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Surface singularities in nematics and some notes on cholesterics (*)

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Résumé. — On présente une analyse topologique des configurations de surface et de volume dans les cholestériques et les nématiques. On rappelle et illustre la procédure de Volovik pour combiner défauts de surface et de volume ; le problème de la torsion dans les cholestériques est discuté suivant le point de vue de Cladis *et al.* Les considérations sont appliquées aux boojums cholestériques. On étudie également la question des textures non singulières, et on propose une texture stable de type soliton pour un cholestérique. Enfin on considère le problème d'un nématique dans une sphère, et on présente les différentes solutions.

Abstract. — A topological analysis of surface and bulk configurations in cholesterics and nematics is given. Volovik's procedure for combining surface and bulk defects is reviewed and illustrated, and the problem of twist in cholesterics is discussed from the point of view of Cladis *et al.* These considerations are applied to cholesteric boojums. Also, the question of non-singular textures is studied and a soliton-like texture for a cholesteric is proposed and is predicted to be stable. Next, the problem of a nematic in a sphere is considered and the various solutions to this problem are given.

1. **Introduction.** — Recently we presented an analysis of the existence and classification of topological defects in cholesteric liquid crystals, particularly those induced by the topology of the boundary (*boojums*) [1]. Here, we wish to present some clarifying remarks about cholesterics, and extend our considerations to the case of nematic liquid crystals. Our discussion is organized as follows. In Section 2, following Volovik [2] we examine the topological invariant which is important for a determination of the bulk structure which can accompany a boundary singularity. It will be seen that in most cases of interest, one's naive intuition is perfectly satisfactory. In considering which structures are actually seen experimentally in cholesterics, the importance of energetics, and in particular the non-local constraints which twist imposes, are emphasized. The twist is seen to affect the structure of boojums very strongly over distances of the order of the pitch. In Section 3, we discuss the importance of non-singular mappings (the Hopf map) on cholesterics. Besides their effects upon singular configurations, as has been discussed by Bouligand *et al.* [3], we note that completely non-singular solitons, with finite energy, are also expected to arise. Characterized by a size the order of the pitch, they can be very loosely described as a nematic soliton embedded in a cholesteric. In Section 4, we discuss the problem of a nematic liquid crystal in a sphere with tangential orientation at the boundary. After delineating all distinct configurations, we conclude that energetically the most favored form is a configuration free of bulk singularities, with the two units of vorticity on the surface separate.

2. **Non-local constraints in cholesterics.** — Consider first the case of cholesterics. The traditional picture of the order parameter in the medium is $G = \mathrm{SU}(2)/Q$ [4] (but see below), where Q is the discrete group of quaternions, and is $H = S^1$ on the boundary if the container is prepared properly. As there are no point defects in the bulk, the possible defects of the boundary are only points, characterized by the homotopy group $\Pi_1(H) = Z$. It is of interest to ask, however, what is the relevant topological quantity if there are point defects in the bulk, so that $\Pi_2(G)$ is non-trivial as well as $\Pi_1(H)$? For simple cases, we would expect that it is just the direct product, but in the most general case the question of how the defects on the boundary will interact with those in the bulk must be taken into account.

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The answer has been given by Volovik [2], who has noted that to include this, one does not characterize the defect by a circle Γ on the boundary of the material surrounding the point (see Fig. 1), but by a section of a spherical surface, whose boundary is the circle Γ, S_Γ. The topological invariant relevant is then $\Pi_2(G, H)$. Use of the exact sequence will demonstrate that if either $\Pi_1(H)$ or $\Pi_2(G)$ is isomorphic to the trivial set, then $\Pi_2(G, H)$ is just $\Pi_2(G)$ or $\Pi_1(H)$, respectively, so the conclusions of ref. [1] concerning cholesterics are unchanged.

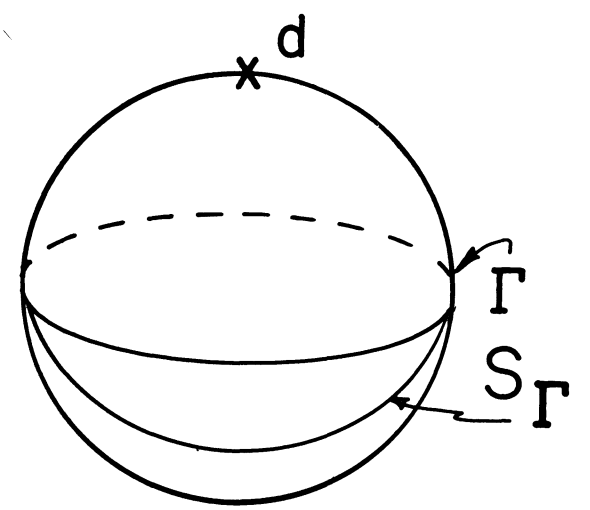

Fig. 1. — A point defect, *d*, surrounded by a line contain, Γ, and a hemisphere S.

The case of a spherical container is a good case in point. There must be two units of vorticity on the surface [5]. We argued that the lowest energy configuration would be an $m = 2$ vortex on the surface with a non-singular bulk texture, which is similar to the boojum known in He^3 (for all radii of the sphere greater than the average intermolecular separation, which is assumed for all cases in this paper). It must be remembered, however, that besides the local order parameter there is a non-local condition of periodicity in the twist which must be imposed.

The importance of including the constraint imposed by the twist has recently been emphasized by Cladis, White, and Brinkman [6], and by Mineyev [7]. Following experimental observations, Cladis *et al.* point out the existence of an $S = +2$ line defect which apparently does not relax to a defect-free state. The specific configuration is of two λ lines entwined in a helical manner. The defect relaxes initially so that there is no singularity in the director, but is stabilized at a point where the relative separation of the λ lines is on the order of the pitch, and hence is singular in the twist axis on a scale of the order of the pitch. The authors note that the stability of the configuration can at least be understood as a result of its being a local minimum of the free energy.

With these considerations in mind, we can sharpen our understanding of boojums in cholesterics. Suppose that in a sphere we begin with the Frank-Pryce texture, so that there is a singularity in both the director and the twist axis. As asserted previously, the singularity in the director will relax, but only over distances of the order of the pitch, remaining singular in the twist axis. Thus the boojums in cholesterics are distinct from both the Frank-Pryce texture and the boojums in He^3-A, where no singularities exist along any axis. This accords much better with the observations of Robinson, Ward and Beevers [8], who found that a radial singularity seemed to be present in droplets where the pitch was much less than the spherulite radius. This *radial singularity* is actually non-singular in the director axis at length scales as small as an intermolecular separation, but exhibits a singularity in the twist axis at length scales of the order of the pitch. This may be stated more simply : a 4π line defect in a cholesteric can only relax to distances of the order of the pitch, rather than over the entire volume of the cholesteric. From the nematic point of view, no singularity exists, but from the cholesteric viewpoint one does. This illustrates the importance of specifying which length scale one is considering when talking of defects. The topological considerations of ref. [1] that were confined to a cholesteric on a surface remain unchanged.

3. **Cholesteric solitons.** — We would now like to discuss the existence of topologically stable textures in cholesterics. The free energy of a cholesteric is given by

$$E = K \int [(\nabla . \mathbf{n})^2 + (\mathbf{n} . \nabla \times \mathbf{n} \pm q)^2 + (\mathbf{n} \times \nabla \times \mathbf{n})^2] \, d^3 r$$

where we define the pitch $q^{-1} > 0$. Consider now as our order parameter only the director field (S^2) — the neglect of the twist component of the order parameter in topological analysis will be justified shortly.

Roughly speaking, the topological invariant which we shall use characterizes maps of all space ($\mathbb{R}^3$ compactified to S^3) onto the director : such maps are classified by the Hopf index $\pi_3(S^2) = Z$ [9]. This is not a precise statement since the ground state of a cholesteric is not homogeneous, so all points at infinity cannot be identified as a single point, which is the usual means of compactification. This does not prevent us from rigorously defining the Hopf index as for a system with a uniform ground state. Consider a closed curve Γ along which $\mathbf{n}$ is constant, and some surface S_Γ with Γ as its boundary. Then the Hopf index is given by [10]

$$Q = \int_{S_\Gamma} h^i \, dS_i ,$$

where

$$h^{\mathrm{i}} = \frac{1}{8\,\pi}\,\varepsilon^{\mathrm{ijk}}\,\varepsilon_{\mathrm{abc}}\,n^{\mathrm{a}}\,\partial_{\mathrm{j}}n^{\mathrm{b}}\,\partial_{\mathrm{k}}n^{\mathrm{c}}\,,$$

and $\mathrm{d}S_{\mathrm{i}}$ is a surface element of S_Γ. The curve Γ is arbitrary; in particular, if at infinity

$$n = (\cos qz,\ \pm\ \sin qz, 0)\,,$$

we can take any curve at infinity parallel to the x-y plane.

The importance of the Hopf index in cholesterics was first noted in a beautiful example due to Bouligand *et al.* [3]. Consider two interlocked ring defects. If each defect belongs to a non-commuting element of the group of quaternions, a theorem of Poenaru and Toulouse [11] prevents their skew crossing, and so the rings are stable. Bouligand *et al.* noted that stability can also arise not because of the singular but because of their non-singular structure. For example, two interlocked J defects cannot separate as their Hopf index, defined above, is one. This is termes as evidence of *double* topological character.

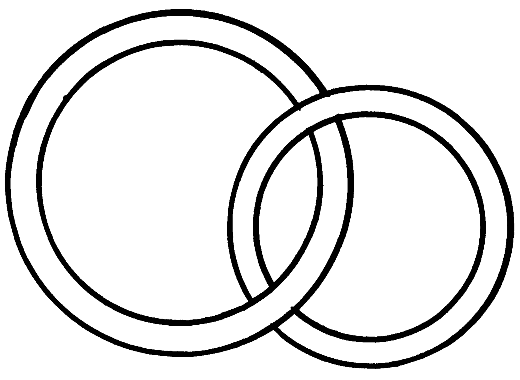

Fig. 2. — Two interlocked ring defects in cholesterics.

We wish to suggest that another, more direct manifestation of double topology is the existence of non-singular textures in cholesterics. By well-known scaling arguments [9], we can immediately rule out such textures if $q = 0$: by taking $x \to \lambda x$ (where λ is a dimensionless parameter), $E \to \lambda E$. But then any configuration with finite energy could not be stable, as by letting $\lambda \to 0$ we take $E \to 0$.

Such scaling arguments do not apply to cholesterics since in order to have a configuration with finite energy, there is also a boundary condition at infinity; viz, $\mathbf{n}.\nabla \times \mathbf{n} = \mp\, q$. Scaling $x \to \lambda x$ violates this condition, so that for any $\lambda \neq 1$, the energy will be infinite. Note that $q \to q/\lambda$ is not permissible, since q is not dependent on a given configuration but is an external parameter characteristic of the medium itself (although of course it will depend on other external parameters, such as the temperature).

Consider now redefining $\tilde{x} = qx$, where $\tilde{x}$ is a dimensionless parameter. Then

$$E = \frac{K}{q}\,\tilde{E} = \frac{K}{q}\int \mathrm{d}^3\tilde{x}((\tilde{\nabla}.\mathbf{n})^2 + $$
$$+ (\mathbf{n}.\tilde{\nabla} \times \mathbf{n} \pm 1)^2 + (\mathbf{n} \times \tilde{\nabla} \times \mathbf{n})^2)\,,$$
$$Q = \tilde{Q}\,.$$

We propose that there exists a configuration of $\vec{n}(\tilde{x})$ which : 1) minimizes $\tilde{E}$, 2) has finite $\tilde{E}$, 3) has $\tilde{Q} \neq 0$. In general we expect that for a given $\tilde{Q}$, such $\vec{n}(\tilde{x})$ should exist for either sign of the medium's chirality. The basis for this proposal rests on the fact that a cholesteric, unlike a nematic, has associated with it an intrinsic length scale q^{-1}, and one therefore naturally expects any solution with nonzero Q to assume a size of the order of the pitch. Thus, the existence of the pitch stabilizes the soliton.

Unfortunately, although by scaling we cannot disprove the existence of these configurations, neither can we prove their existence. Needless to say, the equations for any solution which minimize E are involved; incorporating $\tilde{Q} \neq 0$ appears to make any attempt to find $\vec{n}(\tilde{x})$ analytically hopeless.

Nevertheless, it is interesting to inquire what the properties of these textures would be. As $\tilde{x}$ is dimensionless, $\tilde{E}$, and any reasonable parameter which measures the size of the configuration, are $\sim 0(1)$. Hence for the texture in real space, its size $r = c_1/q$, and its energy $E = c_2\,K/q$, with c_1 and c_2 pure numbers given by the exact $\vec{n}(\tilde{x})$. We now see why the neglect of the twist is justified, as within the texture the director varies too rapidly to define the twist. In essence, the presence of twist stabilizes the soliton at a size of the order of the pitch. (This is analogous to solitons in ^{3}He-A, where continuous superflow is necessary to stabilize solitons [11].)

Such non-singular textures will in general be difficult to observe. As they have only finite energy, thermal fluctuations in practice will render them metastable. Consider the following situation : prepare a defect-free region where q is large. With E small, solitons should be easy to produce. Consider now decreasing q — the size and the energy of any soliton will increase inversely. The rate of decreasing q must be rapid enough so that some solitons survive. For any that do, however, their decay, involving large energies, should be rather dramatic. The soliton's decay will, moreover, be characteristic of such solitons only, as for $q \to 0$ any system of cholesteric defects will go smoothly into the ground state or nematic defects.

4. **Nematics in a sphere.** — Similar observations can be made in nematics. We shall be interested in the case when the director at a surface has a preferred direction : either in the plane of the surface, normal to it (*homeotropic* orientation), or at a spe-

cific angle to the surface normal (*conical* orientation), the last often realized at a nematic-isotropic interface [12]. The case of homeotropic orientation has been discussed previously by one of us [13]. There, it was shown that for a surface with Euler characteristic E, there must be a total index of point defects $q = E/2$. Consider then the case where the director must lie parallel to the surface, or at a fixed angle with parallel component. Here,

$$G = S^2/Z_2\,, \qquad H = S^1/Z_2\,,$$

so that by use of the exact sequence $\pi_2(G, H) = Z \times Z$. As there are point defects in the bulk ($\pi_2(G) = Z$), and point defects on the boundary ($\pi_1(H) = Z$), this result is simply telling us that they can combine in a trivial manner.

Let us then address the problem of nematics in a sphere and ask which configurations are possible topologically and which configurations will be expected to predominate energetically. To begin with, the use of Euler's characteristic tells us there must be two units of vorticity on the surface [5] :

$$\sum_{\text{vortices}} m = E\,.$$

The number of solitons which one can fit onto these boundary point defects is somewhat larger than for cholesterics. First there is the possibility of line disclinations terminating on the surface, the least energetic possibility being four $m = 1/2$ disclinations. Consider next the case where the surface defects coalesce into a single $m = -2$ defect, like the cholesteric boojum. There are two possibilities. One is a defect-free texture within the sphere matching smoothly onto the boundary singularities, like the He^3-A texture. The other is analogous to the Frank-Pryce texture without twist, or better Blaha's vorton [14] point defect, which is now topologically stable, connected by a line defect to the $m = -2$ boundary singularities. Following the analysis of Volovik as mentioned above, there are an infinite number of homotopic classes of such maps isomorphic to Z, corresponding to the strength of the bulk point defect.

In addition to these four configurations, there are two solutions which can result if the vortices separate and migrate to opposite poles. The first, illustrated figure 3*a* relaxes to a non-singular texture, leaving a configuration similar to an onion-skin, as illustrated of the sphere along a diameter. In any plane perpendicular to this diameter, the directors lie in concentric circles in the plane. There is another possibility where the line defect connecting the poles in figure 3*a* relaxes to a nonsingular texture, leaving a configuration similar to an onion-skin, as illustrated in figure 3*b*. In this case, it is as if one boundary defect acts as a source and the other as a sink, with the molecules *flowing* between. On the surface, with the boundary singularities at the poles, the directors lie parallel to the lines of equal longitude; in the interior, the molecules lie parallel to the diameter connecting the poles. The rest of the texture interpolates smoothly between the two. Such a configuration was earlier proposed on the basis of extensive energetic calculations [15]. This texture is not possible if the order parameter in the bulk is a triad, as in He^3-A or cholesterics, since in these cases 2π line defects are not equivalent to the defect-free state.

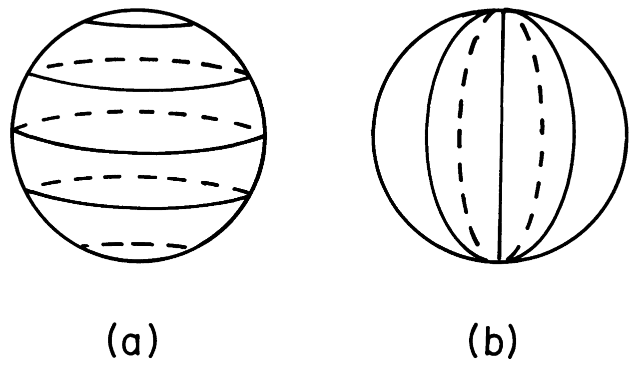

Fig. 3. — *a)* A solution of a nematic in a sphere with a line defect along the diameter connecting the poles. *b)* A solution which is regular everywhere except at the poles; the *onion-skin* texture.

As to which configurations will predominate, it appears that the 2π configurations of figures 3*a* and 3*b* will be favored over those with 4π surface vortices. This is contrary to the case of He^3-A and cholesterics, where the opposite is true. For spheres which are slightly deformed, the onion-skin texture should prevail. The gain in energy over configurations involving line and/or point defects in the bulk is clear, involving energies as $R \ln R$ and R, respectively, where R is the radius of the sphere. The gain relative to the non-singular 4π texture is not as great, for as the energy of a point defect of strength m on the surface goes as $c_1 m^2 \ln R + c_2$, two $m = -1$'s win over one $m = 2$. Here c_2 is due to the energy of the bulk texture, c_1 some constant, so this is only valid for large R. As the only other length of relevance is the intermolecular spacing, this is no difficulty. If one deforms the sphere to a shape approaching that of a pancake, it is expected that then the structure of figure 3*a* will eventually be favored. In no case does it appear that the combinations of the surface defects to form a 4π texture will be preferable.

Candau *et al.* [16] have carried out experiments in which nematic and cholesteric droplets (MBBA, Merck IV-cholesteryl propionate) are immersed in an isotropic solvent (glycerol). The ground state for nematic spherulites with tangential orientation was indeed the *onion-skin* of figure 3*b*, and for cholesterics with pitch much less than spherulite radius observations consistent with Robinson *et al.* [9] were reported. For nematic droplets with homeotropic orientation at the boundaries observations agreed with the analysis of ref. [13].

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