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Molding optical waveguides with nematicons

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We demonstrate that a crosslinked polymer network can permanently mold spatial soliton waveguides into nematic liquid crystals for the confinements of optical signals, paving the way towards a novel bottom-up technology for guided-wave photonics with beam-shaped circuits. Moreover, we show that stabilization through polymerization is effective in quenching the trajectory fluctuations of such spatial solitons –nematicons-, allowing also their real-time use for all-optical signal processing.

Waveguides play an essential role in photonics, as they allow light to travel without diffraction, thereby confining optical signals and maintaining their intensity and transverse profile. Optical fibers in communications and medicine, guided-wave elements in optoelectronics, sensing, and signal processing are among prominent examples. Waveguides are usually fabricated by top-down methods, including light-based ones such as direct laser writing; these methods modify the refractive index distribution of the material by external means and ensure transverse light confinement at desired wavelengths. Conversely, in self-focusing nonlinear optical media, beam self-action can compensate diffraction, forming self-guided beams, also known as spatial solitons.[1, 2] This bottom-up approach is real-time; light-induced soliton waveguides enable to simultaneously realize and monitor guided-wave channels confining other optical signals, as well. While the nonlinear response of most materials to light is rather small and large intensities are required to sustain solitons,[3,4] nematic liquid crystals can support stable solitons, termed "nematicons",[5] at remarkably low, even sub-mW, powers and over large propagation distances (mm).[6-7] Nematicon waveguides thus combine beam self-action and material self-organization with low-power excitation, all beneficial properties for all-optical circuitry. Yet, they tend to suffer from trajectory fluctuations as a function of time and propagation, which remains an unsolved critical issue. In this Communication, we show that the combination of nematic liquid crystals with polymerizable materials provides new opportunities for the fabrication of stable beam-shaped waveguides. We first demonstrate that polymer stabilization is effective in quenching nematicon fluctuations, while it maintains unaltered the possibility of all-optically defining waveguides in real time. Then, we show that nematicon waveguides can be permanently molded into the medium via a crosslinked polymer network and persist after turning off the forming beam. With such findings, we introduce a new platform for guided-wave photonics in

liquid-crystalline matter, a platform with unprecedented versatility and a wealth of possibilities for complex optical circuitry, signal addressing, and processing.

Nematic liquid crystals (NLC) are uniaxial dielectrics, i.e., their optical properties are different for light polarized either in the plane defined by the molecular director and the wavevector (extraordinary polarization) or orthogonal to it (ordinary polarization), making them an important class of materials for various photonic applications.[8] When exposed to light, the NLC molecules experience a torque that tends to reorient them towards the direction of the electric field.[9,10] For finite light beams with bell-shaped profile, this reorientational response leads to a transversely graded distribution of the refractive index for the extraordinary polarization, which gives rise to nematicon waveguides.[5] Such reorientational nonlinearity is saturable and nonlocal, ensuring the stability of nematicons.[5,11] Nematicons can also confine and route co-polarized signals at shorter and longer wavelengths than the soliton itself;[5] hence, they entail an entire class of guided-wave circuits induced and controlled by propagating beams, including bistable elements [12] as well as all-optical routers and interconnects for optical processing.[13–16] Nonetheless, since they stem from beam-induced reorientation competing with the director distribution determined by the sample boundaries, nematicons are subject to restoring elastic forces as well as to temperature and torque fluctuations.[17] As a result, they exhibit trajectory fluctuations in the transverse direction, particularly when high optical powers are used to increase the index contrast or to maintain the confinement over extended propagation distances, the latter typically limited by scattering losses.[18] Early attempts to reduce these fluctuations were carried out by Blach *et al.* in voltage-biased cells with polymer-stabilized nematics,[19] and by Henninot *et al.* in the presence of a longitudinal voltage along the direction of propagation.[20]

Solitons form in a dynamic process. Hence, nematicon waveguides only exist as long as the defining beam propagates in the medium (or shortly thereafter until material relaxation has completed). They exhibit a fully flexible transverse (graded index) profile and

longitudinal shape (trajectory), which can be modified through external stimuli such as electric fields,[13,15,21] as well as through the interaction with other beams,[22,23] offering a wide range of possibilities for realizing three-dimensional circuits and interconnects such as in-plane and skew bends, spirals, junctions and splitters.[5] Hence, it would be extremely beneficial or even strategic to acquire the ability to "freeze" such real-time structures into permanent ones crafted in the medium. Previous work on beam-defined waveguides in polymer-based compounds, however, was based on the use of one and the same beam to simultaneously define and polymerize the guiding channels at a resonant (absorbed) wavelength, hampering the potential of the approach due to attenuation and the consequent lack of size and profile preservation over extended distances.[24-26] Herein, in order to render permanent the nematicon structures in the medium while preserving the full versatility of the technique we decouple soliton generation from the material polymerization for the first time, carrying out an independent/subsequent freezing process with ultraviolet exposure.

The sample geometry and our experimental setup are sketched in **Figure 1**. A planar cell holding a 100 μm-thick layer of NLC was assembled with two parallel polyimide-coated mechanically-rubbed glass slides in order to ensure uniform alignment of the molecular director **n** in the plane *yz* at 45° with respect to *z* (Figure 1(a)). This initial alignment provides a large reorientational nonlinearity in response to extraordinarily-polarized waves. Similarly treated thin cover slides sealed the cell orthogonally to *z* and provided molecular anchoring along *y*. A doped NLC mixture, comprising 4-cyano-4'-pentylbiphenyl (5CB) as the host NLC and photo-polymerizable compound(s) as the guest dopants, was infiltrated into the cell by capillarity (e.g., as in Karimi *et al.*[27]). A *y*-polarized Gaussian input beam of wavelength λ=1.064 μm and waist of about 4 μm was launched in the mid-plane of the sample (at 50μm from the upper and lower interfaces) with a planar phase-front, coupling to extraordinary waves with wave vector along *z*. At sufficient powers, this near-infrared (NIR) beam can induce appreciable molecular reorientation and generate a nematicon.[5,21] A photograph of

such a nematicon excited in undoped NLC by a 4mW NIR beam is shown in Figure 1(c,bottom), as obtained by monitoring the beam evolution in the *yz* plane with an optical microscope and imaging the light scattered towards *x* with a CCD camera. The Poynting vector exhibits a finite walk-off (nearly 7°) due to the birefringence of the host.[21]

The photo-polymerizable guest components used were a monoacrylate 6-(4-cyanobiphenyl-4'-yloxy) hexyl acrylate (**1**; Synthon Chemicals) and a crosslinking molecule 1,6-hexanediol dimethacrylate (**2**; Polysciences), shown in Figure 1(d). Both compounds were used without further purification. The choice of materials was motivated by the earlier work of Shishido's group, [28] showing that polymer stabilization (corresponding to 5CB:**1** mixture) can enhance the so-called Jánossy effect in dye-doped liquid crystals,[29] while crosslinking (corresponding to the 5CB:**1**:**2** mixture) allows inscribing polarization-selective, planar microlens arrays.[30] In an effort to utilize these earlier observations in the context of light-defined waveguides, we addressed the two main challenges described above: (i) quenching fluctuations of soliton trajectories in polymer-stabilized 5CB:**1** mixtures for real-time applications; (ii) molding nematicon waveguides in crosslinked NLC:**1**:**2** mixtures, rendering permanent the confining structures even after turning off the forming beam. In case (i) we chose 10 mol-% concentration of **1** [31] and no photo-initiator [32] in order to slow down the polymerization process as to ease the monitoring of soliton trajectories versus time. In case (ii) 3 mol-% of **2** was added to the 5CB:**1** mixture, as was 0.1 mol-% of UV photo-initiator (Irgacure 651, Sigma-Aldrich) in order to speed up the formation of crosslinked polymer network and molding of the nematicon waveguides.

Typical experimental results on reducing the trajectory fluctuations of nematicons are shown in **Figure 2**. When excited by a 3mW beam (typical excitation level), the nematicon paths exhibited transverse fluctuations in the observation plane *yz*, as displayed in Figure 2(a), plotting several trajectory realizations within an observation window of about 4.5s. The soliton trajectories were dispersed within a two-dimensional section of a cone in *yz*, with

output signal delivered to well separate locations in the transverse coordinates. Such spread in the location of the output signal is certainly detrimental for optical interconnects. We then progressively exposed the sample to an incoherent source of continuous-wave ultraviolet light (UV LED, 365 nm, 5 mW/cm^2), and monitored the soliton trajectories at fixed intervals of 4.5 s versus UV illumination. As displayed in Figure 2(b-c), the fluctuations underwent effective quenching versus irradiation, with a variance in transverse displacement eventually reduced by a factor ≈ 3. The process saturated in about two hours, resulting in a nearly time-independent nematicon. After polymer stabilization and UV exposure, remarkably, the material was still mobile enough (hence nonlinear) to support nematicon formation over and over again, with the same acquired path steadiness. This finding proves the potential of polymer stabilization even for non-dissipative nonlinear optics, i.e., Kerr and Kerr-like effects with non-absorbed light.

We then proceeded to permanent molding of nematicon waveguides. The key was to employ a non-resonant NIR beam for defining the nematicon and UV illumination for crosslinking, i. e., we decoupled waveguide generation and photo-polymerization processes. Therefore, we first all-optically defined and monitored the waveguides before making them permanent by photo-polymerization and eventually operating at various wavelengths/intensities. We performed measurements as before, but due to the presence of the photo-initiator, with shorter UV exposure times at irradiance of ≈ 1 mW/cm^2. **Figure 3**(a)-(c) shows a series of photographs of a 3mW-soliton evolving in the propagation plane of the cell for increasing exposure intervals: for times in excess of 4 minutes, the soliton waveguide lost its real-time character and was completely molded in the sample, without any further changes after prolonged UV illumination. This is apparent in Figure 3(e)-(h), plotting several trajectory realizations for UV exposure times as in Figure 3(a)-(d): the fluctuations reduce and eventually quench within a couple of minutes. For times in excess of 4 minutes, no changes in

trajectory could be observed any longer, as the nematicon waveguide had acquired a permanent nature, i.e., it was carved in the NLC by polymerization.

The results of Figure 3 also show that our material system and polymerization process can be further optimized. For example, the waveguide trajectories of Figure 3(b)-(c) deviate from straight; these deviations can be attributed to localized phase gradients and walk-off changes occurring during the polymerization process as the optical properties (refractive indices and birefringence) of the guest-host evolve during polymerization. Furthermore, new scattering domains appear after complete polymerization, as visible along the beam in Figure 3(d); such domains, analogous to those observed after UV-induced phase separation in NLC-monomer guest-host systems,[33] are likely to increase the disorder in the sample and lower the NLC order parameter. The images in Figure 3 allowed us to estimate an added randomness with defects as large as $\approx 5\mu m$. This was further confirmed by the analysis of several realizations of diffracting NIR beams either ordinarily or extraordinarily polarized in homogeneously UV-cured areas (without soliton channel); increased disorder enhanced scattering and, therefore, diffraction.[34]

In **Figure 4** we illustrate the guiding operation of the nematicon molded waveguide both in the near-infrared and in the visible. The graph in Figure 4(a) shows the measured NIR beam size w versus propagation distance *z* in the sample, comparing the linear diffraction at low power in a homogeneous sample before polymerization (dashed line) with the transverse confinement of an input launched in the nematicon waveguide after UV curing (solid lines). Before exposure to UV light, the weak beam is seen to diffract; after the waveguide has been molded, the in-coupled light is clearly trapped regardless of its input power (e.g., at 50 μW and 3, 10 and 20 mW, respectively), even for excitations much higher than initially employed to launch the nematicon in the system. Noteworthy, despite that polymerization transformed the dynamic scattering of light into scattering from disorder, the throughput of the nematicon waveguide remained of the same magnitude, close to 30%, with an attenuation of 1.54

dB/mm. Finally, in order to prove the relevance of this approach for guided-wave photonics, we launched a visible probe beam at λ=632.8nm into the polymerized sample, with the same linear polarization as that of the NIR input. Figure 4(b)-(c) shows that, when launched outside the nematicon channel, the red laser beam diffracts in the bulk of the sample, with transverse spreading enhanced by the increased randomness; when coupled into the molded waveguide, conversely, the probe is guided along it and propagates along the path previously defined by the NIR nematicon. In the latter case the scattering was almost doubled as compared to the NIR, with propagation losses of ≈ 3.19 dB/mm.

In conclusion, we have demonstrated that judicious doping of nematic liquid crystals with monoacrylates, followed by polymer stabilization, allows reducing and eventually eliminating the fluctuations of spatial soliton trajectories, providing steadiness to real-time light-defined nematicons and the associated optical waveguides. We have shown for the first time that complete UV curing permits to permanently mold soliton-defined guided-wave elements for their subsequent use with optical signals, independently of their power. While material optimization is certainly required and is underway in order to minimize scattering losses, this novel technological platform for bottom-up waveguide fabrication with beam shaped profile paves the way to the realization of light-defined beam-tailored guided-wave photonics in nematic liquid crystals, whereby multiple nonlinear beams could be launched and interact to implement simple as well as complex circuits for signal interconnects and processing, including, e.g., bends,[13,15,22] junctions,[23] multiplexers.[14]

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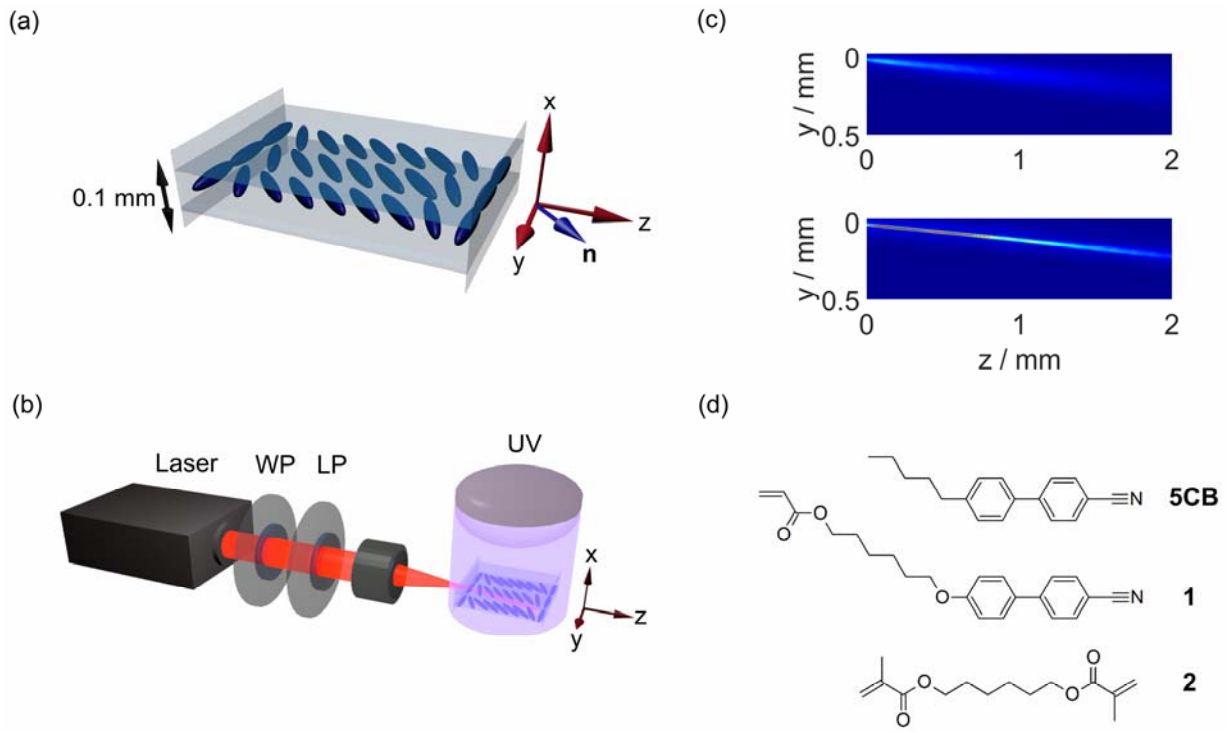

Figure 1. (a) 3D view of the planar cell filled with NLC, (b) basic sketch of the experimental setup, with half-wave plate WP and linear polarizer LP in front of a microscope objective. (c) NIR beam evolution in *yz* when diffracting at low power (50 μW, top) and when self-confined at high power (4 mW, bottom). (d) Molecular structures of the components indicated in the text and used to prepare the guest-host mixtures.

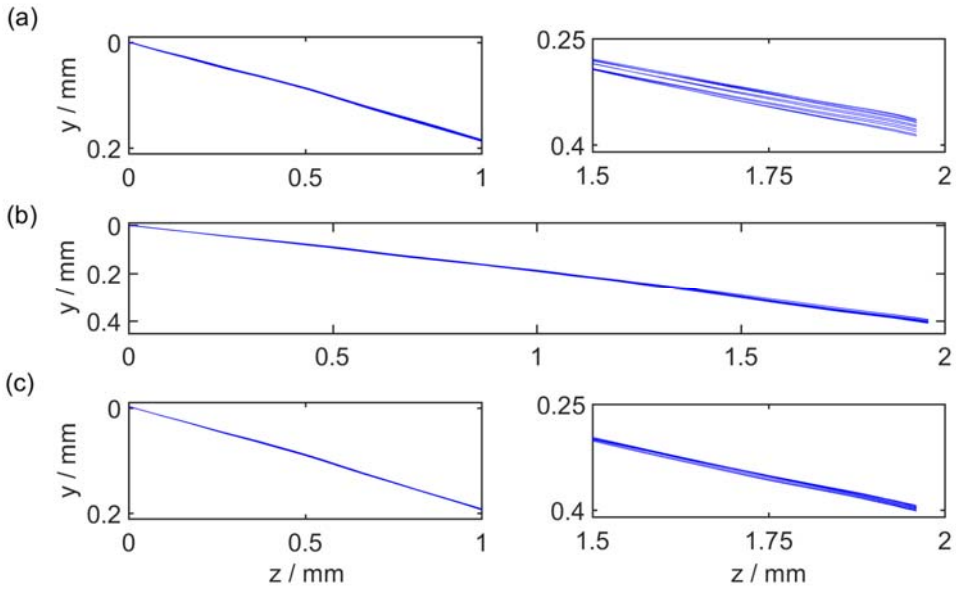

Figure 2. Trajectory realizations of a near-infrared nematicon (fixed input) within a time period of 4.5 s, (a) before UV illumination, (b) after one and (c) two hour exposure, respectively. Compared to (b), the graphs in (a) and (c) are magnified and split into initial and final portions, in order to enhance the resulting visibility of the path dispersion.

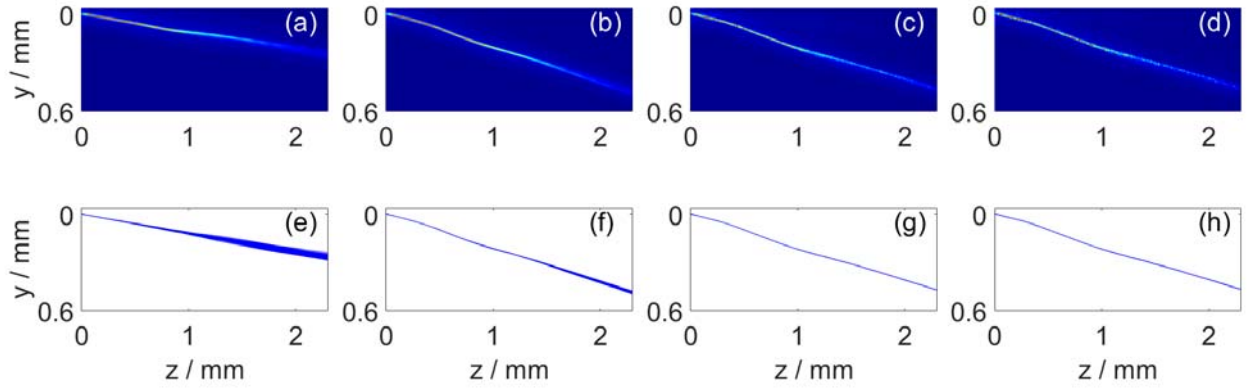

Figure 3. Snapshots of nematicon evolution (top) and trajectory realizations (bottom) in the plane *yz* (a,e) before UV exposure, (b,f) after 120 and (c,g) 240 s of UV exposure, (d,h) after curing.

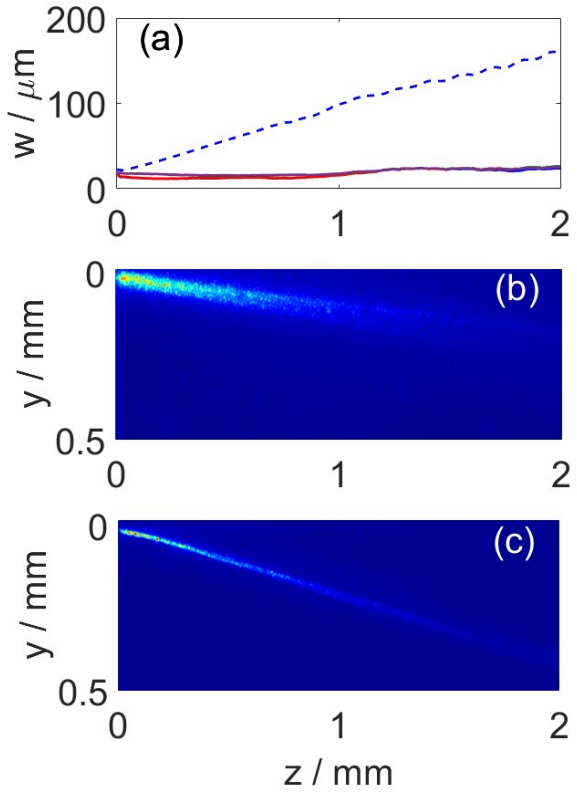

Figure 4. Permanent waveguide operation. (a) Measured NIR beam (1.064 μm) width versus propagation distance before (dashed blue line) and after complete (240s) UV curing (solid lines) for various input powers, from 0.05 (blue) to 3 (red), 10 (green), 20 mW (violet), respectively. The solid lines are nearly indistinguishable owing to guidance. (b) Photograph of the extraordinarily-polarized probe (0.6328 μm) beam diffracting when launched in the bulk of the cured sample. (c) Photograph of the extraordinarily-polarized probe beam confined within the nematicon molded waveguide. The increased scattering associated with added randomness makes the red beam appear much wider in (b) and its intensity in (c) decay faster versus propagation.