Optical waveguiding in bent-core liquid-crystal filaments

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We demonstrate optical waveguiding in recently discovered free-standing bent-core liquid-crystal filaments. The bent-core liquid-crystal molecules in air self-assemble into a cylindrical geometry that is “solidlike” along the radial direction of the filament and liquid in the axial direction of the filament. These filaments are unique not only because they are fluids, but also because they are anisotropic. For this reason, their waveguiding properties not predictable need to be characterized. The light power transmitted through the filament was found to be independent of temperature from 180 °C to near room temperature. Initial defects of newly pulled filaments were found to self-anneal, thus leaving defect-free fibers, where light scattering was found to be insignificant. The absorbance was found to be strongly wavelength dependent in the visible regime and relatively small in the infrared range. A self-assembled optical waveguide with self-annealing fluid properties may have promising applications in optical communications or in optical microchips.

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I. INTRODUCTION

In 1996, it was discovered that not only rod-shape (calamitic) or discotic molecules form liquid crystals, but bent-core (bowlike or banana-shaped) molecules do also.[1] This discovery has opened up a major new and exciting dimension in the science of thermotropic liquid crystals. Seminal findings, with broad implications for the general field of soft condensed matter, include the observations of ferroelectricity and spontaneous chiral symmetry breaking in smectic phases composed of molecules that are not intrinsically chiral [2]. A similarly important observation was that in the B_7 phase [3] (that exhibits helical filaments on cooling from the isotropic fluid), the material can form free-standing strands [4], just like columnar liquid crystals of disk-shaped molecules [5]. These observations were confirmed with other B_7 materials [6] and subsequently it was found that B_2-type banana smectics (polar-tilted fluid smectic phase) also formed strands of fibers [7]. Although the most stable fibers with aspect ratio λ as large as 5000 form in the B_7 phase, fibers in the B_2 phase also have aspect ratios $\lambda > 100$. This is well above the Rayleigh-Plateau limit ($\lambda = \pi$) of Newtonian isotropic fluids [8] and of the nematic and smectic liquid crystals of rod-shaped molecule [9] and of columnar phase of disk-shaped molecules [5], which have $\lambda \sim 3.1$, 4, and 100, respectively. The mechanical and electrical properties of B_7- and B_2-type fibers are relatively well characterized [10–12] and understood [13], but concerning their optical properties, we only know that single filaments are highly birefringent along the axis of the filament [7]. Although one can anticipate that a fiber with a refractive index about 1.6 has waveguiding properties for visible light and recent theory [14] actually predicts that the forward propagation of smectic fibers behaves similarly to standard waveguides, one obviously needs to check these predictions experimentally, especially because it is not trivial whether director fluctuations would not cause significant light scattering. This is of concern because many liquid crystalline materials tend to scatter light strongly over length scales useful for optical filaments due to either director fluctuations or defect formation.

In this paper, we report an experimental study of the waveguiding properties of bent-core filaments. We show that they can guide light both in the crystal and liquid crystalline phases. The results indicate that in the infrared range, bent-core filaments may be interesting candidates for self-assembled and self-annealing organic optical waveguides.

II. EXPERIMENTS

For our studies, we chose the bent-core liquid-crystal material 2-nitro-1,3-phenylene-bis[4-(4–8-alkyloxyphenyliminomethyl) benzoates [4] [see Fig. 1(a)] because these were found [4] to form the most stable fiber. It has a wide range enantiotropic B_7 phase with a phase sequence Cr 116 °C B_7 177 °C I. In the B_7 phase, the molecules form two-dimensional fluid planes or layers that spontaneously self-assemble to concentric layer structure that is solidlike in the radial direction of the filament and liquid in the axial direction [see Fig. 1(b)]. Such filaments have well-defined diameters of a few microns [7]. The experimental setup to pull the fibers and to study the optical waveguiding properties is shown in Fig. 1(c).

Both a 70mW Nd:yttrium aluminum garnet (YAG) Viasho 532nm laser and a 30mW Melles-Griot HeNe 633nm laser were used in the studies, but quantitative measurements were done only at $\lambda = 532$ nm. The linearly polarized beam of the laser is incident upon a 10x microscope objective and then coupled into a 1 m Corning single mode (SMF-28) optical fiber (with an 8 μm core and 125 μm cladding diameter). The free end of the optic fiber was glued into a 1 mm capillary tube. The capillary tube is used to support the free end of the optical fiber in an Instec HS1 heat stage and was mounted into a vertical micrometer zy stage. Acetone was used to remove 6 cm of the plastic coating from the optical fiber end that is inserted into the heat stage to avoid melting. The zy stage is mounted to the deck of an Olympus BX50

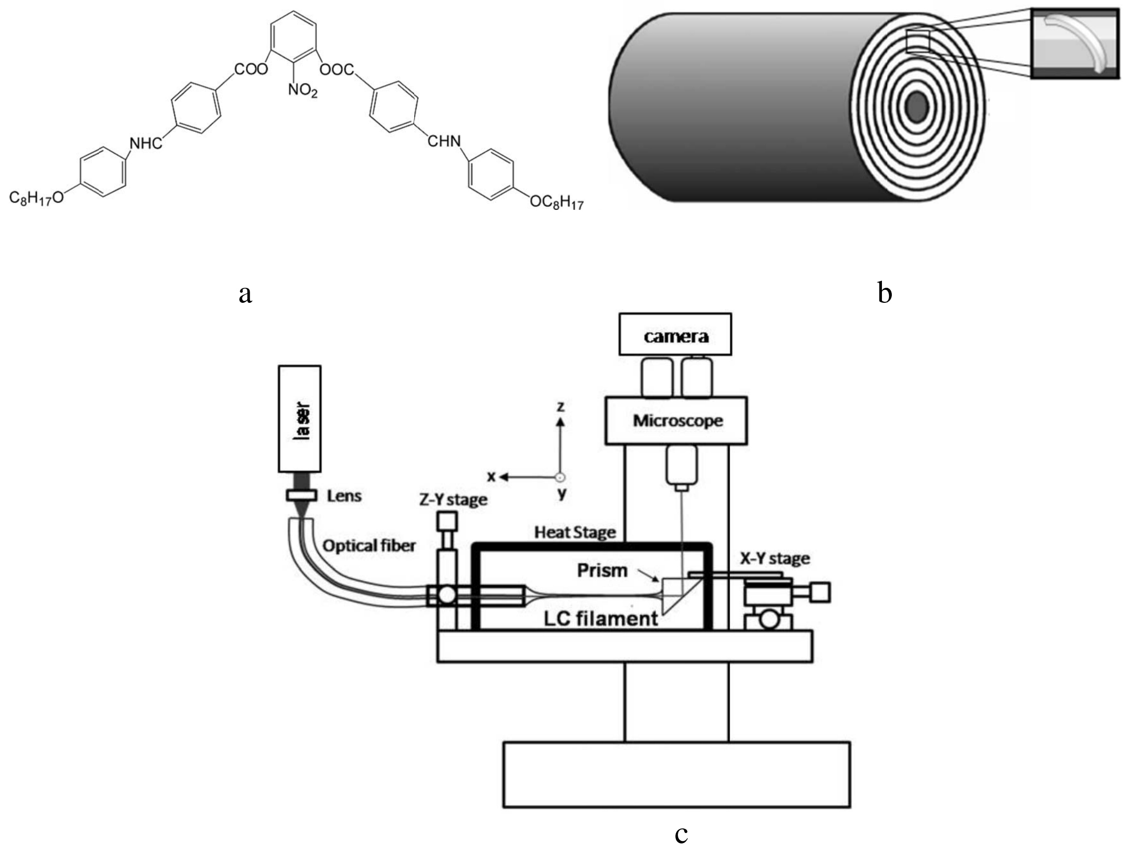

FIG. 1. (a) Illustration of the molecular structure of the bent-core liquid-crystal material 2-nitro-1,3-phenylene-bis[4-(4–8-alkyloxyphenyliminomethyl) benzoates (Ref. [4]). (b) The smectic layer arrangement in the filament (Ref. [7]). (c) Schematic of the experimental apparatus.

microscope. The capillary tube, with the optical fiber inside, was inserted into the heat stage. A 45-45-90 prism was mounted onto an *xy* stage. The top of the prism, the liquid-crystal filament, and the optical fiber can be seen through a window in the heat stage by simply looking at the reflection from the mirrored prism face. The images were recorded with a Sanyo charge coupled device (CCD) VCC-3972P camera and a Sony RDR-HX520 DVD/HDD recorder. Images from each video were extracted using Frameshots Video Frame Capture® program.

The bent-core liquid-crystal material in the crystalline phase was placed onto the tip of the optic fiber in the heat stage. Then, after melting into the isotropic liquid phase, it was cooled to 175 °C, slightly below the isotropic B_7 phase transition. The prism is then translated in the $+x$ direction to touch the optic fiber tip with the liquid-crystal droplet. Once the liquid crystal touches the face of the prism, the sample was pulled back in the $-x$ direction to extrude a liquid-crystal filament as shown in Fig. 2.

Figure 2(a) shows the optical fiber, liquid-crystal filament, and prism face. Figure 2(b) is the image that is reflected from the prism face inside the heat stage. The large bottom laser spot is from the laser light scattered from the optic fiber to the prism. The small top laser spot is laser light that has propagated through the liquid-crystal filament. If the prism was translated in the $\pm y$ direction, the angle between the optical fiber and the liquid-crystal filament increased. Corre-

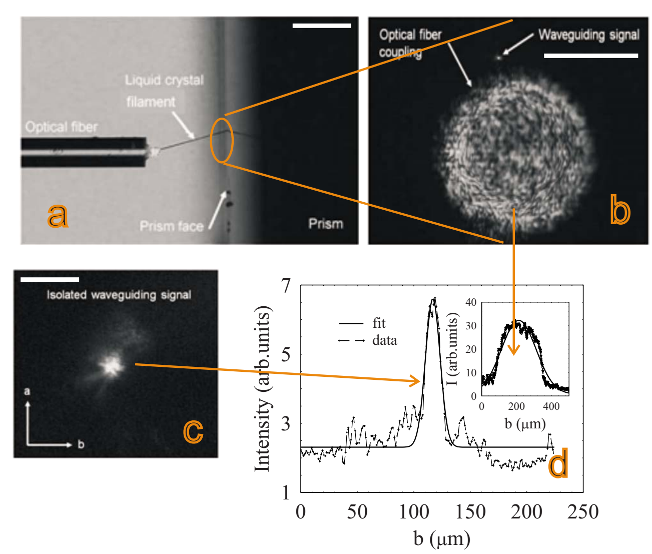

FIG. 2. (Color online) (a) Optic fiber, liquid-crystal filament, prism face held at 170 °C and probed at 633nm. (b) Waveguiding signal from liquid-crystal filament (top small spot) and the coupled light between the optical fiber and the prism (bottom large spot). (c) Isolated waveguiding signal from the bent-core liquid-crystal filament. White bars in (a) and (b) indicate 200 μm length and in (c) 20 μm length. (d) Intensity profile of the beam transmitted from the bent-core liquid-crystal fiber of 6.3 μm (main pane) and from the glass optical fiber (insert).

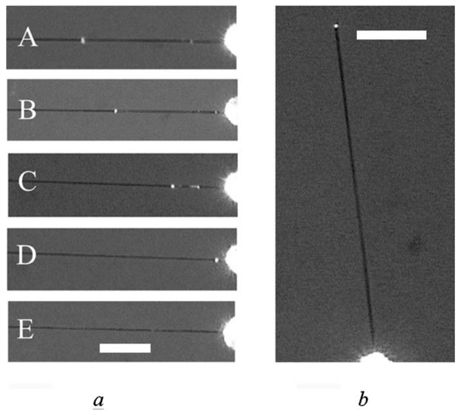

FIG. 3. (a) Translation of a defect (position of defect is a bright spot in the filament) in a newly pulled filament vs time. The time in between images A–E is ~4–5 min. (b) Free-standing filament of bent-core mesogens in the Cr phase, held at 35 °C. White bars indicate 50 μm length.

spondingly, one observes the small top laser spot translate in the same direction showing that it has propagated through the liquid-crystal filament. Interestingly, if the angle of the filament became too large, the filament would slide to decrease that angle. This was observed in the microscope as the small spot would also move in correspondence with the filaments movement. Figure 2(c) is the isolated and cropped image of the transmitted signal from the filament.

To determine the spatial distribution of waveguided signal from the filament, the cropped images from the camera were analyzed. A LABVIEW® program was used to sum the intensity values of the pixels (0–256) in the vertical direction [a direction in Fig. 2(c)]. The summed intensity for each a column is plotted as a function of the b direction in Fig. 2(a). The intensity profile was fitted with a Gaussian distribution on a constant background, $I=Ae^{(-(b-B)^2/C)}+D$, where A, B, C, and D are fit parameters. Three images were processed for each measurement to obtain the average fit parameters. The light spot coming from the fiber actually shows the cross section of the meniscus at the fiber-prism interface, which is due to the fluid nature of the fiber and is larger than of the diameter of the fiber. The background signal is due to the curvature of the meniscus, where the light is strongly scattered. As one can see in Fig. 3(b), the diameter of the light coming out from a broken end of the fiber without meniscus is the same as of the diameter of the fiber and the light is not scattered there. As shown in the insert of Fig. 2(a), the profile of the single mode optical fiber is also Gaussian (there, the width is related to the diverging beam to the prism), indicating that the bent-core liquid-crystal fiber behaves as a single mode fiber. The average transmitted power P was then calculated from these parameters as $P=\int_{-C}^{+C}Ae^{[-(b-B)^2/C]}db$ and was measured as a function of temperature. It was found (figure not shown) that within the measurement error, the transmitted power from the filament appears to remain constant from the top range B_7 phase (177 °C) even through the transition into the crystal phase (116 °C) is down to room temperature. This remarkably wide temperature range is quite unexpected, and promising for applications.

Shortly after pulling a new filament, we observed defects that scatter light and appear as bright spots in Fig. 3(a). These defects are moving along the axis of the filament and eventually get expelled as shown in A–E of Fig. 3(a). This suggests that the filament is self-annealing and it is possible to form a defect-free structure, given sufficient time. To demonstrate the possibility of realizing defect-free filaments even in the crystal phase, a filament was cooled to 35 °C and broken as shown in Fig. 3(b). The filament appears black; the only light escaping is from the input and terminal ends. This demonstrates that the scattering of the light out of the filament surface due to director fluctuations, defects, or other mechanisms is insignificant.

The integrated light intensity of the spot at the function of the length is shown in Fig. 4(a). Although the measured data scatter considerably due to the length dependence of the size

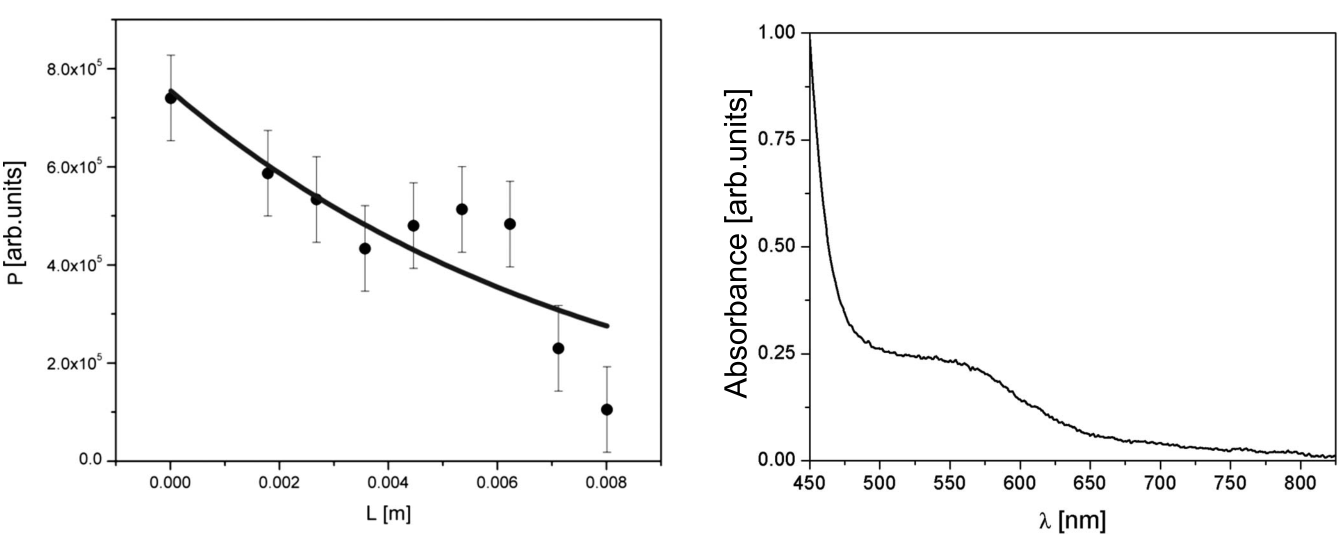

FIG. 4. (a) Transmitted power vs filament length at 175 °C at 532 nm of a 6.5 μm fiber. The experimental data were fitted by an exponential. (b) Absorbance vs wavelength measured by a Ocean Optics spectrophotometer, 5 mm path-length cuvette where 2.1 mg of 2-nitro-1,3-phenylene-bis[4-(4–8-alkyloxyphenyliminomethyl) benzoates] was mixed into 0.5ml of ethyl acetate.

of the meniscus and shape and due to defects, they can be fitted by a single exponential. This shows that the decrease is dominated by the absorption of the light due to the yellowish color of the material. The absorption spectrum of the material was measured by an Ocean Optics spectrophotometer in the 400–800 nm range and shown in Fig. 4(b). One sees that absorbance is high in the blue range (~ 1), moderate in the rest of the visible regime and negligible in the near IR range. This shows that only short fibers of this material can be suitable for guiding light in the visible range, but in the infrared range it might be useful for optical computing or even in telecommunications.

To summarize, optical wave guiding in bent-core liquid-crystal filaments of bent-core molecules was demonstrated for 5–10 μm diameter fluid fibers in centimeter length scales. The temperature dependence of the light power transmitted by the filament was constant even through the liquid crystal to crystal transition. Although waveguiding could be predicted, we found that defect self-annealing leads to monodomain liquid-crystal filaments that cause no observable light scattering neither in the liquid crystal nor the crystal phases. This is partially due to the layer structure that suppresses orientational fluctuations and partially due to bent shape of the molecules that suppress out of layer fluctuations. Although absorbance was shown to exist in the visible regime, it seems to be negligible in the infrared range, where optical communication is usually operating. Finally, we note that besides being a fluid and anisotropic, bent-core filaments have other unique properties to take advantage, such as self-assembly into well-defined diameter and ferroelectricity, which may have interesting nonlinear properties such as second-harmonic generation. Future studies will include measurements of this material in the infrared range and other bent-core materials that are transparent in the visible range.

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