



Universal features of polymer shapes in crowded environments

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ABSTRACT

We study the universal characteristics of the shape of a polymer chain in an environment with correlated structural obstacles, applying the field-theoretical renormalization group approach. Our results qualitatively indicate an increase of the asymmetry of the polymer shape in crowded environment comparing with the pure solution case.

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1. Introduction

In studying the transport properties of polymer fluids, an important role is played by the shape characteristics of a single polymer chain configuration. It is established [1,2], that the typical polymer chain realization does not possess spherical symmetry. A quantity, which characterizes the asymmetrical shape of a polymer chain, is the asphericity ratio \hat{A}_d of the chain configuration. \hat{A}_d attains a maximal value of one for a completely stretched, rod-like configuration, and equals zero for the spherical form (see Fig. 1), thus obeying the inequality: $0 \leq \hat{A}_d \leq 1$. In the limit of long chains this quantity appears to be universal and depends on space dimension d only: $\hat{A}_d > 1/2$ at $d < 4$, $\hat{A}_d = 1/2$ at $d \geq 4$. The size measure of a flexible polymer chain is usually defined by either the mean-squared end-to-end distance R_e or radius of gyration R_G . The ratio of these quantities, the so-called size ratio $g \equiv \langle R_e^2 \rangle / \langle R_G^2 \rangle$ again is an universal quantity ($g > 6$ for $d < 4$, $g = 6$ for $d \geq 4$). The study of these universal quantities characterizing the polymer shape is a subject of a great interest [3–6].

A current problem in polymer physics is the influence of structural obstacles (impurities) in the environment on the universal properties of macromolecules. Such a disordered (crowded) environment can be found, in particular, in a biological cell, composed of many different kinds of biochemical species, which may oc-

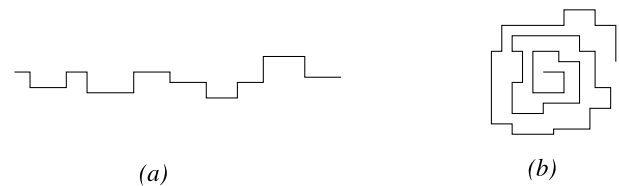


Fig. 1. Schematic presentation of a polymer configurations with asphericity ratio \hat{A}_d value close to one (a) and close to zero (b).

cupy a large fraction of the total volume [7]. In the language of lattice models of polymers, the crowded environment with structural obstacles can be considered as a disordered lattice, where a given fraction of randomly chosen sites are to be avoided by the polymer chain. It has been proven both analytically [8] and numerically [9], that weak uncorrelated disorder that corresponds to the point-like randomly distributed obstacles with concentration far from the percolation threshold does not change the universality class of polymers. In the present study, we address a model, where the structural obstacles of the environment are spatially correlated on a mesoscopic scale [10]. Following Ref. [11], this case can be described by assuming the defects to be correlated on large distances r according to a power law with a pair correlation function $h(r) \sim r^{-a}$. For $a < d$ such a correlation function describes defects extended in space (see [11,12] for further details). The impact of long-range-correlated disorder on the scaling of single polymer chains has been analyzed in previous works [12] by means of the field-theoretical renormalization group (RG) approach. The intrigu-

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ing question of how the shape characteristics of a flexible chain are influenced by presence of such a disordered medium remains, however, still unresolved, and is the subject of the present study.

The layout of the Letter is as follows. In the next section, we describe the model of the flexible polymer chain in a crowded environment, develop its field-theoretical formulation and give a brief description of the field-theoretical RG approach. The results of its application to the present model and finally some conclusions and an outlook are given in the following sections.

2. Field-theoretical description of the model

Let $\vec{R}_n = \{x_n^1, \dots, x_n^d\}$ be the position vector of the n th monomer of the polymer chain ($n = 1, \dots, N$). The shape of a specific conformation of the chain can be characterized [2] in terms of the gyration tensor \mathbf{Q} with components:

$$Q_{ij} = \frac{1}{2N^2} \sum_{n,m=1}^N (x_n^i - x_m^i)(x_n^j - x_m^j), \quad i, j = 1, \dots, d. \quad (1)$$

For the averaged radius of gyration R_G one thus has:

$$\langle R_G \rangle = \frac{1}{2N^2} \left\langle \sum_{n,m=1}^N |\vec{R}_n - \vec{R}_m|^2 \right\rangle = \left\langle \sum_{i=1}^d Q_{ii} \right\rangle = \langle \text{Tr} \mathbf{Q} \rangle. \quad (2)$$

Here and below, $\langle \dots \rangle$ denotes averaging over all configurations of the polymer chain. The spread in eigenvalues λ_i of the gyration tensor measures the asymmetry of a given configuration. In Ref. [5] it was proposed to characterize the shape of macromolecule by ratios of rotationally invariant polynomials in the components of \mathbf{Q} . Let us define the asphericity ratio \hat{A}_d as the quotient of two averaged quantities [5]:

$$\hat{A}_d = \frac{1}{d(d-1)} \sum_{i=1}^d \frac{\langle (\lambda_i - \bar{\lambda})^2 \rangle}{\langle (\bar{\lambda})^2 \rangle} = \frac{d}{d-1} \frac{\langle \text{Tr} \hat{\mathbf{Q}}^2 \rangle}{\langle (\text{Tr} \mathbf{Q})^2 \rangle}. \quad (3)$$

Here, $\bar{\lambda} \equiv \text{Tr} \mathbf{Q}/d$ is the mean eigenvalue of gyration tensor for a given chain configuration and $\hat{\mathbf{Q}} \equiv \mathbf{Q} - \bar{\lambda} \mathbf{I}$ with the unity matrix \mathbf{I} . \hat{A}_d equals zero when all configurations are spherical (eigenvalues of each given configuration are equal) and takes the maximal value of one when all configurations are rod-like (all the eigenvalues equal zero except one). Besides the ratio of averages one may also calculate the so-called mean asphericity, given by an average of the ratio [13]. While Ref. [13] has shown some slight quantitative deviations between the two approaches it confirmed that the qualitative dependence with respect to other parameters, e.g. on the architecture of the molecule is the same in each case.

Passing from the discrete model to a continuous polymer chain description [14] allows to derive a field-theoretical formulation of the asymptotic shape characteristics of a single polymer chain in an environment with correlated structural obstacles. Applying the replica method in order to average the free energy over different configurations of the randomly distributed and fixed obstacles leads to an m -component field theory with a Lagrangian L_{Dis} [12]:

$$L_{\text{Dis}} = \frac{1}{2} \sum_{\alpha=1}^n \int d^d x \left[(\hat{\mu}_0^2 |\vec{\phi}_\alpha(x)|^2 + |\nabla \vec{\phi}_\alpha(x)|^2) + \frac{u_0}{4!} (\vec{\phi}_\alpha(x))^2 \right] - \sum_{\alpha,\beta=1}^n \int d^d x d^d y h(|x-y|) \vec{\phi}_\alpha^2(x) \vec{\phi}_\beta^2(y). \quad (4)$$

Here each replica $\vec{\phi}_\alpha$ is an m -component vector field $\vec{\phi}_\alpha = (\phi_\alpha^1, \dots, \phi_\alpha^m)$, $\hat{\mu}_0$ and u_0 are bare mass and coupling, the coupling of the replicas is described by the correlation function $h(r) \sim r^{-a}$ and both the polymer ($m \rightarrow 0$) and the replica ($n \rightarrow 0$) limits are

implied. For small k , the Fourier-transform $\tilde{h}(k)$ of $h(r)$ behaves as: $\tilde{h}(k) \sim v_0 + w_0 |k|^{a-d}$. Taking this into account, rewriting Eq. (4) in the momentum space, and recalling special symmetry peculiarities of (4) that appear for $m, n \rightarrow 0$ [12], a theory with two bare couplings u_0, w_0 results. Note that for $a \geq d$ the w_0 -term is irrelevant in the RG sense and one restores the pure case (absence of structural defects).

As shown in Ref. [5], computing the shape parameters of long polymer chain can be reduced to computing the critical amplitudes and exponents of the corresponding field-theoretical model. In order to extract the scaling behavior of the model (4) we apply the RG method [15] to get the Green's functions $G_R^{(L,N)}$ renormalized at non-zero mass and zero external momenta. The change of couplings u_0, w_0 under renormalization defines a flow in the parametric space, governed by corresponding β -functions: $\beta_u(u, w) = \frac{\partial u}{\partial \ln l} |_0$, $\beta_w(u, w) = \frac{\partial w}{\partial \ln l} |_0$, where l is the rescaling factor, and $|_0$ stands for evaluation at fixed bare parameters. The fixed points (FPs) u^*, w^* of the RG transformation are given by common zero of β -functions. The stable FP corresponds to the critical point of the system.

Following Ref. [5], the averaged moments of the gyration tensor \mathbf{Q} needed to determine the polymer shape characteristics (2) and (3) can be expressed in terms of the connected Green's function. In particular:

$$\langle Q_{ij} \rangle = -\frac{1}{2} \left(\frac{DT}{2\bar{X}} \right)^{2\nu} \frac{\Gamma(\gamma)}{\Gamma(\gamma + 2\nu + 2)} \frac{G_{ij}}{G_R^{(2)}(0, 0, \{\lambda^*\})}, \quad (5)$$

$$\langle Q_{ij} Q_{kl} \rangle = -\frac{1}{4} \left(\frac{DT}{2\bar{X}} \right)^{4\nu} \frac{\Gamma(\gamma)}{\Gamma(\gamma + 4\nu + 4)} \frac{G_{ij|kl}}{G_R^{(2)}(0, 0, \{\lambda^*\})}, \quad (6)$$

where D, T, \bar{X} are non-universal quantities which will drop out in the final expressions, $\Gamma(x)$ is the Euler gamma-function, ν and γ are the critical exponents of model (4), and the following notations are used:

$$G_{ij} \equiv \left(\frac{\partial}{\partial q^i} \frac{\partial}{\partial q^j} G_R^{(2,2)}(0, 0; \mathbf{q}, -\mathbf{q}; \{\lambda^*\}) \right) \Big|_{|\mathbf{q}|=0}, \quad (7)$$

$$G_{ij|kl} = \left(\frac{\partial}{\partial q_1^i} \frac{\partial}{\partial q_1^j} \frac{\partial}{\partial q_2^k} \frac{\partial}{\partial q_2^l} \times G_R^{(2,4)}(0, 0; \mathbf{q}_1, -\mathbf{q}_1, \mathbf{q}_2, -\mathbf{q}_2; \{\lambda^*\}) \right) \Big|_{|\mathbf{q}|=0}. \quad (8)$$

Here $G_R^{(2,2)}(0, 0; \mathbf{q}, -\mathbf{q}; \{\lambda^*\})$ and $G_R^{(2,4)}(0, 0; \mathbf{q}_1, -\mathbf{q}_1, \mathbf{q}_2, -\mathbf{q}_2; \{\lambda^*\})$ are the fixed point values of the renormalized two-point connected Green's functions with two and four ϕ^2 -insertions, the symbol $|_{|\mathbf{q}|=0}$ indicates that the corresponding expressions are to be taken at all external momenta $\{\mathbf{q}\}$ equal to zero.

The isotropy of the model implies, in particular, that $\langle \text{Tr} \mathbf{Q} \rangle = d \langle Q_{xx} \rangle$, so that:

$$\langle R_G \rangle = d \langle Q_{xx} \rangle. \quad (9)$$

For the mean-squared end-to-end distance $\langle R_e^2 \rangle$ one has [5]:

$$\langle R_e^2 \rangle = -\left(\frac{DT}{2\bar{X}} \right)^{2\nu} \frac{\Gamma(\gamma)}{\Gamma(\gamma + 2\nu)} \frac{(\nabla_{\mathbf{k}}^2 G_R^{(2)}(\mathbf{k}, -\mathbf{k}, \{\lambda^*\}))|_{\mathbf{k}=0}}{G_R^{(2)}(0, 0, \{\lambda^*\})}, \quad (10)$$

where $\nabla_{\mathbf{k}}^2$ means differentiation over components of external momentum \mathbf{k} . The not-universal quantities cancel when the ratio $g = \langle R_e^2 \rangle / \langle R_G^2 \rangle$ is considered:

$$g = \frac{2\Gamma(\gamma + 2\nu + 2)}{\Gamma(\gamma + 2\nu)} \frac{(\nabla_{\mathbf{k}}^2 G_R^{(2)}(\mathbf{k}, -\mathbf{k}, \{\lambda^*\}))|_{\mathbf{k}=0}}{(\nabla_{\mathbf{q}}^2 G_R^{(2,4)}(0, 0; \mathbf{q}, -\mathbf{q}; \{\lambda^*\}))|_{\mathbf{q}=0}} \quad (11)$$

and thus the size ratio is universal quantity. The asphericity ratio \hat{A}_d can be expressed in terms of the averaged moments of gyration tensor (5), (6) as follows:

$$\hat{A}_d = \frac{\langle Q_{xx}^2 \rangle + d\langle Q_{xy}^2 \rangle - \langle Q_{xx}Q_{yy} \rangle}{\langle Q_{xx}^2 \rangle + d(d-1)\langle Q_{xx}Q_{yy} \rangle}. \quad (12)$$

One can again easily convince oneself, that all non-universal quantities in Eqs. (5), (6) cancel when calculating (12), and \hat{A}_d is a universal quantity.

Formulas (11) and (12) are used next in estimating the corresponding universal quantities.

3. Results

To obtain the qualitative characteristics of the critical behavior of the model, we exploit a double expansion in the parameters $\varepsilon = 4 - d$ and $\delta = 4 - a$, assuming them to be of the same order of magnitude. Within this approach it was shown [12], that a single polymer chain in a solvent in an environment with long-range-correlated structural obstacles belongs to a universality class different from the case of a pure solvent. In the field-theoretical renormalization group description, this is reflected by the appearance of a new non-trivial stable long-range-correlated (LR) fixed point besides the usual (pure) one. The coordinates of these FPs and their regions of stability read [12]:

$$\begin{cases} \text{pure FP: } u^* = \frac{3\varepsilon}{4}, w^* = 0 & \text{at } \delta < \varepsilon/2, \\ \text{LR FP: } u^* = \frac{3\delta^2}{2(\delta-\varepsilon)}, w^* = \frac{3\delta(\varepsilon-2\delta)}{2(\varepsilon-\delta)} & \text{at } \varepsilon/2 < \delta < \varepsilon. \end{cases} \quad (13)$$

To estimate the size ratio (11) for the case of a polymer in long-range-correlated disorder, we calculate the function $G_R^{(2,2)}(0, 0; \mathbf{q}, -\mathbf{q}; u, w)$ with two insertions $\phi^2(\mathbf{q}), \phi^2(-\mathbf{q})$. In the first order of the $\varepsilon = 4 - d, (\delta = 4 - a)$ -expansion we find:

$$\begin{aligned} G_R^{(2,2)}(0, 0; \mathbf{q}, -\mathbf{q}; u, w) &= \frac{2}{q^2 + 1} - \frac{4}{3} \frac{1}{q^2 + 1} [uI_1 - wJ_1] - \frac{2}{3} [uI_2 - wJ_2] \\ &\quad + \frac{4}{3} \frac{1}{q^2 + 1} [uI_0 - wJ_0]. \end{aligned} \quad (14)$$

Here I_i, J_i are given by the following one-loop integrals:

$$I_0 = \int \frac{d\vec{p}}{(p^2 + 1)^2}, \quad J_0 = \int \frac{d\vec{p} |p|^{a-d}}{(p^2 + 1)^2}, \quad (15)$$

$$I_1 = \int \frac{d\vec{p}}{(p^2 + 1)((p + q)^2 + 1)}, \quad (16)$$

$$J_1 = \int \frac{d\vec{p} |p|^{a-d}}{(p^2 + 1)((p + q)^2 + 1)},$$

$$I_2 = \int \frac{d\vec{p}}{(p^2 + 1)^2((p + q)^2 + 1)},$$

$$J_2 = \int \frac{d\vec{p} |p|^{a-d}}{(p^2 + 1)^2((p + q)^2 + 1)}. \quad (17)$$

To evaluate g according to Eq. (11), we perform the derivation of $G_R^{(2,2)}$ with respect to the components of the vector \mathbf{q} and expand the loop integrals in ε and δ . Further inserting the FP values (13) we finally receive:

$$g = \begin{cases} g^{\text{pure}} = 6 + \frac{\varepsilon}{16}, & \delta < \varepsilon/2, \\ g^{\text{LR}} = 6 + \frac{\delta}{8}, & \varepsilon/2 < \delta < \varepsilon. \end{cases} \quad (18)$$

Let us qualitatively estimate the change in the size ratio g , caused by presence of the structural obstacles, in three dimensions. Substituting directly $\varepsilon = 1$ into the first line of (18), we have for the

polymer chain in a pure solvent: $g^{\text{pure}} \simeq 6.06$. Let us recall, that the influence of the long-range-correlated disorder is relevant for $a \leq d$ ($\delta \geq \varepsilon$) (see e.g. explanation after Eq. (4)). Estimates of g^{LR} can be evaluated by direct substitution of continuously changing parameter δ into the second line of Eq. (18). One concludes, that increasing the parameter δ (which corresponds to an increase of strength of disorder) leads to corresponding increase of the ratio of the end-to-end to the gyration radii g .

To compute the averaged asphericity ratio using (12), we calculate the function $G_R^{(2,4)}(0, 0; \mathbf{q}_1, -\mathbf{q}_1, \mathbf{q}_2, -\mathbf{q}_2; u, w)$ with four insertions $\phi^2(\mathbf{q}_1), \phi^2(-\mathbf{q}_1), \phi^2(\mathbf{q}_2), \phi^2(-\mathbf{q}_2)$. The resulting expansion is too cumbersome to be presented here and will be given elsewhere [16]. Performing the derivatives with respect to the components of the vectors $\mathbf{q}_1, \mathbf{q}_2$ we find:

$$\begin{aligned} G_{xx} &= 576 + \frac{4028}{15}(u - w), \\ G_{xx|yy} &= 320 + \frac{436}{3}(u - w), \\ G_{xy|xy} &= 128 + \frac{308}{5}(u - w). \end{aligned} \quad (19)$$

Inserting the FP values (13) into Eqs. (19) and recalling the definitions (5) and (6) the result is:

$$\hat{A}_d = \begin{cases} \hat{A}_d^{\text{pure}} = \frac{1}{2} + \frac{15}{512}\varepsilon, & \delta < \varepsilon/2, \\ \hat{A}_d^{\text{LR}} = \frac{1}{2} + \frac{1}{48}\varepsilon + \frac{13}{768}\delta, & \varepsilon/2 < \delta < \varepsilon. \end{cases} \quad (20)$$

Again, let us qualitatively estimate the change in \hat{A}_d caused by the presence of structural obstacles in three dimensions. Substituting directly $\varepsilon = 1$ into the first line of (20), we have for the pure case: $\hat{A}_d^{\text{pure}} \simeq 0.53$. Estimates of \hat{A}_d^{LR} can be obtained by direct substitution of the continuously changing parameter δ into the second line of Eq. (20). Increase of the strength of disorder correlations results in increase of the asphericity ratio of polymers in disorder. This phenomenon can be intuitively understood if one recalls an impact of the long-range-correlated disorder on the mean end-to-end distance exponent ν . Indeed, it has been shown in [12], that such a disorder leads to an increase of ν , and subsequently, to further swelling of the polymer chain. Extended obstacles disfavor return trajectories and as a result the polymer chain becomes more elongated. This elongation then leads to an increase of the asphericity ratio as predicted by Eq. (20).

4. Conclusions and outlook

The universal characteristics of the average shape of a polymer coil configurations in a porous (crowded) environment with structural obstacles have been analyzed considering the special case, when defects are correlated at large distances r according to the power law: $h(r) \sim r^{-a}$. Applying the field-theoretical RG approach, we estimate the size ratio $g = \langle R_e^2 \rangle / \langle R_G^2 \rangle$ and averaged asphericity ratio \hat{A}_d up to the first order of a double $\varepsilon = 4 - d, \delta = 4 - a$ expansion. We have revealed, that the presence of long-range-correlated disorder leads to an increase of both g and \hat{A}_d as compared to their values for a polymer chain in a pure solution. Moreover, the asphericity ratio \hat{A}_d value was found to be closer to the maximal value of one in presence of correlated obstacles. Thus, we conclude, that the presence of structural obstacles in an environment makes the polymer coil configurations to be less spherical. Let us note that the present results, obtained in the first order of an ε, δ -expansion should serve as a qualitative estimate rather than an accurate numerical evaluation. The next step in our analysis will be to obtain the higher order expansions for the quantities of interest and to perform computer simulations in order to confirm these theoretical results by numerical values. Further details of our

calculations as well as a generalization of the presented results to the case of polymers with complex topology will be given elsewhere [16].

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