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Renormalization Group Description of Polymer Excluded Volume

Karl F. Freed[1] **and A. L. Kholodenko**[2]

A description is provided of the chain conformation space renormalization group approach to the treatment of polymer excluded volume. This method is transformed into one of identical form to the t'Hooft–Veltman renormalization method of field theory, thereby enabling comparison with other methods. A summary is provided of recent new results obtained by this technique for the full second-order dependence of the mean square end-to-end distance $\langle \mathbf{R}^2 \rangle$ on chain length and excluded volume as well as a calculation of $\langle \mathbf{R}^2 \rangle$ for a polyelectrolyte chain with added salt.

KEY WORDS: Renormalization group; dimensional regularization; polymer excluded volume; crossover; polyelectrolytes.

1. INTRODUCTION

Renormalization group methods have been shown to provide a valuable approach to the description of excluded volume effects in polymer systems. Early renormalization group works are based on the analogy between the properties of polymers with excluded volume and the critical properties of magnets, the so-called polymer–magnet analogy.(1,2) The complexity of polymer systems and deficiencies in the polymer–magnet analogy have led to the development of direct renormalization group approaches to describe polymer excluded volume.(3–5) Two of these(3,4) direct renormalization approaches used methods taken from field theory, while the chain conformation space renormalization treatment(5) differs considerably by being

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[1] The James Franck Institute and Department of Chemistry, The University of Chicago, Chicago, Illinois 60637.

[2] The James Franck Institute, The University of Chicago, Chicago, Illinois 60637.

based solely within chain space. This chain conformation space method performs the renormalization for the chains of *finite* lengths as opposed to the other field theoretical approaches(3,4) which consider only the asymptotic limit of infinitely long polymer chains.

In order to indicate the relationship between these different direct renormalization methods as well as to develop techniques to attack new fundamental polymer problems, we have converted(6) the chain configuration space renormalization group approach into equivalent formulation in Fourier–Laplace transform space. The transformed theory is then shown(6) to be precisely of the form of a one-component ϕ^4-type field theory with the only departure that the numerical combinatorial factors for individual terms in the perturbation expansion are generated from the excluded volume perturbation series. In this formulation our method is shown to provide the following advancements over the other two direct renormalization group approaches: (a) The procedure follows directly from the excluded volume perturbation expansion; (b) the renormalization is performed for *finite* chain lengths, so the complete crossover dependence(7) on excluded volume and chain length can be described by the theory; (c) insertions(3,4,8) are not required, thereby reducing the computational labor; (d) all diagrams can be readily and efficiently evaluated using t'Hooft–Veltman method.(9,10)

The power of the new renormalization method is illustrated by summarizing our recent work(6) on two problems. The mean square end-to-end distance $\langle \mathbf{R}^2 \rangle$ for a single polymer chain has been evaluated to second order in excluded volume. The exponents are already well known in the limit of long chains from the field theory ($n \to 0$) methods(1,2); however, our second-order calculations generate in addition to these exponents the expansion for the previously unavailable numerical prefactors as well as the full second-order crossover dependence on the excluded volume interaction strength and chain length. In addition, we consider the description of excluded volume effects for a polyelectrolyte chain (i.e., one with charges) for solutions where the salt concentration is sufficiently high so that in zeroth order the polyelectrolyte chain backbone has a reasonable degree of flexibility. This limit is the one of most direct relevance to experiments and has not been considered previously by renormalization group methods. Our results show that there is no stable fixed point for polyelectrolyte excluded volume, so that this important system *cannot* be described by simple scaling theories of excluded volume(2) or other renormalization group methods which are solely geared to the calculation of exponents. The final results are shown to depend on three scaling variables, two of which are related to the two types of interactions, while the third involves the ratio of the chain size to the Debye screening length and in these polyelectrolyte chains is always much greater than unity.

In the next section we briefly review the salient physical concepts from the chain conformation space renormalization group procedure. The Fourier–Laplace transform of relevant quantities then is shown in Section 3 to provide an equivalent representation which can more readily be compared with the standard field theoretic renormalization group approaches. No prior knowledge of the field theoretic methods is, however, assumed or required.

Lastly, in Section 4 we summarize our recent results(6) on the second-order crossover behavior of $\langle \mathbf{R}^2 \rangle$ for a single polymer chain with excluded volume as well as $\langle \mathbf{R}^2 \rangle$ for a polyelectrolyte chain.

2. CHAIN CONFORMATION SPACE RENORMALIZATION GROUP METHOD(5)

While the detailed chemical structure of a polymer chain may at first sight appear rather complicated, in general, we are interested here in only long-wavelength properties on some experimental length scale L which is much larger than microscopic length scale, say, as given by the size of a monomer a. On the scale L it is then permissible to consider simplified models of the polymer chain. Here we employ the well-known model of a continuous flexible Gaussian chain where the backbone is taken as a continuous random walk. There is, however, excluded volume which makes contacts between polymer segments energetically unfavorable.

$\langle \mathbf{R}^2 \rangle$ for a random walk chain depends on the chain length N as $\langle \mathbf{R}^2 \rangle \propto N$, whereas with excluded volume the chain swells to provide the dependence $\langle \mathbf{R}^2 \rangle \propto N^{2\nu}$, $2\nu > 1$. Viewing the chain size as a correlation length we have the analogy with critical phenomena where the chain length N corresponds to $|T - T_c|^{-1}$, the inverse distance from the critical point. De Gennes showed that the description of polymer excluded volume can be written(1) as the $n \rightarrow 0$ limit of the $O(n)$ n-vector model of field theory. Scaling theories of polymer excluded volume(2) have also been developed, in analogy with their use in critical phenomena, to provide insights into various asymptotic domains as well as into the value of exponents in these domains. However, the simple scaling theories provide no information on the important prefactors; *they cannot determine when the asymptotic limit is reached*; and they cannot deal with important crossover behavior that arises when more than one length scale becomes relevant. Renormalization group theory, in principle, provides a method for the calculation of all of the above quantities which are important for polymer systems since it is generally the rule that these polymer systems do not lie in the asymptotic scaling type limit. The majority of other renormalization group approaches (3,4) appear to be patterned after those in critical phenomena(8) and directed solely towards the calculation of exponents, thereby sharing some of the deficiences of scaling theories.

The simple model of a continuous chain with excluded volume is summarized by the dimensionless "Hamiltonian"

$$H_a = \frac{1}{2}\int_0^{N_0} d\tau\, |d\mathbf{C}(\tau)/d\tau|^2 + \frac{1}{2} v_0 \iint_{|\tau-\tau'|\geqslant a}^{N_0} d\tau\, d\tau' \delta[\mathbf{C}(\tau) - \mathbf{C}(\tau')] \tag{1}$$

Here $\mathbf{C}(\tau)$ is the chain configuration as a function of contour length variable τ. The first term on the right-hand side describes chain connectivity, and the second term produces excluded volume effects. The Hamiltonian has three parameters: N_0 the chain length, v_0 the bare microscopic excluded volume, and a the cutoff to remove self-excluded volume interactions.

A perturbation expansion in powers of v_0 is readily shown(11) to be in fact an expansion in powers of $v_0 N_0^{\epsilon/2}$ where $\epsilon = 4 - d$ with d the dimensionality of space. Such an expansion is obviously of little use for $N_0 \to \infty$ when $d < 4$. In addition, the coefficients in this expansion are sensitive to the value of a/N_0, making computed properties dependent on what would appear to be irrelevant microscopic details. The first difficulty can be remedied by considering d to be a continuous variable and expanding ϵ for small ϵ via

$$v_0 N_0^{\epsilon/2} = v_0\left[1 + \frac{\epsilon}{2}\ln N_0 + \frac{1}{2}\left(\frac{\epsilon}{2}\right)^2 (\ln N_0)^2 + \cdots\right] \tag{2}$$

The removal of the sensitive dependence on microscopic details and the determination of methods to extrapolate to ϵ approaching unity are provided by a renormalization group method.

The continuous chain model is physically absurd for small length scales, e.g., lengths less than or on the order of a. However, the model is correct on a scale L for $L/a \gg 1$. Hence, the observable polymer length N may differ from that N_0 in the model (1). But if we, say, double the length of the continuous chain N_0 we must double the length of the real polymer chain N. This can be expressed as

$$N = Z_2 N_0 \tag{3}$$

where Z_2 is a proportionality factor which depends on excluded volume and the dimensionless combination a/L. v_0 describes a binary encounter between segments, whereas there is a macroscopic excluded volume parameter v which describes the cooperative effects of excluded volume on a length scale L. Introducing the dimensionless variables

$$u_0 = v_0 L^{\epsilon/2}, \qquad u = vL^{\epsilon/2} \tag{4}$$

this relationship between macroscopic and microscopic excluded volume is summarized by

$$u = u(u_0, a/L) \tag{5}$$

The partition function for fixed end vector **R**, as calculated from (1), is designated as $G_B(\mathbf{R}, N_0, v_0; a)$, where the subscript B indicates bare unrenormalized quantities. G_B contains microscopic irrelevant details, such as the cutoff a, which should not be present on the macroscopic length scale L for the observable $G(\mathbf{R}, N, v; L)$. However, when normalized both G_B and G should produce the same end vector probability distribution, and therefore these two must be related by a proportionality factor

$$G(\mathbf{R}, N, u; L) = Z(u, a/L) G_B(\mathbf{R}, N_0, v_0; a) \tag{6}$$

where subsequently we employ the limit $a/L \to 0$.

In summary, we have a set of microscopic and macroscopic parameters and quantities which differ but which are interrelated as described(5) above in Eqs. (3)–(5).

The existence of these macroscopic–microscopic relations leads to profound consequences. The microscopic model Hamiltonian (1) and all quantities calculated from it are, of course, independent of the macroscopic observational length scale L. Hence, we have the trivial relationship

$$\left[L \frac{\partial}{\partial L} G_B(\mathbf{R}, N_0, v_0; a) \right]_{N_0, v_0, a} = 0 \tag{7}$$

Inserting the macroscopic-microscopic relations

$$G_B(\mathbf{R}, N_0, u_0; a) = Z^{-1}(u, a/L) G\left[\mathbf{R}, Z_2(u, a/L) N_0, u_0(u, a/L); L\right] \tag{8}$$

into (7) we readily obtain the highly nontrivial renormalization group (RG) equation as

$$\left[L \frac{\partial}{\partial L} + \beta(u) \frac{\partial}{\partial u} + L \frac{\partial \ln Z}{\partial L} + L \frac{\partial \ln Z_2}{\partial L} N \frac{\partial}{\partial N} \right]_{N_0, v_0, a} G(\mathbf{R}, N, u; L) = 0 \tag{9}$$

where the Gell-Mann–Low function is given as

$$\beta(u) = L \frac{\partial u}{\partial L} \bigg|_{N_0, v_0, a} \tag{10}$$

This renormalization group equation summarizes the general analytic structure of the dependence of macroscopic observables like G on the macroscopic parameters.

When the macroscopic u is independent of the observational length scale L, (for $L/a \gg 1$) then the system exhibits the asymptotic universal scaling behavior. This is the sole limit which is accessible to simple scaling theories(2) and is the limit which has been the object of a variety of renormalization group approaches.(3,4) This limit is defined by the condition

$$\beta(u)|_{u=u^*} = 0 \tag{11}$$

where u^* is called the fixed point coupling constant. The use of Eq. (11) simplifies the RG equation (9) enormously. The solution of the RG equation and simple dimensional analysis readily enables the demonstration that G behaves in the scaling limit as(5)

$$\overset{\circ}{G}(\mathbf{R}, N, u^*; L \equiv 1) = N^{\delta - \nu d - 1} f(\mathbf{R}/N^{\nu}) \tag{12}$$

where we use a system of units in which $L = 1$, and the exponents ν and γ are numbers calculated from

$$\frac{2\nu - 1}{2\nu} = L \frac{\partial \ln Z_2}{\partial L}\bigg|_{u=u^*}, \qquad \frac{\gamma - 1}{2\nu} = L \frac{\partial \ln Z}{\partial L}\bigg|_{u=u^*} \tag{13}$$

thus providing the asymptotic scaling law formerly only *postulated* in scaling theories. The RG equation (9) can also be solved for $u \neq u^*$ and thereby provide the important crossover dependence on u and N.(7)

The calculation of u^*, γ, ν and the scaling function $f(\mathbf{R}N^{-\nu})$ requires the determination of u, Z, and Z_2. If, for instance, we consider the perturbation expansion for G_B in powers of ϵ and u_0, it is found that there are many terms in the series which become divergent in the limit that $a/N_0 \to 0$, thereby exhibiting a strong dependence on relevant microscopic detail. Hence, the renormalization constants Z, Z_2, $u_0(u)$ are defined such that the perturbation expansion of the macroscopic G should be independent of microscopic details, i.e., independent of a. The actual computations are carried out with the method of dimensional regularization which leads to terms in the perturbation expansion involving inverse powers of ϵ so that the renormalization constants are defined to absorb all the singular ϵ^{-n} terms.

This chain conformation space renormalization group method has been applied to the calculation of a wide variety of polymer properties including $G(\mathbf{R}, N)$,(5) the static coherent scattering function,(12) the internal vector distribution for linear(13) and ring polymers,(14) the second viral coefficient for polymers,(7) the dynamics of single polymer chain,(15) as well as the concentration dependence of the osmotic pressure(13) and $\langle \mathbf{R}^2 \rangle$ for a labeled polymer chain. In addition, the full crossover dependence on N and excluded volume interaction strength$^{(7)}$ has been given for a number of the above polymer properties. Since much of this material, as well as the detailed theoretical methods, are already published, they are not repeated here. Instead we now turn to the conversion of this chain conformation space renormalization group method into an equivalent formulation$^{(6)}$ which allows direct use of the t'Hooft–Veltman style dimensional regularization and renormalization.

3. T'HOOFT–VELTMAN STYLE VERSION OF CHAIN CONFORMATIONAL SPACE RENORMALIZATION GROUP METHOD$^{(6)}$

Let **k** be the Fourier variable conjugate to the position variable, say the chain end-to-end vector **R**. Likewise, let s' be the Laplace variable conjugate to the chain length N_0. Then, introducing the change of variables $s = s'Z_2^{-1}$ the renormalization relationship (see Eq. (8)) for the Fourier–Laplace transform of G takes the form of$^{(6)}$

$$G(\mathbf{k}, s, u; L) = \left[\tilde{Z}\tfrac{1}{2}k^2 + Zs + \tilde{Z}\Sigma(\mathbf{k}, Z_2 s)\right]^{-1} \tag{14}$$

where Σ is the "self energy" or "mass" operator which contains all effects of excluded volume. Σ may be obtained from G_B as

$$G_B(\mathbf{k}, s, v_0; a) = \left[\tfrac{1}{2}k^2 + s + \Sigma(\mathbf{k}, s)\right]^{-1} \tag{15}$$

The derivation of (14) provides the important relationship

$$\tilde{Z} \equiv ZZ_2^{-1}$$

between the renormalization constants which makes it totally unnecessary to employ insertions$^{(3,4,8)}$ to calculate the polymer exponents. The quantities Z and $\tilde{Z}$ are chosen to remove the singular parts of $Z\Sigma(k, Z_2 s)$ which are proportional to k^2 and s, respectively. These quantities have an identical form to standard t'Hooft–Veltman-type renormalization results for *one-component* massive ϕ^4 field theory.$^{(10)}$ The traditional polymer perturbation expansion$^{(11)}$ for G_B, however, provides *different* numerical weight factors for the individual terms in the perturbation expansion, but this introduces no real difference in the application of the method. Whereas two of the previous direct renormalization group approaches$^{(3,4)}$ are limited to the treatment of infinitely long chains (or $s \to 0$), the above procedure enables us to consider the case of general N (or s) and thereby provide molecular-weight-dependent properties such as those which are necessary in treating crossover properties and are essential in describing polyelectrolyte excluded volume. Our method has the additional decided advantage of enabling the values of many of the diagrams to be directly taken from available text-books on field theory.

This new renormalization group method has been applied to the calculation $\langle \mathbf{R}^2 \rangle$ to second order.(6) The exponent 2ν is already well known, and recalculations of it are of no interest. However, in the asymptotic scaling limit, our method permits the evaluation of the overall coefficient, and this can be found from the scaling limit form(6)

$$\langle \mathbf{R}^2 \rangle = N\left(\frac{2\pi N}{L} \right)^{\epsilon/8 + 15/4(\epsilon/8)^2 + \cdots} \left[1 - \frac{\epsilon}{8} + 0.300\left(\frac{\epsilon}{8} \right)^2 + \cdots \right] \tag{16}$$

Although the exponent $2\nu - 1$ is known to be an asymptotic expansion(16) in ϵ, the prefactor appears to be much more reasonably convergent.

The power of the chain conformation space renormalization group method and its Fourier–Laplace representation is exhibited by the crossover behavior of $\langle \mathbf{R}^2 \rangle$. Let ζ be a scaling parameter which measures the strength of the excluded volume interaction. It is proportional to $N^{\epsilon/2}$ as well as to a nonanalytic function of the excluded volume interaction u. ζ ranges from 0 for the Gaussian random walk to ∞ for the self-avoiding walk limit. To second order in u^* $\langle \mathbf{R}^2 \rangle$ is found(6) to be

$$\langle \mathbf{R}^2 \rangle = N\left(\frac{2\pi N}{L} \right)^{[f(\zeta,u^*)](2\pi N/L)^{-2u^*(1+\zeta)^{-1}} - (3/2)[\zeta(u^*)^2/1+\zeta]}$$

$$\times \left\{ 1 - f(\zeta,u^*) - 0.2294\,\frac{\zeta(u^*)^2}{1+\zeta} - 0.0706\,\frac{\zeta^2(u^*)^2}{(1+\zeta)^2} \right\} \tag{17}$$

where the fixed point coupling constant u^* is

$$u^* = \frac{\epsilon}{8} + \frac{21}{4}\left(\frac{\epsilon}{8} \right)^2 + O(\epsilon^3) \tag{18}$$

and $f(\zeta,u^*) = u^*\zeta[\zeta + e^{-f}(1 + \zeta(1+\zeta)^{-17u^*/4})^{-x}]^{-1}$, $f = -\frac{5}{4}(u^*)^2(\zeta - 1)/(\zeta + 1)$, $x = -\frac{17}{4}u^*$. ζ can be written(6) to second order in terms of u as a function of the parameter $w = u(u^* - u)^{-1}$ as

$$\zeta = \left(\frac{2\pi N}{L} \right)^{\epsilon/2} w(1 + w)^{[+(17/4)u^* - 8(u^*)^2 - (21/4)(17/4)(u^*)^2]}$$

$$\times \exp\left[-\frac{5}{4}(u^*)^2 \frac{(w-1)}{(w+1)} \right] \tag{19}$$

Professor Domb's(17) work on lattice enumeration has not yet established the behavior of the proper scaling function ζ as a function of the interaction strength u, but perhaps the RG result (17), (19) will enable this nonanalytic function to be further investigated by lattice enumeration techniques. Notice that the effective exponent of N in (17) is not only a function of the crossover scaling parameter ζ, but also a function of the chain length N, exhibiting the nonuniversal behavior of $\langle \mathbf{R}^2 \rangle$. It will be interesting to compare this result with those of lattice enumerations since the asymptotic exponent 2ν from (16) to order ϵ^2 is rather close to presumably exact numerical results despite the asymptotic nature of the ϵ-expansion. Hence, computed asymptotic properties to second order should be very accurate.

4. RENORMALIZATION GROUP DESCRIPTION OF EXCLUDED VOLUME IN POLYELECTROLYTES

Polyelectrolytes have a variety of different length scales.(18) First there is a Debye–Huckel screening length κ^{-1}; then b is the spacing between charges; and $\lambda_B = e^2/\epsilon_0 kT$ defines the Bjerrum length. The experimentally relevant limit involves the dimensionless parameters

$$\xi = \lambda_B / b \ll 1 \tag{20}$$

$$\zeta = (b\kappa)^{-1} \lesssim 1 \tag{21}$$

where (20) is the condition that no charge condensation occurs,(19) while (21) is the condition that the chain retains sufficient flexibility so that a model in terms of a flexible backbone chain is valid. Experimental data suggest(20) an empirical form for $\langle \mathbf{R}^2 \rangle$ of a polyelectrolyte chain in this regime as

$$\langle \mathbf{R}^2 \rangle = N^{f(\xi, \kappa^2)} g(\xi, \kappa^2) \tag{22}$$

where the functions f and g are, in general, dependent on the three length scales of the problem, thereby indicating the lack of simple scaling or universal behavior. A theoretical description of the rich behavior implied by (22) is totally lacking. A previous renormalization group treatment(21) considered a jellium model with no explicit dependence on salt concentration. If charge condensation is ignored in this hypothetical model, then the chain would be rodlike, and the model invoked of a flexible chain with unscreened Coulomb interactions is of only academic interest.

We have applied the t'Hooft–Veltman type renormalization description(6) to a flexible polyelectrolyte chain with screened Debye type interactions *in addition* to nonelectrostatic excluded volume interactions. A renormalization calculation indicates the absence of a stable fixed point, hence, the absence of a simple scaling limit. The first order expansion in both electrostatic and excluded volume coupling constants provides $\langle \mathbf{R}^2 \rangle$ as a function of three scaling parameters as(6)

$$\langle \mathbf{R}^2 \rangle = N\left(\frac{2\pi N}{L} \right)^{(\epsilon/8)(\zeta_1/\zeta_1 - 1) - (3\epsilon/20)(\zeta_2/1 - \zeta_2)}$$

$$\times \left[1 - \frac{\epsilon}{8} \frac{\zeta_1}{\zeta_1 - 1} + \frac{3\epsilon}{20} \frac{\zeta_2}{1 - \zeta_2} \left(\ln \zeta_3 + 4\zeta_3^{-1} \right.\right.$$

$$\left.\left. + \gamma - 2 + \zeta_3^{-1}\left(3\gamma - \frac{5}{2} \right) \right) + O(\epsilon^2) \right] \tag{23}$$

with γ Euler's constant. The parameters ζ_1 and ζ_2 depend on κ and provide measures of the two types (excluded volume and electrostatic) of interaction strengths, representing a generalization of the simple ζ parameter(19) for uncharged polymers. ζ_3 can essentially be written as $\langle \mathbf{R}^2 \rangle \kappa^2$ and is therefore a large parameter in the theory and in the domain of interest to experiments. Equation (23) produces the previous results for uncharged polymers when $\zeta_2 = 0$. Note that our calculated expression (23) has the general form of the empirical representation (22) and therefore provides the first systematic theoretical approach which is capable of describing the rich experimental behavior of (22). It should also be noted that the same three scaling variables ζ_1, ζ_2, and ζ_3 appear in the calculation of any other equilibrium long wavelength properties of polyelectrolyte chains. The appropriate method for comparison between theory and experiment involves the calculation of a variety of measurable properties. The theory then proceeds by eliminating ζ_1, ζ_2, and ζ_3 to provide interrelationships between measurable properties as a direct generalization of the analysis(7) of uncharged polymers with a single parameter ζ.

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