Three-dimensional imaging of liquid crystal structures and defects by means of holographic manipulation of colloidal nanowires with faceted sidewalls†

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Received 1st February 2011, Accepted 27th April 2011

to evaporate the isopropanol at ~40 °C on a hotplate. The final mixture consisting of dye-doped CLC and rods was stirred just before the CLC cell preparation to improve the quality of dispersion. FCPM and polarizing microscopy textures show that the GaN nanowire exerts tangential surface boundary conditions on the liquid crystal director, aligns parallel to it, and minor additional local director distortions appear only at the rod's ends (Fig. 1b and e) while the studied structures and defects remain unperturbed.

To promote appearance of dislocations in cholesteric lamellae, wedge CLC cells with a small dihedral angle $\alpha < 2^\circ$ and planar cholesteric structure were fabricated by use of 0.15 mm thick glass plates. The substrates were spin-coated with ~1 wt% solution of poly-vinyl-alcohol (PVA) in deionized water at 5000 rpm followed by baking in an oven for 1 h at 100 °C. Two types of cells were used: for some cells the PVA-coated substrates were assembled with no further treatment, while for the most cells the PVA-coated substrates were unidirectionally rubbed with a velvet cloth to force the liquid crystal molecules to align parallel to the rubbing direction. Wedge-shaped cells were prepared by using 80 μm spacers on one edge of the cell and no spacers on the opposite edge. The cells were then filled with a liquid crystal and sealed with epoxy. The confinement of the CLC into the wedge-shaped cells promoted facile appearance of defects such as dislocations and disclinations, which we have imaged by means of FCPM and classified according to the scheme of Kleman and Friedel.[16] In the so-called λ-disclinations, the material **n**-director field is non-singular because of **n** being parallel to the defect line in its core, so that the singularity is observed only in the immaterial director fields of CLC helical axis **c** and τ orthogonal to both **n** and **c**. In the ι-disclinations, the τ-field is non-singular but **n** and **c** director fields are singular. In χ-disclinations, the singularities are found in **n** and τ director fields but not in the **c**-field.

2.2. Integrated optical setup for imaging and manipulation

To manipulate nanowires, we have utilized holographic optical tweezers (HOT).[15,25] In the HOT setup (Fig. 2), a collimated 5 mm beam from a linearly polarized Ytterbium-doped fiber laser (IPG Photonics, $\lambda = 1064$ nm) is resized by a telescope to slightly overfill the active area of the phase-only spatial light modulator (Boulder Nonlinear Systems). We have used laser trap powers of 35–250 mW, smaller than the CLC realignment threshold. After being reflected off the SLM, the beam is coupled into the back aperture of a 60× oil-immersion microscope objective (NA = 1.42, ~60% transmission at 1064 nm) by using a second telescope. The second telescope (in the so-called 4-f arrangement) also images the phase profile encoded by the SLM to the back focal plane of the microscope objective. The holograms displayed on the SLM create trap patterns in the focal plane of the microscope objective. A dichroic mirror (Chroma) reflects the trapping laser beam into the microscope objective while transmits visible light used for

rod-shaped or needle-shaped nanoparticles aligning with the director to enhance the response to external magnetic fields. Similarly, elasticity-mediated alignment of nanorods in CLCs with optically tunable cholesteric pitch allowed for achieving rotation of colloidal rods by means of optical illumination.

When dispersed in CLCs, the nanowires orient along **n**(**r**) of the equilibrium helicoidal structure (Fig. 1f). As the cholesteric pitch decreases from infinity (nematic) to the used relatively short pitch values of 5 μm with the addition of the chiral dopant (Table 1), the angular distribution of nanowire orientations broadens (Fig. 3), although the width of the distribution remains a fraction of degree even for the CLC with $p = 5$ μm. The reason for the distribution broadening is likely that the transverse size of the nanowire corresponds to a certain angular twist of the equilibrium helicoidal structure of the CLC (360 /p, ~10 degrees for $p = 5$ μm). This results in a weaker elastic suppression of the angular thermal fluctuations of the nanowire in the CLC matrix as compared to the case of the nematic, although the colloidal nanowire still aligns parallel to the local **n**(**r**) corresponding to the center of the nanowire.

We control and probe 3D nanowire orientations and positions in the CLC by use of a combination of holographic optical tweezers working at $\lambda = 1064$ nm, optical transmission microscopy, and a stepper motor controlling the sample's vertical position relative to the focal plane of an objective with 10 nm precision. For optical manipulation, one or two laser traps are positioned at the ends of a nanowire and then used to rotate and move it in 3D. In CLCs, nanowire orientation and position along the helical axis are coupled to each other: translation across the "cholesteric layers" is possible only *v a* rotation following the helicoidal **n**(**r**) (Fig. 1f and Video S1†). The handedness of the helix and the rotation direction determine whether the nanowire moves upward or downward; its translation away from the microscope objective is typically easier than toward it, due to the scattering forces originating from the large nanowire-CLC refractive index mismatch. The measured vertical position z_{nw} of a nanowire along the helix is a linear function of its in-plane orientation angle ϕ_{nw} (Fig. 1f), consistent with the **n**(**r**) of the equilibrium helicoidal structure **n**(z) = {cos (2πz/p), sin (2πz/p), 0}. From the linear fit of the experimental data shown in Fig. 1f by $z_{nw} = \phi_{nw}\, p/(2\pi)$, the measured effective pitch is $p = 5.3 \pm 0.3$ μm, matching that obtained from the FCPM cross-section in the same location (top left inset of Fig. 1f).

In order to apply our technique to imaging of complex 3D director structures and defects, we have constructed wedge cholesteric cells with a dihedral angle about 2 and strong surface anchoring that keeps near-surface cholesteric layers parallel to substrates. Elementary edge dislocations perpendicular to the direction of the thickness gradient introduce additional cholesteric lamellae, in accord with increasing thickness of the wedge (Fig. 4). These dislocations have their cores split into disclination pairs and are often accompanied by other cholesteric defects, forming complex 3D configurations of **n**(**r**).

Although the non-destructive 3D imaging of **n**(**r**) around defects can be achieved by means of holographically manipulated nanowires alone, we use FCPM imaging for the comparative analysis of nanowire positions and orientations relative to **n**(**r**). In the vertical FCPM cross-sections, the nanowires oriented orthogonally to the confocal image give rise to a dark triangular-wedge spreading upward from the position of the rod (Fig. 4a–f), which is due to the scattering of the FCPM excitation light by the high-index GaN nanowire ($n_{GaN} = 2.4$, much larger than the effective CLC indices within 1.5–1.7). Optical translation of a nanowire serves as a single-particle probe of **n**(**r**), allowing one not only to measure the equilibrium pitch but also to map changes in its effective value due to defects, yielding results that are in agreement with FCPM imaging (Fig. 4). The confocal vertical cross-sections in Fig. 4 show sequential translation of the nanowire along a Burgers circuit with the translation across the cholesteric layers implemented by rotating the nanowire. The attempted optical translation of a nanowire across the dislocation (discontinuity) in a layered CLC structure results in stretching of the dislocation resisted by its line tension (for example, when moving the nanowire from point #4 in Fig. 4c to the right we observe that as the dislocation eventually moves to the right and stretches when the nanowire approaches the defect core). A combination of rotational and translational motion

upward or downward in accord with the displacement of the layers to preserve its orientation parallel to the local **n(r)**.

nonsingular disclinations in its core split into the $\lambda^{+1/2}$, $^{-1/2}$ pair. When the nanowire is placed on the λ-disclination side of

Supplementary Information

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