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Single molecular statistics of an optically tweezed polymer: A theoretical consideration

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Abstract

The statistics of a polymer chain has been conventionally investigated mainly by scattering. Instead, we theoretically examine the possibility of obtaining the statistics from single molecular observation. Motivated by this we discuss the statistical distribution of an ideal linear chain with its ends constrained, which could be directly obtained in principle by an experiment on an optically tweezed single molecule. We also derive the scattering function pertinent to the situation. Our consideration could open a way to experimentally study the statistics of a linear polymer chain with the ends fixed to obtain that of a ring polymer in lower dimension.

1. Introduction

The central method to investigate the correlation of polymer chains has been scattering experiments. There, the quantities in the momentum space such as the structure factor are easy to access, which can be efficiently used to obtain the real space information [1].

Recently, single molecular observation of large chain molecules such as DNA and optical tweezing of such molecules have become feasible for us [2–6]. Contrary to the scattering, the recent single molecular observation is basically a real space method. We consider below the possibility to directly obtain the distribution function of the polymer statistics by the single molecular observation. In connection with this we discuss the probability distribution and scattering function of a polymer with its ends constrained by two optical tweezers.

2. Convenient observable in single molecular observation

We consider a chain molecule which can be regarded as composed of N monomers, where the distance between the adjacent two monomers corresponds to a Kuhn length a. We mark any two monomers of the chain in a certain way, possibly by introducing fluorescent dyes. The two marked monomers are called mth and nth monomers. In the single molecular observation of this polymer, we can directly observe thermal fluctuation of a chain molecule by a series of snapshots. From such M snapshots, we obtain the probability of the vector from the nth to mth monomers, or the n-to-m vector $\mathbf{R}_{mn} = \mathbf{r}_m - \mathbf{r}_n$, to be $\mathbf{r}$:

$$g_{mn}(\mathbf{r}) = \frac{1}{M}\sum_{k=1}^{M} \delta^{(3)}\left(\mathbf{r} - \mathbf{R}_{mn}^{(k)}\right) \tag{1}$$

where $\delta^{(3)}$ is the three-dimensional delta function of Dirac. Here, $\mathbf{R}_{mn}^{(k)}$ is $\mathbf{R}_{mn}$ of the kth snapshot.

Since we can expect that the long time average is equal to the statistical average, we can express the above quantity by the distribution function, $P_{mn}(\mathbf{R}_{mn})$, that the n-to-m vector to be $\mathbf{R}_{mn}$.

$$g_{mn}(\mathbf{r}) = \int \mathrm{d}\mathbf{R}_{mn}\delta^{(3)}(\mathbf{r} - \mathbf{R}_{mn})P_{mn}(\mathbf{R}_{mn}) = P_{mn}(\mathbf{r}) \tag{2}$$

From Eqs. (1) and (2), we conclude that we can determine the distribution $P_{mn}(\mathbf{r})$, which governs the statistics of the polymer chain, by the single molecular experiment described above: *We can directly obtain the distribution*

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function $P_{mn}(\mathbf{r})$ for a chain with or without the excluded volume that the n-to-m vector to be $\mathbf{r}$ *in experiments.*

To obtain the quantity in Eq. (1) experimentally, we need to know three dimensional configuration for each snapshots. Experimentally, two-dimensional projection to a certain plane of such three-dimensional configuration is far easier to access. Even from such two-dimensional experiments, we can obtain one-dimensional probability by measuring $R^{(k)}_{mn,i}$ of each snapshot:

$$g_{mn,i}(r_i) = \frac{1}{M}\sum_{k=1}^{M}\delta\left(r_i - R^{(k)}_{mn,i}\right) \tag{3}$$

where i can be x,y, or z and $\delta(r)$ is the Dirac's delta function in one dimension. Here and hereafter, x_i denotes the i component of a vector $\mathbf{x}$.

The quantity is also important especially when the distribution function $P_{mn}(\mathbf{r})$ can be factorized as

$$P_{mn}(\mathbf{r}) = \prod_{i=x,y,z} P_{mn,i}(r_i) \tag{4}$$

This is because in such a case the quantity $P_{mn,i}(r_i)$ can be directly observed via single molecular observation due to the relation (the one-dimensional counterpart of Eq. (2)):

$$g_{mn,i}(r_i) = P_{mn,i}(r_i) \tag{5}$$

When the statistics of a polymer can be cast into the Gaussian distribution of the form

$$P_{mn,i}(r_i) = \frac{\exp\left(-\frac{(r_i-\langle R_{mn,i}\rangle)^2}{2\langle \Delta R^2_{mn,i}\rangle}\right)}{\sqrt{2\pi\left\langle \Delta R^2_{mn,i}\right\rangle}} \tag{6}$$

the single molecular observation quantifying $g_{mn,i}(r_i)$ in Eq. (3) directly probing this quantity.

3. Statistics of a tweezed polymer

With a pair of optical tweezers, we can fix the two ends of a linear chain, possibly virtually without fluctuation. This constraint suppresses the translational Brownian motion of the molecule, which would be an advantage in experiment because of no need to track the molecule moving around in the field of microscope.

In this section, we discuss the statistics of a linear chain with such constraint. First we deal with a polymer with its ends fixed completely without fluctuation, which is followed by the discussion on a polymer with its end fixed but not completely without fluctuation. In this section, we limit ourselves to an ideal chain where the excluded volume effect is neglected, although the experiment suggested in the previous section is not restricted to an ideal chain.

3.1. Polymer chain with its ends fixed without fluctuation

Recently, we discuss the distribution of the n-to-m vector of a linear ideal chain to be $\mathbf{r}$ when the end-to-end distance is fixed to $\mathbf{R}_0$ without fluctuation (Fig. 1) [7] by generalizing the strategy for a ring polymer described in [8]:

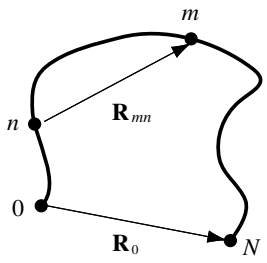

Fig. 1. Random walk with the end-to-end distance fixed to $\mathbf{R}_0$.

$$P^{(0)}_{mn}(\mathbf{r}|\mathbf{R}_0) = \prod_{i=x,y,z} P^{(0)}_{mn,i}(r_i) \tag{7}$$

where $P^{(0)}_{mn,i}(r_i)$ is given by the right-hand side of Eq. (6) with

$$\langle R_{mn,i}\rangle = \frac{m-n}{N}R_{0,i} \tag{8}$$

$$\left\langle \Delta R^2_{mn,i}\right\rangle = \frac{|m-n|a^2}{3}\left(1-\frac{|m-n|}{N}\right) \tag{9}$$

Note that, from Eqs. (8) and (9), we have $\langle \mathbf{R}_{N0}\rangle = \mathbf{R}_0$ and $\langle \Delta\mathbf{R}^2_{N0}\rangle \equiv \sum_{i=x,y,z}\langle \Delta R^2_{mn,i}\rangle = 0$: the end-to-end vector $\mathbf{R}_{N0}$ is fixed to $\mathbf{R}_0$ without fluctuation, as desired. The constraint discussed here is similar in spirit to but incompatible with the works [9–11] appreciated in the context of a polymer chain in networks. Details of the comparison with previous works are given in [7].

The distribution $P_{mn}(\mathbf{r}|\mathbf{R}_0)$ in Eq. (7) could be directly checked through the two dimensional single molecular observation, thanks to Eqs. (3) and (5). The features of this distribution are as follows. Eq. (8) states that the average of the n-to-m vector $\mathbf{R}_{mn}$ scales 'affinely' with the end-to-end vector $\mathbf{R}_0$, or the 0-to-N vector: the average of $\mathbf{R}_{mn}$ is proportional to $m-n$. On the contrary, Eq. (9) predicts that the average of $\Delta\mathbf{R}^2_{mn}$ scales 'non-affinely' with $\Delta\mathbf{R}^2_{N0}$: the variance becomes zero when $m-n$ approaches N (the end-to-end vector is fixed without fluctuation), but scales as $m-n$ when $m-n$ gets smaller or changes 'affinely' with Na^2, the variance of the end-to-end vector of an unconstrained linear chain: *at small distances the monomers do not feel any constraint while at large scales they feel the constraint.*

In the following, we briefly derive the one-dimensional version of Eqs. (7)–(9) in two ways different from and complementary to the arguments in [7]. One way, which is an extension of the arguments for a ring polymer in [12] to a fixed-end polymer, is based on the characteristic function [13] and the other is on the conditional probability. We consider below a one-dimensional N step random walk from $x = x_0$ to $x = x_N$ where the end-to-end distance $x_N - x_0$ is fixed to X. In the first case of the characteristic function each step length $u_i = x_i - x_{i-1}$ $(i = 1,\ldots,N)$ is determined by a Gaussian probability of variance a^2 $(\sim \mathrm{e}^{-u_i^2/2a^2})$ while in the second case of the conditional

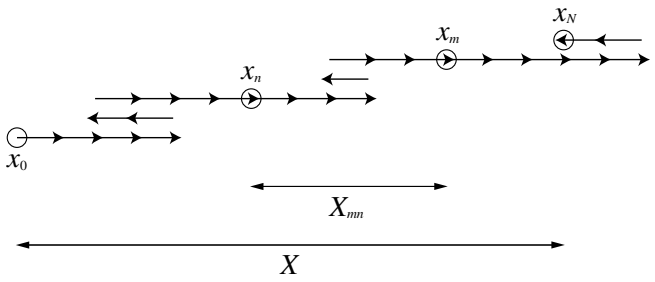

Fig. 2. Random walk in one dimension with the end-to-end distance fixed to X from $x = x_0$ to $x = x_N$ for $n < m$.

probability each step randomly selects either $+a$ or $-a$ as in Fig. 2. Though the two models are different, the resulting probability distribution for the n-to-m vectors are the same as we see below.

3.1.1. Characteristic function

By using the Dirac's delta function, the Gaussian distribution of the step vectors u_i under the desired constraint can be written as

$$P(u) \sim \delta(u_1 + u_2 + \cdots + u_N - X)\mathrm{e}^{-(u_1^2+u_2^2+\cdots u_N^2)/2a^2} \tag{10}$$

The characteristic function is here defined by

$$\phi(k) = \left\langle \mathrm{e}^{i(k_1u_1+k_2u_2+\cdots+k_Nu_N)} \right\rangle \tag{11}$$

where the average is taken by the distribution, $P(u)$. After a straightforward calculation we obtain

$$\phi(k) = \mathrm{e}^{-\frac{a^2}{2}\left(k_1^2+k_2^2+\cdots k_N^2-\frac{1}{N}\left(k_1+k_2+\cdots+k_N+\frac{iX}{a^2}\right)^2\right)} \tag{12}$$

From this expression, we find the average and variance ($m > n$) to recover one-dimensional version of Eqs. (8) and (9).

3.1.2. Conditional probability

The distribution $\overline{P}(X_{mn} \mid X)$ of the distance $x_m - x_n$ to be X_{mn} under the fixed end condition is given by the conditional probability

$$\overline{P}(X_{mn}|X) = \frac{P_{|m-n|}(X_{mn})\widetilde{P}_{N-|m-n|}(X_{mn}, X)}{P_N(X)} \tag{13}$$

where $P_M(x)$ is the standard Gaussian probability of a random walk: $P_M(x) = \exp\left(-\frac{x^2}{2Ma^2}\right)\Big/\sqrt{2\pi Ma^2}$. Here, the probability $\widetilde{P}_{N-|m-n|}(X_{mn}, X)$ is the quantity to be calculated as follows for $m > n$ (a similar derivation is possible also for $n > m$). We consider a one dimensional n-step random walk from $x = x_n$, $x_{n-1}, \ldots$ to $x = x_0$, followed by a fixed step $+X$ to $x = x_N$, which is completed by another $(N - m)$-step random walk from $x = x_N, x_{N-1}, \ldots$ to $x = x_m$: each step in the two random walks selects randomly either $+a$ or $-a$. When the distance $x_m - x_n$ is constrained to a fixed value X_{mn}, every walk consisting of the above two independent random walks (i.e., n-step and $(N - m)$-step walks) plus a fixed step $+X$ should satisfy the following two equations: (1) $M_+ + M_- = n + (N-m)$ and (2) $aM_+ - aM_- + X = X_{mn}$ where M_+ and M_- represent the total numbers of '$+a$'-step and '$-a$'-step in the walk, respectively. If we count the number of distinct ways to realize these equations, we obtain, for $N - m + n$, M_+, $M_- \gg 1$,

$$\widetilde{P}_{N-m+n}(X_{mn}, X) = \frac{\exp\left(-\frac{(X_{mn}-X)^2}{2(N-m+n)a^2}\right)}{\sqrt{2\pi(N-m+n)a^2}} \tag{14}$$

Substituting Eq. (14) into Eq. (13), we obtain one-dimensional version Eqs. (8) and (9).

3.2. Polymer chain with its ends fixed with fluctuation

The optical trapping would imply small fluctuations around a fixed point. If such fluctuations could not be negligible, the distribution of the end points $\mathbf{r}_0$ and $\mathbf{r}_N$ could be expressed as

$$P(\mathbf{r}_0, \mathbf{r}_N) = \frac{\exp\left(-\frac{(\mathbf{r}_0-\mathbf{a})^2}{2\sigma_0^2/3}\right)}{(2\pi\sigma_0^2/3)^{3/2}} \cdot \frac{\exp\left(-\frac{(\mathbf{r}_N-\mathbf{b})^2}{2\sigma_N^2/3}\right)}{(2\pi\sigma_N^2/3)^{3/2}} \tag{15}$$

where $\mathbf{a}$, $\mathbf{b}$ and σ_0, σ_N are the average and the standard deviation of the trapped end points of the 0th and Nth segments, respectively.

With using Eqs. (7) and (15), the distribution of the n-to-m vector to be $\mathbf{r}$ when the end points are fixed by optical tweezers by an average distance $\mathbf{R}$ could be calculated as

$$P_{mn}^{(f)}(\mathbf{r}|\mathbf{R}) = \int \mathrm{d}\mathbf{r}_0 \int \mathrm{d}\mathbf{r}_N P(\mathbf{r}_0, \mathbf{r}_N) P_{mn}^{(0)}(\mathbf{r}|\mathbf{R}_0) \tag{16}$$

with

$$\mathbf{R}_0 = \mathbf{r}_N - \mathbf{r}_0 \tag{17}$$

and

$$\mathbf{R} = \mathbf{b} - \mathbf{a} \tag{18}$$

Then, $P_{mn}^{(f)}(\mathbf{r} \mid \mathbf{R})$ is given in the form

$$P_{mn}^{(f)}(\mathbf{r}|\mathbf{R}) = \prod_{i=x,y,z} P_{mn,i}^{(f)}(r_i) \tag{19}$$

where $P_{mn,i}^{(f)}(r_i)$ is given by the right-hand side of Eq. (6) with

$$\langle R_{mn,i} \rangle = \frac{m-n}{N} R_i = \frac{m-n}{N}(b_i - a_i) \tag{20}$$

$$\left\langle \Delta R_{mn,i}^2 \right\rangle = \frac{|m-n|a^2}{3} + \left(\frac{m-n}{N}\right)^2 \frac{\sigma_0^2 + \sigma_N^2 - Na^2}{3} \tag{21}$$

Comparing Eqs. (9) and (21), we notice that the fluctuation of the optical tweezer could be neglected only when the fluctuations are much smaller than the fluctuation of the linear chain:

$$\sigma_0^2, \sigma_N^2 \ll Na^2 \tag{22}$$

As the fluctuation gets larger and $\sigma_0^2 + \sigma_N^2 - Na^2$ approaches zero, $\langle \Delta R_{mn,i}^2 \rangle$ recovers the 'affine' property. When the fluctuation is very large ($\sigma_0^2, \sigma_N^2 > Na^2$), $\langle \Delta R_{mn,i}^2 \rangle$ is expected to behave in a rather abnormal way although such a situation is probably difficult to be observed by

experiments: $\langle \Delta R^2_{mn,i} \rangle$ scales as $(\frac{m-n}{N})^2(\sigma_0^2 + \sigma_N^2)$. The experimental confirmation of these distinctive regimes and/or the turnover among them would be a challenging future problem.

In what follows, we also derive the scattering function for an ideal chain where its ends are fixed by optical tweezers with possible fluctuations. Although it is not a direct observable in real-space experiments, the corresponding quantity in the wave-number space could be useful (this is just as if one might often analyze time-dependent data in the frequency space to extract the character of the original data) so that we here derive the scattering function for completeness: this quantity could be obtained in principle from experiment once one obtains experimentally the real-space observable $P^{(f)}_{mn}(\mathbf{r} \mid \mathbf{R})$. The scattering function is defined by

$$S(\mathbf{q}) = \frac{1}{N^2} \sum_{m,n=1}^{N} \langle \mathrm{e}^{i\mathbf{q}\cdot\mathbf{R}_{mn}} \rangle = \mathrm{Re}[S_0(\mathbf{q})] \tag{23}$$

where

$$S_0(\mathbf{q}) = \frac{2}{N^2} \int_0^N \mathrm{d}m \int_0^m \mathrm{d}n \langle \mathrm{e}^{i\mathbf{q}\cdot\mathbf{R}_{mn}} \rangle \tag{24}$$

For the Gaussian distribution of the form in Eq. (6), we can directly show

$$\langle \mathrm{e}^{i\mathbf{q}\cdot\mathbf{R}_{mn}} \rangle = \mathrm{e}^{i\mathbf{q}\cdot\langle \mathbf{R}_{mn} \rangle} \cdot \mathrm{e}^{-\frac{1}{6}\mathbf{q}\cdot\mathbf{q}\langle \Delta\mathbf{R}^2_{mn} \rangle} \tag{25}$$

Substituting $\langle \mathbf{R}_{mn} \rangle$ and $\langle \Delta\mathbf{R}^2_{mn} \rangle$ for the constraint in question, given in Eqs. (20) and (21) into this, we obtain

$$\langle \mathrm{e}^{i\mathbf{q}\cdot\mathbf{R}_{mn}} \rangle = \mathrm{e}^{i\tilde{q}_L \widetilde{R}(\bar{m}-\bar{n})} \cdot \mathrm{e}^{-\tilde{q}^2\{(\bar{m}-\bar{n})-c(\bar{m}-\bar{n})^2\}} \tag{26}$$

where q and q_L is the magnitude of the momentum vector $\mathbf{q}$ and its projection to the $\mathbf{R}$ direction, respectively, and c is defined as

$$c = 1 - \frac{\sigma_0^2}{Na^2} - \frac{\sigma_N^2}{Na^2} \tag{27}$$

In the above, n and m are normalize by N ($\bar{n} = n/N$ and $\bar{m} = m/N$) and the momentum and position vectors are normalized by $R_g = \sqrt{Na^2/6}$:

$$\tilde{q} = qR_g, \ \tilde{q}_L = q_L R_g \tag{28}$$

$$\tilde{R} = R/R_g \tag{29}$$

Setting $t = \bar{m} - \bar{n}$ and changing the integration variables from $(\bar{m}, \bar{n})$ to $(\bar{m}, t)$ in Eq. (24) and then changing the order of integrations over $\bar{m}$ and t, we obtain

$$S_0(\mathbf{q}) = 2\int_0^1 \mathrm{d}\bar{m} \int_0^{\bar{m}} \mathrm{d}t F(t) = 2\int_0^1 \mathrm{d}t(1-t)F(t) \tag{30}$$

where

$$F(t) = \mathrm{e}^{i\tilde{q}_L \widetilde{R}\cdot t} \cdot \mathrm{e}^{-\tilde{q}^2(t-ct^2)} = \mathrm{e}^{-Q_-^2} \mathrm{e}^{\tilde{q}^2 c\left(t - \frac{Q_-}{\tilde{q}\sqrt{c}}\right)^2}, \tag{31}$$

where Q_- is defined as

$$Q_- = \frac{\tilde{q}^2 - i\tilde{q}_L\tilde{R}}{2\sqrt{c}\tilde{q}}. \tag{32}$$

With using $Q_+ = 1 - Q_-/(\tilde{q}\sqrt{c})$, or

$$Q_+ = \frac{(2c-1)\tilde{q}^2 + i\tilde{q}_L\tilde{R}}{2\sqrt{c}\tilde{q}}, \tag{33}$$

we further calculate Eq. (30):

$$S_0(\mathbf{q})/2 = \mathrm{e}^{-Q_-^2} \times \int_0^1 \left\{ \frac{Q_+}{\tilde{q}\sqrt{c}} - \left(t - \frac{Q_-}{\tilde{q}\sqrt{c}} \right) \right\} e^{\tilde{q}^2 c\left(t - \frac{Q_-}{\tilde{q}\sqrt{c}}\right)^2} \mathrm{d}t, \tag{34}$$

to obtain the final result:

$$S(\mathbf{q}) = \mathrm{Re}\left[\frac{e^{-Q_-^2}}{c\tilde{q}^2} \left\{ (e^{Q_-^2} - e^{Q_+^2}) + 2Q_+ \left[\mathrm{erfi}(Q_+) + \mathrm{erfi}(Q_-) \right] \right\} \right], \tag{35}$$

which reduces to the expression, Eq. (26), in [7], when $c = 1$ ($\sigma_0^2 = 0, \sigma_N^2 = 0$). Here, we have introduced the imaginary error function as

$$\mathrm{erfi}(Q) = \frac{2}{\sqrt{\pi}} \int_0^Q \mathrm{d}z \exp(z^2)$$

4. Statistics of a ring polymer

If a chain in three dimension whose ends are fixed is projected onto a plane perpendicular to the end-to-end vector $\mathbf{R}_0$, the chain should look like a ring polymer (which has been actively studied [15]) in the two-dimensional space; likewise the statistics of a linear chain fixed at the both ends in two dimension corresponds to that of a ring polymer in one dimension: Instead of synthesizing a ring polymer, we could study a linear chain polymer via optical tweezers and single molecular observation to investigate the correlation of a ring polymer. For example, when $\mathbf{R}_0$ points the z direction, the snapshots on the xy plane is important for the investigation for a ring polymer. From the $g_{mn,x}(r_x)$ measurement from the snapshots on the xy plane for an ideal linear polymer in θ solvent with its ends fixed to $(0, 0, R_0)$, we obtain $P^{(0)}_{mn,x}(r_x) \equiv f(r_x)$ (or equivalent $P^{(0)}_{mn,y}(r_y)$). Then, we obtain the distribution of a ring polymer as $f(r_x)f(r_y)f(r_z)$ to be compared with the result first obtained in [14], which is a special case ($\mathbf{R}_0 = 0$) of Eq. (6) with Eqs. (8) and (9).

For a ring polymer with excluded volume, although the factorization property would be lost, the statistics of a ring polymer in two dimension can be investigated via the xy snapshots, if we find the probability of the projection of $\mathbf{R}_{mn}$ onto the xy plane to be $\mathbf{r}$ (a vector on the xy plane), or the quantity $g_{mn,x}(r_x)g_{mn,y}(r_y)$ through the xy snapshots.

5. Conclusion

In this letter, we theoretically discuss a possible way of studying the statistics of a chain via single molecular observation with optical tweezers *for a chain with or without the excluded volume*. We calculate some observables suitable for an optically tweezed ideal chain. First, we show that two different one-dimensional models of a linear chain with its end fixed lead to the same probability for the n-to-m vector. Second, we calculate the n-to-m probability and scattering function when a linear chain is constrained with a certain fluctuation at both ends. Third, we point out that the statistics of a linear chain fixed at the both ends corresponds to that of a ring polymer in lower dimensions. There should be a lot of difficulties in performing and interpreting the experiments considered theoretically and naively here, which includes, for example, the current low resolution of the standard real space observation and possible differences between the single molecular statistics and the conventional statistics. We modestly wish that our simple ideas would be realized in the near future thanks to efforts with technological developments.

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References

[1] P.G. de Gennes, Scaling Concepts in Polymer Physics, Cornel Univ. Press, Ithaca, 1979.
[2] T.T. Perkins, S.R. Quake, D.E. Smith, S. Chu, Science 264 (1994) 822.
[3] K. Yoshikawa, M. Takahashi, V.V. Vasilevskaya, A.R. Khokhlov, Phys. Rev. Lett. 76 (1996) 3029.
[4] Y. Arai, R. Yasuda, K. Akashi, Y. Harada, H. Miyata, K. Kinosita Jr., H. Itoh, Nature 399 (1999) 446.
[5] Y. Murayama, Y. Sakamaki, M. Sano, Phys. Rev. Lett. 90 (2003) 018102.
[6] S. Araki, T. Nakai, K. Hizume, K. Takeyasu, K. Yoshikawa, Chem. Phys. Lett. 418 (2006) 255.
[7] K. Kawai, K. Okumura, Eur. Phys. J. E 21 (2006) 223.
[8] A.Y. Grosberg, A.R. Khokhlov, Statistical Physics of Macromolecules, AIP Press, New York, 1994.
[9] D.J. Pearson, Macromolecules 10 (1977) 696.
[10] M. Warner, S.F. Edwards, J. Physics A: Math. Gen. 11 (1978) 1649.
[11] H. Benoit, B. Duplessix, R. Ober, M. Daoud, J.P. Cotton, B. Farnoux, G. Jannink, Macromolecules 8 (1975) 451.
[12] J. des Cloizeaux, M.L. Mehta, J. de Physique 40 (1979) 665.
[13] R. Kubo, M. Toda, N. Hashitsume, Statistical Physics II, second ed., Springer, Berlin, 1995.
[14] E.F. Casassa, J. Polym. Sci. A 3 (1965) 605.
[15] For the recent developement, see M.K. Shimamura, K. Kamata, A. Yao, T. Deguchi, Phys. Rev. E 72 (2005) 041804, and references therein.