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Reconfigurable Microlens Arrays via Programmable Liquid Crystal Microdroplet Patterning

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ABSTRACT

In this work, we propose the novel method to simply but precisely manipulate the hemispherical LC microdroplet array on a substrate through controlled nucleation and growth. Moreover, UV light irradiation on a photo-sensitive layer enables precise control of LC droplet location and size across the entire surface for pixelazation. The LC droplet arrays display scattered structural color that varies with the specific size of LC droplets.

1 Introduction

Microscopic droplets are ubiquitous in living systems, often appearing as organelles such as lipid droplets, lysosomes, and peroxisomes. These droplets can synthesize metabolites that help regulate cellular homeostasis. Applying this bio-based architecture to engineered optics is promising because liquid crystal (LC) droplets combine high birefringence with high response with various stimulus (e.g., electric/magnetic field, temperature, pressure, chemicals). Dense arrays of LC surface droplets substantially improve optical performance and enable additional functionalities yet processing methods for LC droplet arrays remain impractical because they require lengthy multi-step processes and high costs.

Here, we present a simple thermodynamic strategy that creates LC microdroplet arrays after thermal cycling processes using LC–polymer bilayer. Heating on the entire system dissolves LCs into the polymer matrix, allowing it to permeate the polymer film. Subsequent cooling induces phase separation, and the separated LC droplets nucleate and growth into an ordered array at the interface between the polymer and the underlying substrate.

Two global parameters govern the outcome: the increase in peak annealing temperature (T_{heat}) facilitate both the higher number of nucleation sites and mean droplet diameter. In contrast, accelerating the cooling rate ($-dT/dt$) only increases the density while decreasing the size.

Additional surface selectivity enables drop by drop control of nucleation sites. A photo switchable layer locally modulates LC adhesion, so droplets form only at preferential light patterned pixels, without any etching, or microfluidic processes. Areal densities exceeding 1×10^5 droplets per square centimeter with less than five percent size variation are readily obtained.

Each surface droplet behaves as a miniature spherical cavity. Refractive index contrast traps internal reflections that yield angle-insensitive structural colors, with the reflected wavelength uniquely encoded by droplet size. The radially aligned nematic LC director adds an electro- and thermally tunable phase delay, allowing rapid, reversible color shifts over entire visible spectrum.

By uniting deterministic geometry, adaptive photonics, and manufacturing simplicity, immobilized LC droplet arrays provide a platform for hyperspectral filters, reconfigurable microlens arrays for three-dimensional imaging, and physically unclonable functions for security tags.

2 Experiment

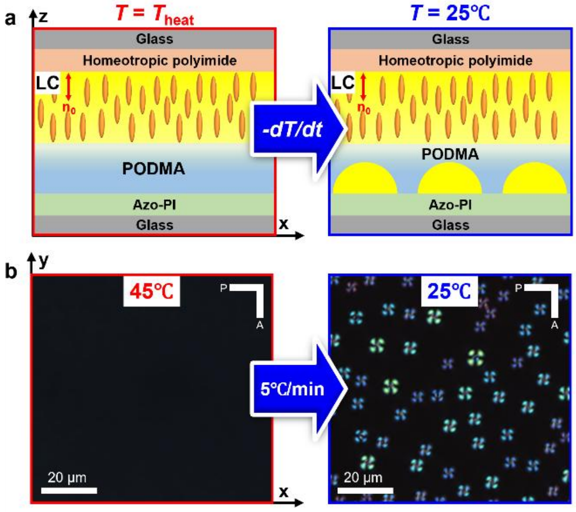

Fig. 1 (a) Thermally driven dissolution of nematic LC into the PODMA matrix at the peak annealing temperature (T_{heat}) followed by phase separation during cooling sequence ($-dT/dt$), yielding hemispherical droplets at the PODMA/Azo-PI interface; schematic cross-section. **(b)** Polarized optical micrographs before (45°C) and after cooling at 5°C/min to 25°C; scale bars 20 µm.

Poly(octadecyl methacrylate) (PODMA) served as the rubbery host polymer. The nematic LC mixture E7 was used as the guest material, which easily dissolves into PODMA layer when heated below its nematic-isotropic transition temperature (T_{NI}). A photo-switchable azobenzene polyimide (Azo-PI) provided nucleation selectivity, and indium-tin-oxide (ITO)-coated glass served as transparent electrodes.

ITO substrates were spin-coated with a 50 nm film of Azo-PI and soft-baked at 120°C for 10 min. A PODMA overlayer was spin-coated and capped with a second ITO slide bearing a homeotropic polyimide. The cell was then filled with E7, heated to 35-55°C ($<T_{\mathrm{NI}}$) for 2 min to dissolve the LC, and subsequently cooled at 3–20°C/min, nucleating hemispherical droplets at the PODMA/Azo-PI interface. Patterned 450 nm light (30-180 s) locally cis-isomerized the Azo-PI, lowering adhesion so that droplets formed only in illuminated pixels. When required, a 0.8-1.2 V/μm electric field applied during cooling preset the internal director orientation (Fig. 1a, b).

3 Results & Discussion

To establish a deterministic, large-area platform for optically active microdroplets, we first quantified how simple thermal parameters govern droplet nucleation and growth. Heating the E7 / PODMA bilayer to $T_{\mathrm{heat}} < T_{\mathrm{NI}}$ dissolved the LC uniformly in the rubbery matrix. As T_{heat} increased from 35 to 55°C, the solubility of E7 in the PODMA matrix rose, loading more LC into the film. This higher LC content subsequently produced both a greater number of nuclei and larger droplet diameters during cooling. In contrast, the subsequent linear cooling rate dictated the final size-density trade off: fast ramps (20°C /min) yielded arrays of ~4 μm droplets at $>1 \times 10^5$ cm^{-2}, whereas slow ramps (3°C/min) produced ~25 μm droplets at one order of magnitude lower density. These two global parameters thus cover three decades of areal density with <5 % size dispersion.

The dependence of droplet size and density on the thermal parameters can be rationalized within a scaling framework based on classical nucleation and diffusion-limited growth. Increasing the maximum annealing temperature increases the solubility of the LC in the PODMA matrix, resulting in a higher number of nucleation sites and larger average radii. Conversely, faster cooling suppresses growth while enhancing nucleation, thereby yielding smaller droplets at higher densities. This trade-off follows a predictable scaling relation, in which the number density varies nearly linearly with the dissolved LC fraction, whereas the droplet radius exhibits a square-root dependence. Such agreement with theoretical expectations highlights the quantitative controllability of our method and confirms that the droplet arrays can be engineered across orders of magnitude in areal density without sacrificing size uniformity.

Spatial selectivity was introduced by photo-patterning the Azo-PI under 365 nm light before cooling. Illumination induces cis-isomerization of Azo-PI, reducing the local work of adhesion and confining nucleation to user defined pixels without etching or microfluidics. Binary images, grayscale gradients, and even stochastic codes were reproduced across large centimeter-scale substrates. Re-exposure to 450 nm light drives cis-to-trans back-isomerization of Azo-PI, restores the local work of adhesion, erases the initial droplet pattern, and enables subsequent irradiation to rewrite new arrays, thereby confirming fully reprogrammable surface chemistry.

Beyond global thermal control, the integration of photo-responsive surfaces enables a richer degree of programmability. High-intensity illumination selectively suppresses nucleation, allowing binary on–off definition of droplet positions, whereas lower intensities modulate droplet size continuously, creating gradients of structural color across the surface. This multi-level control translates directly into optical functionalities: discrete pixel arrays act as reconfigurable microlenses, while continuous size gradients generate broad spectral coverage for hyperspectral filters. Furthermore, the dynamic electro-optic response of each droplet, combined with the rewritability of the photo-switchable interface, offers a unique route to multifunctional devices where permanent microlens arrays, erasable security codes, and adaptive optical elements can all be realized within a single platform.

Each hemispherical droplet behaves as a dielectric Mie resonator. The refractive index contrast between E7 and PODMA supports size-dependent multipolar scattering modes that yield bright structural colors whose peak wavelength scales with droplet radius. Applying an in-plane electric field of 0.8–1.2 V/μm reorients the radial director into a vertical configuration within milliseconds, altering the effective refractive index and shifting the resonance peak, thereby demonstrating rapid, reversible color patterning and tuning.

In summary, thermally driven phase separation, manipulated by light-switchable adhesion, yields ordered yet reprogrammable LC microdroplet arrays whose index-tunable microlenses and size-dependent Mie resonances can be set simply by T_{heat} and $-dT/dt$. This facile platform offers direct routes to spectrally agile optics for all-weather LiDAR and radar receivers, ultrathin multi-focus camera modules, and compact hyperspectral or biosensing elements, underscoring its promise for defense, mobility, imaging, and healthcare applications. Moreover, the intrinsic reconfigurability and rewritability of these droplet arrays provide unique opportunities for security-oriented applications, such as anti-counterfeiting labels and physically unclonable functions (PUFs), which complement their utility in adaptive optics and imaging.

4 Conclusions

We have demonstrated a facile route to pattern hemispherical liquid crystal microdroplets as reprogrammable arrays. Tuning only the peak annealing temperature (T_{heat}) and cooling rate ($-\mathrm{d}T/\mathrm{d}t$) controls nucleation, growth, and areal density over three orders of magnitude while maintaining $< 5\%$ size dispersion. Selective cis/trans photo-isomerization of an azobenzene polyimide adds pixel-level addressability and erasable patterning, yielding droplet lattices that can be rewritten.

Each droplet operates as an electrically tunable Mie resonator whose scattered color encodes its radius; a vertical electric field switches the director within milliseconds, enabling rapid spectral modulation across the visible range. The combination of deterministic geometry, adaptive optics, and single-step reusability positions this platform for immediate translation into index-driven microlenses for all-weather LiDAR and radar, ultrathin multi-focus camera modules, and compact hyperspectral or biosensing elements. Future work will extend the concept to other polymers and LCs, paving the way for wavelength-agile photonics.

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