Fluctuations and Stiffness of Double Stranded Polymers: Railway Track Model

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Many biopolymers are double-stranded with a tight coupling between the filaments. We introduce a simple model to study the consequences. It consists of two worm-like chains coupled by harmonic springs between points of identical contour length and displays a novel behavior: a larger stiffness on large than on small lengthscales. Neglecting local deformations the global persistence length is found to be infinite. Otherwise, worm-like chain behaviour with a higher, renormalized bending stiffness is recovered on large lengthscales. We estimate the coupling contribution to the bending energy of double helices and argue that it could contribute to the stiffness of biopolymers.

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A characteristic feature of many biopolymers is their high bending stiffness. Contour lengths of tens of μm and persistence lengths of the order of one μm as in the case of actin even allow microscopy techniques to be used to directly observe their structure and dynamics [1,2]. In contrast synthetic polymers usually have a persistence length on the nm scale [3]. Many of the physical peculiarities of biopolymers are attributed to their polyionic character, but often there is another distinct feature: the molecules are double- or even multiple-stranded with a tight coupling between the filaments. The bending stiffness of single- and double-stranded DNA, for example, differ by a factor of 25 [4,5].

The purpose of this paper is to study the consequences of backbone coupling in double-stranded macromolecules. The effect is most prominent in two dimensions (2-d). If two strongly linked, incompressible strands are bent, the outer strand must travel a greater distance than the inner one, inducing tension into the links. This effect is non–local and dominates the behavior on large lengthscales. In our model two worm-like chains are coupled by harmonic springs between points of identical contour length, resembling a pair of railway-tracks. The "rails" are free to slide against each other at constant distance. Modifications due to local deformations are included in a second step. For the immediate 3-d extension, a tube like object, we expect qualitatively similar behavior as long as the structure is not twisted. In a physically realistic situation, double or even triple stranded molecules usually wind into a helical structure. Based on the results of the simple strictly 2-d case, we will estimate the contribution of the coupling to the bending energy of a double-stranded molecule wound into a regular helix.

Beyond the molecular lengthscale a polymer of length L is described by its position $\vec{R}(s)$ at the contour point s, $0 \le s \le L$. For an incompressible chain the tangent $\vec{u}(s) = \partial\vec{R}/\partial s$ is a unit vector. In 2-d $\vec{u}(s)$ can be replaced by its angle $\theta(s)$ with the x–axis. The Kratky–Porod or worm-like chain model [6] is characterized by a local bending energy [1]

$$U_{bend} = \kappa/2 \int_0^L ds\ \dot{\theta}^2(s)\ . \tag{1}$$

A worm-like chain is stiff(flexible) on length scales smaller(larger) than the persistence length

$$l_p = \frac{\kappa}{k_B T}. \tag{2}$$

Extending this, we describe double stranded molecules with tight couplings between specific points along the backbones. The distance a between strands is kept constant. This is a reasonable assumption to start with for polymers with backbone diameters of order a and a distance of coupling points smaller than the strand persistence length $l_p/2$. Fig. 1 illustrates the model and the arc–length mismatch, Δ, between the filaments induced by bending. A change $d\theta$ of the direction of the molecule changes Δ by an amount $a\ d\theta$. We emphasize that the contribution of the coupling to the bending energy is non–local, since the arc–length mismatch $\Delta(s) - \Delta_0 = a\ \big(\theta(s) - \theta(0)\big)$ depends on the *integrated* change in orientation. The constant Δ_0 is the overhang of the two strands at one end of the molecule, while $\theta(0)$ corresponds to the rotational degree of freedom of the whole chain. Assuming harmonic coupling $U_{coup} = \frac{1}{2}k \int_0^L ds\ \Delta^2(s)$ and integrating out Δ_0 in the partition function leads to an effective coupling energy,

$$U_{coup}^{eff} = ka^2/2 \int_0^L ds \left(\theta(s) - 1/L \int_0^L ds'\ \theta(s')\right)^2 . \tag{3}$$

[1] For double stranded molecules κ will be twice the bending stiffness of a filament

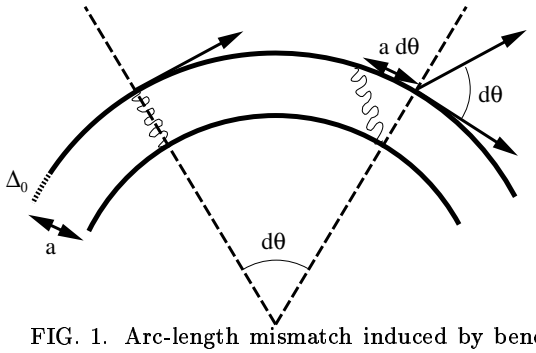

FIG. 1. Arc-length mismatch induced by bending in the railway-track model.

The railway-track model can best be solved by expressing $\theta(s)$ as the Fourier sum

$$\theta(s) = \sum_q \theta_q \cos(qs), \quad q = \{0,\, \pi/L,\, 2\pi/L,\, 3\pi/L, \ldots\}, \tag{4}$$

where the sin modes are omitted due to the boundary condition $(\partial\theta/\partial s)|_{s=0,L} = 0$. Only the modes with $q > 0$ contribute to the total energy

$$U_{bend} + U^{eff}_{coup} = L/4 \sum_{q>0} \left(\kappa q^2 + ka^2\right) \theta_q^2 \quad . \tag{5}$$

since the average θ is subtracted in Eq. (3). U_{bend} and U^{eff}_{coup} are diagonal and quadratic in the modes. The equipartition theorem yields:

$$\langle \theta_q \theta_{q'} \rangle = \frac{2k_BT}{L} \frac{1}{ka^2 + \kappa q^2} \delta_{qq'} \tag{6}$$

The qualitative difference in the behavior of railway-track and worm-like chains becomes obvious when we calculate the *apparent* bending stiffness

$$\kappa^*(q) = \frac{2k_BT}{q^2 L \langle \theta_q^2 \rangle} = \kappa + \frac{ka^2}{q^2} \tag{7}$$

which diverges for small wave vectors. The ratio

$$l^2 = \frac{\kappa}{ka^2} \quad . \tag{8}$$

defines a coupling length. Beyond l the nonlocal contour mismatch dominates over the bending stiffness of the strands. Since the model is Gaussian, other quantities can be obtained as well. The width of the θ distribution for a molecule of length L is given by:

$$\langle \theta^2 \rangle_L = \frac{2k_BT}{L} \sum_{q>0} \frac{1}{ka^2 + \kappa q^2} \sim \begin{cases} L/l_p & \text{for } L \ll l \\ l/l_p & \text{for } L \gg l \end{cases} . \tag{9}$$

Initially, $\langle \theta^2 \rangle_L$ grows linearly in length as for a worm-like chain. For chains with $L \ll l$ the coupling has no effect. For $L \gg l$ the width of the θ distribution saturates at a value $\frac{l}{l_p}$, independent of L. As a consequence, the persistence length $\frac{1}{\frac{\partial}{\partial L}\langle \theta^2 \rangle_L}$ of a railway-track molecule is infinite and the end-to-end distance proportional to L. The extension of the molecule parallel to its average orientation is given by $\langle r_{||} \rangle = L\langle \cos(\theta) \rangle = Le^{-l/2l_p}$. The exponential decrease in length is quite restricted due to the upper limit

$$\epsilon \equiv \sqrt{\kappa k a^2} > k_BT \tag{10}$$

for the validity of the harmonic approximation for the coupling energy. In the range (10) $\langle r_{||} \rangle \geq \frac{1}{2}L$.[2]

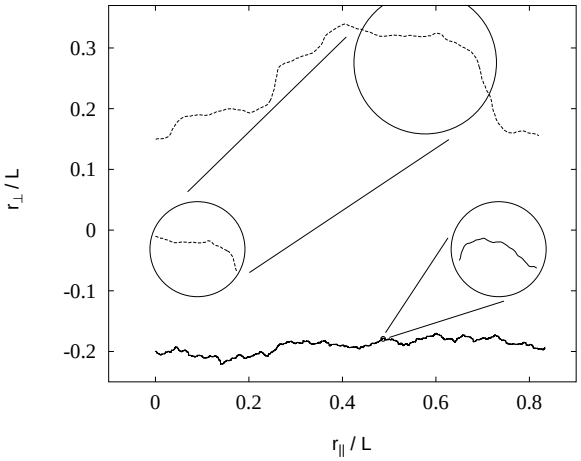

FIG. 2. Typical conformations of a railway-track ($T = \frac{1}{2}\epsilon$, solid line) and a worm-like chain ($T = \frac{3}{2}\frac{\kappa}{L}$, dashed line). The inserts show rescaled parts of the two chains of length $\frac{1}{2}l_p$ which equals the coupling length l in the case of the railway-track model.

In Fig. 2 we have plotted typical conformations of a railway-track and a worm-like chain. They were obtained by drawing random amplitudes for the modes Eq.(4) from Gaussian distributions with widths given by Eq.(6). With the choice $T = \frac{1}{2}\epsilon$ and $T = \frac{3}{2}\frac{\kappa}{L}$ respectivly molecules with identical contour length have similar end-to-end distances. The inserts show rescaled parts of the two chains of length $\frac{1}{2}l_p$ which equals the coupling length l in the case of the railway-track model. This

[2] Since $\langle \Delta^2 \rangle = a^2\langle \theta^2 \rangle$, $\epsilon = k_BT$ implies that the arc-length mismatch is of the order of the width of the molecule. This also explains, why a railway-track molecule cannot crumple: to do so the angle $\theta(s)$ with its mean orientation has to reach at least values of order π. However, even $\langle \theta^2 \rangle \approx \pi^2/4$ is well beyond the range (10) of the harmonic approximation. Therefore, higher-order terms in the coupling energy will become dominant before the molecules crumple.

demonstrates that the coupling has no effect on these lengthscales.

In the continuum limit the correlations along the chain between the deviations from the average orientation are given by:

$$\langle \theta(s)\theta(s') \rangle = l/l_p \ \exp\left(-|s-s'|\,/\,l\right) \quad . \tag{11}$$

This can e. g. be used to calculate the mean square end–to–end distance [7]. Fig. 3 shows the effect of the coupling within the railway-track model for molecules with flexible ($\frac{L}{l_p} = 1000$) and semi-flexible ($\frac{L}{l_p} = 10$) backbones. For large coupling lengths, l, the size of the molecule is given by the well known worm-like chain limit $2l_p\left(L + l_p\left(e^{-L/l_p} - 1\right)\right)$. With decreasing l the long-wavelength modes are damped and the molecule starts to stretch until in the range (10) [7]

$$\langle (\vec{R}(L) - \vec{R}(0))^2 \rangle = L^2 e^{-l/l_p} = \langle r_{||} \rangle^2 \tag{12}$$

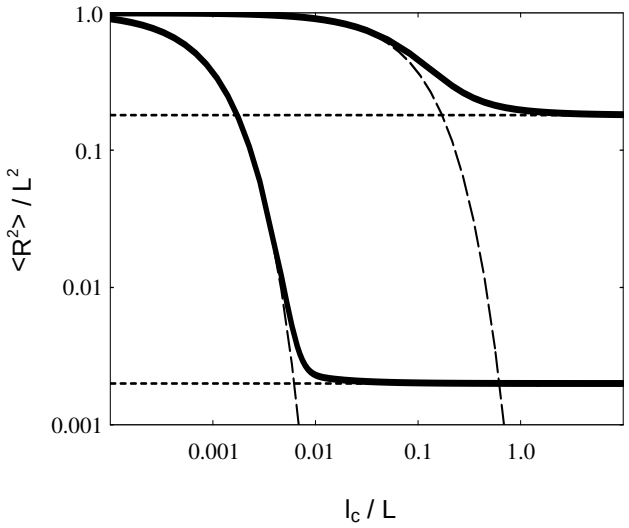

FIG. 3. Mean-square end-to-end distance of railway-track molecules with flexible ($L/l_p = 1000$) and semi-flexible ($L/l_p = 10$) backbones vs. coupling length l/L. Included are the worm-like chain limit and the exponential decrease in the physical limit Eq. (12) with $l < l_p$.

So far only the most simple model of a double stranded polymer was considered. As soon as the arc-length mismatch is too high the original railway-track molecule has no other choice but to bend as a whole. A more realistic model should allow a reduction of the bending induced arc-length mismatch $\Delta(s)$ by local deformations such as filament compression or small variations in the width a of the molecule. To include this, we consider the arc-length mismatch $\lambda(s)$ due to these deformations. The corresponding strain energy is $U_{strain} = \frac{1}{2}\rho \int_0^L ds\ \dot{\lambda}^2(s)$. In the case $\rho = 0$ one has $\lambda(s) = -\Delta(s)$ and the total arc-length mismatch and therefore the effects of the coupling vanish altogether. In general, the coupling energy has the form $U_{coup} = \frac{1}{2}k \int_0^L ds\ (\Delta(s) + \lambda(s))^2 = 0$. For $\rho \neq 0$ the new degrees of freedom $\lambda(q)$ can be integrated out in the partition function and one is left with an apparent bending stiffness [8]

$$\kappa^*_{strain} = \kappa + \frac{ka^2}{l_d^{-2} + q^2} \quad , \tag{13}$$

where we have introduced the new lengthscale, the *de-coupling length* $l_d = \sqrt{\frac{\rho}{ka^2}}$. For large q, we can omit the second term and find again the *local* bending stiffness given by κ. However, now the apparent bending stiffness does not diverge for $q \to 0$. For lengthscales beyond l_d it enters another plateau with a value $\kappa^* = \kappa + \rho$. There the molecule behaves again like a worm-like chain with a renormalized bending stiffness.

So far we have considered strictly two dimensional systems. The immediate generalization to $d = 3$ is a tube-like object of three or more coupled filaments. The qualitative difference between 2-d and 3-d is the occurence of twist and the possibility of averaging out the bending induced arc-length mismatch over the length of a full turn. Consequently, there will be a coupling between bending and torsion. In the limiting case of no spontaneous twist and a high torsion rigidity we expect the same qualitative behaviour for three or more strings in 3-d as for two strings in 2-d. As a possible example we mention microtubuli, rodlike assemblies of ten to fifteen filaments which are the stiffest molecules in the cytoplasm.

Of great interest is the case of two strings in 3-d. The coupling of the filaments introduces an anisotropy into the bending elasticity parallel and perpendicular to the plane of the molecule with a two dimensional worm-like chain of persistence length l_p as a limiting case. This shows that on entropy grounds alone the molecules will prefer to twist. In general, we expect a very complex behaviour. It is quite suggestive to apply the coupling idea to recent observations on actin [1] as the qualitative behaviour of local flexibility and global stiffness is reproduced by the *two* dimensional railway-track model. The question, how this carries over to a helical molecule will require a detailed analysis of the interplay between bending and torsion mediated by the coupling of the filaments. Here, we restrict ourselves to the contribution of the backbone coupling to the large scale bending stiffness of a helical molecule ignoring torsion modes. The results apply to double as well as triple or more stranded molecules.

Consider a helical molecule with a certain repeat distance (pitch) b along the contour. Quite obviously, there will be no significant contribution of the coupling to the bending energy, if $b \ll l$, since the induced arc-length mismatch is averaged out over a distance of b. In the opposite case, $b > l$, we can however expect such a contribution. To estimate the effect we have considered an extension of the railway-track model where the rails are twisted into a double helix. Although the calculations

were carried out for a 2d model the argument also holds for the smooth long scale bend of a molecule in $d = 3$. As a result [8] we find worm-like chain behaviour on large lengthscales with a renormalized bending stiffness

$$\kappa^*_{twist} = \kappa \left(1 + b^2/l^2\right) \ . \tag{14}$$

Eq. (14) implies that backbone coupling in double stranded molecules could cause persistence lengths, which are much larger than those of the individual strands. Furthermore, the signature of the coupling effect is an increase of the bending stiffness with the pitch, b. This is interesting, since the argument that double helices are so much stiffer due to steric repulsions between consecutive turns would point into the opposite direction.

To summarize, we have presented a *model* for coupling effects in double-stranded macromolecules with parameters corresponding to the bending stiffness of the individual filaments, to the coupling strength and to the deformation rigidity of the double-stranded molecule. Our railway-track model displays an interesting new type of behavior for a mechanical model without long-range forces [9]: flexibility on short and stiffness on large lengthscales. Neglecting local deformations the global persistence length was shown to be infinite. Otherwise, worm-like chain behaviour with a higher, renormalized bending stiffness was recovered on large lengthscales. We have estimated the coupling contribution to the bending energy of double helices and argued that it could contribute to the stiffness of biopolymers. Finally we mention two possible extensions of the railway-track model: two coupled sheets in 3-d and the introduction of coupling defects as discontinuities in Δ which alter the preferred direction.

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