Applications of Droplet Microsystems in Optics and Photonics

Subjects: Optics

Contributor: Jin-Kun Guo, W.D.N. Sandaruwan, Jinwei Li, Jinzhong Ling, Ying Yuan, Xin Liu, Qiang Li, Xiaorui Wang

The micro- and nano-machining techniques applied to solid materials have yielded remarkable success in the semiconductor industry by integrating complex functionalities into microscale devices, thus spearheading the modern electronics revolution. Extending similar miniaturization strategies to process and assemble soft matter for creating multileveled functional structures over various length scales presents significant scientific and practical potential. Soft matter, including liquid crystals (LC), colloids, polymers, and biological substances, exhibits widespread influence across nature, living organisms, daily life, and industry. The biomimetic properties, responsiveness to stimuli, and efficacy in controlled release and sensing make soft matter extensively applicable in biology and chemistry.

Keywords: droplet ; liquid crystal ; soft matter ; microfluidics ; laser micro–nano-machining ; 3D printing ; emulsions ; laser injection ; microfabrication ; lab on a chip

1. Introduction

The micro- and nano-machining techniques applied to solid materials have yielded remarkable success in the semiconductor industry by integrating complex functionalities into microscale devices, thus spearheading the modern electronics revolution [1]. Extending similar miniaturization strategies to process and assemble soft matter for creating multileveled functional structures over various length scales presents significant scientific and practical potential [2][3][4]. Soft matter, including liquid crystals (LC), colloids, polymers, and biological substances, exhibits widespread influence across nature, living organisms, daily life, and industry [2][5][6]. The biomimetic properties, responsiveness to stimuli, and efficacy in controlled release and sensing make soft matter extensively applicable in biology and chemistry [7][8][9][10][11][12][13][14][15][16][17][18][19]. Additionally, soft matter offers distinct advantages over solids in optical and photonic applications owing to its inherent adaptability, tunability, and seamless integration capabilities [3][20][21][22][23][24][25]. These unique properties open avenues for pioneering optical designs, adaptive systems, and versatile devices capable of dynamic responses to changing environmental or operational conditions.

Soft matter in mesoscale is profoundly influenced by surface tension, resulting in the spontaneous formation of curved geometries such as spheres or domes when dispersed in other immiscible liquids or air [4]. The curved confinement that encloses soft matter into separate functional units facilitates miniaturization but also poses a barrier to its micro- and nano-machining. However, the three-dimensional (3D) microstructures and the distribution of material composition within the closed confinement are crucial to its optical behavior, playing a pivotal role in the development of applications in optics and photonics. These factors govern the interaction of light, influencing properties such as refraction, reflection, scattering, and the ability to function as optical elements [26]. Therefore, precise micro- and nanofabrication of soft-matter droplets is essential to advance their development and explore their potential.

2. Typical Droplet Structure of Soft Matter

An emulsion droplet is a non-homogeneous liquid structure in soft matter, where one liquid is dispersed in another immiscible liquid in the form of tiny droplets [10]. As illustrated in **Figure 1**, droplets can be categorized based on their hierarchical structure into single emulsions, double emulsions, and multiple emulsions. The simplest form is a single emulsion droplet, for example, water-in-oil or oil-in-water. In cases where more than two phases are involved in emulsion, the dispersed droplets may contain multiple compartments. When two compartments are brought into contact, they can form structures such as Janus, Snowman, and core–shell, determined by the balance of interfacial tensions [27]. The number of inner compartments can exceed one, resulting in a multicore structure. These compartments can either be freely suspended or self-assemble into ordered substructures with the help of their surrounding liquid (**Figure 1**b) [10][28].

Multiple emulsions in soft matter are complex polydisperse systems where both oil in water and water in oil emulsion exist simultaneously (**Figure 1**c).

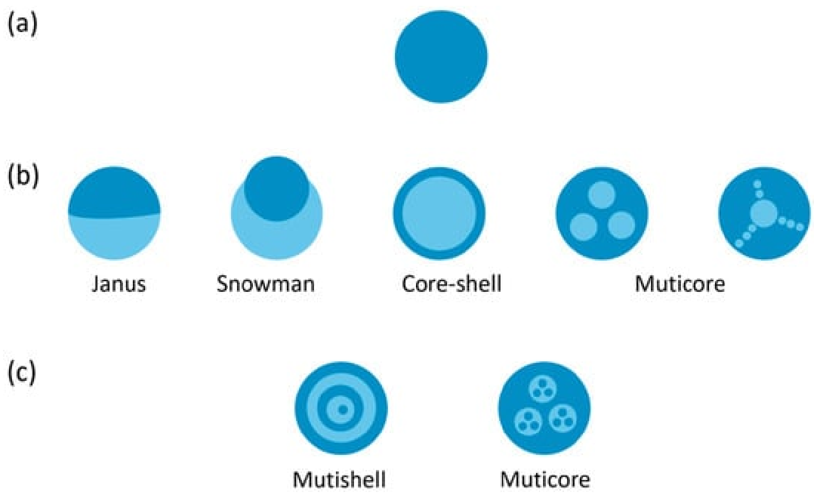

Figure 1. Distinct configuration of single emulsion droplet (**a**), double emulsion droplet (**b**), and multiple emulsion droplet (**c**).

2. Applications of Droplet Microsystems in Optics and Photonics

2.1. Droplet Lasers

The microlaser based on droplets stands out as a promising technology, with its distinctive feature of flexibility and compactness [4][29][30]. The spherical shape of the droplet makes it an ideal candidate to act as a resonance cavity. By incorporating the gain media into the cavity, droplets enable controllable lasering under pump radiation [26]. When employing LC as the active medium, the system acquires dynamic and tunable optical properties, thereby enhancing the versatility and adaptability of the microlaser (**Figure 2**a) [31]. LC molecules in cholesteric phase are noteworthy for their spontaneously self-assemble into periodic helical structures exhibiting a photonic bandgap [32].

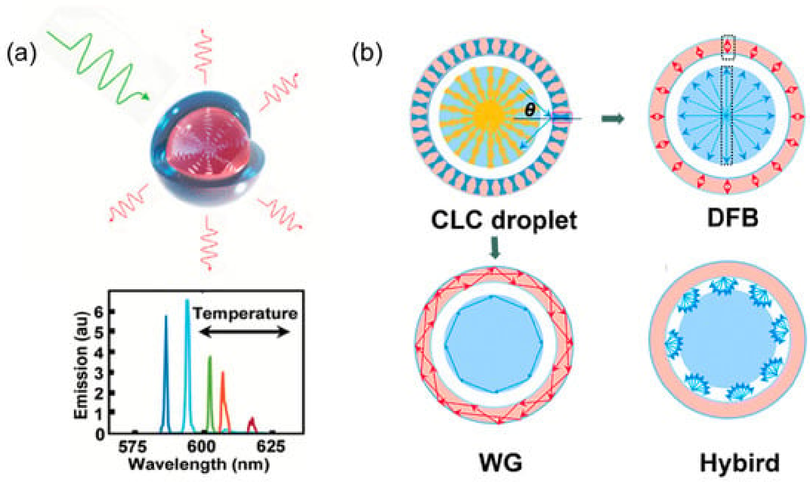

Figure 2. (**a**) Schematics for dye-doped CLC microdroplet laser with internal radial helix director profile. Temperature tunable lasing, indicated by changes in color, occurs via changes in LC molecular helical pitch. (**b**) Cross profile of a CLC triple emulsion droplet. Ray optic schematics for optical resonances: shell and core DFB resonances, TIR-based WG resonances in shell and core, hybrid resonance with optical reflections on Bragg shell. Adapted from Refs. [31][32] with permission from American Chemical Society and Royal Society of Chemistry, respectively.

Since the first discovery of CLC droplet lasers by Musevic et al., there has been substantial progress in developing this system as a tunable wavelength lasing source [33]. The primary focus has been on exploring new cavity design and fabrication approaches to integrate one or more gain media with geometry-dependent modes (**Figure 2**b). Three types of laser resonators can be obtained by altering the droplet structure, such as droplet, core–shell, and multishells, as well as by incorporating gain dyes and CLCs in different cavities. First, once confined CLC within a sphere or a shell with parallel anchoring, LC molecules arrange themselves into a Bragg-onion structure, resulting in laser emitting in band-edge mode (also called distributed feedback or DFB mode) [32]. Second, the spherical cavity contributes to forming Whispery Gallery (WG)-mode lasing based on the light total internal reflections on the boundary of the core and shell [32][34]. Third, once

CLC is confined in the outer shell and fulfills certain conditions, lasing in distributed Bragg reflection (DBR) mode can be achieved [30]. By controlling the spatial coupling between the pump beam and droplet, different modes or wavelengths can be selectively excited [29].

2.2. Waveguide

An optical waveguide is a physical structure designed to confine light within its boundaries, ensuring efficient transmission through mechanisms like total internal reflection and evanescent coupling. This design offers precise control over the trajectory of light, guiding it along specific paths. Notably, droplets can serve as transduction units for light, as demonstrated by Timothy M. Swager et al., who developed optical waveguide-based sensors utilizing complex emulsion droplets [35][36][37]. These emulsion droplets consist of two immiscible liquids with distinct refractive indices, serving as a transduction unit. A unique feature of these emulsion droplets is their ability to undergo changes in geometry or orientation triggered by specific chemical or biological stimuli. This dynamic property allows for the tunability of the refractive index in emulsion droplets, facilitating the manipulation of light out-coupling (**Figure 3**a).

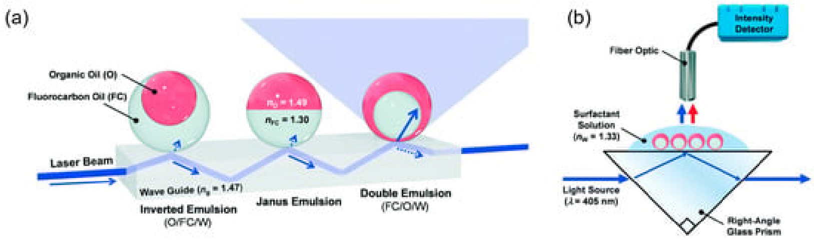

Figure 3. (**a**) Conceptual sketch of the modular waveguide comprising dynamic complex emulsions in three different geometries. Beginning on the left, total internal reflection (TIR) is observed for inverted and Janus emulsions. Upon transition to a double emulsion with the higher index organic phase on the outside, the laser light is out-coupled from the waveguide, thereby increasing light intensity measured above the waveguide. (**b**) Waveguide-based sensing device and optical read-out of changes in droplet geometry. Adapted with from ref. [37] under the terms of Creative Commons attribution 3.0 unported license.

As illustrated in **Figure 3**b, light propagating through a glass waveguide will undergo total internal reflection at the interface when in contact with low refractive index media. However, proximity to a higher refractive index medium can lead to light out-coupling from the waveguide. Researchers can detect and quantify stimuli components by analyzing changes in transmission intensity or scattered light patterns caused by structural changes in droplet droplets. The precise control of emulsion droplets over translation, rotation, and assembly has been extensively demonstrated by using magnetic nanoparticles. These could further enhance the behavior of emulsion droplets as compact, near-real-time waveguides. The dynamic nature of droplets allows for the creation of reconfigurable waveguides, offering adaptability in routing light signals.

2.3. Microlens

Droplets, acting as microscale lenses, harness the optical phenomenon of focusing light through their curved surfaces [38]. This natural effect is akin to the magnification observed when viewing objects through water droplets or the controlled focusing of light within microfluidic systems. A noteworthy application of droplet-based optics is found in liquid microlenses, offering tunable focal lengths achieved through the controlled deformation of liquid droplets. These unique lenses are crafted by combining two immiscible liquids, hydrocarbon, and fluorocarbon, creating bi-phase emulsion droplets. The capability to alter the droplet's geometry allows for the regulation of whether it concentrates or scatters incoming light, providing versatile optical functionalities (**Figure 4**a) [39]. Depending on the setup, the lenses can either provide focused images of objects, as regular lenses do, or magnified virtual images. The geometry adjustment of the droplet can be achieved through external stimuli such as sound waves or electric fields, enabling on-the-fly modifications to the focal length.

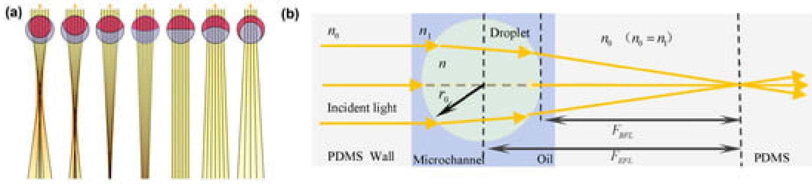

Figure 4. (**a**) Double emulsion droplets with distinct geometries can focus or diverge the light rays. Adapted from Ref. [39] under the terms of Creative Commons CC BY license. (**b**) Light-focusing diagram of a single-droplet microlens within

optofluidic microlens arrays. Adapted from Ref. [40] with permission from American Chemical Society.

In a parallel development, researchers have introduced innovative optofluidic microlens arrays (MLAs) based on water-in-oil droplets featuring a controlled droplet diameter of 200 μm (**Figure 4**b) [40]. These MLAs can be seamlessly self-assembled in a microfluidic chip, and their refractive index can be precisely tuned by adjusting flow rates. This fine-tuning results in an impressive range of focal lengths, spanning from 550 to 5730 μm. Suspended within immiscible liquids, these liquid droplets not only control focal length but also offer high tunability, rapid response times, and a compact, lightweight nature. In contrast to electrowetting lenses, this technology is cost-effective and continually evolving. The MLAs, with their easy integration, find applications in diverse fields, including 3D imaging, stereoscopic analysis, biomedical sensing, and display technology [40][41].

2.4. Display and Information Tags

Droplets exhibit versatility as pixel units in visual presentations, evolving into active or passive displays, as well as information tags. One technological strategy entails utilizing droplets with structural color as photonic pigments, creating patterns for applications such as information display, encryption, and anti-counterfeiting [11][42][43][44]. This phenomenon originates from the interaction between the internal structures of droplets and light, providing advantages such as fade resistance, eco-friendliness, iridescence, high saturation, and intelligent stimulus response compared to methods involving chemical dyes. Diverse technological principles contribute to the formation of structural color in droplets, including the self-assembly of periodic structures using colloids and cholesteric LC within droplets and the creation of specific patterns through photonic cross-communications between droplets in an array (**Figure 5**a) [45][46][47][48][49].

Research has revealed that structural color can be achieved based on total internal reflection and interference at the microscale concave interface within double emulsion droplets with snowman-like and core–shell structures (**Figure 5**b) [50][51][52]. The adjustment of shell thickness and the eccentricity of core–shell structures enable the tuning of structural colors. In addition to structural color, another method involves incorporating dye into the same droplets to impart fluorescent color. By organizing these droplets in an ordered arrangement to form specific patterns, they can display different information under varying conditions, thereby enhancing information security (**Figure 5**c) [53][54][55].

Droplets can also serve as encapsulation units containing colored particles in their cavity. By using electric or magnetic fields to drive the arrangement of particles within the droplets, it becomes possible to adjust light transmittance, achieving information display and switching [56][57]. This approach to reflective displays has advantages such as energy efficiency, a wide field of view, and a relatively fast switching time (~0.14 s). Additionally, it enables the development of displays that can be curved or flexible.

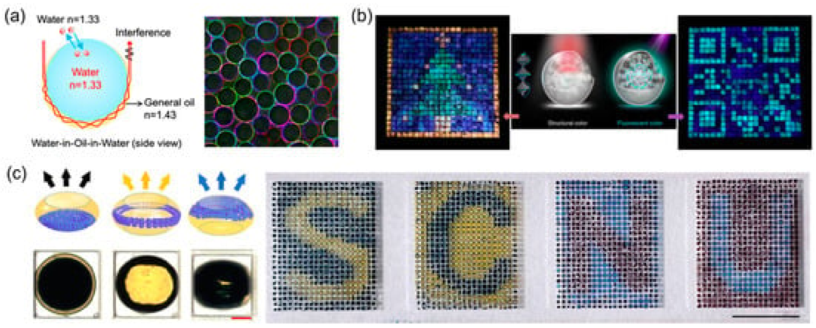

Figure 5. (**a**) Structural coloration in thin-shell emulsion droplets. Reflection optical micrographs of various structural colors in emulsion droplets with different sizes. Adapted from Ref. [50] with permission from American Chemical Society. (**b**) Geminate labels are formed by programming fluorescent CLC droplets, displaying QR code and Christmas tree under different conditions. Adapted from Ref. [53] under the terms of Creative Commons CC BY license. (**c**) A reflective display based on the electro-microfluidic assembly of particles. Adapted from Ref. [57] under the terms of Creative Commons CC BY license.

2.5. Other Applications

Exploring droplet-based sensor development constitutes a noteworthy research avenue. The standard technical strategy involves employing specially designed stimulus-responsive polymers to solidify periodic structures within the droplets. Consequently, the variations in the structural color of these droplets can be harnessed for detecting parameters like temperature, pressure, pH, specific components in solutions (e.g., metal ions, proteins, DNA), and even volatile gases, among others [58][59][60][61][62][63][64][65][66][67]. Such applications necessitate the utilization of smart materials, such as polymers or surfactants, to accomplish the mentioned functions. This characteristic renders this field a focal point in both chemistry and biology.

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