Thermal Fluctuations of Elastic Filaments With Spontaneous Curvature and Torsion

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Abstract

We study the effects of thermal fluctuations on thin elastic filaments with spontaneous curvature and torsion. We derive analytical expressions for the orientational correlation functions and for the persistence length of helices, and find that this length varies non-monotonically with the strength of thermal fluctuations. In the weak fluctuation regime, the persistence length of a spontaneously twisted helix has three resonance peaks as function of the twist rate. In the limit of strong fluctuations, all memory of the helical shape is lost.

Recent advances in the art of micro–manipulation of molecules led to many experimental studies of the elasticity of biomolecules such as DNA, chromatin, proteins, and rodlike protein assemblies. The tacit assumption behind many of the theories is that the elasticity of these biomolecules is of entropic origin and, consequently, they are modeled as random walks. An alternative approach to the modeling of such systems is based on the assumption that the origin of elasticity is energy rather than entropy – there exists a lowest energy equilibrium configuration with associated spontaneous curvature, deviations from which give rise to restoring forces. While such an approach is a straightforward extension of the usual theory of elasticity of thin rods, the description of arbitrary spontaneous curvature and twist involves rather complicated differential geometry and most DNA–related studies considered only fluctuations around the straight rod configuration (see, however,
references [18] and [19]). Following recent studies on the elasticity and stability of thin rods with arbitrary spontaneous curvature and torsion\(^2\), in this work we investigate the effect of thermal fluctuations on the statistical properties of such filaments. We derive the differential equations for the orientational correlation functions of the vectors pointing along the principal axes of the filament, and use them to calculate the correlators and the effective persistence length of an untwisted helix. Analytical expressions for the persistence length of a spontaneously twisted helix are obtained and it is found that this length varies non-monotonically with the amplitude of fluctuations and exhibits resonant–like dependence on the rate of twist. We would like to emphasize that although the present work is motivated by recent studies of biomolecules, its aim is to construct a theoretical framework for the description of fluctuating string–like objects that goes beyond current models of polymer physics, and we do not attempt here to model particular experiments involving single–molecule manipulation.

A filament of small but finite and, in general, non–circular cross–section, is modeled as an inextensible but deformable physical line parametrized by a contour length \( s \) (\( 0 \leq s \leq L \) where \( L \) is the length of the filament). To each point \( s \) one attaches a triad of unit vectors \( \mathbf{t}(s) \) whose component \( t_3 \) is the tangent vector to the curve at \( s \), and the vectors \( t_1(s) \) and \( t_2(s) \) are directed along the axes of symmetry of the cross–section. Note that \( \mathbf{t}(s) \), together with the inextensibility condition \( d\mathbf{x}/ds = t_3 \), gives a complete description of the space curve \( \mathbf{x}(s) \), as well as of the twist of the cross–section about this curve. The rotation of the triad \( \mathbf{t} \) as one moves along the curve is determined by the generalized Frenet equations

\[
\frac{d\mathbf{t}_i(s)}{ds} = -\sum_j \Omega_{ij}(s)\mathbf{t}_j(s) \quad (1)
\]

Here \( \Omega_{ij}(s) = \sum_k \varepsilon_{ijk} \omega_k(s) \), \( \varepsilon_{ijk} \) is the antisymmetric tensor and \( \{\omega_k\} \) are generalized torsions.

The theory of elasticity of thin rods\(^2\) is based on the notion that there exists a stress–free reference configuration defined by the set of spontaneous curvatures and torsions \( \{\omega_0k\} \). The set \( \{\omega_0k\} \) together with Eqs. (1) completely determines the equilibrium shape of the
 filament. Neglecting excluded–volume effects and other non–elastic interactions, the elastic energy associated with some actual configuration \( \{ \omega_k \} \) of the filament is a quadratic form in the deviations \( \delta \omega_k(s) = \omega_k(s) - \omega_{0k}(s) \)

\[
U_{el} \{ \delta \omega_k \} = \frac{kT}{2} \int_0^L ds \sum_k a_k \delta \omega_k^2 
\]

where the bare persistence lengths \( a_1 = E_1 I_1/kT, a_2 = E_1 I_2/kT \) and \( a_3 = E_2(I_1 + I_2)/kT \) (\( T \) is the temperature and \( k \) is the Boltzmann constant) are expressed in terms of the elastic moduli \( E_i \) and the moments of inertia \( I_i \) about the axes of symmetry of the cross–section.\(^5\)

The only limitation on the validity of Eq. (2) is that deformations are small on microscopic length scales, of order of the thickness of the filament. The elastic energy \( U_{el} \{ \delta \omega_k \} \) determines the statistical weight of the configuration \( \{ \omega_k \} \). Calculating the corresponding Gaussian path integrals we find that \( \langle \delta \omega_i(s) \rangle = 0 \) and

\[
\langle \delta \omega_i(s) \delta \omega_j(s') \rangle = a_i^{-1} \delta_{ij} \delta(s - s') 
\]

We conclude that fluctuations of generalized torsions at two different points along the filament contour are uncorrelated, and that the amplitude of fluctuations is inversely proportional to the corresponding bare persistence length.

The statistical properties of fluctuating filaments are determined by the orientational correlation functions, \( \langle t_i(s) t_j(s') \rangle \). In order to derive a differential equation for this correlators, we calculate the variation of \( t_i(s) \) under the substitution \( s \rightarrow s + \Delta s \). Integrating Eq. (4) yields in matrix notation (for small \( \Delta s \)):

\[
t(s + \Delta s) = \left\{ 1 - \int_s^{s+\Delta s} ds_1 \Omega(s_1) + \frac{1}{2} \int_s^{s+\Delta s} ds_1 \int_s^{s+\Delta s} ds_2 \Omega(s_1)\Omega(s_2) + \frac{1}{2} \int_s^{s+\Delta s} ds_1 \int_s^{s_1} ds_2 [\Omega(s_1)\Omega(s_2) - \Omega(s_2)\Omega(s_1)] + \ldots \right\} t(s)
\]

where the last term appears because of noncommutativity of matrices \( \Omega(s_1) \) and \( \Omega(s_2) \) for different \( s_1 \) and \( s_2 \). We multiply the above expression by \( t_j(s') \), average the result and note that for \( s + \Delta s > s > s' \) the fluctuations \( \delta \omega_i(s_1) \) and \( \delta \omega_j(s_2) \) are uncorrelated with the fluctuations of \( t_i(s) \) and \( t_j(s') \). This implies that averages of products of \( \Omega \)'s and \( t \)'s
factorize into products of the averages of Ω's and those of t's. Since the averages of the
terms in the square brackets in Eq. (5) depend only on |s_1 - s_2| and their difference vanishes,
in the limit Δs → 0 this yields

$$\frac{\partial}{\partial s} \langle t_i(s) t_j(s') \rangle = - \sum_k \Lambda_{ik}(s) \langle t_k(s) t_j(s') \rangle$$

(5)

where

$$\Lambda_{ik} = \gamma_i \delta_{ik} + \sum_l \varepsilon_{ilk} \omega_{0l}$$

with \( \gamma_i = \sum_k \frac{1}{2a_k} - \frac{1}{2a_i} \)

The above equations describe the fluctuations of filaments of arbitrary shape and flexibility
and in the following this general formalism is applied to helical filaments.

Consider a helix without spontaneous twist, such that the generalized spontaneous cur-
vatures and torsions \( \{\omega_{0k}\} \) are independent of position s along the contour. In order to
visualize the stress–free configuration of such a filament, it is convenient to introduce the
conventional Frenet triad of unit vectors which consists of the tangent, normal and binormal
to the space curve spanned by the centerline, supplemented by a constant twist angle \( \alpha_0 \)
which describes the rotation of the cross–section about this line. The relation between the
two triads is given by \( \omega_{0i} = \kappa_0 \cos \alpha_0, \omega_{02} = \kappa_0 \sin \alpha_0 \) and \( \omega_{03} = \tau_0 \), where \( \kappa_0 \) and \( \tau_0 \) are
the constant curvature and torsion of the space curve. The rate of rotation of the centerline
about the long axis of the helix is \( \omega_0 = (\kappa_0^2 + \tau_0^2)^{1/2} \), the helical pitch is \( 2\pi\tau_0/\omega_0^2 \) and the
radius is \( 2\pi\kappa_0/\omega_0^2 \). For constant \( \{\kappa_0, \tau_0, \alpha_0\} \), \( \Lambda \) is a constant matrix and \( \langle t_i(s) t_j(s') \rangle \) is given
by \( ij \)–th element of the matrix \( \exp[-\Lambda (s - s')] \). The eigenvalues of the matrix \( \Lambda \) can be
obtained analytically by solving for the roots of a characteristic cubic polynomial but the
resulting expressions are cumbersome and will be presented in a longer report. Here we
discuss only two limiting cases:

Weak fluctuations, \( \sum_i \gamma_i \ll \omega_0 \) In this case

$$\langle t_i(s) t_i(0) \rangle = \left( \frac{\omega_{0i}^2}{\omega_0^2} \right) e^{-s/l} + \left( 1 - \frac{\omega_{0i}^2}{\omega_0^2} \right) \cos(\omega_0 s) e^{-s/l_\phi}$$

(7)

with the decay lengths \( l \) and \( l_\phi \) defined by \( l^{-1} = \sum_k \gamma_k \omega_{0k}^2/\omega_0^2 \) and \( l_\phi^{-1} = \sum_k a_k^{-1} \omega_{0k}^2/\omega_0^2 \).

The physical meaning of this correlator becomes clear by switching off thermal fluctuations
\(\gamma_k = 0\). The first term on the rhs of this equation expresses the fact that the projection of any vector \(t_i\) of the triad on the symmetry axis of the helix is constant, with magnitude \(\omega_0^i/\omega_0\). The projections on the plane normal to this axis with magnitudes \((1 - \omega_0^2/\omega_0^2)^{1/2}\), rotate with angular rate \(\omega_0\). In the presence of weak fluctuations, the axis of symmetry of the helix becomes a random walk and the loss of correlations of its projections along the axes of the triad is described by the factor \(\exp(-s/l)\). In the second term of Eq. (7), \(\exp(-s/2l)\) describes the loss of correlations of the orientation of the plane normal to the axis of the helix.

The angular persistence length \(l_\phi\) results from averaging over the random angle of rotation \((\phi)\) with respect to the axis of the helix, \(\langle \cos[\omega_0 s + \phi(s) - \phi(0)]\rangle = \cos(\omega_0 s) \exp(-s/2l_\phi)\), with \(\langle [\phi(s) - \phi(0)]^2\rangle = s/l_\phi\).

Strong fluctuations, \(\sum_i \gamma_i \gg \omega_0\). In this limit
\[
\langle t_i(s)t_j(0) \rangle = e^{-\gamma_i(s-s')}\delta_{ij}
\]
i.e., the correlators depend only on the bare persistence lengths and memory about the orientation of the vector \(t_i\) decays over contour distance \(\gamma_i^{-1}\). Strong fluctuations destroy all phase coherence and all correlations between different vectors of the triad and lead to complete “melting” of the helix.

We now proceed to calculate the effective persistence length \(l_p\) which controls both the thermal fluctuations of a filament and its elastic response to external forces. It is defined as the ratio of the rms end–to–end separation \(\langle R^2 \rangle\) and the contour length of the filament \(L\), in the limit \(L \to \infty\). The end–to–end vector is defined as \(R = \int_0^L t_3(s) ds\) and thus
\[
l_p = \lim_{L \to \infty} \frac{2}{L} \int_0^L ds \int_0^s ds' \langle t_3(s) t_3(s') \rangle
\]
A simple calculation yields a result valid for arbitrarily strong fluctuations:
\[
l_p = 2\kappa_0^2 \frac{\tau_0^2 + \gamma_1 \gamma_2}{\gamma_1 \cos^2 \alpha_0 + \gamma_2 \sin^2 \alpha_0 + \tau_0^2 \gamma_3 + \gamma_1 \gamma_2 \gamma_3}
\]
For non–vanishing curvature and torsion, this expression diverges in the weak fluctuation limit \(\gamma_i \to 0\) and the shape of the filament is nearly unaffected by fluctuations. Non–monotonic behavior is observed for “plate–like” helices, with large radius to pitch ratio,
For $\gamma_i \to 0$, the effective persistence length approaches zero (recall that $L \to \infty$ in Eq. (4)). Thermal fluctuations expand the helix by releasing stored length and initially increase the persistence length. Eventually, in the limit of strong fluctuations, the persistence length vanishes again (as $\gamma_3^{-1}$) because of complete randomization of the filament. The sensitivity to the constant angle of twist $\alpha_0$ increases with radius to pitch ratio. In the opposite limit of “rod–like” helices $\kappa_0 \to 0$, the effective persistence length approaches $2/\gamma_3$ and becomes a function of $a_1$ and $a_2$ only. Indeed, since straight inextensible rods do not have stored length, their end–to–end distance and persistence length are determined by random bending and torsional fluctuations only, and are independent of twist.

The preceding analysis can be extended to fluctuating filaments with twisted stress–free states characterized by constant curvature $\kappa_0$, torsion $\tau_0$ and rate of twist of the cross–section about the centerline, $d\alpha_0/ds$. The relation between generalized torsions $\{\omega_{0k}(s)\}$ and Frenet parameters $\{\kappa_0, \tau_0, \alpha_0(s)\}$ is given by $\omega_{01} = \kappa_0 \cos \alpha_0$, $\omega_{02} = \kappa_0 \sin \alpha_0$ and $\omega_{03} = \tau_0 + d\alpha_0/ds$. The calculation of the persistence length involves the solution of Eq. (3) with periodic coefficients. Details of the analytical solution will be given elsewhere. For filaments with circular cross–section $a_1 = a_2$, the persistence length is independent of twist. In Fig. 1 we present a plot of the persistence length given in units of the helical pitch $l^* = l^2 \omega_0^2 / 2\pi \tau_0$, on the dimensionless rate of twist $w^* = 2\omega_0^{-1} d\alpha_0/ds$, for a “plate–like” helix with large radius to pitch ratio $\kappa_0/\tau_0$ and ribbon–like cross–section, $a_1 \ll a_2$. Curve 1 corresponds to the case of weak fluctuations, $\gamma_i \ll \omega_0$. Throughout most of the range, the persistence length is independent of the rate of twist but a sharp peak appears at $d\alpha_0/ds = 0$ (see insert), accompanied by two smaller peaks at $d\alpha_0/ds = \pm \omega_0/2$. Note that while in the limit of vanishing pitch, a ribbon–like untwisted helix degenerates into a normal ring, the cross–section of a twisted helix with $d\alpha_0/ds = \pm \omega_0/2$ rotates by $\pm \pi$ and the helix degenerates into a Möbius ring. As the amplitude of fluctuations increases, the central peak transforms into a narrow dip and the two Möbius peaks become broad minima (curve 2). Further increase of fluctuations leads to the disappearance of the Möbius dips and the central dip becomes broad and shallow (curve 3). Finally when $\gamma_i \gg \omega_0$, all dependence of the persistence length
on the spontaneous twist disappears (curve 4). Note that, as expected from the discussion following Eq. (10), the persistence length depends non-monotonically on the amplitude of thermal fluctuations.

In order to understand the origin of the Möbius resonances we note that while the effective persistence length is a property of the space curve given by the Frenet triad, the microscopic Brownian motion of the filament arises as the result of random forces that act on its cross-section and therefore are given in the frame associated with the principal axes of the filament. Since the two frames are related by a rotation of the cross-section by an angle \( \alpha_0(s) \), the random force in the Frenet frame is modulated by linear combination of \( \sin \alpha_0(s) \) and \( \cos \alpha_0(s) \). This gives a deterministic contribution to the persistence length which, to lowest order in the force, is proportional to the mean square amplitude of the random force and therefore varies sinusoidally with \( \pm 2\alpha_0(s) \). The observed resonances occur whenever the natural rate of rotation of the helix \( \omega_0 \) coincides with the rate of variation of this deterministic contribution of the random force, \( \pm 2d\alpha_0/ds \).

In summary, we presented a statistical mechanical description of thermally fluctuating elastic filaments of arbitrary shape and flexibility. We would like to emphasize that the only limitation on the magnitude of fluctuations is that they are small on microscopic length scales, and that our model describes arbitrary deviations of a long filament from its equilibrium shape. The general formalism was applied to helical filaments with and without twist. Strong thermal fluctuations lead to melting of the helix, accompanied by a complete loss of helical correlations. In general, the persistence length is a non-monotonic function of the amplitude of thermal fluctuations. Although through most of its range twist has a minor effect on the persistence length, resonant peaks and dips are observed whenever the rate of twist approaches zero or equals in absolute magnitude to half the rate of rotation of the helix.

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Figure captions

Plot of the dimensionless persistence length $l^*$ as a function of the dimensionless rate of twist $w^*$ for a helical filament with spontaneous curvature $\kappa_0 = 1$, and torsion $\tau_0 = 0.01$ (in arbitrary units). The different curves correspond to different bare persistence lengths: (1) $a_1 = 100$, $a_2 = a_3 = 5000$, (2) $a_1 = 1$, $a_2 = a_3 = 100$, (3) $a_1 = 0.1$, $a_2 = a_3 = 10$, (4) $a_1 = 0.01$, $a_2 = a_3 = 10$. A magnified view of the region of small twist rates is shown in the insert.
