Twist and writhe dynamics of stiff polymers

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This letter considers the dynamics of a stiff filament, in particular the coupling of twist and bend via writhe. The time dependence of the writhe of a filament is $\langle (Wr(t) - Wr(0))^2 \rangle \sim Lt^{1/4}$ for a linear filament and $\sim t^{1/2}/L$ for a curved filament. Simulations, on a simple model of a stiff polymer, are used to confirm the scaling arguments. Fuller's theorem, and its relation with geometric phases, is reconsidered for open filaments.

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Stiff polymers, such as actin filaments or DNA, allow one to study fundamental processes in polymer statics and dynamics using micromanipulation methods. Recent theoretical and experimental work on DNA force-extension curves has shown the importance of the *static* coupling between bend and twist in stiff polymers [1–4]. Here I examine the *dynamics* of twist and writhe fluctuations of a wormlike polymer using scaling arguments; I also introduce a generalized bead-spring model suitable for studying twist and writhe dynamics of polymers; other models for torsionally stiff models do exist in the literature, but are overly detailed, and cumbersome. This "minimum model" of a torsionally rigid polymer is easily generalized to chiral and other low symmetry situations.

For concreteness most of the present article is concerned with the following experimental set up: A stiff polymer, of length L, is held (with some adequate micromanipulation technique for instance magnetic beads) at its two ends. We follow the rotation or "spinning" angle, Ψ of the filament about its ends and ask about the nature of the dynamic correlations that are generated by the Brownian dynamics of the chain. For instance one might measure the two point function, $\langle (\Psi(0,0) - \Psi(L,t))^2 \rangle$. There are two physical processes which contribute to this correlation function. The two mechanisms are *twist* and *writhe*. Twist corresponds to excitation of internal torsional degrees of freedom of the filament at constant filament shape. Writhing is due to the three dimensional geometry of the filament in space. The above experiment is sensitive to the sum of the writhing and twisting contributions: $\Psi(0) - \Psi(L) = W + \phi$ with ϕ the contribution from internal twisting and $W = 2\pi Wr$ the contribution to the angle from the variation of the path in space. This simple additivity is a consequence of White's theorem [5]. Both ϕ and W are individually difficult to measure; They require information of the whole path in three dimensional space. Their sum however can be determined from simpler angular correlation functions.

The dynamics of internal twist degrees of freedom of short filaments are well understood [6] and have been observed in DNA and actin via depolarized light scattering experiments. The *static* writhing geometry of stiff polymers is usually formulated using the theorems of White [5] and Fuller [7]. One of the main questions considered in this paper is the nature of the writhe dynamics, in particular the writhe contribution to $\Psi(t)$, for a long stiff polymer.

It is interesting to note that Fuller's result is closely related to the geometric phase studied in optics and quantum mechanics. This deep analogy will allow us to generalize the idea of writhe to arbitrary open boundary conditions. As reminder to the reader we note that that we can calculate the writhe of a polymer from the tangent, $\mathbf{t}(s)$,which sweeps out a curve on the unit sphere, $\mathcal{S}_2$ as a function of the internal coordinate s. W is just the area (modulo 2π) [7] enclosed by $\mathbf{t}(s)$ on $\mathcal{S}_2$.

Let us assume that we are working with uniform filaments with circular cross sections; the two process of writhing and twisting are controlled by two independent elastic constants, the torsional stiffness, K and the bending stiffness κ. If we work in units such that $k_BT = 1$ these two elastic constants have the dimensions of lengths, and are indeed the persistence lengths for static torsional and bending correlations of the filament. The dynamics obeyed by the torsional and bending modes are, however, different which leads to distinct contributions to Ψ as we shall now demonstrate.

The torsional motion of a straight filament obeys a Langevin equation

$$a^2 \frac{\partial \phi}{\partial t} = K \frac{\partial^2 \phi}{\partial s^2} + \xi(t,s) \tag{1}$$

where the angle $\phi(s,t)$ is the local rotation of the filament in the laboratory frame, ξ is the thermal noise, a is comparable to the radius of the filament, (we neglect all prefactors of order unity and use units in which $\eta = k_BT = 1$, in this system of units time has the dimensions of a volume) so that $\langle \xi(s,t)\xi(s',t') \rangle = 2\delta(s-s')\delta(t-t')/a^2$. The solution of eq. (1) is well known, and can be written in a scaling form, for instance the autocorrelation function

$$\langle (\phi(0,0) - \phi(0,t))^2 \rangle = \frac{L}{K} \mathcal{P}\left(\frac{tK}{L^2 a^2}\right) \tag{2}$$

It is characterized by dynamic correlations of the angle which diffuse along the filament as $l_t = \sqrt{tK/a^2}$. For x small $\mathcal{P}(x) \sim \sqrt{x}$. When the filament can rotate freely about its axis $\mathcal{P}(x) \sim x$ for large x. If rotation of the end $s = L$ is blocked $\mathcal{P}(x) \sim 1$ for large x.

Let us now calculate the writhing contributions to $\Psi(t)$. For a short filament $(L/\kappa < 1)$ oriented along the z-axis the area enclosed by the curve $\mathbf{t}(s)$ is given by $W = 1/2 \int e_z.(\dot{\mathbf{t}} \wedge \mathbf{t}) ds$. The writhe of a filament is signed and averages to zero, $\langle W \rangle = 0$ for a linear polymer at thermal equilibrium. We shall thus consider the statistics and dynamics of $\langle W(t)W(t') \rangle$. The linearized bending modes of the filament obey a Langevin equation

$$\frac{\partial \mathbf{r}_\perp}{\partial t} = -\kappa \frac{\partial^4 \mathbf{r}_\perp}{\partial s^4} + \mathbf{f}_\perp(t,s) \tag{3}$$

with $\mathbf{r}_\perp$ the transverse fluctuations of the filaments and $\mathbf{f}_\perp$ the δ correlated Brownian noise. Using eq. (3) we find that $\langle \mathbf{t}_i,(q,0)\mathbf{t}_j(-q,t)\rangle = \delta_{i,j}\exp(-\kappa q^4 t)/\kappa q^2$ with $\{i,j\}$ x or y. The correlation function $\langle W(t)W(t')\rangle$ is fourth order in $\mathbf{t}$ and can be expanded using Wick's theorem: A short calculation shows that

$$\langle W^2 \rangle \sim L^2/\kappa^2. \tag{4}$$

and that

$$\langle (W(t) - W(0))^2 \rangle \sim l_1(t) L/\kappa^2 \sim L t^{1/4}. \tag{5}$$

with $l_1(t) \sim (\kappa t)^{1/4}$ the characteristic length scale in solutions of eq. (3).

What influence does this writhe fluctuation have on the global motion of a filament. To answer this question consider two extreme, non-physical, cases before coming back to the typical experimental situation. Consider, firstly, the case $K/\kappa \gg 1$; spinning of the end of a polymer can only occur via writhe (the twist degrees of freedom are frozen out). The derivation of eq. (5) has neglected rotational friction: It has been derived using a description of only the *transverse* motions of the polymer. It can only be valid when the rotational friction of a filament is so small that spinning driven by the writhe is able to relax without build up of torsional stress. Similar remarks have recently been made in the propagation of tensional stress in stiff polymers [8]. In this case simple arguments allow one to deduce the validity of the approximation made by balancing the driven motion due to the transverse fluctuations against any additional sources of friction. We shall now apply the same argument to the torsional motion, before checking the results numerically. For the rotational friction to be negligible the free rotational diffusion of a section of filament of length L must be faster faster than the driven writhing motion, implying $t/La^2 > l_1(t)L/\kappa^2$. For filaments which do not satisfy this criterion, *i.e.* when $L > l_W(t) = \sqrt{l_1^3 \kappa/a^2}$, the torsional stress has not had time to propagate between the two ends of the filament [9] and the law (5) does not apply. If we observe now the end of a filament for times shorter than the torsional equilibration time the spinning is due to an end section of length l_W and we should substitute this effective length in eq. (5) to calculate the end motion $\langle \Delta W^2 \rangle \sim l_1^{5/2}/a\kappa^{3/2} \sim t^{5/8}$. It is only after the propagation of the torsional fluctuations over the whole length of the filament that (5) becomes true. We thus hypothesize a scaling behavior for the spinning of a filament with $K/\kappa \gg 1$

$$\langle (\Psi(0,0) - \Psi(0,t))^2 \rangle = \frac{L^{5/3}}{\kappa^{7/3}} \mathcal{Q}\left(\frac{t\kappa^{7/3}}{a^{8/3}L^{8/3}}\right), \tag{6}$$

with $\mathcal{Q}(x) \sim x^{5/8}$ for x small and $\mathcal{Q}(x) \sim x^{1/4}$ for x large, in order to be consistent with eq. (5). This argument is subtle, it is however confirmed by detailed numerical simulations, fig(1), left.

A second extreme case is $K = 0$, as is for instance the case for many simple numerical bead-models of worm-like chains. In this case there is no need for the beads to rotate to follow writhe, the writhe fluctuations are absorbed by the internal twist degree of freedom without cost or dissipation.

Let us now turn to the physical case, $K/\kappa \simeq 1$. From the expressions for l_W and l_t we find that $l_t/l_W = \sqrt{l_1/\kappa} \ll 1$. On the length scale l_t the twist field and the writhe of the filament are in equilibrium and we can add the fluctuations of the twist and writhe degrees of freedom. Beyond l_t the successive section of the polymer are dynamically decoupled: a writhe fluctuation can not be transmitted faster than the signal transmitted by l_t and the second scenario becomes valid with l_t playing the role of a bead size. Beyond l_t the filament can writhe freely without any local consequence on the dynamics of the chain. When l_t reaches L the amplitude of twist fluctuations saturates, while the dynamics of the slower bending modes continues to evolve with $\Psi(0) - \Psi(L)$ varying as eq. (5) until $l_1(t) = L$. The experimental consequences of these remarks are discussed at the end of this letter.

These arguments have been checked by simulations on a discretized bead-spring model: Take $N+1$ spherical beads of diameter a connected by stiff harmonic springs. Each bead is characterized by its position $\mathbf{r}$ and a triad of orthonormal vectors. $\mathbf{M} = \{\mathbf{b}, \mathbf{n}, \mathbf{t}\}$. The vector $\mathbf{t}$ is an approximation to the local tangent to the filament at $\mathbf{r}$. $\{\mathbf{n}, \mathbf{b}\}$ span the normal space of the filament; $\mathbf{n}$ could describe the position of some feature on the surface of the molecule. The springs are not connected to the centers of the beads rather a bead i is linked to its neighbors by connections situated on its surface at $\{\mathbf{r}_i - a\mathbf{t}_i/2, \mathbf{r}_i + a\mathbf{t}_i/2\}$. The energy is

$$\begin{aligned} E = B/2 \sum_i (\mathbf{r}_i - \mathbf{r}_{i+1} - a\mathbf{t}_i/2 + a\mathbf{t}_{i+1}/2)^2 \\ + \kappa/a \sum_i (1 - \mathbf{t}_i \cdot \mathbf{t}_{i+1}) \\ + K/2a \sum_i (1 - \mathbf{n}_i \cdot \mathbf{n}_{i+1} - \mathbf{b}_i \cdot \mathbf{b}_{i+1} + \mathbf{t}_i \cdot \mathbf{t}_{i+1}) \end{aligned} \tag{7}$$

The first term imposes the linear topology of the filament; when B is large the curvilinear length of the filament is $L = Na$. In the simulations we are only interested

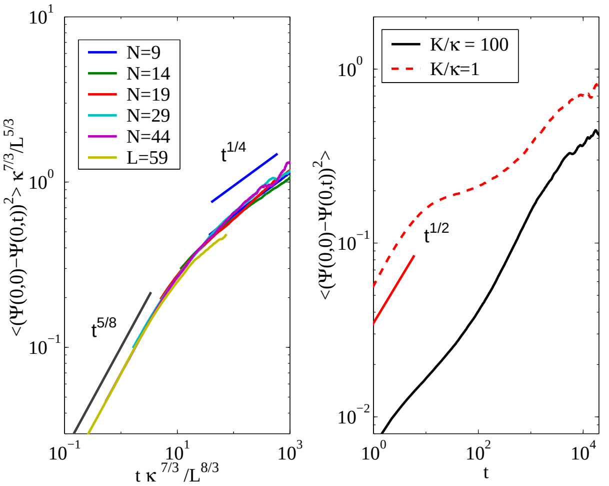

FIG. 1. Left: Dynamics of $\langle(\Psi(t,0)-\Psi(0,0))^2\rangle$ with $K/\kappa = 300$. Right: Dynamics of a curved, semicircular section of filament for two values of K. In the lower curve the dynamics is due to writhe. For the upper curve there is a two time behavior, with firstly twist then writhe relaxing. Simulations are for $L/a = \kappa/a = 35$.

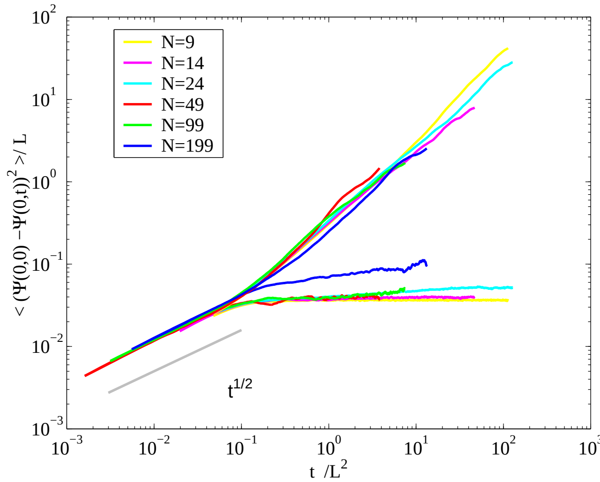

FIG. 2. Dynamics with two different boundary conditions, scaled according to eq(2). At short times the angle evolves as $\langle(\Psi(0,t)-\Psi(0,0))^2\rangle \sim t^{1/2}$. For long times there is either saturation of the twist angle to $\phi^2 = L/K$ (lower curves) or free diffusional rotation of the whole filament according to $\langle\phi^2\rangle \sim t/La^2$ (upper curves). $\kappa = 30a$, L varies from $\kappa/3$ to 6.6κ. $K/\kappa = 1$. Splitting of lower curves is due to writhe.

in the limit of large B. The second term is a bending energy. The third term is the torsional energy. Here by using the full set of vectors $\{\mathbf{t}, \mathbf{n}, \mathbf{b}\}$ we have an *over-complete* description of the geometry. This over-complete description however leads to enormous simplifiction in the discretization of the model (avoiding the use of high spatial derivatives and/or Euler angles to calculate the torsion) and integration of the equations of motion

I integrate the equations of motion for the position and orientation adding friction and thermal noise coupled to the translational and angular velocities, effectively leading to Brownian dynamics. Since we are particularly interested in the writhe dynamics I use boundary conditions where the positions of the ends of the filament are free to move, but the end tangents are constrained via external torques, so that the writhe can be calculated via Fuller's result.

A series of simulations were performed to study the writhe and twist fluctuations of filaments with very high torsional constants to verify the predictions made for the writhe dynamics. When K is large all contributions to Ψ come from the writhing. In the simulation the end $s = 0$ is free, while the motion of the end at $s = L$ is blocked. Fig. (1) shows the the scaling proposed, eq (6), is indeed seen. After a short transient, needed for propagation of torsional stress into the filament the spinning of the filament is dominated by the writhing dynamics so that $\langle(\Psi(0,t)-\Psi(L,0))^2\rangle \sim Lt^{1/4}$ as proposed in the above discussion.

A complementary series of simulations were performed with $K/\kappa = 1$ to study the internal twisting dynamics of very long filaments. This simulation was performed to confirm the intuition [10–12] that twist modes can transport torsional stress even in the presence of large bends in the filament. This stress is carried by rapid spinning of the filament in a slowly evolving tube, rather than the rotation of the whole filament collectively moving against solvent friction, (like the rotation of a rigid crankshaft). Simulations are performed for filaments of varying length with two possible boundary conditions. We analyze the simulations using the scaling form (2). The good scaling exhibited by the upper curve shows that torsion propagates perfectly well over many persistence lengths without hindrance from the tortuosity of the path in space; the writhing is subdominant to the free rotation of the filament in the effective tube. A clear *break down* of scaling is seen in the lower branch and is due to the slow writhing contribution (5) to the angle Ψ over and above the contribution from the twist dynamics predicted by eq. (2).

In order to calculate the writhe of a curve via Fuller's theorem we have, until now, considered paths $\mathbf{t}(s)$ which are closed on $\mathcal{S}_2$. In general this is most inconvenient: there are many experimental situations where one would like to compare the spinning of filaments oriented in an arbitrary direction. We now consider how one might generalize the idea of writhe and spinning to filaments with arbitrary open boundary conditions. Fuller's theorem is closely related to Berry's phase in experiments on polarized light transmission along bent optical fibers [13]. The vector $\mathbf{t}(s)$ corresponds to the local tangent to the fiber, while the vectors, $\{\mathbf{n}, \mathbf{b}\}$ transform in exactly the same manner as the plane of polarization, via parallel transport on $\mathcal{S}_2$. The problem of closing paths in $\mathcal{S}_2$ has close analogies in the treatment of Berry's phase in non cyclic

Hamiltonians. In quantum and optical systems interference phenomena allow one to define the relative phase of a system even if the evolution has not been cyclic [14] via the Pancharatnam connection. This convention closes an open trajectory via a geodesic, the same convention can be shown to have sense for writhing polymers.

We can reproduce the result (5) from a simple scaling argument: The path $\mathbf{t}(s)$ on $\mathcal{S}_2$ is a Gaussian random walk ; On a time scale t each section of length l_1 of the filament re-equilibrates a random walk with radius of gyration on $\mathcal{S}_2$ of $r_\Omega^2 \sim l_1/\kappa$ and area $A_1 \sim \pm l_1/\kappa$. There are $N_1 = L/l_1$ dynamically independent contributions to the writhe variation giving $\langle \Delta W^2 \rangle \sim N_1 A_1^2 = l_1(t) L/\kappa^2$ as found above. This argument generalizes easily to bent filaments, or sharp curves: The pre-existing bend considerably modifies the arguments. Consider a section of filament which bends an angle Φ at zero temperature due to intrinsic curvature. At zero temperature the bent loop has a tangent map which maps to the equator of $\mathcal{S}_2$. Under the map to $\mathcal{S}_2$ a distance of s in real space becomes $\Phi s/L$. At finite temperature there is competition between this constant drift and the thermal agitation which leads to a Gaussian random walk with drift on $\mathcal{S}_2$. For the very shortest times only small scale structure is changing:- the writhe dynamics reduces to the case discussed above. At longer times the stretched path moves collectively:- each section of length $\delta_1 = \Phi l_1/L$ on $\mathcal{S}_2$ moves up or down by $r_\Omega = \sqrt{l_1/\kappa}$. Counting $N_1 = L/l_1$ dynamically independent sections

$$\Delta W^2 \sim N_1 (\delta_1 r_\Omega)^2 = \Phi^2 l_1^2/\kappa L \sim t^{1/2}/L. \tag{8}$$

The exponent for the writhe fluctuations has changed from 1/4 to 1/2 due to the stretching of the configuration in $\mathcal{S}_2$. This prediction is tested in fig. (1), where we have added a spontaneous bending energy, in $\alpha \Sigma_i \mathbf{n_i}.\mathbf{t_{i+1}}$ to curve the filament. We now see two distinct regimes in $t^{1/2}$ due to the intrinsic twist dynamics and then the writhe dynamics; the writhing contribution is much enhanced over its value for linear filaments. It would also be interesting to study the case of closed loops, but the simple scaling argument used here becomes considerably more complicated due to the non-local integral constraint $\int \mathbf{t}(s) \c{s} = 0$ coming from closure. However, recent simulations on a detailed model of DNA dynamics used to study the fluctuations of supercoiled loops seem to give a similar result, $\Delta W r^2 \sim t^{1/2}$ [15].

How might one study the writhing of a stiff polymer experimentally? One possibility, using micromanipulation of single actin filaments, is by reproducing the lower curves of fig. (2). In actin the time scale of relaxation of the torsional and writhing modes are very widely separated. For a 15 μm filament torsional modes relax in about 1 ms while the writhing modes relax on a time scale of several minutes. Measurement of light scattering from anisotropic micro-beads [16] detected by a photo-diode would show two relaxation process. A far more direct method of measuring the writhe is by confocal imaging of a single marked filament followed over several minutes.

This paper has studied the simplest model of a stiff polymer, with uniform cross section and without disorder. The Marko-Siggia energy function [17] already has rich static behavior and one would anticipate that the cross terms in this Hamiltonian could increase the importance of the dynamical effects discussed here. Similarly, any disorder in the ground state is expected to dramatically modify the dynamics, by giving a preformed writhe on $\mathcal{S}_2$ and by modifying the picture of simple spinning of the polymer in its tube due to new dissipative processes; It has recently been argued that a mixture of crankshaft and tube spinning should co-exist even in the case of weak disorder coming from sequence fluctuations in DNA. [10]. The numerical model introduced in this paper has been designed for efficiency and simplicity. By the addition of cross terms in the Hamiltonian is is trivial to generate lower symmetry ground states, such as those required to study the effect of chirality or disorder on the statics and dynamics of stiff polymers.

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