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Analysis of Lattice Chain Data and Test of the Theory of the Expansion Factor for Linear Polymers

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Received May 29, 1980

ABSTRACT: In order to test the theory of the expansion factor α of a linear polymer chain, the estimation of the excluded-volume parameter $z = (3/2\pi\langle R^2\rangle_0)^{3/2}n^2\beta$ and the unperturbed mean-square end-to-end length $\langle R^2\rangle_0 = C_n n a^2$ for lattice chains is reconsidered, where n, β, a, and C_n are the number of segments in the chain, the binary cluster integral for a pair of segments, the effective bond length, and the characteristic ratio, respectively. It is shown that Yamakawa's proposals that $C_n = 1$ and $\beta = \beta_0$, the volume per lattice point, are valid only for chains without short-range effects and that for those with short-range interactions the estimated values of z should be modified by a constant factor depending on the interactions. Approximate theories are compared with Monte Carlo and exact enumeration data. The Domb–Barrett equation for α, a 5(1,1) Padé approximant of α_R for the entire accessible range of z, and the original Flory equation of α_R for $z \gtrsim 5$ are all found to be very good, where the subscript R here refers to the end-to-end distance. The ratios $(\alpha^5 - \alpha^3)/z$ and $(\alpha^5 - 1)/z$ are each apparently constant for $z \gtrsim 5$ but with different proportionality constants, indicating that the asymptotic region is not yet reached for $z \sim 20$.

The excluded-volume effect of flexible polymers in dilute solutions has been well studied[2] but still continues to generate interest. Limiting exponents,[3] delineating the asymptotic behavior of very long chains in very good solvents, have received special attention. Experimentally, of course, only finite chains can be observed and a number of systems are known to deviate from the predicted exponents.[4] However, experimental evidence in general gives strong support to the so-called two-parameter principle.[2]

In the two-parameter scheme, excluded-volume effects on solution properties can be expressed by a single variable z, defined by

$$z = \left(\frac{3}{2\pi}\right)^{3/2} \frac{n^2\beta}{\langle R^2\rangle_0^{3/2}} \tag{1}$$

where n is the number of segments in a chain, $\langle R^2\rangle_0$ is the unperturbed mean-square end-to-end distance, and β is the binary cluster integral for a pair of segments, defined by

$$\beta = \int \left[1 - \exp\left(-\frac{w(\mathbf{R})}{kT}\right)\right] d\mathbf{R} \tag{2}$$

Here $w(\mathbf{R})$ is the potential of mean force of the segment–segment interaction as a function of the relative coordinates $\mathbf{R}$. Many mathematical difficulties are encountered in seeking functional forms of z for various properties. Rigorous results are limited to perturbation series,[2] which have a very limited range and indeed are asymptotic rather than convergent.[5–8] Furthermore, when an attempt is made to compare various approximate theories with experimental data, a fundamental difficulty arises from the fact that the parameter β (or z) is not observable in real solutions. However, a number of numerical studies of lattice chains have been performed,[9,10] and in such cases the above difficulty is eliminated, since β can be identified[11] as the volume β_0 per lattice point for self-avoiding chains.

Yamakawa,[2,12] among others[11–14] who attempted to compare lattice chain data with approximate theories, proposed that we should regard the overlap between bonds due to the reverse step as a part of the excluded-volume effect and take $\langle R^2\rangle_0 = na^2$, with a the bond length and $\beta = \beta_0$ for self-avoiding chains, and then observed that the relation between the expansion factor α and z may be approximated by a single composite curve independent of the type of lattice, as is consistent with the principle of the two-parameter theory. Later, Domb and Barrett[15] pursued this line further and proposed a closed formula for α, which takes account of the first three series coefficients but also of the estimated limiting values based on their lattice work. However, $\langle R^2\rangle_0 = na^2$ for 4-choice simple cubic lattice chains and for this case the data separate from the curve mentioned above.[12] This strongly suggests that the combination of $\langle R^2\rangle_0 = na^2$ and $\beta = \beta_0$ is not always valid for self-avoiding walks, and it thus appears necessary to reconsider the estimation of $\langle R^2\rangle_0$ and β. The main purpose of this paper is to correlate curves

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of α for lattice chains and to compare them with approximate theories.

In the two-parameter theory, the excluded volume is superimposed on a Gaussian chain which includes the effects of all short-range interactions along the chain in the effective bond length. A corresponding lattice model is a self-intersecting chain studied by Alexandrowicz and Accad[14] and by Domb and Joyce,[16] in which a self-intersection of the chain is considered as the excluded-volume effect. In the next section, it is shown that approximate theories can also be compared with data for this model since $\langle R^2\rangle_0$ and β are uniquely determined. If we make our model more realistic, such as 4-choice simple cubic lattice chains in which valence angles are fixed, the effective potential for a pair of segments includes interactions with close neighbors on the chain. Thus, we must take into account the correlation between short-range and long-range interactions in order to evaluate β.[17-19] An attempt along this line was made,[17] but a general recipe is not yet available. A proposal that z with $\langle R^2\rangle_0 = na^2$ and $\beta = \beta_0$ should be modified by a constant factor depending on the type of lattice, for self-avoiding chains with short-range interactions, is shown in section II to be supported by existing data. After establishing the estimation of $\langle R^2\rangle_0$ and z, we examine several approximate theories in section III.

We have not included Monte Carlo results for nonlattice chains[20-23] since there does not thus far seem to be sufficiently good agreement among them.

I. $\langle R^2\rangle_0$ and z for Lattice Chains

In the two-parameter treatment, any restrictions on the bond angles and internal rotations in a polymer chain are included in the effective bond length. The unperturbed bond probability is therefore spherically symmetric. The excluded-volume effect is superimposed on this chain and, as a result, $\langle R^2\rangle_0$ and $n^2\beta$ are characteristic parameters representing short-range and long-range interactions in the chain, respectively.

Lattice chains for which the overlap between bonds due to the reverse step is regarded as a part of the excluded-volume effect correspond to the two-parameter model. If an overlap of units is weighted by w, we obtain from eq 2

$$\beta = \beta_0(1 - w) \tag{3}$$

The Θ state, at which β vanishes, is realized at $w = 1$. In this state all chains are equally weighted whether they have any overlap or not, and the mean-square end-to-end distance is na^2. At $w = 0$, $\beta = \beta_0$ and only self-avoiding walks contribute to the averages, which supports Yamakawa's proposal. Several numerical studies,[9,10] by Monte Carlo and exact enumeration methods, have been performed for $w = 0$, and Alexandrowicz and Accad[14] have treated arbitrary w. All these chains we shall call *class I*. It is noted that for this class α and z are determined numerically and that the relation between them is indeed a proper question to be pursued by theoretical efforts.

Next, we consider random walks for which some of the immediate steps are excluded a priori. If all the chains are equally weighted, the mean-square end-to-end distance is $C_n na^2$. The characteristic ratio C_n[24] is generally larger than 1 and becomes independent of n, if n is large, which corresponds to the Θ state. When zero weight is assigned to chains which have any overlap, only self-avoiding walks contribute to the averages. A pair interaction may be assumed to be expressed in the form of eq 3. We call these walks of *class II*. We observe that $(q - 1)$-choice self-avoiding walks may belong to either of the two classes,

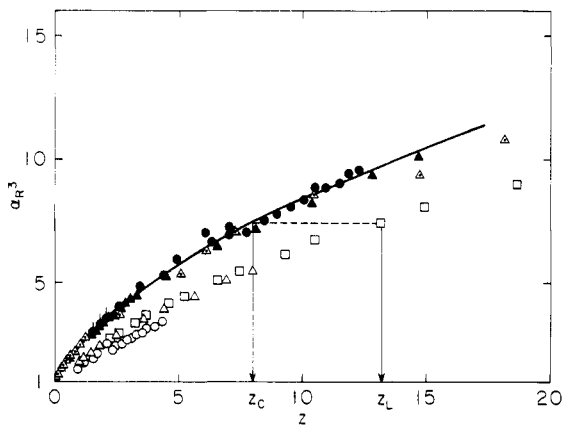

Figure 1. Plot of α_R^3 against z for chains on various types of lattice: (○, ●, ◐) for tetrahedral lattice chains;[25-27] (△, ▲, ◭) for 5-choice simple cubic lattice chains;[28-30] (◬) for 6-choice simple cubic lattice chains;[10] (□) for 4-choice simple cubic lattice chains.[31,32] The filled and center-dotted points are for class I and the open points are for class II. The points with pips show the data calculated from exact enumeration results; others are those from Monte Carlo methods. The solid curve represents the best fit to the data for class I. Arrows show how z_C is determined from values of α_R for class II and the solid curve.

where q is a lattice coordination number, depending on how the unperturbed dimensions are chosen.

Now, a class II chain simulates a real polymer chain more realistically than a chain of class I. We may regard a unit at a lattice point as an atom or at most a monomer unit in the main chain. If k consective bonds of class II chains correspond to one bond of class I, the binary cluster integral must involve k^2 pair interactions. If each pair is independent, this amounts to $k^2\beta$. However, because of correlations with close neighbors along the chain, the total effect is actually smaller than $k^2\beta$. The number of segments is n/k. The correct z is thus

$$z = C_z z_L \tag{4}$$

where z_L is calculated from eq 1 with $\langle R^2\rangle_0 = C_n na^2$ and $n^2\beta = n^2\beta_0(1 - w)$. The correction factor C_z must be constant for large n and smaller than 1. At present, C_z cannot be estimated by computer results.

If the universality of the two-parameter principle holds, the following two conditions must be satisfied: (1) z_L/z_C = constant independent of n; (2) the relation between α_S and z_C for class II should coincide with that of class I, where z_C is to be determined from values of α_R for class II and the relation between α_R and z for class I. The subscripts R and S refer to the mean-square end-to-end distance and radius of gyration, respectively. These conditions are examined in the next section as criteria of validity of the above prediction (and therefore of the two-parameter theory).

For classes I and II in the unperturbed state we must consider overlap of units. But *physically* we can never have intersecting chains, and the Θ state results from favorable weighting of segment–segment contacts. However, as seen above, if we make our model more realistic than class I, we cannot determine z from experiment. Therefore, in this paper we consider only the above two classes.

II. Test of the Two Criteria

In order to test whether the two criteria may be satisfied or not, we must first of all determine values of z_C. In Figure 1, values of α_R^3 for various types of lattice chains are plotted against z (or z_L), where the circles, triangles, and squares represent values obtained from numerical data for tetrahedral,[25-27] 5- and 6-choice simple cubic,[14,28-30] and

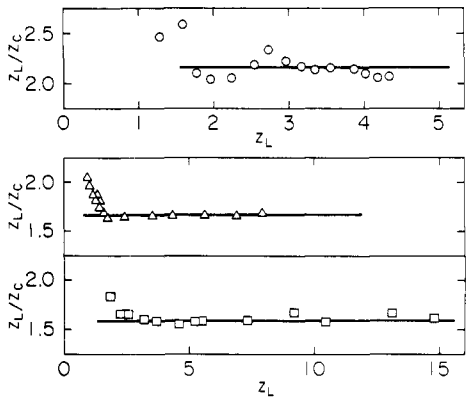

Figure 2. Test of constancy of the ratio z_L/z_C. The symbols are the same as in Figure 1. The solid lines give the best fits to the plotted points for large z_L.

Table I
Values of the Constant C_z and Lower Bound n_1 for Lattice Chains

lattice chain	C_z	n_1
tetrahedral lattice chain	0.46	100
5-choice simple cubic lattice chain	0.60	80
4-choice simple cubic lattice chain	0.63	100

4-choice simple cubic lattice chains,[31,32] respectively. The values for class I are shown by the filled and center-dotted points, while those for class II are shown by the open points. The filled and open points are for self-avoiding chains and the center-dotted points for self-intersecting chains.[14] It is noted that the latter data by Alexandrowicz and Accad cover wide ranges of z, especially near $z \approx 0$, where we have no results for self-avoiding walks.

For the sake of comparison, values calculated from exact enumeration data[26,30] are also plotted in the figure, as the points with pips. Since n is less than 21, only class I chains are considered. For such short chains, we tentatively modify z as

$$z = \left(\frac{3}{2\pi}\right)^{3/2} \frac{(n-1)(n-2)}{\langle R^2\rangle_0^{3/2}} \beta_0 \qquad (5)$$

from the viewpoint that $n^2\beta$ in eq 1 may mean twice the total excluded volume for large n. The points then agree well with those from Monte Carlo calculations, as well as at $z = 0$ for $n = 2$.

A single representation of the points for class I is shown by the solid curve. The open points, which are those for class II, depend on z and the type of lattice.[2,12] In the figure the method of determining z_C from the graph is also depicted.

In order to test criterion 1, values of z_L/z_C for three types of lattice are displayed in Figure 2. The symbols are the same as in Figure 1. The ratios become constant when n exceeds some value n_1, indicating that the criterion 1 is satisfied. The asymptotically constant values C_z^{-1} of the ratios are shown by the horizontal lines. C_z and n_1 are given in Table I and, as expected, C_z is smaller than 1.

Using the given C_z for each lattice, we now evaluate $z = C_z z_L$ and then plot α_S^3 vs. z in Figure 3. The points so determined are shown by the center-dotted circles and triangles.[25-28,33] The other symbols are the same as in Figure 1. The filled points and center-dotted points compose a single curve; i.e., criterion 2 is also satisfied.

It is clear from the above analysis that the relation between α_S and α_R is the same for both classes. Figure 4 directly illustrates a sensitive test of this relation. The symbols are the same as in Figure 1. It is noted that in constructing this figure no assumption is required for the determination of z. The line (1) is the best fit to the data for $\alpha_R^3 > 3$:

$$\alpha_S^3/\alpha_R^3 = 0.91 \qquad (6)$$

This number is very close to the value deduced by Domb and Barrett,[15] who give $\alpha_S^3/\alpha_R^3 = 0.90$. The deviation of the filled points for $\alpha_R^3 < 5$ from eq 6 is due to the short-chain effects discussed later.

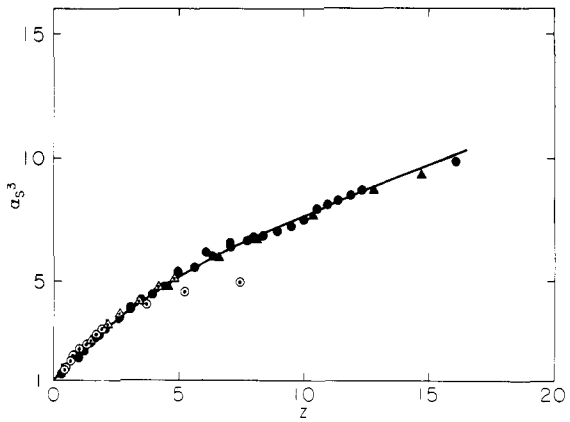

Figure 3. Plots of α_S^3 against z. The center-dotted points[25-28,33] represent data for class II plotted against $z = C_z z_L$. The other symbols are the same as in Figure 1.

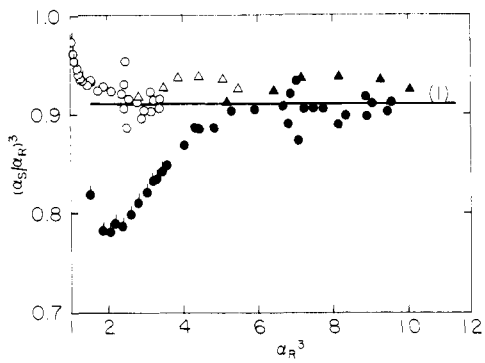

Figure 4. Relationship between α_S^3 and α_R^3. The straight line (1) shows the best fit to data for $\alpha_R^3 > 3$. The other symbols are the same as in Figure 1.

Some comments are now given on the values of n_1 and C_z. If the unperturbed mean-square distance of any two units i and j in a chain has its random-flight value, $\langle R_{ij}^2\rangle_0 = |j-i|a^2$, then strictly $\langle S^2\rangle_0 = (na^2/6)(n+2)(n+1)^{-1}$. A chain longer than about 100 steps is thus required in order that $\langle S^2\rangle_0$ is proportional to n. It appears that n_1 is approximately equal to this lower bound. If the number of segments in a lattice chain of class II equals the number of units, the effective binary cluster integral per segment is $C_z\beta_0$, i.e., $^1/_2$ to $^2/_3$ as large as that for small molecules. If rotational-angle restrictions were also to be taken into account, still smaller values would be expected. Actually, values of β per monomer unit that have been estimated from the measured properties of dilute-polymer solutions are about one-tenth as large as the values predicted by analogy with nonpolymeric solutions.[34,35]

It should be mentioned that the two-parameter scheme has been implicitly assumed by recent workers concerned with excluded-volume effects and asymptotic exponents.[3,36-38] In these papers the binary segment–segment cluster integral is often written as v instead of β.

III. Test of Theories of the Expansion Factor

In sections I and II it has been argued that the theory of α can properly be compared with the data for class I

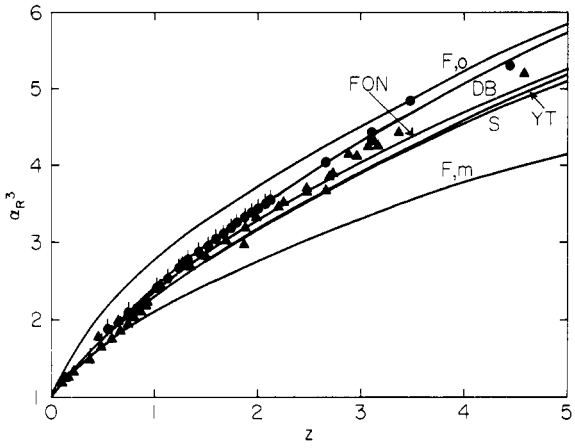

Figure 5. Plots of α_R^3 against z. The center-dotted triangles in Figure 1 are changed into filled ones. The other symbols are the same as in Figure 1. Curve F,o, eq 7 with $C = 2.60$. Curve F,m, eq 7 with $C = {}^4/_3$. Curve YT, eq 8 with $A = 0.428$, $B = 6.23$, and $C = {}^1/_2$. Curve S, eq 9 with $A = {}^{10}/_3$, $B = 2.957$, and $C = 3.513$. Curve DB, eq 10. Curve FON, eq 12.

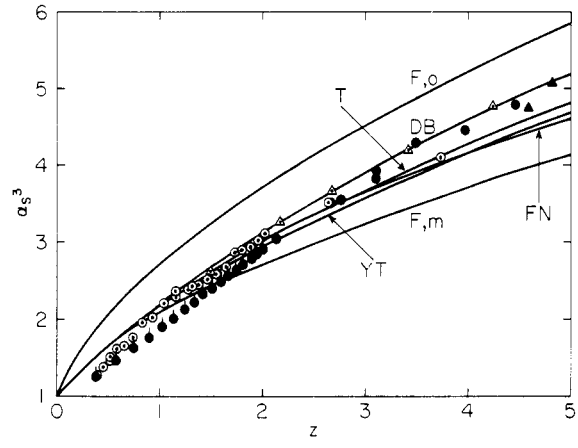

Figure 6. Plots of α_S^3 against z. The symbols are the same as in Figure 3. Curve F,o, eq 7 with $C = 2.60$. Curve F,m, eq 7 with $C = {}^{134}/_{105}$. Curve YT, eq 8 with $A = 0.459$, $B = 6.04$, and $C = 0.46$. Curve T, eq 9 with $A = {}^{67}/_{21}$, $B = 2.501$, and $C = 3.177$. Curve DB, eq 11 with eq 10. Curve FN, numerical results due to Fujita–Norisuye.

and that data for class II will fit with those of class I if we use z_C in place of z_L. In this section we test approximate theories and estimate the behavior of α at large z by using these data. For this purpose we choose six typical expressions for α. They are the Flory equation[39]

$$\alpha^5 - \alpha^3 = Cz \tag{7}$$

the Yamakawa–Tanaka equation[40]

$$\alpha^2 = 1 - A + A(1 + Bz)^C \tag{8}$$

a (2,2) Padé approximant due to Stockmayer[19]

$$\alpha^5 - 1 = Az\frac{1 + Bz}{1 + Cz} \tag{9}$$

the Domb–Barrett equation[15]

$$\alpha_R^2 = \left[1 + 10z + \left(\frac{70}{9}\pi + \frac{10}{3}\right)z^2 + 8\pi^{3/2}z^3\right]^{2/15} \tag{10}$$

with

$$\alpha_S^2/\alpha_R^2 = 0.933 + 0.067\exp[-(0.85z + 1.39z^2)] \tag{11}$$

the Fujita–Okita–Norisuye equation[41]

$$\alpha_R^5 - 0.4931\alpha_R^3 - 0.2499\alpha_R^{-1.332}\sin(1.073\ln\alpha_R) - 0.5069\alpha_R^{-1.332}\cos(1.073\ln\alpha_R) = 2.63z \tag{12}$$

and the Fujita–Norisuye relation[42] for α_S, which is available only in numerical form. Here A, B, and C are numerical constants characteristic of each theory.

First we compare theories of α_R with experiment. An enlargement of Figure 1 for small z is displayed in Figure 5 with curves calculated from the above equations: F,o (eq 7 with $C = 2.60$), F,m (eq 7 with $C = {}^4/_3$),[11] YT (eq 8 with $A = 0.428$, $B = 6.23$, and $C = {}^1/_2$), S (eq 9 with $A = {}^{10}/_3$, $B = 2.957$, and $C = 3.513$),[43] DB (eq 10), and FON (eq 12). The center-dotted points in Figure 1 are now filled. The other symbols are the same as in Figure 1. The Domb–Barrett equation fits all the data well, while curves YT, S, and FON do so for $z < 3$.

Next we examine theories for α_S. Similar plots for α_S are shown in Figure 6. The symbols are the same as in Figure 3. Curves F,o, F,m, YT, T, DB, and FN represent eq 7 with $C = 2.60$, eq 7 with $C = {}^{134}/_{105}$,[45,46] eq 8 with $A = 0.459$, $B = 6.04$, and $C = 0.46$, eq 9 with $A = {}^{67}/_{21}$, $B = 2.501$, and $C = 3.177$,[47] eq 11 with eq 10, and the numerical results of the Fujita–Norisuye treatment, respectively. It

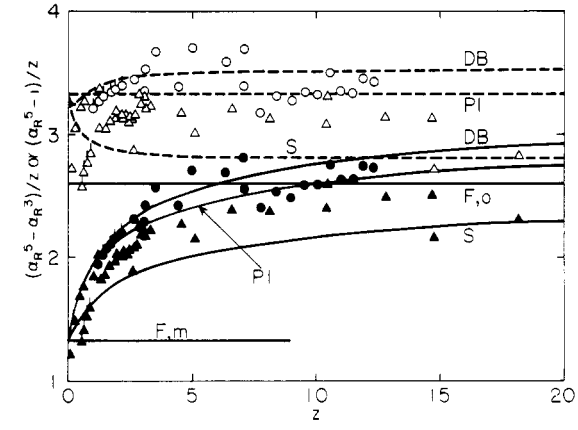

Figure 7. Dependence of $(\alpha_R^5 - \alpha_R^3)/z$ and $(\alpha_R^5 - 1)/z$ on z. The filled and open points are those for class I and the solid and broken curves are those for theoretical values. P1 is a 5(1,1) Padé approximant, eq 9 with $A = {}^{10}/_3$ and $B = C = 0$. The other symbols are the same as in Figure 1.

is noted that a calculation by Gobush, Šolc, and Stockmayer[48] nearly coincides with the curve YT in the range of z shown here. At present, we have no information on class I for $z < 3$ because the available data are limited to self-avoiding walks; for a self-avoiding walk of 100 steps, we obtain $z = 3.3$ (β_0/a^3) > 3. The data for class II plotted against z_C may be useful down to $z \sim 1$, with $n \sim 150$. Therefore, though we cannot compare theory with experiment for $z < 1$, the Domb–Barrett equation may nevertheless be regarded as satisfactory, but the curves YT, FN, and T are adequate only for $z < 3$.

Asymptotically α behaves as

$$\alpha^\nu = Cz \tag{13}$$

The rigorous exact value of the limiting exponent ν is still uncertain,[12,36,49-51] but it is clear that the old Flory value, $\nu = 5$, is very accurate. To examine asymptotic behavior and estimate a constant C, we plot in Figures 7 and 8 observed values of $(\alpha^5 - \alpha^3)/z$ and $(\alpha^5 - 1)/z$ (shown by the filled and open points for class I) and the corresponding theoretical values (shown by the solid and broken curves), respectively. Values of class II are represented in Figure 8 by the large and small center-dotted points. Curve P1 in Figure 7 is a 5(1,1) Padé approximant (eq 9 with $A = {}^{10}/_3$ and $B = C = 0$).[47] The other symbols are the same as those in Figures 1 and 5 for α_R and in Figures

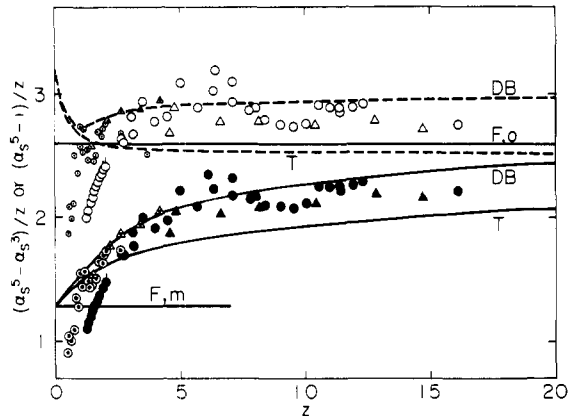

Figure 8. Dependence of $(\alpha_S^5 - \alpha_S^3)/z$ and $(\alpha_S^5 - 1)/z$ on z. The large and small center-dotted points are those for class II. The other symbols are the same as in Figure 7.

3 and 6 for α_S, respectively. Both functions appear to become constant for large z:

$$\alpha^5 - \alpha^3 = Cz \tag{14}$$

with

$$C_R = 2.6$$
$$C_S = 2.2$$

$$\alpha^5 - 1 = Cz \tag{15}$$

with

$$C_R = 3.3$$
$$C_S = 2.85$$

Thus, both equations lead to eq 13 with $\nu = 5$, but with different constants, indicating that the asymptotic region is not reached. The perturbation theory with the estimated values of C suggests that the function $(\alpha^5 - 1)/z$ has a minimum, but this is not certain because of the lack of data and the magnitude of calculational errors.

The Domb–Barrett equations fit the data over the whole range of z shown here, perhaps not surprisingly, since they are based both on the first three series coefficients and on the asymptotic limits, $C_R = 3.44$ and $C_S = 2.90$,[15] very close to eq 15. The 5(2,2) Padé approximant which relies on the first three series coefficients and the asymptotic power $\nu = 5$ deviates from the data for large z. Surprisingly, a simpler 5(1,1) Padé form for α_R, curve P1, is seen to lie closer to the data.[47]

IV. Conclusion

We have reestimated $\langle R^2\rangle_0$ and z for lattice chains and compared approximate theories with numerical calculations for such chains. The conclusions reached are summarized as follows:

(1) z with $\langle R^2\rangle_0 = na^2$ and $\beta = \beta_0(1 - w)$ are valid only for lattice chains without short-range effects.

(2) For chains *with* short-range interactions, the estimated z should be modified by a constant factor dependent on these interactions.

(3) The Domb–Barrett equation for α and a 5(1,1) Padé approximant of α_R are very good over the entire accessible range of z, and the original Flory equation for α_R fits well for $z > 5$.

(4) The functions $(\alpha^5 - \alpha^3)/z$ and $(\alpha^5 - 1)/z$ are each essentially constant for $z > 5$, but the proportionality constants are different. This fact indicates that the asymptotic region is not reached even for $z \sim 20$.

Acknowledgment. The author is grateful to Professor W. H. Stockmayer for helpful discussions and suggestions and for his critical reading of the manuscript. He also thanks Professor H. Yamakawa for his useful comments, encouragement, and interest in the present work. This work was supported in part by the National Science Foundation under Grant No. DMR 76-22985 AO1 of the Polymers Program, Division of Materials Research.

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