Rapid Communication

Randomly charged polymers in porous environment

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We study the conformational properties of charged polymers in a solvent in the presence of structural obstacles correlated according to a power law $\sim x^{-a}$. We work within the continuous representation of a model of linear chain considered as a random sequence of charges $q_i=\pm q_0$. Such a model captures the properties of polyampholytes – heteropolymers comprising both positively and negatively charged monomers. We apply the direct polymer renormalization scheme and analyze the scaling behavior of charged polymers up to the first order of an $\epsilon=6-d$, $\delta=4-a$ -expansion.

Key words: polymers, quenched disorder, scaling, renormalization group

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1. Introduction

Many polymer macromolecules encountered in chemical and biological physics can be considered as long flexible chains. The conformations of individual macromolecules are in general controlled by the type of monomer-monomer interactions. In good solvents, where interactions between monomers are mainly steric, N-monomer homogeneous polymer chains form coil-like structures with the mean-squared end-to-end distance R_e obeying a scaling law [1–3]:

$$\langle R_{\rm e}^2 \rangle \sim N^{2\nu_{\rm coil}},$$
 (1.1)

with a universal exponent depending on space dimension d only [e.g., the phenomenological Flory theory [1] gives $v_{\text{coil}}(d) = 3/(d+2)$]. Note that at d=4, the intrachain steric interactions are rendered irrelevant, and the polymer behaves like an idealized Gaussian chain with $v_{\text{Gauss}} = 1/2$.

The long-range nature of the electrostatic Coulomb interaction between charged monomers produces more complicated effects on polymer conformations. Of particular interest in this context are polyampholytes (PAs) [4] (for a recent review, see e.g.: [5]): heteropolymers comprising both positively and negatively charged monomers. Examples of polyampholytes are proteins and synthetic copolymers bearing acidic and basic repeat groups. The interaction between anionic and cationic groups leads to additional complications in their physical behavior. These polymers usually dissolve only when there is a sufficient amount of salt added, which screens the interactions between oppositely charged segments. The properties of polyampholytes are successfully captured within the frames of a randomly charged polymer model [6, 7, 12]: a linear chain composed of a random sequence of N monomers carrying a charge $\pm q_0$ with a fixed overall charge Q. It is established that the conformational properties of PAs strongly depend both on Q and the quality of the solvent (and thus on temperature T) [6–19]. If positive and negative charges are nearly balanced (Q is small), the attractive Coulomb interaction dominates, and the polymer collapses into a globular (sphere-like) state, as predicted by the Debye-Hückel theory [20]. For polyampholytes with a considerable disbalance between positive and negative charges ($Q > Q_c \sim q_0 N^{1/2}$), Coulomb repulsion will be predominant. They are expected to attain the properties of polyelectrolytes (homogeneously charged polymers) with a stretched configuration governed by the size exponent value $v \approx 1$.

In polymer physics, comprehension of the behavior of macromolecules in the presence of structural disorder is of great importance, e.g., in colloidal solutions [21], near microporous membranes [22], or in the crowded environment of biological cells [23-25]. The density fluctuations of obstacles often lead to spatial inhomogeneity and create pore spaces of fractal structure [26-30]. In the present study we address a model where the structural obstacles of the environment are spatially correlated on a mesoscopic scale [31]. Following reference [32], this case can be described by assuming the defects to be correlated at large distances r according to a power law with a pair correlation function $g(r) \sim r^{-a}$. Such a correlation function describes the defects extended in space, e.g., the cases a = d - 1 (a = d - 2) correspond to lines (planes) of defects of random orientation, whereas non-integer values of a can describe obstacles of fractal structures (see [32-35] for further details). The impact of long-range-correlated disorder on conformational properties of polymer chains has been analyzed in previous works [33-35] by means of the field-theoretical renormalization group (RG) approach. The question how the characteristics of a polymer with long-range Coulomb interaction are effected by the presence of such a long-range-correlated structural disorder remains, however, still unresolved.

In the present study, we aim to develop a direct renormalization group approach to analyze the conformational properties of a random charge model, proposed in references [6, 7, 12] in solution in the presence of long-range-correlated structural obstacles. The layout of the paper is as follows. In the next section, we develop a continuous chain representation of the model and discuss its general properties in detail. In section 3, the direct renormalization procedure is described and the results for the scaling properties of charged polymers are discussed. We close by giving conclusions and an outlook.

2. The model

Following reference [6, 7], we consider a flexible linear chain consisting of N monomers, each carrying a charge $\pm q_0$. If we restrict the ensemble of random charged sequences to have a fixed overall non-zero charge Q, correlations within the positive and negative charge distributions are induced. Denoting by (...) the average over the distribution of charges along the chain, one finds [12]:

$$\overline{q_i} = \frac{Q}{N}, \quad \overline{q_i^2} = q_0^2, \quad \overline{q_i q_j} = \frac{Q^2 - q_0^2 N}{N^2} = \frac{Q^2 - Q_c^2}{N^2}, \quad i \neq j,$$
(2.1)

with
$$Q_{\rm c} = \sqrt{\sum_{i=1}^{N} \overline{q_i^2}} = q_0 N^{1/2}$$
.

with $Q_{\rm c}=\sqrt{\sum_{i=1}^N\overline{q_i^2}}=q_0N^{1/2}$. Let us pass to a continuous model, where the polymer chain in a porous environment is represented by a path r(s), parameterized by $0 \le s \le L$. The probability of each path configuration is given by the Boltzmann distribution function:

$$P(\{\vec{r}\}) = \exp\left\{-\frac{1}{2}\int_{0}^{L} ds \left[\frac{dr(s)}{ds}\right]^{2} - \frac{u_{0}}{2}\int_{0}^{L} ds' \int_{0}^{L} ds'' \delta[\vec{r}(s') - \vec{r}(s'')] - \frac{1}{2}\int_{0}^{L} ds' \int_{0}^{L} ds'' \frac{q(s')q(s'')}{|\vec{r}(s') - \vec{r}(s'')|^{d-2}} - \int_{0}^{L} ds V[\vec{r}(s)]\right\}.$$
(2.2)

Here, the first term in the exponent represents the chain connectivity, the second term describes the short range excluded volume interaction with the coupling constant u_0 , the third term gives the unscreened electrostatic interaction in d dimensions (Coulomb potential), where the function q(s) represents the charges along the chain in a particular configuration, and the last term arises due to the steric interactions between the polymer chain and the structural defects in the environment given by the potential $V[\vec{r}(s)]$. Following [32], we assume the pair correlation function of defects to decay with distance according to the scaling law:

$$\langle V[\vec{r}(s')]V[\vec{r}(s'')]\rangle = w_0|\vec{r}(s') - \vec{r}(s'')|^{-a},$$
 (2.3)

here, $\langle ... \rangle$ denotes the average over different realizations of the disordered environment).

Taking into account that only the two last terms in (2.2) include random variables, we find the averaged partition function $\overline{\langle Z(L) \rangle} = \int \mathcal{D}r \exp(-\mathcal{H}_{\text{eff}})$ with an effective Hamiltonian:

$$\mathcal{H}_{\text{eff}} = \frac{1}{2} \int_{0}^{L} ds \left[\frac{dr(s)}{ds} \right]^{2} + \frac{u_{0}}{2} \int_{0}^{L} ds' \int_{0}^{L} ds'' \delta \left[\vec{r}(s') - \vec{r}(s'') \right] + \frac{v_{0}}{2} \int_{0}^{L} ds' \int_{0}^{L} ds'' \frac{1}{|\vec{r}(s') - \vec{r}(s'')|^{d-2}} + \frac{w_{0}}{2} \int_{0}^{L} ds' \int_{0}^{L} ds'' |\vec{r}(s') - \vec{r}(s'')|^{-a}.$$
(2.4)

Here, we introduce the notation $v_0 \equiv \overline{q(s)q(s')} \sim (Q^2-Q_c^2)$. We thus have a model with three types of intrachain interactions governed by coupling constants u_0 , v_0 and w_0 . From a dimensional analysis of the couplings $[u_0] = L^{-(4-d)/2}$, $[v_0] = L^{-(6-d)/2}$, $[w_0] = L^{-(4-a)/2}$ one easily concludes that for Coulomb interaction, the upper critical dimension $d_c = 6$, whereas for the excluded volume interaction $d_c = 4$, and thus the latter may be neglected in the renormalization group scheme [40–42]. In what follows, we thus restrict our considerations to a model with only two couplings v_0 and w_0 . The sign of the coupling v_0 depends on the overall charge Q of a given polyampholyte: a positive value of v_0 (i.e., $Q^2 > Q_c$) corresponds to the case, where the Coulomb repulsion is predominant in determining the conformation of the polymer chain (and thus the polymer is expected to expand).

Note that in deriving equation (2.4) we restrict the consideration to a simpler case of annealed disorder averaging, taking into account that for an infinitely long single polymer chain in random disorder, the distinction between quenched and annealed averages is negligible [36–39].

3. Renormalization and results

We follow the direct polymer renormalization scheme, developed by des Cloizeaux [2], generalizing it to the case of two intrachain interactions. Working within the frames of the continuous polymer chain model, one encounters problems with various divergences as the polymer length diverges (and thus the number of configurations tends to infinity). However, all divergencies can be eliminated by introducing renormalization factors, allowing to define and directly estimate the physical quantities of interest. The renormalization procedure is related to the existence of universal critical indices and critical factors.

Expanding the theory in the couplings v_0 and w_0 and passing to the Fourier transform according to

$$\left|\vec{r}(s') - \vec{r}(s'')\right|^{-a} \simeq \int d\vec{k} |k|^{a-d} \exp\left\{i\vec{k}\left[\vec{r}(s') - \vec{r}(s'')\right]\right\},$$

we find for the partition function of the system:

$$\overline{\langle Z(L)\rangle} = Z^{0}(L) \left[1 - \frac{4 v_{0}(2\pi)^{-\frac{(d-2)}{2}} L^{3-\frac{d}{2}}}{(4-d)(6-d)} - \frac{4 w_{0}(2\pi)^{-\frac{a}{2}} L^{2-\frac{a}{2}}}{(2-a)(4-a)} \right]. \tag{3.1}$$

Here,

$$Z^{0}(L) = \int \mathcal{D}r \exp \left\{ -\frac{1}{2} \int_{0}^{L} ds \left[\frac{dr(s)}{ds} \right]^{2} \right\}$$

denotes the partition sum of the unperturbed model (Gaussian chain). Note that only the first order terms are kept in the above relation (we restrict ourselves to the so-called "one-loop approximation" of perturbation theory).

The averaged squared end-to-end distance $R_e^2 = [r(L) - r(0)]^2$ of a typical polymer chain configuration may be calculated using the identity:

$$(\overline{\langle R_e^2 \rangle})_{\mathcal{H}} = \left\{ -2d \frac{\partial}{\partial q^2} e^{i\vec{q} [r(L) - r(0)]^2} \right\}_{\mathcal{H}}$$
(3.2)

with:

$$(...)_{\mathcal{H}} = \frac{\int \mathcal{D}r(...) e^{-\mathcal{H}_{\text{eff}}}}{\overline{\langle Z(L) \rangle}}.$$
(3.3)

For the one-loop approximation we find:

$$(\overline{\langle R_{\rm e}^2 \rangle})_{\mathcal{H}} = Ld \left[1 + \frac{4 \, v_0 (2\pi)^{-\frac{d-2}{2}} L^{3-\frac{d}{2}}}{(6-d) \, (8-d)} + \frac{4 \, w_0 (2\pi)^{-\frac{a}{2}} L^{2-\frac{a}{2}}}{(4-a) \, (6-a)} \right]. \tag{3.4}$$

We may thus define a new (renormalized) scale L_R and introduce a swelling factor χ_2 via: $(\langle R_e^2 \rangle)_{\mathcal{H}} = dL_R$, $\chi_2(\nu_0, w_0) = L_R/L$. Remembering that $(\overline{\langle R_e^2 \rangle})_{\mathscr{H}} \sim L^{2\nu}$ and thus $\chi_2 \sim L^{2\nu-1}$ one has:

$$(2\nu - 1) = L \frac{\partial \chi_2(\nu_0, w_0)}{\partial L}.$$
(3.5)

The final renormalization step can be performed by analyzing the virial expansion for the osmotic pressure of a solution of polymers [2]. To this end, we need the contributions to the partition functions $\overline{\langle Z(L,L)\rangle} = \overline{\langle Z_{\nu_0}(L,L)\rangle} + \overline{\langle Z_{w_0}(L,L)\rangle}$ of the system of two interacting polymer chains of the same length L. The dimensionless renormalized coupling constants v, w are thus defined by:

$$v = -\frac{\overline{\langle Z_{v_0}(L,L)\rangle}}{\overline{\langle Z^2(L)\rangle}} L_{\rm R}^{-\frac{d-2}{2}}, \qquad w = -\frac{\overline{\langle Z_{w_0}(L,L)\rangle}}{\overline{\langle Z^2(L)\rangle}} L_{\rm R}^{-\frac{a}{2}}. \tag{3.6}$$

The RG flows of renormalized coupling constants are governed by the functions: $\beta_u = L\partial \ln v/\partial L$, $\beta_w =$ $L\partial \ln w/\partial L$. The corresponding expressions read:

$$\beta_{v} = (6-d)v - \frac{2v^{2}(d-2)}{(8-d)} - \frac{2vw(d-2)}{6-a} = \epsilon v - 4v^{2} - 4vw,$$

$$\beta_{w} = (6-a)w - \frac{2w^{2}a}{(6-a)} - \frac{2vwa}{8-d} = \delta w - 4w^{2} - 4vw.$$
(3.7)

Above, we have performed a double $\epsilon = 6 - d$, $\delta = 4 - a$ expansion, keeping terms up to linear order in these parameters. The fixed points u^* , v^* of the renormalization group transformations are defined as common zeros of the RG functions (3.7). We find three distinct fixed points determining the scaling behavior of a system at different a and d:

Gaussian:
$$v^* = 0$$
, $w^* = 0$, stable for $\varepsilon, \delta < 0$, (3.8)

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Coulomb: $v^* = \frac{\epsilon}{4}$, $w^* = 0$, stable for $\delta < \epsilon$, (3.9)
Disorder: $v^* = 0$, $w^* = \delta/4$, stable for $\delta > \epsilon$. (3.10)

Disorder:
$$v^* = 0$$
, $w^* = \delta/4$, stable for $\delta > \epsilon$. (3.10)

Let us analyze the above results more in detail. The Gaussian fixed point corresponds to the situation, where any monomer-monomer interaction is irrelevant. This happens when we are above the upper critical dimensions of all interactions (d > 6, a > 4). In the case when a > d - 2, the presence of correlated defects plays no role, and the Coulomb fixed point is stable (only the electrostatic interaction is relevant in this case). The fixed point obtained has a positive value, and thus we restore the behavior for $Q > Q_0$ (polyelectrolyte limit). Finally, in the case where a < d - 2, the strongly correlated disorder causes the main effect on the polymer behavior, and the Coulomb interaction is now irrelevant (the Disorder fixed point is stable). The absence of a stable fixed point where both long range interactions are present can be explained using dimensional analysis [see explanation after (2.4)]: at a > d - 2, coupling w_0 becomes dimensionless for d < 6, and thus it is irrelevant in the renormalization group sense, whereas at a < d - 2it is dimensionless for d > 6 and thus the interaction v_0 is irrelevant. The critical exponents v, governing the size measure of a charged polymer in all three situations, described above, are found by evaluating (3.5), rewriting the corresponding expressions in terms of renormalized couplings and finally substituting the values of fixed points listed above. We find:

$$v_{\text{Gauss}} = \frac{1}{2}, \tag{3.11}$$

$$v_{\text{Gauss}} = \frac{1}{2},$$

$$v_{\text{Coulomb}} = \frac{1}{2} + \frac{\epsilon}{8},$$
(3.11)

$$v_{\text{Disorder}} = \frac{1}{2} + \frac{\delta}{8}.$$
 (3.13)

Note that $v_{
m Coulomb}$ coincides with the critical exponent governing the stretching of a polyelectrolyte chain estimated previously within the RG scheme in references [40, 41]. Thus, we conclude that PAs with any $Q > Q_c$ belong to the universality class of polyelectrolyts. For the physically interesting case d = 3 ($\varepsilon = 3$) we may estimate $v_{\text{Coulomb}} \simeq 0.88$. According to (3.13), any neutral chain in solution in the presence of long-range-correlated defects, governed by correlation function with a = d - 2, obeys exactly the same scaling behaviour as a charged polymer with unscreened Coulomb interaction in a pure solvent.

4. Conclusions

We studied the scaling properties of charged polymers (polyampholytes) in a solvent in the presence of structural obstacles spatially correlated on a mesoscopic scale according to a power law $\sim x^{-a}$ [32]. Such correlations can describe extended pore-like defects of fractal structure. Within the randomly charged polymer model, a polyampholyte is considered as a linear chain composed of a random sequence of N monomers, each carrying a charge $\pm q_0$ [6, 7, 12]. The model predicts a contraction of the charged polymer size when the total charge $Q < Q_c$ and an expansion for $Q > Q_c$ (with $Q_c \sim N^{1/2}$). The presence of long-range correlated disorder leads to additional steric interaction between monomers in addition to the long-range Coulomb potential.

Passing to the continuous chain limit with two types of interactions (electrostatic and steric), we developed a direct renormalization group approach by generalizing the scheme of des Cloizeaux [2] and analyzed the peculiarities of conformational transitions, which can be observed in charged polymers in a disordered environment. In the case where a > d - 2, the presence of correlated defects plays no role, and the Coulomb fixed point is stable (only electrostatic interaction is relevant in this case). The fixed point obtained has a positive value, and thus we restore the behavior at $Q > Q_{\mathbb{C}}$ (polyelectrolyte limit [40, 41]). Thus, PAs with any $Q > Q_c$ belong to the universality class of polyelectrolytes. At a < d - 2, the strongly correlated disorder causes the main effect on the polymer behavior, while Coulomb interaction is irrelevant. In particular, one may conclude that any neutral chain in solution in the presence of longrange-correlated defects, governed by correlation function with a = d - 2, follows exactly the same scaling behaviour, as a charged polymer with unscreened Coulomb interaction in a pure solvent.

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Випадково заряджені полімери в пористому середовищі

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Досліджуються конформаційні властивості заряджених полімерів в розчині у присутності структурних неоднорідностей, скорельованих згідно степеневого закону $\sim x^{-a}$. Використовується модель, в якій полімерний ланцюжок представлено як випадкову послідовність зарядів $q_i = \pm q_0$. Така модель описує властивості поліамфолітів – гетерополімерів, що містять як позитивно, так і негативно заряджені групи мономерів. Застосовується підхід прямого полімерного перенормування і аналізується скейлінгова поведінка заряджених полімерів до першого порядку подвійного $\epsilon=6-d$, $\delta=4-a$ -розкладу.

Ключові слова: полімери, заморожений безлад, скейлінг, ренормалізаційна група

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