Hierarchically ordered complex soft materials: where additive manufacturing meets self-assembly

Integration Grant 24IRF-1C003

Tatiana (Tetiana) Orlova

May 16, 2026

ՀՀ ԿԳՄՍՆ
Բարձրագույն կրթության և գիտության կոմիտե

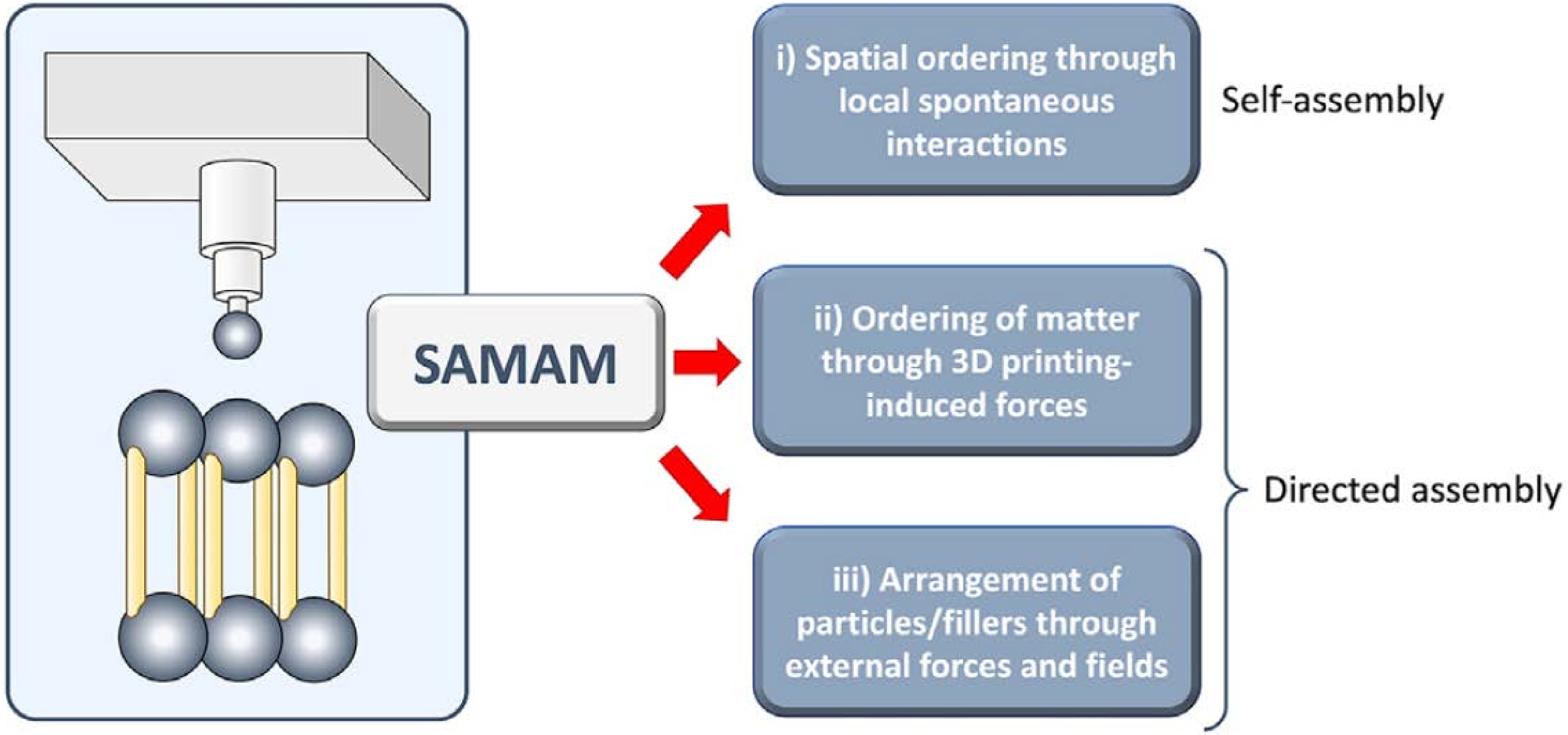
YEREVAN
STATE
UNIVERSITY

The two-part discussion

- Project idea, main goal and research objectives
- Project development and evolution

Project inspiration: SAMAM and DAMAM

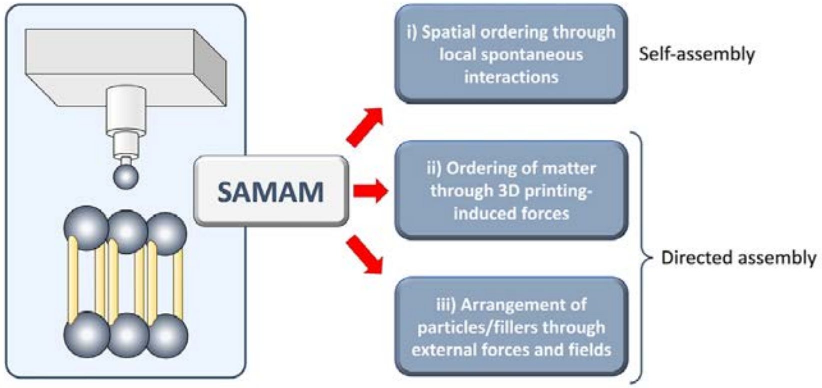
Different approaches to combining additive manufacturing
and self-assembly or directed assembly of matter



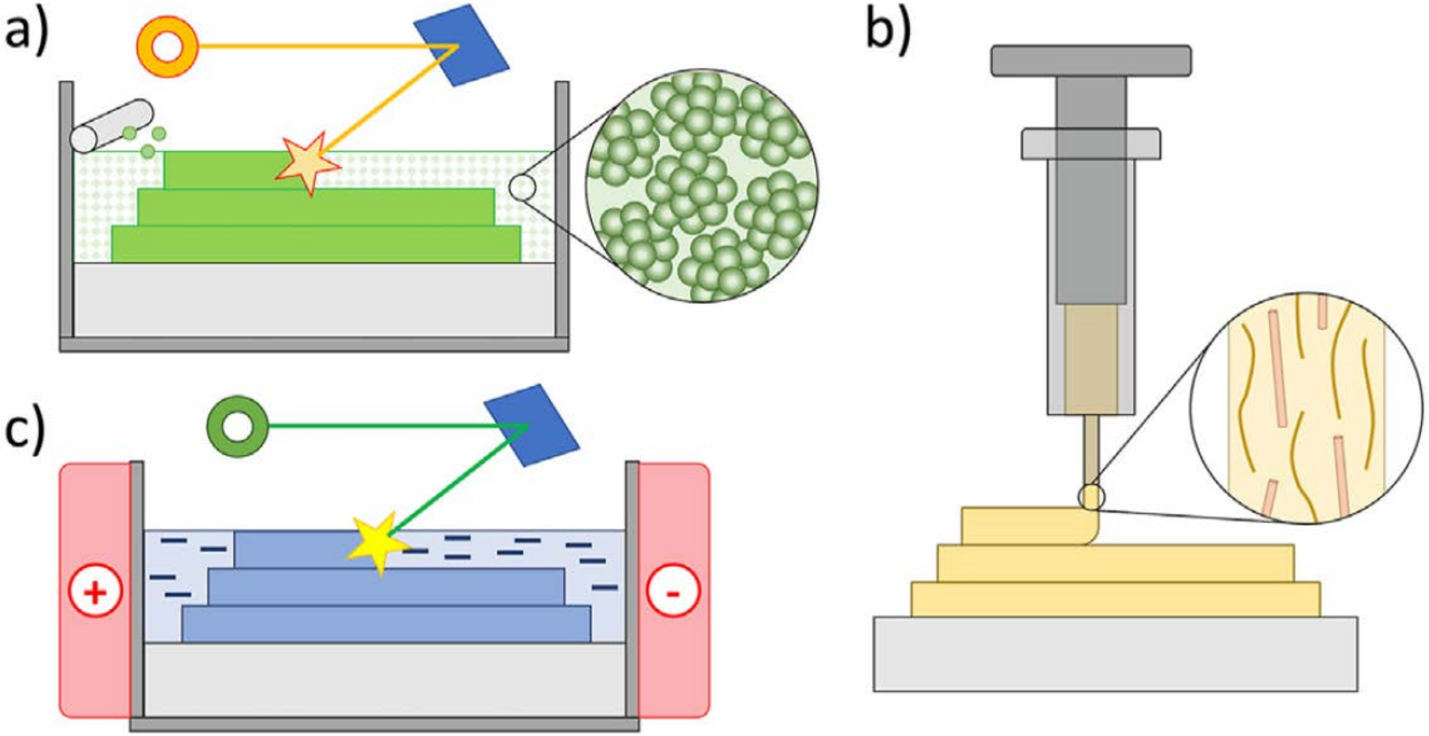
Sola et al. Smart Materials in Manufacturing (2023) 1:100013

Project inspiration: SAMAM and DAMAM

Different approaches to combining additive manufacturing and self-assembly or directed assembly of matter



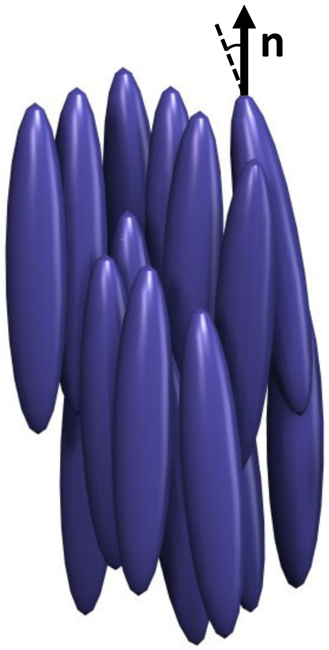
Several experimental approaches as examples



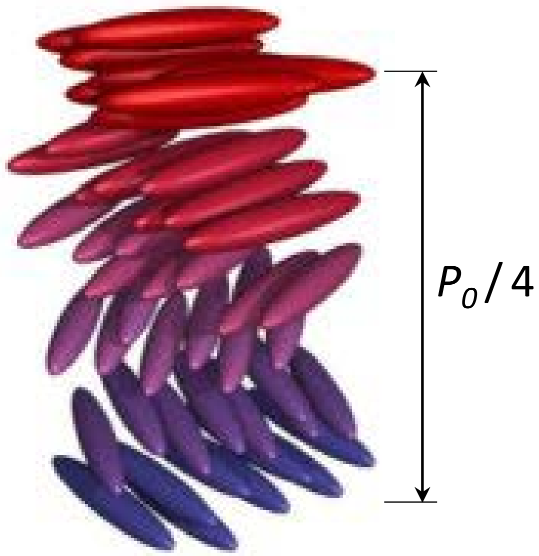
a) local interactions in selective laser sintering, b) 3D printing-enabled SAMAM in materials extrusion, c) field-assisted SAMAM in vat photopolymerization.

Sola et al. Smart Materials in Manufacturing (2023) 1:100013

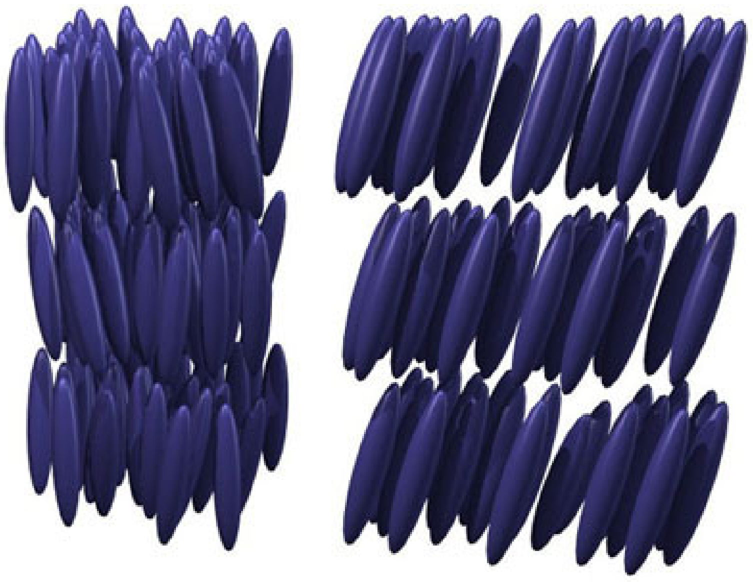
What are liquid crystals?



Nematic N



Chiral nematic N*
(or cholesteric Ch)

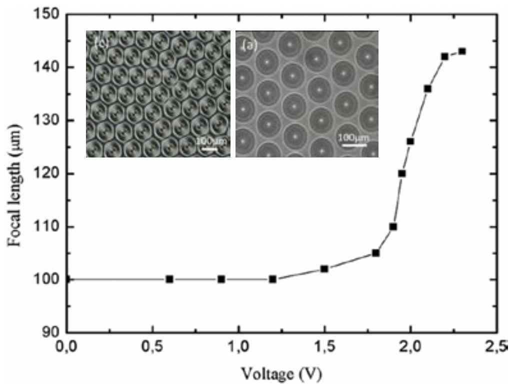


Smectic A and Smectic C

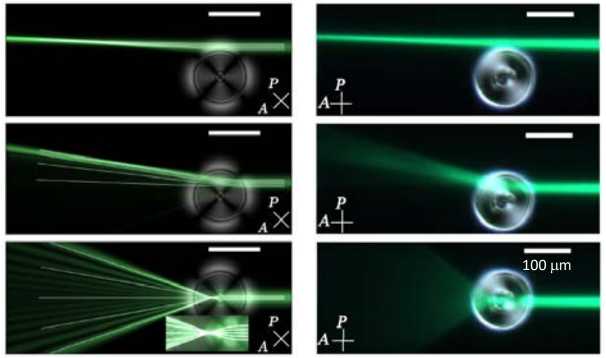
Chiral smectic C*

Self-assembled, particle-like chiral liquid crystal structures for optics, photonics, nano- and micromechanics

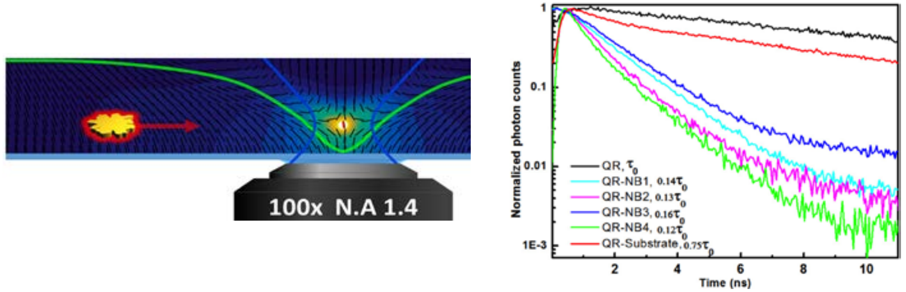
Tunable microlens array with electric field-controlled focal length



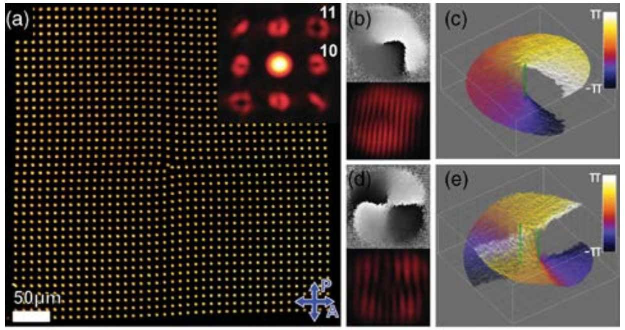
Beam deflection and lensing



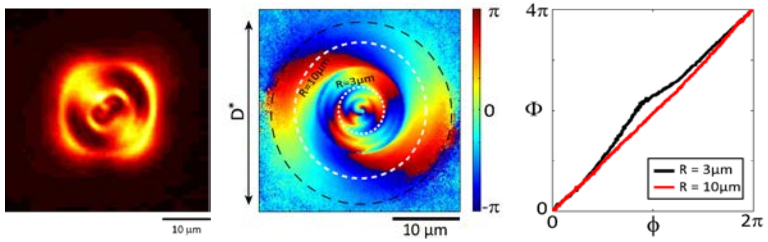
Photoluminescence decay for QRs in topological point singularities co-entrapped with the irregularly shaped disk-like gold nanoparticle



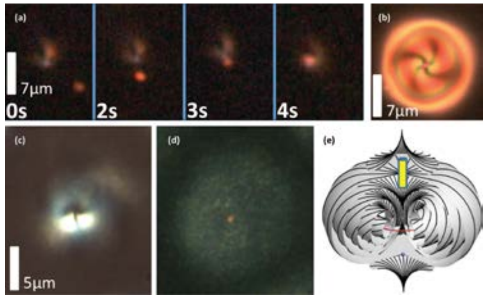
Control of phase singularities by defects in toron gratings



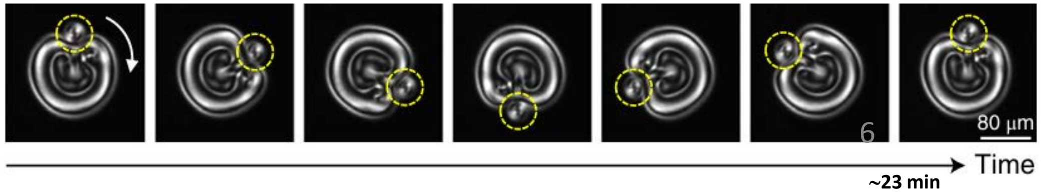
Optical vortex generator



Spatial localization of gold nanoparticles in hyperbolic point defects

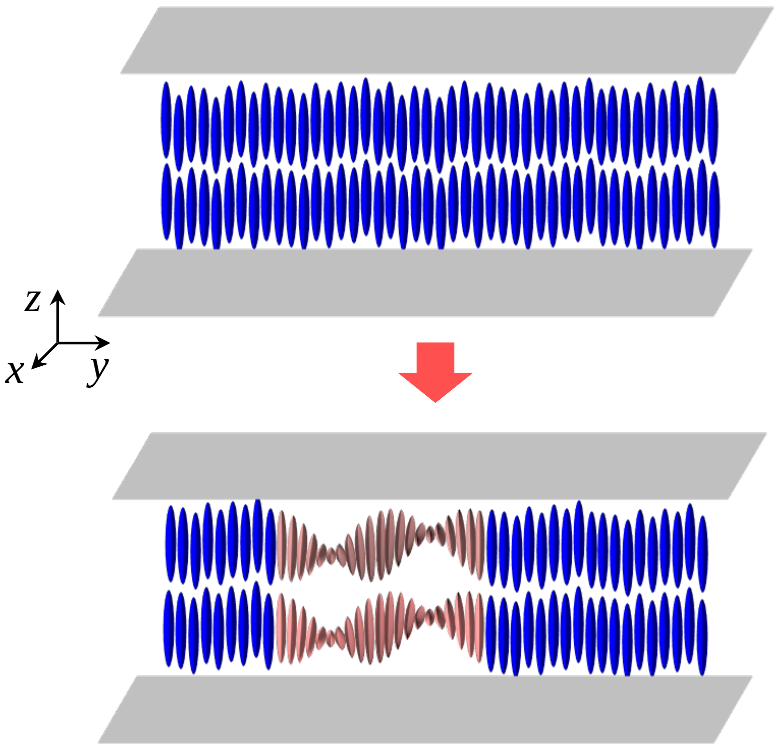


Off-axis rotational transport of a micro-sized cargo

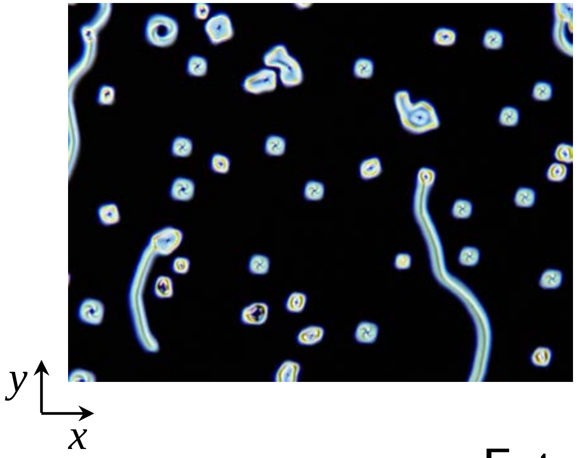


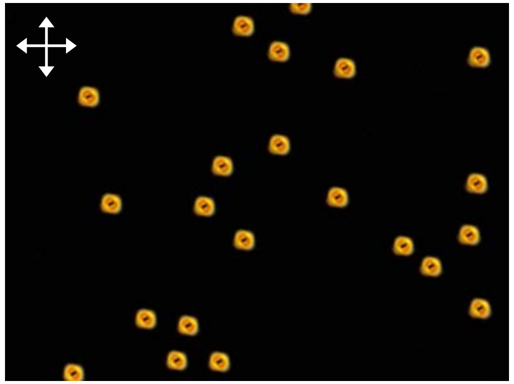
Hamdi et al. J. Appl. Phys. (2011) 110:074902
Hess et al. Phys. Rev. X (2020) 10:031042
Ackerman et al. Phys. Rev. E (2012) 86:021703
Yang et al. J. Opt. (2013) 15:044021
Smalyukh et al. Phys Rev E (2013) 87:032503
Ackerman et al. ACS Nano (2015) 9:12392
Orlova et al. Nature Nanotech (2018) 13:304

Local and nonlocal metastable release of Ch-LC frustration

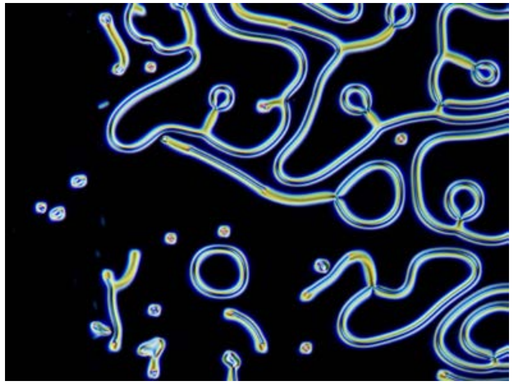


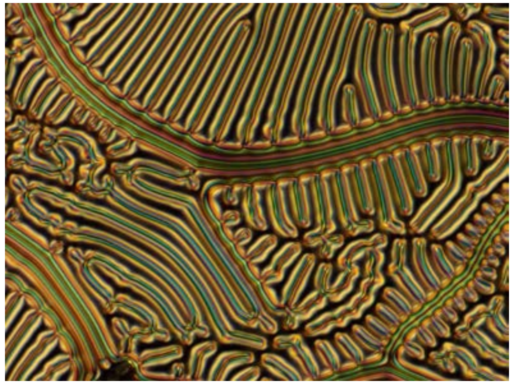
Particle-like elastic excitations





Extended orientation structures





The free energy of the LC director field deformation:

$$F_{def} = \int d^3\boldsymbol{r}\left\{\frac{1}{2}K_{11}(\nabla\cdot\mathbf{n})^2 + \frac{1}{2}K_{22}\left[\mathbf{n}\cdot(\nabla\times\mathbf{n}) + \frac{2\pi}{P_0}\right]^2 + \frac{1}{2}K_3[\mathbf{n}\times(\nabla\times\mathbf{n})]^2\right\} - \int d^2\boldsymbol{r}\,\frac{1}{2}W(\mathbf{n}\cdot\mathbf{n}_0)^2$$

Key parameters controlling topology and geometry of LC solitons

- Liquid crystal chirality
- Liquid crystal elasticity
- Confinement geometry
- LC-substrate interactions
- External field effects
- Nano- and micro-sized particles

Key parameters controlling topology and geometry of LC solitons

- Liquid crystal chirality
- Liquid crystal elasticity
- Confinement geometry
- LC-substrate interactions
- External field effects
- Nano- and micro-sized particles

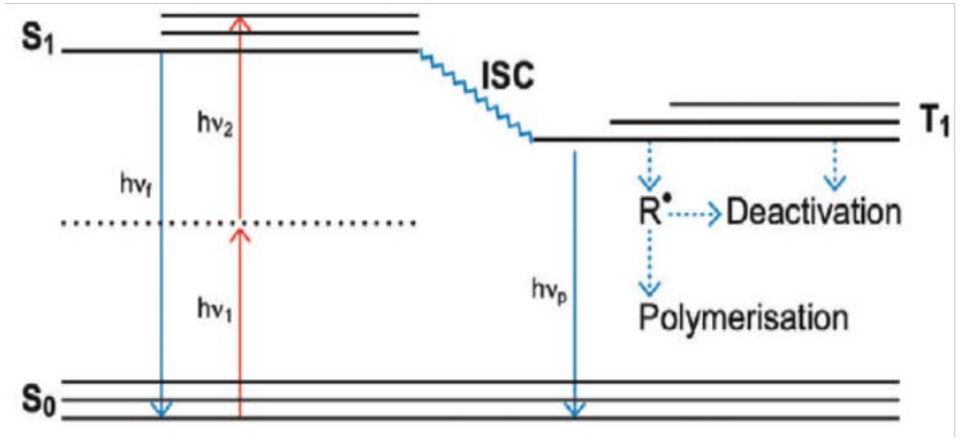
Questions and Perspectives

What if the LC confining substrates acquire a three-dimensional spatial profile?
How does it affect the self-assembly of LC orientation structures?
What if various particles are added into the confining layers?
What if the non-trivial topology of orientation LC structures is stabilized?
What new physical properties and industrial applications can be demonstrated?

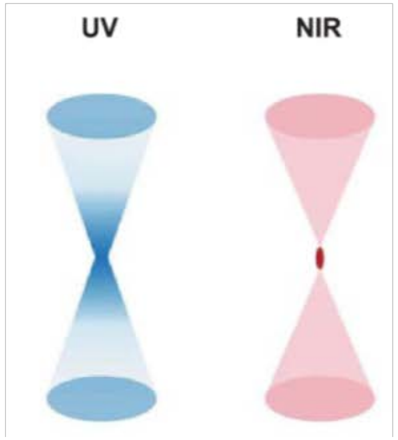
Two-photon polymerization: A high-precision additive manufacturing technology

Working principle of TPP

Two-photon polymerization excited states



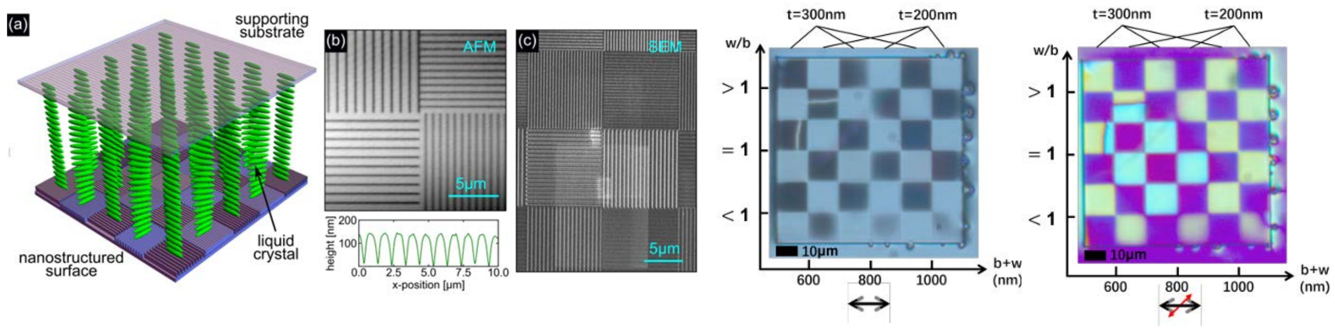
Excitation volume by OPA versus two-photon absorption



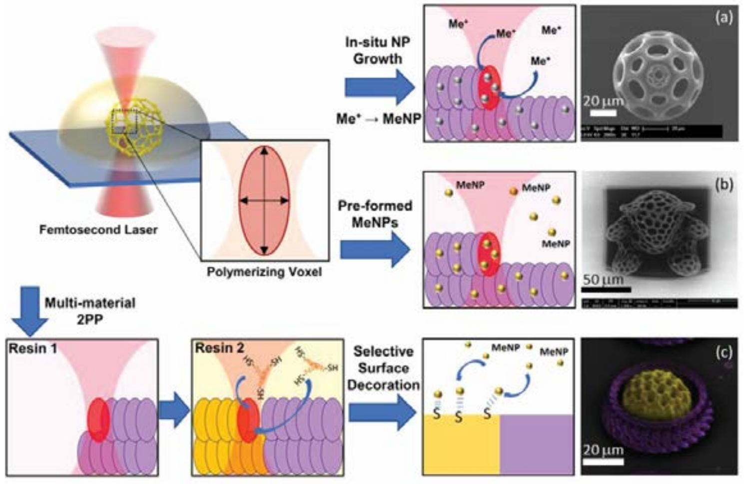
O'Halloran et al., Advanced Science (2023) 10(7):2204072
Jian et al, Int. J. Extrem. Manuf. (2024) 6:012001

Two-photon polymerization: A high-precision additive manufacturing technology

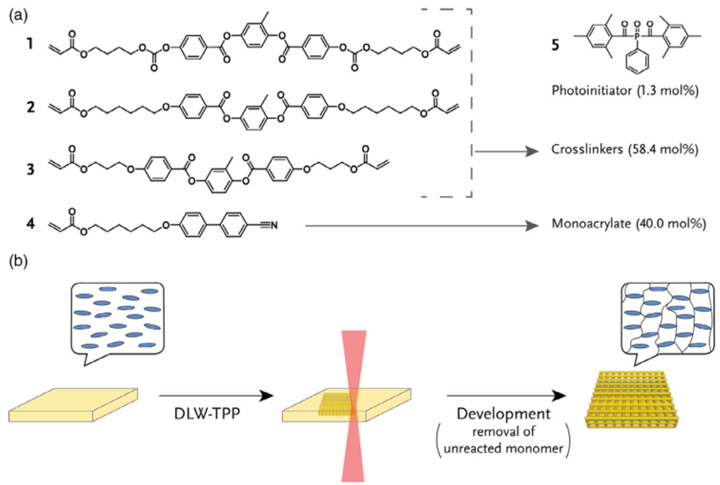
Surface alignment and of nematic LCs by direct laser writing of photopolymer alignment layer

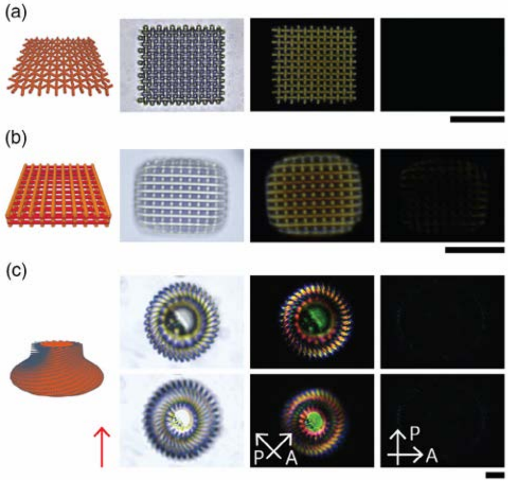


Integrating metal nanoparticles with TPP process



TPP-fabricated temperature-responsive 4D liquid crystal microactuators





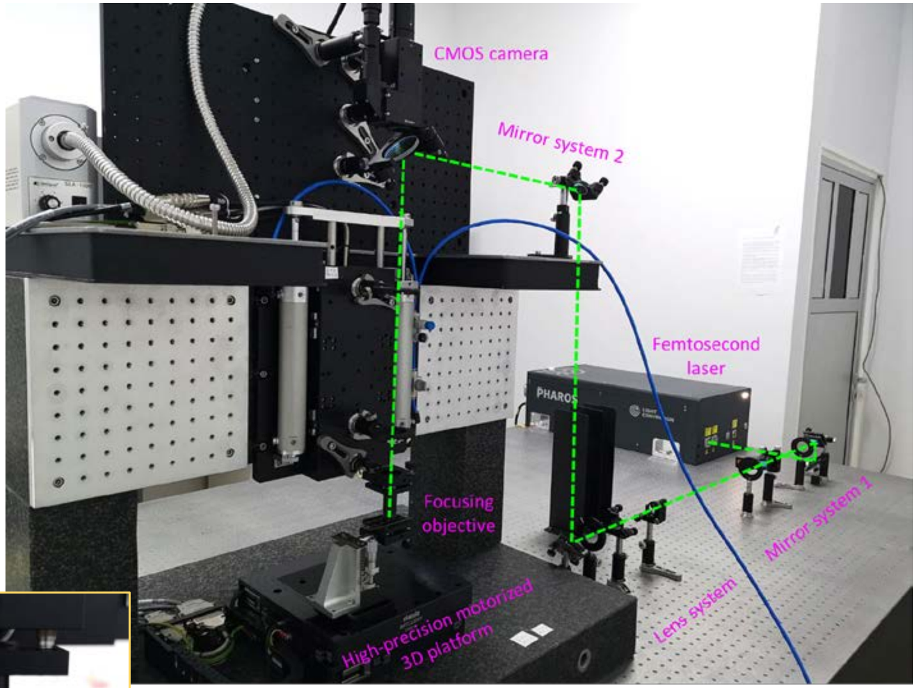
Zhang et al., Opt. Mater. Express (2023) 13(12): 3467
Im et al., Adv. Func. Mater (2023) 33:2211920
del Pozo et al., Small Struct. (2022) 3:2100158

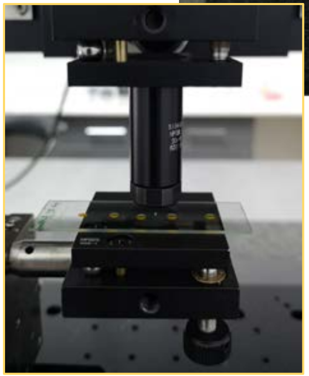
Project work packages

1. Lab space development
2. Numerical model of self-assembled oriented LC structures
3. Self-assembled LC orientation structures driven by TPP-formed substrates
4. Direct laser writing of 3D localized topological structures within the LC volume
5. Laser light-shaped polymer nanocomposites for hybrid substrates
6. Design and creation of multifunctional LC metasurfaces

Optical lab of two-photon polymerization

TPP optical setup based on the femtosecond laser

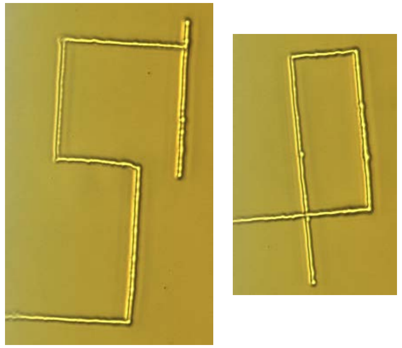




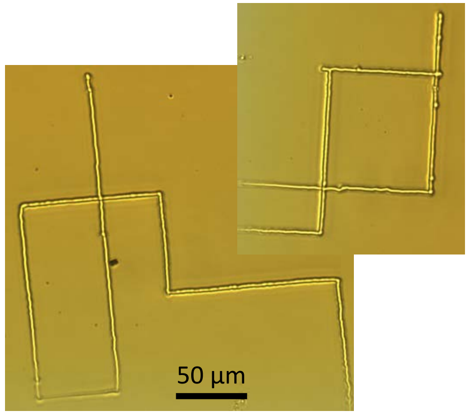
Light beam parameters: 515 nm, 200 kHz,
10W maximal optical power at the laser output.

Manually printed TPP-patterns from
Sona Harutyunyan and Vahan Sargsyan

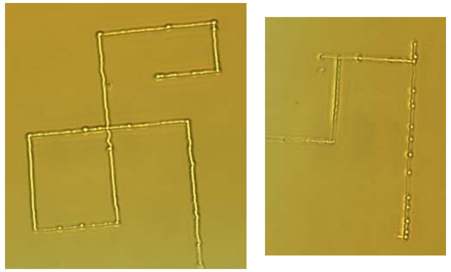
Pulse energy: ≈315 nJ



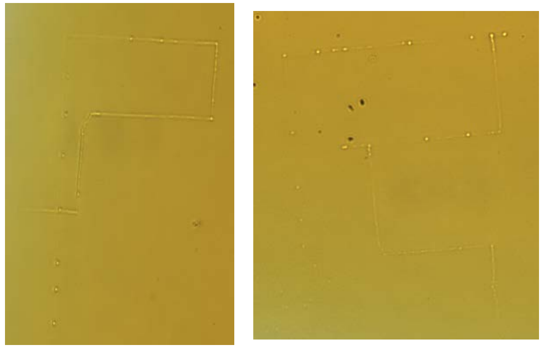
Pulse energy: ≈205 nJ



Pulse energy: ≈92 nJ



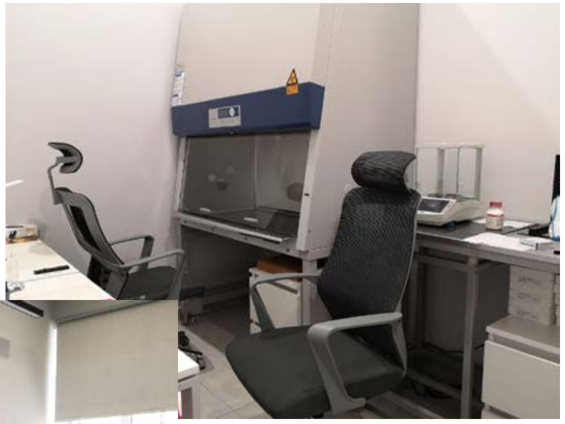
Pulse energy: ≈45 nJ



Recommended printing parameters for the SZ2080 photoresin (FORTH, Greece):

- pulse frequency 50-150 kHz (sweet spot 50-100 kHz);
- pulse energy 50-200 nJ (use 80-120 nJ for start).

New lab space for wet chemistry, sample preparation and characterization



Research articles and conference presentations

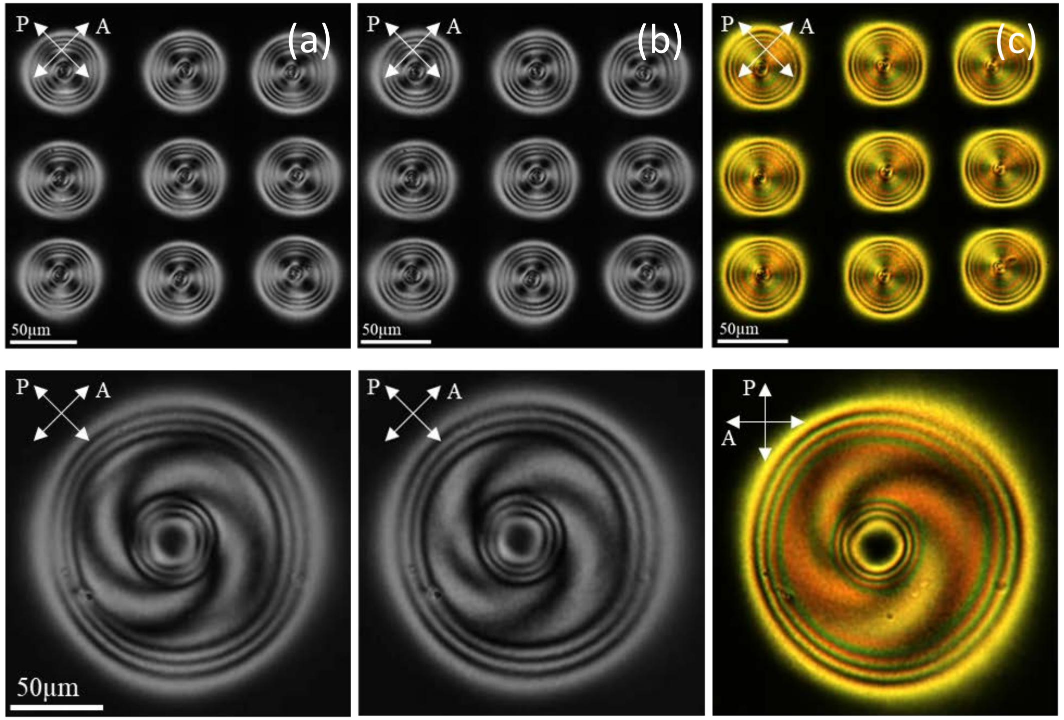
- 3 articles acknowledge support from the Integration grant
  - **T. Orlova**, A.M. Solis, H.R.O. Sohn, T. Madeleine, G. D'Alessandro, I.I. Smalyukh, **M. Kaczmarek**, and J. Brodzki 'Multiscale geometrical and topological learning in the analysis of soft matter collective dynamics', Phys. Rev. Materials 10, 015602, 2026 (**Editor's Suggestion**);
  - E.V. Aksenova, **I.S. Lobanov**, **T. Orlova**, V.M. Uzdin, and A.D. Kiselev, 'Umbilic surfaces as a robust topological probe of 3D solitons in cholesteric liquid crystals', Nanosystems: Phys. Chem. Math. 17 (1), 1–13 (2026);
  - T.V. Reztsov, A.V. Chernykh, **T. Orlova**, and N.V. Petrov, 'A dynamic analysis of toron formation in chiral nematic liquid crystals using polarization holographic microscope', Polymers 17 (13), 1849, 2025.
    - 1 is co-authored with the International partner
      - **T. Orlova**, A.M. Solis, H.R.O. Sohn, T. Madeleine, G. D'Alessandro, I.I. Smalyukh, **M. Kaczmarek**, and J. Brodzki 'Multiscale geometrical and topological learning in the analysis of soft matter collective dynamics', Phys. Rev. Materials 10, 015602, 2026 (**Editor's Suggestion**).

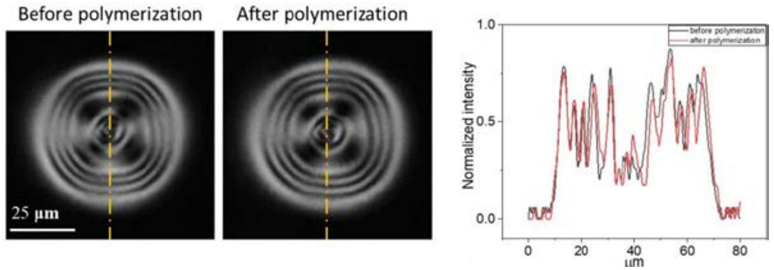
- Other articles related to the project research
  - **D. Darmoroz**, A. Piven, and **T. Orlova**, 'Large supramolecular patterns in photoactive chiral liquid crystals for light-controllable soft micromechanical systems', Small Structures 6 (12), e202500506, 2025;
  - A.N. Shalev, A.A. Misura, A.O. Georgieva, A.V. Chernykh, N.V. Petrov, **T. Orlova**, **I.S. Lobanov**, E.V. Aksenova, V.M. Uzdin, and A.D. Kiselev, 'Polarization-controlled orbital angular momentum of light passing through a cholesteric spherulite', Opt. Lett. 50 (16), 4866-4869, 2025;
  - S.A. Shvetsov, **D.D. Darmoroz**, A. Vasil'ev, **T. Orlova**, **I.S. Lobanov**, and M. Rafayelyan, 'Light-induced isotropic pen for generation of topological solitons and hopfion-toron transition in frustrated chiral nematic films', Chaos Solit. Fractals 199 (3), 116905, 2025.
  - T.V. Reztsov, A.V. Chernykh, A.S. Ezerskii, T. Han, V.R. Gresko, E.G. Tsiplakova, J. Li, M.M. Sergeev, B. Wang, **T. Orlova**, L. Li, S. Makarov, H. Tian, and N.V. Petrov, 'Polarization holographic monitoring for laser treatment diffractive optical elements and metasurfaces fabrication in functional materials photonics', Bull. Russ. Acad. Sci. Phys. 89, S523–S533 (2025);

- Conference presentations
  - *Oral presentation:* **D. Darmoroz,** S. Shvetsov, **T. Orlova**, and M. Rafayelyan, 'Polymer-stabilized topological solitons formed by low-power light beam in a dye-doped chiral nematic liquid crystals'. 9th International Soft Matter Conference, 29 Sep – 3 Oct 2025, Chania, Crete, Greece;
  - *Invited talk:* **T. Orlova**, 'Particle-like elastic excitations in chiral liquid crystals for applications in optics and photonics', Fourth International Sino-Russia Forum on Science and Technology, 23 -27 Oct 2025, Harbin, China.

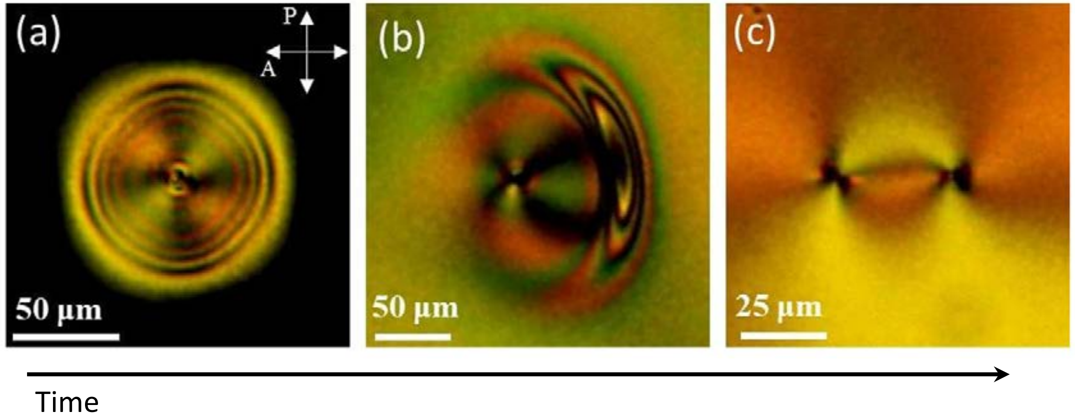
Polymer-stabilized topological solitons formed by low-power light beam in a dye-doped chiral nematic liquid crystal

Polymer network-stabilized ensemble of topological solitons before photopolymerization (a) and after (b,c)





"On-the-fly" polymer network-stabilized defect structures induced by an applied electric field



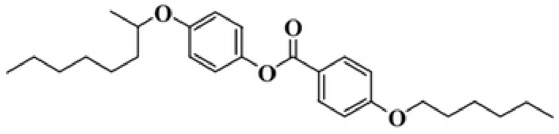
Oral presentation: **D. Darmoroz,** S. Shvetsov, **T. Orlova**, and M. Rafayelyan, 'Polymer-stabilized topological solitons formed by low-power light beam in a dye-doped chiral nematic liquid crystals'. 9th International Soft Matter Conference, 29 Sep – 3 Oct 2025, Chania, Crete, Greece;

Polymer-stabilized topological solitons formed by low-power light beam in a dye-doped chiral nematic liquid crystal

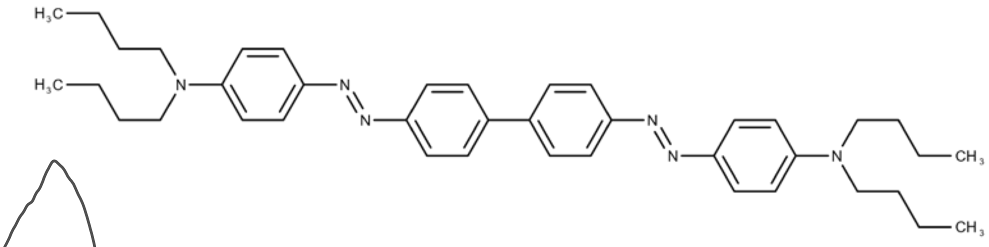
Chiral LC mixture with a dual response to a laser beam (532 nm) with a mW-level optical power and to an applied external electric filed

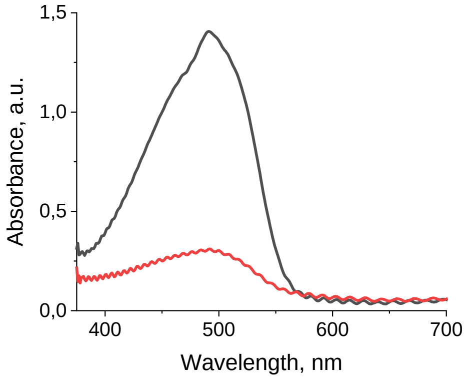
1. Dual-frequency NLC **DP002-113** (HCCH, China)

2. Chiral additive **R811**, 0.22 wt.% (HCCH, China)



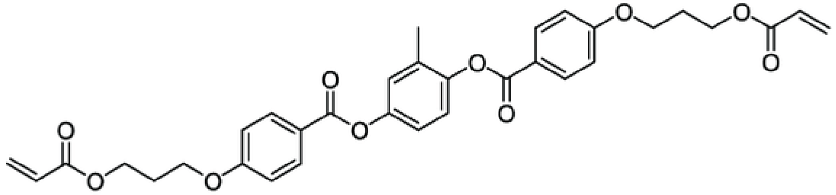
3. **Bis-azobenzene dye**, 0.3 wt.% (Hiap L. Ong)



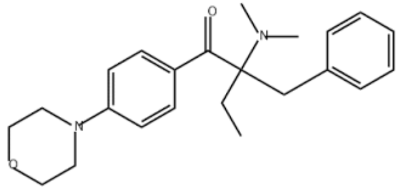


Additives for LC polymerization

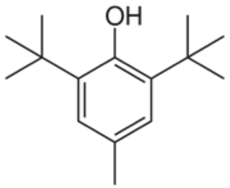
4. Crosslinker, monomer **RM257** (Sigma-Aldrich)



5. Photoinitiator, **Irgacure 369** (Sigma-Aldrich)



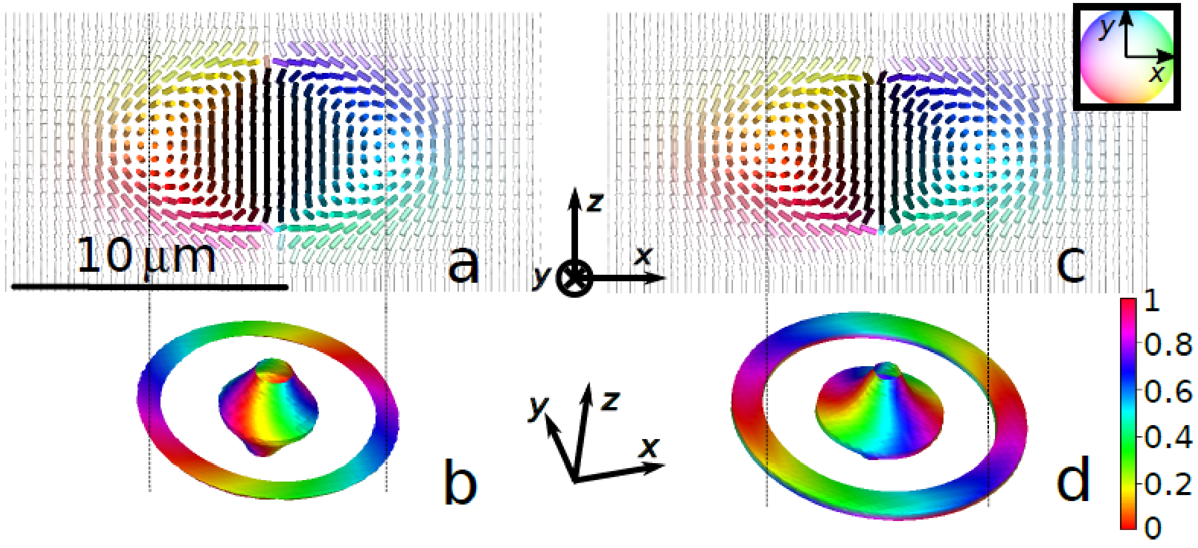
6. **Inhibitor,** butylated hydroxytoluene (Sigma-Aldrich)



A non-ionic LC aligning layer is required

Numerical modeling of 3D solitons in chiral LCs and analysis of umbilic surfaces as a robust topological probe

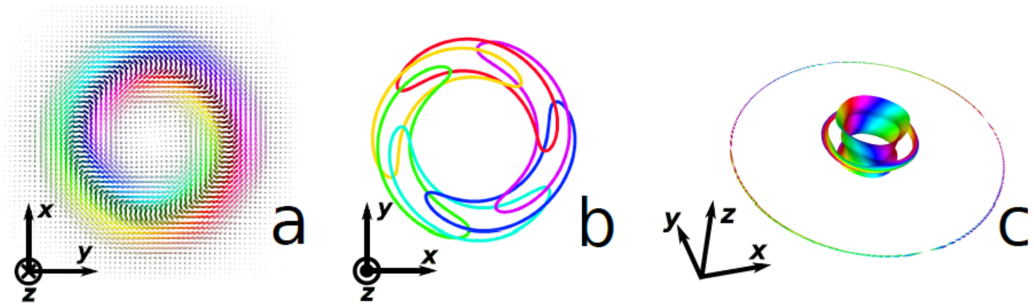
Umbilic surfaces in torons



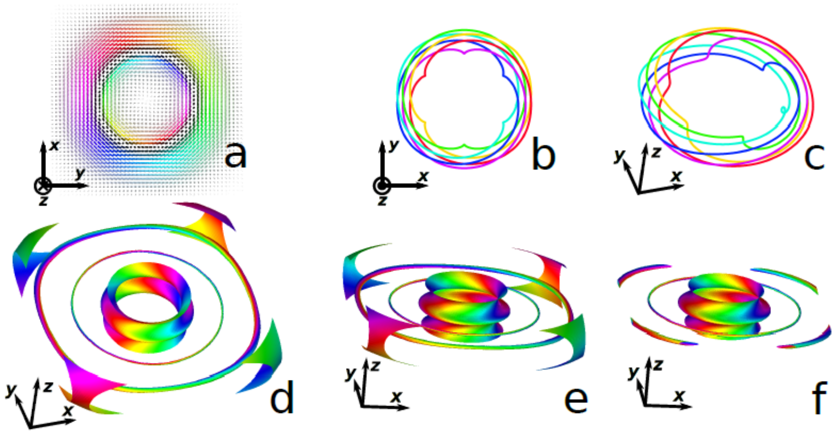
The relaxed configuration obtained by the energy minimization (a,b) and analytical ansatz (c,d)

Umbilic lines are the regions where the gradients of **n** are isotropic in directions orthogonal to **n**. This means that umbilic lines are the cores of vortex structures.

Umbilic surfaces in the CF1-loop structure



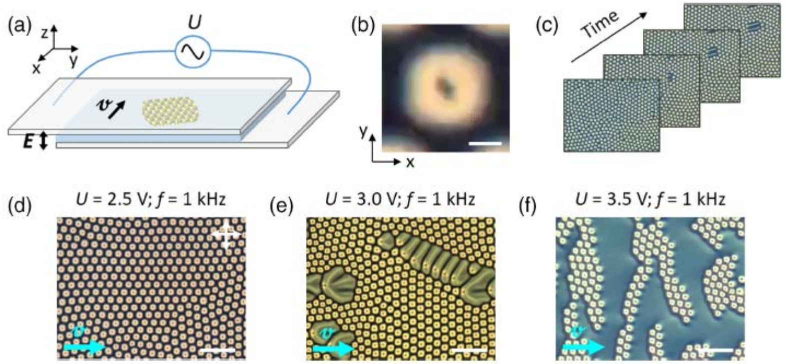
Umbilic surfaces in the CF2-loop structure



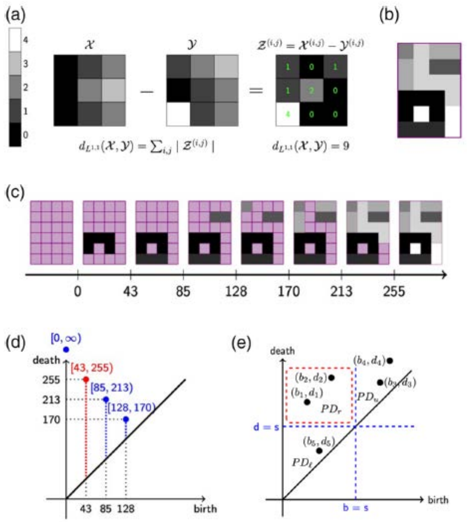
E.V. Aksenova, I.S. Lobanov, **T. Orlova**, V.M. Uzdin, and A.D. Kiselev, ‘Umbilic surfaces as a robust topological probe of 3D solitons in cholesteric liquid crystals’, Nanosystems: Phys. Chem. Math. 17 (1), 1–13 (2026)

Multiscale geometrical and topological learning in the analysis of soft matter collective dynamics

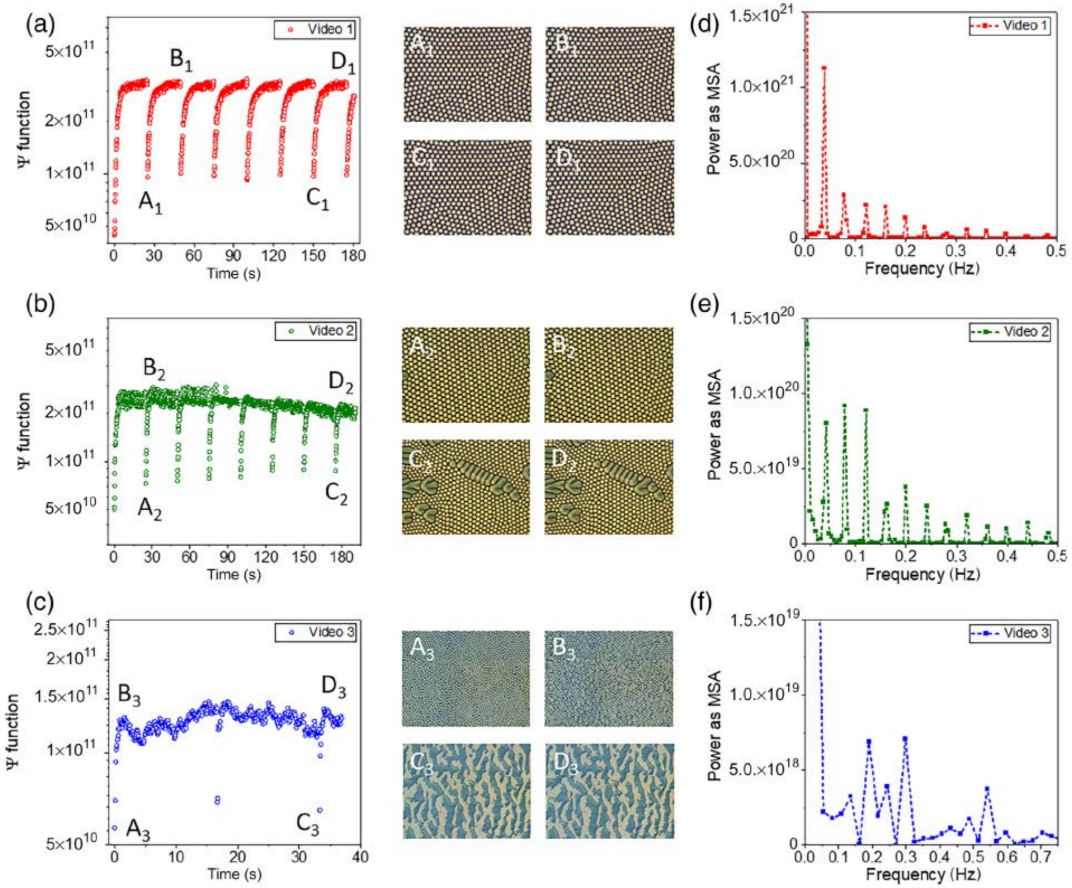
Evolutionary and topological approaches to the analysis of complex soft dynamic systems



Geometric and topological methods for image analysis



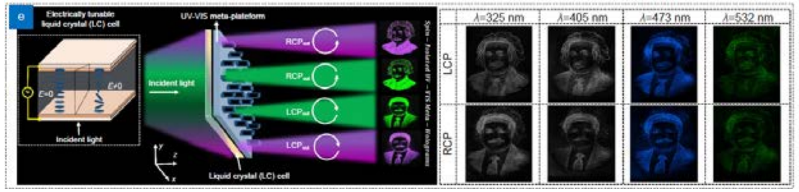
Periodic behavior of the Ψ function, a new topological descriptor reflecting optimized structural heterogeneity



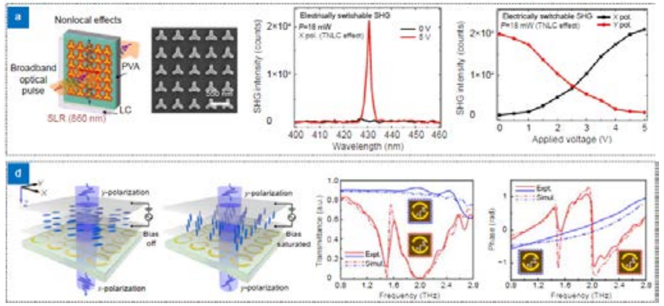
T. Orlova, A.M. Solis, H.R.O. Sohn, T. Madeleine, G. D'Alessandro, I.I. Smalyukh, **M. Kaczmarek**, and J. Brodzki 'Multiscale geometrical and topological learning in the analysis of soft matter collective dynamics', Phys. Rev. Materials 10, 015602, 2026 (**Editor's Suggestion**)

The concept of topological-chiral LC-hybridized metasurfaces

Liquid crystal programmable metasurfaces

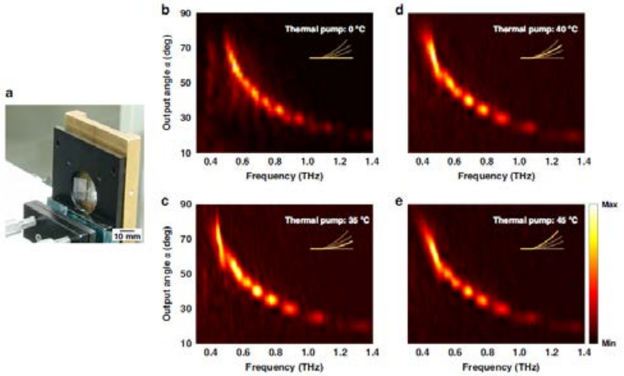


The broadband metahologram operating in both the UV and visible regions



Electrically-driven spectral tuning in the NIR and THz regions

Liquid crystal elastomer metasurfaces for active THz optics



Optical effects demonstrated:

- controllable and broadband wavefront steering;
- tunable azimuthal deflection;
- spatial separation of orthogonal polarization components;
- frequency modulator.

Promising optical and photonic applications of metastable topological structures in chiral liquid crystals

1st Meline Grigoryan
Optics and Photonics Center, Institute of Physics
Yerevan State University
Yerevan, Armenia
meline.grigoryan@ysu.am

2nd Tatiana Orlova
Optics and Photonics Center, Institute of Physics
Yerevan State University
Yerevan, Armenia
tetiana.orlova@ysu.am

***Abstract*—Dynamic control of terahertz (THz) electromagnetic waves remains a significant challenge today due to the limited availability of compact and reconfigurable photonic materials. Metasurfaces provide precise wavefront control at the subwavelength scale but typically remain static after fabrication, limiting their adaptability. In contrast, liquid crystals (LCs) allow tuning through molecular reorientation driven by external fields. However, most LC-integrated metasurfaces utilize the reorientation of spatially uniform LC layers and global modulation of the refractive index.**

Here, we discuss the prospects for integrating topologically structured chiral nematic liquid crystals hosting metastable soliton excitations with metasurfaces. Liquid crystal solitons are capable of providing localized, tunable refractive index modulations with characteristic dimensions on the micron range, comparable to the dimensions of THz metasurfaces. Such hybrid systems can provide spatially structured and dynamically tunable optical response, offering a path to programmable THz photonic elements that go beyond the traditional control of homogeneous liquid crystals and fixed metasurface designs.

***Index Terms*—liquid crystal integrated metasurface, chiral nematic liquid crystal, topological LC soliton, THz photonics, reconfigurable photonic systems, perspective**

I. Structure and properties of metasurfaces

Metasurfaces are artificial planar optical structures composed of arrays of subwavelength elements, often called meta-atoms, that locally alter the amplitude, phase, and polarization of electromagnetic waves [1]–[4]. Unlike conventional optical components such as lenses or diffraction gratings, which rely on wave propagation through bulk materials, metasurfaces operate through spatially varying boundary conditions imposed at an interface. Their fundamental principle is based on the interaction of an electromagnetic wave with a structured surface, where local resonances or guided modes within each meta-atom generate phase shift. These shifts can be engineered to produce arbitrary spatial phase profiles, thus enabling control over wavefront formation. If the spatially dependent phase shift is denoted as $\varphi(x, y)$, then the direction and curvature of the outgoing wavefront are determined by its spatial derivatives. For example, a linear phase gradient $\varphi(x) \propto x$ results in beam deflection, a quadratic dependence $\varphi(x) \propto x^2$ leads to focusing, and an azimuthally varying phase $\varphi(\theta) \propto \theta$ generates vortex beams carrying orbital angular momentum. These properties enable metasurfaces to function as compact optical elements such as flat lenses (metalenses), beam deflectors, holographic phase plates, vortex beam generators, and tunable diffraction gratings. In the terahertz (THz) and infrared ranges, metasurfaces are particularly relevant for applications in imaging, adaptive optics, beam steering, spectroscopy, communication systems, and integrated photonic devices, where conventional bulk components are difficult to implement or dynamically control.

In terms of the materials used, two classes of metasurfaces can be distinguished: metallic and dielectric [3], [5]. Metallic metasurfaces are based on patterned conductive elements such as split-ring resonators, dipole antennas, cross resonators, or

This research was supported by the Higher Education and Science Committee of MESCS RA (Research project 24IRF-1C003).

their central structure remains largely intact, while the peripheral twisted regions expand. This deformation redistributes the spatial variation of the refractive index without changing the topological configuration. This leads to a gradual spatial redistribution of the optical phase contributions. Because the topology is preserved, the optical response changes smoothly rather than abruptly. Alternatively, depending on the parameters of external fields applied, topological reorganization may be induced, leading to discrete transformations such as splitting or annihilation events.

V. Topological chiral LC-hybridized metasurfaces

The integration of metasurfaces with chiral LC films hosting topological solitons can be viewed as a pathway toward hybrid photonic systems with enhanced and reconfigurable functionality [44]. The underlying principle is based on the coupling between the electromagnetic resonance of the metasurface and the spatially varying dielectric medium created by the LC soliton structures. This system can be described within the framework of Maxwell's equations in anisotropic media, $\nabla \times \mathbf{E} = -\partial \mathbf{B}/\partial t$, $\nabla \times \mathbf{H} = \partial \mathbf{D}/\partial t$, with the constitutive relation $\mathbf{D} = \hat{\epsilon}\epsilon_0\mathbf{E}$, where $\hat{\epsilon}$ is the orientation-dependent permittivity tensor determined by the local director field. Spatial variations of the director field associated with LC solitons will lead to local changes in the dielectric permittivity, which, in turn, alter the resonance conditions and the optical response of the metasurface.

In metallic metasurface configurations, capacitive gaps act as regions of highly localized electric fields. If one imagines a toron overlapping such regions, local changes in permittivity can be expected to affect the effective capacitance and, consequently, the resonant frequency. Even relatively small changes in director orientation may lead to noticeable variations in the optical response. Therefore, metallic metasurfaces hybridized with LC solitons are anticipated to be particularly sensitive to localized dielectric perturbations and may be suitable for applications requiring strong local modulation. In dielectric metasurface configurations, electromagnetic fields are distributed over the volume of high-refractive-index resonators rather than confined to narrow gaps. As a result, interaction with the LC soliton will occur over a large region. The presence and motion of torons will modify the refractive index distribution around dielectric meta-atoms, which would lead to smooth and continuous tuning of resonance frequencies and phase response. This suggests that dielectric metasurfaces hybridized with LC solitons will be better suited for applications involving distributed wavefront shaping and continuous phase modulation.

The behavior of such a combined system is expected to be determined by the interplay of several characteristic length scales, including the wavelength λ, the toron diameter D, the metasurface period a, and the LC layer thickness h. Depending on the ratio of these parameters, different physical regimes can be expected to be realized. When $D \ll \lambda$, torons will act as subwavelength perturbations, facilitating the effective medium response without significant scattering. When D becomes comparable to a fraction of the wavelength, torons will behave as optically resolved structures capable of inducing significant phase and amplitude modulation. In the regime $D \approx a$, each toron will interact with a single meta-atom, ensuring pixel-level control, whereas for $D > a$, the interactions will extend over multiple resonators, leading to collective effects.

Furthermore, the dynamic behavior of torons will allow for temporal reconfiguration of the metasurface response. The spatial distribution of permittivity in this case becomes time-dependent, $\epsilon(x, y, t)$, leading to the evolution of phase profiles $\varphi(x, y, t)$. Although the motion of torons is relatively slow, typically on the order of microns per second, this can be expected to be sufficient for smooth control of optical functions such as beam steering, focusing, and diffraction pattern formation. This regime is particularly relevant for adaptive optics and programmable photonic devices.

Finally, collective arrangements of torons may give rise to emergent photonic behavior. If torons form quasi-periodic or ordered arrays with spacing Λ comparable to the wavelength λ, then diffraction and Bragg-like effects can be expected. In contrast, when $\Lambda \ll \lambda$, the system will behave as an effective anisotropic medium with tunable optical properties. By controlling the density and spatial organization of the torons, it will be possible to dynamically tune the system's photonic response.

VI. Conclusion

The overall concept of combining metasurfaces with chiral LC films hosting topological solitons can be considered as a versatile framework for reconfigurable THz photonics. Metallic metasurfaces provide strong localized sensitivity suitable for discrete switching and resonance modulation, while dielectric metasurfaces enable smooth, distributed interactions appropriate for wavefront shaping and adaptive optical components. Liquid crystals provide tunable anisotropy, and topological LC solitons introduce localized, metastable, and movable refractive index structures. The interaction between these elements defines a broad design space governed by dimensionless parameters such as D/λ, D/a, and h/λ.

We believe that, taken together, such systems may form a new class of hybrid photonic media in which electromagnetic boundary conditions, anisotropic molecular order, and topological excitations interact coherently. While the individual components, namely, metasurfaces and topological LC solitons, are well understood, their integration, as outlined here, remains unexplored. The proposed approach may enable the emergence of programmable, topological chiral LC-hybridized metasurfaces with new optical functionalities.

References

[1] N. Yu and F. Capasso, "Flat optics with designer metasurfaces," Nature Mater., vol. 13, pp. 139–150, 2014.
[2] A. V. Kildishev, A. Boltasseva, and V. M. Shalaev, "Planar photonics with metasurfaces," Science, vol. 339, p. 1232009, 2013.
[3] H.-H. Hsiao, C. H. Chu, and D. P. Tsai, "Fundamentals and Applications of Metasurfaces," Small Methods, vol. 1, p. 1600064, 2017.

Kang et al., (2024) 7:230216
Zhuang et al., Light: Science & Applications (2023) 12:14
Long et al., Laser Photonics Rev. (2025) 19:2402167

Planned articles and conference presentations in 2026

Research articles in progress

- A. Osminin, A. Hambardzumyan, **M. Altunyan**, G. Ustinov, A. Muravev, M. Rafayelyan, S. Shvetsov, and **T. Orlova**, 'Digitally programmable light recording of topological liquid crystal structures in photoswitchable chiral-azo-doped nematics'.
- N. Podoliak, T. Madeleine, A.F. de Fazio, A.G. Kanaras, A.M. Solis, **T. Orlova**, G. D'Alessandro, J. Brodzki, and **M. Kaczmarek**, 'Fully reversible assembly and topological analysis of gold nanoparticle networks in liquid crystals'.
- **D. Darmoroz**, E. Aksenova, S. Shvetsov, **I. Lobanov**, A. Kiselev, V. Uzdin, **T. Orlova**, M. Rafayelyan, 'Topological evolution of a toron in an applied electric field and polymer stabilization of non-equilibrium liquid crystal structures'.
- **T. Orlova**, **I.S. Lobanov**, I.M. Tambovtsev, A.D. Kiselev, V.M. Uzdin, and E. Brasselet, 'Light-driven reaction-diffusion spatiotemporal dynamics in photoactive chiral liquid crystals'.

Conference talks in summer 2026

- *Invited talk:* **D.D. Darmoroz**, S.A. Shvetsov, M.S. Rafayelyan, and <u>**T. Orlova**</u>, 'Low-power optical generation and polymer stabilization of topological structures in liquid-crystalline materials', *The 22nd International Conference Laser Optics (ICLO 2026)*, 22-26 June 2026, Saint-Petersburg, Russia.
- *Invited talk:* **M. Grigoryan and** <u>**T. Orlova**</u>, 'Promising optical and photonic applications of metastable topological structures in chiral liquid crystals', *PhotonIcs & Electromagnetics Research Symposium (PIERS 2026)*, 27–31 July 2026, Suzhou, China.
- *Invited talk:* <u>**T. Orlova**</u>, A.Membrillo Solis, H.R.O. Sohn, T. Madeleine, G. D'Alessandro, I.I. Smalyukh, **M. Kaczmarek**, and Jacek Brodzki, 'Spherulite patterns in cholesteric liquid crystals - Geometric and topological analysis of their collective and structural dynamics', *SPIE Optics and Photonics*, 23-27 Aug 2026, San Diego, USA.

Thank you for your kind attention!

Integration Grant 24IRF-1C003
Hierarchically ordered complex soft materials: where additive manufacturing meets self-assembly

ՀՀ ԿԳՄՍՆ
Բարձրագույն կրթության և գիտության կոմիտե

YEREVAN
STATE
UNIVERSITY