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Major Goals: The overarching goal of the project is to develop new approaches to soft mechanical logic devices that can respond to environmental stimuli and perform non-trivial logical computation. During the one year of the project at UMass Amherst, we focused on two major objectives:

1. Understanding the flow of information in ‘wires’ of mechanically coupled bistable elements
2. Developing a platform to enable studies of interactions between photothermally responsive elements that can be used to build computing architectures

Accomplishments: 1. Understanding the flow of information in wires of mechanically coupled bistable elements

Using a combination of theory and experiment, we developed a model system that allows us to study how deformation propagates within a one-dimensional ‘wire’ of bistable elements, a mechanical analog to the flow of current within a wire in traditional electronic computation. Our prototypical bistable element is a buckled beam formed by constraining a thin beam to adopt an end-to-end distance smaller than its natural length. To mechanically couple a series of beams, we place a Hookean spring between the mid-point of each adjacent beam, as shown in Figure 1c (bottom). In our model (Figure 1a), each beam is itself represented by a pair of springs.

For a symmetric beam, buckling to the left or the right has equal energy. However, if we consider a wire with all beams initially buckled to the left (corresponding to the mid-point of each beam being offset from center along the x-direction by an amount -d) and then try to send a mechanical signal down the length of this wire by pushing on the left-most element, the signal may or may not be able to propagate down the length of the chain depending on the parameters of the system. As summarized in Figure 2, we determined whether the signal propagates the full length of the wire (red color) or becomes arrested at some point (purple). Theory and experiment are in excellent qualitative agreement, both showing full propagation for sufficiently short wires with sufficiently stiff interaction springs, while softening the springs limits the distance of propagation. This is an important trade-off to understand, since some finite compliance of the system will be required to do meaningful computation based on the input signals, but will also limit the number of elements through which the signal can travel in the absence of any mechanism for amplification. Notably, the transition from complete to incomplete propagation occurs at several-fold greater spring stiffness in experiment than in theory; we suspect this reflects the possibility for the experimental springs to undergo lateral buckling, which softens their effective response compared to the measured spring constant.

A bias can intentionally be introduced into the system such that buckling of each element in one direction or the other is energetically preferred. In experiments, this is accomplished by printing the beams with a pre-curved state prior to compressing to their final end-to-end separation. In theory, it is achieved by introducing a torsional spring at the center of each bistable beam element with a preferred angle that matches the configuration in either the state buckled to the left or the right. Although the mechanisms are different in detail, both produce similar energy vs. displacement curves (shown schematically in Figure 3A), and the torsional spring constant in theory is adjusted to most closely match the measured energy curves in experiment. As expected, if the system is biased to prefer buckling to the right (asymmetry < 0), a signal can more easily propagate fully from left to right, since each element releases some stored elastic energy as it flips, providing a built in amplification to the signal. However, if the system is biased to buckle left (asymmetry > 0), it becomes more difficult to propagate the signal. Once again, theory and experiment are in close qualitative agreement.

While asymmetry < 0 is an attractive route to enhance signal propagation, we note that the system must also be reset to the higher energy state to allow for subsequent signals to flow, thus understanding the ‘uphill’ flow of signals is also important. In addition, nominally symmetric systems that do not intentionally include bias are likely in many cases to develop a bias over time due to material relaxation/plasticity during storage.

2. Developing a platform to enable studies of interactions between photothermally responsive elements

As a model stimuli-responsive material system, we have focused on photothermally-responsive hydrogel elements formed by lithographic patterning of polymer films containing gold salt precursors that are photochemically reduced to gold nanoparticles. While we ultimately hope to develop architectures similar to those studied in Objective 1, fabrication of such structures from stimuli-responsive materials on sufficiently small size scales to yield responses on time-scales suitable for our studies (e.g., ~ 100 s or less to enable repeated cycling through many operations within accessible experimental windows) has proven challenging to date. However, we have unexpectedly discovered a promising system of photothermal hydrogel particles that hold promise for soft logic devices very similar to those based on bistable mechanical elements, although the interactions differ in detail.

Specifically, we have found that hydrogel nanocomposite disks (HNDs) fabricated in this way and placed at a planar air-water interface are subject to strong Marangoni forces when illuminated with visible light, thanks to absorption of light by the nanoparticles, localized heat generation, and resulting formation of a steady-state temperature gradient. For a single HND under uniform illumination, the temperature field is azimuthally symmetric and therefore the particle experiences no net force. However, two particles in proximity will experience a long-range repulsive interaction (decaying as the inverse of particle separation squared, assuming purely diffusive heat transfer). Similarly, spatial variations in light intensity can give rise to unbalanced forces on the particle, driving motion.

In particular, we find that a non-illuminated region of similar to the HND size serves as a Marangoni optical trap, since a disk at the edge of the region will experience a stronger force pulling it back toward the cooler side of the disk in the non-illuminated trap center. For a sufficiently large trap (with a height approaching the disk diameter), this trapping is stable, allowing the position of the HND to be controlled over time by spatially translating the pattern of light. For a trap with one dimension much smaller than the disk diameter, HNDs do not remain trapped. Remarkably, however, traps with intermediate dimensions give rise to sustained oscillation of the HND around the trap center (Figure 4B). A model that simplifies the HND as a two element lumped thermal system (Figure 4E-F) is able to capture the observed behavior very well and clarifies the underlying mechanism. In short, due to the lag time required to reach thermal steady state, the Marangoni restoring force continues to pull the particle past the mid-point of the trap before the temperature gradient, and resulting force, have time to reverse direction, thus leading to sustained oscillation.

When two trapped particles are held in sufficiently close proximity, the repulsive Marangoni forces between them causes the particles to be pushed off-center in their respective traps. For traps in the oscillatory regime, this results in a frequency locking and anti-phase coupling between the two oscillators (Figure 5).

This system holds potential for the realization of soft mechanical computing elements in at least two respects. First, it is possible to design bistable traps, wherein each HND has two mechanically stable positions of either equal or unequal energy, providing an analog to the bistable beams discussed in Objective 1. Coupled with the repulsive Marangoni interaction, which effectively provides a non-linear interaction spring between HNDs, it should be possible to realize ‘wires’ and more advanced architectures (i.e., logic gates) that mirror the behavior of those discussed above. Second, there has been considerable interest in developing systems of coupled oscillators to perform computational tasks such as pattern recognition, and our platform provides a simple route to define collections of osc

Training Opportunities: The project supported the research, training, and professional development of three PhD students at UMass Amherst (Michelle Berry, Physics, Demi Moed and Hyunki Kim, Polymer Science and Engineering), one visiting PhD student (Yong-Jae Kim, Chemical and Biomolecular Engineering, Korea Advanced Institute of Science and Technology), and a postdoctoral fellow (Ji-Hwan Kang, Polymer Science and Engineering, UMass Amherst).

Results Dissemination: The results are being disseminated through peer reviewed publications (one manuscript accepted, one other currently in preparation) as well as an invited presentation by the PI at the University of Florida Soft Matter Symposium in October 2019, as well as presentations by PhD student Michelle Berry at the 2020 Gordon Conference on Multifunctional Materials and Structures (poster) and the American Physical Society 2020 March Meeting (talk presented via videoconference).

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: The PI and co-PI have had interactions with Phil Buskohl, AFRL, related to the design of multistable architectures and structures that exhibit mechanical computation.

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Project Contribution:
National Academy Member: N

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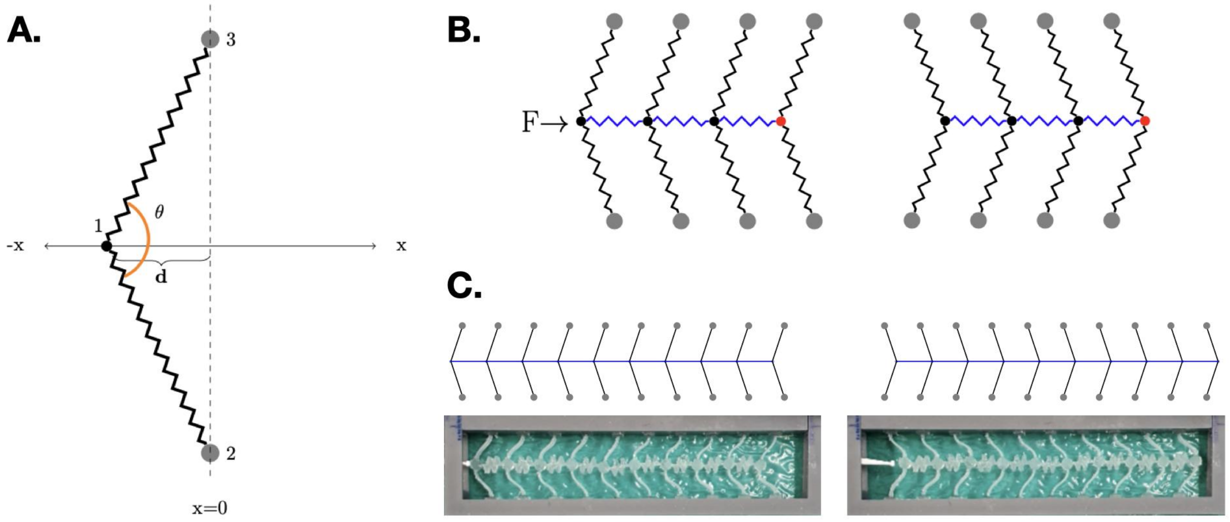

Figure 1. (A) In simulations, the bistable beam is made of two linear springs (black) with stiffness ***s*** joined together by a freely rotating joint (vertex 1). At vertex 1, there is a torsional spring (orange) with stiffness *t* and resting angle θ. The beam is in a stable position when vertex 1 is located at $x = \pm d$. If the torsional spring has a nonzero stiffness, the beam will be biased towards one of the two stable positions. **(B)** A wire is a series of bistable beams connected together by linear interaction springs (blue). The current traveling down the wire is an external push that exerts a force ***F*** on the first beam in the wire. This force causes each beam to transition from one stable position to the other. We determine if the current traveled through the entire wire by determining the displacement of the midpoint of the last beam in the wire (the red vertex). **(C)** A simulated (top) and 3D printed (bottom) wire. On the left, a signal is about to be sent down the wires. On the right, the signal has traveled through both wires.

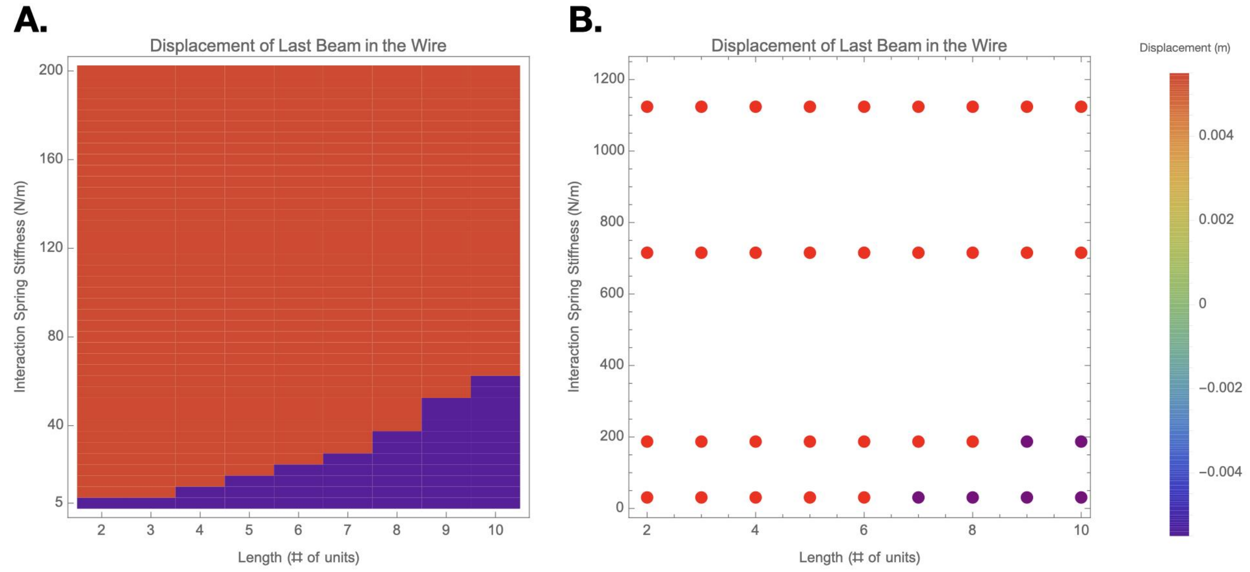

Figure 2. Results for signal propagation when varying the stiffness of the interaction springs and the length of the wire. **(A)** shows results from simulations, and **(B)** shows results from experiments. For both simulations and experiments, the stable positions of the bistable beams are at $x = \pm\ 0.0055$ m. A displacement of + 0.0055 m (red) means that the signal traveled through the entire wire. A displacement of - 0.0055 m (purple) means that the signal stopped traveling at some point along the wire.

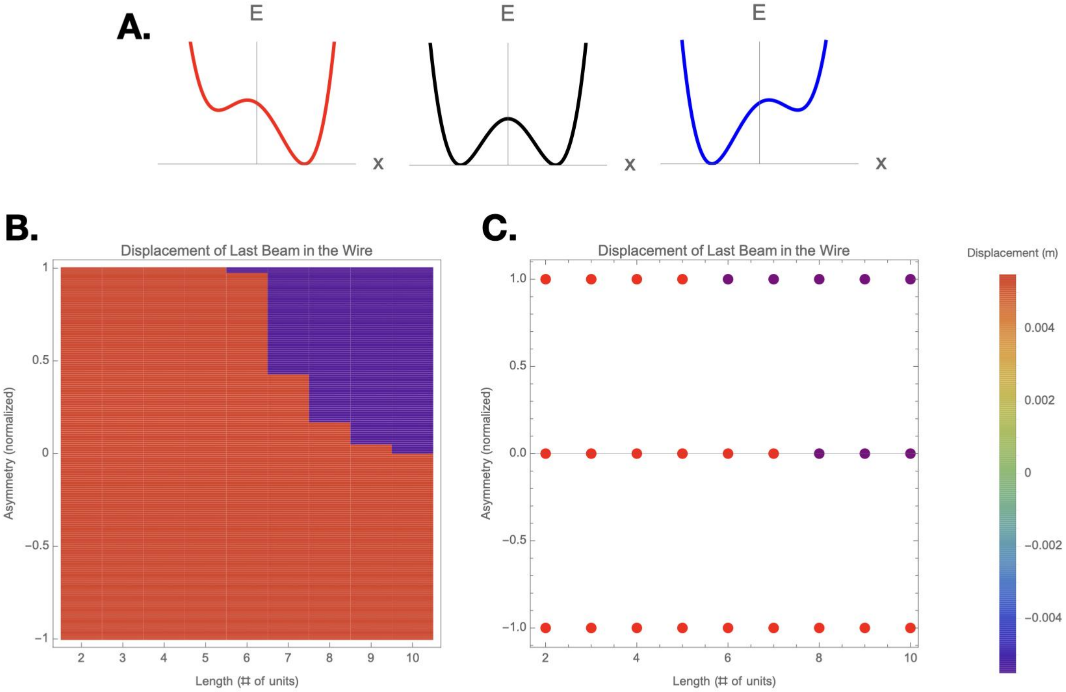

Figure 3. (A) By changing the torsional stiffness *t* and rest angle θ of the torsional springs on the bistable beam, the potential energy of the beam changes. The asymmetry of the bistable beam is defined by the energy difference between the two minima. When the asymmetry is negative (shown in red), sending a current down the wire is like pushing downhill. When the asymmetry is positive (shown in blue), sending a current down the wire is like pushing uphill. The asymmetry is zero when the potential is completely symmetric (shown in black). **(B)** Simulation and **(C)** experimental results from sending a signal down a wire when varying the asymmetry of the bistable beams and the length of the wire. For both simulations and experiments, the stable positions of the bistable beams are at $x = \pm 0.0055$ m. A displacement of + 0.0055 m (red) means that a current traveled through the entire wire. A displacement of - 0.0055 m (purple) means that the current stopped traveling at some point along the wire.

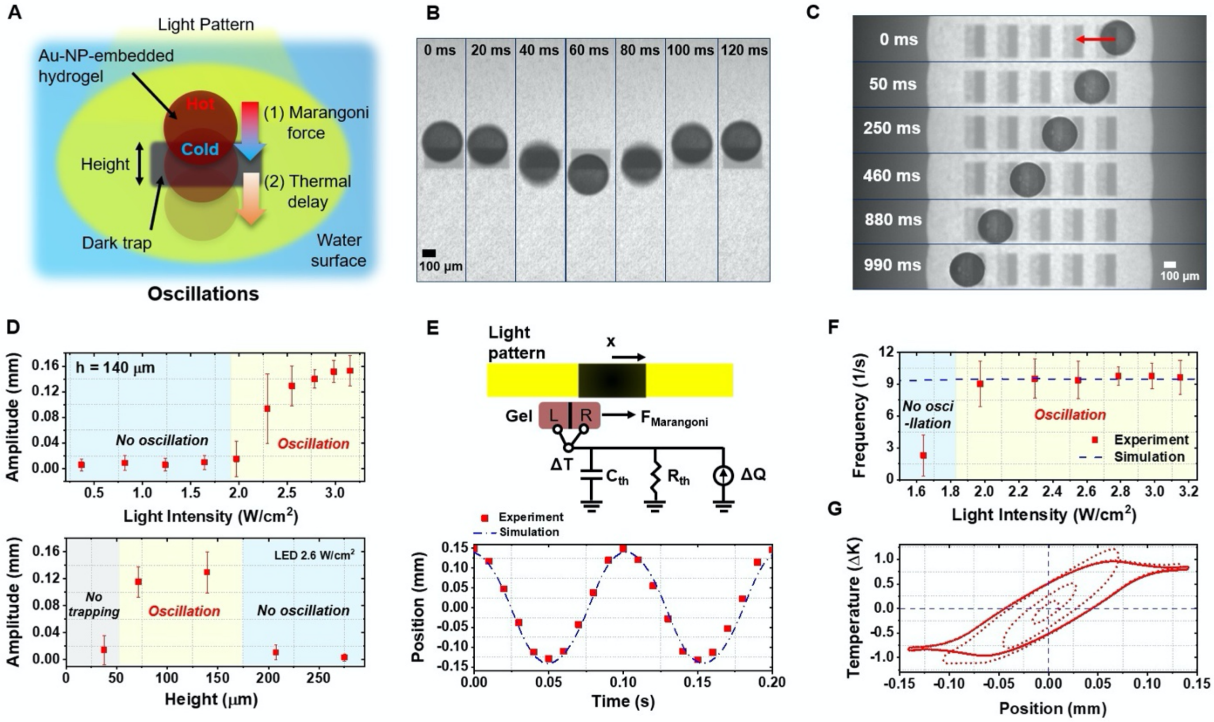

Figure 4. Oscillation of HNDs confined to Marangoni traps. (**A**) Schematic diagram of the trap geometry that drives oscillatory motion. Time-lapse images of an HND undergoing (**B**) oscillation within a symmetric trap and (**C**) directed, stochastic hopping along a row of anisotropic traps (Movie S1). (**D**) Amplitude of HND oscillations with varying light intensity and trap height. (**E**) Schematic diagram of the model (top) along with a comparison of experimental and simulated displacement for two cycles of oscillatory motion (bottom). LED power = 2.8 W/cm^2, trap height 140 µm. (**F**) Comparison between simulated and experimental frequencies with varying light intensity. (**G**) Simulated phase portrait of the oscillator showing position vs. temperature difference.

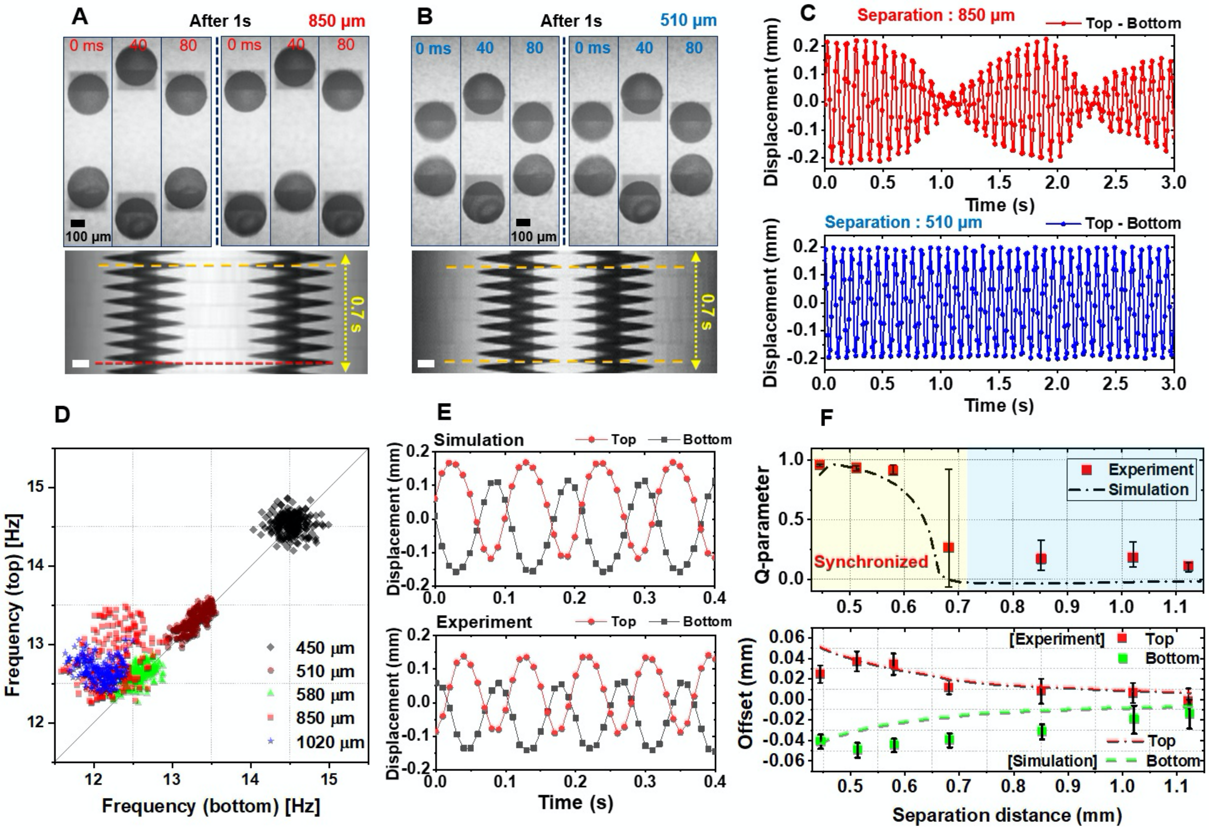

Fig. 2. Distance-dependent coupling of two oscillators. Time-lapse images showing oscillation of two HNDs confined to traps with center-to-center distances of 850 μm (**A**) and 510 μm (**B**). Bottom images are kymographs (0.7 s) of the coupled oscillations. The yellow (red) dashed lines indicate anti-phase (in-phase) synchronization. Scale bars are 100 μm. Time step for each image is 40 ms. (**C**) Differences in displacements of the coupled HNDs with separation distances of 850 μm (top) and 510 μm (bottom). (**D**) Fast Fourier transform results of coupled HND oscillations with varying separation. (**E**) Temporal motions of the HNDs from simulation and experiment on the separation distance of 510 μm. (**F**) Q-parameter and center offsets of the oscillations with respect to HND separation.