Registry No. Polystyrene (homopolymer), 9003-53-6.

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Relationship of the Unweighted Rosenbluth and Rosenbluth Walk to a Polymer Chain at the Θ Point

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ABSTRACT: It is shown that the unweighted Rosenbluth and Rosenbluth (R–R) chains (sometimes called the "true" self-avoiding walk) can be viewed as polymer chains at the Θ point where only second-order cluster-like terms have been included in the partition function. A modified weighting function for the R–R model is proposed that includes only such second-order cluster terms. Such a polymer chain is shown to show normal polymer chain behavior, i.e., chain expansion, a Θ point, and chain collapse. It is suggested that by comparing the results of studies on these chains with those obtained by a normal R–R weighting procedure one should be able to accurately assess the contributions of third-order and higher order cluster terms to polymer chain properties.

I. Introduction

The self-repelling chain or the "self-avoiding" walk (SAW) has been used to model the equilibrium statistics of linear polymers in dilute solutions of good solvents.[1,2] In such a model one enumerates all configurations of an N-step walk where the walks are not allowed to intersect themselves. For the SAW one obtains measures of the size of the chain that differ from those obtained for a random walk model of the chain. For example, the mean squared radius of gyration, $\langle R_g^2 \rangle$, of a SAW is found to be

$$\langle R_g^2 \rangle = N^{\gamma} \tag{1.1}$$

where γ is about 1.2.

Recently Amit et al.[3] suggested that a random walk in which the traveler steps randomly but tries to avoid places he has already visited be called the "true" self-avoiding random walk (TSAW). They point out that this "true" self-avoiding random walker is very different from the normal self-avoiding random walk used in polymer physics to describe a chain in a good solvent, the SAW. Using renormalization group theory they find that the mean squared end-to-end distance, $\langle R^2 \rangle$, obeys an equation of the form

$$\langle R^2 \rangle \sim N \tag{1.2}$$

They also present Monte Carlo data confirming eq 1.2.

Following the work of Amit et al., a number of similar models have appeared in the literature. Some of them have been related to kinetics of chain growth; others have been related to the growth of polymer gels.[4–6] One such model, the kinetic growth walk (KGW) proposed by Majid et al.,[5] is similar to that of Amit et al. in that it includes self-avoidance in the sense of Amit et al. as well as trapping. For this model Majid et al. do not find that eq 1.2 is exact; rather, they find that the mean squared radius of gyration $\langle R_g^2 \rangle$ of the chain obeys the equation

$$\langle R_g^2 \rangle \sim N(\ln N)^{0.2} \tag{1.3}$$

Here we point out the difference in these walks. In the SAW a walker randomly chooses his next step from among all the nearest-neighbor sites whether they have been visited previously or not; if the walker chooses a visited site, the walk stops. In a TSAW the walker visits all nearest-neighbor sites but chooses among those sites with different probability such that the walk favors unvisited sites; in the TSAW the walker is thus never stopped. In the KGW the walker chooses among only unvisited nearest-neighbor sites with equal probability; the walker is only stopped if all nearest-neighbor sites have been previously visited (called "trapping").

Amit et al. suggested (see their note added in proof) that their "true" self-avoiding random walks are identical with the unweighted form of the chains suggested by Rosenbluth and Rosenbluth[7] (R–R) for use in Monte Carlo simulations of polymer chains.[8] In fact, as a result of trapping, the R–R model is more closely akin to the KGW chain.

Many authors have studied the R–R chains in their context as a generator of the normal excluded volume self-avoiding polymer chains.[7–10] In addition, nearest-

neighbor attractive energies have been included in the chain weights in the Monte Carlo chain simulations to estimate the effect of varying solvent–polymer interactions.[8,9] Thus in Monte Carlo simulations using R–R chains with proper weighting one is able to go from the athermal chain region (complete excluded volume) to the Θ region (the excluded volume is approximately compensated for by attractive nearest-neighbor interactions) to the chain collapse region.

In previous Monte Carlo studies of such polymer chains[9] we found that the chains generated by the R–R method with exclusion and energy weightings gave the most accurate data for size measures of the chain near the Θ point of the polymer chain. In the region of the Θ point we were able to generate the longest chains with the least Monte Carlo error (see Figure 2 in ref 9). Thus, the R–R method was used near the Θ point to compute properties where high accuracy and long chains were required.[10,11]

This special property of the R–R chains at the Θ point is due to the near compensation of the weight applied to the R–R chain to make it a correct representation of the self-avoiding walk (an athermal polymer chain) by the weight from the nearest-neighbor interactions at the Θ point. In that earlier paper[9] it was thought that this compensation was largely fortuitous. In this paper we suggest that such a self-compensation is not fortuitous. Rather we suggest that the unweighted R–R chain is a chain with excluded volume and with self-compensating attractive interaction energies in which the third and higher cluster sums have to be set to zero. In Flory's[12,13] sense, this is a chain at the Θ point. From a slightly different point of view we suggest that the unweighted R–R chain is an almost correct representation of a mean field chain with excluded volume and self-compensating energies at the Θ point. In this paper we only consider single-chain properties.

In the next section we describe in some detail the R–R chain generation procedure with interaction energies. We show how eliminating third- and fourth-order cluster terms in self-compensated chains leads to unweighted R–R chains. We also develop a mean field weighting in terms of infinite coordination number which leads to unweighted R–R chains and suggest that this chain is in some sense a representation of a Flory-like chain with excluded volume at the Θ point. In both of the above discussions a modified weighting function for the R–R chain is presented in which only second-order cluster terms are retained. In section 3 properties of chains weighted with this modified one-parameter weighting function are considered. It is shown that these chains show self-exclusion behavior, Θ behavior, and chain collapse.

II. Identification of New Weighting for R–R Chains and Its Meaning near the Θ Point

In this section, we focus on a description of the chains generated by the R–R chain simulation procedure where we include both the effects of weighting which yield normal self-avoiding chains and attractive energies of interaction between nearest neighbors which simulate the effect of temperature and solvent. We also consider how one may modify the weighting of such chains to obtain a meaningful interpretation of the unweighted R–R chains.

Here we consider only chains on a cubic lattice. Monte Carlo chains have been generated on other lattices using this technique but these will not be discussed here[8,9] since the extension to chains on other lattices is obvious.

The partition function, z_0, for a random walk of N steps with no self-reversal (or the so called five-choice random walk), is

$$z_0 = 5^N \tag{2.1}$$

If we generate such chains using a Monte Carlo procedure we assume that each chain would be equally weighted and be given a weight of one. The partition function for N_t such chains is

$$z_0 = \frac{5^N(\text{total weight of the chains})}{N_t} = 5^N \tag{2.2}$$

since the total weight of all generated chains is N_t. Chains generated with the R–R procedure, which are weighted to give the same counting as the random walks with excluded volume, are given a weight w_k for the kth chain in the simulation. According to Rosenbluth and Rosenbluth

$$w_k = \prod_{i=1}^{5} p_i^{n_{ik}} \tag{2.3}$$

where

$$p_i = (5 - i)/5 \tag{2.4}$$

and n_{ik} is the number of sites in the kth chain with i nearest-neighbor contacts found as the chain is created in the Monte Carlo simulation. When we allow nearest-neighbor interactions on the chain (to simulate solvent–polymer interaction differences) of ϵ we have[9]

$$w_k(\phi) = \prod_{i=1}^{5} (p_i^{n_{ik}} \exp(i n_{ik}\phi)) \tag{2.5}$$

where

$$\phi = \epsilon/k_B T \tag{2.6}$$

This equation has been given before.[9] We note that for the kth polymer with N beads and N_{ck} nearest-neighbor contacts

$$N = \sum_{i=0}^{5} n_{ik} \tag{2.7a}$$

$$N_{ck} = \sum_{i=1}^{5} i n_{ik} \tag{2.7b}$$

We have assumed that the polymer is monodisperse (N is independent of k). The number of contacts in the kth chain depends on the chain's configuration and thus on k. The normalized weight for each chain is

$$z_k = z_0 w_k(\phi) \tag{2.8}$$

and the partition function, Z, for all N_t chains normalized to the partition function of the five-choice walker is

$$Z = \frac{1}{N_t z_0} \sum_{k=1}^{N_t} z_k = \sum_{k=1}^{N_t} \frac{w_k(\phi)}{N_t} \tag{2.9}$$

Thus only $w_k(\phi)$ is needed to compute the partition function of the chain.

We rewrite eq 2.3 as

$$w_k(\phi) = \prod_{i=1}^{5} f_i^{i n_{ik}} \tag{2.10}$$

where

$$f_i = (p_i)^{1/i} e^{\phi} \tag{2.11}$$

If we let

$$\delta_i = f_i - f_1 \tag{2.12}$$

we then have for $w_k(\phi)$

$$w_k(\phi) = f_1^{N_{ck}} \prod_{i=2}^{4} (1 + \delta_i/f_1) \tag{2.13}$$

We have shown earlier (see Appendix C, ref 8) that the f_i are near one for $i = 1$, 2, and 3 near the Θ point of these

chains. Thus, near the Θ point for our Monte Carlo chains we expect that δ_2 and δ_3 are small. We may presume that we may expand around these and obtain a cluster-like expansion if f_1 is large compared to all the δ's. A more appropriate expansion in the exponential is also possible where we would have from eq 2.12

$$w_k(\phi) = f_1^{N_{ck}} \exp(\sum_{i=2}^{5} i n_{ik}) \ln (1 + \delta_i/f_1) = f_1^{N_{ck}} \exp(\sum_{i=2}^{5} i n_{ik}\delta_i/f_1) \quad (2.14)$$

We wish, however, to focus on the weight remaining when we have let δ_i for i greater than or equal to 2 to vanish. In this case we obtain for the weight of the R–R chain

$$w_k(\phi) = f^{N_{ck}} \quad (2.15)$$

where we have dropped the index from f_1 for convenience. It is the R–R chain with such a weight whose properties we shall discuss in the next section of this paper.

If we choose $f = 1$ then the chain described by eq 2.15 is the unweighted R–R chain. The unweighted R–R chain can be viewed as a normal self-avoiding walk with nearest-neighbor interactions compensating the exclusion weights in the pair interactions but where contributions from the higher order cluster-like terms are taken into account as the binary cluster term alone.

There is another approach that may be taken to the interpretation of the R–R chains with weights described by eq 2.15. This interpretation can be seen by considering a redefinition of p_i in eq 2.4. In eq 2.4 the p_i are simply the fraction of all possible sites available to a walker as he makes the next step and are given for the five-choice lattice by the values assigned to them by Rosenbluth and Rosenbluth. If we, however, make the ansatz such that we let each p_i be approximated as

$$p_1 = 4/5$$
$$p_i = (p_1)^i \quad (2.16)$$

then we have for eq 2.10

$$w_k(\phi) = \prod_{i=1}^{5} (p_1 e^{\phi})^{n_{ik}} = f^{N_{ck}} \quad (2.17)$$

where we have again left $f_1 = f$. We thus automatically recover eq 2.15 for the weight of each chain. The difference between the p_i in eq 2.16 and those in eq 2.4 can be assessed numerically. The p_2's are close, the p_3's are about 10% different, and the p_4's have a 50% deviation. For p_5 and above the ansatz is of course completely in error. If only p_1 and p_2 dominate the properties of the real chain, the above approach has little error.

The error is smaller for lattices of higher coordination number. For a lattice of arbitrary coordination number σ, we have for p_i

$$p_1 = (\sigma - 2)/(\sigma - 1)$$
$$p_2 = (\sigma - 1 - i)/(\sigma - 1) \quad (2.18)$$

For σ large we obtain for $(p_1)^i$

$$(p_1)^i = \left(\frac{\sigma - 2}{\sigma - 1}\right)^i \approx 1 - i/(\sigma - 1) = p_i \quad (2.19)$$

In the sense that the mean field model is the infinite coordination number model, then the above ansatz is a mean field model.

Thus, we may say in the sense described above, the R–R chain has a weight given by eq 2.17. This chain is self-compensated for $f = 1$ and we suggest that at $f = 1$ we have the Θ point. Strengthening this point of view is the findings of Amit et al. and Madjid et al. that the mean squared end-to-end distance is proportional to N for the TSAW and KGW models (or nearly proportional to N for the KGW model).

At this point we need to consider whether or not the chains we shall use for our further modeling are to be trapped. In the normal R–R walk in a cubic lattice, when a chain's growing end has five unbonded nearest neighbors the chain is given the weight zero and the chain is said to be trapped. The trapped chains have no weight and are not considered in the averaging process. The new weighting proposed in eq 2.15 has no zero for five nearest neighbors. In fact, there are no zeros in the weight at all. In principle, following eq 2.15 would lead us to use chains without trapping.

Generally, authors have had the view that only contributions from second- and third-order cluster terms determine the large-N properties of the chains.[13] Since trapping is a contribution from the fifth-order cluster term in cubic lattice models, one would think that chains with and without trapping would have the same large-N properties. Recently, however, Kremer and Lyklema[14] suggested that TSAW's with trapping (i.e., unweighted R–R chains with trapping) are in the same universality class as SAWS while TSAW's without trapping are in the same universality class as normal random walks.

In consonance with eq 2.15 and the argument of Kremer and Lyklema, we shall use R–R chains without trapping for the data presented in the next section. Chains so produced will contain a few regions of double occupancy when chains are forced to walk out of trapping positions.

We should note that for chains on a cubic lattice Kremer[15] has suggested that only chains with N greater than 50 000 will show the effect of trapping. We have not been able to check this. The Kremer observation is, however, consistent with our results for N less than 10 000, where we have found no difference within the Monte Carlo error in the power law or the approach to the power law for chains with and without trapping. We only find a modest change in the front factor.

In the next section we will show that size properties of the R–R weighted as in eq 2.15 show the expected properties of a chain, i.e., chain expansion, Θ point, and chain collapse.

III. Properties of Weighted R–R Chains with No Third-Order or Higher Order Cluster Terms

We have suggested in the above discussion that the unweighted R–R chains are approximately chains at the Θ point with both excluded volume and nearest-neighbor energy compensation. In this section we present data for the model of the chain with the weighting proposed in eq 2.15. Thus, we generate R–R chains weighted as

$$w_k(\phi) = f^{N_{ck}} \quad (2.15)$$

where we recall N_{ck} is the number of contacts in the kth chain. Of course

$$f = \tfrac{4}{5}e^{\phi} \quad (3.1)$$

for a cubic lattice; for a lattice of coordination number σ, we have of course

$$f = \left(\frac{\sigma - 2}{\sigma - 1}\right)e^{\phi} \quad (3.2)$$

Justification for the study of such a model is manifold. First it is a model of the true self-avoiding walk with interaction between nearest-neighbors; on this account alone

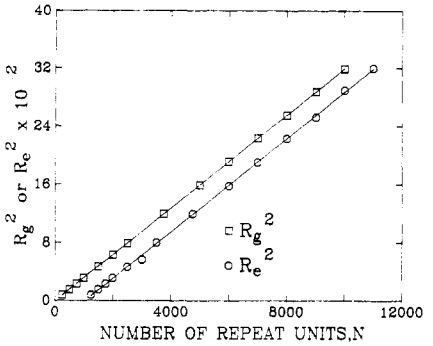

Figure 1. $\langle R_g^2 \rangle$ and $\langle R^2 \rangle$ vs. N for $N = 250$ to $N = 10000$. Data on unweighted R–R chains. $\langle R^2 \rangle$ is offset by 1000 so that $\langle R_g^2 \rangle$ and $\langle R^2 \rangle$ data overlay one another. Notice data are linear and parallel. Scales are exactly a factor of 6 from one another indicating $\langle R^2 \rangle / \langle R_g^2 \rangle = 6$.

it is of interest. Second, as a model of a polymer chain at infinite dilution the modified R–R is a Monte Carlo model of a chain in which only binary cluster terms are considered. A simple reweighting of a set of polymer chains generated by the R–R method would allow one to go from chains with only binary cluster-like terms to chains with all higher order cluster terms; thus studying the effect of the higher order cluster terms on the chain properties might easily be accomplished here. Finally, chains in which only second-order cluster terms are allowed would be expected to be good representations for the so-called two-parameter model of a polymer chain. (Generally these two parameters are χ and characteristic ratio[12,13]—here they are coordination number and ϕ).

The method of chain generation for the Monte Carlo data presented in this section has been described earlier.[8] Only the chain weighting is different. As described earlier,[8] errors for data reported are estimated by using data from about ten different Monte Carlo runs and obtaining the overall averages and the standard deviation. Data with coefficient of variations greater than 5% were not used.

In Figure 1 we show $\langle R^2 \rangle$ and $\langle R_g^2 \rangle$ for the R–R chains with $f = 1$ (unweighted R–R chains). We have studied 40 000 chains of up to length 10 000. In this set 20 runs of 2000 chains each were made. Only for $N = 10000$ were coefficients of variation as large as 5% obtained. For smaller N's coefficients of variation of less than 3% were the rule. To relatively high precision the data apparently fit well to equations linear in N for N from 250 to 10 000. Furthermore, the ratio between $\langle R^2 \rangle$ and $\langle R_g^2 \rangle$ is 6.03 ± 0.04, showing the expected relation between the end-to-end distance and the mean squared radius of gyration at the Θ point.[9,12] The value of $\langle R^2 \rangle / \langle R_g^2 \rangle$ showed no trend as a function of N.

Majid et al. found for the KGW model, a model with trapping, that

$$\langle R_g^2 \rangle \sim N(\ln N)^{0.2} \tag{1.3}$$

An inspection of our data shows that $\langle R^2 \rangle$ and $\langle R_g^2 \rangle$ can be fit to eq 1.3. However, from $N = 250$ to $N = 10000$ the change in either $\langle R^2 \rangle / N$ or $\langle R_g^2 \rangle / N$ is less than 10% and these differences may easily be attributed to small N corrections as found in various models of polymer chains. Such corrections take the form of $A + B/N$, where A and B are constants, for models of polymer chains as random walkers with local structure.[16] For pseudopotential models of chains at the Θ point like those of Fixman and Mansfield,[17] the corrections are of the form $A + B/N^{1/2}$. In Figure 2 $\langle R_g^2 \rangle / N$ vs. $1/N$ is shown. In Figure 3 the same data vs. $1/N^{1/2}$ are shown. In our earlier work on correctly weighted R–R chains at the Θ point we found little to

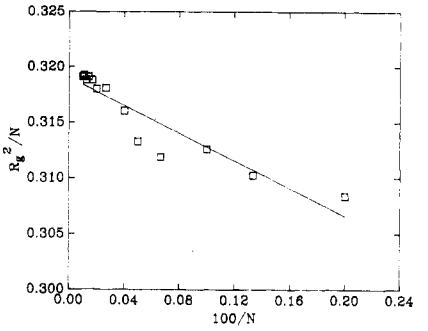

Figure 2. Plot of $\langle R_g^2 \rangle$ vs. $1/N$ from $N = 250$ to $N = 10000$. Same data as in Figure 1.

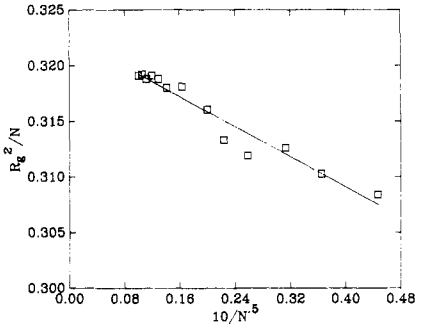

Figure 3. Plot of $\langle R_g^2 \rangle$ vs. $1/N^{1/2}$. Data on unweighted R–R chains. Same data as in Figure 1.

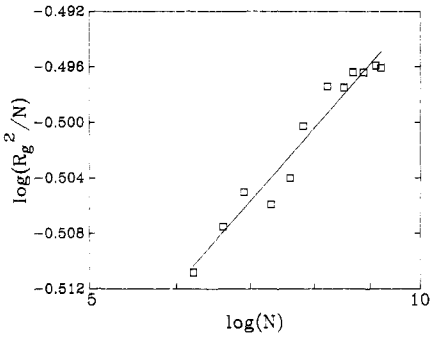

Figure 4. Plot of log $(\langle R_g^2 \rangle / N)$ vs. log (log N). Data on unweighted R–R chains. The solid line is drawn with a slope of 0.2 following Majid et al.[5]

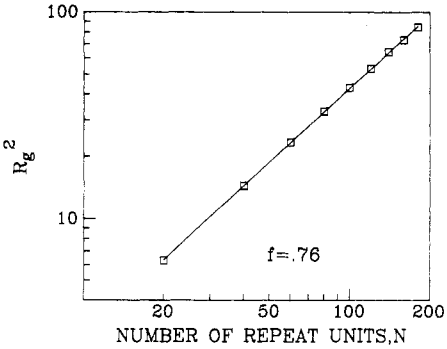

Figure 5. Plot of $\langle R_g^2 \rangle$ vs. N. Data on chains weighted according to eq 2.15 with $f = 0.76$. Solid line is $N^{1.18}$.

distinguish between either of the two choices.[8] In this case the data seem to be fit better by $1/N^{1/2}$. Furthermore, the fit of the $\langle R_g^2 \rangle / N$ data vs. $(\log N)^{0.2}$ in Figure 4 seems also to fit. The data cannot distinguish between these equations.

If these chains are to be Flory-like models of a Θ chain for $f = 1$, one would expect that they show chain expansion for $f < 1$ and chain collapse for $f > 1$.

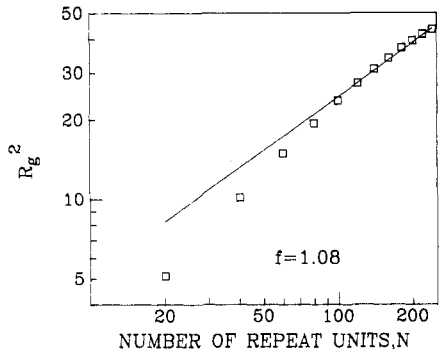

Figure 6. Plot of $\langle R_g^2 \rangle$ vs. N. Data on chains weighted according to eq 2.15 with $f = 1.08$. Solid line is line for $N^{2/3}$.

In Figure 5 we show Monte Carlo data on $\langle R_g^2 \rangle$ for $f < 1$. The data are well represented by an equation of the form

$$\langle R_g^2 \rangle \sim N^{\gamma} \tag{3.3}$$

where $\gamma = 1.18 \pm 0.01$, in agreement with our expectations for chain expansion in a good solvent.

Data for $\langle R_g^2 \rangle$ from Monte Carlo simulations using eq 2.17 for the weight function for $f > 1$ are given in Figure 6. As we found earlier,[9] accurate Monte Carlo data in the chain collapse region are difficult to obtain. For large f, the large coefficient of variation precludes use of data on chains of length of 200 or more. For f just greater than one, one does not approach the asymptotic power law until one gets to very large N and thus obtains larger coefficients of variation. A line for the $^2/_3$ power law is drawn through the large-N data. The data show a general trend toward the $^2/_3$ power line.

Thus we may conclude that if we weight the R–R chains using eq 2.17 the distance parameters in the chains go from good-solvent behavior for $f < 1$ to θ-chain behavior for $f = 1$ to a collapse-chain behavior with $f > 1$. This seems to support our contention that the unweighted R–R is some form of a θ chain.

Such chains with only a variation in f are examples of so-called two-parameter models. The two parameters here are the scale of distance, which put into the Monte Carlo through the lattice and f, which is like Flory's χ.[12,13]

Conclusion

We have suggested that the unweighted R–R walk is a polymer chain at the θ point in a two-parameter model. We propose a two-parameter model of polymer chains which use weighting given in eq 2.17. Such a model shows chain expansion, a θ point, and chain collapse.

We suggest that if higher order cluster-like terms are important in the distinction between a two-parameter model of the θ point and the more realistic modeling of a polymer chain obtained from the R–R chains with the weighting originally proposed by Rosenbluth and Rosenbluth, a careful study of a set of chains created by these two weighting procedures should lead one to obtain a better understanding of the importance of the third-order and higher order cluster terms on equilibrium properties of polymer chains at the θ point. The work of Kremer and Lyklema[14] is suggestive that there is a significant effect of even the fifth-order cluster term.

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