Laser-directed hierarchical assembly of liquid crystal defects and control of optical phase singularities

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Topological defect lines are ubiquitous and important in a wide variety of fascinating phenomena and theories in many fields ranging from materials science to early-universe cosmology, and to engineering of laser beams. However, they are typically hard to control in a reliable manner. Here we describe facile erasable "optical drawing" of self-assembled defect clusters in liquid crystals. These quadrupolar defect clusters, stabilized by the medium's chirality and the tendency to form twisted configurations, are shaped into arbitrary two-dimensional patterns, including reconfigurable phase gratings capable of generating and controlling optical phase singularities in laser beams. Our findings bridge the studies of defects in condensed matter physics and optics and may enable applications in data storage, singular optics, displays, electro-optic devices, diffraction gratings, as well as in both optically- and electrically-addressed pixel-free spatial light modulators.

opological defect lines play a prominent role in many scientific fields[1–15]. For example, optical orbital angular momentum transfer can be enabled by optical vortices, "dark thread" singular lines around which the light's momentum swirls[2–4]. State of the art displays with unprecedented refresh rates and controllable viewing angles[16] are nowadays made of a liquid crystal (LC) in the so-called "blue phase" that contains a periodic three-dimensional (3D) network of defect lines threading through the medium[16–18]. Defects like cosmic strings[14] and vortex lines in electron beams[8] are hard to obtain and study but often can be understood by probing defects in other topologically similar but more easily accessible systems, such as LCs and laser beams[1–5,8,14].

A nematic LC is typically comprised of rod-like molecules that spontaneously orient themselves along a common direction, described by the so-called director $\mathbf{n}$ with nonpolar symmetry ($\mathbf{n} \equiv -\mathbf{n}$), which is the medium's optical axis[1,17,19,20]. Nematic LCs are true anisotropic fluids, which can flow under shear or gravity while maintaining orientational order, although flows, confinement, and various external fields can introduce large-scale spatial changes of this molecular alignment commonly described by the director field $\mathbf{n}(\mathbf{r})$[17]. The fluid nature of LCs - along with the facile response to external fields, surface confinement and temperature changes - often gives rise to various defects, most notably the so-called "disclinations," the defect lines on which orientational order is disrupted and the orientation of LC molecules typically cannot be defined[1,17]. Even the name "nematic" is derived from a Greek word "thread" and refers to ubiquitous defect lines found in these LCs[17]. Disclinations are classified according to their strength s defined as the number of revolutions by 2π that the director makes around the defect core when one circumnavigates the core once[17]. However, typically occurring because of symmetry-brea LC

cross-section is shown in Fig. 1f, in which fluorescence intensity scales as $\propto \cos^6\phi$, where ϕ is the angle between the spatially varying $\mathbf{n}(\mathbf{r})$ and the linear polarization direction of the excitation laser. Using this and other images obtained for various polarization states and cross-sectional planes (supplementary Figs. S3 and S4), we reconstruct the director structure schematically shown in Fig. 1e (see supplementary information for details). Experimental POM (Fig. 2c,e) and 3PEF-PM textures (Fig. 1f) closely resemble the cordramatically as compared to diffraction patterns due to gratings with individual edge dislocations (Fig. 3). The corresponding phase distributions (Fig. 4c) reveal inter-connected screw-edge dislocations: two screw dislocations having elementary charge of opposite signs are connected by an edge dislocation in the phase profile across which the phase has a sharp jump from $-\pi$ to $+\pi$[26–28]. On the other hand, two slightly separated dislocations with the same **b** in a grating shown in the supplementary Fig. S6a produce two off-axis rotating elementary phase singularities of the same sign $N=\pm1$ in the $n_{do}=\pm1$ beams and $2n_{do}$ such defects in the beams of higher order[26]. The angle α

medium within the periodic grating is strongly dependent on the polarization of this beam. The ability to control intensities of different diffraction orders by varying relatively low applied voltages (0.5–2 V) and also ‘‘erasing’’ these gratings at somewhat higher ($\sim$5 V) is of great interest for a number of important applications of phase diffraction gratings[29–33].

Times needed for optical generation of the fingers structures are typically predetermined by the fastest speed of continuous scanning of a focused laser beam using galvano mirrors that still yields reliable generation of uninterrupted fingers (100 μm/s in the case of using a 10x objective). 2D patterns of fingers can be generated within times ranging from a fraction of a second to minutes, depending on complexity of the pattern. The fingers structures can be unwound at applied voltages of 5 V and higher or modified at lower voltages within a typical response time of 5–10 ms, comparable to the res-

One of the major recent research efforts in soft matter and other branches of condensed matter physics is directed at control of topological defects using fields, colloids, optical tweezers, confinement, chirality, and other means[34–44]. Our findings greatly extend the current capabilities and possibly also the utility of such control of defects. Furthermore, using laser generated patterns of defects as a model system, one can obtain insights into how defects in condensed

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(tending to reorient the director to be parallel to the electric field $\vec{}$) overcomes the elastic torque (tending to preserve the initial uniform alignment).[1] When the laser beam̂s waist ***w*** is

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dislocations shown in Figs. 1b, 3a-c, 4a, 5a and supplementary Figure S2a. We have used the theoretical profiles of the layered structure around dislocations obtained in the framework of nonlinear theory of lamellar media[3] to generate fingers gratings with dislocations. For example, to generate grating patterns with an elementary dislocation (Fig. 3a) and with dislocations having larger Burgers vectors (Fig. 3b,c), we have programmed steering of the laser beam along trajectories obtained using the theoretical expression for the layer displacement derived by Kamien and co-workers.[3]

a

periodic arrays of CFs. An example of the minimized CF array obtained from such an initial condition is displayed in the Supplementary Fig. 2b.

3.

We employ the Stokes polarimetry method[8] to characterize phase singularities in the studied diffraction patterns. To do this, we use a setup shown in the Supplementary Fig. S5 that allows us to select beams of different diffraction orders and then map their two-dimensional phase profiles by analyzing the corresponding two-dimensional patterns of Stokes parameters which carry polarization, phase, and intensity information.[9-12] Using a beam splitter BS1, we split the HeNe laser beam into two beams that are then polarized in two orthogonal directions by polarizers P1 and P2. We pass one of the beams through the diffraction grating and then select the diffracted beam of the diffraction order of interest using a mirror. The 2nd beam is used for reference. The diffracted beam and the reference beam are recombined using the beam splitter BS2. We then use a quarter-wave plate and a polarizer P3 for the measurement of different Stokes parameters by measuring the corresponding 2D intensity distributions *I*(

b

c

]Q ": %:BB:GP .Q %Q Y .FCGHP (Q $+(#+84,&4 0) !,26,' 3845%-4 W-L;CF9 2B>J:FG>HM .F:GGP ,:K 5CF?P]eeaXQ

^Q 0A6@MI?=P "Q"QP)6DIH6P "QP)68=MBG?>P Q 3QP)INA>BP Q ,QP 8?:FA6BP .Q (QP 1KCA7@MP !Q 4QP *::P 1QP 1F>J:9>P /Q .QP 6B9 .F6G69P .Q ,Q -DH>86@@M <:B:F6H:9 F:8CB;><IF67@: D=CHCB>8 GHFI8HIF:G C; :@6GH>8 EI6G>D6FH>8@:G >B ;FIGHF6H:9 8=C@:GH:F>8 @>EI>9 8FMGH6@GQ "159 713(44 =;P bdc\Ubdd\ W^\]^XQ

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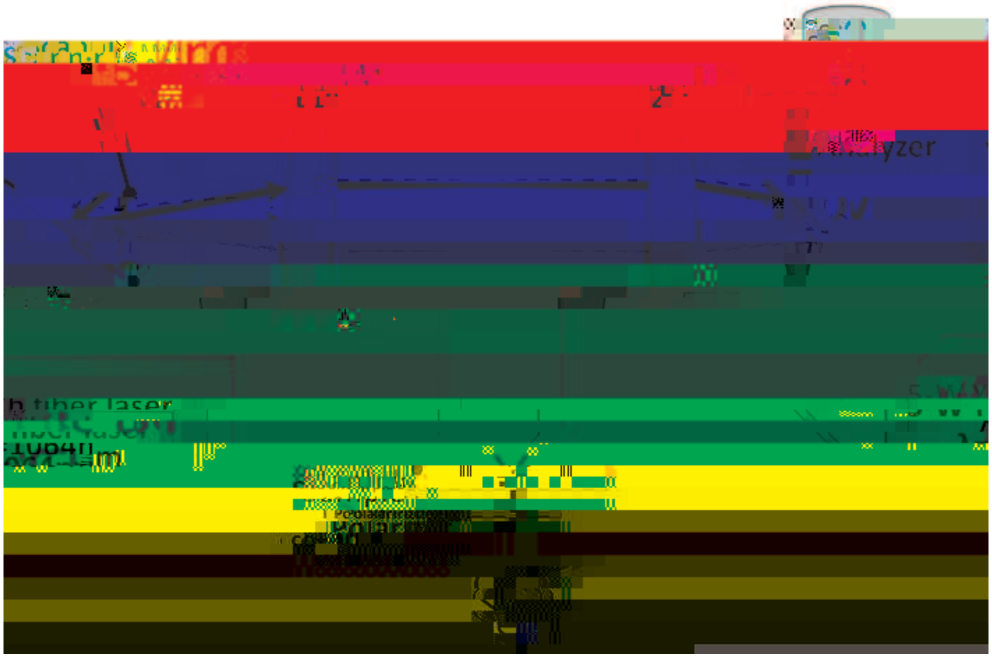
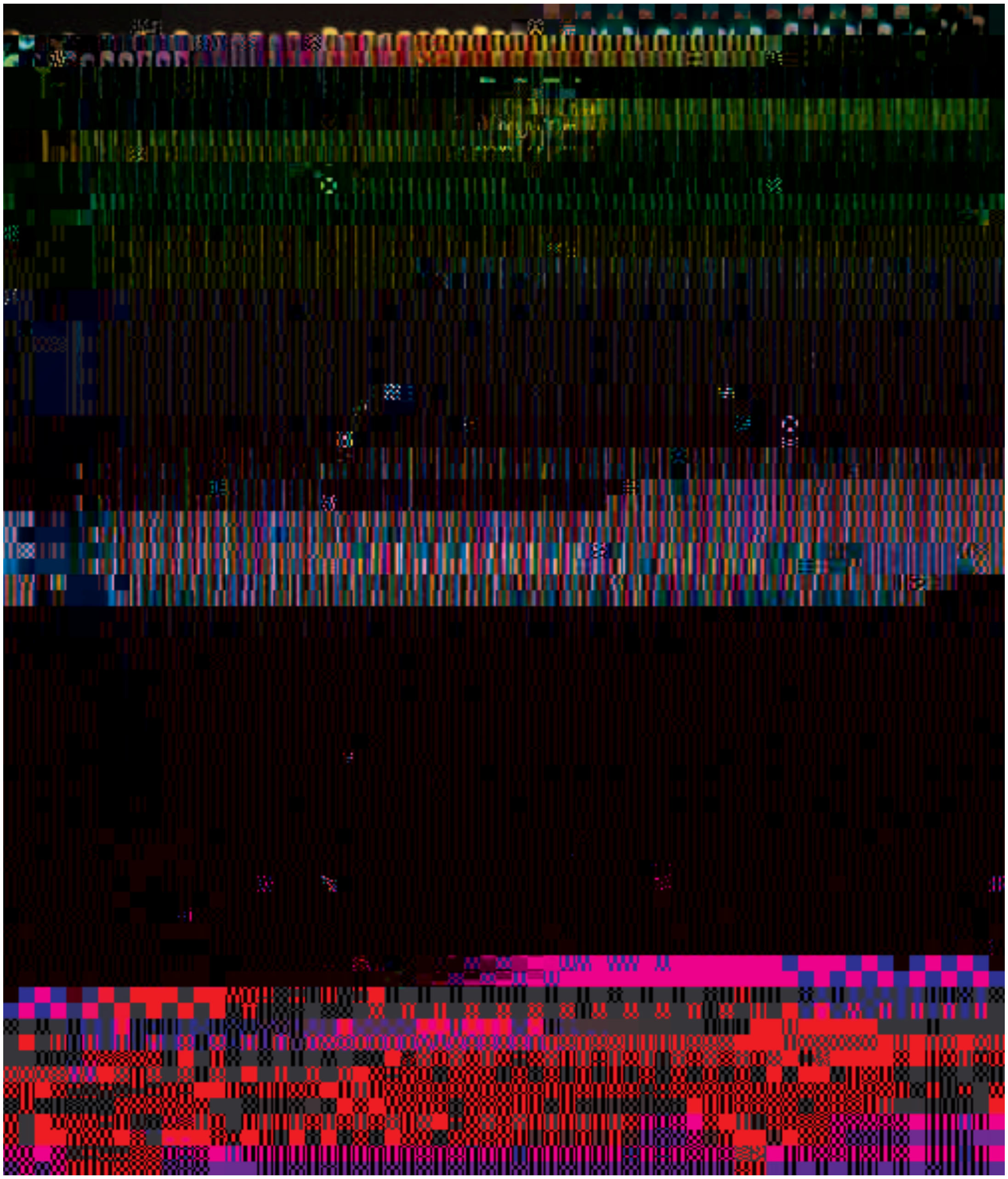
]bQ 0CG?>BP +Q 0QP 36GB:HGCJP +Q 3QP 0>B<I@6F -DH>8GQ #30*3(44 ,/ 015,&4 ?=P ^]eU^cb W^\\]X

]cQ 6G>GH>MP 'Q 3QP 6N=:BCJP 3Q 5IQP 0CG?>BP +Q 0Q 6B9 36GB:HGCJP +Q 3Q -DH>8G C; @><=H 7:6AG K>H= G8F:K 9>G@C86H>CBGQ "15,&4 0. .Q <;>P `^^U`^d W]ee_XQ

]dQ 6IA6BBP 0Q+QP)6@7P "Q+QP +68+>@@6BP *Q&QP 6B9 %6@J:NP #Q(Q .FCD6<6H>CB 9MB6A>8G C; CDH>86@ JCFH>8:G 9I: HC %CIM D=6G:Q "159 713(44 <AP ed]d V ed^c W^\\eXQ

Material/property	K_{11}, pN	K_{22}, pN	K_{33}, pN		n	$\boldsymbol{H_{HTP}}$ of CB-15, μm^{-1}
E7	11.1	7.3	17.1	13.8	0.22	7.3
ZLI-3412	14.1	6.7	15.5	3.4	0.078	6.3





χ , POM image of the grating

!400,' - ' .3#17 +)9 !@9 G8=:A6H>8 C; H=: 0HC?:G DC@6F>A:HFM G:HID IG:9 HC A6D H=: D=6G: C; 9>;;F68H:9 @6G:F 7:6AG(&:,: @6G:F 7:6A >G GD@>H >BHC HKC D6H=G 6B9 H=:B F:8CA7>B:9 7M 7:6A GD@>HH:FG 0] 6B9 0^P F:GD:8H>J:@MQ 1=: @C8? 6B9 'F>G 6F: IG:9 HC G:@:8H H=: 9>;;F68H>CB CF9:F C; >BH:F:GHQ +>FFCFG +]U+_ 6F: G>@J:F A>FFCFG IG:9 HC F:9>F:8H H=: 7:6AQ 1=: 8C@@>A6H>CB C; 7:6AG >G D:F;CFA:9 7M @:BG D6>FG *]U*^ 6B9 *_U*`Q .C@6F>N:FG .]U._ 6B9 H=: 0T` D@6H: 8CBHFC@ DC@6F>N6H>CB GH6H:GQ

"6+23+.) 2%1'6 &+2,/%#3+/.2 +. 3*' 0*#2' /(# &+((1#%3'& $'#-+.&4%'& $7 &'('%32 +. # (+.)'1 0*#2')1#3+.)9 #P .-+ >A6<: C; ;>B<:F D=6G: <F6H>B< K>H= :@:A:BH6FM 9>G@C86H>CBG G:D6F6H:9 7M G:J:F6@ ;>B<:F @6M:FGQ $8 #LD:F>A:BH6@ 9:D:B9:B8: C; H=: 6B<@: 7:HK::B G>B<I@6F>H>:G >B CB: C; H=: 9>;;F68H:9 7:6AG JGQ 9>GH6B8: '& W9>6ACB9GX 6B9 H=: 7:GH ;>H 7M H=: H=:CF:H>86@ DF:9>8H>CB WGC@>9 @>B:XQ 1=: >BG:H G=CKG H=: 8CFF:GDCB9>B< D=6G: DFC;>@: C; H=: ;>FGHUCF9:F 9>;;F68H:9 7:6AQ %P G8=:A6H>8 G=CK>B< HKC HK>GH>B< G8F:K 9>G@C86H>CBG >B H=: D=6G: C; H=: ;>FGHUCF9:F 9>;;F68H>CB 7:6AQ 1=: J:FH>86@ <F::B @>B:G A6F? H=: D=6G: G>B<I@6F>H>:GQ