 10.1039/x0xx00000x

realization of such systems on the microscale. *In this work, we present a new technology that combines two-photon lithography (TPL) with holographic optical tweezers (HOT) to enable the fabrication of micro-sized systems with embedded strain energy.*

TPL is a non-linear photopolymerization process[4] that has found widespread application in the fabrication of complex, true-3D microsystems owing to its high resolution and adaptability to different materials such as polymers[5], metals[6], and ceramics[7]. Optical trapping[8] is a complementary technique that has been widely used by biologists to manipulate nano- and micro-sized objects[9]. This technique has also found applications in microfabrication due to its high precision in positioning microparticles[10,11]. By incorporating a diffractive optical element called a spatial light modulator (SLM), the HOT approach uses a single laser source to generate multiple optical traps that can be independently and simultaneously reconfigured[12]. In this work, a new system is introduced that integrates both the TPL and HOT approaches to allow microelements to be simultaneously printed and deformed to fabricate new polymer microsystems that could not be made using any other approach.

Existing works have utilized TPL-based fabrication and HOT-based manipulation as serial processes in separate systems, such as in the micro-snap-fit mechanism[13] and the micro-screw-wrench[14] by Köhler *et al.*, micro-assembly of non-spherical particles by Ksouri *et al.*[15], and the optical waveguides by Palima *et al.*[16]. Separate TPL and HOT systems are difficult to use because they require challenging post-processing steps to develop the printed objects without washing them away (e.g., complex cage structures are often printed in addition to the intended objects to contain them[14,17]). Furthermore, such systems require the cumbersome task of locating the printed micro-objects under the HOT system after development to then manipulate them. In addition to being difficult to operate, separate TPL and HOT systems are inherently not capable of printing embedded strain energy within structures. The reason is that such systems cannot print new features onto existing objects while they are being deformed by optical tweezers to passively hold the objects in their deformed state and thereby trap the optically induced strain energy within their geometry.

Although others have developed TPL systems integrated with optical-tweezing capabilities (e.g., the system that fabricated the microscale tetherball pole by Dawood *et al.*[18] or the system that enabled the delivery and encapsulation of microspheres in polymer by Askari[19]), no one has combined TPL with HOT. The published integrated systems are only capable of generating a single optical trap to deform printed structures—not multiple independently controlled traps, which HOT systems are capable of generating. As a result, such systems are limited by how they can deform the objects they print. Such objects must be fixed to substrates in order to deform their geometry because a single optical trap produces only a single force, which would displace a free-floating object—not deform it. Additionally, the current integrated systems have not demonstrated the ability to print embedded strain energy within fabricated objects in part because this capability requires sophisticated simulation and automation algorithms to synchronize the TPL and optical tweezing capabilities. The current systems have also only demonstrated the handling of spherical objects. Spherical objects possess the easiest geometry to manipulate using optical tweezers because a sphere requires only one trap to handle and the optimal location to place that trap is commonly known to be at the spheres' centre. Handling other arbitrarily shaped objects in a controlled way often requires multiple optical traps placed at nonintuitive optimal locations to produce the forces and moments required for moving and deforming the object to the desired location and orientation. It is important that these locations and orientations are precise since they must correspond with the location and orientation of new features that must subsequently be printed in conjunction with the previously printed and deformed objects to passively hold them in place and thereby store embedded strain energy.

The new system introduced here combines TPL and HOT for the first time in a single integrated system to enable the fabrication of microsystems with embedded strain energy. The presented TPL/HOT system can simultaneously print multiple microelements of any shape (either free-floating or fixed to a substrate), reorient and hold them in place in a suspension of photopolymer resin, and deform them in a coordinated effort using multiple forces and moments imposed on the elements by groups of automated optical traps. Our approach requires no intermediate chemical development procedure between the TPL and HOT steps, allowing the print-and-deform process to be synchronized and automated. Thus, subsequent features can be printed on top of previously printed features to passively store strain energy in their geometry as optical tweezers are simultaneously holding them in precisely deformed configurations.

To facilitate and optimize the handling, deformation, and actuation of the polymer structures printed by our system, a custom-developed photopolymer chemistry is introduced in this paper. The photopolymer chemistry is notable for its two-photon absorption in the near-infrared (NIR) range, transparency to 532 nm visible light, and readily-available chemical components, which enable printing and deformation of the polymer microsystems fabricated.

We also present an advanced geometric optics tool that generates optical force and torque profiles on micro-bodies of any shape. Simulating such loads has been an area of interest since the discovery of optical tweezers. These simulations are categorized according to the wavelength of the laser beam (λ) and the diameter (D) of the particle being trapped. If a particle is more than an order of magnitude smaller than the wavelength of the laser beam ($D \ll \lambda$, Rayleigh regime), Lorentz force expressions on point dipoles can be used to calculate the induced optical forces[20]. For particles comparable in size to the wavelength of the laser beam ($D \sim \lambda$, Mie regime), more rigorous methods are required to simulate the induced optical forces. One popular method is the *T-matrix* formulation[21] developed by Niemenen *et al*[22]. Another method, which uses optical force density principles to achieve the same objective, was recently introduced by Phillips *et al*[23] to provide a more straightforward alternative. Although these and other methods are capable of simulating optical forces imposed on particles of any shape, they are limited to Mie-regime scenarios. For scenarios of interest to the contributions of this paper in which the printed body is much larger than the wavelength of the trapping laser by at least an order of magnitude (D>>λ, geometric optics regime), ray tracing techniques can be employed to simulate optical forces imposed[24]. Although these techniques have primarily been used to simulate the forces imposed on spherical bodies, others have successfully used ray tracing techniques to simulate the forces imposed on semi-spherical objects such as ellipsoids[24], semi-cylindrical rods[25] and Janus spheres[26]. Ray tracing techniques have been used to simulate the optical forces imposed on bodies of arbitrary shapes[27–29], but defining the geometry of such shapes is typically cumbersome and limited to shapes that can be analytically defined manually. Often times those shapes are approximated for the sake of simplicity and thus the accuracy of the results is compromised. We extend the ray-tracing toolbox provided by Jones *et al*[29] to simulate the accurate optical forces imposed on bodies of any shape that pertain to the geometric-optics regime and are defined using standard computer-aided design (CAD) software, which can be uploaded to the tool as standard stereolithography (*.stl)* files. This tool enables our integrated TPL/HOT approach to rapidly identify the optimal trap locations on printed bodies of arbitrary shapes to deform them in an automated way for embedding strain energy within microsystems, which were not previously possible to fabricate.

Materials and methods

Material

We present a new photopolymer resin that is cured by two-photon absorption using NIR (760 nm) femtosecond pulses but is unreactive to visible continuous-wave (CW; 532 nm) light used for optical trapping. The resin consists of 1.1%wt. TPO-Li (Colorado Photopolymer Solutions), 38.4%wt. ethoxylated (15) trimethylolpropane triacrylate (Sartomer SR9035), and 60.5%wt. deionized (DI) water. The refractive index of the liquid polymer was measured to be 1.3918 and the refractive index of the solid polymer was measured to be 1.4912, a difference significant enough to produce the optical refraction and momentum transfer needed to support optical trapping as shown in the Simulation section. The resin chemistry was initially inspired by Dawood *et al.* who used synthesized MBS as the photoinitiator[18]. However, we present a resin with a commercially-available photoinitiator, TPO-Li as a substitute to MBS, which renders the hybrid TPL/HOT process more widely accessible. The results of the optical characterization tests of this resin can be found in ESI†.

Microfabrication

The microsystems are fabricated in a sample chamber consisting of a microscope slide and a coverslip separated by two pieces of tape of approximately 50 µm thickness each. The microscope slides were functionalized with acrylate for better adhesion of the polymer bodies printed[18]. The hybrid microfabrication process includes printing with TPL and manipulation using HOT, and if required, one or both of these processes are repeated to complete the fabrication process.

The hybrid microfabrication process can be run fully automated. The MATLAB software developed for the control of TPL/HOT system correlates multiple inputs including *(1)* the lithography file (in *.stl* format) representing the structure to be printed, *(2)* the desired locations of the optical traps on the printed bodies, *(3)* printing parameters (e.g., scan rate and laser power), and finally *(4)* calibration values that relate the position and scaling of the scanning mirrors' area, projected hologram's area, and recorded images. This information is loaded into a MATLAB graphical user interface (GUI) using a *.mat* file and the system handles subsequent fabrication steps automatically.

Results and discussion

Simulation

In this section we present a ray-optics-based simulation capable of calculating optical forces and moments on microsystems with any arbitrary shape. This tool (provided in ESI†) is an extension of the open-source and validated Optical Tweezers Software (OTS) published by Jones, Marago, and Volpe[29], but has been advanced here to accept general *.stl* files, to define the geometry of general shapes. The presented MATLAB code facilitates modelling of complex microstructures without the need to manually define different surfaces and their coordinates, which can be a very time-consuming and challenging task for surfaces that are not basic geometric shapes. The improved tool is important because it allows for *a priori* knowledge of where to optimally place optical traps to manipulate and strain TPL-printed parts of arbitrary shape.

In order to demonstrate the capabilities of the simulation tool, optical forces on the irregularly shaped body shown in Fig. 1a-b are studied. The body simulated in the figure is fabricated as an embedded-strain-energy lattice case study and is discussed later in the Fabrication section. Simulation results show that in-plane optical trapping forces are maximized at the edges of the printed micro-bodies due to refraction at those edges (Fig. 1a-b). This refraction and resulting momentum transfer produces a stable potential well that attracts the body to the focal spot of the laser. Furthermore, the simulation tool reveals that, for planar TPL-fabricated microsystems, there is an optimal trapping plane located within the bounds of the microsystem in which axial forces are zero and stable (i.e., 375 nm above the mid-plane of the free-floating body as shown in Fig. 1c), and therefore, optical traps can effectively move the body in-plane without rotating it out of plane. This principle is leveraged in the operation of the TPL/HOT system in that the optical traps are focused slightly above the TPL writing laser's focal spot. Thus, traps are automatically created at the zero-axial-force mid-plane of the printed micro-bodies immediately after they are printed. Furthermore, simulation data shows that the strongest optical trapping occurs at locations where physical edges are located in close proximity. Since physical edge features result in extremum in the force profile, close placement of opposite edge features creates a sign change and steep slope that results in strong and stable trapping (Fig. 1c). However, while optical traps placed on thin features generally support strong optical trapping, it also creates a narrow optical force profile that is not stable over a large range. We favoured 3µm-wide features as they were observed to balance optical trap strength with stability.

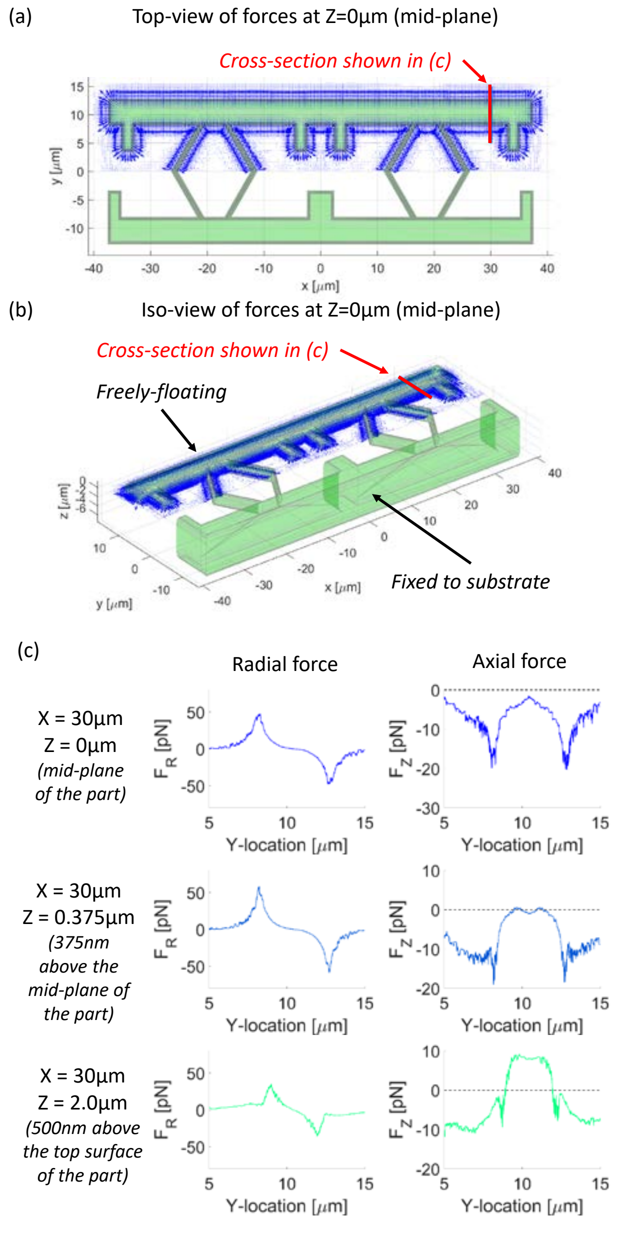

Figure 1. Ray-optics-based force simulations on an irregularly shaped body using a 50 mW, 532 nm optical trap. The effective numerical aperture of the optical trap is 1.3, the refractive index of the medium is 1.3918, and the index of the micro-body is 1.4912. (a) The top view of the force vector field located at the mid-plane of the free-floating half of the body shows forces are maximized at edges. (b) The isometric view demonstrates the tool's ability to calculate out-of-plane forces and moments, which are minimized when the trapping beam is focused close to the mid-plane of the free-floating body. (c) Cross-sectional subplots show radial and axial forces at various Z-heights across the red line shown in (a) and (b).

Fabrication

Our fabrication system consists of three subsystems: the TPL system, the HOT system, and the imaging system (Fig. 2; the system components are described in detail in ESI†). Three case studies that demonstrate the capabilities of the presented hybrid TPL/HOT system are discussed in what follows.

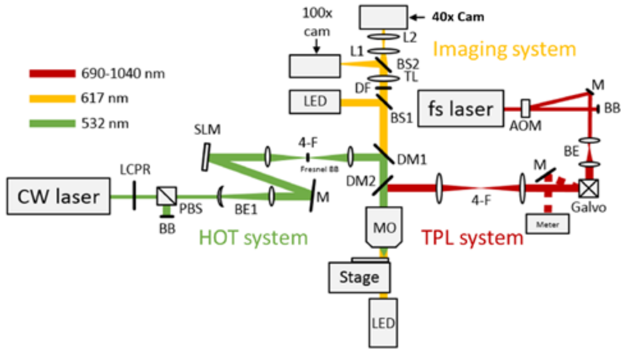

Figure 2. Integrated TPL/HOT system schematic

Microscale jack-in-the-box

The *jack-in-the-box* depicted in Fig. 3a-e and in Vid. 1 from ESI† is a proof-of-concept microsystem capable of storing and releasing strain energy. The fabrication process begins with printing five 3µm-thick support layers using TPL (Fig. 3a) to create a U-shaped box fixed to a substrate. The final layer printed consists of the same U-shaped box but with a vertically oriented lid attached to the box by a compliant hinge as well as a suspended free-floating spring connected to a free-floating disk shaped like a smiley face. The face is then pushed into the box by two optical traps generated by the HOT approach placed on the inner edges of its disk (close to the eyes shown in Fig. 3b). The box lid is then closed with a single optical trap (Fig. 3c). The stored energy in the compressed spring and lid hinge are then released by removing the optical traps holding the system in its deformed configuration. The face hits the lid and pushes it open as the strain energy is converted into kinetic energy (Fig. 3d). Lastly, the lid is brought back to its initial location using a single optical trap and the structure returns to its initial configuration (Fig. 3e).

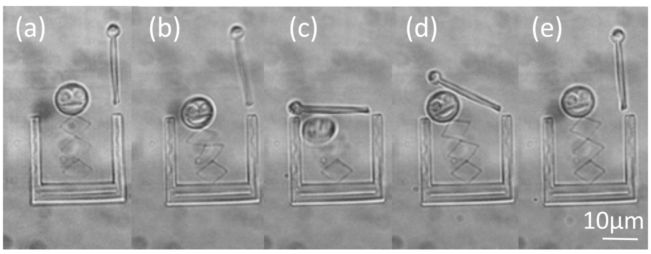

Figure 3. Simultaneous printing and deforming of a microscale jack-in-the-box

Embedded-strain-energy lattice

We now demonstrate the TPL/HOT system's ability to fabricate new metamaterial lattices with embedded strain energy. Such metamaterials could significantly enhance shape-morphing[30], energy absorbing, and deployable[2] applications.

A planar embedded-strain-energy lattice design consisting of 2x2 unit cells was fabricated (Vid. 2, ESI†) using our hybrid microfabrication technique in seven steps. First, an undeformed portion of the lattice (i.e., a single pair of unit cells) was fabricated using TPL with one side fixed to the substrate (Fig. 4a). Second, four optical traps were used to deform the narrow V-shaped flexure elements by actuating the free-floating side of the pair of cells at predefined locations based on *a priori* simulation results (Fig. 1). Third, the TPL system selectively polymerized the resin at four points at the contact interfaces of the two sides of the cell pair thus fusing them together (Fig. 4b). Fourth, a second pair of unit cells was fabricated next to the existing pair using TPL again. The printing location of the second pair of cells was calculated via an image processing routine that uses background subtraction and edge detection in order to find the top of the existing structure. Fifth, the HOT approach was used to bring the second pair of unit cells in contact with the first pair so that the TPL approach can fuse them together (Fig. 4c). Sixth, four optical traps were again used to deform the V-shaped flexure elements of the second pair of printed cells by actuating their top side at predefined locations also based on *a priori* simulation results. Seventh, the TPL system polymerized resin at four points at the contact interfaces of the two sides of the second pair of cells thus fusing them together as well (Fig. 4d).

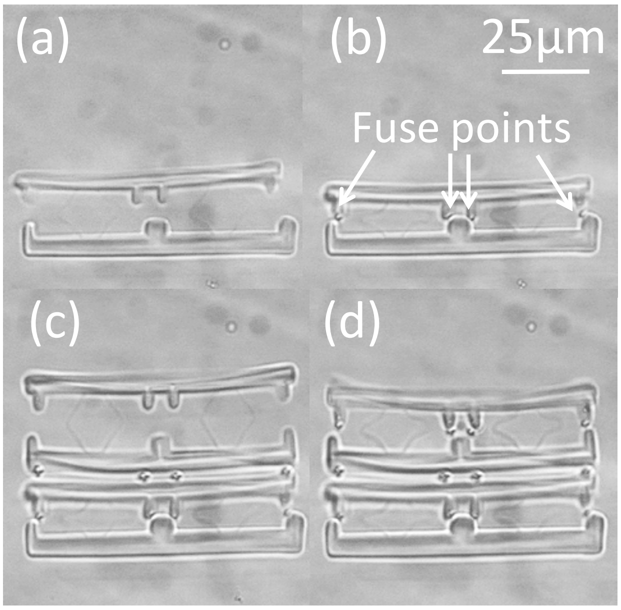

Figure 4. Hybrid microfabrication of an embedded-strain-energy lattice consisting of 2x2 cells. Thin, V-shaped flexure elements, which are difficult to see in the figure, join the sides of each cell as shown in Fig. 1a-b.

The result is a fabricated lattice with strain energy stored in deformed portions of its architecture in a stable equilibrium state. Although experimental data[31–33] has shown that, even on the microscale, some of the energy stored in the lattice will likely diminish gradually as the polymer constituent elements undergo stress relaxation, much of the energy is likely to remain stored for practical long-term use. This energy could be suddenly released if the lattice is impacted in such a way that the fuse points (Fig. 4b) that join the opposing sides of the cells together are cleaved via shearing. Thus, the lattice could cause a projectile that impacts its top surface with sufficient kinetic energy to rebound with even greater kinetic energy after impact. A much larger version of the lattice that consists of many more cells could be used as a shape-morphing or deployable metamaterial that swells in regions that have been fractured. It could also be used as a sensor to detect if and where impact has occurred. Finally, note that although the lattice of Fig. 4d is 2D, the hybrid approach introduced here is not limited to the fabrication of planar 2D structures only. 3D versions of the lattice could be designed and fabricated for practical applications.

Negative Poisson's ratio metamaterial

In our final case study, we apply our new approach to print and actuate a free-floating auxetic (i.e., negative Poisson's ratio[34]) metamaterial to conduct *in situ* measurements of its Poisson's ratio. Auxetic metamaterials are important not only because of their unique deformation behavior[35] but also because of other augmented mechanical properties (e.g., resistance to indentation[35] and enhanced energy absorption[36] for use in impact protection devices[37]). In this work, we fabricated a free-floating auxetic lattice via TPL (Fig. 5a). Six 4 µm-diameter disk-shaped features were added to the ends of the lattice to act as handling locations for the HOT approach. The initial and final locations of the six optical traps used to actuate the lattice were predefined and their displacement was automated so that the Poisson's ratio of the lattice could be measured via image processing (Fig. 5b). The lattice returned to its initial position after removal of the optical traps over numerous cycles due to the strain energy stored in its architecture (Vid. 3, ESI †). The boundaries of the lattice were detected using captured images and the average Poisson's ratio calculated over three cycles actuated within 30% strain was found to be -0.54.

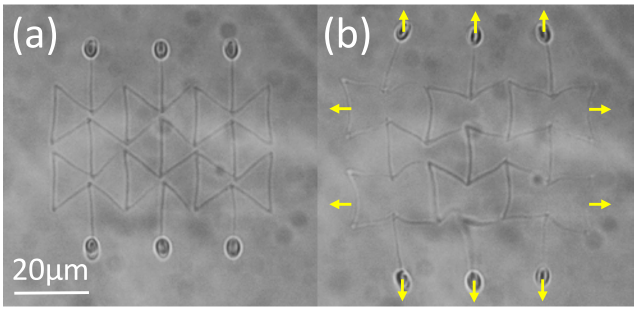

Figure 5. (a) A free-floating auxetic lattice printed using TPL; (b) the lattice is actuated using HOT

Conclusions

This work demonstrated the first integrated TPL and HOT system, which is capable of fabricating microstructures with embedded strain energy. The system can simultaneously print, deform, orient, displace, join, and actuate arbitarily shaped free-floating bodies as desired. We advanced an open-source simulation tool to determine the optimal placement of optical traps for most efficiently handling such bodies. The simulation results indicated that optical traps are most efficient when they are positioned at the edges of bodies and slightly above their mid-plane. Informed by the results of the simulation tool, three example polymer microsystems were fabricated in a photopolymer resin specifically developed for decoupled TPL-based printing and HOT-based handling. Although this work focused on the automated fabrication of planar 2D microstystems with deformed elements, both the simulation tool and hybrid fabrication system could be adapted to enable the fabrication of fully 3D versions of such microstystems as a future work.

Acknowledgements

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Conflicts of interest

There are no conflicts to declare.

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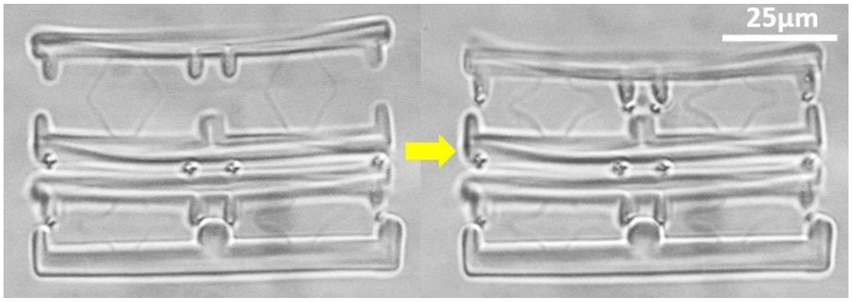

Microstructures with embedded strain energy are fabricated by an advanced approach that combines two-photon lithography with holographic optical tweezers.