Nonlinear Effects in the TGB$_A$ Phase

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We study the nonlinear interactions in the TGB$_A$ phase by using a rotationally invariant elastic free energy. By deforming a single grain boundary so that the smectic layers undergo their rotation within a finite interval, we construct a consistent three-dimensional structure. With this structure we study the energetics and predict the ratio between the intragrain and intergrain defect spacing, and compare our results with those from linear elasticity and experiment.

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The resolution of frustration is a central theme in condensed matter physics. Competing terms in the free energy can favor incompatible spatial organizations, the resolution of which often leads to a rich phase behavior and intricate spatial patterns. The TGB phase of chiral smectics is an ideal example of the resolution of frustration [1]. While the smectic part of the free energy favors a lamellar structure, the chiral nematic part favors a uniform twist of the nematic director. These two structures are incompatible and so the layered structure must be redefined with defects to accommodate the twist. The TGB phase balances the competing interactions by forming a lattice of screw dislocations arranged in twist grain boundaries. In earlier work [2] we investigated the geometry of the TGB phase within a harmonic free energy in which topological defects interact via screened exponential potentials. However, it was argued in [3] and [4] that defects in the same grain boundary interact via power-law potentials when those nonlinearities required by rotational invariance were included in the free energy. In this letter we investigate the rotationally-invariant, nonlinear energetics of the full three-dimensional TGB$_A$ phase, where the smectic blocks between the grain boundaries are smectic-A. We find that intergrain and intragrain interactions of dislocations are both power-law, and we compute the aspect ratio between the intragrain spacing $l_d$ and the intergrain spacing $l_b$. Our computed value is of the same order of magnitude as that computed from the linear theory and measured by experiment. We will comment on the discrepancy and its source in the conclusion.

In an earlier paper [2], we considered the screw dislocation lattice in the TGB$_A$ phase near the transition to the nematic phase. In this regime the dislocation density is low, so the interaction between dislocation cores is negligible. This implies that the lattice structure is completely determined by the elastic energy cost of the smectic distortions created by the screw dislocations. We demonstrated in [2] that these distortion fields can be consistently treated within the harmonic approximation to the elastic free energy. In this approximation, we express the elastic free energy in terms of the Eulerian layer displacement field $u$ and the deviation $\delta n$ of the nematic director from the $z$-axis:

$$F = \frac{1}{2} \int d^3x \left[ B(\partial_z u)^2 + D(\nabla \cdot u + \delta n)^2 + K_1(\nabla \times \delta n)^2 + K_2(\nabla \times \delta n)^2 + K_3(\partial_z \delta n)^2 \right].$$

where $B$ and $D$ are elastic moduli and $K_1$, $K_2$ and $K_3$ are the splay, twist and bend Frank constants, respectively. This free energy is analogous to that of a type II superconductor and the topological defects in a smectic are likewise analogous to Abrikosov vortices. Employing (1) the interaction energy between two parallel screw defects is

$$\frac{F_{\text{int}}}{L} = \frac{Dd^2}{2\pi} K_0 \left( \frac{r}{\lambda} \right),$$

where $K_0$ is the modified Bessel function of order zero, $d$ is the equilibrium layer spacing of the smectic phase, and $\lambda \equiv \sqrt{K_2/D}$.

A twist grain boundary is composed of an array of equidistant parallel dislocations, separated by a distance $l_d$, which force the smectic layers to rotate. On average, the smectic layers drag the nematic director along and allow it to twist. As with screw defects and vortices, the TGB phase is the analog of the Abrikosov vortex lattice phase in superconductors [1, 2], although the lattice structure of defects in the TGB phase is significantly more complicated due to the relative rotation of dislocations in different grain boundaries. We showed in [2] that the linearized interaction energy of two grain boundaries is

$$\frac{F_{\text{int}}}{A} = \frac{Dd^2}{2\pi l_d^2} \lambda e^{-l_d/\lambda}.$$  

Thus the linear theory predicts an exponential interaction between screw dislocations both within the grain boundary and between different grain boundaries. It is simple to check that the elastic interactions of dislocations within a grain boundary are balanced by the interboundary interactions, which leads to a stable ratio $R = l_d/l_b \approx 0.95$ [3], within the experimental range of 0.74–1.08 [3, 4].
Since both smectic and cholesteric ordering are spontaneously broken symmetries, it is essential that our free energy be invariant with respect to rotations. The harmonic free energy is only invariant under infinitesimal rotations; the extra terms needed to render the free energy rotationally invariant under finite rotations lead to nonlinearities that dramatically change the energetics of even an isolated screw dislocation \([4]\). In order to investigate these effects, we work with a rotationally invariant theory that depends on the only Goldstone mode in the system, the director modes are absent. We believe that the essential interaction between the dislocation cores may not be necessary and the former has greater importance than the nonlinear compression energy completely vanishes far away from the dislocation array.

To construct the TGB phase out of individual twist grain boundaries, we confine the layer rotation of the regular LSD to a finite interval, \((-l, l)\) so that the grain boundaries are spaced \(2l\) apart in units of \(l_d/2\pi\), so \(2l = 2\pi l_d/2\pi = \pi l_d\). To do this, we replace the coordinate \(x\) in \([3]\) with an odd deformation function, \(f_l(x)\) which monotonically maps the finite interval \((-l, l)\) to \((-\infty, \infty)\). So that we recover the single grain limit, \(f_l(x) = x\) for large \(l\). The energetics of the confined LSD will impose additional constraints on \(f_l(x)\). These constraints and the optimal choice of \(f_l(x)\) will be the focus of our calculation. Since the energy depends on \(\nu\) we consider:

\[
\begin{align*}
    v_x &= -\sin \left( \frac{\alpha}{2} \right) \frac{\sin y}{\cosh f_l(x) - \cos y} f'_l(x), \\
v_y &= \sin \left( \frac{\alpha}{2} \right) \frac{\sinh f_l(x)}{\cosh f_l(x) - \cos y}, \\
v_z &= \cos \left( \frac{\alpha}{2} \right) - 1.
\end{align*}
\]

The compression elastic free energy per dislocation per unit length in the \(z\)-direction can be found using \([3]\) and the expressions \([3]\):

\[
\frac{F_{\text{compr}}}{l_z} = 2B \sin^4 \left( \frac{\alpha}{2} \right) \frac{B}{2\pi} \left( \frac{l_d}{2l} \right)^2 I_{\text{compr}}[f_l; \alpha; R],
\]

where \(I_{\text{compr}}\) is a dimensionless functional that depends on the deformation function, a short distance cutoff \(\alpha(x)\) required to excise the core from the integration domain \([3]\), and the lattice aspect ratio \(R\). It is interesting that this energy is essentially nonlinear since it depends on the fourth power of the displacement field \(u\).

On the other hand, the curvature contribution to the free energy per dislocation need not have a short-distance cutoff: as we will show, by judicious choice of the deformation \(f_l(x)\), the curvature is everywhere finite. Integrating along the grain boundary direction \(y\) we find:

\[
\frac{F_{\text{curv}}}{2KI_z} = \sin^2 \left( \frac{\alpha}{2} \right) \frac{\pi}{R} \int_0^{\pi/R} dx \left\{ \frac{1}{2} \coth f_l (f_l')^2 - 1 \right\}^2 \\
- \frac{1}{2 \sinh^2 f_l} \left[ (f_l')^2 - 1 \right] f'' + \left( \coth f_l - 1 \right) (f''')^2 \right\} \\
\equiv \sin^2 \left( \frac{\alpha}{2} \right) \frac{I_{\text{curv}}[f_l; R]}{2K}.
\]

The layer rotation angle \(\alpha\) across the grain boundary is set by the topology of the grain boundary and, therefore, does not depend on specific details of the grain boundary model. It is determined by the layer spacing \(d\) and the dislocation spacing \(l_d\) through \(\sin \left( \frac{\alpha}{2} \right) = d/2l_d\). The first term in \([3]\) describes an array of parallel screw dislocations aligned along the \(z\)-direction at \(x = 0, y = 0, \pm l_d, \pm 2l_d, \ldots\) while the second term ensures that the nonlinear compression energy completely vanishes far away from the dislocation array.

We start with the layer distortion field for a linear superposition of parallel equidistant screw dislocations (LSD) \([1, 8]\) and then use it to construct a TGB phase by compressing each of these individual boundaries along the pitch axis to force their rotation into a finite interval. We may then piece together a periodic lattice of these grain boundaries through translation and rotation in order to construct a full, three-dimensional TGB structure. We will calculate the free energy of this structure using \([3]\) to find a minimum energy solution and consequently the geometric parameter \(R \equiv l_d/l_b\) for the system. As noted in \([3]\), an important difference between the nonlinear and the harmonic theories is that in the former the interaction between the dislocation cores may not be neglected: the size and shape of the core region is another variational parameter.

We start with the layer distortion field for a linear superposition of screw dislocations (LSD) in a smectic liquid crystal. Through deformation this will become the displacement field in a single smectic-A block. Choosing the average layer normal to point along the \(z\)-axis, and expressing the displacement \(u\) and coordinates \(x\) and \(y\) in units of \(l_d/2\pi\), we have

\[
u(x, y, z) = -2 \sin \left( \frac{\alpha}{2} \right) \tan^{-1} \left[ \frac{\tanh(x/2)}{\tan(y/2)} \right] + (\cos \left( \frac{\alpha}{2} \right) - 1)z.
\]

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\equiv \sin^2 \left( \frac{\alpha}{2} \right) \frac{I_{\text{curv}}[f_l; R]}{2K}.
\]

Since a single helicoidal screw dislocation has no curvature, we need not excise the core region in the curvature
energy as long as we maintain the screw-like character of the defect. Near \( x = 0 \), \( f_1(x) \approx 0 \) is small and so we may expand the integrand \( I_{\text{curv}} \):

\[
I_{\text{curv}} = \frac{\pi ((f_1')^2 - 1)^2}{2f_1^2} - \frac{\pi ((f_1')^2 - 1)f''_1}{f_1^2} + \frac{\pi (f''_1)^2}{f_1}. \tag{11}
\]

It is evident from the above expression that the curvature energy diverges near \( x = 0 \) unless \( f_1'(0) = 1 \) and \( f''_1(0) = 0 \). Thus we have two additional constraints on \( f_1(x) \). To finish our analysis we must consider the behavior of the energy halfway between grain boundaries at \( x = \pm l \). It is there also that we must take care to match the layer normals in successive blocks so that the nematic director is well defined everywhere.

Near \( x = \pm l \), the compression energy is finite since \( v \) is well behaved. However, the curvature energy is more problematic. Since \( f_1(x) \to \infty \) as \( x \to l \), we can expand the integrand in powers of \( e^{-l} \):

\[
I_{\text{curv}} = 2\pi \sum_{n=1}^{\infty} e^{-2n f_1} [f''_1 - 2n f'_1 + n^2]. \tag{12}
\]

Since \( f_1(x) \) diverges at \( x = l \), we might consider setting the largest term to zero, i.e., the \( n = 1 \) term in (12). Implementing the boundary conditions \( f_1(0) = 0 \) and \( f_1(l) = \infty \), we find:

\[
f_1(x) = \text{sgn}(x) \log \left\{ \frac{\sinh l}{\sinh (l - |x|)} \right\}. \tag{13}
\]

It is straightforward to check that as \( x \to l \), the only nonvanishing term in (12) is the \( n = 2 \) term and that at \( x = l \)

\[
I_{\text{curv}} = \frac{2\pi}{\sinh^4 l}. \tag{14}
\]

We can embellish (13) with a power series around \( |x| = l \) so that the first and second derivatives satisfy the required conditions at the origin:

\[
f_1(x) = \text{sgn}(x) \log \left\{ \frac{\sinh l}{\sinh (l - |x|)} \right\} \tag{15}
\]

\[+ \text{sgn}(x)[a_0 + a_2(l - |x|)^2 + a_4(l - |x|)^4],\]

where \( a_0, a_2 \) and \( a_4 \) may be straightforwardly calculated [14]. These extra terms do not spoil the divergent behavior at \( x = l \) and so it would appear that we have found a low curvature deformation of the original grain boundary. However, our goal is to assemble individual blocks into the TGB structure. This is only possible if the smectic layers match at the midplane. Computing the layer tilt at the midplane, we find that \( v_x \) has a small periodic component, while \( v_y \) attains a constant value corresponding to the rotated layers:

\[
v_x(l, y) = -2\sin \left( \frac{\alpha}{2} \right) \frac{\sin y}{\sinh l}, \tag{16}
\]

\[
v_y(l, y) = \sin \left( \frac{\alpha}{2} \right). \tag{17}
\]

Since \( v_x \) reflects the periodicity of a single dislocation array and in adjacent blocks the dislocation arrays are rotated with respect to each other, the nonvanishing of \( v_x \) at the midplane makes a perfect fit impossible. This might be a consequence of the limited class of transformations considered in our model that allow the smectic layers to relax only in the direction of the pitch axis. A more general model which allows the defects in the layers to ripple in harmony with neighboring grain boundaries might allow for a nontrivial value of \( v_x \) at \( x = \pm l \). Indeed, rippling defects have been considered to explain commensurate twist grain boundary structures [11].

![FIG. 1: Dependence of \( I_{\text{curv}}(R; m) \) on the deformation parameter \( m \).](image-url)

The smectic layers could be matched perfectly if \( f_1 \) were modified to diverge faster than the form in (13). Consider a one-parameter family of deformations based on (13):

\[
f_1(x; m) = m \text{sgn}(x) \log \left\{ \frac{\sinh (l/m)}{\sinh ((l - |x|)/m)} \right\}. \tag{18}
\]

As before, these deformations can be altered so that they satisfy all the necessary boundary conditions. As \( |x| \to 1 \), both \( f''_1(x; m) \) and \([f_1(x; m)]^2 \) diverge as \( (l - |x|)^{-2} \), while \( \exp[-2f_1(x; m)] \sim (l - |x|)^{2m} \). Examining \( I_{\text{curv}} \), we see that as long as \( m \geq 2 \), the curvature remains finite in the whole region. In addition, \( v_x \sim (l - |x|)^{m-1} \), and so for \( v_x \) to vanish at \( x = \pm l \), we must have \( m > 1 \). By numerically evaluating \( I_{\text{curv}} \), we find that the curvature energy is an increasing function of \( m \), as shown in Figure 1. Thus we choose \( m = 2 \) in our calculation of the aspect ratio \( R \). We note that the isolated \( m = 1 \) deformation has a still lower energy, though it is, unfortunately, not allowed because of geometry. In future work we will reconsider the \( m = 1 \) deformation by allowing for more general variations of the dislocation lattice. The equilibrium lattice configuration is determined by minimizing the total free energy density with respect to the dislocation spacing, core size, and parameters characterizing the core shape. The total energy density includes the elastic energy we have discussed, an energetic cost from
the dislocation cores and the crucial chiral contribution which favors twist. We assume that the core energy is proportional to the cross-sectional area \(A\) of the core region, with an energy per unit area \(E\). The chiral energy gain per dislocation \(H\) is independent of the details of the dislocation arrangement, and so for given values of \(E\) and \(H\) we must minimize

\[
f_{\text{tot}} = \frac{1}{4l_d l_b} \left[ 2B \sin^4 \left( \frac{\alpha}{2} \right) \left( l_d/2\pi \right)^2 I_{\text{compr}}(A, R) \right. \\
+ 2K \sin^2 \left( \frac{\alpha}{2} \right) I_{\text{curv}}(R) + EA - H \right]
\]

with respect to \(l_d, l_b,\) and \(A\). Note that the integrals appearing in the compression and layer curvature terms do not depend explicitly on the layer rotation angle \(\alpha\). It is convenient to separate the effects of varying the dislocation density \(1/(l_d l_b) = 1/c^2\) and the lattice aspect ratio \(R \equiv l_d/l_b\). In terms of \(R\) and \(c\), \(l_d = c\sqrt{R}\) and \(l_b = c/\sqrt{R}\). For simplicity, we assume that dislocation core regions are square with sides \(a\) in units of \(c/2\pi\). Our numerical investigation of the \(I_{\text{compr}}[f_l(x), a(x); R]\) suggest that the core, though essential, does not greatly alter the energetics \([12]\). In computing \(I_{\text{compr}}\), the core size has to be reexpressed in units of \(l_d/2\pi\), so that it does not vary as \(l_d\) is varied. In units of \(l_d/2\pi\), the core size is \(a/\sqrt{R}\).

The entire minimization procedure can be formulated in terms of the dislocation density \(1/c^2\), the aspect ratio of the dislocation lattice \(R\), and the core size \(a\). Recalling that \(\sin \left( \frac{\alpha}{2} \right) = d/2l_d\) and assuming \(K/B = d^2\), we have

\[
f_{\text{tot}} = \frac{B d^4}{32\pi^2} \left[ \frac{1}{c^4 R} I_{\text{compr}}(a/\sqrt{R}, R) \right. \\
+ \frac{16\pi^2}{c^4 R} I_{\text{curv}}(R) + \frac{ca^2}{4\pi^2} - \eta \right], \tag{20}
\]

where \(\epsilon \equiv (32\pi^2/Bd^4)E\) and \(\eta \equiv (32\pi^2/Bd^4)H\). When we minimize \(f_{\text{tot}}\) with respect to \(c, a\) and \(R\), we find that the equations for the optimum values of \(R\) and \(a\) only depend on the combination \(\eta^2/\epsilon\). The optimal values of \(R\) and \(a\) computed for \(m = 2\) are given in Figure 3.

We find that the variation in the lattice aspect ratio mainly occurs for large core energies and then asymptotes rapidly to \(R \approx 0.46\) (see Figure 3). In comparison with the linearized theory which predicts that \(l_d \approx l_b\), the model studied here predicts that the repulsion between grain boundaries is stronger than the repulsion between the defects in the same grain boundary and hence \(l_b\) is roughly twice as large as \(l_d\). This may be a consequence of our locking the director to the layer normal near the defect cores \([3]\). Indeed, from Figure 3 we see that the optimal cores are rather large, about 1/6 the defect spacing. In future work we will reintroduce the director modes in a rotationally invariant fashion. This will certainly lower the overall energy and should allow the cores to shrink. As a final aside, we note that were we to choose \(m = 1\) in \([13]\) and simply ignore the director mismatch, we would find \(R \approx 0.85\). That solution will be studied in further work \([12]\).

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[1] S.R. Renn and T.C. Lubensky, Phys. Rev. A 38, 2132 (1988).
[2] I. Bluestein, R.D. Kamien, and T.C. Lubensky, Phys. Rev. E 63, 061702 (2001).
[3] M. Kléman, Points, Lines, and Walls (Wiley, New York, 1983).
[4] R.D. Kamien and T.C. Lubensky, Phys. Rev. Lett. 82, 2892 (1999).
[5] P.-G. de Gennes, Solid State Commun. 10, 753 (1972).
[6] L. Navailles, et al., Phys. Rev. Lett. 81, 4168 (1998).
[7] N. Isaert, et al., J. Phys. II (France) 4, 1501 (1994).
[8] R.D. Kamien, Appl. Math. Lett 14, 797 (2001).
[9] H. Pleiner, Liq. Cryst. 3, 249 (1998).
[10] We find that \(a_0 = \frac{1}{2}(5-5\cot h l- l/\sinh^2 l)\), \(a_2 = -\frac{1}{2}(3-3\cot h l- l/\sinh^2 l)\), and \(a_4 = \frac{1}{2}(1-\cot h l- l/\sinh^2 l)\). Note that as \(l \to \infty\) all of these coefficients vanish.
[11] T.C. Lubensky, T. Tokihiro, and S.R. Renn, Phys. Rev. Lett. 67, 89 (1991).
[12] I. Bluestein and R.D. Kamien, in preparation (2002).