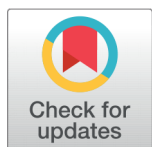


Exploration of Discotic Liquid Crystals for Optoelectronics Applications via Scanning Tunneling Microscope (STM)

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Discotic liquid crystals (DLCs) are a class of liquid crystals that exhibit unique properties that make them attractive for optoelectronic applications. One of their most beneficial characteristics is their charge-carrier mobility, which is facilitated along the stacking axis. This allows for the efficient movement of electrons through the material, making it useful in the construction of electronic devices. DLCs also exhibit self-assembly, self-healing, and solubility in different organic solvents, making them versatile and easy to work with. In this mini-review, we provide an overview of DLCs and their practical applications, with a focus on their charge-carrier mobility (CCM) and alignment. We explore the impact of alignment on the CCM of DLCs and discuss various processing techniques used to achieve alignment, such as zone-casting, Langmuir-Blodgett deposition, solution casting, surface treatment, I.R. irradiation, zone melting, electric field impact, or usage of sacrificial sheets. Understanding and controlling the alignment of DLCs is essential for the successful application of these materials in electronic devices.

Keywords: Discotic liquid crystals (DLCs), Scanning tunneling microscopy, Optoelectronics, Liquid-solid interface, Nanoparticles self-assembly

INTRODUCTION

Discotic liquid crystals (DLCs) are a type of liquid crystal in which the molecules are disc-shaped, with a flat core and a rim that is typically decorated with alkyl chains¹. These molecules self-organize into columnar assemblies that exhibit liquid crystalline behavior, which means that they have properties of both liquids and solids². Unlike conventional nematic or smectic liquid crystals, in which the molecules are rod-shaped or plate-like, respectively, the disc-like shape of the DLC molecules

allows them to form highly ordered stacks of columns that can exhibit long-range positional and orientational order³. The properties of DLCs arise from the interactions between the disc-shaped molecules, which include van der Waals interactions, π - π stacking interactions, and hydrogen bonding⁴. The disc-like shape of the molecules also leads to the formation of polarized charge transfer (PCT) complexes, in which electrons are transferred from the core to the rim of neighboring molecules⁵. These PCT complexes can facilitate charge transport along the stacking direction, making DLCs highly

useful in electronic applications. DLCs can exhibit a range of mesophases, or ordered phases of matter, depending on the temperature and concentration of the material⁶. These mesophases include columnar hexagonal, columnar rectangular, and columnar oblique phases, among others¹⁷. The properties of the mesophase can be tuned by varying the structure of the DLC molecule, as well as the temperature, pressure, and solvent conditions. DLCs have a range of potential applications, including in electronic devices such as solar cells, field-effect transistors, and light-emitting diodes⁸. The highly ordered columnar structures of DLCs can facilitate efficient charge transport, while the self-assembly and self-healing properties of these materials make them easy to work with. Additionally, the solubility of DLCs in various organic solvents makes them highly versatile and compatible with a range of processing techniques⁹.

DLCs have piqued the interest of numerous researchers since their discovery by Chandrasekhar in 1977¹⁰. Considerable investigation and research have been conducted to investigate DLCs in optics and electronic devices. A discotic mesogen possesses a central aromatic core surrounded by three to eight flexible chains⁹. Nematic and columnar phases are the primary dualistic categories of mesophases in DLC arrangement. Nematic phase discs have an orientational direction, whereas columnar phase discs stack into columns based on their orientation. (Figure 1.) Different research groups comprehensively analyze the chemical characteristics of DLCs comprising hexagonal, quadrangular, diagonal, helical, or columnar lamellar flexible configurations¹¹. On the contrary, the initial narrated thermotropic DLCs, indenenes and pseudoazulenes, do not possess flexible chains. Primarily, numerous articles have addressed the strategy and significant DLC configurations and their corresponding features¹². The present

perspective emphasizes the characteristics of DLCs for their future use in optoelectronic approaches, underlining the latest development in the DLCs explored via STM.

Generally, DLCs link via effectual π - π columnlike stacks establishing excessive charge-carrier mobility (CCM). In the columnar stacks, the overlapping of molecular orbitals and the degree of order govern the magnitude of CCM¹³. The charge transference, i.e., in a 1-dimensional stack system, assemble into an arrangement that facilitates film development, which is beneficial for electronic and optoelectronic devices based on organics materials. Additionally, DLC's liquid peculiarity retains its capability to self-heal the structural imperfections (i.e., grain boundaries), making several square millimeters huge size distinct domains of several micrometers¹⁴. Hence, the useful possessions of DLCs comprise long-range self-gathering (directive), processing ease, solvent solubility, self-healing (dynamics), and increased mobility of charge carriers. Anisotropic development driven by microsegregation between flexible chains and rigid cores, or van der Waals attraction between cores, has been proposed as the mechanism by which columnar mesophases grow. (V.W.)^{15,16} Along the column direction, the core-core distance is typically around 3.5Å, whereas the cores center length in adjoining columns is estimated using mesogen size, which generally is about 20-40Å^{17,18}. The flexible alkyl chains around the cores effectively separate column from adjacent column. As a result of DLC columns' increased conductivity along the column axis, they are referred as quasi 1-D conducting wires. There is no doubt that herringbone packing is valid in calamatic L.C.s, which show 2-D charge transport similar to that seen in pentacene or oligothiophene¹⁹. Additionally, DLCs have a greater orbital overlap than calamatics. The arrangement of the discoid

materials' conjugated core determines their electronic properties at the molecular level due to changes in peripheral chains or aromatic cores, which regulate both the solution and the bulk phase self-assembly²⁰. As a result, the core and substituent structure of the materials impact their overall electrical properties. Besides this, the processing technique affects the supramolecular assemblage of DLCs. A brief overview of DLCs and charge mobilities explored via STM is elaborated in the present perspective. Afterward, we highlight specific features linked with DLCs, i.e., configuration and orientation. Then various processing procedures to attain suitable arrangements for efficient, practically applicable device applications were mentioned explored via STM.

Previously, different DLCs have been testified having a considerable number of discoid cores comprising triazine, triphenylene, benzene, pyridine, Hexa-azatrinaphthylene (HATNA), diazatriphenylene, Hexa-azatriphenylene, pyrene, dibenzonaphthacene, dodecaazatrianthracene (DATAN), triazatruxene, or triindole, tricycloquinazoline, perylene, coronene diimide, phthalocyanine (Pc), porphyrin, quinoxalinophenanthrophenazine (TQPP), and Hexa-perihexabenzocoronene (HBC), applicable for various optoelectronic applications²¹⁻²³. The self-assembly and interaction of DLCs were extremely impacted by flexible chains and the central discoid core. For instance, columnar mesophases cannot be formed by substituting six alkylthio or alkyl groups in hexaazatriphenylene. However, mesophase behavior is induced by H-bonding side groups²⁴. H-bonding networks among benzamide side-chain clusters of a phthalocyanine derivative were attributed to the formation of columnar aggregates defined by numerous research group²⁵⁻²⁷. The computational studies attribute this behavior to the discoid cores' intermolecular electrostatic potential²⁸. It

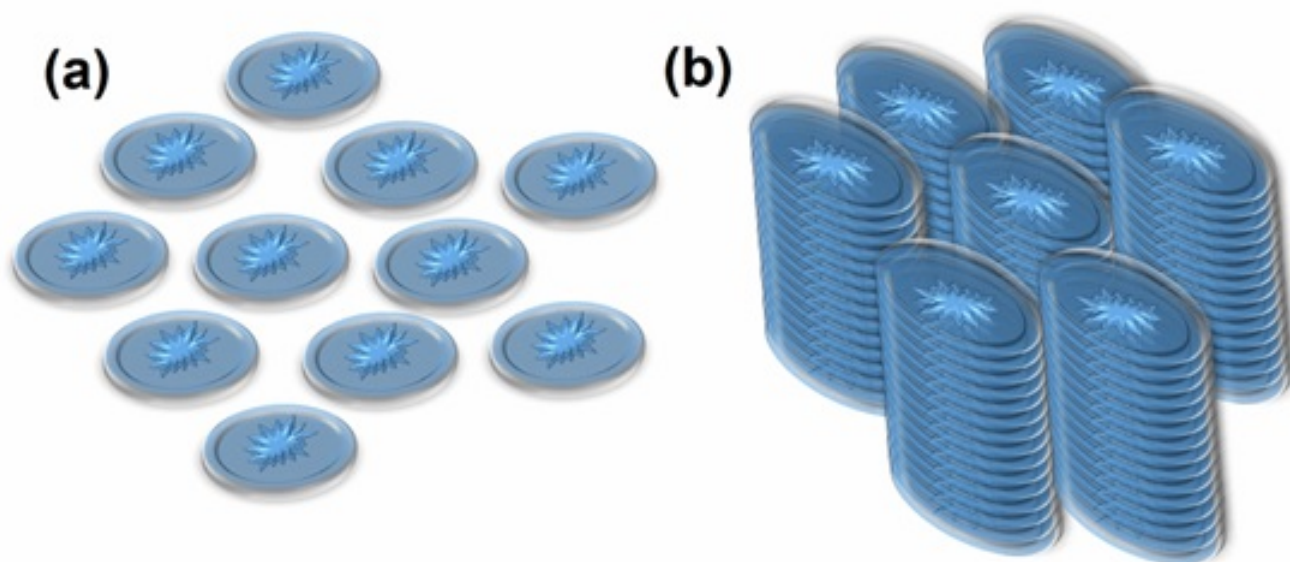


Figure 1. DLCs in (a) nematic and (b) columnar phases aschematic

shows an important role in the stabilization or destabilization of columnar phases. A position modification of the functional group (i.e., carboxylic acid, nitro group, or methyl carboxylate) in dibenzophenazine DLCs sequence from "upper" to "lateral" of aromatic core intensely disturbs the stability and mesophases^{29,30}. Dibenzophenazine and triphenylene derivatives' stability were also influenced due to symmetry. Besides this, the melting temperatures of symmetrical DLCs declined significantly more than their clearing temperatures³⁰. Even though electron-withdrawing groups are substituted in the discoid core to stabilize the mesophases, the fluorescence quantum yield increases.

FACTORS INFLUENCING DLCs

The molecular structure of the DLCs, solvent choice, substrate properties, processing parameters, external stimuli, and annealing can all significantly impact the alignment and organization of DLC films^{31,32}. Understanding these factors and tailoring them to the desired

outcome can lead to highly ordered, aligned, and efficient charge transport in DLC-based devices.

Charge-Carrier Mobilities

Charge-carrier mobility is an important property of semiconducting materials, including DLCs. It refers to the ease with which charge carriers (electrons or holes) move through the material when an electric field is applied³³. In general, higher charge-carrier mobilities result in more efficient electronic devices, as charge carriers can move more quickly and with less resistance, leading to faster response times and higher electrical conductivity. In DLCs, the charge-carrier mobility is influenced by several factors, including the molecular structure of the material, the nature and distribution of the charge carriers, and the local environment of the charge carriers within the material. The mobility of charge carriers in DLCs is typically determined by experimental techniques such as field-effect transistors (FETs), time-resolved microwave conductivity (TRMC), and space-charge-limited current (SCLC) measurements^{34,35}. The mobility of

charge carriers in DLCs is generally lower than in conventional inorganic semiconductors, such as silicon. This is due in part to the relatively disordered molecular structure of DLCs, which can impede the movement of charge carriers through the material. However, researchers have developed several strategies to improve the charge-carrier mobility in DLCs, including the use of high-mobility materials, the optimization of the molecular packing and orientation within the material, and the introduction of dopants or other additives to enhance charge transport^{36,37}. One promising approach to enhancing charge-carrier mobility in DLCs is the use of columnar self-assembly, where molecules are stacked in a highly ordered manner to create pathways for charge-carrier transport. Another approach involves the use of charge-transport additives, such as small-molecule dopants or polymer blends, which can improve the efficiency of charge-carrier transport within the DLC material.

Overall, the charge-carrier mobility in DLCs is an important property that influences the performance of electronic devices based on these materi-

als. Further research into the underlying mechanisms of charge transport in DLCs and the development of new strategies to enhance charge-carrier mobility will be critical for advancing the field of organic electronics. In organic semiconducting materials, the charge carrier's mobility (CCM) may regulate the appropriateness and compatibility with electronic devices³⁸. In crystalline constituents, mobility is intensely affected because it possesses dualistic properties. These planar organic compounds (such as anthracene or pentacene) exhibit advanced mobility in amorphous materials because of their well-organized stacking performance³⁹. Exceptional mobility has been demonstrated in a single crystal device. The main restrictions of this technology are the expense and time required to develop a single crystal. In general, researchers focus and target those systems that self-assemble under wet-processing surroundings such as charge separation in photovoltaic cells, field-effect transistor switching speed, or light-emitting diode intensity (LED)^{40,41}. Various computational and experimental modus operandi has been used to determine the CCM in DLCs. These comprise time-of-flight (TOF) procedure, field-effect mobility, steady-state space charge-limited current (SCLC) or (P.R.) pulse radiolysis time-resolved micro-wave conductivity method (PR-TRMC), PR-TRMC governs CCM at the local level and is comparatively unresponsive to imperfections^{42,43}. They show CCM within spatial constrained domains. It was determined that the defect's presence affected the macroscopic values by using the TOF and field-effect procedures. In SCLC, the CCM is determined by examining the IV characteristics of thin organic films that have been compressed between the injecting electrodes. Previously different researchers elaborately discuss the CCM of various DLCs using TOF15 and PR-TRMC techniques⁴⁴. According to Van de Craats and Warman, a lower CCM

was commonly perceived during the columnar hexagonal phase (C-HP)⁴⁵. The alkyl chains through oxygen atom linked core show low CCM compared to those DLCs in which the chains are attached directly via methylene moiety or through the sulfur. To calculate the maximum CCM, eq. 1 is used, centered on preceding PR-TRMC investigational outcomes on porphyrin, coronene, triphenylene, monoimide, azocarboxyldiimido-perylene, perihexabenzocoronene and phthalocyanine.

$$\sum \mu_{max}.3e^{-83=n} \text{ cm}^2/V \text{ s} \quad (1)$$

Here, n represents the numeral of carbon, nitrogen, and oxygen atoms in the core.

Earlier, Mullen and his fellows proved that CCM anticipated is constrained to core size <40 in Hexaperihexabenzocoronene (HBC). The nature of the substituents associated with discoid core influences supramolecular order, solvability, and, ultimately, CCM of DLCs. The isotropic temperature and interaction between the discoids were lowered near the point of contact. When kept at room temperature, the intramolecular order is maintained at a high level, with a CCM of $0.73 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. When methyl benzenethiol-coated gold nanoparticles are introduced to the C-HP, the conductivity of 2,3,6,7,11-(hexakis-hexyloxy)triphenylene is raised by three orders of magnitude from 10^{-12} to $10^{-99} \Omega^{-1} \text{ cm}^{-1}$. Gold nanoparticle chains have been shown to increase the conductivity by a factor of three to four ($\sim 10^{-6} \Omega^{-1} \text{ cm}^{-1}$). TQPP-[SC₁₂H₂₅]₄, was found to have a saturation hole mobility of $10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, with no mesophase. However, TOF hexapentyloxytriphenylene hole mobility in the (C-HP) reliant on temperature and electric-field having CCM upto $2 \cdot 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Hexahexylthiotriphenylene demonstrated electron and hole mobility in the order of $0.08 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in helical C-HP.⁹⁶ In general, the

lower CCM in the C-HP is due to the increased structural and organizational disorder in the columns caused by the side groups' relatively high vibrational modes freedom. The processing method impacts mobility's reliance on the DLC's morphology. The 2, 3, 6, 7, 10, 11-Hexa(hexyloxy)triphenylene gelation with hydrogen-bonded fibrous groups heightens hole mobility from 4.5×10^{-4} to $1.2 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Temperature and electric field strength do not affect mobility, which is self-regulating. Few researchers claimed that CCM mobility is restricted due to structural imperfections. A defect-free assembly is considered an ideal surface increase in mobility of up to 15 centimeters per second. It has been projected that electron mobilities for lateral hopping exist at 300 K and that this value increases considerably during higher molecule assembly and orientation⁴⁶.

Molecular structure

The molecular structure of the DLCs, such as their shape, size, and chemical functional groups, can significantly influence their properties, including their self-assembly, intermolecular interactions, and charge transport⁴⁷. For instance, DLCs with elongated, rod-like shapes tend to exhibit a high degree of anisotropy in their molecular alignment, leading to highly ordered structures with excellent charge transport properties. On the other hand, DLCs with bulky or irregular shapes can disrupt the molecular packing, resulting in lower mobilities.

Solvent

The choice of solvent used for dissolving the DLCs can impact their molecular packing and alignment on the substrate^{48,49}. For instance, polar solvents such as water or ethanol tend to favor polar interactions, leading to highly ordered and aligned DLC films. In contrast, nonpolar solvents like chloroform or hexane can promote π - π stacking

interactions between the DLCs, leading to more extended molecular packing.

Substrate

The substrate used for depositing the DLCs can significantly impact their alignment and organization. Substrates with high surface energy, such as metals or oxide surfaces, can promote the formation of highly ordered and aligned DLC films⁵⁰. Additionally, surface functionalization with specific chemical groups or self-assembled monolayers can tailor the surface properties to promote specific intermolecular interactions and molecular alignment.

Processing parameters

Various processing parameters such as temperature, pressure, concentration, and deposition rate can significantly impact the alignment and organization of the DLCs⁵¹. For instance, higher temperatures can promote molecular diffusion and reorganization, leading to better-aligned films, while high deposition rates can lead to less ordered structures.

External stimuli

External stimuli such as magnetic fields, light, or electric fields can induce molecular reorientation or alignment, leading to highly ordered DLC films⁵². For instance, magnetic fields can align paramagnetic DLCs, while light can induce photoisomerization or photodimerization, leading to specific molecular alignment.

Annealing

Annealing of DLC films can significantly impact their alignment and organization. Annealing can promote molecular diffusion and reorganization, leading to highly ordered and aligned films^{53,54}. The annealing temperature and duration can significantly impact the degree of molecular reorientation, with higher temperatures leading to

more extended molecular packing and alignment.

Mesophase Engineering

Mesophase engineering in DLCs involves controlling the alignment and organization of the molecules in the liquid crystal phase to optimize their properties for various applications^{55,56}. There are several methods for mesophase engineering, including the use of additives, surfactants, and external fields.

Surfactants can also be used to modify the properties of DLCs. For example, adding surfactants can induce the formation of different mesophases, such as lamellar or hexagonal phases, depending on the nature of the surfactant and its interaction with the DLC molecules⁵⁸. This has been demonstrated in the case of pyrene-based DLCs, where the addition of surfactants led to the formation of hexagonal mesophases with unique optical and electronic properties.

External fields, such as electric or magnetic fields, can also be used to control the alignment and organization of DLC molecules⁵⁹. For example, applying an electric field to DLCs can induce a reorientation of the molecules, which can be used to tune their optical and electronic properties. This has been demonstrated in the case of triphenylene-based DLCs, where the application of an electric field led to a reorientation of the molecules and a change in the color of the material.

Overall, mesophase engineering is an important aspect of DLC research, as it allows for the optimization of their properties for various applications such as electronic and optoelectronic devices. Different techniques were employed to formulate DLC mesophases⁶⁰. Several research groups have already examined the well-organized coordinated self-assembly of amphiphilic HBC derivatives. Trinitrofluorene (TNF) substituents in amphiphilic HBC derivatives from a few micrometers extended coaxial nan-

otubular assembly with 16nm diameter⁶¹. The electron donor graphitic stratum of the -stacked HBC is covered via the electron-accepting TNF layer. It enhances photochemical charge carrier generation, resulting in a rapid photoconductive response. The electron-rich discoid doped with TNF generates charge-transfer complexes (CTC), which leads to the formulation of supramolecular assemblies and mesophases. CTC comprises pentakis(phenylethynyl)phenol and TNF having a 1:1 ratio resulted in diverse mesophases having high clearing temperature^{62,63}. CTC can be synthesized by adding a TNF acceptor series to mesogen having 5 methoxy clusters with functional tail. There are also a lot of different electron donors and acceptors at the end of the functionalization of fluorinated dendrons, which makes them even more useful.

ALIGNMENT OF DLCs

Columnar DLCs tend to align themselves in two distinct arrangements, which are the face-on (or planar) and edge-on (or perpendicular) alignments^{64,65}. In the face-on alignment, the disk-shaped molecules are oriented parallel to the substrate or the surface on which they are deposited, with their long molecular axis lying in the plane of the substrate. This arrangement is favored when the substrate surface is hydrophilic and the DLC molecules have polar substituents or functional groups that can interact favorably with the substrate surface. The face-on alignment can also be achieved by shear or flow alignment during the deposition process. On the other hand, in the edge-on alignment, the disk-shaped molecules are oriented perpendicular to the substrate, with their long molecular axis perpendicular to the plane of the substrate. This arrangement is favored when the substrate surface is hydrophobic and the DLC molecules have nonpolar substituents or functional groups that do

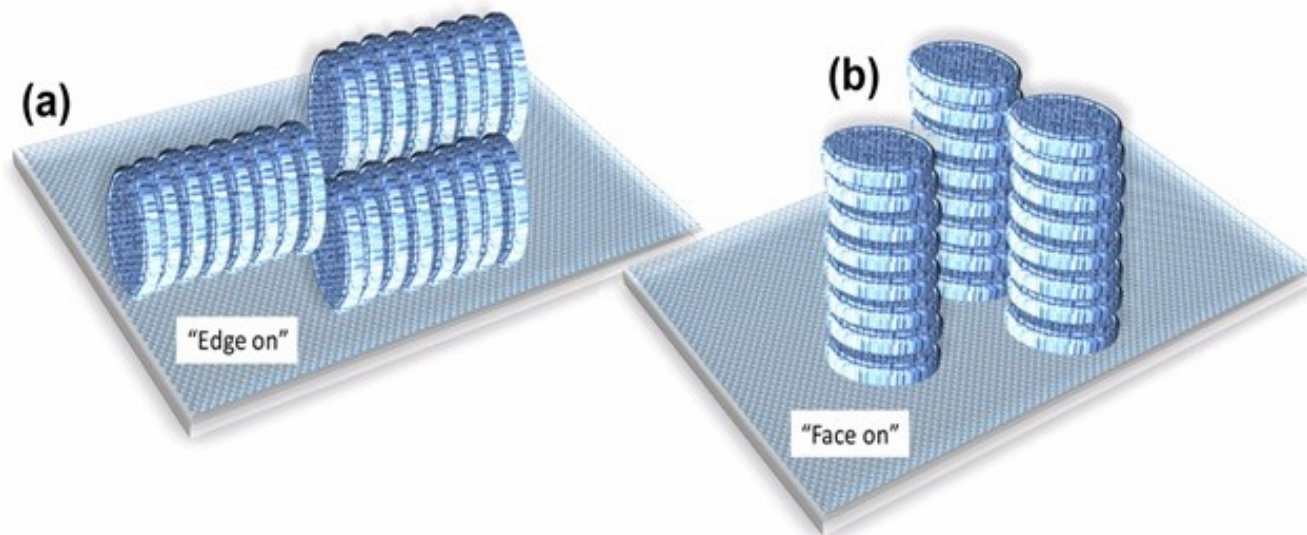


Figure 2. (a) "Edge-on", homogeneous planar unidirectional alignment; (b) homeotropic (face-on) alignment

not interact favorably with the substrate surface⁶⁶. The edge-on alignment can also be achieved by using an external magnetic field during the deposition process.

A homogeneous uniaxial planar structure, also known as edge-on, is when the column axis is parallel to the substrate. In the case of homeotropic orientation, the column axis is perpendicular to the substrate. (Figure 2). The homeotropic orientation is applicable for organic photovoltaics or OLED application, but the unidirectional planar shape is optimal for OFET applications. Guillamat and his colleagues proposed two molecular configurations in rectangular columnar organized mesophases⁶⁷. They tilt slightly in one of two directions: either perpendicular or parallel to the substrate. The experiment has proven that when DLCs are slowly cooled from the isotropic phase, face-on alignment of primary molecular strata works as a nucleation site for additional self-assembly of perpendicular discs⁶⁸. The choice of alignment depends on the specific application and the desired properties of the DLC material. For example, the edge-on alignment can result in higher charge carrier mobility and better electrical conductivity,

while the face-on alignment can result in higher photoconductivity and better optical properties.

Methods of attaining Structural alignment

The capability of switching orientation and structural alignment is difficult to achieve high CCM^{69,70}. DLCs have been aligned using a variety of ways, some of which are detailed below

Zone Casting

In this method, a thin film's homogeneous and uniform alignment is achieved via controlling the concentration gradient formed between the moving supporting substrate and the nozzle. Different casting parameters such as solvent, temperature, concentration, supply rate, substrate velocity, and nozzle/substrate distance may slightly influence the acquired aligned layers^{71,72}. It permits the prearrangement of organic materials homogeneously and consistently. It enables the uniform and constant arrangement of organic materials. Solvent-dissolved DLC is continuously sprayed onto a moving substrate with a temperature gradient using a nozzle. A thin film's homogenous and uniform alignment

is attained via controlling the concentration gradient formed between the moving supporting substrate and the nozzle. Different casting parameters such as solvent, temperature, concentration, supply rate, substrate velocity, and nozzle/substrate distance may slightly influence the acquired aligned layers (Figure 3). Zone casting of HBC-C12 on a PTFE layer resulted in forming a layer having a mobility of $1 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ⁷³. Jiang H hypothesized columnar configuration direction of HBC-C12 has a tenfold upsurge in photoconductivity over the perpendicular orientation⁷³. HBC derivative aligned via the zone-casting technique retains optical anisotropy, which is reversibly (switched on/off) through temperature change leading towards molecules reorientation to an orthogonal stacking arrangement from a tilted stacking arrangement.

Amphiphilic HBC derivatives were attained via Langmuir-Blodgett (L.B.) Deposition. An L.B. film is formed at the water-air interface when hexaalkylhexa-perihexabenzocoronene has an alkyl substituent ended having a carboxylic assembly. The alkyl chain is π - π stacked to generate two separate phases. Another benefit of the benzyl-

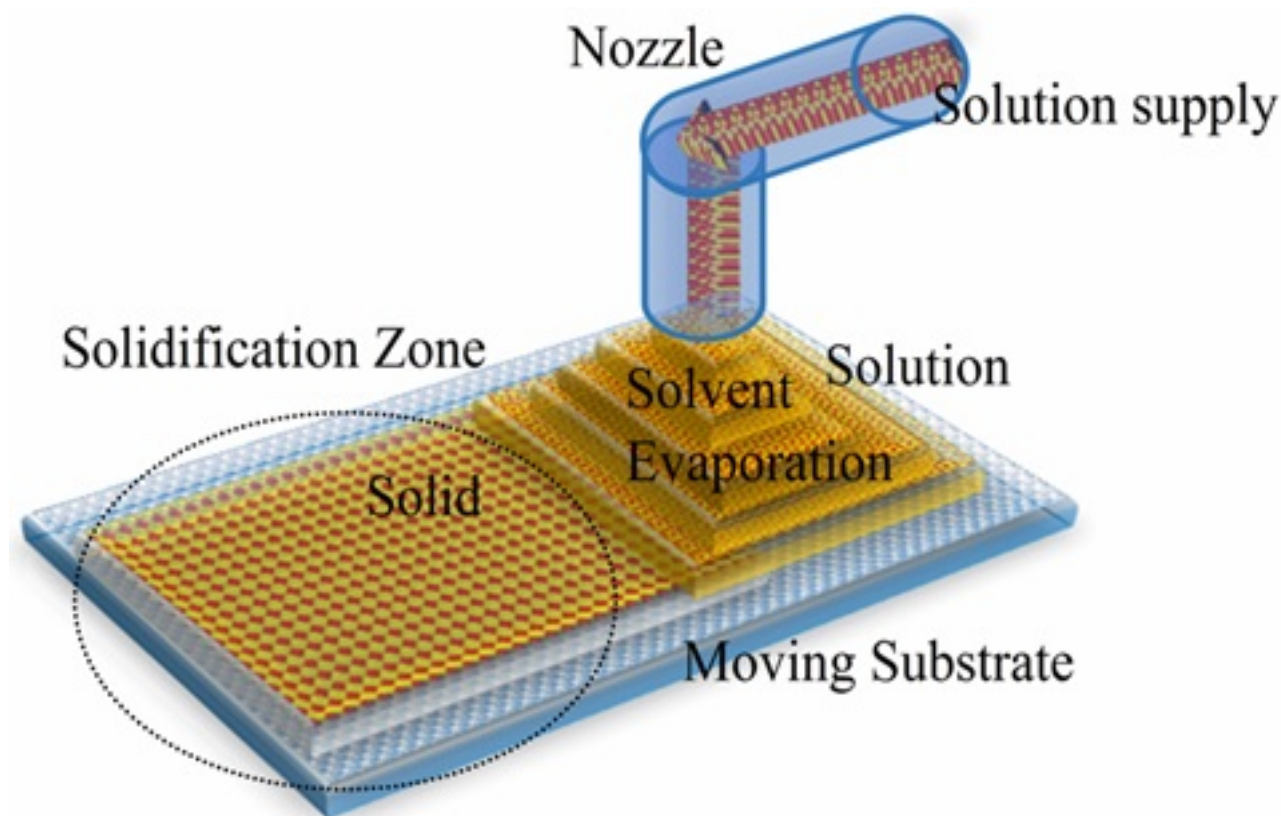


Figure 3. Schematic demonstration of the zone-casting technique

terminated octa-substituted phthalocyanines is that they create highly well-organized L.B. thin films with high thermal stability, despite the edge-on alignment seen in DLCs synthesized using L.B. (one or two triphenylenes connected to TNF)⁷⁴.

Solution-Casting

This method involves continuously spraying a solvent-dissolved DLC onto a moving substrate with a temperature gradient using a nozzle. By controlling the concentration gradient formed between the moving supporting substrate and the nozzle, a thin film's homogeneous and uniform alignment is achieved. For example, zone casting of HBC-C12 on a PTFE layer resulted in forming a layer having a mobility of $1 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. On the substrate, an alternative method for accumulating DLCs involves using oriented polytetrafluoro-

ethylene (PTFE), which can align HBC derivatives such as triphenylene and phthalocyanine⁷⁵. An aligned polyimide stratum is also utilized to collect triphenylene DLCs, similar to the previous example. Solution-casting is another method used to study DLCs via STM. In this method, a solution of the DLC material is prepared and then deposited onto a substrate using spin coating or drop casting techniques. The DLC material in the solution aligns with the substrate and forms a thin film. One approach to achieving alignment in solution-casting is using an oriented polytetrafluoroethylene (PTFE) substrate. PTFE has a high surface energy, which enables it to align HBC derivatives such as triphenylene and phthalocyanine. The DLC material is dissolved in a suitable solvent and then deposited onto the PTFE substrate using spin coating or drop casting techniques. The PTFE substrate aligns the

DLC molecules, resulting in a thin film with a preferred orientation.

Another approach involves using an aligned polyimide layer to collect triphenylene DLCs⁷⁶. The polyimide layer is first aligned by rubbing it with a velvet cloth or using a photoalignment technique. The DLC material is then deposited onto the aligned polyimide layer using spin coating or drop casting techniques. The DLC molecules align with the polyimide layer and form a thin film with a preferred orientation⁷⁷. Overall, solution-casting is a versatile method for preparing DLC thin films with various orientations and alignments. It allows for the deposition of DLCs onto a wide range of substrates and can be used to study the structure and properties of DLCs using various techniques, including STM.

Surface Treatment

Annealing is a widely used treatment for aligning DLCs, where the material is heated above its crystallization temperature and then cooled at a controlled rate to form stable thin coatings⁷⁸. The use of surface treatments such as Ar plasma, UV-ozone, or annealing on an ITO surface can produce well-aligned DLC thin films, which can be favorable for wet conditions and organic thin film formation. In addition, different surface and interface interactions can influence the configuration of DLCs. One example is the use of phosphocyanine (Pc) derivatives with octet oligo(ethyleneoxy) peripheral substituents, which have been found to exhibit different alignments depending on the surface. On a hydrophobic surface, the rod-like liquid crystalline molecule aligns perpendicularly to the substrate to achieve homeotropic alignment⁷⁹. In contrast, when the Pc molecule's outer chains have diverse interfaces, such as those with hydrophilic surfaces, this results in homogeneous or planar alignment, where the molecules align parallel to the substrate. Another treatment method for modifying the alignment of DLCs is through circularly polarized infrared (IR) irradiation. This technique activates the vibrational mode of a mesogen's particular chemical bond, allowing for a specific configuration to be attained based on the incident photon polarization direction and transition dipole moment for the vibrational excitation. This alignment is typically achieved in higher-ordered DLC phases, such as the Colh mesophase, which has a high viscosity. Overall, surface treatments and annealing can significantly influence the alignment and configuration of DLCs, and the use of circularly polarized IR irradiation can provide a means for selectively modifying the alignment of specific chemical bonds in the mesogens. These techniques are important for exploring the potential applications of DLCs in optoelectronics, where precise align-

ment is critical for efficient device performance.

Irradiation

Infrared (IR) irradiation is a technique used to modify the configuration of DLCs for their effective utilization in optoelectronics⁸⁰. The alignment of DLCs can be changed by activating the vibrational mode of a mesogen's specific chemical bond using circularly polarized infrared irradiation. The vibrational excitation is achieved by a specific grouping of the incident photon polarization direction and transition dipole moment⁸¹. This technique is only effective in higher-ordered DLC phases with high viscosity, such as the Colh mesophase. The use of IR irradiation to modify the alignment of DLCs has been reported in studies by Shimizu and colleagues⁵⁰. By changing the alignment of DLCs using IR irradiation, it is possible to optimize their charge-carrier mobility and other properties for use in optoelectronic applications. However, the precise mechanism of this alignment process and the optimal conditions for achieving it are still the subject of ongoing research. Circularly polarized infrared irradiation technique is utilized to modify the configuration of the DLCs. Polarized infrared irradiation changes the DLC alignment by activating the vibrational mode of a mesogen's particular chemical bond. The specific grouping of the incident photon polarization direction and transition dipole moment for the vibrational excitation permits attaining a particular configuration.

Annealing

Annealing is a successful way of aligning the DLCs⁸². For example, an ITO surface treatment using Ar plasma, annealing, or UV-ozone can produce stable DLC thin coatings. The DLC material is heated above its crystallization temperature and then cooled at a regulated degree to manufacturing these films.

Langmuir-Blodgett (LB) deposition

In this method, an L.B. film is formed at the water-air interface when hexaalkylhexa-perihexabenzocoronene has an alkyl substituent ended having a carboxylic assembly. The alkyl chain is π - π stacked to generate two separate phases. Benzyl-terminated octa-substituted phthalocyanines can create highly organized L.B. thin films with high thermal stability.

Magnetic Influence

The mechanism to study DLC via STM influence involves the use of a magnetic field to achieve a uniaxial orientation of the discotic metallomesogens⁸³. In this case, CoS12 DLC was used, which is a diamagnetic material that maintains its interaction even in the presence of an external magnetic field. Lee and colleagues were able to achieve a magnetic uniaxial columnar assemblage of the CoS12 DLC by continuously rotating the sample from isotropic phase to liquid crystalline phase while in the presence of a static magnetic field. The external magnetic field provided an orientation for the DLC, aligning it edge-on. This alignment allowed for the DLC to be studied using STM, which can visualize the surface of the material at the atomic level. This technique is useful for understanding the morphology and structure of DLCs, which is important for their optoelectronics applications. Magnetic alignment is just one of the methods that can be used to achieve alignment of DLCs, and other methods such as surface treatment, zone casting, and Langmuir-Blodgett deposition have also been explored.

Sacrificial Layer Influence

Various surface and interface interactions can change the DLCs' configuration⁸⁴. For example, phosphocyanine derivatives connected to octet oligo(ethyleneoxy) peripheral substituents on a hydrophilic and homogeneous orientation on a hydrophobic sur-

face have previously exhibited different alignments. To study discotic liquid crystals (DLCs) using scanning tunneling microscopy (STM), it is important to first prepare the sample in a way that allows for proper alignment and observation of its properties⁸⁵. One method to achieve alignment is through the use of a sacrificial polymer layer. This method has been demonstrated in an alkoxy phthalocyanine DLC where homeotropic alignment was achieved. The first step in this process involves spin coating or casting a layer of sacrificial polymer onto a substrate. The DLC is then placed on top of the sacrificial layer. The sacrificial layer acts as a temporary substrate that can be removed once the DLC is properly aligned. To align the DLC, thermal annealing is used to induce homeotropic alignment. The sample is heated to a specific temperature to induce the desired alignment, and then cooled to ambient temperature to maintain the alignment. Once the alignment is achieved, the sacrificial layer can be removed through a process known as lift-off. This involves dissolving the sacrificial layer in a solvent that does not affect the DLC. Using a sacrificial layer makes it possible to achieve homeotropic alignment of DLCs, which allows for observation of their properties using STM. This method has been used to study various DLCs and is a valuable tool for developing optoelectronic devices.

DLCs investigated via Scanning Tunneling Microscope (STM)

Scanning Tunneling Microscopy (STM) is a powerful tool for investigating the structure and properties of materials at the nanoscale, including Discotic Liquid Crystals (DLCs)⁸⁶. DLCs exhibit unique properties that make them attractive for use in optoelectronic applications, such as charge-carrier mobility, self-assembly, and solubility in different organic solvents. STM allows for the visualization of DLCs at the molecular level, providing insight

into the structure and arrangement of the disc-shaped molecules. STM imaging can reveal the formation of ordered columnar structures, each consisting of stacked discotic molecules. These columns can be aligned in various orientations, depending on the processing conditions used to prepare the DLC material.

STM can also be used to study the charge transport properties of DLCs. By measuring the tunneling current between the STM tip and the DLC material, it is possible to investigate the movement of charge carriers along the columns of discotic molecules. The charge-carrier mobility of DLCs can be affected by a range of factors, including the alignment of the columns and the presence of defects or impurities in the material. In addition to imaging and charge transport measurements, STM can also be used to manipulate and modify the structure of DLCs. For example, the tip of the STM can be used to apply an electric field to the material, which can induce changes in the alignment of the discotic columns. This can be used to create patterns or structures in the DLC material, which can be useful for constructing electronic devices. Hence, STM is a valuable tool for investigating the structure and properties of DLCs, providing insight into their potential applications in the field of optoelectronics. By understanding the behavior of these materials at the nanoscale, it is possible to develop new and innovative electronic devices that harness the unique properties of DLCs.

Lee and his colleagues suggest an alternate technique for phase-control and electrical measurements of DLCs at the nanoscale⁵¹. Using STM and STS, they investigate the discotic liquid crystalline of ndibenzo[a, c]phenazine and the electron transfer characteristics of self-assembled monolayers formed by a DLC molecule at liquid-solid (L.S.) boundary. As targeted molecules possess an internal dipole moment, the polarity of the substrate underneath may regulate the molecular

polymorphs. They observed two types of orientation, head-to-head and non-head-to-head, which were resistant to negative and positive STM bias. An STM tip may modify their transitions locally as shown in Figure 4(a). Besides this the solvent did not impact the morphology and orientation of molecules. It is due to the low polarity or lack of solvents, co-adsorbed molecules are unaffected by the solvent. Unlike in 2D systems, where substrate effects play an important role, 3D systems emphasize intermolecular attractions, which leads to columnar packing (Figure 4(b)). Additionally, STS results demonstrate that the bandgap of packing I was approximately 400 mV smaller than that of packing II. The STM contrast of packing I is brighter than packing II (Figure 4(c)). According to STM, a brighter contrast indicates higher conductivity, which is in line with this concept. Octanoic acid or n-tetradecane molecular solvent did not alter surface packing in any substantial way. There is an insignificant solvent effect on self-assemblies in a 2D self-assembly system compared to those seen in a 3D design.

Hence, molecules' properties, such as optical, mechanical, heterogeneous catalysis and selectivity, can be influenced by their molecular arrangement, as shown in this study. Numerous 2D materials consisting of self-assembly films have shown fascinating phenomena. Chunli Bai and his colleagues investigate 1,7,13-trialkanoyldecacyclene [TTD] (with n-carbon side chains, n=14 & 18)⁵². According to their research, there is a way to fill two dimensions of a triphenylene core with only three side chains at the interface. This study might serve as a counter-example to previous research that found that three elongated chains would not be enough to occupy the center's area. Figure 5 shows an STM view of a stratum of TTD on a graphite substrate, where two orientation patterns represent the zigzag motifs, as clearly evident. The observed

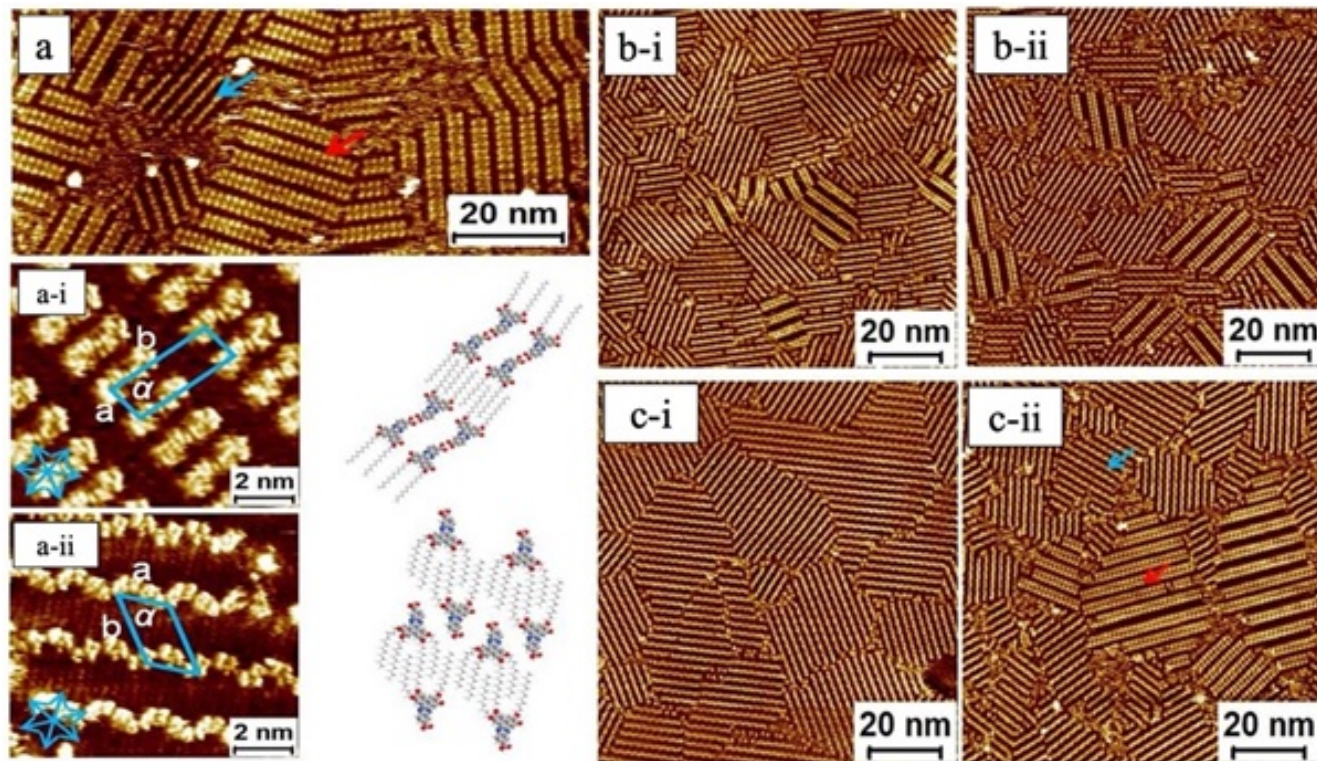


Figure 4. (a) STM images of self-assembly of **1** using 1.0-mM sample solution. a (i, ii) STM image and models of packing-i and packing-ii. STM image of solvent effect: b (i) octanoic acid, (ii) n-tetradecane. c (i, ii) STM images of concentration-dependent surfaces comprised of the polymorphs of **1**. The concentration of sample solutions for (i): 0.01 mM; (ii): 0.5 mM. The red and blue arrows in b) indicate the packing-i and packing-ii motifs, Imaging conditions: E_{bias} , -900 mV; $i_{tunnelling}$, 150 pA. Reprinted with permission⁵¹.

domains have darker and brighter regions, corresponding positions of interdigitated alkyl chains and decacyclene core moieties, respectively.

The TTD molecules were assembled into supramolecular twin chains stabilized by 2D alkyl crystallization. The assembling configurations seen might be explained by the influence of two-dimensional alkane lamella and steric barrier caused by adsorbate-substrate interactions. This could result in a loss of symmetry in nonchiral molecules. Specific geometries favored energetically resulting in structural transformations, i.e., chirality or the disintegration of molecular symmetry, are two examples of this. They can be interpreted in terms of three different driving forces. The initial step is to attain the best possible alignment with the substrate lattice. The second goal is to encour-

age the generation of monolayers in which hydrogen atoms are thought to be in equilibrium. The steric hindrance encourages molecules to attain antiparallel directions, i.e., antiferromagnetic-like triangular networks. Besides this, alkyl chains acquire parallel orientations to achieve maximum local 2D crystallization regardless of the core molecular symmetries. As a matter of fact, molecules may experience spontaneous symmetrical splitting and lose their previous configuration. Decacyclene derivatives would have more steric hindrance due to their larger cores than triphenylene derivatives, and longer side chain would result in more 2D crystallization. In a two-dimensional system, the TTD configuration appears to balance core-substrate intermolecular interactions and surface free energy minimization. A similar arrangement

is used by TTD when four additional methylene units are added; however, due to their longer side chains, the separation and angle were altered. HDTP, a triphenylene derivative with only 12 methylene side chains, retains the uniformity of organic molecules through self-assembly.

Due to their self-assembling characteristics and innovative structural features, liquid crystals are attracting interest from experts in the development of organic light-emitting devices, photovoltaics, and thin-film transistors, along with other scientific disciplines. The polar packing of molecules gives rise to Ferro- or antiferroelectric properties having numerous practical applications, such as piezoelectricity, pyroelectricity, and fast-switching electrooptical devices. It has been predicted that the transition temperature

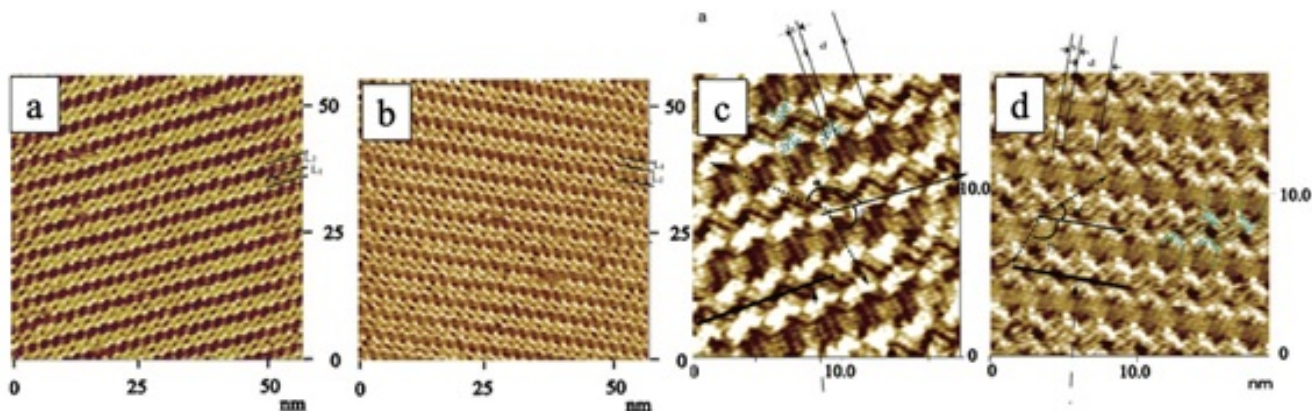


Figure 5. (a) Large-scale image of one of two orientated stripes ($57 \times 57 \text{ nm}^2$). Tunneling conditions: 800 mV, 1.10 nA. (b) STM image of the other of two orientated stripes ($57 \times 57 \text{ nm}^2$). Tunneling conditions: 880 mV, 0.99 nA. (c,d) High-resolution STM images of the TTD assembly. Reprinted with permission⁵².

would decrease with increasing thickness. Still, in a study by Bune et al.,⁵³ a first-order ferroelectric phase was found with a transition temperature nearly identical to the bulk value, challenging this prediction. Antiferroelectric L.C. phase's structure was demonstrated by an STM image, which revealed that the layers are made up of innovative quadruple "zigzag" molecular rows. STM is used to investigate the submolecular structures of 2D assembly of banana-shaped LC molecules, P-n-PIMB (n) 18, 14, and 12, on the HOPG substrate. The findings reveal two types of coexisting molecular arrangements. This surface nano-sized assembling behavioral patterns of liquid crystals may give important information for experimental application and theoretical research^{54–60}.

The scanning tunneling microscope (STM) is a powerful tool that uses an external electric field to control chemical reactions and supramolecular phase transitions at molecular scales. However, it has not been widely used for crystal engineering applications. Khan, S.B and his colleagues demonstrate the use of the directional electric field of an STM to manipulate supramolecular crystallization on a solid surface⁸⁷. They found that a random-tiling assembly of p-terphenyl-3,5,3',5'-tetracarboxylic acid can be transformed

into close-packed periodic assemblies under positive substrate bias conditions at the liquid/solid interface. They were able to tailor the nucleation and crystal growth of the resulting field-induced products in real-time, producing a two-dimensional supramolecular single crystal. The crystals exhibited bright spectroscopic features that were strongly dependent on the STM bias, corresponding to variations in electron density of states. As shown in Figure 6(a), the chemical structure of a TPTC target molecule. While the core A–E of the random tiling displays a negative STM bias, all other closed packed patterns are biased positively. The R and S forms are mirror images of one another at the macro level, and the H (head-to-head) package is a likely third form, all of which share the same cell unit size. The normal direction (n line) of TPTC is perpendicular to the packaging axis (m line) of the close packing motif, which runs in the same plane as HOPG. The blue arrows represent the path of the graphite structure of the grid. The Ebias and itunneling are 0.5 V, 70 pA. The (d) and (e) panels show the random tiling of the five pores on the surface and the percentage of them. Parameters of the unit cells of a, b, and α : 0.9 (± 0.3) nm. They report that an external, orientated electric field of STM can be utilized

to generate STM bias-related new 2D self-assembled structures, which would be otherwise inaccessible by the drop-casting or spin-coating methods.

Optoelectronics applications of DLCs

DLCs have shown great potential in optoelectronics applications due to their unique electronic and optical properties. Here are some of the optoelectronics applications of DLCs. Figure 7 displays the DLCs applications in numerous domains.

Organic Light Emitting Diodes (OLEDs)

DLCs can be used as emissive and conductive layers in OLEDs due to their high carrier mobility, high thermal stability, and strong charge transfer ability. This results in efficient light emission and better device performance⁸⁸.

Organic Solar Cells (OSCs)

DLCs can be used as active layers in OSCs due to their good photoconductivity and high electron mobility. They can also be used as hole blocking and electron blocking layers in OSCs to improve device efficiency⁸⁹.

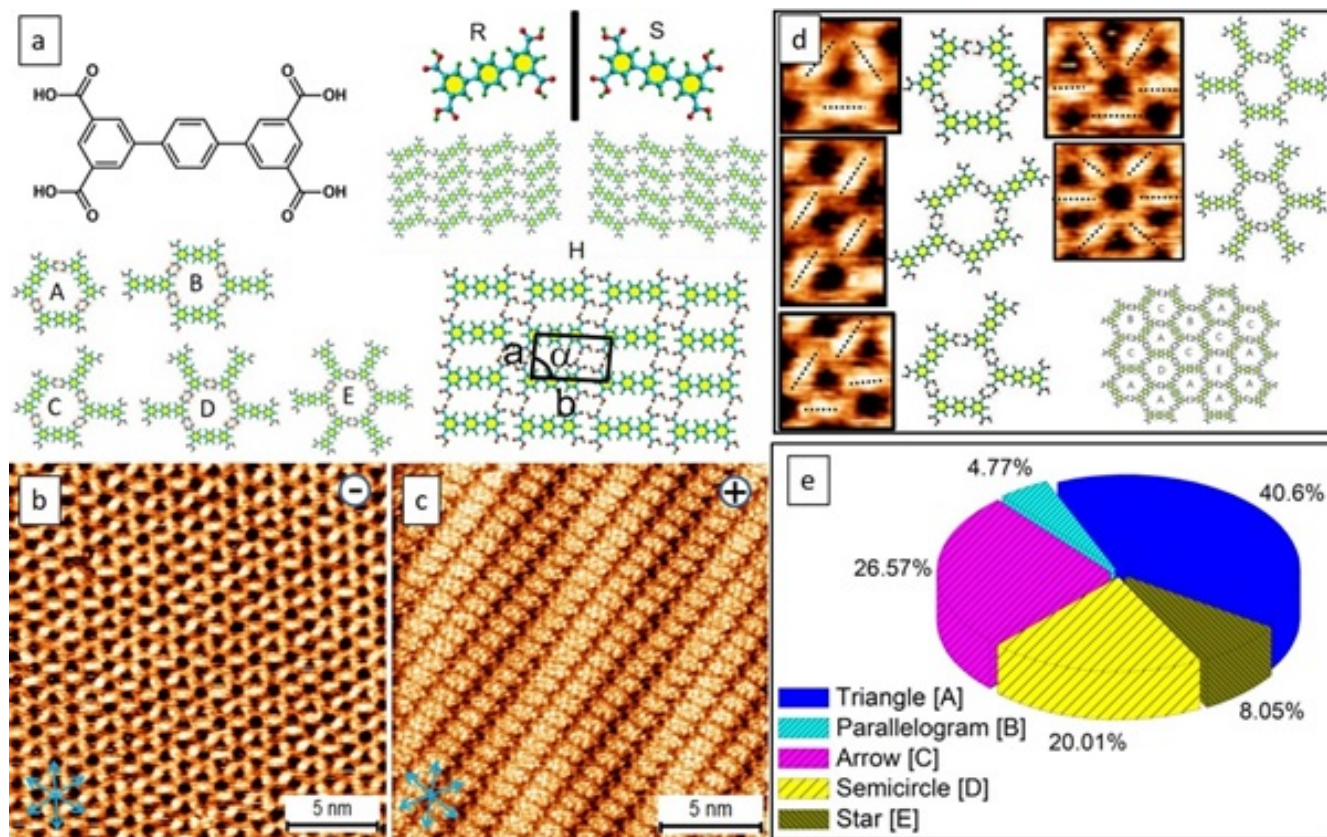


Figure 6. TPTC target molecule chemical structure. (b & c) The close-packed phase features a mirror image (R and S forms) and an expected H (head-to-head) packing with the same unit cell dimensions. The packing axis of the close packing motif goes along one of the normal directions of HOPG below (m line), and a molecular skeleton is 60° from the packing axis so that TPTC lies nearly along another normal direction (n line). Graphite lattice structural direction is shown by blue arrows. 0.5 V, 70 pA for imaging (E_{bias} , $i_{tunneling}$). (d) & (e) demonstrate the random tiling and surface pores. a, b, and α unit-cell parameters: $0.9 (\pm 0.3)$ nm, $1.7 (\pm 0.3)$, $87^\circ (\pm 2^\circ)$. Reprinted with permission⁸⁷

Organic Field-Effect Transistors (OFETs)

DLCs can be used as semiconducting layers in OFETs due to their high carrier mobility and low operating voltage⁹⁰. This results in better device performance and low power consumption.

Photovoltaic Devices

DLCs can be used as photoactive layers in photovoltaic devices due to their high light absorption coefficient and high photoconversion efficiency⁹¹. They can also be used as charge transport layers to improve device performance.

Optoelectronic Sensors

DLCs can be used as sensitive layers in optoelectronic sensors due to their high sensitivity and selectivity towards specific analytes⁹². They can also be used as transducers to convert optical signals into electrical signals.

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Types of DLCs

There are various types of DLCs, including hydrogenated DLC (a-C:H), tetrahedral amorphous carbon (ta-C),

nitrogenated DLC (a-C:N), and fluorinated DLC (a-C:F)^{94,95}. Each type has its unique properties and potential applications in various fields. Here are some examples:

Hexa-peri-hexabenzocoronene (HBC)

HBC is a columnar DLC that has been used in organic light-emitting diodes (OLEDs) as an electron transport material. It has a high charge carrier mobility and can improve the efficiency of OLEDs.

Phthalocyanine (Pc)

Pc is a flat DLC that has been used in organic photovoltaics (OPVs) as a donor material. It has a strong absorption in the visible and near-infrared regions, making it useful for harvesting solar energy. Pc derivatives can also be used as electron transport materials in OLEDs.

Perylene diimide (PDI)

PDI is a planar DLC that has been used in OPVs as an acceptor material. It has a high electron affinity and can improve the efficiency of OPVs. PDI derivatives can also be used as hole transport materials in OLEDs.

Triphenylene (TP)

TP is a columnar DLC that has been used in OLEDs as a hole transport material. It has a high charge carrier mobility and can improve the efficiency of OLEDs.

Tetracene

Tetracene is a planar DLC that has been used in organic field-effect transistors (OFETs) as an active material. It has a high charge carrier mobility and can be used for high-performance electronic applications.

Perylene diimide (PDI) derivatives

PDI derivatives are a class of DLCs that have been widely studied for their potential use in organic solar cells. They exhibit good electron transport properties and high electron mobility,

which can improve the efficiency of solar cells.

Quinacridone derivatives

Quinacridone derivatives are another class of DLCs that have been used in organic solar cells. They have a high extinction coefficient and can absorb light over a wide range of wavelengths, making them useful for harvesting solar energy.

Phthalocyanine derivatives

Phthalocyanine derivatives are a versatile class of DLCs that have been used in a variety of optoelectronic applications, including organic solar cells, organic light-emitting diodes (OLEDs), and photovoltaic devices. They have a high absorption coefficient and can be easily functionalized to tune their optical and electronic properties.

Triphenylamine derivatives

Triphenylamine derivatives are a type of DLC that have been used in OLEDs due to their high hole mobility and good charge injection properties. They have also been used in organic photovoltaics as electron donors.

Boron subphthalocyanine chloride

(SubPc)

SubPc is a DLC that has been used in organic photovoltaics due to its high electron mobility and good stability. It has also been used in OLEDs and other optoelectronic devices.

FUTURE PERSPECTIVES

DLCs have attracted significant attention recently due to their unique properties, including high charge-carrier mobility along the stacking axis, self-assembly, self-healing, and solubility in various organic solvents⁹⁶. These properties make DLCs promising candidates for optoelectronic applications, such as solar cells, organic light-emitting diodes, and field-effect transistors⁹⁷. The use of STM in exploring the structure and properties of

DLCs has emerged as a promising approach for developing efficient electronic devices. STM allows for the visualization of DLCs at the molecular level, providing insight into the structure and arrangement of the disc-shaped molecules. STM imaging can reveal the formation of ordered columnar structures, with each column consisting of stacked discotic molecules. STM can also be used to study the charge transport properties of DLCs and manipulate their structure to create patterns or structures useful for electronic device construction. Overall, exploring DLCs for optoelectronics applications via STM holds great potential for developing innovative electronic devices^{98,99}.

As recent literature explains, this perspective describes the design, synthesis, and morphology of numerous ILCs. Columnar L.C.s provide a variety of ways of achieving tailored structural spiral assemblies. The self-organization process comprises stacking units, self-assembly within twisting columns, and column orientation within prescribed 2D lattices. The non-covalent contacts interact so that the spiral columnar architecture is formed. Another functional property associated with these helical structures is supramolecular chirality, which can be reversibly altered by externally driving the twisted sense. This enables the development of information frameworks of columnar mesophases to switch their chiroptical characteristics¹⁰⁰. Thus, the ultimate goal is to discover how to manage molecular self-assembly and develop novel colloidal systems with specified functions or features closely related to the desired applications through attractive forces. This is an interdisciplinary approach, as the breadth of this research extends considerably beyond the typical boundaries of organic chemistry. This technology applies to various fields, including cosmetics, pharmaceutical formulations, nanomaterials, and catalysis. Additionally, this enables

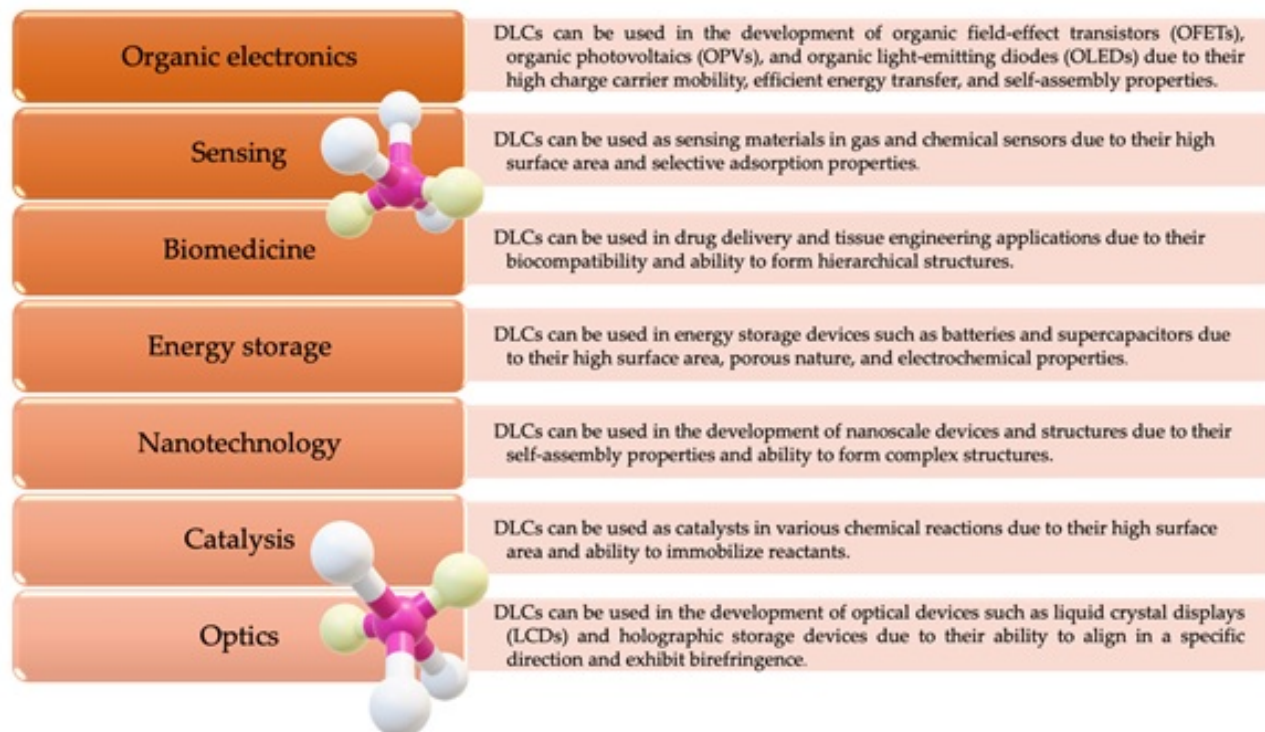


Figure 7. DLCs applications and benefits in different domains.

the production of intermediary systems between homogeneous and heterogeneous mixes, combining the features of homogeneous and heterogeneous catalysis. Consequently, mass transfer between the catalyst and the substrates is significantly enhanced by the water/oil contact, and no stirring is necessary. The separation of reactants and products prevents (or at least reduces) side reactions from occurring¹⁰¹. Additionally, nanotube films can be assembled using established methods of L.C. alignment, such as grooved surfaces, magnetic fields, and patterned electrodes, to control their organization. The nematic matrix is found to order single- and multi-walled carbon nanotubes in L.C. solvents. Other nanometer-sized building blocks can be aligned with L.C. to control organized nanomaterials and their structures. The next stage in developing usable materials and functioning devices is to invent processing methodologies that allow the controlled assemblage of configurations

into larger systems. Single- and multi-walled nanocomposites (SWCNT and MWCNT) have been showing promise building blocks for a broad array of applications, so processes for organizing them are particularly interesting.

CONCLUSIONS

In conclusion, DLCs have emerged as promising candidates for optoelectronic applications, with their unique properties such as high charge-carrier mobility along the stacking axis, self-assembly, and solubility in various organic solvents. The use of Scanning Tunneling Microscopy (STM) in exploring the structure and properties of DLCs has provided valuable insights into the behavior of these materials at the nanoscale. By visualizing DLCs at the molecular level and measuring the tunneling current between the STM tip and the DLC material, STM has enabled the investigation of the charge transport properties of DLCs and their potential for use in electronic

devices. The manipulation of DLCs using STM has also allowed for creating patterns or structures useful for electronic device construction. Overall, exploring DLCs for optoelectronics applications via STM holds great potential for developing efficient and innovative electronic devices.

Exploring discotic liquid crystals (DLCs) for optoelectronic applications through scanning tunneling microscope (STM) provides a promising avenue for developing novel electronic devices. STM enables the observation of molecular arrangement and electronic properties of DLCs at the nanoscale level. Through the manipulation of the DLCs' self-assembly process, it is possible to control their morphology, such as column orientation within prescribed 2D lattices, and tailor their properties for specific applications. The alignment of DLCs through established methods such as zone-casting, Langmuir-Blodgett deposition, or electric field impact, allows for the control of their electronic properties and charge-carrier

mobility (CCM). STM also enables the study of DLCs' supramolecular chirality, which can be reversibly altered by driving the twisted sense. It opens the possibility of developing information frameworks to switch their chiroptical characteristics. The interdisciplinary approach required for developing DLCs for optoelectronic applications extends beyond the typical boundaries of organic chemistry. It applies to various fields such as cosmetics, pharmaceutical formulations, nanomaterials, and catalysis. In conclusion, the use of STM in exploring DLCs offers a promising avenue for developing functional materials and devices for optoelectronic applications.

Conflict of Interest

The authors declare no conflict of interest.

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