Thermal Fluctuations of Elastic Filaments with Spontaneous Curvature and Torsion

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(Received 4 May 2000)

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 nonmonotonically with the strength of thermal fluctuations. In the weak fluctuation regime, the persistence length of a spontaneously twisted helix has three resonance peaks as a function of the twist rate. In the limit of strong fluctuations, all memory of the helical shape is lost.

PACS numbers: 87.15.Ya, 05.40.–a

Recent advances in the art of micromanipulation of molecules led to many experimental studies of the elasticity of biomolecules such as DNA [1–3], chromatin [4], proteins [5], and rodlike protein assemblies [6–8]. The tacit assumption behind many of the theories is that the elasticity of these biomolecules is of entropic origin [9–11] and, consequently, they are modeled as random walks [12]. An alternative approach to the modeling of such systems is based on the assumption that the origin of elasticity is energetic rather than entropic—there exists a lowest energy equilibrium configuration with associated spontaneous curvature [13], 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 [14], the description of arbitrary spontaneous curvature and twist involves rather complicated differential geometry and most DNA-related studies considered fluctuations only around the straight rod configuration [15] (see, however, Refs. [16] and [17]). Following recent studies on the elasticity and stability of thin rods with arbitrary spontaneous curvature and torsion [18], 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 derived, and it is found that this length varies nonmonotonically with the amplitude of fluctuations and exhibits resonantlike dependence on the rate of twist. We 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 stringlike objects that goes beyond current models of polymer physics, rather than to model particular experiments involving single-molecule manipulation.

A filament of small but finite and, in general, noncircular 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 $\mathbf{t}_3$ is the tangent vector to the curve at s, and the vectors $\mathbf{t}_1(s)$ and $\mathbf{t}_2(s)$ are directed along the axes of symmetry of the cross section. Note that $\mathbf{t}(s)$, together with the inextensibility condition $d\mathbf{x}/ds = \mathbf{t}_3$, gives a complete description of the space curve $\mathbf{x}(s)$, as well as of the rotation 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

$$\frac{d\mathbf{t}_i(s)}{ds} = -\sum_j \Omega_{ij}(s)\mathbf{t}_j(s). \tag{1}$$

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

The theory of elasticity of thin rods [14] 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 Eq. (1) completely determines the equilibrium shape of the filament. Neglecting excluded-volume effects and other nonelastic interactions, the elastic energy associated with some actual configuration $\{\omega_k\}$ of the filament is a quadratic form in the deviations $\delta\omega_k(s) = \omega_k(s) - \omega_{0k}(s)$,

$$U_{\mathrm{el}}\{\delta\omega_k\} = \frac{kT}{2}\int_0^L ds \sum_k a_k \delta\omega_k^2, \tag{2}$$

with T the temperature and k the Boltzmann constant. The bare persistence lengths a_1, a_2, and a_3 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 (the precise form of this dependence depends on whether isotropic or anisotropic elasticity is assumed [18]) and are inversely proportional to T. The only limitation on the validity of Eq. (2) is that deformations should be small on microscopic length scales, of order of the thickness of the filament. The elastic energy $U_{\mathrm{el}}\{\delta\omega_k\}$ 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

$$\langle\delta\omega_i(s)\delta\omega_j(s')\rangle = a_i^{-1}\delta_{ij}\delta(s - s'). \tag{3}$$

We conclude that the fluctuations of the generalized curvatures and torsions at two different points along the filament

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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, $\langle \mathbf{t}_i(s) \cdot \mathbf{t}_j(s') \rangle$, where the dot denotes scalar product with respect to the x, y, and z components (t_{ix}, t_{iy}, and t_{iz}) of the vector $\mathbf{t}_i$. In order to derive a differential equation for these correlators, we calculate the variation of $\mathbf{t}_i(s)$ under the substitution $s \rightarrow s + \Delta s$. Integrating Eq. (1) yields in matrix notation (for small Δs)

$$\mathbf{t}(s + \Delta s) = \Big\{ \mathbf{1} - \int_s^{s+\Delta s} ds_1 \, \mathbf{\Omega}(s_1) + \frac{1}{2} \int_s^{s+\Delta s} ds_1 \int_s^{s+\Delta s} ds_2 \, \mathbf{\Omega}(s_1) \mathbf{\Omega}(s_2) + \frac{1}{2} \int_s^{s+\Delta s} ds_1 \int_s^{s_1} ds_2 [\mathbf{\Omega}(s_1)\mathbf{\Omega}(s_2) - \mathbf{\Omega}(s_2)\mathbf{\Omega}(s_1)] + \cdots \Big\} \mathbf{t}(s) , \tag{4}$$

where the last term appears because of noncommutativity of matrices $\mathbf{\Omega}(s_1)$ and $\mathbf{\Omega}(s_2)$ for different s_1 and s_2. We multiply the above expression by $\mathbf{t}_j(s')$, 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 $\mathbf{t}_i(s)$ and $\mathbf{t}_j(s')$. This implies that averages of products of $\mathbf{\Omega}$'s and $\mathbf{t}$'s factorize into products of the averages of $\mathbf{\Omega}$'s and those of $\mathbf{t}$'s. Since the averages of the terms in the square brackets depend only on $|s_1 - s_2|$ and their difference vanishes, in the limit $\Delta s \rightarrow 0$ this yields (for $s - s' > 0$)

$$\frac{\partial}{\partial s} \langle \mathbf{t}_i(s) \cdot \mathbf{t}_j(s') \rangle = -\sum_k \Lambda_{ik}(s) \langle \mathbf{t}_k(s) \cdot \mathbf{t}_j(s') \rangle , \tag{5}$$

where

$$\Lambda_{ik} = \gamma_i \delta_{ik} + \sum_l \varepsilon_{ikl} \omega_{0l} \quad \text{with } \gamma_i = \sum_k \frac{1}{2a_k} - \frac{1}{2a_i} . \tag{6}$$

Together with the initial conditions, $\langle \mathbf{t}_i(s) \cdot \mathbf{t}_j(s) \rangle = \delta_{ij}$, 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 curvatures 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 angle α_0 which describes the rotation of the cross section about this line. The relation between the two triads is given by $\omega_{01} = \kappa_0 \cos\alpha_0$, $\omega_{02} = \kappa_0 \sin\alpha_0$, and $\omega_{03} = \tau_0$, where κ_0 and τ_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 κ_0/ω_0^2. For constant $\{\kappa_0, \tau_0, \alpha_0\}$, $\mathbf{\Lambda}$ is a constant matrix and $\langle \mathbf{t}_i(s) \cdot \mathbf{t}_j(s') \rangle$ is given by the ijth element of the matrix $\exp[-\mathbf{\Lambda}(s - s')]$. The eigenvalues of the matrix $\mathbf{\Lambda}$ 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 paper. Here we discuss only two limiting cases.

Weak fluctuations, $\sum_i \gamma_i \ll \omega_0$.—In this case

$$\langle \mathbf{t}_i(s) \cdot \mathbf{t}_i(0) \rangle = (\omega_{0i}^2/\omega_0^2) e^{-s/l} + (1 - \omega_{0i}^2/\omega_0^2) \cos(\omega_0 s) e^{-s/2l - s/2l_\phi} \tag{7}$$

with the decay lengths l and l_ϕ 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 right-hand side of this equation expresses the fact that the projection of any vector $\mathbf{t}_i$ of the triad on the symmetry axis of the helix is constant, with magnitude ω_{0i}/ω_0. The projections on the plane normal to this axis with magnitudes $(1 - \omega_{0i}^2/\omega_0^2)^{1/2}$ rotate with angular rate ω_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_ϕ results from averaging over the random angle of rotation with respect to the axis of the helix ϕ, $\langle \cos[\omega_0 s + \phi(s) - \phi(0)] \rangle = \cos(\omega_0 s) \exp(-s/2l_\phi)$, with $\langle [\phi(s) - \phi(0)]^2 \rangle = s/l_\phi$.

Strong fluctuations, $\sum_i \gamma_i \gg \omega_0$.—In this limit

$$\langle \mathbf{t}_i(s) \cdot \mathbf{t}_j(0) \rangle = e^{-\gamma_i s} \delta_{ij} ; \tag{8}$$

i.e., the correlators depend only on the bare persistence lengths and memory about the orientation of the vector $\mathbf{t}_i$ decays over contour distance γ_i^{-1}. Strong fluctuations destroy all phase coherence and all correlations between different vectors of the triad and lead to complete melting of the helical structure of the filament.

We now proceed to calculate the effective persistence length l_p which controls both the thermal fluctuations of a filament about its equilibrium configuration and its elastic response to external forces. In the limit $L \rightarrow \infty$, it is defined as the ratio of the rms end-to-end separation $\langle R^2 \rangle$

and twice the contour length of the filament L. The end-to-end vector is defined as $\mathbf{R} = \int_0^L \mathbf{t}_3(s)\, ds$ and thus

$$l_p = \lim_{L\to\infty} \frac{1}{L} \int_0^L ds \int_0^s ds' \langle \mathbf{t}_3(s) \cdot \mathbf{t}_3(s') \rangle . \quad (9)$$

A simple calculation yields a result valid for arbitrarily strong fluctuations:

$$l_p = \frac{\tau_0^2 + \gamma_1\gamma_2}{\kappa_0^2(\gamma_1 \cos^2\alpha_0 + \gamma_2 \sin^2\alpha_0) + \tau_0^2\gamma_3 + \gamma_1\gamma_2\gamma_3} . \quad (10)$$

For nonvanishing 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. Nonmonotonic behavior is observed for "platelike" helices, with large radius to pitch ratio, $\kappa_0/2\pi\tau_0$. For $\gamma_i \to 0$, the effective persistence length approaches zero [see Eq. (9)]. 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 γ_3^{-1}) because of the complete randomization of the filament. The sensitivity to the constant angle of twist α_0 increases with radius to pitch ratio.

In the opposite limit of "rodlike" 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 lengths 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 κ_0, torsion τ_0, and rate of rotation of the cross section about the centerline, $d\alpha_0/ds$. The relation between the generalized torsions $\{\omega_{0k}(s)\}$ and the 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. (5) with periodic coefficients. Details of the analytical solution will be given elsewhere [19]. 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 dimensionless persistence length $l^* = l_p\omega_0^2/\pi\tau_0$, on the dimensionless rate of twist $w^* = 2\omega_0^{-1} d\alpha_0/ds$, for a platelike helix with large radius to pitch ratio $\kappa_0/2\pi\tau_0$ and ribbonlike 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 inset), accompanied by two smaller peaks at $d\alpha_0/ds = \pm\omega_0/2$. Note that, while in the limit of vanishing pitch, a ribbonlike 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). A 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 nonmonotonically on the amplitude of thermal fluctuations.

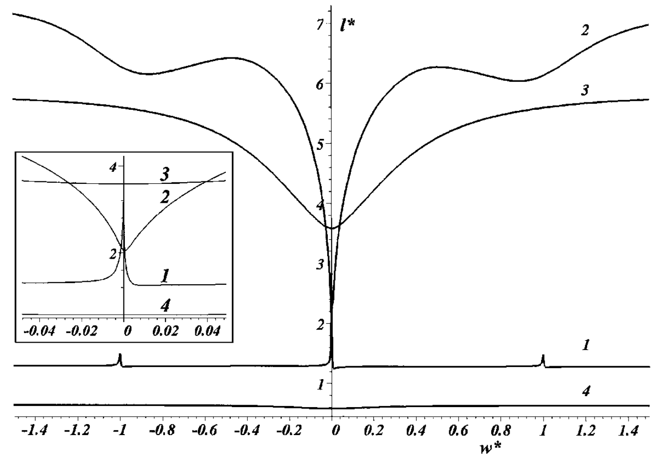

FIG. 1. Plot of the dimensionless persistence length l^* as a function of the dimensionless rate of twist w^* for a helical filament with spontaneous curvature $\kappa_0 = 1$, and torsion $\tau_0 = 0.01$ (in arbitrary units). The different curves correspond to different bare persistence lengths: (1) $a_1 = 100$, $a_2 = a_3 = 5000$, (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 inset.

In order to understand the origin of the Möbius resonances we note that the effective persistence length is a property of the space curve given by the Frenet triad. On the other hand, 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 sinosoidally with $\pm 2\alpha_0(s)$. The observed resonances occur whenever the natural rate of rotation of the helix ω_0 coincides with the rate of variation of this deterministic contribution of the random force, $\pm 2 d\alpha_0/ds$.

In summary, we presented a statistical mechanical description of thermally fluctuating elastic filaments of arbitrary shape and flexibility. We 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 nonmonotonic function of the elastic constants and moments of inertia. 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.

We thank A. Drozdov for illuminating discussions. Y. R. acknowledges support by a grant from the Israel Science Foundation.

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