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LETTER TO THE EDITOR

Free energy of a semiflexible polymer confined along an axis

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Abstract. A continuum model of a fluctuating semiflexible polymer chain confined along an axis is considered. In the regime of strong confinement, configurations with overhangs are negligible, and the partition function is determined by a partial differential equation. An extremum principle for eigensolutions is formulated. The exact solution of the differential equation for a polymer in a harmonic potential is given. A lower bound for the confinement free energy of a polymer in a tube is obtained.

The statistical properties of a semiflexible polymer in a tube have been studied with several equivalent theoretical approaches [1–5], reviewed in [6]. For a cylindrical tube of diameter D the confinement free energy per unit length Δf is given by

$$\Delta f = c \frac{k_B T}{P^{1/3} D^{2/3}} \tag{1}$$

in the regime $P \gg D$ of strong confinement. Here $P = \kappa/k_B T$ is the persistence length, where κ is the bending modulus. The dimensionless constant c is not known exactly but was estimated as 2.46 ± 0.07 in recent computer simulations [7]. From dimensional analysis Δf has the form $k_B T/\lambda$, where λ is a length. As emphasized by Odijk [3, 6], the relevant physical length λ in equation (1) for a strongly confined polymer is not the persistence length P but the collision length or typical distance $\lambda \sim P^{1/3} D^{2/3}$ between points where the polymer touches the tube.

In specifying polymer configurations, it is convenient to use cylindrical coordinates $(x, y, t) = (r, t)$, with the t axis corresponding to the symmetry axis of the tube or confining potential. In the regime of strong confinement, i.e. for sufficiently stiff polymers in sufficiently strong confining potentials, configurations with overhangs are negligible, and r may be regarded as a single-valued function of t , with $\langle (dr/dt)^2 \rangle \ll 1$. The partition function of a strongly confined polymer with axial length t is given by the path integral

$$Z(r, u; r_0, u_0; t) = \int D^2 r \exp \left\{ - \int_0^t dt \left[\frac{1}{2} P \left(\frac{d^2 r}{dt^2} \right)^2 + V(r) \right] \right\} \tag{2}$$

where r and $u = dr/dt$ denote the displacement and slope of the polymer at t , and r_0 and u_0 the same quantities at $t = 0$. The two terms in the exponential function represent the bending energy and the potential energy leading to confinement, both divided by $k_B T$.

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The path integral for a flexible polymer under tension or 'directed polymer' is the same as in (2) except that $(d^2r/dt^2)^2$ is replaced by $(dr/dt)^2$. The partition function $Z(r, r_0; t)$ of the directed polymer satisfies Schrödinger's equation [8-10]. Equation (2) leads to the more complicated partial differential equation [11-14]

$$\left[\frac{\partial}{\partial t} + \mathbf{u} \cdot \nabla_r - \frac{1}{2P} \nabla_u^2 + V(r) \right] Z(r, \mathbf{u}; r_0, \mathbf{u}_0; t) = 0 \quad (3)$$

with boundary equation

$$Z(r, \mathbf{u}; r_0, \mathbf{u}_0; 0) = \delta(r - r_0) \delta(\mathbf{u} - \mathbf{u}_0) \quad (4)$$

at $t = 0$.

In the case $V(r) = 0$ of a free polymer in the unbounded space (x, y, t) , the partial differential equation (3) can be readily solved [14] by Fourier transformation. The exact solution [15] for a polymer confined to the half space $x > 0$ is considerably more complex, since the hard-wall boundary condition cannot be simply satisfied by the method of images. Polymer configurations with a discontinuity in slope cost an infinite energy according to (2) and are completely suppressed. This implies [15] that $Z(r, \mathbf{u}; r_0, \mathbf{u}_0; t)$ vanishes at a hard wall for $\mathbf{u} \cdot \mathbf{n} > 0$. This requirement and the differential equation (3) determine the non-zero but unspecified value of $Z(r, \mathbf{u}; r_0, \mathbf{u}_0; t)$ on the boundary for $\mathbf{u} \cdot \mathbf{n} < 0$. Here \mathbf{n} is a unit vector perpendicular to the boundary surface and directed into the region accessible to the polymer.

In this letter the theoretical approach to strongly confined semiflexible polymers based on the partial differential equation (3) is explored. In general, the equation is difficult to solve in a bounded geometry due to the correlation between r and \mathbf{u} coming from the second term. An extremum principle for eigensolutions is formulated. It may be used to optimize approximate calculations of the free energy of confinement. The exact solution of equations (3) and (4) for a polymer confined by a harmonic potential $V(r) = \frac{1}{2}br^2$ is given. This is an instructive example, and the results for arbitrary length t may be useful in finite-size applications, for example, to liquid crystals. As in the analysis of the quantum harmonic oscillator, the solution can be derived using raising and lowering operators. The case of a polymer in a tube is briefly considered. It is shown that the path integral (2) implies expression (1) for the confinement free energy. The bound $c > \frac{3}{2}$ is derived, using the results for the harmonic potential.

In analysing long polymer chains it is convenient to look for exponentially decaying solutions of equation (3) with the form $\psi(r, \mathbf{u}) \exp(-Et)$. The eigenfunctions ψ_n and eigenvalues E_n satisfy

$$(\mathcal{H} - E_n)\psi_n(r, \mathbf{u}) = 0 \quad \mathcal{H} = \mathbf{u} \cdot \nabla_r - \frac{1}{2P} \nabla_u^2 + V(r). \quad (5)$$

In the long polymer limit the partition function and the confinement free energy per unit length are given by

$$Z(r, \mathbf{u}; r_0, \mathbf{u}_0; t) \approx \psi_0(r, \mathbf{u}) \psi_0(r_0, -\mathbf{u}_0) e^{-E_0 t} \quad t \rightarrow \infty \quad (6)$$

$$\frac{\Delta f}{k_B T} = - \lim_{t \rightarrow \infty} t^{-1} \ln Z(r, \mathbf{u}; r_0, \mathbf{u}_0; t) = E_0 \quad (7)$$

where E_0 is the eigenvalue with the smallest real part. The eigenvalues E_n are, in general, complex. However, for a non-negative confining potential, E_0 is positive and non-degenerate, due to the real, non-negative argument of the exponential function in the path integral (2).

It is convenient to define an inner product (F, G) of two functions $F(r, u)$, $G(r, u)$ by

$$(F, G) = \int d^2r \int d^2u F(-r, u)G(r, u) \quad (8)$$

with $-r$ instead of r in the argument of the function on the left. With this definition the property $(\psi_m, \mathcal{H}\psi_n) = (\psi_n, \mathcal{H}\psi_m)$ follows, for reflection-invariant potentials $V(r) = V(-r)$, from integration by parts. This, together with equation (5), establishes the orthogonality $(\psi_m, \psi_n) = 0$ of the eigenfunctions for $E_m \neq E_n$. These results hold both for soft potentials and the hard-wall boundary condition discussed above.

Since the partial differential equation (5) for the eigenfunctions is, in general, intractable, it is useful to have a variational principle to optimize approximations. It is simple to show that the eigenvalues E_n and eigenfunctions ψ_n defined by equation (5) correspond to extreme values of the functional

$$\mathcal{E}[\Psi] = \frac{(\Psi, \mathcal{H}\Psi)}{(\Psi, \Psi)}. \quad (9)$$

In quantum mechanics any reasonable trial wavefunction furnishes an upper bound to the exact energy of the ground state. However, due to the special inner product (8) in (9) it is not clear that E_0 and ψ_0 correspond to an absolute minimum of $\text{Re } \mathcal{E}[\Psi]$. The question of the nature of the extrema will be taken up again below.

For the harmonic confining potential $V(r) = \frac{1}{2}br^2$ the path integral (2) can be evaluated exactly. As for the quantum mechanical oscillator [8] the path integral is determined by the 'classical' path $r^*(t)$ that minimizes the action. This may be seen by substituting $r(t) = r^*(t) + \xi(t)$ in (2), where r^* satisfies the differential equation

$$P \frac{d^4 r^*}{dt^4} + br^* = 0. \quad (10)$$

A straightforward calculation yields

$$Z(r, u; r_0, u_0; t) = Z(\mathbf{0}, \mathbf{0}; \mathbf{0}, \mathbf{0}; t) \exp[-S^*(r, u; r_0, u_0; t)] \quad (11)$$

where $S^*(r, u; r_0, u_0; t)$ is the action for the path $r^*(t)$. In terms of the dimensionless variables

$$\hat{r} = P^{1/8} b^{3/8} r \quad \hat{u} = P^{3/8} b^{1/8} u \quad \hat{t} = b^{1/4} P^{-1/4} t \quad (12)$$

which eliminate the constants P and b in the path integral (2),

$$Z(\mathbf{0}, \mathbf{0}; \mathbf{0}, \mathbf{0}; t) = \mathcal{N} P b (s_h^2 - s^2)^{-1} \quad (13)$$

$$\begin{aligned} S^*(r, u; r_0, u_0; t) = & \frac{1}{2}(s_h^2 - s^2)^{-1} \{ \sqrt{2}(c_h s_h + cs)(\hat{r}^2 + \hat{r}_0^2) + \sqrt{2}(c_h s_h - cs)(\hat{u}^2 + \hat{u}_0^2) \\ & - 2(s_h^2 + s^2)(\hat{r} \cdot \hat{u} - \hat{r}_0 \cdot \hat{u}_0) - 2\sqrt{2}(c_h s + s_h c)\hat{r} \cdot \hat{r}_0 \\ & + 2\sqrt{2}(c_h s - s_h c)\hat{u} \cdot \hat{u}_0 - 4s_h s(\hat{r} \cdot \hat{u}_0 - \hat{u} \cdot \hat{r}_0) \} \end{aligned} \quad (14)$$

where

$$c_h = \cosh \frac{\hat{t}}{\sqrt{2}} \quad s_h = \sinh \frac{\hat{t}}{\sqrt{2}} \quad c = \cos \frac{\hat{t}}{\sqrt{2}} \quad s = \sin \frac{\hat{t}}{\sqrt{2}} \quad (15)$$

and \mathcal{N} is a dimensionless normalization constant.

Comparing equations (11)–(15) in the long polymer limit $\hat{t} \gg 1$ with (6), (7) yields

$$\frac{\Delta f}{k_B T} = E_0 = \sqrt{2}(b/P)^{1/4} \quad (16)$$

$$\psi_0(r, u) = 2\mathcal{N}^{1/2}(Pb)^{1/2} \exp\left[-\frac{1}{\sqrt{2}}(\hat{r}^2 + \hat{u}^2) + \hat{r} \cdot \hat{u}\right]. \quad (17)$$

The positive correlation between r and u in ψ_0 has a simple physical interpretation, reflecting the tendency of the polymer to lie near the t axis on the average and slope outwards from the axis to a given endpoint r . From equations (11)–(15) the explicit form of the eigensolutions $\psi_n(r, u) \exp(-E_n t)$ is apparent. One sees that the E_n are complex and degenerate, in general, and equally spaced with separation $\exp(\pm i\pi/4)(b/P)^{1/4}$. The $\psi_n(r, u)$ have the form of polynomials in r and u multiplying $\psi_0(r, u)$.

The approximation of neglecting overhangs and a constraint on the total length in the path integral (2) is self-consistent as long as $\langle (dr/dt)^2 \rangle \ll 1$. From equation (12) one sees that this requires $Pb^{1/3} \gg 1$. This inequality defines the regime of strong confinement for a semiflexible polymer in a harmonic potential, analogous to the regime $PD^{-1} \gg 1$ for a polymer in a tube described by (1).

Equations (2), (7), and (16) imply $(k_B T)^{-1} \partial \Delta f / \partial b = \frac{1}{2} \langle r^2 \rangle = 2^{-3/2} b^{-3/4} P^{-1/4}$. Using this result to eliminate the potential parameter b in (16) gives

$$\frac{\Delta f}{k_B T} = 2^{1/3} P^{-1/3} \langle r^2 \rangle^{-1/3} \quad (18)$$

for the confinement free energy per unit length. Note that this expression has the same general form as (1). The general form is independent of the details of the confining potential. However, the value of the dimensionless constant c in (1) does depend on the particular potential.

Since the eigenvalues E_n for the harmonic confining potential are equally spaced, it is natural to look for an equivalent solution in terms of raising and lowering operators. It is straightforward to show that the differential operator \mathcal{H} in (5) with $V(r) = \frac{1}{2}br^2$ can be written in the form

$$\mathcal{H} = \left(\frac{b}{P}\right)^{1/4} (\mathcal{R} \cdot \mathcal{L} + \mathcal{R}^* \cdot \mathcal{L}^* + \sqrt{2}) \quad (19)$$

where

$$\mathcal{R} = \frac{1}{2}(\hat{r} - e^{i\pi/4} \nabla_{\hat{r}} + e^{-i\pi/4} \hat{u} + i \nabla_{\hat{u}}) \quad (20)$$

$$\mathcal{L} = \frac{1}{2}(\hat{r} + e^{i\pi/4} \nabla_{\hat{r}} - e^{-i\pi/4} \hat{u} + i \nabla_{\hat{u}}) \quad (21)$$

and \mathcal{R}^* and \mathcal{L}^* are their complex conjugates. Here \mathcal{R} and \mathcal{R}^* are raising operators that shift the eigenvalue by $\exp(\pm i\pi/4)(b/P)^{1/4}$, and \mathcal{L} and \mathcal{L}^* are lowering operators that shift the eigenvalue by $-\exp(\pm i\pi/4)(b/P)^{1/4}$. This follows from the commutation relations $[\mathcal{R}^*, \mathcal{R}] = [\mathcal{L}^*, \mathcal{L}] = [\mathcal{L}^*, \mathcal{R}] = 0$, $[\mathcal{L}, \mathcal{R}] = \exp(i\pi/4)(\hat{x}\hat{x} + \hat{y}\hat{y})$ and their complex conjugates, which imply

$$[\mathcal{H}, \mathcal{R}] = e^{i\pi/4}(b/P)^{1/4} \mathcal{R} \quad [\mathcal{H}, \mathcal{L}] = -e^{i\pi/4}(b/P)^{1/4} \mathcal{L}. \quad (22)$$

The explicit expressions for E_0 and $\psi_0(r, u)$ in equations (16) and (17) may be derived from (21) and the condition $\mathcal{L}\psi_0 = \mathcal{L}^*\psi_0 = 0$ that the two lowering operators annihilate the ground state. It is much easier to obtain the asymptotic properties in the long chain limit this way than by evaluating the complete path integral.

Of course, the simplest method of all [1, 7, 16] for calculating the free energy per unit length of a semiflexible polymer confined by a harmonic potential is to impose the periodic boundary condition $r(t) = r(t + L)$, transform the action in the path integral (2) to the quadratic diagonal form $\frac{1}{2} \sum_q (Pq^4 + b)a_q \cdot a_{-q}$ by the linear transformation

$r(t) = L^{-1/2} \sum_q \alpha_q e^{iqt}$, and then use the equipartition theorem† $\frac{1}{2}(Pq^4 + b)(\alpha_q \cdot \alpha_{-q}) = 1$. However, it is not easy to obtain the results (11)–(15) for finite chains this way, and for non-harmonic confining potentials, the action cannot be diagonalized by a linear transformation.

Since the question as to the nature of the extrema of $\mathcal{E}[\Psi]$ in (9) has not been settled, it is useful to check the extremum principle for the harmonic confining potential $V(r) = \frac{1}{2}br^2$. For the Gaussian trial function $\Psi(r, u) = \exp(-\alpha\hat{r}^2 - \beta\hat{u}^2 + \gamma\hat{r} \cdot \hat{u})$,

$$\mathcal{E}(\alpha, \beta, \gamma) = \left[\frac{1}{4\alpha}(1 - \gamma^2) + \frac{1}{2\beta}(2\beta^2 + \gamma) \right] \left(\frac{b}{P} \right)^{1/4} \tag{23}$$

Here α, β , and γ are real and satisfy $\alpha\beta > \gamma^2/4$ in order that Ψ remain finite in the limit $\hat{r}, \hat{u} \rightarrow \infty$. The only extremum of $\mathcal{E}(\alpha, \beta, \gamma)$ in the allowed domain of variational parameters is a relative minimum $\mathcal{E} = \sqrt{2}(b/P)^{1/4}$ at $\alpha = \beta = 2^{-1/2}, \gamma = 1$, corresponding to the exact results for E_0 and ψ_0 in (16) and (17). Note that a lower boundary minimum $\mathcal{E} = 0$ is attained for $\alpha \rightarrow \infty, \beta \rightarrow 0$ in the subspace $\gamma = 0$, in which r and u are uncorrelated. Thus in the space of parameters (α, β, γ) , ψ_0 corresponds to a relative minimum but not an absolute minimum.

For an arbitrary central confining potential $V(r)$ that is non-negative and vanishes for $r \rightarrow \infty, E_0 = \mathcal{E}[\psi_0] > 0$. This is a consequence of the non-negative action in the path integral (2). For a trial function of the form $\Psi(r, u) = f(r)g(u)$

$$\mathcal{E}[fg] = \frac{1}{2P} \frac{\int_0^\infty du ug'(u)^2}{\int_0^\infty du ug(u)^2} + \frac{\int_0^\infty dr rV(r)f(r)^2}{\int_0^\infty dr rf(r)^2} \tag{24}$$

One can always choose $f(r)$ and $g(u)$ so that $\mathcal{E}[fg] \rightarrow 0$, as in the previous paragraph. Since $\mathcal{E}[\psi_0] > 0, \psi_0$ clearly does not correspond to an absolute minimum of $\mathcal{E}[\Psi]$. It should be noted that the trial function $\Psi(r, u) = f(r)g(u)$ is unphysical, since it neglects the correlation between r and u due to the first term in \mathcal{H} in equation (5). For trial functions with this form no contribution from the first term in \mathcal{H} appears in $\mathcal{E}[\Psi]$.

We now consider the semiflexible polymer confined in a tube of radius $R = D/2$. In terms of the dimensionless variables

$$\tilde{r} = R^{-1}r \quad \tilde{u} = P^{1/3}R^{-1/3}u \quad \tilde{t} = P^{-1/3}R^{-2/3}t \tag{25}$$

which eliminate the constants P and R in the path integral (2), the differential operator \mathcal{H} in equation (5) becomes

$$\mathcal{H} = P^{-1/3}R^{-2/3} \left[\tilde{u} \cdot \nabla_{\tilde{r}} - \frac{1}{2} \nabla_{\tilde{u}}^2 + \tilde{V}(\tilde{r}) \right] \tag{26}$$

where $\tilde{V}(\tilde{r})$ is infinite for $|\tilde{r}| > 1$ and vanishes for $|\tilde{r}| < 1$. From this expression and equation (7) it is clear that the confinement free energy $\Delta f = k_B T E_0$ has the expected form (1). The approximation of neglecting overhangs and a constraint on the total length in the path integral (2) is self-consistent as long as $\langle (d\tilde{r}/d\tilde{t})^2 \rangle \ll 1$. From equation (25) one sees that this requires $P \gg D$, as stated below equation (1).

The value of the constant c in equation (1) can be obtained, in principle, by solving the differential equation $(\mathcal{H} - E_0)\psi_0 = 0$ with the hard-wall boundary condition mentioned below (equation (3)). So far an exact solution with hard-wall boundary conditions has only been obtained for the half-space geometry [15]. Work on extending this solution to a tube with a rectangular cross section is in progress.

Further work is also required to see whether reliable estimates of c in (1) can be obtained with the extremum principle introduced above. The question of the nature of the extrema

† Note that for the harmonic potential Helfrich's assumption [1, 7] that all the modes are affected equally by the confinement, i.e. that $(\alpha_q \cdot \alpha_{-q})_{\text{confined}}^{-1} - (\alpha_q \cdot \alpha_{-q})_{\text{free}}^{-1}$ is independent of q , is exactly fulfilled.

has not yet been completely settled, and in devising a trial function one must take the hard-wall boundary condition, discussed below equation (3), into account.

Finally, we show that a lower bound for c in (1) follows from the results for the harmonic potential given above. Let us denote the path integral (2) for a polymer in a tube by $Z^{(\text{tube})}$, the path integral with the same endpoints r, u, r_0, u_0 and a harmonic confining potential by $Z^{(\text{har})}$, and the path integral for a polymer in a tube with, in addition, a harmonic potential within the tube by $Z^{(\text{tube+har})}$. Since the paths that contribute to $Z^{(\text{tube+har})}$ are a subset of the paths contributing to $Z^{(\text{har})}$,

$$Z^{(\text{har})} > Z^{(\text{tube+har})} = Z^{(\text{tube})} \left\langle \exp \left(-\frac{1}{2} b \int_0^t dt r(t)^2 \right) \right\rangle^{(\text{tube})} > Z^{(\text{tube})} \exp \left(-\frac{1}{8} b D^2 t \right) \quad (27)$$

where the restriction $|r| \leq D/2$ for the polymer in the tube has been used. Together with equations (7) and (16) this inequality implies

$$\frac{\Delta f^{(\text{tube})}}{k_B T} > \frac{\Delta f^{(\text{har})}}{k_B T} - \frac{1}{8} b D^2 = \sqrt{2} \left(\frac{b}{P} \right)^{1/4} - \frac{1}{8} b D^2 \quad (28)$$

for the confinement free energy per unit length.

Equation (28) also follows from the assumption that the exact eigensolution $\psi_0^{(\text{har})}$ for the harmonic potential yields a lower value of $\mathcal{E}^{(\text{har})}[\psi]$, defined by (5) and (9), than the exact $\psi_0^{(\text{tube})}$ for the polymer in a tube, i.e. $\mathcal{E}^{(\text{har})}[\psi_0^{(\text{har})}] < \mathcal{E}^{(\text{har})}[\psi_0^{(\text{tube})}]$. This implies

$$E_0^{(\text{har})} < E_0^{(\text{tube})} + \frac{1}{2} b \langle r^2 \rangle^{(\text{tube})} < E_0^{(\text{tube})} + \frac{1}{8} b D^2 \quad (29)$$

which, using (7) and (16), is equivalent to (28).

There is a comparable bound in quantum mechanics. Using the ground state of a particle in a box of width D as a trial function in a variational estimate of the ground state energy of the one-dimensional harmonic oscillator, one obtains

$$E_0^{(\text{box})} > E_0^{(\text{har})} - \frac{1}{8} m \omega^2 D^2 \quad (30)$$

in analogy with (28) and (29). Substituting $E_0^{(\text{har})} = \frac{1}{2} \hbar \omega$ in equation (30) and maximizing the right side with respect to ω gives $E_0^{(\text{box})} > \hbar^2 / (2mD^2)$. The exact result $E_0^{(\text{box})} = \pi^2 \hbar^2 / (2mD^2)$ is consistent with this bound but not very close to it.

Choosing b to maximize the right-hand side of (28) for fixed P and D , one obtains

$$\frac{\Delta f^{(\text{tube})}}{k_B T} > \frac{3}{2} P^{-1/3} D^{-2/3} \quad (31)$$

or $c > \frac{3}{2}$ in equation (1). The estimate $c = 2.46 \pm 0.07$ from the computer simulations of Dijkstra *et al* [7] is consistent with this bound.

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