A Facile Stratification-Enabled Emergent Hyper-Reflectivity in Cholesteric Liquid Crystals

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Abstract. Cholesteric liquid crystals (CLCs) are chiral photonic materials with selective reflection in terms of wavelength and polarization. Helix engineering is often required in order to produce desired properties for CLC materials to be employed for beam steering, light diffraction, scattering, adaptive or broadband reflection. Here, we demonstrate a novel photopolymerization-enforced stratification (PES)-based strategy to realize helix engineering in a chiral CLC system with initially one handedness of molecular rotation throughout the layer. PES plays a crucial role to drive the chiral dopant bundle consisting of two chiral dopants of opposite handedness to spontaneously phase separate, and create a CLC bilayer structure that reflects left- and right-handed circularly polarized light (CPL). The initially hidden chiral information therefore becomes explicit, and hyper-reflectivity, i.e., reflecting both left- and right-handed CPL, successfully emerges from the designed CLC mixture. The PES mechanism can be applied to structure a wide range of liquid crystal (LC) and polymer materials. Moreover, the engineering strategy enables facile programming of the center wavelength of hyper-reflection, patterning, and incorporating stimuli-responsiveness in the optical device. Hence, the engineered hyper-reflective CLCs offers great promises for future applications, such as digital displays, lasing, optical storage, and smart windows.

Introduction

Cholesteric liquid crystal (CLC) type structures widely exist in nature, e.g. the condensed phase of DNA, plant cell walls, arthropod cuticles, human dense bone, and many chiral biomaterials [1,2]. The self-organized helical structure possesses unique optical properties [3, 4]. In a conventional CLC, the light is reflected when the wavelength within the medium matches the pitch of cholesteric helical structure [5, 6]. The reflectance is limited to 50% for normally incident unpolarized light due to polarization selectivity. The bandwidth, measured as the width of reflection peak at half height, is typically less than 100 nm due to limited birefringence of the material [5].

Inspired by the nature, scientists have been able to conceptualize versatile photonic architectures, to provide appealing aesthetics and distinctive optical functions. [7]. Tremendous efforts have been paid to engineer CLC materials, or more specifically, to control the helical organization of liquid crystal (LC) directors on the molecular scale. For instance, light-addressable or thermal-responsive chiral dopants allow tuning of the reflection wavelength [8]. Manipulation of the helical pitch direction or distribution provides an accessible route to realize beam steering, light diffraction, scattering or broadband reflection [4, 9]. Coupling helical structures of the opposite handedness can achieve hyper-reflectivity, defined as the ability to reflect both left- and right-handed circularly polarized light (L-CPL and R-CPL) [10]. In crosslinked CLC polymers, the distortion of helical structure generates polarization-dependent pseudo-Bragg reflectors and hyper-reflectivity [11].

In terms of helix engineering, a variety of strategies and tools have been developed to produce the different delicate structures [12,13]. Among all options, photopolymerization-enforced stratification (PES) holds an important position, as the combination of polymer and CLC materials is versatile in LC-based functional devices [13]. PES refers to the formation of layered structures upon the application of photopolymerization in LC/polymer materials. In general, a spatially variant light intensity, or energy absorption of the incident light, or a field induced spatial pattern is required to realize PES. In the context of CLCs, PES has been previously employed to produce flexible temperature-responsive reflectors [8]. However, a UV absorber is required to

realize the vertical light intensity gradient and subsequent stratification. In addition, a non-mesogenic monomer is usually used to form the stratified polymer top layer, which serves as a protecting layer.

In this work, we describe a novel strategy to realize PES via a sequential polymerization and autonomous phase separation mechanism. Enabled by such strategy, hyper-reflectivity, instead of typical cholesteric reflection, successfully emerge from a single-layered chiral CLC system. Specifically, we use two chiral dopants of opposite handedness, with one being polymerizable and the other not. The mixture remains cholesteric with reflection towards CPL of a single handedness, as determined by the majority-rules principle [14]. PES is the key to break down the individual chiral contributions of the opposite chiral additives [13]. Upon photopolymerization, the system undergoes a three-stage phase separation process to form the stratified structure at the end, with two chiral dopants spontaneously phase separating into different layers, resulting into reflections towards CPL of the opposite handedness. This emergent approach to engineer hyper-reflective CLCs not only offers a new route to realize PES, but also enables excellent optical performance, stimuli-responsiveness, as well as the flexibility to adjust the center wavelength of hyper-reflection.

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