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## Molecular Crystals and Liquid Crystals

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N. M. Silvestre <sup>a b</sup> , P. Patrício <sup>c b</sup> & M. M. Telo da Gama <sup>a b</sup>

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<sup>&</sup>lt;sup>a</sup> Departamento de Física da Faculdade de Ciências, Universidade de Lisboa, Lisboa Codex, Portugal

<sup>&</sup>lt;sup>b</sup> Centro de Física Teórica e Computacional, Universidade de Lisboa, Lisboa Codex, Portugal

<sup>&</sup>lt;sup>c</sup> Instituto Superior de Engenharia de Lisboa, Lisboa, Portugal

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### Interactions between Circular Inclusions in Smectic-C Films with Planar Anchoring

N. M. Silvestre<sup>1,2</sup>, P. Patrício<sup>3,2</sup>, and M. M. Telo da Gama<sup>1,2</sup>

<sup>1</sup>Departamento de Física da Faculdade de Ciências, Universidade de Lisboa, Lisboa Codex, Portugal

<sup>2</sup>Centro de Física Teórica e Computacional, Universidade de Lisboa, Lisboa Codex, Portugal

<sup>3</sup>Instituto Superior de Engenharia de Lisboa, Lisboa, Portugal

We investigate analytically the interaction between circular inclusions in smectic-C films with planar anchoring. We present a rigorous solution for an isolated circular colloid with planar anchoring and use it to calculate the interaction between two circular colloids, approximately. At large separations the interaction is quadrupolar and we show that at short range two different regimes may be observed. For weak anchoring, the interaction is always attractive in some directions, which may lead to particle contact or coalescence. However, for strong anchoring, the interaction exhibits a well defined minimum, for colloids at a particular separation and given relative orientation. The equilibrium configuration was found to depend on the anchoring strength.

Keywords: colloidal liquid crystals; topological defects

Pacs Numbers: 61.30.Dk; 61.30.Jf; 82.70.-y

#### I. INTRODUCTION

Colloidal dispersions in liquid crystals have been the subject of numerous studies in recent years [1], due to their novel type of colloidal interaction

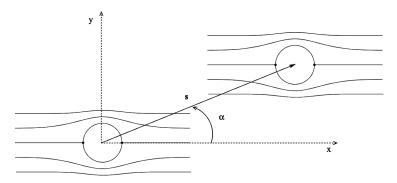
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Address correspondence to P. Patrício, Instituto Superior de Engeharia de Lisboa, Rua Cinselheiro Emido Navarro 1, P-1949-614 Lisboa, Portugal. E-mail: pedro.patricio@dem.isel.ipl.pt

[2], which is mediated by the elasticity of the host medium. Depending on the size, shape and achoring conditions of the colloidal particles, the interactions may be highly anisotropic [3,4], resulting in a variety of self-organized colloidal structures, such as linear chains [5–7], periodic lattices [8,9], anisotropic clusters [10], and cellular structures [11], stabilized, in general, by the presence of topological defects.

In this article, we investigate the interaction between circular inclusions in smectic-C films. In these two-dimensional systems, it is commonly observed a preferred planar anchoring at the colloidal boundaries [7,9,12,13], leading very often to the nucleation of one pair of defects per circular inclusion (defects attach to opposite poles of the colloid). The smectic configuration around isolated colloids has in this case quadrupolar symmetry, and a long-range quadrupolar interaction between the colloids is expected. When several colloids get together, an almost crystalline structure is observed, with well defined separations and orientations between the particles. When only two particles are present, however, experimental studies reveal a range of different equilibrium structures. In particular, Cluzeau  $et\ al.\ [12]$  report a preferred separation  $s\approx (1.33-1.35)R\ (R\$ is the radius of the circular inclusions) and orientation  $\alpha\approx 38^\circ$  (see Fig. 1) while Dolganov and Dolganov [13] report  $s\approx 1.2R$  and  $\alpha\approx 30^\circ$ .

In these articles [12,13], theoretical attempts to interpret the experimental results have been reported. In the limit of strong planar anchoring, the authors derive the equilibrium smectic-C configuration around isolated particles. The configuration when two particles are present is then obtained using the superposition approximation. Exploring these ideas, a preferred relative orientation  $\alpha = \pi/4$  was



**FIGURE 1** Smectic-C configuration around two colloids. s is the separation between the colloids and  $\alpha$  their relative orientation with respect to the far field molecular alignment of the liquid crystal.

found for large colloidal separations, and confirmed the quadrupolar interaction between the particles. However, these calculations also predict that the colloids are attracted until physical contact, at smaller relative orientations. Cluzeau *et al.* [12] suggested that the surface energy, favouring planar anchoring, would introduce a repulsive interaction between the particles at very small distances, and could explain the experimental equilibrium separations.

Here, we extend the theoretical analysis to include weak planar anchoring effects. We also use the superposition principle, but use the analytical solutions for the smectic-C configuration around isolated particles with weak planar anchoring, valid for any anchoring strength. This has led us to the prediction of two regimes at short range. For weak anchoring, the interaction is always attractive in some directions, which may lead to particle contact or coalescence. However, for stronger anchoring, the interaction exhibits a well defined minimum, for certain relative orientations and separations, preventing the coalescence of the colloids. The equilibrium configuration is not unique, but changes with the anchoring strength.

In Section II, we describe the elastic free energy for smectic-C films. In Section III, we obtain analytically the smectic-C configuration around an isolated colloid, which depends on the anchoring strength. In Section IV, we calculate the interactions between two circular colloids, and obtain quantitative results illustrating the different short-range behaviours described above. In Section V, we summarize and make a few concluding remarks.

#### II. MODEL

Smectic C free standing films are modelled by the deformations of the in-layer molecular alignment, or orientational order, represented by a two-dimensional vector field  $\mathbf{c} = (\cos \theta, \sin \theta)$ . The total free energy  $\mathcal{F}$  for a film with thickness l may be written as a sum of two terms:

$$\mathcal{F} = \iint_{\Omega} f_{\kappa} \, dx dy + \iint_{\partial \Omega} ds, \tag{1}$$

where  $\Omega$  is the domain occupied by the liquid crystal, and  $\partial\Omega$  its boundary, whenever a colloid is present. The first term accounts for the elastic distortions of the smectic-C molecular alignment. The bulk free energy density is

$$f_{\kappa} = \frac{1}{2} \left( \kappa_s (\nabla \cdot \mathbf{c})^2 + \kappa_b (\nabla \times \mathbf{c})^2 \right)$$
 (2)

where  $\kappa_s$  and  $\kappa_b$  are the splay and bend elastic constants, respectively. To simplify the theoretical analysis, we will use the one-constant approximation,  $k = \kappa_s = \kappa_b$ . In general, these constants are slightly different. Nevertheless, we expect that our analytical results and conclusions may be extended to many experimental situations. With this approximation, the elastic free energy density becomes

$$f_{\kappa} = \frac{k}{2} (\nabla \theta)^2. \tag{3}$$

The anchoring conditions of the molecular alignment at the surface of the colloids may be taken into account by the second term of the total free energy (Eq. (1)). The surface energy density is

$$f_w = \frac{w}{2} \left( 1 - (\mathbf{c} \cdot \nu)^2 \right),\tag{4}$$

where  $\nu$  is the preferred orientation at the surface of the colloids, and w the anchoring strength. Minimization of the total free energy yields

$$\nabla^2 \theta = 0 \quad \text{in } \Omega \tag{5}$$

$$\left. \frac{\partial \theta}{\partial n} \right|_{\partial \Omega} = \frac{1}{2\zeta} \sin 2(\theta_o - \theta_s), \tag{6}$$

where we have taken  $\nu=(\cos\theta_0,\sin\theta_0)$ .  $\theta_s$  is the value of  $\theta$  on the boundary  $\partial\Omega,\partial/\partial n$  is the derivative along the normal unit vector  $\boldsymbol{n}$  of  $\partial\Omega$  and  $\zeta=k/w$  is the penetration length.  $\zeta$  determines the influence of the bulk elasticity on the molecular alignment at the surface of the colloid: if  $\zeta\to 0$  (strong anchoring) the elastic deformations do not influence the orientation of the molecules at the surface. If  $\zeta\to\infty$  (weak anchoring) the orientation of the molecules at the surface is solely determined by the surrounding liquid crystal.

#### III. ISOLATED CIRCULAR INCLUSIONS

In what follows we consider circular inclusions in otherwise aligned smectic-C films (see Fig. 1). For these systems, experiments reveal that the liquid crystal molecules are parallel to the surface of the colloidal particles. This preferred surface orientation, together with the uniform alignment far from the inclusions, gives rise to the nucleation (for some given  $\zeta < \zeta_0$ ) of two topological surface defects usually called boojums at the poles of the colloidal particles.

We start by considering a single isolated inclusion. For a circular inclusion Burylov and Raikher [14] have obtained a solution for almost

any value of the anchoring strength  $\omega = R/\zeta$  (This solution is valid for  $\zeta > r_c$ , where  $r_c$  is the size of a defect [15]).

$$\theta(r,\phi) = -\arctan\left[\frac{\frac{p_{\omega}}{r^{2}}\sin2\phi}{1 - \frac{p_{\omega}}{r^{2}}\cos2\phi}\right], \tag{7}$$

where we take all lengths in units of the radius of the inclusion;  $p_{\omega}$  is a continuous function of the anchoring strength  $\omega, p_{\omega} = (2/\omega) \times \left(\sqrt{1-(\omega/2)^2}-1\right)$ . In the limit of vanishing anchoring,  $p_{\omega}(\omega \to 0) = 0$  and therefore the solution is simply the constant uniform orientation  $\theta=0$ . For strong anchoring,  $p_{\omega}(\omega \to \infty)=1$ .

#### IV. INTERACTIONS BETWEEN TWO CIRCULAR INCLUSIONS

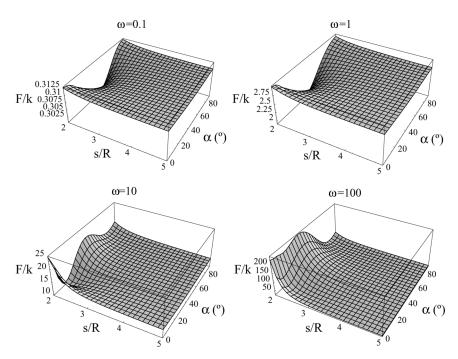
The solution of Eqs. (5) and (6) when two inclusions, at a distance  $\mathbf{s} = (s\cos\alpha, s\sin\alpha)$ , are present is not easy to obtain. However, one can study the interaction between two colloidal particles by considering that the solution is approximated by the superposition of two isolated-particle solutions,  $\theta(r,\phi) \approx \theta_{(0,0)}(r,\phi) + \theta_s(r,\phi)$ . This approximation should be valid for weak anchoring strengths  $\omega$  at any separation, and for strong anchoring when the separation between the colloidal particles is sufficiently large.

By considering that the orientational function  $\theta(r,\phi)$  is just the sum of two solutions of the laplacian equation, the free energy is simply calculated from the surface contributions

$$\mathcal{F} = \frac{1}{2} \int_{\partial \Omega} \left[ \theta \frac{\partial \theta}{\partial n} + \frac{1}{\zeta} \sin^2(\theta_o - \theta) \right] ds \tag{8}$$

where  $\partial\Omega = \partial\Omega_1 \bigcup \partial\Omega_2$ . The free energies given by Eq. (8) were calculated for different values of s,  $\alpha$  and  $\omega$ , with the help of *Mathematica*.

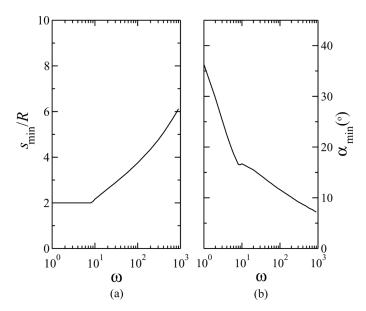
In Figure 2 we show the interaction energy profiles for several values of the anchoring strength  $\omega$ . Note that due to the symmetries of the problem in hand, we need to study the interaction for the first quadrant  $(0 < \alpha < \pi/2)$  only. In every case, we observe a complex anisotropic interaction, with attractive and repulsive orientations. For weak anchoring  $\omega < 10$  the global minimum is at a well defined angle and the particles touch each other, which may lead to coalescence of the colloids. However, for strong anchoring  $\omega > 10$  a repulsion for all orientations arises for short interparticle distances preventing the two inclusions from touching each other.



**FIGURE 2** Interaction energy profiles for several values of the anchoring strength  $\omega$ , as a function of the separation s and relative orientation  $\alpha$ . R is the radius of the circular inclusions.

In Figure 3 we show (a) the equilibrium inter-particle distance  $s_{min}$  and (b) the equilibrium relative orientation  $\alpha_{min}$  as a function of  $\omega$ . Two regimes are clearly distinguished: weak and strong anchoring. In the weak anchoring regime, the liquid crystal surrounding the colloidal particles is only slightly deformed, and the inclusions are allowed to approach each other to minimize the elastic deformations. Our calculations show that this will lead to interparticle contact,  $s_{min} = 2R$ . As the anchoring strength increases, still in the weak anchoring regime, the particles stay in contact, but there will be a re-orientation of the colloidal particles, with a large decrease of  $\alpha_{min}$ .

In the strong anchoring regime, the inclusions possess the ability to actively deform the c-director field. Moreover, the onset of this regime is characterized by the nucleation of two topological defects at the surfaces of each colloid. This leads to the development of a repulsion and  $s_{min}$  increases. Again the elastic energy minimizes



**FIGURE 3** (a) Equilibrium inter-particle distance  $s_{min}$  and (b) equilibrium relative orientation as functions of the anchoring strength  $\omega$ . R is the radius of the circular inclusions.

the overlapping effect of the two deformation coronas by decreasing  $\alpha_{min}$ . However, since defects with equal topological charge repel each other, the nucleation of the defects introduces an angular force that counteracts this reorientation and translates into a decrease in the rate at which  $\alpha_{min}$  decreases.

#### V. CONCLUSIONS

The interaction between circular inclusions in smectic-C films with planar anchoring was investigated. As pointed out by Cluzeau *et al*. [12] we found that, for large separation, the interaction is quadrupolar: the colloids attract each other along a relative orientation  $\alpha \sim \pi/4$ . As the separation between colloids decreases the orientation  $\alpha$  also decreases.

Our calculations, albeit approximate, reveal the existence of two different regimes at short range. For small anchoring strengths, the interaction is always attractive in some directions, which may lead to particle contact or coalescence. However, above a critical anchoring strength, the interaction develops a well defined minimum, for certain relative orientations and separations, preventing the coalescence of

the colloidal particles. In particular, the equilibrium configuration is not unique, changing with the anchoring strength. Although we cannot exclude the presence of other effects, the mechanism proposed in this article provides a good account of the observations reported in the experiments.

Reported in the experiments (Cluzeau et al. [12] report a preferred separation  $s \approx (1.33-1.35)$  R and orientation  $\alpha \approx 38^\circ$  while Dolganov and Dolganov [13] report  $s \approx 1.2R$  and  $\alpha \approx 30^\circ$ ). Recent experiments on ferroelectric smectic-C membranes and droplets [16] report tunable surface properties, controlled by the temperature. It would be interesting to see if the two regimes described here are observed for these more complex systems.

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