Understanding the dynamics of rings in the melt in terms of the annealed tree model

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Abstract
The dynamical properties of a long polymer ring in a melt of unknotted and unconcatenated rings are calculated. We re-examine and generalize the well known model of a ring confined to a lattice of topological obstacles in light of the recently developed Flory theory of untangled rings which maps every ring on an annealed branched polymer and establishes that the backbone associated with each ring follows self-avoiding rather than Gaussian random walk statistics. We find the scaling of the ring relaxation time and diffusion coefficient with ring length, as well as the time dependence of stress relaxation modulus, zero shear viscosity and the mean square averaged displacements of both individual monomers and the ring’s mass centre. Our results agree within error bars with all available experimental and simulation data of the ring melt, although the quality of the data so far is insufficient to make a definitive judgement for or against the annealed tree theory. At the end we review briefly the relation between our findings and experimental data on chromatin dynamics.

Keywords: polymer ring melt, dynamics, annealed tree, lattice animal

(Some figures may appear in colour only in the online journal)

1. Introduction

Recent progress in experimental studies of genome 3D organization in eukaryotic nuclei (see e.g. [1–3]) along with earlier theoretical suggestions [4–6] shed light on the delicate interplay of high density and topological constraints in genome 3D structure and dynamics. This interplay certainly belongs to the most challenging and least understood aspects of polymer physics [7–10]. While there are still many unanswered questions related to the statics and the structure of dense topologically constrained polymer matter, their dynamics are even more difficult. In this paper we consider the simplest problem of this type. We claim our problem to be the simplest because of two decisive simplifications: The first simplification is we examine a prototypical system exhibiting competition between high density and topological constraints—the melt of unknotted and unconcatenated rings. The relation of this model to genome folding was discussed at length (see e.g. [6, 7, 11, 12]), so here we do not discuss that. The relevance of our dynamic results and their potential to clarify experiments on chromatin dynamics is discussed at the very end of our paper. In the main text we work on the model system—on passive dynamics of rings.

Our second crucial simplification comes directly in the context of the ring melt: we will construct the dynamical description based on the recently proposed static Flory-like theory [13]. The origin of that theory goes back to works in [14–17]. As a reminder, the work [14] considered a single loop, having no excluded volume and placed in a lattice of topological obstacles, so that the loop is not threaded by any of the obstacles (see figure 1(a)). In this case, the ring adopts the branched conformation of a tree (also called a lattice animal in this context), with each branch representing a doubly folded section of the ring (figure 1(b)); it was shown [14], that this tree is randomly branched. In [15–18] the authors examined the dynamics of this system.

Since a randomly branched tree has a gyration radius $R$ scaling as $N^{1/4}$, $N$ being the polymerization index (see e.g. [19]), these results do not directly apply to the real rings with excluded volume in 3D (and even more so in 2D), unless $N$ is rather small. Nevertheless, the authors of the work [14] made a very far-sighted comment at the end of their article.
suggestion that ring compaction in the lattice of obstacles is an indication of globular conformations (with gyration radius \( \sim N^{1/3} \)) of the real rings in an unconcentrated melt. Subsequent developments, particularly recent large scale simulations [9, 20] seem to have accepted this idea of compactness (\( R \sim N^{1/3} \)) for each ring in an unconcentrated melt. The Flory-type theory [13] was set to rationalise this finding.

To do so, the work [13] assumed to be in line with [14–17], that crumpled conformation of a ring in an unconcentrated melt can be mapped on a tree, with two important modifications against [14–17]: first, tree branches do not have to be doubly folded down to the monomer scale, they may include (and are likely to include) loops; secondly, the tree is not assumed to be random, rather its branches are viewed as annealed, subject to thermal equilibrium. The latter is described by the special order parameter denoted as \( L \) and is called backbone.

An important piece of evidence in support of the existence of an underlying tree structure for every ring in the melt was provided in a recent simulation work [20]. There, the authors demonstrated quantitatively accurate mapping of the rings on properly chosen trees. We should also mention that tree-like structures were implicated in a chromatin context in the work [21].

To make the present paper self-contained, we include in appendix A a further discussion of \( L \), its meaning and definition. For the same purpose, we discuss in the appendix B the (simpler case of) analogous theory in 2D, where the Flory theory is, as usual, not exact but very accurate.

In this paper, our plan is to use methods of the work [17] to analyse the dynamics of rings viewed as annealed, but not ideal branched objects. In [17], the ideal branched object statistics were considered. The backbone of the ring was assumed to behave as a random walk in space implying the gyration radius \( R \) scaling with the backbone \( L \) as \( R \sim L^{1/2} \). The length \( L \) itself was assumed to scale with the number of monomers of the ring \( N \) and also with exponent \( 1/2 \) i.e. \( L \sim N^{1/2} \), which gives the exponent \( \nu \) governing the spatial size of the ring (gyration radius) \( R \sim N^{\nu} \) to be \( \nu = 1/4 \) [22]. In contrast, the Flory-type theory [13] establishes that rings in the melt represent annealed but not ideal trees, with \( R \sim N^{1/3} \) i.e. \( \nu = 1/3 \) while the backbone scales as \( L \sim N^\rho \) with \( \rho = 1/3\nu_F \simeq 0.567 \) where \( \nu_F = 0.588 \) is the Flory exponent of a regular self-avoiding walk in 3D. The exponents \( \nu = 1/3 \) and \( \rho \simeq 0.567 \) represent compact ring conformations with the backbone following self-avoiding random walk statistics as is shown below. The goal of this paper is to reproduce the main dynamic arguments of [17] for the new exponents \( \nu \) and \( \rho \) and to compare them with the most detailed computer data available [23].

### 2. Calculation details

Here, we derive the time dependence of the stress relaxation modulus, the mean square displacements of monomers and centre of mass and the behaviour of the zero shear viscosity as a function of the ring mass. At first, we briefly summarize the conformational properties of the rings with the annealed tree statistics from [13] and review the ring relaxation dynamics of [17]. Later in section 3 we show how these theoretical predictions compare with experiments [24, 25] and extensive numerical simulations [23] and in section 4 we discuss the relation to dynamics of eukaryotic DNA.

Following [13], the ring is an annealed tree of ‘entanglement’ blobs each consisting of about \( N_e \) monomers, where the entanglement length \( N_e \) is the crossover between the Gaussian and compact regime \( \nu = 1/3 \) for the ring gyration radius \( R \).

\[
R \sim \begin{cases} 
N_e^{1/2} & \text{for } N < N_e \\
N_e^{1/3} N_e^{1/16} & \text{for } N > N_e
\end{cases}
\]

The monomer size and Kuhn segment are assumed for simplicity to be of the same order and are taken as a unit of length. The gyration radius of the Gaussian ‘entanglement’ blob \( N_e^{1/2} \) sets the lattice constant of the topological mesh formed by other rings in the melt. As mentioned above, the backbone of the annealed tree \( L \) scales with the ring length as \( L \sim N^\rho \) with \( \rho = 1/3\nu_F \) where \( \nu_F \simeq 0.588 \) is the Flory exponent of a regular self-avoiding walk in 3D. This says the backbone of the annealed tree is a self-avoiding random walk in space because \( R \sim L^{\nu_F} \sim L^{1/3} \). To summarize, while the annealed tree conformations are characterized by exponents \( \nu = 1/3 \) and \( \rho \simeq 0.567 \), the randomly branched (ideal) trees are described by \( \nu_id = 1/4 \) and \( \rho_id = 1/2 \).

The theoretical work [17] that we follow showed an important difference between relaxation of a ring and a linear polymer. The ring relaxation time is dominated by its backbone as that is the most stable structure of the conformation. Diffusion of the blobs along the backbone of the conformation is modelled as a diffusion of stored length between different branches (side loops) that serve as reservoirs of the
blobs of stored length (figures 1(b) and (c)). This approach tries
to capture the change of the shape of the ring during a diffusion of
a blob along the ring caused by a diffusion of other blobs.

We consider scaling estimates for all quantities examined in
the computational work [23]. Let us start with relaxation time
of the entire ring. Displacement of a single blob to a
neighbouring reservoir at a distance of one lattice constant,
takes about a Rouse time \( t_0 \sim N_e^2 \xi / kT \), where \( \xi \) is the fluid
friction of a monomer. Each blob represents a fraction \( N_i/N_e \)
of the total mass of the ring, which means a single blob displace-
ment to a neighbouring reservoir \( N_i^{1/2} \) away causes the
centre of mass to move a distance \( N_e^{1/2} / (N/N_e) \). There are
about \( (N/N_e)^p \) reservoirs that exchange the blobs between
themselves in time \( t_0 \). The different blob displacements are as-
sumed to be mutually independent (and non-interacting) hence
the mean square displacement of the centre of mass of a ring is
\( \Delta r_{cm}^2 \sim (N/N_e)^p N_e / (N/N_e)^2 = N_e (N/N_e)^{p-2} \). These
displacements with a diffusion coefficient \( D_{cm}^2 \sim \Delta r_{cm} / t_0 \) take place
however along the backbone of the ring. The relaxation of
the whole ring is achieved when the centre of mass is dis-
placed by a distance of the order of the whole backbone, that
is in time \( \tau \sim L^2 / D_{cm}^2 \) which is found to scale as
\[
\tau \sim t_0 (N/N_e)^{p+2}
\]  

From here the scaling of the diffusion coefficient of the
ring in real space is also derived in [17]. In the relaxation time
\( \tau \) the ring is displaced by a distance of its own size and we find
\( D \sim R^{2/\tau} / \tau \) hence
\[
D \sim D_0 (N/N_e)^{2-\rho-2},
\]  
where \( D_0 = N_0 / t_0 \) is the diffusion coefficient of the
entanglement blob.

To find the stress relaxation modulus we use the standard
single polymer dynamics approach [19]. Until time \( t_0 \) the
Gaussian blobs of \( N_e \) monomers are relaxing by Rouse modes
and the stress relaxation modulus decays inversely proportional
to the square root of time. At time \( t_0 \) the stress modulus is
\( kT \) per volume of entanglement blob i.e. \( G(t_0) \sim kT / N_e \).
After \( t_0 \) the relaxation time \( \tau_p \) of the \( p \)-th mode can be cal-
culated as a relaxation time of a ring of \( N/N_e p \) segments
due to the self-similarity of the ring conformation, hence
\( \tau_p \sim t_0 (N/N_e p)^{p+2} \). We can express the number of modes
that relax at time \( t \) as
\[
p(t) \approx (N/N_e) (t_0 / t)^{1/(p+2)}. 
\]  
At time \( t \), segments of length \( N/N_e p(t) \) relax and the stress
relaxation modulus is \( kT \) per volume occupied by such
segments.
\[
G(t) N_e \sim kT (N/N_e)^{-p} \sim kT \left( \frac{t_0}{t} \right)^{1/(p+2)} .
\]  
This holds until the whole ring relaxes \( (t < \tau) \) after which an
exponential decay begins.
\[
G(t) N_e / kT \sim \begin{cases} 
(t/t_0)^{-1/2} & \text{if } t < t_0 \\
(t/t_0)^{-1/(p+2)} & \text{if } t_0 < t < \tau \\
(t/t_0)^{-1/(p+2)} e^{-t/\tau} & \text{if } t > \tau 
\end{cases}
\]  
At intermediate times there is no plateau modulus as for the
linear polymer melts, but the stress modulus decays according
to power-law. This is because the relaxation of a ring continues
in a self-similar manner above the entanglement length, while
the linear chain relaxes only after it reptates out of the
entanglement tube. The separation of timescales in the linear
case, where nothing can be said to relax on the time scale
intermediate to \( t_0 \) and reptation time, is responsible for the
plateau modulus.

Zero-shear viscosity can be calculated from the stress
relaxation modulus as \( \eta_0 = \int_0^\infty G(t) dt \) which is up to a
numerical factor equal to
\[
\eta_0 \sim G(\tau) \sim kT (N/N_e)^{p+1} t_0.
\]  

Three different types of mean-square displacement (MSD) are
computed in [23]:

(a) the MSD averaged over all monomers of a ring
\( g_1(t) = \langle |r(t) - r(0)|^2 \rangle \)
(b) MSD of monomers with respect to the centre of mass
\( g_2(t) = \langle |r(t) - r_{cm}(t) - r(0) + r_{cm}(0)|^2 \rangle \)
(c) MSD of the centre of mass
\( g_3(t) = \langle |r_{cm}(t) - r_{cm}(0)|^2 \rangle \).

Let’s consider \( g_1(t) \). First, the Rouse relaxation, charac-
terized by \( g_1(t) \sim t^{-1/2} \) takes place up to the time \( t_0 \) where
blobs of size \( N_e^{1/2} \) are relaxed. Next, at time \( t \) segments of
length \( N/N_e p(t) \) relax and move a distance of the order of
their size: \( g_1(t_p) \sim N_e (N/N_e p(t))^{2\rho} \). Substituting for \( p(t) \)
from (3), we get
\[
g_1(t) \sim N_e (t/t_0)^{2\rho/(2\rho+2)}. 
\]  
This continues until time \( \tau \) when the whole ring is relaxed and
ordinary diffusion sets off. Hence for the MSD averaged over
all monomers we have
\[
g_1(t)/N_e \sim \begin{cases} 
(t/t_0)^{1/2} & \text{for } t < t_0 \\
(t/t_0)^{2\rho/(2\rho+2)} & \text{for } t_0 < t < \tau \\
(N/N_e)^{2\rho/(2\rho+2)} (t/\tau) & \text{for } t > \tau 
\end{cases}
\]  

The prefactor \( (N/N_e)^{2\rho} \) ensures a smooth crossover between
the intermediate and late regime.

Now consider the MSD of the centre of mass \( g_3 \). On	timescale \( t_p \) a segment of length \( N/N_e p \) moves a distance
of the order of \( (N/N_e p)^\rho \). There are about \( N_e p \) of these
segments in the ring hence the centre of mass moves
\( (N/N_e p)^\rho \) for each of them. Since these displacements
are independent of each other (the relaxation on longer scales
will come at a later time), the total MSD is just \( g_3(t) = N_e p(N/N_e p)^\rho \sim N_e (N_e p(t))^{2\rho-1} \). Using again
the mode time dependence (3) we get
\[
N g_3(t) \sim (t/t_0)^{(2\rho+1)/(2\rho+2)} \]  
At this time \( g_3(t) \) is standard Rouse behaviour at early times. Again after time
\( \tau \) the normal diffusion starts. The MSD of the centre of mass
follows
\[
N g_3(t)/N_e^2 \sim \begin{cases} 
(t/t_0)^{1/(2\rho+2)} & \text{for } t < t_0 \\
(t/t_0)^{(2\rho+1)/(2\rho+2)} & \text{for } t_0 < t < \tau \\
(N/N_e)^{2\rho/(2\rho+2)} (t/\tau) & \text{for } t > \tau 
\end{cases}
\]  
where the factor \( (N/N_e)^{2\rho+1} \) guarantees the smooth crossover.
MSD $g_2$ is the difference between $g_1$ and $g_3$ and it crosses over from Rouse $t^{1/2}$ at early times to a constant at later times $t > \tau$.

3. Results

Let us now compare the results with available data. Experimentally it is difficult to purify a melt of rings of linear contaminants that in sufficient amount have strong effects on stress relaxation [24]. Various studies agree that the critical concentration of the linear chains beyond which the effects on viscosity of the melt become substantial lies well below the concentration of the linear chains [24]. Various studies agree that the critical concentration of the linear chains beyond which the effects on stress relaxation [24] are immobile. In reality, the constraints also move and relax due to the motion of the other chains. To account for this effect, the reptation theory can be adjusted by contour length fluctuations or constraint release [28] to give the exponents

$1 + \rho \simeq 1.567$ for the self-avoiding backbone and 1.5 for the ideal trees. See figure 3. If we take the exponent for the linear melt to be 3.4 we predict $\eta_0 \text{linear}/\eta_0 \text{ring}$ to have the exponent 1.833 and 1.9 for annealed and ideal trees respectively which is still higher than the reported experimental value. The lower exponents found in the experiments can be attributed again to the linear contaminants. The linear contaminants would certainly increase the ring viscosity by threading the rings’ configuration, as noted in [25–27], hence decreasing the exponent of $\eta_0 \text{linear}/\eta_0 \text{ring}$. More experimental data is necessary to quantify the role of the contaminants and to estimate the coefficient of the pure melt of rings more accurately.

Numerical studies of a pure melt of rings [23,27] provide indispensable information for all the calculated dynamical quantities. The numerical results for the diffusion coefficient give scaling $D \sim N^{-2.3}$ which is off by 0.4 from our prediction (2) of the exponent $2\nu - \rho - 2 \simeq -1.9$. However, a similar discrepancy between the effective theory and experiment is found even for melts of linear chains, where the reptation theory predicts $D \sim N^{-2}$ while experimental and numerical results are $D \sim N^{-2.4}$ [23]. In the linear case the difference is attributed to the assumption that the topological obstacles are immobile. In reality, the constraints also move and relax due to the motion of the other chains. To account for this effect, the reptation theory can be adjusted by contour length fluctuations or constraint release [28] to give the exponents that agree with the experiments. At the moment we don’t know how to incorporate the relaxation of the topological constraints in the theory, but if the same corrections are assumed for the rings as found in the linear case, the simulation agrees well: in both cases the theoretical prediction of the exponent is off by 0.4 from the numerics. Using the exponents $\nu_d = 1/4$ and $\rho_d = 1/2$ for the ideal trees, the exponent is $-2$—the same as predicted by the reptation theory for linear melts.

The most important numerical observation is that the diffusion coefficient for the rings and linear melts behaves as $\eta_0 \text{linear}/\eta_0 \text{ring}$. This agrees with both predictions of the ring melt viscosity exponent (6) $1 + \rho \simeq 1.567$ for the self-avoiding backbone and 1.5 for the ideal trees. See figure 3. If we take the exponent for the linear melt to be 3.4 we predict $\eta_0 \text{linear}/\eta_0 \text{ring}$ to have the exponent 1.833 and 1.9 for annealed and ideal trees respectively which is still higher than the reported experimental value. The lower exponents found in the experiments can be attributed again to the linear contaminants. The linear contaminants would certainly increase the ring viscosity by threading the rings’ configuration, as noted in [25–27], hence decreasing the exponent of $\eta_0 \text{linear}/\eta_0 \text{ring}$. More experimental data is necessary to quantify the role of the contaminants and to estimate the coefficient of the pure melt of rings more accurately.
Figure 4. Different relaxation times as a function of ring length $N$. The symbols are numerical results from [23]: diffusive relaxation time (circles); conformational relaxation times: $\tau_{cc}$ (squares) represent decay of the correlation of two vectors connecting monomers 0 with $N/2$ and $N/4$ with $3N/4$, respectively — see details in [23]; $\tau_{R^2}$ represents the decay of the $R^2$ autocorrelation function (diamonds), the solid line has a slope $\rho + 2 \approx 2.567$ (annealed trees) by equation (1), the dashed line has a slope $5/2$ (ideal trees), while the dot-dashed is the best fit of diffusive relaxation time with slope 2.68.

(nearly) identically, but the viscosities diverge dramatically. This suggests decoupling of the relaxation from the diffusion as the ring can relax without diffusing much. This is reproduced by the present theory.

The numerical study presents three relaxation times to characterize the different aspects of relaxation. The diffusive relaxation time and the correlation time of the gyration radius follow each other, while the internal rearrangement time is much smaller and also grows slightly more slowly with the mass of the ring (see figure 4). This can be understood in the context of the present theory as the internal rearrangement by blob diffusion around the ring can take place without the ring diffusing too much. Our calculated exponents for the relaxation time, $2 + \rho = 2.567$ for trees with self-avoiding backbone and 2.5 for ideal trees, are both slightly below the best fit exponent for the diffusive time 2.68. The larger exponent for the trees with self-avoiding backbone is a consequence of their less compact conformation due to self-avoidance hence the ring has to diffuse further to relax.

The time dependence of the mean square displacements exhibit the Rouse behaviour at early times characterized by exponent $1/2$ for $g_1$ and 1 for $g_3$ (figure 5). At intermediate times we found for $g_1$ exponent $2\nu/(2 + \rho)$ which is about 0.260 for the annealed trees and 0.2 for the ideal case. For $g_3$ the exponent is $(2\nu + 1)/(\rho + 2)$ that is 0.649 for the annealed trees and 0.6 for ideal ones.

While $g_3$ agrees well at intermediate times (figure 7), the exponent of $g_1$ (figure 6) is found to be somewhat larger in the simulation [23] (best fit gives about 0.3, not shown). A reason for this numerical observation might be that for the simulated ring lengths the intermediate regime is not long enough to capture the complete crossover to the calculated exponent. Nevertheless, numerically we clearly see a decrease of the exponent at the intermediate timescales. An additional reason can be the assumption that treats the topological obstacles as immobile. As noted above the validity of this assumption at intermediate to late times is questionable as a significant rearrangement already started at the scale of the branches i.e. the scales larger than the lattice constant. This is also suggested by the numerical results of the internal rearrangement relaxation time.

Another recent theoretical and numerical study [29] investigated $g_1$ of a long linear polymer in a dense fractal conformation. Even though the polymer has open ends it is prepared in a conformation with similar properties (gyration radius and contact probability scaling) to those measured for the rings in melt. Then the mean squared displacement of the monomers is measured in the process of equilibration, where an exponent of around 0.38 at intermediate times is found (indicated in figure 6). The reason for comparing this study with the present work and simulation [23] is the common principle of topological constraints governing the genome folding and the conformation of a ring in the melt. The different results from
the two simulations deserve a more in-depth study as they can help us to understand the genome-ring melt correspondence. The analytical part of [29] (predicting exponent of about 0.4) and the present work rely on different assumptions, but both take a mean-field approach. While [29] models the effect of surrounding chains by exploiting the chain’s surface, here we rely on the model of a ring as an annealed tree. Both mean-field approaches neglect density fluctuations that might have an effect on the critical exponents as was demonstrated for the tube model in linear melts [30] and in the recently numerically studied ring in a gel through the effect of self-threading [18]. It is an open question to investigate the sensitivity of the critical exponents of both the statics and dynamics of the melt of rings beyond the mean-field considerations.

Both theories, of the ideal and the self-avoiding backbone, agree very well with the experimental and numerical evidence. Unfortunately, the rather weak dependence of the measurable exponents on $\rho$ in the investigated range does not allow us to conclude in favour of any of the competing theories.

4. Discussion

As we mentioned in the introduction above, the melt of rings in general and its dynamics in particular is a challenging fundamental problem of polymer physics. At the same time, as we also mentioned, this problem appears to have deep connections to the genome 3D structure. Accordingly, here we discuss the possible relevance of our model findings for the genome folding field.

Experimentally, quite a few works investigated the 3D dynamics of chromatin in the cell nuclei of mammals [31–37], drosophila [38, 39] and yeast [40]; genome dynamics in bacterial nucleoid was studied as well [41, 42]. In most cases fluorescent microscopy was used. The common theme of all experimental results is a subdiffusive behaviour, typically with a power close to 0.4: $g_1 \sim t^{0.4}$. Such behaviour is reported for both, parts of the genome itself such as telomeres [31–33] or certain gene loci [33, 40], as well as foreign nanoparticles immersed into the nucleus [34–36, 39]. Typically subdiffusive motion is reported over a limited interval of time—such that the overall displacement during this time is pretty small, usually only a few tens of nanometers, well below 0.1 [$\mu$m].

Although the observation of sub-diffusion is encouraging in terms of the application of polymer models with topological constraints, the detailed quantitative comparison is certainly premature at this stage. The main reason is the fact that genome dynamics in the cell is active, driven and ATP dependent [37, 43]. Active hydrodynamics of chromatin was recently considered in [44]. One interesting observation (see specifically figure 3 in the work [44]) is that chromat dynamics in active and starved cells appear to be different only on length scales in excess of one micron or so. This may give credence to more detailed association of subdiffusive behaviour in folded genomes and in topologically restricted polymer models. On a more cautious note, the goal of our study of polymer dynamics should be to understand their rheological behaviour in terms of moduli dependence on both frequency and wave vector—the information that must be fed into the phenomenological theory of chromatin hydrodynamics [44]. Our present work should be viewed as a step in that direction.

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Appendix A. Gyration radius distribution for a tree and backbone definition—a generalization of Kramers theorem

The Flory-type theory for rings [13] assumes that ring conformation is controlled by the balance of two competing entropic effects. On the one hand, approaching the doubly folded conformation makes it easy to penetrate other rings, which is entropically favourable. On the other hand, double folding the ring is entropically unfavourable. The former factor is described by free energy $\sim R^2/L$, the latter $\sim L^2/N$; their balance is in the heart of the Flory theory [13]. To understand it better, including the meaning of $L$, we derive here the $R^2/L$ result in greater detail.

As usual in the Flory theory, the $R^2/L$ free energy should be viewed as the entropy price of deforming a Gaussian polymer, in this case—a tree with quenched branches.

The entropic price for swelling a tree can be calculated from the probability distribution of gyration radius $R$, particularly the tail of the distribution when $R$ is large. As will be shown below, this also defines the backbone $L$. The approach is modelled after the work [46] (with small corrections in [47, 48]) and [49] [46–48] the considered probability distribution of gyration radius for a linear Gaussian polymer; [49] did the same for a phantom Gaussian ring).

Suppose for simplicity that the functionality of branch points in the tree is just $z = 3$ and there are only branch points and ends, namely, $n$ branch points and $n + 2$ ends, i.e. a total
of \( N = 2n + 2 \) monomers. If \( \vec{r}_i \) are position vectors of these ‘monomers’ then the gyration radius reads

\[
R^2 = \frac{1}{2N^2} \sum_{i,j} (\vec{r}_i - \vec{r}_j)^2. \tag{A.1}
\]

On the tree, every \( \vec{r}_i - \vec{r}_j \) is uniquely represented as the sum of the proper set of bond vectors \( \vec{\eta}_k = \vec{r}_i - \vec{r}_j \), where \( i \) and \( j \) are monomers connected by bond \( k \). Accordingly, gyration radius can be represented as

\[
R^2 = \sum_{k,m} G(k,m) \vec{\eta}_k \vec{\eta}_m, \tag{A.2}
\]

where the indices \( k \) and \( m \) label bonds (unlike \( i, i' \) and \( j \) above, which label nodes or monomers) and \( G(k,m) \) is an interesting \( N - 1 \) by \( N - 1 \) matrix, illustrated in figure A1: each matrix element is given by \( G(k,m) = K(k)M(m) / N^2 \), where \( K(k) \) is the number of monomers on one side of bond \( k \), while \( M(m) \) is similarly the number of monomers on the other side of bond \( m \).

For a Gaussian system (without excluded volume and when ‘bonds’ are long enough to persist length), each bond vector \( \vec{\eta} \) is Gauss distributed and vectors \( \vec{\eta}_k \) and \( \vec{\eta}_m \), with \( k \neq m \), are independent. Accordingly, the formula (A.2) returns the known result for the mean squared average gyration radius of the tree, it involves only diagonal elements \( G(k,k) \) and is given by the trace of that matrix:

\[
\langle R^2 \rangle = a^2 \sum_k G(k,k) = a^2 \sum_{k=1}^{N-1} \frac{K(k)(N-K(k))}{N^2}, \tag{A.3}
\]

which is Kramer’s theorem [50] which can be found also in e.g. [19]; here \( a^2 = \langle \vec{\eta}^2 \rangle \) is the mean squared bond length.

For a Gaussian tree we can do more and find not only the averaged \( R^2 \), but the probability distribution of \( R^2 \), since each bond has the probability distribution \( \sim e^{-\frac{1}{2}a^2} \). The characteristic function of \( R^2 \) reads

\[
\Phi(s) = \langle e^{isR^2} \rangle = A \int e^{-\sum_{k,m} \frac{s}{2a^2} \sum \eta_k \bar{\eta}_m G(k,m)} d^3 \{ \vec{\eta} \}, \tag{A.4}
\]

where the explicit expression for the normalization factor \( A \) is dropped for brevity. Rotating the coordinate system in this \( N = 1 \) dimensional space of \( \{ \bar{\eta} \} \) to the basis of eigenvectors \( \{ \xi \} \) of matrix \( G \), we obtain

\[
\Phi(s) = A \int e^{-\sum_k \frac{s}{2a^2} (1-2is\lambda_k/3)} d^3 \{ \vec{\xi} \}
\]

\[
= \frac{N-1}{\prod_{p=1}^{N-1} (1-2is\lambda_p/3)^{-3/2}}, \tag{A.5}
\]

where \( \lambda_p \) are eigenvalues. From here, finding the probability distribution of \( R^2 \) is a matter of the inverse Fourier transform of \( \Phi(s) \)

\[
P(R^2) = \frac{1}{2\pi} \int e^{-isR^2-\frac{1}{2} \sum_p \ln(1-2is\lambda_p/3)} ds. \tag{A.6}
\]

Since we are interested in the behaviour of \( P(R^2) \) at large \( R \) the asymptotic is controlled by the singularity of \( \Phi(s) \) which is closest to the origin in a complex \( s \)-plane, that corresponds to the largest eigenvalue \( \lambda_{\max} \). In the vicinity of this singularity there is a saddle point which dominates the integral and we can evaluate the inverse Fourier transform integral by the steepest descent. The equation for saddle point location reads \( R^2 \sim \frac{a^2}{2\lambda_{\max}} \). From here the result of the saddle point integration (up to logarithmic corrections) is

\[
P(R^2)_{|R \to \infty} \sim e^{-\frac{a^2}{2\lambda_{\max}}} R^2 \sim e^{-\frac{3a^2}{2\lambda_{\max}}} \lambda_{\max} = e^{-\frac{3a^2}{2\lambda}}. \tag{A.7}
\]

It is difficult to find the maximal eigenvalue \( \lambda \) in general for arbitrary tree configuration, but looking at the equation (A.7) it is natural to imply that \( a^2 \lambda \) is the mean squared average gyration radius up to a numerical factor \( c \): this is what we did in the last step in formula (A.7). Here as above, the mean squared average \( \langle R^2 \rangle \) is the thermal average, given by a formula (A.3), for the given tree i.e. it does not involve the average over different tree topologies. Assuming that the numerical coefficient \( c \), which is a property of any given tree, is of order unity, we drop it for the purposes of scaling analysis in the framework of the Flory theory.

The entropic price for swelling the tree is hence given by

\[
\Delta S(R) = -c R^2 / \langle R^2 \rangle = -R^2 / aL. \tag{A.8}
\]

Since each end-to-end arm in the tree is a Gaussian linear polymer, the latter relation says that \( L \) is a properly (and non-trivially) averaged length of all such arms. In other words we can define \( L \) in terms of eigenvalue \( \lambda \): \( L = 2a\lambda / 3 \). Although the result \( R^2 / L \) for the swelling entropy was known long ago [51], we should emphasize its non-trivial character because when the tree swells, all of its branches swell, not just one.

The result (A.7) or (A.8) is the generalization of Kramer’s theorem [19, 50].

Appendix B. Unconcatenated rings in 2D

In this Appendix, we discuss the statistical equilibrium properties of the unconcatenated ring melt in 2D. The goal is to understand more clearly the underlying annealed tree model, its accuracy and limitations.
In 2D, instead of unconcatenated rings we should think of a system of rings where no ring can be inside another ring. For such a system, the double-folded conformation is not a hypothesis nor an approximation, but an obvious fact. Furthermore the resulting trees are definitely annealed. The Flory theory [13] is generalized straightforwardly in this case yielding $R \sim N^{1/2}$ and $L \sim N^\nu$ with $\nu = 1/2 \nu_{\text{LERW}}$, where $\nu_{\text{LERW}} = 3/4$ is the Flory exponent for a regular self-avoiding walk in 2D. In reality as we explain below based on the works [52–56] $\rho = 1/2 \nu_{\text{LERW}}$, where $\nu_{\text{LERW}} = 4/5$ is the exponent of the loop-erased random walk. If true, this indicates the Flory theory in this case is not exact, but pretty close.

The fact that $\rho = 1/2 \nu_{\text{LERW}}$ in 2D means that the backbone of the ring is a loop-erased random walk with a gyration radius $R \sim L^{1/2 \nu_{\text{LERW}}}$. We will demonstrate this by mapping a melt of linear chains onto a melt of rings and extract the statistics of the backbone of the underlying trees. Numerical simulations [52] and exact calculations [53] of the 2D melt of linear chains show, that chains adopt a compact conformation of a system of rings where no ring can be inside another.

Furthermore the resulting trees are definitely annealed. The ring. For such a system, the double-folded conformation is therefore, described by the same index $\nu_{\text{LERW}}$. This procedure can be repeated for every pair of neighbouring chains.

In this way we map the melt of linear chains in 2D onto the melt of annealed trees in 2D. By circumscribing the ring around each tree, we can also construct the melt of unconcatenated rings. Thus we obtained one-to-one mapping between all three systems—the melt of unconcatenated rings, the melt of linear chains and the melt of annealed trees. All of them are, therefore, described by the same index $\nu_{\text{LERW}}$.

Additionally, the same index also describes a uniform spanning tree [55] (and, therefore, single linear chain or single loop) densely confined inside a region with smooth boundaries. A similar problem of a single unknotted loop in a cavity in 3D was recently studied by simulations [8], the analogy with the melt of rings is confirmed, but perhaps not to the extent of exact mapping as in 2D.

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