Surface-Groove-Induced Azimuthal Anchoring of a Nematic Liquid Crystal: Berreman's Model Reexamined

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To account for azimuthal surface anchoring of a nematic liquid crystal, Berreman [Phys. Rev. Lett. **28**, 1683 (1972)] proposed a simple model attributing the surface anchoring to the elastic distortion of the liquid crystal induced by the grooves of a surface. He showed that the surface anchoring energy is proportional to $\sin^2\phi$, with ϕ being the angle between the director at infinity and the direction of the surface grooves. We argue that his assumption of negligibly small azimuthal distortion of the nematic is not valid. Proper treatment of the azimuthal distortion reveals that the Berreman's model should yield a surface anchoring energy proportional to $\sin^4\phi$. This implies that surface grooves alone cannot contribute to the surface anchoring coefficient in the usual Rapini-Papoular sense.

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The utility of a liquid crystal in applications relies heavily on the fact that the order of a liquid crystal can be controlled by a surface adjacent to it. Such tendency of a surface to align a liquid crystal is called ''surface anchoring'' [1–3], and to achieve desirable anchoring properties is of crucial importance in practical applications such as display technologies. What brings about surface anchoring and how it can be controlled are also of fundamental interest.

The easiest way of achieving an alignment of a liquid crystal along one direction parallel to a surface is to rub the surface in that direction. There has been a longstanding debate on the underlying mechanism of anchoring on rubbed surfaces. Several studies attributed it to the intermolecular interactions between the liquid crystal molecules and the polymer chains constituting the surfaces [4–6], while others discussed the effect of long-range elastic distortion induced by surface grooves or scratches created by the rubbing process. The first theoretical study on the latter mechanism was carried out by Berreman [7], who proposed a simple model; a nematic liquid crystal in contact with a sinusoidal wavy surface. He assumed that the director $\boldsymbol{n}$, a unit vector describing the local orientation of a nematic liquid crystal, at the surface is always parallel to it, and calculated the free energy due to the distortion of $\boldsymbol{n}$ induced by the sinusoidal surface. In the case of equal bend and splay elastic constants ($K_1 = K_3 = K$), the free energy due to the surface per unit area reads

$$f = \frac{1}{4} K A^2 q^3 \sin^2\phi, \tag{1}$$

where A and $q > 0$ are the amplitude and the wave number of the sinusoidal surface, respectively, and ϕ describes the angle between the groove direction and the director at infinity. Since the Berreman's model is simple enough, it has served as a starting point for subsequent numerous theoretical as well as experimental studies concerning the geometrical aspects of surface anchoring [8–10]. Several experimental studies were also devoted to the direct confirmation of the Berreman's argument [11–13]. We also note that the recent rapid development of nanotechnology has opened up the possibility of creating submicron-scale patterned or grooved surfaces to realize certain specific anchoring properties [14–20]. Experimental realization of a submicron-scale surface grooved with sufficient geometrical precision has again provoked interest in the notion of surface anchoring attributable to the geometry of the surface.

In this Letter, we critically reexamine the theoretical treatment of Berreman for the surface anchoring induced by grooves. His theory deriving Eq. (1) involves an assumption that only the tilt distortion is induced by the surface grooves; the azimuthal distortion is negligibly small. We argue that this assumption cannot be valid for general ϕ except for $\phi = 0, \pi/2$; those distortions are of comparable magnitude. Starting from the same setup of the Berreman's argument, we recalculate the surface anchoring energy and find that Eq. (1) needs to be modified in a qualitatively substantial manner.

Let us first describe the model of the system we want to consider and briefly review the theory of Berreman [7]. The Frank elastic energy of a nematic liquid crystal can be described in terms of a director $\boldsymbol{n}$ as [1] $F = \frac{1}{2}\int d\boldsymbol{r}(K_1(\boldsymbol{\nabla}\cdot\boldsymbol{n})^2 + K_2(\boldsymbol{n}\cdot\boldsymbol{\nabla}\times\boldsymbol{n})^2 + K_3(\boldsymbol{n}\times\boldsymbol{\nabla}\times\boldsymbol{n})^2)$. When the distortion of the nematic from the uniform alignment (we take the x direction along this aligned direction) is small enough, we can write down the director as $\boldsymbol{n} = (\sqrt{1-n_y^2-n_z^2}, n_y, n_z) \simeq (1, n_y, n_z)$ and the Frank elastic energy up to quadratic order in n_y and n_z reads

$$F = \frac{1}{2}\int d\boldsymbol{r}(K_1(\partial_y n_y + \partial_z n_z)^2 + K_2(\partial_y n_z - \partial_z n_y)^2 + K_3[(\partial_x n_y)^2 + (\partial_x n_z)^2]), \quad (2)$$

Here we consider a surface groove whose shape can be described by

$$z = \zeta(x, y) = A\sin[q(x\sin\phi + y\cos\phi)], \quad (3)$$

where A, q, and ϕ have been defined above and we assume $Aq \ll 1$. A nematic liquid crystal is filled in the semi-infinite region $z > \zeta(x, y)$. We further assume that the director at the surface tends to lie tangential rather than perpendicular to it, and no preferred direction on the surface is postulated. When $\phi = \pi/2$, the director at the surface should then behave as $n_y = 0$ and $n_z = Aq\cos qx$. With the simplifying assumption that $K_1 = K_3$, the solution of the Euler-Lagrange equations $\delta F/\delta n_{y,z} = 0$ compatible with the above surface behavior of the director reads $n_y = 0$, and

$$n_z = Aq\cos(qx)e^{-qz}. \quad (4)$$

Substituting Eq. (4) into Eq. (2), one can calculate the energy of the director distortion due to the surface groove, that is, the anchoring energy of the grooved surface. The energy per unit area f is $f = \frac{1}{4}K_3A^2q^3$.

In the general case with $\phi \neq \pi/2$, Berreman assumed that the azimuthal variation of the director, n_y is negligibly small as compared with that of n_z. Then, after taking care of the Euler-Lagrange equation $\delta F/\delta n_z = 0$ and the boundary condition at the surface,

$$n_z = Aq\sin\phi\cos[q(x\sin\phi + y\cos\phi)], \quad (5)$$

one finds that Eq. (4) must be replaced by

$$n_z = Aq\sin\phi e^{-qz}\cos[q(x\sin\phi + y\cos\phi)], \quad (6)$$

which yields Eq. (1).

To discuss the validity of the above Berreman's theory, in particular, the assumption of negligibly small n_y, let us consider the equilibrium conditions using the full variational principle; $\delta F = 0$ for infinitesimal variation of the director, δn_y and δn_z. It yields the Euler-Lagrange equations

$$0 = -K_1\partial_y(\partial_y n_y + \partial_z n_z) + K_2\partial_z(\partial_y n_z - \partial_z n_y) - K_3\partial_x^2 n_y, \quad (7)$$

$$0 = -K_1\partial_z(\partial_y n_y + \partial_z n_z) - K_2\partial_y(\partial_y n_z - \partial_z n_y) - K_3\partial_x^2 n_z, \quad (8)$$

together with the condition at the surface [21]:

$$K_1(\partial_y n_y + \partial_z n_z)\delta n_z - K_2(\partial_y n_z - \partial_z n_y)\delta n_y = 0. \quad (9)$$

Next we consider the boundary conditions for n_y and n_z. From Eq. (3), the assumption of the tendency of the director at the surface to lie tangential to the surface reads

$$\partial\zeta/\partial x + n_y\partial\zeta/\partial y - n_z = 0. \quad (10)$$

Equation (10) yields Eq. (5) for n_z up to leading order in Aq, which serves as the boundary condition for n_z. Since $\partial\zeta/\partial x$, $\partial\zeta/\partial y$, and n_z are of the order of Aq, and we have assumed small n_y (here by "small" we imply that n_y is not of order unity), Eq. (10) does not impose any boundary condition on n_y in the leading order in Aq. The following calculation indeed yields n_y of the order of Aq, which justifies *a posteriori* the present treatment for n_y. The fixed boundary condition for n_z, Eq. (5), requires that δn_z in Eq. (9) must be set to zero. Since no condition is imposed for δn_y as noted above, Eq. (9) then results in an additional boundary condition

$$\partial_y n_z - \partial_z n_y = 0 \quad (11)$$

at the surface.

To summarize, what we must do to obtain the equilibrium director profile is to solve the Euler-Lagrange Eqs. (7) and (8) under the boundary conditions (5) and (11). Since Eqs. (7) and (8) are effectively fourth-order linear differential equations due to the coupling between n_y and n_z, four eigenmodes are present in a mathematical sense. However, two of them are incompatible with $n_y = n_z = 0$ at $z = +\infty$. A third one [22] cannot satisfy the boundary condition (11). Thus we are left with one eigenmode that is reasonable in a real physical system. Taking into account the boundary condition (5), one obtains

$$n_z = Aq\sin\phi e^{-qzg_1(\phi)}\cos[q(x\sin\phi + y\cos\phi)], \quad (12)$$

$$n_y = \frac{Aq\sin\phi\cos\phi}{g_1(\phi)}e^{-qzg_1(\phi)}\sin[q(x\sin\phi + y\cos\phi)], \quad (13)$$

with $g_1(\phi) = \sqrt{\cos^2\phi + (K_3/K_1)\sin^2\phi}$. Obviously Eq. (12) satisfies the boundary condition (5) and it is not a difficult task to check that Eqs. (12) and (13) are indeed the solution of the Euler-Lagrange Eqs. (7) and (8) fulfilling the remaining boundary condition (11). In Fig. 1, we show what our director profile, Eqs. (12) and (13), looks like for $\phi = \pi/4$ as compared with Berreman's profile.

From Eqs. (2), (12), and (13), we can straightforwardly calculate the anchoring energy per unit area as

$$f = \frac{1}{4}K_3A^2q^3\frac{\sin^4\phi}{g_1(\phi)}. \quad (14)$$

Several comments are in order: Eqs. (12) and (13) clearly show that n_z, $n_y = \mathcal{O}(Aq)$ and therefore are indeed compatible with the boundary condition (10) up to the leading order in Aq. We also notice that (14) does not contain the twist elastic constant K_2, which indicates that no twist deformation is involved. Indeed, from Eqs. (12) and (13), one can easily check $\partial_y n_z - \partial_z n_y = 0$, which again im-

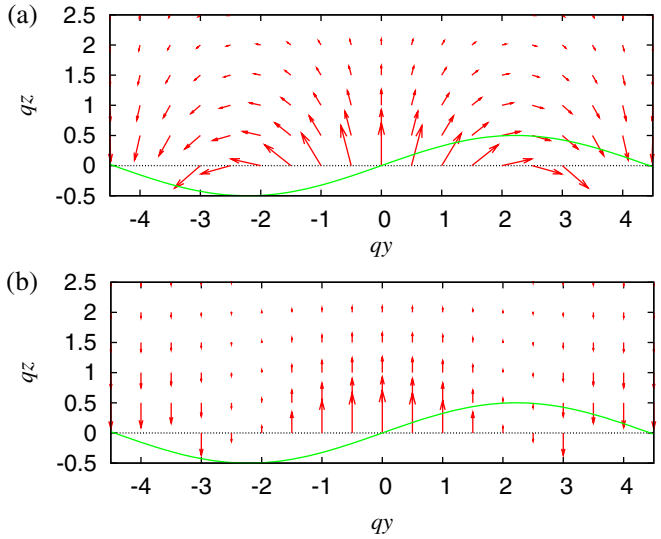

FIG. 1 (color online). (a) Our director profile (n_y, n_z) at $x = 0$, projected onto the $x = 0$ plane. (b) Corresponding Berreman's director profile, with $n_y = 0$. Here we have set $\phi = \pi/4$ and $K_1 = K_3$ is assumed for both cases. The groove profile $\zeta(x, y)$ at $x = 0$ is also shown. The projected vectors are drawn in an exaggerated manner, and the units of length for the projected director are the same for both figures.

plies the absence of twist deformation. Note also that the absence of twist at the surface is nothing but the boundary condition (11).

The most important thing that Eq. (14) implies is that the dependence of the anchoring energy on ϕ is different from that derived by Berreman (1). In a simple case, $K_1 = K_3$, $g_1(\phi) = 1$ yields $f \propto \sin^4\phi$, in sharp contrast to the Berreman's result $f \propto \sin^2\phi$. For clarity, we plot in Fig. 2 Berreman's energy (1) and ours (14) for $K_1 = K_3$. Note that when $\phi(\mathrm{mod}\pi) = 0$ or $\pi/2$, our result (14) agrees with the Berreman's (1), which is clear from the fact that $n_y = 0$ in this case [see Eq. (13)]. On the other

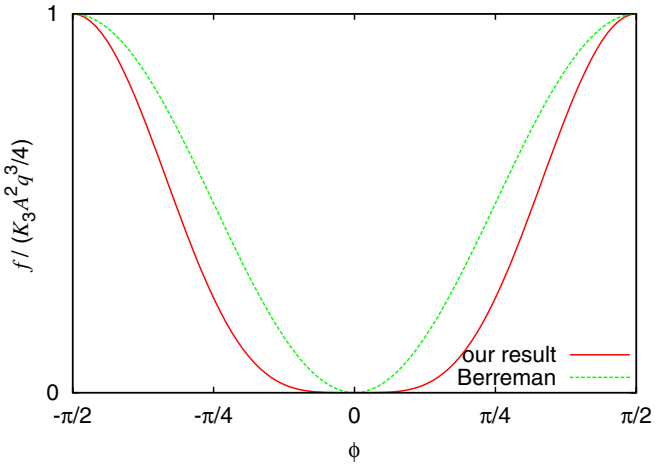

FIG. 2 (color online). Berreman's anchoring energy (1) and ours (14) as a function of ϕ for $K_1 = K_3$.

hand, when $\phi(\mathrm{mod}\pi) \neq 0$ or $\pi/2$, our surface energy (14) is always smaller than the Berreman's energy (1), as seen from Fig. 2. This arises from the fact that the Berreman's director profile, Eq. (6) together with $n_y = 0$, cannot be the equilibrium one. In the case of $K_1 \neq K_2$, the coupling between n_y and n_z prevents $n_y = 0$ from being the solution of the Euler-Lagrange Eqs. (7) and (8). Even when $K_1 = K_2$ (with n_y and n_z being decoupled), although the Berreman's profile now satisfies Eqs. (7) and (8), it is still incompatible with the boundary condition (11).

Almost all of the experimental studies on azimuthal surface anchoring summarize their results using the Rapini-Papoular form of the surface energy [23], i.e., $(1/2)W\sin^2\phi$, or $(1/2)W\phi^2$ in the case of small ϕ (W, the anchoring strength, is $KA^2q^3/2$ in the Berreman's theory). Our result clearly indicates that $W = 0$, that is, sinusoidal surface grooves cannot contribute to surface anchoring in the Rapini-Papoular sense, so long as Aq is small enough to allow the quadratic treatment of the Frank energy (2). One can observe an almost flat energy minimum at $\phi = 0$ in Fig. 2, which clearly reflects the absence of the anchoring of Rapini-Papoular type.

Finally, we comment on why an apparent additional distortion ($n_y \neq 0$) can reduce the Frank elastic energy (for simplicity, we discuss the director profiles in the case of equal elastic constants, $K_1 = K_2 = K_3$). Berreman's profile involves splay, twist and bend deformations, whose contributions to elastic energy are, respectively, given by $f_1^{(\mathrm{Berreman})} = \frac{1}{8}K_1q^3A^2\sin^2\phi$, $f_2^{(\mathrm{Berreman})} = \frac{1}{8}K_2A^2q^3\sin^2\phi\cos^2\phi$, and $f_3^{(\mathrm{Berreman})} = \frac{1}{8}K_3A^2q^3\sin^4\phi$. We note here that near the easy axis ($\phi = 0$), the dominant contribution to the total free energy, up to the order of $\sin^2\phi$, comes only from the splay and the twist modes. The nature of our profile is such that splay and twist deformations are avoided as much as possible; in fact, twist deformation is totally absent and the splay deformation energy, $f_1 = \frac{1}{8}K_1q^3A^2\sin^6\phi$, is nonzero, yet is of the order of $\sin^6\phi$ near $\phi = 0$. The cost of this behavior is a slight increase of bend energy to $f_3 = \frac{1}{8}K_3A^2q^3\sin^4\phi(1 + \cos^2\phi)$, which makes a significant contribution only at angles sufficiently away from the easy axis.

We have revisited the Berreman's theory on the surface anchoring of a nematic liquid crystal induced by surface grooves. We have argued that the assumption in his theory that the azimuthal distortion of the director is negligibly small is not appropriate. Starting from the same setup of the Berreman's argument and properly taking into account the azimuthal distortion of the director, we have shown that the anchoring energy as a function of the angle ϕ between the director at infinity and the groove direction should behave as $\sin^4\phi$, in remarkable contrast with the Berreman's result, $\sin^2\phi$. Berreman's model is the simplest one for the geometry-induced surface anchoring and most of the theoretical studies to deal with the surface anchoring

of geometrical origin begin with, or borrow ideas from it. Moreover, given the recent growing interest in the experimental attempts to utilize microscopically grooved surfaces to achieve desirable anchoring properties and also in the underlying mechanism of surface anchoring, we conclude this paper by emphasizing that the present theories on the geometry-induced surface anchoring and their comparison with experiments must be carefully reconsidered and reexamined.

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[21] One might wonder whether saddle-splay surfacelike elasticity contributes to the boundary condition at the surface. The corresponding energy reads $F_{24} = -\frac{1}{2}(K_2 + K_{24}) \times \int d\boldsymbol{S} \cdot (\boldsymbol{n}\nabla \cdot \boldsymbol{n} + \boldsymbol{n} \times \nabla \times \boldsymbol{n})$, where K_{24} is the associated saddle-splay elastic constant and $d\boldsymbol{S}$ is the vectorial surface element on the boundary. Since $\boldsymbol{n}$ is tangential to the surface in our setup, $d\boldsymbol{S} \cdot \boldsymbol{n} = 0$. The second term does not contribute, either, because $d\boldsymbol{S} \cdot (\boldsymbol{n} \times \nabla \times \boldsymbol{n}) = -d\boldsymbol{S} \cdot (\partial_x n_z \boldsymbol{e}_z + \partial_x n_y \boldsymbol{e}_y)$ which yields zero after integration. In conclusion, the saddle-splay surfacelike elasticity does not contribute to the boundary conditions at the surface. Here we do not discuss the splay-bend surfacelike elasticity (K_{13}); see H. Yokoyama, Phys. Rev. E **55**, 2938 (1997).
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