Directing light with liquid crystal metasurfaces

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Abstract. Liquid crystal metasurfaces self-assemble on polymer substrates regularly patterned by focused ion beam. Periodic deformations of nematic liquid crystal produce distinct colouring and strong diffraction. Applying AC voltage of 4 V across the liquid crystal layer straightens the nematic, suppresses diffraction and increases direct transmission within sub-millisecond switching times. Multi-parametric optimization of the metasurface design substantially enhances the efficiency of diffraction into a single particular channel.

1. Introduction
Optical functionalities of materials based on regular submicro- and nanoscale structures are being intensively studied and various types of planar arrays – metasurfaces – have been proposed and realized in recent years [1]. Being extremely thin, metasurfaces efficiently control light and operate as spectral and polarization filters, wave plates, diffractive elements, transmitting and reflecting lenses. The potential impact of the metasurface concept exceeds by far simple replacement of traditional bulk optical elements with ultrathin analogues. Compact planar design allows creating hybrid systems with metal and semiconductor nanostructures combined with soft matter. Introducing layers of photoactive molecules, polymers and liquid crystals (LCs) one obtains optical elements tunable by low voltage [2], light [3] and temperature [4].

LC-metasurfaces represent a valuable soft matter extension of the optical metasurface concept, as they self-assemble upon specific patterned polyimide (PI) substrates. Periodic LC refractive index modulation on the scale of visible light wavelength gives rise to complex diffraction spectra and remarkably distinct colouring [5]. The important advantage of LC-metasurfaces is the electro-optical tunability [6]. As we show, their optical performance can be reproduced in terms of a relatively simple analytical model which we apply then to optimize the design in order to maximize the efficiency of diffraction into a particular channel.

2. Results
2.1. Fabrication
Display quality glass substrates with ~ 150 nm thin transparent ITO electrodes are covered with about 10 nm thin PI layer by spin-coating followed by annealing at 190°C for 1 hour. A uniform planar LC aligning action of PI is achieved by mechanical rubbing with cotton cloth. As previously [5], FEI Scios dual-beam electron microscope is used for focused ion beam (FIB) patterning with a 0.1 nA beam of Ga⁺ ions accelerated by a voltage of 30 kV. FIB controlled by
digital templates imprints periodic stripe patterns with a 0.5 duty factor and a period from 1 to 8 μm on areas of 0.5 × 0.5 mm² each. Typical fragments are shown in Fig. 1. The LC cells are assembled by stacking the patterned substrate with a chromolane coated one inducing vertical alignment of adjacent LC. Several micrometer thick gaps between the substrates are fixed by plastic spacers and monitored optically. Finally, the cells are filled with Merck E7 nematic LC.

2.2. Optical properties

Optical images of the LC cells are observed with Olympus CX31PF-5 polarized light microscope for the light normally incident on the LC cell and linearly polarized in parallel to the stripes patterned along the PI rubbing direction. Note that the polarization conversion is excluded by the symmetry. One can see in Fig. 2 that the patterned areas exhibit distinct colouring depending on the period. As discussed in Ref. [5], this is explained in terms of a simple model assuming that the pristine rubbed PI induces planar LC alignment while the areas subjected to FIB align the adjacent LC orthogonally to the substrate. The LC in the bulk adjusts according to such inhomogeneous boundary conditions and forms refractive index modulations of the scale of the structure periodicity. Light passing through different areas acquires different phase delay and, at a certain wavelength, a destructive interference causes a transmission dip. For larger periods, the surface induced LC modulation occurs in larger volumes and results in a more pronounced transmission dip at larger wavelengths. Patterns of shorter periods affect only extremely thin LC layers and their effect on the visible light transmission is diminished.

Measurements of light diffraction in transmission geometry on a set up combining Horiba Jobin-Yvon UVISEL 2 ellipsometer and Ocean Optics USB4000 fibre spectrometer confirm that the transmission dip is closely related to diffraction: in all samples the lacking transmitted energy is redistributed into the first strong diffraction orders. Typical data are shown in Fig. 3(a).

Consider this diffraction problem neglecting certain peculiarities of the LC elastic anisotropy [5]. In the so-called one constant approximation [7], the polar angle θ of the LC director satisfies the Poisson equation ∆θ(x, z) = 0 and its solution taking P-periodic values θ₀(x) on one substrate (at z = 0) and π/2 on the other substrate (at z = d) reads as:
Figure 3. Spectra of diffraction efficiencies of 5 µm periodic metasurface in 3 µm thick LC cell: measured (a) and calculated (b) using Eq. (2) with the E7 LC optical dispersion taken from Ref. [8].

\[ \theta(x, z) = a_0 \left(1 - \frac{z}{d}\right) + \frac{\pi z}{2d} + \sum_{m=1}^{\infty} \frac{\sinh[2\pi m(d - z)/P]}{\sinh[2\pi md/P]} \left( a_m \cos \frac{2\pi m x}{P} + b_m \sin \frac{2\pi m x}{P} \right), \]  

where \( a_i \) and \( b_i \) are the Fourier coefficients of the periodic function \( \theta_0(x) \). The main factor determining the diffraction efficiencies \( D_m \) is the spatial modulation of the optical path \( OP \) acquired during light propagation through different areas:

\[ D_m = \sqrt{1 - \left( \frac{\lambda}{mP} \right)^2 \int_0^P dx \ e^{2\pi i \left[ \frac{mx + OP(x)}{P} \right]}}, \]  

where \( n_e \) and \( \Delta n \) are the extraordinary refractive index and the birefringence of the nematic LC correspondingly. To test the model, we calculate the diffraction by a periodic stripe metasurface and obtain good reproduction of the actual experimental features as illustrated by Fig. 3.

2.3. Electro-optics

To study the electro-optical switching of the metasurfaces, we apply 1 kHz rectangular AC voltage across the LC cells. As seen in Fig. 2 on the right, even a voltage of 4 V amplitude ensures complete metasurface bleaching independently of its period. The corresponding transformation of the transmission spectra in Fig. 4(a) shows how “erasing” the periodic LC alignment by the voltage eliminates the transmission dip.

To reveal the switching dynamics we employ an optical set up with a blue 445 nm laser and silicon photo-diode that allows fast resolution of the transmission changes. Typical time-resolved electro-optical responses shown in Fig. 4(b) allow estimating the characteristic switching times. One can see that the switching-on is efficiently controlled by a voltage of a few volts and occurs during 10 milliseconds. It can be considerably accelerated to submillisecond times by applying the voltage up to 10 volts. The switching-off (relaxation) is practically independent of the voltage and its characteristic time is of the order of 10 milliseconds.
2.4. Design optimization

Fast calculation of the optical characteristics using Eq. (2) enables multi-parametric numerical optimization of the LC-metasurface controlled by the patterns imprinted in PI by FIB. Two representative patterns shown in Fig. 5(a) and 5(b) are optimized by MATLAB fminsearch routine to maximize the +1 order diffraction at 500 nm wavelength with the periodicity and thickness kept fixed. The obtained $\theta_0(x)$ profiles induce planar LC alignment on the left side and vertical alignment on the right side. The finer patterning in the middle produces a gradual intermediate transition. For smaller periodicity, the maximum diffraction efficiency still does not exceed 0.5 [Fig. 5(c)] as the director field in the bulk is smoother than at the surface due to the LC elasticity. Increasing the period allows enhancing the efficiency up to 0.7 as illustrated by Fig. 5(d).

**Figure 5.** LC-metasurfaces optimized for the maximum +1 order diffraction at 500 nm wavelength: optimal profiles of the director polar angle at the patterned LC cell interface with periods of 5 $\mu$m (a) and 10 $\mu$m (b) and the spectra of the diffraction efficiencies of the 5 $\mu$m (c) and 10 $\mu$m (d) periodic LC-metasurfaces in a 4 $\mu$m thick LC cell evaluated with Eq. (2).

3. Conclusions

LC-metasurfaces are switched between the diffracting and transmitting states during millisecond characteristic times by applying a voltage of several volts. The optimized transmission and diffraction efficiencies can be on the 0.6–0.7 level. This paves the way for electro-optical blazed grating devices controlling the light propagation direction.

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