Lasing and waveguiding in smectic A liquid crystal optical fibers

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Abstract: We demonstrate a new class of soft matter optical fibers, which are self-assembled in a form of smectic-A liquid crystal microtubes grown in an aqueous surfactant dispersion of a smectic-A liquid crystal. The diameter of the fibers is highly uniform and the fibers are highly birefringent. They are characterized by a line topological defect in the core of the fiber with an optical axis pointing from the defect core towards the surface. We demonstrate guiding of light along the fiber and Whispering Gallery Mode (WGM) lasing in a plane perpendicular to the fiber. The light guiding as well as the lasing threshold are significantly dependent on the polarization of the excitation beam. The observed threshold for WGM lasing is very low ($\approx 75\,\mu J/cm^2$) when the pump beam polarization is perpendicular to the direction of the laser dye alignment and is similar to the lasing threshold in nematic droplets. The smectic-A fibers are soft and flexible and can be manipulated with laser tweezers demonstrating a promising approach for realization of soft photonic circuits.

OCIS codes: (130.5460) Polymer waveguides; (140.3948) Microcavity devices; (230.3720) Liquid-crystal devices; (230.4555) Coupled resonators; (230.7400) Waveguides, slab.

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1. Introduction

Thermotropic liquid crystals (LCs) are formed from anisotropic organic molecules with a shape of a rod or a disc. Depending on the temperature and molecular structure, these compounds spontaneously form various liquid crystalline phases. The most simple is the nematic liquid crystalline (NLC) phase, where the axes of the rod-like molecules are spontaneously aligned into a common direction, called the director $\boldsymbol{n}$. The nematic phase is a positionally disordered liquid phase. By lowering the temperature, the nematic phase usually spontaneously transforms into the smectic-A phase. This phase preserves the spontaneous orientational order, but the LC molecules are in addition positionally ordered into molecular layers, called smectic layers, the layer planes being perpendicular to the director $\boldsymbol{n}$.

There has been a renewed interest in dispersions of NLCs in immiscible fluids and gels, such as water, glycerin, PDMS etc.. Because of chemical incompatibility, the NLC readily forms dispersions of micrometer-sized droplets of perfect spherical shapes. It has been demonstrated that these microdroplets can be used as tunable optical microresonators [1], 3D microlasers [2–4] and Whispering-Gallery Mode (WGM) microlasers [5]. These observations have lead to the idea of using the dispersions of NLC in immiscible fluids as basic photonic elements for hypothetical soft-matter photonic circuits. This idea is at variance with the rapidly developing approaches and technology platforms, where silicon photonics has become one of the most robust and promising platforms for future solid-state photonic circuitry [6,7]. Recently, resonant transport of light from a thin polymer film on glass into a tunable nematic-liquid crystal droplet, floating in water just above the surface of the polymer waveguide, has been demonstrated [8]. The next step towards the realization of all-soft matter photonic circuit is to replace the rigid polymer by a liquid-like optical waveguide that has a potential of self-assembly.

It has been shown that it is possible to assemble freely suspended, sheet-like smectic-A liquid crystal layers in water [9], which could in principle serve as optical slab-waveguides. Another type of structure, corresponding to liquid-like fibers, are the myelin figures which can be grown or self assembled at the interface of lyotropic lamellar LCs and water (see [10] Reissig et al. and references therein). Myelin figures are tube-like structures consisting of cylindrically bend

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water/surfactant bilayers. However, the small difference between the refractive index of the lyotropic myelin figures and their aqueous environment renders these systems unsuitable for waveguiding applications. Recent studies have demonstrated that similar tube-like structures, consisting of coaxially arranged smectic layers, can be grown at the interface of thermotropic smectic-A LCs and aqueous phases containing cationic surfactant micelles [11]. This work has triggered our interest in the photonic properties of these thermotropic myelins, consisting of smectic-A liquid crystal fibers, because of the following reasons: (i) the coaxial organization of smectic layers could give rise to very precise and uniform diameter of the fibers to one molecular length (2-3 nm), (ii) their smectic nature should substantially decrease the degree of light scattering losses and surface smoothness, as the molecular orientational fluctuations are depressed in smectic-A phase, (iii) the coaxial smectic-A fiber should exhibit a topological line defect of the winding number 1 [12] along the major axis of the tube. As the line defect must be in perfect alignment with the cylindrical axis of the fiber due to the molecular order, it is to be expected that the optical modes not only perfectly adjust with this defect, but also have topological singularity of the electromagnetic field.

2. Experiments

We used a common room temperature smectic-A LC compound, 8CB (4-octyl-4'-cyanobiphenyl), for the preparation of the smectic microfibers. The LC was obtained from Synthon chemicals and used as received. The LC compound was doped with 0.01 wt.% of the fluorescent dye 7-diethylamino-3,4-benzophenoxazine-2-one (Nile red, Sigma-Aldrich). We used a cationic surfactant, C16TAB (hexadecyltrimethylammonium bromide, Sigma-Aldrich), for the preparation of the aqueous solution. The critical micellar concentration of this surfactant in water is approximately 1 mM at 25°C, in our experiments we used aqueous solutions with C16TAB concentrations between 10 and 100 mM. The refractive indices of 8CB ($n_o = 1.516$, $n_e = 1.673$) are significantly higher than that of the surrounding medium containing water ($n = 1.33$) and C16TAB ($n = 1.435$). In order to generate the smectic microfibers, 1 mg of the 8CB-dye mixture was sandwiched between two glass plates, which are separated with 100 μm spacers, and the aqueous micellar solution was brought into contact with the LC using capillary forces. Under an optical microscope, one could immediately see very fine microtubes protruding from the LC/aqueous interface into the aqueous phase. A more detailed discussion about the growth of these microtubes is available in our recent publication [13]. The diameter of these tubes or fibers depends on the concentration of the surfactant in the water, the cell thickness and the temperature of the system, and can be varied from a few μm to $\approx 100\,\mu$m. We have found that the fibers can be grown up to centimeter length, provided the 8CB reservoir and the aqueous environment possess appropriate sizes and surfactant concentration. However, the detailed mechanism of the fiber growth is still to be clarified. All experiments were done at 23°C.

For the investigation of the waveguiding and lasing properties of the fibers, the experiments were performed on an inverted microscope (Nikon Eclipse, TE2000-U), using the set-up shown in Fig. 1. The fibers are contained in glass cells (thickness 100 μm), freely floating in the aqueous surfactant solution in which they were grown. The optical setup is basically a combination of two laser tweezers: an infrared fiber laser tweezers operating at 1064 nm (Aresis) and a continuous Ar^+ laser tweezers operating at 514 nm using a 60× high numerical aperture objective. The infrared laser tweezers were used to position the smectic fibers in the aqueous environment. The waveguiding properties of the fibers were studied using the beam of the continuous Ar^+ laser tweezers which was directed to one end of a fiber, where the beam excited the fluorescence from Nile red molecules. In thicker fibers (tens of μm), the fluorescently generated light pattern was directly observable in the optical microscope (see the following section for more details). For the study of the lasing properties of the tubes, lasing was initiated by il-

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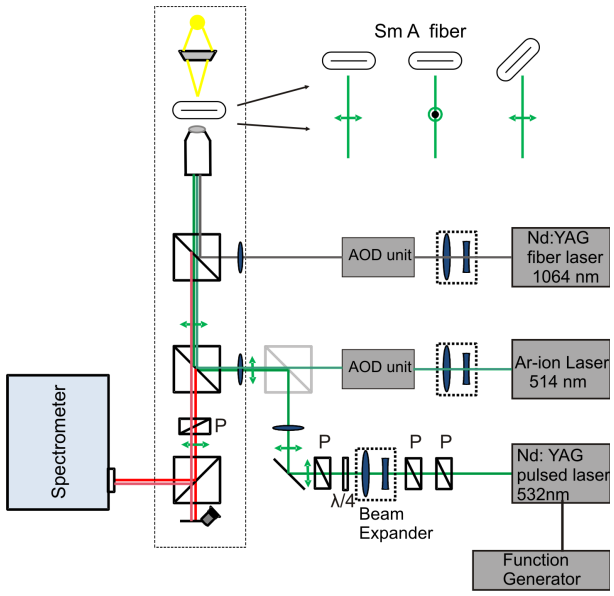

Fig. 1. Schematic diagram of the experimental set-up. The position of the focused infrared beam is controlled by two acousto-optic deflectors (AODs), thus allowing for the manipulation and tweezing of the smectic 8CB fibers in the aqueous C16TAB solution. The green pulsed beam from the doubled Nd:YAG laser is used to induce the fluorescence of the Nile red molecules which are dissolved and captured inside the 8CB fibers. The polarization of the incident beam can be varied with respect to the fiber axis. The continuous Ar^{+} laser is used to study the waveguiding properties of smectic fiber. The imaging spectrometer (Andor, Shamrock SR-500i), equipped with a cooled EM-CCD camera (Andor, Newton DU970N), is used to measure the optical spectrum emitted from the fiber at a 0.05 nm resolution. The CCD camera is used to take photomicrographs of the samples.

luminating the dye-doped fibers under the microscope with a pulsed laser (Nd: YAG 532 nm, Alphalas, Pulselas-A-1064-500) with a 100 Hz repetition rate, a pulse length of 1 ns and a maximum pulse energy of $10\,\mu$J. An additional optics was used to enlarge the illuminating region of the pulsed laser beam to a beam waist of $\approx 20\,\mu$m. An imaging spectrometer with a 0.05 nm resolution (Andor, Shamrock SR-500i) was used to measure the fluorescence and lasing spectra of the fibers, and a cooled EM-CCD camera (Andor, Newton DU970N) was used to take photomicrographs of the fibers.

To understand the results of the wave guiding and lasing in the liquid crystalline microtubes, we have used fluorescence confocal polarizing microscopy (FCPM) [14] to determine the alignment of Nile red molecules and the structure of 8CB microtubes. A Leica TCS-SP2 confocal microscope was used. For the FCPM measurements, the smectic fibers, floating in the aqueous surfactant solution, were contained in glass capillaries with a square cross section slightly larger than the fiber diameter, resulting in an appropriate spatial confinement of the fibers. The Nile red molecules in the fibers align parallel to 8CB molecules. As a result, a maximum fluorescent signal is observed in those regions of the FCPM images in which the 8CB molecules are oriented parallel to the polarization of the exciting laser beam of the confocal microscope.

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3. Results and discussion

Figure 2 shows polarizing micrographs of 8CB microtubes, obtained by myelin-like growth from the interface between 8CB and the aqueous C16TAB solution. Because the indices of refraction of 8CB smectic-A liquid crystal are higher than the refractive index of water, the IR laser tweezers could be used to grab and position the microfiber to a desired place. We noticed that the microfibers are very sensitive to the heat absorption induced by the strong light of the laser tweezers. When locally overheated, the tube ejected a thin jet of 8CB LC, which immediately formed a much thinner microfiber. Because of the layered structure of the smectic-A phase, we conjecture that the LC molecules are oriented perpendicular to the surface of the fiber and the layers are rolled-up in a sequence of concentrically wrapped molecular layers filling the fiber up completely.

The concentric organization of the smectic-A layers was indeed deducted from the inset in Fig. 2(a), where a λ-wave-plate for the red light was added between the sample and the polarizer, with the axis oriented at 45° with respect to the polarization [15]. The yellow and blue colored regions of the LC indicate two orthogonal directions of the local optical axis, which coincides with local molecular orientation. The concentric arrangement of the smectic

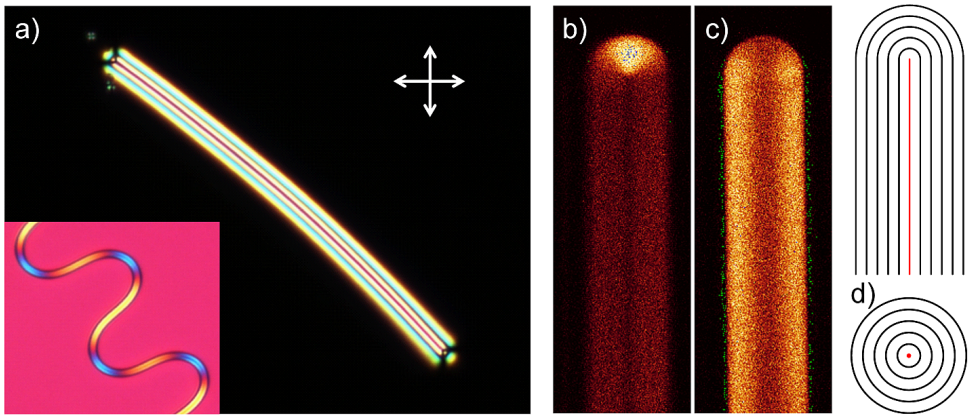

Fig. 2. Polarizing microscopy and FCPM images of smectic-A 8CB microfibers (doped with 0.01 wt.% of Nile red) in aqueous C16TAB solution. (a) 8CB microfiber (diameter $\approx 10\,\mu$m) between crossed polarizers demonstrating the strong birefringence of the smectic fiber. The inset shows a thinner fiber (diameter $\approx 3\,\mu$m), which has adopted a bend S-like shape, between crossed polarizers with the addition of a red (λ) wave plate. The alternating yellow/blue color sequence indicates that the LC molecules, and thus the local optical axis, are always aligned perpendicular to the surface of the fiber. (b) FCPM image of a smectic-A 8CB microfiber (diameter $35\,\mu$m) with the polarization of the exciting light along the axis of the microfiber. High fluorescence intensity is observed at the spherical tip of the fiber. (c) FCPM image of the same fiber with the polarization of the exciting light perpendicular to the axis of the microfiber. High fluorescence intensity is observed along the cylindrical body of the fiber. The distribution of the fluorescence intensities in both images indicates that the Nile red molecules, and thus the LC molecules, are oriented perpendicular to the surface of the fiber. (d) Schematic drawings of the coaxial arrangement of the smectic layers in a microfiber. Top: cross section parallel to the fiber axis. Bottom: cross section perpendicular to the fiber axis. The red line or dot indicates the topological line defect. The rodlike LC molecules (not shown in the drawings) are oriented perpendicular to the layer planes and the fiber surface.

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layers was additionally confirmed by taking the FCPM images of the 8CB microtubes doped with Nile red, shown in Fig. 2(b) and 2(c). The Nile red molecules are known to orient in the LC matrix so to have the brightest fluorescence when the polarization of the excitation light is set parallel to the LC director [16].

These observations lead to a clear confirmation of the internal structure of the smectic-A 8CB fibers, which is schematically depicted in Fig. 2(d). Each fiber is formed of a sequence of coaxial and rolled-up smectic-A layers and there are two hemispherical caps at each end of the fiber. Inside the caps, the smectic layers must form a sequence of nested hemispherical molecular layers, forming a highly birefringent and perfectly hemispherical lens with no topological defects. However, by considering the whole microfiber, there must be a topological line defect in the core of the fiber, extending all along the fiber and connecting the centers of the two hemispherical caps. One can see from Fig. 2 that the fibers are very uniform in diameter and there is no structural defect visible throughout the length of the fiber. The ends of the fibers are perfectly rounded, as shown in Fig. 2(b) and 2(c).

There are two interesting features of these microfibers regarding their photonic properties: *(i)* They are high optical quality and highly birefringent optical microfibers with a topologi-

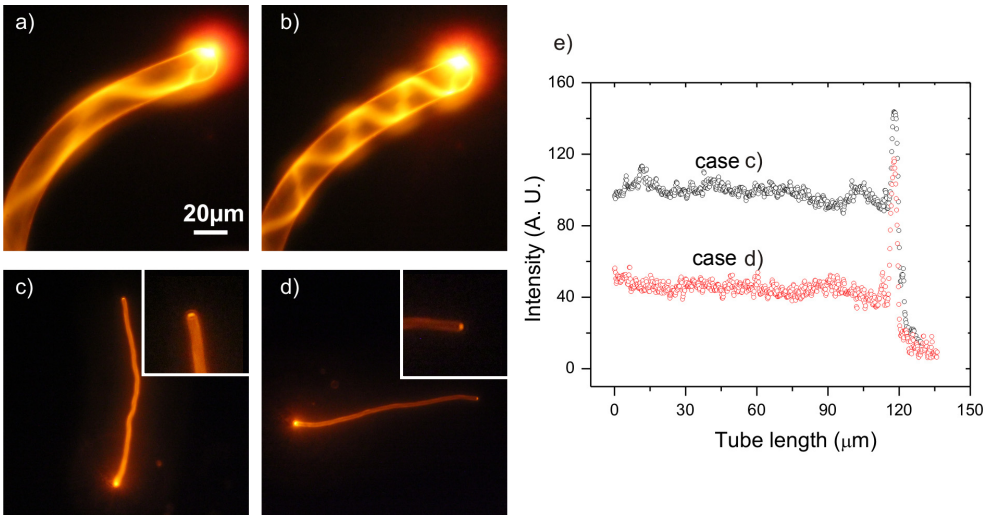

Fig. 3. Light guiding through 8CB smectic-A microfibers doped with 0.01 wt % of Nile red. The concentration of C16TAB surfactant in the surrounding aqueous medium is 20 mM. The power of the applied continuous Ar^+ laser beam for fluorescence excitation is 1 mW. (a-b) A focused Ar^+ beam is positioned at different points at the hemispherical cap of a 20 μm thick fiber. The spiral-shaped trajectories of the guided light are clearly visible because of the fluorescence. (c-d) Light guiding by a thin ($\approx 2\,\mu$m) 8CB microfiber. The fluorescence is excited at the lower (c) or left (d) end of the microfiber using different polarizations of the Ar^+ beam. The insets show the respective other end of the fiber at a higher magnification, the bright spot at the fiber end indicating the leaking of the guided light. The angle between the major axis of the microfiber and the polarization of the Ar^+ beam is 0° in (c) and 90° in (d). The corrugated appearance of the thin fiber is a result of some random bending and the small thickness of the fiber. There is no indication for an axial variation of the fiber thickness. (e) The intensity I of emitted fluorescent light as a function of position (i.e. length) along the thin microfiber in (c,d) remains fairly constant. Note the intensity peak at the end of the microfiber, corresponding to the bright spot shown in the insets in (c,d).

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cal line defect in the core. Having a topological line defect in the core, these microfibers are likely to support Gaussian-Laguerre eigenwaves [17]. It was demonstrated recently by Brasselet et al. [18] that a Gaussian beam is transformed into a Laguerre-Gaussian beam after passing through a microdroplet of a nematic LC with a topological point defect. The topological defects of the matter are therefore expected to be imprinted into the electromagnetic waves as phase singularities. *(ii)* The ends of the fibers are capped with birefringent hemispheres acting as perfect spherical lenses.

It is rather difficult to observe the light-guiding properties of smectic-A microfibers in the geometry that we are using, because it is difficult if not impossible to launch the external light into the fiber and analyze the transmitted light at the other end of the fiber. Therefore, light guiding properties of 8CB microfibers were studied by positioning a focused beam of the Ar^+ laser tweezers on the fluorescently labeled 8CB microfiber and observing the propagation of fluorescently generated light along the fiber by measuring the fluorescent light, leaking-out from the fiber. In this way, we obtain an indirect information on the light-guiding properties of the microfibers that gives us a qualitative figure of the quality of wave-guiding in the fiber. We first concentrated on the wave-guiding properties of rather thick smectic-A microfibers, shown in Fig. 3(a) and 3(b), where an intense fluorescence pattern was observed along the fiber. Changes in the fluorescent patterns were clearly visible as we moved the focus of the Ar^+ beam along the hemispherical cap of the microfiber. A significant amount of the fluorescent light in

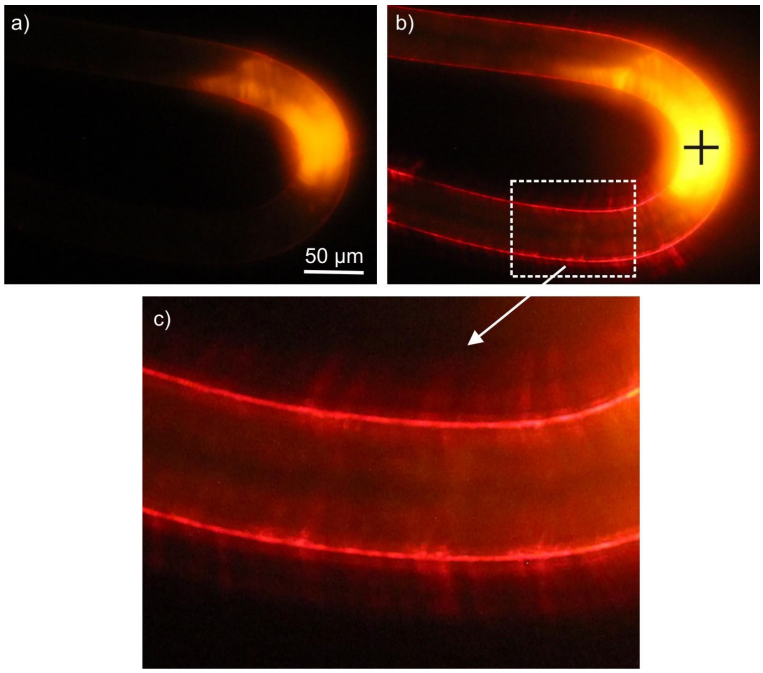

Fig. 4. An example of lasing from a large, $\approx 50\,\mu$m-diameter Nile red-doped 8CB microfiber in a 100 mM C16TAB water solution. The power of the pumping laser is below the lasing threshold in (a) and above the threshold in (b). In both cases the pumping pulsed laser beam is illuminating a $\approx 20\,\mu$m diameter region encircling the black cross. The polarization of the pumping light is along the tangent to the fiber at this position. Note the very distant laser speckles in (b), shining from the surface of the fiber at hundreds of μm separation (see enlarged section shown in (c)). Thinner fibers (thickness of a few μm) showed essentially the same behavior.

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the fiber got internally reflected at apparently very steep angles with respect to the major axis of the fiber (Fig. 3(a) and 3(b)), but this pattern in fact corresponds to a "spiralling" of light along the inner surface of the LC/aqueous interface. In thinner 8CB microfibers, such as the one shown in Fig. 3(c) and 3(d), the fluorescent light is emitted uniformly across and along the fiber and a bright spot is observed at the other hemispherical end of the fiber (insets in Fig. 3(c) and 3(d)). Figure 3(e) shows the measured intensity of the emitted fluorescent light along the microfiber for different polarizations of the excitation beam. No substantial drop ($< 5\%$) of the intensity was observed along the $\approx 130\,\mu$m overall length of the fiber and a bright spike of light was clearly seen at the exit of the fiber. When the polarization of the excitation beam matches the major axis of the tube, we observed substantially enhanced fluorescence emission in comparison to the other polarization.

After investigating the structure and wave guiding in the tubes, their lasing characteristics were studied by exciting the fluorescence in microtubes at a randomly chosen position along the tube. The beam waist of the pulsed and frequency doubled Nd:YAG laser was kept at $\approx 20\,\mu$m thus illuminating a substantial part of a $50\,\mu$m thick 8CB microfiber doped with Nile red dye. At low pumping levels, the whole volume of the microtube, including its surface, was nearly uniformly illuminated, and, while slowly increasing the pumping laser energy, the intensity of the illuminated region gradually increased up to a certain threshold. Then we could clearly observe the onset of very intense and monochromatic reddish speckles shinning from a very thin layer at the 8CB microtube/aqueous interface, as shown in Fig. 4. The intensity of these speckles rapidly increased with increasing pumping energy. The appearance of these speckles is a clear indication of the onset of lasing in the 8CB microfiber.

It is reasonable to assume that the lasing originates from the resonant Whispering Gallery Modes (WGMs) of light, circulating along the perimeter of the microfiber because of the total internal reflection (TIR) at the 8CB/aqueous interface. In Whispering Gallery Mode resonators, resonant condition is achieved by light circulation along the circumference of the cavity, such as a microsphere, micro-toroid, microfiber etc. Because of TIR, the WGM cavities exhibit large Q-factors, enabling low-threshold lasing with very small mode volume [19, 20]. In this case the laser light is emitted out of a resonator in a plane containing the circular path of resonant light bouncing of the cavity's interface due total internal reflection. When such WGM lasing is observed under a microscope, one can usually see strong coherent light shining towards the observer from the thin interfacial region of the cavity [1]. In our case, we have a high refractive index fiber with a circular cross-section which represents a cylindrical microresonator, with WGM modes that are resonantly circulating by TIR in a plane perpendicular to the axis of the cylindrical cavity (i.e. the fiber's axis). In this case, the WGM lasing is observed as a thin "sheet of light" emanating from the interface, which is shown in detail in Fig. 4c. One should note that such picture is observed for all angles of observation because of the cylindrical symmetry of the resonator.

The spectrum of speckles was analyzed at various pumping levels and is shown in Fig. 5(a) and 5(b). We observed a set of sharp spectral lines superposed on a broad fluorescent background, indicating the onset of lasing. These lines can clearly be attributed to the WGMs, circulating along the circumference on the inner side of the smectic-A fiber-water interface. The spectrum is very similar to the WGM lasing observed in nematic LC droplets with either radial or bipolar internal structure and a topological point defect in the center [5]. Similar spectrum of WGM lasing modes was recently reported for dye-doped polymer fibres [21].

As we observed significant effects of light polarization on the wave guiding in the smectic-A microtube, we also investigated lasing characteristics for different orientation of linearly polarized pumping beam with respect to the long axis of the microfibers. When the polarization of the pumping beam is parallel to the geometric axis of the fiber, the lasing threshold is

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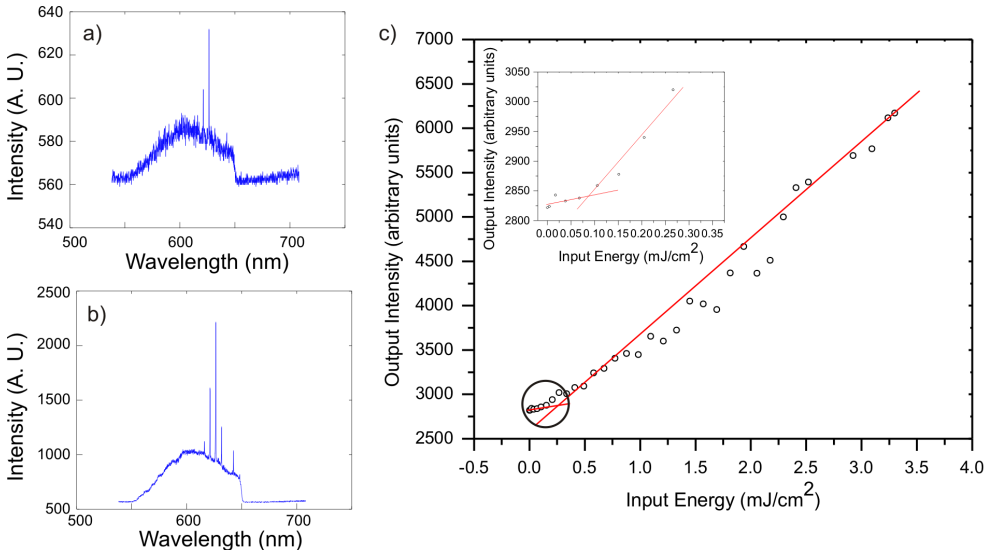

Fig. 5. The WGM lasing from the Nile red dye-doped smectic A microfiber of 8CB in 100 mM C16TAB water solution. (a) The lasing spectrum just above the threshold for lasing.(b) The lasing spectrum well above the threshold. In both cases the polarization of the pumping pulsed laser is along the major axis of the microfiber. The cut-off at 650 nm is due to the IR cut-off filter inserted in the dual beam laser tweezers. (c) Intensity of the emitted spectral line as a function of the laser excitation energy. The inset shows the region around the lasing threshold with a higher resolution. The lasing threshold for this geometry (polarization of exciting beam along fiber axis) is at $\approx 75\,\mu\mathrm{J/cm^2}$ and the laser line width at the threshold is ≈ 0.2 nm.

$\approx 75\,\mu\mathrm{J/cm^2}$. This value is comparable to the lasing threshold for WGM lasing in spherical nematic droplets as reported by Humar and Muševič [5] and is also comparable to recently reported low-threshold lasing in dye-doped polymer fibers [21]. When the polarization of the pumping beam makes an angle of 45° or 90° to the fiber axis, the observed thresholds are at $\approx 0.9\,\mathrm{mJ/cm^2}$ and $\approx 2\,\mathrm{mJ/cm^2}$, which is one or two orders of magnitude higher.

4. Conclusions

This work demonstrates for the first time optical wave-guiding and WGM lasing from organic fiber-like liquid crystalline structures, self assembled at the interface between a thermotropic smectic-A liquid crystal and a cationic surfactant aqueous solution. Together with previously reported tunable liquid crystal microcavities and dye-liquid-crystal microlasers, this is a demonstration of another member from a family of new photonic microdevices realized from soft matter in just past four years. This family of photonic devices is entirely self-assembled from complex liquids and uses surface tension and material elasticity engineering to tune the devices' shape and functionality. Unlike hard matter photonic devices, soft matter devices are assembled from ordered complex fluids, where the topology of the confined matter is important. In future, these devices could form structured soft-matter that could be morphed by light of the laser tweezers and could be used to process light similar to the hard-matter photonic circuits. We expect that the topological properties of these soft matter microstructures could have potential impact on the topology of the light field, resulting in an entirely new class of topological photonic devices. Furthermore, the advantage of soft-matter approach is the ease of manufacturing

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photonic microobjects with perfectly smooth interfaces and in the future one could envisage programmable growth and healing of soft matter photonic circuitry mimicking the processes in living organisms.

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