Dynamical eigenmodes of a polymerized membrane

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Abstract. We study the bead-spring model for a polymerized phantom membrane in the overdamped limit, which is the two-dimensional generalization of the well-known Rouse model for polymers. We derive the exact eigenmodes of the membrane dynamics (the ‘Rouse modes’). This allows us to obtain exact analytical expressions for virtually any equilibrium or dynamical quantity for the membrane. As examples we determine the radius of gyration, the mean-square displacement of a tagged bead, and the autocorrelation function of the difference vector between two tagged beads. Interestingly, even in the presence of tensile forces of any magnitude the Rouse modes remain the exact eigenmodes for the membrane. With stronger forces the membrane becomes essentially flat, and does not get the opportunity to intersect itself; in such a situation our analysis provides a useful and exactly solvable approach to the dynamics for a realistic model flat membrane under tension.

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1. Introduction

Bead-spring models for linear polymers, wherein the polymer is modeled as a sequence of beads connected by springs, play a central role in the theory of polymer dynamics. For a linear bead-spring polymer chain, with position $\mathbf{R}_n$ of the $n$th bead, $n = 1, \ldots, N$, the potential energy is given by $U = \frac{k}{2} \sum_{n=1}^{N-1} (\mathbf{R}_n - \mathbf{R}_{n+1})^2$, in which $k$ is the spring constant. Within the scope of bead-spring models, the Rouse model [1] holds a special status, as it provides a framework for linear polymers by which virtually any static and dynamical quantity of interest can be calculated analytically [2, 3]. In the Rouse model the dynamics of the beads is formulated in the overdamped limit. For solving the Rouse model one considers the so-called Rouse modes (rather than the equations of motion for the individual beads). Using the mode amplitude correlation functions, the quantities of interest for a phantom chain can be analytically tracked by reconstructing them from the modes [2, 3]. The Rouse model has been shown useful for self-avoiding linear polymers [4], and in showing that the dynamics of a tagged bead in a linear bead-spring model is described by the generalized Langevin equation (GLE) [5, 6].

It is therefore a natural question to ask if the analytical solution of the Rouse model can be generalized to higher dimensions, e.g., making it useful for the dynamics of polymerized membranes. There is a considerable amount of interest in the properties of polymerized membranes [7]. The interest stems not only from the point of view of a fundamental understanding, but also because of their importance in biology and chemistry. Instances of polymerized membranes with a fixed connectivity can be found in the 2D-cytoskeletons of cells [8]–[10] and graphite oxide sheets [11]–[13].

Static properties of polymerized membranes as a two-dimensional extension of (one-dimensional) linear polymers came into fashion in the late 1980s. At that time the driving question centered on the equilibrium properties of polymerized membranes, in particular, the scaling of the radius of gyration of a polymerized membrane with its lateral size,
while the corresponding scaling behavior for phantom and self-avoiding linear polymers were already well-known. Sophisticated renormalized group approaches were developed to this end [14]–[17], accompanied by computer simulations [18]–[26]. A notable outcome of these studies is that the radius of gyration $R_g$ of a membrane scales with its lateral size $N$ as $R_g \sim \log N$ for phantom, and as $R_g \sim N$ for self-avoiding membranes.

In contrast to the equilibrium properties, the dynamical properties of membranes have been studied with less intensity. Apart from some scaling analysis [27]–[29], the bulk of the research on the dynamics of polymerized membranes is heavily dominated by computer simulations [30]–[33], leaving exact analytical results on the dynamics of membranes a relatively open area. In this paper we attempt to fill this void—we consider two-dimensional square-polymerized phantom membranes embedded in a three-dimensional space, both in the absence and the presence of tensile forces—and perform a Rouse mode analysis. We show that the Rouse modes are the exact eigenmodes of the membrane dynamics. Akin to the Rouse model for bead-spring linear polymers, our exercise allows us to solve exactly for the static and dynamic properties of phantom membranes; in this process also deriving the exact expression for the scaling of $R_g$ as a function of the membrane’s lateral size $N$ (which confirms the $R_g \sim \log N$ scaling), as well as showing increased fluctuations at the edge of the membrane [34, 35]. In particular, we draw the reader’s attention to the exact solution under tensile forces, for which the membrane essentially encounters a flat geometry. Unlike a phantom membrane in equilibrium, in such a situation the membrane becomes essentially flat, does not get the opportunity to intersect itself, and therefore provides a highly useful and exactly solvable approach to the dynamics for a realistic model flat membrane. We also note that the methods are generalizable to arbitrary internal and spatial dimensions.

The structure of this paper is as follows. In section 2 we describe the dynamical equations without forces, and their diagonalization by the Rouse modes. In section 3 we derive the scaling of the radius of gyration, the mean-square displacement of a tagged bead and the autocorrelation function of a vector connecting two beads. In section 4 we incorporate tensile forces in the dynamical equations and derive the new Rouse modes that diagonalize them. We finally end the paper with a discussion in section 5.

2. Diagonalization of equations of motion for the polymerized membrane and the Rouse modes

We consider a rectangular polymerized membrane, for which $N = L_1 \times L_2$ beads are connected in a perpendicular topology, with $\mathbf{R}_n(t)$ denoting the spatial position at time $t$ of the bead internally labeled as $n$. Naturally for a membrane it is convenient to take two numbers $n = (n_1, n_2)$ to label the beads with $n_i = 1, \ldots, L_i$. The potential energy for a square-polymerized membrane is a simple extension of the Hamiltonian for a linear polymer in two internal dimensions

$$U = \frac{k}{2} \sum_{n_1, n_2=1}^{L_1-1, L_2} (\mathbf{R}_{n_1, n_2} - \mathbf{R}_{n_1+1, n_2})^2 + \frac{k}{2} \sum_{n_1, n_2=1}^{L_1, L_2-1} (\mathbf{R}_{n_1, n_2} - \mathbf{R}_{n_1, n_2+1})^2$$

for some spring constant $k$.
In the absence of externally applied forces, in the overdamped limit the dynamics of each bead of the membrane is described by
\[
\frac{d\mathbf{R}_n}{dt} = -\frac{1}{\zeta} \frac{\partial U}{\partial \mathbf{R}_n} + g_n,
\]
where $\zeta$ is the friction coefficient of the solvent and $g_n$ is the thermal force on the $n$th bead. The thermal forces are uncorrelated between different beads, as well as in time, i.e.,
\[
\langle g_m(t) \cdot g_n(t') \rangle = \frac{6k_B T}{\zeta} \delta_{mn} \delta(t - t'),
\]
with Boltzmann constant $k_B$ and temperature $T$.

For a linear polymer with positions $\mathbf{R}_n(t)$ of beads $n = 1, \ldots, N$ at time $t$, the Rouse modes are given by [2, 3]
\[
X_p(t) = \frac{1}{N} \sum_{n=1}^{N} \cos \left[ \frac{\pi(n - 1/2)p}{N} \right] \mathbf{R}_n(t),
\]
with $p = 0, \ldots, N - 1$ and the inverse given by
\[
\mathbf{R}_n(t) = \sum_{p=1}^{N-1} \cos \left[ \frac{\pi(n - 1/2)p}{N} \right] X_p(t).
\]
The Rouse modes for the membrane are quite similar to those of the linear chain: since topologically the internal directions (connectivity) are orthogonal, the Rouse modes will be products of Rouse modes for a linear polymer. Below we introduce the following definitions for an elegant notation and proof of the independence of the Rouse modes.

First we define
\[
\beta_{p_i} = \begin{cases} 
2 & \text{if } p_i = 1, \ldots, L_i - 1 \\
1 & \text{if } p_i = 0 
\end{cases}, \quad \beta_p = \beta_{p_1} \beta_{p_2}
\]
and
\[
f_{p_i}(n_i) = \cos \left[ \frac{\pi p_i(n_i - 1/2)}{L_i} \right], \quad f_p(n) = f_{p_1}(n_1)f_{p_2}(n_2).
\]
The orthogonality relation at the basis of the proof for the Rouse modes for a linear polymer chain can then be generalized for the membrane
\[
\frac{1}{N} \sum_{n} \beta_p f_p(n) f_q(n) = \delta_{pq},
\]
where the summation is taken over all allowed values of $n$. The Rouse mode amplitudes and their corresponding inverse are then given by
\[
X_p(t) = \frac{1}{N} \sum_{n} f_p(n) \mathbf{R}_n(t),
\]
\[
\mathbf{R}_n(t) = \sum_{p} \beta_p f_p(n) X_p(t).
\]
The position of the center-of-mass $\mathbf{R}_{cm}(t)$ at time $t$ simply equals $X_0(t)$.

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Next, we note that when the equations of motion from equation (2) for the beads are expressed one gets a term for each internal dimension equal to that of a term for a bead in a linear polymer. Taking the time derivative in equation (9) and plugging in the equations of motion for the beads and then the inverse we get

$$\frac{dX_p(t)}{dt} = -\alpha_p X_p(t) + G_p(t)$$

(11)

with

$$\alpha_p \equiv 4k\zeta \left( \sin^2 \frac{\pi p_1}{2L_1} + \sin^2 \frac{\pi p_2}{2L_2} \right)$$

and

$$G_p(t) = \frac{1}{N} \sum_n f_p(n) g_n(t),$$

(12)

i.e., the Rouse modes diagonalize the equations of motion. This set of linearized differential equations can be solved just as for a linear polymer by using the correlation function for the thermal forces given by equation (3). By using the orthogonality relation in equation (8)

$$\langle G_p(t) \cdot G_q(t') \rangle = \frac{6k_B T}{\zeta N \beta_p} \delta_{pq} \delta(t-t'),$$

(13)

so that the following two-point correlation functions for the eigenmodes can be derived:

$$X_{00}(t) \equiv \langle [X_0(t) - X_0(0)]^2 \rangle \equiv \langle [R_{cm}(t) - R_{cm}(0)]^2 \rangle = \frac{6k_B T}{\zeta N t}$$

(14)

$$X_{pq}(t) \equiv \langle X_p(t) \cdot X_q(0) \rangle = \frac{3k_B T}{\zeta N \alpha_p} \alpha_p \exp \left[ -\alpha_p t \right] \delta_{pq} \quad \text{with} \quad p \neq 0.$$  

(15)

Just like the case of a linear polymer, any correlation function of interest for the membrane can be derived using equations (11)–(13). We address a few of them in section 3.

3. A few properties of interest for a membrane

Throughout this section, for the sake of simplicity, we restrict ourselves to square membranes with $L_1 = L_2 = L$, and thus $N = L \times L$.

3.1. Scaling of the radius of gyration $R_g$

The radius of gyration squared for the membrane is defined by

$$R_g^2 \equiv \frac{1}{N} \sum_n \langle [R_n(t) - R_{cm}(t)]^2 \rangle$$

$$= \frac{1}{N} \sum_n \sum_{p \neq 0} \sum_{q \neq 0} \langle X_p(t) \cdot X_q(t) \rangle \beta_p \beta_q f_p(n) f_q(n),$$

(16)

in which equation (10) is used for the second line. Having plugged in equations (8) and (15), the squared radius of gyration reduces to

$$R_g^2 = \frac{3k_B T}{\zeta N} \sum_{p \neq 0} \frac{1}{\alpha_p}.$$  

(17)

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The summation (17), using the definition of $\alpha_p$ (12), can be split into two parts:

$$R^2_g = \frac{k_B T}{k} \left( \frac{3}{2N} \sum_{p=1}^{L-1} \sin^{-2} \left[ \frac{\pi p}{2L} \right] + \frac{3k}{N} \sum_{p_1=1}^{L-1} \sum_{p_2=1}^{L-1} \frac{1}{\alpha_p} \right) = \frac{k_B T}{k} (\mathcal{I} + \mathcal{J}). \quad (18)$$

The first term in equation (18) can be evaluated exactly: $\mathcal{I} = (N - 1)/N$. For the second term we take the long polymer limit $N \gg 1$ so that $\alpha_p$ can be expanded and the sum approximated by an integral with $x_i = p_i/L$, where the integral is over a unit square in the positive quadrant excluding the small area near the origin. The second approximation we make is replacing the area of integration by that of the positive quadrant of a unit circle excluding the small area near the origin, which can be solved analytically

$$\mathcal{J} = \frac{3}{\pi^2} \int \frac{dx}{x^2} = \frac{3}{2\pi} \int_{1/L}^{1} \frac{dr}{r} = \frac{3}{4\pi} \log N. \quad (19)$$

The radius of gyration squared for a square membrane in the long polymer limit thus becomes

$$R^2_g = \frac{k_B T}{k} \left( \frac{N - 1}{N} + \frac{3}{4\pi} \log N \right). \quad (20)$$

A comparison between the two terms in equation (20) shows that the second one is bigger than the first one for $N > 61$. In figure 1 we present a comparison between the analytical result (20) and the exact evaluation of equation (17) for $L$ up to $L = 10^4$. Our result (20) confirms the $\log N$ scaling of $R^2_g$ obtained earlier by field-theory methods [14]–[17].

### 3.2. Mean-square displacement of a tagged bead

Let us define $\Delta R_n(t) \equiv R_n(t) - R_n(0)$ for the $n$th bead. We use equation (10) to write

$$\langle \Delta R_n(t)^2 \rangle = \langle (X_0(t) - X_0(0))^2 \rangle + 2 \sum_{p \neq 0} \sum_{q \neq 0} \beta_p \beta_q f_p(n) f_q(n) \{X_{pq}(0) - X_{pq}(t)\}. \quad (21)$$
Using equations (14) and (15) this simplifies to
\[
\langle \Delta R_n(t)^2 \rangle = \frac{6k_B T}{\zeta N} t + \frac{6k_B T}{\zeta N} \sum_{p \neq 0} \frac{\beta_p f_p(n)^2}{\alpha_p} \{1 - e^{-\alpha p t}\}. \tag{22}
\]
At short times \(kt/\zeta \ll 1/8\) the exponential in the second term can be expanded, and the sum is exactly evaluated. In comparison to the second term the first one can be neglected, thus obtaining
\[
\langle \Delta R_n(t)^2 \rangle = \frac{6k_B T}{\zeta} t \sum_{p \neq 0} \frac{\beta_p f_p(n)^2}{\alpha_p} = \frac{6k_B T}{\zeta} t, \tag{23}
\]
which agrees with the mean-square displacement of a free bead, and confirms the wisdom that at short times the beads do not feel the connectivity.

At very long times the exponentials in equation (22) reduce to zero, the second term becomes a fixed number independent of \(t\) (but dependent on the location of the bead: the sum is largest when the bead is located at the corner of the membrane, and equals \(6k_B T (\log N/\pi - \pi/4 + 2/3)/k\)); i.e., for \(kt/\zeta \gg N \log N/\pi\) the first term of equation (21) dominates, and the motion of the tagged bead becomes simply diffusive, with the same diffusion coefficient as that of the center-of-mass.

At intermediate times the mean-square displacement of the tagged bead will depend on its location on the membrane. We work out three different cases when the tagged bead is (a) the central bead of the membrane, (b) located in the middle of an edge, and (c) a corner bead. The enhanced mobility of edge and corner beads has already been reported for self-avoiding membranes [34, 35].

(a) For the mean-square displacement of a bead at the center of the membrane \(\langle \Delta R_m(t)^2 \rangle\) the sum in equation (22) will only be over even values of \(p_i\). In the long polymer limit the sum can be converted to an integral with \(x_i = p_i/L\), such that
\[
\langle \Delta R_m(t)^2 \rangle = \frac{6k_B T}{\zeta} \int_0^1 dx_1 \int_0^1 dx_2 \frac{1}{\alpha_p} \{1 - e^{-\alpha_p t}\} = \frac{6k_B T}{\zeta} \int_0^{kt/\zeta} dt' \int_0^1 dx_1 \int_0^1 dx_2 e^{-\alpha_p t'}. \tag{24}
\]
We proceed with equation (24) by splitting the exponentials, resulting in two identical integrals which can be evaluated in terms of the modified Bessel function of the first kind \(I_\alpha(z)\) and making a change of variables.
\[
\langle \Delta R_m(t)^2 \rangle = \frac{6k_B T}{\zeta} \int_0^{kt/\zeta} dt' \left( \int_0^1 dx \exp \left\{ -\frac{4kt'}{\zeta} \sin^2 \left[ \frac{\pi}{2} x \right] \right\} \right)^2 = \frac{6k_B T}{k} \int_0^{kt/\zeta} dt' e^{-4t' I_0^2(2t')} \tag{25}
\]
To characterize the moderate time behavior of equation (25) we look at the asymptotic expansion \(I_\alpha(z) = \exp(z)/\sqrt{2\pi z}\) and evaluate the integral so that
\[
\langle \Delta R_m(t)^2 \rangle = \frac{3k_B T}{2\pi k} \left( \log \left[ \frac{kt}{\zeta} \right] + C_m \right), \tag{26}
\]
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where
\[ C_m = \lim_{x \to \infty} -\log[x] + 4\pi \int_0^x dt \, e^{-4t} I_0^2(2t) \approx 4.04. \quad (27) \]

(b)–(c) The same can be done for the mean-square displacement for a bead located in the middle of an edge \( \langle \Delta R_e(t)^2 \rangle \) and corner \( \langle \Delta R_c(t)^2 \rangle \). The corresponding calculations are similar to those in (a), and they yield
\[ \langle \Delta R_e(t)^2 \rangle = \frac{3k_B T}{\pi k} \left( \log \left[ \frac{kt}{\zeta} \right] + C_e \right), \quad (28) \]
where
\[ C_e = \lim_{x \to \infty} -\log[x] + 2\pi \int_0^x dt \, e^{-4t} (I_0^2(2t) + I_1^2(2t)) I_0(2t) \approx 2.47, \quad (29) \]
and
\[ \langle \Delta R_c(t)^2 \rangle = \frac{6k_B T}{\pi k} \left( \log \left[ \frac{kt}{\zeta} \right] + C_c \right), \quad (30) \]
where
\[ C_c = \lim_{x \to \infty} -\log[x] + \pi \int_0^x dt \, e^{-4t} (I_0^2(2t) + I_1^2(2t))^2 \approx 1.47. \quad (31) \]

Note that all constants \( C \) can be evaluated to an arbitrary precision and that the approximations (25), (27) and (29) are valid for \( 4kT/\zeta \ll N/\pi^2 \). In figure 2 we compare the exact evaluations from equation (22) and the corresponding approximations (23), (26), (28) and (30).

3.3. Autocorrelation function of a vector connecting two beads

In the most general case the vector \( \mathbf{r}_{mn}(t) \equiv \mathbf{R}_m(t) - \mathbf{R}_n(t) \) connects two beads with internal coordinates \( m \) and \( n \) at time \( t \). The autocorrelation function of this vector \( D_{mn}(t) \equiv \langle \mathbf{r}_{mn}(t) \cdot \mathbf{r}_{mn}(0) \rangle \) can be expressed in terms of the eigenmodes
\[ D_{mn}(t) = \frac{3k_B T}{\zeta N} \sum_{p \neq 0} \beta_p \alpha_p |f_p(m) - f_p(n)|^2 \exp[-\alpha_p t]. \quad (32) \]
Like in the case for the mean-square displacement of a tagged bead, the behavior of \( D_{mn}(t) \) depends on time and the internal positions of the beads, hence we only briefly outline the calculation procedure and state the results. We focus on two extreme cases: (a) \( D_c(t) \), where the vector connecting the two beads located at the opposite corners of the membrane, and (b) \( D_a(t) \), where the vector connects two beads at positions \( n \) and \( n + a \) somewhere in the middle of the membrane for \( a/L \equiv |a|/L \ll 1 \). We will generalize these results qualitatively for the cases when the locations of the beads and the distances between them are arbitrary.

Short times: for times \( 4kT/\zeta \ll 1/8 \) the exponent is roughly 1, therefore the functions are constant. In the case where the two beads are close neighbors the sum can be evaluated as \( D_a(t) = k_B T(3/2 + \log a)/k \), which is exact for \( a = 1 \), and a rough estimate \( D_c(t) = 6k_B T \log N/(\pi k) \) follows from a calculation similar to that performed for the radius of gyration.
Figure 2. The mean-squared displacement $\langle \Delta R_n(t)^2 \rangle$ for beads in a square membrane as a function of scaled time $kt/\zeta$. The sum in equation (22) was exactly evaluated for $k_B T/k = 1$ for beads at the center of the membrane, at the middle of an edge, and at a corner for $N = 301^2$ and the result is represented by the circular (red), triangular (green), and square (blue) data points respectively. The cyan diamonds represent the exact evaluation for a bead with internal position $n = (151, 5)$ to show the transition that the mean-squared displacement for a bead makes from one region to another. For very short times $kt/\zeta \ll 1/8$ the mean-squared displacement for all beads $\langle \Delta R_n(t)^2 \rangle \sim t$, thus behaving like that of a free bead as shown in equation (23), which is represented by the dashed line in (a). For very long times the mean-squared displacement for all beads $\langle \Delta R_n(t)^2 \rangle \sim t/N$ behaves like that of the mean-squared displacement of the center-of-mass, which is represented by the dotted line in (a). In the intermediate time regime the mean-square displacement for beads $\langle \Delta R_n(t)^2 \rangle \sim \log[kt/\zeta]$ as in agreement with equations (26), (28) and (30)—the corresponding approximations are valid for $kt/\zeta \ll N/\pi^2$—and are represented by the red, green, and blue solid lines in (b) respectively.

Intermediate times: for $a/L \ll 1$ the sum for $D_a(t)$ can be expanded in $a$ and evaluated as

$$D_a(t) = \frac{3\zeta k_B T a^2}{8\pi k^2} \frac{1}{t}.$$  \hspace{1cm} (33)

Further, it can be shown that the summation for $D_c(t)$ equals that of $-\langle \Delta R_c(t)^2 \rangle$ up to a constant, such that

$$D_c(t) = D_c(0) - \frac{6k_B T}{\pi k} \left( \log \left[ \frac{kt}{\zeta} \right] + C_c \right).$$  \hspace{1cm} (34)

Long times: for $a/L \ll 1$ and $kt/\zeta \gg N/\pi^2$ only the lowest mode contributes to the summation, therefore, expanding the correlation function in equation (32) yields

$$D_a(t) = \frac{6k_B T a^2}{kN} \exp \left[ -\frac{\pi^2 k}{\zeta N} t \right],$$  \hspace{1cm} (35)

while

$$D_c(t) = \frac{48k_B T}{\pi^2 k} \exp \left[ -\frac{\pi^2 k}{\zeta N} t \right].$$  \hspace{1cm} (36)

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Figure 3. The scaled autocorrelation function $D_{mn}(t)/D_{mn}(0)$ of the vector connecting beads with internal position $m$ and $n$ in a square membrane as a function of scaled time $kt/\zeta$. The sum in equation (32) was exactly evaluated with $k_B T/k = 1$ for two beads at positions $[(251, 251), (250, 251)], [(251, 254), (258, 251)], [(1, 1), (501, 501)],$ and $[(251, 281), (281, 251)]$ in the membrane with $N = 501^2$, with the results shown by the circular (red), triangular (green), square (blue), and diamond (cyan) data points respectively. Note that the first two pairs of beads are very close to each other in the middle of the membrane whereas the third pair are two beads in opposing corners of the membrane; i.e., the first two cases correspond to $D_a(t)/D_a(0)$, while the third one corresponds to $D_c(t)/D_c(0)$. The function $D(t) \sim \log[kt/\zeta]$, as in agreement with equation (34), corresponding to the blue solid line in (b), for a duration that is larger when the beads are connected by a larger vector $a$. For very small vectors $a$ the behavior will shift to $D(t) \sim 1/t$, as derived in equation (33), corresponding to the red and green solid lines in (a), before $kt/\zeta \sim N/\pi^2$. The function $D(t)$ for the last pair shows a transition from logarithmic behavior to $D(t) \sim 1/t$ before the diffusive time regime.

It is interesting to note the differences in behavior for $D_a(t)$ and $D_c(t)$ at intermediate times. The reason behind this difference is as follows. For $D_a(t)$ the beads are close and as a result they quickly become ‘aware’ of each other’s presence. For $D_c(t)$, on the other hand, the beads do not become ‘aware’ of each other’s presence until almost $kt/\zeta \sim N/\pi^2$. Based on this observation we expect that when the beads are neither very close nor very far, there will first be a logarithmic decay (34) for $D_{mn}(t)$, followed by a $1/t$ power-law decay (33) before the terminal exponential decay (35) and (36) sets in. The above results are verified by comparing the approximations to the exact evaluation of equation (32) for different pairs of beads in figure 3.

4. Dynamical eigenmodes in the case of external tensile forces

In this section we concentrate on presenting the exact eigenmodes when the membrane is stretched in two perpendicular directions by forces $F_1$ and $F_2$ applied at the edges of the membrane.

Adding tensile forces to the system ensures that each bead has its own mean position, around which it fluctuates due to thermal forces. For large enough tensile forces the beads are far enough away from each other, and self-intersection of the membrane is avoided. Such a situation therefore mimics the behavior of a realistic flexible membrane under
tension. The system is still exactly solvable by introducing the following term to the Hamiltonian

\[ U_F = \sum_{n_1,n_2=1}^{L_1-1,L_2} \mathbf{F}_1 \cdot (\mathbf{R}_{n_1,n_2} - \mathbf{R}_{n_1+1,n_2}) + \sum_{n_1,n_2=1}^{L_1,L_2-1} \mathbf{F}_2 \cdot (\mathbf{R}_{n_1,n_2} - \mathbf{R}_{n_1,n_2+1}), \]

making the exercise of this section useful for practical purposes.

The mode amplitudes and their inverses are once again defined by equations (9) and (10), but the following term \( \mathbf{H}_p \) will be added to the right-hand side of the differential equation in equation (11) in order to solve for the dynamics of the mode amplitudes:

\[ \mathbf{H}_p = \frac{1}{\zeta N} \sum_n f_p(n) [\mathbf{F}_1(\delta_{n_1,L_1} - \delta_{n_1,1}) + \mathbf{F}_2(\delta_{n_2,L_2} - \delta_{n_2,1})] \]

\[ = \frac{1}{\zeta} \left[ \delta_{p_00} \{ f_{p_1}(L_1) - f_{p_1}(1) \} \frac{\mathbf{F}_1}{L_1} + \delta_{p_0j} \{ f_{p_2}(L_2) - f_{p_2}(1) \} \frac{\mathbf{F}_2}{L_2} \right]. \]

Note that spatial symmetry for the modes is broken, and that equation (15) is replaced by similar equations, but also depending on the spatial components \( i \) and \( j \) of the vectors.

\[ X_{piqj}(t) \equiv \langle X_{pi}(t) X_{qj}(0) \rangle = \frac{\mathbf{H}_{pi} \mathbf{H}_{qj}}{\alpha_p^2} + \frac{k_B T}{\zeta N \beta_p \alpha_p} \exp \left[ -\alpha_p t \right] \delta_{pq} \delta_{ij} \quad \text{with} \quad p \neq 0. \]

The assumption \( \mathbf{F}_1 \perp \mathbf{F}_2 \) is necessary in order for the modes to be the exact dynamical eigenmodes. If the tensile forces are not orthogonal, then it will result in correlations between some of the modes (and a correspondingly parallelogram-like structure of the membrane).

It is of course interesting to investigate the transverse fluctuations of the membrane under lateral tension. Two observables we consider are the effective thickness and the additional surface area under thermal undulations. A quantity that can be used as a measure for the thickness of the membrane is the standard deviation of the relative height of the bead in the middle of the membrane with respect to its mean position. In the limit of a continuous very large membrane, this thickness is given by

\[ D = \sqrt{\langle R_{m}^2 \rangle} = \sqrt{\frac{k_B T}{4k}}. \]

We verified numerically that the fluctuations increase for beads near the edge of the membrane, in line with self-avoiding membranes \[34, 35\]. Note that this thickness is unaffected by the strength of the tensile forces.

A much more involved calculation is needed for the area of the membrane. Consider a very large square membrane with \( N = L^2 \) beads and perpendicular tensile forces \( F_1 = F_2 = F \) of equal strength. We consider the case where the temperature is low enough, or the tensile forces are strong enough, such that the fluctuations of the beads around their equilibrium are small compared to the distances between two neighboring beads. The total area of the membrane can be written as the sum of areas of all triangles between the three beads with indices \( (i, j), (i+1, j) \) and \( (i, j+1) \) plus the sum of areas of all triangles with indices \( (i+1, j), (i, j+1) \) and \( (i+1, j+1) \); the expectation value for this area, in
the continuum limit, is then given by
\[ \langle A \rangle = \frac{F^2 (L - 1)^2}{k^2} + \frac{k_B T (2L^2 - L)}{8k}. \]  
(41)

Equations (40) and (41) collectively imply that under the application of tensile forces, irrespective of the force magnitude, the out-of-plane degrees of freedom decouple from in-plane degrees of freedom. Moreover at strong stretching \( F \gg k_B T / k \) the main contribution to the area of the membrane comes from the stretching of the individual springs. As already mentioned, at strong stretching there is no difference between a phantom and a self-avoiding membrane, and therefore we expect the same for the increase in area for a self-avoiding membrane under strong stretching forces.

The eigenmodes also give access to dynamical information. For instance, to obtain the mean vector connecting two neighboring beads in the membrane \( \Delta \mathbf{R}_i(t) \equiv \mathbf{R}_{n_i+1,n_2}(t) - \mathbf{R}_{n_i,n_2}(t) \) with \( n_1 = 1, \ldots, L_1 - 1 \) and \( n_2 = 1, \ldots, L_2 \), we use \( \langle \mathbf{X}_p(t) \rangle = \mathbf{H}_p / \alpha_p \), which can be proved by solving the differential equation in a similar fashion as for equation (39), such that
\[ \langle \Delta \mathbf{R}_1(t) \rangle = \sum_p \beta_p \frac{H_p}{\alpha_p} [f_p(n_1 + 1, n_2) - f_p(n_1, n_2)], \]  
(42)
yielding \( \Delta \mathbf{R}_1(t) = \mathbf{F}_1 / k \). The same is true for neighbors in the other internal direction so that for orthogonal tensile forces, on average, the membrane obtained is a flat rectangular structure. (The above results are obtained by using
\[ \sum_{p=1}^{L-1} \sin^{-1} \left[ \frac{\pi p}{2L} \right] \sin \left[ \frac{\pi np}{L} \right] \cos \left[ \frac{\pi p}{2L} \right] = \frac{L}{2} \quad \text{for } n = 1, \ldots, L - 1, \]  
(43)
where the summation is over odd values of \( p \)). Further, the random thermal forces are Gaussian distributed with a variation given by equation (3) such that \( \langle \mathbf{X}_{p_i}(0) \rangle \) and \( \langle \mathbf{X}_p(0) \rangle \) can be used to derive the first two moments for every mode amplitude. We use these results to generate a random typical configuration for a membrane, as shown in figure 4.

5. Discussion

The Rouse model has proved to be a cornerstone for polymer dynamics. We generalized this model with two internal coordinates, for which the entity becomes a phantom membrane. We have shown that a set of eigenmodes can be used to solve the membrane dynamics in the overdamped limit and demonstrated that many interesting properties can be analytically derived for the membrane using certain relations for these eigenmodes. Furthermore, we have shown that adding large enough tensile forces to the system mimics the behavior of a realistic flexible membrane under tension: the eigenmodes can still be exactly solved analytically, making the analysis useful for practical purposes.

We also note, although the following is not relevant to studying the properties of a phantom membrane, that the entire analysis presented in this paper is easily generalized for any number \( d \geq 1 \) of internal coordinates. For every internal coordinate, the Rouse modes and inverse get a factor \( f_p(m_i) \), \( \beta_p \) is a product over \( d \) factors, and \( \alpha_p \) becomes a summation over \( d \) terms. By generalizing the potential energy so that all neighbors are connected, equations (8)–(15) remain valid in their current forms. It can also be shown
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Figure 4. A randomly generated membrane consisting of $25 \times 25$ beads in a three-dimensional space, with orthonormal tensile forces, $k_B T = 1/2$, and $k = \zeta = 1$. The black lines connects neighboring beads. Due to tensile forces the membrane adopts a rectangular lattice-like structure, where thermal forces push the beads out of their mean positions.

that adding a tensile force for each internal coordinate still makes the system analytically solvable. Similarly, the analysis can also be extended to incorporate periodic boundaries (resulting in a torus or cylindrical geometry, as appropriate, when the internal dimension $d = 2$).

References

[1] Rouse P E Jr, 1953 J. Chem. Phys. 21 1272
[2] Doi M, 2001 Introduction to Polymer Physics (Oxford: Oxford University Press) reprinted
[3] Doi M and Edwards S F, 2001 The Theory of Polymer Dynamics (Oxford: Clarendon) reprinted
[4] Panja D and Barkema G T, 2009 J. Chem. Phys. 131 154903
[5] Panja D, 2010 J. Stat. Mech. L02001
[6] Panja D, 2010 J. Stat. Mech. P06011
[7] Nelson D, Piran T and Weinberg S, 2004 Statistical Mechanics of Membranes and Surfaces (Singapore: World Scientific) reprinted
[8] Lipowsky R and Sackmann E, 1995 Structure and Dynamics of Membranes, Handbook of Biological Physics vol 1 (Amsterdam: Elsevier)
[9] Picart C and Discher D E, 1999 Biophys. J. 77 865
[10] Sackmann E, 2002 ChemPhysChem 3 237
[11] Hwa T, Kokufuta E and Tanaka T, 1991 Phys. Rev. A 44 R2235
[12] Spector M S, Naranjo E, Chiruvolu S and Zasadzinski J A, 1994 Phys. Rev. Lett. 73 2867
[13] Wen X et al, 1992 Nature 355 426
[14] Kantor Y, Kardar M and Nelson D R, 1986 Phys. Rev. Lett. 57 791
[15] Kantor Y, Kardar M and Nelson D R, 1987 Phys. Rev. A 35 3056
[16] Essafi K, Kownacki J-P and Mouhanna D, 2011 Phys. Rev. Lett. 106 128102
[17] Kownacki J-P and Mouhanna D, 2009 Phys. Rev. E 79 R040101

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[18] Koibuchi H, Kusano N, Nidaira A and Suzuki K, 2004 Phys. Rev. E 69 066139
[19] Münkel C and Heermann D W, 1995 Phys. Rev. Lett. 75 1666
[20] Plischke M and Boal D, 1988 Phys. Rev. A 38 4943
[21] Radzihovsky L and Toner J, 1995 Phys. Rev. Lett. 75 4752
[22] Mori S and Komura S, 1996 J. Phys. A: Math. Gen. 29 7439
[23] Liu D and Plischke M, 1992 Phys. Rev. A 45 7139
[24] Abraham F F, Rudge W E and Plischke M, 1989 Phys. Rev. Lett. 62 1757
[25] Kroll D M and Gompper G, 1993 J. Physique I 3 1131
[26] Zhang Z, Davis H T and Kroll D M, 1993 Phys. Rev. E 48 R651
[27] Muthukumar M, 1988 J. Chem. Phys. 88 2854
[28] Wiese K J, 1998 Eur. Phys. J. B 1 269
[29] Wiese K J, 1998 Eur. Phys. J. B 1 273
[30] Popova H and Milchev A, 2008 Phys. Rev. E 77 041906
[31] Boal D, Levinson E, Liu D and Plischke M, 1989 Phys. Rev. A 40 3292
[32] Drovetsky B Y, Chu J C and Mak C H, 1998 J. Chem. Phys. 108 6554
[33] Babu S B and Stark H, 2011 Eur. Phys. J. E 34 136
[34] Gompper G and Kroll D M, 1992 J. Physique I 2 663
[35] Abraham F F and Nelson D R, 1990 Science 249 393