

Abstract Book

OLC2021 Okinawa

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Tutorial Abstracts

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Indium tin oxide (ITO)-free optoelectronic devices have been discussed for a number of years in the light of a possible indium shortage as demand rises. In particular, this is due to the largely increased number of flat panel displays and especially liquid crystal displays (LCDs) being produced for home entertainment TV and mobile technologies. While a shortage of primary indium seems far on the horizon, nevertheless, recycling has become an important issue, as has the development of ITO-free electrode materials, especially for flexible liquid crystal devices. We will thus summarize the present situation of primary and secondary indium production, the main uses of indium as well as the prerequisites and possibilities for recycling[2]. The main contenders for new electrode technologies are then discussed, starting with non-carbon ITO-replacement technologies[3], such as metal nanowires, conductive polymers and ultra-thin metal films. In the discussion an emphasis will be placed on carbon-based materials for LCDs, including composite approaches. These will include films of single-wall and multi-wall carbon nanotubes[4,5], and graphene, graphene oxide (GO) and reduced graphene oxide (rGO)[6,7]. At present, some of these technologies already fulfil the technical specifications demanded from ITO with respect to transmittance and sheet resistance, albeit not in relation to cost and large-scale production. Promising approaches in terms of cost versus performance seem to lie in carbon-based composite materials, combining nanotubes and graphene with metal nanowires or conductive polymers.

Even once a performance equivalent replacement technology for ITO is found, it is unlikely that we will see ITO-free devices taking over the market soon afterwards. For this to happen, the new materials either need to be considerably better or clearly cheaper than ITO, because the latter is, and has been for many years, the established industry standard with all the necessary production infrastructure and R&D knowledge in place. Moreover, a deviation from this standard will be a slow process that can only be initiated via a considerable step-increase in performance or cost decrease, or for example via environmental legislation, or with the development of novel display technologies, which cannot be realised with conventional ITO electrode techniques. One of such technologies could be flexible devices on polymer substrates, which cannot be processed by ITO sputtering techniques. In this case, solution processing could pave the way for electrodes either of metal nanowire networks or of carbon-based materials, or indeed composite materials of both. In this case, it is likely that price will be the decisive factor, which could favour carbon-based technologies. Especially, future large-area applications with environmental impact, such as energy saving or energy-harvesting windows, could tip the balance towards carbon-based electrode materials. Yet, it does not seem likely that within the next few years, a drastic move towards ITO-free technologies will be observed.

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T2

Computer simulations and our understanding of liquid crystal behavior

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Notwithstanding the scientific and technologic importance of liquid crystals, the relation between a certain molecular structure and the possibility of forming liquid crystal phases is still far from being understood. The first generation of theories and computer simulations of liquid crystals have made drastic and often contrasting assumptions on the model representation of constituent mesogens and on the type of intermolecular interactions (e.g purely attractive in Maier-Saupe type and purely hard repulsions in Onsager models)[1, 2]. Computer simulations of liquid crystals, that started with simple lattice models, have upgraded over the years to off-lattice models where molecules are replaced by relatively simple shape objects endowed with attractive and repulsive interactions and, more recently, to very realistic fully atomistic models.[3]

In this tutorial talk we shall briefly summarize the main features of these categories of models showing some applications [4-6] and highlighting their strengths and limitations. The contribution of different interactions to the phase behaviour as well as open problems will be discussed.

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T3

Optical imaging of complex topological defect structures in nematics

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Frustrated nematics often described by term topological soft matter in their orientational order-parameter fields exhibit stable & metastable point defects, disclination lines, and solitons. In such systems often conflicting effects of confining geometries, surface anchoring, external fields, chirality, and elasticity are the source of frustrations. Basic concepts and approaches needed to model complex topological structures and predict their optical images will be summarized in the following topics:

- Constrained nematics: chirality, elasticity, confinement, geometry, anchoring, and external fields via phenomenological approach lead to complex structures.
- Topology of nematic defects: points, lines, loops, braids, and solitons.
- Optical imaging: polarization microscopy and multi photon fluorescent microscopy.
- Simulated POM approaches [1-6]: Jones approach, improved ray tracing including refraction & focusing, beam propagation, diffractive transfer matrix, plane wave expansion, and numerical solving of Maxwell Equations (FDTD).

The supports by the Slovenian Research Agency (P1-0099) and EU COST action EUTOPIA (CA17139) are acknowledged.

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T4

Collisions of topological defects

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Abstract: Since the discovery of liquid crystals, a great attention was paid to the identification of topological defects which very often play the role of fingerprints allowing to recognize different mesophases.

In this lecture, after an introduction to topological defects, we will report on a recent progress in studies of collisions of nematic monopoles [1] and disclinations. We will outline methods that allow to generate, set into motion and bring to collisions these point and linear defects.

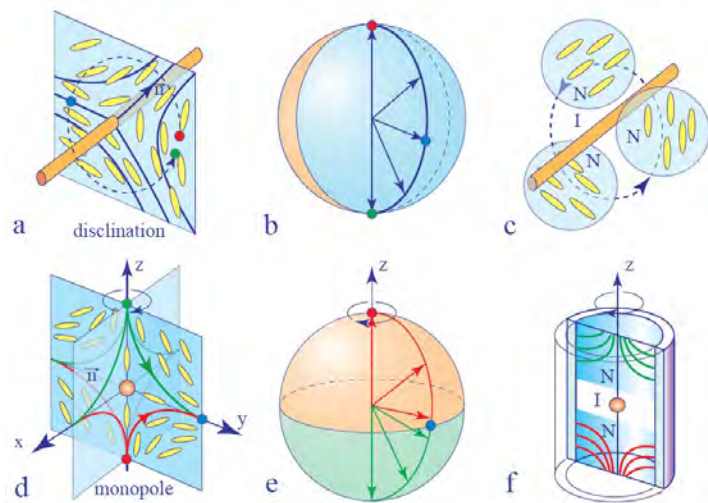


Fig. 1 Topology of disclinations and monopoles

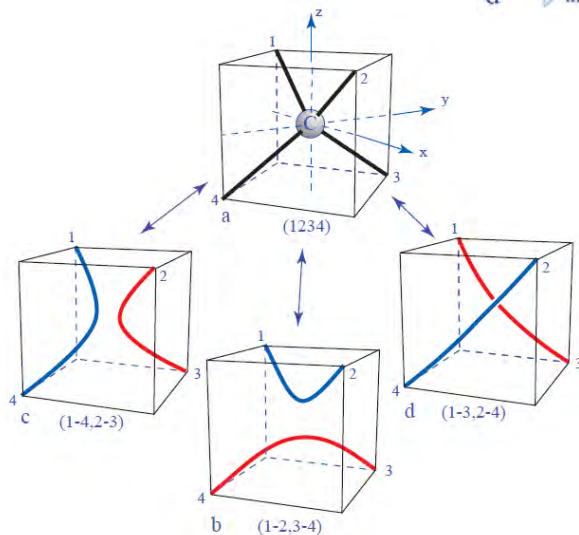


Fig. 2 Rewiring of colliding disclinations

In the case of monopoles, we will show how to control the outcome of collisions: annihilation or avoidance.

In the case of colliding disclinations, we will point out that the resulting rewiring can be controlled too.

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The Pancharatnam-Berry Phase (PBP) or the geometric phase of polarized optical waves are an optical embodiment of a universal behavior of transverse vectorial waves [1]. Unlike the ordinary phase changes associated with propagation of waves in space, the PBP develops instantaneously upon encountering anisotropic media according to the internal geometry of the medium. This property makes the PBP look rather mysterious at first glance.

Liquid crystals as responsive optically anisotropic media offer a unique ground, on which the PBP can be generated, manipulated and exploited for practical use. Since the PBP does not require a large optical path length to develop an arbitrarily large phase difference, there arises an engineering opportunity to replace conventional optics by “thin film optics” and further to open up an entirely new domain of optical technology employing a new form of optical wave such as vortex waves [2].

The photoalignment is the best suited surface-alignment method for liquid crystals to achieve the PBP liquid-crystal devices. It is just straightforward, in principle at least, to form microscopically resolved alignment patterns as required for PBP [3]. We have experienced a great deal of technical sophistication over the past three decades since its inception in the early 1990’s. Our toolbox is now well equipped to deal with difficult challenges to realize the potential of PBP.

In this tutorial talk, I try to offer a layman’s overview of PBP and the relevant photoalignment techniques with a view to lowering the hurdle of entry to this exciting field.

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T6

Liquid crystalline chiral photonic crystals for ultrafast (linear and nonlinear) photonics

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Abstract

Bulk/crystalline reorientation by an external field resulting in changes in the optical properties of the liquid crystals and trigger the desired operations such as polarization switching and control in ubiquitous devices. These processes are characterized by response times in the milliseconds to microseconds time scales and the fundamental optical physics underlying these processes are now quite well understood.

In this lecture, I will focus on the relatively less-explored electronic ultrafast [ps and fs] response of LC molecules to an optical field and unravel some myths about liquid crystals in general and revealed the uncanny abilities of fully developed cholesteric liquid crystals to modulate ultrafast laser pulses in particular. Owing to the unique combination of photonic crystal and liquid crystalline properties, and their extraordinary thicknesses with period number $\gg 1000$, the performance characteristics of CLC as a fully developed chiral photonic crystals in the ultrafast time scales rival most state-of-the-art materials for all optical self-compression/stretching and polarization switching of optical vector fields of common (linear, elliptical) or complex (azimuthal, radial) polarization states of picosecond and femtosecond laser pulses.

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Oral Abstracts

PL1

Title : Toward New Applications of Reactive Mesogens for Quantum-Dot Displays and Anticounterfeiting

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Reactive mesogens (RMs) have been widely used for producing anisotropic thermal, optical, and electrical properties resulting from the liquid crystalline order of monomer units through polymerization. The RMs enable to construct a rich variety of self-assembled mesoscopic structures and phases dispersed within amorphous matrices or on solid substrates so that they open new applications such as quantum-dot (QD) displays with a wide color gamut and an artificial iris with self-regulating capacity for a human eye. In realizing the QD displays, uniformly dispersed QDs in a host medium is one of the critical issues to acquire the homogeneous emission together with the low light leakage over the entire area of an individual pixel and to suppress the aggregation-induced quenching as well. Thus, it remains a challenge to achieve highly uniform dispersion of the QDs in well-defined patterns for light emission with the high intensity the high color purity.

In this presentation, we describe two representative types of new applications of the RMs, one of which is the uniform dispersion of the QDs in the host RM matrix with the molecular order and the other is the coloration technology for anticounterfeiting. Basically, these two examples share common features of the optical anisotropy inherent to the RMs in solidified films. Further applications of the RMs in biomimetics are also discussed.

PL2 **Laser speckle reduction solution with a liquid crystal diffuser**

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Laser-based displays suffer from speckle noise due to the random interference patterns of scattered coherent light from rough surfaces. Commonly utilized solution, such as moving diffusers, creates time-varying speckle patterns that were averaged on the observer's retina or the image sensor. This solution requires the use of motorized parts and can be bulky with the potential risk of mechanical failure. Hereby, I will present a liquid crystal diffuser that reduces speckle noise by over 90%. It is electrically driven, compact, and with no motorized parts. The randomized, time-varying domains with mismatched refractive indices of the liquid crystals produce varying speckle patterns. A near zero speckle contrast is achieved.

K1

Title : Guiding optical solitons by liquid crystal topology

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Different types of topological defects and solitons are found in practically all branches of physics, ranging from optics to fluid mechanics, cosmology, particle physics, and condensed matter. However, solitons of these very different types rarely co-exist and interact with each other and with other topological entities, like singular defects [1-3]. Here we develop a system that hosts co-existing optical solitons dubbed nematicons or similaritons [2,3] and topological solitonic localized structures or singular defects in the molecular alignment field, including Skyrmions, disclinations and twist walls [1]. We demonstrate a large diversity of interactions between such optical solitons and topological solitons and defects. These interactions are enriched by a host of nonlinear effects and the host medium's tendency to minimize the overall free energy due to orientational liquid-crystal elasticity when the perturbations of director-field alignment due to nematicons are in proximity to the director field configurations of the topological solitons and defects. The interplay of nonlinear effects that leads to the stabilization of these different solitons individually also causes highly nonlinear responses. Our findings show that such nonlinear interactions yield exceptionally rich behaviors that can be controlled by external stimuli, including light itself, and that may find practical applications ranging from beam steering to light localization.

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Limitations and Opportunities for the Fourth Generation Optics

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Diffraction waveplates (DWs), also referred to as geometrical phase optics, or Pancharatnam-Berry phase optics, or polarization gratings, are new, Fourth-Generation optics (4G optics), developed to overcome the size, weight, and efficiency limitations of conventional refractive or diffractive optics, including those based on metamaterials, and speed limitations of switchable optics technologies [1,2]. DWs, however, have unique features that may appear as limitations for applications involving unpolarized and spectrally broadband light. Examples of such applications are augmented reality glasses and adaptive ophthalmic lenses, along with high resolution thin and ultralight space telescopes, and even diffractive solar sails.

DWs offer effective means for overcoming those limitations due to absence of scatter inherent to metamaterial optics such as metalenses, and due to versatility in customization of their diffraction spectra. We will present different designs of DWs and their system that perform focusing, beam deflection, and more complex beam shaping optical functions in a thin-film format, and we will discuss large aperture high efficiency broadband imaging systems made possible by the use of DWs.

The maturation of the technology pioneered by BEAM Co. over 20 years ago now is at the stage of scaling-up production and cost reduction which is being obtained by automation of coating and optical recording processes. Improvement in fabrication technology are progressing to accommodate complexity of novel DW optics components requiring “random patterning” for switchable holograms and beam shapers, spectral customization and multifunctionality, and further enhancement of their capabilities with reflective geometrical phase optics [3].

Yes, there are issues to resolve and overcome in the development of millisecond-fast, micrometer-thin, 1V switchable and practically weightless large aperture optics, but there is no alternative in the horizon to DWs since “There is no Moore’s Law in Optics” [4].

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K3

Liquid crystal metamaterials for light guiding

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Liquid crystal metamaterials have a potential for a range of optic and photonic applications, as they can control the flow-of light by the spatially variable material birefringence, which –in principle- can include also negative refractive eigen-indices. In this work, we present the control of light by selected liquid crystal metamaterial structures using FDTD and FDFD based numerical modelling. Specifically, we show the role of topological defects in liquid crystal metamaterials which we show can perform as attractors and deflectors for light. Also we demonstrate the emergence of light edge states as determined by anisotropic metamaterial structures. More generally, this work is aimed at developing novel tuneable approaches for guiding and controlling the flow of light at the wavelength level.

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K4

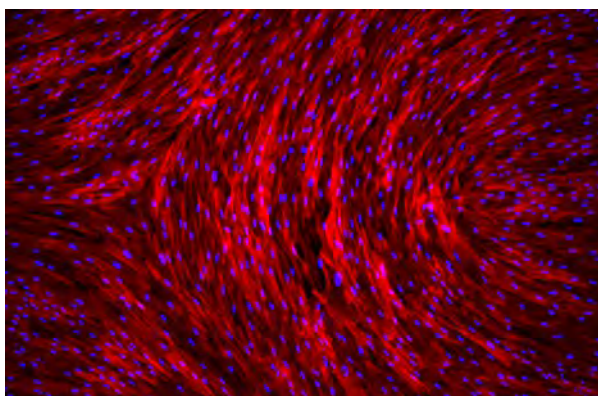
Photopatterned liquid crystal elastomers to control microparticles and living tissues

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Controlling spatial location of microparticles and living cells is one of the challenges of modern technology. Here we demonstrate that photoaligned liquid crystal elastomer (LCE) coatings [1,2] could be used as an effective approach to control dynamically the placement of microparticles [3] and to control cells in living tissues [4], such as the monolayers of human dermal fibroblasts shown in the figure. The director patterns inscribed into the LCE result in a deterministic response of the coating profile when activated by temperature, light, or humidity. The spatially-varying reversible profile is used to move the microparticles in the plane of the substrate. Anisotropic swelling of the LCEs in contact with the aqueous cell growth medium



produces surface profiles that are capable to align the living tissues [4]. The LCE aligns the tissues, causes a strong spatial variation of cell phenotype, their surface density, number density fluctuations. It also allows for a faster directional growth of tissues which is important for wound healing applications.

The work is supported by Office of Sciences, Department of Energy, grant DE-SC0019105.

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K5

Effect of the presence of the dye Sunset Yellow on the stabilization of the amphiphilic-lyotropic uniaxial and biaxial nematic phases

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In the talk we will discuss the interactions of the anionic azo dye Sunset Yellow (SSY) with dodecylalkylammonium bromide (DAABr) surfactants in lyotropic mixtures. In particular the effect of the addition of the dye in the lyotropic mixture have been investigated for the stabilization of the different lyotropic nematic phases. Sunset Yellow doped mixtures of DAABr surfactants, with different head-group sizes, were studied by polarizing optical microscopy, laser conoscopy, and small-angle x-ray scattering. Interactions between ionic species on the micelles' surfaces affect the stabilization of different lyotropic nematic phases by modifying the packing of the surfactant molecules in the micelles, the surface curvature of the micelles, and the micellar shape anisotropy. By modifying those interactions, the stabilization of different nematic phases becomes possible. Moreover, the temperature range of the biaxial nematic phase in the partial phase diagrams can be controlled. This can be done mainly in two ways, by considering the kosmotropic and the chaotropic properties of the surfactant head groups and of the counterions or ions of the electrolytes present in the lyotropic mixtures: (a) using a surfactant and choosing ions with different degree of the kosmotropic or chaotropic character with respect to it; or (b) using surfactants with different head-group sizes to change the degree of the kosmotropic or chaotropic character of the surfactant head group and known electrolyte ion. The latter way was followed in the present study. Thus, the interactions between the SSY, which has chaotropic character with two ionic groups at its periphery, and dodecylalkylammonium bromide surfactants with different head group character was investigated. It was found that the extent of the interaction of Sunset Yellow with DAABr surfactant head groups on micelle surfaces affects the stabilization of different nematic phases, by causing the change in the micellar structural parameters. The results also indicated that the micelle surface curvature should be different in the three nematic phases. The surfactant packing parameter and the area per surfactant head group should have certain values for the stabilization of the nematic phases, especially the biaxial one [1].

\$ TÜBİTAK, BAP, INCT-FCx, NAP-FCx, FAPESP, CNPq, CAPES.

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In contrast to the conventional approach entailing sandwiched liquid crystal (LC) cells in the LC–solid interface techniques for LC-based biosensing, here we alternatively demonstrate a simpler platform for label-free and quantitative detection of biomolecules (Fig. 1). This unique LC-on-a-single-substrate scheme enables efficient detection of proteins, such as the model protein bovine serum albumin (BSA) [1–4], as well as rapid screening and bioassay of DNA hybridization [1] and cancer markers such as the carcinoma antigen 125 (CA-125) [2,3]. Experimental results based on three different probe mesophase systems—namely, nematic [1,2], cholesteric [4], and nematic/polymer [3]—spin-coated on a biomolecule-immobilized glass substrate will be reported to manifest the feasibility for easier and time-efficient establishment of a biosensing platform. The working principle of both the cell platform and the LC-on-a-single-substrate scheme stems from the disrupted LC alignment and, in turn, reduced orientational order by the immobilized biomolecules; i.e., the analyte, in the interface. Comparatively addressed will be the pros and cons of both approaches for application in LC–solid biosensing.

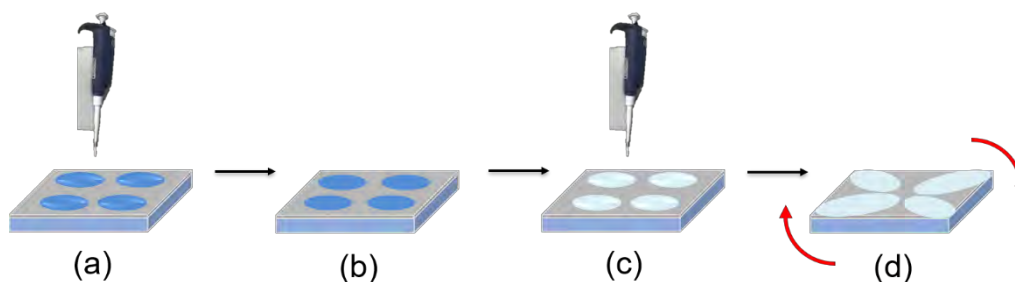


Fig. 1. Steps to fabricate an array of biomolecular samples on a single DMOAP-coated glass substrate and successively layer LC films atop the biomolecular samples with various concentrations.

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K7

Cellulose nanocrystals (CNCs) colloidal liquid crystals in water: slight changes, huge differences

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Cellulose nanocrystals (CNCs) are very well known to form aqueous colloidal lyotropic liquid crystalline phases in water [1]. The colloidal CNCs mesomorphic system is due to the presence of anisometric nano rods. The formation of colloidal liquid crystals in aqueous solutions was first reported for rod-like tobacco mosaic virus solutions [2] and in 1949, Onsager [3] used entropic arguments to explain the formation of a nematic phase from a disorder phase of rods. For CNCs, in addition to the nematic phase, a cholesteric structure was also reported [4]. The formation of tactoids from an isotropic phase was found to be at the genesis of a high-density cholesteric phase that deposits at the bottom of the vials. Here, we present the coexistence of a nematic low-density phase coexisting with an isotropic high-density phase, at the isotropic-nematic phase separation, in CNCs aqueous solutions. Furthermore, atactoids were observed in the upper birefringent phase that after freeze drying the sample translate into holes in solid cellulose nanosheets. The negative tactoids present needle-shapes with very low axial ratios compared with the axial ratio of about 50 presented by the CNCs. The appearance of the nematic phase inversion and the formation of the atactoids were interpreted taking into account the size and the charge polydispersity of the samples. Our work opens new horizons on the preparation of liquid crystalline phases from CNCs lyotropic systems with diverse optical functionalities.

Acknowledgements

This work was carried out under the Project inactus – innovative products and technologies from eucalyptus, Project N.º 21874 funded by Portugal 2020 through European Regional Development Fund (ERDF) in the frame of COMPETE 2020 nº246/AXIS II/2017 and by FEDER funds through the COMPETE 2020 Program, National Funds through FCT - Portuguese Foundation for Science and Technology and POR Lisboa2020, under the projects numbers POCI- 01-0145-FEDER-007688 (Reference UID/CTM/50025), PTDC/CTM-REF/30529/2017 (NanoCell2SEC) and COST Action EUTOPIA.

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We report dynamic retarder for asymmetric optically compensated LCDs such as IPS, FFS-LCDs and ECB, VA-LCDs. Fig.1 shows the optically compensated LCDs such as IPS, FFS, and ECB, VA mode, where retarder(compensator,+A-plate)we used in this research is the same as LC cells for the active LCD and the dynamic retarder is used by applying voltages for changing its properties. When, the azimuthal angle of LCD cell's axis, $\phi_1 = \pi/4$ and that compensator (retarder), $\phi_2 = 3\pi/4$ and they are crossed in the x-y plane and the phase retardation of both elements $\delta_1 = \delta_2$, then the optical system is symmetric [1-3]. Contrary, in the IPS(FFS) $\phi_2 = 3\pi/4 - \alpha$ the optical system is asymmetric and in the OCB(VA) when $\delta_2 \rightarrow \delta_2 - \alpha$ the optical system is asymmetric [1-3]; this is done by slating the retarder. All the alterations of $\alpha(V)$ are done by applying voltage to the element; this the meaning of the proposed dynamic retarder [1-3].

In the single cell and the proposed asymmetric optically compensate (OC) IPS(FFS)-LCD, the normalized output transmission respectively reads:

$T_1 = \cos^2(2\phi_1)$ (Normally white) and $T_2 = \sin^2(2\phi_1 + 2\alpha)$ (Normally black), and for ECB(VA) -LCD

$T_1 = \cos^2(2\delta_1)$ and $T_2 = \sin^2(2\phi_1 + 2\alpha)$ respectively.

$T_2 = I_0 \cos^2(2\phi_1 + 2\alpha)$ that has a geometrical phase advancement compared to the symmetric and single IPS(FFS)-LCD whose $I_1 = I_0 \cos^2(2\phi_1)$ and two-fold fast speed response in the switching off process. The similar results were obtained in the asymmetric OC ECB-LCD.

We have developed the computer controlled dynamic retarder equipped with an instrument for measuring electro-optical performance of the proposed asymmetric OC-LCDs.

The functions of the proposed dynamic retarder are as follows:

1. Commonly in the both of the proposed asymmetric optically compensated system it is possible to control the starting and ending time of the dynamic retarder,
2. In the T_2 of OC-IPS(FFS), real situation is not normally black, however making normally black is possible with the dynamic retarder; an example is demonstrated in Fig.2, the same example is also in the reference [1].

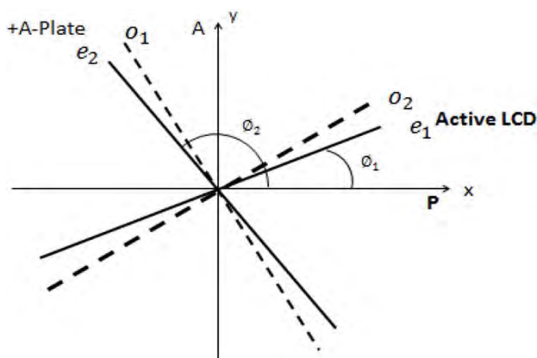


Fig. 1 Our optical system

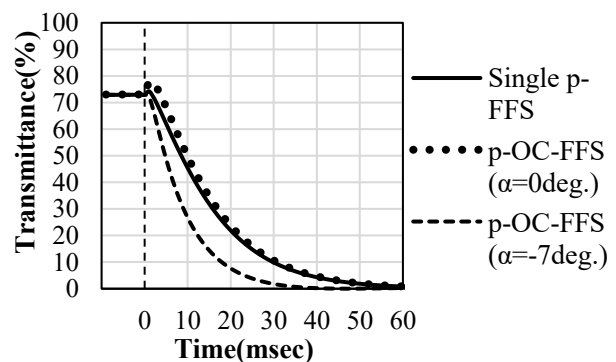


Fig. 2 Simulated t-T curves

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The hydrodynamic assembly of colloidal rods is an effective strategy for the synthesis of soft materials with structural anisotropy. In this work, cellulose nanocrystals (CNC) are used as model colloidal rods on two representative microfluidic devices, to probe the role of shearing and planar extensional flows on the alignment of CNC dispersions [1]. Characterization of the flow field by micro-particle image velocimetry is coupled with flow-induced birefringence analysis to quantify the deformation rate–alignment relationship. The deformation rate required for CNC alignment is $4\times$ smaller in extension than in shear. The birefringence signal derived from the CNC alignment in shear and extension can be scaled on a single master curve using a Peclet number that accounts for the shear and extensional viscosity of the solvent fluid, respectively. Our results will provide the basis to decouple the orientation dynamics in industrially relevant flows, where a combination of shear and extensional rates is usually present and interparticle interactions also play a crucial role. Quantification of the differences between shearing and extensional kinematics at aligning colloidal rods establishes important guidelines for the manufacture of structured soft materials.

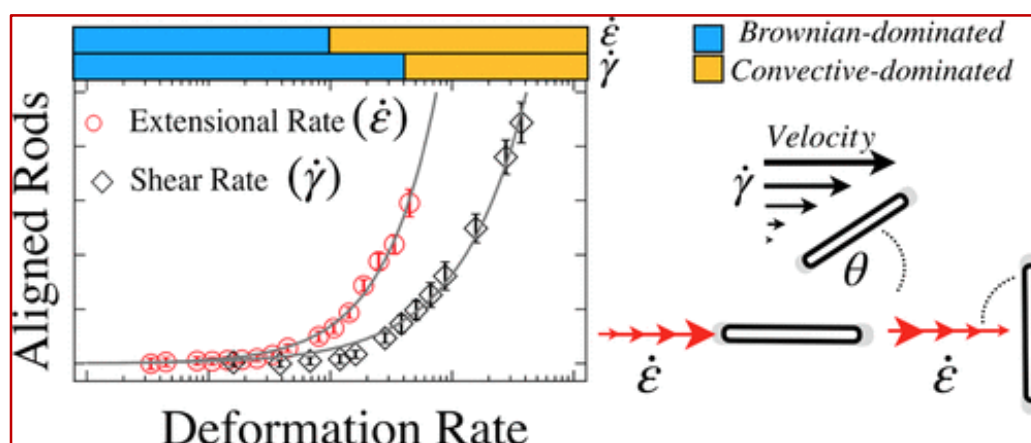


Figure 1: (Left) Number of aligned rods vs. deformation rate. (Right) Schematic of the effect of the shear and extensional rate on the orientation angle of a colloidal rod.

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Title : "WONDERLITE®Dx" - Light control window for automotive glazing.

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AGC has launched light control window "WONDERLITE®Dx". It has been adopted to automotive panoramic roof. PDLC (Polymer Dispersed Liquid Crystal) film, used as a light control material, is integrated to laminated glass and it is world first adoption of PDLC as automotive exterior window for mass-produced car.

Light control window has switchable function between transparent state and opaque state. When it is applied to automotive roof, we can get both high exhilaration and privacy and it contributes to cabin comfort. Characteristic of PDLC is high switching speed, so it brings sense of wonder. Actually, very positive feedback was given from the market.

In order to realize commercialization, of course liquid crystal material itself is required to have high performance (optical performance, high switching speed, high durability and so on), but specification of automotive manufacturer is focused on laminated glass, as final product. So laminated glass design is also important. For example, automotive window is 3D curved glass, so adaption to 3D shape is one of important point.

AGC has already launched SPD (Suspended Particle Dispersion) type light control window [1], and has knowledge of product design, glass structure, film integration technology, lamination process etc. By combination of PDLC material and glass technology, we could reach to meet requirements of automotive manufacturer.

In this paper, we introduce mainly the performance, value of WONDERLITE®Dx, and total product design of laminated glass which light control film is integrated, in order to meet requirement for automotive exterior glazing.

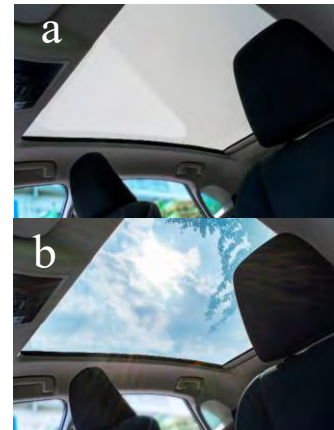


Fig. 1 Appearance of WONDERLITE®Dx
a: Off state b: On state

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LCs are widely used in every type of displays such as screens of TVs and smart phones. Their unique electro-optic properties also make them a useful material for non-display applications such as SLMs (spatial light modulators) and beam steering devices.

The simulation estimating the final performance and optimizing the design parameters is indispensable technique in designing and developing the LC devices and it facilitates clarifying causes of defects in production process of them. Recent progress in the computer technology makes the simulation reliable and useful. I would like to insist on the necessity and the effectiveness of the computer simulation with some examples carrying out a commercial simulator package [6].

The simulator consists of two major functions, which determines the orientation and behavior of the director under the given electric field by the numerical calculation based on the continuum theory [1,2], and which evaluates the optical response of the director model aligned by the first function solving Maxwell

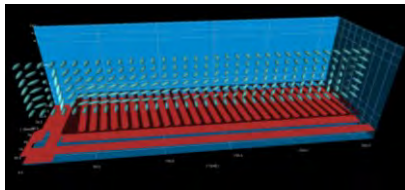


Fig.1. Director orientation of FFS cell.

equation of the electromagnetic field. Fig.1 shows a director alignment calculated by FEM in the three-dimensional model of an FFS cell. Its viewing performance may be evaluated by the transmission matrix algorithm [3,4], which represents the polarization propagation in the optically anisotropic medium. Fig.2 shows the

direction of light emitted from an LC polarization grating in a non-mechanical beam steering device of which optical configuration is illustrated in Fig.3. The azimuth of the nematic director varies 180 degrees uniformly during a period of $2\mu\text{m}$. The configuration repeats periodically along the x -direction. The optical calculation is performed by the algorithm called RCWA, in which both the dielectric function $\varepsilon(x)$ and the electromagnetic field $\mathbf{E}(x)$ are expanded in Fourier series and coupled together through Maxwell equation.

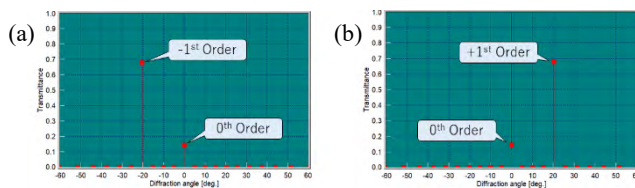


Fig.2. Transmittance vs. direction regarding the incidence of (a)left-handed and (b)right-handed circularly polarized light.

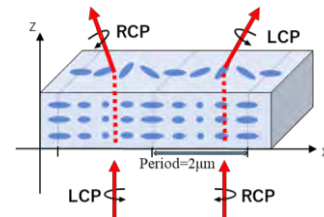


Fig.3. Optical configuration of PG.

Examples of the application of the simulation in design process of LC optical devices will be presented in the session. The theoretical foundations of the algorithms will be reviewed briefly.

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New Brightest Transparent Screen Using Tilted Cholesteric Liquid Crystals for Rear Projection

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Abstract: We have developed a new brightest transparent screen using diffraction and scattering by tilted cholesteric liquid crystals. It controls the direction of a rear projection light to realize both high brightness and transparency. We expect that it could expand outdoor AR applications for stores and vehicles.

Transparent screen displays are in high demand for augmented reality (AR) use of a large number of viewers. A rear projected type is preferable for outdoor use where a projector light is from indoor projector. However, a conventional scattering type transparent screen has a tradeoff between brightness and transparency.

We have developed a new transparent screen realizing high brightness and transparency, by using tilted cholesteric liquid crystals in Fig.1. We controlled the grating pitch of the cholesteric liquid crystals to diffract oblique projection lights to observers without color separation. In single-pitch grating, a projector light is diffracted in specific directions according to the various wavelengths, resulting in color separation. To prevent it, we applied the diffraction grating having various grating pitches distribution. We controlled the cholesteric liquid crystals grating pitch with a varied distribution in the thickness direction, by the spontaneous orientation of liquid crystals. We designed the surface energy at the air interface and at the substrate interface independently to achieve different tilt angles at the each interface. As shown in Fig.2, the tilt angle at the air interface is lower than that at the substrate interface, resulting in larger grating pitch at the air interface than that at the substrate interface, from $0.9\mu\text{m}$ to $1.4\mu\text{m}$. Thus, all visible lights can be diffracted to the observer. Also, the waving alignment generated by self-assembly of liquid crystals scatters lights to produce an optically real image. As a result, transparent screen without color separation can be realized.

Fig. 3 shows the projection image (incident light angle is 45°) and the transparent image of a background object, comparing our pitch gradient type and the scattering type transparent screens. Our screen shows much brighter image and higher transparency (low haze). We believe that it is the most appropriate for outdoor AR use such as store windows and vehicles to project advertisements.

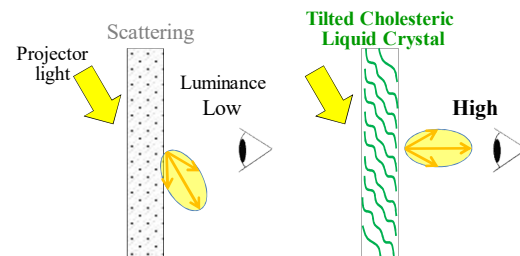


Fig. 1 Conventional type and our proposal

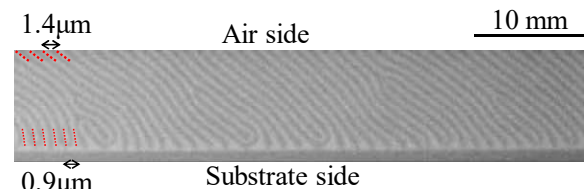


Fig. 2 Cross sectional SEM image

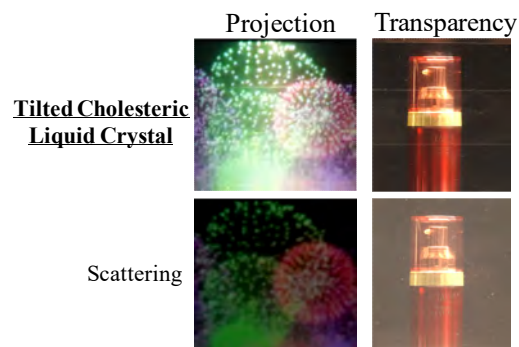


Fig. 3 Photos of projection and transparent image

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Development of Full Color Transflective and Reflective IGZO-TFT LCDs with Twisted-VA Modes

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In this report, we review our developed transflective and reflective LCD technologies, which provide high quality color image, low power consumption and good visibility under various light circumstances [1] [2].

We clarified that our developed reflective LCD technology, based on a new twisted-VA mode and an electrode with silver alloy micro reflective structures (MRS), had excellent optical properties such as high reflectance, high contrast ratio and high NTSC ratio. Flicker-less 1Hz driving was also achieved with applying IGZO-TFT technology and optimal designs of the MRS electrode, LC material and alignment film material. Some prototypes with the reflective LCD technology showed high quality video image and low power consumption. Considering the visibility in dark environments, we developed a novel transflective LCD technology based on the reflective LCD technology with the twisted-VA mode. We found that the twisted-VA mode indicated good optical performance when cell gaps of reflective and transmissive areas were same. Accordingly, between the both areas in a pixel in our transflective LCD, there was no step which caused instability of LC alignment. In addition, fabrication process of our transflective LCD became simpler than that of the conventional, which needed the step between the areas. We successfully introduced the transflective LCD technology into full color 31.5-inch IGZO-TFT LCDs the mass production of which has been started since 2020. Figures 1(a) and 1(b) show photographs of the product. We believe that the transflective and reflective LCD technologies will contribute to expanding a lineup of new LCD applications.

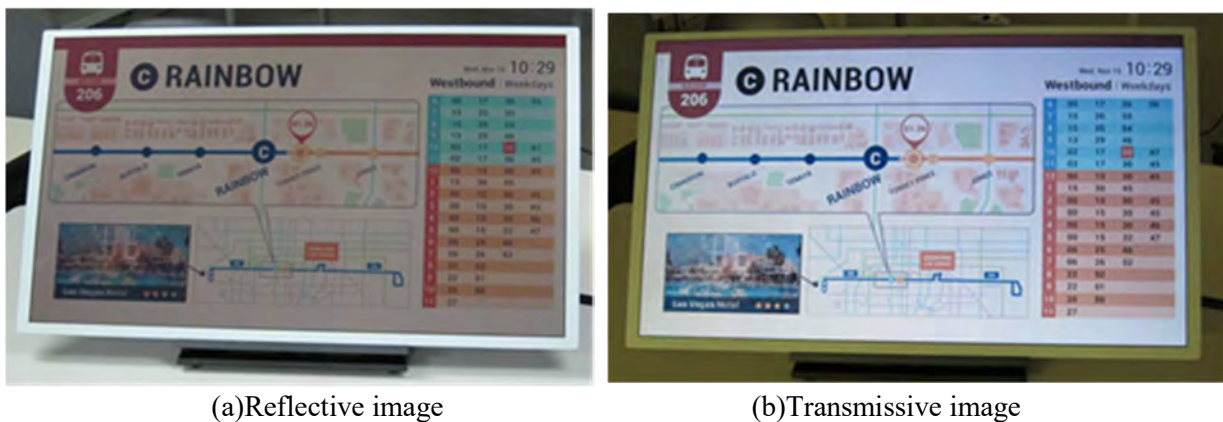


Figure 1. Full color transflective 31.5-inch IGZO-TFT LCD

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Application of Liquid Crystal Materials in the Microwave and Millimetre Wave Range for 5G and Satellite Communication Antennas

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Liquid Crystal (LC) has been subject of research in the radio frequency (RF) community for about 25 years [1]. Various efforts have been undertaken to exploit LC's unique features to produce tunable components in the microwave and millimetre wave range. Following about 10 years of research at TU Darmstadt, ALCAN Systems have developed an LC-based flat panel antenna technology applicable to frequencies from about 10 to above 77 GHz, currently used for satellite communication (SatCom) antennas and mobile/5G antennas in the Ku, K and Ka band (between 12 and 30 GHz) [2]. Due to the unique properties of LC, the concept can be extended to higher frequencies [3] even into the THz range.

In this invited talk, well aware of the material, chemistry and optics focused audience, we will visit the principles of wave propagation in RF circuits and their differences from typical optical applications, and visit the functional principles of array antennas and the benefit of using LC based components within them. This will conclude with an introduction of the LC antenna concepts used by ALCAN to enable low cost SatCom and terrestrial communications (such as but not limited to 5G applications).

At last, the particular challenges for LC materials in antenna applications will be discussed. Unlike in optics, dielectric loss of the material ($\tan\delta$) plays a relatively big role for antenna applications. This is because signal attenuation directly translates into added noise. Lowering the losses lead to surprisingly low-loss materials with the drawback of very high viscosities, resulting in very long switching times. A high tunability (that is anisotropy) of the material can reduce device sizes significantly, in turn reducing metallic losses. And last but not least, managing environmental influences such as temperature variation, will be discussed.

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Photo-triggered mass transport driven from the film surface

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Most polymer films have a free surface (surface exposed to air). Recent studies have emphasized the importance of the free surface in many aspects of polymer processing. We have recently shown that the free surface plays the critical role in controlling the mesogen alignment of side chain liquid crystalline polymers (SCLCPs) [1]. A skin layer (typically 20 nm thickness) or inkjet-printed pattern at the free surface is able to regulate the mesogen orientation of SCLCP films of micrometers thickness [2,3]. The mesogen orientation can be switched by the photoresponsive top surface both in the in-plane [2] and out-of-plane [3] directions.

Another interesting aspect is the surface effect for the induction of surface morphing.

We have recently demonstrated the photoinduced Marangoni flow occurs by UV light irradiation at inkjet-printed regions, resulting in on-demand induction of surface morphology at any timing and at selected areas [4]. These results inspired us to attempt the surface relief grating (SRG) formation of a light-inert SCLCP film only from a photoresponsive surface skin layer prepared by LS method (Fig. 1.) [5].

A large mass transfer was actually induced upon patterned UV light irradiation. It was found that an LS skin layer of Az-SCLCP can induce 20 times thicker mass transfer of the light-inert SCLCP film existing underneath to generate a SRG structure. This fact indicates that the mass transfer is driven in large part by the surface effect of Marangoni flow. Thus, a deeper understanding is obtained for our previous results with photoresponsive SCLCPs [6].

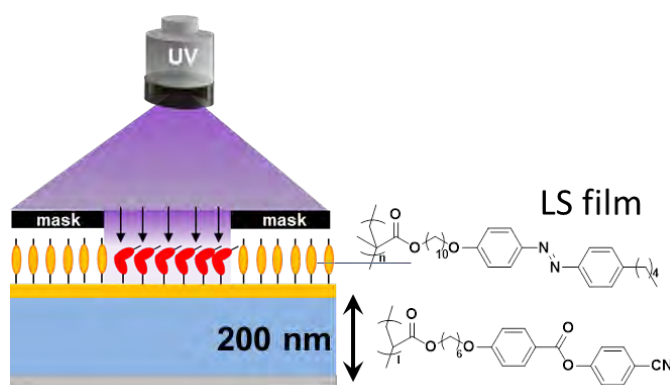


Fig. 1. Schematic of a LC polymer SRG system possessing a photoresponsive skin layer only at the free surface.

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Optical tomography of momentum-resolved emission of dye-doped monocrystalline blue phase (I)

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Cholesteric blue phase (BP) of a chiral liquid crystal is an iconic example of self-organized three-dimensional photonic crystal formed by soft matter. Usually BP exists in a narrow temperature range of 0.5-2.0 K between an isotropic and cholesteric phases, but this can be extended by use of polymer network implemented. There are three types of BPs of a different crystallographic symmetries (BP III, BP II and BP I) observed upon cooling. Well aligned and oriented monodomain BP layers are essential for practical applications¹. Common method for examination of homogeneity is observation of so-called Kossel patterns obtained under illumination with monochromatic light. Here we report a novel approach of studying optical properties of monocrystalline BP I (includes Kossel patterns), which is three dimensional momentum-resolved tomography of the emission. Our samples can be doped with various luminescent dyes (like P580), which allows for direct observation of the dispersion relation of light emitted from the inside of the BP.

Optical properties as emission or reflectivity spectra for BP strongly depends on directionality^{2,3}. The method proposed by us gives direct access to reciprocal space of optical diffraction. The obtained results are three-dimensional maps in the space of energy and momentum (in both k_x and k_y direction). The cross-section of this map in the energy- k_x space reveals presence of the photonic gap. On the other hand the cross-section of the k_x - k_y space results in observation of Kossel patterns for any wavelength within the spectral emission range of the dye with which the sample was doped.

The optical tomography of BPs opens up new possibilities for research. It allows the study of quality and orientation of the crystal, determination of the lattice constant with better precision and observation of full dispersion relation of transmitted or emitted light by an angle-resolved experiment. It will help in better understanding of the interactions between the light and blue phase and will give the opportunity to observe new physical phenomena, including quantum ones, related to it.

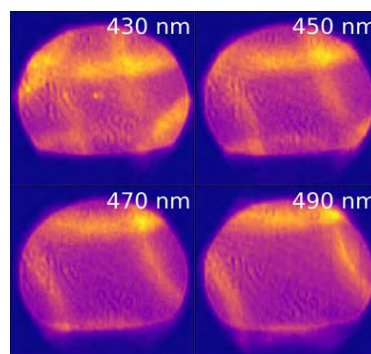


Fig 1. Kossel patterns obtained by momentum-resolved tomography.

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Optical solitons and chirality-enhanced nonlinear optical response in frustrated cholesterics

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During the past 20 years, seminal works [1,2,3] by the groups of Prof. Assanto and Prof. Karpierz have established liquid crystals as an invaluable platform to study spatial optical solitons, which correspond to self-focused laser beams propagating without diffracting thanks to nonlinear optical effects. Indeed, liquid crystals are associated with a giant nonlocal response to external fields leading to the robust formation of accessible optical solitons called nematicons at beam powers of typically a few mW. Such optical solitons have mostly been studied in achiral nematic liquid crystals with a well-defined uniform far-field optical axis, in which the laser-induced perturbation of the molecular orientational field allows the compensation of diffractive effects. Although a few preliminary works [4,5] have shown the possibility of generating similar self-focused beams in chiral liquid crystals, the role of chirality in the nonlinear optical response of liquid crystals have not yet been fully elucidated.

In this contribution, I will present our recent contribution [6] to this topic: chirality can boost the nonlinear optical response of frustrated cholesterics cells with homeotropic boundary conditions, thus leading to the formation of optical solitons at lower power than the nematicons of purely achiral samples. Such solitons are associated with an interesting bouncing pattern thanks to total internal reflections between the confining plates of the sample (see image below). I will explain the origin of this chirality-enhancement effect based on the formalism of Green functions, and if time allows I will also present additional applications of this novel physical effect.

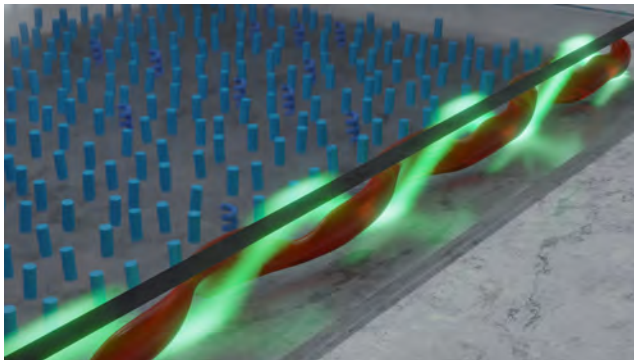


Figure: Simulated bouncing optical soliton (in green) in a frustrated cholesteric cell. The red domain corresponds to the part of the LC bulk with twist distortions of natural handedness.

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Multistable ring-shaped liquid crystal configurations through patterned planar photo-alignment

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Patterned liquid crystal (LC) configurations find widespread applications in functional devices such as lenses, gratings, displays and soft-robots. Photo-alignment at the confining surfaces offers an attractive way to steer the self-assembly of hierarchical LC superstructures in the bulk [1-3]. By combining a rotating planar alignment at one substrate with uniform planar alignment at the opposing substrate, defects in the LC are usually created. Herein we use projection illumination to define a ring-shaped photo-alignment pattern at the bottom substrate with a 360° azimuthal rotation (when moving from the inner radius r_{in} to the outer radius r_{out} Fig. 1(a)) [2]. For this alignment configuration, the formation of four different metastable structures is observed:

- structure A in which a ring-shaped region is formed with an out-of-plane (vertical) orientation perpendicular to the substrate,
- structure B which has a single disclination loop and a 180° twist at the inner region of the photo-patterned ring ($r < r_{in}$),
- structure C which has no discontinuities but a 360° twist in the inner region ($r < r_{in}$),
- structure D in which two disclination loops are observed.

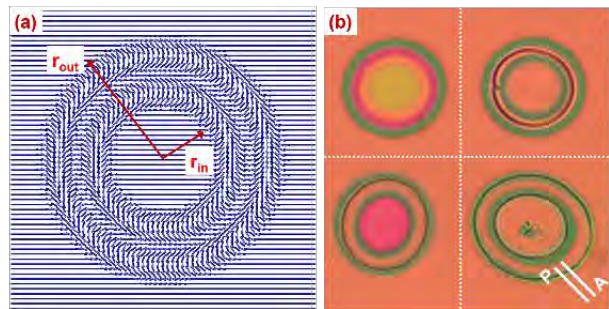


Fig. 1 alignment pattern; POM image

Even though the alignment patterns at the top and bottom substrates define planar alignment and induce twist conflicts, bulk configurations without singular disclinations are also observed. Discontinuities in the director configuration can be avoided by creating a localized region with close to vertical director orientation in the bulk (structure A). Moreover, the closed-loop photo-alignment pattern also allows to avoid disclinations by introducing a twist deformation in the unpatterned inner region of the circles ($r < r_{in}$). One disclination line can shrink and thereby induce a 180° twist in the central region (structure B) or both disclination lines can disappear by introducing a 360° twist (structure C).

The LC director configurations are simulated through finite element Q-tensor simulations and the optical transmission is simulated using a generalized beam propagation method.

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Imprinting the vectorial nature of light in liquid crystals

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Among the properties of light that dictate its mechanical effects, polarization has held a special place since the mechanical identification of the photon spin by Beth in 1936. Nowadays, little surprise might be expected from the mechanical action of linearly polarized weakly focused (paraxial) beams on transparent and homogeneous dielectrics. Still, here we unveil vectorial optomechanical effects mediated by the material anisotropy and the longitudinal field component inherent to real-world beams. Experimentally, this is demonstrated by using a nematic liquid crystal that is prone to exhibit a sensitive and reversible effect and our results are generalized to vector beams, as depicted in the figure below. This corresponds to an alternative strategy compared to other approaches of laser processing of materials in presence of structured light fields using glassy or polymeric materials, whose lack of reversibility prevent from developing single-snapshot imaging of structured light. Our results thus represent as a first step towards developing liquid crystals based vectorial polarimeters. Also, recalling that the light-driven reorientation of liquid crystals is related to giant optical nonlinearities, we show that, in contrast to the usual self-focusing situation studied since the early 1980s, it is possible to create multiple self-induced lenses from a single beam which appears worth exploring in the context of spatial optical solitons in liquid crystals.

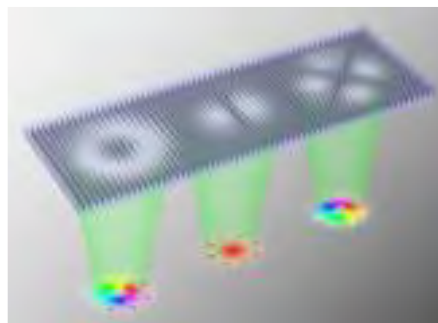


Illustration of the optomechanical recording of vectorial optical structures into a liquid crystal layer.

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Title : Synthesis and Characterization of Photochromic Controllable Mesomorphic Diarylethene Derivatives

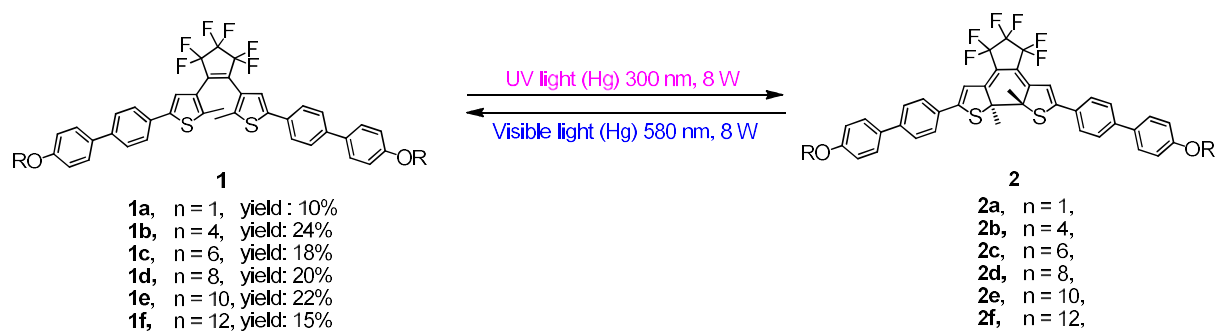
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A series of photochromic mesomorphic diarylethene derivatives, denoted as compounds **1a-f**, were synthesized and characterized.^[1-2] The colorless hexane solution containing **1** turned blue with 330-nm light and reversed to colorless by visible light at 580 nm. The conversion from the open-ring form to the closed-ring form reached a photostationary state under UV irradiation, with 99% as closed form **2**. The photochromic behavior of **1** was investigated in solutions and thin films by nuclear magnetic resonance spectroscopy (NMR) and UV-visible spectroscopy. Both open and closed forms showed liquid crystalline (LC) properties, which was investigated using polarizing optical microscopy (POM), differential scanning calorimetry (DSC), and powder X-ray diffraction (XRD). The compound **2a** with short methyl group exhibited excimer/exciple formation as well as aggregation induced emission phenomena in the THF. Formation dynamics of intermolecular excimer/exciple was studied by UV-vis spectroscopic method.



Scheme 1. The photoswitching of diarylethene derivatives **1**.

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Ultrafast collective motions of liquid-crystalline azobenzene molecules observed by ultrafast time-resolved electron diffraction

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We performed ultrafast time-resolved electron diffraction measurements to demonstrate the formation of higher-orientation of the liquid-crystalline (LC) azobenzene molecules on the picosecond timescale by linearly polarized ultraviolet (UV) pulse [1,2]. The photoisomerization of azobenzene molecules is the representative dynamic phenomena in soft matter. However, the ultrafast structural dynamics of the molecules during isomerization of azobenzene molecules and their subsequent response have not been well explored because of their complex and ultrafast nature. We found that the ultrafast higher-orientation is caused on the timescale of ~ 100 ps through the trans-to-cis photoisomerization of the azobenzene molecules. Figure 1 shows the schematic illustration of ultrafast collective motions of LC azobenzene molecules by the trans-to-cis photoisomerization of a azobenzene molecule. This is the fastest collective motion observed in the molecules in soft matter. Our observations are consistent with simplified molecular dynamics calculations that revealed that the molecules are aligned with the laser polarization axis by their cooperative motion after photoisomerization. The insight obtained in this study advances the fundamental chemistry of photoresponsive molecules in soft matter as well as their ultrafast photomechanical applications.

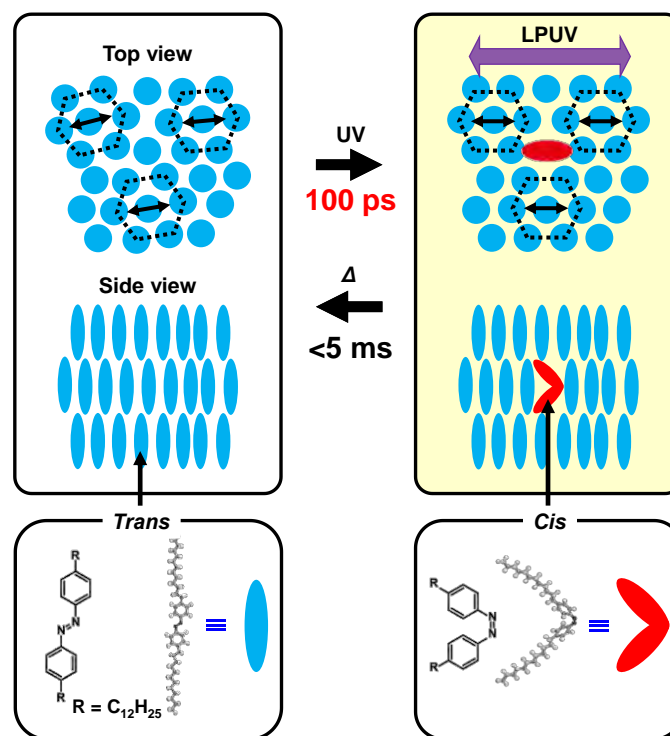


Fig. 1 Schematic illustration of ultrafast collective motions of LC azobenzene molecules

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Nature's light manipulation strategies, especially those at the origin of bright iridescent colours, have fascinated humans for centuries. Many arthropods display polarization sensitive iridescent colours for communication and aid its camouflage against predators. This phenomenon is due to the hierarchical Bouligand-type structures that exhibit helical organization and periodic surface structure. Knowledge about the interplay between the morphology, composition and optical appearance of biological photonic systems can provide inspiration for novel artificial photonic devices. Inspired by the beetle of *Chalcothea smaragdina*, here we demonstrate a scalable route for the manufacture of custom-tailored photonic cellulose paper with hierarchical architectures. We combine the nanoscale self-assembly of cellulose nanocrystal with microscale imprinted surface grating-like structure, exhibiting dual structural colour that derived from the helical matrix and circularly polarized light diffraction.^{1, 2} Besides, we further extended cellulose nanocrystal as ink for transfer imprinting lithography to present a micropatterned photonic cellulose paper for retroreflection.³ We disentangle the hierarchical cellulose printing in different steps: the microscale surface structure of the patterned template, the drying dynamics of the nanoscale cellulose self-assembly and, finally, the coupled optical properties of bulk films. The resulting photonic cellulose paper is particularly important to understand how hierarchy affects the properties of chiral light-matter interactions in living organism, not only provides inspiration for designing of bio-mimetic artificial counterparts, but also allows to further understand their biological significance in nature.

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Quasi Twisted Nematic Mode Using Asymmetry Polar Anchoring in Hybrid Aligned Cell

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A “twisted hybrid aligned (THA) [1]” cell or a “hybrid twisted nematic (HTN) [2]” cell has been proposed for a low driving voltage. In this study, we have designed the HTN cell using asymmetry polar anchoring strength, that is, strong polar anchoring of the planar surface and weak polar anchoring of the homeotropic surface. We can obtain the same LC director distribution as that in a conventional 90° TN cell, therefore, we call the cell a quasi-twisted nematic (Q-TN) cell. LC director distributions and electro-optical properties in the Q-TN cell were numerically analyzed.

Figure 1 shows a schematic model of the Q-TN cell. Azimuthal easy axes of two substrates are orthogonal and strong (infinite) azimuthal anchoring strength. When the polar anchoring of the homeotropic surface ($W_{p,d}$) becomes lower than critical one (W_c), the tilt angle on the surface is 0°. In the case of quasi homogeneous cell, W_c is estimated as K_{11}/d [3]. When the high voltage is applied, the LC the LC director configuration becomes the same as the HTN. The LC director distribution is numerically analyzed by minimizing a total free energy of the cell using a finite difference method. A transmittance is estimated using Jones matrix, as a function of normalized applied voltage. In this study, elastic constant ratio $K_{11}:K_{22}:K_{33}=1:0.75:1.4$ is used, that is very usual nematic LC physical property for display devices. W_c is estimated as K_{11}/d if $K_{33}=2K_{22}$. Figure 2 shows tilt angle θ and twist ϕ angle at the center of the cell and transmittance in the Q-TN. The LC reorient without the threshold voltage. ϕ ($2/d$) decreases with the voltage rapidly, then the polarization rotation of the incident light does not occur and transmittance decreases with very low field. The driving voltage V_{10} is about $0.35V_{th}$ ($V_{th} = \pi \sqrt{(K_{11} + \frac{K_{33}-2K_{22}}{4})/\Delta\epsilon\epsilon_0}$), comparing to $1.8V_{th}$ in the conventional 90° TN cell.

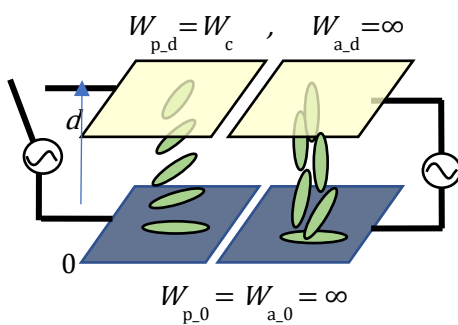


Fig. 1 Schematic model of Q-TN cell in off- and on states.

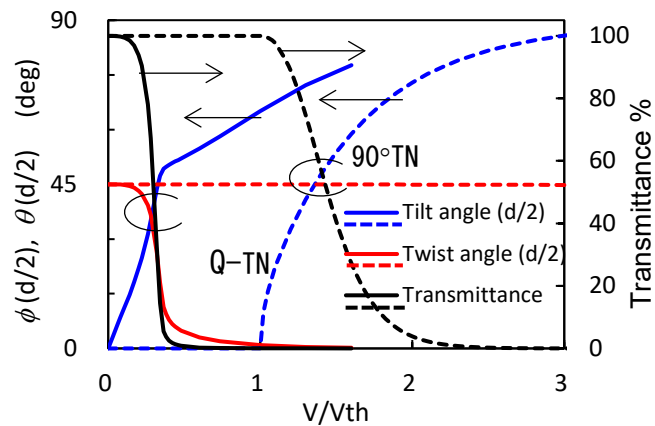


Fig. 2 Tilt and twist angle at the center of the cell and transmittance in the TN and the Q-TN. Δn , λ : 1st Morgan minimum condition.

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In-fiber switchable polarizer by capillary-aligned liquid crystals

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We present an electrically switchable polarizer that was embedded inside of standard telecommunication fiber (SMF-28). Following on our prior work [1,2], a periodic array of nanometric sized holes were opened fully through the fiber cladding by ultrashort pulsed laser nano-exposition. The spatial pitch of holes ($\Lambda = 1072$ nm) was selected to generate a second order Fiber Bragg grating with a resonant reflection at ~ 1550 nm, exhibiting very small polarization dependence.

The injection of 5CB nematic liquid crystal material (NLC) into the hole array was done via the capillarity. Due to the elastic energy minimization, the liquid crystal molecules were oriented predominantly along the long axis of holes that were positioned perpendicular to the propagation direction of waveguiding light along the fiber axis. This strong anisotropy of the NLC gave rise to high birefringence in the Bragg resonance (figure 1) resulting in stronger and wider stopbands forming due to high value of effective refractive index for the extra-ordinary polarized light (e-mode) relative to the ordinary polarization (o-mode). The strong Bragg stop band (1552 – 1556.5 nm) for the extraordinary, polarized light further provided high polarization dependent losses on the long wavelength side of the resonance (figure 1), resulting in a strong extinction ratio of up to 20 dB extending over a ~ 5 nm band. A moderate insertion loss of < 1 dB was noted arising between the two polarization states of the shifted stopbands. The spectral responses of the grating followed the thermo-optic response of the NLC to further offer dynamic tuning and switching of the polarization extinction response without increase in the insertion loss.

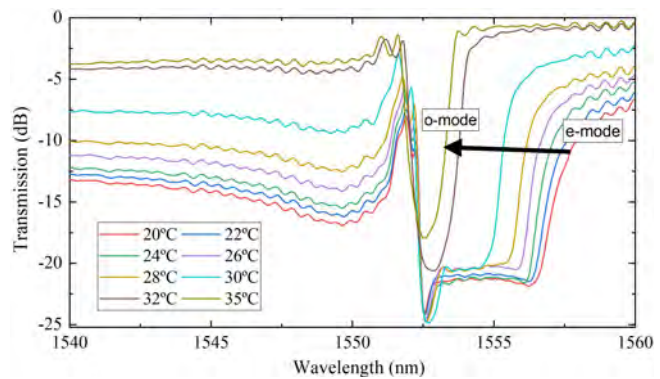


Figure 1. The thermo-optic response of 2nd order hollow FBG ($\Lambda = 1072$ nm, 1000 nanoholes) filled with NLC showing a series of transmission spectra recorded for parallel polarization over temperature range of 20°C to 35°C.

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High open circuit voltage in double chiral ferroelectric liquid crystals comprising an extended π -conjugated unit

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The most remarkable characteristic of liquid crystalline (LC) semiconductors is that macroscopically homogenous thin films can be produced by solution processes [1,2]. Our research group synthesized ferroelectric liquid crystals comprising oligothiophene units and chiral side chains to exhibit a bulk photovoltaic effect and rotation of the polarized plane of electroluminescence.

Recently we have reported a bulk photovoltaic effect in a polarized smectic phases of chiral phenylterthiophene derivatives (*R*)-**1** and (*R,R*)-**2** (Fig. 1(a)). In the polarized smectic phase, the internal electric field produced by the electrical macroscopic polarization promoted photocarrier generation and transport [3,4]. By doping fullerene derivatives in (*R*)-**1**, an open circuit voltage over 0.5 V was generated in a symmetrical cell consisting of ITO anode and cathode. By the inversion of the poling DC bias, the polarity of the photovoltaic effect could be changed [5]. In the polarized smectic phases of the compound (*R,R*)-**2** doped with fullerene derivatives, the external quantum efficiency exceeded 80 % and the open circuit voltage reached 1.0 V in a symmetrical cell using ITO electrodes (Fig. 1(b)).

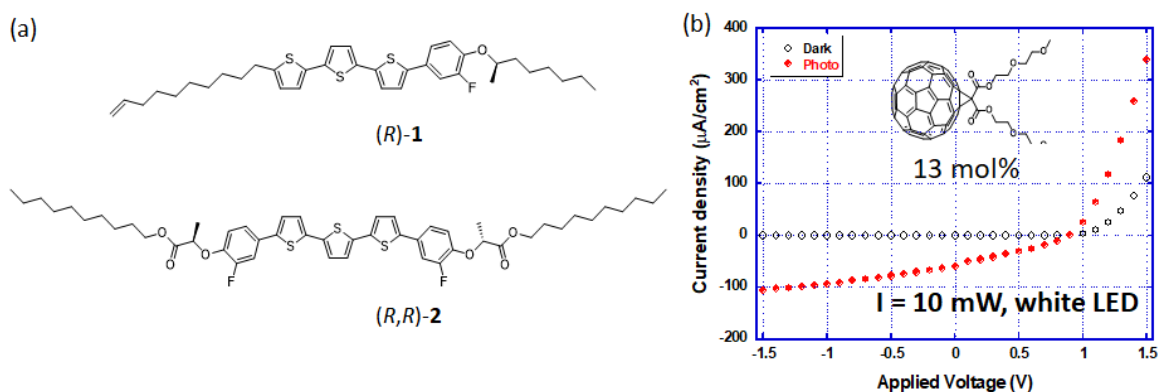


Figure 1(a) Molecular structures of chiral liquid crystalline semiconductors. (b) J-V characteristics in the polarized smectic phase of (*R,R*)-**2** doped with a fullerene derivative for illumination of blue light (5 mW/cm²)

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Hyperbolic Metamaterials with Nematic Liquid Crystals and Dye in Spontaneous Emission Engineering

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Hyperbolic metamaterials (HMMs) are special class of engineered metamaterials which exhibit hyperbolic dispersion for interactions with electromagnetic waves, and they possess non-trivial electromagnetic properties depend on the spatial projection of their nanostructure. Their unique properties such as: negative refractive index, near-zero and indefinite permittivity or permeability indices, backward wave, create completely new perspectives of manipulation of electromagnetic radiation [1-3]. In this work we investigated hyperbolic metamaterials with nematic liquid crystals (NLCs + Dye) in spontaneous emission engineering. The vacuum isofrequency surface of propagating waves in an isotropic media is spherical and is given by:

$$k_x^2 + k_y^2 + k_z^2 = \left(\frac{\omega}{c}\right)^2 \quad (1)$$

In HMMs which are anisotropic uniaxial media, the tensors ϵ and μ are given by:

$$\hat{\epsilon} = \text{diag}[\epsilon_{xx}, \epsilon_{yy}, \epsilon_{zz}] \quad (2) \quad \hat{\mu} = \text{diag}[\mu_{xx}, \mu_{yy}, \mu_{zz}] \quad (3)$$

There are two types of HMMs that depend on which components of dielectric tensor are negative – Type I have one component negative ($\epsilon_{\parallel} > 0$ and $\epsilon_{\perp} < 0$), while the Type II have two negative components ($\epsilon_{\parallel} < 0$ and $\epsilon_{\perp} > 0$).

The most important consequence of unbounded dispersion of Type I and Type II of HMMs is a divergent photonic density of states (PDOS) resulting in an enhancement in the spontaneous emission of dipole emitters placed in the vicinity of HMM leading to a metamaterial based broadband Purcell effect and in the result the possibility of control of gain/absorption in tunable hyperbolic metamaterials. The idea and design of the transducer developed on the basis of the HMM structure with a layer of liquid crystal and dye could allow to obtain controlled emission enhancement by photon excitation, which is simplified as shown in Fig. 1.

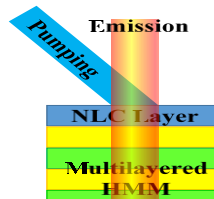


Fig. 1. HMM transducer design with a layer of NLC and dye to achieve emission enhancement.

Properties of HMM cells were simulated and the next check experimentally. Simulations of HMM structure properties as well as Purcell factor calculations were performed using by the Lumerical Software. The obtained gain can be described quantitatively using the Purcell factor which could be measured enhancement in the total fluorescence intensity as well as by Ramann Scattering Spectroscopy. We propose the constructions of the transducer based on the multilayered hyperbolic metamaterials Ag/SiO₂ structure with an additional layer of nematic liquid crystals with high optical anisotropy and dye used to control the gain of spontaneous emission associated with the Purcell effect more than 10 times.

This work has been supported by The National Centre for Research and Development under grant No. TECHMATSTRATEG1/347012/3/NCBR/2017(HYPERMAT) in the course of "Novel technologies of advanced materials - TECHMAT-STRATEG."

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Observation of Polymer Structure and Electric-field Response of Liquid Crystal Director in Composite Films Consisting of Liquid Crystal and Highly Oriented Polymer

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The composite films consisting of liquid crystal and unidirectionally oriented polymer exhibit light control function that are transparent without an electric field and become turbid with an electric field. Since the index ellipsoids of both the oriented polymer and the liquid crystal in the film match well without an electric field, the composite film is remarkably transparent even viewed from an oblique angle. The film is prepared via photopolymerization-induced phase separation of precursor consisting of liquid crystals, monomers, and a photoinitiator. In this study, the phase separation process and the resulting polymer structure in the composite films doped with fluorescent monomer were observed nondestructively by using confocal laser-scanning microscope (CLSM). Furthermore, by doping the fluorescent monomer and a dichroic dye together, a relationship between the polymer structure and the azimuthal distribution of liquid crystal director in the film were observed using CLSM equipped with a polarization rotation unit.

It was found that the highly oriented pillar-shaped structure of polymer-rich phase was generated in an initial phase separation process, and the observed image contrast between the polymer phase and the liquid crystal one increased with progress of phase separation while maintaining the pillar-shaped structure. It is suggested that a highly oriented pillar-shaped polymer structure (Fig. 1(a, d)) in the direction perpendicular to the cell substrate surface was formed via spinodal decomposition due to the large anisotropy in the coefficients of the gradient energy and the diffusion.

The initial state of the liquid crystal director in the film was a uniform homeotropic alignment (Fig. 1(b, e)), but it was found that the liquid crystal directors in each domain partitioned by the pillar-shaped polymer structures were tilted in different azimuthal directions by application of an electric field normal to the cell substrate surface (Fig. 1(c)). Interestingly, mono-domains were observed in the thickness direction under an electric field, suggesting that the light control function of this film is due to a new mechanism rather than the conventional transparent-scattering mode by refractive index-mismatching.

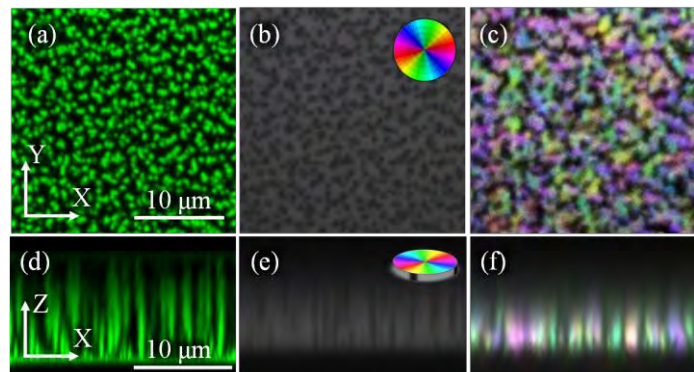


Figure 1 XY (a, b, c) planar and XZ (d, e, f) cross-sectional images of polymer structures (a, d) and azimuthal distribution of LC directors without (b, e) and with (c, f) application of electric field to a highly oriented liquid crystal / polymer composite film.

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These days, topological defects in LCs have been paid much attention to because they could potentially control the structure and motion of LCs. Intentionally formed topological defects by the photo-alignment layer could control the alignment of LCs and can be utilized for various thin optics. When a droplet is formed with LC, different types of topological defects were formed inside and on the surface. These droplets and spheres have been studied intensively as an ‘active matter,’ where the object could move around spontaneously like a living object by external energy sources.

In this study, we accidentally found a unique crystallization phenomenon while studying a new category of active matters consisted of LCs, which could move around in a surfactant solution due to an induced convective flow inside and outside the droplets while gradually dissolving into it. We found the photo-controlled motion of the LC droplets by promoting adsorption and desorption of molecules [1,2] and showed a clockwise/anti-clockwise rotation under the light.[3] During this study, we found a crystallization of a chemical was triggered by light at the topological defect in a pure LC droplet (Fig.1), even though the chemical was dissolved outside the LC droplet. Under the light irradiation whose wavelength matches the absorption of chemicals, a crystal was formed inside an LC droplet. It grew at the center of the droplet (topological defect) with a branched shape like an ice crystal. This is the first demonstration of the topology-induced crystallization under the geometrical frustration.

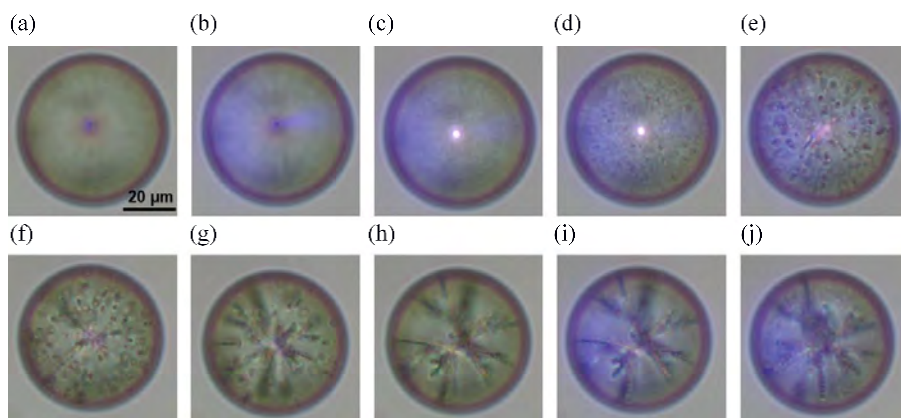


Fig.1 The snapshots of a 5CB droplet in an SDS solution with p-nitrophenol (0.01 wt%) under the on-off operation of the UV light is shown; (a) before irradiation, (b) - (e) 0, 30, 60 and 90 s after the UV was turned on, (f) - (h) 0, 60, and 120 s after the UV light was turned off, (i) and (j) 0 and 30 s after the UV light was turned on again.

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Understanding structure formation and related phase transitions is a ubiquitous problem in many scientific disciplines, with significant academic and practical implications. Recently, resonant X-ray scattering has shown that the cholesteric blue phase (BP) I with body-centered cubic (bcc) symmetry shows twinning on the (211) plane, which is commonly found in atomic crystals with the same symmetry [1]. While X-ray measurements can provide high-resolution diffraction data, it is not easy to perform microscopic observations in a specified area of the sample or in-situ observations while applying a field. Here, a detailed optical analysis of twinning in BP I was performed through a combination of wide-field and Kossel pattern observations.

A BP material mixture was filled into a sandwich cell with a cell-gap of $\sim 5 \mu\text{m}$, and temperature-controlled to show BP I. Polarized optical microscope images were obtained together with Kossel patterns using a high-magnification objective lens. Also, an electric field was applied in the cell-normal direction to investigate *in-situ* the evolution of twin domains.

Figure 1 shows Kossel patterns from a monodomain and twinned BP I with (110) lattice plane orientation. In addition to the four Kossel lines from (110) planes creating a pattern with two-fold symmetry, six lines are observed in the twinned domain, attributed to a shared (211) plane. We discuss how the Kossel pattern can be analyzed to obtain the twin elements of BP I, and a reveal a twin-induced pinning effect that occurs as the BP I undergoes electrostriction by an electric field.

Acknowledgements

We acknowledge Prof. Hirotsugu Kikuchi, JNC Corporation and DIC Corporation for providing materials, and Prof. Jun-ichi Fukuda for providing the order parameter data of BP I. This work was partly supported by MEXT KAKENHI (19H02581, 20H00391, 20H04672, 20K21154), MEXT LEADER Program, and NSFC (65005008).

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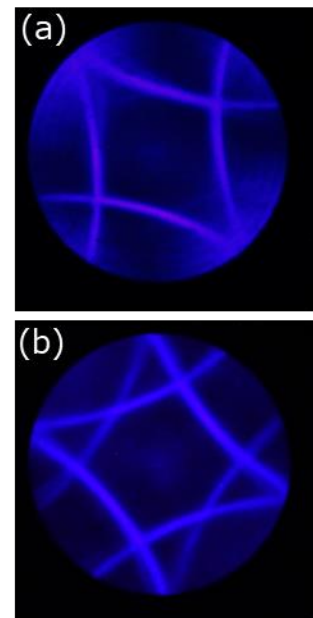


Figure 1 Kossel pattern of a (a) monodomain and (b) twinned BP I.

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Structural colors, originating from a photonic nanostructure with a periodicity in the visible-wavelength range, are ubiquitous from animals to plants. Interestingly, some tropical fish can dynamically change their structural colors by modulating the periodic distance of crystalline guanine sheets cofacially oriented in their cytoplasm. Inspired by such a sophisticated system, we recently reported that an aqueous dispersion of a tiny amount of inorganic nanosheets (<0.5 vol%) exhibited freely tunable structural colors [1].

The building block of our dynamic photonic crystal is a titanate nanosheet (TiNS, Fig. 1a), featuring ultra-thin (0.75 nm) and extra-wide (several μm) dimensions [2]. Owing to its dense negative charges on its surface, the strong electrostatic repulsion works between TiNSs in addition to the van der Waals attraction. These competitive repulsive and attractive forces allow for interlocking TiNSs in a cofacial manner at a fixed relative position, as described in the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory [1,3]. Therefore, TiNSs form a liquid-crystalline lamellar structure in water, although the periodicity of as-prepared TiNSs is too small to exhibit a structural color (Fig. 1b, left) [4]. We found that the periodicity could be drastically enhanced up to 675 nm by reducing the excess amount of ions in the dispersion of as-prepared TiNSs because of the increment of the electrostatic repulsion between TiNSs (Fig. 1b) [1]. To our surprise, the resultant dispersion composed of >99.5 vol% water and <0.5 vol% TiNSs exhibited vivid structural colors, which could be easily modulable from UV (370 nm) to even NIR (1,750 nm) regions upon varying the TiNS content (0.50–0.09 vol%) (Fig. 1c). The obtained photonic crystals quickly modulated their structural colors upon various external stimuli, such as temperature, magnetic field, and pH, because of their fluidity. Furthermore, we succeeded in fixing the photonic structure as a hydrogel, so that the obtained photonic hydrogel changed its color in response to a mechanical force [5]. More recent findings will be also discussed in the presentation.

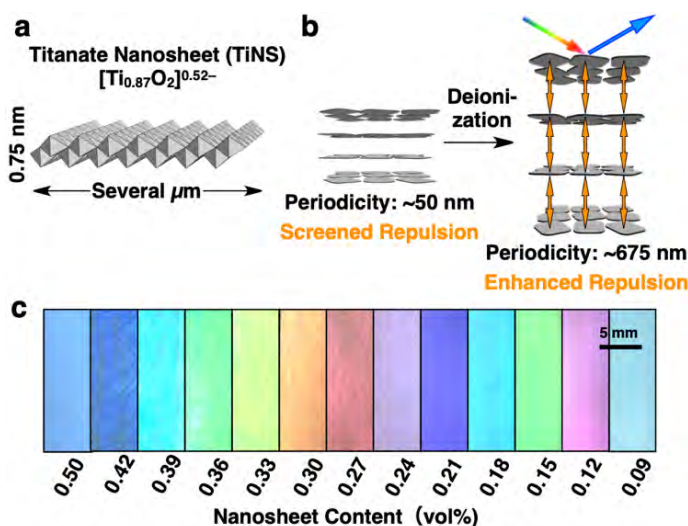


Fig. 1. (a) Schematic image of titanate nanosheet (TiNS) and (b) expansion of TiNS distance by deionization. (c) Real images of the dynamic photonic crystals with various TiNS contents.

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Bulk Photovoltaic Effect for Polar Columnar Liquid Crystals

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The bulk photovoltaic effect (BPVE) involves direct current generation in noncentrosymmetric semiconductors or insulators under light irradiation. The BPVE is fundamentally different from the conventional photovoltaic effect observed in p-n junctions. Recently, the BPVE has attracted wide research interest due to its unique features, such as being ultrafast responsive, being dissipation-less, and having a low noise photocurrent. Thus far, the BPVE has been studied mainly for inorganic ferroelectrics such as oxides, chalcogenides and hybrid perovskites, while studies on the BPVE in organic materials are quite limited.^{1,2,3} In general, rationally designing such polar organic materials is difficult because molecules prefer to assemble symmetrically, resulting in nonpolar assemblies.

Recently, we reported a hexagonal columnar liquid crystal with a bowl-shaped π conjugated molecule, subphthalocyanine (SubPc-SC₁₂). Although individual columnar assemblies of SubPc-SC₁₂ are intrinsically polar, the as-prepared sample exhibits no SHG signal, indicating that no macroscopic polarization was developed. On the other hand, just by cooling the sample from its isotropic melt under an application of DC e-field, the resultant assembly became SHG active. Interestingly, the obtained polar columnar assemblies exhibited the BPVE over a wide range of wavelengths up to 650 nm (Figure 1). Furthermore, just by sandwiching this columnar assembly between two ITO electrodes, the resultant device reaches a light-on/off ratio, $I_{\text{Light}}/I_{\text{Dark}}$, as high as 6.6×10^3 , indicating that the polar columnar assemblies with SubPcs are promising for photodetectors.⁴ In this presentation, we also discuss the relationship between BPVE and the macroscopic polarization developed by SubPc-SC₁₂.⁵

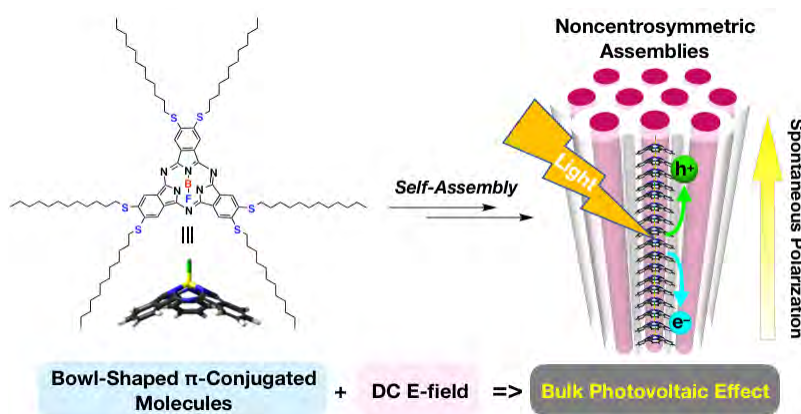


Figure 1: Schematic illustration of bulk photovoltaic effect for the columnar assembly of SubPc-SC₁₂

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01

Light-melt adhesives: Structural design and adhesive performances

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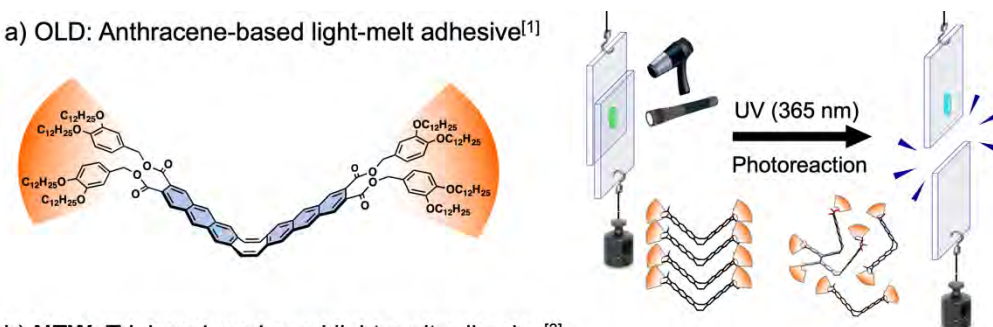
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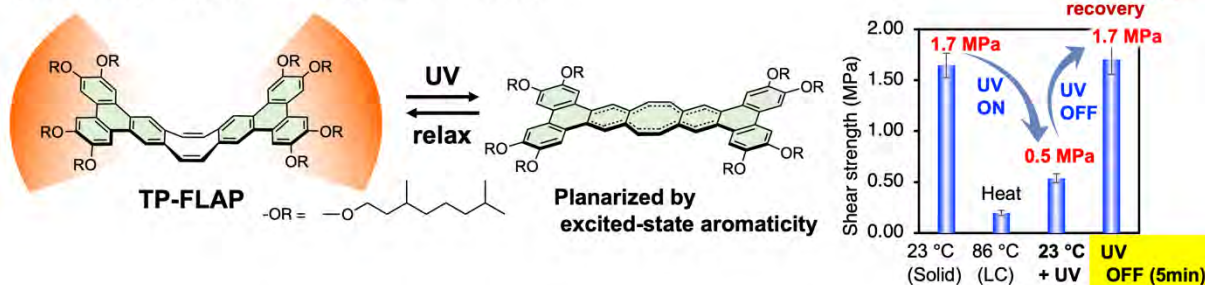
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Light-melt adhesive offers a new application of columnar liquid crystals.^{1,2} The previously reported flapping anthracene molecule shows a high cohesive force originating from the tightly stacked V-shaped columnar π -stacking, while the UV-induced dimerization of the anthracene moieties leads to disorganization of the ordered phase, providing a non-adhesive fluid.¹ However, the anthracene-based adhesive has some remaining problems, which hamper practical applications in industry. To solve the problems, here we have developed a triphenylene-based light-melt adhesive (TP-FLAP).³ TP-FLAP has the following advantages: 1) Novel working principle for realizing the UV-induced melting behavior. Flapping dynamics of the V-shaped molecules is induced by excited-state aromaticity, which leads to rapid destruction of the columnar packing without any photoreaction. 2) Spontaneous recovery of the adhesive strength (instant reworkability) by simply stopping the UV irradiation. 3) Large-scale synthetic approach established by only three-step reactions from a commercially available compound and easy reprecipitation process. Molecular design, adhesive performance, and the working mechanism revealed by ultrafast electron diffraction analysis⁴ will be introduced.

a) OLD: Anthracene-based light-melt adhesive^[1]



b) NEW: Triphenylene-based light-melt adhesive^[3]



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A Photovisible Capacitor with Enormous Capacitance Tunability Realized by A Polar Nematic Fluid

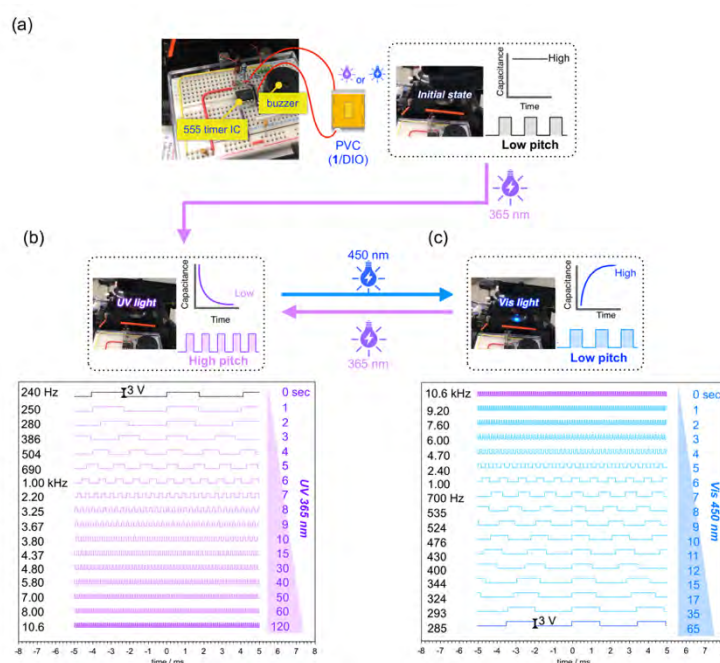
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In early 2017, a new nematic polymorph with highly polar order has been found in archetypal molecules, RM734 [1] and DIO [2] which usher in an entirely new era in the history of liquid crystals. A common significant characteristic of them is a colossal dielectric permittivity ($>10,000$) in a polar nematic phase. Needless to say, they have vast potential for innovative applications by the proactive use of its gigantic dielectric properties. For example, with the aid of the anomalously large dielectric permittivity, just a usual sandwich LC cell with a micron-gap may work as an electric condenser with a useful capacitance value. In this work, we demonstrate an epoch-making photovisible electric condenser (PVC) by doping DIO with a photo-trigger molecule, resulting in an extremely wide phototunability in capacitance between $0.34 \mu\text{F}$ and 7.0 nF . To the best of our knowledge, this is the first PVC using LC with such a wide capacitance tunability. To prove the usefulness of our PVC, we fabricated an electric sound oscillator using PVC. The sound pitch ranging over the entire range from 240 Hz to 10.6 kHz within the human audibility could be controlled remotely, rapidly, reversibly, and drastically by light stimuli (Fig. 1). Such an unprecedented photo-tunable feature with a fluidic nature may bring innovation in the state-of-the-art flexible devices, such as for soft robotics, etc. and may open the new field for applicational uses of LCs other than displays.

Fig.1: Demonstration of the photo-tunable sound oscillator utilizing PVC. a) Left panel: the appearance of the setup built with a timer IC and PVC, connected to an electric buzzer. Right panel: the relation between the capacitance and the generated electric pulses from the oscillator in the initial state. b,c) Upper panels: PVC upon UV/VIS illumination, shown with an expected alteration in the capacitance and the generated pulses. Bottom panels: evolution of the output as a function of illumination time. Experimental condition: $T = 55.6 \text{ }^\circ\text{C}$, UV (365 nm , $I = 2.3 \text{ mW cm}^{-2}$), Vis (450 nm , $I = 3.0 \text{ mW cm}^{-2}$).



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03

Lattice orientation of cholesteric blue phases imposed by planar surface anchoring

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Cholesteric blue phases (BPs) are complex three-dimensional structures that a highly chiral liquid crystal can exhibit. They are composed by an array of double twist cylinders (in which orientational order is twisted along all directions perpendicular to the cylinder axis) interwoven by line defects of orientational order (disclination lines). In addition to drawing academic interest as frustration-induced order, BPs have been gaining greater attention because of their potential for practical optical applications including light manipulation making use of chiral submicron periodic structures. However, preparation of large-scale monodomain BPs, necessary for applications, is still a challenge. Recent attempts to obtain monodomain BPs rely on rubbed or patterned surfaces, and the understanding of the behavior of BPs in contact with such surfaces is important. Motivated by a recent experimental study clarifying the lattice orientation of BPs imposed by planar surface anchoring [1], we numerically investigate the behavior of BPs in contact with surfaces enforcing planar anchoring.

Our work uses the Landau-de Gennes theory describing the orientational order of the liquid crystal by a second-rank tensor χ_{ij} and the free energy of the liquid crystal as a functional of χ_{ij} with the bulk and the surface contributions. We focus on the following two cases that are commonly realized in experiments: (i) Blue Phase I (BP I) with its (110) plane parallel to the surfaces, and (ii) Blue Phase II (BP II) with its (100) plane parallel to the surfaces. By systematically evaluating the free energy as a function of the angle θ between the lattice axis of BPs and the easy axis of the surface anchoring, we find that in both cases BPs adopt a specific lattice orientation with a single value of θ , and that the anchoring strength on the order of 10^{-5} J m^{-2} is sufficient to lock the lattice orientation. Our numerical findings are consistent with some of the experimental observations, but not all; the experimental θ depends on the thermal process. Therefore, how BPs orient in response to the treatment of the surfaces still needs further theoretical and experimental studies.

This work is supported by JSPS KAKENHI (Grant numbers: JP17H02947 and JP21H01049) and JSPS Bilateral Joint Research Project between Japan and Slovenia.

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04

Theory of Aqueous Liquid Crystals

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Recently water-dispersible liquid crystalline molecules have received significant attention as new materials for not only display but also food science and biomaterials [1]. Typical examples are chromonic liquid crystals, graphene oxide, and surfactant-covered carbon nanotubes, etc.

In this paper, we introduce a mean-field theory to describe nematic (N) ordering and phase separations in aqueous low-molecular-weight liquid crystalline molecules (LCs). We consider a physical bonding (or a hydrogen bonding) between LC and solvent (water) molecules. Our theory predicts that the hydrogen bonds between the unlike molecules stabilize the N phase and take place novel phase separations on the temperature-concentration plane. As increasing the number of functional groups on the LCs, capable of forming the physical bond with the solvent molecule, the stable N phase region is broadened on the phase diagram. We also find a miscibility window of the N phase and closed-loop coexistence curves with upper and lower critical solution temperatures near the N phase [2].

Figure 1. Schematic representation of the dispersion of the m -clusters with solvent molecules. Each LC molecule carries $f(=4)$ identical functional groups capable of forming a physical bond with the solvent molecule. The open circles show the free solvent molecules and the closed circles show the solvent molecules bonded on the functional group. The number on the LC molecule is the number m of the bonded solvent molecules, which become a part of the LC molecules.

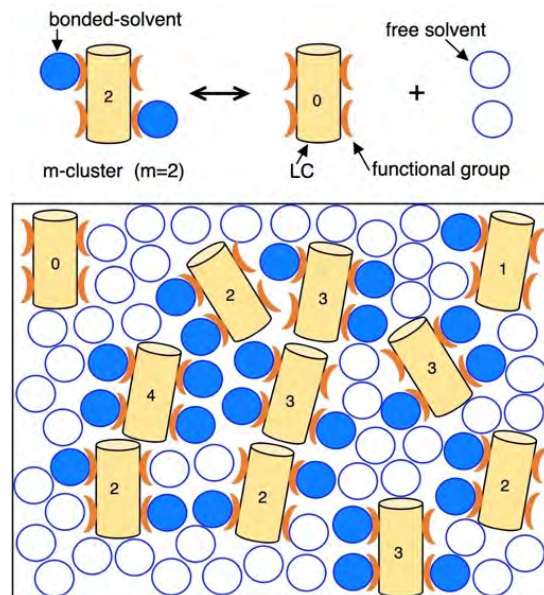


Fig.1

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05

Flow induced deformation and orientation of micelles in phantasmagoric nematic liquid crystals near continuous I-N phase transition

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In this paper, we investigate the flow induced the shape deformation and the orientation rotation motion in phantasmagoric nematic liquid crystals in the vicinity of the continuous Isotropic (I) - Nematic (N) phase transition. We measure the response of the flow birefringence under applying sinusoidally modulated flow field. For the phantasmagoric liquid crystal systems, shape transformation of the amphiphilic supermolecules promotes I-N phase transition in addition to the order-disorder transition for the orientation of super molecules. In the isotropic phase, order parameter fluctuation can be observed by the depolarized VH scattering. It is evident that the relaxation frequency linearly diverges toward I-N phase transition temperature (T_{IN}), and the critical temperature T^* is almost equivalent to T_{IN} , $T_{IN}-T^*\sim 0$. It is confirmed that the I-N transition shows the continuous phase transition. On the other hands, even in the isotropic phase, the susceptibility of the external field should diverge near continuous I-N phase transition, due to the large order parameter fluctuation and the critical slowing down near T_{IN} .

External flow should couple with the orientation and shape of the micelles. It is obvious that the large flow induced birefringence can be widely identified until $T - T_{IN} \sim 10^\circ\text{C}$. We measure the response time in the flow birefringence under the sinusoidally modulated flow field and discuss the origin and dynamics of the flow induced birefringence. The response time of the flow birefringence is 3-order slower than the relaxation time of the order parameter fluctuation observed in DLS measurement even in the isotropic phase near continuous I-N transition point.

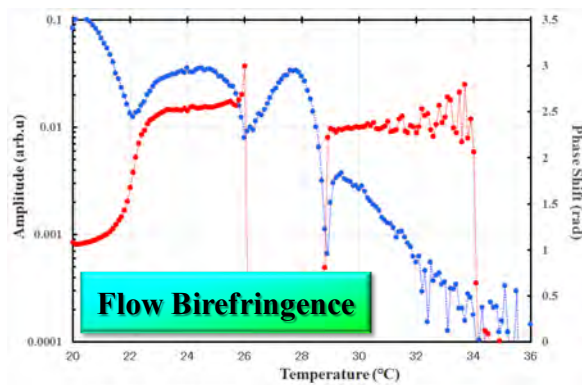


Fig.1 Temperature dependence of flow birefringence for $\phi=8\%$ and $\alpha=2.2$. Blue circles indicate the amplitude, and red circles phase shift.

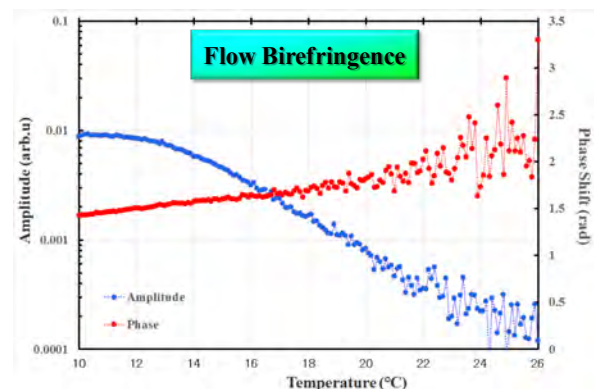


Fig.2 Temperature dependence of flow birefringence for $\phi=8\%$ and $\alpha=2.6$. Isotropic phase is always stable, but the critical phenomena evidently observed in whole temperature range.

06

Formation of surface structures by photoinduced molecular diffusion

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Photocontrol of surface topology is key to developing highly functional materials and devices in the field of optics, mechanics, and electronics. Although many researchers have reported photoinduced surface structures [1], it is still challenging to induce surface structures without complicated processes using photoresponsive dye molecules and polarized light. Recently, we revealed that a surface relief structure was formed in the polymer film by patterned photopolymerization, based on a concept of molecular diffusion [2]. Here, we investigated the detailed relationship between the surface relief structure and molecular alignment induced by the molecular diffusion.

A mixture composed of an anisotropic monomer, a crosslinker, and a photoinitiator was prepared as a photopolymerizable sample. The solid-state sample mixture was melted above its isotropic temperature (150 °C) on a hot stage, injected into a glass cell (thickness, 2-3 μm), and cooled down to the photopolymerization temperature of 100 °C. The photopolymerization was conducted using patterned UV light with bright and dark stripes (Fig. 1). The confocal laser micrographs showed formation of surface relief structures in the obtained film. Polarized optical microscopy revealed that birefringence was highest at the boundaries between bright and dark areas. According to the comparison of the degree of the molecular alignment with the surface relief structure, the shape and height of the formed structures were affected by molecular motion induced by patterned photopolymerization [3].

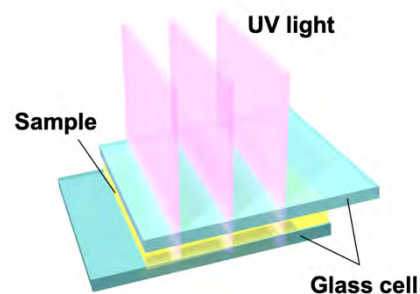


Fig. 1 Schematic of patterned-photopolymerization

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Soft Elasticity Effects in Nematic Elastomers Revealed by Various Types of Deformation

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Ideal nematic elastomers (NEs) are theoretically expected to exhibit a liquid-like behavior with zero shear modulus due to the coupling of nematic director and rubber elasticity, which is termed “soft elasticity”. [1] Many real NEs have finite shear modulus, but they show a low plateau stress over finite strain range when stretched normal to the initial director, or when polydomains are uniaxially stretched, [2] both of which have been regarded as the indication of soft elasticity effect. Most of the earlier studies have investigated the mechanical behavior of the NEs exclusively by uniaxial stretching normal to the initial director. However, the characterization using uniaxial stretching provides a limited basis to understand the large deformation behavior of NEs, because it is only a particular one among all accessible deformations. We characterize the large deformation behaviors of NEs using various types of deformation including uniaxial stretching at oblique angles relative to the initial director, biaxial stretching and torsion.

Figure 1 shows the nominal stress – stretch relations of a monodomain NE with planar alignment in uniaxial stretching at various stretching angles (θ). The θ is the angle between the uniaxial stretching and initial director. The stress-plateau width (λ_E) increases with θ whereas the plateau vanishes at $\theta < 24^\circ$. The θ dependence of λ_E is successfully explained by a scenario of the soft elasticity theory, i.e., a planar stretching via director rotation. [3]

In biaxial stretching of polydomain NEs, we observe that the two orthogonal true stresses are equalized independent of strain biaxiality. [4] This unusual feature means that the polydomain NE sheet undergoes the shape change at no extra energy cost under the constant true biaxial stress conditions, indicating a liquid-like behavior. We also observe a pronounced effect of soft elasticity in a polydomain NE cylinder under torsion.

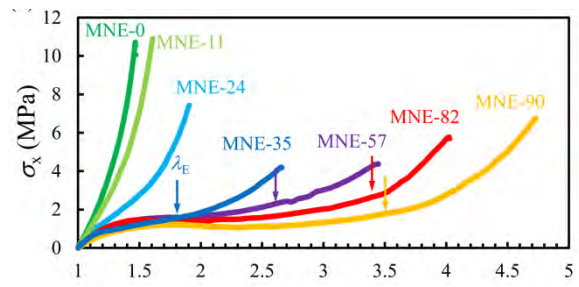


Fig. 1. Uniaxial nominal stress versus stretch for a monodomain NE with planar alignment at various stretch angles.

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Marangoni convection driven by temperature gradient near an isotropic-nematic transition point

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When temperature or concentration field is not uniform in a fluid surface, a surface tension gradient is generated. This often results in the formation of a convective flow, which is called Marangoni convection. In the convection due to the non-uniformity of the temperature field, the flow from high to low temperature region has been typically observed in liquid surfaces, because the surface tension increases with the decrease of the temperature in typical liquids. This tendency, however, is not necessarily observed in the liquid crystalline (LC) materials, and especially in the temperature near isotropic (I)-nematic (N) transition point, a singular behavior with a drastic decrease and increase of the surface tension has been reported [1]. Hence, we considered that the flow from high to low-temperature region would not be always driven in the surface of LC. To clarify this, in this study we observed the Marangoni flow in the N LC material of 7CB under the temperature gradient. The gradient was applied by the experimental system illustrated in Fig.1, and the flow field near the air interface was analyzed by fluorescence photo-bleaching method as shown in Fig.2. In the N phase, the flow away from the interface was observed in low-temperature side, and the flow toward the interface in high-temperature side. This flow distribution indicates the existence of the convection near the air interface. On the other hand, in the I and the coexistence state of the I and N phases, the flow direction was opposite from that in the N phase. This difference in the flow direction is resulted from the singularity of the temperature dependence of the surface tension near the I-N transition point. The detail about the relation between the surface tension gradient and the flow will be reported in presentation.

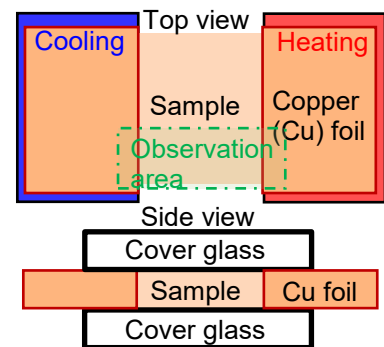


Fig.1 Schematics of experimental setup.

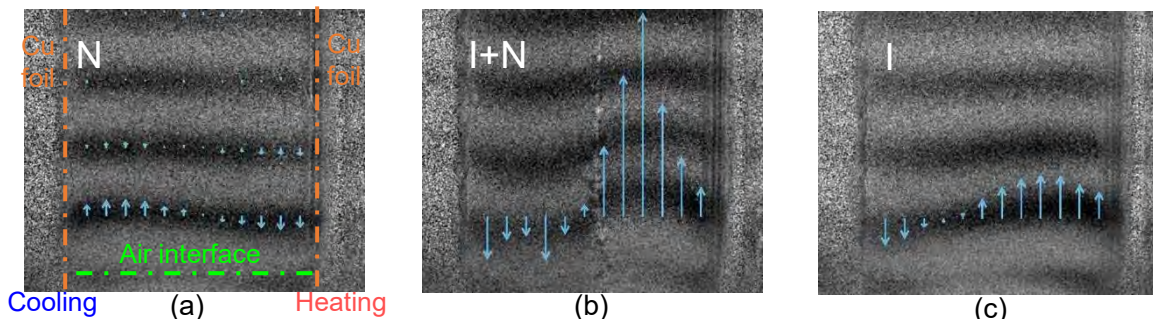


Fig.2 Measurement results of flow field by photo-bleaching method. The field is indicated by aqua arrows, and the temperature gradient was set to be 35K/mm. The dye of C6-NBD Ceramide was added for the fluorescence observation. (a) N phase (Averaged temperature in the sample T_0 is 36°C). (b) I-N coexistence phase ($T_0=42^\circ\text{C}$). (c) I phase ($T_0=47^\circ\text{C}$).



Development of novel spectroscopy systems to directly observe microscopic translational molecular dynamics in various liquid crystal phases

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Unique properties and functionalities of liquid crystal phases originate from the coexistence of microscopic molecular orders and molecular mobility. Understanding the microscopic molecular dynamics in association with the order is highly important to microscopically elucidate the macroscopic functionalities. Recently, we developed synchrotron-radiation-based quasi-elastic scattering spectroscopy using multi-line time-domain interferometry (TDI) of Mössbauer gamma rays [1]. The multi-line TDI system allows us to determine the intermediate scattering function in the angstrom and nanometric spatial scales and in time scales between few nanoseconds and sub-microsecond, in which molecular translational motions occur in many liquid crystal phases [1]. So far, molecular dynamics in nematic, cholesteric, smectic A, and cholesteric blue phases has been measured [2-4]. In this presentation, we introduce new application results on molecular translational dynamics studies for smectic B and smectic C phases. In both cases, we could selectively determine the time scales of the microscopic molecular translational motions across the smectic layer and inside of the layer, respectively. It was revealed that the molecular dynamic behaviour in smectic B phase is apparently different from these in smectic A and C phases due to the creation of the bond-orientational order in smectic B phase.

In this presentation, we also introduce another new synchrotron-radiation-based spectroscopy system using nuclear Bragg monochromator. Recently, it was demonstrated that the new system allows for the determination of molecular dynamics between 100 picoseconds and 10 nanoseconds. It is expected that the molecular dynamics can be measured in most of liquid crystal phases and high-temperature isotropic phases by combination of the two spectroscopy systems. We discuss the possible applications of these new dynamics measurement systems on liquid crystal phases. This work was supported by JST CREST Grant Number JPMJCR2095, Japan.

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Ferroelectric liquid crystals are partially ordered anisotropic fluids with ferroelectricity. Although ferroelectric nematic phase composed of rod-like molecules with electric dipole was predicted theoretically, its experimental realization has not been reported for a long time. Recently, some experimental groups have succeeded in synthesizing molecules which possibly show ferroelectric nematic order [1-2].

In this work, we numerically study ferroelectric nematic phase formation with a simple molecular model. A conventional nematic phase does not have polar order even if the rod-like molecules have the electric dipoles along their long axes. It is considered that a pair of the rod-like molecules are placed side-by-side and oriented toward the opposite directions because of the dipole-dipole interaction. This local alignment cancels the polar order.

We employ rod-like molecules with a bulky central part. We expect that the side-by-side alignment is avoided by the bulky central part and the polar order is not canceled. Each molecule is given as an array of the Lennard-Jones particles. It is known that the system exhibits isotropic-nematic-smectic transitions by changing the temperature and/or the packing fraction when the molecule is long enough. In our model, the molecules are composed of eight particles, and the central two particles are enlarged by the ratio θ . Each rod-like molecule has an electric dipole along its long axis at the center by introducing the positive and negative electric charges to the central two particles. We carry out molecular dynamics simulation under the NVT ensemble. Although spontaneous orderings have not been observed, our molecular systems possibly show ferroelectric nematic order by employing aligned initial conditions. Figure 1

shows the dependences of the nematic order P_2 and the polar order P_1 against the packing fraction ρ . The particle size ratio is changed from $\theta = 1.0$ to $\theta = 1.5$. In the system of $\theta = 1.5$, we observed the ferroelectric order ($P_2 > 0, P_1 > 0$) when the packing fraction is large. This work was supported by JST, CREST Grant Number JPMJCR2095, Japan.

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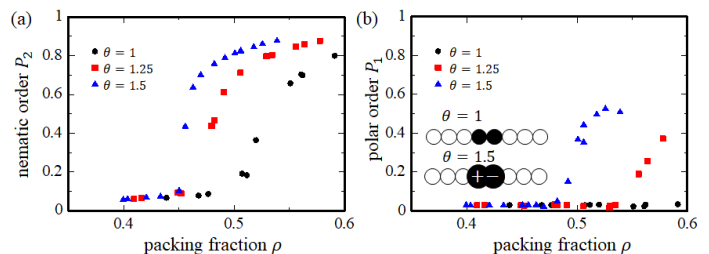


Fig. 1 Dependences of the (a) nematic P_2 and (b) polar order P_1 on the packing fraction ρ . The particle size ration θ is changed.

Controlling the temperature range of ferroelectric nematic phases based on eutectic phenomenon

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A fluorinated liquid crystal with 1,3-dioxane skeleton (C3DIO) exhibits ferroelectricity and abnormally large dielectric constants reaching 10,000 in the ferroelectric nematic phase (N_F phase which used to be called MP phase)[1]. However, the N_F phase of

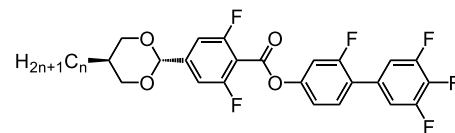


Figure 1 Chemical structure of C_nDIO .

C3DIO is mainly a monotropic phase which appears during the cooling process and is therefore thermodynamically metastable, while the temperature range of the thermodynamically stable enantiotropic N_F phase is very limited. Therefore, an enlargement of the temperature range of the thermodynamically stable N_F phase is strongly desired. Here, we present an enlargement of a thermodynamically stable temperature range of N_F phase by freezing point depression using eutectic phenomenon appeared through mixing a series of fluorinated liquid crystals (C_nDIO ($n = 1, 2, 3, 4$), Fig. 1) with different alkyl chain length.

Dielectric properties of C2DIO/C3DIO and C1DIO/C4DIO mixtures with different mixing ratios were measured in homeotropic alignment ITO glass cells at different temperatures in heating processes.

Temperature dependences of dielectric constants in C1DIO/C4DIO mixture with different mixture ratio are shown in Fig.2. Liquid crystal phases or coexisting phases showing a relative dielectric constant of 1,000 or more at 100 Hz were regarded as an N_F phase. In the C2DIO/C3DIO mixture, a temperature range of N_F phase of C2DIO/C3DIO=70/30 wt. ratio mixture was extended to 17 °C. For the C1DIO/C4DIO mixture, melting point of C1DIO/C4DIO=20/80

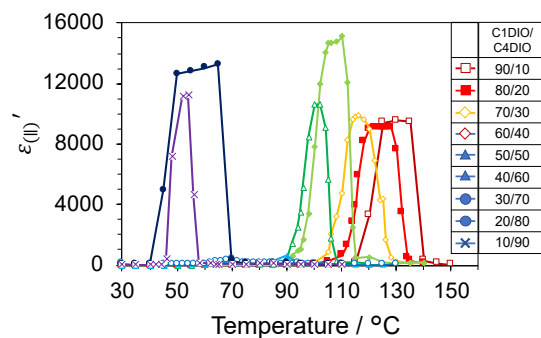


Figure 2 Temperature dependences of dielectric constants in C1DIO/C4DIO mixture with different wt. ratio.

wt. ratio mixture was reduced by 21 °C from that of pure C4DIO, and the temperature range of the N_F phase was extended to 20 °C.

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O12

Development of weak anchoring surface for nematic liquid crystal reorientation modified with silane coupling reagents

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Introduction

With the development of communication devices, the number of personal smartphones and PC devices with liquid crystal (LC) displays is rapidly increasing, thus decreasing of the power consumption of LC displays is crucially demanded. In general, the LC contacting with the substrate surface is strongly anchored to the surface, resulting inhibition of reorientation of LC by electric field, therefore, the development of weak anchoring cells is a promising way to decrease power consumption. In the present work, we performed weak anchoring of LC and glass substrate by modification of silane coupling reagents and examined the effect of chemical structure of silane coupling reagents.

Experiments

Ethyltrimethoxysilane (**C2**) and 3-Glycidyloxypropyltrimethoxysilane (**GTMS**) were used for surface modification of glass substrates. Silane coupling reagents and water were dissolved in ethanol with pH controlled by acetic acid or hydrochloric acid. The solution was stirred at room temperature for 12 hours to hydrolyze the methoxy groups of silane coupling reagents, followed by stirring at 130 °C for 5 hours for polymerization. The resulting liquid was diluted by cyclopentanone, and glass substrates were spin coated with the solution at 3,000 rpm. After the coating, the substrates were heated under reduced pressure at 130 °C for 1 hour. A mixture of liquid crystal (JC1041XX: 5OCB = 1: 1 (w/w)) was filled in a 10- μ m thick cell composed of the modified glass substrates. The anchoring effect were evaluated by polarizing optical microscopic (POM) observation under magnetic field with azimuth rotation.

Results and Discussion

From POM observation, the LC in both silane coupling modified and unmodified cell showed planar orientation at a temperature range of nematic phase. The director of LC in **C2** modified cell rotated accordingly with the rotation of magnetic field, while no change was observed in unmodified glass cell. Furthermore, we observed LC in the cell modified with **C2** hydrolysis rate of 30% (**C2-30**) and 100% (**C2-100**) at the first step of synthesis to compare the effect of chemical structures. The samples of **C2-30** and **C2-100** highly and poorly responded to the magnetic field, respectively, indicating that the remaining C-O bonds reduced the anchoring strength. LC cells were also synthesized from **GTMS** with C-O bond at side chain to see the effect of C-O bonds for anchoring. This **GTMS** modified cell showed weaker anchoring than the **C2** modified cell, supporting that the silane coupling reagents with C-O bonds is responsible for reducing the anchoring strength between LC and glass substrate.

Acknowledgment This work was supported by JST-CREST Grant Number JPMJCR1424 and JSPS Grant Number 18H03920.

Superior electro-optical response in Organic molecules doped in the nematic liquid crystal device: present status and future perspectives.

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Abstract: A superior electro-optical response has been achieved by doping organic N-benzyl-2-methyl-4-nitroaniline (BNA) and Morpholinium 2-chloro-4-nitrobenzoate (M2C4N) in nematic liquid crystals (LCs) reported in this study. The BNA and M2C4N-doped LC cells have a five-fold fall time (τ_{off}) and threefold faster than the pristine LC cell, respectively. The possible reason behind the τ_{off} could be the remarkable decrements in the rotational viscosity and threshold voltage by 45% and 27%, respectively, and a strong additional restoring force resulted from the spontaneous polarization electric field BNA. Contrastingly, the dielectric anisotropy ($\Delta\epsilon$) of the LC mixture increases by 17% and 7%, respectively, with M2C4N and BNA dopants. M2C4N dopant induces a tremendous dielectric anisotropy because the phenyl-amine/hydroxyl in M2C4N induces a robust intermolecular interaction with LCs. Additionally, BNA dopant roots a strong absorbance near the wavelength of 400 nm that filters the blue light. Density Functional Theory(DFT) calculations explain the results mentioned above and confirm that BNA and M2C4N increase the dipole moment, polarization anisotropy, and hence $\Delta\epsilon$ of LC mixture. These results indicate that M2C4N doping can be used to develop a high $\Delta\epsilon$ of LC mixture, and BNA doping is suitable to fabricate a fast response and potentially anti-ultraviolet (UV) applications, such as anti-UV smart windows, glasses, and promising application fields of LC devices and displays.

Keywords: Organic molecule; liquid crystal; fast response time; density functional theory(DFT).

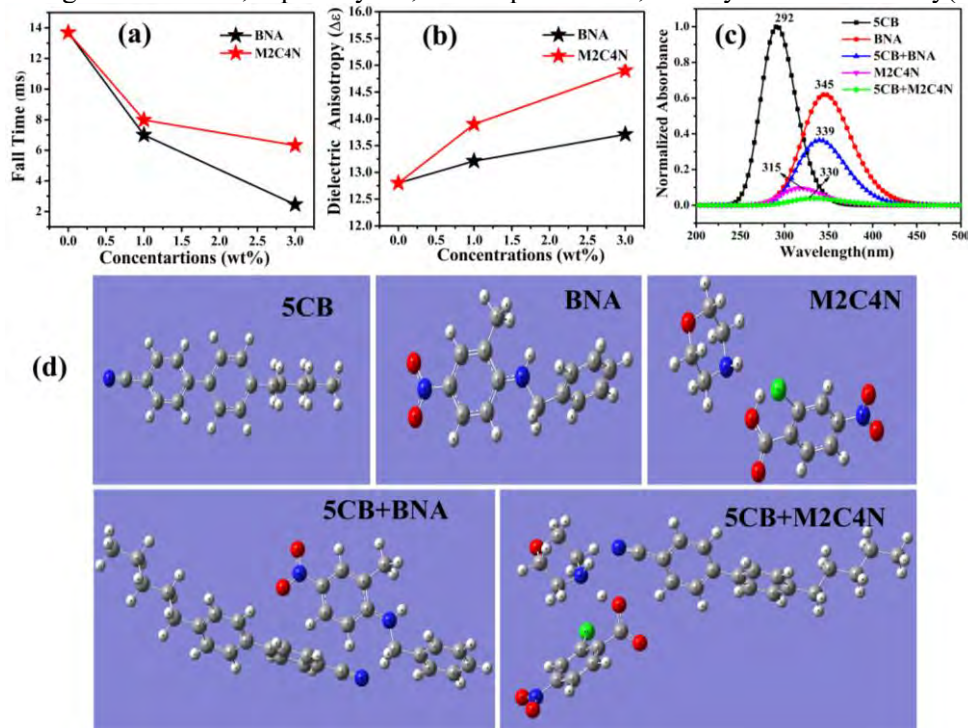


Fig.1. (a) τ_{off} , (b) $\Delta\epsilon$ as functions of BNA and M2C4N-doped LC cells at 1 kHz, respectively, (c & d) Theoretical absorption spectra and optimized geometries for 5CB, BNA, M2C4N, 5CB + BNA, and 5CB + M2C4N by Density Functional Theory (DFT).

Reference:

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Large aperture liquid crystal lens using the hybrid alignment layer with titanium dioxide nanoparticle and polyimide composite

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ABSTRACT

The hole-patterned LC lens is an attractive option because it possesses excellent tunable focal length capability, realizes any sizes of optical aperture easily and only requires simple fabrication and addressing scheme [1]. The hole-patterned liquid crystal (LC) lens has been fabricated using the hybrid alignment layer with the rutile titanium dioxide (TiO₂) nanoparticles (NPs)- coated polyimide (PI). Electro-optical properties of the pure PI and hybrid PI- LC lenses such as the TiO₂ NPs-doped concentration of the hybrid alignment was set to 0.1, 0.5 and 1.0 wt% are demonstrated and compared. The interference fringes and focal lengths of the fabricated LC lenses are measured to compare the operation voltage and lens power. The wavefront error and the full width at half maximum (FWHM) of the focusing spot of the LC lens are determined to analyze the lens and focusing qualities. The response time of the LC lens is determined by a transient transmission measurement. The imaging performance of the LC lens is recorded.

The experimental results introduce the utilization of the hybrid PI-layer decreases the turn-off time of the LC lens by ~2x, due to the increased interaction between the LCs and alignment layer. The appropriate TiO₂ NP-doped concentration also decreases the turn-on time of the LC lens by ~4x in comparison with the pure PI-LC lens. Notably, it does not degrade the focusing and lens qualities. However, the hybrid alignment layer decreases the lens power and increases the operating voltage of the lens, possibly due to the rough PI surfaces from TiO₂ NPs aggregation and the increased screen effect. The imaging performance of the hybrid PI-LC lens has been demonstrated practically through focusing on three objectives at different distances. While varying the applied voltage, the lens cell has functioned on three objects at different positions with tunable focusing.

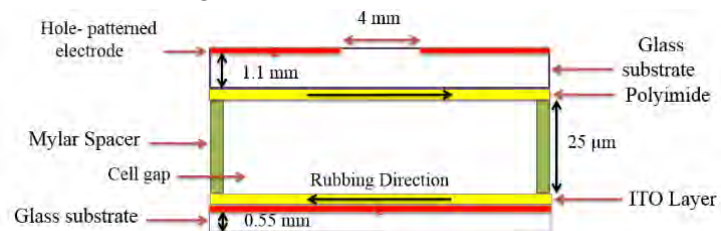


Fig. 1. Schematic of the hole-patterned LC lens structure.

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Bragg diffraction of light at oblique helicoidal cholesteric structures

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Chiral mixtures with a small bend elastic constant exhibit a peculiar oblique helicoidal cholesteric (Ch_{OH}) state when acted upon by an electric field [1-2]. Both the period P and the cone angle θ of the Ch_{OH} are tunable by the applied field in a broad spectral range. The Ch_{OH} state forms in chiral mixtures composed of flexible dimers which exhibit bent molecular conformations (see, e.g. [3]). At the normal incidence of light, reflection from the Ch_{OH} half-pitch $P/2$ is used for the first in-situ measurements of bend elastic modulus of the chiral material [4]. At the oblique incidence of light, reflection from two periodicities associated with the full P and the half-pitch $P/2$ is tunable by the electric field and the angle of incidence. The full and the half-pitch bandgaps have distinct polarization characteristics, Fig.2. The work is supported by the NSF grant ECCS-1906104.

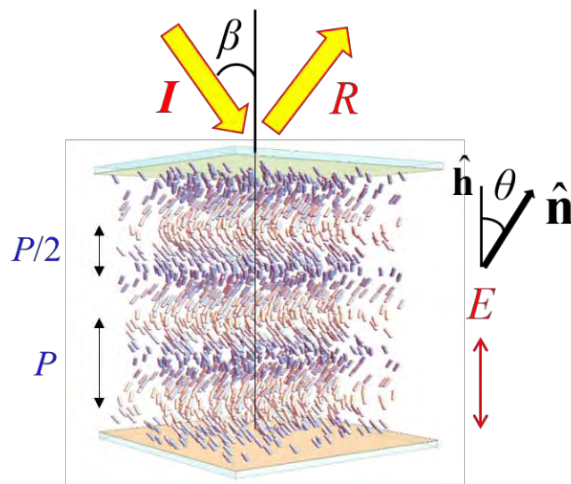


Fig. 1. Oblique helicoidal cholesteric structure with the pitch P and the cone angle θ tunable by applied electric field \mathbf{E} . At the oblique incidence of light, two periodicities associated with the full P and the half-pitch $P/2$ are observed in reflection. Notation for angles: β is the angle of incidence, θ is the cone angle.

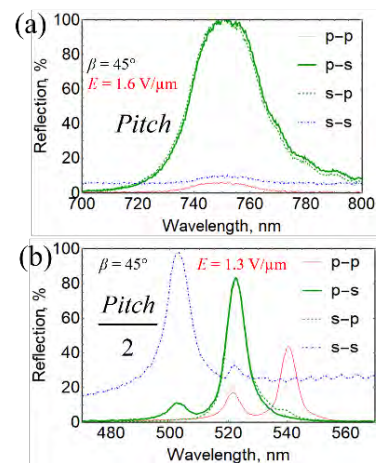


Fig. 2. Polarization characteristics of reflection from (a) the full P and (b) the half-pitch $P/2$ in the planar Ch_{OH} cell, $d = 23 \mu\text{m}$, at the angle of incidence of polarized light $\beta = 45^\circ$.

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The Two-dimension Diffraction and the Milky Haze of a Large-scale Topological Defect Array in Nematic Liquid Crystal

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Abstract: Large array of topological defects in nematic liquid crystal (NLC) can be applied as smart windows. The haze is a mixture of three optical effects, diffraction, scattering of light, and lensing effects. The diffraction and lensing effects are significant when the defects are periodically arranged. Sometimes the sharp glitter due to the diffraction and the split multiple image due to lensing may lead to uncomfortable visual experiences. The scatter generates the soft, milky feature which is friendly to the viewer. Therefore, in this research, large, dense, messy topological defect network is generated in NLC test cell. The director field and diffraction pattern are analyzed carefully. We developed new experiment and analysis methods to distinguish the glitter of diffraction and milky texture of scattering. Our new device may lead to smart windows with better visual experiences. In addition, topological defect arrays have innovative applications, such as optical vortex generation and phase gratings. It can be used for dynamic beam steering, which is essential for holography [1].

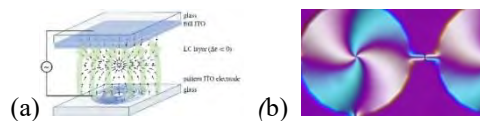


Figure 1: NLC operated by vertically aligned mode (a) The test cell structure. (b) Each pad pixel has one defect which topological charge of +1 at the center and one hyperbolic hedgehog with topological charge of -1 between the pads

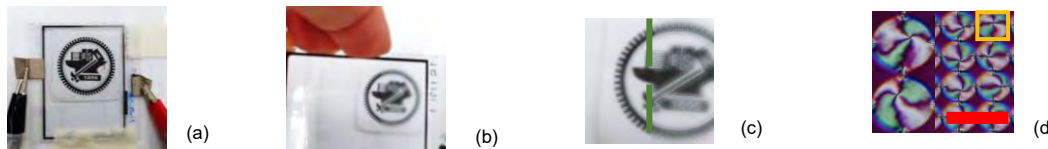


Figure 2: (a) test cell test cell lay on the picture with applied voltage (b) the test cell lifted (c) the visual blur caused by different pixel size (d) POM image of the test cell

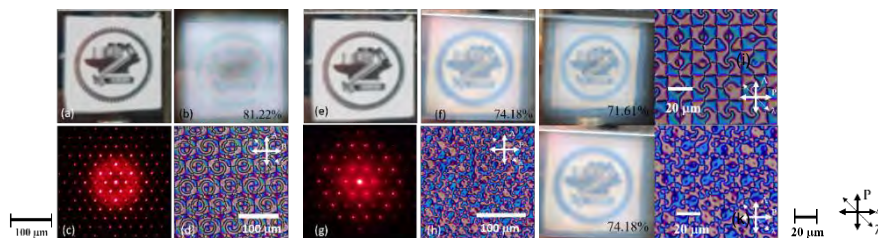


Figure 3: optical and visual effect vs. pixel size

(a-h) and lattice structure (i-l) with annulus shaped electrode. (a-d) 50 μm pixel; (e-h) 20 μm pixel; (i)(j) square lattice; (k)(l) hexagonal lattice. (a)(e) test cell without voltage (b)(f)(i)(k) test cell with 16 V (c)(g) diffraction pattern (d)(h)(j)(l) POM image of AC voltage applied.

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Preferred Photo-Induction of $Ia3d$ Cubic Phase in ANBC-22/Azo Binary MixtureShoichi Kutsumizu^{1*}, Tomonori Kondo¹, Masamichi Ogawa¹, Yuka Mizutani¹, Yohei Miwa¹, Yasuhisa Yamamura², and Kazuya Saito²¹ Department of Chemistry and Biomolecular Science, Faculty of Engineering, Gifu University, Japan² Department of Chemistry, Faculty of Pure and Applied Sciences, University of Tsukuba, Japan

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Thermotropic bicontinuous cubic (Cub) and smectic C (SmC) liquid crystalline (LC) phases form three-dimensionally (3D)- and one-dimensionally (1D)- organized packing structures, respectively. LC states often change their organizations in response to various external stimuli such as temperature, UV light, and so on. One of our main interests is to control the transformation from 1D SmC to 3D Cub structures using light because such transformations accompany dramatic changes in optical and mechanical properties [1,2]. The gyroid-type $Ia3d$ structure (Fig. 1a) is a famous Cub organization seen in a wide range of materials. In this organization composed of seemingly rod-like but actually anti-spindle-like molecules, packing frustration of such molecules is essential for the control of the transformation.

The system examined in this study comprises two components, an $Ia3d$ Cub LC molecule (here denoted ANBC-22, the upper part of Fig. 2) and an azobenzene molecule of a similar structure (Azo, Fig. 2). The system exhibits a SmC phase for all Azo amounts, and just above the phase, an $Ia3d$ phase is formed at 19 mol% Azo while an ' $Im3m$ '-like chiral cubic phase (Fig. 1b) appears at 9 and 15 mol% (see the lower part in Fig. 2). UV-irradiation of the SmC phase of these three mixtures induced the $Ia3d$ cubic phase (Fig. 3 for 15 mol%). Since the *trans-cis* photoisomerization of Azo molecules increases the system's entropy, the transitions seem to mimic being heated. What to note here is, however, at 9 and 15 mol% of the Azo, the induced phase is not the actual high-temperature phase. The mechanism will be proposed and discussed.

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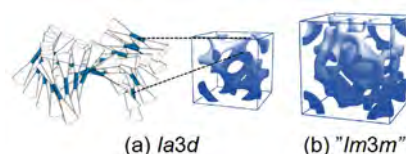
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Fig. 1 Packing structures of two Cub phases, where molecular core regions are painted in blue. In (a), helical arrangement of anti-spindle-like molecules is also illustrated.

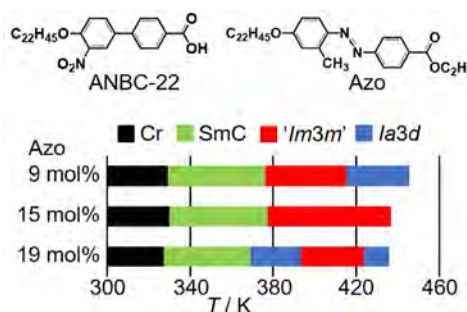


Fig. 2 (Upper) Chemical structures of two components constructing the binary mixture examined. (Lower) Transition schemes of mixtures containing three different Azo amounts.

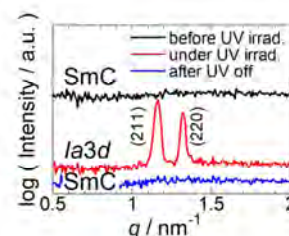


Fig. 3 Photo-induction of $Ia3d$ phase as seen from X-ray diffraction for 15 mol% Azo mixture at 369 K ($q=|\mathbf{q}|$; \mathbf{q} , scattering vector).

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Cholesteric phases have one-dimensional helical structures. A cholesteric layer between two parallel interfaces with tangential molecular alignment affects visible lights propagating parallel to the layer normal. When the helical pitch is comparable to the light wavelength, the light with the same helicity as the cholesteric helical structure is fully reflected. The cholesteric phase is easily curved; e.g., a cholesteric shell between two tangentially aligning interfaces can work as three-dimensional chiral photonic crystals [1]. Microfluidic devices enable us to fabricate mono-dispersed droplets and core-shell emulsions. We have developed microfluidic techniques to fabricate and manipulate capsules with high-melting-point and/or high-viscosity materials [2]. Cholesteric shells with a dye in the inner phase exhibit omnidirectional laser action [1,3]. Besides, cholesteric shells with luminol in the inner phase work as chemiluminescence sensors for hydrogen peroxide [4].

Here we present the techniques and materials for the manipulation of cholesteric shells. First, we developed the synthesis method of ordered porous film to immobilize cholesteric shells, as shown in Fig. 1 [5]. We found that a topologically essential defect in the cholesteric shell leads to the self-assembly of particles dispersed in the shell. It can induce “mutualistic self-assembly” in the cholesteric shell with magnetic nanoparticles [6]. We focused on magnetically controllable droplets of liquid crystalline nitroxide radicals (LC-NRs) even without any metal elements [7,8]. We succeeded in the synthesis of a new low-melting-point LC-NR showing a cholesteric phase. Owing to the temperature dependence of selective reflection that the cholesteric phase shows, the cholesteric shells with the new LC-NR work as magnetically migratable temperature sensors.

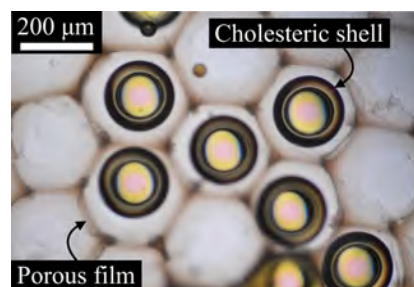


Fig. 1 Photograph of cholesteric shells immobilized in porous film.

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Quantification of internal strain in bent silicone elastomers via selective reflection of cholesteric liquid crystals

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Flexible devices composed of soft materials, such as polymers, elastomers, and liquid crystals have attracted a lot of interest as a next-generation device. An obstacle for developing flexible devices is deterioration of their performance during bending. Enhancing bending durability of devices requires appropriate design based on bending behavior of soft material components. Although many researches have reported on the bending behavior of soft materials [1-3], internal strain in bending soft materials has not been measured due to experimental constraints. Measuring internal strain is essential to understand the bending behavior, leading to optimized device design.

To measure internal strain in bending soft materials, we focus on selective reflection of a cholesteric liquid crystal (CLC). The selective reflection arises from a self-organized helical structure of CLC, according to $\lambda = nP$ (where λ , n , and P represent reflection wavelength, refractive index, and helical pitch, respectively). Mechanical strain changes the helical pitch (P) and reflection wavelength (λ) [4], thus internal strain induced by bending of soft materials can be evaluated via reflection wavelength change of CLC. In previous work, we fabricated a CLC sensor that can detect internal strain in soft materials [5]. Herein, using the CLC sensor, we quantified internal strain in a bending polydimethylsiloxane (PDMS) film which is a common soft material (Fig. 1). As a result, internal strains in the bending PDMS film were asymmetric: 19% compression and 17% tension occurred in bending outer and inner surfaces, respectively. Surprisingly, large bending of the PDMS film caused 7% compression in the middle of the film thickness where strain was assumed to be zero. This work will help us design flexible devices with high bending durability.

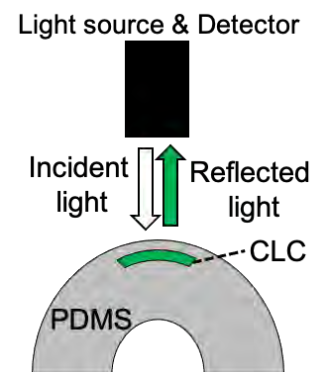


Fig. 1 Measurement of internal strain in a PDMS film via selective reflection of CLC.

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Liquid crystal elastomers, LCE [1], are weakly crosslinked polymer networks with molecular units often called “mesogens” that induce liquid crystal, LC, phases. In the case of nematic LCE, the attractive interaction between mesogens tend to align their long molecular axes along each other. Since the network is crosslinked, the molecular units are not mobile as in the low molecular weight, Mw, nematic LC. Instead, the nematic tensor order parameter couples to the strain field via the polymer network, leading to novel actuation and morphing applications.

Here, we present polarized microscopy study of domain textures of nematic LCE. A polydomain texture is known to spontaneously form upon cooling from the isotropic state, at which the polymerization has proceeded. Unlike the schlieren texture on low Mw nematic LC, the nematic domains cannot coarsen due to the frozen network topology by chemical crosslinks. As a result, the nematic domains with a characteristic length typically of ~ 1 micrometer are stabilized, showing opaque appearance macroscopically due to the multiple scattering from the birefringent domains. It is revealed via polarized fluorescence microscopy that the characteristic domain texture with specific orientational correlation of nematic directors exists (Fig). This qualitative feature has been theoretically expected by Uchida [2] and experimentally revealed here for the first time. This observation method can be useful for characterizations of domain transformation under strain and upon nematic-isotropic phase transition [3,4].

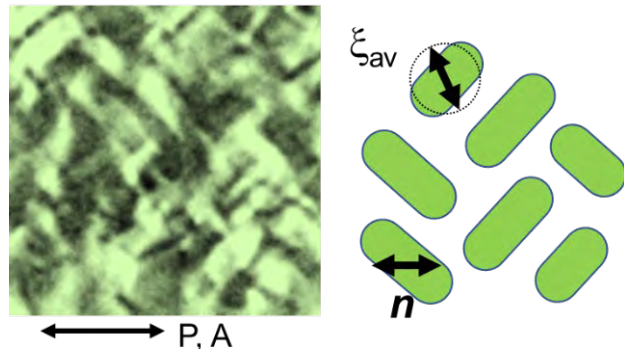


Fig. Polarized confocal fluorescence microscopy image of polydomain nematic LCE ($7.6^2 \mu\text{m}^2$). Domains with n parallel to polarizers (P,A) appear bright according to the dichroism of probe dyes. Note the characteristic orientational correlation in $\pm 45^\circ$ to n and domain size of $\xi_{\text{av}} \sim 1 \mu\text{m}$.

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Acknowledgements: This work was partly supported by JSPS KAKENHI Grant Number 20K20967. The authors are grateful for many useful communications from E. M. Terentjev and M. O. Saed.

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Super-Monodispersed Cholesteric-Liquid-Crystal Polymer Microparticles with Three-Dimensionally Controlled Molecular Orientation

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In spherically confined liquid crystal systems, molecular alignment can be three-dimensionally controlled by surface anchoring. Thus, the liquid crystal microparticles exhibit unique optical properties depending on molecular alignments. In particular, cholesteric-liquid-crystal (Cho-LC) microparticles have drawn attention because of their three-dimensionally controlled helical alignment of mesogens and their optical properties based on the helical alignment. In this work, we developed monodispersed polymer particles composed by the Cho-LC with precisely controlled helical axis orientation. We report that such monodispersed polymer microparticles can be easily prepared by the conventional dispersion polymerization. In the polymerization process, monodomain of Cho-LC was spontaneously formed in the polymer particles. To date, the mechanism for the LC alignment in the polymer particles during dispersion polymerization has been still unclear. Here, we first report the detailed mechanism to form the controlled helical-axis orientation of the Cho-LC in the polymer particles.

We obtained the super-monodispersed Cho-LC polymer particles with a diameter of $2.6 \pm 0.1 \mu\text{m}$. We confirmed that the monodomain of the Cho-LC orientation was formed in the particles. In addition, we succeeded in controlling the orientation of helical axis of the Cho-LC in the particles, and radial helical-axis orientation was formed in the particles. The reporting Cho-LC polymer particles can be easily and efficiently prepared by the common way, dispersion polymerization, which is industrially utilized to manufacture polymer particles. Due to the controlled helical axis orientation of Cho-LC, the particles showed unique optical properties and can have highly potential application as optical materials with recognition abilities of a reflection color and a circular polarization of light.

Unidirectional alignment of polymer-grafted ZnO nanorods in liquid-crystalline polymer films

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Inorganic nanotubes and nanorods have attracted much attention due to their anisotropic properties different from bulk materials. Of these anisotropic inorganic materials, ZnO nanorods show excellent properties, e.g. wide bandgap, thermal and chemical stability, and anisotropic photoluminescence. Controlling the alignment of ZnO nanorods is essential for developing functional devices by utilizing their great potential. Although the alignment control of nanorods has been attempted by many methods [1–3], it is still a challenge to develop sophisticated methods for aligning nanorods over a large area. We have recently proposed a simple method for the unidirectional alignment of ZnO nanorods on an alignment layer [4]. In this method, ZnO nanorods grafted with liquid-crystalline (LC) polymers are aligned cooperatively with surrounding nematic LC molecules. In this study, we demonstrate the unidirectional alignment of polymer-grafted ZnO nanorods in micrometer-thick LC polymer films.

Polymer-grafted ZnO nanorods were mixed with a polymerizable host LC monomer and a photoinitiator, and ultrasonically dispersed. A glass cell was fabricated by adhering a pair of glass substrates coated with an alignment layer. The sample mixture was injected into the cell at its isotropic temperature, cooled down to LC temperature, and subsequently kept for 24 h. The photopolymerization was conducted by the irradiation with ultraviolet (UV) light. The polarized optical microscopy and polarized UV-visible absorption spectroscopy suggested that the nanorods were aligned together with host LC moieties along the rubbing direction of alignment layer. The alignment of the nanorods was also confirmed by transmission electron microscopy. Thus, the unidirectional alignment of polymer-grafted ZnO nanorods in LC polymer films was successfully concluded.

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A calamitic liquid crystal (LC) composed of rod-shaped (calamitic) molecules is known to be transformed into chiral nematic LC with a helical structure upon doping with chiral molecules. The effect of molecular chirality for the calamitic LCs has been well investigated thus far. The helical structures can be also observed in the columnar LCs mainly formed by disc-shaped molecules. While helical columnar LCs have attracted widespread research attention^[1], the correlation between molecular chirality and the helical structure induced is not perfectly understood.

Recently, we found that a type of octahedral metallomesogen molecules, **Ru-C8** (Fig. 1a), having propeller-shaped chirality at the molecular core ($\Delta\Lambda$ chirality) exhibits the hexagonal columnar LC possessing helical structure at room temperature. While the experimental studies revealed that the basic information about the helical structure, its details were clarified by performing all-atom molecular dynamics (MD) simulation. The structure obtained from MD simulation shows that the molecules stack with both the molecular position and C_3 axis rotating along the column axis (Fig. 1b). It indicates that the structure was classified as a hybrid of two major proposed helical stacking types. Further analysis based on the MD simulation results highlights the interplay of the steric repulsion between the chiral cores, π -stacking, and dipolar interaction in the helix formation.^[2] This work was supported by JSPS KAKENHI Numbers JP19H02537 and 19H05718.

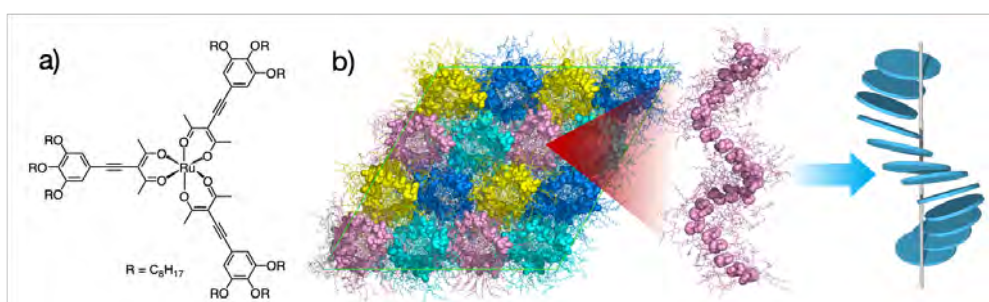


Fig. 1 (a) Molecular structure of Δ -**Ru-C8** and (b) top view (left) and side view of a single column (center) of Δ -**Ru-C8** after 200 ns of MD simulation and schematic of a single column structure

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Patterning the orientation of liquid crystals is a well-known way to encode arbitrary optical phase profile of a geometric nature within physically flat optical elements, which has led to the advent of a nowadays mature liquid crystal photonic technology. One of the simplest practical example is that of geometric phase lenses, whose convergent or divergent nature can be controlled by the handedness of the incident circularly polarized light. Recently, such geometric phase lensing met the nonlinear optics of liquid crystals in the framework of so-called ‘nematicons’, which deal with self-trapping of light as a results of localized optical induced liquid crystal reorientation, as reported by Jisha and coworkers [1]. The latter work discusses the experimental observation of self-trapping of light in a particular light-matter interaction geometry interpreted as being the result of propagation dependent spin angular momentum transfer from light to matter. This eventually leads to alternating in-plane twisting of the director field, thereby realizing a continuum nonlinear version of a previous linear optics, static, discrete one [2]. Still, the direct identification of the basic physical process at play—the nonlinear geometric phase lensing—remains elusive so far. Here we report on its experimental identification based on a simple experimental scheme, which should foster further works on the nonlinear spin-orbit photonics of liquid crystals.

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Terahertz waves have attracted significant attention for many years, mainly because of their promising applications including post-5G communication technologies and terahertz sensing/imaging such as nondestructive testing [1,2]. Liquid crystal (LC) materials are important for tunable devices in terahertz frequency because of their controllability with low drive voltage and low power consumption. However, many LC materials usually exhibit large dichroism in the terahertz region. This dichroism can cause some deformation of polarization state in the LC-based terahertz wave control devices. Previously, we showed that a hydrogen-bonded LC exhibits no dichroism at 2.5 THz [3]. We believe that the hydrogen bonding has important role not only in supramolecular LCs [4] but also in LCs for terahertz devices. In this study, we introduce the hydrogen-bonded LC to the twisted nematic (TN) LC device and investigate operation properties of the TN LC device in the terahertz frequency region.

Fig. 1 indicates the transmittance of TN LC device using hydrogen-bonded LC PC29 (LCC, Yamanashi Japan). The polarization state of terahertz wave passing through the TN device was measured by using an optically pumped gas laser as a continuous wave terahertz source. We placed the TN LC device between two wire-grid polarizers, and the intensity of the terahertz wave was detected by a pyroelectric detector. In fig. 1, we show the terahertz wave transmittance as a function of analyzer angle ψ_A . We set the angle of polarizer $\psi_P = 90$ deg, and the analyzer angle $\psi_A = 0$ deg corresponds to the crossed Nicols. Since the minimal transmission is almost zero, linear polarized terahertz wave was rotated almost 90 deg in the TN LC device keeping its polarization state. Furthermore, owing to the isotropic losses of the PC29 at 2.5 THz, the polarization direction is switched without changing intensity by applying electric field to the TN device (see on state in fig.1). We believe that utilization of the hydrogen-bonded LCs in the terahertz tunable devices is powerful technique for creating applications such as ultra-fast terahertz wireless communication and terahertz imaging.

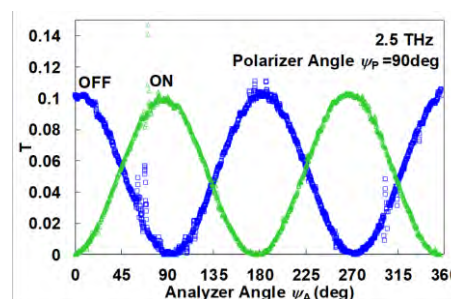


Fig. 1 Transmittance of TN LC device using hydrogen-bonded LC at 2.5 THz.

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Optical waveguides consist of a core and a cladding. To confine the light in the core of the waveguide, it is required that the refractive index of the core n_{core} is larger than the refractive index of the cladding n_{clad} . Nematic liquid crystals (LC) are birefringent and have an ordinary refractive index n_o and an extraordinary refractive index n_e . These indices can be used to build a confined waveguide in the LC bulk.

Using a novel patterned photoalignment technique we demonstrate the ability to define specific alignment orientations at the substrates of a nematic LC cell that will lead to complex 3D configurations. In these configurations there are well-defined regions in which the LC is oriented in such a way that the effective refractive index is higher than in the neighborhood, leading to the occurrence of an optical waveguide. [1]

More specifically, we demonstrate the fabrication of ring shaped regions with rotational symmetry. The 3D configuration is structured in such a way that the optical properties of the waveguide are maintained along the entire ring. [2]

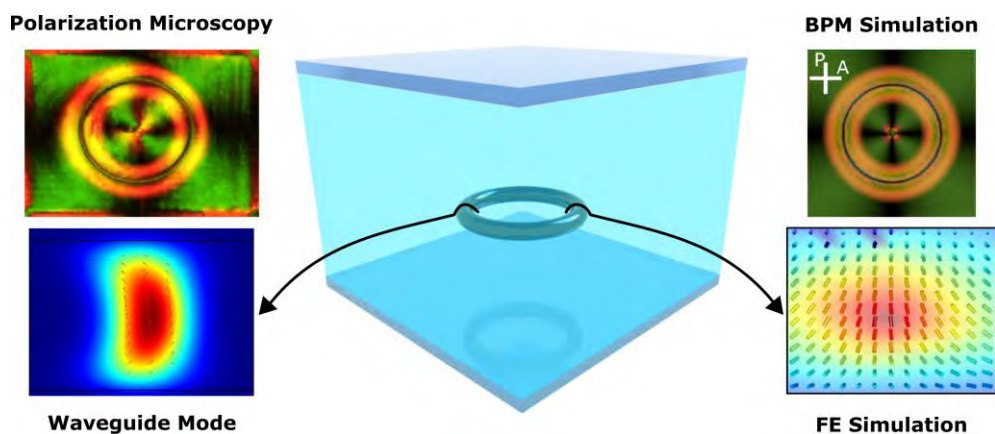


Fig. 1: LC based ring resonator investigated using polarization microscopy, Finite Element (FE), Beam Propagation Method (BPM) and waveguide mode simulations.

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Nematic liquid crystals (NLCs) doped with nanoparticles are used in various photonic systems. Depending on the specific type of nanoparticles (metallic, ferroelectric, dielectric, etc.) as well on their size, shape, concentration or surface coating [1–4] NLCs doped with nanoparticles may exhibit different properties. Metallic nanoparticles show great promise in device applications due to their ability to improve liquid crystal performance. An interesting effect in a composite consisting of gold nanoparticles and a NLC matrix is spontaneous formation of the photonic structure that was observed two years ago [5]. A self-creating one dimensional photonic crystal (Fig. 1) may allow a design as well fabrication of spectrally adjustable optofluidic photonic devices with a broad tuning range. This self-assembly process is completely reversible and can be controlled by modifying diameter of the confining space. Consequently, a period of the observed self-organizing structure depends on the capillary diameter and varies in the range from 10 to even 60 μm . Additionally, the whole system is characterized by high refractive index contrast as the refractive index is being changed between nematic phase and isotropic phases. A significant difficulty in studying properties of the periodic photonic structure is its stabilization in a very narrow temperature range. The use of cross-linking process helps to obtain the stability of the photonic system structure at lower temperatures [6]. For this purpose, a nematic liquid crystal doped with gold nanoparticles was combined with a small amount of a mixture consisting of a monomer and a UV-sensitive activator. In this work we present a way for constructing and developing enhanced temperature stability of self-organizing photonic structures based on nematic liquid crystals.



Fig. 1. One dimensional photonic crystal in nematic liquid crystal doped with gold nanoparticles

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Tunable liquid crystal optical microcavities for topological photonics

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The performance of novel kind of a tunable optical microcavity consisting of a nematic liquid crystalline birefringent optical medium enclosed in a typical Fabry-Perot resonator has been studied [1]. The large freedom of molecular reorientation, allow for convenient control of eigenmodes of light propagating through optical cavity while driven by relatively weak external electric fields. With the ability to manipulate the permittivity tensor and, therefore, effective refractive indices for different polarizations of light it is possible to tune the energy splitting between cavity modes which strongly influences the transmission, reflection and luminescence (if doped with a dye). Our observations can be interpreted in terms of spin-to-orbital angular momentum conversion, which conserves the angular momentum of the excited photons when they convert from one spin to the other through the spin-orbit coupling of light [2] (where “spin” represents polarization of light). Our novel device allows for generation and observation of various topological light polarizations like so-called persistent spin helix, i.e. polarization stripes [3] and complex polarization whirls like first and second-order merons and anti-merons [4] (**Figure**). In the future it allows for the integration of Bose-Einstein exciton-polariton condensates into the room-temperature operating devices.

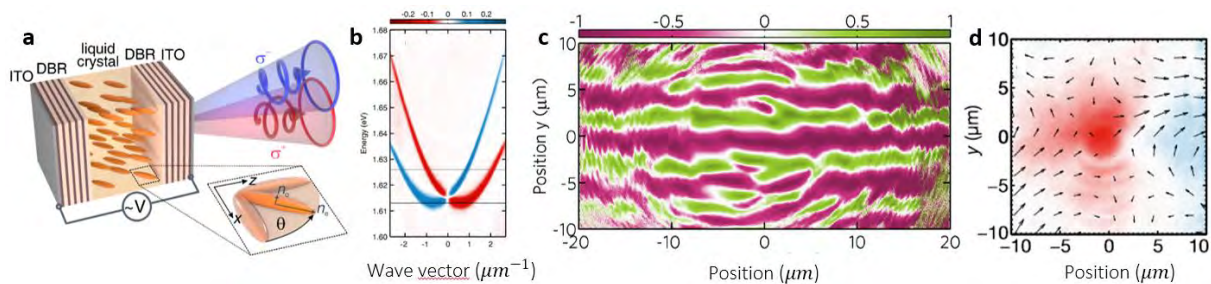


Figure. a) Scheme of the tunable LC microcavity and b) dispersion relation of circularly polarized light transmission [2]; c) observation of so-called persistent spin helix of light [3] and d) topological state of light: second order anti-meron of light polarization [4].

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O29

Title : Reflective optical components based on chiral liquid crystal for head-up displays

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Augmented reality and head-up displays have inspired new applications for flat optics. One of the ways to realize a virtual image in such displays is to use flat diffractive components based on chiral liquid crystal (CLC). CLC is a soft material that can self-organize into a periodic helical structure that strongly reflects visible light. Combining this property with a variation of the optical axis in a liquid crystal layer results in a reflective diffractive optical component that can diffract the light with a high efficiency over large angles and is polarization sensitive. The before-mentioned variation of the optical axis is achieved using a photo-alignment technique, which allows us to reach a sub-micrometer period. Using this method, we can obtain a range of reflective photonic components that can be used in head-up display systems. In the past, we reported diffraction gratings that are only a few micrometers thick and have an efficiency of over 90% [1]. In this work, a range of new reflective optical components, such as reflective lenses, is realized and investigated. We also further investigate the theory behind the simple reflective diffraction grating and suggest an analytical model to estimate the director configuration in the bulk of such a structure and to simulate optical behavior of the device [2].

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We report the formation of high optical power microlenses (Fig. 1) in the near-surface region of the liquid crystal layer. Such microlenses, possessing a very small focal length f at a rather large aperture A ($f/A \sim 2$), are able to focus the light into spots of a characteristic size comparable with the wavelength (Fig. 2). Using numerical modeling, a specific profile of patterning of an LC alignment surface by an ion beam [1] is proposed to provide the orienting properties necessary for the formation of an array of microlenses with a focal length comparable to the LC cell thickness. The proposed microlens arrays are produced and their optical properties are discussed.

Optical measurements of the focal length of the microlenses with a microscope give a value of $\sim 35 \mu\text{m}$. Thus, the F-number (the ratio of the focal length f to the aperture A) for the obtained lenses ($f/A \cong 2.3$) turns out to be higher than the simulated one, but, nevertheless, allowing classifying the microlenses as fast (wide-aperture).

The new microlenses can be useful for novel applications in LC photonics, for example, for the formation of periodically distributed intense optical fields inside the LC layer itself. These high-power fields enable optical pumping in LC microlasers and opto-optical control of the liquid crystals themselves.

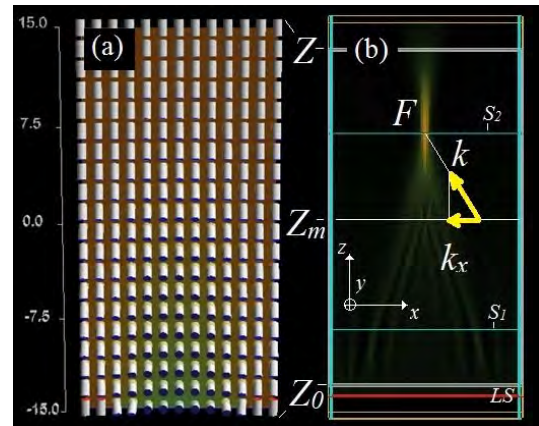


Fig. 1. a) Model of a 30 μm thick LC layer with a profiled distribution of the director in the x-direction on the surface Z_0 , providing the properties of a cylindrical microlens; b) light intensity distribution inside the LC layer calculated by FDTD method

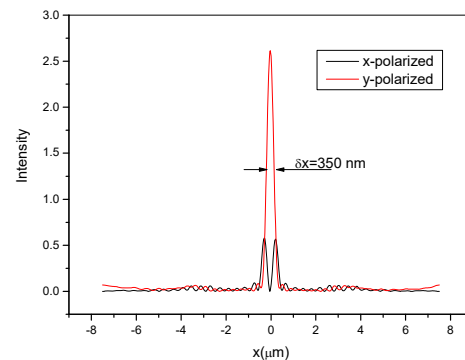


Fig. 2. Light intensity distribution at a wavelength of 450 nm in the focal plane for the calculated cylindrical LC microlens having 15 μm width.

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Selective light diffusion of polymer network liquid crystals formed through photopolymerization induced phase separation by nonuniform photoirradiation

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Light diffusers are fundamental but indispensable in optical and photonic applications, such as beam homogenization, optical attenuation, and privacy protection. Such applications demand different properties in light scattering, and various light diffusers have been developed using polymer network liquid crystals (PNLCs), such as hemispheric and unidirectional light diffusers [1,2]. PNLCs basically consist of phase-separation structures of liquid crystals (LCs) and polymers at a (sub)micron scale, and are versatilely applicable using the features of LCs, namely, anisotropic molecular structures and extrinsic-stimulative responses. We have developed diffusive and diffractive structures through a self-assembly process or photopolymerization induced phase separation (PPIPS) conducted by nonuniform photoirradiation technique [3,4].

Using this technique, we fabricated here light diffusers differently depending on optical polarization, scattering direction, and temperature. Figure 1 shows snapshots of light diffusion selective at a specific polarization azimuth to a specific scattering direction. The PNLCs were formed through PPIPS conducted by projection of anisotropic speckle patterns onto the raw mixtures of nematic LCs (5CB) and mixed monomers (AH600, DCPA, Ho, etc.). As shown in Fig. 2, micron-scale anisotropic domains were formed, and LC molecular orientation was uniaxially ordered in the domains. The vertically anisotropic domains produced light scattering intensive specifically in the horizontal direction, and the uniaxially orientation order of LC molecules in the domains linearly polarized the light scattering, as shown in Fig. 1. The nonuniform photoirradiation techniques are flexible and easy but practical for developing various advanced light diffusers.

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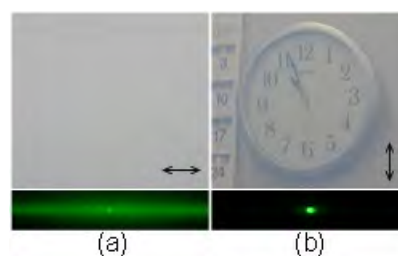


Fig. 1 Snapshots of wall clock looked through the sample and light distribution of green laser scattered from the sample, when the direction of light polarization is (a) horizontal and (b) vertical, as expressed by arrows.

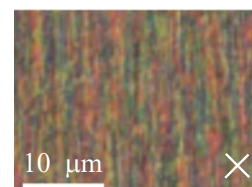


Fig. 2 Anisotropic phase separation structure of PNLC observed polarizing optical microscope at $\pm 45^\circ$ crossed-Nicols state, as expressed by cross.

Fast and Slow Tuning of Molecular Recovery Response of Cholesteric Liquid Crystal Elastomers in Layered System

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Liquid crystal elastomers (LCEs), with strong coupling of molecular orientation order and macroscopic elasticity of polymer networks, changes their material properties of molecular reorientation under external stimuli, leading to realize stimuli-responsive applications that work in the field of soft-robotics, flexible sensors, and so on. Although such molecular reorientation is very fast when stimulus applied, the recovery response after stimulus removal is relatively slow due to the slow relaxation of polymer networks and molecular orientation order. Very recently, a synthetic approach has succeeded to tune the speed of recovery response; in some case, however, the modification of molecular structures might deteriorate materials properties. Considering the strong coupling between the molecular orientation and materials shape of a LCE, the recovery response can be tuned by layering it with other layers with desired mechanical properties. Here we have investigated this layered system to arbitrary tune the molecular recovery response of LCEs. For visualizing molecular orientation order facilely, we employed a cholesteric liquid crystal as an internal LCE layer because cholesteric medium, has helically twisted molecular orientation and reflective color appearance due to Bragg reflection depending on the helicity.

The synthesized cholesteric LCEs clearly showed reflection color; and the color was blue-shifted under tensile elongation reversibly (Figure 1). For evaluating molecular recovery response, we applied square-wave strain on the film. The single-layered LCE exhibited typical viscoelastic response behavior after stimulus removal. On the other hand, the film layered by rubbery materials of polydimethylsiloxane (PDMS) showed fast recovery response with no residual strain both in microscopic molecular orientation and macroscopic shape (Figure 2). Furthermore, by changing external layers, we could tune the speed of recovery response, *e.g.* $\ll 1$ s or > 6 months, without any modification of molecular structure of LCEs. We demonstrated the cholesteric LCEs acting as a sensor detecting a spatiotemporal strain by using a commercially available camera.

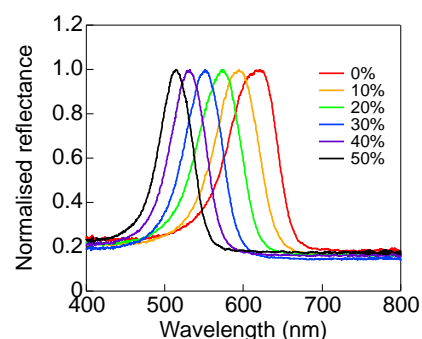


Figure 1. Reflection spectra of the cholesteric LCE under tensile strains of 0–50%.

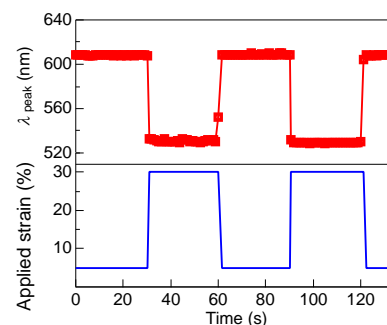


Figure 2. Time courses of the change in reflection wavelength of the PDMS layered film under 5–30% tensile strains.

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Confocal Observation of Reconnecting disclination lines in Nematic Liquid Crystal

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In nematic liquid crystals, singularities in the director field appear as disclination. Disclination lines in three-dimensional liquid crystal move with deformation in observable timescales. The motion of disclination lines is expected to depend not only on the location and shape of the disclination but also on the director profile around disclination. Currently, the relationship between the motion of disclination lines and the director profile has been actively studied also in biological systems such as microtubule bundles, bacterial populations, and morphogenesis.

Here we aim at direct three-dimensional measurements of dynamics of disclination lines. Using fluorescent dyes and confocal microscopy, we successfully observed the three-dimensional dynamics of disclination lines (Fig. 1). We analyzed reconnections, which are a characteristic phenomenon of disclination lines, and determined a scaling law describing the distance between disclinations. Further, we found asymmetry in the speed of the two disclinations toward the reconnection. A possible cause of the asymmetry is the difference in the curvature of the two disclination lines, but this alone is found to be quantitatively insufficient. Consideration of the director profile around the disclinations suggests that the asymmetry in the director profile gives a major contribution to the asymmetry in the motion.

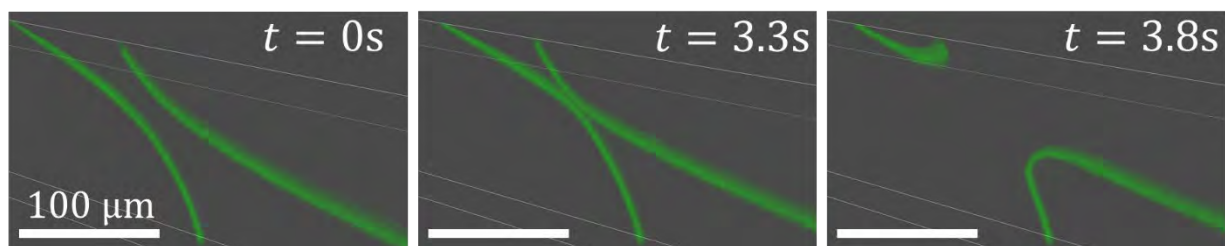


Fig. 1 Three-dimensional confocal microscopy images of reconnection of disclination lines in nematic liquid crystal

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Yamamoto et al. recently invented the principle and design of the “*Slippery interface*”, which has zero (or almost zero) anchoring at the substrate interface, and the response of the vertically aligned ferroelectric liquid crystal (VA-FLC) was drastically improved (low driving voltage)[1]. Furthermore, we utilized the localization of ionic liquid to the electrode by polar interaction, and fixed the ionic liquid at the electrode part by gelling. As a result, we achieved further reduction of driving voltage[2], but there is a problem that the slow response component was dominated in the transmittance. If the structure could be divided in some way and the co-operative motion of helix unwinding by applied electric field could be localized, it is expected to lead to a high-speed response. In this paper, we tried to utilize nano-phase separation[3] composed of helical filament of B4 phase of bent-core molecules and rod-like liquid crystals for dividing the helical structure of FLC. Combining the ionic liquid gel and this nano-phase separation, we evaluated the electrooptic behavior of vertically aligned FLC.

Ionic liquid gel used was a mixture of CDBA6•Cl and 1-ethyl-3-methylimidazolium bis (fluorosulfonyl) imide (50 g/L). Bent-core molecule (P-8-OPIMB) was ca. 1wt% mixed into FLC, and finally ionic gel was 20-50 wt% mixed into FLC mixture. This sample was inserted 3.5 μm -thick IPS cell, and applied electric field treatment ($\pm 4 \text{ V}/\mu\text{m}$) was performed for several hours at 70°C in order to accumulate the ionic liquid sol on electrode. Afterwards, ionic liquid was fixed on the electrode as a ionic liquid gel by cooling process. Electrooptic measurement was performed under the application of step-wise electric field.

Figure 1 shows the electro-optic response of VA-FLC without ionic liquid gel (violet), with ionic liquid gel (green), and with ionic liquid gel and bent-core liquid crystals (red) in 3.5 μm -thick IPS cell under the applied step-wise electric field ($+1.5\text{V}/\mu\text{m} \rightarrow 0 \rightarrow -1.5\text{V}/\mu\text{m} \rightarrow 0$). In VA-FLC with ionic liquid gel, transmittance drastically increases, which suggests the reduction of driving voltage by introduction of ionic liquid gels.

Compared to VA-FLC only with ionic liquid gel, furthermore, a higher ratio of fast response components in transmittance was found in VA-FLC with ionic liquid gel and bent-core liquid crystals. This result suggests that the helical structure of FLC was divided due to the nano-phase separation and co-operative motion of helix unwinding becomes faster.

Acknowledgement

This study was supported by JST-CREST (Grant No. JPMJCR1424).

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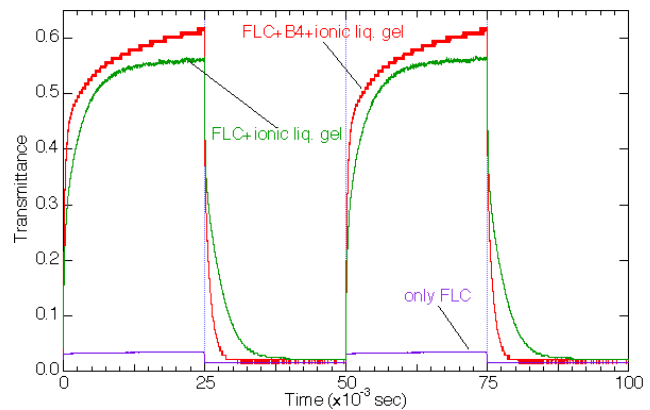


Fig. 1 : Electro-optic transmittance of three types of VA-FLC in 3.5 μm -thick IPS cell under the applied step-wise electric field of 10 Hz at 65°C.

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Diffraction gratings are fundamental optical components that are used in many fields of science. Nematic liquid crystals (NLCs) are interesting for tunable diffraction gratings because the optical properties can be controlled by external fields. For the study of the optical property of gratings, fabrication of a spatially periodic director field is crucial. Recently, we found a simple method for micropatterning the director field in NLCs in the macroscopic scale using homeotropically aligned NLCs with negative dielectric anisotropy. Due to ions charged at the interfaces, the micropattern is spontaneously formed at the reorientation of the director field by an electric field. Moreover, the director configuration exhibits defect structures often called umbilics, which are useful for optical vortex generation. [1] In this system, it is possible to control the emergence of pattern formation by modifying the surface anchoring condition. An example is shown in Fig. 1. When the director starts to reorient at the threshold, we can observe an array of umbilical defects. Such a highly self-organized pattern is interesting for both far- and near-field diffraction. In this presentation, we report near-field diffraction of NLC gratings consisting of topological defects arrays.

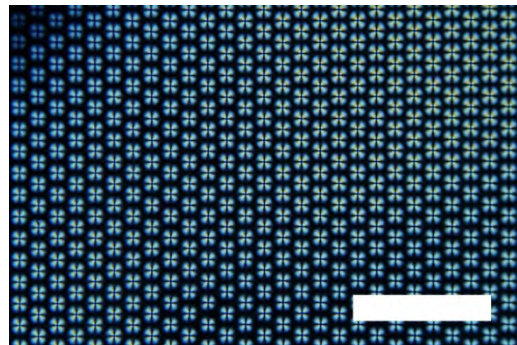


Fig. 1 An array of topological defects induced by an electric field observed under crossed polarizers. Scale bar, 400 μm .

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Helical Nanopores to Induce Chiroptic Function – Use of Empty Spaces Created by Helical Nanofilaments

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The B4 phase of some bent-core liquid crystals is an optically active state induced by the non-trivial chiral superstructure of helically-twisting smectic layers. This helical structure, the so-called helical nanofilament (HNF), is now understood as a result of the coupling of the saddle-splay curvature and the chirality induced by the spontaneous molecular tilt in the smectic layers. HNFs are also known for their excellent chiroptic functions, such as huge circular dichroism (CD) and optical rotation (OR) signals. The vividly- blueish optically segregated domains of the B4 phase has been reported already in the first works by Watanabe and Takezoe et al., and since then it has been dealt as one of the symbolic chiral states of the bent-core liquid crystals [1].

Here in this work, we established an epoch-making method based on bottom-up templating for the fabrication of a chiral nanoporous film that provides a chiral environment (chiral nano-space) to induce the chiroptic functions [2-3]. HNFs of a bent-core molecule were utilized as a three-dimensional mold, and thus the fabricated chiral nanoporous film has an inverse nano-helix structure. Upon refilling this inverse helix with an “achiral” nematic liquid crystal, distinct CD signals appeared due to the transfer of chirality from the inverse helix to the achiral nematic liquid crystal.

The formation of the chiral superstructure was confirmed by the induction of CD signals. In addition, the induced CD signals can be readily modulated by external stimuli, such as the application of heat or an electric field. Amazingly, by refilling the chiral nanoporous film with a fluorescent nematic liquid crystal, it exhibits stimuli-responsive circularly polarized luminescence (CPL). The proposed approach has huge potential for practical applications, such as for chiroptic modulators and switches and biological sensors.

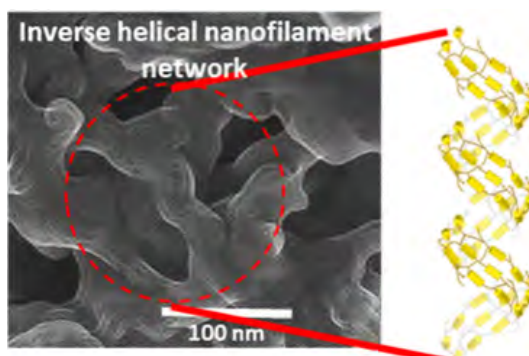


Fig. 1 Helical polymer structure fabricated in the inversed helix replicated from HNFs of the B4 phase of the bent-core liquid crystal, P7.[3]

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Inorganic nanosheets obtained by exfoliation of layered crystals have been investigated as intriguing nanomodules for various functional materials. Under a certain condition, nanosheets dispersed in a solvent are spontaneously ordered to form liquid crystal phase with regulated superstructures^[1]. In this study, we prepared the layered perovskite nanosheet liquid crystals with lamellar-like periodic structure that show structural colors and immobilized them in polymer gel by in-situ polymerization technique,^[2] in view of applications as stimuli-responsive soft materials.

The perovskite nanosheets (Inset of Fig. 1) were synthesized using the method reported previously.^[1] To obtain the composite gels, the monomer (*N*-isopropylacrylamide; NIPAm), crosslinker and photoinitiator were dissolved into the nanosheet colloid and the mixture was irradiated with UV-light to proceed photo polymerization.

The nanosheet colloid showed pearl-like purple structural color, giving the maximum reflectance wavelength (λ_{\max}) of 555 nm in the visible reflectance spectrum. As the monomer was added, λ_{\max} shifted to 521 nm, indicating the slight change of the interactions between nanosheets. Even after polymerization, the structural color was retained ($\lambda_{\max} = 684$ nm), indicating successful immobilization of the superstructure of the liquid crystalline nanosheets. In the final state, considering the Bragg-Snell law, the distance between the nanosheets is calculated as 257 nm. The gel possessed high mechanical toughness with the compressive breaking stress of up to 3 MPa due to the reinforce effect by the nanosheets. The color of the gel was changed from light pink to green and blue by mechanically compressing or stretching the gel (Fig.1). The mechanochromic response was reversible, fast (response time less than 1ms), and highly sensitive to very weak stress of 1 kPa.



Fig. 1 The photograph of the gel with varied structural color during a stretching test and the schematic structure of the perovskite nanosheet.

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Hexagonal magnetite (Fe_3O_4) nanoplates are of great interest because of their marvelous properties and wide range application in functional magnetic fluids and smart nanosensors. To control the dispersion of Fe_3O_4 nanoplates, polymers could be utilized to modify Fe_3O_4 nanoplates to induce the synergistic effects of Fe_3O_4 and polymers. According to our previous research, iron(III) oxide ($\alpha\text{-Fe}_2\text{O}_3$) particles and Fe_3O_4 particles with anisotropic shapes could be covered by thick polymers via surface-initiated atom transfer radical polymerization (SI-ATRP)^[1]. In the present study, hexagonal Fe_3O_4 nanoplates obtained from the wet chemical reduction of $\alpha\text{-Fe}_2\text{O}_3$ nanoplates were modified by poly(methyl methacrylate) (PMMA) chains *via* SI-ATRP shown in Fig. 1a. Such PMMA-modified Fe_3O_4 nanoplates indeed showed good dispersion stability, lyotropic liquid-crystalline (LC) properties, and a fast response to an external magnetic field in ionic liquids. In particular, a TEM image in Fig. 1b indicates that the PMMA-modified Fe_3O_4 nanoplates can show a nematic columnar structure after applying an external magnetic field.

The nematic columnar structure along magnetic field direction is a new example for the magnetic responsiveness of lyotropic LC PMMA-grafted Fe_3O_4 nanoplates in compare with the stacked alignment of unmodified Fe_3O_4 nanoplates in Fig. 1c. Ultrasmall-angle synchrotron X-ray scattering measurements also verified the uniaxially aligned nematic columnar structure of PMMA-modified Fe_3O_4 nanoplates in ionic liquids under an external magnetic field^[2] as shown in Fig. 1d. The research results might be closely connected with the performance improvements of functional magnetic fluids and the design of new devices.

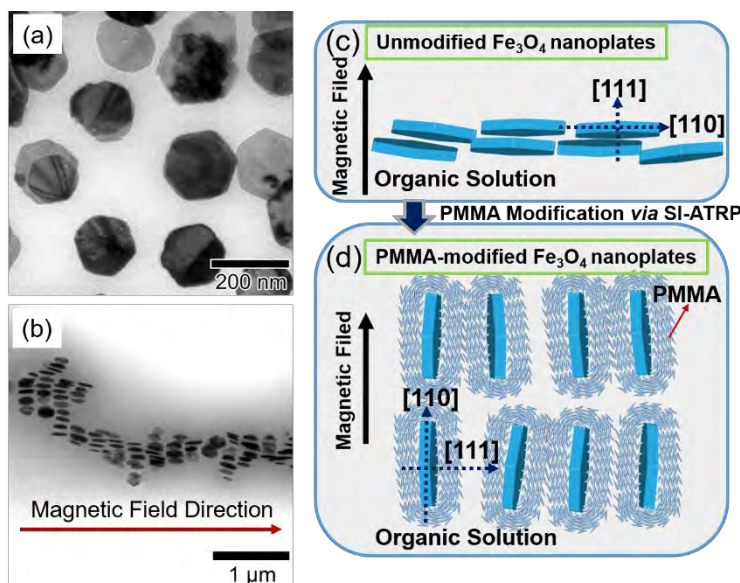


Figure 1. TEM images of (a) unmodified Fe_3O_4 nanoplates, and (b) PMMA-modified Fe_3O_4 nanoplates with nematic columnar structure under magnetic field. The most plausible model: (c) A stacked alignment of unmodified Fe_3O_4 nanoplates, and (d) a uniaxial alignment structure of PMMA-modified Fe_3O_4 nanoplates under magnetic field.

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Self-assembly of rod-shaped organic molecules is versatile to prepare various layered nanostructures, as investigated using smectic liquid crystals and amphiphilic systems. The unique feature of these molecules lies in the formation of layered molecular assemblies associated with the side-by-side intermolecular interaction. This feature gains considerable attention in the field of organic semiconductors, because it enables the formation of smooth channel/gate-insulator interfaces to obtain high-performance organic field-effect transistors (OFETs).

We have demonstrated that the ability to form thin layers is significantly improved using a class of rod-shaped π -conjugated molecules substituted by a single alkyl chain [1]. These materials afford the formation of a bilayer-type structure, being reminiscent of cell membranes. Recently, we have shown that these materials even afford the formation of self-assembled single molecular bilayers (SMBs), whose thickness uniformity extends over the area as large as wafer size scale (**Fig. 1**) [2-4]. To produce SMBs, a novel concept, geometrical frustration, is adopted in the film-formation technique. In this technique, two molecules with different alkyl chain lengths are contained in the solution [2]. The use of the mixed solution can effectively suppress the interlayer stacking while maintaining the intralayer molecular arrangements between π -conjugated molecules [2, 3]. The obtained SMBs afford the OFETs showing efficient two-dimensional carrier transport [4]. This finding should open a new route to SMB-based ultrathin super-flexible electronics.

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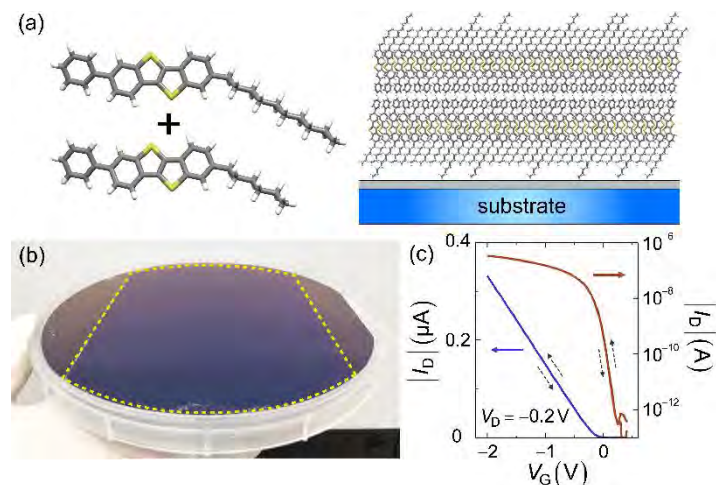


Fig. 1 Formation of the wafer-scale single molecular bilayer (SMB). (a) (Left) Schematic for two kinds of liquid crystalline organic semiconductors (Ph-BTBT- C_n) with different alkyl chain lengths. (right) Schematic for geometrical frustration induced by chain-length disorder. (b) A photograph of the wafer-scale SMB. (c) Typical transfer characteristics of the SMB-based OFET.

040

Photo-induced phase transitions of calamitic-discotic bimesomorphic azobenzene-triphenylene liquid crystals

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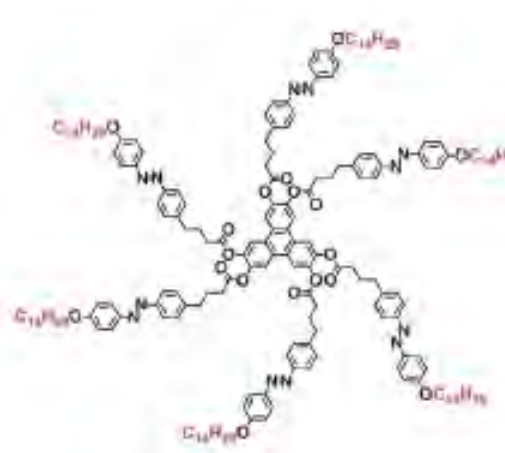
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A novel type of liquid crystal, calamitic-discotic bimesomorphous azobenzene-triphenylene derivative (**1-C14-3**) which exhibit both calamitic SmA and discotic Col_r mesophases by way of the change of molecular conformation [1, 2]. have also a potential as a photo-responsive material evidenced by the fact that **1-C14-3** shows photo-induced bimesomorphous phase transitions as shown in Fig. 1. To clarify the mechanism, GI-SAXS method was applied to the aligned cells with homeotropic and planar alignments in the SmA phase.

Fig. 2 shows GI-SAXS images of homeotropically aligned SmA phase. It was found that the photo-irradiation makes the layer spacing a bit shorter, indicating the interdigitation of alkyl chains among layers gets less, probably due to the trans-cis photoisomerization of azobenzene units and thus, the stability of SmA phase decreases under photo-irradiation. Successive cooling leads to the formation of Col_r phase in a manner of arrangement for the columnar axis which is comparable to the case of thermal phase transition.

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1-C14-3

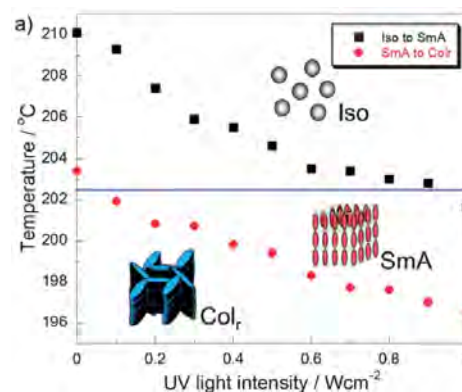


Fig. 1 Phase transition diagram under photo-irradiation at 365 nm.

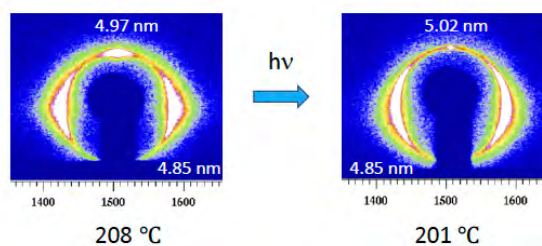


Fig.2 GI-SAXS images of homeotropically aligned SmA phase obtained on cooling from Iso phase.

Polarization-Maintainable Axially Polar Ferroelectric Columnar Liquid Crystal Phase Realized by Introducing Chirality

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Axially polar ferroelectric columnar (AP-FCLC) phases, in which the polarity along the column axis is switchable by applying an external electric field, and is maintained even after the electric field is removed, have attracted much attention. If the polar direction of each column is switchable with a nano-sized electrode, an ultra-high density memory device can be achieved. Recently, many examples were reported as AP-FCLC phases, and their hysteresis loops and/or switching current peaks were shown as evidence of ferroelectricity. However, most examples are done at high frequencies that polarization relaxation cannot keep up with, and very few studies focus on polarization maintenance (PM) after removal of the electric field. We would like to distinguish between the phase that shows PM and the phase that does not show PM, and believe that PM is the most important property to realize the above-mentioned memory devices. So, since 2005, in liquid crystal (LC) urea compounds, we have been aiming for realization of a polarization-maintainable AP-FCLC phase that perfectly maintains its polarization even after removal of the voltage.

In this talk, we report on a polarization-maintainable AP-FCLC phase and the mechanism for realizing the phase. The six achiral alkyl chains in *N,N'*-bis(3,4,5-trialkoxyphenyl)ureas **Urea-n** were replaced with (*S*)-citronellyl groups in (*S*)-**1**. Surprisingly, the columnar LC phase of (*S*)-**1** showed perfect ferroelectricity. Furthermore, the polarization directions were switchable by applying a rectangular wave voltage, and the induced polarization was maintained after removal of the electric field.

Second harmonic generation (SHG) experiments were performed to investigate the polarization states of **Urea-10** and (*S*)-**1**, respectively. Both compounds in the columnar phase showed strong SHG signals as soon as 100 V_{DC} was applied. After turning off the voltage, the SHG signal for **Urea-10** disappeared immediately, but that for (*S*)-**1** was maintained at the same level for at least 10 h. Helical column formation caused by the introduction of chirality is assumed to play an important role in achieving the ferroelectricity.

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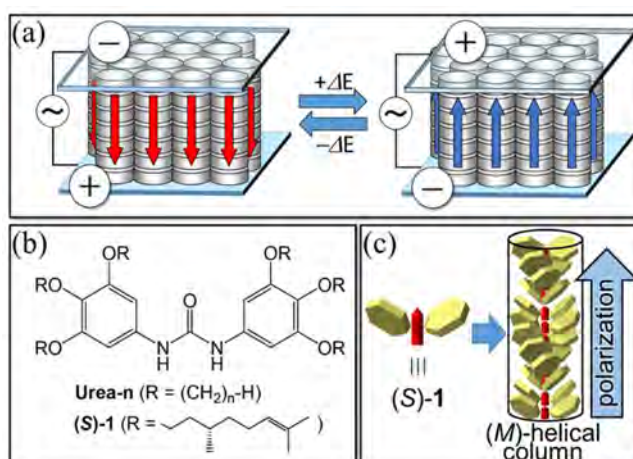


Fig.1 (a) switching in AP-FCLC phase, (b) molecular structures of ureas, and (c) helical column of (*S*)-**1**.

Poster Abstracts

P1

Title : Development of Novel Chiral Molecules with High Helical Twisting Power

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When a chiral molecule is added to a nematic liquid crystal, a cholesteric phase with a helical structure is formed. The force that induces the helical structure is called helical twisting power (HTP). The twisting power of the chiral azobenzene compounds is decreased, Δ HTP, by ultraviolet irradiation to cause trans-cis photoisomerization of the azobenzene compounds. We have reported reversible photoswitching of transparent and light-scattered states by controlling the balance of HTP between photochromic and nonphotochromic chiral dopants [1]. The development of chiral molecules with large HTPs and Δ HTPs is essential to achieve highly efficient switching with a small amount of dopant. Recent studies have shown that binaphthyl derivatives exhibit high HTPs and high Δ HTPs. In this study, we synthesized several compounds with alkyl chains and liquid crystalline units introduced into binaphthyl derivatives (Fig.1) and studied their HTP and Δ HTP values.

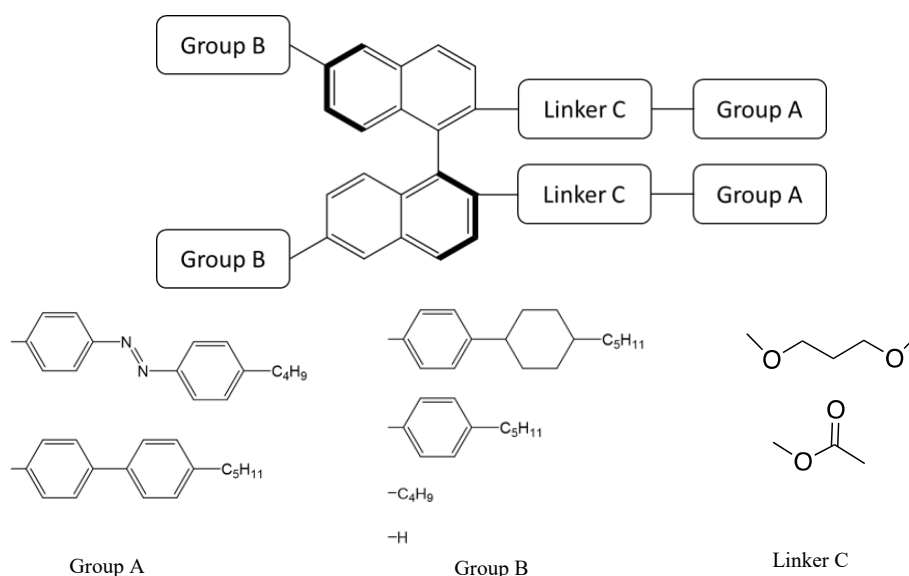


Fig.1 Molecular structures of photochromic and nonphotochromic chiral compounds.

Reference

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Analysis of Angular Dependence of Band Edge Wavelengths in Cholesteric Mirror and Bragg-Berry Cholesteric Deflector

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Patterning of the liquid crystal director in cholesteric liquid crystals (ChLCs), or control in the structural phase of the helical structure, enables circularly polarized light to be diffracted [1]. Although the selective reflection phenomenon in a conventional ChLC mirror has been the subject of thorough investigation and is well understood, recently a new simple approximation model for estimating band edge wavelengths of selective reflection from ChLCs for oblique incidence has been proposed [2]. More recently, the approximation model has been developed for ChLC based holographic optical elements, in which the band edge wavelengths and bandwidth of the selective diffraction are determined from simple equations [3]. In this study, we investigate angular dependence of the band edge wavelengths in ChLC mirrors and deflectors using the approximation model and two-dimensional finite-difference time-domain (FDTD) method.

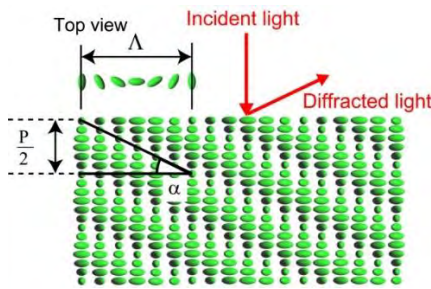


Fig. 1 Schematic of Bragg-Berry cholesteric deflector.

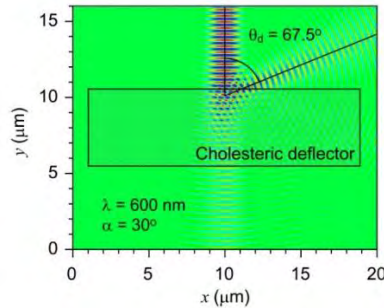


Fig. 2 FDTD calculation of diffraction from Bragg-Berry cholesteric deflector.

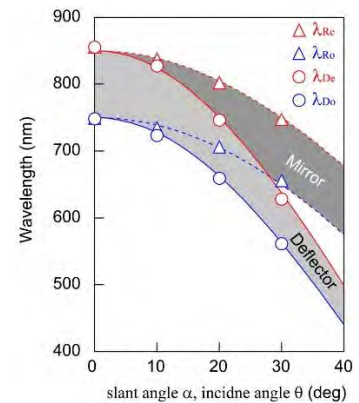


Fig. 3 Band edge wavelengths of reflection and diffraction from cholesteric mirror and Bragg-Berry cholesteric deflector.

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Aggregation Enhanced Room-Temperature Phosphorescence with Dual Emission from Rod-Like Gold(I) Complexes

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Organic materials with aggregation-induced emission (AIE) are of increased interest due to the broad practical application in optical devices and probes. Many types of organic AIE-active molecules (AIEgens) have been reported. In particular, gold complex is one of the most attractive AIEgen, owing to the ability to form dimeric compound via aurophilic interaction. We have reported gold complexes with AIE properties, of which luminescence behavior is highly sensitive to not only the molecular structure but also the structure of molecular aggregate.¹ Therefore, controlling the aggregated structure is necessary to obtain the desired luminescence properties by molecular design.

In this study, a series of rod-like gold complexes were synthesized (Figure 1), and effects of the molecular structures on their photophysical properties were investigated. All gold complexes showed strong emission in the crystal, but only weak emission was observed in the solution. The emission properties of gold complexes in the crystal are affected by molecular structure. As shown in Figure 2, in crystal, complexes containing biphenyl moiety (B2-5 and B3-5) exhibited an emission band at ~500 nm. On the other hand, crystal of the complex containing cyclohexylphenyl moiety (CP-5) showed dual emission: a new emission band appeared in the shorter wavelength region. The relative intensities of the dual emission bands could be controlled by excitation wavelength; thus, we could control the emission color of CP-5 crystal.

The lifetime measurements revealed that the observed emission from the gold complexes in crystal was phosphorescence. In the crystal, even at room temperature in air, they showed high phosphorescence quantum yield (~23%). This intense room-temperature phosphorescence is considered due to the tight packing structure of the crystal which prevents the internal molecular motion and the oxygen penetration.

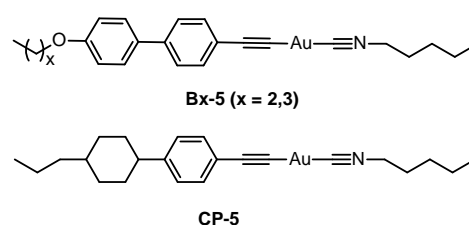


Figure 1. Molecular structure of gold complexes used in this study

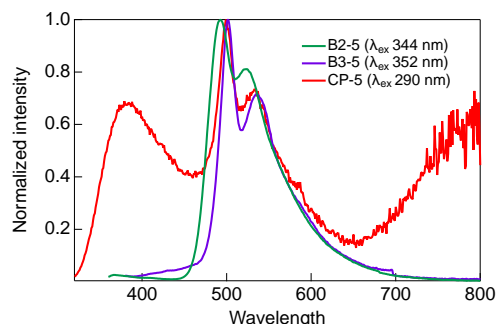


Figure 2. Emission spectra of gold complexes in the crystal

References

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P4

Dynamic coupling of gel network and nematic orientation order

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We studied the dynamic coupling between orientation fluctuation of liquid crystals and conformation of polymers in the nematic gel (NG). The pre-cursor of the NG is made by heptyl-cyanobiphenyl (7CB), 8% hexyl-cyanobiphenyl acrylate (LC-A), and 0.6/0.8% reactive mesogen (RM257) (similar mixture used in the previous study[1]). We made the two NG by changing the cross-link density, high cross link density (LCLD NG) and low density (HCLD NG). A small amount of DMPAP used as a polymerization initiator. We cured the sample by illuminating with UV lights (365 nm, 880 $\mu\text{W}/\text{cm}^2$) for 30 minutes. We measure the dispersion relation before and after polymerization of LCLD NG and HCLD NG (Fig.1 (a) and (b)).

(1) Before polymerization, both dispersion relations of the orientation fluctuation for LCLD and HCLD NG are almost equivalent, and relaxation time is proportional to the inverse square of q ($\sim q^{-2}$) as predicted by the hydrodynamic theory.

(2) After polymerization, dispersion relation of LCLD becomes about 5 times slower, but still proportional to $\sim q^{-2}$. On the other hand, it should be noted that the relaxation time becomes independent of q in the low q region for HCLD.

In order to investigate the spatial distribution of polymer network in NG, we measured the position dependence of the retardation of the NG by 2D spectrometer, because the polymer

concentration directly relate the scalar order parameter of the orientation order. Since the wavelength dependence of the transmitted light intensity should oscillate dependent on the birefringence, we can analyze the size of the inhomogeneity of the spatial distribution of the polymer. Fig.2 show the observed image for transmitted light intensity of LCLD NG and HCLD HG (Fig.2 (a) and (b)). It can be confirmed that the size of the inhomogeneity of polymer reaches 0.1mm for LCLD NG whereas less than 0.01mm for HCLD NG. In conclusion, in the cross-linking point density for HCLD NG is enough to be percolated and sufficient to confine the orientation fluctuation. This is because the relaxation frequency is independent of q in low q region as seen in the dispersion relation of polymerized HCLD NG.

References [1] C.C.C hang, et al. Phys. Rev. E 56, 595 (1997)

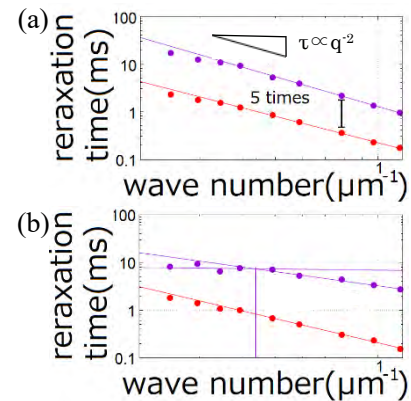


Fig.1 Dispersion relations
(a) LCLD NG (b) HCLD NG (30°C)

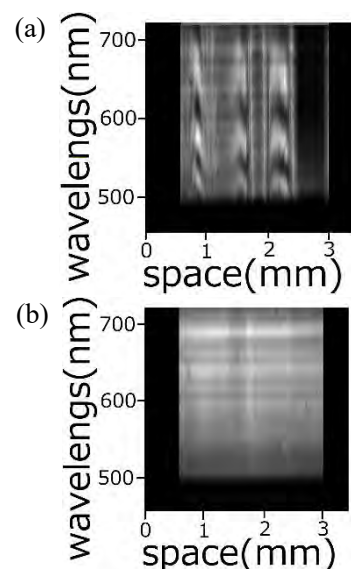


Fig.2 images by 2D spectroscope
(a) LCLD NG (b) HCLD NG (44°C)

P5

Shape classification of liquid crystal elastomers with twist director configurations

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Introduction

Various types of deformation are thermally induced in liquid crystal elastomer (LCE) as a result of the coupling of LC orientation and rubber elasticity by tuning the director configuration. The LCE ribbons having twist director configurations (TNE) undergo a large change in helical pitch and diameter of the helical shape in response to temperature variation [1]. However, it remains to be elucidated what governs the helical shape. We investigate what type of parameter governs the shape of the TNE ribbons, and classify the shapes depending on the geometrical features.

Experiment

The TNE ribbon specimens were made by using the method in Ref. [1]. The twist angle (χ) was varied as 90, 135 and 180° by tuning the amount of chiral dopant. A ribbon specimen was cut out from the TNE sheets so that the long axis had an angle (θ) with respect to the mid-director direction (Fig. 1). The angles θ were varied from -90° to +90°.

Result & Discussion

We observe the shape changes driven by temperature variation in each TNE ribbon. We find that the helical shapes at fixed temperature are classified into four types depending on θ , regardless of χ (Fig. 2). The classifications are based on the helical handedness (right or left) and the outer surface (top or bottom). The top and bottom surfaces are defined according to Fig. 1. In a previous study [1], the helical handedness depending on θ was characterized at $\chi=90^\circ$, but the distinction of the outer surface (top or bottom) was missing. The shape classification in Fig. 2 is also confirmed by the finite element simulation for the TNE ribbons using a planar LCE with the same composition as a unit element.

Reference

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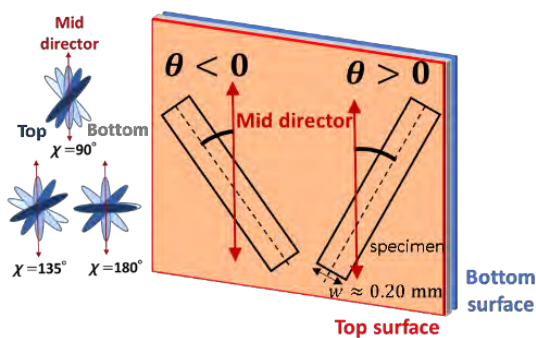


Fig. 1 Geometry of TNE ribbon specimens

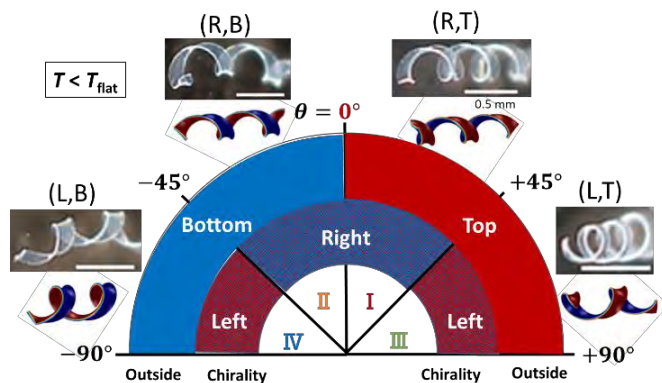


Fig. 2 Shape classification of TNE ribbon.

P6

Mechanoresponsive PDMS that reversibly changes fluorescence in sub-MPa stress

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PDMS is a silicone elastomer that is widely used in biotechnology and electronics fields due to its biocompatibility, flexibility, transparency, and easy processability. In this study, we have developed PDMS elastomers that can quantitatively evaluate weak sub-MPa stress by ratiometric fluorescence analysis. In the PDMS, a flexible fluorescent force probe (FLAP) has been chemically introduced into cross-linking positions of the polymer chain network. The FLAP molecule changes the fluorescence spectrum quickly and reversibly by its conformational conversion, thus FLAP can be used for real-time analysis of nanoscale stress concentration^[1]. Stress-strain curve and viscoelasticity of PDMS with different composition ratios have been analyzed to reveal the relationship between mechanical properties and fluorescence response. The fluorescence response by compression as well as stretching will be discussed.

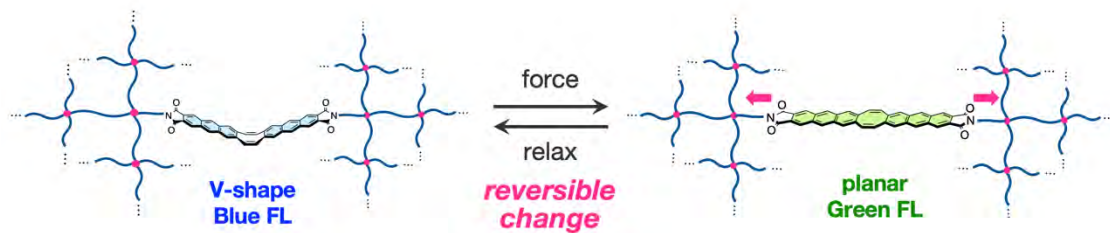


Fig. 1. Force-responsive reversible fluorescence change of the FLAP molecule.

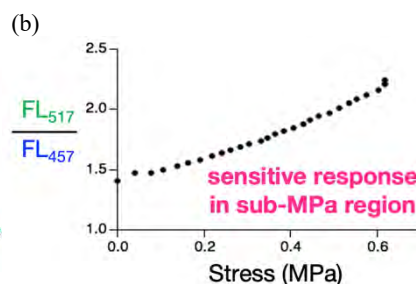
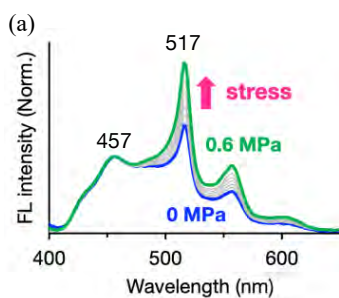


Fig. 2. (a) Fluorescence spectral change and (b) ratiometric fluorescence analysis of the PDMS elastomer during a tensile test.

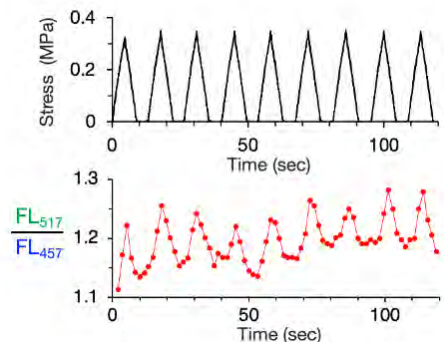


Fig. 3. Reversible fluorescence response of the PDMS elastomer during stretching and shrinking.

References

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Dynamic Control of Array Structure Based on Fe₃O₄ Magnetic Nanoparticles by Hybridization with Liquid-Crystalline Organic Dendron

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Magnetic nanoparticles (NPs) are attracting special attention because of their unique magnetic properties different from those of the bulk states. In addition, their magnetic properties are affected by their interactions dependent on the interparticle distances. Therefore, formation of the array structure based on magnetic NPs enables us to control their magnetic properties, being still a challenging problem due to strong magnetic interaction between magnetic NPs in such magnetic NPs-based colloidal crystals. Liquid-crystalline (LC) organic dendron is one of the most representatives to show a thermotropic LC behavior by self-assembling into columnar and spherical dendrimer-like structures. In our previous studies, we have developed the dendron-modified inorganic NPs to form LC cubic structures by surface modification of NPs with LC dendrons [1,2]. In the present study, we applied this technique to Fe₃O₄ magnetic NPs to control the array structure based on magnetic NPs. The schematic image of the material design is shown in **Fig. 1**. A synthetic procedure for the dendron-modified Fe₃O₄ NPs is as follows: Firstly, COOH-modified Fe₃O₄ NPs were synthesized by a thermal decomposition method followed by a ligand exchange reaction with a terminal COOH-substituted phosphonic acid. Then, an amidation reaction was carried out between the COOH group on the surface of Fe₃O₄ NPs and an amino group of LC dendrons. **Fig. 2** shows TEM images of COOH- and dendron-modified Fe₃O₄ NPs. Since both the particle diameters were calculated as 6.8 ± 0.6 nm, the size and shape was scarcely influenced by the reaction. On the other hand, interparticle distance increased to 12.9 nm from 8.7 nm and well-ordered hexagonal array were observed after the reaction. The self-assembling property was successfully introduced into the Fe₃O₄ NPs. The self-assembling behavior and the magnetic property of LC dendron-modified Fe₃O₄ NPs will be discussed.

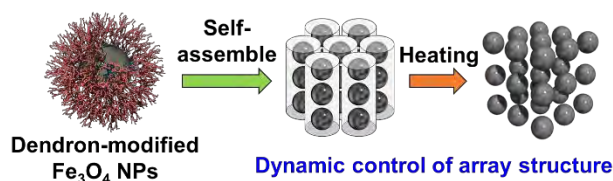


Fig. 1 Schematic illustration of dynamic control of array structure based on Fe₃O₄ NPs.

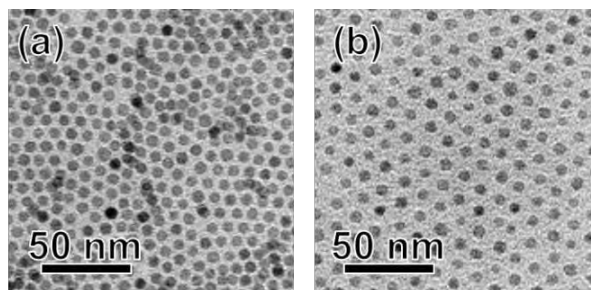


Fig. 2 TEM images of (a) COOH-modified and (b) dendron-modified Fe₃O₄ NPs.

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P8



Title : Synthesis of Polymerizable Tetrabenzocoronene Mesogen Carrying with Diacetylenic Side Chains

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Polymerizable tetrabenzocoronene (TBC) derivatives were designed and synthesized in our work, the structure is shown as Figure 1. TBC compound have a good electron transport ability caused strong π - π interaction. All TBCs can be polymerized by UV irradiation and form the homogenous film, which is proposed to show the greater charge mobility than TBC monomer. Synthetic strategy of TBCs is using Katz-modified Mallory half-photocyclization method irradiated by 48 W of 365 nm mercury lamp, using FeCl_3 to process the cyclodehydrogenation, and then do esterification with diacetylenic side chains to obtain final compound. The purity of compounds is investigated by Nuclear Magnetic Resonance spectroscopy (NMR) and Infrared (IR) spectroscopy. The characteristics are investigated by differential scanning calorimetry (DSC), X-ray diffraction, fluorescence spectroscopy, and UV-vis spectroscopy.

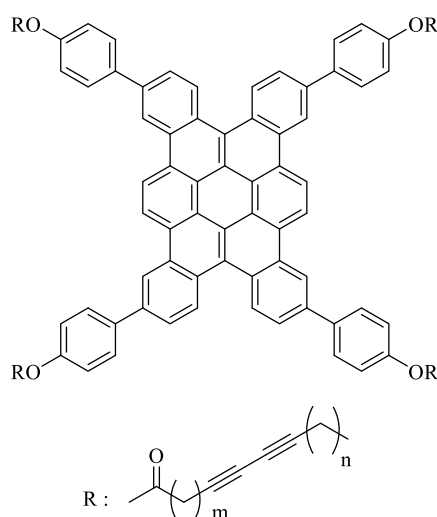


Figure 1. The structure of TBC

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P9

Mechanical Response of Molecular Orientation in Liquid Crystal Elastomers Controlled by Multi-Layered System

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Liquid crystal elastomers (LCEs), in which mesogenic orientation are strongly coupled with polymer network, have attract much attention to realize stimuli-responsive applications. For example, the mechanical elongation leads to macroscopic deformation of elastomer in coincidence with mesogenic reorientation and of materials properties. This process is well-known as soft-elastic response. Typically, the soft-elastic response is very fast when stimulus applied; however, after the stimulus removal, the macroscopic shape and microscopic orientation exhibits slow recovery due to the viscoelastic behavior based on a molecular structure of CLEs. This means that the limited tunability on the speed of recovery response has tended to narrow their application range. Very recently, we have found that a simple materials design concept that is an LCE layered with other films showing desired mechano-responsive behavior enables to arbitrary tune the speed of recovery response even in the region of soft-elastic response. This initial finding indicates that microscopic mesogenic orientation is governed by the deformation behavior of other external layers. In this study, we investigated the relationship between the mesogenic reorientation of the inner layer of LCEs and the external other layers under applied strain. More specifically, we examined the effect of anisotropic deformation of the outer layers on the three-dimensional mesogenic reorientation behavior of LCEs. To facilely quantify the mesogenic reorientation with high spatiotemporal resolution, we employed cholesteric liquid crystal medium as an internal LCE layer. The helically twisted mesogenic orientation in the medium, exhibiting Bragg reflection at a certain wavelength matched with the helical pitch, allows us to visualize three-dimensional mesogenic reorientation under deformation.

The cholesteric LCEs sandwiched by other layers were fabricated by photopolymerization of mixtures of a LC monomer, a chiral dopant, a plasticizer, a crosslinker and an initiator in a glass cell attached with the external layers. The multi-layered film exhibited a bright reflection, and the color was blue-shifted by application of tensile strain. We evaluated the strain in thickness from the reflection wavelength and investigated the effects of mechanical properties of external layers on the three-dimensional mesogenic reorientation behavior.

P10

Two-step fluorescence color change of stress-responsive polyurethanes that incorporate a flapping molecule

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We have recently developed flapping molecules, FLAP [1], with various functions and applications. For example, anthracene-based FLAP works as a ratiometric force probe that shows a fluorescence switch at the low threshold (*ca.* 100 piconewton) [2]. Relaxed bent conformation of FLAP emits a blue fluorescence in polymeric environments, while the stressed planar conformation shows a green fluorescence in stretched polymers. The FLAP force probe reversibly shows the stress-induced fluorescence change without covalent bond scission, and therefore it realizes quantification of local stress concentration in entangled polymer chain network before structural damage.

Here we discovered a novel two-step fluorescence color change during the tensile test of FLAP-incorporated polymers. A new flapping fluorophore containing phenazine wings (FLAP1) was chemically doped as a crosslinker of segmented polyurethanes (Figure 1). The fluorescence color changed from blue to green under lower stress (-12 MPa), and then it further changed to yellow under high stress (-57 MPa). Mechanism of the two-step fluorescence change will be discussed (Figure 2).

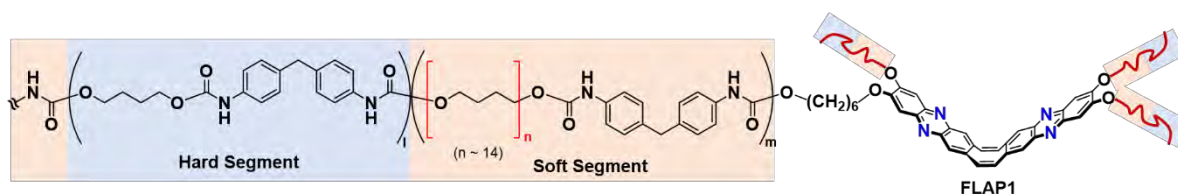


Figure 1. Chemical structure of the FLAP1-doped segmented polyurethane.

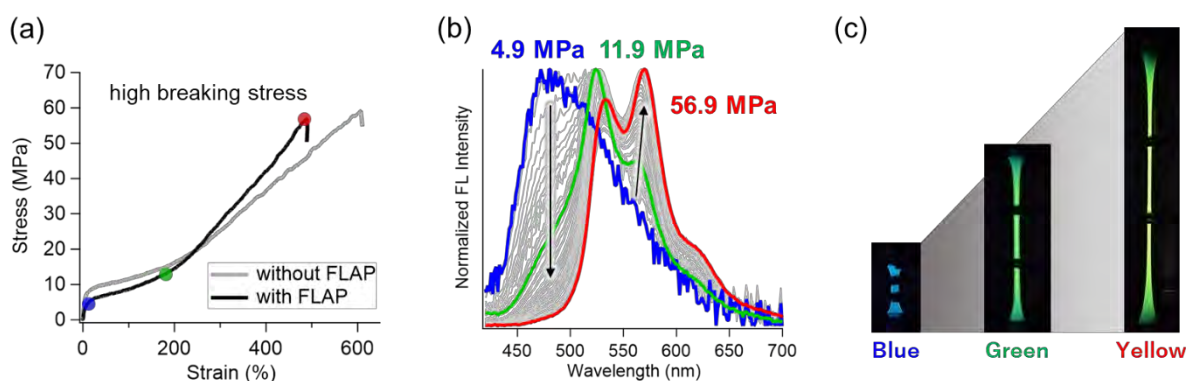


Figure 2. (a) Stress-strain curve of the FLAP1-doped polyurethane. (b) Fluorescence spectral change during the tensile test. (c) Photographs of the stretched specimen showing the two-step fluorescence color change.

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Introduction: Water solution of $C_{12}E_5$ forms a lamellar phase in a wide region of concentrations. We were interested in the dynamic coupling between the undulation fluctuations of lamellae and the Brownian motion of colloidal particles [1]. We investigated the dispersion relations for both modes by using the dynamic light scattering (DLS). We try to clarify the nature of the dynamic coupling between two motions by changing the concentration of colloidal particles.

Results: We used of the mixture of the lamellar phase made of the water solution of $C_{12}E_5$ and water dispersion of colloidal particles. Lamellar phase consists of the water solution of $C_{12}E_5$ ($\varphi=6$ wt%; inter-membrane distance 62.5nm) and colloidal particles dispersion (latex beads, $\Phi=0.15$ wt%; diameter 20 nm, negatively charged).

As shown in Fig.1, two types of relaxation modes were observed simultaneously in the autocorrelation function of the mixture. We analyzed the dispersion relation of both fast and slow modes by changing the scattering angle (Fig.2). Since the relaxation frequency of the fast mode of the mixture (green closed circles) is very close to that of pure colloid dispersion (blue closed circles), then fast mode is identified to be Brownian motion of colloidal particles. Red circles is the dispersion relation of the undulation fluctuation of the pure lamellar phase. Slow mode can also be identified to be the undulation fluctuation of the lamellar of mixture. We found that the undulation fluctuation of the lamellar should be slowing down due to the existence of the colloid particles. Finally, we studied effect of concentration of colloids on the undulation fluctuations of the lamellar (Fig. 3). It should be noted that the undulation fluctuations of the lamellar is slowing with increase of Φ .

Conclusion: The deceleration of the undulation fluctuations in the mixture should be caused by decrease of the layer compression modulus. Namely, the presence of colloidal particles between membranes prevents direct collisions between adjacent membranes and weakens the steric interaction between them.

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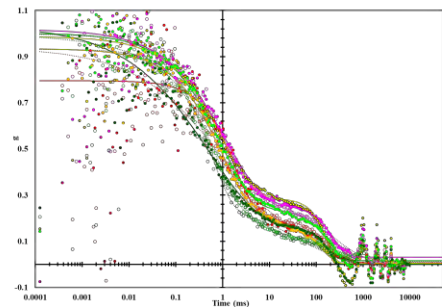


Fig. 1 Autocorrelation function of the mixture of the lamellar phase and colloidal dispersions. Each line represents the scattering angle dependences.

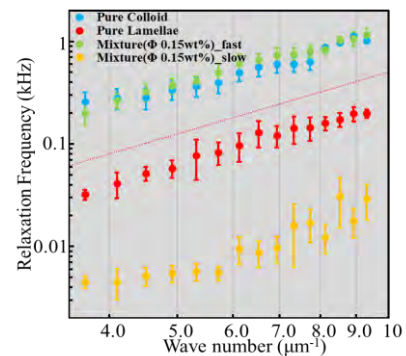


Fig. 2. Dispersion relation of pure colloidal dispersion, pure lamellar phase, and the mixture.

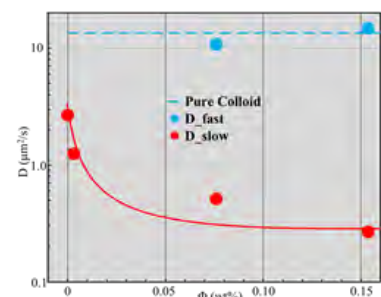


Fig. 3. Concentration Φ dependence of diffusion constants for fast and slow modes.

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Inorganic nanosheets have been highlighted as new-type colloidal liquid crystal (LC) systems since their rediscovery¹ and have been applied for anisotropic and tough gel materials suitable for artificial muscles.² However, it was difficult to precisely optimize the nanosheet LC property due to the difficulty in controlling the particle size and the lack of structural designability. In this situation, we recently found that monodisperse nanosheets synthesized by bottom-up method² form a string-like columns of stacked nanosheets and columnar nematic liquid crystal phase. In this study, we synthesized columns of stacked nanosheet stabilized by long-chain alkylammonium (dodecyltrimethylammonium chloride [C₁₀TMACl]) with the aim of using them as actuator devices.

The wide angle X-ray scattering (WAXS) results of the columns of stacked titanate nanosheets with tetramethylammonium (TMA) as the interlayer cation are shown in Fig. 1a. A sharp peak at $d_{001}=1.7$ nm is observed, indicating that the nanosheets are stacked with a period of 1.7 nm. Since the thickness of the titanate nanosheets is 0.75 nm, the interlayer distance is calculated as 0.95 nm that matches the size of hydrate TMA. By the addition C₁₀TMA (8.0 times amount of the cation exchange capacity [CEC] of the titanate nanosheets) (Fig. 1d), d_{001} increases to 2.9 nm (interlayer distance of 2.15 nm). Considering that the length of the all-trans C₁₀TMA⁺ is about 1.54 nm, the C₁₀TMA⁺ is intercalated as a bilayer structure with a 44° tilt in the interlayer. As the amount of C₁₀TMA⁺ increases (Fig. 1b, c, d), the full width at half maximum of the d_{001} peak decreases, indicating the amount of intercalated C₁₀TMA⁺ increases.

When the samples added with 8 times CEC of C₁₀TMA was observed with polarized optical microscope, interference colors due to birefringence. Based on these results, we conclude that columnar nematic LC of stacked nanosheets with interlayer C₁₀TMA⁺ are formed.

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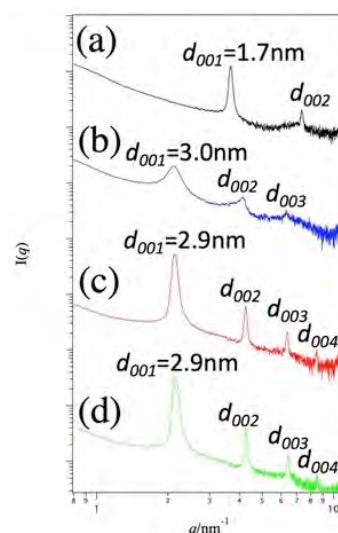


Fig. 1 WAXS patterns of the nanosheets added with C₁₀TMACl. The added amount is (a) 0, (b) 4, (c) 6, and (d) 8 times of the CEC of the nanosheets.

P13

Photoreduction Synthesis of Gold Nanosheet in Hyperswollen Lyotropic Lamellar Phase

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Nanosheets (NSs) synthesis in lyotropic lamellar phases have attracted attentions because of their high versatility. We have developed the synthesis method of NSs inside bilayers of hyperswollen lyotropic lamellar phases (HL phases), whose bilayers with several nm thickness are separated from each other by several hundred nm [1]. We can use them as independent two-dimensional (2-D) reaction fields. These reaction fields can prevent NSs from aggregation because of the intervals. We named this synthesis method ‘two-dimensional reactor in amphiphilic phase (TRAP) method.’ We have successfully synthesized hydrophobic NSs inside the hydrophobic TRAPs in aqueous amphiphilic solutions [2]. We expected that the unexplored hydrophilic TRAPs enable us to synthesize hydrophilic NSs. As the next target of the NSs synthesis, gold is the suitable because it is synthesized by photoreduction in the hydrophilic reaction field. Gold has face centered cubic lattice, whose growth is isotropic. Therefore, the synthesis of the gold NSs (AuNSs) acts as a test of new TRAP method applicable to other materials like other crystalline materials with cubic lattices, organometallic compounds, and amorphous materials.

Here, we show the facile synthesis of uniformly thin AuNSs in a sodium 4-octylbenzenesulfonate solution by irradiation with UV light. The horizontal width and thickness of the obtained AuNSs measured by atomic force microscopy (AFM) are 1500 ± 560 nm and 2.18 ± 0.50 nm, respectively, as shown in Fig. 1. The full width at half maximum (FWHM) of each of the X-ray diffraction (XRD) peaks of the AuNSs was broader than that of the Au fine particles synthesized without TRAPs. The Williamson-Hall plots indicate that the crystallite size of the AuNSs is smaller than that of Au fine particles. Moreover, the selected area electron diffraction (SAED) pattern measured by transmission electron microscope (TEM) also indicates that the AuNSs are polycrystals consisting of small crystals; the SAED pattern shows concentric circles. These results indicate that the uniformly nanosized Au particles are likely to fuse laterally in the confined thin 2-D reaction field in the HL phase, and they only grow laterally, as shown in Fig.1. The hydrophilic TRAP method could be applied to other hydrophilic NSs because the precursors are accumulated due to rather physical confinement than chemical specificity.

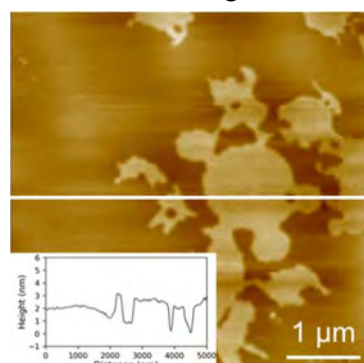


Fig. 1 AFM image and cross-section analysis of one of the AuNSs.

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Fabrication of nematic liquid-crystal necklaces and their physical properties

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Various kinds of self-assembly of liquid crystals, i.e., nematic droplet, nematic shell, cholesteric shell and liquid crystal fiber, are abundantly studied for the applications to a micro-laser, a micro optical resonator, and so on. Among these self-assemblies, nematic droplets demonstrate Whispering Gallery Mode (WGM) resonance, which is useful for a micro optical resonator [1]. In this study, we succeeded to fabricate the connected nematic liquid-crystal droplets with tethers, called as a liquid crystal necklace [2].

Liquid crystal necklace was made with a microfluidics device [3]. 5 % aqueous solution of polyvinyl alcohol (PVP 72000, Merck) and 4-cyano-4'-Pentylbiphenyl (5CB; TCI) were used as continuous and dispersed phases, respectively. Necklace structures with various droplet diameters (ca. 20 – 300 μm) were formed by tuning the capillary diameter of a microfluidics device or flow rates of both liquids. The tether part was so thin that it could not be observed by an optical microscopy, however we could confirm the connection between tethers and droplets by means of the laser tweezers. The necklace structure was not broken by the nematic-isotropic phase transition induced by the increase in the temperature. The structure was also preserved by the change in the molecular alignment of liquid crystals inside the droplet from parallel to homeotropic alignment.

The detailed structure and the physical properties of tethers were investigated. By the observation of a confocal microscopy and a polarization microscopy, it was revealed that tethers are made of the complex of PVP and 5CB, and in addition, both of them align along the long axis of a tether. The elastic constant of the tether was determined by using laser tweezers to stretch the tether. It is about 8 kPa, and thus it is similar to the elastic constant of a PVA hydrogel [4].

The WGM circulating inside individual droplets in the necklace structure were also observed. However, no light could be seen being guided through the tethers. For the light transport to tethers, the stabilization of tethers and usage of chiral nematic liquid crystal will be helpful.

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Iridescent Colors in Aqueous Solutions of Chromonic Disodium Cromoglycate Doped with Chiral Additives

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Understanding the mechanism underlying the self-organization of chromonic molecules into lyotropic liquid crystals (LCLCs) has drawn much attention in recent years, as it may provide more insights into the research for biological and medical applications [1]. This is probably because of their similarity to DNA liquid crystals. This report investigated the phase behavior and the helical twisting powers of chiral solutions of disodium cromoglycate (DSCG) in water [2-3]. The addition of water-soluble chiral dopants to the nematic phase of DSCG readily turned it into the cholesteric phase. The cholesteric pitch was measured by Grandjean–Cano wedge cell method. The microscopic structure was characterized by X-ray diffraction and freeze-fracture transmission electron microscopy (FF-TEM). In general, the twisting power is weak, and the shortest pitch (ca. 2.9 μm) was obtained using *trans*-4-hydroxy-L-proline. However, one can still observe the iridescent colors of some of the cholesteric samples at certain viewing angles [4].

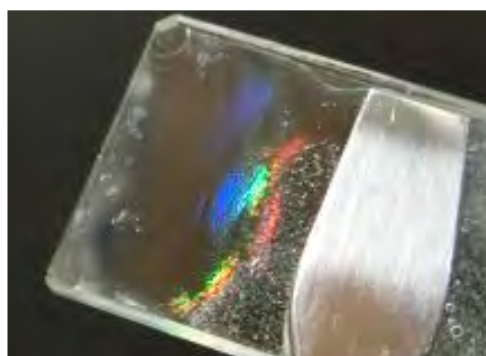


Fig. 1 Iridescent colors due to the selective reflections in an unaligned sample of disodium cromoglycate in water doped with *trans*-4-hydroxy-L-proline 26 wt%.

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High-performance liquid-crystal-loaded terahertz wave variable phase shifter with non-radiative dielectric waveguide structure

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To realize ultra high-speed terahertz (THz) wireless communication, antennas that support transmission/ reception in multiple directions become indispensable, and high-performance phase shifters are increasingly required. Liquid-crystals (LCs), which have excellent characteristics such as large anisotropy even in the THz region and good response to an external electric field, are expected for THz applications in recent years. In the region of light, most experiments allow light waves to pass through a free space before arriving at the device. On the other hand, unlike light, the THz waves have a longer wavelength of about 1 mm, and when they are radiated into free space, a large amount of leaky-wave and diffraction wave arises. In previous study, a transmission-type THz wave phase shifter has been reported. In this case, the transmission loss is enormous at about -15 dB, and there is an unsolved problem of satisfying both large phase change and fast response [1]. Furthermore, we have reported a transmission-line-type THz wave phase shifter using a non-radiative dielectric (NRD) waveguide[2]. In this study, we propose a high-performance LC-load THz wave variable phase shifter with the NRD waveguide structure, which simultaneously performs short response times and continuous phase control.

Fig. 1 shows the cross-section diagram of the device and a simulation result of propagating electric field distribution at 350 GHz. In this study, we used high-density polyethylene (HDPE) as low loss dielectric material for the transmission line and PTFE sheet as spacers for the LC cell. With simulation results, we can see that the THz wave is radiated from a rectangular waveguide (WR2.2) and completely confined in the dielectric waveguide without leaky-wave and diffraction wave at the air part or PTFE spacers. Moreover, as shown in Fig. 2, the THz wave variable phase shifter having response times of about 1 s to several ms and phase change near 270 degrees is realized. Detail will be debated on site.

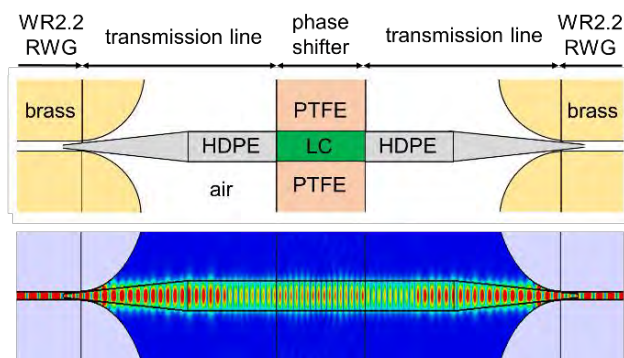


Fig. 1 Cross-section diagram of the device and simulation result of electric field distribution at 350 GHz

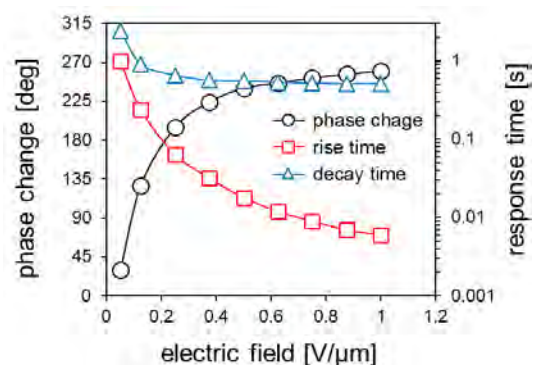


Fig. 2 Response characteristics of the phase shifter at 350 GHz

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P19

Emitting Color-switching in TN display using transparent UV range polarizer

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A conventional liquid crystal display (LCD) is non-emissive type and polarizer films with the transmittance of about 45% or less. Recently, Mochizuki and Morita have proposed a novel transparent emissive LCD which was fabricated by using a UV range polarizer, a TN liquid crystal cell and a polarized blue light emitting film [1]. Both the polarizer and the emitting film have high transmittance over 85% in the visible region, therefore the LCD shows a blue emission on the transparent background. In this study, we have proposed the emissive color switching LCD using the polarizer and the emitting film.

Figure 1 shows a schematic model of the emissive LCD and a principle of the emission color switch. A small amount of fluorescent dichroic dye is dissolved in the TN cell. In the off-state, the polarization direction and the absorption axis of the dye in the TN cell is parallel and bright emission from the LC cell can be obtained. Moreover, the polarization direction of the transmitted UV light is rotated at 90° and is not absorbed in the external blue emitting film, that is “LC emitting state”. In the on-state, the emission light intensity decreases and the UV light passes through without the polarization rotation, then the external emission film absorbs the UV light and emits a bright light, that is “emitting film excited state”. We used Coumarin 6 as a guest of fluorescent dichroic dye [2]. When without the UV irradiation, the LCD is slightly yellowish because of the absorption of Coumarin 6 in visible region. Using a UV-LED ($\lambda_{\max}=365$ nm), the emission spectrum was measured and switching property is shown in Fig. 2. The peak wavelength changed from 525 nm of Coumarin 6 to 440 nm of the emitting film and the emission color was successfully switched from green to blue. Dichroic ratio (DR) of Coumarin 6 in the LC is about 6 and higher DR might improve the color difference more.

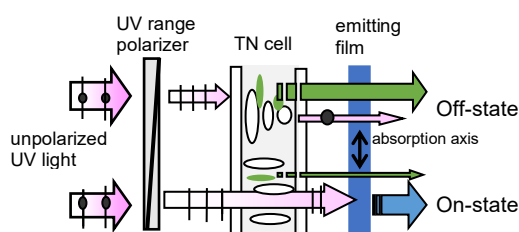


Fig.1 Principle of emission color switching

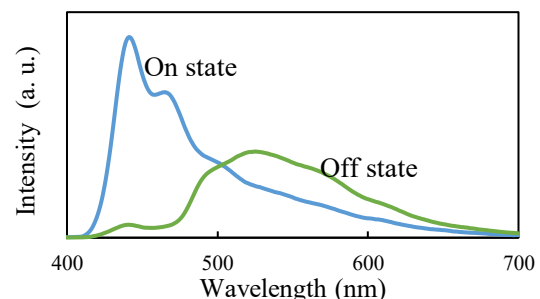


Fig.2 Emission spectra in off- and on-state of LCD.

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Continuous rotation of a cholesteric liquid crystalline droplet by a circularly polarized optical tweezers

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The manipulation of micrometer-scaled objects is important for utilizing them as micromechanical devices and to elucidate their microscopic properties. Optical manipulation methods including optical tweezers have been intensively studied and one can translate and rotate micro-objects with accuracy of sub-microns. Not only the manipulation of optically isotropic objects but also that of optically anisotropic ones has been studied. In case of birefringent objects, one can rotate them via linear and circularly polarized light [1]. One of typical optical birefringent spherical objects is a liquid crystalline (LC) droplet. The continuous spinning of a nematic LC droplet has been reported under the irradiation of focused circularly polarized laser beam [2,3]. The direction of the rotation is controlled by that of a circular polarization of the beam.

In this research, we studied the opto-mechanical response of a cholesteric liquid crystalline (ChLC) droplet to a circularly polarized optical tweezers. The micrometer-sized droplets were prepared by vigorous mixing of a nematic liquid crystal and chiral dopant R-811 in water. The ChLC droplets with various sizes (diameter d) and helical pitch p (strength of chirality) were trapped by an optical tweezers. Their response was studied by varying the direction of circularly polarized light (CPL). Although the alignment of LC molecule within a droplet depends on the relative ratio d/p [4], the optically induced rotation is asymmetric to the direction of CPL irrespective to their inner molecular alignment. This asymmetric response enables us to detect the chirality of droplets and separate them within a microfluidic device.

For a droplets of $d/p < 1$, a twisted bipolar droplet [4] exhibits asymmetric response: the rotation appears only for a right-circularly PL (RCPL). For a droplet of $d/p > 1$, a radial spherical droplet [4] also rotates only by RCPL. In the case of $d/p \sim 1$, the rotation is induced by both RCPL and LCPL. Its direction of the rotation is simply determined by the chirality of ChLC in this case. The intermittent rotation is also observed for a linearly PL, which has already been reported by Yang *et al.* [5]

This work is supported by JSPS KAKENHI (JP20H02947).

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P21

Specific molecular orientation and structure induction at the interface between nematic liquid crystal and azobenzene polymer film

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A monolayer of azobenzene compound or polymer on the surface can induce reversible alignment switching of nematic liquid crystals (NLCs) between homeotropic and planar modes upon visible/UV light irradiation, which is called the command surface [1]. To gain insight into the molecular process at the interface, Ubukata et al. investigated model hybrid LB films composed of 5CB and an Az polymer [2]. On the other hand, we recently fabricated a setup for the *in-situ* measurement of grazing incident small angle X-ray scattering (GI-SAXS) that can evaluate the structure at the substrate surface. In the present work, this X-ray analysis tool is applied to evaluate the structure at the interface between NLC and alignment layer.

In this study, liquid crystal molecules (5CB) were placed on an alignment film of side-chain azobenzene polymer (P5Az10MA) (Figure 1), and GI-SAXS measurements were performed to evaluate the structure from the vicinity of the interface to the bulk (Figure 2).

For the structural analysis, we designed a 35- μm thick glass cell (Fig. 2). A P5Az10MA spin-coated film (thickness: 60 nm) alone gave the out-of-plane peaks, indicating a smectic layer formation in the in-plane direction. Interestingly, after 5CB is injected, the out-of-plane peaks disappeared and the peaks up to the third order appeared in the in-plane direction (Fig. 2). This result indicates that, upon 5CB injection into the homeotropically aligned P5Az10MA, an orientation inversion to the in-plane direction accompanied by a highly ordered smectic hybrid layer formation of 5CB and P5Az10MA occurred at the interface. The details will be reported at the presentation.

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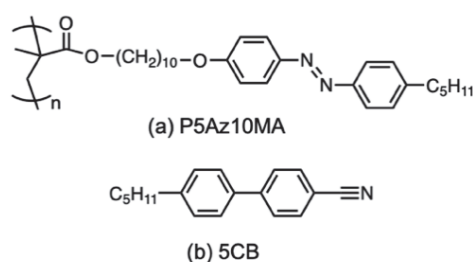


Figure 1. Structures of P5Az10MA (a) and 5CB(b)

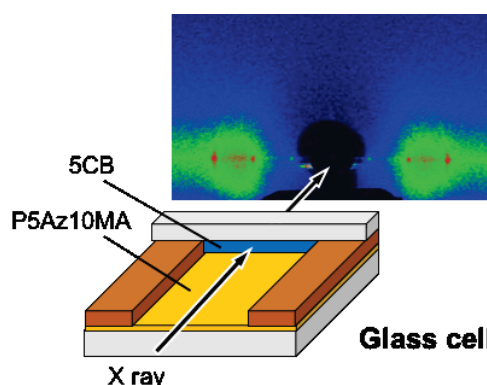


Figure 2. The glass cell used in GI-SAXS measurement and the image of interface structure formed by P5Az10MA and 5CB.

Ultrafast time-resolved electron diffraction with double-pulse-excitation applied to the light-melt adhesive based on a flapping triphenylene liquid crystal

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We performed ultrafast time-resolved electron diffraction measurements with single-pulse-excitation and double-pulse-excitation to investigate the mechanism of the photoinduced melting phenomena of the light-melt adhesive, which is composed of a flapping triphenylene liquid crystal (TP-FLAP).

Ultrafast time-resolved electron diffraction measurements enable us to directly monitor the molecular-level structural dynamics under photoexcitation on the femto-to-picosecond timescales [1]. The method with single-pulse-excitation monitors the photoinduced event starting from the ground state (with all molecules taking a bent form), which does not necessarily represent the photoinduced behavior of the TP-FLAP molecules under continuous irradiation. In this study, we compared the time-evolution of the differential electron diffraction patterns from the molecules excited by single near-ultraviolet (UV) pulse and double near-UV pulses. Note that time-resolved electron diffraction measurements with double-pulse-excitation can monitor the photoinduced event starting from the excited state (with some excited molecules taking a planar form). Consideration of these excitation conditions leads to further understanding of the photoinduced melting behavior of the TP-FLAP liquid crystal.

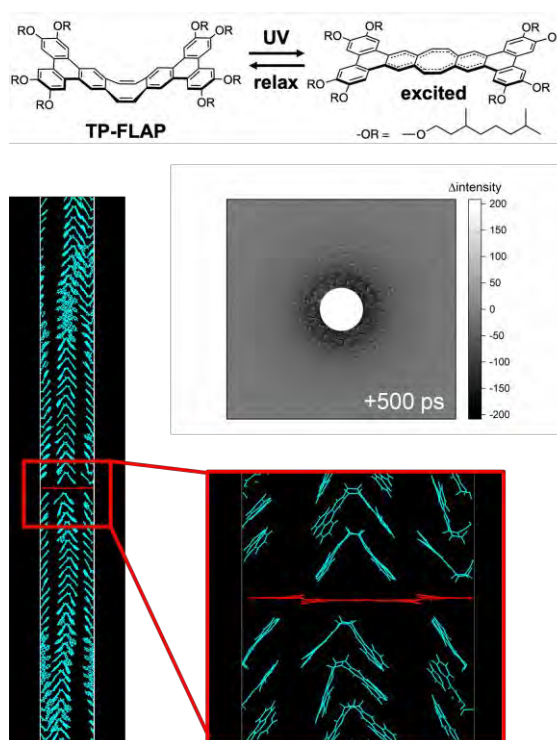


Fig. 1 TP-FLAP molecule shows an ultrafast bent-to-planar conformational change in the excited state (top). In the columnar packing structure, a fraction of the molecules are planarized by a single pulsed near-UV photoexcitation, which modulates the differential electron diffraction patterns (bottom).

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P23

Development of a photoreversible adhesive with two adhesion states using poly(methyl methacrylate) containing an azobenzene liquid-crystal plasticizer showing smectic E phase

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We investigated the photoplasticization behavior and photoinduced adhesion properties of poly(methyl methacrylate) mixed with azobenzene liquid-crystal plasticizers exhibiting different phase structures, i.e., nematic, smectic E, and crystalline phases. It was found that photoplasticization, which is the photoinduced reduction of glass transition temperature and hardness of polymers,^{1,2} could be repeatedly induced by alternate irradiation with ultra-violet (UV) and visible light, regardless of the phase structures of the photoresponsive plasticizers. Upon photoplasticization under UV-light irradiation, all samples exhibited the first adhesion state with weak adhesion strength, namely a pressure sensitive adhesion (PSA) state. Upon irradiation of the photoplasticized samples with visible light, the samples exhibited a second adhesion state by recovering their elastic moduli to the initial levels. The adhesion strength of the samples in the second adhesion state was significantly influenced by the phase structures of the photoresponsive plasticizers. When a photoresponsive plasticizer exhibited the smectic E phase, which is a highly ordered liquid crystalline phase, the adhesion strength was remarkably larger than that of the case using the plasticizers showing nematic and crystalline phases. This result was reasonably explained in terms of the suppressed bleed-out of the photoresponsive plasticizers from the polymer, and the good mechanical properties of the sample stemming from the characteristics of the smectic E phase. Through the reversibility of a photoplasticization process, we achieved a photoinduced reduction of the adhesion strength by UV irradiation of the samples in the second adhesion state. Thus, a photoreversible adhesive with two adhesion states could be developed by the photoplasticization of poly(methyl methacrylate) containing an azobenzene liquid-crystal plasticizer showing the smectic E phase.

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P24

Precision polymerization of a degradable block copolymer containing a nematic liquid-crystalline polymer block

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Block copolymers have attracted great attention as nanotemplates by using sub-100 nm structures formed by microphase separation. It is necessary to control the alignment of microphase-separated structures to obtain desired functions. Directed self-assembly of block copolymers have been developed to obtain well aligned nanostructures along physical or chemical guide patterns [1,2]. We have recently proposed a simple method for unidirectionally aligning microphase-separated structures simply by introducing a nematic liquid-crystalline polymer block, where the molecular alignment of liquid crystals induces the aligned nanostructures without fine guide patterns [3]. In this study, we designed a block copolymer consisting of degradable polymer and nematic liquid-crystalline polymer blocks towards developing hollow nanotemplates. We present the synthesis of the block copolymer and evaluation of their fundamental properties.

Poly(lactic acid) (PLA) was selected as a degradable polymer. Polymethacrylate having a phenyl benzoate moiety as a side-chain mesogen, PMA(4OPB), was used as a nematic LC polymer. A block copolymer PLA-*b*-PMA(4OPB) was synthesized by atom transfer radical polymerization (ATRP) using a PLA macroinitiator and a methacrylate monomer. A homopolymer PMA(4OPB) was also synthesized by ATRP for comparison. Gel permeation chromatography analysis indicated that PLA-*b*-PMA(4OPB) and PMA(4OPB) with narrow molecular weight distributions were successfully obtained. The homopolymer PMA(4OPB) exhibited a nematic phase according to differential scanning calorimetry and polarized optical microscopy. The block copolymer PLA-*b*-PMA(4OPB) also showed LC properties similar to the homopolymer. The degradation behavior of the polymers was investigated by immersing polymer films in a solution of trifluoroacetic acid and subsequently monitoring the changes in the molecular weights. PLA was completely degraded, while PMA(4OPB) almost maintained the molecular weight. Furthermore, the molecular weight of PLA-*b*-PMA(4OPB) was decreased. These results suggest that the PLA block in the block copolymer was selectively degraded.

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P25

Two-dimensional molecular orientation directed by scanning wave photopolymerization with digital light processor

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The key for designing high-performance optical devices is the fine and precise control of molecular orientation in liquid-crystalline (LC) materials. In particular, cycloidal molecular orientation in the LC films enables the fabrication of diffractive optical elements [1]. So far, cycloidal patterns are generally induced by the polarization holographic technique based on photoalignment processes using the interference of orthogonally polarized beams [2]. However, this technique has challenges such as low processability, necessity of polarized laser beams, and difficulty of large-area patterning. Recently, we have developed a novel method for molecular orientation, termed scanning wave photopolymerization (SWaP) [3,4]. This method utilizes molecular diffusion triggered by spatiotemporal photopolymerization, and thus allows us to direct large-area and two-dimensional (2D) molecular orientation without polarized light. In this study, we present the fabrication of LC polymer films with 2D patterns by SWaP with a digital light processor and investigate the optical property of the resultant films.

As a photopolymerizable sample, a mixture of an acrylate monomer A6CB, a crosslinker HDDMA, and a photoinitiator Irgacure 651 was used (Fig. 1). The glass cell was prepared

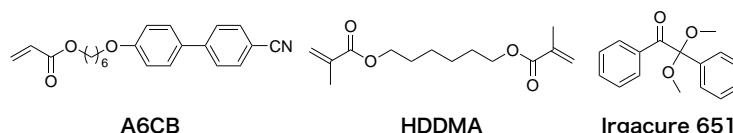


Fig. 1 Chemical structures of compounds used for photopolymerization.

by adhering a pair of ultrasonically cleaned glass substrates with 2- μm -thick silica spacers. The sample mixture was injected into the glass cell and irradiated with UV light by scanning the rod-shape patterns by digital light processor to carry out SWaP. The polarized optical microscopy revealed the cycloidal molecular orientation was induced in the LC films. Moreover, the LC film with the cycloidal orientation exhibited selective diffraction of circularly polarized light.

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Patterning Topological Defects in Nematic Liquid Crystals Using Area-Selective Surface Modification

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The control of the director field is crucial for liquid-crystal-based optical applications. Utilization of spontaneous pattern formation is an interesting approach, while it is usually difficult to expand the uniform domain in the macroscopic scale. In this regard, it is expected that the assistance of surface fabrication is useful for producing highly self-organized patterns. [1] In this study, we explore an efficient approach for controlling self-organized pattern formation of topological defects in nematic liquid crystals (NLCs). A typical result is shown in Fig. 1. By selectively modifying the surface, we prepare two homeotropic regions with different anchoring strength which are also capable of generating a square array of topological defects. (Fig. 1A) Here, a standard transmission electron microscopy (TEM) grid is tested as a mask in modifying the surface. The NLC used is MBBA. Under no electrical voltage, the whole area shows a homeotropic alignment. (Fig. 1B) On the other hand, when the electric field is increased, we observe square arrays of umbilical defects. We obtain a quite uniform domain in the area where the surface is modified, while the outside area shows multiple domains. (Fig. 1C) Since the diameter of TEM grids is around 3 mm, it provides a sufficiently large area for optical experiments. In this presentation, we show experimental results which are obtained by changing the sample thickness and the size of TEM grids.



Fig. 1 (A) Schematic illustration of surface modification. (B) A homeotropic texture under no electric field (C) A highly self-organized pattern of defects arrays induced by an electric field obtained by the assist of surface modification. Scale bar, 1 mm.

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Enhanced electric field tunability and thermal stability of photonic crystal fibers filled with gold nanoparticles-doped cubic blue phases liquid crystals

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Blue phase liquid crystals (BPLCs) are interesting materials that create unique self-assembling cubic structures in two phases BP I and BP II or occur as an isotropic-like ‘fog phase’ BP III. BPLCs exist between cholesteric and isotropic phases in a very narrow range of temperatures (~ 0.1 - 5.0 K) and are characterized by outstanding properties such as 3D Bragg reflections, optical isotropy, and polarization insensitivity in a macroscopic scale for the wavelengths outside their resonance bands [1, 2]. Currently, BPLCs belong to the most promising materials for perspective applications not only in fast-switching LC displays, but also in advanced photonic structures such as photonic crystal fibers (PCFs) [3, 4]. However, so far only a few papers have focused on studies in the latter field, considering mainly characterization of spectral properties of BPLC-filled PCFs. Moreover, these complex photonic structures still have significant limitations in optical communication such as not fast enough response times as well a necessity to use high driving voltage due to a relatively large cladding diameter of the PCF.

This work expands our preliminary observations by carrying out studies of optical properties of gold nanoparticles-doped cubic BPLCs infiltrating PCFs. It has been shown that the examined photonic system can provide promising tunable properties, enhancing the external electric field sensitivity and temperature stability. This is due to the presence of gold nanoparticles with an appropriate organic coating in a liquid crystal matrix. The presented results indicate lowering the threshold voltage and increasing the operating temperature range of the investigated photonic system. Furthermore, it turns out that a control of the light intensity for specific wavelengths can be obtained. Moreover, a range of wavelengths corresponding to low transmission (photonic bandgap) depends on the input light polarization and can be controlled by an external electric field.

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Observation of three-dimensional structure in electrohydrodynamic convective patterns of cholesteric liquid crystals by fluorescent confocal microscopy

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Electrohydrodynamic convective (EHC) patterns in nematic liquid crystal (NLC) have been studied as a typical non-equilibrium open system. Since cholesteric liquid crystal (ChLC) exhibit helical structure due to chirality, the convective patterns in ChLC are expected to be more complex than those in NLC. Various kinds of characteristic patterns have been reported: stripe, square lattice, eddy ones^{1,2}). The ratio of a helical pitch p to the cell thickness d is the parameter which governs the type of the observed patterns. A pattern which appears first depends on p/d : a stripe pattern appears first for small value of p/d , and a square lattice pattern for large p/d .

EHC patterns are usually observed under a polarizing microscope. However, the obtained images are averaged ones of the three-dimensional pattern along vertical direction. Recently, fluorescent confocal polarizing microscopy (FCPM) has been developed and applied to three-dimensional observation of local orientation in LC³). In this study, we observed the three-dimensional structure of the stripe pattern and the square lattice pattern in ChLC by FCPM.

FCPM makes it possible to acquire local orientation of LC in three-dimension under a polarized laser by adding small amount of fluorescent dye which orients parallel to the LC. The used ChLC in this study is a mixture of a chiral dopant, an NLC with negative dielectric anisotropy and a surfactant. The pitch was adjusted by the concentration of the chiral dopant. This sample was sealed in a planar cell and an AC electric field was applied perpendicular to the cell surfaces.

In the absence of electric field, the structure exhibits a stack of flat layers of ChLC parallel to the cell surfaces. The one layer corresponds half helical pitch. Above a certain threshold voltage, the layers begin to undulate in a sinusoidal form. Their magnitude increases and the layers become rectangular form with increase of electric field. Further increase of layer deformation results in the increase of the number of layers. Such deformation occurs in one dimension for the stripe pattern and in two dimensions for the square lattice pattern. In addition, we dispersed micrometer-sized particles in ChLC and tracked their positions to visualize the convective flow. For example, they rotate around the lattice points with a constant angular velocity in the square lattice pattern.

This work is supported by JSPS KAKENHI (JP20H02947).

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P29

Photoinduced Charge Behaviors in a Liquid Crystal Mixture on Different Measurement Frequencies

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We proposed a photoinduced charge measurement of Liquid Crystal (LC) mixtures by irradiating a light which is a selected wavelength by monochromator [1]. In this research, we investigated a change of photoinduced charge in an LC mixture by changing the measurement frequency.

Figure 1 shows the block diagram of the measurement. We irradiated the light from Xe lamp to cells through the monochromator that can scan the wavelength from 280 nm to 780 nm by 10nm step. We applied ± 10 V triangular waveform voltage to an LC cell by changing the measurement frequency from 10 Hz to 0.1 Hz and measured the displacement current to obtain the total charge which is calculated from the entire area of voltage versus current curve in an LC cell on each wavelength. A photoinduced charge was calculated from the difference of total charge between with and without light exposure conditions. We prepared MLC2132 (Merck) as an LC mixture which was injected into an empty cell having the SiN on ITO glass substrates.

Figure 2 shows the wavelength dependency of photoinduced charge of the LC cell by changing the measurement frequency. We found that the photoinduced charges of each frequency suddenly started to increase at 360nm, and the photoinduced charge amount increased by decreasing measurement frequency. We also could see the obvious peak wavelength of 330 nm on 0.1 Hz. The results suggest that the photoinduced charges are mobile ions which have a mobility in an LC mixture and the light exposure of 330 nm is a sensitive wavelength for a degradation of the LC mixture.

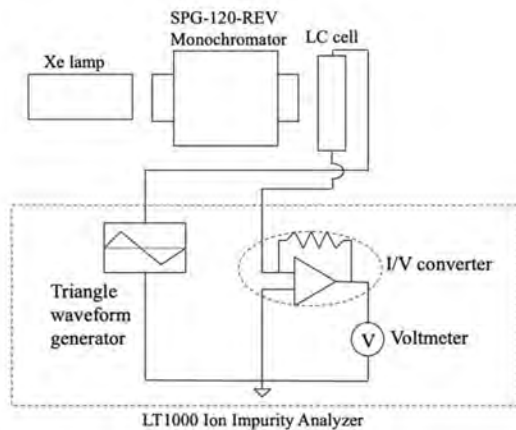


Fig. 1 Block diagram of photoinduced charge measurement

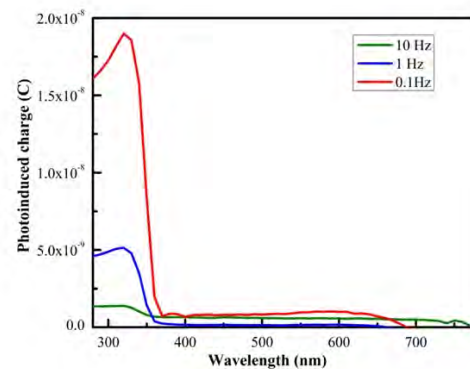


Fig. 2 Wavelength dependency of photoinduced charge on different measurement frequency

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Charge Transport Property of Butterfly-like Shape Liquid Crystals Based Fused-Thiophene as Organic Semiconductors

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In recent decades, great advances in the chemistry of carbon- and heteroatom-based π -conjugated systems have contributed substantially to the emergence of organic electronics and optoelectronics research domains,[1] as evidenced by the large quantity of synthesized semiconducting molecular and macromolecular systems for a variety of technological and medical applications. To improve the mobility of charges in discotic systems, research attentions have focused on modifying the periphery (nature and type of chains) and/or changing the molecular-core symmetry and size. Beside disc-like molecules, there have been recently few reports on board-like molecules, pseudo-discs also possessing large extended π -systems and being able to stack, exhibiting high electronic charge transport mobility competing with those from conventional DLCs.[2] Structurally related butterfly-like shape molecules specifically made from fused-thiophene aromatics also stack up into columnar mesophases, and were found to exhibit outstanding optical properties as well as enhanced electron-conducting properties.[3]

In this study, mesomorphous butterfly-like shape molecules based on cyclopentadithiophen-4-one and benzodithiophene-4,8-dione core moieties were synthesized. These butterfly-like shape molecules spontaneously self-organize into columnar hexagonal mesophase. The charge carrier mobility was measured by time-of-flight transient photocurrent technique in the mesophases. They display hole-transport performances with positive charge mobility in the $10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ range, consistent with the high degree of ordering and stability of the columnar superstructures. One mesogen in particular with benzodithiophen-4,8-dione core (BDTD) shows outstanding ambipolar charge carrier transport with both high electron ($\mu_e = 6.6 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and hole ($\mu_h = 4.5 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) mobility values.[3]

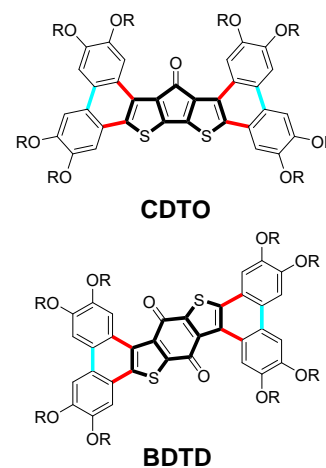


Fig. 1. Board-like and butterfly-shaped fused-thiophene columnar liquid crystalline semiconductors synthesized by successive **Suzuki** (bonds in red) and **Scholl** (bonds in blue) tandem reactions, **BDTD** and **CDTO**, this work (R is alkyl chain, $\text{C}_n\text{H}_{2n+1}$).

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Viscosity probing function of flapping fluorophores and their optical properties in a nematic liquid crystal

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Fluorescent molecular viscosity probes are widely used in materials science and cell biology.^[1] We have recently reported flapping fluorophores, FLAP, that show viscosity-probing functions by the analyses of ratiometric fluorescence (FLAP0)^[2] and fluorescence lifetime (FLAP1) (Figure 1).^[3] These molecules take a V-shaped conformation as a stable form in the S_0 ground state, and they show a V-shaped-to-planar conformational change in the S_1 excited state. Since the excited-state planarization dynamics is considerably suppressed in highly viscous media, local viscosity can be estimated by the analyses of fluorescence properties.

Here we demonstrate that these flapping viscosity probes are more sensitive than BODIPY- C_{12} ^[4], a well-studied molecular rotor working as a fluorescence viscosity probe^[3b], particularly in a low viscosity range of 0.3-3 cP. Moreover, local viscosity of a nematic crystal, 5-CB, has been analyzed by using these flapping fluorophores, and compared the results with the reported bulk viscosity of 5-CB (Figure 2). Temperature-dependent viscosity change will be also discussed in relation to the phase transition behavior of 5-CB.

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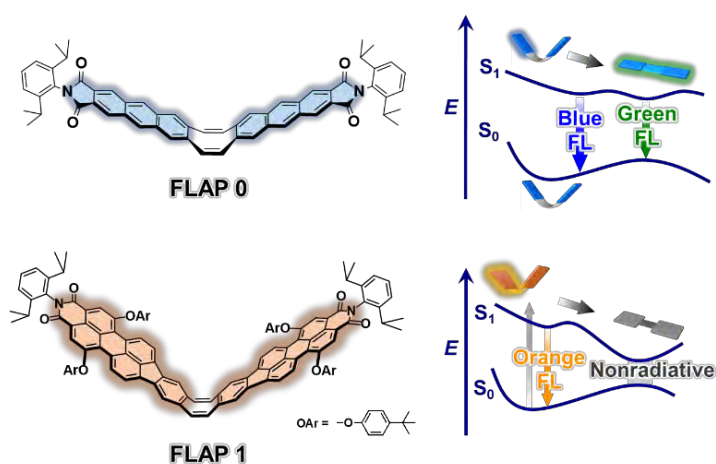


Figure 1. (left) Chemical structures of FLAP0 and FLAP1 (right) Excited state dynamics of each FLAP.

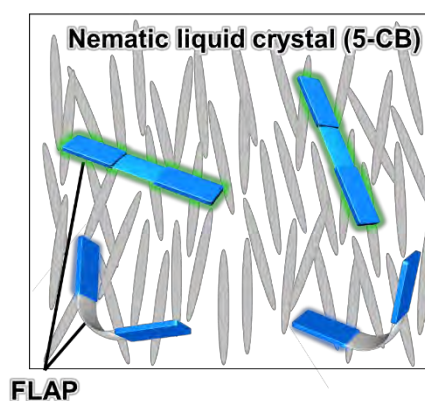


Figure 2. Flapping viscosity probe dispersed in 5-CB.

Comparison of Characteristics between Electro-sprayed Cholesteric Droplets and Cholesteric Cell

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In recent years, a different approach of utilizing liquid crystal which is in droplet form has received much attention from researchers in various fields. In this study, we compare the characteristics of cholesteric liquid crystal (CLC) droplets using electro-spray method with CLC in a conventional cell, and investigate the temperature induced phase transition of the droplets. To fabricate the CLC droplets, we prepared CLC which was a mixture of nematic LC (E44) and 30 wt% of chiral dopant (S-811). We then set up an electro-spray apparatus in which a glass substrate treated with coating agent was placed in between a needle electrode connected to a high voltage source and an earthed plate electrode, as shown in Fig. 1. About a drop of CLC was applied to the tip of the needle electrode and a negative DC high voltage of -5kV . The CLC was sprayed to the glass substrate and droplets that were formed were then observed using a polarized microscope. Figure 2 shows transmittance spectra of CLC in a conventional cell and CLC droplet at 28°C . Although both samples show selective reflection in their transmission spectra, the center wavelengths are different. A more detailed approach to the results and discussion will be presented at the venue.

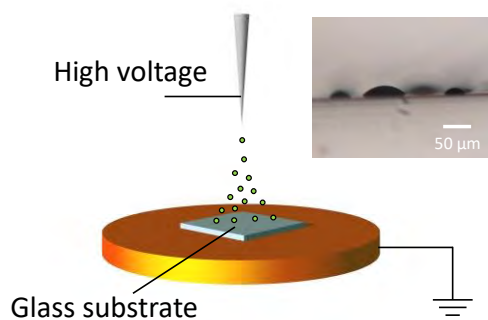


Fig. 1 Schematic of fabrication of electro-sprayed CLC droplets

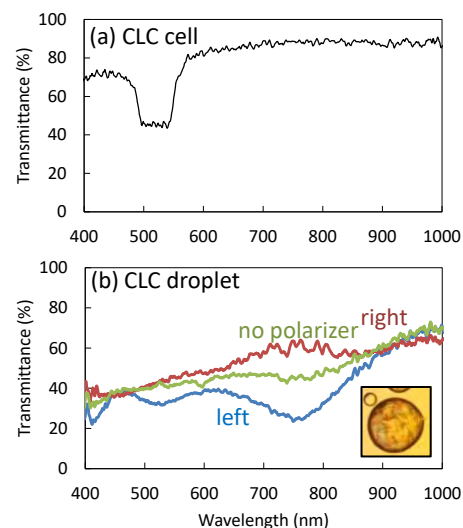


Fig. 2 Transmission spectra of CLC cell and CLC droplet.

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Abstract:

Liquid crystals (LCs) possess long-range molecular interactions, which can be utilized for macroscopic structural change and motion. Due to this macroscopic change, LCs has been regarded as a good platform for an active matter in recent years. Although there have been various studies on the structure, the dynamics information on the LC motion is still lacking. We studied how fast and how long this molecular interaction propagates during a phase transition by observing a change in a refractive index with a high time resolution. ^[1] Here, we clarified the initial dynamics of the molecules leading to the phase transition by the patterned-illumination time-resolved phase microscopy (PI-PM), which could be used for the observation of the dynamic processes via the images of the refractive index change with a high time resolution.

A sample was N-(4-methoxybenzylidene)-4-butylaniline (MBBA) used as purchased and was put into an LC cell (E.H.C) with a rubbed polyimide films inside for the planer alignment. By matching a polarization of an illumination light shined onto the sample and the direction of LC orientation, we could obtain an image of the change in the extraordinary refractive index (Δn_e), and the change in the ordinary refractive index (Δn_o) can be imaged by making them vertical.

Figure 1(a) and (b) show the sequences of images for each change in the refractive index. The white regions in the stripe pattern in Fig. 1(a) corresponds to an increase of the refractive index, while a decrease of the refractive index was observed in the stripe pattern in Fig. 1(b). Fig. 1(c) shows the average time responses for each refractive index change obtained by analyzing the images. Δn_e response rose quickly after the pulse irradiation, on the other hand, Δn_o response started to rise from around 10 ns, respectively. Both of the signals increased rapidly up to 20 ns followed by an further slow increase up to 2 μ s. In the previous study, spatially inhomogeneous LC phase transition was reported in photo-induced phase transition. ^[2] Based on the above results and reference, we suppose that the 1st rising components until 20 ns correspond to a local phase transition process caused by a molecular disordering in the direction parallel to the liquid crystal orientation, and the subsequent 2nd rising components until 2 μ s correspond to an expansion process of the phase transition region.

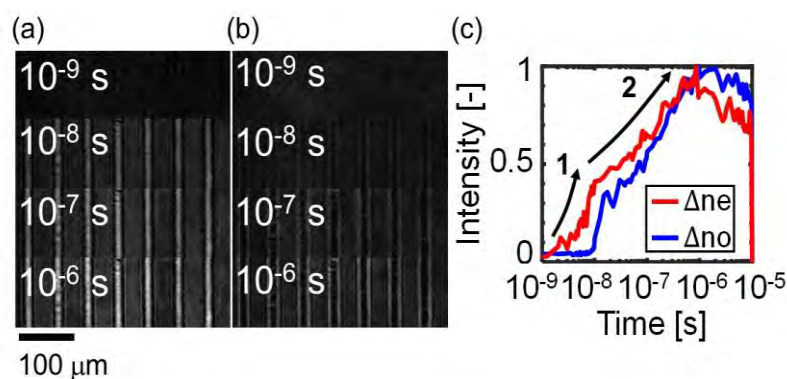


Fig.1 Time sequences of the phase imaging for (a) Δn_e and (b) Δn_o and (c) the corresponding average time responses.

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Polymerization Induced Fluorescence Enhancement of Aggregation-Induced Emission Dye-Doped Liquid Crystals

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Introduction: Aggregation-induced emission enhancement (AIEE) is a phenomenon where the fluorescence intensity of a dye increases on becoming a solid [1]. Recently, a class of liquid crystalline AIEE dyes based on 2-(2-hydroxyphenyl)benzothiazole (HBT) have been synthesized that show high quantum efficiencies with large Stokes shifts due to excited-state intra-proton transfer [1]. Considering the AIEE nature of the dye, further enhancement of the fluorescence can be expected by polymerizing the liquid crystal (LC) host. Here, we employed a photo-polymerizable LC as the host material and performed in-situ measurements of the fluorescence intensity as the host was polymerized.

Fabrication method: The molecular structure of the AIEE dye, HBT-C5Ph is shown in Fig 1. A polymerizable LC mixture was prepared by mixing a nematic mixture (LCC, E7), a reactive mesogen (BASF, LC242), and HBT-C5Ph at a ratio of 69:30:1. A reference material without the reactive mesogen was also prepared. The LC precursor was filled in a sandwich cell with uniform homogeneous alignment and a cell-gap of 10 μ m. The fluorescence intensity was measured as a UV-LED with peak intensity at 385 nm (Thorlabs, M00268607) was irradiated on the cell.

Results and discussions: Figure 2 shows the evolution in the fluorescence spectrum upon irradiation of UV light. The fluorescence intensity of the polymerizable sample (Fig. 2(a)) shows an increase by approximately 18 % with a time constant of 4.5 s, which is not observed in the non-polymerizable reference (Fig. 2(b)). The increase is attributed to the increased viscosity in the polymerized material, which inhibits the molecular motion that causes non-radiative dissipation of energy.

Conclusion: A liquid crystalline AIEE dye was found to increase its fluorescence intensity when the host LC was polymerized.

Acknowledgment: This work was partly supported by MEXT KAKENHI (19H02581, 20H00391, 20K21154) and MEXT LEADER Program.

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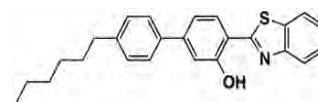


Fig.1 Structure of HBT-C5ph

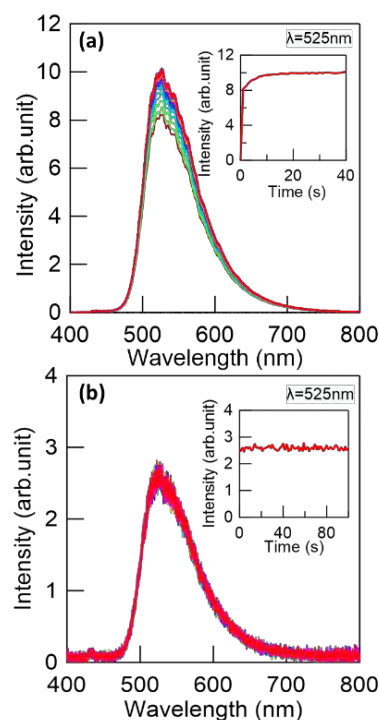


Fig. 2 (a) Fluorescence spectrum of HBT-C5ph doped in the polymerizable LC and (b) non-polymerizable.

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Introduction

Ideal liquid crystal elastomer (LCE) is theoretically expected to deform at no energy cost when the director rotates because of the coupling of LC alignment and rubber elasticity, which is called “soft elasticity”. Real nematic elastomers (NEs) can be elongated uniaxially at very small constant force when stretched at large oblique angles relative to the initial director,[1] which has been considered as an indication of soft elasticity. However, the details of the soft elasticity feature in real NEs remain to be characterized. We investigate the dynamic mechanical properties of the NEs to which a static strain close to the onset of the plateau regime is applied.

Experiment

The thiol-acrylate main-chain NEs with planar- and polydomain alignments (designated as C6CT-M and C6CT-P, respectively) were prepared by using the method in Ref [1]. The rectangular monodomain NEs (C6CT-M- θ) were cut out from the sample sheet so that the long axis had an angle (θ) with respect to the initial director. The dynamic mechanical measurements were performed in the stretched state with various degrees of static elongation (λ_s).

Results and discussion

Figure 1(a) shows the quasi-equilibrium stress-elongation relationships of C6CT-M-90 and C6CT-P. Both specimens indicate quasi-plateau regime over finite strain range which results from the accompanying director rotation toward the stretching axis. Figure 1(b) displays the angular frequency dependence of loss tangent ($\tan \delta$) for each specimen at $\lambda_s = 1.01$ and 1.05. In the linear viscoelasticity regime ($\lambda_s = 1.01$), $\tan \delta$ for each specimen exhibits a definite peak at a characteristic frequency which reflects the dynamics of director. Importantly, a small increase in λ_s induces a considerable increase in the peak height ($\tan \delta_{\max}$) for each specimen, and the $\tan \delta_{\max}$ values reach almost unity. This marked increase in energy dissipation property indicates the soft mode feature of NEs becomes more obvious as the applied stretch approaches the onset value of the stress-plateau regime.

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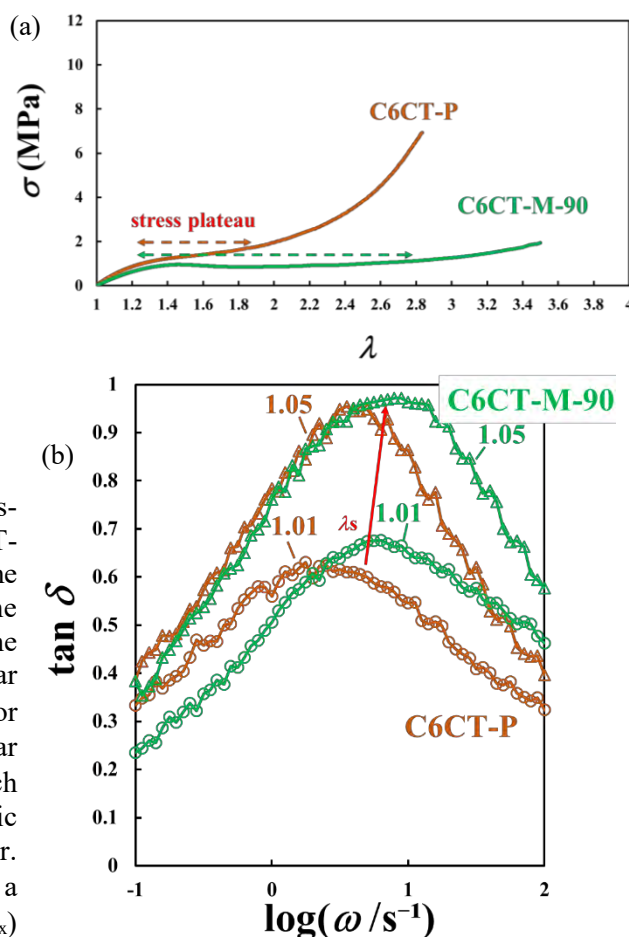


Fig. 1 (a) Nominal stress-elongation relation (b) ω dependence of $\tan \delta$ of C6CT-Poly and C6CT-Mono at various λ_s . The numerals indicate the values of λ_s .

Realization of a Room-Temperature Ferroelectric Columnar Liquid Crystal by Introducing Branched Alkyl Chains.

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Ferroelectrics are materials in which polarization caused by displacement of atoms/molecules or alignment of dipoles is switched by an external electric field and maintained after removal of the field. Though ferroelectrics are applied to actuators, sensors, non-linear optics, most of them are currently based on crystalline materials. Some of them contain toxic lead or rare metal elements. On the other hand, organic ferroelectric columnar liquid crystals (FCLCs) are promising materials that are more eco-friendly and highly processable, and show lower power consumption compared to crystalline materials. Achieving FCLCs is still challenging because electrostatic repulsion occurs between adjacent polar columns in a syn-parallel manner after removal of the field. Further, the temperature ranges exhibiting FCLCs were mostly above 100 °C. In our previous study, thermodynamically stable FCLC compound **1** was generated by introducing chiral alkyl chains because a tightly wound helical structure promotes the polarization maintenance (Fig. 1a). However, the interactions that were too strong inhibited its polarization switching at room temperature [1]. In this study, to realize room-temperature FCLCs, we focused on achieving both polarization maintenance and switching at room temperature.

Introducing branched alkyl chains is an effective approach for lowering liquid crystalline temperatures and expected to suppress the molecular thermal motion and prevent the depolarization. The polarization-switchable temperatures of columnar liquid crystalline compounds **2** (63–143 °C) and **3** (20–165 °C) were lower compared to that of **1** (105–150 °C). Further, second harmonic generation (SHG) measurements revealed that induced polarization after removal of a direct current 100 V was partially maintained, indicating lower-temperature FCLCs (**2**: 63–103 °C and **3**: 20–60 °C) (Fig. 1b). Notably, compound **3** shows an unprecedented room-temperature FCLC and maintains the polarization for at least 18 h. Dynamics of depolarization in compounds **2** and **3** were further investigated by switching current measurement, and depolarization activation energies of **2** and **3** were calculated to be 111 and 104 kJ mol⁻¹, respectively. These energies correspond to that of an FCLC benzene-1,3,5-tricarboxamide (108 kJ mol⁻¹) [2]. Thus, we concluded that **2** and **3** generated kinetically stable (slow relaxation) FCLCs.

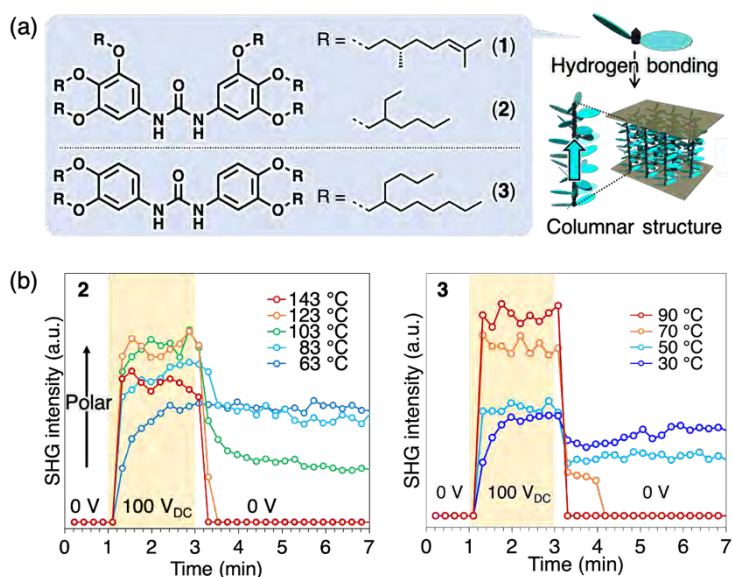


Fig. 1 (a) Molecular structures of **1–3**. (b) Polarization maintenance behaviors of compounds **2** and **3**.

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Synthesis of nanosheet liquid crystal/elastomer nanocomposite by using gel template

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Silicone elastomers are important elastomeric materials in rubber industries. Due to its heat resistance and chemical stability, it is used as a medical device and sealing material. To improve mechanical properties and gas barrier properties, compounding inorganic fine particles with silicone elastomers has been investigated.¹⁻³ Especially, the inorganic nanosheets obtained by exfoliation of a layered material is attracting attention as anisotropic nanofillers.⁴⁻⁹ However, it is generally difficult to obtain completely exfoliated nanosheets in a non-aqueous solvent that is required for ideally compounding with silicone elastomer. Here, we investigated the synthesis of liquid crystalline nanosheet/silicone elastomer nanocomposite by using the nanosheet liquid crystal composite gel as a template.

An elastomer sample without nanosheets was synthesized by heating the mixture of monomer (diethoxydimethylsilane, tetraethoxysilane) and the catalyst (water, dibutyltin diacetate). As a result, a transparent, strong, and elastic sample was obtained. To synthesize the liquid crystalline nanosheet/silicone elastomer nanocomposite, a composite gel of nanosheet liquid crystal and polydiethylacrylamide (pDEAA) was first prepared. The gel was then impregnated with the monomer solution. The gel remained transparent at this stage, indicating that the introduction of the silicone monomer into the gel was successful. The polymerization of silicone monomer in this monomer-impregnated gel is now underway and the results will be presented at the poster session. The formation of homogeneous composites is expected after optimization of polymerization conditions.

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Self-organization in a nematic phase of Au nanorods modified with liquid-crystalline organic dendrons

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Gold nanorods (AuNRs) show a unique localized surface plasmon resonance (LSPR) depending on their sizes, aspect ratios, and interparticle interactions. The absorption of AuNRs derived from LSPR changes when AuNRs are aligned in an ordered structure, which can result in an enhancement of electric field between AuNRs. On the other hand, liquid-crystalline (LC) organic dendrons are one of the representative materials to form self-organized LC structure. Therefore, we focused our attention on the dendron modification of AuNRs to form AuNRs-based thermotropic LC structure,^{1,2)} showing structure-dependent dynamic control of their optical properties, by introduction of the self-assembling property into AuNRs.

Dendron-modified AuNRs were synthesized in a following two-step procedure. Firstly, cetyltrimethyl ammonium bromide-stabilized AuNRs (CTAB-AuNRs) were prepared. Then, CTABs on the surface of AuNRs were exchanged by 16-mercaptohexadecanoic acid (MHA) having a COOH-moiety to produce COOH-modified AuNRs (COOH-AuNRs). Fig. 1 shows a molecular structure of NH₂-substituted dendron used in this study. Secondly, surface modification of the COOH-AuNRs with the dendrons were performed by forming an amide bond between the COOH-groups on the surface of the AuNRs and NH₂-moieties at the terminal of the dendron molecules in the presence of a condensation agent. The resulting dendron-modified AuNRs (dendron-AuNRs) were characterized by TEM, UV-Vis and SAXS.

Fig. 2 shows TEM images of CTAB-AuNRs and dendron-AuNRs. The size of AuNRs was 34.6 ± 3.9 nm in the longer axis direction and 12.9 ± 1.4 nm in the shorter axis direction. Here, the surface-to-surface distance of CTAB-AuNRs was 3.5 nm, while that of dendron-AuNRs expanded to 4.2 nm by dendron-modification. Therefore, the dendron-modification of AuNRs successfully proceeded, maintaining the mainlining the rod shape. For the evaluation of thermally orientation of dendron-AuNRs, temperature-dependent SAXS measurements were carried out. Three broad scattering peaks (d_1 , d_2 , and d_3) derived from the shorter axis of the AuNRs were observed at 30 °C. As the temperature rose, they were shifted to the small angle side. Because the reciprocal ratio of d_1 , d_2 , and d_3 at 150 °C is found to calculate as 1: 2: 3, our dendron-AuNRs must form a nematic phase above 150 °C.

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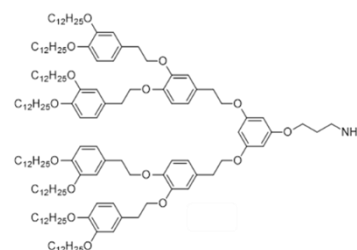


Fig. 1 A molecular structure of NH₂-substituted dendron.

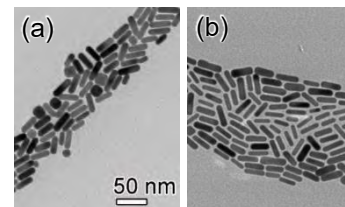


Fig. 2 TEM images of (a) CTAB-AuNRs, (b) dendron-AuNRs. The scale bar in (a) is common to (b).

Optical Properties of Cholesteric-Liquid-Crystal Elastomers with Precisely Controlled Molecular Orientation by Photopolymerization

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Cholesteric liquid crystals (CLC) intrinsically form a helically twisted molecular orientation. Based on the helical orientation, CLC have a periodic distribution of refractive index; thus, they show unique optical properties such as reflection and diffraction. So far, we fabricated elastomers with a three-dimensional polymer network using CLC to create strain responsive optical materials, exhibiting both selective reflection and rubber elasticity. This would have a great potential to be used as mechanosensing materials. For further extending our concept, we have focused on the molecular orientation patterns of cholesteric liquid crystal. In this study, we found that the irradiation of a spatially patterned light to prepare the CLC elastomer by photopolymerization induced a structure with a periodic change of in-plane refractive index, and that the obtained CLC elastomer showed diffraction. Here, we report that the optical and mechanical properties of the CLC elastomer films.

Figure 1 shows the molecular structure of the compound used for fabrication of elastomers. A mixture of liquid crystal monomer (**HAB**), chiral monomer (**CHA**), crosslinking agent (**HDDA**), plasticizer (**5CB**), and initiator (Irgacure 651) was sealed in a glass cell with 6- μm cell gap, and the elastomer was prepared by photopolymerization. The molecular orientation in the elastomer was observed by a polarizing optical microscopy (POM). In the elastomer films, we observed a striped periodic structure (Figure 2a). When tensile strain was applied to the film, the structural period increased. Diffracted light measurements of the CLC film revealed that the diffraction angle decreased in proportion to the tensile strain (Figure 2b).

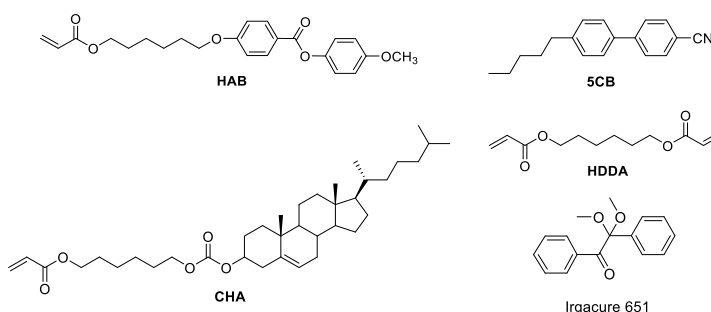


Figure 1. Molecular structures of materials used in this study.

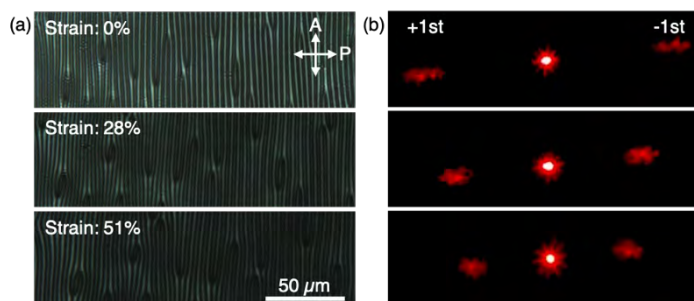


Figure 2. (a) POM images of the CLC elastomer under applied strain. (b) CCD images of the transmitted and diffracted beams incident on the CLC film.

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Cholesteric-Liquid-Crystal Polymer Particles with Controlled Orientation of Helical Axis and Their Application to Deformation Analyses

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Flexible, stretchable soft materials have attracted much attention for realizing soft robots, wearable electronics, etc. One of the large issue for the development of soft materials is quantitative analysis of deformation with large strain region. Recently, digital image correlation (DIC) technique is well-known as a practical method to analyze the deformation behavior. In DIC, a random pattern, called a speckle pattern, is formed by spraying particles on a surface to be analyzed; and then, one can visualize deformation behavior with high spatiotemporal resolution by monitoring the displacement of the speckle patterns. Nowadays, two-dimensional analysis by DIC enables to quantitatively investigate deformation behaviors even with complexed shape changes such as bending and twisting. For further development, three-dimensional analysis of deformation has been highly desired.

As an origin of speckle patterns, cholesteric liquid crystals (Cho LCs) might be useful. The Cho LCs form helically twisted molecular alignment, and exhibit selective reflection at a certain wavelength depending on its helical pitch. Since the helical pitch of Cho LCs can be easily controlled, Cho LCs have a potential application to multi-colored speckle pattern, allowing us to monitor the three-dimensional deformation of soft materials by the DIC technique. However, typical Cho LCs show angular-dependent reflection; thus, the color of speckle patterns depends on viewing angles, interfering with monitoring the three-dimensional deformation by the multi-color DIC. Recently, we have succeeded in preparing a monodispersed polymeric Cho-LC particles with radial orientation of the helical axis. The particle with the radially oriented helix exhibited omni-directional selective reflection, and the speckle pattern formed by the Cho-LC particles can be visualized without any angular-dependence. In this study, the Cho-LC polymer particles were dispersed in a film of polydimethylsiloxane, and deformation behavior of the film was observed. We demonstrated that the Cho-LC polymer particles with the radial helical-axis orientation is useful for the multi-color DIC to monitor the three-dimensional deformation of soft materials.

Inorganic nanosheets are plate-like particles having a high aspect ratio and a large specific surface area. They are obtained by exfoliation of an inorganic layered crystal and it is used as a polymer filler, etc. Silylation of nanosheets is effective to modify the property and functions of nanosheets. However, it is still challenging to precisely control the modification ratio and to obtain highly exfoliated modified nanosheets.¹ Here, we demonstrate a novel silylation process of nanosheets to overcome these problems by using a degradable polymer gel.

According to the previous report, 0.5wt% hexaniobate nanosheets/water colloids were synthesized.² The monomer (acrylic acid; 6.44×10^{-3} mol), degradable cross-linker (3,9-divinyl-2,4,8,10-tetraoxospiro[5.5]undecane; 6.44×10^{-4} mol) and photopolymerization initiator (2-hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone; 5.47×10^{-4}) mol were dissolved in hexaniobate/water colloid, followed by photopolymerization to immobilize the nanosheets in the polymer hydrogel. The solvent of the hydrogel was exchanged with 0.01M HCl to exchanged the original counter cation of the nanosheets propylammonium to proton. The solvent was then exchanged with *N,N*-dimethylformamide (DMF). Then, the DMF solution of a silylating agent (3-mercaptopropyl(dimethoxy)-methylsilane) was introduced into the gel, and the silylation reaction was allowed to proceed by heating at 100°C for 7 days. After the reaction, the gel was sealed in a pressure-resistant container with water and heated at 140°C for 6 h to decompose the gel and recover the silylated nanosheets.

In the IR spectrum of the unmodified nanosheet (Fig. 1a), the peaks due to propylammonium (the counter cation of the nanosheet) is observed. After the proton-exchange, silylating reaction and decomposition/removal of the acrylic acid gel (Fig. 1b), stretching vibrations of Nb-O-Si (●:1016 cm^{-1}), Si-O-Si (○:1085 cm^{-1}), Si-CH₃ (◎:1259 cm^{-1}) and C-O (⊙:1463 cm^{-1}), while the amide group C=O stretching vibration (1720 cm^{-1}) due to acrylic acid gel disappeared. The peak of PA⁺ disappeared due to ion exchange. Thus, grafting of the silylating agent and gel degradation were confirmed.

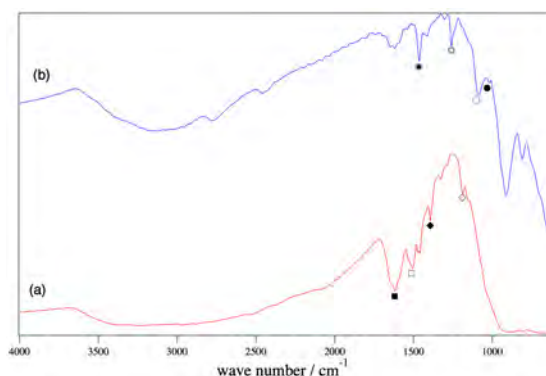


Fig.1 IR spectra of (a) original PA⁺-Nb₆O₁₇ nanosheet (b) H⁺-Nb₆O₁₇ silylated nanosheet

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P42

Title : Novel Liquid Crystal Droplets for the Detection of Bacterial Toxins

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Typically, liquid crystals (LCs) are most well known for their use in LC displays (LCDs). In recent years however, LC droplets have received increased exposure as a result of their numerous applications, including biosensing. Research into LCs as biosensors is driven by several factors, namely the sensitivity of LCs to physical and chemical reactions at the surface and the resultant change in anchoring strength, leading to changes in the optical properties of the LC¹. Additionally, the birefringent properties of LCs mean that no labelling of the target biomolecule is required to produce an optical signal. Their potential for low-cost, sensitive sensors make them extremely interesting to study².

Here we describe a biosensing system incorporating phospholipid-coated nematic LC droplets immobilised in a 1wt% agarose gel. Monodisperse lipid-coated LC droplets of the order of $\sim 10\mu\text{m}$ are produced using microfluidic devices with a flow-focusing configuration. Disruption of the lipid monolayer produces an alignment switch from homeotropic (radial) to planar (bipolar). Such switching of the LC alignment produces a measurable optical response when observed using POM as demonstrated in Fig 1. We describe said switching of LC droplets when suspended in an agarose gel matrix, which takes us a step closer to our goal of a dipstick biosensor. Furthermore, we describe the immobilisation of chiral LC droplets in the same gel matrix, with the view to utilise the selective reflection properties of chiral LCs to create biosensors capable of producing colour changes visible with the naked eye.

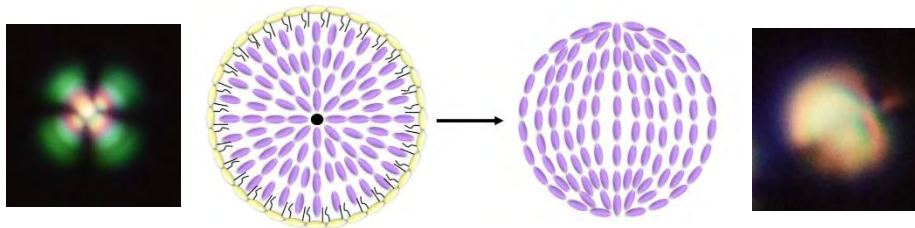


Fig. 1 Switch in alignment of an LC droplet from homeotropic (radial) to planar (bipolar) upon disruption of the lipid monolayer.

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A viologen salts are functional compounds that responds to various external stimuli such as electric field and light, and may exhibit liquid-crystalline (LC) properties depending on their molecular design. In particular, the ionic site in viologen salts have a large effect on the LC properties since the ionic LCs undergo phase formation under the influence of both van der Waals forces and electrostatic interactions [1]. Therefore, the molecular structure of ionic site must be carefully designed. Recently, it has been reported that π -extended viologens exhibit a SmA phase [2], and that a large difference in the length of the alkyl chains at both ends of the mesogens results in a decrease in the LC-isotropic phase-transition temperature ($T_{LC-Iso.}$) [3]. We have synthesized two kind of viologen salts, as shown in Fig. 1 (a) and (b), for application in photoresponsive materials. **12NN12Tf₂N** and **12NPN12Tf₂N** which have symmetric molecular structures, exhibited SmB phase. In this study, we designed and synthesized asymmetric viologens to obtain extended viologens with low order in the LC phase.

An asymmetric **12NNP12Tf₂N** obtained in this study exhibited SmA and SmC phases. The crystalline-LC phase-transition temperature ($T_{Cryst-LC}$) of **12NNP12Tf₂N** was confirmed to be below room temperature. **nNNP12Tf₂Ns** ($n = 8, 10, 12, 14, 16$) varied the length of the alkyl chain attached to the pyridine to increase the asymmetry were synthesized. SmA and SmC phases were observed in the **nNNP12Tf₂Ns** ($n = 8, 10, 12, 14, 16$). UV light was irradiated on **12NNP12Tf₂N** sandwiched in a glass cell. When **12NNP12Tf₂N** was irradiated with UV light above 60°C, which is the temperature of the LC phase, a dark field was observed in the polarized optical microscopy.

Acknowledgment This work was supported by JST CREST Grant Number JPMJCR2095, Japan

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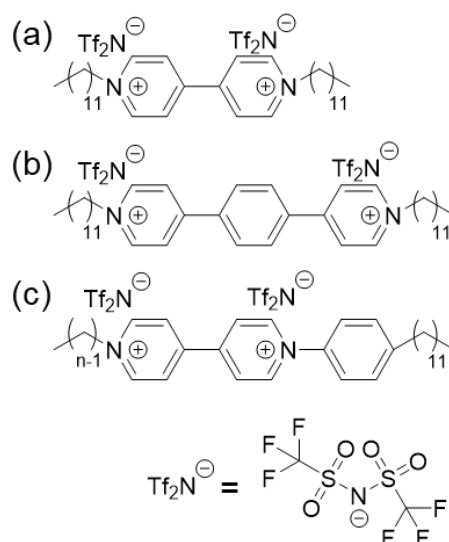


Fig. 1. Structures of viologen compounds. (a) **12NN12Tf₂N**, (b) **12NPN12Tf₂N**, and (c) **nNNP12Tf₂N**.

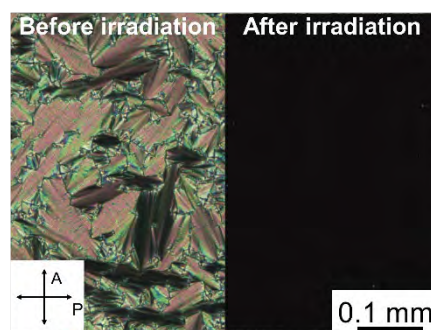


Fig. 2. A polarized optical micrograph of **12NNP12Tf₂N** films before and after UV irradiation (60 °C, 19 mW/cm², 180 min).

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Flapping molecule (FLAP) has a force-probing function with a reversible fluorescence spectral change (Figure 1). This function of FLAP is based on a ground-state conformational change without covalent bond cleavage, and the mechanical response has been studied in crystalline^[1] and polymeric^[2] environments.

However, conventional anthracene-based FLAP (**FLAP1**) cannot probe local force in the presence of solvent molecules (namely, in solution or gel) due to spontaneous planarization in the excited state.^[3] In this study, we synthesized a new FLAP force probe bearing two pyrene units (**FLAP2**) that showed a dual fluorescence even in solution, emitted from both the bent and planar forms.

First, **FLAP2** was chemically incorporated into main chains of a polyurethane elastomer without solvents, and the stretch-induced fluorescence response was demonstrated during tensile testing up to 8.7-MPa stress (Figure 2a,b). Next, an organogel were prepared by swelling the polyurethane elastomer with mesitylene. Although the breaking stress of the organogel was low (0.57 MPa), stretch-induced fluorescence response of **FLAP2** was observed (Figure 2c,d). These findings will offer a new approach to quantification of local stress concentration in soft materials.

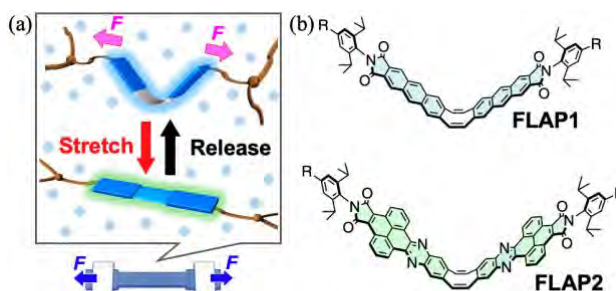


Figure 1. (a) Flapping force probes incorporated into a polymer chain. (b) Chemical structures of anthracene-based FLAP (**FLAP1**) and pyrene-based FLAP (**FLAP2**).

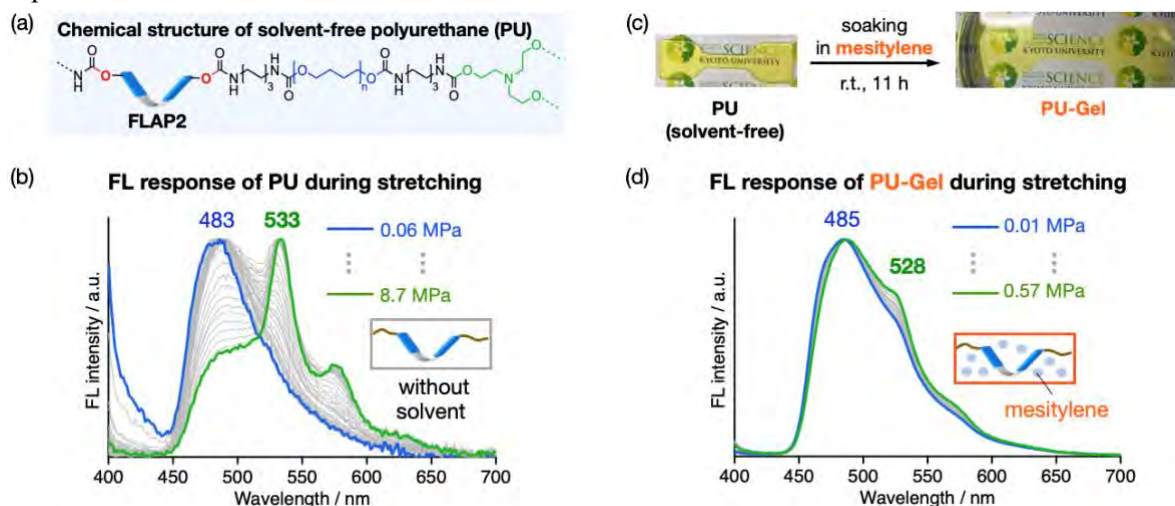


Figure 2. Chemical structure and fluorescence (FL) response of the polyurethane before and after swelling.

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P45

Uniformly oriented cast film of hydroxypropyl cellulose liquid crystal

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It is known that HPC aqueous solution has a cholesteric liquid crystal phase in which the light of a certain wavelength can be reflected selectively. The molecules in the cholesteric phase align as a spiral structure that gives rise to the selective reflection. The dominant wavelength of the reflection coincides with the pitch of the spiral. And a bright and colored reflection depending on the pitch can be observed.

One of the main problems is how to make a sample with uniformly well-oriented molecules. So far, samples have been prepared by applying pressure to the liquid crystal, injecting it into a test-cell and aligning it with rubbing stress with surface of the cell. This conventional cell is made of glass. The reflected light of this cell, however, was not circularly polarized and the orientation of the cell was not very good.

In this study, liquid crystal cast films with high crystallinity and good alignment was prepared and compared with the conventional cells. By drawing the liquid crystal on the slide glass slowly using gravity, a sample with very good orientation was obtained. The reflected light of this cast film is circularly polarized and shows clear optical activity. Furthermore, as a result of polarization analysis, uniform and large optical anisotropy was observed throughout the cast film. The observation results with a polarizing microscope are shown in Fig.1. The left photos are the result of a sandwich cell and the right photos are the result of a cast film, respectively.

Clearly uniform anisotropy is observed in the cast film. From these results, it was found that the orientation of the cast film was very good.

Since cast film is open on one side, addition and removal of additive ions can be easily performed, so application to variable color devices can also be expected.

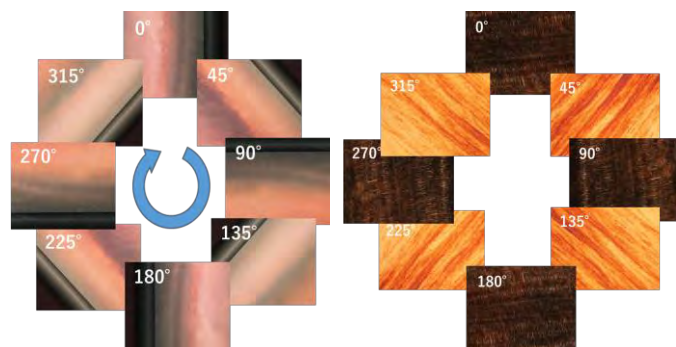


Figure 1: Polarized microscope observation of the films.

Improvement of Switching Speed in Liquid Crystal Millimeter Wave Phase Shifter Under Bias Magnetic Field

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Millimeter-Wave (MMW) is expected to be used for next generation high speed wireless communication standard. Since the MMWs are extremely high frequency region, they have physical characteristic of strong straightness like a light beam. Then, control devices such as phased array antenna are indispensable for MMW applications. Phase shifter is the most important element for phased array antenna. Some liquid crystal (LC) MMW phase shifters have been reported, which are expected as low power consumption and low cost device.^[1,2] We have investigated LC MMW phase shifter that has simple structure by replacing a part of dielectric of microstrip line (MSL) with liquid crystal.^[3] However, since this device has very thick LC layer with several hundred μm , it has very long recovery time. In this study, we investigated operating characteristics of the LC MMW phase shifter under bias magnetic field to improve the recovery speed.

Fig.1(a) shows the structure of LC MMW phase shifter. This device consists of MSL substrate and dielectric substrate including electrode. After combine both substrates, LC material is injected into the cavity fabricated in the MSL substrate. Fig.1(b) shows LC molecular orientation state under bias magnetic field and electric field, which corresponds to ON or OFF state. ON and OFF states are switched by electric field under a constant and small enough bias magnetic field. Magnetic field is applied parallel to the initial orientation direction of the liquid crystal molecules, and electric field is applied perpendicular to the magnetic field. Fig.2 shows the phase change properties of LC phase shifter from ON state (7.9V) to OFF state (0V) under various magnetic biases. It is seen that the recovery speed of this device is improved by increasing applied magnetic field. Recovery time of 290 seconds under 0 gauss magnetic field decreases to 26 seconds under 1000 gauss magnetic field.

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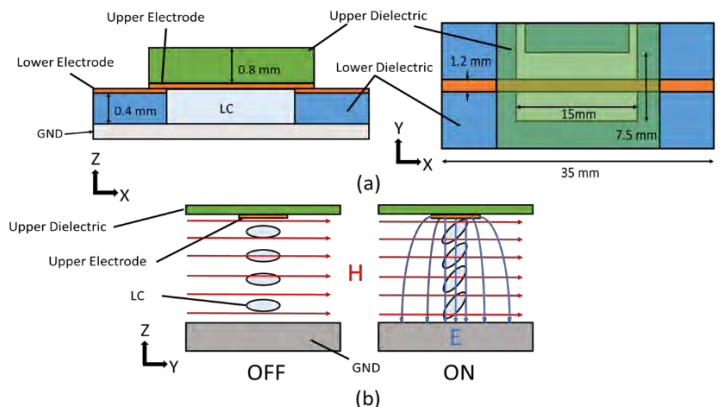


Fig.1 (a)Structure of LC MMW phase shifter, (b)Orientation of LC molecule under ON or OFF state bias.

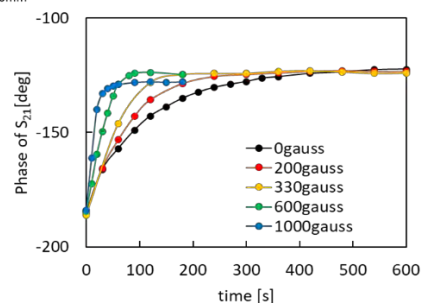


Fig.2 Response property under magnetic bias.

Generation of Focused Vortex Beam by Liquid Crystal Spiral Zone Plate with Homeotropic and Planer Patterned Alignment Layer

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Introduction: Liquid-crystal-based diffractive optical elements (LC-based DOE) are attractive as active and flat optics [1]. LC-based DOEs with patterned alignment such that the azimuthal orientation of the LC rotates around a specific point on a substrate (i.e., q -plates) can generate optical vortices (OVs) for circularly polarized light incident on the device. On the other hand, LC-based DOEs with the LC orientation patterned in the polar angle direction can generate OVs for linearly polarized light. Here, we demonstrate homeotropic and planar alignment patterning on a substrate by depositing a silane coupling agent through a photolithographic approach. A LC spiral zone plate (SZP) is fabricated, and the device is confirmed to show a clear dependence on linear polarization, in contrast to standard q -plates.

Methods: A spiral zone pattern was patterned on a photo-resist film (TOK, OFPR-800 LB) coated on an ITO glass substrate by photolithography. A self-assembled monolayer (SAM) of a silane coupling agent (TCI, Octadecyltrimethoxysilane) was vapor-deposited on the substrate, after which the photo-resist was removed to create regions with and without the SAM. A planer alignment layer (DIC, LIA-03) was then coated on the region without the SAM. A counter substrate with the SAM coated on the entire surface was prepared, and sandwiched to a cell-gap of $\sim 9.0 \mu\text{m}$. A nematic LC material (LCC, E7) was injected into the LC cell.

Results: Figure 1 shows a polarization microscope (POM) image of the sample, which clearly shows regions of homeotropic and hybrid-alignment. A linearly polarized light with a wavelength of 532 nm was incident on the sample, and the diffraction image at the focus of the SZP was observed by a CCD camera (Fig. 2). A vortex beam reflecting the topological charge of the SZP was generated when the azimuth direction of incident linearly polarized light was parallel to the long axis of the hybrid-aligned LC. For perpendicular polarization, the diffraction disappeared, leaving the light unaffected.

Conclusion: A LC SZP with homeotropic and hybrid patterned alignment was fabricated, and used to generate a focusing OV for linearly polarized light.

Acknowledgment: The authors thank DIC Corporation for providing materials. This work was partly supported by MEXT KAKENHI (20H00391), and Osaka University Innovation Bridge Grant.

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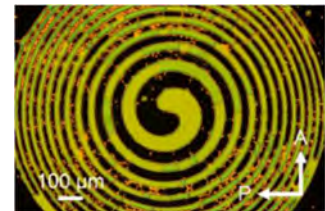


Fig.1 the POM image of the LC SZP

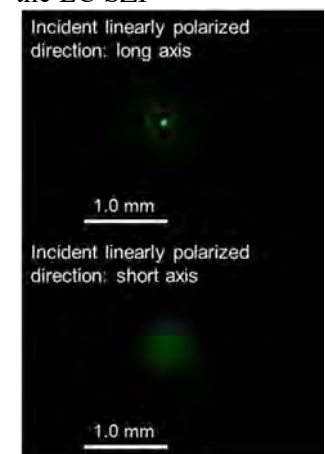


Fig. 2 the diffraction image from the sample

Reverse mode smart window by UV irradiation through hole patterned photomask

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A reverse mode LC cell which is clear in a voltage-off state and a light scattering state in a voltage-on has been proposed. Some mechanisms of the light scattering have been reported, one is by a microlens array system by using hole patterned electrode [1]. Another is a refractive index mismatching between a polymer matrix and liquid crystal domains. Light scattering properties have been also reported by using both light scattering mechanisms [2]. In this study, we have improved a reverse mode property by using two-step UV irradiation method [3] with a hole patterned photomask.

An n-type LC ($\Delta n = 0.237$, $\Delta\epsilon = -5.07$) of 95wt% and reactive mesogen of 5wt% were mixed and the mixture was homeotropically aligned in the cell with the thickness of 9 μm . The hole diameter was 20 μm and the holes occupy 50% of the photo mask area. The first UV irradiation (365 nm, 20 mW/cm^2 , 2 J) was performed through a hole patterned photomask. Following, the second UV irradiation (4 J) was uniformly performed without the photomask. A concentration difference of a polymer matrix in the LC cell is caused by two-step UV irradiation, as shown in Fig. 1. The electro-optical property was measured and compared to that in the cell by conventional one-step UV irradiation (6 J). Threshold voltage of the LC cell using photomask was lower than that of a LC cell without photomask, as shown in Fig. 2. The polymer poor region causes the low threshold voltage. Moreover, threshold voltage difference between polymer rich and poor regions causes the micro lens array effect, which might supply the light scattering.

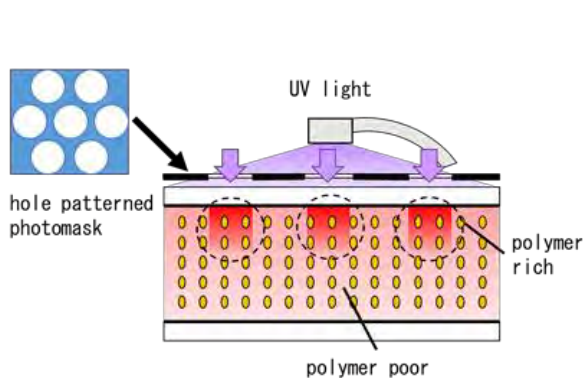


Fig. 1 Schematic of UV irradiation with photomask

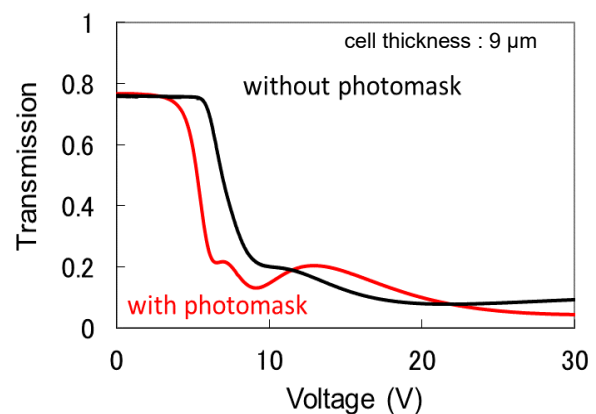


Fig. 2 Electrooptical properties of reverse mode LC cell

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Elastic constant of Hydrogen Bonded Liquid Crystals Fashioned Between 4-*m*-Alkyl Cyclohexane Carboxylic Acids and 4-*n*-alkylbenzoic Acids (*n*BA)

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Recent years, hydrogen bonded liquid crystals (H-bonded LC) have been applied to millimeter-wave and terahertz applications due to a small absorption coefficient or its anisotropy in those regions [1]. The H-bonded LC cell is used by applying AC (~ kHz) voltage as well as a conventional LC device. However, there are a very few papers reported physical properties of H-bonded LC which are required for the electrical driving. In this study, we have investigated physical properties especially bend, twist and splay elastic constants, K_{11} , K_{22} and K_{33} , of H-bonded LCs from the viewpoint of the device application.

A series of H-bonded LCs fashioned between 4-*m*-alkyl cyclohexane carboxylic acids (CCA) and 4-*n*-alkylbenzoic acids (*n*BA) are used, as shown in Table 1. The concentration of H-bonded LC mixtures is controlled to be a nematic phase in room temperature. The LC with *n* of 3 only showed a smectic phase. Dielectric constant and transmittance were measured in both homogeneous and twisted oriented cell as a function of applied voltage (1 kHz), then the theoretical curve was fitted to measurement results to obtained physical properties, for example of *n*=5 as shown in Fig. 1, in which fitting parameters of Δn , $\Delta\epsilon$, K_{11} , K_{22} and K_{33} were 0.13, 0.74, 20, 8 and 29, respectively. All $\Delta\epsilon$ showed positive and was lower than 0.8. We obtained K_{11} , K_{22} and K_{33} of H-bonded LC mixtures with different alkyl chain length *n*, as shown in Fig. 2. K_{11} and K_{33} showed odd-even effect of alkyl carbon number. K_{33}/K_{11} , decreases with *n*. K_{22} slightly increases with *n* and odd-even effect did not clearly show. K_{22}/K_{11} is about 0.3-0.5, which is small compared to usual LCs of about 0.5-0.7.

Table 1 H-bonded LC mixtures with *m*CCA and *n*BA

3CCA	4CCA	5CCA	2BA	<i>n</i> BA (<i>n</i> =2, 4-10)
16.6	16.6	16.6	20	30 wt%

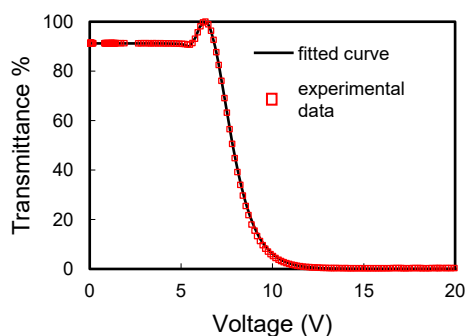


Fig. 1 Theoretical VT curve fitted to measurement result in the TN cell with *n* of 5.

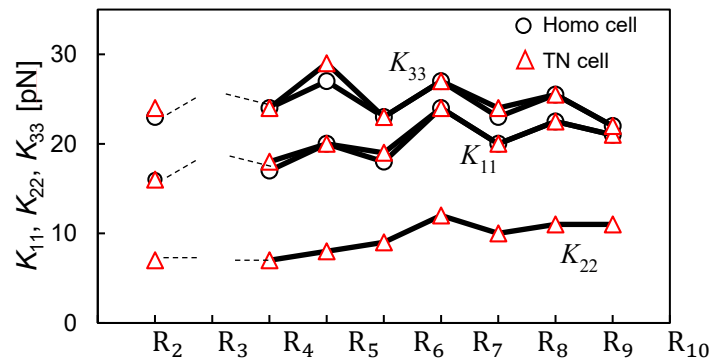


Fig. 2 K_{11} , K_{22} , K_{33} of H-bonded LC mixture with different alkyl chain length *n*.

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Polydimethylsiloxane (PDMS), characterized by high optical transparency and low surface energy is widely recognized as an interesting, high-quality organic material for micro- and opto-fluidic functional systems. Another advantage of applying PDMS is its cheap and easy processing. PDMS-based photonic devices are typically fabricated with use of standard soft-photolithography. In this way, complex microstructures can be successfully manufactured at much lower cost when compared to traditional glass- or semiconductor-based components for integrated optical systems. Moreover, air-channels formed in PDMS can be easily filled with liquid materials, what is a principle of optofluidics. In our work traditional fluids are replaced with liquid crystals (LCs) and advantage of proposed solution stems from high anisotropy of such materials, whose properties can be easily tuned by external electric field (due to high electrooptic effect), as well as by high-power optical fields (due to their significant optical nonlinearity). In this context electrical and optical tuning along with specific polarization properties of LC:PDMS elements are easy to be demonstrated. In this communication several designs and practical realization of LC:PDMS tunable devices together with different cheap and efficient methods for mold fabrication are presented. In addition, possibility of eutectic liquid electrodes application is shown.

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P51

Molecular liquid crystal adhesive that melts by light: Mechanism elucidation of the photomelting function

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Recently we have demonstrated a new application of photoreactive columnar liquid crystals, that is, temporary adhesive that melts by light.¹ Here, we have developed a new light-melt adhesive composed of a flapping triphenylene (TP-FLAP)-based columnar liquid crystal.² Unlike the previously reported light-melt adhesive based on anthracene units,¹ the TP-FLAP adhesive does not require any photoreaction for the photomelting event, but shows excited-state aromaticity. The adhesive strength is recovered simply by stopping the light irradiation. Here, molecular-level mechanism of the macroscopic photomelting behavior is discussed based on the method of the ultrafast electron diffraction analysis.³

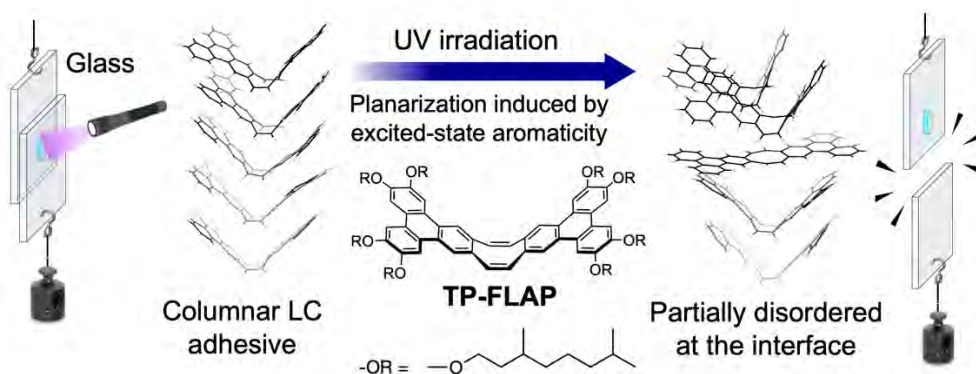


Fig. 1 Light-melting diagram by flattening of TP-FLAP.

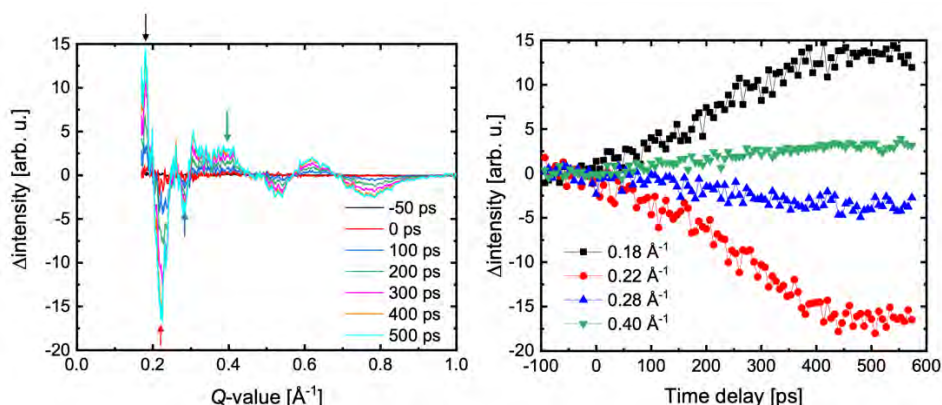


Fig. 2 UV-pulse-induced electron diffraction analysis of the TP-FLAP adhesive.

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A triphenylene derivative which has six 4-tetradecyloxyazobenzene units connected via propylene-ester linkages exhibits both calamitic and discotic LC phases [1, 2]. Furthermore, it is so interesting that this bimesomorphy of LC phase transitions is realized in a photonic as well as a thermotropic manner. Therefore, Sm-Col phase transition mechanism is so much important to consider its functional properties. Recently, it was found that by synchrotron radiation XRD at SPring-8 for the aligned samples, the Sm-to-Col phase transition takes place in relation to a face-to-face interaction among triphenylene rings within a layered structure of Sm phase and this indicates that it is strongly affected by the length of alkyl chains as shown in Fig. 1 [3]. In this communication, phase transition behavior of the alkyl homologues is shown to compare to that of the thermal phase transitions.

Fig. 2 shows phase diagram of **1-C12-3** and **1-C14-3** under the photoirradiation ($\lambda = 365$ nm). For **1-C12-3** no specific temperature was found at which Col-SmA-Iso isothermal phase transition is controlled by photo-intensity and only Col-Sm phase transition was observed, while **1-C14-3** exhibits such an isothermal photo-induced phase transition sequence.

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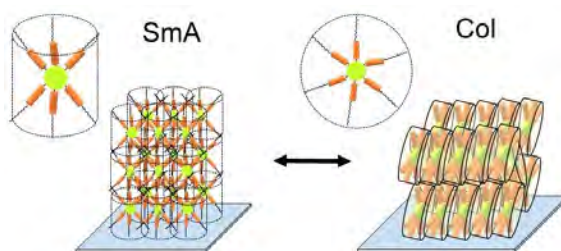
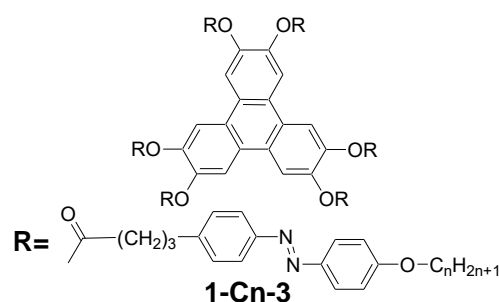


Fig.1 Indicated Sm-Col phase transition with molecular con-formation change.

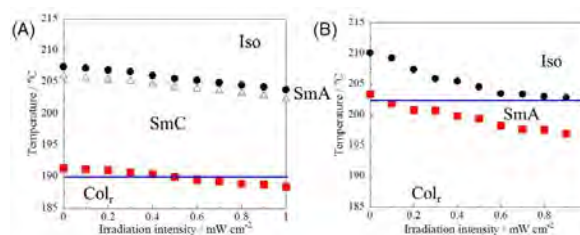


Fig.2 phase diagram of (A) **1-C12-3** and (B) **1-C14-3** under the photoirradiation ($\lambda = 365$ nm).

Synthesis of Liquid-Crystalline Viologens Exhibiting Photo-Responsive Properties

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Viologens exhibits a reversible redox reaction with color change when an electric field is applied. The most stable state of a common viologen is a colorless dication, which can be reduced to a blue-violet radical cation by one-electron reduction and to an orange quinoid state by two-electron reduction. The one-electron reduction can also be induced by UV irradiation. In other words, viologens also function as photo-responsive sites. Moreover, several cases have been reported in which liquid crystalline (LC) phase was observed by designing the molecular structure such as the length of alkyl chains and the counter anions [1]. If viologens are given LC properties, the change in alignment of mesogens may be induced due to the changes in intermolecular interactions by photoirradiation. In this study, we synthesized the LC viologens (**nNPNn-X** (**n**: length of alkyl chain, **NPN**: rigid moieties in viologen, **X**: counter anion)) shown in Fig. 1(a), in which an aromatic ring was introduced between two pyridine rings, in order to obtain LC viologens with both high birefringence and photo-responsive properties.

The obtained **nNPNn-Tf₂Ns** ($n = 10, 12, 14, 16, 18, 20$) exhibited a SmB phase. **12NPN12-Tf₂N** films in a glass cell were observed by a polarized optical microscope (POM) before and after UV irradiation. Under cross polarized light, no change in texture was observed after UV irradiation at room temperature in the crystalline state, but UV-light irradiation at 175°C, which is the temperature range of the LC phase, resulted in a dark-field image. It was found that a decrease in the LC-isotropic phase-transition temperature was observed by photoirradiation. However, the blue-violet color caused by the formation of radical cations was not observed. Next, we prepared a polymer with the **NPN** moieties in the main chain (Fig. 1(b)). **NPN-polymer** with a large molecular weight was prepared. **NPN-polymer** did not show LC phase. The films were changed from colorless to purple when irradiated with UV light even in air.

Acknowledgment

This work was supported by JST CREST Grant Number JPMJCR2095, Japan

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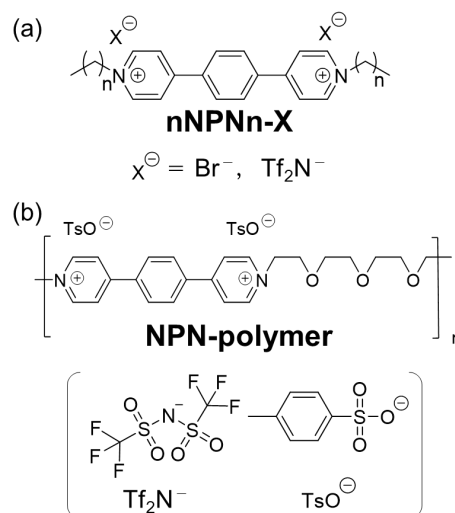


Fig. 1 Chemical structures of LC viologens (a) and viologen-containing polymers (b) synthesized in this study.

Ultrafast Time-Resolved Electron Diffraction Capturing Layer Stacking Dynamics of Graphene Oxide

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We demonstrated ultrafast time-resolved electron diffraction measurements on a nanosheet liquid crystal, graphene oxide (GO), to understand their layer stacking dynamics.

Layer stacking of two-dimensional nanosheet materials, *e.g.*, graphene and transition metal dichalcogenides, is critical in controlling their physical and transport properties. GO also undergoes layer stacking along with its photo- and thermal-induced reduction process; however, the mechanism and dynamics during the layer stacking have not been clarified yet. In previous study, we have reported the ultraviolet (UV) photo-induced reduction mechanism (removal of epoxy-group oxygen) of GO using ultrafast time-resolved electron diffraction measurements [1]. In the present study, we demonstrated ultrafast time-resolved electron diffraction to monitor the structural dynamics during the layer stacking of GO induced by UV photoexcitation. The experimental results are combined with the density functional theory calculations, which reveals that AB-stacking of graphitic domains of GO layers coincides in ~ 40 ps on photo-induced removal of the epoxy-group oxygen from the basal plane of GO *via* overlapping of the π -electronic orbits of GO layers (Fig. 1).

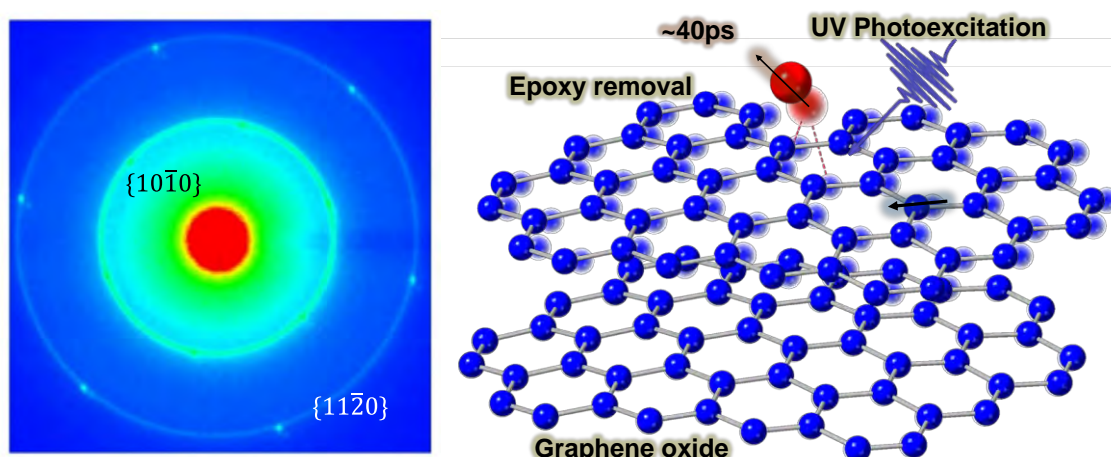


Fig. 1 Electron diffraction pattern from GO thin film (left) and the schematic illustration of the photo-induced layer stacking of GO (right).

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Scanning wave photopolymerization: effect of molecular diffusion of liquid-crystalline monomers on polymerization behavior

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Control of molecular alignment in liquid-crystalline (LC) polymer films is key to developing high-performance devices with optical, electronic and mechanical functions. As a molecular alignment method, application of mechanical stress is the simple and powerful way for fabricating one-dimensional alignment over large area. On the other hand, a photoalignment method has attracted much attention due to its clean and non-contact processes. Photoirradiation with linearly polarized light causes a photochemical reaction of photoresponsive dyes and induce the alignment of the dyes and surrounding LCs depending on the polarization direction [1]. This advanced technology has already achieved various applications such as state-of-the-art displays. Recently, we have proposed a novel photoalignment method based on molecular diffusion caused by the polymer concentration gradient, termed scanning wave photopolymerization [2-4]. In this study, we report a specific polymerization behavior in scanning wave photopolymerization. The effect of molecular diffusion on the photopolymerization behavior was investigated by measuring the molecular weight of the polymers obtained under various photopolymerization conditions.

We prepared a photopolymerizable sample by mixing a methacrylate monomer **M6BACP** and a photoinitiator Irgacure 651 (Fig. 1). The sample was injected into a hand-made glass cell, and irradiated with a scanned UV slit light. Polarized optical microscopy and polarized UV-vis absorption spectroscopy revealed LC moieties in the films aligned parallel to the light scanning direction. The molecular weight and monomer conversion of the resultant polymers were evaluated

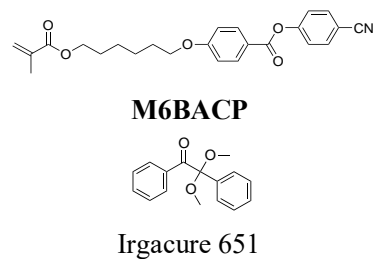


Fig. 1 Chemical structures of the sample used in this study.

by gel permeation chromatography (GPC). The polymer films fabricated by scanning wave photopolymerization and static photopolymerization showed different conversion and molecular weight distribution. These results suggest that molecular diffusion caused a specific polymerization behavior.

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Controlled Formation of Topological Defects of Liquid Crystals in Micro-Chambers

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Topological defects are structural singular points, and they have attracted much attention in the field of liquid crystal (LC) these days. Although the topological defects themselves occupy a small portion of an LC in structure, they could control the overall structure, motion, and activity.^[1,2] They have been studied in terms of the physical properties and extended for practical applications for optics. Since biological cells form their alignment to make topological defects, the concept was extended for understanding the structure and motion of biological phenomena.

We have studied the molecular alignment change of LCs around topological defects from the interests in the molecular interactions there. For a double emulsion made of an LC as a shell, the photo-induced phase transition was induced from a topological defect center.^[1] We have recently developed a new microscope to study the micro-scale behavior of the molecular orientation changes around topological defects and found that the molecular orientation angle and ordering occurred step-by-step, instead of changing them at the same time.^[2] For the basic understanding of the topological defects, it is desired to have an isolated topological defect in a controlled manner. Inspired by the works by Kim and Serra works,^[3,4] there is a possibility that the topological defects could be stabilized by micro-structures, and we have developed a more robust and simple method to produce isolated topological defects with high reproducibility by using pre-designed microfabricated polydimethylsiloxane (PDMS) chambers. The micro-chambers were prepared by capping a glass substrate on PDMS. The topological defects were stabilized by optimizing the size and the shape and satisfying the boundary conditions inside each chamber, which was controlled by the air-plasma treatment on the surfaces. We found the optimal conditions for the reproducible and stable formation of topological defects; a circular shape with its diameter <10 μm and its depth <15 μm and the glass side was treated by air-plasma, providing a right balance of the elastic force to form topological defect at the center. **(Figure)**

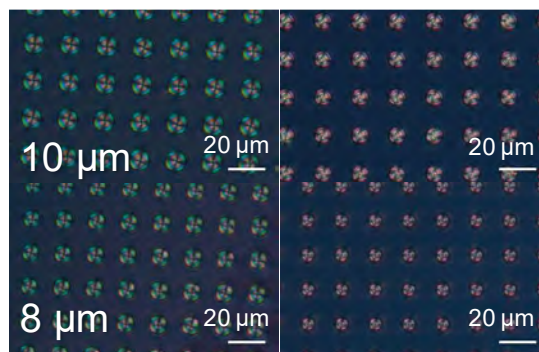


Figure The demonstration of the isolated topological defects using micro-chambers. The number indicated was the diameter of the chamber for each row. The depths were different for each column.

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Banana-shaped liquid crystalline molecules are attracting attention as molecules with ferroelectricity and antiferroelectricity. Many theoretical and simulation studies have predicted phase diagrams on the temperature-bent angle plane [1-3]. However, the phase diagrams of binary mixtures of banana-shaped liquid crystalline molecules with different bent angles have not been reported yet. In this paper, based on the previous theory [3] for nematic ordering of a banana-shaped molecule, we construct a mean-field theory for the two-component systems of banana-shaped molecules.

We calculate the orientational order parameters of the banana-shaped molecules and study the nematic-isotropic phase transitions, depending on the concentration (volume fraction) and temperature. We find novel phase diagrams on the temperature (τ) and concentration (ϕ_1) plane. We also discuss uniaxial and biaxial nematic phases by calculating biaxial order parameters.

Figure1. Phase diagram on the temperature-concentration plane for the binary mixture of banana-shaped molecules with the bent angle $\theta_1 = 90^\circ$ and $\theta_2 = 180^\circ$. The solid curve shows the first-order isotropic (I)-uniaxial nematic phase (N_U^i , $i = a, b, c$) transitions [3]. The closed circle shows the Landau point (LP). Below the LP, the first-order phase transition between two uniaxial nematic phases appears.

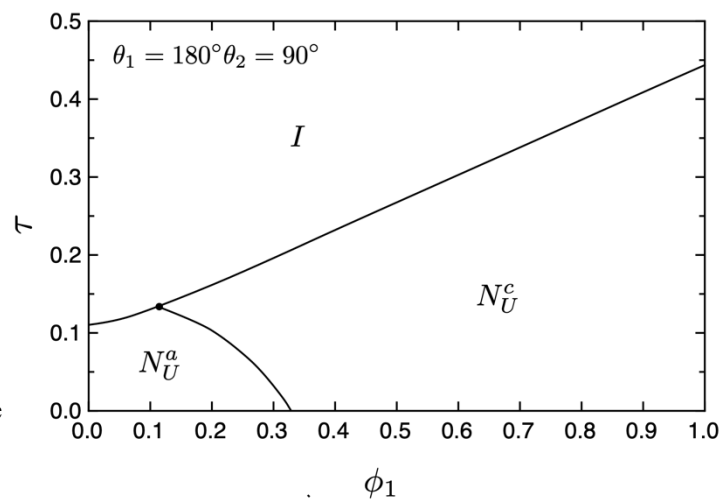


Fig.1

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Electroconvection controlled by patterned electrodes

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Liquid crystal, such as *N*-(4-Methoxybenzylidene)-4-butylaniline (MBBA), under AC voltage is an excellent showcase of pattern formation [1]. Close to the bifurcation point, the parallel roll pattern is observed whose typical length scale is determined by the gap width of a cell, and the pattern with larger wave number is excited while increasing the applied voltage. However, we found that they can be controlled by spatial modulation of the electric fields induced by patterned electrodes. Appropriate spatial modulation of convection will be useful for active smart devices. In this talk, we explore the relationship between the pattern of an electrode and the electroconvection pattern of MBBA.

The pattern of insulated region was given by photoetching of ITO layer, as shown in Fig. 1(a). Here, the diameter of the circles without ITO layer was 50 μm and the lattice constant, the distance between the centers, was 55 μm . We also prepared the pattern with different lattice constants. We used a cell composed of an unetched and a patterned plates, while the gap width was 27 μm . Some regions of the patterned plate were homogeneously coated with ITO. This region is denoted as H-region and used as a reference for the patterned region denoted by P-region. The electrode had uniform planer alignment of MBBA. MBBA layer was sandwiched in the cell, then, AC voltage (peak-to-peak voltage V_{pp}) with 100 Hz was applied. In our condition, a steady parallel roll pattern appeared at $V_{pp} = 19$ V at H-region. When $V_{pp} = 26$ V, the roll pattern showed spatial as well as temporal modulation at H-region, i.e., defect turbulence, as shown in Fig. 1(b). On the contrary, the lattice-like convective pattern was formed at P-region while the temporal modulation was suppressed as shown in Fig. 1(c). This suppression of defect turbulence was effective for the smaller lattice constant of the electrode pattern. In this talk, we will show the effect of the different patterned electrodes on the electroconvection, and will describe possible mechanisms and their applications.

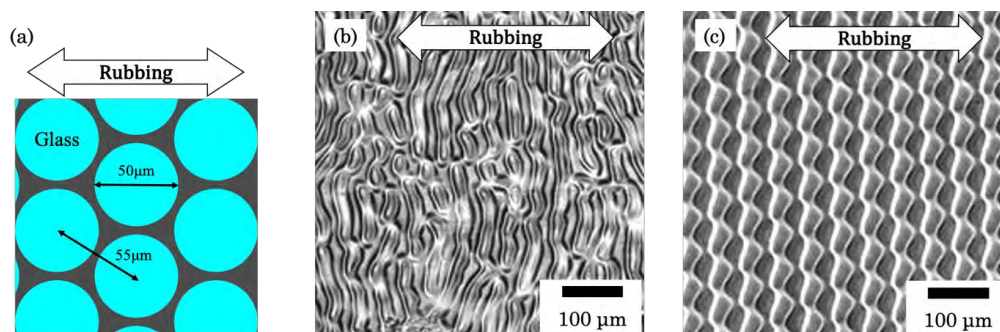


Figure1: Electroconvection patterns. (a) Schematic image of the patterned ITO glass slide. Microscope images of the electroconvection with $V_{pp} = 26$ V at (b) H-region and (c) P-region.

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Nonlinear optical molecular reorientation of oligothiophene-doped fluorinated liquid crystals

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Liquid crystals (LCs) are functional materials that exhibit unique properties such as fluidity and optical anisotropy [1]. Their optical properties vary by controlling the molecular orientation using external stimuli such as light and electric fields. In particular, the oligothiophene dye-doped LCs reorient parallel to the incident polarization direction upon irradiation with a high-intensity laser beam [2-5]. This process, called a photophysical process, occurs only when the LCs are irradiated above a threshold intensity; thus it has attracted much attention for applications as optical switching and light dimming. However, the threshold intensity for the molecular reorientation is so high that practical applications are greatly restricted. In this study, we investigate the effect of host LCs on the nonlinear optical molecular reorientation using fluorinated LCs, which aims to enhance the sensitivity of molecular reorientation.

Host LCs were prepared by adding a fluorinated liquid crystal to 5CB with various ratios. An oligothiophene dye was doped into the host LCs. The mixtures were injected into glass cells whose surfaces were treated with a homeotropic alignment layer. Polarized optical microscopy and polarized ultraviolet-visible absorption spectroscopy confirmed that the LC and dye molecules were homeotropically aligned. When a laser beam was incident on the cells, diffraction rings appeared on a screen based on the molecular reorientation. We revealed that adding the fluorinated liquid crystal to 5CB decreased the light intensity at which the first ring appears, indicating that the sensitivity for the photoinduced molecular reorientation was successfully enhanced.

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