

3D microlasers from self-assembled cholesteric liquid-crystal microdroplets

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Abstract: We demonstrate a tunable and omnidirectional microlaser in the form of a microdroplet of a dye-doped, cholesteric liquid crystal in a carrier fluid. The cholesteric forms a Bragg-onion optical microcavity and the omnidirectional 3D lasing is due to the stimulated emission of light from the dye molecules in the liquid crystal. The lasing wavelength depends solely on the natural helical period of the cholesteric and can be tuned by varying the temperature. Millions of microlasers can be formed simply by mixing a liquid crystal, a laser dye and a carrier fluid, thus providing microlasers for soft-matter photonic devices.

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1. Introduction

Here we demonstrate, for the first time, full 3D tunable lasing in laser dye-doped, cholesteric liquid-crystal (CLC) microdroplets, 15-50 μm in diameter, embedded in an isotropic carrier fluid. These droplets are spontaneously self-assembled in a fraction of a second by dispersing a small amount of the cholesteric liquid crystal in a non-miscible fluid, such as glycerol. Due to the chirality of the CLC, a strong modulation of the refractive index is obtained in the radial direction, thus forming a radial Bragg (onion) resonator [1]. As a result of the symmetry, such a structure exhibits a photonic band gap (PBG) for an arbitrary value of the index contrast between the layers and the omnidirectional lasing from the CLC microdroplets is achieved by pulsed optical pumping.

To realize a 3D Bragg cavity we employed the soft-matter approach, using cholesteric liquid crystals [2] as a material for the spontaneous formation of a spherical microcavity with radial modulation of the refractive index. For non-miscible, soft-matter materials, a spherical shape

appears naturally because of the surface tension, which tends to reduce the amount of surface for a given volume of material. On the other hand, chiral nematic liquid crystals provide a natural periodic modulation of the refractive index, with a birefringence of the order of ~ 0.1 – 0.2 . In a cholesteric liquid crystal, rod-like molecules are locally oriented with their axis parallel to each other in a direction called the director [2]. Due to the molecular chirality, the director spontaneously twists in a direction perpendicular to the long molecular axes. The periodicity of this helical and birefringent structure (the pitch), depends on the molecular chirality and is in the range from ~ 100 nm to several micrometers. Under white-light illumination, cholesteric liquid crystals are often characterized by the fascinating iridescent colors of the reflected light. Because of their helical modulation and birefringence, these CLCs exhibit a 1D photonic band-gap for light, traveling along the helical axis of the CLC, if it is circularly polarized and of the same handedness as the CLC helix. For circular polarization of the opposite handedness, the light is transmitted.

The first observations of band-edge lasing in CLCs were independently demonstrated by Kopp et al. [3] and Taheri et al. [4, 5]. Since then, it has been shown in a number of experiments on dye-doped cholesteric liquid crystals that a low-threshold mirrorless lasing can be achieved at the photonic band edge of a laser dye-doped CLC, when it is optically pumped with pulsed laser light [6–9]. A recent review of this subject can be found in Ref. [10]. The helical birefringent planar structure of the cholesteric liquid crystal acts as a mirrorless 1D microcavity and therefore defines the direction of the lasing. The planar cholesteric microlaser therefore radiates light in a preferential direction, and this can be exploited in laser LC display devices [10]. We have further developed the idea of mirrorless lasing in a thin planar CLC by considering the geometry, where the cholesteric layers are wrapped into a microsphere. In this case, the helix is now originating from the center of the microsphere in all possible radial directions, as shown in Fig. 1. In this geometry, there should be a PBG for the light going out in any direction, and therefore the lasing should also be in all directions.

The development of low-threshold microlasers has attracted increasing interest in recent years, not only because of the fundamental questions of spontaneous emission and photon localization in artificially created structures, but also for potential commercial applications in integrated photonics. Narrow-linewidth, ultra-small mode volume and tunability are highly desirable attributes for these novel laser sources. Of particular interest are lasers based on optical microcavities, which confine the light into a small volume by either total internal reflection at the microresonator's interfaces or by Bragg reflection from a periodic dielectric structure. So far, lasing has been achieved in a number of different dielectric microstructures, such as photonic crystal defect microcavities [11], micropillar cavities [12], whispering-gallery microcavities [13], a 3D photonic crystal of the liquid-crystal blue phase II [14], and has been combined with plasmon resonances in nanosized laser sources [15, 16].

Spherical Bragg-reflector microcavities, based on an alternating series of low- and high-refractive-index material concentric shells, are particularly appealing because of their perfect rotational symmetry in 3D, so the photonic band gap (PBG) is expected to be independent of the direction of light propagation and the light is strongly confined in all directions [1, 17–19]. A number of Bragg-onion resonator structures have been realized in 2D using standard planar lithography and the lasing has been demonstrated [20–22]. The natural step now would be to go into three dimensions. However, unfortunately, it is quite difficult to manufacture any 3D solid-state microcavity [23]. There have been several attempts to produce 3D Bragg-onion resonators by chemical synthesis [24] or by combining etching and chemical vapor deposition [19], but to our knowledge lasing has not been achieved in these solid-state structures.

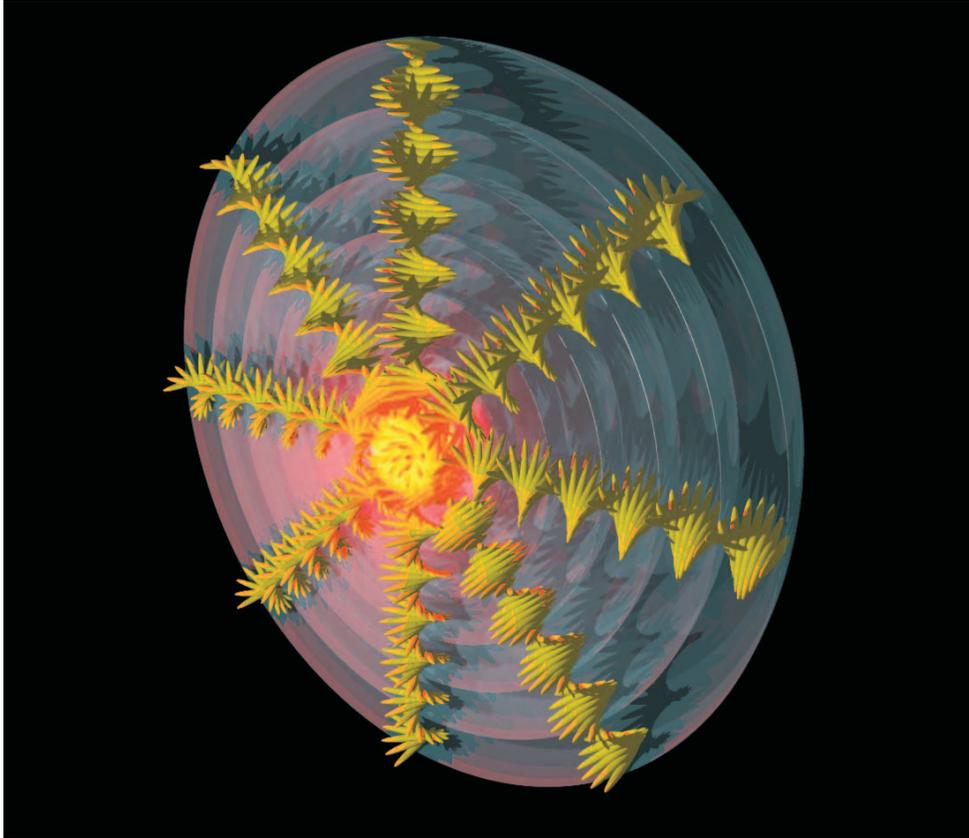


Fig. 1. The schematic view of the arrangement of CLC molecules in a cholesteric microdroplet with parallel anchoring of the LC molecules at the surface. The helical structure of the liquid crystal originates from the center of the droplet and gives rise to concentric shells of constant refractive index. This dielectric structure is optically equivalent to the well-known Bragg-onion optical microcavity.

2. Experimental setup

Two different dye-doped cholesteric liquid-crystal mixtures were used as the active medium. The low-birefringence mixture that was used in most of the experiments was prepared using the MLC-7023 liquid crystal ($n_e = 1.53$, $n_o = 1.46$; Merck) with 25.5wt% S-811 chiral dopant and 0.2wt% fluorescent dye 7-diethylamino-3,4-benzophenoxazine-2-one (Nile red, Sigma-Aldrich). The high-birefringence mixture was prepared using the MLC-2132 liquid crystal ($n_e = 1.77$, $n_o = 1.51$; Merck) with 26wt% S-811 chiral dopant (Merck) and 0.5wt% laser dye 4-dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM, Exciton). In both cases, the concentration of chiral dopant was chosen so that the longer wavelength edge of the cholesteric PBG overlaps with the emission maximum of the dye used. The position of the PBG of each mixture was determined by measuring the spectrum of white light reflected from the 30- μm -thick planar cell containing the mixture.

A small quantity, typically a few percent, of dye-doped cholesteric liquid crystal was mechanically mixed with glycerol forming small droplets of different sizes. No special mixing protocols were used, as the dispersion readily forms, even when mixing it manually. Glycerol

was used because it imposes parallel anchoring of the CLC molecules at the interface and at the same time it has a refractive index that is high enough to suppress the whispering-gallery modes in the case that a low-index CLC was used. The mixture of dye-doped cholesteric liquid-crystal droplets in glycerol was introduced into a 150- μm -thick glass cell using capillary force, and the cell was sealed with Torr Seal glue. The glass cell was made of two glass slides of 1 mm thickness, and the cell-gap was controlled by 150 μm spacers. A typical cholesteric droplet with a reduced concentration of the chiral dopant (thus having its helical period in the micrometer range, easily seen under the microscope) is shown in Fig. 2(a) and 2b. One can clearly see the alternating light-dark concentric shells, which are due to the radial modulation of the index of refraction. The radial period corresponds to one half of the cholesteric pitch. From these images we can deduce the internal structure of the droplet, which is the same as the one anticipated and shown in Fig. 1. The structure of the droplet is known to be of the spherulite-type with a radial defect, which is a $s = 2$ disclination line going from the center of the droplet to the surface [25,26]. Such a $s = 2$ disclination line defect has been observed in many experiments on CLC droplets [27–29], although a $s = 1$ diametrical defect structure, where the defect line runs diametrically from one surface to the other has also been reported [27, 30]. The reason for the appearance of the radial $s = 2$ or diametral $s = 1$ defect line is not well understood. Nearly all of the droplets smaller than approximately 100 μm had this perfect spherulite structure, without any additional defects or imperfections. A single CLC microdroplet was observed under an optical microscope (Fig. 2(d-f)) and illuminated by a pulsed laser. The pumping laser was focused through a 20x objective to a waist of $\sim 100 \mu\text{m}$, thus illuminating the selected droplet. An actively Q-switched doubled Nd:YAG laser with a pulse length of 1 ns, a repetition rate of 200 Hz and a maximum pulse energy of 10 μJ was used as the excitation source (Alphaslas, Pulselas-A-1064-500).

3. Results and discussion

When increasing the pumping pulse energy, at one point, a bright red laser spot suddenly appeared in the center of the droplet, as shown in Fig. 2(e). This is obviously the threshold for lasing and by further increasing the pump-light intensity, the radiating light becomes very strong. In the spectrum of the light, emitted from the center of the droplet, we reveal a single and sharp spectral line, positioned at $\sim 600 \text{ nm}$. The threshold power for 3D lasing of the CLC microdroplets is determined by measuring the intensity of the emitted spectral line as a function of the excitation energy (Fig. 3(a)). We observed a typical threshold behavior (Fig. 3(b)) with a threshold energy of 20 nJ for a 1 ns pumping pulse, focused to uniformly illuminate a 40 μm diameter droplet. The linewidth of this laser line is typically $\sim 0.1 \text{ nm}$, as shown in Fig. 3(c). This is comparable to the width of the laser lines in thin planar cholesteric layers and the liquid-crystal blue phase II [14]. In our case, the Q-factor for smaller droplets is limited by the number of layers, and the threshold for lasing increases with a decreasing microdroplet diameter. As a result, the threshold intensity depends on the diameter of the microdroplet (Fig. 3(d)) and the smallest droplets made of high-birefringence cholesteric liquid crystal that were still lasing were 15 μm in diameter. Nearly all the droplets larger than this diameter emitted laser light when excited. We measured the average power of a CLC Bragg laser to be up to 0.05 mW at a 200 Hz repetition rate. For bigger droplets, strong thermal orientational fluctuations of the CLC are probably the limiting factor for obtaining narrower linewidths. We can observe these low-frequency orientational fluctuations, even with the naked eye as, a characteristic "flickering" of the light intensity, emitted from the resonator's center. We observed that when the acquisition time of our spectrophotometer is set below $\sim 50 \text{ ms}$, the linewidth of the laser light is below the resolution of the spectrophotometer (0.05 nm), and the position of the laser line clearly fluctuates with time.

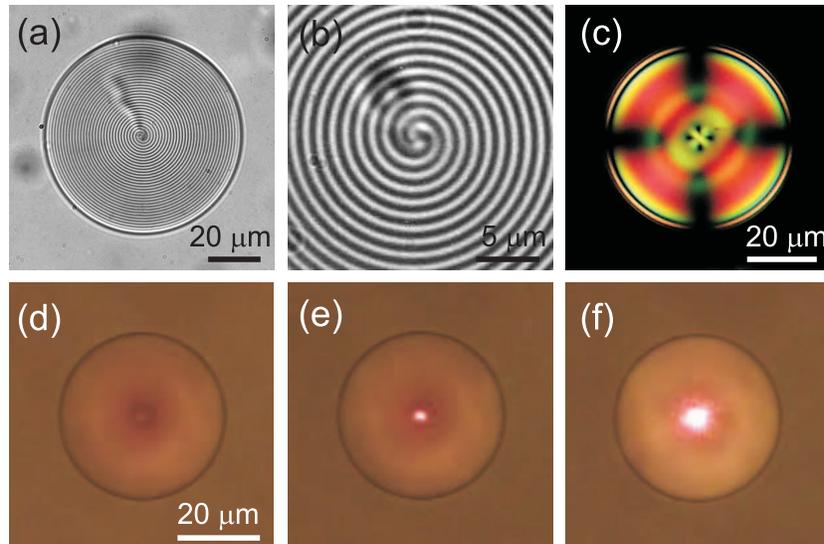


Fig. 2. (a) A typical cholesteric droplet with a pitch $p = 2.2 \mu\text{m}$ in glycerol. The light and dark concentric shells are due to the spatial variation of the refractive index of the cholesteric liquid crystal in the radial direction. (b) Close up of the center of the cholesteric droplet, when viewing in the direction parallel to the disclination line. (c) Cholesteric droplet with PBG in the visible range of light, viewed under crossed polarizers and white-light illumination. (d-f) (Media 1) Omnidirectional (3D) lasing in a cholesteric droplet illuminated by laser pulses ($\lambda = 532 \text{ nm}$) and a weak white background illumination. (d) Below the lasing threshold (1.6 mJ/cm^2) the droplet is fluorescing uniformly. (e) Just at the threshold for lasing (1.9 mJ/cm^2), a bright spot of radiating monochromatic light can be observed in the center of the droplet. (f) Lasing becomes very intense at a high pump power (12 mJ/cm^2).

The position of the laser line, emitted from CLC microdroplet, was compared to the reflection spectra, as measured on thin planar layers of the same material. This is shown in Fig. 4, and we see that the lasing from the CLC microdroplets occurs at the edge of the photonic band gap of the planar CLC material, indicating a typical band-edge lasing. The lasing mechanism for our CLC microdroplets is therefore the same as in planar CLC lasers that do not contain a defect.

Polarization of the emitted light from the CLC microlasers was also investigated. A single droplet was observed using a low numerical aperture objective and a CCD camera with an exposure time of 1 s. We used a circularly polarized laser beam to optically pump the droplet. A polarizer was inserted between the sample and the objective, and the collected intensity was measured as the polarizer was rotated. We found that the intensity did not depend on the orientation of the polarizer, meaning that the emitted light is not strongly linearly polarized. In the next experiment, a quarter-wave plate was inserted in between the sample and the polarizer and the intensity was again measured as the polarizer was rotated. If the light were to be circularly polarized, the quarter-wave plate would turn it into linear polarization, which would be detected by the polarizer rotation. However, we again did not detect any intensity modulation with the polarizer rotation. From both experiments we can conclude that in the time window of our experiment (i.e., 1 s exposure time) the light is neither linearly nor circularly polarized. We did not measure in detail the polarization of the light emitted along the defect line. In all the experiments the defect line was oriented randomly.

Lasers are characterized by their highly directional emission, but in some special cases, such

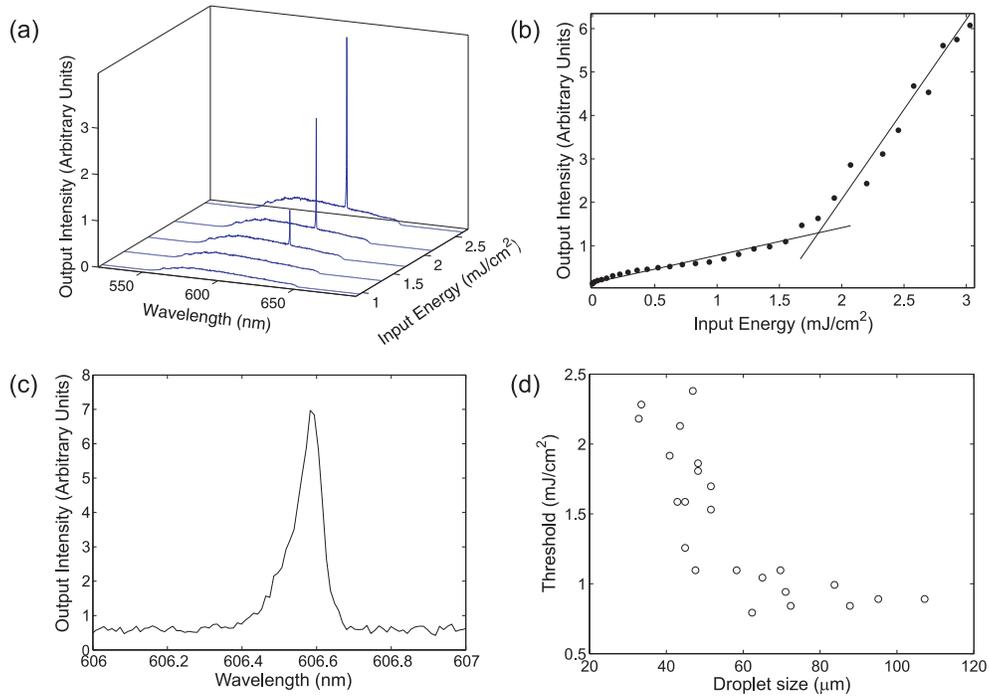


Fig. 3. Lasing characteristics of a single droplet of dye-doped CLC. (a) The spectra of light emitted from the center of the CLC microdroplet at different energies of the pumping pulse. (b) The radiated laser-light intensity as a function of the input-pulse energy density. The threshold for lasing is clearly seen at ~ 1.8 mJ/cm². (c) Magnified lasing spectrum showing a laser linewidth of ~ 0.10 nm. (d) The threshold for lasing as a function of the diameter of the CLC microdroplet. All the spectra were measured using an imaging spectrometer with a 0.05 nm resolution (Andor, Shamrock SR-500i) and cooled EM-CCD camera (Andor, Newton DU970N).

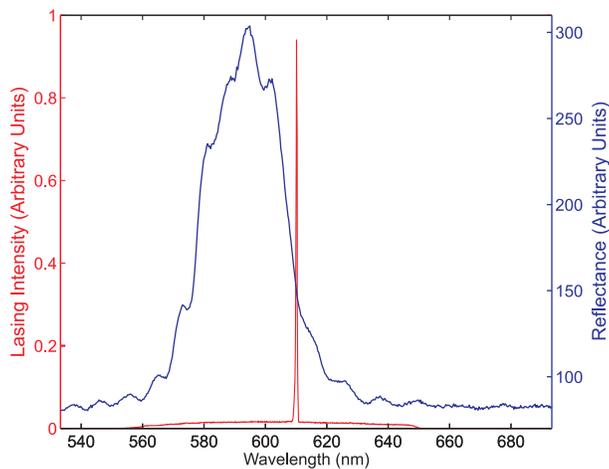


Fig. 4. Lasing spectrum of a single CLC droplet compared to the reflection spectrum of a 30 μ m planar cell filled with the same CLC mixture. The reflection spectrum was measured for light propagating along the helix of the CLC.

as holography, sensing or imaging, a point source of coherent light that emits light in all directions is a better choice or even necessary. The Bragg-onion laser with omnidirectional dielectric cladding is a natural candidate for such a source. We have therefore measured the angular dependence of the laser emission intensity and wavelength. This was done by introducing a glycerol dispersion of CLC microlaser droplets into a cylindrical glass tube. The angular dependence of the emitted light from a single droplet was measured by rotating the CCD camera, or an optical fiber guiding the collected light into the spectrometer, around the tube. The results shown in Fig. 5(a) clearly indicate that the intensity is highly uniform across the entire solid angle. The wavelength of the emitted light is also independent of the direction of the emission. We have not measured in detail the emitted intensity along the direction of the defect itself.

Furthermore, liquid crystals have large response to external stimuli, such as electric field and temperature. In an earlier work, we have shown that electrically tunable whispering-gallery-mode microcavities can be made from nematic liquid crystal droplets in a polymer [31]. The cholesteric onion laser is also highly tunable, as the cholesteric pitch usually depends on the temperature [32] and can also be set by the chiral dopant concentration. In a mixture of nematic LC and a chiral dopant at the right concentration, together with the proper dye, lasing from UV to IR has been achieved in planar cells of CLCs [33, 34], and it should also be possible to achieve this in CLC onion microdroplets. By changing the temperature we can, in our case, tune the emission by ~ 35 nm (Fig. 5(b)). The spectral shift is almost linear with temperature (3.5 nm/K) and completely reversible. Phototunability has also been demonstrated in cholesteric lasers [35] and should be interesting for application in phototunable spherical lasers.

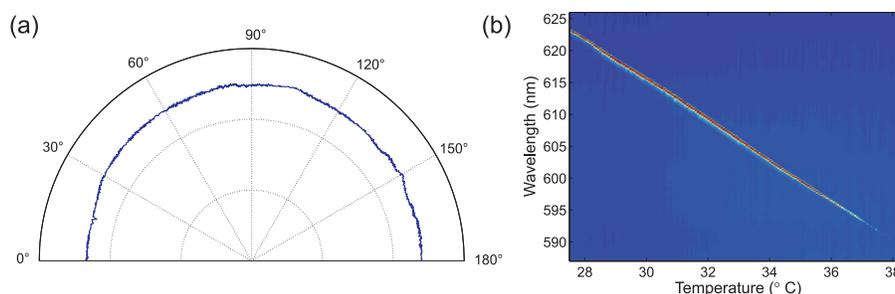


Fig. 5. (a) Lasing intensity from a single $50 \mu\text{m}$ CLC droplet as a function of the angle of rotation of the photodetector around the axis of the cylindrical tube, containing the microdroplets. (b) Lasing spectra as a function of temperature. At higher temperatures the laser line is shifted outside the optimum wavelength region of the dye used, so the laser emission ceases.

4. Conclusion

In conclusion, we have demonstrated 3D lasing from dye-doped cholesteric microdroplets with a Bragg-onion configuration of the refractive index. The lasing wavelength is determined solely by the cholesteric pitch. The laser light is emitted from the center of the CLC microdroplet in all directions, thus the laser is acting as a coherent, point-like, and omnidirectional source of light. Because of the temperature dependence of the helical pitch of the cholesteric, the microlaser wavelength is tunable by changing the temperature, the tuning range being several tens of nanometers. A number of applications of the cholesteric onion microlasers is anticipated, such as holography, telecommunications, optical computing, imaging, sensing and even as a material for paints or light sources that emit coherent light in all directions. By coating the droplet with a protective shell or by polymerizing the liquid crystal itself, a more mechanically stable mi-

crolaser could be made, useful for example in biological imaging. Further studies could include a fluorescent/plasmon particle as an active core, nonlinear core material for second-harmonic generation, suppression of spontaneous emission and coupled regular arrays of thousands and even millions of CLC microlasers. We anticipate that by using better materials and optimized material parameters, such as the concentration of the chiral dopant and of the fluorescent dye, it will be possible to further reduce the size and increase the tunability and the functionality of the cholesteric onion lasers. The CLC onion microlasers could also be combined with optical fibre waveguides to collect radiating light into the waveguides. The proposed procedure of making a CLC onion microlaser by mechanical mixing is simple and straightforward and produces millions of microlasers in a fraction of a second.

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