Topology of Smectic Order on Compact Substrates

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Smectic orders on curved substrates can be described by differential forms of rank one (1-forms), whose geometric meaning is the differential of the local phase field of density modulation. The exterior derivative of the 1-form is the local dislocation density. Elastic deformations are described by superposition of exact differential forms. Applying this formalism to study smectic order on a torus as well as on a sphere, we find that both systems exhibit many topologically distinct low energy states that can be characterized by two integer topological charges. The total number of low energy states scales as the square root of the substrate area. For a smectic on a sphere, we also explore the motion of disclinations as possible low energy excitations, as well as its topological implications.

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The problem of smectic order on a curved substrate naturally arises in a variety of contexts, such as block copolymer films coated on a curved substrate [1–4], colloidal particles immersed in a smectic liquid crystal [5] with a strong tangential boundary condition, smectic polymer vesicles, as well as Turing patterns on a sphere [6]. Last but not least, a flexible charged polymer adsorbed onto an oppositely charged curved surface [7] may also form an equal-distance layer pattern on the surface.

The main purpose of this Letter is to study the low energy smectic states on curved substrates with a minimal number of defects, which are experimentally most relevant at low temperature. We shall find that smectic order both on a sphere and on a torus exhibit many topologically distinct, nearly degenerate, and minimally defected states. These states can be classified by two integer topological charges, which are related to substrate topology as well as to a disclination pattern. The total number of these states is proportional to $\sqrt{A}/\ell_0$, where A is the substrate area and ℓ_0 the preferred layer spacing. Their energy differences scale sublinearly with the system size. For smectic on a sphere, by exchanging defects pairs in an appropriate manner, two integer charges can be systematically changed. Interestingly, the corresponding process realizes the Euclidean algorithm for finding the greatest common divisor of two integers. Our results are complementary to a recent work by Santangelo *et al.* [1], which addresses the energetic interplay between smectic order and substrate curvature. A more detailed version of our analysis is presented elsewhere [8].

The local phases of density modulation of a smectic order at two nearby points x and $x + dx$ are related by $\Theta(x + \delta x) \approx \Theta(x) + \psi_\alpha(x)dx^\alpha$. While the phase field Θ is generically not globally well defined, its differential $\psi(x) = \psi_\alpha dx^\alpha$, called a 1-form in modern differential geometry [9], nevertheless still is. A simple geometric reasoning shows that the norm of ψ is proportional to the reciprocal of layer spacing ℓ: $|\psi| = \sqrt{g^{\alpha\beta}\psi_\alpha\psi_\beta} = 2\pi\ell^{-1}$. The nonlinear strain $w(x) = \frac{1}{2}(\frac{\ell_0^2}{4\pi^2}|\psi|^2 - 1)$ measures the dilation or compression of smectic layers. This strain is identical to the one used in Ref. [1] and reduces to the well-known nonlinear strain for smectic order in flat space [11,12]. The total free energy of the system is the sum of the strain energy density $Bw^2/2$ and the Frank free energy, appropriately generalized to curved space [13].

The integral of ψ along a loop γ is the net phase change $\Delta\Theta$ as one walks around γ; i.e., the dislocation charges, multiplied by 2π, enclosed by the loop. Using the Stoke's theorem this can in turn be related to the 2D integral over the region D enclosed by γ:

$$\Delta\Theta = 2\pi N_\gamma = \oint_\gamma \psi = \int_D d\psi, \tag{1}$$

where the two form $d\psi = (\partial_x\psi_y - \partial_y\psi_x)dx \wedge dy$ is the exterior differential of ψ. $d\psi$ therefore must be the dislocation density. Hence a dislocation-free smectic state is described by a 1-form satisfying $d\psi = 0$. Such a 1-form is called *closed*. By contrast, a 1-form $\psi = d\Theta$ that is the differential of a function Θ is called *exact*.

An exact form is closed. If the phase field Θ is globally defined, we have $\psi = d\Theta$, and $d\psi = d^2\Theta = (\partial_1\partial_2 - \partial_2\partial_1)\Theta dx^1 \wedge dx^2 \equiv 0$; that is, the dislocation density vanishes everywhere. This obvious result is called Poincaré's lemma. Its converse holds in flat space: If $d\psi = 0$ everywhere, the phase field Θ is globally defined: $\psi = d\Theta$, and the integral equation (1) along arbitrary loop always vanishes.

The converse of Poincaré's lemma may fail in space with different topology [14]. De Rham's theorem of cohomology [15] identifies the number of independent closed-but-not-exact forms with that of nonretractable loops on the manifold. A torus, for example, has two nonretractable loops, as shown in Fig. 1(a). The most general closed 1-form on a torus is given by

$$\psi_{N_\theta,N_\phi} = N_\theta d\theta + N_\phi d\phi + d\Phi(\theta, \phi). \tag{2}$$

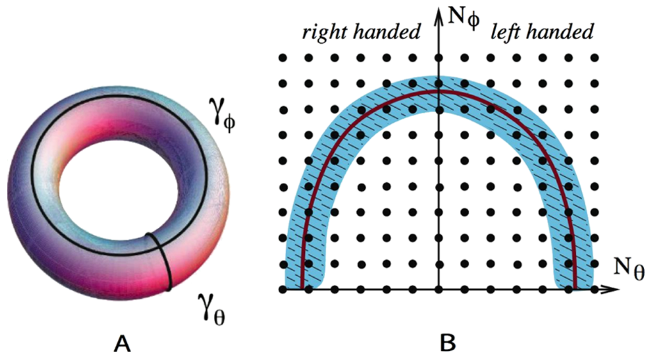

FIG. 1 (color online). (A) Two cycles on a torus. (B) Smectic states on a torus with distinct global dislocation charges are shown as grid points in the upper-half (N_θ, N_ϕ) plane. The curve (ellipse) is the loci of vanishing strain $w = 0$, as determined by Eq. (3). States in the shaded region have low strain energy and are approximately degenerate.

The integral of ψ along loops γ_θ and γ_ϕ give, respectively, $2\pi N_\theta$ and $2\pi N_\phi$. Equation (2) is exact only if both N_θ and N_ϕ vanish. In order for Eq. (2) to describe a defect-free smectic state on a torus, both N_θ and N_ϕ must be integers. The exact form $d\Phi(\theta, \phi)$ in Eq. (2) describes an elastic deformation of smectic layers relative to the state with $d\Phi = 0$. It can be ignored for a sufficiently thin torus.

We define two states to be topologically equivalent if they can be brought into each other by continuous deformation that does not break any smectic layer. The topological properties of a defect-free smectic state on a curved substrate are therefore characterized by a closed-but-not-exact differential form. Similarly, any excitation or distortion of the smectic pattern that cannot be relaxed away by elastic deformation is a topological defects. According to this definition, therefore topological defects should be identified as the source of *internal strain*, a concept thoroughly explored in the continuous theory of defects in crystals [16]. We note, however, that this definition of topological equivalence is more refined than the one used in the homotopy theory topological defects [17,18]. Consequently, the topological charges discussed in this work may not be truly topological invariants according to the homotopy theory. A more detailed discussion of this issue can be found in [8].

Toroidal smectic states with different charges (N_θ and N_ϕ) are clearly topologically distinct. Two integers (N_θ and N_ϕ) share essential similarity with usual dislocation charges but encode global properties of *defect-free* smectic states on a torus. Hence they shall be called *global dislocation charges* [19]. Each defect-free smectic state can therefore be represented as a point in the half lattice of integers (N_θ and N_ϕ) [20], as shown in Fig. 1(b).

The nonlinear strain associated with states Eq. (2) on a thin torus (with $d\Phi$ set to zero) is given by

$$w = \frac{\ell_0^2 N_\theta^2}{(2\pi R_\theta)^2} + \frac{\ell_0^2 N_\phi^2}{(2\pi R_\phi)^2} - 1. \qquad (3)$$

As shown in Fig. 1(b), the equation $w = 0$ traces out an ellipse in the (N_θ, N_ϕ) plane. States satisfying $|w| \leq \ell_0/\sqrt{A}$ [shaded in Fig. 1(b)] have the total strain energy bounded by $B\ell_0^2/2$, which is independent of the system size. A simple calculation [8] also shows that the total Frank free energy scales sublinearly with the system size. Therefore all of these states are approximately degenerate for a large system. The total number of these low energy states is given by the area of the shaded region in Fig. 1(b) and scales as the square root of the substrate area:

$$\mathcal{N}\left(F_B \leq \frac{1}{2}B\ell_0^2\right) = 2\pi\frac{\sqrt{A}}{\ell_0}, \qquad (4)$$

We now turn to smectic order on a sphere. According to the Gauss-Bonnet-Poincaré theorem, the total disclination charge of a smectic order on a sphere must be two. Let us write $\psi = |\psi|\hat{n}$, where $\hat{n} = \hat{n}_\alpha dx^\alpha$ is the unit 1-form describing the smectic layer normal, while $|\psi|$ is the norm of ψ. Taking the exterior differential of ψ, as well as using the Leibniz rule, we find $d\psi = (d|\psi|) \wedge \hat{n} + |\psi| d\hat{n}$. Note that $d\hat{n} = (\partial_1 \hat{n}_2 - \partial_2 \hat{n}_1) dx^1 \wedge dx^2$ is a 2-form describing bending deformation of the layer normal. Hence, if there is no dislocation ($d\psi = 0$) and the layer spacing is constant ($d|\psi| = 0$), the bending deformation of the director field is strictly forbidden ($d\hat{n} = 0$). This implies that the bending constant K_3 is effectively infinity in a dislocation-free smectic [21].

A spherical nematic in the limit of infinite bending rigidity is characterized by a one-parameter family of degenerate ground states, where four $+1/2$ disclinations sit on a great circle and form a rectangle with an arbitrary aspect ratio [22]. For a given bending-free nematic state, we can start from disclination cores and grow, layer by layer, a dislocation-free smectic pattern with equal layer spacing. We will, however, have to fine-tune the layer spacing ℓ so that the smectic pattern can be fit onto the sphere with given radius R. This fine-tuning results in a small strain of order of ℓ_0/R and a total strain energy $F_B \approx B\ell_0^2$, which does not scale with the system size. Since the Frank free energy of all of these states is the same by construction, the total free energy is approximately degenerate.

Several numerical studies of spherical smectic orders formed by diblock copolymer films confined on a sphere have been reported recently [2–4]. By slowly annealing the system, three classes of low energy smectic states were found [2]: latitudinal states [23], where all layers are circles of constant latitude, spiral states, where all smectic layers are spirals around the two poles, and quasibaseball states, where four $+1/2$ disclinations are well separated on a great circle. All of these states are dislocation-free. There may be, however, at most one layer termination at each disclination core.

Consider a quasibaseball state, shown in Fig. 2(a), with four disclinations sitting on the equator. We stereographically project the pattern from the south pole to the complex

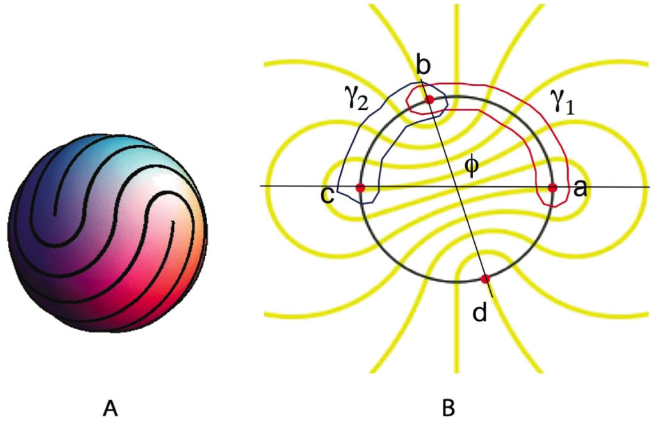

FIG. 2 (color online). (A) A quasibaseball state, with four $+1/2$ disclinations sitting on the equator. Each disclination core has one layer termination. Only two disclinations are visible here. (B) The same state stereographically projected onto the complex plane. The equator is mapped to the unit circle. The red contour encloses defects a and b and intersects 12 smectic layers in total. The blue contour encloses defects b and c and intersects 8 layers in total. Hence this quasibaseball state is labeled by two integer charges ($N_1 = 12$ and $N_2 = 8$).

plane, such that the equator is mapped into the unit circle. The image smectic pattern is shown in Fig. 2(b), with four defects located at $(a, b, c, d) = (1, e^{i\phi}, -1, e^{-i\phi})$, respectively. The segment ab of the unit circle intersects $N_1/2$ smectic layers. (A layer terminated at disclination a or b is counted as a $1/2$ intersection.) Likewise, the segment bc of the unit circle intersects $N_2/2$ smectic layers. Note that by definition both N_1 and N_2 are non-negative. Without loss of generality we choose $N_1 \le N_2$. Two integer charges (N_1 and N_2) completely determine the global properties of a quasibaseball state. Latitudinal states are special cases of quasibaseball states with $N_1 = 0$ [24], while spiral states correspond to the regime $N_1 \ll N_2$.

It is clear from Fig. 2(a) that all smectic layers intersect the equator vertically. The arc lengths s_{ab} and s_{bc} are therefore given by

$$s_{ab} = R\phi = N_1\ell, \qquad s_{bc} = R(\pi - \phi) = N_2\ell,$$

from which we find

$$\frac{N_1}{N_2} = \frac{\phi}{\pi - \phi}, \qquad \ell = \frac{\pi R}{N_1 + N_2}, \tag{5}$$

where $\ell \approx \ell_0$ is the layer spacing. The equilibrium value of the angle ϕ is therefore completely determined by the two charges N_1 and N_2. Large fluctuations of ϕ necessarily induce a significant change of layer spacing and therefore is energetically penalized. More importantly, Eq. (5) also shows that, in all low energy states, $N_1 + N_2$ is completely determined by the sphere radius R and the layer spacing ℓ. Since N_1 can take an arbitrary integer value from 0 to $\pi R/\ell$, the total number of low energy smectic states on a sphere is approximately given by

$$\mathcal{N}(F_B \le B\ell_0^2) = \frac{\pi R}{\ell_0} + 1 \approx \frac{\sqrt{\pi A}}{\ell_0}, \tag{6}$$

i.e., also scales as the square root of the sphere area.

To obtain an analytic description for quasibaseball states, we invoke the Hodge decomposition theorem [15] of differential forms on a compact manifold. The closed 1-form ψ can be expressed as the sum of the real part of an analytic form ψ^c and an exact form $d\alpha$:

$$\psi(x, y) = \mathrm{Re}\,\psi^c(z) + d\alpha(x, y), \tag{7}$$

where $\psi^c(z) = f(z)dz$, with $f(z) = f(x + iy)$ a meromorphic function, while $\alpha(x, y)$ is a smooth (but not analytic) function on the z plane. The topological properties of the smectic order on a sphere is completely encoded in $f(z)dz$. The exact form $d\alpha$ describes elastic deformation and should be chosen to minimize the total free energy.

The complex meromorphic form ψ^c that describes a spherical smectic with four disclinations at a, b, c, and d can be shown to be [8]

$$\psi^c = \frac{Ae^{-i\alpha}}{\sqrt{(z-a)(z-b)(z-c)(z-d)}}dz. \tag{8}$$

As shown in Fig. 2(b), let us construct two loops γ_1 and γ_2 that enclose the segments ab and bc, respectively. Integration of ψ along the loops γ_1 and γ_2 yield two topological charges N_1 and N_2 that we defined above:

$$2\pi N_k = \oint_{\gamma_k} \psi = \mathrm{Re} \oint_{\gamma_k} \psi^c, \qquad k = 1, 2. \tag{9}$$

Solving Eq. (9) for A and α, we obtain

$$Ae^{-i\alpha} = \left(\frac{\pi N_2}{K(\cos\frac{\phi}{2})} - i\frac{\pi N_1}{K(\sin\frac{\phi}{2})}\right)e^{i\phi/2}, \tag{10}$$

where $K(\cdot)$ is complete elliptic integral of the first kind. Equations (8) and (10) completely determine the meromorphic form ψ^c for given charges N_1 and N_2. We note that the integral of ψ^c Eq. (8) defines the (multiple-valued) inverse function of a doubly periodic elliptic function. Two integer charges N_1 and N_2 are the real parts of two periods of this elliptic function [8].

Spiral states are especially interesting because of their closely bound defects pairs. As illustrated in Fig. 3, consider a spiral state with charges ($N_1 = 2, N_2$), align the sphere so that one pair of defects a and b sit near the north pole. Let us fix the south pole and elastically twist the whole sphere, together with all smectic layers, around the z axis by an angle π, so that the defects pair a and b at the north pole exchange their positions. The branch cut [shown blue in Fig. 3(a)], which connects a and d (invisible in Fig. 3) before the twist operation, after the twist operation connects b and d instead, shown red in Fig. 3(b). The corresponding integer charge associated with this branch cut is N_2 before the twist and becomes $N_2 + N_1$ after the twist. Hence a counterclockwise twist of the defects pair a and b by angle π leads to the following transformation of

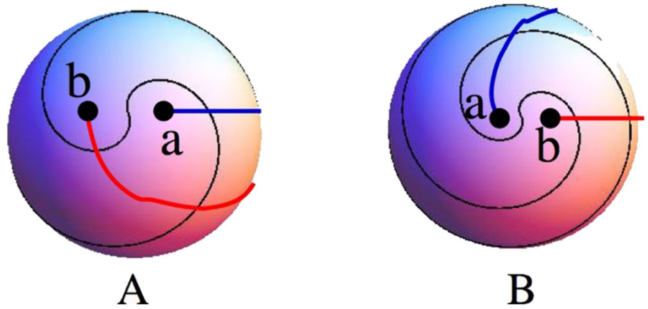

FIG. 3 (color online). Starting from a state (N_1, N_2) with $N_1 = 2$, we twist the sphere counterclockwise by π so as to exchange the defects pair a and b and obtain a state $(N_1, N_2 + N_1)$. These two states are therefore topologically equivalent. This twist operation changes the layer spacing by a factor of N_1/N_2.

the topological charges:

$$(N_1, N_2) \to (N_1, N_2 + N_1). \tag{11}$$

Obviously clockwise twist of the pair (a, b) leads to a different transformation:

$$(N_1, N_2) \to (N_1, N_2 - N_1). \tag{12}$$

Therefore, if defects are allowed to move, states (N_1 and $N_2 \pm N_1$) are topologically identical to the state (N_1, N_2). As long as $N_1 \ll N_2$, this twist manipulation induces only a small change in layer spacing and therefore costs little strain energy. The twist of closely bound defects pair is therefore a low energy excitation in spiral states.

It is important to note that, even if N_1 and N_2 are comparable, a twist of defects pair is still topologically possible but costs large strain energy. The two charges (N_1 and N_2) of a quasibaseball states are therefore stabilized by both topological and energetic barriers. Furthermore, defects twisting can be successively applied. Starting with a state with $N_1 \leq N_2$, we twist an appropriate defects pair so as to obtain new charges ($N_1' = N_1$ and $N_2' = N_2 - N_1$). If $N_1' \leq N_2'$ still holds, we simply repeat the same twist operation. If, after the twist, we find $N_1' \geq N_2'$, we twist a different pair so that we have a new state $(N_1', N_2' - N_1')$. In every step, one of two integers (N_1 or N_2) is reduced. This process therefore must end after finite steps, where one of the two integer charges vanishes. The final state is therefore a latitudinal state. More interestingly, this process of charge reduction is precisely the *Euclidean algorithm* for finding the greatest common divisor $\gcd(N_1, N_2)$ between two integers (N_1 and N_2). Therefore the nonvanishing charge of the final state must be $\gcd(N_1, N_2)$. Hence, if disclinations are allowed to move, an arbitrary low energy smectic state on a sphere is topologically identical to a certain latitudinal state, with only one nonzero integer topological charge.

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[1] C. D. Santangelo *et al.*, Phys. Rev. Lett. **99**, 017801 (2007).
[2] T. L. Chantawansri *et al.*, Phys. Rev. E **75**, 031802 (2007).
[3] J. F. Li *et al.*, Eur. Phys. J. E **20**, 449 (2006).
[4] P. Tang *et al.*, Phys. Rev. E **72**, 016710 (2005).
[5] C. Blanc and M. Kleman, Eur. Phys. J. E **4**, 241 (2001).
[6] C. Varea, J. L. Aragon, and R. A. Barrio, Phys. Rev. E **60**, 4588 (1999); P. C. Matthews, *ibid.* **67**, 036206 (2003).
[7] A. Y. Grosberg, T. T. Nguyen, and B. I. Shklovskii, Rev. Mod. Phys. **74**, 329 (2002); S. Y. Park, R. F. Bruinsma, and W. M. Gelbart, Europhys. Lett. **46**, 454 (1999); E. M. Mateescu, C. Jeppesen, and P. Pincus, Europhys. Lett. **46**, 493 (1999); R. Netz and J.-F. Joanny, Macromolecules **32**, 9013 (1999); **32**, 9026 (1999).
[8] X. Xing, arXiv:0806.2409.
[9] The application of differential form and exterior calculus to defected condensed matter systems dates back to Dzyaloshinskii and Volovik, as well as to Julia and Toulouse. See [10] and references therein.
[10] I. E. Dzyaloshinskii, *Gauge Theories and Density of Topological Singularities*, Les Houches Lecture Notes (North Holland, Amsterdam, 1980).
[11] P. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1993).
[12] P. Chaikin and T. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, England, 1995).
[13] See Ref. [8] for a more detailed discussion on energetics of smectic order on curved substrates.
[14] Such a failure can be seen only at the global scale. Locally, a manifold resembles flat space; hence, the converse of the Poincaré lemma always holds locally.
[15] M. Nakahara, *Geometry, Topology, and Physics* (Taylor & Francis, London, 2003); T. Frankel, *The Geometry of Physics* (Cambridge University Press, Cambridge, England, 2004); M. Stone, online lecture notes, http://webusers.physics.uiuc.edu/~m-stone5/.
[16] E. Kroner, *Continuous Theory of Defects*, Les Houches Lecture Notes (North-Holland, Amsterdam, 1980).
[17] M. Kleman and J. Friedel, Rev. Mod. Phys. **80**, 61 (2008).
[18] N. D. Mermin, Rev. Mod. Phys. **51**, 591 (1979).
[19] Similar integer charges also appear in crystalline orders on compact manifolds, such as carbon nanotubes and spherical crystals.
[20] The state $(-N_\theta, -N_\phi)$ is the same as (N_θ, N_ϕ).
[21] This result applies to arbitrary surface, since the above discussion does not refer to any metric structure.
[22] H. Shin, M. J. Bowick, and X. Xing, Phys. Rev. Lett. **101**, 037802 (2008).
[23] These are called hedgehog states in Ref. [2].
[24] For a latitudinal state $N_1 = 0$, the contour γ_1 in Fig. 2 can be shrunk to a single point and gives no information about the global property. There is then only one contour γ_2 left, which gives one integer charge.