

Boulder, CO



iLCEC 2023



Welcome Reception:



*University Memorial Center, South Terrace
Sunday (October 8), 5:15-7:00
Drinks, heavy appetizers provided*

Conference Banquet:



*Chautauqua Dining Hall (900 Baseline Road, Boulder CO 80503)
Tuesday (October 10), 6:00-8:00
Buses leave UMC promptly at 5:30
Buses return to UMC and conference hotels at 8:00
Meal provided, cash bar*

SUNDAY, OCTOBER 8 (University Memorial Center, Glenn Miller Ballroom)

12:00-13:00 Registration

LCE and Robotics I – Prof. White

13:00-13:35 **Shengqiang Cai** (UCSD)
"Recent applications of liquid crystal elastomers"

13:35-14:00 **Carlos Sánchez-Somolinos** (CSIC-UNIZAR)
"Enabling Functionality and Miniaturization in Soft Robotic LCEs through Additive Manufacturing"

14:00-14:25 **Danqing Liu** (Eindhoven University of Technology)
"Interactive liquid crystal polymers for 2D soft robotic functions"

14:25-14:40 **Joselle McCracken** (University of Colorado, Boulder)
"Thickness-Dependent Topography of Liquid Crystalline Elastomer Conical Actuators Under Compressive Load"
J.M. McCracken, T.J. White

14:40-14:55 Break (Coffee and Snacks available)

Computational & Experimental Mech I – Prof. Liu

14:55-15:30 **Kaushik Bhattacharya** (California Institute of Technology)
"Mechanical properties of isotropic-genesis polydomain nematic elastomers"

15:30-15:45 **Daniel Duffy** (University of Cambridge)
"LCE Cones: Lifting, Loading, and Buckling"
D. Duffy, J.S. Biggins, J.M. McCracken, T.S. Hebner, T.J. White

15:45-16:05 **Renee Zhao** (Stanford)
"Liquid Crystal Elastomer – Liquid Metal Composite: Ultrafast, Untethered, and Programmable Actuation by Induction Heating"

16:05-16:20 **Milan Wilborn** (Harvard University)
"Inverse Gaussian Shape Morphs based on LCE films"
M. Wilborn, P. Qu, S. Li, F. Streicker, J. Aizenberg

16:20-16:55 **Shu Yang** (University of Pennsylvania)
"Programming shape morphing and chirality switching by liquid crystalline elastomer microparticles"

16:55-17:10 **Morgan Barnes** (University of Cambridge)
"Microstructures and Surface Instabilities in Liquid Crystal Elastomers"
M. Barnes, F. Feng, J. Biggins

17:10-19:00 **Welcome Reception – South Terrace, University Memorial Center**
Heavy appetizers and drinks (provided)

MONDAY, OCTOBER 9 (University Memorial Center, Glenn Miller Ballroom)

7:30-8:30 Registration

Molecular Engineering of LCEs – Prof. White8:30-9:15 **Dirk J. Broer** (Eindhoven University of Technology)
"On the history of reactive mesogens"9:15-9:40 **Javier Read de Alvaniz** (UC Santa Barbara)
"Exploration of white-light responsive liquid crystal elastomers"9:40-10:00 **Emily Davidson** (Princeton University)
"Controlling Phase Behavior With Precise Liquid Crystalline Oligomers"
*C.L.C. Chan, S. Maguire, E. Ostermann, E. Davidson*10:00-10:15 **Charlie Lindberg** (University of Chicago)
"Relax, everything is under control: Leveraging modularity for precision dynamic bond placement in liquid crystal elastomers"
*C. Lindberg, E. Ghimire, C. Chen, S. Lee, N.D. Dolinski, J.M. Dennis, S. Wang, J.J. de Pablo, S.J. Rowan*10:15-10:30 **David Kennedy** (University of Colorado, Boulder)
"Molecular Engineering of Mesogen Interactions in Liquid Crystalline Elastomers"
D. Kennedy, T.J. White

10:30-10:45 Break

10:45-11:20 **Yue Zhao** (Université de Sherbrooke)
"Actuation Functions Enabled by Crosslinker in Liquid Crystal Elastomer Structure"11:20-11:45 **Taylor Hebner** (University of Oregon)
"Spatial Patterning of Material Properties in Liquid Crystalline Elastomers for Complex and Dynamic Actuation"
*T. Hebner, C.N. Bowman, T.J. White*11:45-12:00 **Tim White** (University of Colorado, Boulder)
"Mark Warner's Contributions to Our Field and the ILCEC"12:00-13:00 **Conference Photograph @ 12:00, Lunch, Buffet Lunch (Provided), Hang Posters****Nonlinear Deformation & Impact Resistance – Prof. Davidson**13:00-13:35 **Helen Gleeson** (University of Leeds),
"Auxetic liquid crystal elastomers: how they work"
*H.F. Gleeson, D. Mistry, T. Raistrick, A. Street, M. Reynolds, S. Berrow, Z. Wang*13:35-14:00 **Eugene Terenjev** (University of Cambridge)
"Pressure sensitive adhesion of nematic elastomers"14:00-14:25 **Takuya Ohzono** (AIST)
"Optimal conditions for efficient photomechanical response in nematic elastomers"
*T. Ohzono, E. Koyama*14:25-14:50 **Thao (Vicky) Nguyen** (Johns Hopkins University)
"Modeling the Viscoelastic Behavior of Liquid Crystal Elastomers"14:50-15:15 **Kenji Urayama** (Kyoto University)
"Exploring Nonlinear Elasticity in Nematic Elastomers: Beyond Simple Stretching?"

15:15-15:30 Break

To (Be) LCE or Not (To Be) LCE? – Prof. Gleeson15:30-15:55 **Ivan I. Smalyukh** (University of Colorado, Boulder)
"Towards Hopfionic Haptics"
*I.I. Smalyukh, J. Peixoto, D. Hall, D.J. Broer, D. Liu*15:55-16:20 **Albert Schenning** (Eindhoven University of Technology)
"Thermoplastic Liquid Crystal Elastomers"
*A. Schenning, D.J. Mulder, S. Lugger*16:20-16:45 **Ryan Hayward** (University of Colorado, Boulder)
"High-performance photomechanical materials via polymer-templated growth of aligned microcrystal arrays"16:45-17:00 **Hongye Guo** (University of Cambridge)
"Main-chain nematic side-chain smectic composite liquid crystalline elastomers"
H. Guo, M.O. Saed, E.M. Terentjev

TUESDAY, OCTOBER 10 (University Memorial Center, Glenn Miller Ballroom)

7:30-8:30 Registration

3-D Printing of LCE – Prof. Lagerwall

8:30-8:55 **Howon Lee** (Seoul National University)
"Programming Molecular Order of a Liquid Crystal Elastomer with Magnetic-Field-Assisted DLP Printing"

H. Lee, Y. Wang, J. An, H. Kim, S. Ko

8:55-9:15 **Devin Roach** (Sandia National Laboratories)
"Multi-Planar 4D Printing and Deformation Control Strategies for Liquid Crystal Elastomers"

D. Roach, J. Herman, A. Cook, T. White, B. Kaehr

9:15-9:35 **Suk-kyun Ahn** (Pusan National University)
"Dual Morphing of Direct-Ink-Written Liquid Crystal Elastomers Under Heat and Moisture"

K. Kim, K. Kim, S. Ahn

9:35-9:50 **Klaudia Dradrach** (University of Cambridge)
"3D-printed flat sheets of LCE morphing into intrinsically curved folds"

K. Dradrach, F. Feng, M. Barnes, M. Zmyslony, J. Biggins

9:50-10:05 **Wei-Ting Hsu** (National Taipei University of Technology)
"The fabrication of liquid crystal networks structures using two-photon technology"

W.-T. Hsu, H.-J. Chou, Y.-C. Cheng

10:05-10:20 **Caitlyn Krikorian (Cook)** (Lawrence Livermore National Laboratory)
"Light driven morphing of printed liquid crystal elastomers"

C. Krikorian, M. Ford, D. Porcincula, R. Telles-Arriaga, J. Mancini, Y. Wang, M.H. Rizvid, C.K. Loeb, B. Moran, J.B. Tracy, El. Lee, J. Lewis, S. Yang

10:20-10:35 Break

Hierarchical Structure in LCE – Prof. Schenning

10:35-11:00 **Jan Lagerwall** (University of Luxembourg)
"Cholesteric Liquid Crystal Elastomer Sheets and Fibers for Mechanochromic Strain Sensing"

Y. Geng, R. Kizhakkidathazhath, J.P.F Lagerwall

11:00-11:25 **Ana Almeida** (Universidade Nova de Lisboa)
"Cellulose-based anisotropic networks for moisture and temperature time sensors"

A. Almeida, N. Monge, J.P. Canejo, P.L. Almeida, M.J. Godinho

11:25-11:40 **Yoo Jin Lee** (Texas A&M University)
"Self-Assembled Microactuators Using Chirality of Liquid Crystal Elastomers"

Y.J. Lee, M.K. Abdelrahman, M.S. Kalairaj, T.H. Ware

11:40-11:55 **Alexis Phillips** (University of Colorado, Boulder)
"Color change in cholesteric liquid crystalline elastomers"

A.Phillips, J. Chen, T.J. White

11:55-13:00 **Buffet Lunch (Provided), Poster #1-30 Presenters Available for Discussion 12:30-13:00****LCE and Motility in Robotics – Prof. Kuenstler**

13:00-13:25 **Ravi Shankar** (University of Pittsburgh)
"Motility from Liquid Crystallinity"

13:25-13:50 **Antonio DeSimone** (The Biorobotics Institute, Scuola Superiore Santanna)
"Morphing and motility of LCE films"

A.DeSimone, L. Teresi

13:50-14:10 **Hao Zeng** (Tampere University)
"Self-Oscillating Materials: Enabling Self-Regulated Robotic Functions"

J. Yang, Z. Deng, Y. Nemat, H. Zeng

14:10-14:25 **Jacopo Movilli** (Harvard University)
"Encoding life-like multimodal locomotion in photo-responsive microstructures"

J. Movilli, A. Wilborn, J.T. Waters, F. Stricker, A. Balazs, J. Aizenberg

14:25-14:40 **Mason Zadan** (Carnegie Mellon University)
"Wireless and Thermoelectric Actuation Methods for Liquid Crystal Elastomer Based Soft Robots"

M. Zadan, J. Wang, Y. Song, D.K. Patel, Z. Li, L. Yao, S. Kumar, C. Majidi

14:40-14:55	Sean Lee (The University of Chicago) "Soft liquid crystal elastomer with integrated stretchable Joule heater" <i>S. Lee, S.J. Rowan, S. Wang</i>
14:55-15:10	Break
LCE and Material Processing – Prof. Shankar	
15:10-15:35	Atsushi Shishido (Tokyo Institute of Technology) "Photoalignment patterning of liquid crystals by scanning wave photopolymerization"
15:35-16:00	Zachariah A. Page (The University of Texas at Austin) "Liquid Crystalline Elastomers as Photoswitchable Adhesives" <i>Y. Wu, B.D. Clarke, K.M. Liechti, Z.A. Page</i>
16:00-16:20	Alexa Kuentler (University of Illinois Urbana-Champaign) "Liquid Crystal Elastomer Nanocomposites as a Platform for Optically- Addressable Materials"
16:20-16:35	Lovish Gulati (Max Planck Institute for Medical Research) "3D director alignment of liquid crystalline structures with magnetic fields" <i>L. Gulati, F. Giesselmann, P. Fischer</i>
Poster Session	
16:35-17:20	Poster Presentations - #1-30
17:30	Transit via bus from UMC to Chautauqua Dining Hall
18:00-20:00	Conference Banquet (Chautauqua Dining Hall) <i>Appetizers and dinner (provided)</i> <i>Cash bar for alcoholic beverages</i>

WEDNESDAY, OCTOBER 11 (University Memorial Center, Glenn Miller Ballroom)

7:30-8:30 Registration

Emerging Frontiers in LCE – Prof. Li8:30-9:05 **Arri Priimagi** (Tampere University)
"Liquid crystal network actuators that "adapt" (?), "self-regulate" (?) and "learn" (?)"9:05-9:30 **Chinedum Osuji** (University of Pennsylvania)
"Limit Cycle LC Elastomer Actuation for Temperature Regulation and Programmed Thermal Transport"*C. Osuji, Z. Liu, Y. Zhao*9:30-9:45 **Asaf Dana** (Texas A&M)
"Liquid crystal elastomer based entangled active matter"*A.Dana, M.K. Abdelrahman, T.H. Ware*9:45-10:00 **Stuart Berrow** (University of Leeds)
"The Effect of Spacer Length on the Properties of Auxetic Liquid Crystal Elastomers"*S. Berrow, R.J. Madle, H.F. Gleeson*10:00-10:15 **Friedrich Stricker** (Harvard John A. Paulson School Of Engineering And Applied Sciences)
"Light driven liquid crystalline actuators with multiple phase transitions"*F. Stricker, M. Wilborn, J. Movilli, J. Aizenberg*10:15-10:30 **Laurens Theobald de Haan** (South China Normal University), G. Zhou
"Soft electricity-responsive liquid crystal polymer materials for switchable surfaces"*L.T. Haan, G. Zhou*

10:30-10:45 Break

Biology and Bioinspiration – Prof. Priimagi10:45-11:10 **Taylor Ware** (Texas A&M University)
"Liquid Crystal Elastomers: Artificial Muscles and Collective Action"11:10-11:35 **Kelly Burke** (University of Connecticut)
"Synthesis of Liquid Crystalline Networks for Biomaterials Applications"*A. Tulli IV, K.A. Burke*11:35-11:50 **Nathaniel Skillin** (University of Colorado, Boulder)
"Stiffness anisotropy coordinates supracellular contractility driving long-range myotube-ECM alignment"*N. Skillin, B.E. Kirkpatrick, K.M. Herbert, B.R. Nelson, G.K. Hach, K.A. Gunay, R.M. Khan, F.W. DelRio, T.J. White, K.S. Anseth*11:50-12:05 **Zixuan Deng** (Tampere University)
"A light-fueled cilium"*Z. Deng, H. Zhang, A. Priimagi, H. Zeng*12:00-13:00 **Buffet Lunch (Provided), Poster #1-30 Presenters Available for Discussion 12:30-13:00****LCE Materials Chemistry and Processing – Prof. Osuji**13:00-13:25 **Christopher Bowman** (University of Colorado, Boulder)
"Reconfigurable Shape Change in Liquid Crystalline Elastomers Enabled by Stimulus Orthogonality in Dynamic Covalent Networks"*T.S. Hebner, T.J. White, C.N. Bowman*13:25-13:50 **Min-Hui Li** (Institut de Recherche de Chimie Paris, CNRS, Chimie ParisTech, Université PSL)
"Electroactive Bi-functional Liquid Crystal Elastomer Actuators"*Min-Hui Li, G. Liu, Y. Deng, G. Nguyen, C. Vancaeyzeele, A. Bulet, F. Vidal, C. Plesse*13:50-14:05 **Kristin Lewis** (University of Colorado, Boulder)
"Supramolecular Liquid Crystalline Elastomers"*K. Lewis, J. Hoang, S. Aye, D. Kennedy, T.J. White*14:05-14:20 **Elina Ghimire** (University of Chicago)
"Trainable liquid crystal elastomers via the controlled synthesis and dynamic covalent exchange of aza-Michael reactions"*E. Ghimire, C.A. Lindberg, T.D. Jorgenson, C. Chen, N.D. Dolinski, S.J. Rowan*14:20-14:35 **Jin-Hyeong Lee** (Pusan National University)
"Body Temperature Actuation of Exchangeable Liquid Crystal Elastomers"*J.-H. Lee, D.-G. Kim, S.-K. Ahn*

14:35-14:50	Hongshuang Guo (A. Priimagi presenting) (Tampere University) "Halogen-bonded shape memory liquid crystal polymer" <i>H. Guo, R. Puttreddy, H. Zeng, A. Priimagi</i>
14:50-15:05	Break
Computational & Experimental Mech II – Prof. Burke	
15:05-15:30	Rafael Verduzco (Rice University) "Understanding the effect of liquid crystal content on the phase behavior and mechanical properties of liquid crystal elastomers" <i>M. Barnes, S. Cetinkaya, A. Ajnsztajn, R. Verduzco</i>
15:30-15:55	Gregor Skačej (University of Ljubljana) "Photo-responsive liquid crystal elastomers: A molecular Monte Carlo study" <i>G. Skačej, L. Querciagrossa, C. Zannoni</i>
15:55-16:15	Kai Yu (University of Colorado, Denver) "Enhanced Energy Dissipation of 3D Printed Liquid Crystal Elastomers and Their Material Structures"
16:15-16:30	Thomas Raistrick (University of Leeds) "Direct and indirect evidence of biaxial order in auxetic liquid crystal elastomers" <i>T. Raistrick, Z. Wang, A. Street, M. Reynolds, Y. Liu, R. Mandle, H.F. Gleeson</i>
16:30-16:35	Closing Remarks

International Liquid Crystalline Elastomer Conference
October 8-11, 2023
Boulder, CO USA

Abstracts

- **Oral presentations (listed in order)**
- Poster presentations

Recent Applications of Liquid Crystal Elastomers

Shengqiang Cai

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Liquid crystal elastomers have been intensively explored in various applications in the recent decade. As a stimuli-responsive material, liquid crystal elastomer has been used to construct soft robots of different forms, active morphing structures and autonomous devices. Moreover, the unusual mechanical properties of liquid crystal elastomer arising from its special molecular structures have been recently exploited for some novel applications such as adhesion, shock adsorption and soft dielectric actuator. This talk will demonstrate some recent applications of liquid crystal elastomer recently explored in my group, including liquid crystal elastomer based multifunctional tensegrity structures, liquid crystal elastomer base adhesive, and electrically active liquid crystal elastomer. I will also discuss our understandings of the performance of liquid crystal elastomers in those applications.

Enabling Functionality and Miniaturization in Soft Robotic LCEs through Additive Manufacturing

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The ability to structure smart materials in three dimensions can provide soft robotic systems with sensing and actuation capabilities, as well as other advanced functionalities. In particular, digital fabrication techniques can achieve all of this with high reproducibility, scalability, and flexibility in design, making them a key enabling technology for the future development of soft robotic systems with potential impact in various technological areas, including microfluidics, photonics, and biomedicine. Liquid crystalline elastomers (LCEs) are particularly interesting materials in soft robotics as they have demonstrated great potential for implementing complex shape-morphing structures when exposed to stimuli such as heat, light, or humidity. Recent developments in LCE fabrication have enabled the creation of actuators and devices with digitally defined director patterns, enabling programmed shape morphing. [1-3] However, challenges remain, such as the limited availability of functionalities and the miniaturization of structures. This lecture presents recent efforts carried out at the Advanced Manufacturing Laboratory, with an emphasis on advancing new functionalities and achieving finer feature sizes. [4-6]

References

- [1] CP. Ambulo et al. ACS Appl Mater Interfaces 9, 37332, (2017).
- [2] M. López-Valdeolivas et al. Macromol Rapid Commun 39, 1700710 (2018).
- [3] A. Kotikian et al. Adv Mater 30, 1706164 (2018).
- [4] M. Javadzadeh et al. Adv. Mater. 35, 2209244 (2023).
- [5] L. Ceamanos, et al., J. Mater. Chem. B, 11, 4083-4094 (2023).
- [6] L. Ceamanos, et al., ACS Appl. Mater. Interfaces, 12 (39), 44195–44204 (2020).

Acknowledgments

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Interactive liquid crystal polymers for 2D soft robotic functions

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We propose the use of a liquid crystal polymer network for soft robotics where the various molecular accessories are assembled in the two dimensions of a coating. For instance, the LCN surface deforms dynamically into a variety of pre-designed topographic patterns by means of various triggers, such as temperature, light and the input of electric field¹. These microscopic deformations exhibit macroscopic impact on, for instance, tribology, haptics, laminar mixing of fluids in microchannels and directed cell growth. Another robotic-relevant function we brought into the LCN coating is its capability to secrete liquids under UV irradiation or by an AC field². This controlled release is associated with many potential applications, including lubrication, controlled adhesion, drug delivery, and agriculture, antifouling in marine and biomedical devices, personal care and cosmetics. With this we bring together a tool box to form two dimensional soft robots designed to operate in area where man and machine come together.

References

- [1]. Dongyu Zhang, Jacques Peixoto, Yuanyuan Zhan, Mert O Astam, Tom Bus, Joost JB van der Tol, Dirk J Broer, Danqing Liu*, *Advanced Materials*, 2023, 35, 2209729.
- [2]. Danqing Liu, Nicholas B Tito, Dirk J Broer, *Nature communications*, 8:1526.

Thickness-Dependent Topography of Liquid Crystalline Elastomer Conical Actuators Under Compressive Load

J. M. McCracken¹, T. J. White^{1,2}

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Liquid crystal elastomers (LCE) are soft actuators capable of lifting loads hundreds of times their own mass.¹ Azimuthally oriented LCE, +1 disclinations, are known to buckle out of plane as they undergo order to disorder thermotropic transitions.² The conical geometries that result show significant increases in absolute load and work performed with increased element thickness.³ Recently, we described a direct stacking approach to prepare 32+ layered LCE elements that actuate at lower temperatures, carry loads greater than 1 N, and respond to transient compression by leaping out of plane.⁴ Optical profilometry in conjunction with transparent loading of conical LCE actuators enables topographic studies of these stacked elements of varying thickness. This technique has found utility in determining the exact load that induces buckling.⁵ It further allows for deeper characterization of LCE cones' response to compression. Numerous topographic trends emerge from these studies, such as wrinkling waveforms and particular modes of failure, that depend on thickness. These carry implications for mechanics and further applications for stacked LCE actuators.

References

- [1] Wermter, H. & Finkelmann, H. Liquid crystalline elastomers as artificial muscles. *e-Polymers* 1, 21974586 (2001).
- [2] Warner, M. Topographic Mechanics and Applications of Liquid Crystalline Solids. *Annu Rev Condens Matter Phys* 11, 125–145 (2020).
- [3] Guin, T. et al. Layered liquid crystal elastomer actuators. *Nat Comm.* 9, 2531 (2018).
- [4] McCracken, J. M., Hoang, J. D., Herman, J. A., Lynch, K. M. & White, T. J. Millimeter-Thick Liquid Crystalline Elastomer Actuators Prepared by-Surface-Enforced Alignment. *Adv Mater Technol* (2023) doi:10.1002/admt.202202067.
- [5] Duffy, D., McCracken, J. M., Hebner, T. S., White, T. J. & Biggins, J. S. Lifting, loading, and buckling in conical shells. (2023).

Mechanical properties of isotropic-genesis polydomain nematic elastomers

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This talk will provide an overview of recent experimental observations and theoretical studies of isotropic-genesis polydomain nematic elastomers. We begin by recalling the recent observation of an in-plane liquid-like behavior in sheets subjected to biaxial loads, and explain how this is a generalization of the well-known semi-soft elasticity [1]. We explain the mechanism behind this phenomenon through a combination of experimental observation and high fidelity micromechanical models [1,2]. We use these insights into developing a macroscale model that is appropriate for engineering scale [3]. We then use this model to study various phenomena including Hertz contact [4], torsion [5] and adhesion [6]. We conclude by discussing ongoing work on the high strain-rate behavior.

References

- [1] H. Tokumoto, H. Zhou, A. Takebe, K. Kamitani, K. Joiho, A. Takahara, K. Bhattacharya and Kenji Urayama, *Science Advances* 7, abe949 (2021)
- [2] H. Zhou and K. Bhattacharya, *Journal of the Mechanics and Physics of Solids* 153, 104470 (2021)
- [3] V. Lee, A. Wihardja and K. Bhattacharya, *Journal of the Mechanics and Physics of Solids* 179, 105369 (2023)
- [4] A. Maghsoodi, M.O. Saed, E.M. Terentjev and K. Bhattacharya, *arXiv*, 2305.00255 (2023)
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- [6] A. Maghsoodi and K. Bhattacharya, In preparation (2023)

LCE Cones: Lifting, Loading, and Buckling

D. Duffy¹, J. S. Biggins¹,

Joselle M. McCracken², Tayler S. Hebner², Timothy J. White²

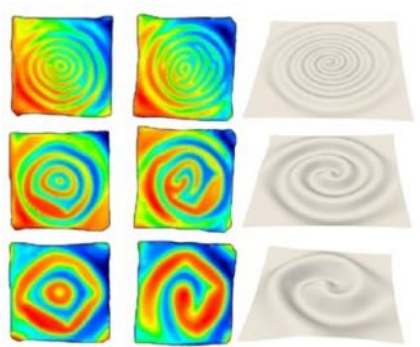
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Liquid crystal elastomer films that morph into cones are strikingly capable lifters. Thus motivated, we combine theory, numerics, and experiments to reexamine the load-bearing capacity of conical shells, and explore the lifting capacity of their deeply buckled states. Most pre-existing simulation platforms are ill-suited for such shape-morphing problems, so to fill this gap we present our open-source and freely available code MorphoShell, which is tailored specifically towards shape-morphing problems, as we demonstrate using a variety of LCE examples from recent work [1-3].

We then show [4] that a cone squashed between frictionless surfaces buckles at a smaller load, even in scaling, than the classical Seide/Koiter result. Such buckling begins in a region of greatly amplified azimuthal compression generated in an outer boundary layer with oscillatory bend. Experimentally and numerically, buckling then grows sub-critically over the full cone. We derive a new thin-limit formula for the critical load, proportional to thickness^{5/2}, and validate it numerically. We also investigate deep post-buckling, finding further instabilities producing intricate states with multiple Pogorelov-type curved ridges arranged in concentric-circles or Archimedean spirals. Finally, we investigate the forces exerted by such states, which limit the performance of conical LCE lifters, and find that a many-ridged state can lift to a quantized ladder of heights, with potential for robust soft mechanisms or soft computation.



Intricate spiral and concentric-circle patterns of curved ridges in our experiments (L, M) and simulations (R).

References

- [1] D. Duffy, J. S. Biggins, *Soft Matter* 16, (2020), 10.1039/D0SM01192D
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Liquid Crystal Elastomer – Liquid Metal Composite: Ultrafast, Untethered, and Programmable Actuation by Induction Heating

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Abstract: Liquid crystal elastomers (LCEs) are a stimuli-responsive material which has been intensively studied for applications including artificial muscles, shape morphing structures, and soft robotics, due to its capability of large, programmable, and fully reversible strains. To fully take advantage of LCEs, rapid, untethered, and programmable actuation methods are highly desirable. Here, we report a liquid crystal elastomer – liquid metal (LCE – LM) composite, which enables ultrafast actuations and high heating programmability by eddy current induction heating. The composite consists of LM sandwiched between two 3D-printed LCE layers via direct ink writing (DIW). When subject to a high-frequency alternating magnetic field, the composite can be actuated in milli-seconds. By moving the magnetic field, the eddy current can be spatially controlled for selective actuation. Additionally, sequential heating is achievable by programming the LM thickness distribution in a specimen. With these capabilities, the LCE – LM composite is further exploited for multimodal deformation of a pop-up structure, on ground omnidirectional robotic motion, in water targeted object manipulation, and crawling.

Inverse Gaussian Shape Morphs based on LCE

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Changes in curvature are used in nature from the swimming of soft-bodied organisms to the pumping of fluids by internal organs. Due to limitations in fabrication resolution and actuation mechanisms, realizing such systems synthetically remains challenging. However, achieving complex and dynamic mechanical response within an artificial single material system will vastly widen the design space. Stimuli-responsive soft actuators are highly sought-after material systems due to their potential for application across a wide range of fields such as soft pumps, biomedical robotics, and extreme-terrain exploration.

We rationally design liquid crystal (LC) systems at the molecular level to develop magnetically aligned liquid crystal elastomers (LCEs) with unusual phase behavior and corresponding macroscopic mechanical response. We report the formation of a smectic C-chevron phase upon polymerization which leads to elastomers that undergo first contraction then elongation along the axis of the director in response to monotonically increasing stimuli.

Combining the unusual molecular packing of LCEs with radial director fields patterned via magnetic alignment, we create macroscopic (cm-scale) films capable of forming dynamic Gaussian shape-morphs (e.g., flat – saddle – dome). Through small and wide-angle X-ray scattering measurements, we characterize the influence of magnetic fields on spatially patterning mesogen orientation, scalar order parameter, and phase within a large LCE film. By simply tuning magnetic field conditions during fabrication, we exert high resolution control over these parameters, and demonstrate a rich mechanical library, in which actuators of the same material are able to access a variety of unique deformations and nonmonotonically achieve multiple Gaussian curvatures in response to increasing stimuli, validated by finite element analysis. This system exhibits great potential for the creation of motile actuators capable of functional motions such as swimming, walking, and pumping.

Programming shape morphing and chirality switching by liquid crystalline elastomer microparticles

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Materials that can change shapes in a programmable fashion upon stimuli are of interests for applications such as soft robotistic, smart wearables, 3D displays, and topological materials. Among different responsive materials, liquid crystal elastomers (LCEs) with intrinsic anisotropy have been attractive. Here, we report spatially programming the out-of-plane and in-plane bending in elastomer films embedded with shape-changing LCE microparticles, leading to complex shape morphing^[1]. Further, by encoding such particles in a honeycomb lattice, their continuous shape change in response to temperature leads to lattice reconfiguration, from a right-handed chiral state to achiral one, then to a left-handed chiral state, without breaking the translational symmetry. Accordingly, the sign of rotation of the polarized light passing through the lattices changes as measured by time-domain terahertz (THz) spectroscopy. Further, we can locally alter the chirality in the adjacent domains using near-infrared (NIR) light illumination^[2].

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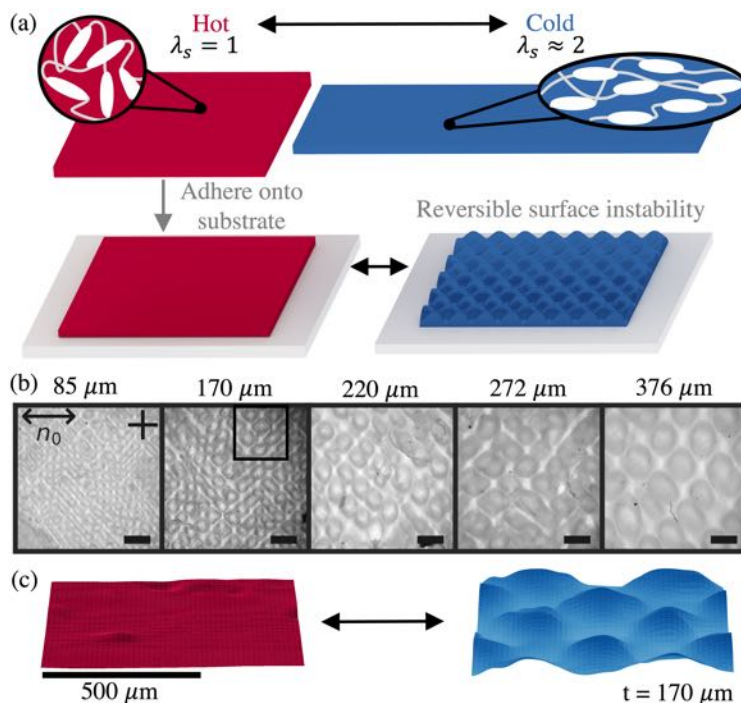
Microstructures and Surface Instabilities in Liquid Crystal Elastomers

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Liquid crystal elastomers (LCEs) are soft phase-changing solids that exhibit large reversible contractions upon heating, Goldstone-like soft modes, and resultant microstructural instabilities. We heat a planar LCE slab to isotropic, clamp the lower surface then cool back to nematic. Clamping prevents macroscopic elongation, producing compression and microstructure. We see that the free surface destabilizes, adopting topography with amplitude and wavelength similar to thickness. To understand the instability, we numerically compute the microstructural relaxation of a "non-ideal" LCE energy. Linear stability reveals a Biot-like scale-free instability, but with oblique wavevector. However, simulation and experiment show that, unlike classic elastic creasing, instability culminates in a cross-hatch without cusps or hysteresis, and is constructed entirely from low-stress soft modes. The LCE surface instability is an exemplar of how mechanical instabilities provide a useful route to complex and switchable shape changes without correspondingly complex fabrication. Finally, the switchable high amplitude topography associated with the instability suggests applications in smart surfaces. For example, a droplet sitting atop the surface instability demonstrates both a much higher contact angle and much stronger pinning than one sitting on a chemically identical smooth surface, as is characteristic of Wenzel wetting states on rough surfaces. Such hydrophobic pinning is observed on rose petals, which have a strikingly similar topography. Further smart-surface applications could include switchable aerodynamics, haptics, adhesion, and friction.



On the history of reactive mesogens

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Since their development in the 80's of last century, reactive mesogens (RM's) form a versatile class of soft materials. Advancing on the liquid crystal elastomer technology as developed by Heino Finkelmann et al., the frozen-in molecular order of the low molar mass liquid crystal monomers that is preserved during their network forming photopolymerization led to a wealth of new applications. In these early days, the focus was mainly on di-acrylate-based systems, but also di-epoxides and thiol-ene based RMs were used to form well-ordered polymer systems with nematic or smectic-type of molecular organization. Initially, at the Philips Research Laboratories, the RMs were developed for their low linear thermal expansion coefficient thus reducing thermal stresses in their use as protective coating on telecommunication fibers. But soon it was found that the polymer networks produced by the RM's demonstrated fascinating optical properties, such as high birefringence and polarization selective reflectivity, which drew the attention of developers in the then emerging flat panel display industry and their suppliers. Typical examples of new applications were polarizing beam splitters, birefringent films improving liquid crystal displays on their viewing angle, non-absorbing polarizer improving the displays on brightness and efficiency, and polarizing gratings as produced by polarization holography. But also, beyond display optics, new applications could be found, such as smectic polymer networks with nanometer pores capable to separate organic species on size and charge. And more recently, the possibility to trigger the molecular network by heat, light or electricity, the polymers change shape, surface structure or porosity leading to, among others, self-sustaining oscillators, cilia based micro-transport devices and responsive haptic surfaces.

The lecture will attempt to place the development of RMs in a historical perspective and discuss several of the early-day applications, some of them even been forgotten but still with the potential to inspire those new in the field.

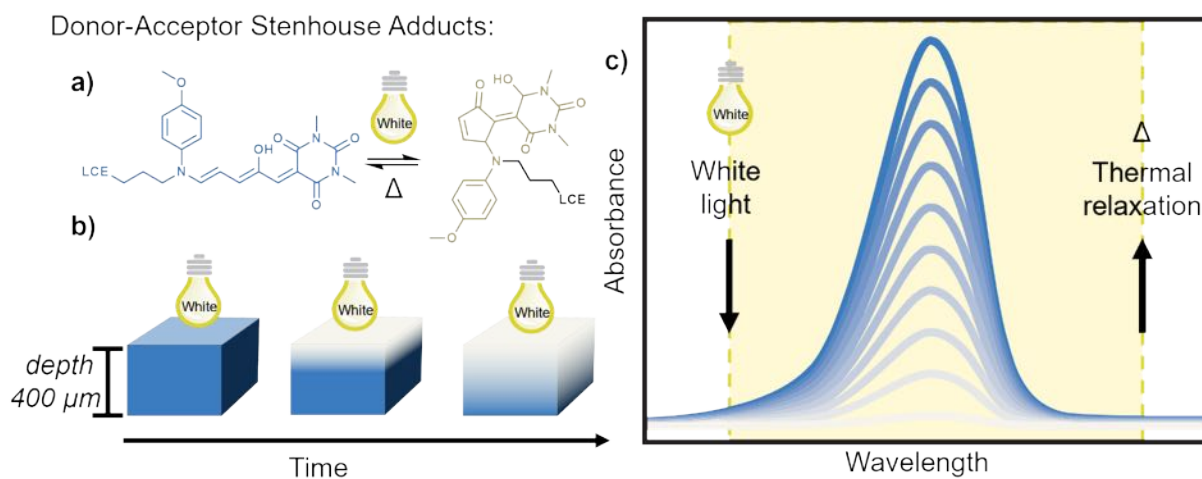
Exploration of white-light responsive liquid crystal elastomers

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Photons have multiple enabling advantages to control chemical reactions, processes and stimuli-responsive materials. In this seminar, I will discuss our groups effort to design and develop a new class of negative photochromic molecules termed donor-acceptor Stenhouse adducts (DASA), their incorporation into liquid crystal elastomers and subsequent effort to unlock their potential to create photoresponsive materials.



Controlling Phase Behavior With Precise Liquid Crystalline Oligomers

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The actuation and soft elastic behavior of liquid crystalline (LC) polymers have led to their use in applications ranging from soft robotics to elastocaloric devices when networked into elastomers. However, the behavior of a main-chain LC polymer differs markedly from that of its constituent monomer(s) and is influenced by factors including molecular weight, sequence, and molecular weight dispersity. LC oligomers and polymers of controlled length and sequence are challenging to achieve through typical step-growth LC synthetic strategies. As such, we demonstrate a LC synthetic approach based on iterative exponential growth (IEG) to prepare a series of monodisperse triazole-linked LC oligomers of precise lengths; this synthetic strategy is further amenable to the synthesis of sequence-defined LC co-oligomers and co-polymers. We synthesize several novel IEG-compatible mesogens, and explore the effects of increasing molecular weight on the thermal transitions of these discrete LC materials. Synthesized materials are molecularly characterized via nuclear magnetic resonance and gel permeation chromatography, and their properties and phase behavior examined using differential scanning calorimetry, polarized optical microscopy with in-situ heating/cooling, and X-ray scattering measurements. We find that we can tune crystalline, liquid crystalline, and metastable liquid crystalline phase transitions via oligomer length and sequence. Alternating copolymers appear to adopt columnar order not found in either homo-oligomer. These results are contrasted with the behavior of comparable species prepared via conventional step-growth polymerization. These studies highlight important factors affecting LC phase transitions, establishing design rules for tailored and precise LC behavior.

Relax, everything is under control: Leveraging modularity for precision dynamic bond placement in liquid crystal elastomers

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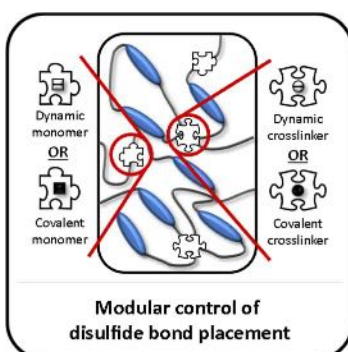
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Dynamic liquid crystal elastomers (LCEs) are a class of elastomeric networks that leverage liquid crystalline domains and dynamic covalent chemistries to produce adaptive stimuli response. Many previous works have explored the incorporation of a vast array of dynamic chemistries into LCEs to impart specific material properties depending on the nature of the dynamic bond being incorporated. However, given the variety of synthetic pathways that have been utilized to build these bonds into LCE networks, even when considering a single type of dynamic bond, it is rather difficult to directly compare these networks to one another in terms of dynamic bond placement throughout the network. To study the effects of dynamic bond topology in LCEs, a series of materials were synthesized by employing a generalizable approach that leverages tandem thiol-ene/yne chemistry where dialkene components form linear linkages while dialkyne components form crosslinks. The versatility of this approach allows dynamic bond placement to be systematically varied while holding constant other network characteristics such as crosslinking density, glass transition (T_g), and LC transition (T_{NI}). To determine how dynamic topology affects the material properties of LCEs, thermomechanical characterization was performed across a range of temperatures. At intermediate temperatures, thermomechanical properties are found to be largely similar; however, at elevated temperatures (160-180 °C) differences in relaxation activation energies begin to emerge based only on the placement of the dynamic bond within the network demonstrating that intentional dynamic bond placement can be used to tune stress relaxation times without affecting the LC character of the network.



Molecular Engineering of Mesogen Interactions in Liquid Crystalline Elastomers

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Conventional liquid crystal monomers were designed to ensure stability in optical elements subject to wide temperature swings. Comparatively, in implementations in liquid crystalline elastomers, recent literature reports detail the reduction and sharpening of stimuli-response temperature in utilizing non-traditional liquid crystallin monomers.¹ This contribution details ongoing systematic research effort to examine the structure-property relationships regarding core structure, pendant group presence, and aliphatic chain length. The effort spans monomer synthesis, oligomeric preparation, and ultimately the fabrication of LCE from distinctive starting materials.

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Actuation Functions Enabled by Crosslinker in Liquid Crystal Elastomer Structure

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In this talk, I will discuss the rational choice of crosslinker to endow liquid crystal elastomer (LCE) actuator with a specific function. A few years ago, we synthesized a main-chain LCE bearing cinnamyl units in the structure for photocrosslinking. The use of light allows for spatially selective or patterned crosslinking of monodomain film with uniaxially aligned mesogens. This function for optical inscription of actuation domains makes it easy to fabricate actuators that can undergo programmed shape changes or light-driven movements. Afterwards we carried out several follow-up studies by synthesizing various LCEs with the same basic polymer structure but different crosslinkers aiming for new functions. These include: 1) using anthracene as crosslinker, a uniformly photocrosslinked monodomain film can be photo-de-crosslinked in selected regions, which allows an actuator to be optically reconfigured to deform or move differently; 2) with dynamic covalent crosslinker (Diels-Alder reaction involving furan and bismaleimide), the alignment of mesogens can be “self-locked” by slow formation of dynamic bonds without the need for a separate step of photo- or thermal crosslinking, which imparts the actuator with the melt- or solution-reprocessability as well as bulk 3D shape morphing; and 3) through incorporation of an AIEgen in the dynamic crosslinker, it can act as a fluorescent probe to “report” the crosslinking state and thus the actuation efficiency.

Spatial Patterning of Material Properties in Liquid Crystalline Elastomers for Complex and Dynamic Actuation

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Liquid crystalline elastomers (LCEs) are functional materials capable of large, complex deformation. The deformation of LCEs is contingent on both the properties of the polymer network and the liquid crystalline phase behavior of mesogenic components. Here, we detail strategies for manipulating the polymer chemistry of LCEs to control phase behavior and introduce the opportunity for post-functionalization of the network.¹ Findings from these fundamental studies allow for precise tuning of material properties and, analogously, the responsive nature of the LCE.

Leveraging this ability to manipulate material properties, we implement new approaches to pattern spatial deformation of LCEs as illustrated in Figure 1. In one instance, through-thickness variation of actuation properties is achieved by covalently bonding multiple LCE layers, enabling snap-through deformation in response to heat.² In another approach, we achieve selective two-dimensional spatial patterning of properties throughout a single material, which gives rise to thermomechanical deformations exhibiting Gaussian curvature.³ The complexity of these tunable responses, in combination with heat transfer considerations, allows for programming of functional behavior that manifests as various forms of dynamic actuation such as leaping of LCE elements and continuously changing curvature of flat sheets.

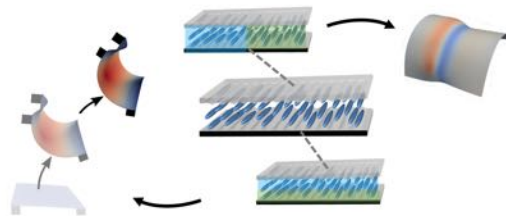


Figure 1. Complex programming of LCE actuation is enabled by spatial patterning of material properties.

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Mark Warner's Contributions to Our Field and the ILCEC

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We will take a moment to celebrate and reflect on Prof. Mark Warner's immense contributions to our field and to this conference. Mark passed away on December 24, 2021.

Auxetic liquid crystal elastomers: how they work

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Auxetic liquid crystal elastomers were first reported in 2018 [1], presenting a family of acrylate materials that become thicker, rather than thinner under strain beyond a material-dependent threshold. The auxetic response occurs at a molecular level and it was clear from the beginning that the mechanism was very different from conventional (re-entrant) auxetic materials which are cleverly engineered, porous materials in which the pore size increases on strain. We confirmed that auxetic liquid crystal elastomers (LCEs) conserve volume on strain. It was also clear from the very first report that this family of LCEs differs from other LCEs in that they deform via a so-called mechanical Freedericksz transition (MFT) rather than through the more usual semi-soft elastic response. Indeed, the auxetic response was linked to the MFT from the earliest reports [2].

Recent work has elucidated the mechanism behind the auxetic response in LCEs and has, at the same time, determined what is actually occurring in materials that undergo an MFT [3,4]. The auxetic, nematic LCEs become biaxial immediately they are strained, with the biaxiality growing as the strain increases. The uniaxial order parameter reduces, a feature long-associated with materials that undergo an MFT. At a sufficiently high strain, the optically positive biaxial system undergoes a transition to an optically negative one; when viewed in a well-aligned planar sample, this transition appears as a zero-birefringence state that bounds a sharp, 90° change in the director orientation. Thus, the MFT is not at all akin to a Freedericksz transition, but is instead a continuous evolution of biaxial order in a strained LCE – the name is rather misleading and it is perhaps time for a new one! The auxetic response is associated with the observed biaxiality; reconstituting the orientational distribution function from the order parameters [3] shows a population that is out of plane and a sufficiently large response will cause an increase in thickness beyond a specific threshold strain, as observed experimentally.

Understanding the auxetic response in LCEs allows the design rules for such materials to be asserted. This talk will describe the evidence for the auxetic response mechanism, the design rules that have been determined so far, and will consider some of these materials' potential applications.

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Pressure sensitive adhesion of nematic elastomers

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Traditionally, the practical interest in LCEs was based on their unique large-stroke actuator performance; this effect is well understood. More recently, the enhanced vibration damping of LCE has generated potential practical applications; the fundamental understanding of this effect is not yet complete, although it is certain that it is related to the internal rotational viscosity of the nematic director. The third practically relevant effect, of the enhanced reversible adhesion of LCEs, is very rapidly moving towards industrial applications; here the understanding of mechanisms leading to enhanced adhesion are understood even less, although there is an expectation that ‘adhesion is related to internal dissipation’. Here we will discuss the details of this physical effect, examine how the pressure-sensitive adhesion of LCE surface depends on the material characteristics and test conditions, reversibly diminishing in the isotropic phase, and present the ‘mesoscopic model’ of the mechanism of this effect that is based on deformation gradients and stress relaxation in the polydomain LCE structure, at the micron length scale.

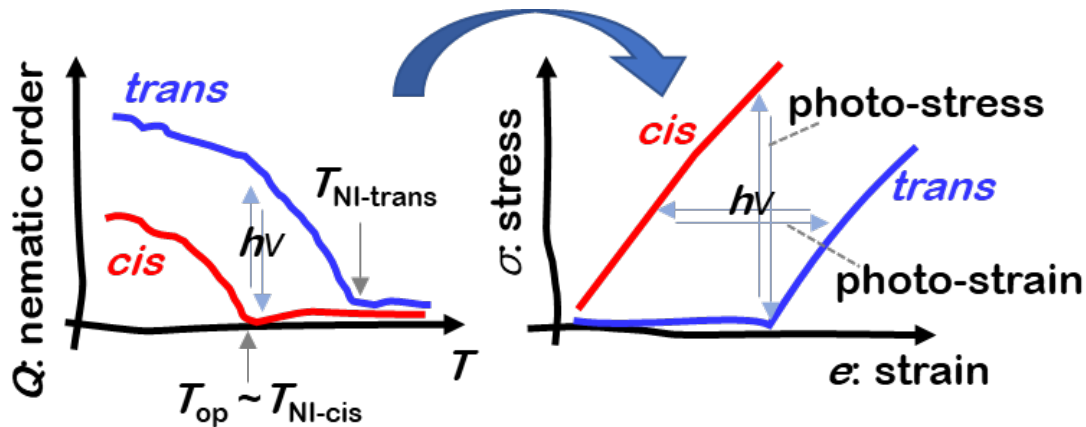
Optimal conditions for efficient photomechanical response in nematic elastomers

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Photoresponsive nematic liquid crystal elastomers (LCEs) containing azobenzene units have potential applications in light-controlled soft actuators [1]. Photo-isomerization of the trans to bent cis azobenzene form disturbs the nematic order, modulating the mechanical properties between soft and entropic elasticity [2-4]. Controlling and optimizing the modulation range are critical for the application of LCEs. In this study, by investigating a series of photo-LCEs with various phase transition temperatures, which affect the correlation between the scalar nematic local order parameter Q and the mechanical properties, we explore how the thermodynamic properties and operating conditions affect the efficiency of conversion of light energy to mechanical changes [5]. The generation of photo-stress or photo-strain under constant strain or stress, respectively, is maximized when the operating temperature (T_{op}) is close to the nematic-isotropic transition temperature of the cis-rich state (T_{NI-cis}) (Figure). Under these conditions, the light-induced change in Q is also maximized, which is further supported by a simple model that considers the light-induced shift of the nonlinear curve of the temperature-dependent Q along the temperature axis. The present results provide useful guidelines for optimizing the materials and operating conditions for the efficient conversion of light energy into mechanical work in photo-LCEs.



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Modeling the Viscoelastic Behavior of Liquid Crystal Elastomers

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Liquid crystal elastomers (LCEs) exhibit complex behavior, including semi-soft stress response, rate-dependency, anisotropy, and large deformation induced by the isotropic temperature transition. These unique features render LCEs highly promising for various applications, such as programmable soft robotic actuators, vibrational dampers, and impact protection applications. In order to fully leverage the potential of LCEs, predictive modeling is needed to efficiently design and optimize LCE structures to achieve the desired performance. This presentation will describe our efforts to develop a generalized viscoelasticity model for monodomain nematic liquid crystal elastomers (LCEs). The model attributes the rate-dependent behavior of LCEs to the viscous rotation of the director and the viscoelastic deformation of the network chains. Furthermore, we developed a robust implementation of the model for finite element analysis. By employing our model, we have investigated how the interplay between viscous director rotation and viscoelastic network deformation contributes to the observed enhanced dissipation behavior in experimental settings. Additionally, we have examined the influence of realistic boundary conditions, the heterogeneous distribution of director orientation, and other structural features of the director on the rate-dependent stress response and hysteresis.

Exploring nonlinear elasticity in nematic elastomers: beyond simple stretching?

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The nonlinear elasticity of nematic elastomers is distinguished by its soft elasticity, arising from the correlation between deformation and director realignment.[1] Soft elasticity effect manifests as a pronouncedly low stress plateau over a finite strain range in stress-strain relations under simple (uniaxial) stretching, a process that facilitates director rotation in the stretching axis.

Although simple stretching is a particular mode among all physically feasible deformations, experimental studies exploring other deformation types have been relatively scarce. Understanding the nonlinear elasticity under varying deformation modes is crucial for not only a comprehensive grasp of liquid crystal elastomer mechanics, but also considering their diverse deformation patterns during practical use.

This presentation introduces our recent experimental investigations on biaxial stretching, torsion and crack properties of polydomain or monodomain nematic elastomers. In biaxial stretching, we found that the true stresses in both directions remain identical during polydomain-to-monodomain transition regardless of biaxial strain ratio, and they solely depend on the loading plane area.[2] In terms of torsion, we focus on the correlation between shear and normal stresses, and the associated buckling phenomena. Furthermore, we have investigated the unique strain-field near the crack-tip in nematic elastomer sheets.

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Towards Hopfionic Haptics

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Topological order and phases represent an exciting research frontier that emerged recently [1]. In soft condensed matter systems, hopfions [2], heliknotons [3] and a variety of other topological solitons have been recently demonstrated and attract a great deal of interest [1]. This lecture will discuss how such 3D topological particle-like can be realized in polymer responsive networks, as well as how the topology of the solitonic director field configurations can be used in enabling surface topography actuation, with potential haptics applications.

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Thermoplastic Liquid Crystal Elastomers

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A new class of thermoplastic liquid crystalline elastomers is presented as “smart” materials with stimuli-responsive, self-healing, and recyclable properties [1]. The smart materials are based on segmented liquid crystalline polythiourethanes (Figure 1). The dynamic structural character of the hydrogen bond crosslinks can be used to construct programmable, self-healing and recyclable soft actuators.

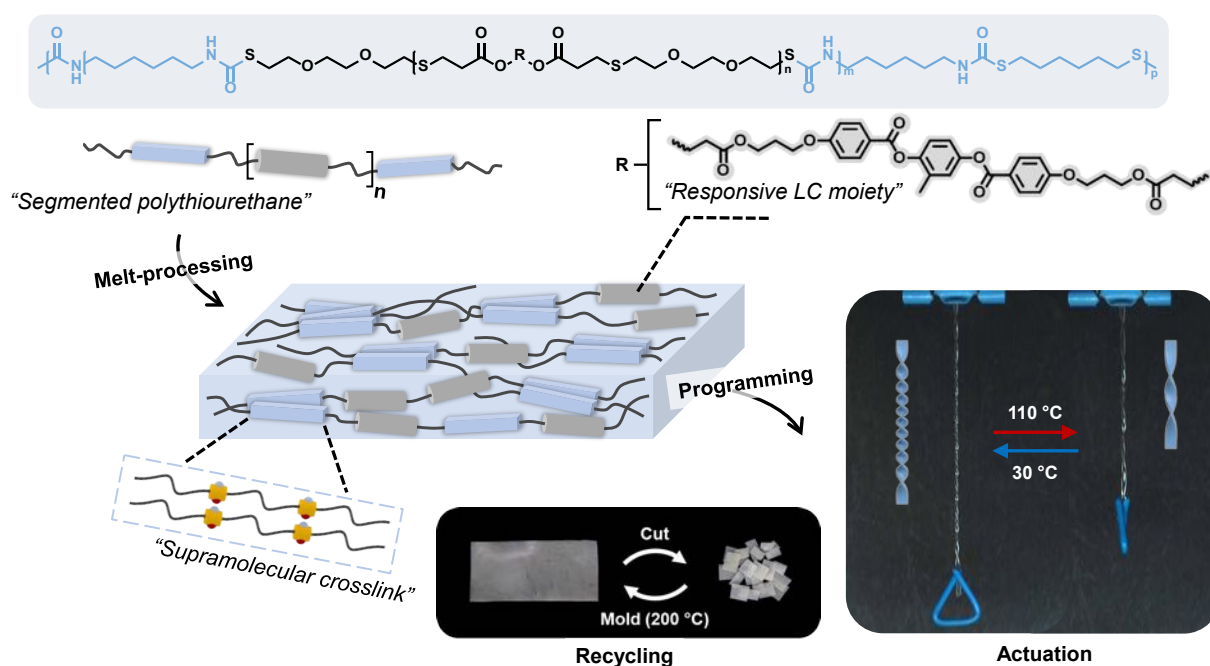


Figure 1. Responsive, recyclable thermoplastic liquid crystal elastomers [2].

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High-performance photomechanical materials via polymer-templated growth of aligned microcrystal arrays

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Materials capable of directly converting photon energy into mechanical deformation offer promise in a wide variety of contexts including adaptive optics, remotely operated swimmers, and actuators controlled via lightweight optical cables that resist corrosion and electromagnetic interference. Organic photoswitches offer significant potential, thanks to their ability to undergo large changes in molecular geometry following photochemical reactions, and their highly tailorable absorption spectra. Pioneering work using liquid crystal elastomers and networks as hosts for photochromic molecules have been of great interest in this context thanks to their excellent processability and mechanical properties, coupled with reasonable work densities. Single crystals of small photomechanical molecules offer the potential for substantially greater work output due to the high density and ordering of photochromes that they present, but have been hampered by poor mechanical stability. We have recently focused on developing hybrid materials that combine many of the advantages of these disparate approaches. In particular, we have studied the growth of micrometer-scale photomechanical diarylethene crystals within the pores of polymer membranes [1]. By nearly matching the mechanical stiffness of the crystals, and biaxially controlling their orientation in a desirable way, such materials exhibit photomechanical work outputs surpassing the best previously reported systems, while enabling facile processing into mechanically robust samples with cm-scale lateral dimensions.

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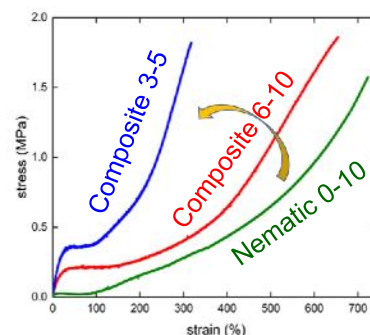
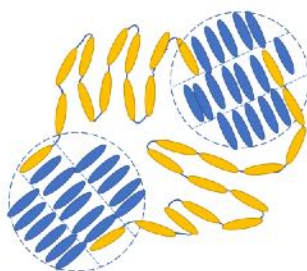
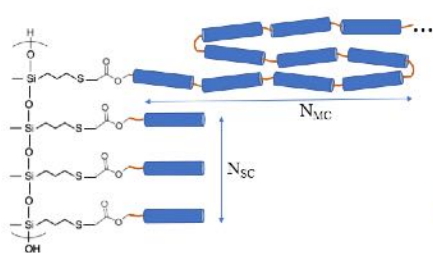
Main-chain nematic side-chain smectic composite liquid crystalline elastomers

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A composite liquid crystalline elastomer is designed, combining main-chain and side-chain mesogenic polymers in the network, and resulting in micro-phase separated regions of nematic and smectic ordering in the macroscopically homogeneous elastomer. A range of different fractions of the components is explored, from fully nematic main-chain system, across to fully smectic side-chain elastomer. Thermal phase transitions of both phases coexisting in the material can be detected by calorimetry, and the nematic/smectic structure is investigated by X-ray scattering. The tensile stress-strain data reveals the key effect of such a multi-phase composite, where the nematic fraction adds ductility while the smectic fraction increases the modulus and mechanical stiffness. Varying the composition, we were able to control a broad range of mechanical properties of this material type, and achieve temporary programming of the director orientation by 'locking' the deformed state in the smectic domains.



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Programming Molecular Order of a Liquid Crystal Elastomer with Magnetic-Field-Assisted DLP Printing

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Shape memory polymers (SMPs) and hydrogels are two major materials classes of stimuli-responsive materials that have been widely used in 4D printing. SMP can memorize its original printed shape and restore it when needed, but its stimuli-responsive deformation is only one-way because mechanical processing is always required for shape programming. Hydrogels can deform reversibly depending on various environmental changes, but they suffer from extremely slow time-scale for the shape change due to a quadratic scaling law for the diffusive water molecule migration process and are not suitable for applications where surrounding water is not available. Recently, liquid crystal elastomers (LCEs), combining polymeric elasticity with liquid crystalline anisotropy, have received growing attention as a new class of a 4D printing material that can overcome the limitations of SMPs and hydrogels.

LCEs can show reversible stimuli-responsive deformation driven by rearrangement of molecular orientation of liquid crystal (LC) molecules. However, alignment of LC molecules has been achieved primarily using mechanical extension or viscous shear, which significantly limit its applicability in additive manufacturing. Here, we present a DLP system capable of printing an LCE structure while selectively programming molecular orientation of LC molecules using a magnetic field. We report a LCE precursor solution that maintains a nematic phase at room temperature. Using a custom-built DLP printing system with an integrated magnetic field generator, LC molecules are aligned in the desired orientation and selectively cured by digital light projection. Therefore, LC orientations can be freely encoded in a printed structure. This allows for spatial patterning of LC orientations independent of the geometry of the printed structure. Considering the advantages of DLP printing over other additive manufacturing methods, this approach can offer unmatched opportunities for programming various modes of programmable shape deformation of responsive architected materials.

Multi-Planar 4D Printing and Deformation Control **Strategies for Liquid Crystal Elastomers**

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Fabrication pathways for liquid crystal elastomer (LCE) have remained a topic of intensive research interest in recent years. Photopatterning and two-stage reaction techniques were pioneering methods which enabled molecular patterning and fabrication of LCE networks in few, relatively simple steps. Recently, additive manufacturing (AM), or 3D printing, has become another promising approach for facile production of LCE networks. The combination of 3D printing with responsive materials, such as LCE, has led to a new generation of smart structures that not only possess a static shape but also can change their shape over time. This process is termed 4D printing, with the fourth dimension being time. The focus of this talk will be on the development of new 4D printing techniques to generate a new class of complex, multi-planar LCE geometries. One of the most notable 4D printing methods, direct ink write (DIW), enables simple fabrication of complex LCE structures by coupling the printing process with the LCE alignment step. This occurs due to the shear forces generated during extrusion through the DIW nozzle. By combining this approach with multiple materials, novel shape transformations can be achieved. For example, by combining LCE with the unique properties of another smart material called shape memory polymers (SMP), the shape transformation can be locked in place. Furthermore, LCE/SMP composites offer superior mechanical properties in both the deformed and undeformed state for functional engineering applications. The structures produced using this method, however, can only perform 2D to 3D shape transformations. Many smart structures applications require more complex 3D shapes to be fabricated. For this reason, we will discuss a recently developed printing method called embedded 4D printing. Here, LCE is extruded into a gel matrix to create complex 3D architectures which can generate unique 3D to 3D shape transformations. This approach enables LCE molecular programming in any 3D cardinal direction. The ability to 4D print complex 3D LCE structures without the need for supports opens new avenues for the design and development of functional and responsive systems such as soft robotics, biomedical devices, and advanced materials engineering.

Dual Morphing of Direct-Ink-Written Liquid Crystal Elastomers Under Heat and Moisture

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Multiresponsive soft actuators that respond to (combinations of) different environmental stimuli, can offer a wide range of complex motions and adaptable functions similar to living systems.^[1,2] Herein, we present direct-ink-written (DIW) liquid crystal elastomers (LCEs) that can undergo two distinct shape-morphing when exposed to heat and moisture. The temperature response is due to the intrinsic phase transition of LC, while the moisture response is imparted by ionizable tertiary amine moieties in the LCE as explained in our previous work.^[3] Various LCE structures are printed by the DIW method, with demonstrating reversible shape morphing in orthogonal or synergistic manner in response to heat and moisture. The ability to achieve dual and complex shape morphing can broaden the potential applications of LCEs in soft robotics, sensors, responsive architectures, and textiles.



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3D-printed flat sheets of LCE morphing into intrinsically curved folds

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Previously, there has been much work on designing and fabricating LCE sheets with planar director profiles that morph into curved surfaces on actuation. Importantly, such sheets can morph into intrinsically (Gauss) curved surfaces [1], lending their actuation great strength, as exemplified by the lifting power of the LCE cones that form actuation of +1 defect patterns. Looking beyond LCEs, intrinsically curved folds are used to lend form, strength and function to a host of natural and engineered surfaces, including the lily-pad in Fig. 1 a, and also ubiquitous deployable structures such as tents and umbrellas. Here, I will present the basic kinematic rules for describing and classifying ICFs, which provide insight into when they are and are not rigid, and also into their utility as mechanisms and deployable structures [2]. I will also demonstrate, theoretically and experimentally, how to create a patterned LCE sheet that will morph into a surface containing an ICF on heating (Fig. 1 b). Such actuation is superficially like a bending bi-layer, but endowed with the mechanical strength of intrinsic curvature, and offers great potential for biomimetic engineering applications. Our fabrication is via 3D extrusion printing, which is enabled by the development of a new “slicer” algorithm for printing arbitrary complex director patterns. Finally I will highlight how several LCE ICFs can be combined to create a strong Gaussian gripper (Fig. 1 c – e), capable of lifting many times its own weight.

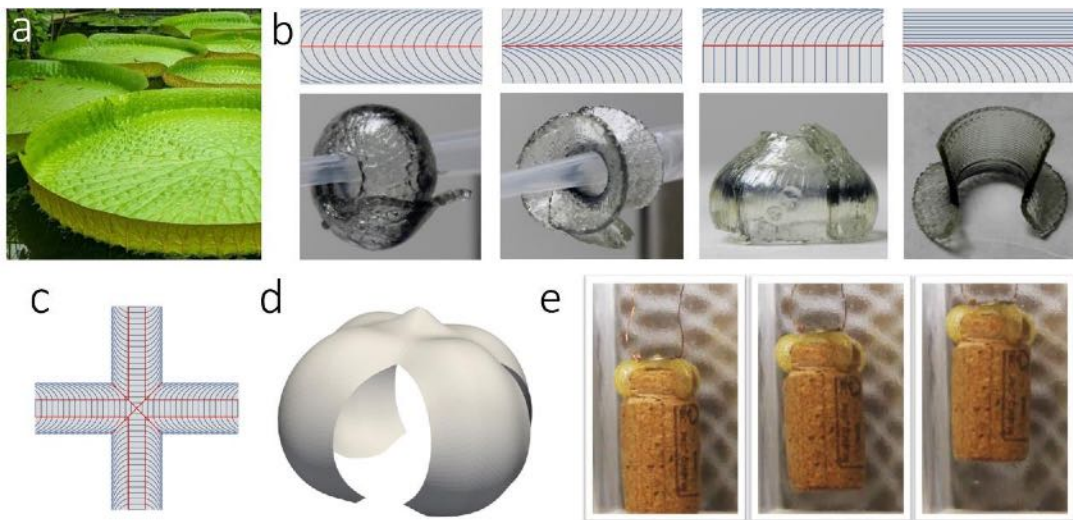


Figure 1 a – water lily-pad; b – director patterns and actuated shaped of ICF: symmetric positive, symmetric negative, asymmetric positive and asymmetric negative, respectively; c – pattern of the gripper combining positive asymmetric folds; d - simulated shape of the gripper; e - gripper's performance during experiments.

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The fabrication of light crystal network structure using two-photon technology

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Liquid Crystal Networks (LCNs) can undergo macroscopic deformations, such as thermal actuation and anisotropic expansion, due to different molecular arrangements under external stimuli[1-2]. Traditional LCN elastomer processes mainly involve aligning LCN using two substrates (top and bottom) and curing them into thin films using a single light beam, making it challenging to fabricate LCN microstructures of arbitrary complex shapes[3]. In this study, we employed dual-photon technology to manufacture elliptical microstructures. By coating polyimide (PI) on the glass surface, we controlled the alignment of liquid crystals in a single direction, thereby constructing vertically aligned LCN elliptical microstructures. We used polarized optical microscopy to observe the correct arrangement of the liquid crystal network structure. Finally, we subjected the elliptical structures to repeated irradiation with green laser light (532 mW) to measure their expansion/contraction and verify if the alignment matches the expected results.

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Light driven morphing of printed liquid crystal elastomers

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Light as a stimulus for shape change is useful for untethered localized actuation and subsequent locomotion. Applying this phenomenon to architected three-dimensional structures enables actuation modes not previously achieved¹. Thus, the development of liquid crystal elastomer (LCE) direct ink write (DIW) feedstocks tailored not only for light responsivity, but also for printing of architectures with overhangs or tall features opens the door for applications such as search-and-rescue or medical robots/devices. Here, we report our approach to tailoring LCE gold nanorod (AuNR) composite DIW feedstocks to maintain photoresponsivity while increasing the printability towards complex architectures. We evaluated the advantages and disadvantages of thiol vs acrylate based LCE networks in curing kinetics, molecular weight, chain flexibility, and AuNR dispersibility with respect to printability and photothermal actuation. The resulting ink could be programmed during DIW printing and locally reprogrammed with near IR light post printing to induce novel behaviors like oscillation, crawling, rolling, and transportation of mass. Pairing light stimuli with computer vision results in untethered training and direction of the printed LCE structure's movement, increasing functionality and applicability towards being used as soft machines.

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Cholesteric Liquid Crystal Elastomer Sheets and Fibers for Mechanochromic Strain Sensing

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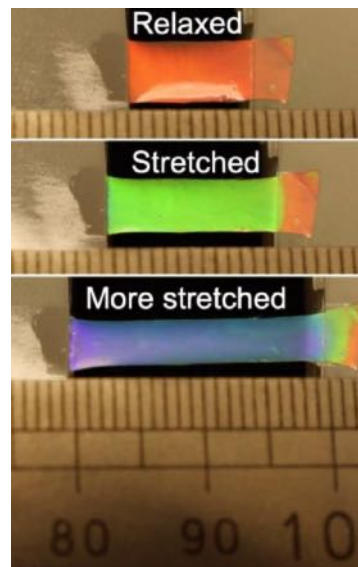
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I will report on our recent and on-going efforts to make cholesteric liquid crystal elastomer (CLCE) sheets [1] and fibers [2,3] in scalable procedures, with good control and wide tunability of the relaxed state helix pitch and good visibility of the color regardless of background. In case of sheets we are also developing a process suitable for laymen, requiring no special equipment or expert knowledge. I will also describe our efforts to bring these materials to the market, for applications in structural health monitoring [4] (crack detection and monitoring, detection of residual strains after catastrophic events like earthquakes, ...) and in textiles (from medical treatment to fashion).

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Cellulose-based anisotropic networks for moisture and temperature time sensors

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Cellulose ribbons, if produced from liquid crystalline solutions, can be responsive to moisture [1]. Also, plants can program cellulose-based dead tissues to respond to external stimuli, usually water [2]. One of these examples is the awns of the *Erodium* fruit, which present fantastic coil and uncoil motions in response to humidity. Due to its intrinsic curvature, the straight awns existing in the *Erodium* plants acquire the shape of a right-handed helix after leaving the fruit with the seed. When in contact with water, the awn unwinds, allowing the seed to bury in the soil. This mechanism was recently applied to spread seeds from other kinds of plants [3]. In this work, we found structures resembling the characteristics of liquid crystalline elastomers at the genesis of these movements. While swollen in water, ribbons isolated from *Erodium* awns are transparent, presenting birefringence between cross polars. Depending on the moisture quantity, these ribbons change reversible shape from left to right-handed helices. These movements result from anisotropic cellulose-based materials organized in layers that contract differently upon the presence of humidity.

Nature, cellulose-based inspired networks will be described, being some examples and mechanisms discussed.

Acknowledgments

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Self-Assembled Microactuators Using Chirality of Liquid Crystal Elastomers

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Developing actuators that undergo reversible, programmed shape changes at microscale presents a complex challenge. Although machines with sensors, processors, and motors can be designed,^[1] the system has limitations in miniaturization due to the functional components. To achieve such responsive, programmed shape change at the microscale, stimuli-responsive polymers offer advantages. As stimuli-responsive polymers function as both sensors and actuators,^[2,3] they offer advantages in developing microactuators. However, developing processing strategy to effectively control the shape changes of microactuators still remains challenging.

Liquid crystal elastomers (LCEs) are promising stimuli-responsive polymers for achieving microactuators with programmed shape change. LCEs that undergo simple expansion or contraction have been developed by utilizing uniaxial compression forces. More complex shape changes have been achieved by applying magnetic fields across LCEs. In this work, we introduce new tools to develop LCE microactuators using the spontaneous self-assembly of chiral nematic monomers within molds. Chiral nematic monomers self-assemble asymmetrically across the thickness of microactuator. Asymmetric molecular orientation within the structure results in bending actuation in response to temperature. LCE microactuators with 0.05 wt% chiral dopant exhibited bending from flat to an angle of $27.2 \pm 11.3^\circ$ at 180 °C (Figure 1). In addition to chirality, protrusions were introduced onto LCEs to break the symmetry of the structure. Breaking the symmetry influences molecular orientation and determines the bending direction of actuators. The presented fabrication strategy controls the bending of microactuators in both magnitude and direction without complex setups and allows for easy scaling up to arrays of actuators.

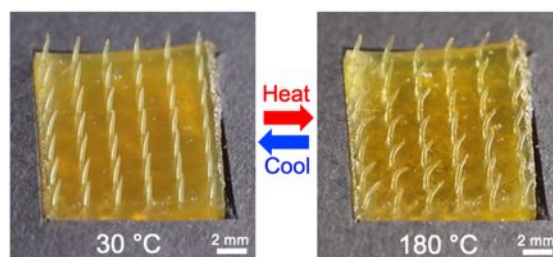


Figure 1. Reversible, controlled bending of LCE microactuators in response to temperature.

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Color change in cholesteric liquid crystalline elastomers

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Soft optical materials are being explored in the automotive, aerospace, and consumer markets. Cholesteric liquid crystalline elastomers (CLCEs) are a unique class of soft optical materials that self-organize into a helicoidal structure. The periodicity of the refractive index across the thickness of the material forms a 1-D photonic bandgap resulting in selective reflection.

This presentation will detail the use of surface alignment to make CLCEs with optical quality necessary for use in ophthalmic devices or the built environment. Specifically, we will report our recent focus on adjusting the modulus to control the tunability of the reflection and how the optical properties can be switched in response to mechanical and electrical stimuli.

Motility from Liquid Crystallinity

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Patterned disruption of orientational order in liquid crystalline elastomers (LCE) is a versatile platform for eliciting dramatic shape transformations in response to stimuli. Nascent to this actuation is also the ability to generate work-densities that compare favorably against natural muscles [1]. Accessing the response of LCEs to power motility in robotic applications requires a) sustaining continuous actuation cycles and b) accelerating the actuation rates to generate relevant dynamic responses.

Photomechanically active LCE that transduce light into mechanical work can continuously sustain actuation-relaxation cycles when the material repetitively couples-decouples from the optical stimulus during its photoactuation. This can occur either through periodic self-shadowing [2] or through modulation of the optical stimulus [3]. In judiciously designed chiral configurations, actuation cycles can be harnessed for motility in mg-scale microrobots. Complementing this, thermally driven actuation in (supercoiled) chiral geometries sustains motility using actuation-relaxation cycles that are generated from thermal gradients [4]. Thermal responsiveness of LCE also lends itself to electrically powered actuation using Joule heating from innervated electrodes. Utilizing them in shell-like geometries is key to exploiting mechanical non-linearities. High-speed actuation in LCE shells that undergo snap-through present microrobots with motility powered with frugal electrical sources [5].

These examples also highlight the need to harness the responsiveness of LCE in freeform geometries. The emergence of additive manufacturing of LCE that encode molecular order in 3-dimensional freeforms is a key enabler [6]. Biomimetic microrobots and manipulators become feasible for operation in domains ranging from the human body, engineered spaces, and natural environments.

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Morphing and motility of LCE films

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Liquid Crystal Elastomers (LCEs) are polymeric materials exhibiting large spontaneous deformations in response to a wide range of stimuli. These include temperature changes, light, electric and magnetic fields [1]. At least in the case of nematic elastomers, the basic mechanism underlying their responsiveness is the uniaxial distortions induced by the isotropic to nematic phase transition. These can be modeled in the framework of nonlinear elasticity, thanks to the multiplicative decomposition of the deformation gradient into the product of an elastic and an inelastic part [2,3].

The flexibility in the possible actuation strategies makes LCEs interesting candidate materials for several applications. At the same time, the variety of physical mechanisms relevant for their response makes LCEs a very interesting system for multi-physics modeling. Morphing and motility, in particular, require the solution of complex nonlinear systems resolving the interaction of a structure exhibiting large deformations with the surrounding fluid (fluid-structure interaction problems), thermoelastic and photoelastic coupling, electro-magneto-elastic coupling, etc. In this talk, we will review some of our recent efforts towards an integrated framework for the computational modeling of the multiphysics response of thin films of LCEs, by discussing the solution of specific problems arising in applications.

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Self-Oscillating Materials: Enabling Self-Regulated Robotic Functions

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Self-oscillation is a phenomenon in which a mechanical structure sustains periodic motion despite the absence of a periodic stimulus. Over the past decade, this field has experienced rapid growth as researchers investigate various responsive systems with different geometries and self-oscillating modes. The potential applications encompass the creation of autonomous micro-robots capable of walking, swimming, and the ultimate dream of flying. During this presentation, I will share our insights into the mechanism of light-induced self-oscillation. Additionally, I will showcase our recent achievements in the laboratory, including a liquid crystal elastomer capable of non-reciprocal self-oscillation underwater [1], an ultra-sensitive actuator that oscillates in response to omnidirectional sunlight [2], and a dandelion structure that self-oscillates within a flowing wind [3].

Furthermore, my talk will emphasize the realization of flying robots. I will present examples of passive fliers, such as dandelion-like and spiral seed inspired gliders. To conclude, I will outline a set of crucial points necessary to achieve the ultimate goal of smart material-based hovering robots – the active mode of flight (though not implemented yet).

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Encoding life-like multimodal locomotion in photo-responsive microstructures

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Living systems express propulsion at the microscale mostly due to non-reciprocal motions. At Low Reynolds number (drag-dominated regime), motility is achieved only when displaying non-reciprocal shape transformations (scallop theorem).¹ Likewise, terrestrial propulsion escape friction forces by exploiting non-reciprocal movements, resulting in directional motility (*e.g.*, metachronal wave).² While non-reciprocal motions are well established in microorganisms (*e.g.*, cilia/flagella in bacteria, legs in millipedes), artificial micro walkers or swimmers are currently facing multiple challenges to achieve such deformations and, therefore, motility. Liquid crystal elastomers (LCEs), not only present the structural integrity but also can be polymerized with well-defined molecular alignments to encode complex, macroscopic motions in a non-reciprocal fashion.³ The latter is a crucial feature as it also allows for inter-media locomotion at the microscale in synthetic matter, encompassing both aquatic and terrestrial environments. By using photo-responsive mesogens in pillar-based single-material LCE microstructures, we investigate the emergence of multimodal locomotion in different media. The control of speed and directionality will be evaluated as a function of the angle of irradiation, intensity and type of light. We believe that the combination of non-reciprocal behavior and external control will help advance the creation of soft robotics and intelligent materials that perform controlled motion under dissipative conditions.

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Wireless and Thermoelectric Actuation Methods for Liquid Crystal Elastomer Based Soft Robots

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Liquid crystal elastomer (LCE) is a class of loosely crosslinked shape memory polymers that exhibit reversible shape change during transitions between liquid and crystalline states. These transitions, referred to as the nematic to isotropic phase transition, are often triggered by thermal stimuli. Recently, LCE has been explored for soft robotic applications to replace more traditional actuation methods. These soft robotic applications have commonly used approaches such as joule heating and near infrared (NIR) light as actuation methods. Currently these approaches have two main drawbacks: inefficient energy conversion and a lack of closed loop control. To address these challenges, new actuation approaches are needed for LCEs. In this talk I focus on our recent works that have addressed both challenges. First, soft and stretchable thermoelectric devices (TED), which convert current across oppositely doped semiconductors into temperature differentials and vice-a-versa, are integrated with LCE to act as Peltier heaters and coolers for LCE actuation. These LCE-TED transducers enable closed loop control, which is lacking in many LCE soft robotic approaches [1]. LCE-TEDs double as energy harvesters through the Seebeck effect to recover energy in between actuation cycles and when in contact with environmental heat sources. Separately, we introduce WASER (wireless actuation for soft electronics-free robots) as an approach for more efficient and non-line of sight soft robotic actuation. WASER enables 2.4GHz beamforming through an antenna array for localized heating. LCE is coated with liquid metal-based patch antennas for wireless heating and actuation, establishing a fully wireless non-line of sight soft robot [2][3].

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Soft liquid crystal elastomer with integrated stretchable Joule heater

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Robots play important roles in modern society from enabling rapid mass production in manufacturing to precise surgical maneuver. The field of robotics has seen remarkable advancements in last century and continues to challenge the limits of what could be possible. One such new frontier is the idea of bio-inspired “soft robots” whose constituent components possess mechanical properties on the order of those found in living things. While conventional robots are unable to adapt well to changes in their environments due to the limited number modality of actuation afforded by their rigid bodies, soft robots incorporate compliant actuators to achieve more degrees of freedom. The thermotropic liquid crystalline elastomer (LCE) has garnered a lot of attention as a potential actuating material to build such soft robots due to its in-plane actuation, two-way shape memory, appreciable power density, and high actuation efficiency. In this work we utilize a facile fabrication technique to leverage the utility of dynamic chemistries and intrinsically stretchable Joule heaters in the production of soft robots. We incorporate a post-synthetically processed dynamic disulfide LCE and separately fabricated stretchable joule heaters together using silane surface chemistry via CVD. The method is a versatile way to adhere different popular soft electronic substrates, such as PDMS, to the dynamic disulfide LCE. In addition, the robust adhesion of two functional layers allows for combination before or after the strain-alignment of dynamic disulfide LCE, which leads to multimodal actuation depending on the device structure.

Photoalignment patterning of liquid crystals by scanning wave photopolymerization

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Hierarchical control of two-dimensional (2D) molecular alignment patterns over large areas is essential for designing high-functional organic materials and devices. However, even by the most powerful current methods, dye molecules that discolor and destabilize the materials need to be doped in, complicating the process. We present a dye-free alignment patterning technique, based on a scanning wave photopolymerization (SWaP) concept, that achieves a spatial light-triggered mass flow to direct molecular order using scanning light to propagate the wavefront. This enables one to generate macroscopic, arbitrary 2D alignment patterns in a wide variety of optically transparent polymer films from various polymerizable mesogens with sufficiently high birefringence (>0.1) merely by single-step photopolymerization, without alignment layers or polarized light sources (Figure 1). A set of 150,000 arrays of a radial alignment pattern with a size of $27.4 \mu\text{m} \times 27.4 \mu\text{m}$ were successfully inscribed by SWaP. This dye-free inscription of microscopic, complex alignment patterns over large areas provides a new pathway for designing higher-performance photonic and robotic devices.

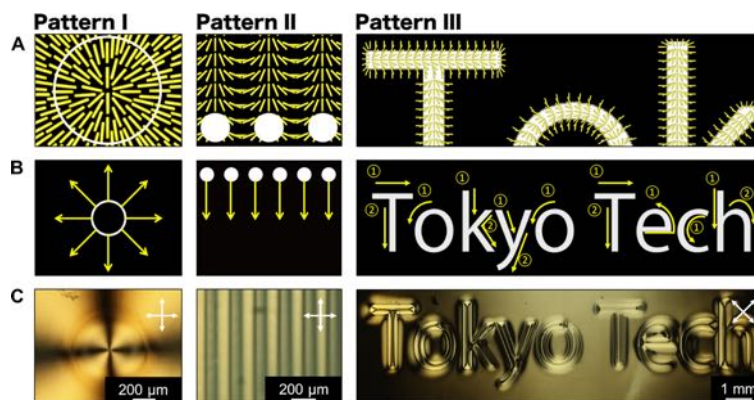


Figure 1. Arbitrary alignment patterns developed by spatiotemporal scanning of light [2].

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Liquid Crystalline Elastomers as Photoswitchable Adhesives

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Abstract

The incorporation of light-responsive components into liquid crystalline elastomers (LCEs) has enabled spatiotemporal control over dramatic changes in physical properties, including volumetric changes for actuation and (meso)phase changes for adhesives. To the latter point, photoswitchable adhesives have been recognized for their potential to enable patterned dry transfer of materials from their growth substrate to a functional surface with little-to-no waste generation or contamination. For example, this would prove transformative with the transfer of 2D semiconductors to silicon for next generation electronic devices. However, the development of effective photoswitchable adhesives for this purpose remains in its infancy and thus necessitates a systematic examination of LCEs to identify key structure-reactivity relationships. This presentation will focus on our efforts to create photoresponsive acrylate-based LCEs and characterize mesophase changes that occur upon exposure to different wavelengths of light at variable temperatures. Additionally, the use of polarized optical microscopy to study the effect of azobenzene-containing resin formulation on room temperature photoswitchability to this end will be described. Finally, the presentation will be concluded with discussing the effects of light-induced mesophase changes on adhesive properties along with a future outlook.

Liquid Crystal Elastomer Nanocomposites as a Platform for Optically-Addressable Materials

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Liquid crystal elastomers (LCEs) are an attractive platform for dynamic shape morphing due to the large directional strains produced along the director upon the loss of mesogenic order. While robust mathematical frameworks allow for *a priori* patterning of a given director field to achieve a desired target shape upon deployment, this strategy cannot be generalized to materials systems where high-resolution surface alignment is impractical or impossible. Alternatively, a more general strategy for prescribed shape morphing is to program the magnitude of local strain via localized deformation in a sheet of constant director orientation. In this work, we demonstrate a method to pattern local photothermal heating – and thus strain – in thin, planar nematic LCE sheets via spatial patterning of gold nanoparticles (AuNP). Using a combination of theory, simulation, and experiment, we show that this method can be used to generate discontinuous and continuous strain profiles that drive out-of-plane buckling into specific shapes. Additionally, we explore the utility of this system for triggering buckling instabilities for photoinduced wrinkling, snapping, and motion. Finally, this patterning methodology is extended to non-canonical geometries including cylindrical fibers and exploited to control photoactuation in 3D space.

3D director alignment of liquid crystalline structures with magnetic fields

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The director is key to liquid crystalline systems and their behavior, such as their optical and mechanical response. Several techniques have consequently been developed to align liquid crystalline mesogens. Surface alignment techniques are well established in creating 2D director profiles. Electric and magnetic field alignment techniques offer, in principle, the ability to tune the director profile in three dimensions [1]. Similarly, magnetic fields have been used to pattern director profiles, voxel by voxel [2]. Common to most studies that use magnetic field alignment of the director is that relatively strong magnetic fields, e.g. 0.3 T, are required and that the alignment is generally slow and often requires high temperatures.

In this work, we have successfully achieved control over the director in the desired 3D orientation at room temperature with weaker magnetic fields. This will make it easier to pattern the complex director profile in liquid crystalline structures and to thereby achieve unique mechanical properties. Additionally, we describe an optical characterization technique that can be used to determine the alignment of the director in 3D. It permits director changes to be observed in real time. The important parameters, such as field strength, surface anchoring effects, coatings and temperature are explored to determine the key drivers that facilitate the alignment of the director by magnetic fields in our LC system. The role of the viscosity, magnetic anisotropy and elastic energy are discussed. The lower magnetic fields in our system will facilitate applications and further studies.

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Liquid crystal network actuators that “adapt” (?), “self-regulate” (?) and “learn” (?)

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Biological systems, viewed within the materials science perspective, are excessively complex. They are adaptive, multifunctional and -responsive, dissipative, self-regulating, and capable of evolving and learning from their past experiences. Hence, biological systems have provided a great source of inspiration for scientists aiming to design synthetic functional materials that in some sense can be deemed “intelligent“. Liquid crystal network (LCN) and hydrogel -based soft actuators provide a rich platform for simplistically mimicking some of the properties of natural systems, as demonstrated – focusing the attention to our own work only – by examples displaying “life-like” features resembling autonomous action,[1] adaptation,[2] homeostasis,[3] and learning.[4] Yet again, all these systems fall way short on their natural counterparts in terms of complexity, capability to respond to environmental cues, and to evolve based on past experiences. Having this in mind, is the terminology used to describe their behavior justified or helpful? The aim of this talk is to reflect on this question, using our own work on light-responsive LCNs as an example.

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Limit Cycle LC Elastomer Actuation for Temperature Regulation and Programmed Thermal Transport

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Regulating thermal transport is important in a diverse range of situations. In living systems, endothermic species such as birds and mammals are able to maintain their bodies at metabolically favorable temperatures by actively generating and dissipating heat. In engineered systems, controlling thermal transport is critically important to optimize efficiency and safety. The realization of heat switches that can passively regulate temperature by toggling heat flow represents one such area of interest. We explore the use of thermomechanical actuation in LC elastomers as a route to establish limit cycle temperature responses and, thereby, the realization of a passive heat switch. Temperature-dependent force generation on a small widget due to the thermomechanical response of liquid crystal elastomer is coupled with a distance-dependent magnetic restoring force of the widget for a fixed warm surface. Under appropriate conditions, the resulting motion is time-dependent and exhibits limit cycle behavior, with heat periodically transferred to a nearby cold surface. The ability to achieve limit cycle responses depends on a relatively sensitive balance between the force-generation characteristics of the LC elastomer, the force generated by the magnet, and the thermal transport properties of the system. We examine the behavior over a range of experimentally accessible parameters, and construct a minimal model that satisfactorily captures the different response regimes observed. We anticipate that self-sustained cyclical thermal transport of this variety will have application in waste heat harvesting. In particular, the thermal oscillator realized here is of interest for waste heat recovery using solid-state piezoelectric devices.

Liquid crystal elastomer based entangled active matter

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Systems in which individual components convert free energy into mechanical work are commonly referred to as active matter. Physically entangled active matter is an emerging area of both living and man-made systems, where mechano-functionality of the collective emerges through physical interaction of individual elements. Liquid crystal elastomers (LCEs) are a class of stimuli responsive polymers that undergo large, reversible, anisotropic shape change in response to different stimuli, including heat and light, making them ideal candidates for many applications, especially as deployable devices. LCE films fabricated using surface-alignment techniques with prescribed director rotation along the thickness (twisted nematic) exhibit three-dimensional conformations in response to variations in temperature. The addition of magnetic particles enables internal motion of the individual units under a rotating magnetic field. The combination of the induced motion from the magnetic field along with the temperature-induced bending facilitates ribbon entanglement into a macroscopic multifunctional solid. Namely, single ribbon motion was found to exhibit a significant transition across a temperature range of approximately 100 degrees from a simple oscillatory motion to complex trajectories that visit larger amounts of positions in the field. Tuning of the nematic twisting angles (between 0 and 90 degrees) and the offset angles relative to the axes of the surface director (between 0 and 45 degrees) allowed for tuning the critical temperatures for entanglement along with the mechanical properties of the structure. The outcomes of this project are expected to open the way for new active matter systems composed of dynamic elements, where rich material physics can be uncovered and applied towards the development of a variety of unique autonomous devices.

The Effect of Spacer Length on the Properties of Auxetic Liquid Crystal Elastomers

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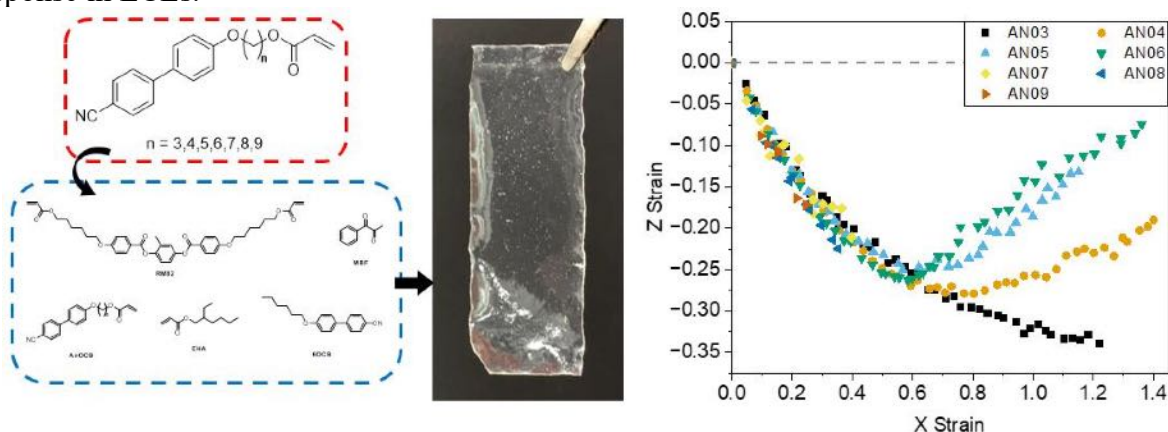
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Auxetics are materials possessing a negative Poisson's ratio, i.e. they get thicker when subject to strain.[1] They typically consist of porous structures, with deformation of the pores during strain causing the auxetic response.[2,3] However, liquid crystal elastomers (LCEs) displaying auxetic behaviour have been reported, realizing the first examples of a non-porous, synthetic, molecular auxetic material.[4,5] The mechanism of the auxetic response is attributed to an out-of-plane rotation of the nematic director under strain, and a resulting emergence of biaxial character within the elastomer.[4,5]

The auxetic LCE can be described as predominantly a side-chain LCE, in which the mesogens are attached to the polymer backbone by a flexible chain known as a spacer. In non-auxetic LCEs and polymers, studies have shown that variations in the length of this spacer can have pronounced impacts on material properties.[6-8] However, as yet the studies of potential structure-property relationships in auxetic LCEs have been minimal.

This work will discuss the synthesis of a series of cyanobiphenyl acrylate monomers with varying spacer lengths, and their subsequent incorporation into liquid crystal elastomers of formulations consistent with the auxetic LCE previously reported. The impact of the change in spacer length in these elastomers on properties such as the phase transition temperatures and the nematic order parameter of the LCEs will be reported. Crucially, the impact of spacer length on the auxetic behavior of the LCEs will be addressed to further our understanding of the auxetic response in LCEs.



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Light driven liquid crystalline actuators with multiple phase transitions

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Over millennia of evolution nature has mastered the ability to control and convert stimuli into a multitude of signals or macroscopic outputs often based on multiple materials working in concert. Synthetically however most materials are based on binary systems where one input results in one specific output with orthogonality providing multi responsive systems. Achieving complex and dynamic mechanical responses to one or multiple stimuli within a single material system would vastly widen the synthetic design space. Specifically soft material actuators are sought after materials in fields like biomedical robotics, wearable devices, and extreme-terrain exploration. However, due to limitations in fabrication resolution and actuation mechanisms, realizing such a system remains challenging.

Here we present a magnetically aligned liquid crystal elastomer (LCE) system showing multiple phase transitions with opposite deformation along the alignment director. Through two independent stimuli (light and heat) a multitude of states can be achieved. Furthermore, simply tuning magnetic field during fabrication, demonstrate high resolution of the alignment resulting in a wide mechanical library capable to access a variety of unique deformations. This system is highly promising for remote controlled actuators capable of multidirectional motion, rotation, and object manipulation.

Soft electricity-responsive liquid crystal polymer materials for switchable surfaces

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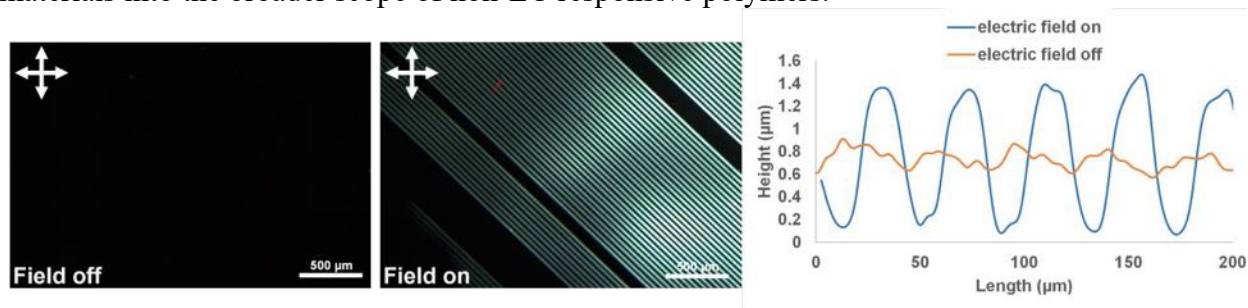
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Liquid crystal polymers are a prime candidate for the preparation of stimuli-responsive surfaces. Especially interesting are systems which respond to electric fields, as such materials are highly applicable in the development of useful products[1]. Realignment of the mesogenic units in a liquid crystal polymer network by DC electric fields leads to large shape deformations of the material[2][3], which is an interesting effect with many potential applications. However, the materials need to be above the glass transition temperature for this effect to be observed, which is well above room temperature for the polyacrylate liquid crystal materials which are generally used.

In this project, new electricity-responsive soft polymer materials based on liquid crystals are developed. The materials are made through a Michael addition reaction between an electricity-responsive monoacrylate reactive mesogen, a diacrylate linker, and a tetrathiol. The diacrylate and tetrathiol form the network, while the monoacrylate mesogen provides the electricity responsiveness. Due to a lack of a high T_g polymer backbone, the materials have a T_g well below room temperature and can easily deform without the need for heating. The ratio between monoacrylates and diacrylates can be used to regulate the crosslinking of the material, and as such the formulation of the reaction mixture has dramatic effects on the behavior of the final film. The new materials are synthesized as thin films on glass surfaces bearing ITO electrodes, which are used to apply in-plane DC electric fields. Polarized optical microscopy is used to study the realignment of the molecules, while contactless 3D surface profilometry is used to study the shape and extent of the deformation. Surprisingly, a formulation was found which results in an isotropic solid film which shows reversible molecular reorientation and surface deformation when the electric field is applied (see Figure). This finding could take these materials into the broader scope of non-LC responsive polymers.



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Liquid Crystal Elastomers: Artificial Muscles and Collective Action

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Aligned liquid crystal elastomers (LCEs) respond to temperature changes by changing form in programmed manners. We see this well-known property as a tool to create actuators and a way to assemble materials and devices. For example, LCE artificial muscles may enable the treatment of stress urinary incontinence. As a soft actuator that can respond to temperatures just beyond body temperature, these materials are well suited to manipulate soft tissues. An artificial LCE sphincter could normally help block urine flow and then be triggered by photothermal heating from an IR light to allow voiding. In a rabbit model of incontinence, the force required to cause urine leakage can be modulated by over 30% based on the temperature of the LCE. We will also discuss the challenges that remain for LCEs as long-term implanted devices. There are also opportunities to use the shape change of LCEs beyond actuators. We will describe using shape morphing ribbons comprised of LCEs to create solids that self-assemble, modulate volume, and disassemble on demand. This is achieved by inducing dispersions of LCE ribbons to mechanically interlock. This approach mimics the aggregations of fire ants, worms, and caterpillars where many 'responsive' units can transiently aggregate to generate new collective functions. Ribbon shape, which is readily controlled in LCEs, and concentration dictate if aggregation occurs and govern the global mechanical properties of the solid that forms. The collective solid can self-assemble after injection. This approach to injectable materials enables new microstructures, which in turn may enable new applications of LCEs.

Synthesis of Liquid Crystalline Networks for Biomaterials Applications

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Hydrogels are desired for use as 3D scaffolds in tissue culture due to their tunable mechanical properties and high-water content that facilitates nutrient exchange. A challenge with existing hydrogels is that, though the natural extracellular matrix of tissues may be ordered, there are a limited number of biocompatible ways to incorporate anisotropy within a hydrogel. Liquid crystalline polymer networks (LCNs) combine ordering and elasticity, which may provide an opportunity to generate anisotropic hydrogels. The application of LCNs as culture substrates has primarily focused on exploiting actuation in two-dimensional (2D) culture on films or in three-dimensional (3D) culture on solid scaffolds. Although LCNs have shown good biocompatibility, ordered microstructure, and tunable mechanical properties, the hydrophobicity of mesogens has hindered their use in polymers that can be cross-linked into hydrogels for 3D cell encapsulation. This work investigates poly(ethylene glycol)-based LCNs, and the ability of the liquid crystalline (LC) phases to affect cell behavior. We report on the synthesis and characterization of water-soluble LC polymers using alkyne-azide cycloaddition (AAC) chemistry. An alkyne-terminated mesogen (**5yHBP**) was polymerized with azide-terminated poly(ethylene glycol) (**PEG**) to impart flexibility and increase the hydrophilicity of the LC polymers. The strong hydrophobic core of **5yHBP** results in LC phase formation, while enough hydrophilic **PEG** content imparts aqueous solubility. The LC polymers can be cross-linked in aqueous media using a tetrafunctional dibenzocyclooctyne (DBCO)-terminated **PEG** via strain-promoted AAC chemistry, which was selected for its high efficiency and cytocompatibility. The resulting networks maintain LC phases in both the dry and hydrated states after cross-linking. Human mesenchymal stem cells (hMSCs) were encapsulated in the hydrogels successfully by cross-linking the LC polymers in the presence of a cell suspension. The results showed excellent viability and proliferation of the encapsulated hMSCs, suggesting potential for LC polymer hydrogels in tissue culture applications.

Stiffness anisotropy coordinates supracellular contractility driving long-range myotube-ECM alignment

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Long-range cellular alignment is critical for muscle tissue development and regeneration.^[1] Muscle stem cells rely on an anisotropic extracellular matrix (ECM) to regenerate functional, aligned myofibers following injury.^[2] However, little is known about the collective cellular response to mechanical anisotropy due to difficulty in preparing anisotropic materials that also lack topography. Here, we used monodomain and isotropic liquid crystalline polymer networks (LCNs) as cell culture substrates to study the effects of mechanical anisotropy, without the influence of topography, on long-range cellular alignment.

Through live-cell imaging and advanced image analysis, we assessed how C2C12s interact with the LCN substrate, neighboring cells, and nascently deposited ECM over increasing length scales to generate long-range order. C2C12 myoblasts seeded on monodomain LCNs (mLCNs) were randomly oriented until they reached a critical cell density threshold, upon which they collectively polarized with the nematic director. As myoblasts differentiated and fused to form multinucleated myotubes, collective cell-ECM flows emerged and increased the orientation-order parameter of the culture. After five days in differentiation conditions, C2C12 myotubes were highly aligned over long distances (>4 mm) on mLCNs with an impressive orientation-order parameter of 0.92. Myotube order was significantly higher on stiffer mLCNs and further scaled with the degree of stiffness anisotropy. In contrast, myotubes cultured on isotropic LCNs that lack stiffness anisotropy did not develop long-range order. Taken together, our multi-scale analysis of collective cellular dynamics reveals new mechanisms by which cell populations are influenced by biophysical interactions to orchestrate distinct coordinated behaviors in development and regeneration.

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A light-fueled cilium

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Biomimicry of natural cilia or flagella to endow active materials with sophisticated functions has been exploited immensely in microfluidics. Though complex cilia-like motions have been kinetically studied, i.e., non-reciprocal motion in a cilium and metachronal wave in cilia synchronization,^[1] these conventional artificial cilia are fundamentally different from their biological counterparts as they are operated in equilibrium states.^[2] Here, we report an optically powered soft material strip that can perform non-reciprocal, self-sustained oscillation under water. The orthogonal excitations allow for piecewise control over the strip deformation, further being able to fully program the non-reciprocity (stroke patterns and handedness), which significantly affects the fluidic pumping efficiency. The autonomous interaction between active matter and surrounding liquids, enables spontaneous microfluidic pumping, translocation of a micro-object by liquid propulsion, and more importantly, coupling between two oscillating strips through liquid medium interaction. The results can open new possibilities for stimulating novel designs for responsive soft robots that possess bio-like functions at their non-equilibrium states.

Keywords: liquid crystal elastomers; photoactuator; artificial cilia; non-reciprocal motion; self-sustained oscillation

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Reconfigurable Shape Change in Liquid Crystalline Elastomers Enabled by Stimulus Orthogonality in Dynamic Covalent Networks

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The ability of liquid crystalline elastomers (LCEs) to undergo stimuli-responsive deformations stems from the disruption of anisotropic structural order within polymer networks. The shape behavior that an LCE exhibits upon actuation depends on the mesogen orientation patterning. Typically, mesogen orientation within the LCE is enforced during initial synthesis of the polymer network such that the LCE retains the programmed orientation and associated properties. However, this approach leaves minimal opportunities for reprogramming due to the permanence of the covalent crosslinks. Therefore, the combination of covalent adaptable networks (CANs) with LCEs is highly desired as a means to enable reconfigurable anisotropic patterns within the material and, as a result, reprogrammable shape changes.

When using dynamic covalent bonds in LCEs, stimulus orthogonality must be considered to ensure that actuation and network reorientation are activated independently since CANs rely on external stimuli to activate bond exchange. In this work, we demonstrate one approach in which LCEs are designed for thermal actuation and photo-activated network reorientation. [1] Using photo-responsive allyl sulfide bonds, the network undergoes reprogramming by mechanically deforming the network and activating bond exchange. The resulting LCE undergoes thermal actuation, exhibiting complex actuated shapes that are not accessible by surface or mechanical alignment alone. In a second demonstration, LCEs are programmed for photoresponsive actuation and thermally activated bond exchange. Notably, in these LCEs, using diarylethene enables thermally stable photomechanical actuation, allowing for thiol-anhydride bond exchange to take place in the nematic phase without compromising photoresponsive properties. [2]

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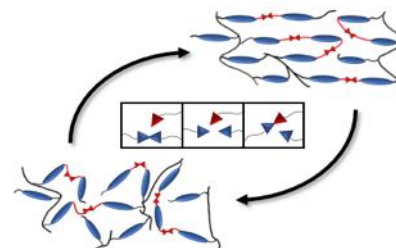


Figure 1. Dynamic bonds enable reprogramming in liquid crystalline elastomers

Electroactive Bi-functional Liquid Crystal Elastomer Actuators

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Liquid crystal elastomers (LCEs) show promising potentials as smart actuators, for example, those contracting/expanding linearly like mammalian muscles.^[1] Direct heating and light illumination are the most used activation mode in LCE actuators because LCEs are based on thermotropic or phototropic liquid crystals.^[1,2,3] However, in the actuator world, electrical energy is the most convenient and in demand stimuli. Indeed, the nature does use electrical impulses between nerves and muscles to trigger actuation with extraordinary efficacy, and electrical stimulation is also more widely utilizable as driving forces in industrial devices.

We present here a trilayer electroactive LCE (eLCE) by combining two smart materials, LCE and ionic electroactive polymer device (i-EAD), which is bi-functional and can perform either bending or contractile deformations under low voltage stimulation. By applying a voltage of ± 2 V at 0.1 Hz, the redox behavior and associated ionic motion in the two conducting polymer electrodes of i-EAD provide bending deformation of the device up to a bending strain difference of 0.8%. On the other hand, by applying a voltage of ± 6 V at 10 Hz, the ionic current-induced Joule heating triggers the muscle-like contractile response of the central ion-conducting LCE component, a linear contraction ratio of 20% being obtained without load. Moreover, a load of 150 times of eLCE weight can be left with a 20% strain. This approach of combining two smart polymer technologies, LCE and i-EAD, in a single device, is promising for the development of smart materials with multiple degrees of freedom in soft robotics, electronic devices, and sensors.

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Supramolecular Liquid Crystalline Elastomers

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Liquid crystalline elastomers (LCEs) are lightly crosslinked networks that combine the elastic properties of rubber with the anisotropic properties of liquid crystals. These materials exhibit large deformations, when exposed to a stimulus such as heat, due to the disruption of liquid crystalline order.¹ LCEs, however, are currently limited for functional applications due to a weak coupling of stimuli and response. To address this limitation, our group has demonstrated that the inclusion of liquid crystalline mesogens formed with supramolecular bonds results in improved actuation properties.²

Here, we analyze how the intra-mesogenic supramolecular bonds behave throughout the phase transition of LCEs. Dimers are formed within the crosslinks through the combination of benzoic acids and are shown to be disrupted with temperature and throughout the nematic to isotropic phase transition.³ These dimerized mesogens are included with both three ringed and two ringed mesogens, which exhibit reduced intermolecular interactions. This allows us to probe how both the surrounding network and heat affect the disruption of H-bonds.

This effort is also focused on the structure-property effect as we vary the type of supramolecular mesogen to understand the connection between H-bond strength and actuation dynamics. Benzoic acid derivatives are included with molecules containing functional groups such as pyridines to form single hydrogen bonds. The performance of the LCEs can be controlled by varying the strength of the hydrogen bonds and the mesogenic intermolecular interactions. Overall, we are working to understand the influence of H-bond interactions on LCE properties and tailor this response through molecular engineering.

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Trainable liquid crystal elastomers via the controlled synthesis and dynamic covalent exchange of aza-Michael reactions

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Trainability in soft materials can be defined as the material's ability to reorganize its constituent units in response of macroscopic mechanical deformation such as external environmental stress. Liquid crystal elastomers (LCEs) are lightly crosslinked polymer networks that combine the mechanical properties of elastomers with the anisotropy of small molecule liquid crystals. LCEs possess the unique capacity to rearrange their mesogenic units in response to mechanical deformation, making them suitable as trainable materials. Furthermore, addition of dynamic covalent bonds in LCEs allows for reprogrammability and reconfigurability. In this work, we studied the influence of various hydrogen-bond donor catalysts on aza-Michael reactions in LCEs. First, we demonstrated that by tuning the electronics of the catalyst, the amine-acrylate reactions could be significantly enhanced. Building upon this knowledge, we catalyzed the synthesis of aza-Michael LCEs using a one-pot reaction, incorporating a liquid crystalline diacrylate monomer, an amine-containing chain extender and crosslinker. Additionally, the dynamic exchange behavior of amine-acrylate adducts in LCEs was investigated through stress relaxation and creep measurements, providing insights into the resulting training responses. Moreover, we studied the ability to selectively activate or deactivate the thermal reprogrammability of LCEs by post-synthetically introducing or removing the catalyst through swelling and soxhlet extraction, respectively. This dynamic exchange control facilitated the induction of reprogrammability while maintaining desired network stability. Overall, this work contributes to the development of LCEs as trainable materials with adaptable responses to external stimuli, opening avenues for their application in various fields.

Body Temperature Actuation of Exchangeable Liquid Crystal Elastomers

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Conventional liquid crystal elastomers (LCEs) often require high actuation temperature, usually above 65 °C, which limit practical applications of LCEs. To address this limitation, various methods have been employed to tune and reduce the actuation temperature of LCEs, including adjusting mesogen constituents, alignment conditions, and network structures.^[1-7] Recent studies have shown promising results in achieving actuation temperatures that are sufficiently low, approaching or even below ambient temperature.^[8] However, these studies have primarily focused on conventional LCEs with permanent crosslinkers. In this study, we have successfully synthesized exchangeable LCEs (xLCEs) incorporating dynamic thiourea bonds with body-heat actuation capability based on our previous report.^[9] In particular, the impact of molecular structure and composition of LCE building blocks on the mechanical, viscoelastic, relaxation and actuation properties of the xLCEs is systematically investigated. Notably, effective modulation of actuation temperature with a wide range that includes body temperature is demonstrated (**Figure 1**). The reprocessable LCEs capable of reversible shape morphing triggered by body heat have significant potential for expanding LCE applications in wearable, haptic and biomedical devices.

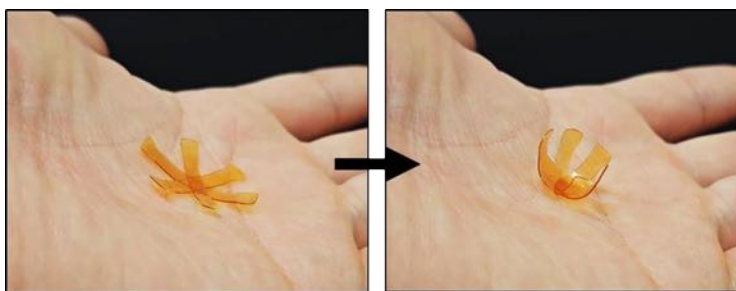


Figure 1. A palm-heat actuation of an exchangeable LCE actuator.

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Halogen-bonded shape memory liquid crystal polymer

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(presented by Arri Priimagi)

Stimuli-Responsive Liquid Crystalline Elastomers (LCEs) are a class of materials that can undergo significant changes in their properties and shape in response to external stimuli. In particular, harnessing the power of halogen bonding (XB), a non-covalent interaction between an electron-deficient halogen atom and a Lewis base, holds great potential for developing advanced LCEs with unique functionalities. In our study, we present a novel approach utilizing a liquid crystalline network comprising both permanent covalent crosslinks and dynamic halogen bond crosslinks. By incorporating XB into the LCE matrix, we achieve a reversible thermo-responsive shape memory behavior. The I···N halogen bond, a widely studied motif in crystal engineering, plays a pivotal role in enabling temporary shape fixation at room temperature and subsequent shape recovery when exposed to human body temperature. Notably, our research showcases the remarkable versatility of shape programming in these halogen-bonded LCE networks through simple human-hand operation. Additionally, we propose a micro-robotic injection model that allows for complex 1D to 3D shape morphing in aqueous media at 37 °C. Through meticulous structure-property-performance studies, we establish the indispensable contribution of the I···N crosslinks in driving the shape memory effect observed in our halogen-bonded LCEs. The introduction of halogen bonding expands the toolbox for designing smart supramolecular constructs with tailored mechanical properties and thermoresponsive behavior, opening up exciting possibilities for future medical devices and other advanced applications. ^[1]

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Understanding the effect of liquid crystal content on the phase behavior and mechanical properties of liquid crystal elastomers

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Liquid crystal elastomers are stimuli-responsive, shape-shifting materials. They typically require high temperatures for actuation which prohibits their use in many applications, such as biomedical devices. In this work, we demonstrate a simple and general approach to tune the order-to-disorder transition temperature (T_{ODT}) or nematic-to-isotropic transition temperature (T_{NI}) of LCEs through variation of the overall liquid crystal mass content. We demonstrate reduction of the T_{NI} in nematic LCEs through the incorporation of non-mesogenic linkers or the addition of lithium salts, and show that the T_{NI} varies linearly with liquid crystal mass content over a broad range, approximately 50 °C. We also analyze data from prior reports that include three different mesogens, different network linking chemistries, and different alignment strategies, and show that the linear trend in T_{ODT} with liquid crystal mass content also holds for these systems. Finally, we demonstrate a simple approach to quantifying the maximum actuation strain through measurement of the soft elastic plateau and demonstrate applications of nematic LCEs with low T_{ODT} s, including the first body-responsive LCE that curls around a human finger due to body heat, and a fluidic channel that directionally pumps liquid when heated.

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Photo-responsive liquid crystal elastomers: A molecular Monte Carlo study

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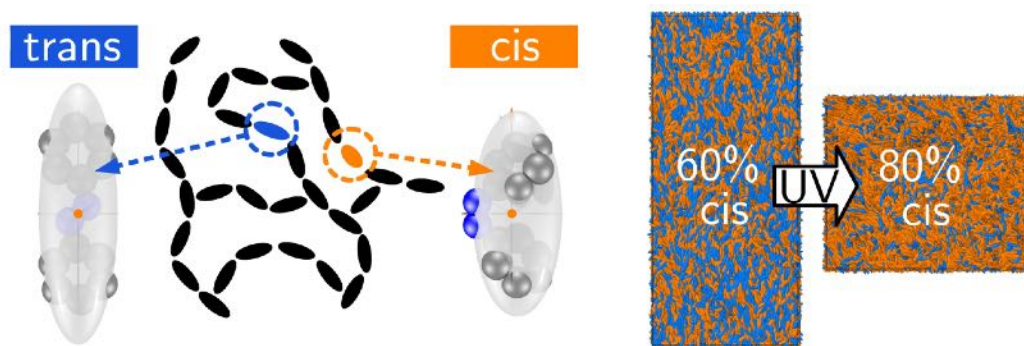
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We investigate main-chain liquid crystal elastomers formed by photo-responsive azobenzene units [1] with different populations of *trans* and *cis* conformers. We study their macroscopic properties as well as their molecular organization through extensive Monte Carlo simulations at coarse-grained level where both types of conformers are represented by soft-core biaxial Gay-Berne particles [2] with size and interaction energy parameters obtained by fitting the corresponding azobenzene moiety represented at atomistic level. Increasing the fraction of *cis* conformers, as through near-UV irradiation, turns out to shift the nematic-isotropic transition to a lower temperature, while generating internal stress in a clamped sample. Further, an analysis of pair distributions shows that the immediate surroundings of a bent *cis* molecule are slightly less dense and more orientationally disordered in comparison with those of a *trans* conformer. Comparing nematic and smectic elastomers, actuation in the smectic phase proved less effective, disrupting the smectic layers to some extent but preserving the orientational order of azobenzene moieties [3].



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Enhanced Energy Dissipation of 3D Printed Liquid Crystal Elastomers and Their Material Structures

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Polymer foams and architected metamaterials are increasingly being used to protect valuable assets against mechanical shock and vibration. Compared to conventional metal structures, these lightweight materials provide several advantages such as large recoverable strains under repeated loading and excellent chemical resistance for prolonged usage in harsh environments. 3D printing enables the creation of innovative metamaterials with tailored unit cell geometry, providing enhanced design flexibility to control their mechanical properties.

In this talk, I will introduce our recent work focusing on the 3D printed material structures for enhanced energy dissipation. In our study, the recently emerged liquid crystal elastomers (LCEs) are adopted as the base material for the 3D printing of energy dissipative material structure. LCEs represent a transformative energy absorption material with mesogens built on the chain backbone. In addition to the viscoelastic relaxation of polymer chains, the mechanically induced mesogen rotation provides an additional mechanism for energy dissipation. We use Digital Light Processing (DLP) 3D printing to carefully control foam periodic meso-structure. In both quasi-static and dynamic loading conditions, the LCE lattices show enhanced dissipation in uniaxial compression compared to the non-LCE foams. The lattice geometry plays a prominent role in determining the dissipation ratio. When increasing the connectivity, the deformation is translated from bending-dominated to stretching-dominated, which generates higher axial strain in the struts and higher energy dissipation of the lattice. The findings hold great potential for harnessing the capabilities of 3D printing using advanced polymer materials to develop the next generation of protective foams.

Direct and indirect evidence of biaxial order in auxetic liquid crystal elastomers

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Recently, some LCEs have displayed an auxetic response, [1,2] which means that they become thicker when stretched. Until the discovery of auxetic LCEs, auxetic materials were made by designing porous structures into otherwise positive Poisson ratio materials. However, LCEs are auxetic on a molecular level, which has long been a goal of the auxetic community.

Auxetic LCEs, have been found to deform via a ‘mechanical Fréedericksz transition’, characterised by an apparent sharp rotation of the director at a critical strain, in contrast to the more typical ‘semi-soft elastic response’. [3] The apparently discontinuous nature of the ‘mechanical Fréedericksz transition’, together with measurements of the order parameters *via* Raman scattering, suggest that biaxial order may be important in auxetic LCEs. [2,4,5] We present direct evidence of biaxial order emerging in strained auxetic LCEs observed *via* conoscopic microscopy. [5] We show that the observations complement results from Raman spectroscopy, which probes the orientational order of the LCE as a function of strain, and reveal the emergence of biaxial order. Calculation of the orientational distribution functions shows that this facilitates the auxetic response *via* out-of-plane rotation of the mesogenic units. [2]

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International Liquid Crystalline Elastomer Conference
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Abstracts

- Oral presentations (listed in order)
- **Poster presentations**

Poster #1

Donor Acceptor Stenhouse Adducts for deep, white light photo-mechanical response of liquid crystal elastomers.

Jesus Guillen Campos¹, Mykhailo Pevnyi², Sara Sandlass³, Cassidy Tobin³, Minwook Park¹, Peter Palffy-Muhoray² and Javier Read de Alaniz¹.

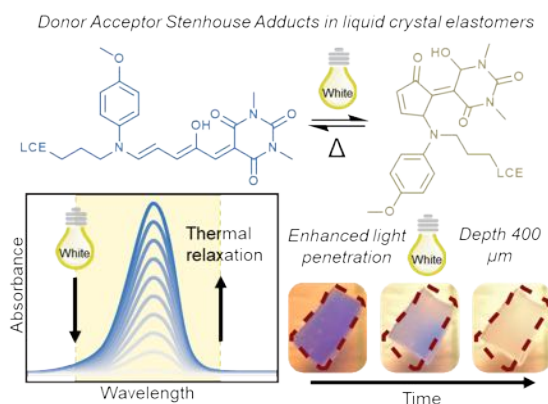
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The ability of spatial-temporal control of motion and optical properties of micro soft robots is an attractive feature of light responsive liquid crystal elastomers (LCEs), however, the use of this material for real-world applications has been constrained by the practicality of the used wavelengths of light utilized and the robustness of the actuators. The wide-spread use of UV or near UV triggered photoswitches like azobenzene require the use of high energy, wavelength specific lasers, while the penetration depth is limited to a couple of microns below the surface limiting the effective use of the photo-responsive molecule to LCE thicknesses in the tens of microns scale. Here we report for the first introduction of donor-acceptor Stenhouse adducts (DASAs) in LCEs to enable reversible, deep-penetrating, and practical white light induced motion with full penetration of this films up to 400 μm thick. Introduction of DASA is done *via* a post-alignment functionalization where the nematic phase is retained with the photoswitch self-aligning along the director upon introduction enabling photo-thermal and photo-chemical actuation.



Monodomain Nematic Liquid Crystal Elastomer of Arbitrary Thickness Through PET-RAFT Polymerisation

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Liquid crystal elastomers (LCEs) can possess several interesting properties, such as significant changes in shape upon application of stimuli [1-3], or auxetic behaviour [4]. The alignment of mesogens within the elastomer determines the extent to which these interesting phenomena are observed [3,4]. Without attempts to align the mesogens, polydomain LCEs, in which order exists in localized microscopic domains, are isolated. The lack of macroscopic order can eliminate the occurrence of phenomena such as auxetic or stimuli-responsive behaviour [3]. Consequently, the alignment of mesogens to yield monodomain LCEs, in which alignment is maintained throughout the sample, is desirable.

Several techniques have been developed to achieve monodomain alignment. One such technique is the use of surface alignment, which relies on mesogens interacting with surface coatings and in doing so being topologically aligned [2]. Surface alignment has many advantages over other techniques, such as enabling complex director patterns to be obtained [2]. However, the surface alignment technique is limited to low sample thicknesses (typically 100 μm), as the affinity for the mesogens to align decays with increasing distance from the alignment substrate [2].

Photoinduced electron/energy transfer reversible addition fragmentation chain transfer (PET-RAFT) polymerisation is a means of achieving living free-radical polymerisations [5]. It allows for a polymerisation to be stopped and started by the application or removal of a light source [6]. In this work, we present results that demonstrate the exploitation of this stop-start capability to produce monodomain LCEs of up to 300 μm thickness, aligned via surface alignment.

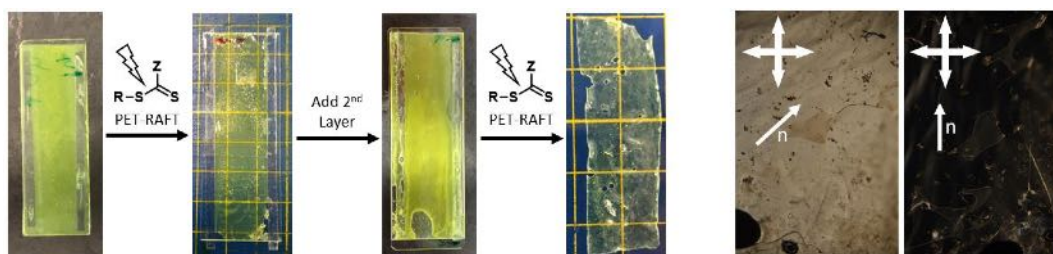


Figure 1 – The PET-RAFT process employed in this work, and the resulting planar alignment of the LCE.

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Poster #3

Enhancing the Electromechanical Response of Dielectric Liquid Crystalline Elastomer Actuators

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Liquid crystalline elastomers (LCEs) are crosslinked polymer networks made up of directionally oriented mesogens. Previous research has demonstrated the ability to use thermal or optical stimuli to interrupt this orientation to produce a directional actuation response [1]. The compelling stimuli-response of LCE exhibit large deformation, at times significant work, but at low cycle frequencies. Accordingly, this and prior examinations have considered the utilization of electrical stimuli to induce direct mechanical effects to realize faster and potentially more efficient actuation in LCEs.

An initial report detailed the utilization of LCEs as dielectric elastomer actuators (DEAs) [2]. The inherent anisotropy of the elastic modulus and the dielectric constant in LCEs leads to an anisotropic actuation response that can be spatially programmed. Through adjustment of the composition the achievable strain of LCE DEAs was increased to 20% [3]. By further investigating new materials chemistries and compositions for DEAs with LCEs, we look to gain a deeper understanding of composition-property-performance relationships governing these materials. This presentation will detail these relationships with the goal of minimizing energy losses and improving the achievable deformation in DEAs made with LCEs.

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Liquid crystal elastomer fibers for active textiles

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Liquid crystal elastomers are a class of shape-changing polymers capable of undergoing reversible shape changes in response to changes in external stimuli. Our goal is to create liquid crystal elastomer fibers with high actuation strain and then integrate them with commonly used fabrics to make active and wearable textiles. A two-step reaction is used for the synthesis of the fibers. The first reaction is the Michael addition of a nematic diacrylate monomer with a dithiol chain extender. The resulting thiol-terminated liquid crystal oligomer can be extruded, creating long fibers on a rotating mandrel. During the printing phase, the fibers undergo a photocuring process to crosslink the aligned oligomers. These aligned fibers demonstrated a $36\% \pm 6$ reversible actuation strain between 25°C and 60°C . The fibers are then transferred to a sewing machine bobbin to be used as the bottom thread in a commercially available sewing machine. Design patterns are fed to the machine for fast and automated sewing of complex patterns onto the fabric. For example, when an intricate pattern of concentric squares is sewn on top of a cotton fabric a resulting 3D cone shape is formed when actuated. Additionally, sewing a layer of stainless-steel fibers under the LCE design can allow us to actuate local areas of the fabric remotely via electrothermal stimuli. Therefore, by integrating these functional fibers into textiles can introduce complex shape change in the fabrics and this has the potential to be used in biomedical applications such as compression garments and e-textiles.

Aligned liquid crystalline elastomers for directional adhesion

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Pressure sensitive adhesives (PSAs) are ubiquitous in everyday life. Conventional PSAs are nonreactive adhesives and require low pressure to stick to a surface. Here, we are interested in directional PSAs, in which the peel force to remove the adhesive is substantially greater along one direction than another. Prior research has realized directional adhesion by introducing patterns of modulus stiffness, surface topography, or chemical functionality. Herein, we detail distinctive opportunities to realize directional adhesion in monolithic, aligned liquid crystalline elastomers (LCEs). LCEs are crosslinked polymer networks with unique anisotropic mechanical properties associated with their unidirectional alignment of molecules along the director axis. When aligned, these materials exhibit classic elastic behavior when tensile load is applied along the director; however, when tensile mechanical load is applied orthogonal to the director, the liquid crystalline molecules reorient towards the load direction. This reorientation results in a highly nonlinear stress-strain response and a distinct means of anisotropic energy dissipation, otherwise known as soft elasticity. In this work, we demonstrate a LCE PSA that exhibits distinct directional adhesion. The peel strength is locally controlled based on the geometry of the aligned liquid crystalline molecules and the resulting energy dissipation. We show that varying the material stiffness dictates the peel strength and peel anisotropy.

Poster #6

Evaluating the Impact of Soft Elasticity on Crack-Tip Strain Fields in Nematic Elastomers

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Ideal nematic liquid crystal elastomer (LCE) is theoretically expected to deform without extra energy cost through the director rotation. This feature is known as soft mode or soft elasticity, achieved by combining LC alignment and rubber elasticity [1]. Indeed, real nematic LCEs with polydomain director alignment can undergo a transition to a globally aligned monodomain state under the application of exceedingly small tensile force.

Experimental studies on soft elasticity for LCEs have primarily focused on deformations with uniform strain distributions. Little experimental work has been done on deformations with nonuniform strain distributions, such as those found in cracks. Understanding the crack properties is crucial from both academic and practical viewpoints, given that the crack growth leads to fracture. Our previous work characterized both stationary and dynamic cracks in isotropic (non-LC) elastomers, revealing unique crack-tip properties using a digital image correlation (DIC) technique [2].

In the current study, we focus on the characterization of nonuniform strain fields around the crack-tip of nematic elastomers. We prepared thiol-acrylate main-chain polydomain LCEs using the methods in Ref [3] and used Styrene-Butadiene Rubbers (SBR) for comparison. Our results demonstrate LCEs exhibit distinct features in the crack-tip strain field, originating from the properties of soft elasticity.

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Slidable Polyrotaxane Networks in Liquid Crystal Elastomers: Toward Extreme Damping Materials

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Damping for structural stability and noise reduction plays a crucial role in a wide range of applications such as mechanics and construction. Conventional damping materials rely on isotropic elastomers, but recent studies suggest anisotropic elastomers such as liquid crystal elastomers (LCEs) can outperform the isotropic elastomers.^[1,2] Herein, we synthesize a new class of LCE incorporating slidable polyrotaxane (PR) cross-linkers (PR-LCE) to achieve extreme damping.^[3] In particular, we investigate viscoelastic and mechanical properties as well as damping performances of PR-LCEs with both tensile and compressive test at various temperatures. The results are compared with conventional LCEs and isotropic elastomers. Interestingly, both the soft elasticity and damping performances of the PR-LCE are significantly affected by the concentration of PR crosslinkers and the number of cyclodextrin rings within PR crosslinkers. In addition, the energy dissipation of PR-LCE foams with various pore sizes are further examined. The unique energy dissipation of LCE with slidable cross-links can be promising for creating high-performance shock and sound absorbing materials.

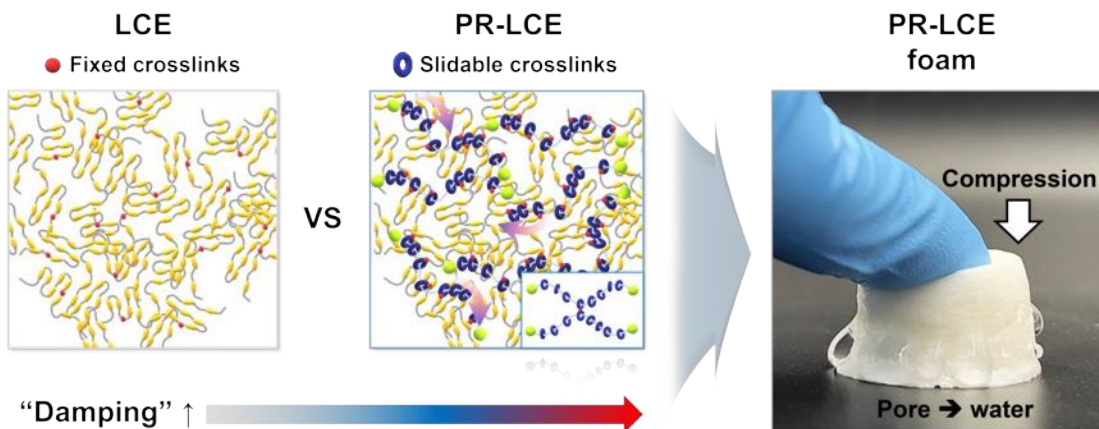


Fig. 1. Schematic of slide-ring effect for LCE incorporating PR crosslinker(left) and a photograph of PR-LCE foam fabricated via salt leaching(right)

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Poster #8

Selective Patterning of Auxetic Liquid Crystal Elastomers

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Auxetic materials show the property of a negative *Poisson's ratio*, under strain they will become thicker in at least one transverse direction. Auxetic materials show an increased indentation resistance as well as increased energy absorption when compared to non-auxetic materials^[1].

Some acrylate-based liquid crystal elastomers (LCEs) show auxetic behavior when a critical threshold strain is reached and a thickening in one transverse direction is observed^[1]. Such materials represent the first synthetic molecular auxetic materials; they maintain constant volume under strain in contrast to most other auxetic materials where the behavior is observed due to an increase pore volume within the material. To date, the auxetic acrylate LCEs have been synthesized via an uncontrolled free-radical polymerization, but this limits some of the geometries that can be readily achieved.

Photo-induced electron/energy transfer reversible addition fragmentation chain transfer (PET-RAFT) polymerization, a 'living' form of free radical polymerization^[2], was used in conjunction with a photomask to selectively polymerize the liquid crystal elastomer until it was dimensionally stable, allowing for the unpolymerized mixture to be washed away. The 'gaps' could then be filled with a differently oriented LCE, allowing for complex patterns.

We describe ~100 μm thick nematic LCEs fabricated with planar alignment for an auxetic response in the thickness or homeotropic alignment for an auxetic response in the width^[3]. LCEs containing domains of planar alignment rubbed at arbitrary angles as well as domains cured in the isotropic phase can be produced using this method and their mechanical properties are discussed.

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Poster #9

Novel Methods in Controlling the Crystallinity and Rare Mesogen Alignment in Liquid Crystalline Elastomer Thin Films

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Liquid crystal elastomers (LCEs) are at the forefront of scientific interest due to their distinctive blend of liquid crystals' anisotropic properties and rubbers' elasticity. Their potential for stimuli-responsive actuation applications, including artificial muscles or soft robotics, is being widely studied, and their aptness for applications like anisotropic tissue engineering scaffolds, which benefits from their biocompatibility, anisotropic features and elasticity is also noteworthy.^[1] This study introduces innovative techniques to fine-tune LCE's alignment patterns, crystallinity, and stiffness. We use organic solutions of varied concentrations of photochemical crosslinkable liquid crystal free mesogens to modulate the soaking-in uptake of free mesogens in preformed LCEs, achieving efficient control over crystallinity and stiffness. On the other hand, while the programmable mesogen orientation in LCE thin films through mechanical stretching is well-established,^[2] achieving vertical or helical alignments relative to the film surface via mechanical stretching still presents a challenge due to lack of proper tools. The second part of the work presents a solution to this challenge via chemically bonding LCE thin films to thiol-coated, surface-functionalized glass slides via a sandwiching assembly. By vertically stretching this assembly, liquid crystal mesogens were aligned perpendicular to the film's surface, which were confirmed via in situ optical microscopy and spectroscopy. Additionally, twisting the sandwiching glass slides resulted in twisted mesogen alignments in LCEs. In summary, this research paves the way for advanced methods in controlling LCE crystallinity and alignment, making it a valuable contribution to new LCE applications.

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Poster #10

Elastocaloric Effects in Amorphous and Liquid Crystalline Elastomers

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Elastocaloric cooling is a promising emergent alternative technology to vapor-compression refrigeration. Here, we focus on the potential of amorphous polymer networks containing appreciable liquid crystalline (LC) content and supramolecular hydrogen bonds to exhibit elastocaloric behavior through mechanotropic phase transitions upon deformation. Due to the intermolecular interactions from LC constituents and incorporation of supramolecular bonds, these materials are compelling candidates to achieve first-order mechanotropic phase transitions. Through a two-step thiol-Michael oligomerization/supramolecular chemistry approach, we tune the nematic to isotropic transition temperatures (T_{NI}) of these liquid crystal-rich amorphous polymer networks to occur at or below room temperature. Further, we find these networks manifest enhanced elastic recovery in response to strain cycling accompanied by minimal mechanical hysteresis. The large order and entropy changes observed upon deformation yield significant elastocaloric temperature changes in response to minimal stress and strain applied to the polymer network due to exothermic contribution of breaking hydrogen bonds. Our work provides foundational insights for the mechanistic understanding of elastocaloric properties of LC-rich polymer networks as well as formulation-based opportunities for tuning the properties of LCE for commercial solid-state cooling.

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Poster #11

Phase Separation of Homopolymer Blended Liquid Crystal Elastomers for Creation of Porous Structures

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By introducing pores into liquid crystal elastomer (LCE), porous LCE has more advantages than solid materials, such as lower density, higher processability, and larger surface areas^{1,2}. The porous structure of LCE is more adaptable for incorporating functional materials with different properties. As a 3D scaffold for cell cultures, the interconnected pores of LCE can provide suitable mass transport for cells to access nutrients, gases and waste management³⁻⁵. The significantly increased surface area in the LCE can further enhance the photoresponse to increase bending angle⁶, and electrothermal⁷ or humidity⁸ effect to achieve a greater degree of deformation. These pores are usually introduced by gas release to generate free volume⁶, sacrificial metal scaffold⁴, or particle leaching methods^{5,7}, with micron-sized and randomly distributed. In this work, homopolymers are introduced into the LCEs as sacrificial domain through blending, which can provide customizable material properties, but avoid complex synthesis procedure as required for covalently bonding polymers. It will provide a fundamental understanding of the self-assembly and phase separation behavior of homopolymer blended liquid crystalline polymer system, and the effect of molecular weight and ration on domain distribution and orientation in the resulting porous LCEs.

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Poster #12

Morphological Study of the Order-Disorder Transition in Liquid Crystalline Elastomers Containing Supramolecular Mesogens

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Liquid crystalline elastomers (LCEs) consist of lightly crosslinked networks which combine the elastic properties of rubbers and the molecular anisotropy of liquid crystals (LCs). When aligned LCE networks are subjected to external stimuli, to include heat, these materials can undergo large deformations due to the order-disorder transition (ODT) of the liquid crystalline phase.¹ In order for these materials to be incorporated into functional applications, such as soft robotics, the stimuli-response of these materials require enhancement. Our lab recently synthesized and demonstrated LCE networks containing both conventional mesogens and LC mesogens formed from supramolecular bonds. These networks show an enhanced ODT response over networks only containing conventional LC mesogens during actuation testing.²

In this work, a morphological study of the network ODT was probed by conducting in situ heat treatments during synchrotron X-ray scattering experiments. Simultaneous small/wide angle X-ray scattering (SAXS/WAXS) allowed for phase confirmation and distinguishing the contribution from conventional and supramolecular mesogens when undergoing the ODT during heating experiments. By varying both the concentration and conventional mesogen, we are able to probe the influence of how network chemistry and hydrogen bonding impact the stimuli response properties of these LCE networks.

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Poster #13

Metrologies for tracking heterogenous deformation of main-chain liquid crystal elastomers

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Liquid crystal elastomers (LCEs) couple liquid crystalline regions (*i.e.* mesogens) into polymer networks that result in unique functionality for shape memory and dissipation applications. These networks may be actively manipulated via external fields such as stress, electromagnetic, or thermal to locally orient the mesogen phases and enable a specific material response. The rotation of mesogen groups in response to an external stress creates a non-linear stress-strain response and large hysteretic behavior. The evolution of mesogen re-orientation is coupled with the elastomeric network topology, initial nematic order, applied load and loading rate. Better measurements of these structure-property transitions are needed to develop molecular and structural design pathways. In this work, we investigate the relationship between mesogenic order and viscoelasticity for polydomain LCEs. A high-level understanding of these relationships is developed using uniaxial tensile tests with digital image correlation to capture the inhomogeneous evolution of strain as the microstructure order in the specimen changes with the degree of stretch.

Poster #14

Analysis of orientation and deformation of liquid crystal polymer spheres controlled by electric field

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Liquid crystal networks (LCNs) exhibit anisotropic mechanical response properties, whereby external stimuli can induce changes in their cross-linked networks, leading to macroscopic deformations. Conventional planar or surface alignment methods have limitations in designing three-dimensional structures and alignments [1-2]. In this study, we utilize glycerin-water solutions as aqueous environments to create liquid crystal microdroplets, where the formation of LC droplets is predominantly governed by surface tension [3]. Due to the anisotropic alignment of LCN, such as radial or bipolar alignment, the LC microspheres undergo a transition from a spherical shape to an elliptical shape during deformation. By employing light stimuli, we successfully demonstrate the transformation of a 20 μm diameter LCN microsphere into an ellipsoid with a major axis of 23 μm and a minor axis of 17 μm , achieving a maximum deformation rate of 20 %. Such light-induced micro spherical actuators hold promise for various applications in the field of bioscience, enabling advancements in areas such as microfluidics, biomedical engineering, and biological studies.

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Poster #15

Control and Analysis of Mechanical Response in Liquid Crystal Networks: Formation and Deformation of Three-Dimensional Spherical Structures

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Liquid crystal networks (LCNs) exhibit anisotropic mechanical response properties, whereby external stimuli can induce changes in their cross-linked networks, leading to macroscopic deformations. Conventional planar or surface alignment methods have limitations in designing three-dimensional structures and alignments [1-2]. In this study, we utilize glycerin-water solutions as aqueous environments to create liquid crystal microdroplets, where the formation of LC droplets is predominantly governed by surface tension [3]. Due to the anisotropic alignment of LCN, such as radial or bipolar alignment, the LC microspheres undergo a transition from a spherical shape to an elliptical shape during deformation. By employing light stimuli, we successfully demonstrate the transformation of a 20 μm diameter LCN microsphere into an ellipsoid with a major axis of 23 μm and a minor axis of 17 μm , achieving a maximum deformation rate of 20 %. Such light-induced micro spherical actuators hold promise for various applications in the field of bioscience, enabling advancements in areas such as microfluidics, biomedical engineering, and biological studies.

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Poster #16

Multi-Material Printing of Liquid Crystal Elastomer with Integrated Stretchable Electronics

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Liquid crystal elastomer (LCE) has grown in popularity in recent years as a stimuli-responsive material for soft actuators and shape reconfigurable structures. To make these material systems electrically responsive, they must be integrated with soft conductive materials that match the compliance and deformability of the LCE. This study introduces a design and manufacturing methodology for combining direct ink write (DIW) 3D printing of soft, stretchable conductive inks with DIW-based “4D printing” of LCE to create fully integrated, electrically responsive, shape programmable matter. The conductive ink is composed of a soft thermoplastic elastomer, a liquid metal alloy (eutectic gallium indium, EGaIn) and silver flakes, exhibiting both high stretchability and conductivity (order of 10^5 S m^{-1}). Empirical tuning of the LCE printing parameters gives rise to a smooth surface ($<10 \mu\text{m}$) for patterning the conductive ink with controlled trace dimensions. This multi-material printing method is used to create shape reconfigurable LCE devices with on-demand circuit patterning that could otherwise not be easily fabricated through traditional means, such as an LCE bending actuator able to blink a Morse code signal and an LCE crawler with an on/off photoresistor controller. In contrast to existing fabrication methodologies, the inclusion of the conductive ink allows for stable power delivery to surface mount devices and Joule heating traces in a highly dynamic LCE system. This digital fabrication approach can be leveraged to push LCE actuators closer to becoming functional devices, such as shape programmable antennas and actuators with integrated sensing.

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3D Printing Magnetically Aligned Liquid Crystalline Elastomers

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3D printing via direct ink write (DIW) is an established method to prepare spatially complexly aligned liquid crystalline elastomers (LCE).^{1,2} To date, DIW printing of LCEs is limited by the direct association of alignment and print path. Vat polymerization techniques including digital light processing (DLP) have also been considered in the preparation of LCE. Prior research have detailed the preparation of unaligned (polydomain) LCE to explore their exceptional damping and impact resistant properties.^{3,4} Until recently, DLP printing of LCEs have not retained alignment. Here, we detail research in which we utilize a magnetic field of relatively low strength to align LCEs during DLP printing. We show that fundamental variables such as thickness, magnetic field exposure time, and material composition affect the alignment properties. We print aligned LCE into a variety of structures.

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Vector Slicer for rapid generation of paths for 3D printing

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Fig. 1. Evertor director pattern as designed (A) [1], and sliced and fabricated (B). The actuation of the evertor turns the flat annulus inside out, with its original inner radius becoming the outer one.

The field of 3D printing of liquid crystal elastomers has been rapidly developing since the first demonstrations were shown back in 2017. Since then, a large focus has been put on the material aspect of the process, both in terms of synthesis and fabrication of composites. However, most of the printed demonstrations are limited to a few simple designs, with uniaxial and azimuthal director patterns being the most common.

The reason for that is that despite the 3D printing of LCEs being built on extensive knowledge of additive manufacturing, it does not readily benefit from the slicing software, which is developed with replicating the designed geometry in mind, while LCEs additionally require control over the local printing direction. In order to address this issue, we developed the Vector Slicer – tool that allows for the generation of planar patterns with control over both the geometry and the director pattern [2]. The slicer opens 3D printing to director patterns which were previously only accessible using photoalignment, allowing for the fabrication of thick film actuators with director-encoded functions.

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Poster #19

Lifted Surfaces via Direct Ink Write Printing of LCEs

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Liquid crystal elastomers (LCEs) can be spatially patterned to localize stimuli-response and realize out-of-plane deformation. Classically, this is well-studied in the case of a director profile described as an azimuthal +1 defect that transforms from a flat sheet into a cone when heated. Inspired by recent predictions¹, we have explored the ability to prepare surfaces that lift out of plane in printed LCE. We utilize multi-material direct ink write printing, where inactive elastomers and LCEs are co-printed into a single material system. This form of 3-D printing allows for deformations that lift flat surfaces using LCEs. These lifting deformations are analyzed mechanically to see to what extent the geometrical incompatibility between the active and inactive parts of the print affects the force output of the LCE. In addition, more complex 3-D shape changes are shown in prints with different geometries such as elongated rectangles or corners. Finite element analysis modeling confirms the shape changes seen experimentally and provide insight into the forces that give rise to these complex deformations.

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Leveraging Low-Intensity Light for Omnidirectional Self-Oscillator with Scalability Potential.

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Self-oscillators driven by light and based on stimuli-responsive soft materials have garnered significant attention due to their potential in a wide range of robotic functions, including adaptation, autonomous locomotion, and energy conversion. However, the scalability and implementation challenges arising from the high energy density and unidirectionality of the excitation light beam have hindered their application. Here, we introduce a self-oscillator model employing a lampshade-like smart material assembly that responds to light stimuli. This assembly leverages a low-phase transition temperature liquid crystal elastomer as the photomechanical component (Figure 1a) [1], enabling twisting movements when exposed to low-intensity and incoherent light (Figure 1b). By incorporating a lampshade frame composed of four spiral segments, an innovative negative feedback mechanism is achieved, with an equal amount of light being shadowed to sustain oscillation under a constant light field from omnidirectional excitation ($0^\circ - 360^\circ$ azimuth and $20^\circ - 90^\circ$ zenith) (Figure 1c). To showcase the concept's versatility, we fabricated oscillators of different sizes with diameters ranging from 6 mm to 50 mm, demonstrating the potential for scalable implementation [2]. This research offers new insights into the rapidly evolving soft material-based micro movements field.

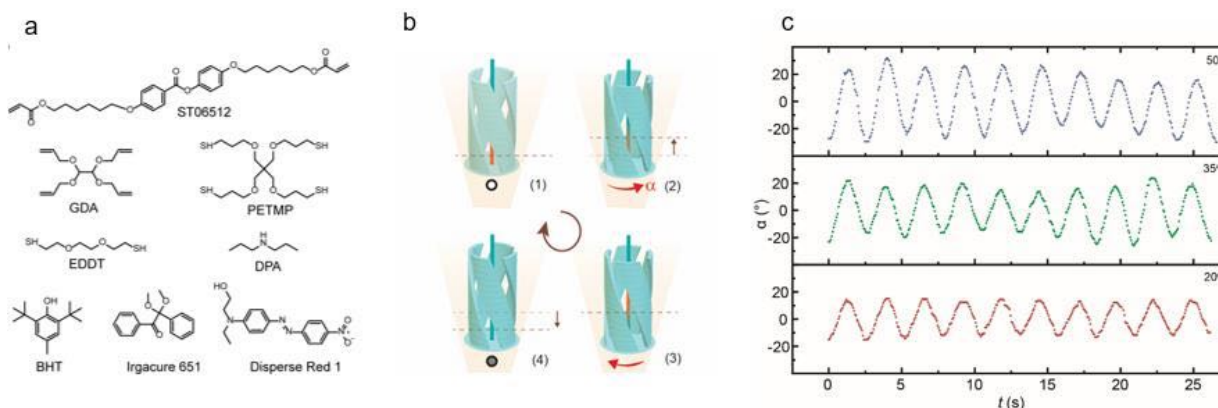


Figure 1. (a) Compounds used for fabricating the light-responsive LCE. (b) The self-shadowing mechanism for the self-oscillation through lampshade rotation. (c) Oscillation at different zenith angles.

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Poster #21

Finite Element Approximation of a Membrane Model of Liquid Crystal Polymer Networks

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Starting from the classical 3D trace formula energy of liquid crystal polymer networks of Bladon, Warner and Terentjev [1], we derive a 2D membrane energy as the formal asymptotic limit of the 3D energy and characterize the zero energy deformations in the spirit of [2]. The membrane energy lacks convexity properties, which lead to challenges for the design of a sound numerical method. We discretize the problem with conforming piecewise linear finite elements and add a higher order bending energy regularization to address the lack of convexity. We prove in [3] that minimizers of the discrete energy converge to zero energy states of the membrane energy in the spirit of Gamma convergence; this includes the presence of creases. We solve the discrete minimization problem via an energy stable gradient flow scheme. We present computations showing the geometric effects that arise from liquid crystal defects as well as computations of nonisometric origami, including origami with incompatible stretching across creases that goes beyond theory [4].

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Poster #22

Liquid crystal, carbon nanotube and polymer mixed-phased-composites for accumulation-responsive soft materials

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Mixtures of liquid crystals (LCs) and carbon nanotubes (CNTs) have been shown to act as switches in response to an electric field [1]. These composite systems can display resistive memory, mimicking neuronal behavior such as synaptic plasticity and providing opportunities to develop new classes of neuromorphic materials [2]. Typically, miniscule amounts of nanotubes are mixed into an alcohol solvent, diluted, tediously dispersed into liquid crystal solutions, and evaluated using small gap (< 5 um) electrodes. Our efforts to reproduce results with these mixtures has been less than successful. Here, we tune out the challenges of CNT solubility in LCs and investigate LC mixtures that are saturated with CNTs. These “two-phase” systems display reproducible hallmarks of neuromorphic materials such as memory, reset and accumulation in response to pulsed electric fields. We show electric-field induced CNT solubilization in LC “solvents” and parallel field alignment are the dominant mechanisms governing response. These effects are not restricted to micron gapped electrodes but are apparent across mm distances and translated into polymer dispersed liquid crystals, useful for device level applications. Incorporation of this mechanism into liquid crystal elastomers provides opportunities for adaptive behavior-based soft robotics. Overall, this work provides a foundation to develop tunable, neuromorphic materials that can remember and respond to electrical cues.

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Photochemically-driven 4D printed LCE swimmer

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Biomimicry of movements, such as swimming or crawling, of a variety of organisms have been extensively studied, both mathematically [1] and experimentally [2] by researchers from all over the world, with a focus on soft-robotics application in the last years. Among the different locomotion gaits, swimming is certainly one of the most interesting and it is quite compatible with soft or shape changing materials applications. In the shape changing materials category, hydrogels and liquid crystal elastomers (LCE) have been recently used to recreate swimming motion, but in both cases is difficult to create fast actuating, free swimming systems. The hydrogels suffer a relatively slow mass transfer and diffusion process, while the LCEs can be divided in two categories: dye containing materials that can react fast and nicely simulate swimming systems, but typically need a strong focused light source and, successively, a tracking system [4]; and azobenzene containing materials that have relatively slow actuation times, considering the photochemical effect in water, not compatible with swimming systems [5].

Here we present a novel swimming system inspired by the motion of ephyrae, that can overcome the previously cited issues and swim under a moderate light power (40-100mW/cm²). The new developed system, incorporating azobenzene molecules, has been prepared by direct ink writing. The system can fast react (less than 1 second) to UV light under water, thanks to the softness of the matrix, and regain its original shape by using visible light. The photoinduced deformation for swimming is majorly ascribed to the photochemical effect.

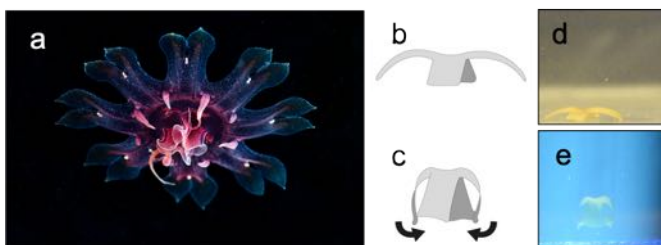


Figure 1: a) Photo of an ephyra organism (Photo by Alexander Semenov, Reproduced with permission under a Creative Commons CC BY-NC-SA 2.0 License); actuation principle of the system (b-c) and actual images of the swimming system before (d) and after UV (e) irradiation

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Acknowledgments

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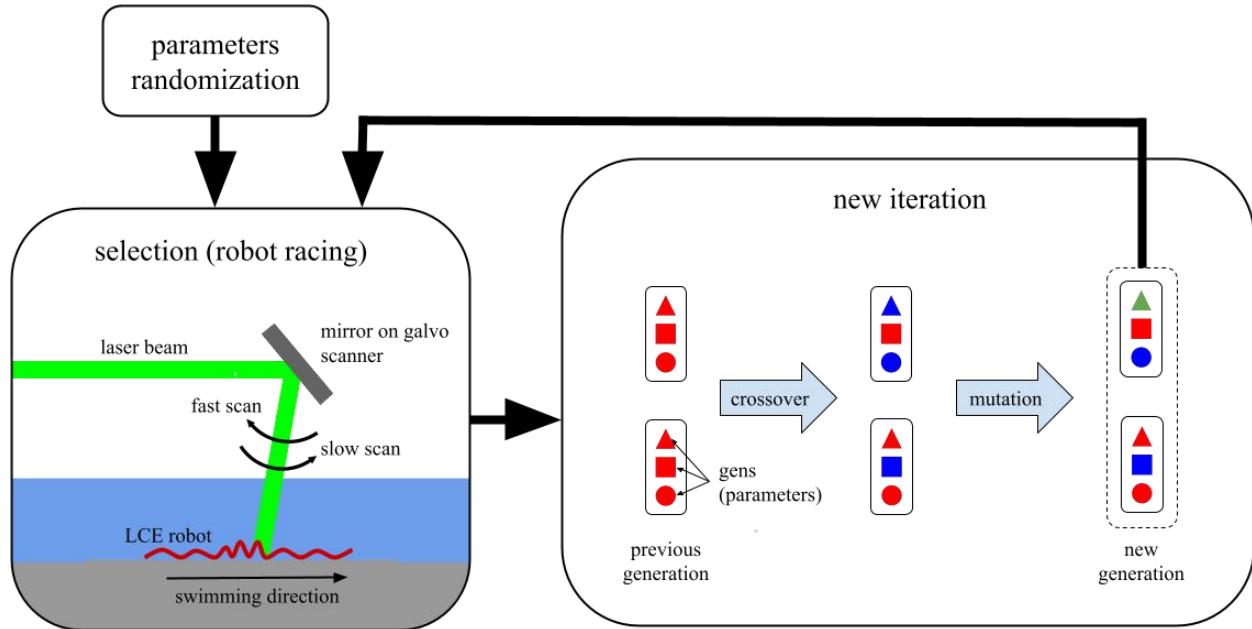
Optimizing underwater locomotion of light-driven soft robots

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The theory of evolution has led to a paradigm shift in our understanding of life, driving advancements in optimization methodologies across various fields. This research focuses on applying genetic algorithms (GA) to optimise the speed of centimeter-long, light-driven swimming robots designed for underwater locomotion. The design is inspired by our previous research [1], robots are made of liquid crystal elastomers (LCEs) and are propelled and controlled by lasers. The main objective is to determine the optimal combination of parameters, including length, LCEs' molecular orientation, thickness, and laser beam properties, that can maximize the robots' speed. Through the use of genetic algorithms, this research aims to unlock the potential for achieving heightened performance and efficiency in light-driven swimming robots, paving the way for improved underwater locomotion capabilities.



We apply a genetic algorithm, treating the robot's parameters as genes within the algorithm. The initial generation of robots is randomly generated, and subsequent generations are created by measuring the speed of each robot and selecting the fastest ones for crossover. Additionally, a mutation operation introduces the possibility of changing one of the robot's parameters. This iterative process allows for the evolution of faster swimming robots over successive generations. Through the use of genetic algorithms, we aim to optimize the swimming robots' speed by mimicking the principles of natural selection. The results of the optimization process provide insights into the most effective parameter combinations for achieving maximum speed.

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Poster #25

Liquid Crystal Elastomer Based Dynamic Device for Urethral Support: Potential Treatment for Stress Urinary Incontinence

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Stress Urinary Incontinence (SUI) is the involuntary leakage of urine affecting nearly 50% of adult women. Severe SUI is treated with the insertion of synthetic or autologous sling underneath the urethra to provide needed support. A limitation with current midurethral slings is their fixed level of support, which doesn't allow urethral support adjustment for voiding. Here, we report the 3D printing fabrication of a novel device based on liquid crystal elastomers (LCEs) capable of changing shape in response to temperature increase induced by transcutaneous IR light. A scar-tissue phantom model was used to characterize shape change of the LCE-based device in a constrained environment. An *in vitro* urinary tract model was designed to study the efficacy of the LCE-based device to support continence and adjust sling support with IR illumination. The LCE device achieved $5.6\% \pm 1.1\%$ actuation when embedded in agar gel. The corresponding device temperature was $44.9^\circ\text{C} \pm 0.4^\circ\text{C}$, and the surrounding agar temperature stayed at $42.1^\circ\text{C} \pm 0.4^\circ\text{C}$. Time to leak 20mL volume in the *in vitro* urinary tract model significantly decreased ($p < 0.000$) from $5.2\text{min} \pm 1\text{min}$ to $2\text{min} \pm 0.5\text{min}$ when an LCE cuff was sutured around the urethra and when it was illuminated with IR light, respectively. Finally, the device was acutely implanted and tested for induced changes in female multiparous rabbits. Abdominal force required to cause leakage significantly ($p = 0.01$) increased with LCE-CB cuff implantation around the rabbit urethra. The force decreased significantly ($p = 0.023$) when the device was illuminated with IR light through the tissue.

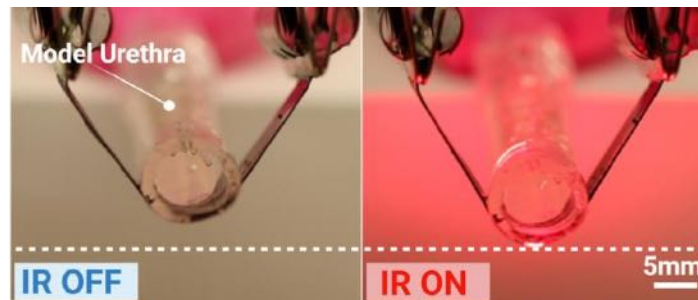


Figure 1. Model Urethral is supported with an LCE based device (left). The LCE device is illuminated with IR light allowing it to change shape, reduce urethral support, and allow facial voiding (right).

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Poster #26

Bioinspired Fast and Slow Twitch Muscles via Liquid Crystal Elastomer Twisted Fibers and Bundles

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The human body has fast and slow twitch muscles acting in unison to perform complex tasks. Here, we use twisted liquid crystal elastomer fibers to mimic fast and slow twitch behavior in muscles. Rectilinear liquid crystal elastomer fibers contract upon heating. This research is concerned with twisted LCE fibers and bundles. The stimuli-response of these systems spans a distinct geometric phase space. The fibers start in a rectilinear geometry and then spontaneously coil when heated. The stimuli-response of LCEs changes the twist density of the LCE fibers. The response of the fibers to heat is strongly dependent on the fibers' morphological constraints (e.g., the twist density). A material with a fixed twist density can have different geometric actuation responses based on the load or force that needs to be generated. Using spontaneous coiling for actuation leads to fast actuation responses, an increase in actuation stroke, and the ability to optimize work. Combining fast- and slow-twitch fibers in a device can add asymmetry and facilitate more complex designs.

Poster #27

Amplifying helical liquid crystal assembly to macroscale actuation

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Strategies for translating molecular structures to the macroscale for controllable motion are key for applications in soft robotics, complex actuation, and studying nature-mimetic systems. Here, we demonstrate a liquid crystal elastomer system that shows reversible photothermal-triggered helical motion, induced by cholesteric self-assembly of a chiral dopant within a liquid crystal matrix. The handedness of the actuated helix can be tuned by varying the enantiomeric excess of the dopant and the composition of the film. Helical actuation is shown to be attainable even without the use of any surface alignment techniques and combinations (which are often key for directing liquid crystal anisotropic motion in a helical conformation), enabling surface alignment variations to be used as an additional but not necessary lever for modulating helical actuation geometry. Self-assembling dopants are shown to be able to direct liquid crystal ordering, introducing an approach for amplifying self-assembled helices to the macroscale.

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Dandelion-Inspired, Wind-Dispersed Liquid Crystal Elastomer Actuator Controlled by Light

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The emergence of stimuli-responsive polymers has engendered a profusion of materials tailored to the realm of small-scale, wirelessly controlled soft-bodied robots. However, transcending the confines of conventional robotic movements exhibited by synthetic systems, such as walking, swimming, and jumping, achieving flight in the air through dispersal, gliding, or even hovering remains an uncharted domain for responsive materials. Such unexplored frontiers confront the materials domain with formidable challenges, epitomized by the exacting requirements concerning actuator performance, lightweight construction, and effective aerodynamic design. In this context, the present study unveils a liquid crystal elastomer-based porous structure, endowed with the remarkable ability to disperse and execute lift-off and landing maneuvers with the assistance of wind. Inspired by the dandelion seed, this design manifests various biomimetic characteristics, including high porosity, lightweight composition, and the generation of separated vortex rings when subjected to a steady wind flow. Surpassing its natural counterparts, this artificial seed features a pliable actuator fabricated from a light-responsive liquid crystalline elastomer, inducing reversible opening and closing actions of its bristles upon exposure to visible light. This shape-morphing capability empowers the manual calibration of critical parameters such as terminal velocity, drag coefficient, and wind threshold to optimize dispersal. The study showcases the successful realization of optically controlled wind-assisted lift-off and landing maneuvers, alongside light-induced local accumulation. These findings present pioneering avenues for the development of miniature wirelessly controlled devices, enabling passive navigation across vast aerial expanses.

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Mechano-optical response behavior of cholesteric elastomers in various deformation modes

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Cholesteric liquid crystal elastomers (CLCE) are unique materials that combine the rubber elasticity with the optical properties of cholesteric liquid crystals (CLC). A marked feature of CLCE is that the characteristic wavelength of selective reflection of CLC is reversibly varied in response to the application and release of deformation.¹⁻⁵⁾ This mechano-optical response behavior has been primarily investigated by uniaxial extension^{1,2)}, and equibiaxial extension (equivalent to uniaxial compression)³⁻⁵⁾ normal to the helical axis.

Exploring the mechano-optical response under various deformation modes is vital to understand comprehensively the physical properties of CLCE, and to consider their diverse deformation patterns during practical applications. In this study, we have employed four deformation types, i.e., uniaxial extension, compression, planar extension, and swelling via solvent, to characterize the optical response behavior of a CLCE film with the helical axis oriented in the thickness direction. In planar extension which is intermediate between uniaxial and equibiaxial extension, the sample is stretched in one direction while maintaining the dimension in the other direction.

In uniaxial extension and compression, and planar extension, the resultant film-thickness reduction causes a shift of selective reflection band toward shorter wavelength. Notably, the shift amount depends on the deformation mode when compared at the same film-thickness reduction. In addition, the selective reflection property is preserved throughout compression, while it vanishes in both uniaxial and planar extension. The vanishing of the selective reflection property in planar extension occurs at a higher stretch compared to that in uniaxial extension. Swelling, which increases each dimension but anisotropically, drives a shift of selective reflection band toward longer wavelengths. However, the shift amount is less than expected by affine displacement.

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Poster #30

Molecular engineering of polymer stabilizing network to enhance the electro-optic response of polymer stabilized cholesteric liquid crystals

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The cholesteric liquid crystalline (CLC) phase self-organizes into a periodic structure and in the planar orientation, exhibits a selective reflection. These materials are widely considered for applications in optics and photonics, including emerging applications in immersive displays [1]. Polymerization of a small concentration of monomer can stabilize the CLC phase (polymer stabilized CLC, or PSCLC). We have recently been exploring an ion-mediated mechanism to displace the polymer stabilizing network. It is well known that liquid crystal mixtures retain ions such as Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , Zn^{2+} , and Al^{3+} . These ions can negatively impact the performance of liquid crystal devices (response time, image sticking, color staining, voltage holding ratio) [2]. However, these impurities can be leveraged in this mechanism. Accordingly, this presentation reports that preparation of polymer stabilizing networks with rational incorporation of ion-trapping groups, such as 4-acryloxyterpyridine (TPy-Ac), 4-((6-(Acryloyl)oxy)benzoic-15-crown-5-ether (OBA-15C5), and 4-((6-(Acryloyl)oxy)benzoic-18-crown-6-ether (OBA-18C6) (Fig.1). The polar nature of these comonomers can capture transition and alkali metal ions. This presentation will detail the ion-material coupling and the resulting impact on electro-optic performance.

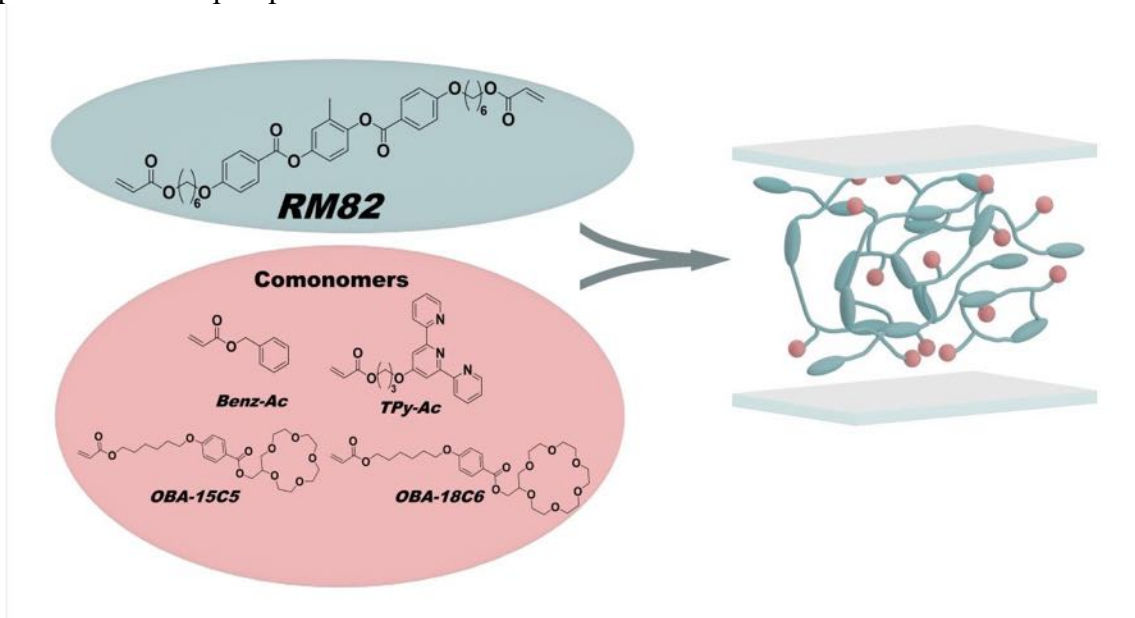


Figure 1. An illustration of how the difunctional liquid crystalline monomer RM82 (blue) and the monofunctional ion-trapping comonomers (red) for a loosely crosslinked polymer network within the sample

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Shape memory perspective of liquid crystalline elastomers

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Unable to attend

Shape memory polymers (SMP) and liquid crystalline elastomers (LCE) are two parallel classes of stimuli-responsive shape morphing materials. The former emphasizes the freedom to repeatedly program the shape morphing pathways in a more or less arbitrary manner whereas the latter focuses mainly on the use of a liquid crystalline transition for reversible shape morphing. Despite the differences, there seems to be similar principles to borrow from each field to gain synergy. This talk will focus on the use of principles well known in the SMP to broaden the design versatility of LCE, including rapid motion reprogrammability and ultrafast digital productions of LCE actuators.

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Porous Liquid Crystalline Networks as cell instructive scaffolds

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Control of the collective cells organization and their correct differentiation are key steps to reproduce biological tissues in vitro. So far, many attempts to prepare polymeric scaffolds that mimic the biological environment have been done, trying to reproduce the mechanical properties or the extracellular matrix composition and topography. To date, cell differentiation protocols required scaffolds bearing micro/nanometric features or stimulated ones (by mechanical stretching or electric field). However, the complexity of these methodology strongly limits their wide application.

A completely different approach, described in this communication, for the alignment and differentiation of several cell lines, exploits Liquid Crystalline Networks. Materials with anisotropic molecular structure and tunable rigidity have been prepared by photopolymerization of acrylate-based LCs and tested as support for cell growth (from human fibroblasts, C2C12 myoblasts and cardiomyocytes). We demonstrated LC polymers to present several advantages with respect to commercial scaffolds such as an improved adult-like dimensions and a more mature cell function of human induced pluripotent stem cell-derived cardiomyocytes (hiPSC-CMs) in a shorter time. [1] Furthermore, LC organization was demonstrated able to drive specific cells arrangements during fibroblast and myoblast cultures, as shown in Figure 1. Cell alignment was found to follow the nematic director thus demonstrating as the LC order inside the material can be translated to a living organism. [2,3] Eventually, myotube maturation has been investigated with electrophysiological studies showing as muscular fibres cultured on our LCs present better functional features with respect of those obtained on standard supports. [4]

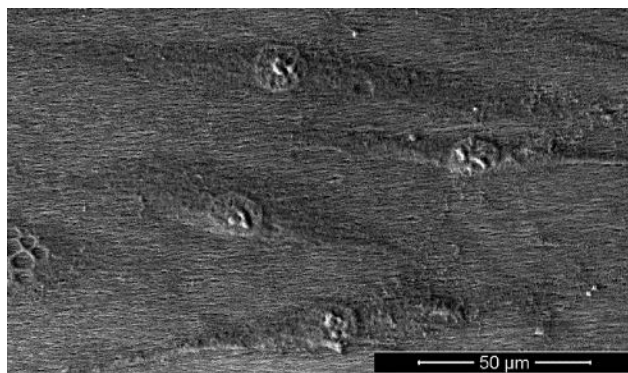


Figure 1. SEM image of aligned fibroblasts on a LC scaffold

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Towards Differentiation in Untethered Liquid Crystal Elastomer Microactuators

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Soft actuators have received tremendous attention due to their potentials to revolutionize many applications ranging from artificial muscles to biomedical applications. The creation of biomimetic actuators at microscopic length-scale with tailor-made functionality is broadly sought after, but challenging to achieve. Multipotent cells have the ability to differentiate into cell types with various shapes, surface patterns, and functions within one particular lineage. Particularly, simple force fields are used in either a discrete or a synergic way to induce the event of cell differentiation. Inspired by cell's ability to differentiate, we developed a modular soft manufacturing based on droplet microfluidics, using different force fields to orthogonally program the shape, surface pattern, and microactuation of an untethered liquid crystal elastomer (LCE) microactuator towards bioinspired soft microrobotics. Hydrogel microfibers containing arrays of prepolymer microparticles were fabricated via single emulsion droplet microfluidics. By fine-tuning fiber dehydration in gravitational field, various particle geometries including sphere, spindle, rod, dumbbell, wormlike, can be customized in a highly tunable manner. Moreover, a novel dehydration phenomena of the hydrogel fiber- at room temperature, using only water, taking few minutes - was investigated to induce stress-driven buckling, resulting in various patterns on hydrogel. After polymerization in a magnetic field to encode the mesogenic orientation, the resulting LCE microactuators with different shapes, mesogenic information, and surface patterns displayed distinct deformations upon heating above T_{n-i} , which were validated with FEA simulations. We foresee that these responsive LCE microactuators can be widely used in a myriad of applications e.g. micromotors, microswimmers, dynamic assembly etc.

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Thermomechanical Coupling in Monodomain and Polydomain Liquid Crystal Elastomers

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Liquid crystal elastomers (LCEs) are rubbery polymer networks with liquid crystal mesogens embedded into their polymer chains. These materials can generate large deformation and mechanical work upon temperature change or light illumination, making them ideal candidates for new applications including soft robots, metamaterials, and other stimuli-responsive devices. However, despite the rapid development in the chemistry, manufacturing and applications of LCEs, the fundamental thermomechanical coupling between the microscopic temperature-controlled processes and the macroscopic mechanical behavior of these materials remains poorly understood when it comes to combining theories and experiments. It remains further unclear how the modern fabrication methods such as the widely used “two-step polymerization” affects this thermomechanical coupling. This talk will present our recent progress in understanding the thermomechanical behavior of nematic LCEs under both mechanical loads and prescribed temperature. Through experiments with quantitatively controlled fabrication parameters, we show a nontrivial effect of the applied pre-stretch during polymerization on the thermomechanical behavior of nematic monodomain and polydomain LCEs. To understand the experimental observations, we establish a classical free energy-based continuum theory that incorporates the entropic elasticity of the polymer network and the long-range directional molecular interaction between mesogens. These efforts are aimed to advance the fundamental knowledge of liquid crystal elastomers, bring together communities of relevant research fields, and further expand the large-scale applications of these materials.

Anisotropic stress concentration behavior of liquid crystal elastomers

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With increasing demand of application, the problem of stress concentration in liquid crystal elastomers (LCEs) cannot be neglected. Nematic liquid crystal elastomers (NLCEs) have unique anisotropic soft elastic behavior due to effects of liquid crystal mesogens. Based on Ericksen-Leslie theory, an anisotropic constitutive framework is developed to describe the transversely isotropic soft elastic behavior of uniaxial NLCEs. Then we studied stress concentration problems of NLCEs thin films with defects like circular and elliptical holes through analytical and finite element methods. Our results indicate that the stress concentration of NLCEs is more significant compared with ordinary elastomers when the stretch direction is parallel to director. These unique behaviors are mainly caused by elastic anisotropy and non-uniform directional rotation at hole edge. Besides, the material characteristic direction has a strong impact on stress concentration behaviors: when the stretch direction is not along the director, the stress and strain concentration factor get smaller. This inspires us that by designing the distribution of directional vectors at hole edge to decrease stress concentration. This research might provide solutions for material designing which will help to decrease the stress concentration of NLCEs.

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Additive manufacturing of liquid crystal elastomer actuators based on knitting technology

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Liquid crystal elastomer (LCE) exhibits large and reversible deformability originating from the alignment of liquid crystal mesogens. Additive manufacturing provides a high degree of control over the geometry and deformability of LCE actuators by locally controlling the alignment of liquid crystal mesogens during the shaping process. However, currently applied additive manufacturing methods usually introduce chemical cross-linking to fix the structure and alignment of the LCE actuator, making it difficult to change the deformation and structure after shaping and impossible to recycle. It still remains a challenge to customize LCE actuators with both diverse three-dimensional deformability and recyclability. In this work, a new strategy is developed to exploit knitting technique to additively manufacture LCE actuators through the entanglement of LCE fibers. The obtained LCE actuators are fabric-structured with designed geometry and deformability. By accurately adjusting the parameters of the knitting patterns as modules, diverse geometry is pixel-wise designed, and complex three-dimensional deformations including bending, twisting and folding are quantitatively controlled. In addition, the fabric-structured LCE actuators can be threaded, stitched, and reknitted to achieve advanced geometry, integrated multi-functions and efficient recyclability. This approach allows the fabrication of versatile LCE actuators with potential applications in smart textiles and soft robots.

Liquid Crystal Elastomer Tubes as Artificial Vasculature to Support Organoid Growth

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Organoids—3D lab-grown tissue that can mimic organs *in vivo*—have attained great popularity as they can be used to study the diverse and complex biology of organ development *in vitro*. These organoids lack an internal vasculature and thus do not completely grow/exhibit the rich multicellular architecture of the organs they mimic. The organoids therefore currently rely on diffusion for transport of nutrients and waste products^[1], thus impeding cell survival at the organoid core (necrosis). As a potential means of overcoming this limitation, we explore the use of liquid crystal elastomer (LCE) tubes as active artificial vasculature in organoids. LCEs can exhibit dramatic shape changes in response to temperature changes or light irradiation, related to the loss/recovery of long-range orientational chain order in the LCE network. Recently, we reported the production of LCE tubes of arbitrary length using a continuous three-phase coaxial flow microfluidic process^[2]. We attain the tubes by flowing an oligomeric LCE precursor liquid between two immiscible aqueous phases, along with surfactants in each phase to reduce the interfacial tension to the point where the tubular flow is stable over tens of centimetres. We then crosslink the flowing oligomer into an LCE by *in-situ* UV irradiation. We also demonstrated reversible actuation with consequent reduction of the interior space of the tubes upon heating. We are now targeting on the light-driven sequential localized actuation of the tubes along their length to use them as peristaltic pumps in organoids, and also tailoring tube porosity for efficient liquid exchange between tube interior and exterior.

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Microscopic Actuation for Macroscopic Aggregation

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An important area of materials research is the development of adaptive structures capable of sensing, reconfiguring, and self-healing with minimal intervention. Inspiration for the development of such structures is found in the survival strategies demonstrated by many insects. For example, fire ants and blackworms aggregate to mitigate environmental threats(1, 2). This is achieved through each insect's ability to attach and detach to its neighbors. In order to mimic this behavior, liquid crystal elastomer (LCE) ribbons (250 μm x 6 mm x 50 μm in cross-section) are programmed to reversibly actuate from a straight to a coiled configuration. By heating these LCE strips and allowing them to aggregate and mechanically interlock with each other, structures capable of reversibly transitioning from a liquid-like state to a solid-like state are achieved. Rheological data of these structures demonstrate an increase in yield stress from 23 Pa at 50 °C to 329 Pa at 150 °C. Along with this increase in yield stress, a reversible negative thermal expansion of 30% is observed. Notably, the mechanical properties of the aggregate are controlled by tuning individual ribbon properties. By increasing the length of the individual ribbons from 3 mm to 12 mm, yield stress increases from 200 Pa to 725 Pa, and by increasing the offset angle from 0° to 45°, a decrease in yield stress from 459 Pa to 38 Pa is observed. This synthetic aggregate's ability to reversibly transition from low-yield stress to high-yield stress on demand enables various functionalities such as clogging, targeting, and self-healing.

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Finite element model for transient response of photoactive LCE beams

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Light actuation is one of the preferred and advantageous approaches to remotely induce and control deformations in soft materials such as photoactive liquid crystal elastomers (LCEs). Several studies have been carried out in the literature that proposed mathematical and numerical models based on linear beam theory [1] and large deformation theories (without considering inertial terms) [2] to predict the equilibrium and transient response of thin LCE beams under light illumination. However, it is still necessary to develop more efficient numerical models based on large deformation theory considering inertial term to investigate the phenomena such as self-oscillation in LCE beam [3,4] subjected to constant light illumination. In current study, we have developed a coupled multi-physics model considering the inertial term to predict dynamic response of photoactive LCE beams under illumination. The developed model is implemented in COMSOL software through weak PDE interface. We first test our model against an experiment [5] in which a double-clamped thin nematic LCE beam is subjected to UV light, and the stress is generated in the beam due to induced contraction. We also investigate the self-oscillation phenomenon in LCE beam due to sequentially actuation of top and bottom surfaces and self-shadowing effect. Our results show that constant light illumination can pump energy into the system and trigger the self-oscillating phenomenon. Furthermore, we perform parametric studies to investigate the effect of beam's length and light intensity on amplitude and frequency of oscillation under illumination.

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Liquid crystal network micro-actuators using light

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Miniaturization in micro-robotics is a key issue to achieve micro-interventions in the medical field regarding hard-to-access areas. In this context, remote actuators are often used, but in the case of photo-actuators, an embedded actuation is preferred to reduce health risks and avoid light absorption. This work presents two strategies of fabrication for photo-actuated micro-actuators using liquid crystal networks [1]. First, stereolithography printing of beam actuators, then, multi-material printing of micro-actuators using direct laser writing of liquid crystal network (LCN) and a glassy polymer photoresist.

To achieve high-resolution printing of Liquid Crystal Network, we use stereolithography [3] with a stabilized liquid crystal resin using 54-Cyano-4'-pentylbipheny (5CB) to avoid crystallization of the resin during the printing process. The alignment is obtained by gluing rubbed glass slides on the building platform of the printer and the container. With this technique, we obtain actuated LCN beam structures as a first step toward more complex bio-inspired structures.

Multi-material Direct Laser Writing has been explored in the last few years [2]. Here we apply this technique to Liquid Crystal Network. First, LCN muscle-like structures are printed inside alignment cells [3]. The design has anchors to have good adhesion to the bases that are printed afterward. After the development of the LCN structures, non-photo-active bases are printed with the photoresist. The bases are used as a support to be able to integrate this actuator as a building block. The photo-induced contraction is measured regardless of the base anchoring, proving the ability of the actuator to perform simple actions.

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Coupled Photo-Mechanical Simulation of Nematic Liquid Crystal Elastomers with Patterned/Varying Alignments

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Light-responsive liquid crystal elastomers (LCEs) have gained significant attention in the recent times as stimuli-responsive actuators. The light-responsive elastomers are formed by doping the elastomeric liquid crystalline network with the azo-benzene based mesogens. The combined effect of elastomeric network's weak cross-linking and the mesogen's order-disorder transition results in certain exotic behaviors as mentioned below.

1. The stretching of poly-domain LCEs results in the uniaxial alignment of the mesogens.
2. The rise in temperature leads to nematic-isotropic transitions generating large strains.
3. The weak cross-linking of the elastomers coupled with *trans*-to-*cis* isomerization of the azo-benzene can lead to photo-activated alignment changes of LCE upon illuminating with polarized light sources and large strains (photo-mechanical coupling).

In this study, we have developed a user element subroutine (UEL) using the commercial finite element software, Abaqus to simulate the photo-mechanical coupling of these nematic elastomers subjected to mechanical stress and illumination. To model the photomechanical coupling using the finite element method, the minimization of the free energy approach is implemented considering Corbett and Warner's formulation [1] for polymeric network elastic behavior and Maier-Saupe theory-based formulation [2] for nematic liquid crystal behavior. A system of linear equations with displacements, order-parameter and the alignment direction as primary variables is developed by linearizing the non-linear system. The developed model is benchmarked by comparing the simulation of the exotic behaviors of the 2D LCE sheet with the work of Bai and Bhattacharya [2]. The developed framework is further extended to simulate the coupled photomechanical response of LCEs with patterned/varying alignment distributions, made possible by the developed finite element framework.

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