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Pierre-Gilles de Gennes: Beautiful and mysterious liquid crystals

Pierre-Gilles de Gennes : Beaux et mystérieux cristaux liquides

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ABSTRACT

The articles of Pierre-Gilles de Gennes on liquid crystals published between 1968 and 1972 in the *Comptes rendus de l'Académie des sciences* appear today as models of conciseness. On barely three pages, with a few words and equations, de Gennes was able to give answers to enigmas posed by strange and often beautiful phenomena occurring in liquid crystals. Thread-like disclinations in nematics and cholesterics, dislocations in smectics or milky aspect of nematics due to light scattering are a few examples of them. Pierre-Gilles de Gennes was able to examine them from heights accessible only to him, to explain them and also to find how they are related to phenomena occurring in other ordered systems. His article on "Singularities permitted in ordered phases" laid the foundation for future work on the classification of topological defects. Our aim is to illustrate issues raised by de Gennes by examples taken from very recent experiments. By doing this, we prove that de Gennes' achievements remain valid and valuable today.

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RÉSUMÉ

Les articles de Pierre-Gilles de Gennes sur les cristaux liquides publiés entre 1968 et 1972 dans les *Comptes rendus de l'Académie des sciences* frappent par leur concision. Sur trois pages à peine, avec peu de mots et d'équations, de Gennes esquissait les solutions d'énigmes posées par les étranges et souvent beaux phénomènes observés dans les cristaux liquides ; filiformes discinclinaisons dans les nématiques, dislocations dans les smectiques ou l'aspect laiteux des nématiques n'en sont que trois exemples. Pierre-Gilles de Gennes était capable de les examiner d'une hauteur á lui seulement accessible, de les expliquer et de les relier aux phénomènes se produisant dans d'autres systèmes ordonnés. Dans son article *Types de singularités permises dans une phase ordonnée*, de Gennes jetait les bases de travaux futurs sur la classification des défauts topologiques. Notre ambition est ici d'illustrer les problèmes considérés par de Gennes avec des exemples tirés de travaux très récents. De cette manière, nous montrons que les idées de Pierre-Gilles de Gennes restent toujours vivantes.

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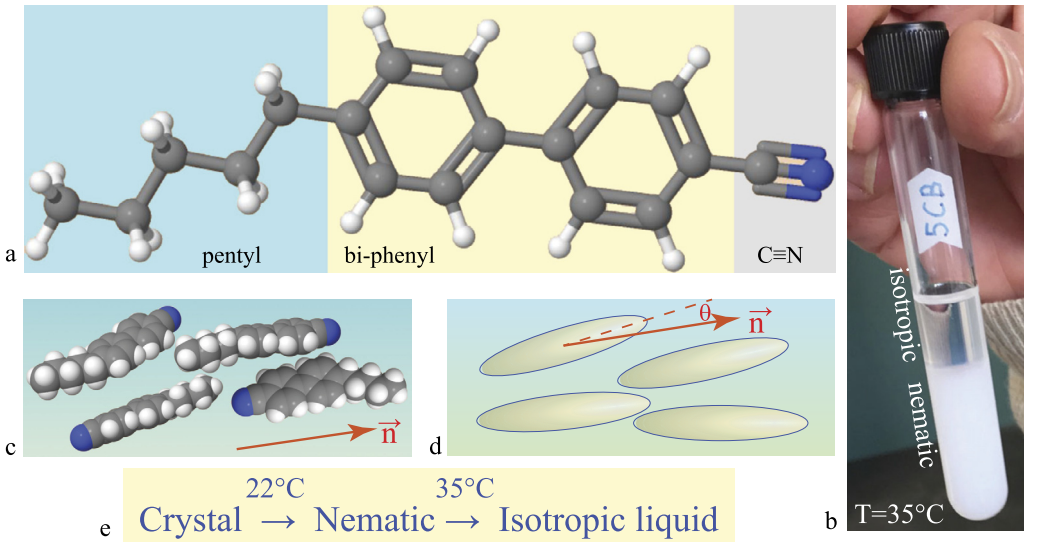

Fig. 1. 5CB – a stable synthetic compound displaying the nematic phase at room temperature [12]. a) The hybrid molecular structure of 5CB; taken separately, pentane and bi-phenyl would be respectively liquid and crystalline at room temperature. b) View in unpolarised white light of the nematic–isotropic phase transition occurring at $T = 35\,^{\circ}\mathrm{C}$. The turbid aspect of the nematic phase will be discussed in section 6. c) Uniaxial orientational order of elongated molecules. d) Nematic order of ellipsoids – quadrupolar objects. e) Phase sequence of 5CB.

1. Introduction

1.1. The fascinating complexity and beauty of liquid crystals

The articles of Pierre-Gilles de Gennes published in the *Comptes rendus de l'Académie des sciences* [1–9] are like his drawings, in which with a very few lines he was able to express the subtle beauty of the model. In his articles, using very few words and equations, de Gennes introduced new theoretical concepts making understandable experimental facts. Our aim here will be to provide experimental illustrations to de Gennes' articles and by this means to show how complex and beautiful is this physical reality explained with a few words and equations.

De Gennes was fascinated by the complexity and the beauty of liquid crystals and he expressed his feelings in the motto of the first edition of his *opus magnum* "The Physics of Liquid Crystals" [10]: *Liquid crystals are beautiful and mysterious and I am fond of them for both reasons*. The reader understands now why such a unusual title was chosen for this article.

1.2. Scenery and characters

Most of the articles on liquid crystals published by de Gennes in the *Comptes rendus* between 1968 and 1972 were written in the "Laboratoire de physique des solides" in Orsay. Attracted there by Jacques Friedel (see chapter 13 "*Orsay, les dix glorieuses*" in Ref. [11]), de Gennes created first a team dealing experimentally and theoretically with superconductivity. Then, in 1968, he turned his attention to liquid crystals. Although studies in this field had been initiated in France already half a century ago by George Friedel (grandfather of Jacques), de Gennes felt that with his knowledge of systems with long-range order, expressed by scalar, vectorial or complex-order parameters, he would be able to shed a new light on the structures and properties of liquid crystals. Several senior scientist (Madeleine Veyssie, Georges Durand, Étienne Guyon, Maurice Kléman, Olivier Parodi) and a swarm of enthusiastically acting PhD students (they were too many to list them here) joined de Gennes, and an avalanche of new results was triggered. For most of them, working under the auspices of de Gennes was the chance of their scientific life.

2. Reminder on liquid crystalline phases

2.1. Long-range orientational order in nematics

As already stated, our aim here is to relive the difficulty of reading extremely concise de Gennes' papers by means of simple experimental illustrations. To start with, let us introduce the compound known as 5CB (see Fig. 1) that we will quote below very often. In 1968, when de Gennes started to work on liquid crystals, many other compounds such as MBBA or PAA were known to display the nematic phase, but they were not easy to use in experiments because either the temperature range of the nematic phase was too high (PAA) or they were chemically unstable (MBBA). In contradistinction, the 5CB, synthesized in 1973 by G. Gray et al. [12], had none of these drawbacks, so that today it is used (in mixture with other

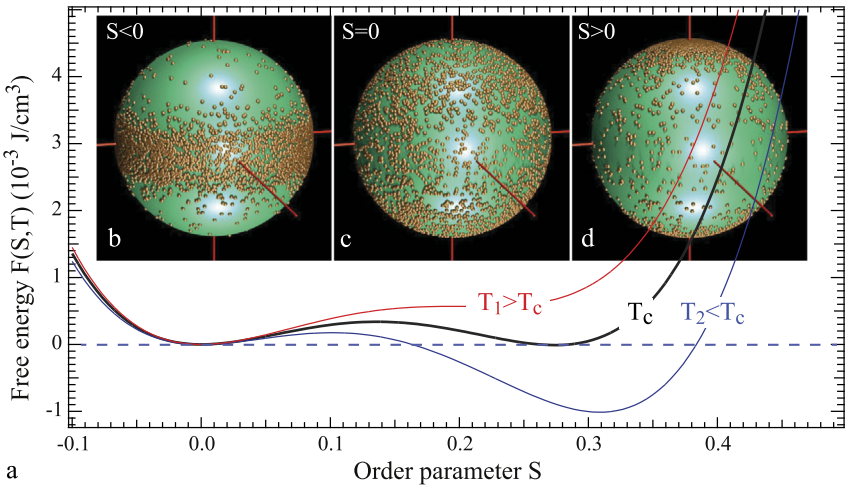

Fig. 2. Landau–de Gennes theory of the nematic → isotropic transition. a) Plot of the free energy expression given by Eq. (2) for three values of the temperature T. b–d) Visualisation of molecular orientations typical for $S < 0$, $S = 0$, and S> 0. The dots on the unit sphere represent the orientations of the individual molecules. The distributions of molecular orientations for $S > 0$ and $S < 0$ are not related by any symmetry. For this reason, the free-energy expansion (Eq. (2)) contains the nonzero third-order term.

substances) for the production of liquid crystals displays. Indeed, in the phase sequence of 5CB (see Fig. 1e), the nematic phase occurs at room temperature and chemically this compound is very stable.

When a sample of 5CB in its nematic phase is slowly heated in a test tube, even a layman will notice a spectacular phenomenon: the nematic → isotropic phase transition that occurs at 35 °C as shown in Fig. 1b. Its first-order character can be inferred from the obvious optical discontinuity as well from the density discontinuity: the isotropic phase floats above the nematic phase because its density is lower. As shown in Fig. 1c, in the nematic phase, the 5CB molecules are free to rotate around their long axes that are aligned, on average, in some arbitrary common direction called **n**. For this reason, the nematic phase is often represented as a system of aligned uniaxial ellipsoids. During the nematic → isotropic transition, this long-range orientational order is broken and the 5CB molecules are entirely free to rotate.

The first-order character of the nematic → isotropic transition was understood in 1958 by W. Maier and A. Saupe [13] in terms of a mean-field theory (analogous to the Curie–Weiss theory of the ferromagnetic → paramagnetic transition), in which a scalar order parameter $S = (3\langle\cos^2\theta\rangle - 1)/2$ played the main role (θ is defined in Fig. 1d as the angle between individual molecules and their common direction **n**).

2.2. Landau–de Gennes theory of the nematic → isotropic transition

One of the first major achievements of de Gennes consisted in his introduction of the tensorial order parameter S_{ij} and construction of the Landau-type free-energy expansion in terms of invariant powers of S_{ij} and of its gradients [14]:

$$F = F_o + \frac{1}{2}A(T)S_{ij}S_{ji} - \frac{1}{3}BS_{ij}S_{jk}S_{ki} + \frac{1}{4}(S_{ij}S_{ji})^2 + \frac{1}{2}L_1\partial_i S_{jk}\partial_i S_{jk} + \frac{1}{2}L_2\partial_i S_{ik}\partial_j S_{jk} \quad (1)$$

By this means, de Gennes established a bridge between liquid crystals and other ordered systems for which the Landau theory of phase transitions was applied previously. In the case of usual, optically uniaxial nematics, the tensorial order parameter can be written as $S_{ij} = S(n_i n_j - \delta_{ij}/3)$ and the free energy expansion takes a very simple form:

$$F = F_o + \frac{1}{2}A_o(T - T^*)S^2 - \frac{1}{3}BS^3 + \frac{1}{4}S^4 + \ldots \quad (2)$$

Let us stress that here S is nothing else but the scalar order parameter used by Maier and Saupe.

The most important feature of this expansion is that it contains the nonzero third order term, which is allowed by the lack of symmetry upon the change of the sign of S (see Figs. 2b and d). For this reason, the nematic → isotropic transition is of the first order (Fig. 2).

2.3. Cholesteric phase

When a chiral impurity is dissolved in 5CB, the alignment of the molecules in one common direction **n** is replaced, in the simplest case, by a helical configuration shown in Fig. 4c, which is the fingerprint of the cholesteric phase. The pitch p of the helix depends on the concentration c and tends to infinity when $c \to 0$. For this reason, the cholesteric phase can be seen as a chiral version of the nematic phase.

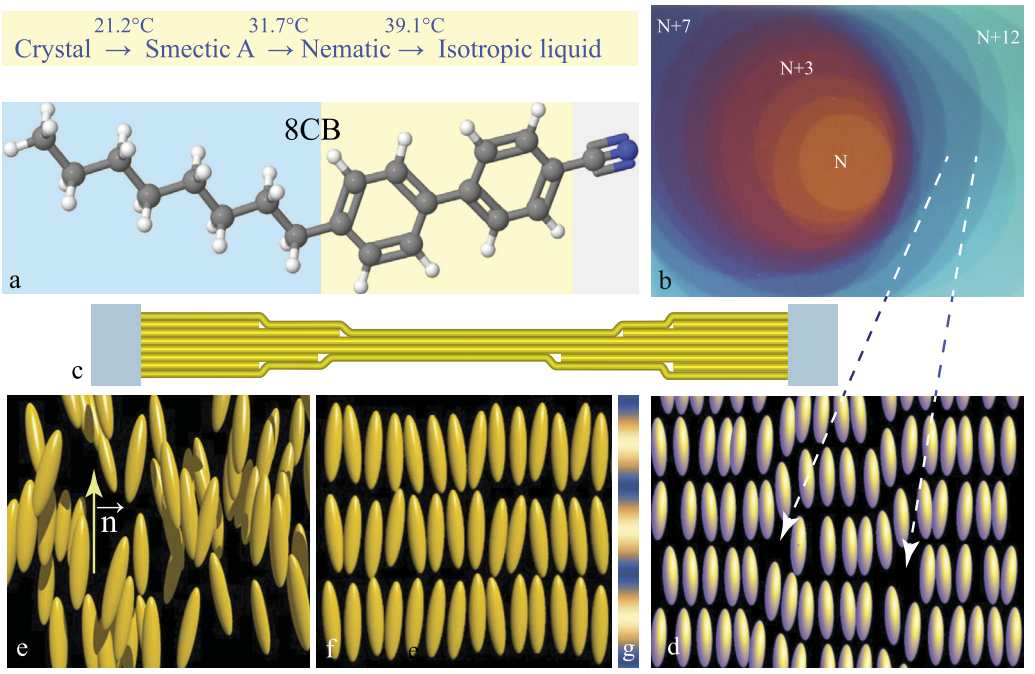

Fig. 3. Smectic A phase. a) Molecular structure of 8CB and its phase sequence. b) View in a reflecting microscope of a free-standing smectic A film. Colour discontinuities correspond to unitary changes in the number of smectic layers. c) Cross section of a free-standing smectic film suspended on a rigid frame. d) Schematic view of wedge dislocations in the free-standing film. e–f) Birth of the lamellar order during the nematic → smectic A transition. g) The order parameter resulting from the breaking of the translational symmetry: periodic modulation of the mass density.

2.4. Lamellar order in the smectic A phase

The 5CB compound is a member of a homologous series of mesogenic molecules *n*CB that differ by the length n of the aliphatic chain. For $n = 8$, that is to say, with the compound known as 8CB, beside the nematic → isotropic phase transition discussed above, another spectacular phenomenon can be easily observed: the existence of free-standing films, similar to soap bubbles, that can be drawn on rigid frames of arbitrary shapes.

Like soap bubbles, smectic films display beautiful interference colours (see Fig. 3b). However, unlike in soap bubbles, their spectrum is not continuous, but discrete, as it can be inferred from the picture in Fig. 3b. This is the fingerprint of the smectic A phase, which can be seen as a stack of liquid layers with molecules orthogonal to them (Fig. 3d). In the free-standing film shown in Fig. 3b, the number N of smectic layers is small and it increases one by one from the central orange circular field toward the meniscus connecting the film to the frame. By the way, let us remind that the name "smectic" was given to this phase by Georges Friedel because the Greek word $\sigma\mu\eta\gamma\mu\alpha$ means "soap".

In 8CB (see the phase sequence in Fig. 3a), the smectic phase occurs upon cooling of the nematic phase. During this phase transition, the orientational order parameter $S_{ij} = S(n_i n_j - \delta_{ij}/3)$ is preserved, but the continuous translational symmetry is broken in the z direction taken parallel to **n**. The mass density becomes a periodic function of z and can be represented as a Fourier series:

$$\rho(z) = \rho_0 + \mathrm{Re}\{\rho_1 e^{i\varphi} e^{iq_0 z}\} + \dots \tag{3}$$

De Gennes remarked that the complex order parameter $\Psi = \rho_1 e^{i\varphi}$ resulting from this symmetry breaking is analogous to the one of superconductors [15]. As we will see in section 4.2, this analogy was very fruitful.

3. Disclinations

3.1. Cano wedge

The article entitled "Structure des cloisons de Grandjean–Cano" [1] starts with the following sentence by way of an introduction: "*Inséré dans une fente de clivage (…), un cristal liquide cholestérique montre au microscope polarisant une succession de plages séparées par des bords très nets.*" Its meaning is explained in the capture of Fig. 4. Indeed, when observed in a polarising microscope (Fig. 4a), a wedge made of a cleaved mica sheet and filled with a cholesteric liquid crystal displays a system of stripes separated by colour discontinuities. In equilibrium, in the absence of surfaces, the director field of the cholesteric would adopt the twisted configuration depicted in Fig. 4c, characterised by an equilibrium twist wave vector

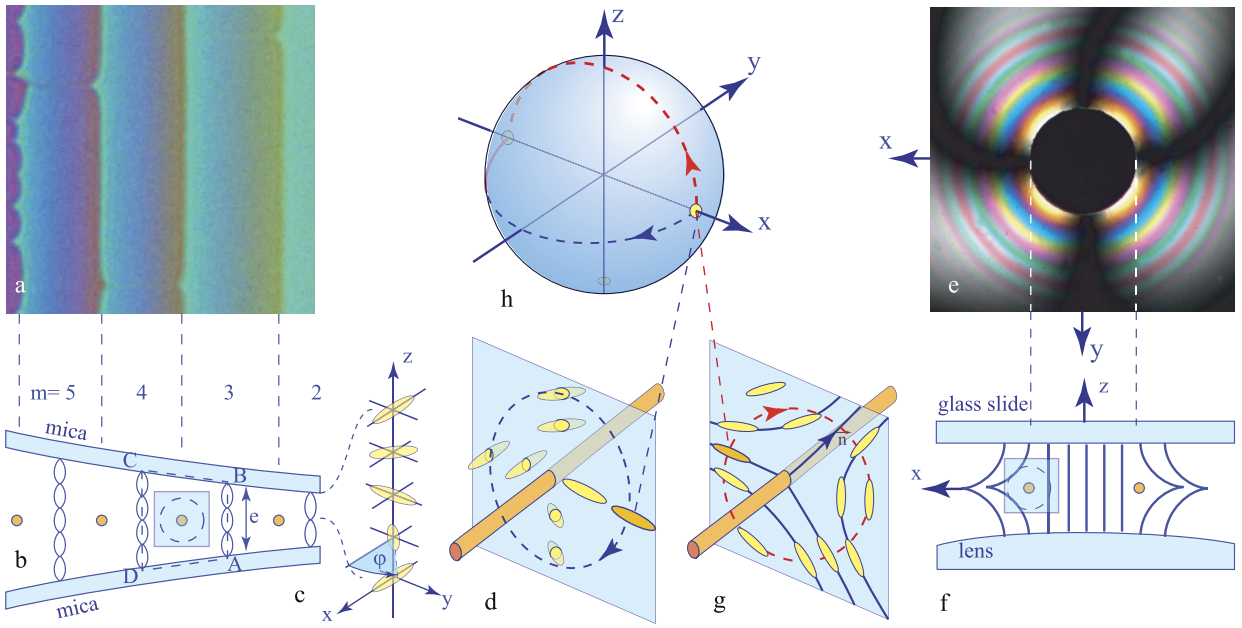

Fig. 4. Disclinations – linear topological defects in nematic and cholesteric mesophases equipped with a quadrupolar order parameter $S_{ij} = S(n_i n_j - \delta_{ij}/3)$. a) Grandjean–Cano texture in a wedge filled with a cholesteric. View in a polarising microscope. b) The wedge is formed inside a partially freshly cleaved mica sheet. Surfaces of mica induce planar anchoring parallel to the x axis. Molecules rotate by $m \cdot \pi$ inside the gap. c) Perspective view of the cholesteric helix. d) Detailed view of molecular orientations in the vicinity of the disclination separating the strips with $m = 3$ and $m = 4$. Molecular orientations encountered on a the circular dashed loop are mapped on the unit sphere in picture h where they form the semi-circular blue trajectory passing from x, through $-y$, to $-x$. e–g) Disclination loop in a nematic filling the gap between a lens and a glass slide. The glass surfaces induce homeotropic anchoring parallel to the z avis. As shown in Figs. 5c and d, homeotropic anchoring is also induced by the air/nematic interface at the lateral meniscus. e) View in a polarising microscope with crossed polarisers. f) xz cross section of the sample shown in e. In the centre (inside the black area in e), the orientation of molecules **n** is uniform and parallel to the z axis (optical axis). In the periphery (outside of the black area in e), the orientation of the molecules rotates by π from the bottom to the top. g) Detailed view of molecular orientations in plane intersecting the disclination loop. The molecular orientations encountered on the circular dashed loop are mapped on the unit sphere in picture h, where they form the semi-circular red trajectory passing from x, through z, to $-x$.

$\mathbf{q}_o = (\pi/p_o)(0, 0, 1)$ parallel to the z axis. If φ is the angle between the director **n** and the x axis, then the cholesteric helix is described by

$$\varphi(z) = q_o z = \pi \frac{z}{p_o} \tag{4}$$

In the wedge, the molecules on the mica surfaces are oriented in the direction parallel to the x axis. If, by convention, one sets $\varphi(0) = 0$ on the lower mica surface, then on the upper surface one must have $\varphi(e) = m\pi$ with $m = 0, 1, 2, \ldots$ Using Eq. (4), one finds that the equilibrium configuration exists only for discrete values of the thickness: $e_m = mp_o$. Everywhere else, the wedge is either too thick or too thin. The elastic energy associated with these departures from the equilibrium configuration of the cholesteric helix can be written as:

$$F_m = \frac{K}{2}\left(q_o - \frac{m\pi}{e}\right)^2 \tag{5}$$

K, known as the elastic constant of Oseen–Frank, is of the order $5{\cdot}10^{-12}$ N. F has then the dimension N/m and can be interpreted as a force per unit length. Let $e_{m,m+1}$ be the thickness at the frontier between the stripes m and $m+1$. This frontier is thus submitted to two forces, acting on it from its two sides, and from their balance

$$\frac{K}{2}\left(q_o - \frac{(m+1)\pi}{e_{m,m+1}}\right)^2 = \frac{K}{2}\left(q_o - \frac{m\pi}{e_{m,m+1}}\right)^2 \tag{6}$$

De Gennes finds the thickness $e_{m,m+1}$ at the frontier:

$$e_{m,m+1} = p_o[m + (1/2)] \tag{7}$$

The second result of the de Gennes' paper concerns the structure of the field $\varphi(x, y, z)$ at the frontier: this field has a linear topological singularity called by de Gennes "disinclination", which is known today in French as "disinclinaison" and in English as "disclination". Its nature is explained in Figs. 4d and h. Let us consider a closed trajectory of arbitrary size and

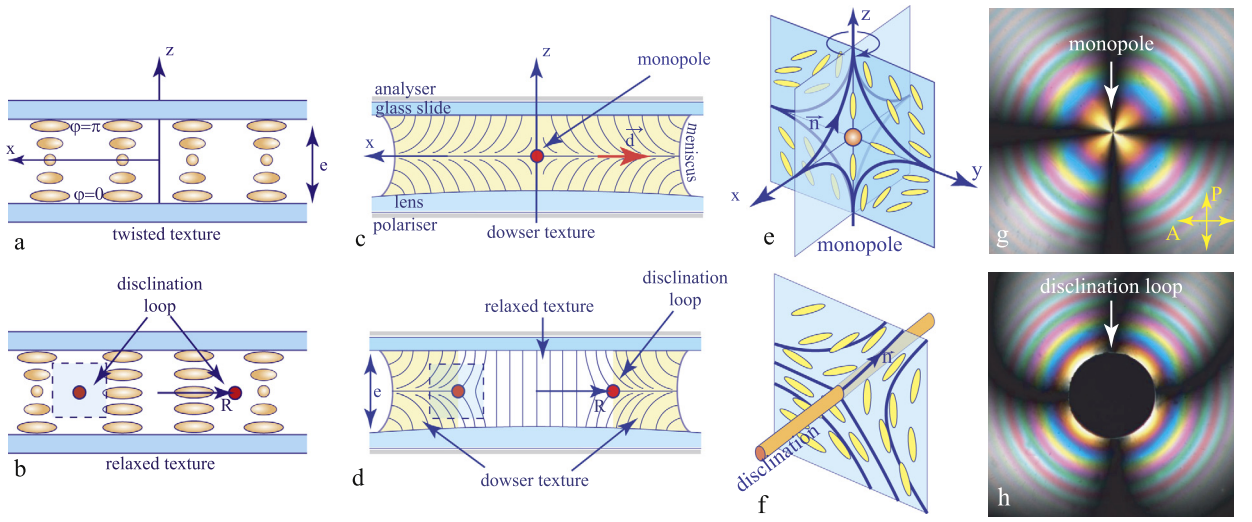

Fig. 5. Nucleation and expansion of a disclination loop. a–b) Geometry of the problem considered by Friedel and de Gennes in Ref. [3]. a) Twisted texture. Molecular orientation **n** rotates by π between the limit surfaces. b) Due to the presence of the disclination loop, the twisted texture is relaxed in the area inside the loop. c–h) Recent experiments analogous to the one proposed by Friedel and de Gennes. c) Texture of the director field **n** inside a drop squeezed between a lens and a glass slide. As lines of the director field **n** have the shapes of a wooden dowser tool, we call it "the dowser texture". The director **n** rotates by π between the lower and upper glass surfaces. Its distortion will be released by nucleation and expansion of the disclination loop. d) Upon reduction of the sample's thickness e below 1 μm, the monopole is transformed into a dislocation loop, which expands. The expansion of the loop is driven by the reduction of the distortion energy – the area of the distortionless homeotropic texture is growing at the expense of the dowser texture. e) Detailed view of the director field in the vicinity of a singular point called monopole. f) Director field in the vicinity of the disclination. g–h) Expansion of the disclination loop nucleated from the monopole observed under the polarising microscope with crossed polarisers.

shape, but necessarily surrounding this line: the quadrilateral ABCD in Fig. 1b or the small circular loop. Orientations of the director **n** (or of molecules) encountered on the small loop in Fig. 4d are mapped on the unit sphere in Fig. 4h. Starting from the orientation parallel to x $\mathbf{n} = (1, 0, 0)$, one follows the red dashed line and arrives at the end of the loop with $\mathbf{n} = (-1, 0, 0)$. Due to the fact that the order parameter of the nematic phase is quadrupolar – $S_{ij} = S(n_i n_j - \delta_{ij}/3)$ – there is no mismatch. Let us remind that the name "nematic" (*nématique* in French) stems from the Greek word $\nu\eta\mu\alpha$ – thread. It was given to this mesophase by Georges Friedel because disclinations that appear as threads under the microscope are ubiquitous; they are fingerprints of this phase. In practice, they are so easy to generate that if one wants to handle them in a controlled manner, special methods must be used, as we will see below.

3.2. Generation of a disclination loop

The second paper, entitled "Boucles de disclination dans les cristaux liquides" deals precisely with the issue of the controlled generation of disclination loops in nematics. It was written with J. Friedel, whose book on dislocations was and still remains an unavoidable reference in this field [16,17]. The authors consider a nematic slab contained between two parallel surfaces with planar anchoring and ask the question: what happens when the azimuthal direction of the anchoring is fixed on one surface (for example, $\varphi(0) = 0$) and is growing on the other surface (for example, to $\varphi(e) = \pi$)? (see Fig. 5a.) The guess of the authors is that the resulting twist distortion $\partial\varphi/\partial z = \pi/e$ can be released by generation and subsequent expansion of a disclination loop shown in Fig. 5b. The loop is submitted to two opposite forces. On the one hand, it has a tendency to shrink because of its own tension $T \approx (\pi K/4)\ln(e/a)$ (see also section 4.2.2 in Ref. [10]). The corresponding centripetal Laplace force per unit length is T/R. On the other hand, the gain in the elastic energy per unit area in the distorsionless area inside the loop results in a centrifuge force per unit length $(K/2)(\pi/e)^2 e$.

As the critical radius of the loop resulting from the balance of these forces turns out to be proportional to the thickness e

$$R_c \approx e\frac{1}{2\pi}\ln\left(\frac{e}{a}\right) \tag{8}$$

de Gennes and Friedel suggest that the nucleation of disclination loops should be possible upon a sufficient reduction of the sample's thickness e. Here we will illustrate these ideas of de Gennes and Friedel with an experiment performed in a different though analogous geometry depicted in Figs. 4c and 5. As the strained director field **n** in Fig. 5c has locally shapes similar to a wooden dowser tool, we call it "the dowser texture". Globally, the director field **n** has a symmetry of revolution around the z axis and contains a singular point called monopole that will be discussed later (Fig. 5e). Using the same arguments as those of de Gennes and Friedel, one can expect that, upon a sufficient reduction of the sample's thickness, a disclination loop can nucleate in the distorted dowser texture and subsequently expand for the benefit of the distortionless

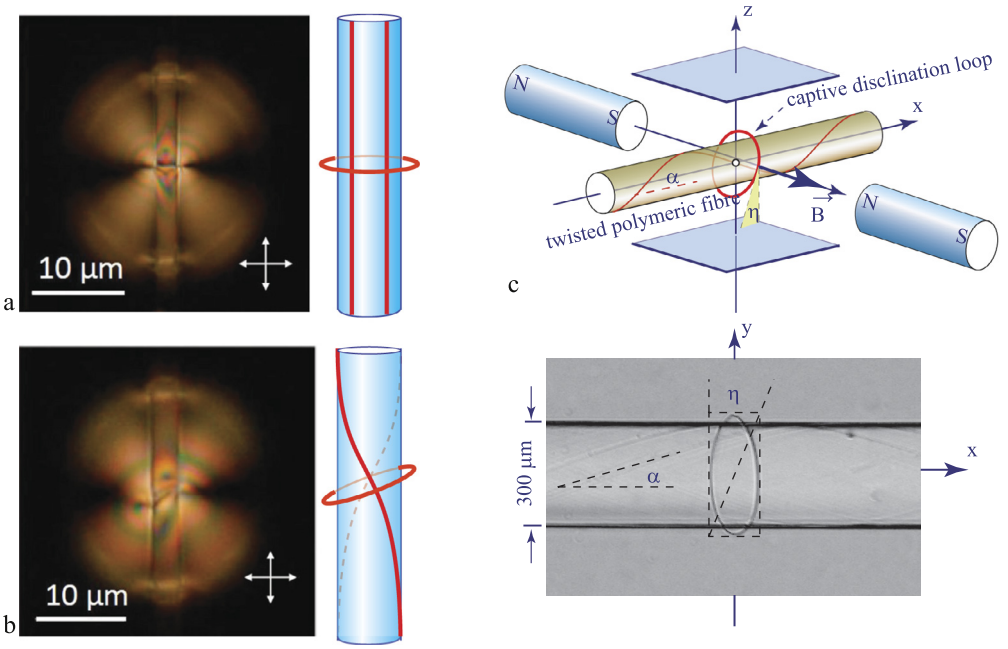

Fig. 6. Captive disclination loops. a) Nematic droplet on a cellulosic fibre electrospun from an isotropic HPC/water solution. The disclination loop threaded on the fibre is orthogonal to the anchoring axial direction (red lines). b) The same experiment with a fibre electrospun from an anisotropic HPC/water solution. The tilt of the loop unveils the helicoidal planar anchoring on the fibre's surface. c) Experiment with a disclination loop threaded on a nylon fibre (fishing line 300 µm in diameter) unveils the chirogyral effect: the tilt of the disclination loop induced by mechanical torsion of the fibre. d) View from the z direction. The tilt angle η is proportional to the torsion angle α.

homeotropic texture. Figs. 5g and h show that actually the expected disclination loop is nucleated and expands. Let us emphasize that the nematic monopole acted as a nucleation centre. In fact, upon the decrease of the thickness, the nematic monopole has been transformed into the disclination loop. This transformation conserves the topological charge of the monopole.

3.3. Collapse of a disclination loop, the dowser texture

After its expansion, the disclination loop does not disappear but stays at rest in the vicinity of the air/nematic meniscus. The radius R of such a *peripheral* disclination loop is thus close to the radius of the drop. We can thus ask an inverse question: *Is it possible to make shrink this peripheral disclination back to the monopole?* Knowing that R_c is proportional to the drop's thickness, it should be possible to increase e so much that the critical radius R_c given by Eq. (8) would become greater than the radius of the drop R. Paradoxically, this experiment was made by accident and we were surprised by its result: for e larger than $\approx R/2$, the peripheral disclination shrank into a monopole and the distortionless homeotropic texture was replaced by the dowser texture.

Let us stress that the dowser texture was well known previously under the name of *quasi-planar texture*, which was unduly believed to be replaced always by the homeotropic one.

Knowing that, in spite of its metastability, the dowser texture can be preserved indefinitely provided that the thickness e is not reduced too much, we were able to work with it and to unveil its remarkable properties. As we will see below, several of them are related to the de Gennes' articles published in the *Comptes rendus*. Before that, let us mention briefly another recent experiment with dislocation loops.

3.4. Captive disclination loops

All surfaces Σ of the nematic droplet kept by capillarity between the lens and the glass slide (see Fig. 5c) impose the homeotropic anchoring conditions on the director field: $\mathbf{n}$ orthogonal to Σ. For topological reasons, the nematic droplet must thus contain either a monopole or a charged disclination loop equivalent to the monopole. We have seen above that when the thickness e is large enough, the disclination loop collapses into the monopole. However, when the droplet is pierced by a polymeric fibre, the disclination loop can be threaded on it. In this case, the collapse process is hindered by the elastic repulsion of the disclination by the fibre surface. In equilibrium, the disclination levitates at some height h above the fibre's surface (see Fig. 6).

Systèmes (d = 3) :	Superfluides	Smectiques A	Ferro (Heisenberg)	Nématiques	Texture «sourcier»
Go	(Jauge)	Translations (— axe optique)	Rot. 3 dim.	Rot. 3 dim.	Rot. 2 dim.
Param. d'ordre :	F. d'ondes	Mod. densité	Aimentation	Anisotropie tensorielle	Champ «sourcier»
{ S }	$\psi = \psi' + i\psi''$	$\psi = \psi' + i\psi''$	M_α	$Q_{\alpha\beta}$	$e^{i\varphi}$
n	2	2	3	5	1
δ	1 (vortex)	1 (dislocations)	1 ou 0	1 ou 0 ⟷	0

Fig. 7. Types of singularities allowed in ordered phases. d is the dimension of space in which the system exists, n is the number of components of the order parameter, and δ is the dimension of the permitted singularities which is 0 for points (monopoles) and 1 for lines (dislocations, disclinations or vortices). (Adapted from Ref. [4]).

It has been pointed out in Ref. [18] that such levitating disclination loops act as probes of the fibre surface. Experiments with tiny nematic droplets pierced by cellulosic or spider silk fibres have shown that the disclination loop is tilted with respect to the fibre surface when the fibre is chiral (see Figs. 6a and b).

The same effect, called chirogyral, has been detected using mechanically twisted nylon fibres (see Figs. 6c and d) [19]. As expected from theoretical considerations, the tilt angle η is proportional to the twist angle α.

4. Topological defects in systems with an order parameter

In 1991, the Nobel Prize in Physics was awarded to Pierre-Gilles de Gennes "*for discovering that methods developed for studying order phenomena in simple systems can be generalized to more complex forms of matter, in particular to liquid crystals and polymers*".

Among others, this sentence refers to the work of de Gennes on analogies between systems equipped with order parameters resulting from broken symmetries. The article "Types de singularités permises dans une phase ordonnée" published in the *Comptes rendus* in 1972 [4] contains the very first classification of topological defects in systems with an order parameter. In spite of its importance, this paper remains paradoxically almost unknown (10 citations listed by Google Scholar). Using the group theory, de Gennes arrives at results represented in Ref. [4] as a table reproduced here in Fig. 7 in which d is the dimension of space in which the system exists, while n is the number of components of the order parameter. For given d and n, de Gennes points out that the dimension δ of the permitted singularities has to satisfy the following inequalities:

$$d - n \leq \delta \leq d - 2 \quad \text{and} \quad 0 \leq \delta \tag{9}$$

4.1. Uniaxial nematics in three dimensions

In section 3, we dealt with the three-dimensional nematic uniaxial phase ($d = 3$), in which the quadrupolar order parameter S_{ij} (or Q_{ij} using the notations of Ref. [4]) has only $n = 4$ components instead of $n = 5$. Using the de Gennes rules (Eq. (9)), one obtains $\delta = 0$ or 1 as in the case $n = 5$. The corresponding point and the linear singularities correspond to disclinations and monopoles discussed above (see Figs. 5c and d).

4.2. Analogy between superconductors and smectics A

The de Gennes' table lists also two systems with the complex order parameter $\Psi e^{i\varphi}$: superfluids (or superconductors) and smectics A. Its meaning in the case of the layered smectic A phase is represented graphically in Fig. 8a as a sinusoidal modulation of the mass density in the z direction. The phase φ has thus a simple physical meaning: its change $\delta\varphi$ corresponds to the translation δz of layers. Knowing this, the wedge (Fig. 8b) and screw dislocations (Fig. 8c) appear as 2π defects of the phase φ on trajectories surrounding them. Dislocations in smectics A are therefore similar to vortices in superfluids or superconductors, which are also characterised by a 2π defects of the phase φ of the wave function.

De Gennes knew that in superconductors of type II submitted to a large-enough magnetic field **B**, a system of vortices is generated, so he asked: "*how can one generate a similar system of dislocations in smectics A?*" The answer he found seems to be simple: "*this should occur in smectics A made of chiral molecules*", but it is much more difficult and space-consuming to explain it. To understand why in smectics the chirality plays the role similar to the magnetic field, we will recommend to the reader the reading of de Gennes' paper "An analogy between superconductors and smectics A" [15]. This remarkable conjecture of de Gennes, enounced in 1972, was proved to be true in 1989 by Goodby et al. [20] on the example of the chiral smectic phase known today as the Twist Grain Boundary Phase (TGB). In Fig. 8f, we show that in the TGB phase screw dislocations are assembled in walls for reasons that were understood by Lubensky and Renn [21]. Thanks to them, the tendency of chiral molecules to adopt a twisted texture is satisfied.

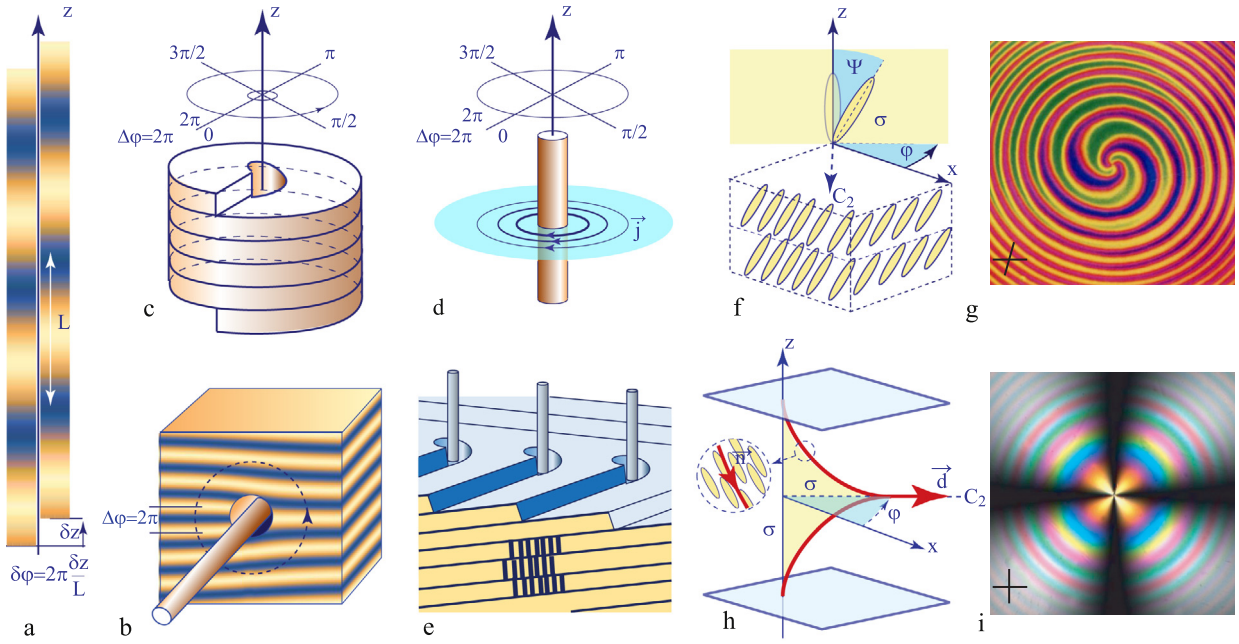

Fig. 8. Systems with the complex order parameter $\Psi e^{i\varphi}$. a) In smectics A, the variation $\delta\varphi$ of the phase correspond to the translation δz of smectic layers. b) Wedge dislocation in smectics A. On the trajectory surrounding the dislocation, the phase varies by 2π [9]. c) Screw dislocation in smectics A. d) Vortex in a superconductor. De Gennes stressed its topological analogy with dislocations in smectics A [15]. e) Structure of the Twist Grain Boundary (TGB) phase predicted by de Gennes on the basis of the superconductor–SmA analogy[15]. f) Smectic A–smectic C transition [6]. The order parameter resulting from the tilt of molecules is $\Psi e^{i\varphi}$. g) View under a polarising microscope of a free-standing smectic C film. It contains in its centre a $\Delta\varphi = +2\pi$ singularity. h) The order parameter of the dowser texture considered as a two-dimensional system is a 2D unitary vector field **d** equivalent to $e^{i\varphi}$. i) A $\Delta\varphi = +2\pi$ singularity of the dowser field observed between crossed polarisers.

4.3. Smectic A smectic C phase transition

In 1972 de Gennes tackled also the issue of the smectic A → smectic C transition represented schematically in Fig. 8f [6]. It breaks the symmetry of revolution around the z axis normal to smectic layers and results in appearance of another complex order parameter $\Psi_t e^{i\varphi_t}$. Its amplitude Ψ_t and phase φ_t correspond respectively to the tilt and azimuthal angles. Thanks to this second-order parameter, the smectic C phase beside dislocations can also have disclinations – lines around which the phase φ_t has a 2π defect. They are easy to observe in free-standing smectic C films by means of a polarising microscope with slightly decrossed polarisers (see Fig. 8g).

4.4. Polarisation of the chiral smectic C phase

In terms of the group theory, the smectic A → smectic C transition lowers the symmetry from $D_{\infty h}$ to C_{2h}. Only the mirror plane σ (the plane of the tilt) and the C_2 axis (around which the tilt occurred) are left. A remarkable phenomenon predicted by R.B. Meyer occurs when molecules are chiral so that the mirror symmetry is broken. The remaining symmetry of the system C_2 is so low that it allows the existence of a spontaneous polarisation **P** parallel to the C_2 axis. Following suggestions of Meyer, the team of chemists (L. Strzelecki, L. Liebert, and P. Keller) attracted to our lab by de Gennes, synthesised a chiral material having the smectic C* phase [22]. As expected by Meyer, this phase was polarised.

5. Remarkable properties of the dowser texture

5.1. Order parameter $e^{i\varphi}$

The last scheme in Fig. 8h represents a more detailed view of the dowser texture introduced previously in Fig. 5. Its symmetry C_{2v} can be seen as obtained by the breaking of the symmetry $D_{\infty h}$ of a nematic with a homogeneous orientation along the z axis. The resulting order parameter is simply $e^{i\varphi}$, with the phase φ representing the azimuthal orientation of "the dowser tool". Alternatively, it can be also represented by a 2D unitary vector field $\mathbf{d} = (\cos\varphi, \sin\varphi)$.

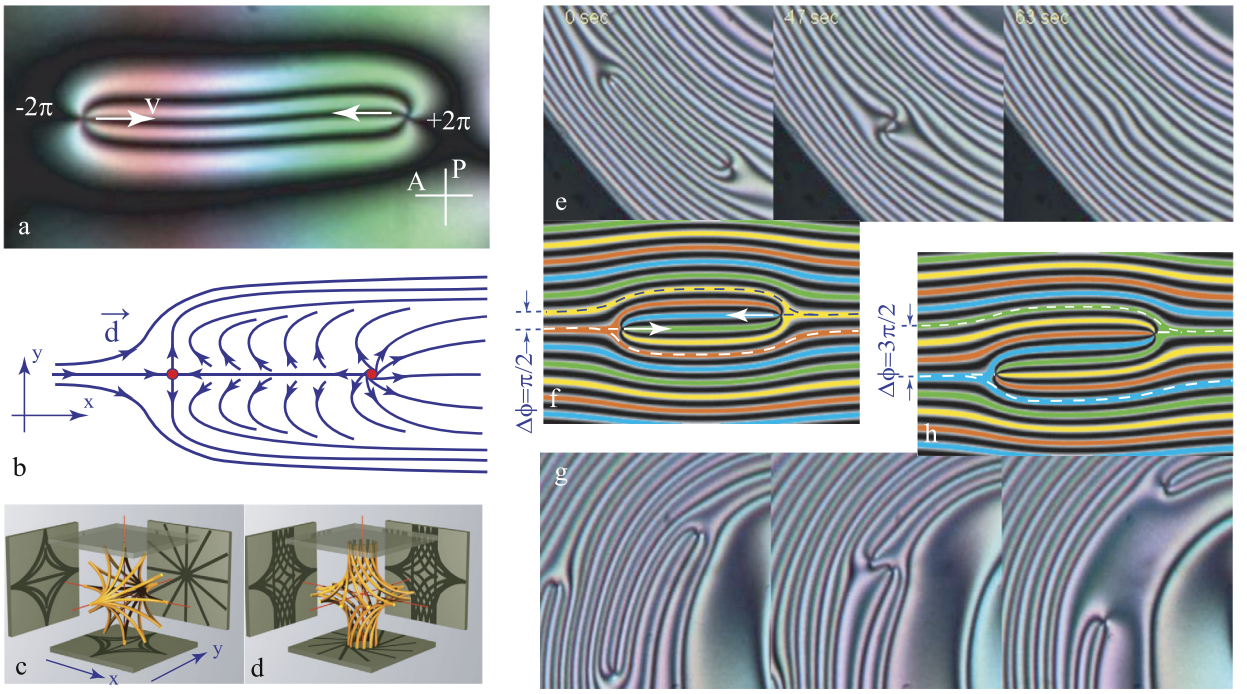

Fig. 9. Nematic monopoles as singularities of the dowser texture. a) A view under the polarising microscope of a pair of the $+2\pi$ and -2π singularities of the dowser texture. b) The dowser field **d** inferred from the picture a. c–d) Three-dimensional structure $\mathbf{n}(\mathbf{r})$ of nematic monopoles corresponding to singularities a and b. e) Collision of a pair of $+2\pi$ and -2π defects resulting in their annihilation. f) The phase field $\varphi(x, y)$ of the pair shown in e. The colour code used in this picture is: red – φ mod $2\pi = 0$, yellow – φ mod $2\pi = \pi/2$, green – φ mod $2\pi = \pi$ and blue – φ mod $2\pi = 3\pi/2$. The phase difference between the two defects is $\Delta\varphi = \pi/2$. g) Collision of a pair of $+2\pi$ and -2π defects without annihilation. h) The phase field $\varphi(x, y)$ of the pair shown in g. Annihilation is avoided because the phase difference $\Delta\varphi = 3\pi/2$ is larger than π.

5.2. Monopoles as singularities of the dowser field

In terms of the de Gennes' classification, we have thus $n = 1$ (see the last column added to the de Gennes' table in Fig. 7). As the dowser field **d** can evolve only in the xy plane of the nematic layer, this system can be considered as two-dimensional with $d = 2$. Using the de Gennes rules (Eq. (9)), one arrives at the conclusion that the dowser field can have only $+2\pi$ and -2π point singularities. In Fig. 9a, we show a pair of these singularities seen in polarised light. The corresponding dowser field $\mathbf{d}(x, y)$ is drawn in Fig. 9b. However, when the dowser texture is considered in $d = 3$ dimensions of the nematic layer (see Fig. 9c), the $+2\pi$ and -2π point singularities of the dowser field from Fig. 9b appear as monopoles – the point singularities of the director field **n** or of the equivalent quadrupolar order parameter S_{ij} (or Q_{ij}) [23].

It is worthwhile to mention that the concept of monopoles is much older than the de Gennes' paper. Nabarro quotes, in Ref. [24], published a few months after the de Gennes' work, the work of Poincaré on point singularities in solutions to differential equations [25] as well as the paper of Feldtkeller [26] on singular points in ferromagnetics. (In his table, de Gennes indicates that monopoles are permitted also in Heisenberg ferromagnetics.)

In nematics, monopoles predicted by de Gennes and Nabarro were observed first in capillaries with homeotropic boundary conditions [27] and later in the vicinity of inclusions [28]. In the light of a recent work [29,30], the dowser texture appears as a natural universe of nematic monopoles for the following reasons:

1) it is possible to generate them in a controlled manner as pairs of the $+2\pi$ and -2π defects of the dowser field,
2) it is possible to make them move on isophasic trajectories orthogonal to gradients of the phase **grad** φ,
3) it is possible to observe their annihilation when these motions lead to encounters of $+2\pi$ and $-2\ \pi$ pairs.

Figs. 9e–h show that the result of such collision depends on the distance of trajectories in terms of the phase difference $\Delta\varphi$. Annihilation occurs when $\Delta\varphi < \pi$ (see Figs. 9e and f), while for $\Delta\varphi > \pi$ the annihilation is avoided (see Figs. 9g and h).

5.3. Polarisation of the dowser texture

The conjecture of R.B. Meyer on polarisation of the chiral smectic C phase was formulated during his postdoctoral visit in our lab. Discussions with de Gennes about the nature and consequences of the smectic A $\rightarrow$ smectic C transition were certainly useful for him in this matter; nevertheless, the sources of this brilliant idea are older. Indeed, already in 1969, R.B.

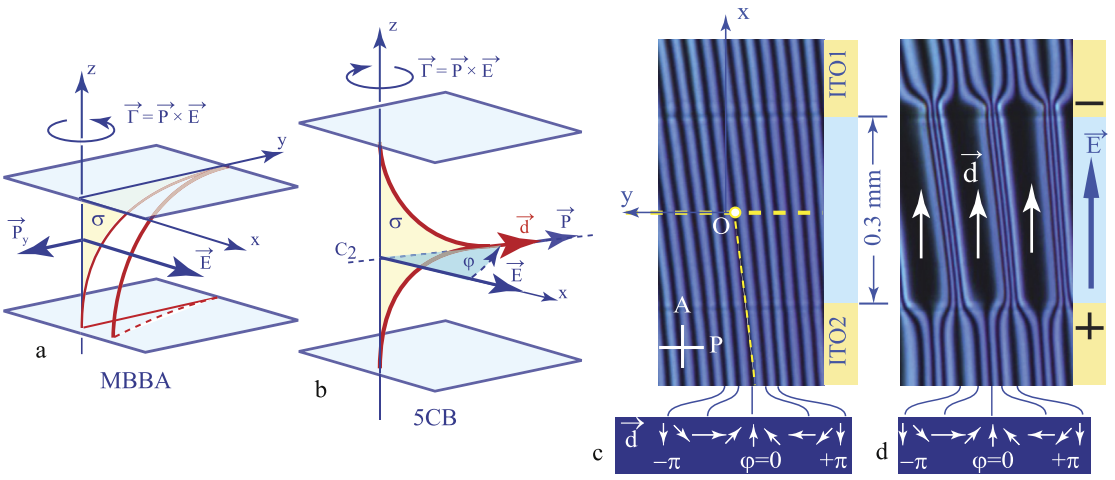

Fig. 10. Flexo-electric polarisation of the dowser texture. a) Geometry of the generic experiment of Dozov et al. [34]. b) The symmetry of the dowser texture C_{2v} allows the existence of the flexo-electric polarisation **P**, which must be collinear with the two-fold symmetry axis C_2. Its orientation – parallel or antiparallel to the dowser field **d** – depends on the material. c–d) The experimental evidence for the flexo-electric polarisation in CB5 (nematic material). The experiment is made in two stages. c) The dowser is first wound up by elliptical flows: $\varphi = qy$. c) The wound-up dowser texture is then submitted to an electric field **E** parallel to x. As a result, black interference fringes with **d** parallel to **E** are enlarged. This means that in 5CB **P** is parallel to **d** as shown in b. The result of the same experiment made with MBBA is inverse: **P** is antiparallel to **d**, in agreement with the first experiments by Dozov et al. [34].

Meyer published his paper on "Piezoelectric effects in liquid crystals" [31], in which he predicted that distortions of the director field **n** in nematics can generate an electric polarisation.

The great merit of de Gennes was to express this idea of Meyer in a more concise and accessible form in his book [32] and by this means to make it well known. The first experiment proving the existence of flexo-electricity was performed in 1971 by Schmidt, Schadt, and Helfrich [33]. Later, in 1982, I. Dozov, P. Martinot-Lagarde, and G. Durand [34] made a different experiment using the geometry depicted in Fig. 10a. Here, the distortion of the director field is due to different boundary conditions on limit surfaces: **n**//**z** at the bottom and **n**//**y** on the top. The director field is thus symmetrical with respect to reflection in the mirror plane $\sigma(yz)$ and the flexo-electric polarisation **P** has to be also parallel to it. The electric field applied in the x direction produced a torque which deformed the director field as shown.

The Meyer's idea has been tested recently using the dowser texture represented schematically once again in Fig. 10b [35]. As already mentioned above, the dowser texture is symmetrical not only with respect to reflections in the mirror plane σ containing the director field, but also with respect to rotations around the two-fold axis C_2 parallel to **d**. The corresponding symmetry group C_{2v} allows the existence of a polarisation **P** that must be collinear with the two-fold axis C_2. As we will see below, the direction of **P**, parallel or antiparallel to **d**, depends on the nematic material.

Using the notations introduced by de Gennes [32], the polarisation per unit area of the nematic layer of can be expressed as

$$\mathbf{P}_{\mathrm{fe}} = \int_0^e [e_1 \mathbf{n}\vec{\nabla}\cdot\mathbf{n} + e_3(\vec{\nabla}\times\mathbf{n})\times\mathbf{n}]\,\mathrm{d}z \tag{10}$$

In the approximation where the director field of the dowser texture is represented as

$$\mathbf{n} = [\sin(\pi z/e)\cos(\varphi), \sin(\pi z/e)\sin(\varphi), \cos(\pi z/e)] \tag{11}$$

one obtains:

$$\mathbf{P}_{\mathrm{fe}} = \mathrm{P}_{\mathrm{fe}}\mathbf{d} \quad \text{with } \mathrm{P}_{\mathrm{fe}} = \frac{\pi}{2}(e_3 - e_1) \text{and } \mathbf{d} = [\cos(\varphi), \sin(\varphi)] \tag{12}$$

The polarisation of the dowser texture should obviously be detected from its response to an electric field applied in the plane xy of the nematic layer. This response can be easily detected when the field is applied to a wound up dowser texture, for in such a case all possible orientations of the dowser field with respect to **E** are tested simultaneously. For this purpose, the dowser is first wound up by elliptical flows as shown in Fig. 10c. Subsequently, it is submitted to an electric field **E** that deforms it as shown in Fig. 10d. As black isochromes with **d** parallel to **E** are enlarged (favoured), we can conclude that in 5CB **P** is parallel to **d** as shown in Fig. 10b. The result of the same experiment made with MBBA was inverse: **P** was found to be antiparallel to **d** in agreement with the experiment of Dozov et al. [34] (see Fig. 10a).

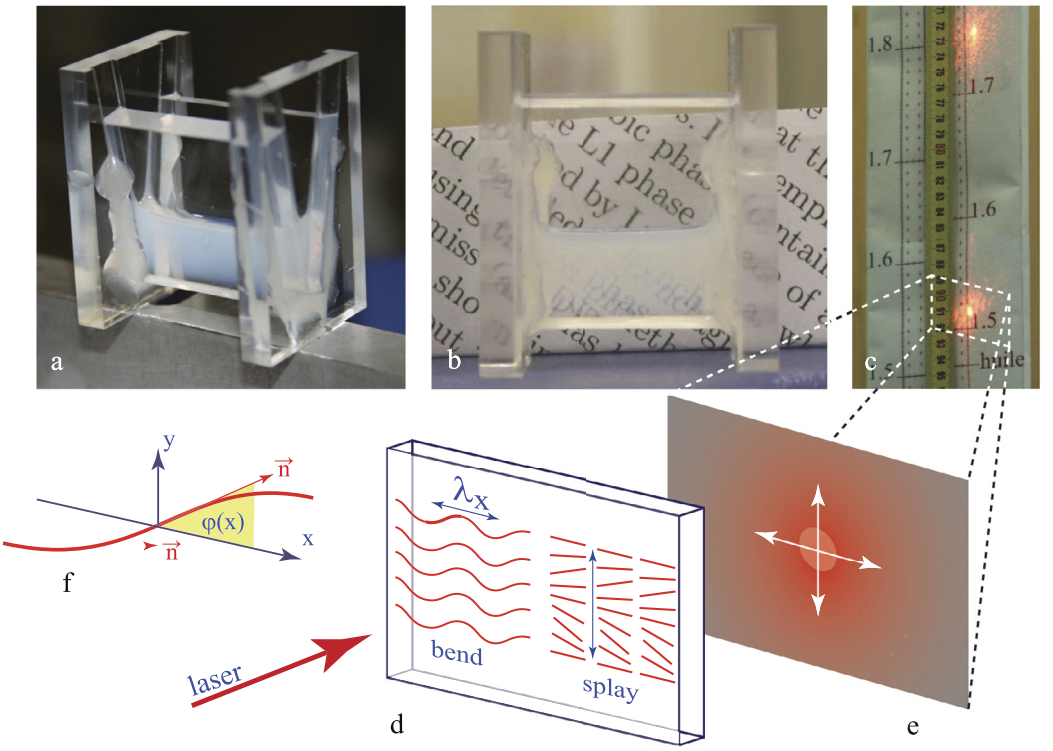

Fig. 11. Scattering of light by nematics. a) In spite of its homogeneous orientation inside the prismatic vessel the nematic 5CB has a milky aspect due the light scattering by thermal fluctuation of the director field. b) Classical evidence of the birefringence of the nematic – double image of the text seen through the nematic prism. c) Measure of the refractive indices of 5CB by means of the nematic prism: $n_e = 1.52$ and $n_o = 1.75$. d) Relationship between the fluctuations of the director field and the scattered light. (This elegant class-room experiment with the nematic prism stems from by H.-S. Kitzerow.)

6. Scattering of light by nematics

When one examines for the first time a sample of a nematic contained in a transparent vessel, such as the glass test tube shown in Fig. 1b or the prism made of Plexiglas shown in Fig. 11, its milky aspect suggests that it could be a biphasic emulsion in which the light is scattered on inhomogeneities of the refractive index. This belief was discarded shortly after the discovery of liquid crystals and was replaced by other hypotheses, which unfortunately were not more successful.

In the first sentence of his article "Fluctuations d'orientation et diffusion Rayleigh dans un cristal liquide nématique" [2], de Gennes discards the so-called "swarm" model, invoking the existence of a fluctuating short-range order of molecular orientations. With his knowledge of fluctuations in systems with order parameters (magnets, superfluids, superconductors, or crystals) de Gennes understood that in nematics the light scattering is due to thermal fluctuations of the long-range uniaxial quadrupolar order parameter $S_{ij} = S(n_i n_j - \delta_{ij}/3)$ (or, using the notations of Ref. [2], $Q_{ij} = Q(n_i n_j - \delta_{ij}/3)$) or, more precisely, to fluctuations of its dielectric permittivity tensor ϵ_{ij}, proportional to S_{ij}.

To understand why fluctuations of the director are coupled with light, a very simple experiment with a nematic prism depicted in Fig. 11 is sufficient. First of all, it provides a clear evidence of a very strong birefringence of nematics. From the double refraction of a laser beam by the nematic prism, one determines the ordinary and extraordinary indices, which in the case of 5CB are $n_o = 1.52$ and $n_e = 1.75$. The dielectric permittivity tensor, expressed by de Gennes as

$$\epsilon_{ij} = \bar{\epsilon}\delta_{ij} + \epsilon_a\left(n_i n_j - \frac{\delta_{ij}}{3}\right) \tag{13}$$

has therefore a very strong anisotropy $\epsilon_a = \epsilon_{//} - \epsilon_\perp \approx n_e^2 - n_o^2$.

Next, this experiment shows that (1) each of the two refracted beams is surrounded by a halo of scattered light-fluctuating speckles, (2) the time-averaged intensity I($\mathbf{q}$) of the scattered light decreases with the angular distance from direct beams, that is to say with the length q of the scattering vector $\mathbf{q}$ (see Figs. 11c and e). In Fig. 11d, we show two modes of fluctuation of the director $\mathbf{n}(\mathbf{r})$ around its mean orientation $\mathbf{n}_o//\mathbf{x}$. In the mode labelled "bend", the director $\mathbf{n}$ stays in the plane xy and its angle with $\mathbf{x}$ varies as $\varphi(x) = \varphi_o \cos(q_x x)$.

In the second sentence of his paper de Gennes, proposes to calculate the amplitudes of these fluctuations in the frame of the continuous elastic model. The cornerstone of this model is the statement that deviations from the nematic ground state,

i.e. from a uniform orientation of molecules $\mathbf{n} = \mathbf{const}$, result in an increase in free energy. In the de Gennes' expression of the free energy density (Eq. (1)) introduced in section 2.2, this elastic energy is given by the two last gradient terms. Let us note however that, as the Landau–de Gennes theory was posterior to the paper on Rayleigh scattering, de Gennes used another classical expression of the elastic energy density proposed first by C.W. Oseen [36] and perfected later by F.C. Frank [37]:

$$F_{\mathrm{el}} = \frac{1}{2}K_{11}(\vec{\nabla}\cdot\vec{n})^2 + \frac{1}{2}K_{22}[\vec{n}\cdot(\vec{\nabla}\times\vec{n})]^2 + \frac{1}{2}K_{33}[\vec{n}\times(\vec{\nabla}\times\vec{n})]^2 \tag{14}$$

For the bend mode, expressed as

$$\vec{n} = \{\cos[\varphi_{\mathrm{o}}\cos(q_x x)], \sin[\varphi_{\mathrm{o}}\cos(q_x x)], 0\} \tag{15}$$

one obtains $\mathrm{F_{el}} = (\mathrm{K_{33}}/2)\varphi_{\mathrm{o}}^2 q_x^2$ and using the equipartition rule applied to a volume Ω of the nematic, one gets

$$\langle\varphi_{\mathrm{o}}^2\rangle = \Omega\frac{kT}{Kq_x^2} \tag{16}$$

Subsequently, de Gennes finds that the intensity of the scattered light is proportional to $\langle\varphi_{\mathrm{o}}^2\rangle$ and, finally, explains the observed $I(q) \sim 1/q^2$ dependence.

It is important to emphasize that prior to the publication of this paper in the *Comptes rendus*, intense experimental and theoretical studies of the light scattering by nematics were under way under auspices of de Gennes in the team of George Durand. The results of this work were published in articles under the signature "Orsay Liquid Crystal Group". Later on, the turbidity of nematics due to the scattering on fluctuations of the quadrupolar order parameter was studied by D. Langevin and M.-A. Bouchiat [38], who found that it depends strongly on the orientation of **n** with respect to the wave vector **q** and to the polarisation **i** of the incident beam. In particular, the turbidity of the nematic is strong when the **n** is orthogonal to **q**, which actually is the case in the experiment with the nematic prism (see Fig. 11).

7. Epilog

It was a great honour for me to have been selected by Marie-Anne Bouchiat and Jacques Villain as one of the contributors to the historical issue of the *Comptes rendus Physique* on the most remarkable authors who have published in this glorious journal. It was also a pleasure for me to deal with articles of Pierre-Gilles de Gennes to whom, like so many other PhD students working in the 1970s under the auspices of de Gennes, I am deeply indebted for his generous support. I had the chance to meet him thanks to Etienne Gyuon, my PhD adviser and thanks to Monsieur Jacques Friedel who showed me the way leading to the world of the scientific research.

In this article, the name of R.B. Meyer is quoted several times, because de Gennes had a deep esteem for Meyer's scientific skills and quoted him very often. R.B. Meyer, like Sir C. Frank, W. Helfrich, F.R.N. Nabarro, N.A. Clark, P. Cladis, P. Pershan, and so many other foreign scientists came to our lab for visits attracted by the auras of J. Friedel, of P.-G. de Gennes, and of the renowned Orsay Liquid Crystal Group. All young researchers benefited greatly from interactions with them. Working under the auspices of Friedel and de Gennes had also magic effects during travels and conferences, because we were always welcomed everywhere. As an example, I could quote the warm hospitality of Alfred Saupe in Kent (USA), who knew de Gennes and had a great esteem for him since his beginning in the field of liquid crystals. For the same reason, the doors of The Geilo School in Norway, founded by T. Rieste and Arne Skjeltorp, were widely open to the young coworkers of de Gennes.

The scientific legacy of de Gennes counts about 500 articles, that is to say, two orders of magnitude greater than the collection of a few articles discussed here. This legacy is discussed in its whole extension in the Memorial Book *P.G. de Gennes' Impact on Science* [39,40].

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