Check for updates

View Article Online
View Journal

Soft Matter

Accepted Manuscript

This article can be cited before page numbers have been issued, to do this please use: G. Petriashvili, M. De Santo, R. Barberi, R. J. Hernandez and G. Cipparrone, *Soft Matter*, 2017, DOI: 10.1039/C7SM01068K.

This is an Accepted Manuscript, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this Accepted Manuscript with the edited and formatted Advance Article as soon as it is available.

You can find more information about Accepted Manuscripts in the author guidelines.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard Terms & Conditions and the ethical guidelines, outlined in our author and reviewer resource centre, still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this Accepted Manuscript or any consequences arising from the use of any information it contains.

ROYAL SOCIETY OF CHEMISTRY

rsc.li/soft-matter-journal

View Article Online
DOI: 10.1039/C7SM01068K

Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.

Soft Matter Accepted Manuscript

Soft Matter

ARTICLE

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

www.rsc.org/

Mixed emulsion of liquid crystals microresonators: towards white laser systems

Gia Petriashvili[a], Maria Penelope De Santo[b], Raul Josue Hernandez[c], Riccardo Barberi[b] and Gabriella Cipparrone[b]

Microdroplets systems have attracted great interest because of their wide range of applications, easiness in processing and handling, feasibility in developing miniaturized devices with high performances and large flexibility. In this study, a stable emulsion based on different dye-doped chiral liquid crystals droplets has been engineered in order to achieve simultaneous omnidirectional lasing at different wavelengths. To obtain the mixed emulsion of dye doped Bragg onion-type microresonators the twofold action, as surfactant and droplets stabilizer, of the polyvinyl alcohol dissolved in water has been exploited. Multi wavelengths lasing in all directions around the mixed emulsion is demonstrated. By water evaporation, a plastic sheet including different types of chiral droplets is also obtained, retaining all the emission characteristic of the precursor emulsion. A relevant feature is the large flexibility of the preparation method that enables an easy and full control of the lasing spectrum addressing towards white laser systems. However, the simplicity of the procedure based on a single-step process as well as the high stability of the mixed emulsion is a relevant result, envisaging strong potentiality for developing easy and friendly technologies useful in the field of identification, sensing, imaging, coating and lab on chip architectures.

Introduction

Microdroplets[1-4] are entities with enormous practical impact in several branches of science (as chemistry, biology, physics) and for industrial, environmental and biomedical applications (food foams and emulsions, cosmetics and paints, drug encapsulation and delivery, etc.). They represent a very promising high-throughput technology addressing small volume devices with high performances. Microdroplets can be, for example, natural self-contained microreactors with very low materials consumptions suitable to compartmentalize and to isolate reactants,[1] or microresonators with very high quality factor, providing interesting photonics applications.[2-4] With respect to this last aspect, isotropic and anisotropic fluids have been used with the aim to develop microdroplets based lasers systems.[6-14] Generally, lasing occurs in whispering gallery modes:[2,6-11] the drops are microcavities and the light emitted from a dye is confined and amplified within the droplets by total internal reflections. Several microfluidics droplets-based lasing systems have been investigated in the last decade demonstrating the high flexibility and great capabilities of these devices in developing lab on chip instruments for photonics, spectroscopy, sensing, etc..[2,7-9, 10,12]

In case of chiral anisotropic fluids, like cholesteric liquid crystals,[13] the supramolecular arrangement of the fluid within the droplets provides another interesting photonic device, that is a 3D microlaser,[14,15] based on a “Bragg onion type” microresonator.[16] Such device is constituted by an alternating sequence of concentric shells of low and high-refractive-index materials, and due to their perfect rotational symmetry in 3D, the photonic band gap is independent from the direction of light propagation. Microlasers can be obtained creating small droplets of chiral nematic doped with a fluorescent dye in an isotropic immiscible fluid like water or glycerol.[14,15,16] Simple stirring, shaking processes or microfluidic systems have been used to generate LC droplets emulsions.[14,15,17] Further, the use of surfactants has enabled to control the anchoring of the liquid crystal molecules at the interfaces. Such boundary condition plays, in fact, a relevant role in determining the internal supramolecular arrangement.[18] Due to the liquid crystals hydrophobicity, the droplets acquire a spherical shape in water, while the internal supramolecular helicoidal structures spontaneously self-organize in geometries that depend on the combination of chirality, elasticity, and interface properties.[18] The most favourite configuration, in case of planar orientation of the LC molecules at the interface and droplet size larger than the cholesteric pitch, is the one with radial configuration of the helix axes, where the cholesteric layers are bent in concentric spherical surfaces with a single defect in the centre, exhibiting spherically symmetric

a. *Institute of Cybernetics, Georgian Technical University, 0175 Tbilisi, Georgia.*
b. *Department of Physics and CNR-Nanotec, University of Calabria, 87036 Rende (CS), Italy.*
E-mail: gabriella.cipparrone@fis.unical.it.
c. *Nanotec-CNR, sede Cosenza, 87036 Rende (CS), Italy.*
† Footnotes relating to the title and/or authors should appear here.
Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/x0xx00000x

View Article Online
DOI: 10.1039/C7SM01068K

Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.

Soft Matter Accepted Manuscript

Bragg reflection.[13,15,18] Optically, such droplet structure is equivalent to concentric alternating shells of dielectrics with different refractive index (Bragg-onion type microresonators). 3D omnidirectional microlaser have been demonstrated exploiting chiral nematic microparticles.[15] The laser is a band edge type, the emitted wavelength is positioned at the band edge of the chiral material stop-band.[19] Based on the high sensitivity to external stimuli of the liquid crystals, wavelength tuning is one of the main features of such kind of lasers and several strategies have been investigated.[20] Further, the combination with polymeric materials has enabled to develop polymerized 3-D microlasers, acquiring a higher stability with a loss in tunability, and paintable dye-doped chiral nematic emulsions.[21,22,23]

Based on the materials and the procedures exploited to create LC droplets, up to now, to our knowledge, emulsions including simultaneously LC droplets with different dopants (fluorescent dyes) or dopants concentration (chiral agents) have been never reported in literature. In fact, polymerized cholesteric droplets or spherical shells are used in order to achieve mixed suspensions displaying multi-colored patterns.[24] Indeed, the possibility to create such kind of colloidal system based on mixed emulsions could introduce some advantages to develop flexible multicolour laser sources, as well as devices for sensing, coating and imaging, etc.; however, neither production nor stability is straightforward.

Polyvinyl alcohols (PVAs) are commonly used as surfactants for liquid crystals devices to promote parallel orientation of the liquid crystal molecules at the interface with the surrounding medium (fluid or solid substrates). Moreover, PVA is a water soluble, nontoxic, biodegradable polymer, that is also widely employed as an emulsifier for oils and polymers.[25,26] PVA has been used successfully as protecting agent in suspension polymerization and as a steric stabilizer in the dispersion polymerization of aniline, methyl methacrylate, styrene, and acrylic acid.[27] The suspension or emulsion stability depends on several parameters, as the PVA molecular weight, the concentration, the degree of hydrolysis and the distribution of hydroxyl groups in the macromolecules.[25] All these factors also influence the interfacial tension between the aqueous phase (the vinyl alcohol groups interact favourably with water) and the monomer, affecting the droplets size.

In the present work, the twofold function of the PVA, as planar surfactant for chiral LC devices and as steric stabilizer for LC droplets, is exploited in order to create multicolour colloidal lasers based on mixed emulsions of dye doped chiral liquid crystal (Ch-LC) microdroplets lasing at different wavelengths. To achieve these results three chiral nematic mixtures with different chiral dopant concentrations are combined with different fluorescent dyes. However, since the different chiral mixtures are miscible, the presence of the proper PVA concentration, that prevents droplets coalescence, enables to obtain stable mixed emulsions of cholesteric droplets with different pitches including also proper dyes for lasing. Different combinations of sensitizer-emitter dyes, providing the Forster coupling effect, are used to obtain emission in the blue, green and red regions simultaneously, excited by the same pumping wavelength. Omni-directional and simultaneous lasing at three different wavelengths from the emulsion is demonstrated. In addition, the mixed emulsion is transformed, after water evaporation, in a PVA film including Ch-LC droplets, demonstrating the possibility to build up the same device in shape of a plastic sheet. We, therefore, present a successful control of a self-assembly route enabling the scalable, low-cost and single-step production of a stable mixed emulsion of Ch-LC droplets including different dopants and we demonstrate a very simple and easy way to achieve a multicolour laser device.

Experimental section

Materials

The selected nematic liquid crystal is MLC-7023 (extraordinary refractive index n_e=1.53, ordinary refractive index n_0=1.46), the chiral dopant is MLC- 6248 (right handed) (both from Merck). Three different fluorescent dyes, Uvitex (Niopik), Coumarin 6 and Nile Red (Sigma-Aldrich), and chiral nematic mixtures are prepared in order to achieve lasing at three different wavelengths under optical pumping of the same laser wavelength in the UV region, by combining the cholesteric pitch and the dye emission. The following mixtures are selected and prepared:

1) Blue Ch-LC - 99.6%wt. [68% wt. MLC-7023 + 32%wt. MLC-6248] + 0.4%w blue dye Uvitex;

2) Green Ch-LC - 99.5%wt. [74%wt. MLC-7023 + 26%wt. MLC-6248] + [0.3%wt. blue dye Uvitex + 0.2%wt. Coumarin 6];

3) Red Ch-LC - 99.4%wt. [78%wt. MLC-7023 + 22%wt. MLC-6248] + [0.2%wt. blue dye Uvitex + 0.2%wt. Coumarin 6 + 0.2%wt. Nile Red].

Emulsion preparation

The liquid matrix is prepared mixing uniformly water and PVA (Sigma Aldrich) at a temperature of 90°C. The selected weight percentage of water and PVA is 90%wt. and 10%wt., respectively. For weight percentage of PVA higher than 11% wt., aggregates start to form. The refractive index of the PVA/water solution is 1.34.[28] Successively, a small quantity of the three mixtures, about (1%wt.) is added to the blend including water and PVA, at room temperature. The glass vial (diameter 1.0cm, height 2.5cm) containing the blend and the Ch-LC mixtures is then subjected to a shaking process at 20Hz for 40s, at a temperature of 40°C in a laboratory vortex mixer.

Polymeric film preparation

To obtain a uniform thin film of dye doped Ch-LC droplets encapsulated in the polymeric matrix, the emulsion is drop casted on a glass slide and left to dry for 48 hours at room temperature. Then, it is carefully detached from the glass substrate. The polymeric film including the Ch-LC droplets is freestanding, flexible and mechanically resistant to small stresses.

This journal is © The Royal Society of Chemistry 20xx

View Article Online
DOI: 10.1039/C7SM01068K

Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.

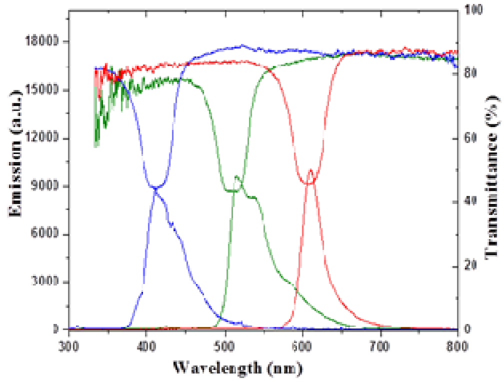


Fig. 1 Transmission (top) and fluorescence (bottom) spectra for the three dyes doped chiral liquid crystal mixtures.

Results and discussion

Initially, the optical properties of the three cholesteric mixture and of the fluorescent dyes are investigated. In Fig. 1 are reported the transmission spectra (using unpolarized light) of the Ch-LCs, and the fluorescence spectra of the dyes used in the three types of mixtures, showing their proper relative position in order to achieve lasing at the longer band edge of the three types of microresonators. Only a narrow range of wavelengths is selectively reflected by the chiral materials, providing the amplification of the light produced by the fluorescent dyes. The presence of Uvitex in mixture 2 and of Uvitex and Coumarin in mixture 3, enables the Förster-type energy transfer,[29] using Uvitex molecules as donors in the first case and the Uvitex and Coumarin combination as donors in the second case. Due to this photochemical process, the same UV pumping wavelength can be used in order to achieve simultaneous lasing in the blue, green and red region where the three stop-bands of the Ch-LCs are located.

The microdroplets emulsion is obtained by adding a small amount (about 1%) of each mixture to the water-PVA blend, Fig. 2. Due to the LC hydrophobicity, the Ch-LCs droplets acquire a spherical shape. The glass vial containing the blend is, then, subjected to a shaking process, resulting after some tens of seconds in a mixed emulsion including three types of Ch-LC droplets with diameters mainly ranging between 10μm - 60μm. The optical microscope image acquired between crossed polarizers displays for each droplet the typical Maltese cross, due to the isotropic arrangement of the supramolecular helicoidal structures confined within the droplets. Based on the LC hydrophobicity and the presence of PVA, the LC molecules orient parallel to the water interface promoting the formation of an internal radial configuration of the cholesteric helices, i.e. where the cholesteric layers are bent in concentric spherical surfaces with a single defect in the centre. Of course, this configuration is obtained in the case of droplets with a radius larger than the cholesteric pitch,[18] that is the case treated in the present work (the cholesteric pitch of the Ch- LC ranges from 280nm-blue mixture to 420nm-red mixture). The microemulsion obtained after the shaking process includes radial droplets, each having different pitch and proper dyes inclusion. In Fig. 3a is reported the image of a droplet in the emulsion, acquired with the confocal microscope, including Ch-LC with micrometer sized pitch in order to display the internal “onion structure” of the cholesteric layers. In Fig. 3b is reported an optical microscope image acquired in reflection mode showing the emulsion prepared with the three LC mixtures. The image shows droplets reflecting light of the three expected colours: blue, green and red. The central coloured spot is fixed and independent from the droplets orientation, due to the omnidirectional reflectance proper of the Bragg onion structures of the microresonators;[15] the displayed colours ($\lambda = n_{av}p$) depend on the pitches (p) of the supramolecular helicoidal structures within the droplet, induced by the percentage of the chiral dopant of the three mixtures (Fig. 1), and the average refractive index of about 1.48 for all mixtures.[13,19]

It must be stressed that this task is not so obvious since the three mixtures added to the water/PVA blend are miscible. In order to achieve emulsions including droplets of the three types of mixtures, one of the main drawbacks to be inhibited is the coalescence, i.e. the process of thinning and disruption of the liquid film between the droplets that results in the fusion of two or more droplets into larger ones. The driving force for coalescence is the surface or film fluctuations which results in close approach of the droplets whereby the van der Waals forces is strong, thus, preventing their separation.

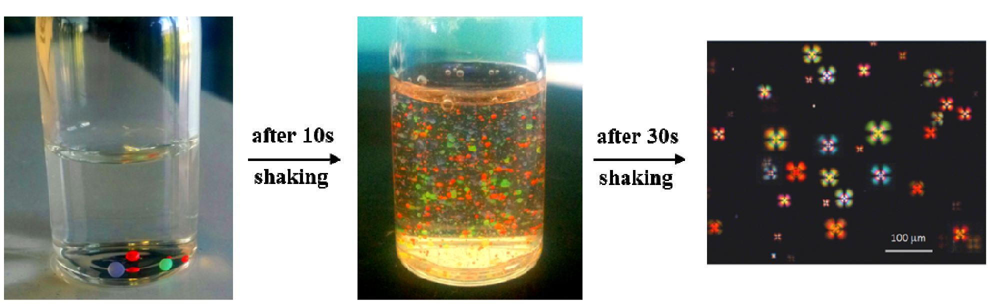


Fig. 2 Schematic representation of the emulsification procedure used to create the mixed emulsion including droplets of the three types of dyes doped Ch-LC mixtures. The last image is acquired using an optical microscope in transmission mode between crossed polarizer.

Soft Matter Accepted Manuscript

View Article Online
DOI: 10.1039/C7SM01068K

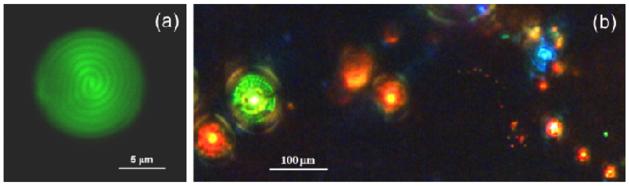


Fig. 3 a) Confocal microscope 3D image acquired on a long pitch (p≈1.5μm) dye doped droplet with a radius of ≈5μm. The measurements are performed by means of a confocal laser scanning microscope Leica TCS SP8; b) optical microscope image of the emulsion acquired in reflection mode.

The stabilization of dispersions of liquid/liquid types, against flocculation and/or coalescence requires the presence of an energy barrier between the droplets that prevents their closer approach. Two general mechanisms of stabilization can be applied. The first one refers to electrostatic stabilization and it is based on charge separation and formation of electrical double layers. This is the case of ionic surfactants or emulsifiers.

An alternative approach involves non-ionic surfactants, the involved mechanism is usually referred as steric stabilization. This is the case of PVA. The acetate groups give PVA its amphipathic character; on a hydrophobic surface (as our liquid crystals) the polymer absorbs with preferential attachment of the acetate groups on the surface, leaving the more hydrophilic vinyl alcohol segments dangling in the aqueous medium. These stabilising chains extend from the surface, creating a layer with a thickness d that can be several nanometers.[27] When two droplets approach to a distance shorter than 2d, the chains may undergo to an overlap, such overlap is unfavourable leading to a strong repulsion.

Good stability of the mixed emulsion is obtained for a percentage of PVA in water in the range 8-11% in weight. While for higher concentrations the PVA water solution is too dense to allow the creation of microdroplets through mechanical shaking, for lower concentrations the mixed emulsion is unstable and coalescence of the droplets occurs. Based on the drafted droplets formation during the shaking process, the emulsion contains three types of microresonators including proper dyes (Fig. 4) that, pumped with the same UV light source, enable to achieve laser emission at different wavelengths in the red, green and blue ranges, namely: 425nm, 516nm and 610nm. For lasing experiments, a Nitrogen laser is used as pumping light source. The pulse wavelength, width, and repetition rate are 337nm, 4 ns, and 5Hz, respectively. The pumping energy is about 150 μJ/pulse, the pumping beam is focused (focal length of the lens, 5cm) on a “cuvette” containing the mixed emulsion. The cuvette has a square base (1cmx1cm) and height of 3cm; the section of the beam at the beam waist within the sample has an area of about $5x10^{-3}$ cm^2. The experimental investigation is performed at room temperature. The typical exposure times are tens of minutes, and any significant photodegradation of the PVA has been detected from the absorption spectra. An optical fibre, coupled to a spectrometer (AVASPEC-2048, Avantes) with spectral resolution of 1 nm collected the light emitted from the sample. Based on the photonic features of the mixed emulsion and on the omnidirectional lasing proper of the dye doped Ch-LC microresonator, simultaneous lasing at different wavelengths is expected in all directions around a “cuvette” containing the emulsion.

In Fig. 5 are reported the spectra measured in front of the “cuvette” along the pumping direction (F) and on a side perpendicular to the pumping beam direction (S). The emission from the “cuvette” in both directions highlights the presence of three stable laser lines at 425nm, 515nm and 610nm. All lines are located in the region near the longer wavelength edge of the photonic band-gap of the three Ch-LCs mixtures (Fig. 1), as expected for Bragg lasing.[19] Laser lines supported by whispering gallery modes are not observed in the investigated spectral range, probably due to the moderate index contrast between the droplets and the PVA/water environment. In each spectrum the intensity of the three lines is also comparable, as expected due to the same percentage of the three Ch-CLs mixtures used to build-up the emulsion. The pump beam line at 337 nm appears together with the emitted lines, showing that their widths are comparable. However, the intensities of the lines as well as their height with respect the pumping line in [S] and [F], depend on the detector position. In the first case the fibre is placed at 1cm from the cuvette, while in the second case is at a distance of 10cm in order to minimize the detected intensity of the transmitted pumping beam.

Although the operating principle of the method and its feasibility is demonstrated for only three wavelengths in the blue red and green, it must be highlighted its great flexibility and easiness to make possible a full control of the lasing spectrum. The position, as well as the number of laser lines and their intensity, can be easily tailored by properly preparing the dye doped Ch-LC mixtures according to the position of the photonic band-gaps and the emission spectra of the fluorescent dyes, by the number of dye doped Ch-LC mixtures and by their concentration to build up the PVA/water emulsion. The reported procedure paves the way to optimize the fabrication of an omnidirectional white laser source.

In order to demonstrate the feasibility in manipulating and arranging this composite system and the versatility of the photonic device, the emulsion is transformed, after water evaporation, in a plastic sheet containing the dye doped Ch-LC microdroplets. Fig. 6a shows an optical microscope image acquired in transmission mode of the plastic label, including droplets with different cholesteric pitch and dyes. Even during this process, the PVA chains around the droplets preserve their separation, and enable to obtain a plastic or paintable[23] option of the same device.

Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.
Soft Matter Accepted Manuscript

This journal is © The Royal Society of Chemistry 20xx

View Article Online
DOI: 10.1039/C7SM01068K

Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.

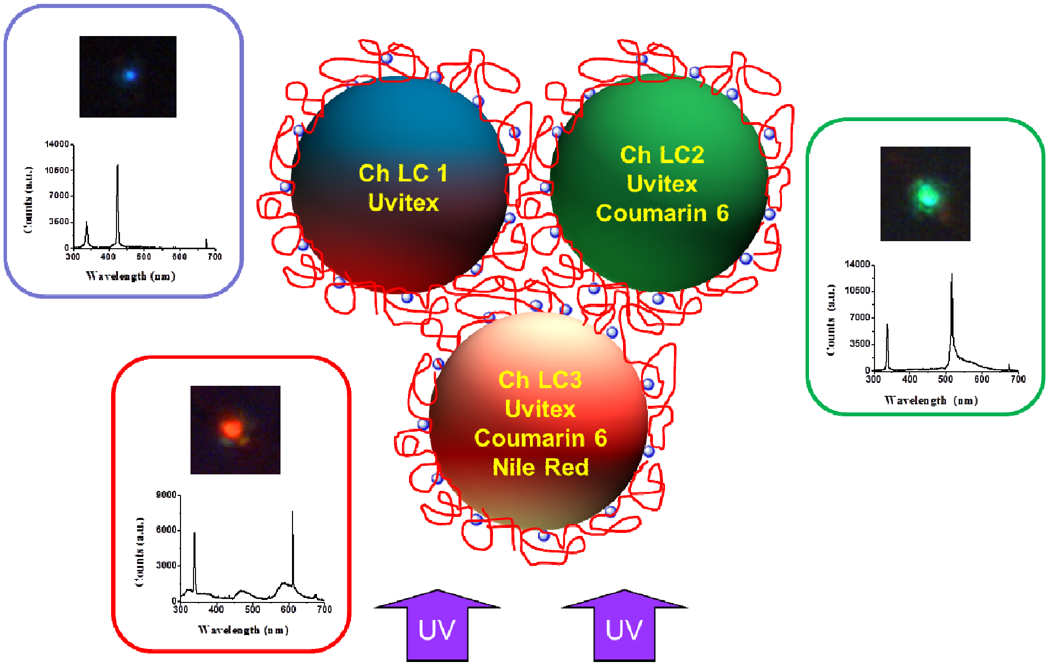


Fig. 4 The three types of droplets in the mixed emulsion including proper dyes combination. The scheme shows that each droplet excited by the same pumping UV source exhibits lasing in the blue, green and red region of the VIS spectrum. In the boxes the lasing spectra displayed for the three emulsions excited by the pumping UV Nitrogen laser beam at 337nm.

Fig. 6a, obtained using an optical microscope, as well as Fig. 6b, obtained using a confocal microscope, clearly show two droplets in contact between them that are deformed by the mechanical stress occurring during the formation of the polymeric film, while droplets coalescence is inhibited. The emission from the polymeric layer excited by the Nitrogen laser shows three stable laser lines at 421nm, 516nm and 615nm (Fig. 6c), demonstrating that the optical quality and the photonic features of the microresonators, even after encapsulation in the polymeric matrix, are well preserved. Fig.6d shows a picture of the flexible polymeric film. It must be noted that also in the case of chiral droplets encapsulated in a polymeric matrix, even if such kind of composed material is well known (polymer dispersed liquid crystals) and several methods have been developed in order to produce them,[30,31] the present procedure offers the unique capability to achieve a polymeric matrix where different types of droplets are dispersed. The above feature allows to easily engineer the optical materials by properly tailoring transmission, reflection and absorption properties in order to manipulate light intensity, wavelength and polarization, besides the already shown control of the photonic performances.

Moreover, since the liquid crystal phase in the shape of droplets is always maintained in both devices (emulsion and polymeric film), also the tunability functions related to the action of external stimuli on the liquid crystal properties and on its supramolecular arrangement within the droplets, can be exploited to control and tune the optical and photonic properties of the microdevices giving, for example, the opportunity to introduce electro-optical functionalities to the photonic system.

Conclusions

In summary, a very simple and flexible method to create a multicolour laser system based on a mixed emulsion of Ch-LC droplets has been presented. The main task successfully achieved is to have an emulsion with droplets of Ch-LC

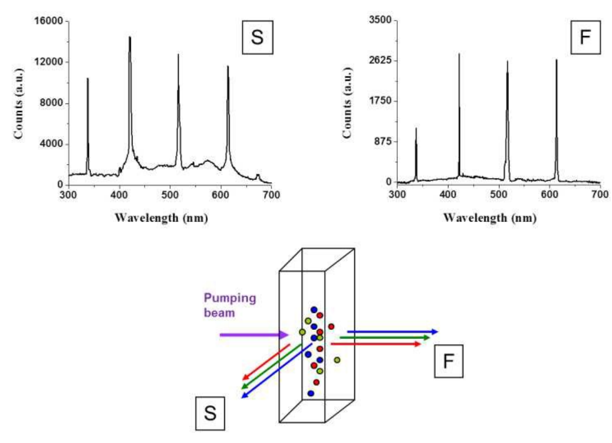


Fig. 5 Lasing spectra from the mixed emulsion when excited by the pumping beam at 337nm. Spectrum F is acquired along the same direction of the pumping beam, spectrum S is measured in a direction perpendicular with respect to the pumping one.

Soft Matter Accepted Manuscript

View Article Online
DOI: 10.1039/C7SM01068K

Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.

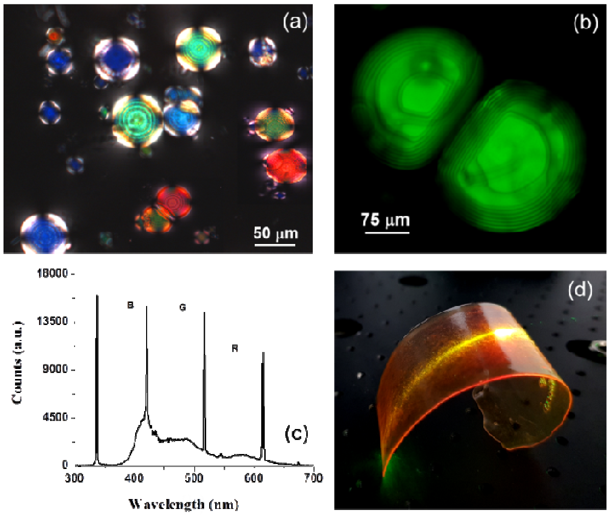


Fig. 6 a) Optical microscope image of the polymeric film embedding different types of dye doped Ch-LC droplets, between crossed polarizers; b) confocal microscope image displaying two Ch-CL droplets in contact; c) lasing spectrum from a PVA film encapsulating dyes doped Ch-LC droplets after water evaporation, when the film is pumped with the Nitrogen laser beam at 337 nm, d) flexible film.

including different dopants, even at different concentrations. The strategy adopted to substantially improve the stability of the composite material is to exploit the steric stabilization of the droplets obtained through the proper concentration of PVA, a surfactant generally used in LC devices to promote parallel orientation of the LC molecules at the interface with liquids or solids. Based on the peculiarity of the Bragg onion-type microresonators displayed by the chiral droplets, simultaneous lasing at three wavelengths (blue, green and red) in all directions around a "cuvette" containing the mixed emulsion has been successfully achieved. This good result is connected to the high stability of the mixed emulsion, the proper selection of the materials (LC and fluorescent dyes) and the combination of photoinduced processes. This work provides an effective method to create omnidirectional multicolour laser systems with very high flexibility. Photonic properties of the devices can be easily engineered (full control of the laser spectrum). The fluidic nature of the photonic device is extremely useful in offering opportunities for reconfigurability and manipulation, and easy integration into optofluidic systems and lab on chip devices. A very simple procedure enables to transform the mixed emulsion in a solid polymeric label where the droplets are embedded, preserving their optical and photonic properties. The emulsified materials can be easily coated on glass, plastic or metallic substrates offering interesting opportunities for coatings, optical security devices, sensing, imaging and optical technologies, etc. Moreover, for some of these applications where narrow droplets size dispersion is not a strict condition, the feasibility to generate a large amount of mixed emulsion in only a single step process represents a great advantage with respect to microfluidic procedures where one droplet at a time is generated. Finally, mixtures including different types of dopants (as for example metallic and/or magnetic nanoparticles[32]) can be also used to produce the mixed emulsion in order to change or increase the material functionalities by controlling plasmonic and magnetic properties besides the demonstrated optical (and photonic) ones.

Acknowledgements

G. Cipparrone and R.J. Hernandez thank COST Action MP1205 "Advances in Optofluidics: Integration of Optical Control and Photonics with Microfluidics" for supporting their research activities. All authors are grateful to Mauro L.D. Bruno for technical support. This work has been partially supported the Calabrian Innovation Pole for Materials and Production Technologies, Calpark, Italy.

Notes and references

1 A. Huebner, S. Sharma, M. Srisa-Art, F.Hollfelder, J. B. Edel, A. J. deMello, Lab on a Chip, 2008, 8(8), 1244 and A. B.Theberge, F.Courtois, Y.Schaerli, M.Fischlechner, C.Abell, F. Hollfelder, W. T.Huck, Angewandte Chemie International Edition, 2010, 49(34), 5846 and C. Martino, A. J. deMello, Interface Focus, 2016, 6(4), 20160011 and S. Mashaghi, A. Abbaspourrad, D. A.Weitz, A. M.van Oijen, Trends in Analytical Chemistry, 2016, 82, 118.
2 Li, Zhenyu, D. Psaltis, Microfluidics and Nanofluidics, 2008, 4(1-2), 145.
3 R. Zilionis, J. Nainys, A.Veres, V.Savova, D. Zemmour, A. M. Klein, L. Mazutis, Nature Protocols, 2017, 12(1), 44.
4 J. H. Jang, S. Y. Park, Sensors and Actuators B: Chemical, 2017, 241, 636-643.
5 R. K. Chang, A. J. Campillo, Optical Processes in Microcavities, World Scientific Publishing Co., USA, 1996.
6 X. Qian, J. B. Snow, H. M. Tzeng, R. K. Chang, Science, 1986, 231(4737), 486.
7 S. K. Y. Tang, Z. Li, A. R. Abate, J. J. Agresti, D. A. Weitz, D. Psaltis, G. M. Whitesides, Lab on Chip, 2009,9(19), 2767.
8 M. Saito, H. Shimatani, H. Naruhashi, Opt. Express, 2008, 16(16), 11915.
9 S. K.Tang, R. Derda, Q.Quan, M.Lončar, G. M. Whitesides, Optics Express, 2011, 19(3), 2204.
10 Y. Wang, H.Li, L.Zhao, B. Wu, S. Liu, Y. Liu, J. Yang, Optics & Laser Technology, 2016, 86, 61.
11 S. Yang, Y. W. Van Duong Ta, R. Chen, T.He, H. V. Demir, H. Sun, Scientific Reports, 2016, 6, 27200.
12 H. G.Lee, S.Munir, S. Y. Park, Materials & Interfaces, 2016, 8(39), 26407 and M.R. Foreman, J.D. Swaim, F.Vollmer, Advances in Optics and Photonics, 2015, 7, 168.
13 G. Chilaya, Chirality in liquid crystals. Springer, New York, 159, 2001.
14 I. Muševič, Liquid Crystals Reviews, 2016, 4(1) and M. Humar, Liquid Crystals, 2016, 43, 1937.
15 M. Humar, I. Muševič , Optics Express, 2010, 18, 26995.
16 Y. Xu, W. Liang, A. Yariv, J. G. Fleming, S. Y. Lin, Optics Letters, 2003, 28, 2144.
17 P. Z.Sun, Z.Liu, W. Wang, X.Wang, H. M. Zhang, Y. X.Lu, Z. G.Zheng, Optical Materials Express, 2016, 6(4), 1256 and L. J. Chen, L. L.Gong, Y. L.Lin, X. Y.Jin, H. Y. Li, S. S. Li, C. J. Yang, Lab on a Chip, 2016, 16(7), 1206.
18 D. Seč, T. Porenta, M. Ravnik, S.Žumer, Soft Matter, 2012, 8, 11982 and Y. Bouligand, F.Livolant, Journal de Physique, 1984, 45, 1899.

Soft Matter Accepted Manuscript

This journal is © The Royal Society of Chemistry 20xx

Please do not adjust margins

View Article Online
DOI: 10.1039/C7SM01068K

19 V. I. Kopp, B. Fan, H. K. M. Vithana, A. Z. Genack, Optics Letters, 1998, 23 (21), 1707 and I. P. Ilchishin, E. A.Tikhonov, Progress in Quantum Electronics, 2015, 41, 1.
20 M. Ozaki, M.Kasano, T.Kitasho, D. Ganzke, W.Haase, K.Yoshino,. Advanced Materials, 2003, 15(12), 974 and L. Saadaoui, G. Petriashvili, M.P. De Santo, R. Hamdi, T. Othman, R. Barberi, Optics Express, 2015, 23, 22922 and G. Petriashvili, M.A. Matranga, M.P. De Santo, G. Chilaya, R. Barberi, Optics Express, 2009, 17, 4553.
21 M. Humar, F.Araoka, H.Takezoe, I. Muševič, Optics Express, 2016, 24(17), 19237.
22 R. J.Hernández, C.Provenzano, A.Mazzulla, P.Pagliusi, M.Viola, G. Cipparrone, Liquid Crystals Reviews, 2016, 4(1), 59.
23 D.J. Gardiner,. S. M. Morris, P. J. W. Hands, C. Mowatt, R. Rutledge, T. D. Wilkinson, H. J. Coles, Optics Express, 2011, 19, 2437.
24 Y.Geng, J.Noh, I.Drevensek-Olenik, R. Rupp, G.Lenzini, J. P. Lagerwall, (2016). Scientific Reports, 6, 26840.
25 C. A. Finch Polyvinyl Alcohol: Properties and Applications. John Wiley & Sons, New York, 1973.
26 T. Okaya, A. Suzuki, K. Kikuchi, Colloids Surf., 1999, 153, 123 and K. Yuki, M. Nakameae, T. Sato, H. Murayama, T. Okaya, Polymer International, 2000, 49, 1629.
27 J. Stejskal, P. Kratochvil, M.Helmstedt, Langmuir, 1996, 12, 3389 and O.K. Kim, K. Lee, K. Kim, B.H. Lee, S. Choe, Colloid and Polymer Science, 2006, 284, 909 and J. Hong, C.K. Hong, S.E. Shim Colloids and Surfaces A, 2007, 302, 225 and H. Minami, A. Kimura, K. Kinoshita, M. Okubo, Langmuir, 2010, 26, 6303.
28 R.J. Sengwa, S. Sankhla, S. Sharma, Indian Journal of Chemistry, 2007, 46A, 1419.
29 A. Chanishvili, G. Chilaya, G. Petriashvili, R. Barberi, R. Bartolino, G. Cipparrone, A Mazzulla, R. Gimenez, L. Oriol, M. Pinol, Applied Physics Letters, 2005, 86(5), 051107.
30 P. S. Drazic, Liquid Crystal Dispersions, World Scientific, Singapore, 1995.
31 L. G. Chen, R. A. Shanks, Liquid Crystals, 2007, 34, 1349.
32 Y Li , J.J.Y. Suen, E. Prince, E. M. Larin, A. Klinkova, H. Thérien-Aubin, S. Zhu, B. Yang, A. S. Helmy, O. D. Lavrentovich, E. Kumacheva, Nature Communications, 2016, 7, 12520.

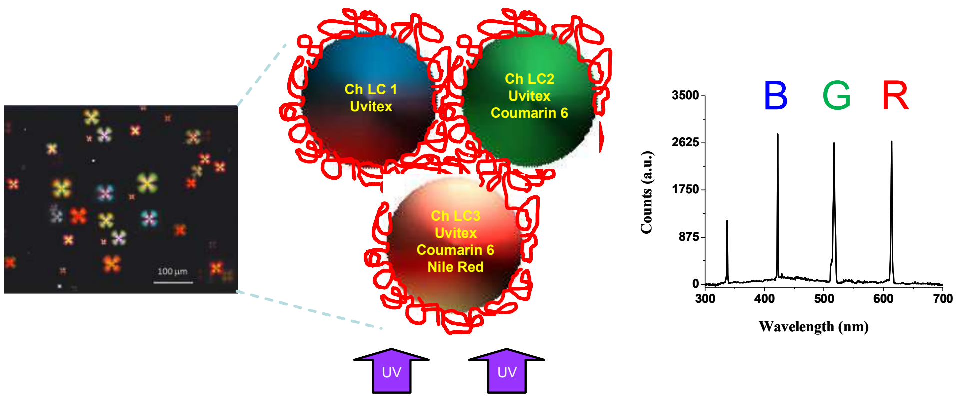
Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.
Soft Matter Accepted Manuscript

This journal is © The Royal Society of Chemistry 20xx

View Article Online
DOI: 10.1039/C7SM01068K

A table of contents entry:

Mixed emulsions based on dyes-doped chiral liquid crystals droplets are developed to create multicolor laser systems with full control of the lasing spectrum.



Published on 26 July 2017. Downloaded by University of Newcastle on 27/07/2017 19:27:24.

Soft Matter Accepted Manuscript