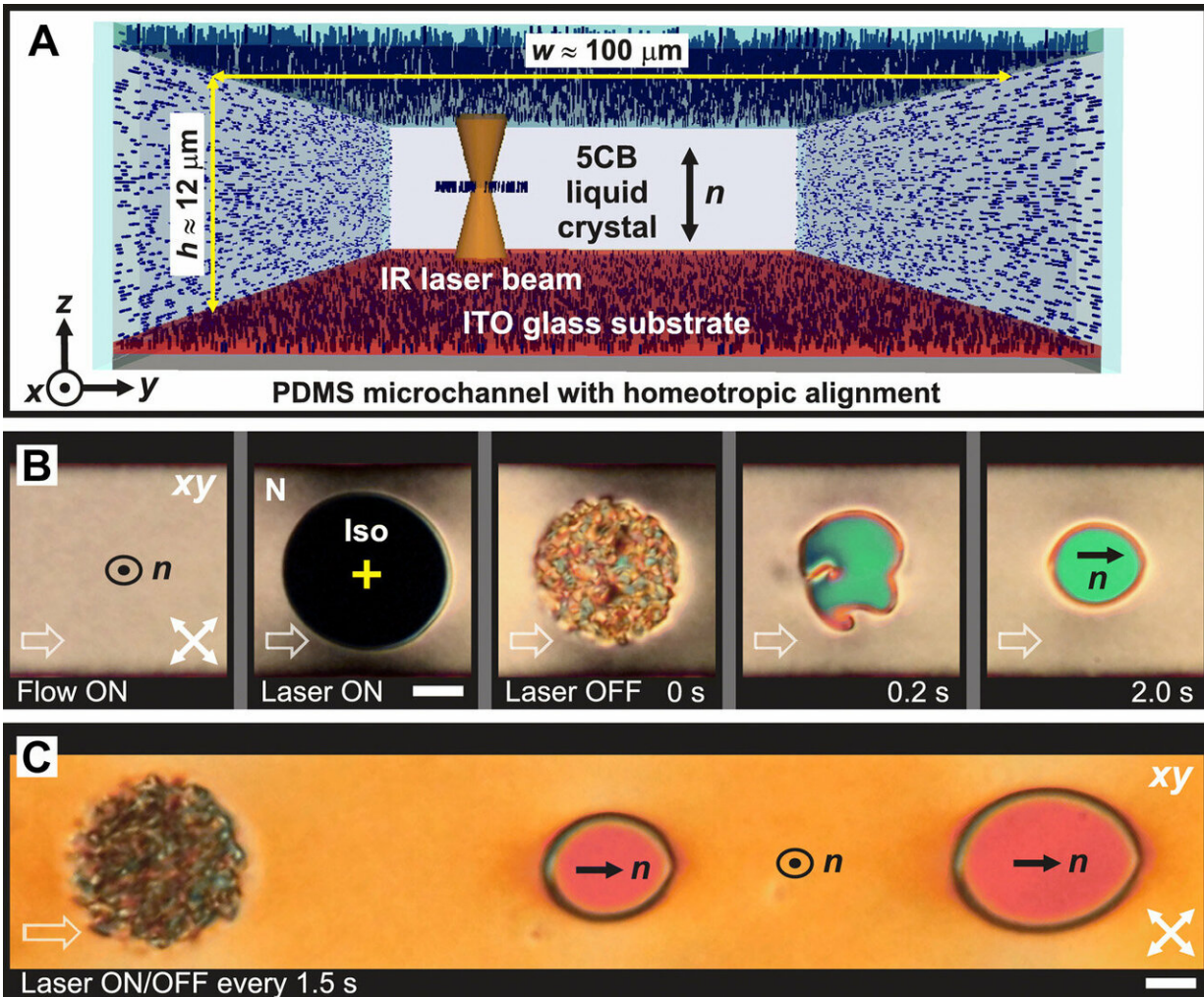


# Sculpting stable structures in pure liquids

February 21 2019, by Thamarasee Jeewandara



Nucleation of orientation phase domains in pressure-driven nematic microflows. (A) Schematic illustration of a channel with homeotropic anchoring on the top and bottom surfaces used in the experiment; IR, infrared; ITO, indium tin oxide. (B) The nematic in a channel looks black between crossed polarizers in the absence of flow and gains visible birefringence due to flow-driven director distortion that traps a domain of the flow-aligned state (also called the dowser

state from here on);  $n$  denotes the nematic director. Strongly absorbed light of the laser tweezers heats the NLC, creating an isotropic (Iso) island that is quenched into the nematic (N) phase when the laser is switched off. The dense tangle of defects coarsens into a single defect loop that traps a flow-aligned dowsler state, identifiable as a green area at low velocity. (C) The laser-induced nucleation of dowsler domains can be automated and their shape can be dynamically controlled by tuning the flow parameters. Crossed double arrows indicate the orientation of the polarizers. White empty arrows in the bottom left corners indicate direction and qualitative velocity of the flow throughout the paper. Credit: Science Advances, doi: 10.1126/sciadv.aav4283

Oscillating flow and light pulses can be used to create reconfigurable architecture in [liquid crystals](#). Materials scientists can carefully engineer concerted [microfluidic](#) flows and localized [optothermal](#) fields to achieve control on [nucleation](#), growth and shape of such liquid domains. In comparison, [pure liquids](#) in thermodynamic equilibrium are structurally homogeneous. Experimental work based on theory and simulations have shown that if the liquids are maintained in a controlled state of nonequilibrium, the resulting structures can be indefinitely stabilized.

Sculpted liquids can find applications in microfluidic devices to selectively encapsulate solutes and particles into optically active compartments to interact with external stimuli for a variety of medical, healthcare and industrial applications. In a recent study published in *Science Advances*, Tadej Emeršič and co-workers in Slovenia and the USA developed pure [nematic](#) liquid crystals (NLC), where they dynamically manipulated defects and reconfigurable states of the materials by the simultaneous application of multiple external fields.

Solid materials can exhibit distinct [structural phases](#) simultaneously, a property that can be manipulated to [engineer functionality](#). However, in pure liquids [at equilibrium](#), such structural phases that correspond to

grain boundaries and defects do not arise. While liquids exhibit a number of attractive features including the [ability to wet surfaces](#), demonstrate high diffusion coefficients and absolute compliance, it is challenging to include additional functionalities to liquids due to their inherent homogeneity. Complex behavior is observed in multicomponent synthetic and biological mixtures and the resulting structures are difficult to manipulate since they occur in out-of-equilibrium situations. Such situations generally involve multiple components with sharp miscibility and gradients between [hydrophilic and hydrophobic](#) domains as well.



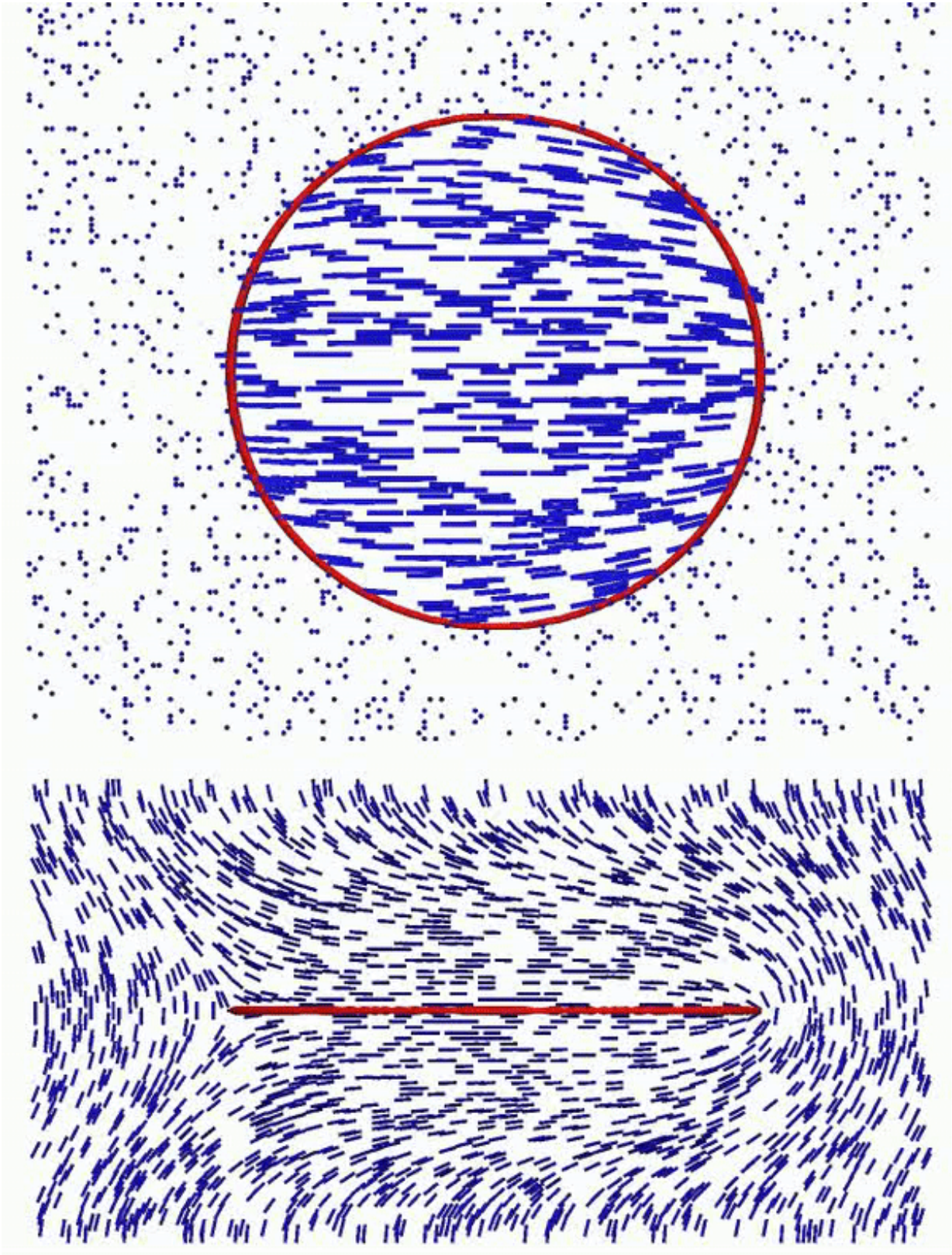
Expansion and contraction of laser-nucleated dowser domains in a moderate nematic microflow. The lifetime of the domain is proportional to the critical velocity and the initial size. Recorded under crossed polarizers at 30 fps, the view field size is  $480 \mu\text{m} \times 120 \mu\text{m}$ . Credit: Science Advances, doi: 10.1126/sciadv.aav4283

Scientists have developed [active matter](#) in the form of living colonies and bioinspired synthetic counterparts. They printed hydrophobic/hydrophilic domains on to liquid mixtures by relying on [surfactant nanoparticles](#) and controlled non-equilibrium systems to demonstrate the motion and transition between different rheological regimes. Liquid crystals (LCs) are [an ideal system](#) to study the

phenomena of interest, such as spontaneous symmetry breaking, topological defects, orientation ordering and external stimuli based phase transitions.

[Nematic liquid crystals](#) (NLCs) are the simplest form of [liquid crystal](#) molecules without orderly positions, and they differentiate from pure liquids at the level of the molecular orientation. NLCs have a range of properties that allow them to [serve as microreactors](#) and conduct inherent polymerization reactions for intriguing future applications. Current work in the field is still experimental, for example, [nematic flows in microfluidic environments](#), which highlight the potential cross-talk between topological defects in different fields of velocity and molecular orientation.

In this work, the scientists observed the phase interface with NLCs for the first time, experimentally accomplished by generating polar-phase domains that were controlled by combining microfluidic confinement, fluid flow rates and [laser pulses](#) in practice. Emeršič et al. used the single-component nematic material [pentyl-cyanobiphenyl](#) (5CB) in all experiments performed in linear microfluidic channels with a rectangular cross-section. The scientists fabricated the channels with polydimethylsiloxane (PDMS) relief and indium tin oxide (ITO)-coated glass substrates using [standard soft lithography](#) procedures. They then filled the microfluidic channels with 5CB in its heated isotropic phase and allowed it to cool down to the nematic phase, before beginning the flow experiments. The scientists also chemically treated the microchannel walls to engineer a strong [homeotropic](#) surface to anchor the 5CB molecules.



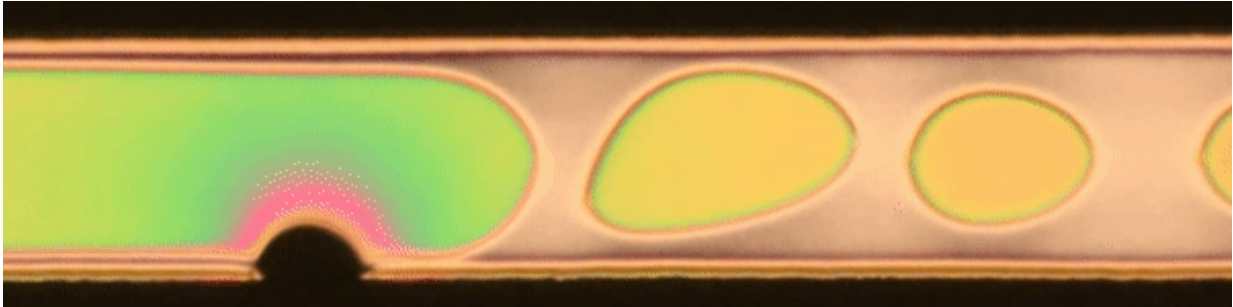
Growing and shrinking dowsers domains in numerically simulated nematic microflows. Simulation of a laser-induced defect loop in a channel either undergoing expansion or shrinkage, subject to a strong or a weak pressure-driven flow. Top: top view of the channel showing defect loop. Bottom: side view showing the evolution of dowsers structure. Elastic constants of 5CB are adopted in the calculation. Credit: Science Advances, doi: 10.1126/sciadv.aav4283

The work represented an ideal experimental model of a quasi-two-dimensional (2D) orientation material phase. In the initial stationary state within a microfluidic channel, the heated material appeared black. When the flow was turned on, depending on the velocity of flow, the [birefringent](#) appearance changed from black to bright colors. The flow aligned domains evolved in this way to either grow or annihilate with flow velocity.

The [materials scientists](#) named the flow regime the 'bowser state' due to the bowed profile of the material and the flow aligned state as the 'dowsers state' due to its analogy to the so-called [dowsers field in nematostatics](#), where nematostatics is the charge density of elastic nematic materials, analogous to electrostatics. The dowsers state has an anisotropic orientation with its own elastic behavior, topological defects and [solitons](#) (a solitary wave packet that maintains its shape while propagating at constant velocity). In comparison, the bowser state is effectively isotropic and simple in the simplified 2D view. The scientists were able to control the shape, splitting and coalescence of these phase domains.

Emeršič et al. conducted all experiments at room temperature, driving and controlling fluid flow in the microchannel with a pressure-driven microfluidic flow control system. They studied the flow regimes, reorientation dynamics and flow-driven deformations of 5CB in the

microchannels using polarized light microscopy. The scientists built [laser tweezers](#) around the inverted optical microscope with an IR fiber laser operating at 1064 nm as a light source, and a pair of acoustic-optic deflectors driven by a computerized system to precisely manipulate the beam.



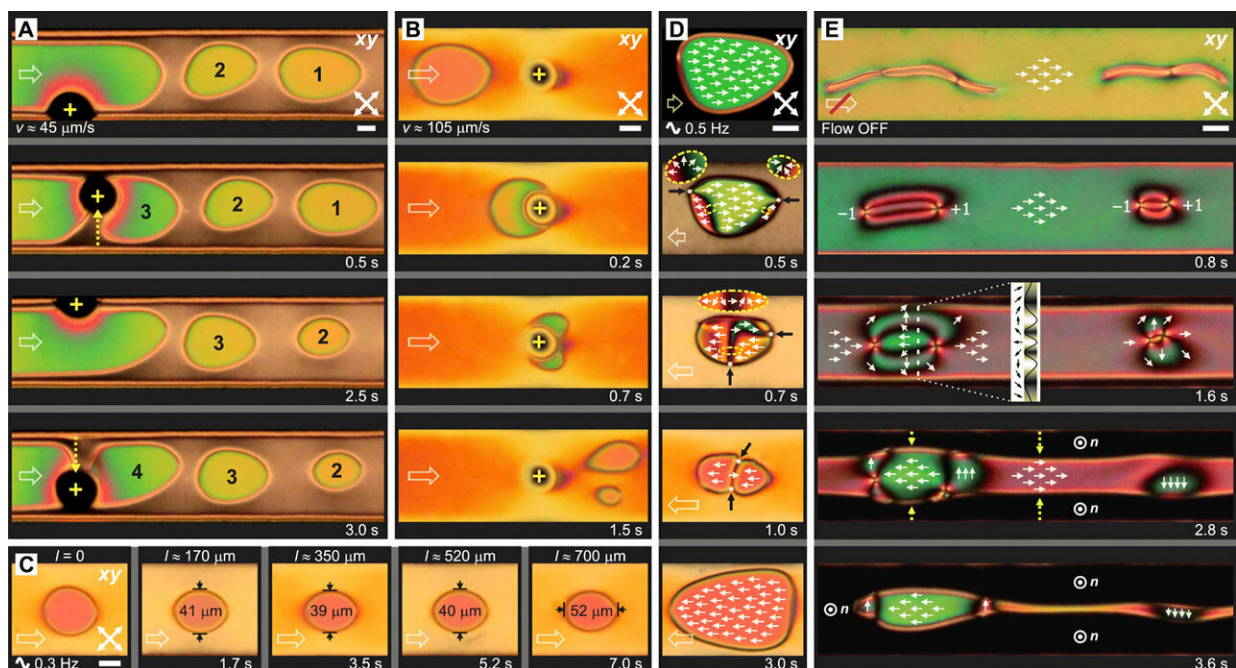
Producing a steady stream of dowser domains by chopping the bulk dowser state with a moving laser spot. By moving an isotropic island of laser-heated nematic phase transversely across the phase boundary between the dowser and bowser state (black) one can produce a uniform train of dowser domains. Recorded under crossed polarizers at 30 fps, the view field size is  $480 \mu\text{m} \times 120 \mu\text{m}$ . Credit: Science Advances, doi: 10.1126/sciadv.aav4283

In the study, the flow-aligned dowser state was stable under strong flows but unstable in weak flow. Depending on the flow velocity, the dowser domains could grow and shrink in the experiments as seen in numerical simulations. The scientists calculated the criteria for growth and shrinking of domains in time and indicated how the domains grew, shrunk or annihilated along the channel.

By carefully applying the laser tweezers, the scientists showed that a steady stream of domains could be produced by dissecting the original bulk dowser with a moving laser spot, where the laser melted the sides of

the material's phase boundary. A growing [domain](#) at higher flow velocity could thus be split longitudinally in half, with a static laser beam at low light intensities.

The laser tweezers allowed dynamic control of the size, number and lifetime of generated dowser domains, which were further manipulated by modulating the periodic flow velocity. For instance, under uniform flow, the dowser field aligned uniformly along the direction of flow to either grow or shrink, depending on the regime of the velocity. The scientists were able to tune and actively control the flow as a constant-sized domain that could be stably maintained for more than ten seconds.



Systematic reshaping of dowser domains under laser action and oscillatory flows. (A) Moving the laser beam transversely across the bulk dowser pinches off a uniform “train” of the domains. (B) A static beam at a low power of 80 mW generates a small isotropic region that cuts a large dowser domain longitudinally in half. (C) The shape and size of the domain can be maintained over long time and length scales by periodically modulating the driving pressure around the



value that induces the desired average flow rate. (D) Under an alternating flow, a dowser domain reverses orientation every time the flow direction is changed. The reorientation creates surface point defects and realigning fronts, visible under the microscope as a rapid color change. The energetically unfavorable “old” orientation shrinks into a narrow  $2\pi$  soliton and pinches the domain boundary (black arrows). (E) Sufficiently rapid flow reversal creates point defect pairs connected by solitons. With the flow turned off, the characteristic length goes to infinity, and the solitons expand, revealing their signature profile in transmitted light intensity (inset). In a slow residual flow, flow-aligned parts shrink more slowly than parts with unfavorable orientation. Scale bars, 20  $\mu\text{m}$ . Credit: Science Advances, doi: 10.1126/sciadv.aav4283

Furthermore, in the model developed by Emeršič et al., they showed how the flow direction could be reversed for the dowser domain, leading to a rapid reversal of orientation from the previous state of equilibrium. In addition, the dowser field could couple to external magnetic and electric fields and gradients of the channel thickness to determine the control, [flow](#) steering and optical tuning of the 5CB nematic material. The scientists observed the direct response to the [external stimuli](#) clearly through birefringence in the study and determined this to be a suitable method to measure the viscoelastic and rheological properties of the material.

Emeršič et al. envision the possibility of conducting chemical reactions in such enclosed volumes in practice, as previously shown with [liquid crystal templates](#). In addition to that, based on the principles outlined by Emeršič and co-workers, a 3D printing system can be engineered to contain liquids, within which complex and out-of-equilibrium structures can be created and stabilized. The experimental models developed in this study using standard thermotropic LCs are also transferrable to active and biological materials with nematic behavior. The proposed and demonstrated method is a technical tool in materials science, with

potential applications in biophysics, chemistry and chemical engineering.

**More information:** Tadej Emeršič et al. Sculpting stable structures in pure liquids, *Science Advances* (2019). DOI: [10.1126/sciadv.aav4283](https://doi.org/10.1126/sciadv.aav4283)

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