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13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT

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RPPR Final Report

as of 08-May-2023

Agency Code: 21XD

Proposal Number: 77190SM

Agreement Number: W911NF-21-1-0068

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Final Report for Period Beginning 01-Feb-2021 and Ending 31-Jan-2023

Title: Polymer Chemistry: Soft Mechanical Logic

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STEM Degrees: 2

STEM Participants: 4

Major Goals: The overarching goal of the project was to develop new approaches to soft mechanical logic devices that can respond to environmental stimuli and perform non-trivial logical computation. During the two year period of the project, we made substantial progress on six major objectives:

1. Characterizing the behavior of mechanical 'transistors' that can gate the flow of information through 1-dimensional mechanical 'wires' as a building block for performing non-trivial logical computations.
2. Understanding how modifications to the geometry of a mechanical linkage influence the topology of its configuration space.
3. Using our approach to design configuration space topology of mechanical linkages to gate propagation of signals down a chain.
4. Establishing a new approach for (sub-)micrometer-scale 3D printing that will facilitate the fabrication of small-scale mechanical logic devices.
5. Developing logic gates using Marangoni optical trapping and repulsive interactions between photothermally responsive elements.
6. Understanding the stability of interacting elements in optical traps to identify fundamental limitations of large-scale logic architecture.

Accomplishments: See attachment.

Training Opportunities: The project contributed to the research, training, and professional development of one PhD student at Syracuse University (Michelle Berry, Physics), one PhD student at UMass Amherst (David Limberg, Polymer Science and Engineering) two PhD students at CU Boulder (Joseph Roback, Chemical Engineering; Rishabh Tennankore, Materials Science and Engineering), and one postdoctoral fellow at CU Boulder (Nabila Tanjeem, Chemical Engineering). Berry successfully defended her PhD in June of 2022 and is now a data scientist at Kroger, Limberg successfully defended his PhD in February of 2022 and is now a research scientist at DuPont. Tanjeem completed her postdoctoral fellowship in July of 2022 and is now an Assistant Professor of Physics at California State University Fullerton.

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Results Dissemination: The results have been disseminated through:

- ? Peer reviewed publications (six published papers and one accepted, one manuscript currently in preparation).
- ? Six contributed presentations by project personnel, at the American Physical Society March Meetings in 2021 and 2022 and the European Symposium for Photopolymer Sciences (ESPS) in 2021.
- ? 11 invited presentations by PI Hayward (Penn State University, Northwestern University, University of Houston, American Chemical Society Fall 2021 Meeting, Smart and Tough Gels IUTAM Symposium, University of Akron, University of Maryland, University of Colorado Boulder, Materials Research Society Fall 2020 Meeting, Lebanon Valley College, University of Wisconsin Madison) and 5 invited presentations (University of Connecticut, Yale, University of Bristol, New York University, Simons Center for Geometry and Physics at Stony Brook University) by co-PI Santangelo.

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: Ryan Hayward

Person Months Worked: 2.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Co PD/PI

Participant: Christian Santangelo

Person Months Worked: 2.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Michelle Berry

Person Months Worked: 15.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: David Limberg

Person Months Worked: 15.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Joseph Roback

Person Months Worked: 15.00

Project Contribution:

National Academy Member: N

Funding Support:

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as of 08-May-2023

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Nabila Tanjeem

Person Months Worked: 7.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Graduate Student (research assistant)

Participant: Rishabh Tennankore

Person Months Worked: 2.00

Funding Support:

Project Contribution:

National Academy Member: N

ARTICLES:

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Peer Reviewed: Y

Publication Status: 1-Published

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Date Submitted: 8/31/21 12:00AM

Date Published: 4/1/21 6:00AM

Publication Location:

Article Title: Coupled oscillation and spinning of photothermal particles in Marangoni optical traps

Authors: Hyunki Kim, Subramanian Sundaram, Ji-Hwan Kang, Nabila Tanjeem, Todd Emrick, Ryan C. Hayward

Keywords: Responsive materials, Marangoni force, coupled oscillators, active matter

Abstract: Cyclic actuation is critical for driving motion and transport in living systems, ranging from oscillatory motion of bacterial flagella to the rhythmic gait of terrestrial animals. These processes often rely on dynamic and responsive networks of oscillators—a regulatory control system that is challenging to replicate in synthetic active matter. Here, we describe a versatile platform of light-driven active particles with interaction geometries that can be reconfigured on demand, enabling the construction of oscillator and spinner networks. We employ optically induced Marangoni trapping of particles confined to an air–water interface and subjected to patterned illumination. Thermal interactions among multiple particles give rise to complex coupled oscillatory and rotational motions, thus opening frontiers in the design of reconfigurable, multiparticle networks exhibiting collective behavior.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of the American Chemical Society

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Publication Identifier: 10.1021/jacs.1c11022

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Date Submitted: 8/4/22 12:00AM

Date Published: 3/1/22 7:00AM

Publication Location:

Article Title: Triplet–Triplet Annihilation Photopolymerization for High-Resolution 3D Printing

Authors: David K. Limberg, Ji-Hwan Kang, Ryan C. Hayward

Keywords: high resolution 3D printing, triplet-triplet annihilation, photoinitiation

Abstract: Two-photon polymerization (TPP) currently offers the highest resolution available in 3D printing (?100 nm) but requires femtosecond laser pulses at very high peak intensity (?1 TW/cm²). Here, we demonstrate 3D printing based on triplet-triplet-annihilation photopolymerization (TTAP), which achieves submicron resolution while using a continuous visible LED light source with comparatively low light intensity (?10 W/cm²). TTAP enables submicrometer feature sizes with exposure times of ?0.1 s/voxel without requiring a coherent or pulsed light source, opening the door to low-cost fabrication with submicron resolution. This approach enables 3D printing of a diverse array of designs with high resolution and is amenable to future parallelization efforts.

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Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Advanced Materials

Publication Identifier Type: DOI

Publication Identifier: 10.1002/adma.202105758

Volume: 34

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Date Submitted: 8/4/22 12:00AM

Date Published: 11/1/21 6:00AM

Publication Location:

Article Title: Shape-Changing Particles: From Materials Design and Mechanisms to Implementation

Authors: Nabila Tanjeem, Montana B. Minnis, Ryan C. Hayward, Charles Wyatt Shields

Keywords: shape changing materials, particle fabrication, self assembly, responsive systems

Abstract: Demands for next-generation soft and responsive materials have sparked recent interest in the development of shape-changing particles and particle assemblies. Over the last two decades, a variety of mechanisms that drive shape change have been explored and integrated into particulate systems. Through a combination of top-down fabrication and bottom-up synthesis techniques, shape-morphing capabilities extend from the microscale to the nanoscale. Consequently, shape-morphing particles are rapidly emerging in a variety of contexts, including photonics, microfluidics, microrobotics, and biomedicine. Herein, the key mechanisms and materials that facilitate shape changes of microscale and nanoscale particles are discussed. Recent progress in the applications made possible by these particles is summarized, and perspectives on their promise and key open challenges in the field are discussed.

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Journal: Physical Review E
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevE.106.044212
Volume: 106 Issue: 4 First Page #:
Date Submitted: 5/4/23 12:00AM Date Published: 10/1/22 6:00AM
Publication Location:

Article Title: Mechanical signaling cascades

Authors: Michelle Berry, Yongjae Kim, David Limberg, Ryan C. Hayward, Christian D. Santangelo

Keywords: mechanical computation, dynamics of networks, additive manufacturing, nonlinear waves

Abstract: Mechanical computing has seen resurgent interest recently owing to the potential to embed sensing and computation into new classes of programmable metamaterials. To realize this, however, one must push signals from one part of a device to another and do so in a way that can be reset robustly. We investigate the propagation of signals in a bistable mechanical cascade uphill in energy. By identifying a penetration length for perturbations, we show that signals can propagate uphill for finite distances and map out parameters for this to occur. Experiments on soft elastomers corroborate our results.

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Journal: Physical Review E
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevE.106.055002
Volume: 106 Issue: 5 First Page #:
Date Submitted: 5/4/23 12:00AM Date Published: 11/1/22 6:00AM
Publication Location:

Article Title: Controlling the configuration space topology of mechanical structures

Authors: M. Berry, David Limberg, M. E. Lee-Trimble, Ryan Hayward, C. D. Santangelo

Keywords: smart materials, mechanical meta materials, topology, mechanical computation

Abstract: Mechanical computing has seen resurgent interest recently owing to the potential to embed sensing and computation into new classes of programmable metamaterials. To realize this, however, one must push signals from one part of a device to another and do so in a way that can be reset robustly. We investigate the propagation of signals in a bistable mechanical cascade uphill in energy. By identifying a penetration length for perturbations, we show that signals can propagate uphill for finite distances and map out parameters for this to occur. Experiments on soft elastomers corroborate our results.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
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Publication Identifier Type: DOI Publication Identifier: 10.1039/D2SM00369D
Volume: 18 Issue: 34 First Page #: 6384
Date Submitted: 5/4/23 12:00AM Date Published:
Publication Location:

Article Title: Robust folding of elastic origami

Authors: M. E. Lee-Trimble, Ji-Hwan Kang, Ryan C. Hayward, Christian D. Santangelo

Keywords: self-folding structures, smart materials, origami

Abstract: Self-folding origami, structures that are engineered flat to fold into targeted, three-dimensional shapes, have many potential engineering applications. Though significant effort in recent years has been devoted to designing fold patterns that can achieve a variety of target shapes, recent work has also made clear that many origami structures exhibit multiple folding pathways, with a proliferation of geometric folding pathways as the origami structure becomes complex. The competition between these pathways can lead to structures that are programmed for one shape, yet fold incorrectly. To disentangle the features that lead to misfolding, we introduce a model of self-folding origami that accounts for the finite stretching rigidity of the origami faces and allows the computation of energy landscapes that lead to misfolding. We find that, in addition to the geometrical features of the origami, the finite elasticity of the nearly-flat origami configurations regulates the proliferation of po

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Acknowledged Federal Support: Y

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Partners

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I certify that the information in the report is complete and accurate:
Signature: Ryan Hayward
Signature Date: 5/4/23 2:08PM

Characterizing the behavior of mechanical ‘transistors’

We studied a system of two connected 4-bar linkages that either blocks a signal, or allows it to pass, depending on the state of a switchable component (Fig. 1A, center). This system, nicknamed the “flip-flop,” was simulated using a combination of rigid beams and linear springs with tunable stiffnesses. The flip-flop has two zero modes that correspond to the top or bottom half shifting laterally. When all components of the flip-flop are infinitely rigid, only one half of the linkage can move at one time. With finite stiffness components, a finite energy barrier resists moving both halves of the flip flop at the same time (Figure 1B). We coupled the two zero modes of the flip flop to mechanical wires (Figure 1A) to allow the flip-flop to gate a mechanical signal. In the configuration shown, a signal sent along the top wire is allowed to fully propagate. To gate the signal, we send a signal down the bottom wire which shifts the bottom half of the flip-flop to the right and locks the top half in place.

A 3D printed physical system was first designed as a prototype using a flexible resin on a commercial stereolithography (SLA) 3D printer (Figure 1C). While the compliance of individual components could be adjusted by tuning thickness, the device did not effectively gate mechanical signals due to the low material stiffness. Fabricating the device monolithically using a stiffer material would no longer allow the joints to bend and bistable beams to buckle as desired. The device was thus redesigned using rigid LEGO pieces that could be assembled by hand. These experiments showed that locking strength is high when the compliance of the device is low, but introducing just one compliant beam dramatically reduces the locking strength. Thus, this mechanism is likely not well-suited for constructing mechanical logic with stimuli-responsive elements.

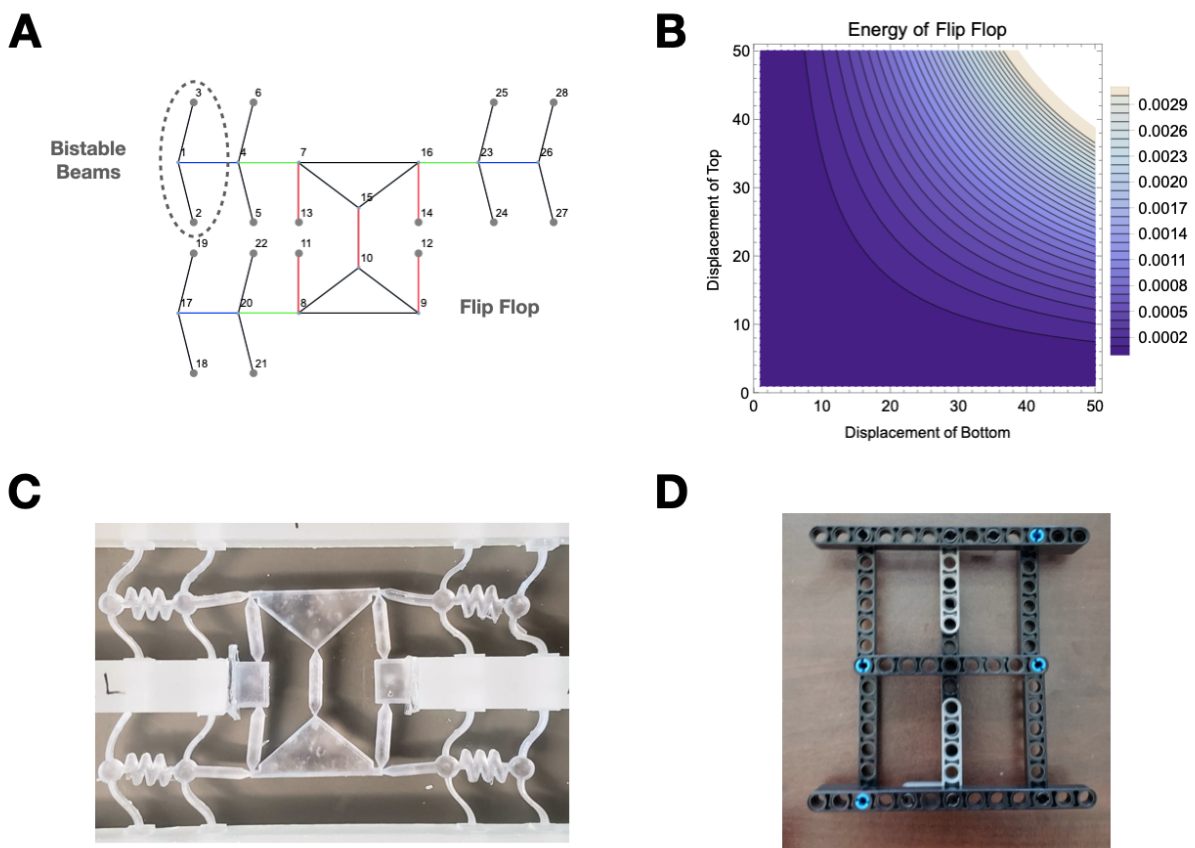


Figure 1. (A) A flip-flop inserted into a mechanical wire. In this configuration, the flip-flop will act as a transistor controlled by the bottom left wire. (B) The energy landscape of the flip-flop. A positive displacement of the top or bottom half corresponds to that half shifting to the right by some amount. (C) The experimental realization of the design in (A) using a soft, elastic material. (D) A flip-flop built using rigid Lego Technic parts.

Understanding geometry-dependent configuration spaces of mechanical linkages

As experiments revealed that the flip-flop was not as robust as needed for computations, we broadened our strategy to identify alternative linkages that may offer improved performance. The key feature that allows the flip-flop to behave as a transistor is that it has a critical point in its configuration space, where two different branches in its one degree-of-freedom motion intersect. Beyond simply designing linkages that behave as transistors and logic gates, the ability to engineer the configuration space of a linkage to have a desired number of critical points connected in a specific fashion provides a route to a general class of computing devices known as ‘finite state machines’. To rationally design such devices from mechanical linkages, we first need to fully characterize how linkage geometry controls its configuration space.

To design linkages with at least one critical point, we connected two simple one degree of freedom linkages and with a single rigid bar. This resulted in a larger linkage that still had one degree of freedom. The length of the additional rigid bar is a function of the positions of the two original linkages, and this function tells us exactly how to connect the two linkages to create a critical point.

We first focused on joining two rotors--rigid bars with one free end and one fixed end--to create a four bar linkage. By tuning the lengths of the bars and positions of the fixed vertices, we developed systematic rules governing how critical points in the configuration space can be eliminated to yield two disconnected branches (Figure 2). We then added a third rotor using the same process, and generalized our approach for designing the topology of the configuration space to higher dimension (Figure 3).

Each of the simulated linkages can be fabricated using LEGOs. Further, by inserting a custom 3D printed piece that can buckle or stretch as the center linkage, the linkage can transition between disconnected pathways on the configuration space beyond a critical force. As shown in Figure 4, the linkage can freely move along the lowest energy pathway (orange). Bending the center beam allows the linkage to access its mirror image structure from state c to f. Ongoing measurements are focused on determining the energy barrier to transition between states as a function of beam length and compliance.

In addition, we prepared bistable beams that allow the linkage to be switched from one configuration to another. As shown in Figure 5a-b, when the beam is in its long state, the mechanism is confined to the region of configuration space outside of the central triangle. By applying force to shorten the beam, it now accesses the region within the triangle (5c and d), which was inaccessible previously. This is accomplished without disassembling and reassembling the device, opening a route to stimuli-responsive mechanisms that dramatically change their deformation pathways in response to an input stimulus.

This work was published in *Physical Review E* in 2022 [1].

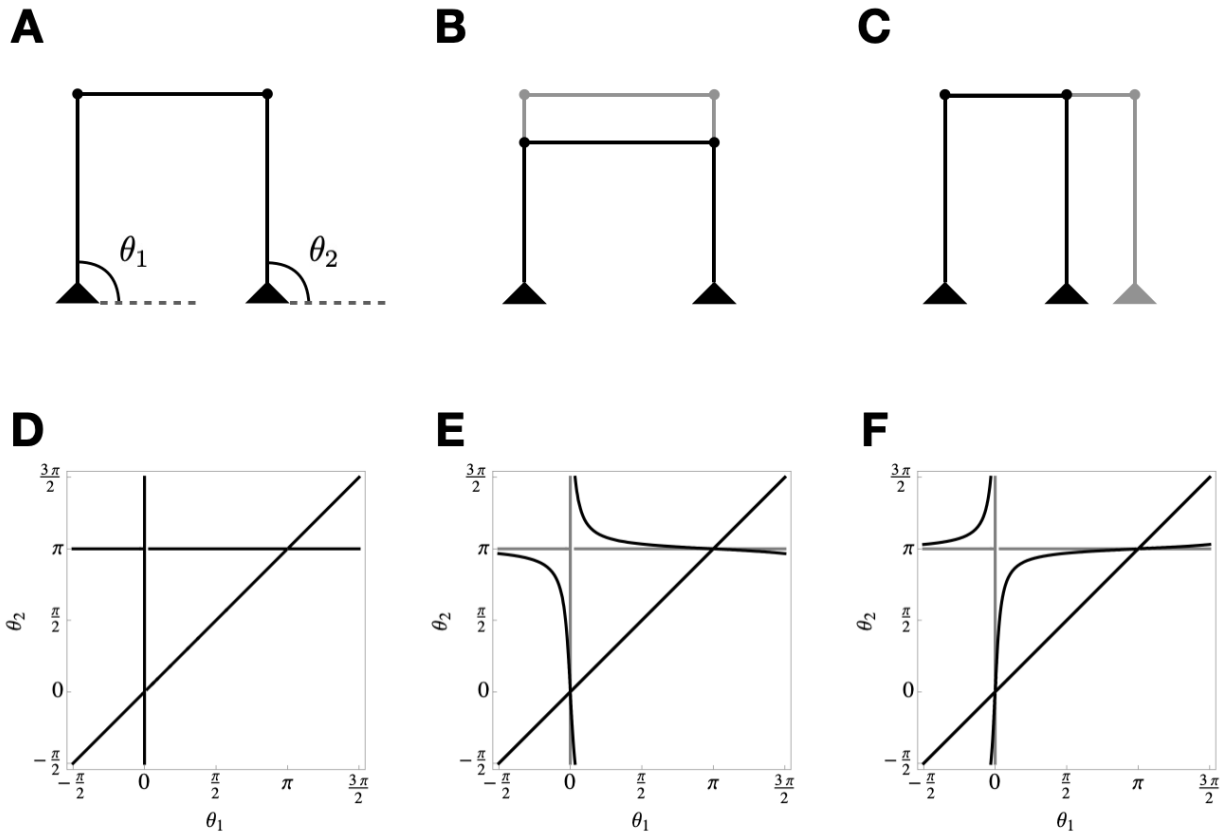


Figure 2. Four bar linkages with each beam the same length (A), with the left and right beams shorter (B), and with the middle beam shorter and the fixed vertices moved closer together (C). The original (D), and modified (E, F) configuration spaces are plotted as functions of the angular position of the two rotors. One of the critical points (top left intersection of black lines) present in D is destroyed by both changes, but leading to different connectivities of the resulting configuration spaces in E and F.

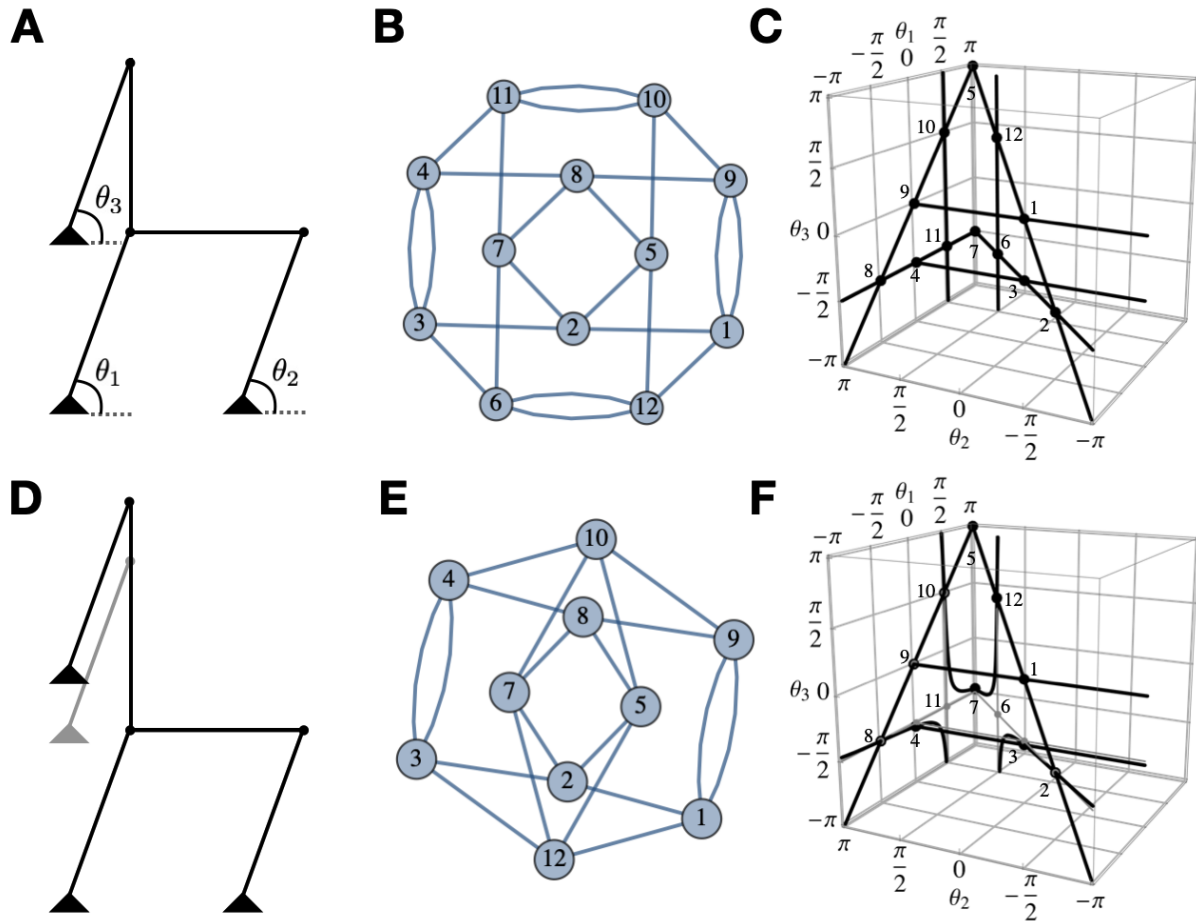


Figure 3. (A) A linkage consisting of three rotors attached together by two additional beams. All beams and rotors are the same length. (B) A graph and 3D plot (C) showing how the critical points are connected. (D)-(F) Modifying the linkage by lengthening one of the beams and moving the fixed point of one rotor as sketched (D) destroys two of the critical points (6 and 11), restructuring the graph (E) and configuration space (F).

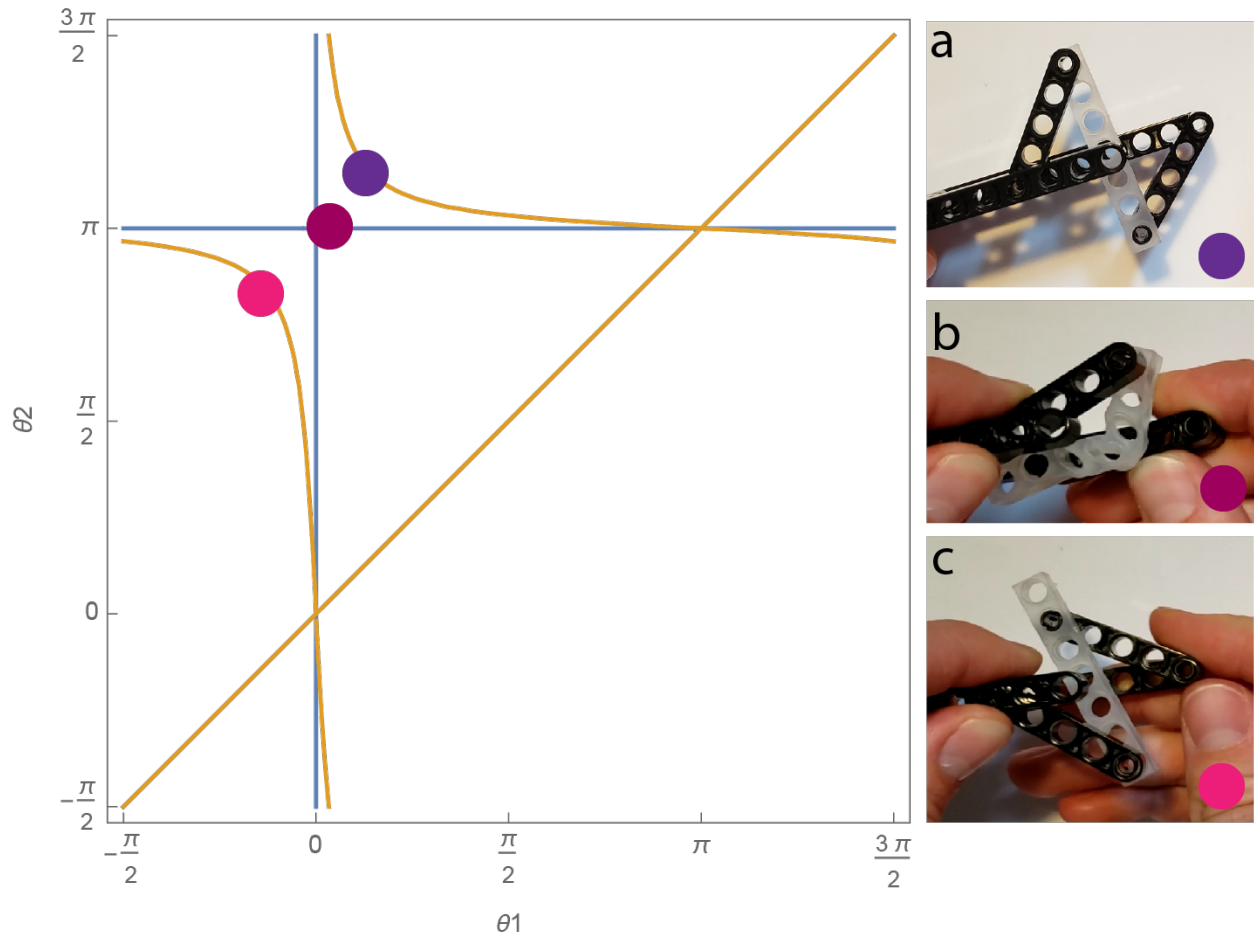


Figure 4. Inserting a flexible middle beam enables a snap buckling transition between disconnected paths in the configuration space from Figure 2E. a) The beam angles are $\pi/4$ and $5\pi/4$ and the middle beam is at its rest length, as denoted by the purple point in the configuration diagram at left. b) The flexible beam buckles under applied force, allowing the rotor angles to approach 0 and π , corresponding to the magenta spot on the phase diagram, where the destroyed critical point was previously located. c) After snapping through to the other branch (pink spot), the rotor angles are $-\pi/4$ and $3\pi/4$ and the center beam regains its rest length.

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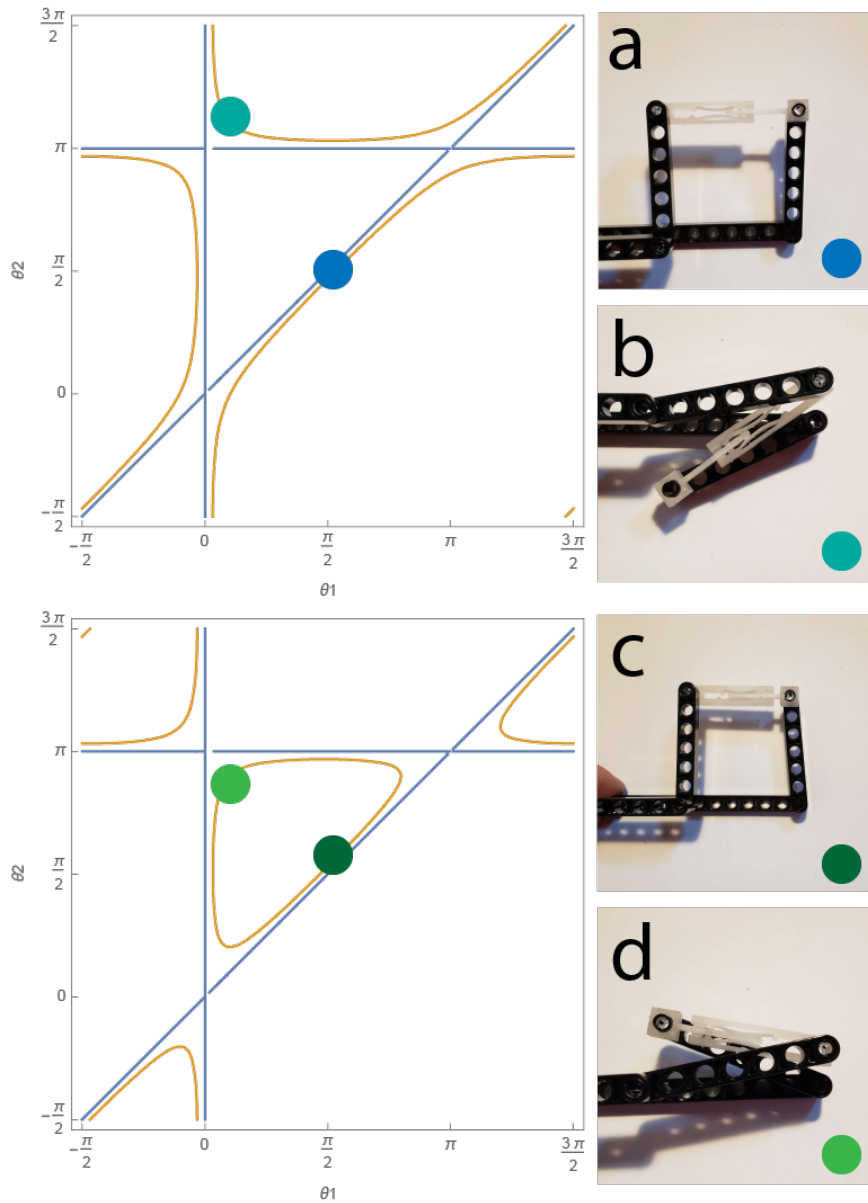


Figure 5. Beams with bistable lengths allow transition between different configuration spaces by application of a critical force. a-b) The variable-length beam is longer than other beams and the linkage follows the path outside of the central triangle. c-d) Shortening the variable beam now constrains the linkage to the branch inside of the central triangle.

Gating signal propagation in mechanical chains

Building upon the a systematic approach to engineer the configuration space topology for mechanical linkages consisting described in the previous section, we rationally gate mechanical signal propagation along a chain of linkages known as the KL (Kane-Lubensky) chain. Specifically, we chose a 'spinner'-type KL chain which is shown in Figure 6a in gray.

In the spinner phase of the KL chain, a full cycle consists of the soliton traveling back and forth across the chain once, and the KL chain returning to its initial configuration. After one full cycle, each rotor in the KL chain has rotated by 2π , with each rotor rotating by π each time the soliton passes. Here, we will show that these additional components can act as a gate by opening a gap in the full 2π rotation of the KL chain rotors, thereby obstructing the passage of the soliton.

By using the critical value set and region plots, we can determine how to keep or remove critical points in a linkage's configuration space by changing the lengths of the bars in the linkage. Using this ability, we can design a linkage that, when attached to a KL chain, acts as a gate that will block/allow the soliton to fully propagate.

By changing the position of fixed vertex 3, we can switch between the gated and un-gated states of our system. This allows us to control the propagation of a soliton in the Kane-Lubensky chain. When $D < 5r$, the system is in the "red" regime and when $D > 5r$ it is in the "blue" regime. This choice determines whether the KL chain rotors wind around fully or not. Note that the projection of the configuration space in the θ_1 - θ_2 plane never changes shape, but that the change in how the branch points split into hyperbolas determines whether the full range of angles is accessible to the system or not.

Changing the position of that vertex changes a specific length in the linkage. Changing that length causes the critical point in the linkage with $D = 5r$ to decompose in one of two ways. We were able to determine that specific length change due to a general analysis of how to add/remove critical points in the configuration space of a linkage.

We constructed just the gate (highlighted components of the linkage shown Figure 6) out of LEGO pieces, as shown in Figure 7. The specific design was chosen to be compatible with previous experimental realizations of the KL chain. The soliton can fully propagate if each rotor in the KL chain can fully rotate. When testing the LEGO device, we focus on the motion of the blue LEGO piece. In the un-gated state, that blue piece can spin all the way around. In the gated state, the blue piece gets "stuck" and cannot access the angles around $\pi/4$.

These results were also included in our recent publication [1].

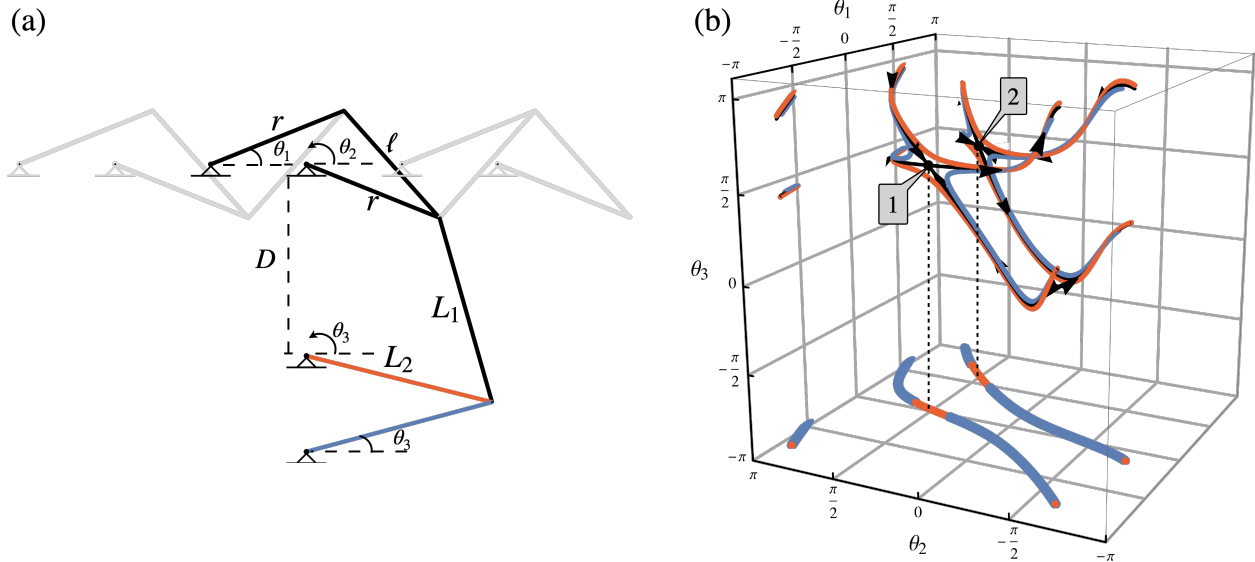


Figure 6. (a) A gated and ungated Kane-Lubensky chain controlled by the length D . (b) The configuration space at and near the critical point as a function of the three rotor angles, and the projection of that configuration space onto the q_1 - q_2 plane. When $D = 4$, the linkage is in the red “un-gated” configuration. When $D = 6$, the linkage is in the blue “gated” configuration.

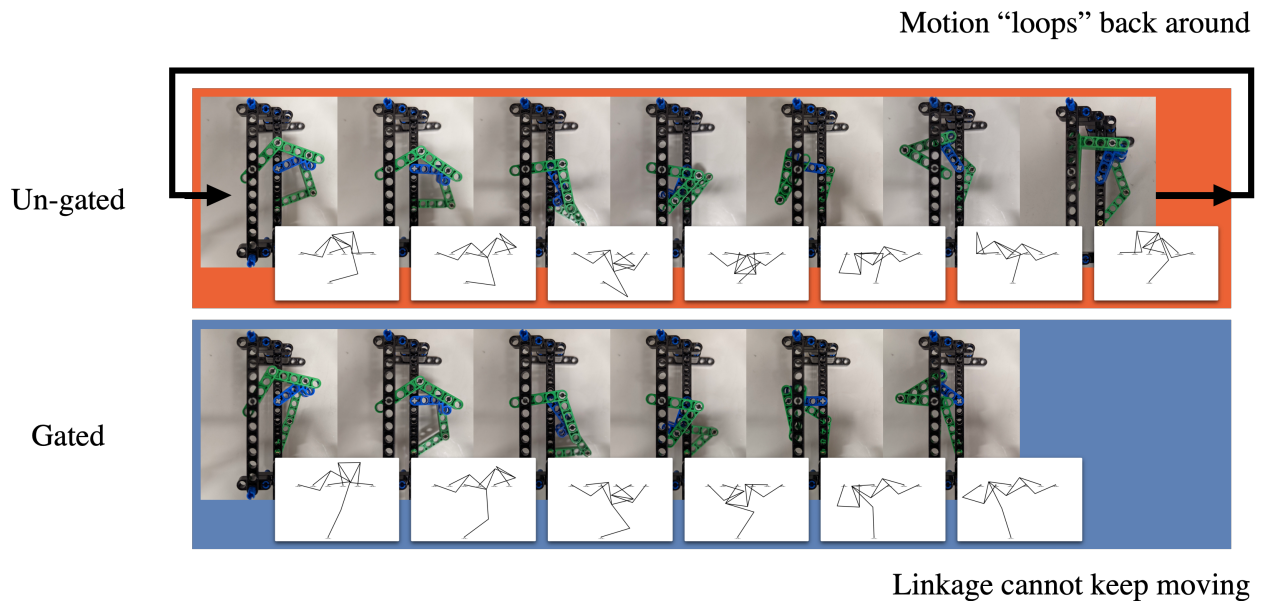


Figure 7. Top row (red): Ungated device made from LEGOs with the corresponding simulation. This device can continue rotating and return back to its initial position, as indicated by the arrow. Bottom row (blue): Gated device made from LEGOs with the corresponding simulation. This device gets stuck in the configuration shown in the last frame and is forced to reverse direction in order to continue moving.

Development of a new approach to (sub-)micro-scale 3D printing

While custom fabrication of macro-scale mechanical devices can easily be achieved using commercial 3D printers, it is important to consider how to extend fabrication of these devices to smaller scales. Two-photon polymerization (TPP) is currently the dominant approach for micro-scale 3D printing, offering resolution down to ~ 100 nm, but it suffers from low volumetric write speeds and very high cost of components ($> \$0.5$ M for a commercial Nanoscribe printer). This cost is dominated by the femtosecond pulsed laser that is required to reach high peak light intensities (~ 1 TW/cm²) and associated advanced optics.

Triplet-triplet annihilation (TTA) offers an alternative pathway to achieve photon upconversion that requires significantly lower peak light intensities (~ 10 mW/cm²), which can be achieved with a continuous wave LED light source and microscope optics. We developed an approach to use TTA for photopolymerization (TTAP), as shown in Figure 8. Green light excites the photosensitizer, which enters its triplet excited state and transfers its energy to a triplet acceptor. Two triplet acceptors annihilate to generate a singlet acceptor, which transfers its energy to a photoinitiator to initiate polymerization. Excitation of the photoinitiator is therefore dependent on light intensity squared, which limits the excitation volume to the ellipsoidal focal point of light, where light intensity is above a critical value.

By using a digital micromirror array device (DMD) to project light patterns, the spot size can be varied to print features of various sizes as shown in Figure 9. These graphs show how the resulting feature size changes with respect to projected spot size for two different exposure times and various light intensities. Low light intensities result in no polymerization with smaller projected spot sizes, while similar intensities overexpose larger projected spots. Features can be printed to match the projected spot size by controlling both light intensity and exposure time, as shown by points laying on the line $y = x$. The smallest features shown measure 0.6 ± 0.1 μm .

Arbitrary 3D designs can be printed at this resolution, as shown in Figure 10. Figure 10a and e show the selected designs as printed by a commercial 3D printer. These designs were sliced into voxels and assigned coordinates as shown in 10b,f and printed using TTAP. Figure 10c-d shows the resulting object printed with 41 mW/cm² intensity and 0.15 s exposure time per voxel. The stair-steps in Figure 10d are spaced by one voxel and measure 0.6 μm in width, showing that submicron resolution is maintained. Figure 10e-h shows that more complex open strut-like features can be printed successfully using the same light intensity and 0.1 s exposure time per spot.

This work was published in the *Journal of the American Chemical Society* in 2022 [2].

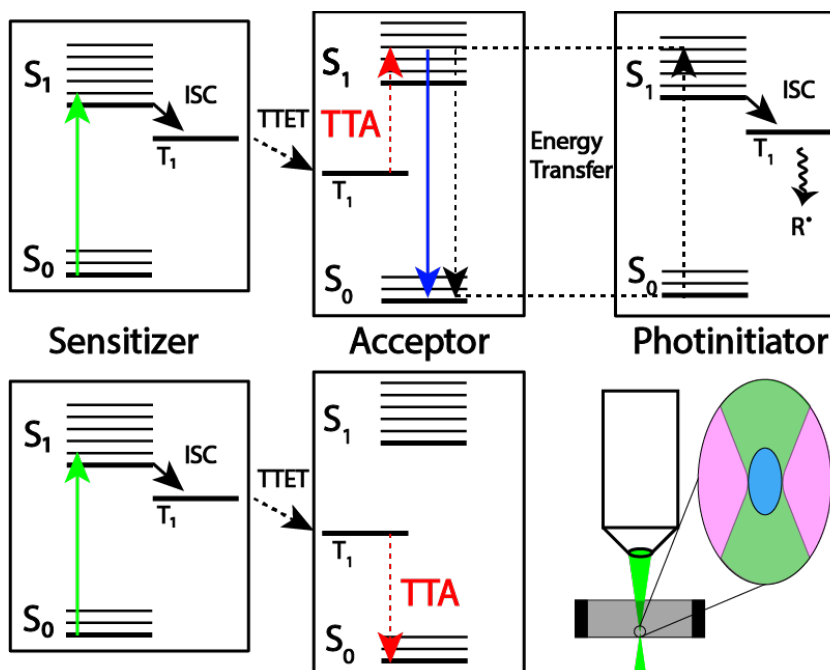


Figure 8. A schematic illustration of triplet-triplet annihilation photopolymerization (TTAP). TTA occurs, then energy is transferred to the photoinitiator ultimately inducing radical generation, which starts polymerization. This polymerization is limited to the focal point of the light beam as it requires a critical concentration of triplet acceptors, which can only be found in regions with sufficiently high light intensity.

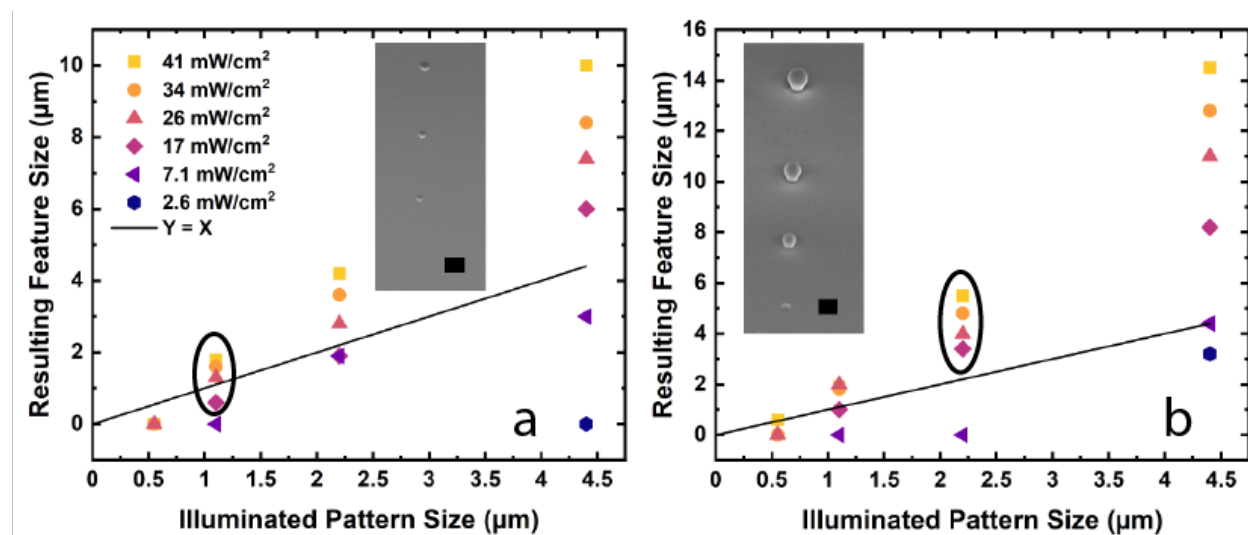


Figure 9. a) Graph of projected light pattern size from micromirror array vs resulting feature size for 0.1 s illumination time. No polymerization was observed for any points laying on the $y = 0$ line. Note that no polymerization was observed under these conditions for 0.55 μm spot size. The line $y = x$ provided to guide the eye. Inset shows the circled features with a scale bar of 5 μm . b) A similar graph of projected light pattern size from micromirror array vs resulting feature size for 0.2 s illumination time. Submicron projection (0.55 μm) yields features measuring 0.6-0.7 μm wide at the highest light intensity.

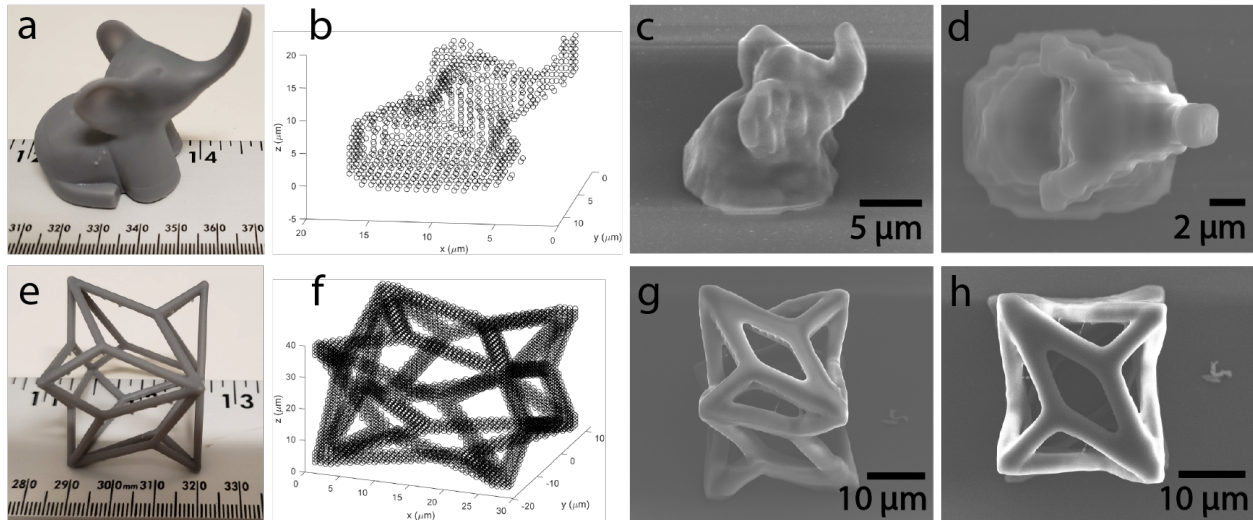


Figure 10. a) Left: elephant model printed using a commercial 3D printer. Ruler shows size in millimeters. b) Distribution of planned exposures after voxelization of the original design. Resulting feature using TTA photopolymerization as visualized using SEM, taken (c) at a 45° tilted angle from the vertical and (d) from directly above. e) Strut design printed using a commercial 3D printer. f) Point distribution after voxelization and resulting feature fabricated by TTA photopolymerization visualized by SEM g) with a 30° tilt angle and h) from directly above.

Marangoni Logic Gates

We employed Marangoni forces between hydrogel nanocomposite disks (HNDs) under patterned light illumination to demonstrate fundamental logic elements (AND, OR, and NOT gates). The HNDs were fabricated using lithographic patterning of polymer films containing gold salt precursors that are photochemically reduced to gold nanoparticles. Circularly-shaped HNDs (diameter $d = 375 \mu\text{m}$), when placed at a planar air-water interface and illuminated with visible light, form a temperature gradient around themselves due to light absorption by the gold nanoparticles and thermal diffusion. A small non-illuminated circular region (with a size nearly equal to that of the HND) serves as a Marangoni optical trap because of the higher surface tension near the cooler part of the HND (Figure 11A). Additionally, when placed in proximity, multiple HNDs experience long-ranged repulsive interactions arising from their individual temperature fields, with a repulsive force that is anticipated to decay as the inverse of particle separation squared (assuming purely diffusive heat transfer) (Figure 11B)

Using Marangoni optical traps and repulsive interactions, we have designed a bistable element in which an HND can be positioned at either side of an “H”-shaped trap (Figure 11C). The HND represents an output signal whose value, ‘0’ or ‘1’, is determined by the position of the input signals and the geometry of the bistable element. For example, for a trap with a smaller side length h , the output signal moves to ‘1’ when the input signal is placed at ‘1’. However, for a larger value of h , the output signal remains at ‘0’ when the input signal is ‘1’.

We examined signal transmission for a variety of trap heights h (Figure 12A) and consistently observed that signal transmits from ‘0’ to ‘1’ when $h \leq 190 \text{ mm}$. As seen in this plot, signal transmission occurs within a few hundred milliseconds. Based on the characterization of the bistable element geometry, we then demonstrated simple logic elements - AND, OR, and NOT gates (Figure 12B).

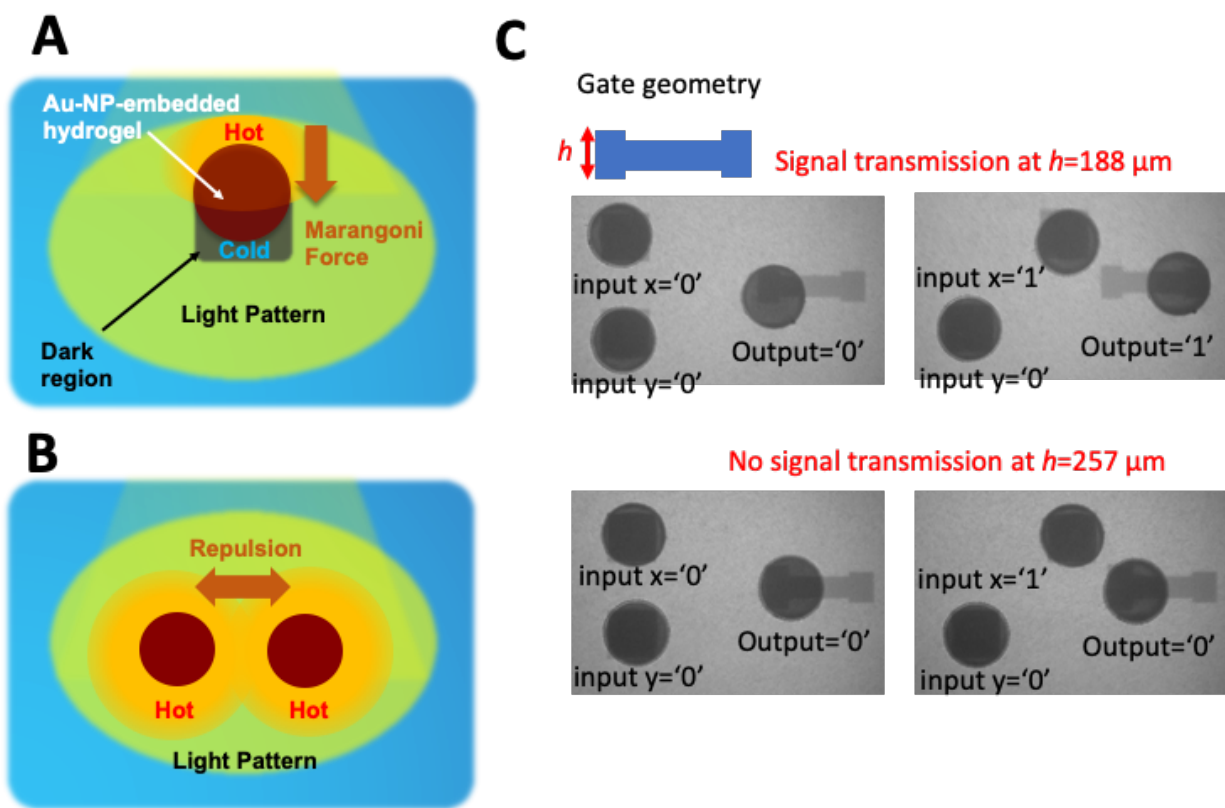


Figure 11. Schematic illustrations of (A) an HND in a Marangoni optical trap and (B) Marangoni repulsion between nearby HNDs. (C) An HND held in a bistable 'output' trap is triggered by the presence of only a single 'input' particle in the closer ('1') state for sufficiently small trap height h (top), but not for larger values (bottom).

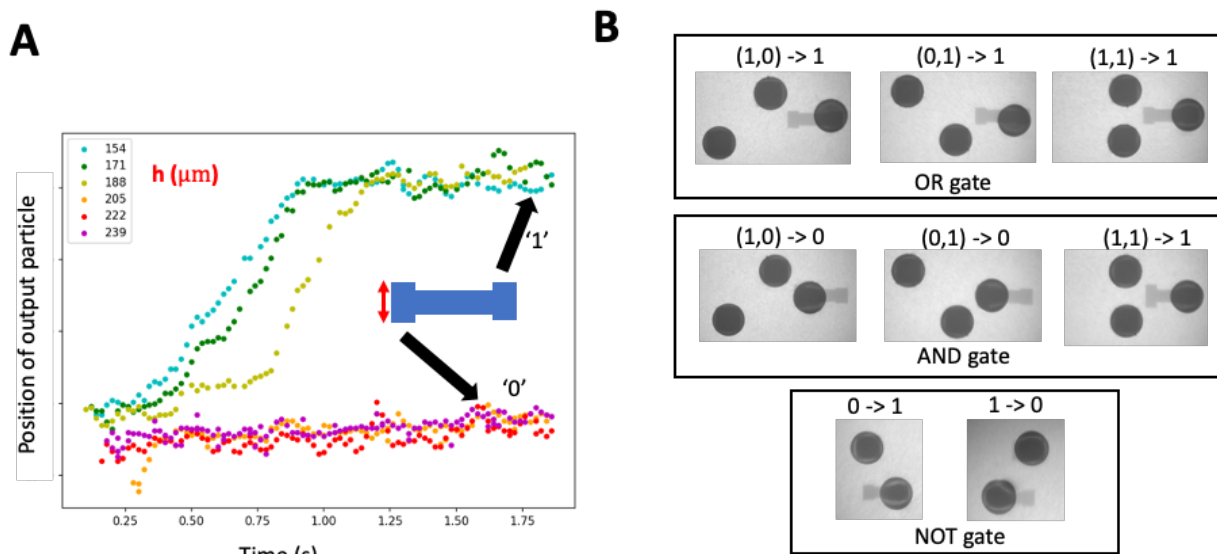


Figure 12. (A) Output particle position vs. time after assigning a single input particle to position 1. (B) Logical OR, AND, and NOT gates achieved with different trap geometries.

Stability of multi-particle Marangoni trap architectures

To engineer nested logic architecture that will allow us to perform complex computations, it is essential to combine multiple logic elements. Because our system relies on both interparticle repulsive potential and Marangoni trapping potential, the stability of the gates is determined by the interplay between these two forces. We find that when multiple logic gates are arranged in a 2D geometry, the long-ranged repulsive “potential” (although the forces between particles are not truly derived from a conservative potential, in the quasi-static case considering only thermal diffusivity, the Marangoni forces can be treated as arising from a pseudo-equilibrium potential) from the neighboring particles can overcome the trapping “potential”, resulting in an escape of one or more particles from corresponding traps. To understand this fundamental limitation, we placed N particles ($N = 2,3,4,5,6$) in equally-spaced traps around a ring geometry and gradually reduced the diameter of the ring. We found that for a critical separation distance D_c , at least one particle becomes unstable in its trap (Figure 13A). Interestingly, we find that D_c increases with the number of particles N , consistent with our expectation that a repulsive potential that scales as $\sim 1/r$ will cause the energy to increase in a superextensive manner with the system size (Figure 13B).

Our experiments demonstrate a new capability of performing logic operations at an air-water interface. Because the repulsive and trapping potential can be precisely tuned by the geometry of illuminated light patterns, the system is promising for designing robust and fast logic gates. The fundamental limitations regarding the stability of the traps indicates that careful considerations are required when scaling the system up to engineer complex logic architecture. These considerations may involve adjusting light intensity, interparticle separation distance, and trap geometry in order to achieve optimal performance while maintaining a stable spatial arrangement of logic gates. We are currently preparing a manuscript describing the findings in this and the previous section [3].

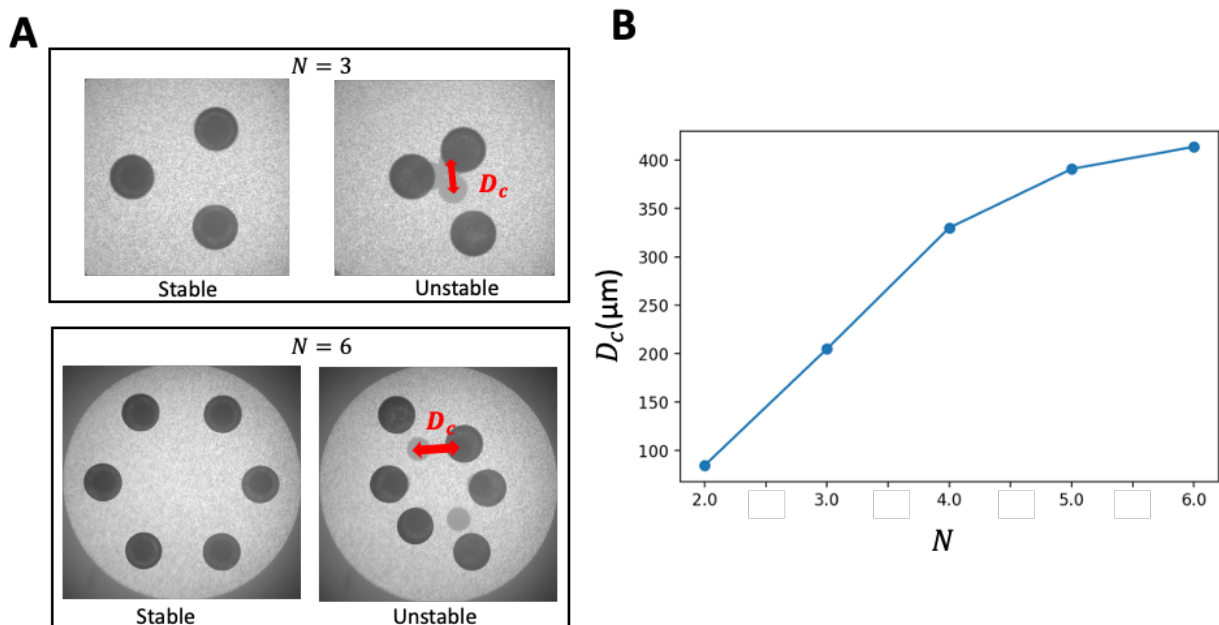


Figure 13. (A) Images showing loss of trap stability upon bringing HNDs closer than a critical value D_c which (B) increases with the number of trapped particles N in a ring.

[1] Berry, M., Limberg, D., Lee-Trimble, M.E., Hayward, R.C., Santangelo, C.D., “Controlling the configuration space topology of mechanical structures”, *Phys. Rev. E.*, 106(5), 055002 (2022). DOI: 10.1103/PhysRevE.106.055002

[2] D. Limberg, J.-H. Kang, R.C. Hayward, “Triplet–Triplet Annihilation Photopolymerization for High-Resolution 3D Printing”, *JACS*, 144, 12, 5226-5232 (2022). DOI: 10.1021/jacs.1c11022

[3] N. Tanjeem, R.C. Hayward, “Interactions between photothermal hydrogel particles in Marangoni optical traps”, manuscript in preparation (2022).