

All-Optically Driven Optofluidic Light Modulator

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Liquid crystals (LCs) are widely recognized for unique physical and optical properties and remarkable ability to modulate light beams. Traditional methods for actuating LCs by electric fields rely on the use of metallic electrodes, which face limitations in the spatial resolution of LCs actuation and their effective integration into optofluidic devices. In this work, an innovative approach is presented to realize a fully integrated optofluidic light modulator, capable of modifying the optical properties of a light beam propagating through an optical waveguide and coupled with a microfluidic channel. Specifically, the optoelectronic properties of lithium niobate (LN) are leveraged to control the orientation of LCs molecules confined in the microfluidic channel without the need for fixed metallic structures, but solely exploiting light-induced electric fields. It is demonstrated that the light-driven orientation of the LCs can efficiently modulate both the intensity and polarization of the light beam propagating through the waveguide, while the desired amplitude and time-constant of the output optical signal can be achieved by tuning the compositional properties of the LN substrate. The modulation of the guided beam is completely defined and controlled by a pump light source, assuring a high degree of reconfigurability and compatibility of the final optofluidic lab-on-a-chip system.

molecules was achieved solely by controlling their flow within microfluidic channels, paving the way for high-potential optical applications, integrated into fluidic circuits.^[8–10] Recently, the use of LCs has also broadened to biosensing and biomedicine,^[11–13] research areas that are greatly benefitting from microfluidic technology. Despite these remarkable results, the capability to reorient LCs with the flow is not fast or tunable enough to compete with standard full-optical applications. In addition, this method suffers from the need for external, bulky pumps, contrary to integrated optofluidic devices, and it has not yet been coupled efficiently with photonic structures, such as optical waveguides. In principle, these limits can be easily overcome by orienting LCs molecules with the use of electric fields, as witnessed by the breakthrough of technologies based on LCs displays.^[14] However, metallic electrodes are required to properly create the desired electric fields, and their integration in microfluidic circuits heavily limits the degree of reconfigurability and

versatility of the final device. Among others, optoelectronics represents a promising alternative way for generating electric fields able to efficiently actuate liquid crystals. In this case, virtual electrodes are entirely created and shaped by light beams on the surface of optically active materials, causing strong light-induced electric fields that can be used to actuate liquid crystals.^[15]

Ferroelectric crystals presenting either the pyroelectric^[16,17] or the bulk photovoltaic effect^[18] have been shown to be optimal substrates for optoelectronic applications.^[19] Indeed, in these materials photogenerated electric fields can be easily tailored by

1. Introduction

Liquid crystals (LCs) have contributed significantly to the growth of the fields of optics and fluidics, due to their liquid nature and unique interaction with light, becoming ubiquitous materials in these research areas.^[1–3] With the advent of microfluidics, LCs have demonstrated their potential to investigate several interesting phenomena, such as unique flow patterns and dynamics,^[4,5] the emergence of topological states,^[6] and super-screening effects.^[7] In these studies, the reorientation of LC

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laser beams, achieving values up to 10^6 V m^{-1} with moderate light intensity ($I > 10^2 \text{ W m}^{-2}$). LCs of different nature have been controlled using optothermal heat and pyroelectric fields, as in the case of ferroelectric nematic liquid crystals.^[20] The photovoltaic effect has achieved ever more success, as shown by the development of nematic liquid crystal cells driven by light,^[21–23] the manipulation of the chiral liquid crystals, the induction of controlled defects, the manipulation of liquid crystal droplets and the development of optically addressable light modulators.^[24–28] These methods have also been recently investigated in microfluidic configurations, with microchannels directly integrated into ferroelectric crystals,^[29] opening new manipulation opportunities for lab-on-a-chip devices.^[30,31] Among ferroelectric crystals, lithium niobate (LN) is well known for its optical properties,^[32] making it a key material for photonic applications,^[33–36] as well as for a new concept of optofluidic devices.^[37–39] In particular, the coupling of optical systems and microfluidic channels in LN has boosted the investigation of physical phenomena based on light-matter interaction occurring inside microfluidic circuits.^[40–42] Besides sensing applications, the incorporation of optical waveguides into a microfluidic circuit allows the local and detailed characterization of the physical properties at the liquid–liquid and liquid–solid interfaces.^[43] The realization of optical waveguides in this material has been widely investigated, achieving performant and complex designs, as interferometric interactions,^[41] using various experimental approaches.^[44–47] Furthermore, the integration of microfluidic channels in lithium niobate is increasingly becoming comparable to other conventional materials and various techniques have been demonstrated to be efficient in microstructuring this material.^[48–50]

In this work, we take advantage of the synergy between liquid crystals and lithium niobate to realize an all-optically driven light modulator that is fully integrated into an optofluidic device. With this approach, the limitations typical of bulky LCs-based systems are overcome, assuring a high degree of reconfigurability of the system as well as the possibility of integrating it into portable lab-on-a-chip devices. Specifically, we employ LCs to control the optical response of the optofluidic device and, in a reverse direction, an external light source to actuate the orientation of the liquid crystal. Similar to a pump-probe experiment, the light carried by the waveguide acts as an optical probe, whose polarization and transmission are modulated by the orientation of the liquid crystal molecules; at the same time, a laser pump beam is exploited to induce, at the surface of the fluidic channels, electric fields strong enough to reorient the LCs optical axis.

2. Working Principle

The working principle of the optofluidic device is based on the ability of a nematic liquid crystal contained in a microfluidic circuit to modulate a light beam that propagates inside a waveguide and crosses a microfluidic channel (see Figure 1a). Similarly to technologies based on spatial light modulators, the light propagating in the device's optical waveguide is modulated by the orientation of the liquid crystal molecules. However, in this work, the electric fields used to rotate the optical axis of the liquid crystal are not created using metallic electrodes but rather induced by the illumination of the cover of the microfluidic device. In

fact, the microfluidic circuit has been realized by engraving an undoped congruent lithium niobate substrate x-cut, then sealing the fluidic circuit with a z-cut iron-doped lithium niobate crystal (Fe:LN), as depicted in Figure 1a. The typical photovoltaic effect of this material leads to the creation of a photocurrent upon proper illumination, caused by the migration of photoexcited charge carriers along the polar axis of the material (c- or z-axis). The presence of iron within the lithium niobate cover significantly enhances the photovoltaic response of the material with respect to the undoped substrate, since iron ions act as donors (Fe^{2+}) and acceptors (Fe^{3+}) of charge carriers. Therefore, illumination of the optofluidic device with a visible laser beam ($\lambda = 532 \text{ nm}$, see Figure 1a) leads to net charge accumulation on the two main faces of the Fe:LN cover crystal, in correspondence of the illuminated area, resulting in a remarkable electric field inside the material. The temporal evolution of the light-induced electric field can be determined by the one-center model,^[51] following the relations

$$E_{PV}(t) = E_{SAT}(1 - e^{-\frac{t}{\tau_{PV}}}) \quad (1)$$

$$\tau_{PV} = k \cdot r^{-1} \cdot I^{-1} \quad (2)$$

where τ_{PV} is the photovoltaic time constant and I is the light intensity. The reduction degree of the crystals, defined as the ratio between the concentration of iron sites $r = \frac{C_{\text{Fe}^{2+}}}{C_{\text{Fe}^{3+}}}$, while $k = 6 \cdot 10^3 \text{ s W m}^{-2}$ is a constant depending on the material properties (see Supporting Information for further details). For $t < \tau_{PV}$, the charge accumulation and E_{PV} are growing linearly with the illumination time t and inversely with τ_{PV} . For $t > \tau_{PV}$, the field saturates to a fixed electric field E_{SAT} , which depends on the crystal properties and the acceptors concentration $C_{\text{Fe}^{3+}}$.

In the absence of illumination, once injected inside the optofluidic device the molecules of the liquid crystal are oriented along the main direction of the channel (Figure 1b), assuming a semi-planar anchoring on LN surfaces^[4,22] with an angle relative the flow direction which could vary from 0° to 45° . When the green laser (pump source) is switched on to illuminate the device, the light-induced electric field starts to grow, and because of the crystallographic alignment of Fe:LN, its evanescent component propagates outside the material mainly perpendicular to the c-faces of the crystal.^[51] The interaction between $E_{PV}(t)$ and the liquid crystal leads to a temporal-dependent rotation of the LC molecules, whose optical axis aligns with the external electric field (Figure 1c), as described in Ref. [22]. The reorientation of the liquid crystal strongly affects the light beam propagating in the optical waveguide (called probe beam, $\lambda = 633 \text{ nm}$) and crossing the microfluidic channel. Compared to other microfluidic configurations,^[52] the probe beam is integrated into the microfluidic device, allowing for fully optofluidic lab-on-a-chip operation. When the probe beam propagating inside the input waveguide faces the microfluidic channel and travels through the LCs layers, its ordinary and extraordinary components experience a phase shift caused by the LC birefringence. In fact, the channel filled with LC acts as a waveplate, changing the polarization of the beam. As a consequence, the transmitted signal in the output waveguide exhibits a rotation in the polarization direction with respect to the input one.^[23] The magnitude and dynamics of this effect can be estimated by monitoring the intensity of the

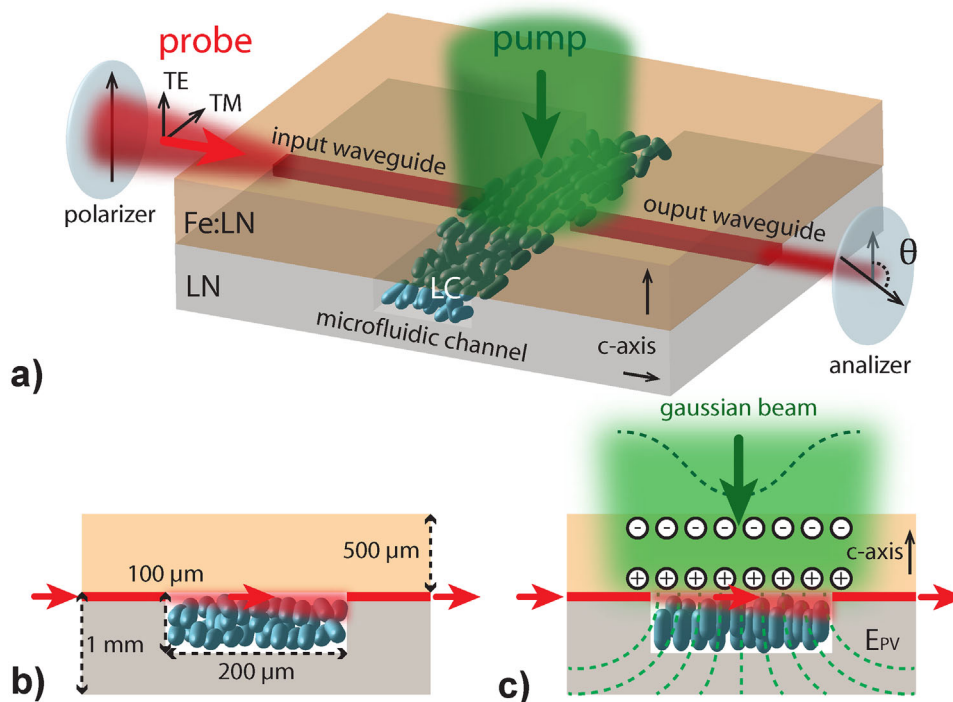


Figure 1. Working principle of the optofluidic device. a) Schematic of the optofluidic device. A probe light beam (633 nm) is polarized and then coupled into the optical input waveguide. Outcoming light from the input waveguide interacts with the liquid crystal inside the channel. After the propagation through the microfluidic channel, the light is coupled into the output waveguide. The transmitted light intensity is finally measured with a photodiode after passing through the analyzer at an angle θ with respect to the polarizer. Panels (b) and (c) show two states of the liquid crystal inside the channel before and after the actuation of the photovoltaic field with 532 nm light from the top. It is worth mentioning that the alignment of the LC molecules could be not completely planar and a pre-tilt could be present.

output light in time (I_{probe}) through a detector (photodiode) and a polarizer (called *analyzer* in the following), placed at the exit of the output waveguide (see Section 5 for further details). Specifically, θ represents the angle between the main axis of the analyzer and the input polarization of the probe beam.

3. Results and Discussion

As a first step, we have verified the response of our optofluidic device by monitoring the intensity of the probe beam I_{probe} while the main microfluidic channel is filled with an optically inactive liquid. In Figure 2a, we show the temporal evolution of I_{probe} in the presence of distilled water in the channel. As expected, water does not affect the polarization of the waveguided beam, and the intensity of the signal remains constant within 5% for several minutes, both in the presence and in the absence of the green pump light. In contrast, if the microfluidic channel is filled with 5CB liquid crystal, the intensity of the output beam increases up to about 40%, when the pump beam is switched on (Figure 2b). These tests confirm that the pump laser and the presence of the liquid crystals are fundamental requirements to observe a variation in the intensity of the probe beam, which is caused by the rotation of the LC optical axis triggered by the presence of the photovoltaic electric field.

Figure 2c shows the temporal evolution of I_{probe} during illumination with the pump laser for two different polarization states of the light entering the device. The TE and TM states indicate

that the polarization of the input light beam is vertical or horizontal, respectively. Before the pump laser is switched on the LC molecules are more parallel to the TM polarization plane, whereas they progressively align with the TE axis under the action of the light-induced electric fields. It is important to note that the output signals (I_{probe}) are normalized with respect to their initial intensity obtained in the absence of the light-induced electric fields. In fact, since initially the extraordinary axis of the LC molecules is more aligned along the horizontal direction than the vertical one, a pronounced *refractive index matching* between the waveguides and the liquid is achieved for a TM polarized light. As a consequence, the initial intensity for the TM polarization state is 10^2 times higher than TE one. For the same reason, when the analyzer inserted in front of the detector is set with the optical axis parallel to the polarization of the input beam, the light-induced rotation of the LC molecules results in a progressive decrease (increase) of the probe intensity for a TM (TE) polarized beam, as reported in Figure 2c. When the saturation field E_{SAT} is achieved, the liquid crystals are fully reoriented along the electric field lines and the transmitted intensity of the probe signal remains constant. In contrast, if the analyzer is rotated by $\pi/2$ with respect to the initial polarization of the light, I_{probe} follows the behavior reported in Figure 2d. In this configuration, the polarizer and the analyzer are perpendicular and, if the LC molecules are perfectly aligned horizontally, a null transmitted intensity is expected before switching on the pump laser. However, we observed a non null I_{probe} , caused by a non perfect planar alignment of the

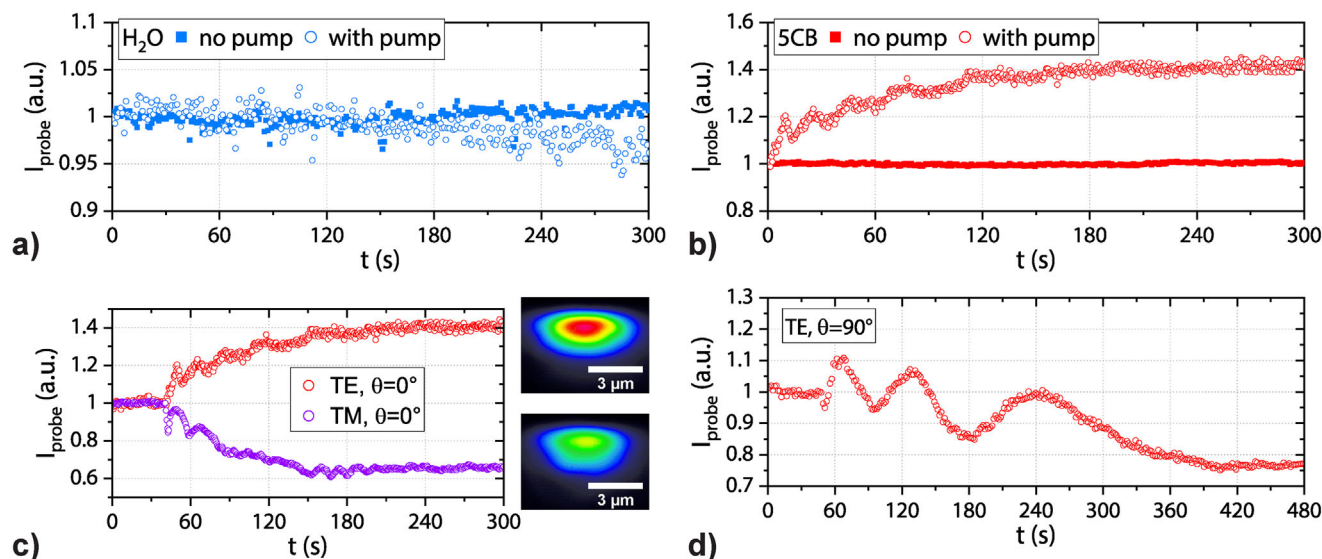


Figure 2. Measurements to characterize the pump-probe system. a) Measurements with inactive liquid, i.e., distilled water, injected in the channel instead of 5CB liquid crystals. b) Same as (a) but for 5CB inside the channel. In both (a) and (b), the graphs compare the temporal evolution of the probe with and without the pump (starting at $t=0$ s). c) The graph shows the signal for two opposite initial polarization states, TE and TM. The two insets are near-field images of the single modes carried from the waveguide for the two polarizations. d) Similar to the (c) with the initial polarization TE and the analyzer set at 90° compared to the initial polarization. In each figure, the transmitted signal from the output waveguide is normalized to the initial signal.

liquid crystals. This is not surprising considering that the fluidic circuit was used as-engraved and no further surface treatment was performed to promote the LC planar alignment. Moreover, it is worth mentioning that the probe beam does not illuminate the entire depth of the liquid crystal stream, but only a thin layer of approximately $20\text{ }\mu\text{m}$ below the Fe:LN crystal.^[43] This means that only the LC layer in proximity to the channel cover interacts with and influences the propagation of the probe beam and even slight anisotropic anchoring of LC to the LN surfaces becomes noticeable in the measurements. The signal oscillations of Figure 2d are due to a change in the light polarization introduced by the LC molecules, as also previously reported and investigated in Refs. [23, 53]. The detailed analysis of this phenomenon is beyond the scope of this work, but a qualitative description of it can be obtained by considering the Jones matrices^[54] (as shown by Figure S1 and dedicated section in the Supporting Information). The temporal evolution of the light signal recorded by the analyzer follows a sine function, whose argument is proportional to the angle ω between the incident light polarization and the orientation of the liquid crystals director. In first approximation, ω linearly depends on the applied electric field, which in turn follows the exponential law reported in Equation (1). Consequently, the oscillation period is expected to become longer with time, as confirmed by the experimental data, whereas the intensity of subsequent maxima decrease due to the progressive alignment of the LC molecules along the vertical axis.

3.1. Photovoltaic-Driven Change of the Beam Polarization

The temporal evolutions of I_{probe} in Figure 2 resemble the behavior of the photovoltaic field described by Equation (1)–(2). Since

the dynamics of liquid crystal reorientation is much faster than the typical constant time of the photovoltaic field,^[52] it is expected that the modulation of the probe beam is governed by photovoltaic phenomena.

The time constant τ_{PV} is proportional to the intensity of the pump light, I_{pump} , and the reduction degree of the material, r . Therefore, to elucidate the role of these parameters in controlling the time scale of the LC reorientation, first we compare the performances of two optofluidic devices presenting Fe:LN covers with different reduction degrees. In Figure 3a,b the intensity of the probe beam is reported as a function of the illumination time for the two devices, characterized by a reduction degree of the cover of $r_a = 0.03$, $r_b = 0.20$, respectively. It should be noted that the intensity of the transmitted beam starts to increase at 240 s (Figure 3a) and 30 s (Figure 3b), that is, the time at which the pump beam was turned on. In both curves, the light intensity follows an exponential trend with time, as confirmed by the good agreement achieved by fitting the experimental data to the function $I_{\text{SAT}}(1 - e^{-t/\tau})$ (blue lines). As expected, the timescales of the two processes are different, and we find $\tau_a = 510 \pm 7$ s and $\tau_b = 61.0 \pm 0.7$ s for the device with the Fe:LN cover more and less reduced, respectively. The ratio between the time constants is equal to $\tau_a/\tau_b = 0.120 \pm 0.002$, which is compatible with the inverse ratio of the reduction degrees, $r_a/r_b = 0.15 \pm 0.02$, confirming the major role played by the photovoltaic effect in determining the dynamic of I_{probe} . Concerning the maximum gains in the transmitted light intensity I_{SAT} , we obtain for the two devices $I_{\text{SAT},a} = (0.38 \pm 0.05)$ and $I_{\text{SAT},b} = (0.40 \pm 0.02)$. The agreement between these values is consistent with the hypothesis of a direct relation existing between the photovoltaic effect and the LC reorientation.

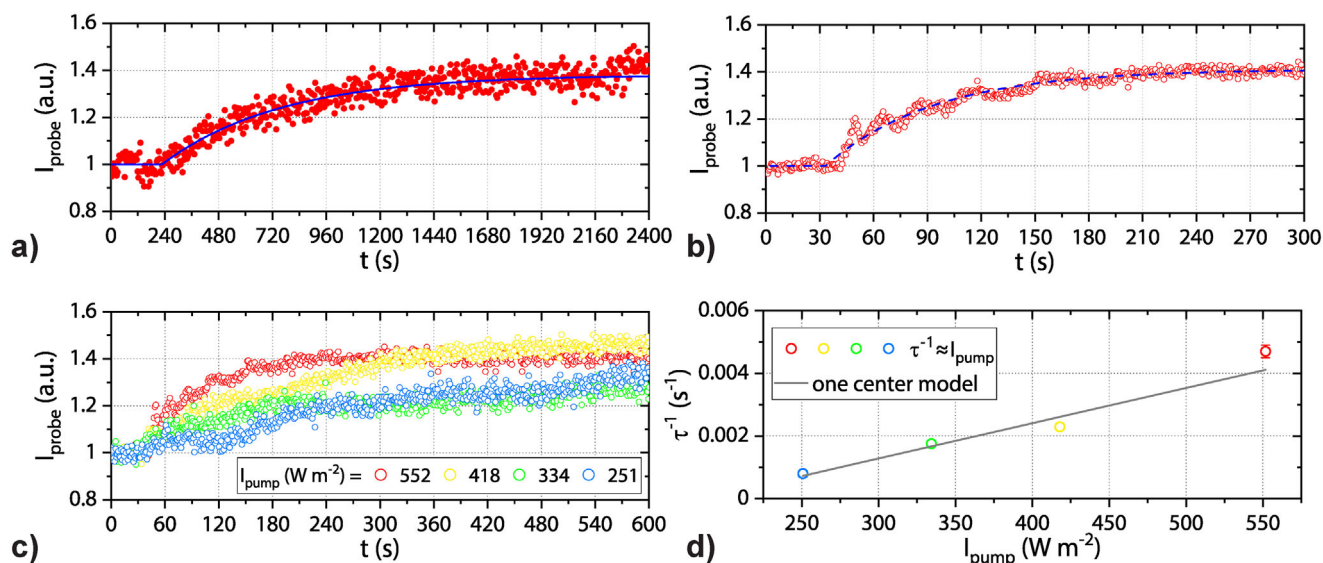


Figure 3. Optical modulation of the probe signal by tailoring of the photovoltaic electric field. Graphs (a) and (b) show similar behavior of the probe signal at different timescales for two Fe:LN covers with different reduction degrees $r_a = 0.03$ and $r_b = 0.20$ (filled and empty red dots, and solid and dashed blue lines), respectively. The blue lines are the interpolation with the exponential growth of Equation (1), where the E_{SAT} is replaced with another constant I_{SAT} . The fitted results are $\tau_a = 510 \pm 7$ s and $I_{\text{SAT},a} = 0.38 \pm 0.05$, and $\tau_b = 61.0 \pm 0.7$ s and $I_{\text{SAT},b} = 0.40 \pm 0.02$. c) The graph reports the probe signal for different pump light intensities. The time constants τ obtained from the fitting for a timescale below 60 s are plotted in graph d). The same color code is used for (c) and (d). The linear fit, the gray dashed line, shows the linear expected dependency from the one-center model, with a slope $1.2 \pm 0.2 \cdot 10^{-5} \text{ W}^{-1} \text{ m}^2 \text{ s}^{-1}$, and an intercept compatible with 0. It is worth noting that the slope agrees with the theoretical value $kr^{-1} = 3.3 \cdot 10^{-5} \text{ W}^{-1} \text{ m}^2 \text{ s}^{-1}$. In each of the experiments presented in this Figure, the initial polarization of the probe beam is TE.

In fact, the amplitude of the photovoltaic field is proportional to the concentration of the acceptor centers, which for the two devices are $C_{\text{Fe}^{3+},a} = (18.2 \pm 2.6) \cdot 10^{18} \text{ cm}^{-3}$ and $C_{\text{Fe}^{3+},b} = (15.6 \pm 0.8) \cdot 10^{18} \text{ cm}^{-3}$. Being very close to the acceptor concentrations, the final reorientation of the LC molecules is expected to be similar for the two devices: this is confirmed by the recorded values of I_{SAT} . This analysis highlights the possibility of controlling the device's efficiency and actuation speed by controlling the material composition properties, namely the r and the iron concentration, which can be easily performed with suitable thermal treatments.

From Equation (2), the second parameter useful to control the photovoltaic constant τ_{PV} , and thus the dynamics of I_{probe} , is the intensity of the pump source I_{pump} . The latter was varied using different output powers of the pump laser (i.e., 66 mW, 50 mW, 40 mW, 30 mW, respectively) and keeping unvaried the size beam. Figure 3c shows the variation in the probe signal over time for different values of I_{pump} , where it is possible to observe that the temporal growth of I_{probe} is slowed down by decreasing the pump intensity. Fitting the data to $I_{\text{SAT}}(1 - e^{-t/\tau})$, the time constants τ of each curve can be derived and the results are reported in Figure 3d as a function of the pump power (i.e., intensity). The inverse of the time constants, τ^{-1} scale linearly with I_{pump} , as expected by the photovoltaic one-center model discussed in Section 2, demonstrating the feasibility of modulating the intensity of a probe light by acting directly on the pump source. In the next sections, the pump intensity I_{pump} and the reduction degree r are fixed at the maximum values used in this work.

3.2. Control of the Light Polarization

In the previous section, we have demonstrated that the intensity of the output light I_{probe} undergoes a variation due to the rotation of the 5CB liquid crystal upon the arising of a light-induced photovoltaic electric field. The variation of I_{probe} is due to the interaction of the probe beam with the rotating liquid crystal,^[22] and depends on the initial polarization of the probe light beam. In this section, we investigate in detail how the synergy between the photovoltaic field and the LC molecules can be employed to control the polarization of a probe light beam. In particular, the measurements carried out with a 5CB liquid crystal in the main fluidic channel are compared with those obtained by using an optically inert liquid as water.

The experimental procedure can be summarized as follows. First, we determine the polarization direction of the output probe beam, ϕ_0 , in the absence of the E_{PV} , which could differ from the input beam polarization due to a non-perfectly planar alignment of the LC molecules inside the channel. To do this, we use a fixed polarization of the input beam (TE) and we rotate the analyzer, to find the maxima and minima of the transmitted light intensity. The intensity of the probe beam, I_{probe} , is measured for different angles θ between the main axis of the analyzer and the initial polarization direction of the light beam. Specifically, the analyzer was rotated from $\theta = 0$ to $\theta = 2\pi$, by steps of $\pi/8$. At each step, the signal I_{probe} is acquired for 30 s and 60 s, in the case of water or 5CB filling the fluidic channel, respectively. The experimental data obtained with the pump laser switched off are displayed in Figure 4a–c for the 5CB and the water, respectively.

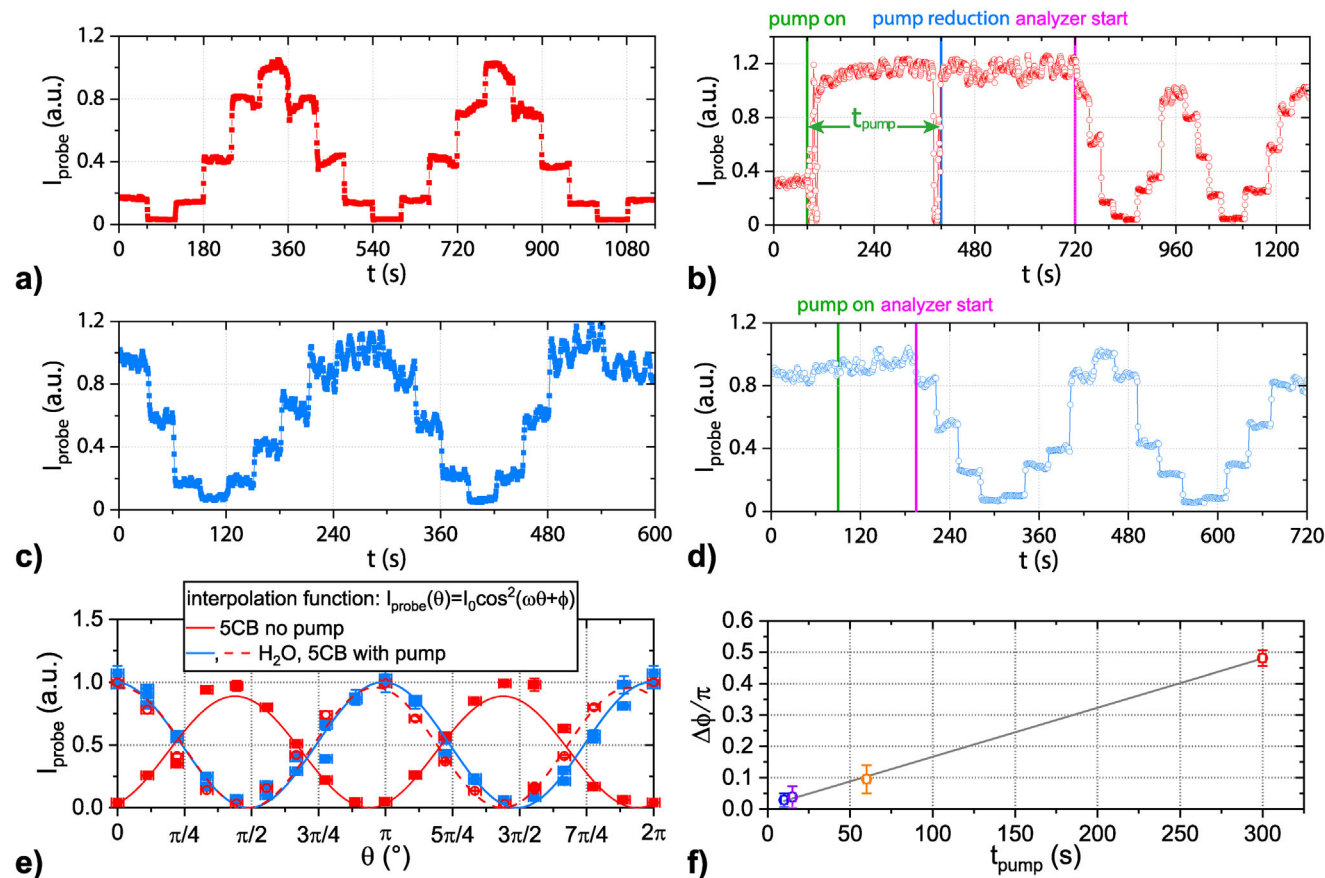


Figure 4. Polarization analysis of the probe signal. The graphs show the transmitted probe intensity during a 2π rotation of the analyzer with steps of $\pi/8$. Graphs (a) and (b) show the signal achieved when 5CB are injected in the fluidic channel, with and without the pump illumination, respectively. Graphs (c) and (d) report the equivalent graphs for the case of water filling the channel. e) The intensity of the probe signal, I_{probe} , is plotted as a function of the angle θ between the analyzer and the initial polarization direction of the beam (i.e., the angle between the polarizer and the analyzer), for water and liquid crystal (blue and red points) with and without pump (filled squares and empty circles). f) The variation shift of the probe signal, $\Delta\phi$, presents a linear behavior with the pump time t_{pump} , which is the time interval between the switching on of the pump (pump on in green) and the moment when the pump intensity is reduced (blue lines). The linear fit reveals a slope $0.0156 \pm 0.00003 \text{ s}^{-1}$ and intercept 0.01 ± 0.01 , with $R^2 > 0.996$.

In both cases, after 8 rotation steps, the initial value of I_{probe} is recovered, indicating that the probe beam polarization has not changed during the measurement. Notably, in the case of the 5CB molecules, the maximum of I_{probe} is achieved after rotating the analyzer by 5 steps, indicating that for this specific measurement, a pre-tilt of the liquid crystals of about 30° was present. Then, the pump laser is switched on for $t_{\text{pump}} = 300 \text{ s}$ to establish the light-induced electric field and during this temporal interval, the output I_{probe} is monitored with the analyzer set at $\theta = 0$. As expected, when the fluidic channel is filled with water I_{probe} does not vary in time (Figure 4d, $t = 90\div 180 \text{ s}$), whereas it follows an exponential trend in the presence of the LC molecules (Figure 4b, $t = 80\div 400 \text{ s}$), as discussed before. The illumination time was set to 300 s to ensure that the light-induced photovoltaic field had reached its saturation value. After this time, the intensity of the pump beam is not switched off but rather reduced with a filter $OD = 5$ (blue line in Figure 4b), to avoid the erasure of E_{PV} caused by the dark conductivity^[55] and maintain its value constant during the subsequent measurements with the rotating analyzer. In Figure 4b, after the reduction of the pump intensity we waited almost 300 s before starting to rotate the analyzer, to ver-

ify the stability of the signal and demonstrate the effectiveness of our approach in maintaining the E_{PV} constant. After the creation of E_{PV} and the consequent rotation of the LC molecules, the analyzer was rotated from 0 to 2π by steps of $\pi/8$, as shown in Figure 4b ($t = 720\div 1280 \text{ s}$) and Figure 4d ($t = 180\div 720 \text{ s}$) for 5CB and water in the channel, respectively. Already from a first interpretation of the graph, it appears evident that the maximum of the transmitted light signal is achieved for the analyzers set at $\theta = 0$ and $\theta = \pi$, meaning that the light after passing through the water or the reoriented LC molecules is TE polarized. This is particularly evident in Figure 4e, where the mean intensity of the probe beam is reported as a function of θ (i.e., the rotation steps of the analyzer). The data show that the arrangement of the water molecules under the presence of the photovoltaic field does not vary the polarization of the probe beam, being water an optical isotropic medium. In contrast, when the fluidic channel is filled with 5CB, a remarkable variation in the polarization axis of the probe beam is observed when the photovoltaic field reaches its saturation value. The results fit the Malus law $I(\theta) = I_0 \cdot \cos^2(\omega\theta + \phi)$, where the parameter ϕ represents the variation of the light polarization with respect to its initial

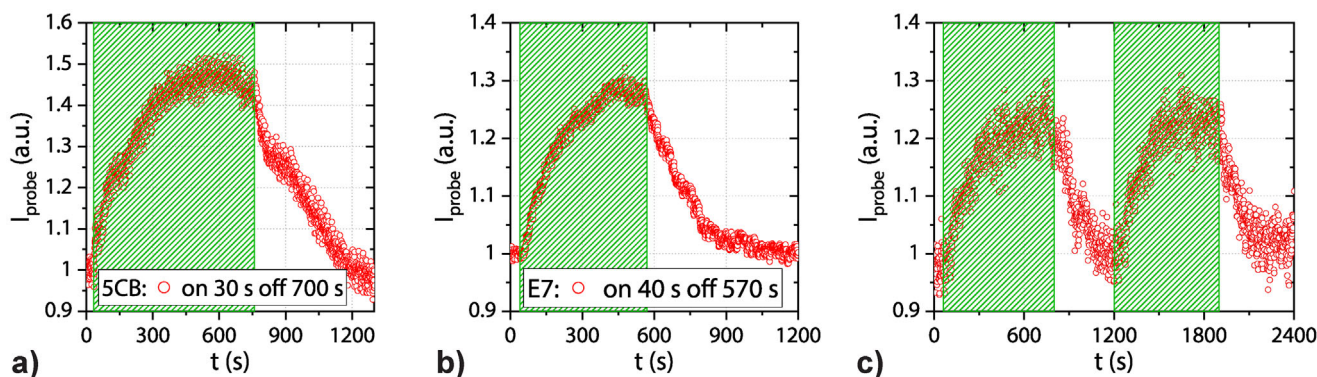


Figure 5. Experimental results for pump intervals, defined as the temporal interval during which the pump beam illuminates the Fe:LN (green region in the graphs). a) Probe signal during a pump interval between 30 s and 700 s for 5CB liquid crystal. b) Probe signal during a pump interval between 40 s and 570 s for E7 liquid crystal. c) Probe signal during two consecutive pump intervals (60–800 s) and (1200–1900 s) for 5CB liquid crystal.

state, and it is expected to be proportional to the orientation of the LC molecules inside the fluidic channel.^[22] In Figure 4e, in the case of water filling the main fluid channel, the shift results null with and without the pump illumination; on the contrary, for 5CB the shift induced by the photovoltaic effects results $\phi = (0.557 \pm 0.006) \pi$, before switching on the pump beam, and $\phi = (0.01 \pm 0.01) \pi$ after reaching the saturation value of E_{pv} . It should be emphasized that this polarization change is achieved by applying to the liquid crystal the maximum photovoltaic electric field achievable with the employed Fe:LN, that is the saturation values E_{SAT} . In this case, we also observed a remarkable increase in the output intensity of I_{probe} due to the alignment of the LC molecule along the vertical direction and the consequent refractive index matching.

Since the orientation of the LC molecules is determined by the amplitude of the applied electric field, it is reasonable to expect a different $\Delta\phi = \phi - \phi_0$ for different illumination times with the pump source. In fact, by varying t_{pump} , electric fields with different amplitudes $E_{pv}(t_{pump})$ are created on the Fe:LN cover, resulting in different alignment of the LC molecules. In Figure 4f, we report an example of the $\Delta\phi$ achieved by illuminating the Fe:LN cover of the optofluidic device for different times t_{pump} , from 10 s up to 300 s. The raw data and the analysis to obtain these values are reported in Figure S2 in the Supporting Information. The polarization shift exhibits a linear dependence on the illumination time, demonstrating the capability of the hybrid LC-LN device proposed in this work to tune the polarization of a light beam via the photovoltaic effect. Also worth noting is the fact that the process can be sped up or slowed down according to the desired applications, by varying the reduction degree of the optically active Fe:LN cover or the intensity of the pump light source. As final remark, we would like to highlight that the achieved light polarization (LC rotation) can be kept stable as long as desired by applying the same approach we used during the experiments, that is by using a very attenuated laser beam to maintain the light-induced electric field constant and impede the spontaneous reorientation of LC. To this aim, it is sufficient to use a very weak green light source, with an intensity of about 0.5 W m^{-2} , which is commercially available at low cost and with very compact size.

3.3. Multiple Actuation

A key feature of the optofluidic light modulator presented in this work is the reversibility of the photovoltaic effect driving the LC reorientation. In addition to this, the final device presents a high degree of customization, being possible to substitute the 5CB used in this work with other liquid crystals presenting different physical and chemical properties, as in the case of E7. Figure 5 shows the variation of I_{probe} with time in the presence and absence of pump illumination when the fluidic channel is filled with 5CB (Figure 5a and 5c) and E7 (Figure 5b). The green areas of the graphs highlight the time intervals when the pump beam is maintained on, whereas the white areas indicate the absence of the pump illumination. In Figure 5a, I_{probe} increases thanks to the photovoltaic-driven reorientation of LC molecules until 700 s, when the pump is turned off and I_{probe} starts to decrease, reaching its initial values after only 500 s. It is intuitive to deduce that during this relaxation interval, the liquid crystals are reorienting back to the initial state. However, this phenomenon cannot be directly attributed to the photovoltaic field, since the dark conductivity responsible for the erase of E_{pv} in the absence of light is characterized by constant times at least one order of magnitude larger than τ_{pv} .^[55] A possible mechanism underlying the spontaneous reorientation of liquid crystal molecules could be a screening effect similar to that recently reported in the literature for Fe:LN crystals.^[31,56] Indeed, once the photovoltaic current is no longer flowing in the Fe:LN crystal due to the switching off of the pump light, the liquid crystal molecules closed to the Fe:LN cover rearrange to create a dipole counteracting the evanescent component of E_{pv} , resulting in a progressive and fast decay of its amplitude inside the underlying fluidic channel. In these conditions, the liquid crystal molecules lying far away from the Fe:LN surface start to reorient, aligning to the underlying LC molecules anchored to the bottom of the channel. This could lead to a domino effect aligning the liquid crystals to the initial state layer by layer. A detailed model and explanation of this mechanism is beyond the scope of this paper, although it represents an interesting outlook for future investigations in the field of liquid crystals. The observed mechanisms emerging from the interaction of the 5CB molecules with the photovoltaic field are also confirmed by

further experiments performed using other nematic liquid crystals, such as E7, as shown in Figure 5b. In addition, these probe signal curves are reproducible over time and for multiple pump illuminations, as reported in the signal of Figure 5c. As a final remark, it is worth mentioning that the time constants observed in this work for aligning the LC could be further reduced by speeding up the creation of the light-induced electric field. This can be achieved by properly tuning some experimental parameters, such as the reduction degree of the Fe:LN crystal or the intensity of the pump light, thus achieving sub-second times for the light-induced LC rotation. On the other hand, the switch-off time is governed by the viscous and elastic properties of the used LC type,^[23] as well as the thickness of the LC layer that interacts with the electric field. Thus, the decrease in the fluidic channel depth would allow also for reducing the switch-off time down to the sub-second range.

4. Conclusion

This work demonstrates the feasibility of realizing a fully integrated optofluidic light modulator by combining liquid crystals (LCs) with lithium niobate (LN) crystals. An optical waveguide created on the surface of a LN substrate is coupled with a microfluidic channel filled with nematic LCs, whose molecules reorient under the action of an external electric field. The intensity and polarization of the beam propagating through the waveguide are controlled by the LC orientation, which in turn is optically driven by the photovoltaic effect. In fact, the electric fields induced by light at the surface of Fe-doped LN crystals are employed to control the time scale as well as the maximum rotation of the LC molecules, thus determining the polarization and the intensity of the waveguided beam. Thanks to the use of this new miniaturized optofluidic configuration, the modulation is enhanced up to 40% of the guided beam, compared to the 10% of previous configurations.^[22,23,52,57] In addition to the intensity, the system enables also tailoring the polarization shift of the guided beam, by varying the exposure time of the pump illumination used to create the surface electric field.

The absence of fixed metallic structures to control the LCs alignment increases the degree of reconfigurability of the final devices, facilitating its integration in optofluidic lab-on-a-chip devices where both the sensing of the LC alignment and/or the control of light polarization represent key features.^[58] For instance, the proposed device could be used to detect a change in LCs orientation due to the interaction with a biological target or control light-polarization triggered reactions by rotating LC at will.^[59,60] The approach herein presented is compatible with different kinds of LC, such as the widely employed 5CB and E7, as well as for the simultaneous control of multiple waveguides and more complex photonic structures. The full reversibility of the entire process was proven, enabling multiple consecutive actuation experiments. In addition, the Ti-diffused waveguides as well as the photovoltaic effect in lithium niobate have already been demonstrated to work at wavelengths beyond the visible range in the near-infrared spectrum.^[44,61] This extends the technological applications of the device, although care must be taken in the choice of the LC type and the wavelength, to limit optical absorption by the LC in the infrared, which is related to molecular vibrations. It is also worth noting that the working principle of the proposed

device also holds if the LN substrate hosting the waveguides is substituted by other materials, including Si, LN on Si, and SiO₂-based devices. In conclusion, this innovative light-driven liquid crystal modulation system based on lithium niobate represents a significant step forward in the development of the next generation of optofluidic devices. The elimination of metallic electrodes, combined with the ability to precisely control the light-induced alignment of LC molecules, offers promising new possibilities for a wide range of applications in integrated photonics, microfluidics, and sensing technologies.

5. Experimental Section

The device used in this study comprises two primary components: the lower monolithic LN platform, which hosts the optical waveguides and channels, and the Fe:LN cover. The optical setup is also divided into two parts: one for the probe and the other for the pump, as shown in Figure 6a. The setup and fabrication procedure of the device are presented in detail below. 5CB and E7 were injected into the microfluidic channel through standard inlet and disposable syringes.

Optical Setup: The probe light source consists of a He-Ne CW laser ($\lambda = 632.8$ nm and maximum power 1 mW by Coherent Inc., Santa Clara, CA, USA) which is butt-coupled to the input waveguide by an objective lens (O1, Achromat-20X/0.35 WD 0.17 by Olympus, Tokyo, Japan); similarly, an identical objective (O2) is employed to collect the light from the output waveguide after the transmission across the channel and the analyzer (A). The polarization of the light source is controlled by a half-waveplate ($\lambda/2$) and a polarizer (P). The light signal collected by the second objective is driven to a photodiode and amplified by a lock-in amplifier (SR830 by Stanford Research Systems, Sunnyvale, CA, USA), which also controls the optical chopper (C) located after the polarizer. Part of the intensity of the probe (approx. 10%) is collected by a photodiode (PD1) and normalized in the lock-in-amplifier to take into account power fluctuations of the laser source. The pump laser consists of a CW diode laser ($\lambda = 532$ nm and maximum power 66 mW). The power of the laser is controlled by a rotational attenuator (OD). The beam is expanded by a telescope and then focused onto the cover in correspondence of the fluidic channel crossing with the waveguide. It is worth noting that mirrors (M) and two telescopes (TL1, TL2) are also employed to adjust the beam waists when needed.

Fabrication of the Bottom Substrate: The optical waveguides are Ti-indiffused into 3-inch commercial x-cut LiNbO₃ wafers (LN, thickness: 1 mm, by Crystal Technology Inc., Palo Alto, CA, USA), resulting in single-mode waveguides in the visible range. The process consists of three main steps: lithography, Ti deposition and lift-off, and thermal diffusion. For the lithography, the photoresist S1805 (Dow Chemical, Midland, MI, USA) was used and the titanium was deposited via magnetron sputtering. The lift-off process left only titanium stripes on the wafer. Titanium stripes of 5.2 ± 0.3 μm width and 41 ± 5 nm thickness, as the one in Figure 6b, were diffused at 1030°C for 2 h in O₂ atmosphere inside a tubular furnace (F-VS 100-500/13 Carbolite Gero, Neuhausen, Germany). Secondary ions mass spectrometry and Rutherford backscattering spectrometry reveal that the dopant concentration follows an in-depth semi-Gaussian profile with a mean diffused depth $h_{wg} = 1.13 \pm 0.05$ μm and maximum titanium concentration at the surface of $(1.06 \pm 0.04) \cdot 10^{21}$ cm⁻³. The refractive indices variations induced by Titanium doping are $\Delta n_e = (1.12 \pm 0.03) \cdot 10^{-2}$ and $\Delta n_o = (0.66 \pm 0.03) \cdot 10^{-2}$ for extraordinary and ordinary indices, respectively. A detailed description of the lithographic process with all parameters and the titanium profile analysis are given in Ref. [62]. The final waveguides are z-propagating and single-mode with Gaussian profiles in both transverse directions with an average diameter along the y-axis of $w_0 = 3.6 \pm 0.3$ μm (see Figure 1d). The propagation loss of 4 ± 2 dB cm⁻¹ at 632.8 nm was measured by a cut-back technique. Several waveguides at a distance of 500 μm were diffused to avoid cross-talk between waveguides. A 200 μm wide channel was diced after the diffusion of the waveguides to split them into two sets of self-aligned waveguides. A Disco Dad 321 precision saw (Disco Corporation, Tokyo, Japan) was used for the mechanical micromachining

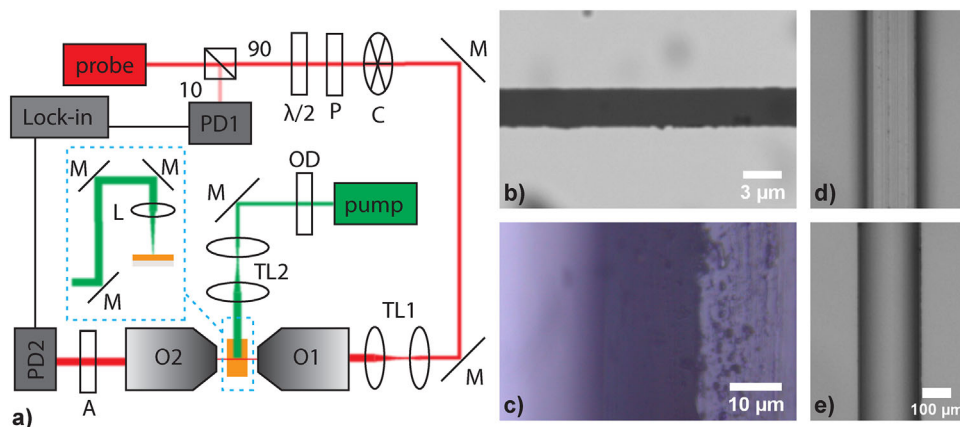


Figure 6. Experimental details of the fabrication procedure and the setup. a) Top view schematics of the optical setup described in the text. The part highlighted in light blue shows the side view of the pump beamline. b) Confocal microscope image of a titanium stripe on the LN before the diffusion. c) Reflection microscope image of the NOA68 glue used to seal the two substrates, from left to right the Fe:LN, the NOA68 in black, and the pure LN. d, e) Confocal microscope image of the microchannel's bottom surface and top edge, respectively.

equipped with a polymeric blade, coated with diamond particles (blade diameter of 56 mm and nominal thickness of 200 μm) at a rotating speed of 10000 rpm and a cutting speed of 0.2 mm s^{-1} . A complete characterization of the surface roughness with an atomic force microscope and scanning electron microscope is given in Ref. [48].

Fabrication of the Upper Substrate: The two Fe:LN samples with a total iron concentration of $C_{\text{iron}} = 18.8 \cdot 10^{18} \text{ cm}^{-3}$, 0.1 % mol and congruent composition were grown at the University of Padova by using the Czochralski technique. The growth direction was along the z-axis of the material with a pulling rate of 2 mm h^{-1} . To obtain a single domain structure, the boule was poled in an air atmosphere at 1200°C. Using X-ray diffraction, the crystalline quality of the grown samples was checked to be compatible with commercial crystals, and the orientation of the axis of the boule was determined. Consequently, the samples were diced into two samples used with the main face perpendicular to the c-axis (z-cut). The samples were polished using a Logitech PM5 lapping machine to achieve optical quality surfaces. After the growth process and without any treatment, the control of the reduction degree r was achieved by reduction thermal treatment of varying time at 500°C in a gas mixture of Ar (98%) and H_2 (2%). The value of r is measured by optical absorption with a Jasco V-670 spectrophotometer in the range 330–2000 nm.^[63]

Assembly Process: The device is finally obtained by sealing the two substrates. Before the gluing process, two holes for the inlet and outlet are drilled in the iron-doped cover. The sealing process is performed by distributing a UV curable glue (NOA68, by Norland Optical Adhesive Inc., Cranbury, NY, USA) all around the two substrates kept in tight contact. In this way, the glue flows inside the thin air gap between the two substrates due to the capillary pressure. According to Langelier et al.,^[64] the abrupt changes in hydraulic resistance at the edge of the channel confine the advancing glue front, thus avoiding the risk of flooding the microfluidic channel. Finally, the entire device is exposed to UV light for 7 min, obtaining the final device.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

light modulation, liquid crystals, lithium niobate, optofluidics, photovoltaic effect

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