

Phase Transitions in Liquid Crystals With Disk-Like and Graphene Molecules

Naskalova O.V.

Department of physics and ICT
Chechen state university
Grozny, Russia
naskalova_o@mail.ru

Larin E.S.

Research institute of physics
Southern Federal University
Rostov-on-Don, Russia
eslarin@mail.ru

Kuznetsova E.M.

Institute for Mathematics, Mechanics, and Computer Science in the name of I.I. Vorovich
Southern Federal University
Rostov-on-Don, Russia
esmit@mail.ru

Abstract—The research of possible types of phase diagrams in liquid crystals with discotic mesogens and “graphene molecules” was carried out within the framework of the Landau theory of phase transitions. The possible types of ordering of mesogens in such systems are considered.

Keywords—liquid crystals, discotic, nematics, mesophases

I. INTRODUCTION

In everyday life, an increasing number of electronic devices are used, in which various states of a liquid-crystalline substance are used to display information. The development of this direction was largely promoted by calamite (rod-like) liquid crystals (liquid crystal, LC), while discs (disk-like) initially had only limited use (LC-screen films, gas sensors, etc.), but in recent years they are considered as promising concepts of the latest devices. In recent times, a lot of research papers have appeared devoted to the attempt to understand the relationship between the structure – property relation, with a view to further producing substances with necessary mesomorphic and other physical properties and, for example, using them as anisotropic organic semiconductors in organic field-effect transistors (OFETs), organic light emitting diodes (OLEDs) and organic photovoltaic devices (OPVs) [1–4].

The study of liquid crystals based on discotic mesogens began in 1923, when the German scientist D. Vorlander suggested the possibility of the existence of column-type liquid-crystal phases. D. Vorlander studied substances consisting of flat molecules such as triphenylene and perylene. These molecules are planar polycyclic aromatic hydrocarbons consisting of three or more benzene rings [5, 6]. Unfortunately, he was not able to observe the manifestation of mesomorphism (the existence of liquid-crystalline phases) for these complexes, which were later recognized as typical basic units of many discotics. The experimental breakthrough happened in 1977, when S. Chandrasekhar published the results on the mesomorphic properties of aromatic

hydrocarbons derived from benzene. These data today are considered as the birth of a liquid crystal – discotic [7].

II. THE STRUCTURE OF DISCOTIC LIQUID CRYSTALS

Let us briefly consider the structure of the main phases of discotic. It is possible refer to more detailed description of the structures and their characteristics obtained in the investigation by X-ray diffraction and a polarizing optical microscope [8].

Discotic liquid crystals are composed of molecules with a disc-shaped core, which is surrounded by alkyl, sufficiently long chains. Particular interest is presented by discotic mesogens formed by an aromatic nucleus surrounded by aliphatic chains that do not contain benzene rings. In such compounds, a large number of phase transitions are usually observed [9–11]. Therefore, disk-shaped systems are of great interest from the point of view of basic research in the physics and chemistry of liquid crystals.

The packaging of disc-shaped mesogens in liquid crystals in 1D-column is the most common. Ideally, the columns have infinite length, and the molecules in the column have only close-range order. The mesogens with their solid, relatively flat nuclei self-organize into columns. This ordering is caused by the steric factor and $\pi - \pi$ interactions between the nuclei [12]. High entropy and disorder of flexible aliphatic chains on the periphery prevent the formation of a 3D crystal. Column packing also leads to nano- segregation between the nuclei and tails.

There are different types of ordering in the columns. “Distributed columns” with irregular packing of disks, “ordered columns” where the nuclei are equidistant, while flexible alkylated tails are disordered, as well as “lateral columns” with disk nuclei inclined relative to the axis of the column. All of these columns do not represent the correct 1D translational order and therefore can be considered as 1D-fluids.

Nematic phases – these are the simplest liquid crystal phases, formed as states that have a long-range orientational order (molecules, columns, etc.), but there is no longitudinal translational order. For discotic molecules, various nematic phases are known.

(Figure 1)

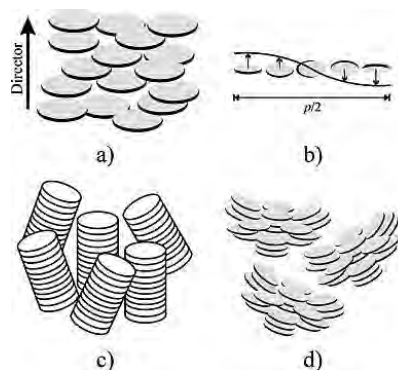


Fig. 1. Various nematic phases of discotic liquid crystals: (a) nematic discotic N_D ; (b) cholesteric discotic N_D^* ; (c) column nematic N_C ; (d) lateral nematic phase N_L [13]

In discotic, the nematic mesophase N_D is built from single, flat molecules that have complete translational and orientational freedom around their main short axes. However, their longitudinal axes have an average oriented order (Fig. 1a). There is also a chiral discotic variant with a nematic phase, known as the cholesteric N_D^* phase (Fig. 1b), which is formed by the chiral discotic mesogen or after the addition of chiral molecules to the achiral discotic. Figure 1a shows a spiral visible nematic structure, where the director's area is continuously bent normally towards local director n (r) (Fig. 1b) with a pitch p of the helical structure.

N_C column nematic mesophase is formed from columns as building blocks (Fig. 1c). For example, the variants of an electron donor doped by an electron acceptor are possible [14]. In this case, the columns are ordered by charge transfer interactions. The formation of a 2D grid is impeded by the molecules with different lengths of their chains. In this case, the columns are lined up parallel to each other in the columnar nematic mesophase and therefore show the translational short range order and orientational long range order.

A lateral nematic phase of N_L (fig. 1d) is also possible. In this case, the discotic molecules form large disc-shaped blocks that behave like the N_D nematic. Their local ordering is combined with the absence of long-range translational order. The nematic phases described above are uniaxial (axial) ones.

In the case of disco-shaped liquid crystals, the formation of a biaxial (biaxial) nematic phase is possible in the compounds of rod-like and discotic mesogens [15]. Such compounds of molecules have been studied by many groups of scientists using computer modeling [16–18]. The simulations have shown the emergence of a biaxial nematic phase. However, experimentally the similar structures have not yet been obtained.

Liquid crystals with discotic molecules are also interesting for obtaining new ferroelectric materials with chiral molecules.

Ferroelectric crystals have a non-zero electric polarization even in the absence of an electric field. Such spontaneous

polarization is denoted as P_s . If the P_s direction can be switched by an external electric field between two stable states of free energy, the material is called a ferroelectric. Pyro- and ferroelectricity is possible only in phases with symmetry belonging to ten polar point groups: C_s , C_n ($n = 1, 2, 3, 4$, and 6) and C_{nv} ($n = 2, 3, 4$, and 6). Since ferroelectricity requires long-range dipole correlations, it was believed that this physical phenomenon is inherent only in the crystalline state.

However, in 1974, the R.B. Meyer discovered that the smectic (C) phase with chiral rod-shaped molecules has a polar symmetry C_2 . It explains the existence of spontaneous electric polarization in the liquid-crystalline state (Fig. 2). The work of Meyer et al. [19] discovered a completely new field of research, namely, ferroelectric and later antiferroelectric liquid crystals [20, 21].

For the first time ferroelectric column liquid crystal (FCLC) with C_2 type symmetry was observed by H. Bock and W. Helfrich [22] in 1995 for dibenzopyrene, which has a disk shape with chiral and strongly polar carboxyl side chains (Fig. 2a). The observation of stable spontaneous polarization of P_s along the columns in FCLC was first reported in [23] in 2012.

	Introduction of chirality	Packing of polar molecules
Rod shape	Chiral SmC* LCs C_2	Bent-core LCs C_{2v}
Disc shape	Chiral Disc LCs C_2	Conical & Bowl LCs $C_{\infty v}$

Fig. 2. Liquid crystal phases with polar point symmetry groups that allow the appearance of spontaneous P_s polarization: (a) chiral smectic (SmC*) phase with C_2 and P_s symmetry along the C_2 -axis (normal to the figure surface); (b) inclined chiral molecules having a disk shape, forming a column with symmetry C_2 and P_s along the C_2 -axis (normal to the figure surface); (c) the smectic phase with achiral banana mesogens with C_{2v} and P_s symmetry along the lateral direction (C_2 -axis); (d) conical molecules or molecules in the form of a bowl that form a column with symmetry $C_{\infty v}$ and P_s , directed along the axis of the column (axis C_{∞}) [21].

III. THERMODYNAMIC POTENTIAL MODEL

The purpose of this research was to consider the orientational behavior in liquid crystals with discotic mesogens during thermotropic (temperature change) and lyotropic (change in the shape of mesogens and concentration) phase transitions.

With the change in the temperature or molecular geometry of the mesogens in discotic liquid crystals, the phase transitions from the isotropic (completely disordered) phase to the ordered discotic nematics N_D and various columnar N_{Col} phases can be observed (Fig. 3). Nematic discotic phases may be uniaxial N_D and biaxial N_{Db} , by analogy with rod-like liquid crystals [24]. The uniaxial N_D nematic phase has point

macroscopic symmetry $D_{\infty h}$. The biaxial N_{Db} nematic phases arise from the N_D phase as a result of the molecular biaxiality of the discotic molecules, which prevents their rotation around the normal line (n director) to the plane of the disk. In this case, the point macroscopic symmetry is D_{2h} .

We have carried out a theoretical study of transitions in liquid crystals with discotic molecules from the isotropic to nematic phases N_D , N_{Col} , N_L , N_{Db} within the framework of the Landau theory of phase transitions. The stability of mesophases in liquid crystals with discotic molecules is determined by various types of competing interactions, which are responsible for the self-organization of the columns and their spatial ordering. The appearance of spontaneous orientation of hard disk-shaped nuclei leads to the anisotropy of the shape of blocks containing a certain number of mesogens. We choose the traceless tensor of the 2nd rank as the leading order parameter in the model under consideration, [25]:

$$Q_{ik} = \frac{1}{2}\eta_1(3\eta_i\eta_k - \delta_{ik}) + \frac{\sqrt{3}}{2}\eta_2(l_i l_k - m_i m_k).$$

The order parameter, which is transformed by the same irreducible representation, can be any material tensor of the 2 rank ξ_{ik} . It can be the deformation tensor u_{ik} . Then the Landau thermodynamic potential, describing the interaction of two order parameters, has the following form:

$$\Phi_{int} = \gamma Q_{ik} \xi_{kl}.$$

We choose the system of axes l_i , $i = x, y, z$ (l_i – single vectors along x, y, z accordingly), in which Q_{ik} – diagonal tensor, then we obtain:

$$Q_{xx} = -\frac{1}{2}(\eta_1 - \sqrt{3}\eta_2), Q_{yy} = -\frac{1}{2}(\eta_1 + \sqrt{3}\eta_2), Q_{zz} = \eta_1,$$

$$Q_{ik} = 0, \text{ when } i \neq k; i, k = x, y, z.$$

In this case it is possible to show that a real rational basis of invariants is composed of 2 variants and has the form of:

$$I_1 = \text{spur}(Q_{ik})^2 = \frac{3}{2}(\eta_1^2 + \eta_2^2); I_1 = \text{spur}(Q_{ik})^3 = \frac{3}{2}(\eta_1^3 - 3\eta_1\eta_2^2).$$

In this system of axes the tensor ξ_{kl} can also be led to diagonal form with own values ξ_1 and ξ_2 . In this case it has the form of:

$$\Phi_{int} = \gamma(\eta_1\xi_1 + \eta_2\xi_2).$$

With the limitation up to the 6th degree, we choose as the full model of the unbalanced thermodynamic potential [24]:

$$\begin{aligned} \Phi = & a_1 I_1 + a_2 I_1^2 + a_3 I_1^3 + b_1 I_2 + b_2 I_2^2 + c_{12} I_1 I_2 + \\ & + \gamma(\eta_1\xi_1 + \eta_2\xi_2) + \alpha(\xi_1^2 + \xi_2^2). \end{aligned} \quad (1)$$

The stability of the model of thermodynamic potential (1) is determined by the following conditions: $a_3 > 0$, $b_2 > 0$, $\alpha > 0$. Further, we will consider the model with $a_2 > 0$, $\gamma < 0$. In this case, model (1) assumes that variable parameters depending on the conditions on the thermostat are presented by two phenomenological parameters a_1 and b_1 .

The system of equations of state that determines the extremal points of the thermodynamic potential (1) and has the form of

$$\frac{\partial \Phi}{\partial \eta_i} = 0, \text{ where } i = 1, 2$$

and determines four different phases:

$$I: \eta_1 = \eta_2 = 0; \xi_1 = \xi_2 = 0;$$

$$N_{col}: \eta_1 > 0, \eta_2 = 0; \xi_1 > 0, \xi_2 = 0;$$

$$N_D: \eta_1 < 0, \eta_2 = 0; \xi_1 < 0, \xi_2 = 0;$$

$$N_{Db}: \eta_1, \eta_2; \xi_1, \xi_2;$$

Figure 3 presents a schematic view of nematic phases in a polymer discotic, described by the thermodynamic potential (1).

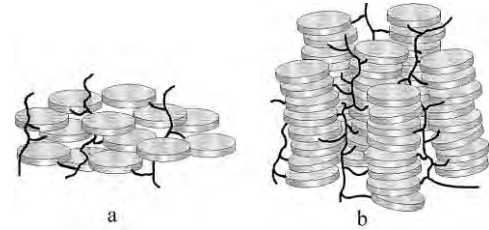


Fig. 3. Schematic representation of nematic phases in discotic: (a) N_D phase; (b) N_{col} phase [26]

The detailed analysis of the phase diagram of the potential $F(\eta_1, \eta_2)$, conducted by methods of the theory of catastrophes, showed that two types of phase diagrams are possible in the space of variables a_1, b_1 (Fig. 4).

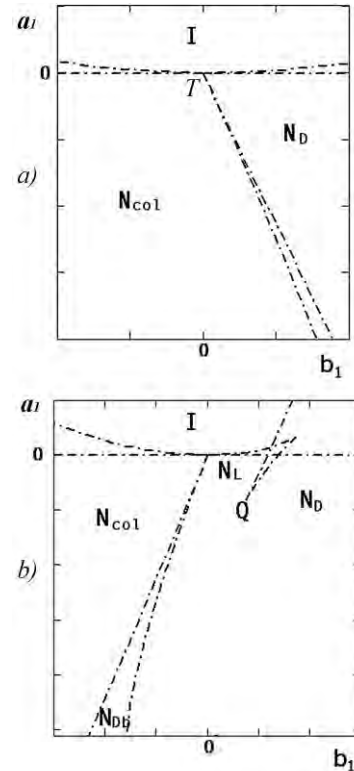


Fig. 4. Phase diagrams of the thermodynamic potential model P (1): (a) for $c_{12} > 0$; (b) at $c_{12} < 0$. The dashed lines are the lines of phase stability.

IV. RESULTS AND DISCUSSION

1. $I-N_{col}$, $I-N_D$, and N_D-N_{col} phases are the transitions of the 1st type. On the phase diagram, there is an area of coexistence of phases I and N_{col} , N_D and N_{col} , as well as I and N_D (Fig. 4a), within which the line of transition of the 1st type passes. The lines of phase transitions of the 1st type converge at a three-phase point T . The topological features of the phase diagram coincide with the experimentally observed ones, if we assume that the variable parameters a_1 , b_1 depend on temperature and weight concentration of monomers [26, 27].

2. It is important to note that a phase transition of the 1st type between the N_D and N_{col} phases at $c_{12} < 0$ splits into a sequence of phase transitions of the 2nd type: $N_D - N_{Db}$ and $N_{Db} - N_{col}$ (Fig. 4b). In this case, the three-phase point T in the phase diagram (Fig. 4a) becomes four-phase (Fig. 4b).

3. The condition for the formation of columnar phase N_{col} is: $\eta_1 > 0$, $\zeta_1 = u_{zz} > 0$, $\zeta_2 = u_{xx} = u_{yy} = 0$. Then, due to local deformation stresses of the medium, the discotic mesogens oriented along the director n tend to be ordered into the columns. At the same time, the geometric dimensions of discotic mesogens and their number in columns will determine the height of the columns in the N_{col} phase. Depending on the length / diameter of the columns, the appearance of the smectic phase along with the columnar ordering is possible. The stability of a particular ordering will be determined by the ratio of the length / diameter of the columns [28-30].

4. The phases N_L ($\eta_1 > 0$; $\zeta_1 = u_{zz} > 0$) and N_D ($\eta_1 < 0$; $\zeta_1 = u_{zz} < 0$) are isomorphic and an isomorphic first-order phase transition occurs between them, ending at the critical point Q (Fig. 4b). Above the critical point Q , a continuous change in the structure of the N_L phase to the N_D phase occurs due to the destruction of the disk-shaped blocks.

5. The formation of the biaxial N_{Db} phase occurs as a result of the change in the average position of the centers of gravity of the discotic molecules. The increase in biaxiality leads to a decrease in the value of $|u_{zz}|$.

6. The breakthrough of Heim and Novoselov in 2005 of two-dimensional graphene gave rise to a rapid growth in the creation of electronic devices of a new type [32, 33]. Numerous studies of graphene have shown the uniqueness of its physical, electrical, optical, mechanical, thermal, and other properties. This, in turn, is of interest in conducting fundamental research on systems containing graphenes and "graphene molecules" dissolved in them.

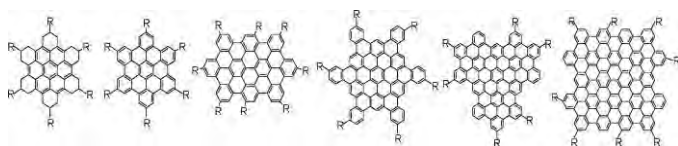


Fig. 6. Schematic view of large graphene macromolecules (mesogens) [31].

The work [31] includes various schemes for the synthesis of huge molecules of various sizes and symmetries with a solid graphene nucleus (Fig. 6). The disc shape of such molecules (mesogens) allows considering the possibility of the appearance of various columnar phases in the system during

the thermotropic transition. Two-dimensional graphene serves as the nuclei of such a mesogen [34].

V. CONCLUSION

The breakthrough of American scientist S. Chandrasekhar achieved more than 40 years ago on the ability to self-organize disk-shaped molecules into column mesophases stimulated numerous studies. Especially promising is that some of the discotic liquid crystals can be considered as organic semiconductors. The synthesis of such compounds has generated a huge structural diversity, and good solubility provided an advantage over solid semiconductors.

The future use of disc-shaped graphene molecules in electronics or photonics depends on improving the mobility of the transfer of internal charge, controlling the ordering of graphene super molecules in thin films, and the processes of creating devices. These requirements involve the combined efforts of chemists, physicists and engineers. Further miniaturization of devices to the molecular scale is possible; however, the key issue is devoted to the method how various electronic elements may be combined and assembled into ordered nanostructures. The carbonization of graphene molecules provides a good prediction of structures and electronic properties.

These graphene macromolecules can also be transformed into 1D carbon nanotubes with a well-controlled graphene layer orientation and solid-state pyrolysis process directed to the substrate. Semiconductor nanotubes can be used, for example, in field transistors.

In addition, chemical modification of the units of these special carbon nanotubes or filling them with various pore substances can lead to complex nanostructures that are interesting for the creation of new miniature electronic devices.

Another promising area is the preparation and investigation of solutions, suspensions of liquid crystalline compounds and carbon nanotubes (graphenes). During the course of the research [35] various types of phase diagrams in such systems were considered within the framework of the phenomenological theory of phase transitions. In this case, the variety of options are realized: discotics (disk-like) - nanotubes (rod-like); disk-like - graphene macromolecules (disk-like); calamite liquid crystals (rod-like) - graphene macromolecules (disk-like), etc.

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