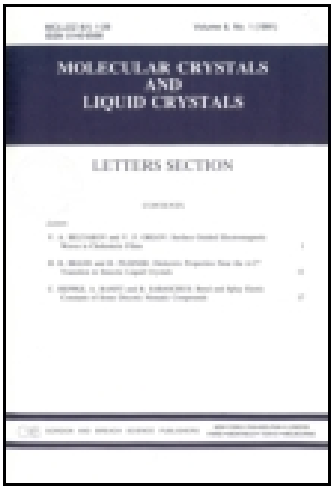

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Topological Soft Matter for Optics and Photonics

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Topological Soft Matter for Optics and Photonics

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Liquid crystal colloids are interesting for a variety of mechanisms—including self-assembly, optical-tweezers assisted assembly, topology, and material flow—that can be used to create various complex optical and photonic structures. Here, we present a brief overview of liquid crystal colloidal structures, as recently achieved by numerical modeling and experiments. Central to the structures are complex conformations of topological defects, as they can bind, stabilize, or distort the structure. Using topological and geometrical arguments, we show that the defects can be controllably rewired and imprinted, for example by using optical tweezers. We show that 3D colloidal crystals can be assembled from elastic dipoles of spherical beads in nematic liquid crystals or via inherently inhomogeneous order profiles in bulk and confined cholesteric blue phases. Colloidal crystals are generalized to close-packed colloidal lattices, which we show can serve as natural templates for defect networks. Finally, photonic bands are calculated for selected structures and possible defects in the structure are discussed.

Keywords Colloidal crystals; nematic liquid crystals; blue phases; photonic crystal; self-assembly

1. Introduction

Liquid crystals are excellent examples of soft matter where large susceptibility to external stimuli together with the optical anisotropy was used to develop display applications. Recently, the orientational ordering of liquid crystals constrained by nontrivial geometries revealed complex topological structures—topological soft matter—which have attracted a lot of attention [1–3]. This field covers systems from chirality stabilized nematic defect structures [4], colloidal nematic dispersions [5, 6], to porous materials permeated by liquid crystals [7]. The ability to form or even self-assemble complex optically birefringent super structures that are highly responsive to external stimuli has a potential for new optical and photonic applications, such as electrically tunable topological shaping of light [8], waveguides [9], or as a longer-term-goal as optical transistors [10].

A promising approach to novel liquid crystalline structures is by self-assembly of colloidal particles in liquid crystalline solvents, as achieved by the generated strong inter-particle potentials of multiple symmetries. Numerous structures can be assembled in

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nematic liquid crystals already with simple spherical particles where interactions are mediated either by localized nematic defects or by nematic braids where entangling of particles by disclinations stabilizes the structures [3, 11]. A special situation occurs when defects are used as traps for small colloidal particles [12]. Defects can be stabilized either by the confining geometry [13] or by the complex chiral ordering like in blue phases [14]. In such phases strongly chiral nematic fluid exhibits periodic 3D modulation of the average molecular orientation [15].

The generated liquid crystalline structures can be used to mold the flow-of-light at various levels. For example, nematic droplets can be used as highly tuneable optical resonators [16] or as 3D omnidirectional lasers [17], nematic defect lines can manipulate the orbital angular momentum of light at the micron scale in nematic films [18], and plasmonic complex fluids of nematic-like and helicoidal self-assemblies of gold nanorods could be used as plasmonic polarizers [19].

In this paper, we give a brief overview of the main topological and geometrical aspects of colloidal structures appearing in chiral and achiral orientational fields of the liquid crystal, with particular focus on three-dimensional colloidal structures including 3D nematic and 3D blue phase colloidal crystals. Modelling of the flow-of-light includes FDTD calculations of the transmittance of 1D photonic crystals and photonic band calculations in blue phase I and II.

2. Methods and Theory

Material and optic-photonic properties of the nematic structures are approached by extensive numerical modeling based on the minimization of the Landau-de Gennes free energy [20]. The simulation method discretizes the simulation volume on a regular (cubic) grid, and then calculates the spatial profile of the nematic order parameter tensor Q_{ij} as conditioned by the particles, geometry, surfaces, or possible external fields, like optical tweezers (for more, please see [21]). Such approach turns out to be very well able to predict or explain various structures in nematic liquid crystal colloids at qualitative or even quantitative level when compared to experiments, including systems like nematic shells [22] or nematic colloidal platelets [23].

Optical and photonic properties of the nematic structures are calculated in parallel by two approaches: (i) Finite Difference Time Domain (FDTD) simulations of the light field propagation and (ii) photonic band calculations (Fig. 1). The approaches are complementary and are used depending on the considered problem. In the finite-difference time-domain method, the electric and magnetic fields are spatially discretized onto a lattice and then time-evolved according to Maxwell's equations [24, 25]. In our studied liquid crystal samples, we assume no free charges and no currents and also neglect magnetic anisotropy of the liquid crystal. In photonic band calculations, we solve the eigen-problem for the electic field eigenstates in a general 3D periodic unit cell of a nematic [26]. Effectively, we perform a Fourier expansion of the tensor order parameter field, map it to the dielectric permittivity tensor, and then use the Bloch theorem to formulate the eigenproblem, which finally we solve numerically.

3. Control of Topological Defects

Colloidal particles dispersed in liquid crystalline hosts are naturally accompanied by the topological defects, which compensate for the distortion field typically imposed by the particle surfaces. To show the location and structure of the defects, which are typically

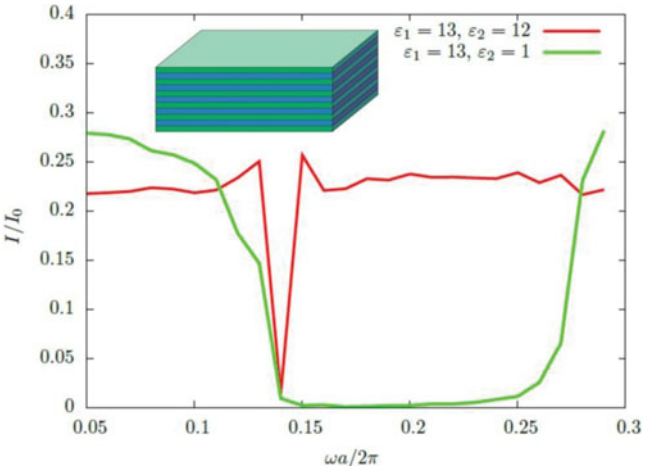

Figure 1. Example of a calculation performed by the FDTD method—the intensity trasmittance I/I_0 through a 1D photonic crystal. Note the formation of band gaps of different widths, depending on the dielectric contrast of the two stacked materials with dielectric permittivity ε_1 and ε_2. a is width of two layers. These results are in full agreement with [27].

$-1/2$ disclinations for particles with homeotropic anchoring or surface boojums for particles with planar anchoring, we usually draw the isosurfaces of constant nematic degree of order. Further structure of the defects can be revealed if drawing other local characteristics, such as splay-bend or twist parameter (for more see [28]). Figs. 2A and 2B show a colloidal dimer of two particles with strong hometropic anchoring in a π–twisted nematic cell. Interestingly, the defect conformation that binds the particle-pair in Fig. 2A can be changed—rewired—to form new different defect conformations as indicated in Fig. 2B. In the π-cell there turn out to be four distinct rewiring sites (A, B, C, D), which each carry the symmetry of a tetrahedron [29, 30]. Changing the defect conformation then corresponds to a solid rotation of any combination of the tetrahedrons. One should note, that upon the rotation of the tetrahedrons, the nematic field and the exact location of the disclination lines can further relax to some extent and effectively distort the tetrahedrons, yet the general defect conformation remains unchanged.

A strong approach to controlling the defect conformation is by external fields, where in particular optical laser fields integrated into an optical tweezers setup prove to be a strong tool for manipulation [31]. Fig. 2C shows a colloidal particles with hometropic anchoring as an elastic dipole in a planar cell, where the originally -1 hedgehog defect in the form of a small ring (upper panel) is manipulated by the Gaussian laser beam to form a defect loop (middle panel), which can be expanded to larger lengths by further gradual repositioning of the laser beam (lower panel).

The concept of manipulating topological defects via local geometric rewiring rules, which are then further subjected to minimum energy conditions, can be naturally expanded also to two-dimensional and three-dimensional colloidal structures. Fig. 3 shows two-dimensional close-packed colloidal crystal in a $\pi/2$ twisted nematic cell where complex conformations of $-1/2$ disclination lines emerge as a result of confinement imposed on the nematic by particles and cell walls. Calculation in Fig. 3 is an example of a possible structure and was performed in a periodic rectangular unit cell with total of 2 particles. Using 2D arrays of particles naturally reveals distinct regions—such as overcrossing or undercrossings—of disclination line segments where they can be locally rewired. Note,

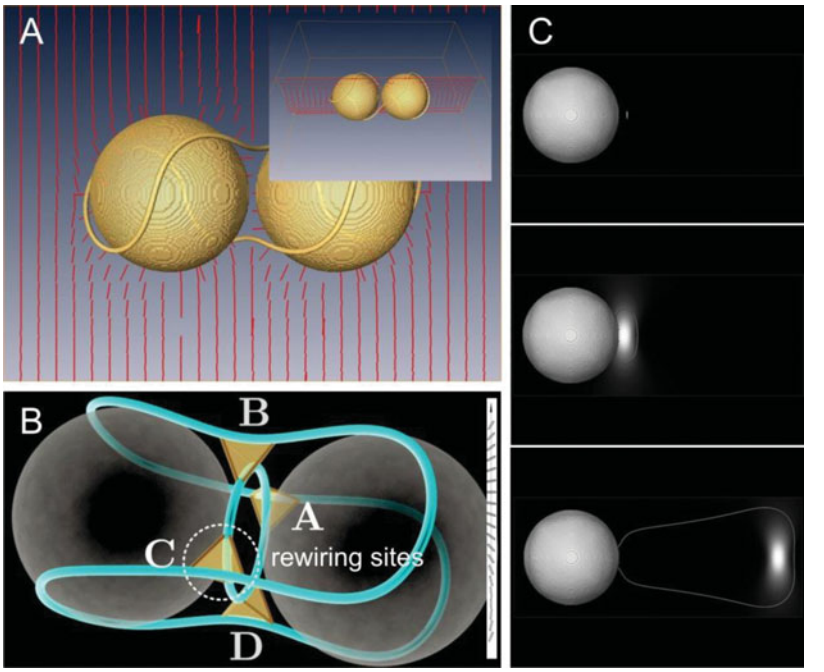

Figure 2. Manipulation of topological defects. (A) Colloidal dimer of 1μm sized particles in a π-twisted nematic cell. Inset shows the full simulation box. (B) Scheme of the four rewiring sites, where the general conformation of the disclination (or disclinations) can be manipulated. (C) Manipulation of a -1 point defect (in the form of a small ring) of an elastic dipole by optical tweezers in a planar nematic cell. Upper panel shows undistorted elastic dipole, middle panel shows the change of the point defect upon illumination close to the particle surface and lower panel shows the defect—now in the form of a long loop—upon gradual stretching with the tweezers.

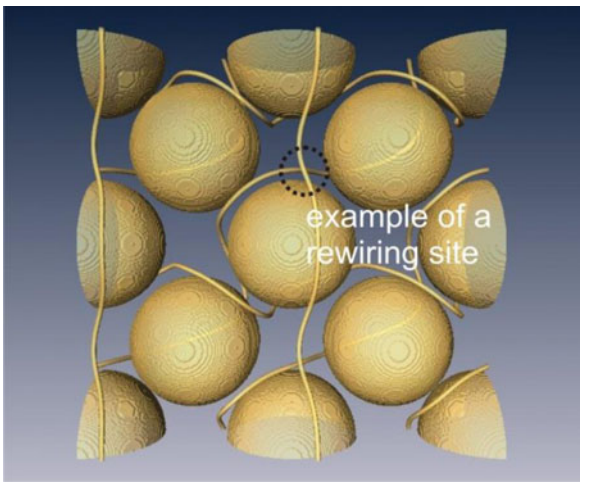

Figure 3. Two-dimensional close-packed colloidal crystal in a $\pi/2$ twisted nematic cell with distinct regions—the rewiring sites—where defect conformation can be controllably manipulated.

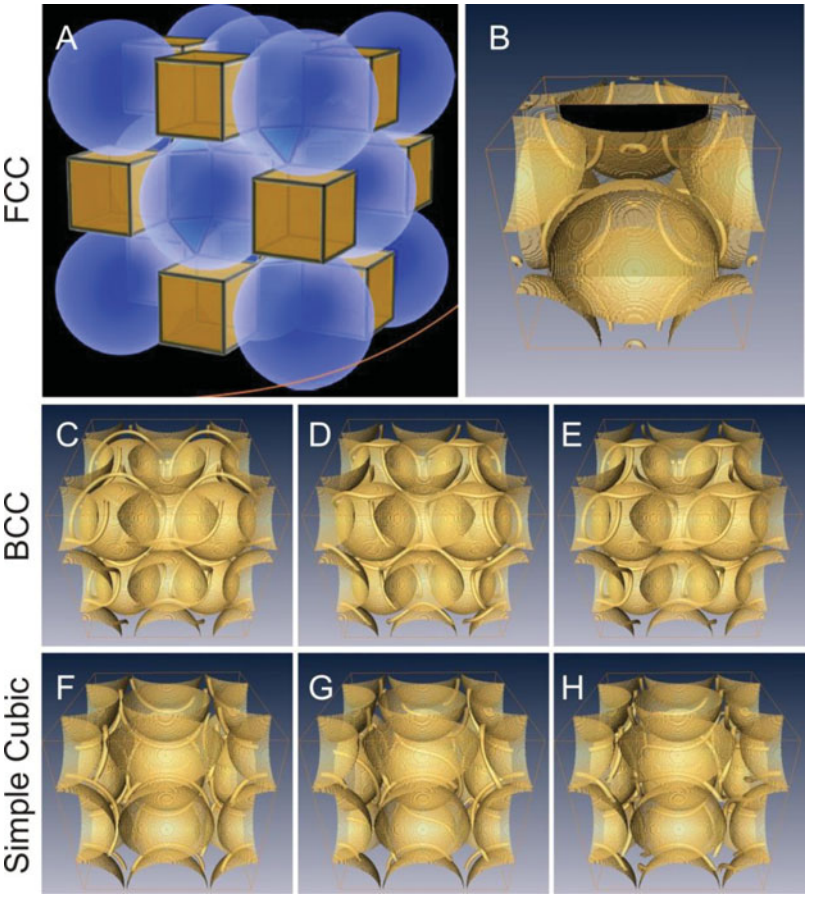

Figure 4. Defect networks in close packed colloidal opals with different packing symmetry: (A, B) face centered cubic, (C–E) body centered cubic and (F–H) simple cubic. Particles surfaces with homeotropic anchoring generate local voids which can be decomposed as rewiring sites in the shape of cubes or tetrahedrons as shown in (A).

however, that this local rewiring change can affect the global topology of the defect conformation providing means for directly controlling the topology of these soft matter materials. Indeed, knots and links can be created and manipulated in such 2D colloidal structures, as shown in [3].

4. Reconfigurable Defect Networks in Nematic Colloidal Opals

Nematic colloidal opals—close packed lattice of colloidal particles infused by nematic liquid crystal—are exciting candidates for soft matter topological photonic materials [32, 33]. We focus on a densely packed cubic lattice (face centered cubic, body-centered cubic or simple cubic) of spherical colloidal particles with homeotropic boundary conditions. The complex arrangement of conflicting orientations imposed by particle surfaces generates a network of defects, more specifically $-1/2$ disclination lines, which span through the colloidal material as shown in Fig. 4. The conformations of the network are very diverse

and show strong hysteresis, primarily dependent on the actual formation dynamics of the network. In Figs. 4B–4H different initial profiles of the nematic order parameter tensor were used to generate different network conformations in the considered colloidal opal lattices.

Defect networks observed in colloidal opals can be decomposed into elementary building blocks, taken as segments of the defect network which are subjected to distinct rewiring rules (see Fig. 4A). Two types of the rewiring sites are observed: the tetrahedron (same as shown for dimers and 2D nematic colloids in Figs 2 and 3) and a cube. In the tetrahedron two line segment span each between the two corners of the imaginary tetrahedron, whereas in the cube there are 4 line segments connecting the eight corners of the cube. Connecting corner-to-corner different cubes and tetrahedrons in a periodic pattern, as shown in Fig. 4A, topologically fully reconstructs the defect network. Indeed, different corner-to-corner connections of the imaginary effective rewiring sites—the cubes or tetrahedrons—reproduce different opal lattices shown in Fig. 4. Such approach allows for algorithmic characterization and enumeration of the network. Also, it can be very efficiently used in numerical modeling for construction of reliable initial conditions.

5. Three-Dimensional Colloidal Crystals for Photonics

Photonic crystals are optic elements that rely on a periodically repeating modulation of the dielectric permittivity. And regular colloidal structures, such as colloidal crystals, are today explored as an important route for creating such photonic materials via a self-assembly process. Here, we show two self-assembly approaches for building three-dimensional colloidal crystals based on: (i) cholesteric blue phase liquid crystal hosts and (ii) regular achiral nematics.

Blue phase are three-dimensional ordered structures found in highly chiral liquid crystals, and are composed of an array of $-1/2$ disclinations, interspersed by double twist cylinders [13], as shown in Fig. 5A (blue phase I – BP I) and Fig. 5B (blue phase II–BP II). Because of the highly inhomogeneous orientational profile, particularly in the cores of the disclinations, regions emerge within the blue phase unit cells that can effectively trap colloidal particles [34]. These trapping sites are periodic because of the inherent periodicity of the blue phases and 3D colloidal crystals can be stabilized [35], as shown in Fig. 5E. The exact locations of the trapping sites importantly depend on the size and shape of the partices as well as on the anchoring strength at the particle surfaces. Blue phases are themselves naturally forming photonic crystals, with the periodic modulation of the dielectric permittivity stemming from the periodic modulation of the optical axis [36]. In Fig. 5C and 5D, we show the calculated photonic bands of the blue phase I and blue phase II, respectively, as calculated with the photonic band approach. One observes distinct local band gaps, which depend strongly on the dielectric anisotropy of the chiral nematic, but no full band gaps.

Three-dimensional colloidal crystals can be realized also in regular nematics [37]. The structure was numerically predicted to consist effectively of antiparallel chains of nematic elastic dipoles and is shown in Fig. 5F. The calculated unit cell is tetragonal with basis and is in good agreement with experiments. The 3D nematic dipolar colloidal crystal responds interestingly to an external electric field, with the response crucially dependent on nematic having positive or negative dielectric anisotropy. In a positive dielectric anisotropy nematic, giant electrostriction of the colloidal crystalline is observed upon applying the

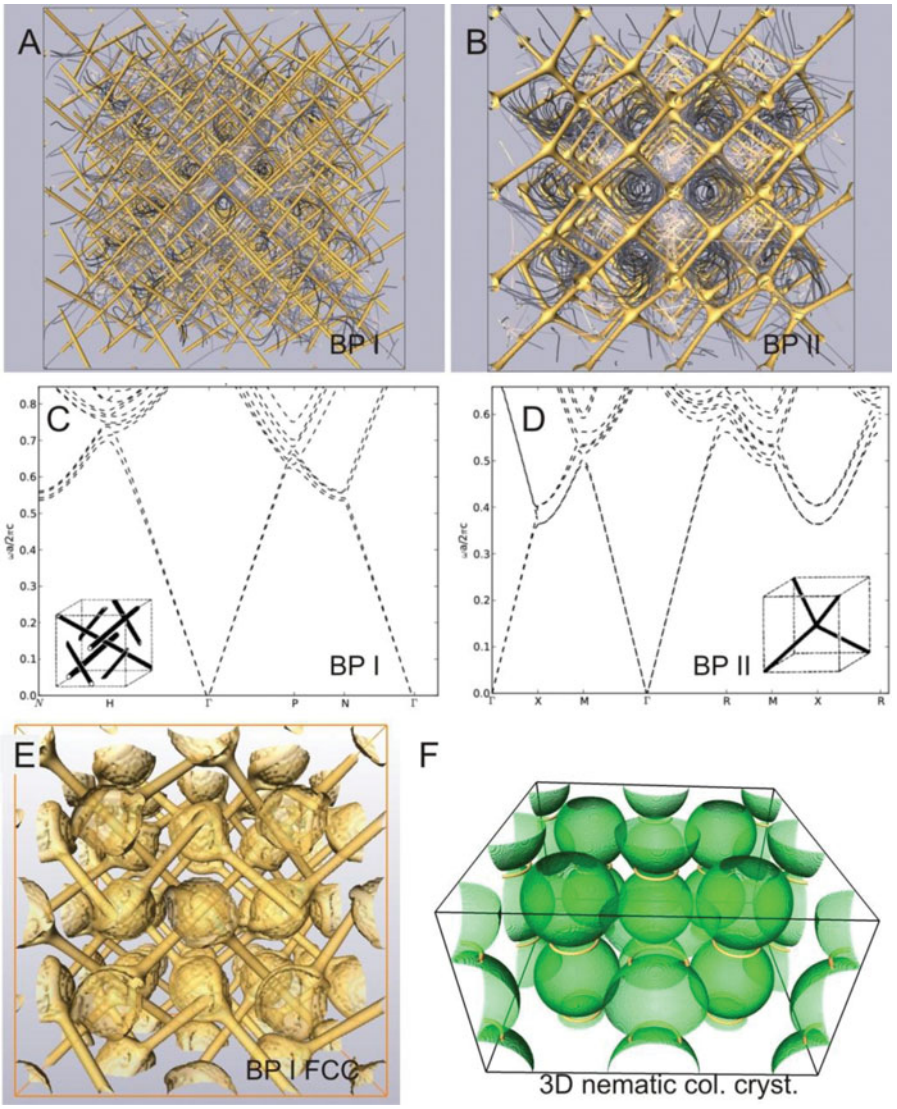

Figure 5. Three-dimensional photonic crystals from periodic nematic and chiral nematic. (A) Unit cells $4 \times 4 \times 4$ of blue phase I and (B) blue phase II. Disclination lines are drawn in yellow, whereas gray streamlines show the director with the distinct double twist cylinders. (C) Calculated photonic bands of blue phase I and (D) blue phase II. (E) Face centered cubic colloidal crystal formed in blue phase I. $2 \times 2 \times 2$ unit cells are shown. (F) Thee-dimensional dipolar colloidal crystal in (achiral) nematic.

external electric field with the changes in the lattice constants as large as few 10%. In negative dielectric anisotropy nematic, however, the colloidal crystallite rotates as whole with the observed angles of rotations up to 30° at fields of 0.1V/μm. This demonstrates a new class of electrically highly responsive soft materials, with directly electric-field tuneable photonic properties.

6. Conclusions

In conclusions, we show that nematic and chiral nematic colloids are materials, which are conditioned by the design and control of topological defects, acting as one of the modern platforms for topological soft matter, with direct implications for complex optics and photonics. Topology of the defect can be controlled via local rewiring sites, which by-symmetry behave as tetrahedrons or cubes of defect line segments dressed with the director field. Examples of such rewirings are shown for a particle dimer and 2D colloidal crystal in twisted nematic cells. Methods for calculation of photonic bands and light field propagation are discussed, giving examples of transmittance calculations of a 1D photonic crystal and photonic band structure of blue phase I and blue phase II. Specifically, three-dimensional colloidal structures are presented: (i) colloidal opals as percolated network of disclinations, (ii) 3D blue phase colloidal crystals, and (iii) 3D nematic dipolar colloidal crystals with remarkable giant electrostriction and electro-rotation. Finally, the demonstrated tuneability and controllability of the nematic colloidal structures is emerging as a potent route towards tuneable and by-optical-anisotropy conditioned complex optics and photonics.

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