Universal size ratios in complex polymers: Radius of gyration vs hydrodynamic radius

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I. INTRODUCTION

In statistical description of polymers, a considerable attention is paid to the universal quantities describing equilibrium size and shape of typical conformation adapted by individual macromolecule in a solvent. In particular, many physical properties are manifestations of the underlying polymer conformation, including the hydrodynamic properties of polymer fluids, the folding dynamics and catalytic activity of proteins etc. As a size measure of a single macromolecule one usually considers the mean square radius of gyration \( R_g^2 \), which is directly measurable in static scattering experiments. Denoting coordinates of the monomers along the polymer chain by \( \vec{r}_n \), \( n = 1, \ldots, N \), this quantity is defined as:

\[
\langle R_g^2 \rangle = \frac{1}{2N^2} \sum_{n,m} \langle (\vec{r}_n - \vec{r}_m)^2 \rangle ,
\]

and is thus given by a trace of gyration tensor \( \mathbf{Q} \). Here and below, \( \langle \ldots \rangle \) denotes ensemble average over possible polymer conformations. Another important quantity that characterizes the size of a polymer coil is hydrodynamic radius \( R_H \), which is directly obtained in dynamic light scattering experiments. This quantity was introduced based on the following motivation [11]. According to the Stokes-Einstein equation, the diffusion coefficient \( D \) of a spherical particle of radius \( R_s \) in a solvent of viscosity \( \eta \) at temperature \( T \) is given by:

\[
D = \frac{k_B T}{6 \pi \eta R_s}
\]

where \( k_B \) is Boltzmann constant. In order to generalize the above relation for the case of molecules of more complex shape, their center-of-mass diffusion coefficient \( D \) is given by Eq. (2) with \( R_s \) replaced by \( R_H \). The latter is given as the average of the reciprocal distances between all pairs of monomers:

\[
\langle R_H^{-1} \rangle = \frac{1}{N^2} \sum_{n,m} \left\langle \frac{1}{|\vec{r}_n - \vec{r}_m|} \right\rangle .
\]

Namely, \( R_H \) is related with the averaged components of the Oseen tensor \( \mathbf{H}_{nm} \) characterizing the hydrodynamic interactions between monomers \( n \) and \( m \) [12]. To compare \( R_g^2 \) and \( R_H^{-1} \), it is convenient to introduce the universal size ratio

\[
\rho = \sqrt{R_g^2/R_H},
\]

which does not depend on any details of chemical microstructure and is governed by polymer architecture. In the present paper we restrict our consideration to the ideal (Gaussian) polymers, i.e. monomers have no excluded volume, which corresponds to the case of so-called \( \Theta \)-solvents. This approach allows to obtain the exact analytical results for the set of universal quantities characterizing conformational properties of macromolecules. In particular, for a linear Gaussian polymer chain the exact analytical result for the ratio \( \rho \) in \( d = 3 \) dimensions reads [13, 14]:

\[
\rho_{\text{chain}} = \frac{8}{3\sqrt{\pi}} \approx 1.5045.
\]

The universal ratio of a Gaussian ring polymer was calculated in Refs. [14, 16, 17] and is given by

\[
\rho_{\text{ring}} = \frac{\sqrt{2\pi}}{2} \approx 1.2533.
\]

The validity of theoretically derived ratios \( \rho_{\text{chain}} \) and \( \rho_{\text{ring}} \) was confirmed in several simulation studies [15, 17, 18].

Polymer macromolecules of complex branched structure attract considerable attention both from academical and applied perspective, being encountered as building blocks of materials like synthetic and biological gels, thermoplastics, melts and elastomers. High functionality of polymers provides novel properties with applications in diverse fields like drug delivery, tissue engineering, super-soft materials, and antibacterial surfaces etc. On the other hand, multiple loop formation in macromolecules is often encountered and plays an important role in biological processes such as stabilization of globular proteins or transcriptional regularization of genes. [31]
In this concern, it is of fundamental interests to study conformational properties of complex polymer architectures. The distinct example of branched macromolecule is the so-called rosette polymer \[^{33}\], containing \( f^c \) linear chains and \( f^r \) closed loops (rings), radiating from the same branching point (see Fig. 1). Note that for \( f^r = 0 \) one restores architecture of a star polymer with \( f^c \) functionalized linear chains radiating from a central core, for which an exact analytical result is known for the size ratio (Ref. \[^{33}\]):

\[
\rho_{\text{star}} = \frac{8\sqrt{f(3f^c - 2)}}{3(f^c)^2\sqrt{\pi}}(\sqrt{2} - 1)(\sqrt{2} + f^c).
\]  

(7)

The estimates for \( \rho_{\text{star}} \) have been also obtained by numerical Monte-Carlo simulations \[^{35}\]. Using molecular dynamics (MD) simulations, Uehara and Deguchi derived the universal size ratios for macromolecules such as single ring \( (f^c = 0, f^r = 1) \), tadpole \( (f^r = 1, f^c = 1) \) and double ring \( (f^r = 2, f^c = 0) \) \[^{17}\]. The overview of existing literature data for universal size ratios obtained in analytical \( \rho_{\text{theory}} \) and numerical \( \rho_{\text{sim}} \) investigations are listed in Table I.

The aim of the present work is to extend the previous analysis of rosette-like polymers \[^{33}\], by thoroughly studying their universal size characteristics. For this purpose we apply the analytical theory, based on path-integration method, and extensive numerical molecular dynamics simulations. The layout of the paper is as follows. In the next section, we introduce the continuous chain model and provide the details of analytical calculation of the universal size ratios \( \rho \) for various polymer architectures applying path integration method. In section IV we describe the numerical model and details of MD simulations. In the same section we present numerical results and compare them with our theoretical predictions. We draw conclusions and remarks in section IV.

### II. ANALYTICAL APPROACH

#### A. The model

Within the frame of continuous chain model \[^{30}\], a single Gaussian polymer chain of length \( L \) is represented as a path \( \bar{r}(s) \), parameterized by \( 0 < s < L \). In this model the Hamiltonian of a polymer chain reads:

\[
H = \frac{1}{2} \int_0^L ds \left( \frac{d\bar{r}}{ds} \right)^2.
\]  

(8)

We adapt this model to more complicated branched polymer topologies, containing in general \( f^c \) linear branches and \( f^r \) closed rings (see figure 1). In the following, let us use notation \( f = f^c + f^r \) for total functionality of such structure. The corresponding partition function of rosette polymer is given by:

\[
Z_{f^c,f^r} = \int d^d \{ \bar{r} \} D_{f^c,f^r} e^{-\frac{1}{2} \sum_{i=1}^f \int_0^L ds \left( \frac{d\bar{r}_i}{ds} \right)^2},
\]  

(9)

where \( d^d \{ \bar{r} \} \) denotes multiple path integration over trajectories \( \bar{r}_i(s) \) \( (i = 1, \ldots, f) \) assumed to be of equal length \( L_1 = L \), and the factor \( D_{f^c,f^r} \) determines the topology of the molecule:

\[
D_{f^c,f^r} = \prod_{i=1}^f \delta(\bar{r}_i(0)) \prod_{j=1}^{f^r} \delta(\bar{r}_j(L) - \bar{r}_j(0)).
\]  

(10)

Here, the first product of \( \delta \)-functions reflects the fact that all \( f^c + f^r \) trajectories start at the same point (central core), and the second \( \delta \)-functions product up to \( f^r \) describes the closed ring structures of \( f^r \) trajectories (their starting and end points coincide).

Within the frame of presented model, the expression for the mean square gyration radius from Eq. (11) can be rewritten as:

\[
\langle R_g^2 \rangle = \frac{1}{2(fL)^2} \sum_{i,j=1}^f \int_0^L \int_0^L ds_2 ds_3 \langle |\bar{r}_i(s_2) - \bar{r}_j(s_1)|^2 \rangle,
\]  

(11)

#### Table I: Literature data for the universal size ratio for different polymer topologies, derived using analytical theory \( \rho_{\text{theory}} \) and numerical simulations \( \rho_{\text{sim}} \). The theoretical values for tadpol and double ring architectures were calculated on the basis of our general analytical result, cf. Eq. (19).

| Topology    | \( f^c \) | \( f^r \) | \( \rho_{\text{theory}} \) | \( \rho_{\text{sim}} \) |
|-------------|----------|----------|--------------------------|--------------------------|
| Chain       | 1        | 0        | 1.5045 Eq. (5)           | 1.5045 ± 0.0005 [18]     |
| Ring        | 0        | 1        | 1.253 Eq. (6)            | 1.253 ± 0.013 [17]       |
| Star        | 2        | 0        | 1.150 Eq. (7)            | 1.28 [35]                |
| Star        | 3        | 0        | 1.40 Eq. (7)             | 1.11 [35]                |
| Star        | 4        | 0        | 1.33 Eq. (7)             | 1.04 [35]                |
| Tadpol      | 1        | 1        | 1.415 Eq. (20)           | 1.380 ± 0.021 [17]       |
| Double ring | 0        | 2        | 1.217 Eq. (21)           | 1.215 ± 0.011 [17]       |
Below we will apply path integration approach to calculate the hydrodynamic radius:}

\[
\langle R_H^{-1} \rangle = \frac{1}{(fL)^2} \sum_{i,j=1}^f \int_0^L \int_0^L ds_2 ds_1 \langle |\vec{r}_i(s_2) - \vec{r}_j(s_1)|^{-1} \rangle,
\]

where \(\langle \ldots \rangle\) denotes averaging over an ensemble of all possible configurations defined as:

\[
\langle \ldots \rangle = \frac{1}{Z_{f,c,f,r}} \int d\vec{r} \, D_{f,c,f,r}(\ldots) \times -\frac{i}{2} \sum_{i,j=1}^f \int ds_2 (|\vec{r}_i(s_2)|^2)^2 \ . \tag{13}
\]

\[\text{B. Calculation of hydrodynamic radius and universal size ratio}\]

The crucial point in the calculation of the hydrodynamic radius is utilization of the following equality \[37\]:

\[
|\vec{r}|^{-1} = (2\pi)^{-d} \int d\vec{k} 2^{d-1} \pi^{d-1} |\vec{k}|^{d-1} \Gamma \left( \frac{d-1}{2} \right) k^{1-d} e^{i\vec{k} \cdot \vec{r}}. \tag{14}
\]

where \(\Gamma(x)\) is Gamma function. Applying the above expression to Eq. \[12\] allows to rewrite the mean reciprocal distance from the definition of \(R_H\) as

\[
\langle |\vec{r}_i(s_2) - \vec{r}_j(s_1)|^{-1} \rangle = (2\pi)^{-d} \int d\vec{k} 2^{d-1} \pi^{d-1} |\vec{k}|^{d-1} \times \Gamma \left( \frac{d-1}{2} \right) k^{1-d} e^{i\vec{k} \cdot (\vec{r}_i(s_2) - \vec{r}_j(s_1))}. \tag{15}
\]

Below we will apply path integration approach to calculate the mean reciprocal distances. To visualize different contributions into \(\langle |\vec{r}_i(s_2) - \vec{r}_j(s_1)|^{-1} \rangle\), it is convenient to use the diagrammatic technique (see Fig. 2). The final result is obtained by performing the integrations over the restriction points \(s_1\) and \(s_2\). The analytic expressions, corresponding to diagrams \((1)-(5)\) on Fig. 2 lead:

\[
\langle |\vec{r}_i(s_2) - r_i(s_1)|^{-1} \rangle_{(1)} = \int_0^L ds_2 \int_0^{s_2} ds_1 \left( s_2 - s_1 \right)^{\frac{d}{2}} ,
\]

\[
\langle |\vec{r}_i(s_2) - r_j(s_1)|^{-1} \rangle_{(2)} = \int_0^L ds_2 \int_0^{s_2} ds_1 \left( s_2 - s_1 \right)^{\frac{d}{2}} ,
\]

\[
\langle |\vec{r}_j(s_2) - r_i(s_1)|^{-1} \rangle_{(3)} = \int_0^L ds_2 \int_0^{s_2} ds_1 \left( s_2 - s_1 \right)^{\frac{d}{2}} ,
\]

\[
\times \left( s_2 - s_1 - \frac{(s_2 - s_1)^2}{L} \right)^{\frac{d}{2}} ,
\]

\[
\langle |\vec{r}_i(s_2) - r_j(s_1)|^{-1} \rangle_{(4)} = \int_0^L ds_2 \int_0^{s_2} ds_1 \left( s_2 - s_1 - \frac{(s_2 - s_1)^2}{L} \right)^{\frac{d}{2}} ,
\]

\[
\times \left( s_2 + s_1 - \frac{s_2^2}{L} - \frac{s_1^2}{L} \right)^{\frac{d}{2}} ,
\]

\[
\langle |\vec{r}_j(s_2) - r_j(s_1)|^{-1} \rangle_{(5)} = \int_0^L ds_2 \int_0^{s_2} ds_1 \left( s_2 - s_1 - \frac{s_2^2}{L} \right)^{\frac{d}{2}} .
\]

Note that in the above equations the factor \(\Gamma\left(\frac{d-1}{2}\right)\) has been omitted. Also, each diagram in Fig. 2 is associated with the corresponding combinatorial factor. Namely, the contribution \((1)\) in above expressions is taken with the pre-factor \(f^c\), contribution \((2)\) with \(f^{(d-1)/2}\), \((3)\) with \(f^c\), \((4)\) with \(f^{(d-1)/2}\) and the last contribution \((5)\) with the pre-factor \(f^c f^c\). Summing up all contributions from Eq. \[16\] with taking into account corresponding pre-factors we finally obtain the expression for the hydrodynamic radius of a rosette structure:

\[
\langle R_{h,\text{rosette}} \rangle = \frac{\Gamma\left(\frac{d-1}{2}\right)}{\Gamma\left(\frac{d}{2}\right) \sqrt{2}} 12 (f^c + f^r)^2 \sqrt{L} \times
\]

\[
\left[ -6 f_r \pi \left( \sqrt{2} f^r - 1 \right)^2 + 16 \left( \sqrt{2} - 1 \right) f^c \left( \sqrt{2} + f^c \right) + 3 f^c f^r \left( 10 \arcsin \left( \frac{\sqrt{5}}{5} \right) - \pi + 4 \right) \right]^{-1} . \tag{17}
\]

The expression for the mean square gyration radius of a rosette architecture is \[33\]:

\[
\langle R_{g,\text{rosette}}^2 \rangle = \frac{L d}{12 (f^r + f^c)^2} [f^r(2f^r - 1) + 2f^c(3f^c - 2) + 8f^c f^r] . \tag{18}
\]

Finally, using Eqs. \[17\] and \[18\], we calculate the the universal size ratio \[4\] of rosette-like polymer architecture in Gaussian approximation:

\[
\mu_{\text{rosette}} = \frac{\sqrt{6} d \Gamma\left(\frac{d-1}{2}\right)}{72 (f^r + f^c)^2 \Gamma\left(\frac{d}{2}\right) \times}
\]

\[
\sqrt{6} (f^c)^2 \sqrt{6} f^c f^r + 2 (f^r)^2 - 4 f^c - f^r \times
\]

\[
\left[ -6 f_r \pi \left( \sqrt{2} f^r - 1 \right)^2 + 16 \left( \sqrt{2} - 1 \right) f^c \left( \sqrt{2} + f^c \right) + 3 f^c f^r \left( 10 \arcsin \left( \frac{\sqrt{5}}{5} \right) - \pi + 4 \right) \right] . \tag{19}
\]
For $f^c = 1$ and $f^r = 1$ we find analytic expression for the so-called tadpole architecture:

$$\rho_{\text{tadpole}} = \frac{\sqrt{22}}{96\sqrt{\pi}} \left[ 3\pi + 28 + 30 \arcsin \left( \frac{\sqrt{5}}{5} \right) \right] \approx 1.415. \tag{21}$$

In Fig. 3 we plot calculated theoretical values of the universal size ratio vs number of functionalized chains for stars comprised of linear polymers with $f^c > 0$, $f^r = 0$ (red symbols) and ring polymers $f^r > 0$, $f^c = 0$ (blue) as well as rosette polymers with equal number of grafted linear chains and rings $f^r = f^c > 0$ (purple). For all architectures we observe decrease in $\rho$ with increasing functionality. In the next subsection we compare our theoretical predictions with the result of MD simulations.

### III. NUMERICAL APPROACH

#### A. The method

Numerical data in this work have been obtained from MD simulations. We consider simple three-dimensional numerical model of a rosette polymer consisting of arms which are $f^c$ linear chains and/or $f^r$ ring polymers. Each arm is composed of $N$ sizeless particles of equal mass $m$ connected by bonds. We study ideal (Gaussian) conformations of rosette polymers in dilute solution corresponding to conformations of real rosette polymers at dilute $\theta$ solvent conditions. In our numerical model the connectivity along the polymer chain backbone is assured via harmonic potential

$$V(r) = \frac{k}{2}(r - r_0)^2, \tag{22}$$

where $k = 200 k_B T/b^2$ is the interaction strength measured in units of thermal energy $k_B T$ and $r_0$ is the equilibrium bond distance.

The molecular dynamics simulations were performed by solving the Langevin equation of motion for the position $\mathbf{r}_i = [x_i, y_i, z_i]$ of each monomer,

$$m\ddot{\mathbf{r}}_i = \mathbf{F}_i - \zeta \dot{\mathbf{r}}_i + \mathbf{F}^{\text{stochastic}}_i, \quad i = 1, \ldots, fN, \tag{23}$$

which describes the motion of bonded monomers. Forces $\mathbf{F}_i$ in Eq. (23) above are obtained from the harmonic interaction potential between Eq. (22). The second and third term on the right hand side of Eq. (23) is a slowly evolving viscous force $-\zeta \dot{\mathbf{r}}_i$ and a rapidly fluctuating stochastic force $\mathbf{F}^{\text{stochastic}}_i$ respectively. This random force $\mathbf{F}^{\text{stochastic}}_i$ is related to the friction coefficient $\zeta$ by the fluctuation-dissipation theorem $\langle \mathbf{F}^{\text{stochastic}}_i(t) \mathbf{F}^{\text{stochastic}}_j(t') \rangle = k_B T \delta_{ij} \delta(t - t')$.

The friction coefficient used in simulations was $\zeta = 0.5 m \tau^{-1}$ where $\tau = [mb^2/(k_B T)]^{1/2}$ is the unit of time. A Langevin thermostat was used to keep the temperature constant. The integration step employed to solve the equations of motions was taken to be $\Delta t = 0.0025 \tau$. All simulations were performed in a cubic box with periodic boundary conditions imposed in all spatial dimensions. We used Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [38] to perform simulations. Simulation snapshots were rendered using Visual Molecular Dynamics (VMD) [39].

#### B. Results

Simulations of rosette polymers were performed for the following number of monomer beads per arm $N = 100, 200, 400, 800, 1600$ and $6400$. The number of arms for star polymers composed of solely linear chains (i.e. with $f^r=0$) and ring polymers (i.e. with $f^c = 0$) were varied in the range between 1 to 4. In the case of rosette polymers which are hybrid polymer architectures comprised
of linear chains and ring polymers we considered two arm functionalities with \( f^c = f^r = 1 \) and 2. To increase conformational sampling each simulation was carried out with 50 identical molecules in a simulation box. In the course of simulations the universal size ratio was measured, cf. Eq. (4). In the numerical calculation of quantities like \( \rho \) a crucial aspect is finite degree of polymerization \( N \) that we are dealing with in simulations, while theoretically obtained values of \( \rho \) hold in the asymptotic limit \( N \to \infty \). Thus, the finite-size effects (or corrections to scaling) should be appropriately taken into account.

For the size ratio of an ideal linear chain, this correction is given by

\[
\rho = \rho_\infty (1 + aN^{-\Delta}),
\]

where \( \rho_\infty \) is the asymptotic value obtained at \( N \to \infty \), \( a \) is non-universal amplitude, \( \Delta \) is the correction-to-scaling exponent for \( \theta \)-solvent is \( \Delta = 1/2 \) [15] whereas for good solvent conditions is \( \Delta \approx 0.53 \) [18]. In our numerical analysis we use Eq. (24) to obtain the universal size ratio in the asymptotic limit for all considered architectures. For this purpose we plot \( \rho \) vs correction-to-scaling term \( N^{-1/2} \) and get \( \rho = \rho_\infty \) for \( N \to \infty \).

In Fig. 4 we display the results of our MD simulations for two “benchmark” systems which are Gaussian linear chain (red circles) and Gaussian ring (blue circles). For both architectures systematic increase in the size ratio is observed with increasing value of \( N \). In the asymptotic limit \( N \to \infty \) we obtain \( \rho_{\text{chain}} = 1.499 \pm 0.005 \) and \( \rho_{\text{ring}} = 1.244 \pm 0.004 \). These numerical values with very good accuracy reproduce known theoretical results. The latter are given by Eq. (5) for linear chains and by (6) for rings. The complete list of numerically derived universal size ratios and their comparison to theoretical values can be found in Table II.

In Fig. 5 we show numerically derived universal size ratios as a function of \( N^{-1/2} \) for more complex architectures. We investigated conformations of stars comprised of linear chains, stars of ring polymers and rosette polymers with equal number of grafted linear and ring chains. For all architectures we observe systematic approaching to asymptotic values predicted by theory with increasing value of \( N \) per arm. For stars of linear chains with
which matches theoretically predicted value for this type of polymer from Eq. (21). For rosette polymers with $f^c = 2$ and $f^r = 2$ our simulations provide $1.295 \pm 0.018$ which is comparable with the corresponding value calculated from the formula given in Eq. (19). The full list of calculated values of $\rho$ is in Table I.

### IV. CONCLUSIONS

We have studied by combination of analytical theory and molecular dynamics simulations conformational properties of rosette polymers which are complex macro-molecules consisting of $f^c$ linear chains (branches) and $f^r$ closed loops (rings) radiating from the central branching point. Our focus was on characterizing structure of these polymers in theta solvents. For this purpose we investigated basic structural quantities such as the mean square radius of gyration $R^2_g$, the hydrodynamic radius $R_H$ and most importantly the universal size ratio $\rho \equiv \sqrt{R^2_g/R_H}$. Our calculations demonstrated gradual decrease in $\rho$ with increasing functionality $f = f^c + f^r$ of grafted polymers. The analytical results are in perfect agreement with our numerical simulations data. Since both quantities $R^2_g$ and $R_H$ are directly accessible via correspondingly static and dynamic scattering techniques we hope that our results will stimulate further experimental studies on the behavior of complex polymer architectures in solutions.

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