

Theory of Polymer Dynamics

1 Introduction

This document summarizes information related to the construction of theoretical models representing polymers. It is mainly drawn from the book by Doi [4].

2 The freely jointed chain

From [4]. We start with a simple model: a chain consisting of N links, each of length b_0 and able to point in any direction independently of each other. The conformation of the model is represented by $(N + 1)$ position vectors $\{R_n\} = \{R_0, R_1, \dots, R_N\}$, or by the set of bond vectors $\{r_n\} = (r_1, r_2, \dots, r_N)$, where $r_n = R_n - R_{n-1}$.

Since the bonds are independent, the configuration distribution is defined as

$$\Psi(\{\vec{r}_n\}) = \prod_{n=1}^N \psi(\vec{r}_n)$$

for vector of constant length b_0

$$\psi(\vec{r}) = \frac{1}{4\pi b_0^2} \delta(|\vec{r}| - b_0)$$

we then normalize such that

$$\int \psi(\vec{r}) d\vec{r} = 1$$

If the bond vectors are presented using spherical coordinates with angles θ and ϕ , then the configuration distribution of the chain is given by

$$\psi(\theta^N, \phi^N) = \left(\frac{1}{4\pi}\right)^N \prod_{i=1}^N \sin(\theta_i)$$

This is due to the fact that the surface element of the sphere is given by the determinant of the Jacobi matrix, J , of spherical transformation for each $R_i = (x, y, z)$, namely

$$\begin{aligned} x &= b_0 \sin(\theta) \cos(\phi) \\ y &= b_0 \sin(\theta) \sin(\phi) \\ z &= b_0 \cos(\theta) \end{aligned}$$

which is $|J| = b_0^2 \sin(\theta)$, to get the probability that a bond vector is in the range $d\theta d\phi$, we divide $|J|$ by the surface area of the sphere which is $4\pi b_0^2$ to get $\sin(\theta)/4\pi$ for each bond vector. Since the bond vectors are independent, we get the multiplicative expression for ψ .

Any average quantity $B(\theta, \phi)$ that depends on the configuration of the chain in equilibrium will be written as

$$\langle B \rangle = \int \int B \psi d\theta^N d\phi^N$$

The above property of expected value of a quantity $g(x)$ given that we know only the distribution function $f(x)$ is called the **Law of the unconscious statistician**.

$$E[g(x)] = \int_{-\infty}^{\infty} g(x) f(x) dx = \int_{-\infty}^{\infty} g(x) dF(x) dx$$

where $F(x)$ is the cdf (if known).

2.1 The end-to-end vector

We define the end-to-end vector as $\vec{R} = \vec{R}_N - \vec{R}_0 = \sum_{n=1}^N \vec{r}_n$. The quantity $\langle R^2 \rangle$ characterizes the length of the chain. The standard deviation of the end-to-end vector

$$\bar{R} = \langle R^2 \rangle^{0.5} = \langle (R_N - R_0)^2 \rangle^{0.5} = \sqrt{N} b_0$$

since

$$\langle R^2 \rangle = \left\langle \left(\sum_{n=1}^N r_n \right)^2 \right\rangle = \sum_{n=1}^N \sum_{m=1}^N \langle r_n r_m \rangle + \sum_{n=1}^N \langle r_n^2 \rangle + 2 \sum_{m>n}^N \langle r_n r_m \rangle = \sum_{n=1}^N \langle r_n^2 \rangle = N b_0^2$$

therefore, $\langle R^2 \rangle^{0.5} = \sqrt{N} b_0$. Here we used $\langle r_n r_m \rangle = \langle r_n \rangle \langle r_m \rangle = 0$. Note that this is just the sum of variances of the bond vectors (each of which has variance of b_0^2)

3 Distribution of the end-to-end vector

We denote $\Phi(R, N)$ as the probability distribution function that the end-to-end vector of a chain of N beads is R .

$$\Phi(R, N) = \int \int \int \dots \int \delta(R - \sum_{n=1}^N r_n) \Psi(\{r_n\}) dr_1 dr_2 \dots dr_N$$

in short, we take all chain configurations for which the end-to-end vector is exactly R . Using the Fourier transform identity for the delta function

$$\delta(r) = \frac{1}{(2\pi)^3} \int e^{ikr} dk$$

see wikipedia. where, after some manipulation and using polar coordinates for solving the integral, we get

$$\Phi(R, N) = \frac{1}{(2\pi)^3} \int e^{ikR} \left(\frac{\sin(kb)}{kb} \right)^N dk$$

Continuing with a series of approximations to the integral above, we get

$$\Phi(R, N) = (3/2\pi Nb^2)^{1.5} \exp\left(-\frac{3R^2}{2Nb^2}\right)$$

4 The Gaussian Chain

From [4]. We consider a chain whose bond length has a Gaussian distribution

$$\psi(r) = \left[\frac{3}{2\pi b^2} \right]^{3/2} \exp\left(-\frac{3r^2}{2b^2}\right)$$

in 3D, with bond-length variance $\langle r^2 \rangle = b^2$. The Gaussian chain represent the model of beads on a strings, with the harmonic potential energy

$$U_0(\{R_n\}) = \frac{3k_B T}{2b^2} \sum_{n=1}^N (R_n - R_{n-1})^2$$

Since the sum of two Gaussian random variables is again a random variable, it turns out that the distribution of the bond between any two beads m and n is Gaussian with

$$\Phi(R_n - R_m, n - m) = \left[\frac{3}{2\pi b^2 |n - m|} \right]^{3/2} \exp\left[-\frac{3(R_n - R_m)^2}{2|n - m|b^2}\right]$$

and for any m and n

$$\langle (R_n - R_m)^2 \rangle = |n - m|b^2$$

From the formula of the distribution, we can immediately see that setting $R_N = R_m$, meaning placing two beads at the same position, gives us the encounter probability at steady state, namely $A|n-m|^{(3/2)}$, with A some constant of normalization.

We can thus immediately see that when setting $R_n = R_m$ we get

$$\Phi(0, |n - m|) = (3/2\pi b^2 |n - m|)^{1.5}$$

which is the steady state probability of encounter, which drops with the distance

5 The center of mass

The center of mass is defined as the mean bead position over time

$$cm(t) = \frac{1}{N+1} \sum_{i=0}^N R_i(t)$$

therefore, the differential equation for the center of mass is

$$\frac{dcm}{dt} = \frac{1}{N+1} \sum_{i=0}^N \frac{dR_i}{dt} = \frac{1}{N+1} \sum_{i=0}^N f_i(t)$$

6 The source of the Hookian behavior

From [2]. Let us look at the probability density function of the end-to-end vector being length r at equilibrium

$$P_{eq}(r) = \left(\frac{3}{2\pi(N-1)b^2} \right) \exp(-3r^2/2(N-1)b^2)$$

The Helmholtz free energy $A = U - TS$, with U the internal energy and S the entropy, of the system in equilibrium at temperature T is given by

$$A = -KT \ln(Z)$$

with Z the partition function, which is defined as

$$Z = c \int e^{(\mathbb{K} + \phi)/(KT)}$$

with c some constant, \mathbb{K} is the kinetic energy of the system, ϕ is the potential energy, over the phase space of the system.

When the length of the chain is changed by a small amount, the change in the Helmholtz free energy is

$$dA = -KT d \ln P_{eq} = \frac{3KT}{(N-1)b^2} (r \cdot dr)$$

We also know that the change in the Helmholtz free energy is related to the tension F^c of the chain by

$$dA = (F^c \cdot dr)$$

equating these two quantities we get

$$F^c(r) = -\frac{3KT}{(N-1)b^2} r \quad N - large$$

therefore, we see that the tension behaves as a Hookian spring. If we set $N = 2$ we get the spring constant between any two beads.

We can therefore write the probability density function for the end to end vector using the Hookian spring constant

$$P(r) = \left(\frac{k}{2\pi KT} \right)^{1.5} \exp \left(-\frac{kr^2}{2KT} \right)$$

7 Centralizing the chain

The differential equation describing the dynamics R_n in $3D$ is

$$\frac{dR_n}{dt} = -\frac{3D}{b^2} (2R_n(t) - R_{n-1}(t) - R_{n+1}(t)) + f_n(t)$$

If we shift the chain so that its new center of mass always lays at the origin, we get

$$\frac{d(R_n - cm)}{dt} = -\frac{3D}{b^2} (2R_n - R_{n-1} - R_{n+1}) + f_n - \frac{1}{N+1} \sum_{i=0}^N f_i$$

for such a new system, the new center of mass, cm^* should have zero derivative, indeed.

$$\frac{dcm^*}{dt} = \frac{1}{N+1} \left[\sum_{i=1}^N \frac{dR_i}{dt} - (N+1) \sum_{i=0}^N \frac{1}{N+1} f_i \right] = \vec{0}$$

Of course, this process is only valid post simulation. It seems not plausible to simulate a random process such that its center of mass remains in one point.

Subtracting the center of mass from the position of each bead should not affect the distribution of bond lengths, and it is indeed obvious to see it by subtracting two consecutive beads equation that the center of mass cancels out. However, the distribution of bead position is affected.

since each f_n is normally distributed with std $\sqrt{2D}$ and mean 0, the random variable $\frac{1}{N} \sum_{i \neq n}^N f_n(t)$
[UNFINISHED]

8 The distribution of the bond lengths

The bond length behaves like the central χ^2 with d degrees of freedom, where d is the dimension of the problem. In $3D$, since each $\vec{r}_i = [x_i, y_i, z_i]$ has 3 coordinates, each i.i.d normally distributed with $\langle x_i^2 \rangle = \langle y_i^2 \rangle = \langle z_i^2 \rangle = \frac{b^2}{3}$, the length squared $\|\vec{r}_i\|^2 = x_i^2 + y_i^2 + z_i^2 \sim \chi^2(3)$ with variance $\mu = b^2/3 + b^2/3 + b^2/3 = b^2$ which is the sum of variances.

To calculate the cumulative distribution function (CDF) of the bond length, we use result from the analysis of quadratic forms [6] (chapter 3). From the CDF we will obtain the *survival probability*, which its first moment is the *mean first passage time* for the two ends of the polymer.

For the polymer chain, the bond length is a random variable, $b \sim \mathcal{N}(0, \sigma_b)$

9 The dynamics of bond length

In simulations we would like to determine Δt such that no 'blow-ups' will occur. For this end, we constrain the mean of squared difference between bond length in consecutive steps to lay below some predefined small value.

The result show that

$$\gamma = \ll \|r_n(t + \Delta t) - r_n(t)\|^2 \gg = \left(\frac{3D\Delta t}{b}\right)^2 + 6D\Delta t$$

[UNFINISHED]

10 The Rouse Chain

From [4] Chap. 4. The Rouse model was suggested as the basis for polymer dynamics in dilute solution. Let $\{R\} = \{R_1, R_2, \dots, R_N\}$ be the coordinates of the beads of the chain. The motion of the beads is described using the Smoluchowski equation

$$\frac{\partial \Psi}{\partial t} = \sum_n \frac{\partial}{\partial R_n} H_{nm} \left[k_B T \frac{\partial \Psi}{\partial R_m} + \frac{\partial U}{\partial R_m} \Psi \right]$$

11 Mean First End To end Encounter Time Of the Rouse Chain

From [1].

12 Eigenvalues of the Rouse Ring

Connecting bead 1 and N to form a ring, we now search for the eigenvalues of the new *Rouse ring* matrix, R , harboring such a connection. For this end, we have to calculate the determinant of the matrix

$$D_N = |R - \lambda I| = \begin{vmatrix} x & -1 & 0 & 0 & \dots & -1 \\ -1 & x & -1 & 0 & \dots & 0 \\ 0 & -1 & x & -1 & \dots & 0 \\ \cdot & & & \cdot & & \cdot \\ \cdot & \cdot & & & \cdot & \\ \cdot & & & -1 & x & -1 & 0 \\ 0 & & & & -1 & x & -1 \\ -1 & \cdot & \cdot & \cdot & 0 & -1 & x \end{vmatrix}_{N \times N}$$

with $x = 2 - \lambda$. We can apply to the matrix of size $(N-1) \times (N-1)$ the same procedure as before and try to solve it recursively. The boundary conditions now read

$$D_1 = x; D_2 = x^2 - 1$$

According to the solution in section ?? and in [5], we have

$$D_{N-1} = \frac{\sin(N\theta)}{\sin(\theta)}$$

The relationship between D_N and D_{N-1} is found by developing the determinant of D_N according to the last column

$$D_N = x(-1)^{2N}D_{N-1} + (-1)(-1)^{2N-1}D^* + (-1)(-1)^{N+1}D^{**} = xD_{N-1} + D^* + (-1)^{N+2}D^{**}$$

with the determinant D^* and D^{**} defined as the minors

$$D^* = \begin{vmatrix} x & -1 & 0 & 0 & \dots & 0 \\ -1 & x & -1 & 0 & \dots & 0 \\ 0 & -1 & x & -1 & \dots & \\ \vdots & & & \vdots & & \\ \vdots & & & \vdots & & \\ \vdots & & & -1 & x & -1 \\ -1 & & & 0 & -1 & \end{vmatrix}_{(N-1) \times (N-1)} \quad D^{**} = \begin{vmatrix} -1 & x & -1 & 0 & \dots & 0 \\ 0 & -1 & x & -1 & \dots & 0 \\ \vdots & & & \vdots & & \vdots \\ \vdots & & & \vdots & & \vdots \\ \vdots & & & -1 & x & -1 \\ 0 & & & & -1 & x \\ -1 & & & & 0 & -1 \end{vmatrix}_{(N-1) \times (N-1)}$$

We calculate the determinants D^* and D^{**} by minors according to the last row to get

$$D^* = (-1)(-1)^{2N-2}D_{N-2} + (-1)(-1)^{N-1+1}(-1)^{N-2} = (-1)^{2N-1}[D_{N-2} + 1]$$

$$D^{**} = (-1)(-1)^{2N-2}(-1)^{N-2} + (-1)(-1)^{1+N-1}D_{N-2} = (-1)^{3N-3} + (-1)^{N+1}D_{N-2}$$

Therefore,

$$\begin{aligned} D_N &= xD_{N-1} + (-1)^{2N-1}[D_{N-2} + 1] + (-1)^{N+2}[(-1)^{3N-3} + (-1)^{N+1}D_{N-2}] \\ &= xD_{N-1} + (-1)^{2N-1}[D_{N-2} + 1] + (-1)^{4N-1} + (-1)^{2N+3}D_{N-2} \\ &= xD_{N-1} + D_{N-2}[(-1)^{2N-1} + (-1)^{2N+3}] + (-1)^{2N-1} + (-1)^{4N-1} \\ &= xD_{N-1} - 2D_{N-2} - 2 \end{aligned}$$

Substituting the solution for D_{N-1} and D_{N-2} , we get

$$D_N = \frac{x \sin(N\theta) - 2 \sin((N-1)\theta) - 2 \sin(\theta)}{\sin(\theta)} = 0$$

Further simplification of D_N leads to

$$\begin{aligned} D_N &= 2 \frac{\cos(\theta) \sin(N\theta) - [\sin((N-1)\theta) + \sin(\theta)]}{\sin(\theta)} \\ &= \frac{2}{\sin(\theta)} [\cos(\theta) \sin(N\theta) - 2 \sin(\frac{N\theta}{2}) \cos(\frac{(N-2)\theta}{2})] \\ &= \frac{4}{\sin(\theta)} [\cos(\theta) \sin(\frac{N\theta}{2}) \cos(\frac{N\theta}{2}) - \sin(\frac{N\theta}{2}) [\cos(\frac{N\theta}{2}) \cos(\theta) + \sin(\frac{N\theta}{2}) \sin(\theta)]] \\ &= -4 \sin^2(\frac{N\theta}{2}) \end{aligned}$$

Equating $D_N = 0$ we get the solutions

$$\theta_p = \frac{2\pi p}{N}$$

for $p = 0, 1, \dots, (N - 1)$

Therefore, the eigenvalues of the Rouse ring are

$$\lambda_p = 2(1 - \cos(\theta_p)) = 2(1 - \cos(2\pi p/N))$$

Since $\cos(\frac{2\pi\phi}{N}) = \cos(\frac{2\pi(N-\phi)}{N})$, we have eigenvalues multiplicities. In our case, setting $\phi = p$ we have $\cos(\frac{2\pi p}{N}) = \cos(\frac{2\pi(N-p)}{N})$ for $p = 1, 2, 3, \dots, (N - 2)$. For the end-values, $p = 0$ and $p = N - 1$ the eigenvalues are unique.

13 The Pair Correlation function

According to [3]. To investigate the spatial distribution of segments in the Rouse polymer we introduce the segment pair correlation function, $g(r)$, defined as follows. If we focus on the n^{th} segment, we define $g_n(r)$ to be the average segment density at position r from segment n . If we write R_n ($n = 1, 2, 3, \dots, N$) for the position vectors of the segments, then we can express $g_n(r)$ as

$$g_n(r) = \sum_{m=1}^N \langle \delta(r - (R_m - R_n)) \rangle$$

the average is a spatial average, so in 3D we have to divide by the volume $r + \delta r$ to get the concentration. The time correlation function, $g(r)$ is then defined as the average of the segment pair correlation function, $g_n(r)$.

$$g(r) = \frac{1}{N} \sum_{n=1}^N g_n(r)$$

In addition, we define $g(q)$, the Fourier transform of $g(r)$

$$g(q) = \int e^{iqr} g(r) dr = \frac{1}{N} \sum_{n=1}^N \sum_{m=1}^N \langle \exp[iq(R_m - R_n)] \rangle$$

The pair correlation function is thus also called the radial distribution function. In the simplest terms it is a measure of probability of finding a particle at a distance r away from a given reference particle.

14 Radius of Gyration

From the definition of the Fourier transform of the pair correlation function for small q , we can define the length called radius of gyration.

15 The Rouse ring

Construction of the Rouse ring is done by setting the first bead at random position x_0 , adding a phantom bead $N + 1$ and setting its position to x_0 . Then, we connect bead 1 and the phantom bead by a random walk, $w(t)$, of N steps using the Brownian bridge.

The **Brownian bridge** which starts at x at time 0 and ends in point y at time t_0 is given by the general form

$$x(t) = \sqrt{2D}w(t) - (t/t_0)[\sqrt{2D}w(t_0) - y + x] + x \quad 0 < t \leq t_0$$

For the Rouse ring we set $x = y$ and calculate the probability function

$$Pr(x(t) < X) = Pr(w(t) < (X - x)/\sqrt{2D} + (t/t_0)w(t_0))$$

so, the probability is given by the probability that the random walk, $w(t)$, is lower than the line $(X - x)/\sqrt{2D} + (t/t_0)w(t_0)$. This can be calculated using the cylindrical sets $C(t, [a, b])$, which has a Wiener measure

$$Pr(C(t, [a, b])) = \frac{1}{\sqrt{2\pi t}} \int_a^b e^{-x^2/2t} dx$$

setting $a = 0, b = (X - x)/\sqrt{2D} + (t/t_0)w(t_0) = \alpha + \beta t$, results in

$$F = Pr(C(t, [0, \alpha + \beta t])) = \frac{1}{\sqrt{2\pi t}} \int_0^{\alpha + \beta t} e^{-x^2/2t} dx$$

The probability distribution is then

$$\frac{\partial F}{\partial x} = f = \frac{1}{\sqrt{2\pi t}} [e^{-(\alpha + \beta t)^2/2t}]$$

in three-dimensional space we have

$$f(t) = \left(\frac{1}{\sqrt{2\pi t}} \right)^3 [e^{-3(\alpha + \beta t)^2/2t}]$$

Now that we have the probability density in equilibrium of the Brownian bridge we can calculate average quantities related to the positions of the beads.

The mean square distance between bead 1 and $1 < j \leq N$ is given by the relation

$$\langle r \rangle^2 = \int r^2 f(r) dx$$

[UNFINISHED]

16 Encounter probability in the Rouse ring

Following the method in [1]. First we transform the Rouse ring system of equation into normal coordinates using the Rouse ring eigenvalues found in Section 12. We transform to normal coordinates by $u_p = \sum_{n=1}^N \alpha_p^n R_n$, where α_p are the eigenvectors. We then represent our boundary conditions that bead j and bead k are in the same position, using the normal coordinates representation. The probability density function in the normal coordinates should then satisfy the forward Fokker Planck equation. The solution to the probability density function should then be expanded as an infinite sum of the form

$$p(x, t) = \sum_{i=1}^{\infty} a_i w_{\lambda_i^\epsilon}(x) e^{-\lambda_i^\epsilon t D} e^{-\phi(x)}$$

where $w_{\lambda_i^\epsilon}(x)$ and λ_i^ϵ are the eigenvectors and eigenvalues of the forward Fokker Planck operator outside of the domain, and a_i are coefficients. Integrate the probability density according to x , over the domain without the absorbing boundary (the encounter sphere), to get a function of time. An integration of the resulting function from time $t = 0$ to ∞ is the first moment of the survival function, which is the mean first encounter time (i.e the first time the polymer's ends are at the absorbing sphere).

17 Center of mass diffusion of the rouse ring

By summing the rows of the Rouse ring matrix, we see that the differential equation describing the diffusion of the center of mass does not change in relation to the chain's organization. Inner loops in the ring do not change the center of mass diffusion. Therefore

$$\frac{dcm}{dt} = \frac{1}{N} \sum_{n=1}^N g_n$$

we follow calculation by [Unfinished]

18 Generalized Rouse Chain

Assuming a chain with N beads, in which each bead is connected to its nearest neighbors and at most $N - 2$ beads with spring constants k_{ij} between bead i and j . the configuration distribution of the chain is given by

$$\psi = \prod_{i=1}^{N-1} \prod_{j=1}^{N_i} \left(\frac{k_{ij}}{2\pi KT} \right)^{1.5} \exp \left(-\frac{k_{ij} r^2}{2KT} \right)$$

with N_i the number of connections of bead i , with multiplication over unique pairs.

19 Simulations

19.1 Choosing Δt

The dynamics of beads is governed by the equation

$$\frac{dP}{dt} = -\frac{3D}{b^2}RP + \sqrt{2D}f_n$$

if we represent the Rouse matrix in its diagonal form

$$R = Q^{-1}\Lambda Q$$

where Q is an orthonormal matrix with the eigenvectors of R in its columns, and Λ is a diagonal matrix with the Rouse eigenvalues in the diagonal, we get

$$\frac{dP}{dt} = -\frac{3D}{b^2}Q^{-1}\Lambda Q + \sqrt{2D}f$$

multiplying from the left by Q and setting $S = QP$ we get

$$\frac{dS}{dt} = -\frac{3D}{b^2}\Lambda S + \sqrt{2D}Qf$$

19.1.1 The case with no noise

if we omit the noise term and solve for S the numerical scheme is

$$S(t + \Delta t) - S(t) = \left(-\frac{3D\Delta t}{b^2}\Lambda\right)S(t)$$

taking the norm of both sides and dividing

$$\frac{\|S(t + \Delta t) - S(t)\|}{\|S(t)\|} = \left\| \left(-\frac{3D\Delta t}{b^2}\Lambda\right) \right\| = \frac{3D\Delta t}{b^2}\lambda_{max} = \frac{12D\Delta t}{b^2}$$

where for the Rouse matrix $\lambda_{max} = 4$. Demanding that the quotient be smaller than 1 we have

$$\frac{12D}{b^2} \leq 1 \implies \Delta t \leq \frac{b^2}{12D}$$

19.1.2 The case with noise

using the same procedure as above,

$$\begin{aligned} \frac{\|S(t + \Delta t) - S(t)\|}{\|S(t)\|} &= \left\| \left(-\frac{3D}{b^2}\Lambda\right)S + \sqrt{2D}Qf \right\| / \|S\| \leq \left(\frac{3D}{b^2}\|\Lambda\| + \sqrt{2D}\|Qf\| / \|S\| \right) \\ &= \leq \frac{3D}{b^2}\lambda_{max} + \sqrt{2D}\frac{\|Q\|\|f\|}{\|S\|} = \frac{12D}{b^2} \leq \frac{3D}{b^2}\lambda_{max}\Delta t + \sqrt{2D}\Delta t \end{aligned}$$

Demanded that the quotient be smaller than 1 we have

$$\frac{12D}{b^2} \leq 1 \implies \Delta t \leq \frac{b^2}{12D + \sqrt{2D}b^2}$$

19.2 Relaxation time

The relaxation time is determined according to the formula

$$\tau_p = \frac{b^2 \Delta t}{12d^2 \sin^2(\frac{p\pi}{2N})} \quad (1)$$

References

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