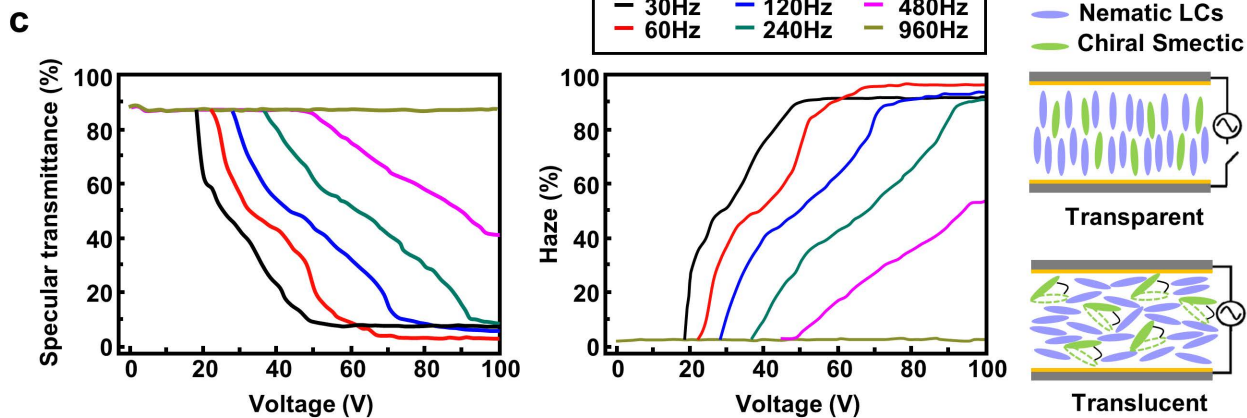
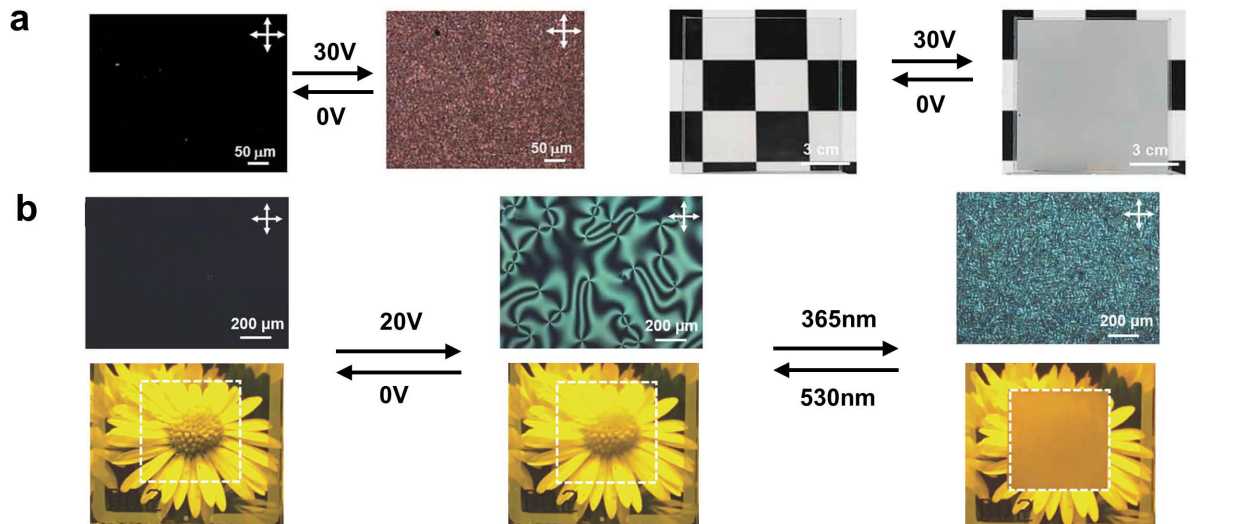




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Electrohydrodynamic instabilities for smart window applications

Yuanyuan Zhan^a, Han Lu^a, Mingliang Jin^{a,b} and Guofu Zhou^{a,b,c}

^aSCNU-TUE Joint Lab of Device Integrated Responsive Materials (DIRM), National Center for International Research on Green Optoelectronics, South China Normal University, Guangzhou, P. R. China; ^bGuangdong Provincial Key Laboratory of Optical Information Materials and Technology & Institute of Electronic Paper Displays, South China Academy of Advanced Optoelectronics, South China Normal University, Guangzhou, P. R. China; ^cShenzhen Guohua Optoelectronics Tech. Co. Ltd., Shenzhen, China

ABSTRACT

Electrohydrodynamic instability in liquid crystals is investigated for smart window applications. By comparing different species of ions doped in liquid crystals, the positive charge of the organic part of the salt is the origin of the generation of vortices and leads to the activation of electrohydrodynamic instability. By replacing conventional electrolytes with zwitterions, compared to electrolyte-doped liquid crystals, zwitterion-doped liquid crystals have a broader range of optimal frequency from 10 Hz to 5 kHz. The devices can be switched between the transparent state and the light scattering state for thousands of times without showing any fatigue. A scattering colour is obtained by incorporating a dichroic dye in zwitterion-doped liquid crystal. A patterned device is designed and shows a localised light scattering and colour effect in the presence of an electric field. This strategy will make electrohydrodynamic instability possible to enrich the applications across smart windows, projection screens and information displays.

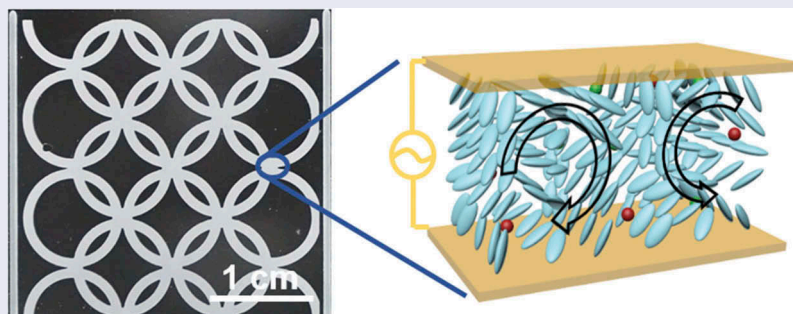
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Electrohydrodynamic instability; zwitterion; dichroic dye; smart window; information display



1. Introduction

Electrohydrodynamic instability (EHDI) was first discovered in the 1960s [1]. Since then, it has been drawing much attention from scientists for applications such as liquid crystal (LC) display and beam steering [2,3]. Under a sufficiently high electric field strength, dynamic scattering mode can be induced based on the generation of turbulence originated from ions oscillate between electrodes and leads to light scattering [4]. In the early stage, investigation on EHDI in LC was mainly focused on William Domains, generally driven by a voltage of direct current field based on the injection of ions from electrodes or impurities from the bulk LC [5–7]. With emerging of smart window technology [8–15], light scattering mode is demanding in practice. One option is to reduce electrical conductivity of LC mixture by the generating ions to the host LC and to apply an

alternating electric field with a low frequency to oscillate ions to form polydomains [16–18]. For instance, creating zwitterions by illuminating UV can trigger EHDI and therefore generate light scattering effect [19]. This strategy creating a light scattering state by using multiple stimuli is promising in saving energy for smart window applications. Hereby, we thoroughly investigate EHDI by varying species of ions and incorporating dyes in LCs.

2. Experimental section

2.1. Materials

Sorbitan monooleate (J & K Scientific), sodium dodecyl sulphate (Aladdin), decyltrimethylammonium bromide (Sigma-Aldrich), dodecyltrimethylammonium bromide (Sigma-Aldrich), hexadecyl trimethyl

ammonium bromide (Sigma-Aldrich), trimethyloctadecylammonium bromide (Sigma-Aldrich), (Lauryldimethylammonio)acetate (Sigma-Aldrich) and Reichardt's dye (Sigma-Aldrich) were used as ions to dope into a host LC. Negative LCs HNG30400-200 ($T_i = 94^\circ\text{C}$) and dichroic dyes RL013 and non-dichroic dye C1 were purchased from Jiangsu Hecheng Advanced Materials Co., Ltd. Fluorescent dye K160 was purchased from Risk Reactor.

2.2. Sample preparation

Commercial cells of 10 μm of cell gap with or without patterns (XGH3030-10, Shenzhen Guohua Optoelectronics Tech. Co., Ltd.) were used for experiments. The samples that used for all the measurements are 3 cm by 3 cm in width and length, unless otherwise specified. The cells were filled with an LC mixture by capillary suction at 110°C . The samples were cooled slowly to 75°C and held for 2 h to eliminate any thermal instability before further cooled down to room temperature. For the comparison of different species of ions in activating EHDI, each sample contains 2.7×10^{-6} mol of ions. Samples only doped with dye was prepared by incorporating 0.3 wt% of the dye in a host LC. Coloured samples that can trigger EHDI were prepared by doping 0.3 wt% of dichroic dye and 0.1 wt% of zwitterionic Reichardt's dye in a host LC.

2.3. Sample characterisation

The textures of samples were checked using polarised optical microscopy (POM) (Leica DM2700P) combined with heating and cooling system (Linkam32). Transmission spectra were recorded with a UV/VIS/NIR spectrometer (PerkinElmer 950 and R1 Series, Shanghai Ideaoptics Instrument Co., Ltd). Angle-resolved light scattering spectra were performed by angle-resolved fibre optical spectrometer (R1 series, Shanghai Ideaoptics Instrument Co., Ltd). The alternating electric field with a sine wave was provided by a function generator (AFG1022, Tektronix). The electric signal from the function generator was amplified by a high-voltage linear amplifier (A400, FLC Electronics). The output voltage was measured by an oscilloscope (TBS2000, Tektronix).

3. Results and discussions

We chose a negative LC ($\Delta\epsilon = -8.3$) with a birefringence of 0.149 (Supporting Information, Table S1 and Figure S1), in the presence of a small amount of cationic electrolyte hexadecyltrimethylammonium bromide (CTAB); initially, on polarised optical microscope, the LC is black which is corresponding to its homeotropic alignment between two electrodes perpendicular to the substrates (Figure 1(a)). By applying an alternating current (AC) field, the birefringence of LC is changed and multi-domains can be observed [19]. This is due to that the long axis of the LC molecule can be reoriented perpendicular to the electric

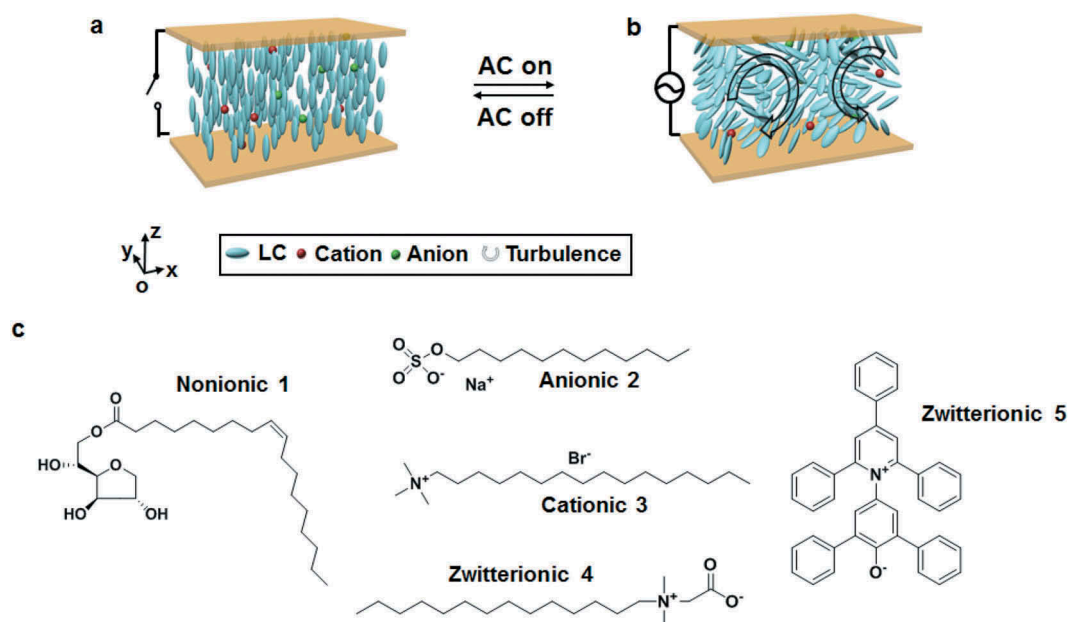


Figure 1. (Colour online) Schematic representation of electrohydrodynamic instability (EHDI) (a) at 0 V and (b) under the application of an alternating electric field. (c) Chemical structures of four different species of ions: the non-ionic 1, the anionic 2, the cationic 3 and the zwitterionic 4, 5.

field line, and at the same time, ions start oscillating between electrodes (Figure 1(b)). The electrical force exerted on the ions can be transferred to the LC by the exchange of momentum. Analogous to thermal-induced Rayleigh–Bernard instability, the LC molecules are set in motion and EHDI is triggered.

We first investigate the influence of species of electrolyte on the activation of EHDI. By comparing three different species of commercial electrolytes, non-ionic molecule **1**, anionic molecule **2** and cationic molecule **3** (Figure 1(c)), only the sample containing cationic electrolyte has a dramatic transmittance decrease during applying the field (Figure 3(a), Supporting Information, Figure S2). This result can be explained by that the positive charge is the main source of movable charge carrier (Supporting Information, Figure S3). The argument is that negative Br^- ions in molecule **3** are barely soluble in the bulk LC mixture, and they also tend to be strongly attracted to opposite charges in the conductive indium tin oxide layers. The positive CTA^+ ions are relatively more soluble in the LC mixture because the N^+ charge is separated by those three methyl groups from the opposite charges and is less attracted. In addition, the aliphatic tails can help the positive CTA^+ ions dissolve more easily in the host LC. On the contrary, anionic molecule **2** can poorly dissolve in the bulk nematic LC though the tiny amount is doped and therefore cannot initiate EHDI, while as expected, the non-ionic molecule **1** cannot activate EHDI due to the lack of charges in the molecule.

The modest solubility of electrolyte in host LCs and accumulation of ions onto electrodes tend to reduce the lifetime of devices. In order to solve those problems, hereby, we propose to replace electrolyte with zwitterion. A zwitterion is a molecule with multi-functional groups of which one possesses a positive and the other possesses a negative electrical charge, and the entire molecule appears electrically neutral [20]. In our previous research, we created zwitterions in the LC upon exposure to UV light, and the effect of EHDI in the presence of zwitterions is comparable to that of LC doped with conventional electrolyte. The principle behind is that zwitterions can oscillate between electrodes following the polarity change of the applied AC electric field and generate chaotic vortices, resulting in macroscopic light scattering. Hereby, we replace the electrolyte with zwitterions (molecules **4**, **5**). Transmittance measurement on UV-Vis spectra at 550 nm shows that zwitterion **5** is more efficient in triggering EHDI, and we ascribe this to the larger molecular size of zwitterion **5** which can easily create vortices when oscillating between electrodes (Figure 2(b), Supporting Information, Figure S4). Therefore, we mainly utilise zwitterion **5** to study EHDI in this work.

The zwitterion **5** shows a maximum absorption around 726 nm in LCs (Supporting Information, Figure S5). To reduce the colour effect originated from the absorption of the zwitterionic dye, we use LC cells with a cell gap of 10 μm . EHDI is characterised on POM and demonstrated macroscopically. When blending a certain amount of the zwitterion with LC, in the absence of electric field, LC shows a homeotropic alignment between electrodes and the sample appears transparent suggesting the zwitterions mixing with LC uniformly (Figure 2(a,c,e)). Transmission spectra show no absorption of the zwitterion dye at field-off state. When the sample is driven by the AC field with stepwise changing the voltage at a frequency of 1 kHz, light scattering occurs. POM image shows that when the voltage birefringence of LC is changed (Figure 2(b)), it leads to a difference of refractive index in each vortex. Those vortices generate light scattering (Figure 2(d)). Based on ASTM D1003-00, Standard Test Method for Haze and Luminous Transmittance of Transparent Plastics, the haze measured at 30 V_{rms} is 93.68%, indicating that most of the light is scattered at an angle greater than 2.5° from the normal.

When switching on the AC field, the EHDI is developed within 100 ms and followed by the second stage approximately 1 s where the LC molecules speed up to maximum velocity. This result is consistent with the observation that the output current is gradually increasing after switching on the field. Therefore, the rise time of turbulence generation is related to the amount of mobile ions and their velocity. The disappearance of EHDI takes 200 ms (Figure 2(g)). The relaxation of the light scattering is related to reorientation of LC molecules under the influence of the surface anchoring force exerted by the homeotropic alignment layers.

We investigated angle-resolved light scattering by varying an angle of incident light from the normal by measuring the transmitted light intensity as a function of the angle of incidence (Supporting Information, Figure S6). Figure 2(h) shows that at the threshold voltage of 10 V_{rms} activating EHDI, the transmitted intensity decreases with increasing incident angle, which is because the effective path length of the light within the sample increases and therefore more of the light is expected to be scattered with increasing angle. Below the threshold, only negative LC is reoriented by the field and the sample is nearly 100% transparent. However, with further increase in voltage up to 20 V_{rms} , the transmitted intensity shows angular independence, indicating that increase of voltage gives rise to smaller domains with LC molecules dynamically oriented along all directions and a higher velocity of the turbulence.

Next, we compare EHDI in the cationic electrolyte-doped and zwitterion-doped LCs. Transmittance

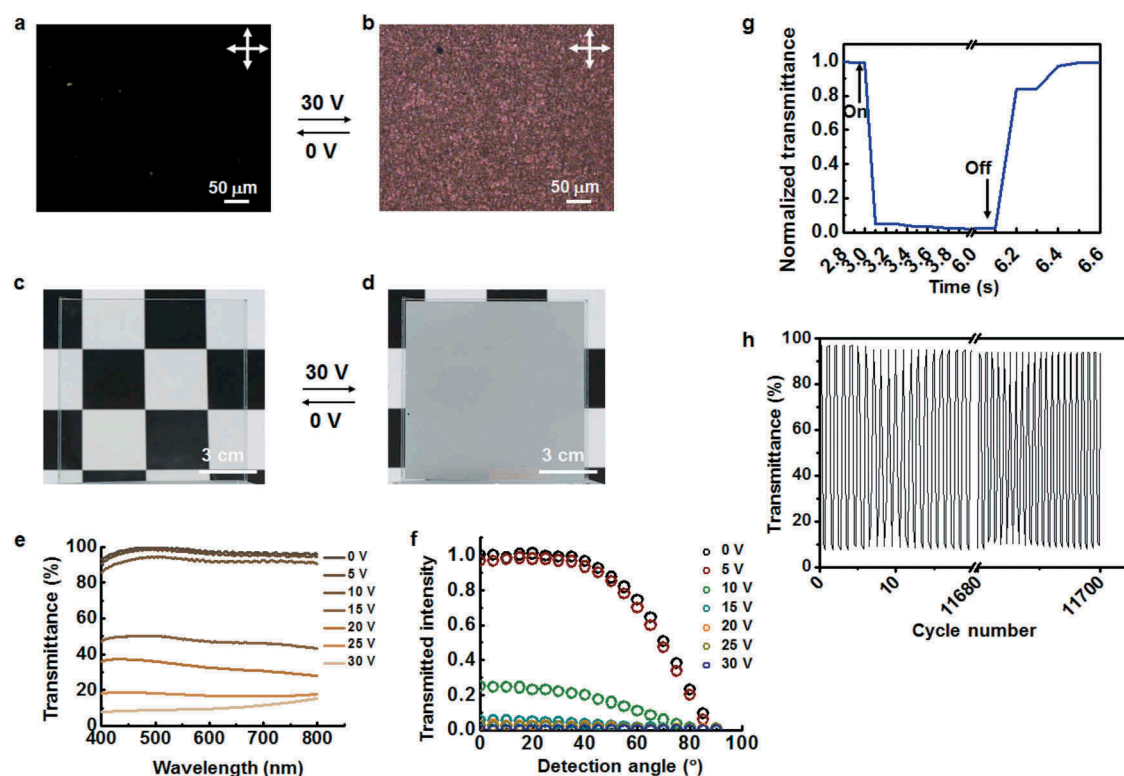


Figure 2. (Colour online) Polarised optical microscope image showing the cell at (a) field-off state and (b) field-on state. Macroscopic images showing the cell switched from (c) the initial transparent state in the absence of the field to (d) the field-on light scattering state. The size of the sample is 9 cm $\times$ 9 cm in width and length. The checkboard background is behind the sample with a distance of 10 cm. (e) Transmittance spectra of LC mixture doped with zwitterion at different voltage at 550 nm. (f) Angle-resolved transmitted light intensity measurement. (g) Response time of the window between switch-on and switch-off. The frequency applied is 1 kHz. (h) Thousands of switches between the transparent state and the light scattering state measured on UV/Vis spectroscopy.

measurement indicates that cation-LC and zwitterion-LC show a comparable light scattering effect (Figure 3(a)). The threshold voltage for triggering EHDI in zwitterion-LC is slightly higher, which might be due to the large molecular size of the zwitterion. The current generated from the movement of charge carriers in zwitterion-LC is approximately three times as much as that in cation-LC at a voltage of 30 V_{rms} (Figure 3(b)). This result suggests that less accumulation of the zwitterions on the electrodes takes place. With the measured current, we are able to estimate the power consumption of the window. Following the equation of power $P = V \cdot I$, where V is the rms voltage and I is the rms current passing through the window, the window of 9 cm by 9 cm demonstrated in Figure 2(d) consumes 0.649 W of power, and for 24 h, the energy consumed is 0.016 kWh (Supporting Information S7).

The optimal frequency is also investigated by giving the samples a frequency sweep under a sine wave signal generated from function generator. Figure 3(c) shows that the optimal frequency in zwitterion-LC appears to be ranging from 100 Hz to 5 kHz, whereas cationLC has

an optimal frequency ranging from 10 to 200 Hz. This difference is originated from the conductivity of the LC mixture which can be regulated by the amount of ions. We proved that by reducing the amount of zwitterions approximately 16 times, the cut-off frequency for EHDI can be lowered and the optimal range can be narrowed (Supporting Information, Figure S8).

Notably, EHDI is dependent on temperature [21]. We investigated the thermal behaviour of EHDI by giving samples heating and cooling treatment (Figure 3(d)). Upon heating up to 85°C where the LC mixture is still nematic, the transmittance increases are shown on both samples. This can be attributed to the decrease in birefringence of LCs and the increase of the conductivity of LC mixture when the temperature increases. Upon cooling, zwitterion-LC shows a reversible transmittance change and can go back to the initial value. Cation-LC shows a transmittance increase during the cooling process. We ascribe this to the consecutive adsorption of ions to the electrodes started from the heating process, and therefore, the amount of movable cations decreases during cooling. At room temperature, zwitterion-LC can be switched more than 11,700 times consecutively since

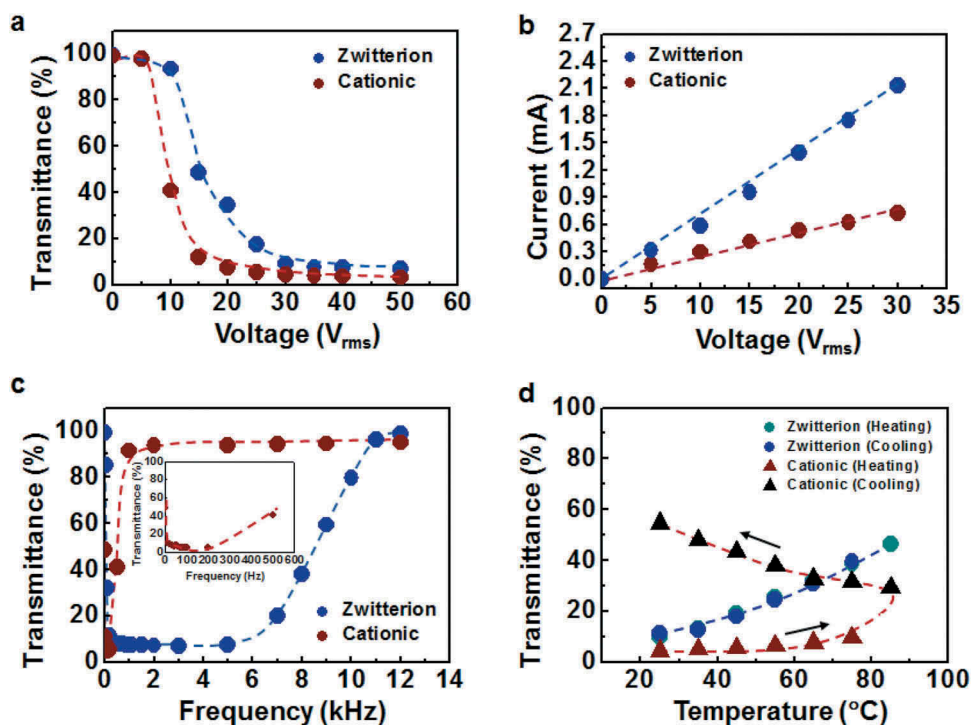


Figure 3. (Colour online) Comparison between cationic electrolyte-doped and zwitterion **5**-doped LCs in EHD. (a) Light scattering upon the application of voltage at a frequency of 1 kHz. Transmittance was measured at 550 nm. (b) Current passing through the samples. (c) Frequency-dependent transmittance at 30 V_{rms} . (d) Temperature-dependent light scattering upon heating and cooling process, respectively. The sample was applied with a continuous field is 30 V_{rms} at 1 kHz.

during applying the field no heat is generated from the motion of charges. At elevated temperature, the sample can also be switched reversibly without showing any fatigue compared to cation-LC. During the UV-Vis measurement with an applied voltage of 30 V_{rms} for consecutive 25 h, the sample appears to be stable (Supporting Information, Figure S9).

In our previous zwitterionic light-responsive EHD, the colour effect can only be created when photochromic zwitterionic merocyanine was created under the illumination of UV light. And localised scattering was obtained by placing a mask between the UV source and the sample, which is not convenient for smart window applications [19]. Therefore, we propose to develop a coloured device without extra external stimulus by incorporating dichroic dye into zwitterion-LC. Initially, the dichroic dye can be well aligned with host LC in a homeotropic fashion [22], and therefore, less colour can be exhibited. When addressing the electric field, colour appears based on the molecular orientation of the dichroic dyes. The scattering colour is characterised on UV/Vis spectroscopy. A dye RL013 with a high dichroic ratio (DR) of approximately 12.1 is mixed with LCs containing zwitterion, showing a maximum absorption at 560 nm (Supporting Information, Figure S10). To avoid the absorption band

of the dye, light scattering was measured at a wavelength of 700 nm. At the field-off state, the dichroic dye dissolved in LC mixture aligns parallel to the director of LC molecules and shows high transmittance at 560 and 700 nm (Figure 4(a,e)). When applying the AC field, light scattering is induced, and in the meantime, the colour effect is exhibited due to the motion of the dye involved in the vortex (Figure 4(b,e)). Above the optimal frequency, for example, 5 kHz, EHD steps into dielectric regime and the ions stop oscillating while the dichroic dye within LC can still be reoriented parallel to the electrodes. Therefore, the scattering can be erased while the sample appears purple (Figure 4(c,f)). Transmittance at 560 nm decreases because of the dichroic absorption of the dye, while the transmittance at 700 nm keeps the same due to the inactivation of EHD (Figure 4(f)). A designed device with patterns is shown in Figure 4(d) (Supporting Information, Figure S11). By patterning conductive indium tin oxide electrodes, scattered colour state with specific patterns can be obtained. Taking a closer look, the scattering can be sharply observed, indicating a high resolution of the device. Another example with a low DR of approximately 3.1 is investigated and demonstrated (Figure 4(h-i)). With decreasing the DR at field-off state, the sample shows a lower transmittance at the

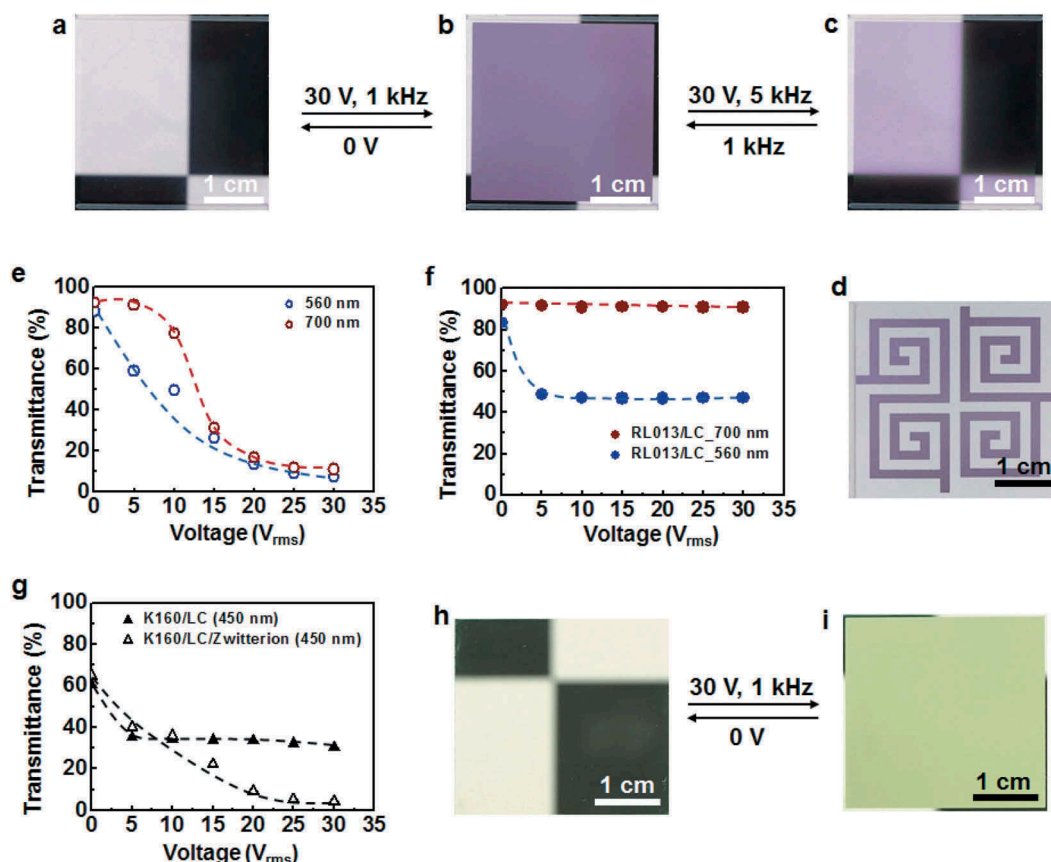


Figure 4. (Colour online) Colour window based on EHDI. Demonstration of the coloured window (a) at off-state, (b) switched to a coloured scattering state in the presence of the AC field of 30 V_{rms} at 1 kHz and (c) adjusted to a coloured state at 5 kHz. (d) Designed device with patterned ITO electrodes switched on to the coloured scattering state by the electricity. (e) Transmittance of the sample containing LC doped with RL013 and zwitterion at a wavelength of 560 and 700 nm, respectively. (f) Transmittance of LCs doped with RL013. (g) Comparison of transmittance change in K160 doped systems. (h, i) Light scattering switched between the transparent state and the scattered colour state.

wavelength of maximum absorption. For instance, K160 shows approximately 25% less absorption than RL013 (Figure 4(g)). In comparison, in dielectric regime, the non-dichroic dye-doped LC shows a small decrease of absorption when increasing the voltage (Supporting Information, Figure S12).

4. Conclusions

In conclusion, by comparing different species of the electrolyte, we have demonstrated that positive charge of the organic part of the salt plays a role in generating turbulence and activating EHDI. By replacing electrolyte with charge neutral zwitterion, the optimal frequency of EHDI can be adjusted to the range of 10 Hz to 5 kHz, and the scattering can be reversibly switched by temperature in the presence of the AC field. This result suggests less accumulation of zwitterions on the electrodes, especially at elevated temperatures. We also demonstrated a coloured device by incorporating

dichroic dye in zwitterion-LC. The device showed a distinguished contrast of colour between field-off and field-on state. The colour effect in the dielectric regime is dependent on the DR of the dye. By patterning the conductive electrodes, a localised scattering colour has been demonstrated. Based on this thorough investigation on EHDI in ion-doped LC system, we are able to develop devices for smart window applications.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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Light-Driven Electrohydrodynamic Instabilities in Liquid Crystals

Yuanyuan Zhan, Albertus P. H. J. Schenning, Dirk J. Broer, Guofu Zhou,* and Danqing Liu*

The induction of electrohydrodynamic instabilities in nematic liquid crystals through light illumination are reported. For this purpose, a photochromic spiropyran is added to the liquid crystal mixture. When an electrical field is applied in the absence of UV light, the homeotropic liquid crystal reorients perpendicular to the electrical field driven by its negative dielectric anisotropy. Upon exposure to UV light, the nonionic spiropyran isomerizes to the zwitterionic merocyanine form inducing electrohydrodynamic instabilities which turns the cell from transparent into highly scattering. The reverse isomerization to closed-ring spiropyran form occurs thermally or under visible light, which stops the electrohydrodynamic instabilities and the cell becomes transparent again. It is demonstrated that the photoionic electrohydrodynamic instabilities can be used for light regulation. Local exposure, either to drive the electrohydrodynamics or to remove them enables the formation of colored images.

1. Introduction

Liquid crystals (LCs) are an interesting class of materials, especially for the large optical and dielectric anisotropy which forms the basis for a number of electro-optical effects.^[1] Electrically tunable birefringence is used for most liquid crystal displays.^[2] For example, patterned LC alignment results in switchable gratings and optical lenses.^[3] Light scattering effects, in which transparent and scattering states are modulated through an electric field, are presently the basis for the development of a variety of applications ranging from reflective displays to smart

windows.^[4] Such devices are most often based on mixtures of polymer and liquid crystals.^[5] For example, polymer dispersed liquid crystals (PDLCs) in which the scattering is based on the refractive index mismatch between a phase separated LC constituent and a polymer binder in the field-off state,^[6] while in polymer stabilized liquid crystals (PSLCs), the scattering appears in the field-on state and originates from a combination of polydomain formation and a refractive index mismatch between the LC and a liquid crystal polymer additive.^[7] The electro-optical performances in both PDLCs and PSLCs strongly rely on the delicate control over the polymerization process and the choice of polymer and liquid crystals is critical.^[8,9]

Polymer-free scattering devices are therefore appealing because of their simplicity of fabrication.^[10]

In this work, we reevaluate an old principle based on electrohydrodynamic instabilities (EHDI) when an LC is subjected to an electric field. EHDI in LCs was discovered and reported already in the 1960s and was initially used to make scattering-based displays.^[11,12] EHDI requires the coexistence of two events, the reorientation of aligned LC and the motion of charge carriers under an electric field. Generally, homeotropically aligned LCs with a negative dielectric anisotropy are used that realign to a planar orientation.^[13] Charge carriers are often added to increase the conductivity and thereby reduce the switching voltage of the EHDI effect.^[14] In the current work, we propose to generate the ionic species by light. Thereto, we use a photochromic dye to create charge carriers by light. Using light as a contactless and remote stimulus would enable us to fabricate dual responsive and patterned addressable colored scattering devices by local exposure.^[15,16]

For our experiments, an LC mixture with a negative dielectric anisotropy ($\Delta\epsilon = -8.3$) was selected. In a first reference experiment, hexadecyltrimethylammonium bromide (CTAB) was added as electrolyte to study EHDI of the LC mixture. The CTAB-doped LC mixture is placed in a cell constructed with two glass plates provided with transparent electrodes and homeotropic alignment layers. The LC is initially homeotropically aligned (Figure 1a,c) and shows a high light transmittance (Figure 1e). When applying an external alternating electric (AC) field of 20 V_{rms} across the cell, the initially homeotropically oriented liquid crystals tilt 90° toward a polydomain planar orientation. Meanwhile, the mobile charge carriers start oscillating under the influence of the AC field generating chaotic turbulence (Figure 1b,d.). Consequently, the LC strongly scatters light and completely shields the

Y. Zhan, Prof. A. P. H. J. Schenning, Prof. D. J. Broer, Prof. G. Zhou, Dr. D. Liu
SCNU-TUE Joint Lab of Device Integrated Responsive Materials (DIRM)
National Center for International Research on Green Optoelectronics
South China Normal University
Guangzhou 510006, P. R. China
E-mail: guofu.zhou@m.scnu.edu.cn; d.liu1@tue.nl

Y. Zhan, Prof. A. P. H. J. Schenning, Prof. D. J. Broer, Prof. G. Zhou, Dr. D. Liu
Laboratory of Functional Organic Materials and Devices (SFD)
Department of Chemical Engineering and Chemistry
Eindhoven University of Technology
Groene Loper 5, 5612 AE Eindhoven, The Netherlands
Prof. A. P. H. J. Schenning, Prof. D. J. Broer, Dr. D. Liu
Institute for Complex Molecular Systems (ICMS)
Eindhoven University of Technology
Groene Loper 5, 5612 AE Eindhoven, The Netherlands
Prof. G. Zhou
Shenzhen Guohua Optoelectronics Tech. Co. Ltd.
Shenzhen 518110, China

 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adfm.201707436>.

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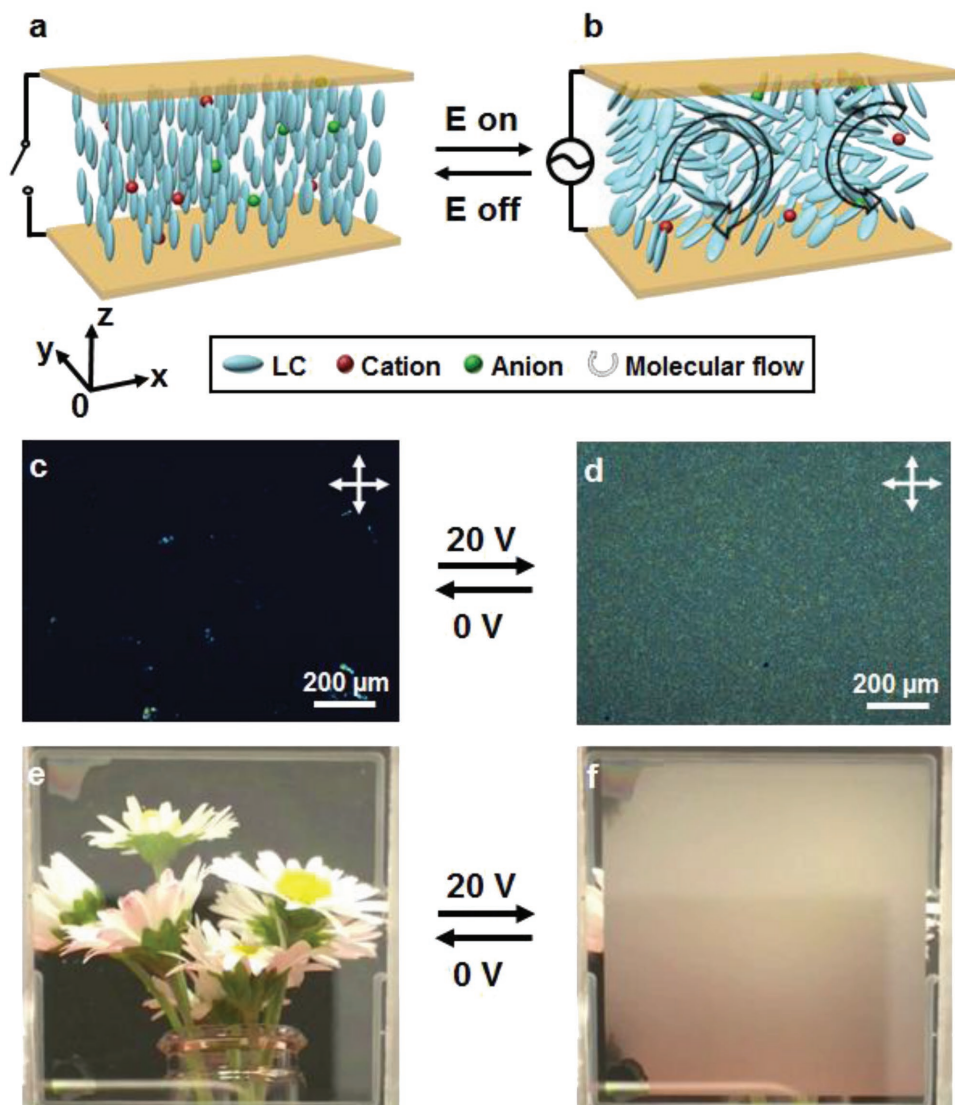


Figure 1. Electrohydrodynamic instability in a CTAB-doped LC mixture. Schematic representation of a) homeotropically aligned LC mixture in a cell at 0 V_{rms} and b) under an alternating electric field of 20 V_{rms} at 50 Hz showing EHD. Cross-polarized optical microscopy image showing c) initial homeotropic orientated LC at 0 V_{rms} and d) turbulence patterns formed in the LC at 20 V_{rms} and 50 Hz. e) The transparent state at 0 V_{rms} showing the flowers placed behind the cell, and f) the light scattering state under 20 V_{rms} at 50 Hz which hides the flowers. The voltage described in the pictures is the root-mean-square voltage. The cell gap is 10 μm .

background (Figure 1f). The EHD effect stops when the electric field is switched off and the initial homeotropic LC orientation and highly transparent state reverts immediately within 10 s.

In order to fabricate dual responsive colored scattering devices, we added the photochromic 1',3'-dihydro-1',3',3'-trimethyl-6-nitrospiro[2H-1-benzopyran-2,2'-(2H)-indole], further denoted as the spiropyran, instead of CTAB to the LC mixture. The spiropyran aligns with the LC host (Figure 2a,d; Table S1 and Figure S2, Supporting Information).^[17] Moreover, it can be switched reversibly by UV light from the nonionogenic ring-closed spiropyran form (SP) to the blue zwitterionic merocyanine open form (MC) (Figure S1, Supporting Information). Based on this, we can reversibly modulate the scattering and color of the LC device by light and electricity. As with the SP-doped sample,

when applying an external AC field, SP-doped LC is reoriented perpendicularly to the electric field line resulting in a planar alignment (Figure 2b). Since the alignment layer does not give a preferred reorientation direction, a random polydomain texture is formed which is characterized by point disclinations connected by line disclinations. The director describes a complex rotation on its transition from one domain into the other which becomes visible when observed between cross polarizers in optical microscopy. Since the LC is not homogeneously aligned in the mono-domain fashion, minor light scattering appears yet the sample still exhibits highly optical transparency (Figure 2h). When addressed with UV light ($\lambda = 365$ nm) at low intensity (18 $mW\ cm^{-2}$) for 5 s the nonionogenic closed-ring SP form converts to the ionic open-ring MC form (the MC conversion is

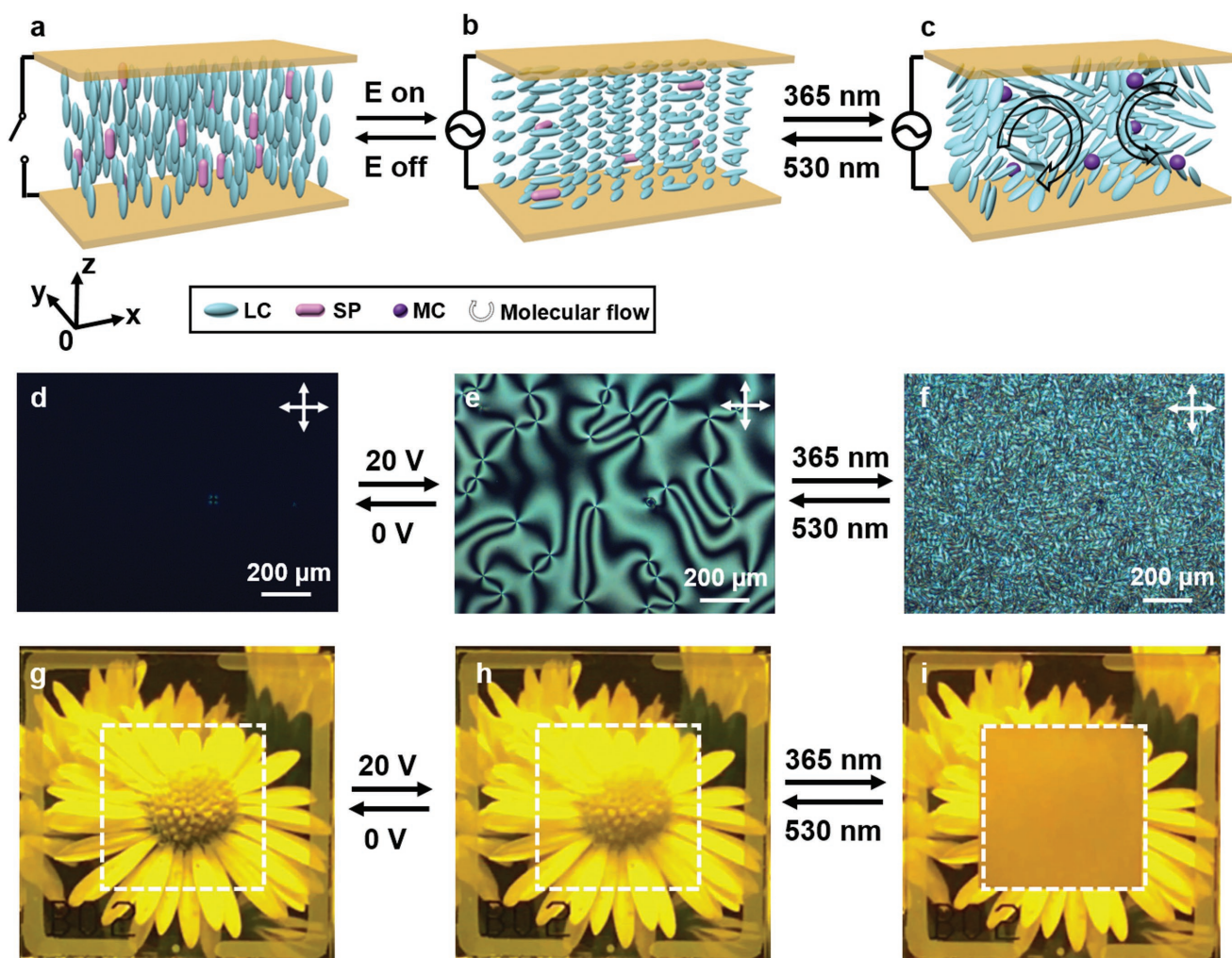


Figure 2. Light-induced EHDI. Schematic illustration of a) initial homeotropically aligned SP-doped LC at 0 V_{rms} , b) LC realigns to random planar polydomain under the AC field of 20 V_{rms} at 50 Hz, c) UV exposure converts SP to MC and initiates EHDI. d–f) The corresponding cross-polarized optical microscopic images of (a)–(c), respectively. Photos show that the sample is g) transparent without electric field, h) slightly scattering under the electric field, and i) highly opaque when illuminated with UV light. The samples are kept in the laboratory with yellow light to avoid MC to SP back reaction. The dotted square highlights the active area of the sample with the dimension of 1 cm $\times$ 1 cm. The voltage described in the pictures is root-mean-square voltage. The cell gap is 9 μ m.

given in Figure S3, Supporting Information). Consequently, the formed MC oscillates with the electric field which in combination with the realigned LCs triggers the EHDI (Figure 2c,f). Hence, strong light scattering is observed and the scattering cell shields the background (Figure 2i). The light scattered state is also colored correlating to the absorption spectrum of the MC isomer (Figure S1, Supporting Information). The EHDI and the corresponding light scattering stay for 10 min after switching off the UV irradiation (Figure S4, Supporting Information). The transparent state can be formed again by exposing the sample to green light at $\lambda = 530$ nm which stimulates the back isomerization to the SP form. Further switching off the external electric field, the LCs matrix recovers to the original homeotropic alignment.

To analyze the light-induced EHDI principle in more detail, we investigate first the input voltage and the corresponding output current (Figure 3a, Figure S5, Supporting Information). Before switching on the EHDI by UV light, a low current

of 2 μ A is measured at 5 V_{rms} which is comparable with LC mixture without SP.^[18] Upon UV exposure, the current increases significantly by a factor of 4 as the evidence of the formation of charge carriers upon the conversion of SP to the MC isomer. Next, we estimate the influence of electric field frequency and strength during UV light exposure.^[19] Figure 3b shows the optimal frequency to trigger the EHDI is 50 Hz. Below this value, the MC oscillation increases with frequency and a larger turbulence is generated and retained. Above the optimal frequency, the MC oscillation begins to lag behind the oscillation field polarity. Consequently, EHDI effect decreases and eventually disappears. Next, the scattering is characterized by stepwise changing the AC voltage from 0 to 30 V_{rms} at 50 Hz (Figure 3c). The scattering was measured at a wavelength of 700 nm to avoid the absorption band of MC. Under ambient conditions without UV illumination, the sample exhibits a constant high optical transmission with increasing voltage. Only when voltage

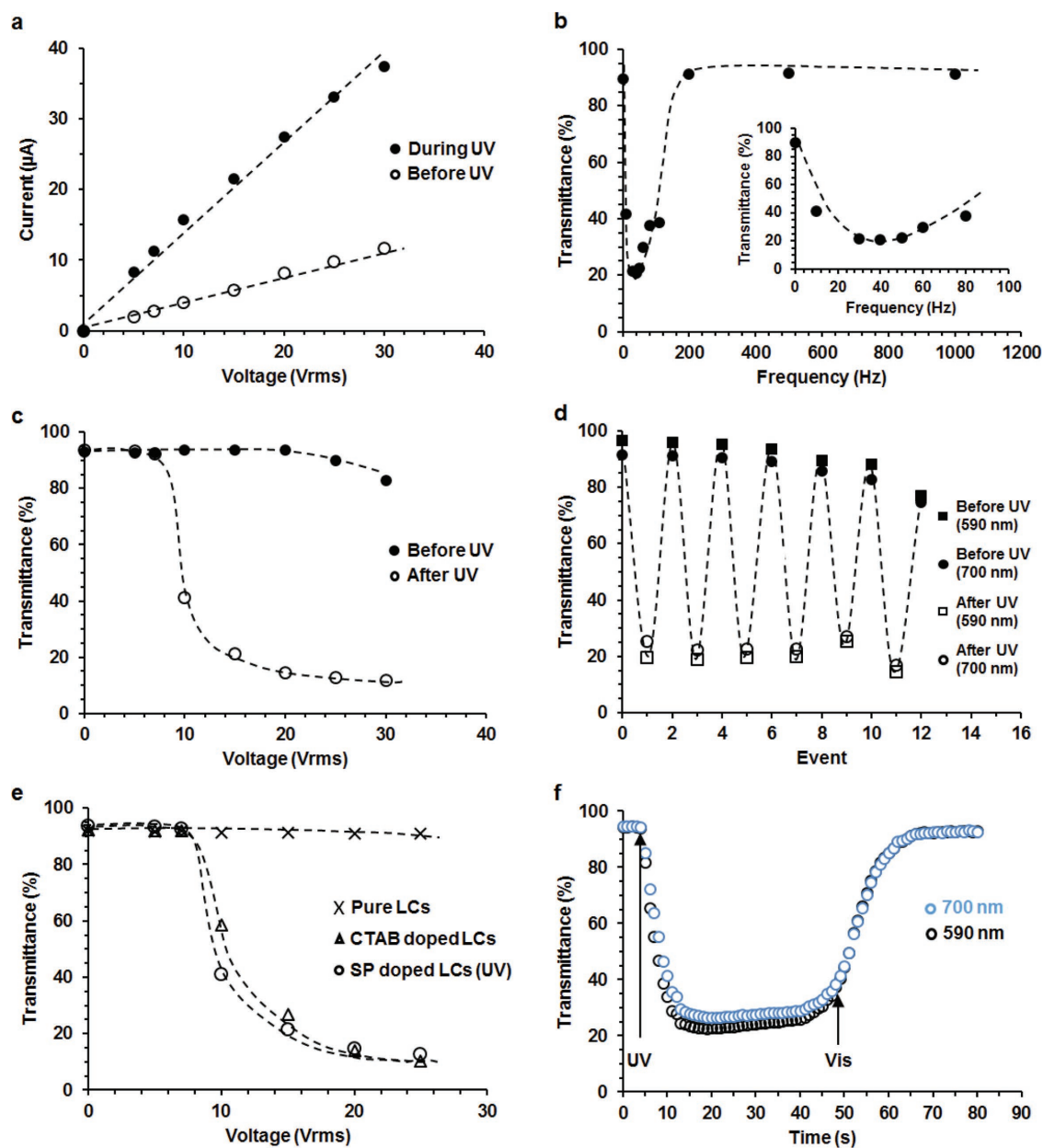


Figure 3. Characterization of light-induced EHD. a) Current generated in the sample before and after EHD is initiated. b) Influence of frequency on the light scattering. c) Voltage-dependent transmittance measured before and after UV light irradiation. The applied AC voltage is $20 V_{rms}$ at 50 Hz. The time interval between every two cycles is 2 min. d) Six switching cycles under the alternating UV and green light irradiation. The applied AC voltage is $20 V_{rms}$ at 50 Hz. The time interval between every two cycles is 2 min. e) Comparison of the transmittance at 700 nm between samples with pure LC, electrolyte CTAB and SP-doped LC. f) Response kinetics of spiropyran conversion and corresponding scattering under UV and green light exposure. The exposure conditions for all experiments are 18 mW cm^{-2} at 365 nm and 7 mW cm^{-2} at 530 nm. The cell gap is $9 \mu\text{m}$.

exceeds $30 V_{rms}$, the transmittance of the sample decreases slightly. Upon UV illumination, the transmittance drops dramatically to 16% at $20 V_{rms}$ as the result of the EHD effect. Note that in the experiments, the ultraviolet–visible spectra are taken directly after UV illumination.

By alternating the UV and green light exposure, the cell can be switched between the scattering and transparent state (Figure 3d). When measuring the transmittance at 590 nm which is the absorption maximum of MC it can be seen that the transmittance in the scattering state is low while it is high in

the transparent state (Figure S6, Supporting Information). This clearly shows that the scattering is caused by the formation of MC while in the transparent state the SP form is present. We further notice that the sample does not completely recover to its initial transmittance after several switching cycles which might be ascribed to a small fraction of MC still present after green light exposure.^[20] It should be noted that the sample can fully relax back to the initial state at an elevated temperature. When comparing the EHD effect between samples with pure LC, electrolyte CTAB-doped LC and SP-containing LC upon

applying an AC field, the pure LC sample as expected does not exhibit EHDI effect (Figure 3e). The light exposed SP–LC and CTAB–LC devices show comparable transmittance change suggesting similar EHDI behavior. The transmittance can also be adjusted by the AC field strength, as seen in Figure 3c,e. Above the threshold voltage, the LC breaks into small domains and moderate light scattering is induced. Further increasing the voltage generates the chaotic turbulence in LCs and results in the maximum light scattering.

In order to quantify the response kinetics, we measured the time-resolved transmittance of SP in the LC and the light-induced scattering by taking ultraviolet–visible spectra at 590 and 700 nm, respectively. Results are given in Figure 3f. It is obvious that the LC light scattering occurs simultaneously with the SP to MC conversion without any phase lag. The back relaxation from MC to SP and the ceasing of light scattering are also synchronized and take 10 min in the dark (Figure S4, Supporting Information). The back reaction can be accelerated to 20 s by exposing the sample to green light. For reference,

we performed an identical experiment with SP dissolved in tetrahydrofuran as solvent (Figure S7, Supporting Information). Results indicate that LCs matrix does not influence the isomerization of the photochemical reaction of spiropyran.

In order to demonstrate a light-rewritable patterned scattering device, we performed a mask-wise UV exposure as illustrated in Figure 4a–d (Movies S1–S3, Supporting Information). The projected flower pattern gives a conversion from transparent to blue and opaque, which is caused by the absorption of MC isomer and the EHDI-induced light scattering, respectively. The scattering pattern can be erased when addressed with a high frequency voltage, for example, 1 kHz (Figure 3b) or by switching off the electric field. Both methods remove the EHDI-induced light scattering while keeping the light-induced MC pattern (Figure 4e). The entire pattern can be erased by green light converting the colored MC isomer to the SP colorless form. The printed pattern can also be erased locally through focused green light (Figure 4f). Currently, most e-writer tablets provide a global erasing possibility. Based on our technology, we

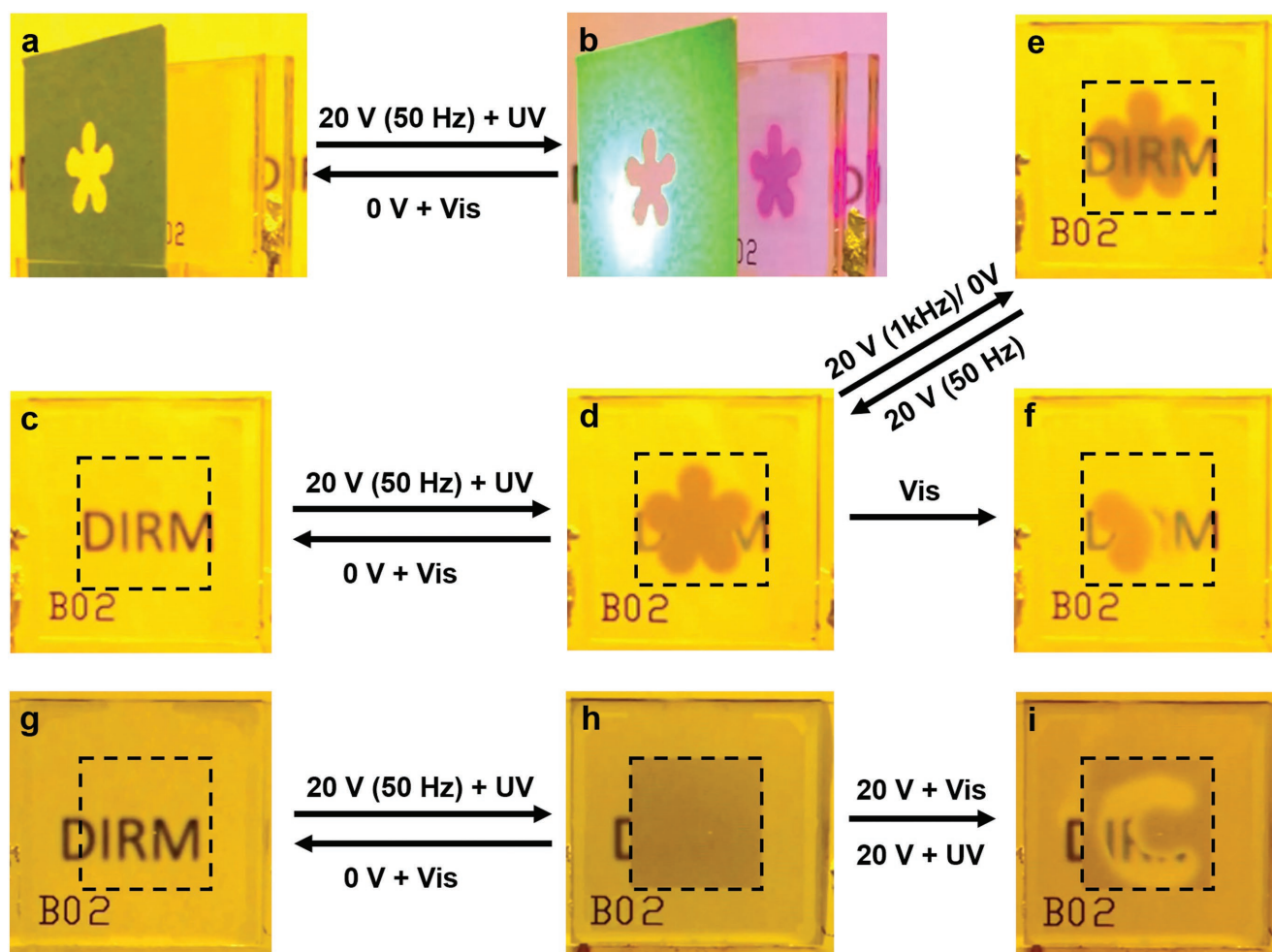


Figure 4. Photos show that EHDI can be locally generated and erased. a,b) The blue opaque flower is generated. c,d) The corresponding front view of (a) and (b), respectively. e) The blossom is partially erased by focused green light. f) The blue opaque flower turns into transparent when addressed with high-frequency voltage or switching off the AC field. g–i) Direct writing on the EHDI background through focused green light. All the samples are kept in the laboratory with yellow light to prevent MC to SP back reaction. The electric active area is highlighted by the dotted square. The voltage described in the pictures is root-mean-square voltage. The cell provides homeotropic alignment and the cell gap is 9 μm .

propose a complementary solution to partially erase the display while keep the desired information on the display as shown in Figure 4f. Besides displaying preprogrammed information, for example, by patterned electrodes or by masked exposure, direct writing is enabled by using a focused light source (Figure 4g–i). In this experiment, the colored EHDI scattering provides the background while the focused green light partially erases EHDI and leaves information on scattering sample.

In conclusion, we have presented a new approach in initiating electrohydrodynamic instabilities in liquid crystals under light illumination. EHDI can be triggered or removed by alternating UV and green light exposure within tens of seconds. We especially explore the light scattering effect originated from EHDI in which the sample can be modulated from nearly 100% transparent state to 16% transmittance. The light scattering from EHDI exhibits a strong shielding effect which is proposed to be useful in smart window applications. Using light to trigger EHDI provides possibilities to localize light scattering by mask-wise exposure or by local writing with a focused beam. We anticipate the use of the effect for an information display or a smart window in which the messages can be written, stored, and erased, either locally or globally. In addition, this technology possesses large potential applications, such as the dye-doped colored windows, segmented triggered window, and e-paper.

2. Experimental Section

Materials: 1',3'-Dihydro-1',3',3'-trimethyl-6-nitrospiro[2H-1-benzopyran-2,2'-(2H)-indole] (spiropyran) was obtained from Sigma-Aldrich. CTAB was purchased from Sigma-Aldrich. LCs with negative dielectric anisotropy were obtained from Jiangsu Hecheng Advanced Materials Co., Ltd. (HNG30400-200, $T_{N-I} = 94$ °C, $\Delta\epsilon = -8.3$, $\Delta n = 0.149$). The conventional EHDI mixture contains 0.05 wt% CTAB and 99.95 wt% LCs. The light-induced EHDI mixture contains 0.3 wt% of spiropyran and 99.7 wt% LCs.

Sample Preparation: Commercial cells (S100A090uT80, Instec; and XGH3030-10, Guohua Star) were used for experiments. The cells were filled with LC mixture at by capillary suction at 120 °C. The samples were cooled slowly to 80 °C and held for 2 h to eliminate any thermal instability before further cooled down to room temperature.

Sample Characterization: Samples were checked by optical microscope equipped with crossed polarizers (Nikon Ci Eclipse). Transmittance of the sample was measured by UV–vis–NIR spectrometer (PerkinElmer 750 and Ocean Optics HR2000+). The alternating electric field with a sinusoidal wave function was provided by a function generator (33220A, Agilent). The electric signal from the function generator was amplified through a high-voltage linear amplifier (F20A, FLC Electronics). The output voltage was measured by an oscilloscope (DSOX3032T, Keysight). A LED lamp (M365L2 and M530L3-C2, Thorlabs) was used to provide monochromatic light.

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.

Keywords

electrohydrodynamic instabilities, light scattering, liquid crystals, photochemistry

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