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Graphene oxide liquid crystals for reflective displays without polarizing optics†

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The recent emergence of liquid crystals of atomically thin two-dimensional (2D) materials not only has allowed us to explore novel phenomena of macroscopically aligned 2D nanomaterials but also has provided a route toward their controlled assembly into three-dimensional functional macrostructures. Using flow-induced mechanical alignment, we prepared flakes of graphene oxide (GO) in different orientational orders and demonstrated that GO liquid crystals (LC) can be used as rewritable media for reflective displays without polarizing optics. With a wire or stick as a pen, we can make the surface of GO LC reflective and bright, and we can then manually draw lines, curves, and any patterns with dark appearance. The contrast between bright and dark features is due to anisotropic optical responses of ordered GO flakes. Since optical anisotropy is an intrinsic property of 2D structures, our observations and demonstration represent one of many potential applications of macroscopically aligned 2D nanomaterials.

A variety of atomically thin two-dimensional (2D) materials have been synthesized or fabricated since the preparation of single-layer graphene by dry mechanical exfoliation.^{1–5} Beside their great potential in basic and applied research, their optical properties and photonic applications have attracted considerable attention.⁶ Two-dimensional nanomaterials have found applications in diverse areas such as transparent contacts, high-speed broadband photodetectors, high-efficiency light-emitting diodes and flexible solar cells.^{6–8} With the development of wet exfoliation,⁹ a large quantity of discrete

layers of 2D materials can be routinely obtained and suspended in solvents, and the recent emergence of liquid crystals (LCs) of 2D nanomaterials—a 3D self-assembly of suspended 2D flakes, has created a new opportunity for their novel applications.¹⁰ Liquid crystals of 2D materials not only allow people to study the basic interaction among flakes and their 3D macroscopic order, but also provide a means to assemble flakes in 3D, creating new structures with desired functionalities. Liquid crystals of graphene oxide (GO) are the first reported and studied among 2D nanomaterials.^{10–12} In this communication, we systematically investigated the optical properties of GO liquid crystals, and we discovered that mesoscopic order of GO liquid crystals can be revealed by scattering or transmission of unpolarized light. By using a simple stylus as a writing and alignment tool, we demonstrated reflective displays as new photonic applications of GO LCs: arbitrary patterns can be created, preserved, erased, and viewed without any polarizing optics.^{13,14}

Ink writing on a white paper and chalk writing on a blackboard are excellent examples of reflective displays, and they represent some of the oldest information recording and communication technologies. The modern electronic version of a reflective display is widely used in portable devices such as Kindle and Boogie Board.^{13–15} Compared with back-illuminated LC displays, it has the advantages of being low cost and consuming little or no energy because it makes use of ambient light and does not require polarizing optics. Kindle and Boogie Board represent two mature reflective display techniques. Kindle utilizes the difference in electrophoresis between black and white pigment particles embedded in microcapsules,¹⁵ while Boogie Board makes use of the structural transition between planar and focal conic textures of cholesteric LCs.^{13,14} The Bragg periodicity of cholesteric molecules in the planar structure is responsible for the strong reflection, and the reflective display demonstrated here is based on the anisotropic optical response of GO flakes. Because optical anisotropy is an inherent property arising from the shape anisotropy,^{16,17} orientation-dependent light scattering and the related application in reflective displays can

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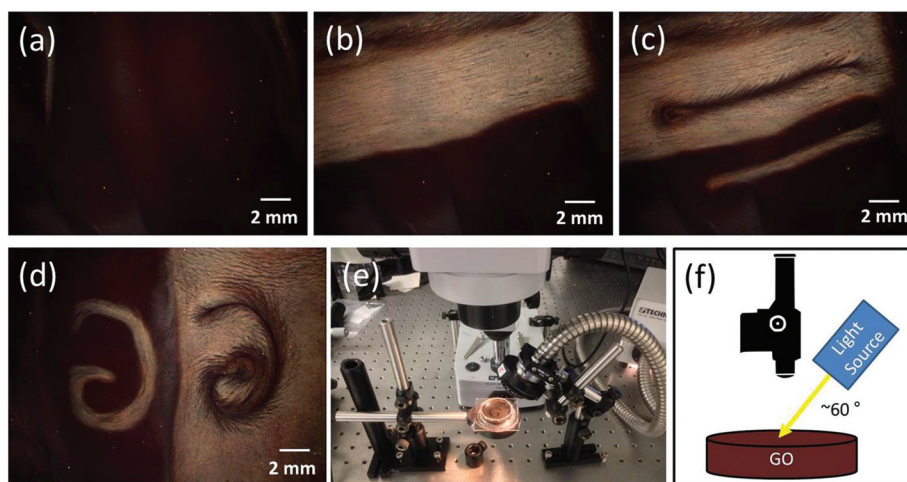


Fig. 1 Two ways to create and display arbitrary features on the surface of graphene oxide (GO) liquid crystals (LCs). (a) A clear, dark surface with weak light back scattering. (b) A bright surface region created from the dark surface. (c) Straight and (d) curved lines created in both dark and bright regions. (e, f) Photo and schematic of the experimental setup showing how the GO LCs are illuminated and imaged.

be observed and realized using other 2D materials. The recent demonstration of the manipulation of the orientation of GO flakes under an electrical field further paves the way for the controlled assembly of 2D nanomaterials and their new applications in many fields of science and technology.¹⁸

Graphene oxide flakes are synthesized in two separate laboratories using slightly different procedures, but both procedures use a similar chemical exfoliation based on Hummers' method.^{19–21} GO liquid crystals are prepared by dispersing flakes in de-ionized water. Fig. 1a–d show that arbitrary and high-contrast surface textures can be created on a GO LC surface. These textures can be conveniently captured using a reflective imaging setup as shown in Fig. 1e and f. As can be seen, there are two ways these surface textures can be created and imaged: reflective bright textures on a dark background surface or dark features on a reflective bright surface. Unless stated otherwise, no polarizers are used to create these images.

Fig. 2 and its four insets show additional examples of surface textures and symbols. Like most things that require external light illumination, the visual rendition of such manually created surface textures is strongly dependent on the lighting and imaging conditions. In this case, the photos were taken under natural sunlight with a commercial Canon camera (EOS60D). The size of the GO flakes might also be a factor that causes the difference in brightness and contrast between Fig. 1 and 2. The average size of the GO flakes in Fig. 2 is 8 μm ,¹⁹ while the average size of the flakes in all of the other figures is 15 μm .²⁰

These surface textures and letters are drawn manually using a thin stick or wire in exactly the same way as writing with a pen or stylus. Fig. 3 shows how a GO LC is prepared for writing and how writing will affect the orientation of the GO flakes. Fig. 3a and b represent two types of featureless surfaces with weak light scattering. In Fig. 3a, the GO flakes are randomly oriented in three directions, resulting in an optically isotropic GO solution, and in Fig. 3b the GO flakes are vertically



Fig. 2 More examples of surface features: vortex-like structure and four letters. A vortex was created on a dark GO LC surface by making the liquid turn clockwise in a petri dish. The letters "G," "D," "U" and "T" were created on bright GO LC surfaces.

oriented, but their directors are freely oriented in the X-Y plane. The GO LC in Fig. 1a is an excellent example of a flake suspension depicted in Fig. 3b. In order to create a large-area, bright reflective surface, we can simply sweep the surface with a stick parallel to the surface, which will turn flakes and make them aligned nearly parallel to the surface (GO flake directors in the Z direction). Fig. 3c illustrates the effect of this surface sweeping on initially randomly oriented GO flakes, and it should be noted that this creation of bright surfaces works regardless of the initial orientations of GO flakes. Fig. 1b is a good example of a bright surface created from a dark one. To create a dark line on the bright reflective surface, we can hold the stick vertically in the GO LC and then slide it in any direction, as shown in Fig. 3d. This movement aligns flakes vertically along the path of sliding, and this is the way in which the dark line and curve in bright regions in Fig. 1c and d are created. In contrast, the bright line and curve in Fig. 1c and d

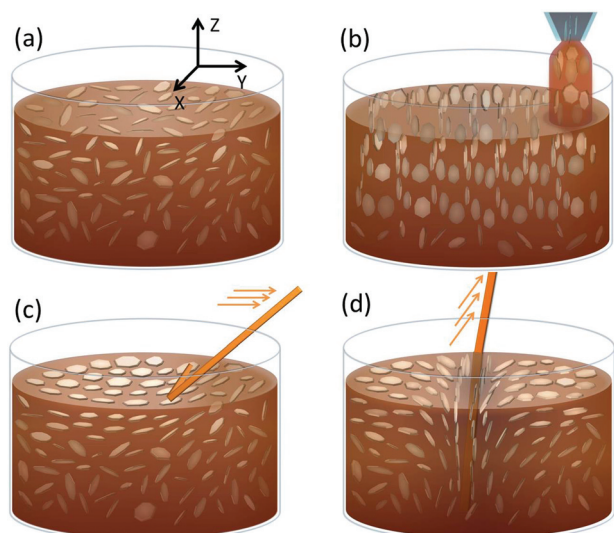


Fig. 3 Schematics of orientation of GO flakes before and during writing. (a) An isotropic GO liquid suspension with randomly oriented flakes. Z is defined perpendicular to the liquid surface. (b) Creation of a clear, dark LC surface by injecting a GO flake suspension from a capillary glass tube onto a petri dish. The directors of flakes (perpendicular to the GO flakes) are oriented in the X - Y plane. (c) Creation of a bright surface by sliding a stick along the LC surface. The flake directors become aligned in the Z direction. (d) Creation of a dark line by putting the stick vertically in the liquid and sliding it horizontally.

are created in the same way as the bright surface is created, but with a smaller flat tip.

The illustrated orientations of GO flakes in Fig. 3d can be conveniently confirmed using polarizing optics. Fig. 4a–c show the reflective image of a dark line as well as its transmission

images between two crossed polarizers. The vertical alignment of the GO flakes along the line is evident because the line becomes bright in transmission when both polarizers are rotated 45 degrees from the line but becomes dark when one of the polarizers is parallel to the line. This alignment of the GO flakes is further confirmed by their anisotropic optical absorption. Fig. 4e and f show optical transmission images when the incident light is polarized perpendicular to or parallel to the plane of the flakes.¹⁷ The alignment of GO flakes can be most conveniently observed in transmission without using any polarizers; as shown in Fig. 4d, the line becomes brighter than the surrounding region, indicating that the apparent dark line in reflection is actually more optically transparent. It becomes apparent that the stronger optical transmission through the GO flakes aligned by the stick is not due to a lower concentration of flakes than the surrounding region. Like reflective displays, GO LC can be equally used for transmissive display without polarizing optics.

The creation of a bright surface such as that shown in Fig. 3c can be considered to be an action of erasure. In combination with the writing in Fig. 3d, dark surface textures on a bright background can be generated and erased repeatedly. To demonstrate this rewritable property of GO LCs, we performed a series of write–erase tests. As can be seen in Fig. 5, after each erasure the previous patterns become almost invisible in reflection. However, from the transmission images between cross-polarizers before and after writing shown in Fig. 5a and f, the effect of writing is still visible. This obvious contrast between the reflection and transmission images occurs

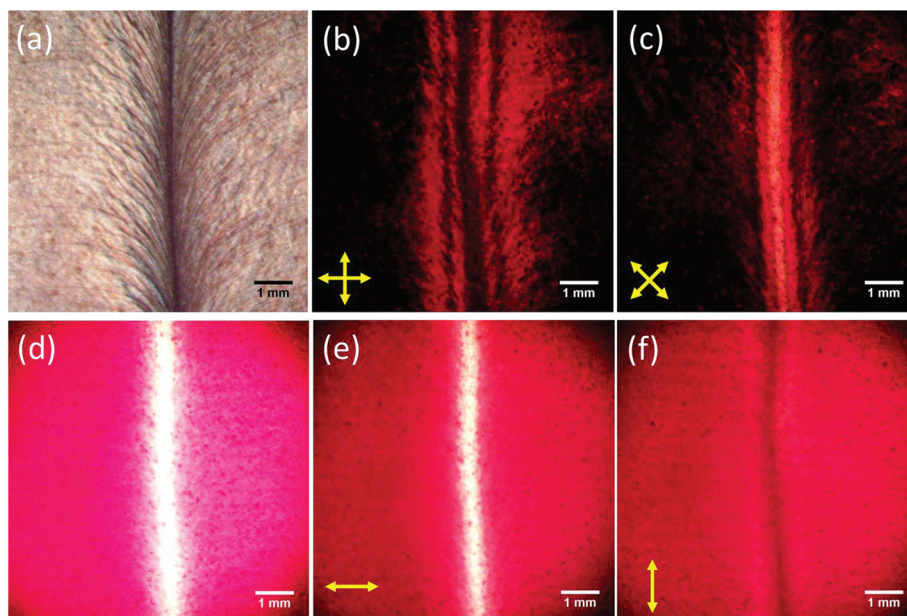


Fig. 4 Images of a straight line in reflection and transmission. (a) Reflection and (d) transmission with unpolarized light. (b, c) Transmission images between two cross polarizers. (e, f) Transmission images with incident light polarized (e) perpendicular to or (f) parallel to the line.

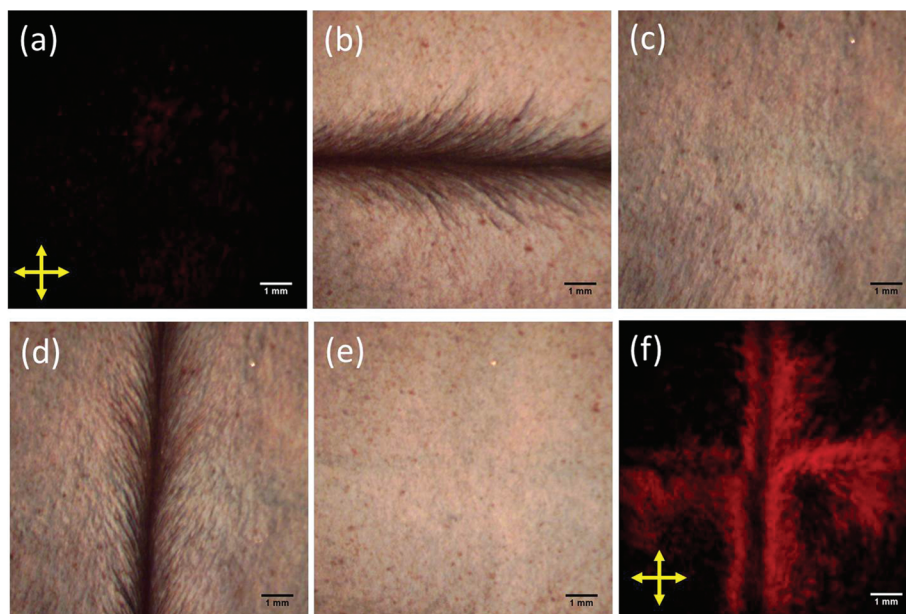


Fig. 5 Rewritable and erasable characters of GO LC. (a) Transmission image of nearly isotropic LCs between two cross polarizers before line drawing. (b–e) Reflective images of the same GO LC surface after the sequence of write–erase–write–erase. (f) Transmission image of GO LCs in (e).

because the action of erasure only aligns the GO flakes near the surface, and the reflective images are only sensitive to surface GO flakes, while the writing aligns the GO flakes both near and below the surface. Local erasure of small features can be achieved using a pen with a smaller tip.

The alignment of GO flakes is induced by shear flow created by relative motion between the stick and the liquid. Such alignment has been demonstrated with GO LCs and nanowires.^{17,22–24} Although the active moving object in our case is the stick rather than the LC, the effect is the same. As reported by Guo *et al.*,²² the GO flakes will form a concentric layer driven by homeotropic anchoring on the surface of the stick and become aligned in the wake of flow behind the stick. This picture of alignment is also valid when the path of the stick is curved, as shown in Fig. S1.† The flow alignment of the GO flakes is also used to create vertically oriented GO flakes, and as shown in Fig. 3b these flakes are aligned along a glass capillary tube when the GO solution is injected into a petri dish. Their orientations remain the same when the liquid spreads over the dish, however the flakes are not vertically aligned as well as those by the stick alignment, as is evident from some white background in Fig. 1a. The imperfect alignment by flow can also be seen from a low order parameter of ~ 0.4 as compared to ~ 0.8 or larger in typical liquid crystals.¹⁷ In contrast, the aligned GO flakes such as those shown in Fig. 4 exhibit an order parameter as high as 0.7.

As mentioned above, the flakes aligned perpendicular to the LC surface appear dark in reflection, while those parallel with the LC surface show stronger reflection and appear bright. Note that this relatively strong reflection is due to the reflection from multiple GO flakes, which can form domains,

so the flake schematics in Fig. 3 do not necessarily represent individual flakes. Such mesoscopic ordering of GO flakes is a result of their mutual interactions, and it is a basic and unique property of all LCs. Because the lateral size of the GO flakes is on the order of $10\ \mu\text{m}$, which is much larger than the wavelength of visible light, such a stack of GO flakes or flake domains can be treated as a reflective mirror. The only difference is that unlike spectral reflection from a flat mirror, the scattering from GO flakes is very diffusive: the brightness and contrast of surface features do not strongly depend on the illumination and imaging directions. We believe that this diffusive reflection is mainly caused by two things: (1) GO flakes are not perfectly flat—they may have wrinkles or folding structures, and (2) the flakes have different orientations and are not perfectly aligned parallel to the surface, although their average directors are in the Z direction.

The dark appearance of GO flakes perpendicular to the surface indicates weak back scattering of light, which is due to a combined effect of shape anisotropy, dielectric anisotropy, and the macroscopic order of the GO flakes. First, the back scattering is weak because the effective light scattering cross-section is small when light is incident on the edges of the GO flakes. This mechanism is similar to the reason that surfaces with vertically grown wire or wire-like nanostructures are strongly absorptive and appear black.²⁵ Second, because the out-of-plane dielectric constant or refractive index is smaller than the in-plane value, light with out-of-plane polarization will have less back scattering, which will reduce the total back scattering of light. Finally, it is well known that strong light scattering is caused by a medium's density and structural or index fluctuation. Aligned domains of GO flakes will become

more transparent and exhibit less light scattering compared with randomly oriented flakes, as shown in Fig. 3a. This property has been frequently observed in LCs,^{26,27} and has been successfully used in smart windows: the window is transparent and clear when the domains of LCs are aligned by an electrical field, but it becomes diffusive and milky when domains are randomly oriented.²⁸

High optical anisotropy is a unique property of atomically thin 2D materials, in which the in-plane dielectric constant is typically larger than the out-of-plane constant. This property is not shared by many other nanomaterials, such as nanowires.²⁵ Graphene is an excellent example;¹⁶ its out-of-plane refractive index is close to one, but the in-plane index has a larger real component as well as a large imaginary part, and this polarization-sensitive absorption has been used to make broadband polarizers. Graphene oxide flakes have inherited graphene's optical anisotropy, although the precise index of individual GO flakes has not been reported,^{17,29} and it may depend upon the synthesis method, among other factors. The bright and dark appearance of the line pattern shown in Fig. 4e and f provides direct evidence of anisotropic optical absorption of GO flakes. The strong transmission with unpolarized light in Fig. 4d is mainly due to the reduced optical absorption in Fig. 4e, although another factor is the reduced scattering of the incident light through this more-ordered region of GO flakes.

To summarize the above discussion, the contrast between bright and dark features in reflective displays of GO LCs is a result of multiple contributions from scattering, absorption, and transmission, and Fig. 6a and b provide another excellent example of such an effect. First, as anticipated from any reflec-

tive display, the vertically aligned GO flakes appear dark regardless of the polarization of incident light, but a closer look reveals that the line is darker and thicker when the polarization is parallel to the line. Based on the difference between the in-plane and out-of-plane optical constants, the backscattered light should be stronger when the incident light is parallel to the line or planes of the GO flakes, which should create a relatively brighter color for the line in Fig. 6a than in Fig. 6b. We believe that multiple scattering from flakes below the LC surface is responsible for this discrepancy. Incident light that is perpendicular to the line will penetrate deeper below the surface, and some of the light will be back scattered from flakes or the bottom of the petri dish below the surface, eventually contributing to the total back-scattered light and making the line lighter.

Such a combined effect can be used to understand the evolution of contrast as a function of GO weight concentration. Fig. 6c–h show the images of a similar line in GO liquids with low to high concentrations. As can be seen, the contrast becomes more visible only when the concentration reaches 0.05 wt%, and the contrast becomes diminished at concentrations lower than 0.05 wt% or higher than 0.4 wt%. The contrast is low for the low-concentration GO suspension because both reflection and absorption are weak due to lower numbers of aligned GO flakes. At the concentration of 0.05 wt%, the line does not appear dark because of low optical absorption, as well as the considerable back-scattered light from below the surface, as discussed above. For very high concentrations, the viscosity of the GO solution increases so significantly that it becomes difficult to change and align the orientations of the GO flakes.

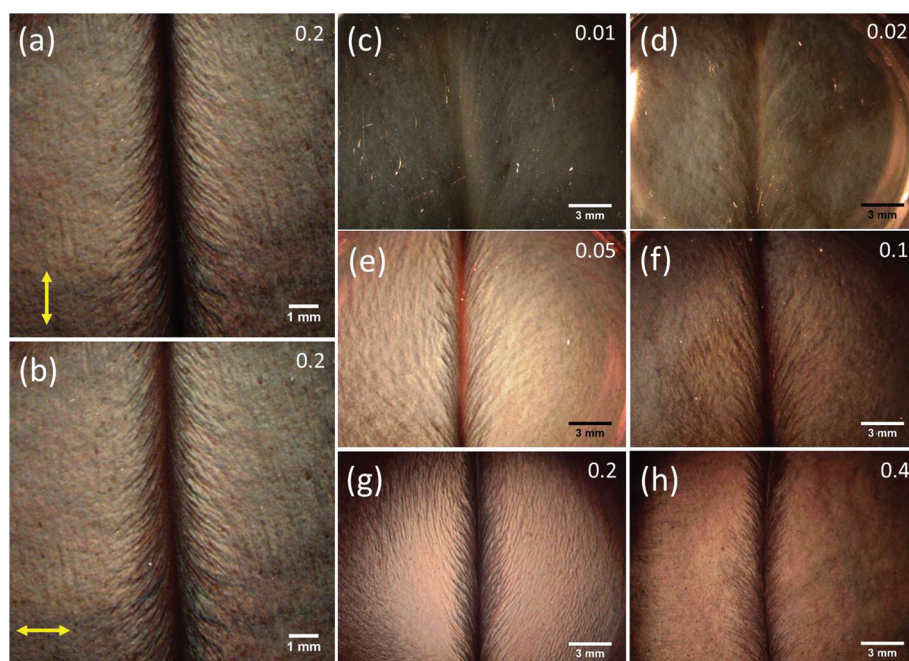


Fig. 6 Reflective images of a straight line in GO solution under polarized light or with different weight concentrations. (a, b) Under incident light with polarization parallel to (a) and perpendicular to (b) the line. Concentration: 0.2 wt%. (c–h) GO solutions with increasing concentrations from 0.01 to 0.4 wt%.

Previous studies have shown that a GO flake solution with a weight concentration <0.05 wt% is isotropic and that solutions with higher concentrations begin to exhibit a higher fraction of the nematic phase.^{17,18,22} Such concentration-dependent phase transitions can be conveniently observed by monitoring the dynamics of aligned GO flakes, as shown in Fig. S2–4.† As can be seen, the initially bright line becomes dimmer and disappears in about 90 s for a solution with 0.01 wt%, but the alignment persists longer for solutions with higher concentrations, and no obvious decay is observed when the concentration reaches 0.05 wt%. Such a concentration-dependent retention time allows us to choose the right concentration of GO flakes for the right application.

The above observations show that ordered structures of GO flakes can be temporarily created in otherwise isotropic solutions. Conversely, an isotropic phase can be generated in a nematic LC phase. Such isotropic phases are actually frequently created when we prepare 0.2 wt% GO LCs for many experiments in this work. To do that, we manually shake the solution randomly using the petri dish. Except for GO flakes near the surface that have been partially aligned parallel to the surface, flakes inside the solution are believed to have random orientation as shown in Fig. 3a. Such an isotropic phase shows a relatively strong scattering of light but exhibits no birefringence, as indicated in Fig. 4 and 5. If we shake the dish by the hand and make the liquid spin slowly in one direction, a vortex-like pattern as shown in Fig. 2 will be generated, provided that the concentration is higher than 0.4 wt%. Note that the liquid near the wall of the petri dish shows weak back scattering because it does not move as the rest of the solution.

We demonstrate repeated manual writing of dark features on a bright background in Fig. 5, and a similar demonstration of bright features on a dark background, as shown in Fig. 1, can also be accomplished, with the exception that the erasure or preparation of a dark surface requires the extraction and injection of GO solution through a capillary tube. The recent demonstration of alignment of GO flakes using an electrical field has solved this erasure problem.^{18,30} Using a similar technique that has been employed in Boogie Board and other LC displays, GO flakes can be aligned vertically between two transparent electrodes by applying a voltage, and thus a device similar to Boogie Board or an electronic blackboard can be demonstrated. It should be noted that the electrical field cannot fix the directors of GO flakes, it can only align them in a pattern shown in Fig. 3b if an electrical field is applied between the top and bottom surfaces of the LC.

Conclusions

In conclusion, we created GO flakes with different macroscopic ordering of orientation using mechanical flow alignment. We systematically studied their optical responses, including back scattering, absorption, and transmission under different polarizations of light. A relationship between scattering and orientation of GO flakes was established and explained, and a

reflective display using GO LC as a rewritable medium was demonstrated. Because shape and optical anisotropy are the basic properties of 2D materials, the observed phenomena and demonstrated applications are applicable to any 2D nanomaterials. The creation of macroscopically ordered structures from individual flakes enables new opportunities for basic research and device applications of 2D nanomaterials.

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