A Toroidally Folded Monomolecular DNA Chain

—From the perspective of colloidal physics—

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DNA は荷電高分子として振る舞い静電相互作用を通して溶液中の様々なイオンとの間に複合体を作っている。溶液条件によってはこのような複合体は分子全体を覆い DNA のセグメント同士が凝集したグロビュールと呼ばれる構造を作ることがある。T4DNA の場合、典型的なグロビュールはトロイド構造をしており、このことから、グロビュールへの折りたたみ転移は分子鎖上でのループ形成とループ構造同士の凝集という二つの過程を通して理解できる。本発表では折りたたみ転移の平衡論における一般論を展開し、また、wormlike chain モデルに基づいてトロイド構造の持つべき自由エネルギーを導く。これらより得られた理論的予想は実際の実験における DNA の折りたたみ転移挙動をよく記述するものとなっている。

1   Abstract

DNA is a double-helical polyelectrolyte molecule that has an entire length ranging from several nanometers to centimeters. In principle, DNA is a very stiff molecule, and the chain shorter than its persistence length ($\simeq$ 50nm) behaves as a stiff rod. On the other hand, most of natural DNA molecules have the sizes much longer than this length, and they exhibit a nature of soft matter even in the level of a monomolecular chain. For example, a single T4 DNA (57μm) chain undergoes the transition from a fluctuating coiled state to a compact condensed state (a coil-globule transition) in the presence of various ionic species. Such a compact state is, after all, an aggregate of a number of DNA segments that are chemically linked with each other one-dimentionally. Typically, a condensed structure of T4 DNA exhibits a toroidally folded ring structure. In this case, the coil-globule transition of DNA may be characterized by the following two processes; the formation of loops and their aggregation along a single chain.

For a DNA molecule, each monomer size is significantly smaller than the persitence length,

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and thus, a continuous model works satisfactorily. The chain conformation is then characterized by l_s, the total length of the string segments and $\rho(\nu, \ell)$, the existing probability density of the aggregate of ν loops with the mean loop length of ℓ. The combination of string segments and rings in this chain is given as

$$W = \frac{\prod_{\nu,\ell} l_s{}^{\rho(\nu,\ell)}}{\prod_{\nu,\ell} \rho(\nu,\ell)!} \tag{1}$$

Using a free energy of a ring $f(\nu, \ell)$ and that of a string segment per unit length f_s, the total free energy for the chain with a rings-on-a-string structure is given as

$$F[\rho] = \int_A \rho f \, d\nu \, d\ell + k_B T \int_A \rho(\ln(\rho/l_s) - 1) \, d\nu \, d\ell - f_s(L - l_s) \tag{2}$$

where the domain A is given as $\{(\nu, \ell) | 1 \leq \nu < \infty, 0 \leq \nu\ell \leq L - l_s\}$. L is the entire length of the chain. The conservation of mass is written as

$$\int_A \rho\nu\ell \, d\nu \, d\ell = L - l_s \tag{3}$$

In this presentation, we derive the free energy of a ring structure $f(\nu, \ell)$ on the basis of a wormlike chain model, and solve the minimization of the total free energy. A ring is formed by a polyelectrolyte chain, and is stabilized by complex with (multivalent) counterions, which leads to the charge neutralization of this ring. We model this ring as a structure where only its surface is electrically charged. Applicability of this framework to an actual experiment, especially the dependence of the transitional behavior on the ionic strength is discussed. In principle, the folding transition of T4 DNA is all-or-none type transition in a low-salt condition, and the partially folded chain appears stably only in a high-salt condition. The present formalism semiquantitatively explains the experimental trend with a fairly reasonable setting of parameters.

References

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