Anomalous Elasticity of Polymer Cholesterics

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We show that polymer cholesterics have much longer pitches than comparable short molecule cholesterics, due to their anomalous elasticity. The pitch $P$ of a chiral mixture with concentration $c$ near the racemic (non-chiral) concentration $c^*$ diverges like $|c - c^*|^{-\nu}$ with, according to our very precise calculations [1], $\nu = 1.43 \pm 0.04$ (for short molecule cholesterics $\nu = 1$). The short molecule law is recovered for polymers of finite molecular length $\ell$ once the pitch is longer than a length that diverges like $\ell^{\gamma}$ with $\gamma = 0.67 \pm 0.01$. Our predictions could be tested by measurements of the pitch in DNA [2].

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Nature is chiral. In short-chain chiral molecules this leads to structures so numerous that we must borrow words from Greek in order to categorize them properly. The simplest of these structures is the cholesteric, in which the molecules lie perpendicular to a “pitch” axis, about which they rotate as one moves through space along it.

The standard mean field theory of cholesterics [3] predicts that the cholesteric pitch $P$ diverges according to $P \propto |c - c^*|^{-1}$, where $c$ is the concentration of some chiral additive and $c^*$ the concentration of the racemic (i.e., non-chiral) mixture.

In short molecule cholesterics, this mean field theory is exactly correct; all effects of fluctuations can be absorbed into perfectly finite renormalizations of the effective parameters of the mean-field theory. However, this is certain not to be the case for polymeric cholesterics, since it has been shown [4] that, as in smectics [5], the effects of thermal fluctuations on polymer nematics lead to infinite renormalizations of the elastic constants (for infinitely long polymers). This is known as “anomalous elasticity”.

Here we investigate the effect of anomalous elasticity on polymer cholesterics, of which DNA is a classic example [6]. We find that once the pitch predicted by mean field theory ($P_0$) is much greater than the intrinsic length $\xi_N$ beyond which the elasticity becomes anomalous, the actual pitch $P$ is given by:

$$P = \xi_N \left( \frac{P_0}{\xi_N} \right) ^{\nu} \times \mathcal{O}(1),$$

with $\nu = 1.43 \pm 0.04$. This rapidly diverging pitch reflects a sort of fluctuation suppression of the bare chirality, a phenomenon that occurs [7] in chiral membranes as well, although the effect found here is quantitatively much stronger than in chiral membranes, being an algebraically diverging correction in our case, in contrast to a logarithmic correction in membranes. For $P_0 << \xi_N$, $P = P_0$. For polymers whose interactions are primarily steric entropic (i.e., due to their meandering and bumping into each other), reference [4] showed that $\xi_N = L_P^{A/3} / a^{1/3}$, where $L_P \equiv \kappa / k_B T$ is the orientational persistence length for a single isolated polymer, (with $\kappa$ the polymer bend modulus), and $a$ is the mean polymer spacing.
In addition, we consider the effects of a finite (but long) polymer length on both polymer nematics and cholesterics. We find that these were incorrectly treated in [4]. Here, we determine the correct behavior of the renormalized Frank elastic constants as a function of polymer length $\ell$ for polymer nematics, which proves to be:

$$K_1 = \rho_0 k_B T \ell, \quad K_{2,3} = K_{2,3}^0 \left(\frac{\ell}{\ell_0}\right)^{\phi_{2,3}},$$

where $\phi_2 = 0.20 \pm 0.01$ and $\phi_3 = 0.15 \pm 0.02$, respectively. Here,

$$\ell_0 \equiv \left[\frac{K_{2,3}^0 (K_{2,3}^0)^3}{(k_B T)^2}\right]^{1/2} a^2 \xi_N^N,$$  \hspace{1cm} (3)

with $K_{2,3}^0$ the bare Frank constants and $a$ the mean polymer spacing. In addition, $\rho_0$ is the mean polymer density. For entropic steric polymers as described earlier, $\ell_0 = L_p^3/a^2$.

The value of $K_1$ was originally predicted by Meyer [8].

For polymer cholesterics, this finite polymer length cutoff leads to a crossover back to the mean field law equation $P \propto P_0$ once the actual pitch $P$ exceeds a length

$$\xi^L_{\perp}(\ell) = \xi^N_{\perp} \left(\frac{\ell}{\ell_0}\right)^{\gamma},$$

where $\gamma = 0.67 \pm 0.01$. Mean field theory [3] predicts that the pitch is proportional to $K_2$ for fixed chirality; hence, in this large pitch ($P >> \xi^L_{\perp}(\ell)$) regime, the pitch becomes $\ell$ dependent, diverging in the same way with $\ell$ as $K_2$; i.e.,

$$P(\ell) = P_0 \left(\frac{\ell}{\ell_0}\right)^{\phi_2}.$$ \hspace{1cm} (5)

This surprising prediction could be tested experimentally in DNA by using enzymes to cut the DNA while keeping other microscopic properties fixed.

The above predictions for the behavior of the pitch in a polymer cholesteric with long, but finite polymers, can be summarized for $P_0 >> \xi^N_{\perp}$ by the scaling law

$$P = P_0^{\nu} h \left[\frac{P_0}{P_0^{\nu}(\ell)}\right],$$

(6)
where \( h(x \to 0) \to (\xi_1^N)^{1-\nu} \times \mathcal{O}(1) \), \( P_c^c(\ell) = \xi_1^N \left( \frac{\xi_1^L(\ell)/\xi_1^N}{(1/\nu)} \right) \), and \( h(x \to \infty) \to (\xi_1^N x)^{1-\nu} \). Our full predictions for the pitch \( P \) are plotted in Figure 1.

The derivation of the above results begins by formulating the fully rotationally invariant, anharmonic theory of a polymer in a liquid crystal matrix. After [9], we expect that at long distances, this will also describe polymer liquid crystals in an isotropic solvent.

The free energy has four terms: 1) A term aligning the polymers with the surrounding nematic matrix. 2) The Frank free energy for distortions of this surrounding matrix. 3) The entropy of mixing for polymer ends. 4) A chiral term which generates spontaneous twist. In addition, as emphasized in past treatments [10,9,4], polymer nematics are distinguished from short-molecule nematics by a constraint relating the polymer density to the local polymer tangent field \( \vec{m} \), defined to point parallel to the local unit polymer tangent with magnitude equal to the polymer areal density in the plane perpendicular to \( \vec{m} \).

In terms of \( \vec{m} \) and the usual nematic director \( \hat{n} \), the polymer-nematic coupling is [11]

\[
F_{\text{pol}} = \frac{1}{2} \int d^3x \, I_{\mu\nu} \left( m_\mu - \rho_0 n_\mu \right) \left( m_\nu - \rho_0 n_\nu \right)
\]

where \( I_{\mu\nu} \) is diagonal and \( I_{xx} = I_{yy} \) (here and hereafter, \( \hat{z} \) is the average polymer direction). Defining the vector \( \vec{t} \) in the \( xy \)-plane via \( \vec{m} = \rho \hat{z} + \vec{t}, \, t_z = 0 \) and writing \( \vec{n} = \left[ \delta n_x, \delta n_y, \sqrt{1-(\delta \vec{n})^2} \right] \), (7) becomes

\[
F_{\text{pol}} = \frac{1}{2} \int d^3x \left\{ \frac{E}{\rho_0^2} \left( \rho - \rho_0 \sqrt{1-(\delta \vec{n})^2} \right)^2 + B(\vec{t} - \rho_0 \delta \vec{n})^2 \right\},
\]

where \( E = \frac{I_{zz}}{\rho_0^2} \) and \( B = I_{xx} \). It was shown in [4] that in the infinite polymer case the only relevant anharmonic term comes from the second order term in the expansion of \( \sqrt{1-\delta \vec{n}^2} \).

The only relevant terms [4] in the Frank free energy are

\[
F_{\hat{n}} = \frac{1}{2} \int d^3x \left\{ K_1 (\nabla \cdot \delta \vec{n})^2 + K_2 (\nabla \times \delta \vec{n})^2 + K_3 (\partial_z \delta \vec{n})^2 \right\}.
\]

The entropy of mixing of polymer heads and tails, in the limit of long polymers (where the density of heads and tails is low) should be just that of an ideal solution; hence the
contribution to the free energy is just \( k_b T \) times this entropy [9]:

\[
F = k_b T \rho_H \ln \rho_H + k_b T \rho_T \ln \rho_T.
\]

Expanding this about the equilibrium density \( \rho_0 = \rho_H \ell \) gives

\[
F_{\text{ends}} = \frac{1}{2} \int d^3 x G(\rho_H - \rho_T)^2
\]

where \( G = \frac{\partial^2 f}{\partial (\rho_H - \rho_T)^2} = k_b T(\langle \rho_H \rangle^{-1} + \langle \rho_T \rangle^{-1})/4 = k_b T \ell/2 \rho_0 \).

Now we turn to the constraint. Conservation of polymer requires \( \nabla \cdot \vec{m} = \rho_H - \rho_T \).

This becomes, to leading order

\[
\partial_z \rho + \nabla \perp \cdot \vec{t} = \rho_H - \rho_T.
\]

Note that as \( \ell \to \infty \), the quadratic term in (10) forces the constraint \( \nabla \cdot \vec{m} = 0 \) to hold everywhere, as expected.

Following [10,9,4], we introduce a phonon-like field through \( \vec{t} = \rho_0 \partial_z \vec{u} \), and add an

“incompatibility field” \( \omega \) defined by \( \delta \rho = -\rho_0 \nabla \perp \cdot \vec{u} + \omega \). Using these definitions and equation (11) to write \( \rho_H - \rho_T \) in terms of \( \vec{u} \) and \( \omega \), and integrating out \( \delta \vec{n} \), we are led to the

full free energy:

\[
F = \frac{1}{2} \int d^3 x \left\{ E \left[ \nabla \perp \cdot \vec{u} + \frac{1}{2} (\partial_z \vec{u})^2 + \omega \right]^2 + G(\partial_z \omega)^2 \right\} + F_{\vec{n}}[\partial_z \vec{u}].
\]

In the limit \( \ell \to \infty \), \( G \to \infty \), and \( \omega \) is forced to be independent of \( z \). In this situation, \( \omega \) can be eliminated by a change of variables \( \vec{u} = \vec{u}' + \nabla \perp \Omega(x,y) \), where \( \nabla \perp^2 \Omega = -\omega \), and

equation (12) reduces to precisely the model considered by [4]. It was shown there that, for nematic polymers below four dimensions (like most experimental systems), thermal

fluctuations always invalidate harmonic elastic theory at sufficiently long length scales.

Harmonic elastic theory only applies when the length scale under consideration in the

\( xy \)-plane is less than \( \xi^N_\perp \), or that in the \( z \)-direction is less than \( \xi^N_z \), where

\[
\xi^N_\perp = \left( \frac{(K_2^0)^{3/2}(K_3^0)^{1/2}}{E_0 k_b T} \right)^{1/(4-d)}, \quad \xi^N_z = \xi^N_\perp \sqrt{\frac{K_3^0}{K_2^0}}
\]
where $K_2^0$, $K_3^0$ and $E_0$ are the bare values of the elastic constants, and we’ve generalized from 3 to d dimensions. For polymers that interact primarily through steric repulsion (i.e., by bumping into each other as they meander), this becomes [12], in $d = 3$ (the physically relevant dimension [13]), $\xi_N^N = P^{4/3}/a^{1/3}$ and $\xi_N^z = P^{5/3}/a^{2/3}$.

For length scales longer than $\xi_N^N$ and $\xi_N^z$ in the appropriate directions, the equilibrium linear response and correlation functions of a nematic polymer are characterized by wavevector dependent elastic moduli $E(\vec{q})$, $K_2(\vec{q})$, and $K_3(\vec{q})$, given by

$$K_{2,3}(\vec{q}) = (q_z\xi_z^N)^{−\eta_{2,3}} f_{2,3} \left[ \frac{(q_z\xi_z^N)\zeta}{q_{\perp}\xi_{\perp}^N} \right] \propto \begin{cases} q_z^{−\eta_{2,3}}, & (q_z\xi_z^N)\zeta \gg q_{\perp}\xi_{\perp}^N \\ q_{\perp}^{−\eta_{2,3}/\zeta}, & (q_z\xi_z^N)\zeta \ll q_{\perp}\xi_{\perp}^N \end{cases}$$

$$E(\vec{q}) = (q_z\xi_z^N)^{\eta_\perp} f_E \left[ \frac{(q_z\xi_z^N)\zeta}{q_{\perp}\xi_{\perp}^N} \right] \propto \begin{cases} q_z^{\eta_\perp}, & (q_z\xi_z^N)\zeta \gg q_{\perp}\xi_{\perp}^N \\ q_{\perp}^{\eta_\perp/\zeta}, & (q_z\xi_z^N)\zeta \ll q_{\perp}\xi_{\perp}^N \end{cases}$$

(14)

where $f_{E,2,3}$ are universal scaling functions, and the universal exponents $\zeta$, $\eta_\perp$, $\eta_2$, and $\eta_3$ satisfy two exact scaling relations, which, in $d = 3$, read $\eta_\perp + \eta_2 + \eta_3 = 1$ and $\zeta = 1 + \eta_2^2 - \eta_3^2$. This behavior is what we mean by “anomalous elasticity”; the usual, constant elastic modulus behavior we will refer to as “conventional elasticity”, with no pejorative condescension intended. In reference [4], the numerical values of $\eta_2$ and $\eta_3$ were calculated from an $\epsilon = 4 - d$ expansion; the best resulting numerical values in $d = 3$ [14] are $\eta_2 = 0.31 \pm 0.02$ and $\eta_3 = 0.24 \pm 0.02$. Henceforth numerical values and error bars for all other exponents will be obtained by deriving exact expressions for them in terms of $\eta_2$ and $\eta_3$, and then using the just-quoted numerical values and error bars in those exact expressions.

When the length $\ell$ of the polymers, and hence the elastic constant $G$ in (12) are finite, this behavior ultimately gets cut off at sufficiently long length scales, beyond which the conventional (i.e., non-anomalous) elastic behavior of a conventional nematic is again recovered. To calculate this length, it is first instructive to consider the harmonic approximation, in which we can integrate out $\omega$. We thereby obtain, in Fourier space,

$$F = \frac{1}{2} \int d^3 q \left\{ \frac{G^2 q_z^4}{(Gq_z^2 + E)^2} (q_{\perp} \cdot \vec{u})^2 + K_2 q_z^2 (q_{\perp}^2 \delta_{ij} - q_i q_j) u_i u_j + K_3 (q_z^2 u)^2 \right\}. \quad (15)$$

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One can see that if $Gq_z^2 > E$ this action reduces to the infinite polymer action of [12,9]. In other words, at short distance scales, even finite polymers appear infinite. The crossover to finite length effects will occur when $Gq_z^2 = E$. This corresponds to a length scale $\xi_z^L(\ell) = \sqrt{G/E}$. At distances longer than this, the finite length effects should become important. At long distances though, finite polymers appear only as points, and so we expect the system to behave as a short molecule liquid crystal.

We would now like to see how the the breakdown of conventional elasticity theory changes this length scale. The crossover to conventional elasticity still occurs at the momentum at which $E = Gq_z^2$. This is modified from the harmonic result just derived since $E$ becomes momentum dependent itself. A priori, one might fear that $G$ acquires a nontrivial momentum dependence as well; however, this does not happen, as the following argument shows: The free energy (12) is invariant under $\omega \rightarrow \omega + h$ and $\vec{u} \rightarrow \vec{u} - \frac{1}{\partial_1} h \vec{x}_\perp$. Since the effective theory must have the same symmetry, any graph that renormalizes any quadratic term in the free energy $F$ involving $\omega$ must be absorbable into the renormalization of the coefficient of $(\nabla_\perp \cdot \vec{u} + \omega)^2$ i.e., into the renormalization of $E$. In other words, there are no graphs “left over” to renormalize $G$ which thus has only trivial renormalizations. Therefore we are able to find the fluctuation corrected crossover length $\xi_z^L$. Using $E = E_0(q_\perp \xi_\perp^N)^{\eta_\perp/\zeta}$, and converting $q_z$ to $q_\perp$ by $q_\perp \xi_\perp^N = (q_z \xi_z^N)^{\zeta}$, we have

$$\xi_z^L(\ell) = \xi_\perp^N \left[ \frac{G}{E_0(\xi_z^N)^2} \right]^{\frac{\zeta}{\eta_\perp}} = \xi_\perp^N \left( \frac{\ell}{\ell_0} \right)^\gamma = \xi_\perp^N \left[ \frac{\ell a^2}{2P^3} \right]^\gamma$$

(16)

where $\gamma = 0.67 \pm 0.01$ and $\ell_0$ is given by (3). The final equality holds only for steric interactions, although the $\ell$ dependence is universal.

Replacing $q_\perp$ with $(\xi_z^L)^{-1}$ in the expressions (14) for $K_{2,3}(q)$ yields the length-dependent $K_{2,3}$ (2) with $\phi_{2,3} = \eta_{2,3}/(1 + \eta_2 + \eta_3)$. $K_1(\ell)$ can be obtained from $K_1(\ell) = E (q_\perp = 1/\xi_z^L, q_z = 0) / (q_z^P(\ell))^2$ [8], with $q_z^P(\ell)$ obtained as described above.

To study the effect of chirality, we add the usual chiral term

$$F_{\text{chiral}} = \int d^3x (c - c^*) \hat{n} \cdot \nabla \times \hat{n}$$

$$\approx \int d^3x \left\{ (c - c^*) \left[ 1 - (\partial_z \vec{u})^2 \right] \nabla \times \partial_z \vec{u} - 2(c - c^*) \partial_x u_x \partial_y u_y \right\}$$

(17)
where $c$ is proportional to the concentration of a particular chiral species and $c^*$ is the value of $c$ at which the handedness of the phase changes from left to right. In the second, approximate, equality, we have done a few integrals by parts.

The pitch of the resulting phase is gotten by balancing the twist term with the chiral term. That is $q^P = (c - c^*)/K_2$. Hence we need the renormalized values of $c$ and $K_2$.

We first note that the chiral term does not get renormalized on length scales $<< P$, the pitch. On these length scales, we are considering the renormalization group flow of $(c - c^*)$ near $(c - c^*) \approx 0$. Fortunately, the only graph that can renormalize the chiral term is a tadpole which comes from the anharmonic term in (17) itself (the other terms in (12) have chiral symmetry). This is already proportional to $(c - c^*)$, so we need not consider the correction to the propagator coming from the quadratic term in (17) to leading order in $c - c^*$. Moreover, this correction is simply proportional to the real space mean squared director fluctuation $\langle \delta \vec{n}^2(\vec{x}) \rangle = \langle |\partial_z \vec{u}(\vec{x})|^2 \rangle$ which is finite in $d = 3$. Thus $(c - c^*)$ acquires no divergent renormalizations. This is in contrast to [7], where the corrections were divergent because, in membranes, $d = 2$ and $\langle \delta \vec{n}^2(\vec{x}) \rangle$ does diverge in that case, leading to a non-trivial renormalization of $(c - c^*)$.

Finally, we calculate the pitch of polymer cholesterics, given the chirality $(c - c^*)$. In mean field theory the pitch is found by balancing the twist term $K_2(\nabla_\perp \times \partial_z \vec{u})^2$ with the source term $(c - c^*)\nabla_\perp \times \partial_z \vec{u}$. The resulting pitch is the inverse of the momentum at which $\delta n(\vec{q}) = \mathcal{O}(1)$, or, in other words, $q_z u(\vec{q}) = \mathcal{O}(1)$. This gives the familiar mean field result $q_{\perp}^P \sim (c - c^*)/K_2^0$ for the wavenumber $q_{\perp}^P \equiv 2\pi/P$ of the twisted structure, where $P$ is its pitch. Since the source term for twist is not renormalized and the effect of fluctuations can, by construction, be absorbed into effective, momentum-dependent Frank constants, we simply replace $K_2^0$ with $K_2(\vec{q})$. Now balancing the twist and source in the effective theory, we find

$$K_2(q_{\perp})q_{\perp}^2[q_z u(\vec{q})]^2 = (c - c^*)q_{\perp}[q_z u(\vec{q})].$$ (18)
Using $K_2(q_\perp, q_z = 0) = K_0^2(q_\perp \xi^N_\perp)^{-\eta_2/\zeta} \times O(1)$, $q_z u = O(1)$, and $q_\perp^P \equiv 2\pi/P$ we find equation (1) of the introduction, with $\nu = \frac{\zeta}{\zeta - \eta_2} = \frac{2 + \eta_2 - \eta_3}{2 - \eta_2 - \eta_3}$, where we have used the scaling relation for $\zeta$. Once the pitch is greater than $\xi^L_\perp(\ell)$, it obeys the conventional, short molecule cholesteric law $P \propto P_0$ because the elasticity is again conventional. However, $P$ now depends on the polymer length since $K_2$ does (as in (2)). Using (18) again to determine the pitch yields the $\ell$ dependent pitch given in equation (5).

DNA provides an excellent experimental example of a polymer cholesteric. The DNA sample in [15] had an orientational persistence length of $L_P = 600 \text{Å}$ and mean polymer spacings $a = 35 \text{Å}$. The typical length of their DNA molecules was 4 mm! Using our estimates for the crossover lengths $\xi^N_\perp$ and $\xi^L_\perp$, we find that $\xi^N_\perp \approx 1500 \text{Å}$ and $\xi^L_\perp \approx 4 \times 10^4 \text{Å}$, which leaves a respectable range of length scales (1.4 decades) to observe the scaling behavior in (1). While the nonlinear effect is due to long molecules, the chirality of the system could be changed by adding short molecules into the mix, if they could be dissolved. By cutting up the DNA with enzymes, it should be possible to move from the nonlinear to linear regime by shortening $\xi^L_\perp$, and thereby observe the $\ell$ dependence (5) of the pitch.

The fact that DNA can have a rather long pitch of $\sim 10^4 \text{Å}$ [2] could be partially explained by our result. Taking the above numbers, and a “bare” cholesteric pitch for DNA of 7000Å, equation (1) predicts that the actual pitch will be doubled from this bare value.

In some cases [16] DNA shows hexatic order in the plane orthogonal to $\hat{n}$. This “N+6” [17] structure leads to an additional chiral coupling [18], which might tremendously increase the pitch. This may explain why no twist is observed in these systems [2]. This is currently under investigation [19].

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Figure Captions

Fig. 1. Pitch $P$ as a function of the bare pitch $P_0$. For actual pitches $P$ below $\xi^N_\perp$, harmonic elasticity theory applies. For larger pitches, anharmonic effects become important, leading to a nonlinear dependence of $P$ on $P_0$. For pitches longer that $\xi^L_\perp$, the finite length of the polymers cuts off the anharmonic effects, and we return to a linear relation again, with a new slope.
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