Toroids versus racquets: the collapse of semiflexible polymers of finite thickness

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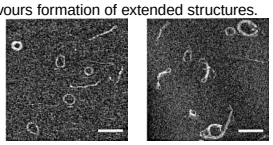
Abstract. *We calculate the minimal energy shapes of a semiflexible polymer in a poor solvent. Following Schnurr, MacKintosh et al. our conformational energy includes the bending elastic component and the surface energy. We take into account the finite thickness of the molecule and reconsider the relative stability of rod-like, toroidal and "racquet" conformations. The main result is presented as a phase diagram computed for relatively short effective length. In agreement with earlier results in the zero-thickness approximation, thin filaments collapse into toroidal shapes. However, beyond a critical thickness, the kinetically preferred racquet state also turns out to be the ground state.*

Introduction

The conformation of the semiflexible polymer (e.g., F-actin or DNA) is determined by the balance of the surface energy and the bending energy. The first is responsible for tendency of the polymer to form the most compact state in a poor solvent while the second favours formation of extended structures.



AFM images of condensates formed with pBR322 and cationic pegylated-poly(amido-amine) [2]. Scale bars = 300 nm.

The polymer's resulting shapes, also observed experimentally, are represented by rods, racquets and toroids. Numerical simulations of the condensation process reveal existence of long-lived intermediate states [5, 4].

The conformational energy of such shapes was computed in [6] under assumption that the polymer has negligible thickness. It was concluded that final states are always toroids.

Here we show that taking into account thickness of the molecule can lead to the situation when the rod or racquets may become more preferable from energetical point of view, thus they may be considered as final stable states.

Toroids

For a molecule of length L, the bending energy is

$$U_{bend} = \frac{B}{2}\int_0^L \varkappa^2(s)ds,$$

where B is the bending stiffness and $\varkappa(s)$ is the curvature of the centreline.

For a N-fold bundle of length L_N, the surface energy can be approximated as

$$U_N = \gamma\alpha_N L_N,$$

where α_N is the coordination number ($\alpha_1 = 3$, $\alpha_2 = 5$, $\alpha_3 = 6, \ldots$), γ is a coefficient.

In what follows, all the lengths and energies will be presented in the dimensionless form by normalizing them on the *condensation length* $L_c = \sqrt{B/\gamma}$ and the *condensation energy* $U_c = \sqrt{B\gamma}$, respectively.

Consider a tube with helical centreline $x = \rho\cos\varphi$, $y = \rho\sin\varphi$, $z = \frac{g}{2\pi}\varphi$, its curvature $\varkappa = \rho\left(\rho^2 + \frac{g^2}{4\pi^2}\right)^{-1} = const$, g is the pitch parameter. The length of the helix is given by

$$\lambda = \sqrt{\rho^2 + \frac{g^2}{4\pi^2}}\,\varphi.$$

To find the length of one coil λ_1, we first compute a root which is close to 2π of the equation

$$\rho^2\sin\varphi_1 + \frac{g^2}{4\pi^2}\varphi_1 = 0. \qquad (1)$$

The surface energy for the thick toroid $1+$ (the length of the centreline exceeds the length of one coil but less then two coils):

$$u_{s1+} = \alpha_1\lambda_1 + (\alpha_2 - \alpha_1)(\lambda - \lambda_1) = \\ = (2\alpha_1 - \alpha_2)\lambda_1 + (\alpha_2 - \alpha_1)\lambda.$$

The conformational energy is a sum of the bending and surface energy

$$u_{1+} = \frac{\lambda\rho^2}{2\left(\rho^2 + \frac{g^2}{4\pi^2}\right)^2} + \varphi_1\sqrt{\rho^2 + \frac{g^2}{4\pi^2}} + 2\lambda. \qquad (2)$$

Assuming that the successive coils are tightly packed, we can find the (squared) distance between the closest points on the centreline

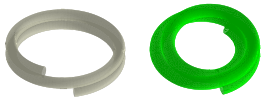
$$\delta^2 = \frac{g^2}{4\pi^2}\varphi_1\left(\varphi_1 - 2\tan\frac{\varphi_1}{2}\right). \qquad (3)$$

Eliminating the radius ρ from Eq. (2) with help of Eq. (1) and expressing the parameter g as function of the thickness δ from Eq. (3), we obtain the conformational energy $u_{1+} = u_{1+}(\lambda, \delta; \varphi_1)$. To detect its minimum, we solve $\frac{\partial u_{1+}}{\partial\varphi_1} = 0$ for the angle φ_1.

Now the optimal comformational energy can be represented as function of λ and δ.

The surface energy for the thick toroid $2+$ (number of coils lies between 2 and 3)

$$u_{s2+} = \alpha_2\lambda_1 + (\alpha_3 - \alpha_2)(\lambda - 2\lambda_1) = \\ = (3\alpha_2 - 2\alpha_3)\lambda_1 + (\alpha_3 - \alpha_2)\lambda.$$



Left: Optimal toroid $1+$ ($\lambda = 12$, $\delta = 0.25$, $\rho = 1.24$). Right: Toroid $2+$ ($\lambda = 13$, $\delta = 0.25$).

Racquets

We model the racquet head as a planar elastica. The tangents to its centreline are antiparallel and orthogonal to the end force. The centreline end points are separated by the thickness diameter δ so that the tube touches itself to continue as a bundle of straight rods. The head may be either at one or both ends.

The bending energy of the head is computed as [8]

$$u_{head} = \frac{4\xi^2(k)}{\chi}\left[(2k^2 - 1) + \frac{\delta}{\chi}\right],$$

where χ stands for the length of the head's centreline and $\xi(k)$ is a function of the elliptic modulus k, $1/\sqrt{2} \le k \le 1$, which can be found from the boundary condition

$$\frac{\delta}{\chi} = 2\frac{\eta(k)}{\xi(k)} - 1. \qquad (4)$$

The shape of a head can be either 1) Ω-like (with two inflection points) or 2) U-like (without inflection points). In the first case we have

$$\xi(k) = 2K(k) - F\left(\frac{1}{k\sqrt{2}}, k\right),$$

$$\eta(k) = 2E(k) - E\left(\frac{1}{k\sqrt{2}}, k\right),$$

and in the second case

$$\xi(k) = F\left(\frac{1}{k\sqrt{2}}, k\right), \quad \eta(k) = E\left(\frac{1}{k\sqrt{2}}, k\right).$$

Here K, F and E are standard notations for the elliptic integrals.

The total conformational energy for the single and double racquets are respectively

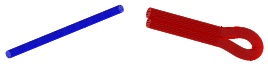
$$u_1 = u_{head} + \frac{1}{2}(\chi + 5\lambda), \quad u_2 = 2u_{head} + 2(\chi + \lambda), \qquad (5)$$

Given the thickness δ, we express the head's length χ from Eq. (4) and substitute it into Eq. (5) which allows us to obtain the total energy as function of k and the parameters δ and λ

$$u_1 = v_1 + v_2 + \frac{5}{2}\lambda, \quad u_2 = 2v_1 + 4v_2 + 2\lambda,$$

where $v_1 = \frac{8}{\delta}[2\eta(k) - \xi(\eta)][(k^2 - 1)\xi(k) + \eta(k)]$ and $v_2 = \frac{\delta\xi(k)}{2[2\eta(k) - \xi(k)]}$.

Minimization of u_1 and u_2 relative to k gives the optimal conformation.



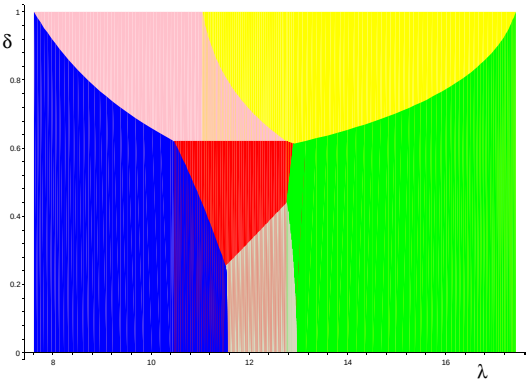
Left: Rod ($\lambda = 10$, $\delta = 0.5$). Right: Single-head racquet ($\lambda = 12$, $\delta = 0.5$).

Left: Single-head racquet ($\lambda = 11$, $\delta = 0.7$). Right: Double-head racquet ($\lambda = 13$, $\delta = 0.7$).

Phase diagram

The parameters of optimal toroids $1+$, $2+$ and racquets were computed for a range of length λ and thickness δ and their conformational energies were compared. The regions on the parameter plane were found where the energy of a particular conformation is minimal.

The results are presented below as a phase diagram.



blue: rods

red: thick single racquets (Ω-like)

wheat: toroids $1+$ (thick helices)

pink: thick single racquets (hairpins)

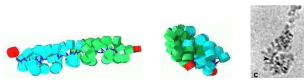
green: toroids $2+$

yellow: thick double racquets (staples)

Concluding remarks

It is shown on a simple model that excluded volume effects can make racquet-like conformations preferable for polymers with relatively large persistence length. There exists a range of the polymer's effective length for which the racquet-like conformations are the ground states even for relatively thin molecules. Numerical experiments with a bead-spring model support this result [5, 7].

Computer simulations of the 30-nm chromatin fibre reveal racquet-like structures with hairpins [3]. If the bending energy cost of sharp kinks is smaller than that we have used in this work, then the conformations with hairpins become even more competitive with respect to toroids.



Left & Middle: Simulation of the chromatin: stretched fibre and hairpin [3]. Right: Cryoelectron microscopy of unfixed, unstained polynucleosomes [1]. Note a sharp kink of the fibre.

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