

Research Article

Reduced ionic effect and accelerated electro-optic response in a 2D hexagonal boron nitride planar-alignment agent based liquid crystal device

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Abstract: The presence of excess free-ion impurities in liquid crystals (LCs) gives rise to a number of problems in the electro-optical liquid crystal displays (LCDs), *e.g.*, slow electro-optical responses and image sticking effects. Here we experimentally present that the two-dimensional (2D) hexagonal boron nitride (*h*-BN) nanosheet can serve as a planar-alignment agent and as an ion-capturing agent at the same time in an electro-optic LC device. The 2D *h*-BN nanosheet is employed as a planar-alignment agent on one side of an LC cell, where the standard planar-aligning polyimide (PI) layer is used on the other side of the cell. The LC exhibits uniform planar-alignment in this *h*-BN/PI hybrid device. It is found that the free-ion impurities in the LC are significantly suppressed in this *h*-BN/PI hybrid cell compared to that in a standard PI/PI LC cell. The free-ion density is reduced in the hybrid cell due to the 2D *h*-BN nanosheet's ion-capturing process. The reduction of ionic impurities results in an accelerated electro-optic response of the LC in the *h*-BN based hybrid cell—which may have potential application for faster electro-optic devices.

1. Introduction

The existence of excess free ions [1,2] in liquid crystals (LC) causes complications in electrooptical liquid crystal displays (LCDs). The ion-related problems in LCDs include slow responses, short-term flickering, and long-term image sticking effects [3–9]. These free-ions generally come from the LC's chemical synthesis process, the LC cell's conductive electrodes [10,11], and the polyimide (PI) alignment layers [12]. Therefore, investigating the influence of the free ions on the LC's electrical, mechanical, and electro-optical properties [13–20] is an essential area of fundamental and applied research. Over the last decade, there has been another exciting direction of research which shows that the colloidal dispersion of nanomaterials, such as ferroelectric nanoparticles [21,22], titanium nanoparticles [23], carbon nanotubes [24–26], graphene [27–32], and fullerenes [23,33,34] in the LC can suppress the free-ion concentration significantly by the ion-trapping process. The ion-capturing process by doping nanomaterials in the LC can enhance the overall performance of LC-based electro-optic devices [23,30,35]. Here we present the fabrication of an electro-optic LC cell with the two-dimensional (2D) hexagonal boron nitride (h-BN) nanosheet as a planar-alignment agent on one side and the standard PI alignment layer on the other side of the cell. We then show that the presence of this 2D h-BN planar-alignment nanosheet can greatly suppress the free-ion concentration in the LC. Consequently, this h-BN/PI hybrid LC cell exhibits a faster electro-optic response, higher optical transparency, and an enhanced dielectric anisotropy than a commercial PI/PI LC cell. This investigation also falls in the current theme of research of the application of 2D materials in various optoelectronic devices [36–38].

2. Experiments, results, and discussion

In this section, we present (a) the fabrication of the *h*-BN based planar LC cell, (b) the ion-trapping phenomenon by the *h*-BN alignment agent, (c) the effect of free-ion reduction on the electro-optic switching of the LC, (d) the optical transmission spectroscopy of the *h*-BN based cell, and finally, (e) the effect of free-ion reduction on the anisotropy of the LC.

2.1. Fabrication of the 2D h-BN based LC device

Alternating boron and nitrogen atoms in a regular sp^2 -bonded honeycomb pattern with a lattice spacing of 1.44 Å [39] form the 2D h-BN nanostructure—which is an ultra-flat insulator [40] and has very high structural [41], thermal [42], and chemical [43] stability. To fabricate the h-BN/PI cell, chemical vapor deposition (CVD) grown monolayer h-BN film on a copper foil (commercially obtained from Graphene Supermarket, Inc) was transferred onto an indium tin oxide (ITO) coated glass substrate using the standard polymethyl-methacrylate (PMMA) assisted wet transfer method [44,45]. After the transfer process, no rubbing was done on the *h*-BN film. This *h*-BN/ITO glass slide and a unidirectionally rubbed planar-aligning PI layer on an ITO coated glass slide were placed together to make the *h*-BN/PI hybrid cell with an average cell-gap, $d = 21.5 \,\mu\text{m}$. The 2D *h*-BN nanosheet and the PI layer faced each other inside the cell. The standard PI/PI planar cell ($d = 20 \pm 0.5 \,\mu\text{m}$, $A = 1 \,\text{cm}^2$) was commercially obtained from *Instec*, Inc. We fabricated another cell with a rubbed PI layer on one side and a plain ITO glass slide (with no h-BN or PI) on the other side. All the three cells, the commercial PI/PI cell, the hybrid h-BN/PI cell, and the PI/Glass cell had the same PI material (KPI-300B, Kelead Photoelectric Materials Co., Ltd.) for consistency. All these three cells were filled with liquid crystal E7 $(T_{\rm NI} = 60.5 \ ^{\circ}{\rm C}).$

The quality of planar-alignment of the LC in all the three cells was studied by rotating the cells under a crossed polarized optical microscope (Olympus BX61). Figure 1(a) schematically shows the commercial PI/PI cell. The micrographs in Figs. 1(b) and 1(c) present the LC texture in the standard PI/PI cell, where the nematic director, \hat{n} is at 45° (bright) and 0° (dark), respectively, with respect to the polarizer. Figures 1(d) and 1(e) present the micrographs of the LC in the PI/Glass cell, where the rubbing direction of the PI alignment layer at one side is at 45° and 0°, respectively, with respect to the polarizer. These two micrographs clearly show *nonuniform* bright and dark textures unlike the micrographs in Figs. 1(b) and 1(c). Therefore, the absence of a planar-alignment agent on the other side in the cell disturbs the uniform planar-alignment of the LC.

The *h*-BN/PI hybrid cell is schematically presented in Fig. 1(f) where the 2D *h*-BN nanosheet is employed as the planar-alignment agent on one side, and the standard unidirectionally rubbed planar-aligning PI is used on the other side. The bond length between B and N in the *h*-BN lattice is 1.44 Å [39]. The bond length between two carbon atoms in a hexagonal benzene ring is 1.40 Å. Therefore, this nano-architectural symmetry between the hexagons in both these species leads to an epitaxial interaction between them, and the benzene rings of the LC coherently align on the hexagonal lattice of *h*-BN [46]. This type of epitaxial interaction also exists between the nematic LC and the hexagonal lattice of other different nanomaterials, such as graphene [47–55] and carbon nanotube [56,57]. This spontaneous alignment mechanism of the LC on the hexagonal lattice is the fundamental principle behind designing the LC cell employing the 2D *h*-BN as a planar-alignment agent. This spontaneous planar-alignment of the LC and the *h*-BN lattice is illustrated in Fig. 1(g). This epitaxial interaction between the LC and the *h*-BN lattice is illustrated in Fig. 1(h) confirms the presence of monolayer *h*-BN on the copper foil before transferring onto the ITO coated glass substrate.

On the 2D h-BN crystal, the LC molecules can obtain three degenerate planar orientations separated by 60 degrees due to the three-fold rotational symmetry of the hexagonal structure. It is



Fig. 1. (a) A schematic representation of the standard PI/PI cell. Micrographs of LC E7 in the PI/PI cell under the crossed-polarized microscope with the director \hat{n} at (b) 45° (bright) and (c) 0° (dark) with respect to the polarizer. Micrographs of the PI/Glass cell filled with LC E7 under the crossed polarized microscope where the PI's rubbing direction is at (d) 45° and (e) 0° with respect to the polarizer. (f) A schematic representation of the *h*-BN/PI hybrid cell containing the unidirectionally rubbed PI alignment on one side and the 2D *h*-BN nanosheet on the other side of the cell. (g) the planar-alignment of nematic LC molecules (ellipsoids) on 2D *h*-BN (honeycomb structure) is illustrated. The epitaxial interaction between the LC and the *h*-BN lattice is schematically shown by matching the LC's benzene rings on the *h*-BN-honeycomb structure. (h) Raman signal of monolayer *h*-BN on copper foil. Micrographs of LC E7 in the *h*-BN/PI hybrid cell under the crossed-polarized microscope with the director \hat{n} at (i) 45° (bright) and (j) 0° (dark) with respect to the polarizer.

still possible to achieve uniform planar-alignment in-between two *h*-BN layers (without any PI) in an LC cell [58]. However, that process requires several steps to align two *h*-BN layers in the same direction to avoid the degenerate planar orientations in the cell [58]. Here, we have used one rubbed PI layer and one *h*-BN layer in the cell. The unidirectionally rubbed PI alignment layer at one side of the hybrid cell enforces the LC to choose the alignment on the *h*-BN surface parallel to the PI's rubbing direction. See Fig. 1(f). This mechanism minimizes the elastic distortion of the nematic director field in the cell and reduces the three-fold degeneracy to the uniaxial alignment of the LC on the *h*-BN layer. We have previously shown [58] that two *h*-BN layers can be successfully employed as the planar-alignment agent to fabricate a planar LC cell—which shows the classic electrically-controlled birefringence effect. In this present work, we show that the presence of the *h*-BN nanosheet as a planar-alignment agent on one side of the cell reduces the free ion concentration in the LC significantly. This ion-reduction results in an accelerated dynamic electro-optic effect and an enhanced dielectric anisotropy of the LC.

Figure 1(i) exhibits a bright uniform texture of the LC in the *h*-BN/PI cell under the microscope when \hat{n} is at 45° with respect to the crossed polarizers. When the cell is rotated by 45°, \hat{n} becomes parallel to the polarizer, revealing a *dark* uniform texture in Fig. 1(j). This result confirms that the 2D *h*-BN can serve as a planar-alignment agent on one side of the cell due to the hexagonal symmetry matching between the LC's benzene rings and the *h*-BN's honeycomb structure.

2.2. Measurement of free ions in the h-BN/PI cell

Now that we have established that the 2D *h*-BN nanosheet retains the uniform planar-alignment of the LC in the cell, we next show that the free-ion concentration is greatly reduced in the *h*-BN/PI cell than that in the standard PI/PI cell. The free-ion concentration, n_i in LC E7 in the PI/PI cell and the hybrid h-BN/PI cell was measured from the transient ion current, I_{ion} generated by inverting the polarity of the applied voltage across the cell [22,52,59]. When a square-wave voltage changing from + V to -V (*i.e.*, the voltage polarity is inverted) is applied across the cell, the LC molecules do not rotate as the director rotation depends only on the *magnitude* of E, and not on its *polarity* [60]. However, inverting the voltage polarity triggers the motion of the ions in the LC towards the opposite electrodes—which results in a transient ion current, I_{ion} in the cell. A square-wave peak-to-peak voltage of 20 V (i.e., +10 V to -10 V) at 1 Hz was applied using an Automatic Liquid Crystal Tester (Instec, Inc.) to generate I_{ion} as a function of time in both the cells as shown in Figs. 2(a), 2(b), and 2(c) at T = 25 °C, T = 40 °C T = 55 °C, respectively. Note that I_{ion} reaches its peak value when the positive and negative ions meet approximately at the middle of the cell. Finally, I_{ion} decays to zero when the positive and negative ions reach the opposite electrodes—which can be seen in Figs. 2(a), 2(b), and 2(c). The total free-ion transport in the cell then was calculated by taking the area under the I_{ion} vs. time curve. The free-ion concentration, $n_i = (\int_0^t I_{ion} dt)/A d$ was extracted using the known cell-gap d and active area A. Figure 2(d) presents n_i in both cells as a function of temperature, depicting that n_i is substantially suppressed (more than 75%) in the *h*-BN/PI hybrid cell. As a result, the ion-conductivity, σ_i is significantly declined in the *h*-BN/PI hybrid cell, as shown in Fig. 2(e).



Fig. 2. Ion current, I_{ion} as a function of time for LC E7 in the two cells at (a) 25 °C and (b) 40 °C, and (c) 55 °C after the polarity of the voltage is inverted across the cells. (d) Free-ion concentration, n_i as a function of temperature for LC E7 in the two cells listed in the legend. (e) Conductivity due to ion-transport, σ_i as a function of temperature for LC E7 in the two cells listed in the legend. Typical error bars are shown. Schematic representations: (f) the presence of free ions (small spheres) in the LC in the PI/PI cell, (g) the presence of fewer ions in the LC media due to some trapped ions on the *h*-BN layer.

In the existing reports in the literature [21–35], the nanomaterials (in the form of a colloidal dispersion) can trap free ions in the LC. Since the 2D *h*-BN nanosheet is directly exposed to the LC as a planar-alignment agent, it captures a significant amount of free ions in the *h*-BN/PI

hybrid cell in our experiment. Figures 2(f) and 2(g) schematically illustrate the ion-capturing process. It is important to point here that the PI/Glass LC cell also showed n_i values similar (within 5%) to that of the standard PI/PI cell. Therefore, the reduction of free ions is attributed to the *h*-BN's ion capturing process.

2.3. Dynamic electro-optic response of the h-BN/PI cell

The reduction of free ions in the LC results in a faster dynamic electro-optic response of LC [22,23,27,28,50,54]. Since the h-BN/PI hybrid cell shows a significant decrease in the free-ion concentration, we have studied the dynamic electro-optic response of this hybrid cell and compared that with the PI/PI cell. The dynamic electro-optic response of the LC in both the cells was studied using an optical setup with a 5-mW He-Ne laser beam ($\lambda = 633$ nm) sent through a polarizer, the LC cell (where \hat{n} was oriented at 45° with respect to the polarizer), a crossed analyzer, and into a photodetector. The output of the photodetector was connected to a digital storage oscilloscope to detect the change in the transmitted intensity through the cell as a function of time when a square-wave voltage of 25 Hz was applied across the cell. Transmittance responses were studied for several applied voltages much higher than the orientation threshold switching voltage, V_{th} . In Fig. 3(a), the left y-axis represents the normalized transmitted intensity of LC E7 in both the cells as a function of time when a square-wave voltage (30 V) was applied. The right y-axis shows the applied square-wave voltage (0 to 30 V) across the cell. After the voltage is turned on, transmitted intensity through the cell decreases. The time it takes to drop from 90% to 10% of its maximum value is defined as the optical switching on, τ_{on} . After the voltage is turned off, the transmitted intensity increases, and the optical switching off, τ_{off} is defined by the time it takes to rise from 10% to 90% of its maximum value. These two switching times, τ_{on} and τ_{off} are described as [61]

$$\tau_{\rm on} \propto \frac{\gamma_1 d^2}{\Delta \varepsilon \, \varepsilon_0 \, V^2 - K_{11} \, \pi^2}; \, \tau_{\rm off} \propto \frac{\gamma_1 d^2}{K_{11} \, \pi^2} \tag{1}$$

where γ_1 is the rotational viscosity, *d* is the cell-gap, $\Delta \varepsilon$ is the dielectric anisotropy, *V* is the applied voltage, ε_0 the is free space permittivity, and K_{11} is the splay elastic constant.



Fig. 3. The dynamic electro-optic switching response of LC E7 in the two cells. (a) Right *y*-axis: applied square-wave voltage profile. Left *y*-axis: normalized transmitted intensity as a function of time when a peak-to-peak voltage ($V_{pp} = 30$ V) is turned off at t = 0, and then turned on at t = 20 ms, for the two cells listed in the legend ($T = 20^{\circ}$ C). (b), (c) τ_{off} and τ_{on} , respectively, as a function of V_{pp} for the two cells listed in the legend. (d) Optical transmission as a function wavelength for the LC-filled PI/PI cell and the LC-filled *h*-BN/PI cell listed in the legend. The visible wavelength range is shown in the *x*-axis.

Figures 3(b) and 3(c) show τ_{off} and τ_{on} , respectively, for both the cells as a function of peak-to-peak voltage, V_{pp} . Clearly, τ_{off} is ~ 30% faster in the *h*-BN/PI hybrid cell despite its little higher cell-gap. If the backflow in the cell is neglected, τ_{off} is purely driven by the elastic interaction between the LC and the alignment substrate. A reduction of free ions lessens γ_1 [22,27,28] and also increases interactions between the LC molecules and the alignment layers [23]. Therefore, τ_{off} in the *h*-BN/PI hybrid cell is faster than the PI/PI cell. On the other hand, the driving voltage, V is the dominating factor for τ_{on} when $V >> V_{th}$ [53]. In our case, $V_{th} \approx 0.8$ V, and the applied voltage ranges from 20 V to 45 V. Since $V >> V_{th}$ here, V is the dominating factor for τ_{on} , and any small change in γ_1 has a minimal effect on τ_{on} . Therefore, Fig. 3(c) shows a very little change in τ_{on} . The total dynamic response time of the LC is defined as $\tau = \tau_{on} + \tau_{off}$. In the entire applied voltage range studied (20 V to 45 V), τ is always faster for the *h*-BN/PI hybrid cell. For $V_{pp} = 20$ V, τ is 15% faster and for $V_{pp} = 45$, τ is 25% faster in *h*-BN/PI hybrid cell.

2.4. Optical transmission spectroscopy of the electro-optic h-BN/ITO cell

The optical transmission spectroscopy was carried out for the LC-filled PI/PI cell and for the LC-filled *h*-BN/PI cell separately employing the FLAME-S-XR1-ES (*Ocean Optics, Inc.*) spectrometer and DH-2000-BAL UV-VIS-NIR (*Ocean Optics, Inc.*) light source. The transmission spectra for these cells in a wide wavelength range (300 nm to 1000 nm) are shown in Fig. 3(d). The *h*-BN/PI cell indicates more optical transparency than the commercial PI/PI cell. The thickness of the rubbed planar-aligning PI layer is about 60 nm, and the same of a 2D *h*-BN film is around 0.3 nm [62]. Replacing one PI alignment layer with the 2D *h*-BN nanosheet on one side decreases this effective layer thickness to less than 1 nm—which results in a reduced transmissive loss through this electro-optic LC device over a broad spectral range.

2.5. Enhanced dielectric anisotropy in the h-BN/ITO cell

The existence of a large number of foreign particles in the LC generally promotes disorder in the system and subsequently decreases the LC's scalar order parameter [63]. Free ions are also foreign charged particles and exist as impurities in the LC. The reduction of free ion impurities results in an increase in the scalar order parameter by enhancing the intermolecular association in the nematic phase [64]. The dielectric anisotropy in the nematic phase is defined as $\Delta \varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$, where ε_{\parallel} are the dielectric components parallel and perpendicular to \hat{n} , respectively. In the nematic phase, LC's dielectric anisotropy is proportional to the scalar order parameter [60], and therefore, an increased $\Delta \varepsilon$ indicates an improved orientational order.

The dielectric behavior of LC E7 in both the cells as a function of E (f = 1000 Hz) was investigated by the capacitance measurement technique [65]. Figure 4(a) shows the dielectric constant, ε as a function of E at T = 30 °C for both the cells. The LC in the *h*-BN/PI hybrid cell exhibits an enhanced value of $\Delta \varepsilon$ due to the reduction of free ions in this cell. Figure 4(b) exhibits the pretransitional behavior of $\Delta \varepsilon$ for LC E7 in both the cells. The result indicates that the orientational order of the LC is improved when the ionic impurities are greatly suppressed. This result is consistent with a recent report in the literature [64] that showed that the ion-trapping effect by TiO₂ nanoparticles in an LC enhances its $\Delta \varepsilon$ by improving the orientation order of LC molecules.

Note that $V_{\text{th}} = \pi \sqrt{K_{11}/\varepsilon_0 \Delta \varepsilon}$, and our additional measurements reveal that the splay elastic constant K_{11} increases due to the reduction of free-ions in the *h*-BN/PI cell. Since both K_{11} and $\Delta \varepsilon$ increase by a similar factor, the effective change in V_{th} is minimal.

It might be attractive to the LC community to know how the LC parameters are altered by reducing the ion concentration. The data shown in Fig. 4(b) can be used to obtain several parameters [66] of the pristine E7 and its purified counterpart whose ions are partially in captivity.



Fig. 4. (a) Dielectric constant, ε as a function of applied rms field *E* (*f* = 1000 Hz) in the nematic phase (*T* = 30 °C) of LC E7 in the two LC cells listed in the legend. (b) Pretransitional behavior of dielectric anisotropy, $\Delta \varepsilon$ of LC E7 in the two cells listed in the legend.

3. Conclusions

The appearance of the uniform planar-alignment in the h-BN/PI cell and its absence in the PI/Glass cell is a clear indication that 2D h-BN nanosheet can serve as a planar-alignment agent. While retaining the uniform planar-alignment in the cell, the 2D h-BN nanosheet considerably diminishes the free-ion concentration in the LC by the ion-capturing process. This result of ion-capturing by h-BN may have potential applications for refining the LC to remove excess ionic impurities without further chemical synthesis. The reduction of ionic impurities results in a faster electro-optic response and a higher anisotropy of the LC in the h-BN/PI cell. The overall optical transmission through the h-BN/PI is also improved due to the lower absorption by the 2D h-BN nanosheet.

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Disclosures

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