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Swelling of Subchains of a Single Polymer Chain with Excluded Volume in Two and Three Dimensions: A Monte Carlo Study

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ABSTRACT: The expansion of subchains of a single excluded volume chain in two and three dimensions was studied using the bond-fluctuation model. The average expansion factor of subchains containing n segments shows a maximum for $n < N$, where N is the number of bonds of the whole chain. The subchains are more swollen than an isolated chain containing the same number of segments, and this excess swelling is most pronounced in the center of the chain. Both the behavior of the average expansion factor and the excess swelling can be described using the analytical results of Duplantier and of Schäfer and Baumgärtner, particularly in three dimensions.

1. Introduction

In this work we focus on single isolated chains with excluded volume which are related to chains in a very dilute solution under good solvent conditions. An isolated chain with excluded volume exhibits larger mean dimensions than do chains unperturbed by this long-range interaction.[1] The chain is swollen, and its mean square end-to-end distance $\langle R_N^2\rangle = \langle(\vec{r}_0 - \vec{r}_N)^2\rangle$ obeys the scaling law for large N

$$\langle R_N^{\ 2}\rangle \sim N^{2\nu} \tag{1}$$

where N is the number of chain segments, $\vec{r}_i$ is the position of monomer i ($i = 0, 1, ..., N$), and ν is the correlation length exponent. In three dimensions renormalization group calculations yield $\nu = 0.588 \pm 0.001$[2] whereas in two dimensions the exact value $\nu = {}^3/_4$ is known.[3] Without excluded volume (denoted in the following by the subscript 0) $\langle R_N^2\rangle_0$ behaves as $\langle R_N^2\rangle_0 \sim N$. The overall expansion of the whole chain, described by the expansion factor $\alpha^2 = \langle R_N^2\rangle/\langle R_N^2\rangle_0$, is well-known and was studied by various authors (see for example refs 4 and 5 and references therein).

Less knowledge exists concerning the expansion of a given part of the chain in relation to the expansion of the whole chain, i.e. the swelling of subchains. A subchain of a chain is defined as a part consisting of $n = |j - i|$ segments ($n \leq N$) between the monomers indexed by i and j ($i, j = 0, ..., N$). A long discussed problem is whether the subchains of a given length n swell uniformly independent of their position in the chain[6] or not. Early works performed both analytically[7,8] and numerically[9,10] showed that the mean square end-to-end distance $\langle R_{i,j}^2\rangle = \langle(\vec{r}_i - \vec{r}_j)^2\rangle$ of the subchain is a rather complicated function of i and j; i.e. the expansion is not uniform. In contrast to that both the popular blob model[11−13] and its modified versions[14−17] assume that internal parts of the chain are less swollen than the total chain; their swelling is independent of their position in the total chain. But such uniform swelling models were criticized by some authors.[18,19]

Computer simulations[20−25] gave some evidence of a dependence of the subchain expansion on the position in the chain. It was found that a subchain is more swollen than an isolated chain containing the same number of bonds. This excess swelling of subchains would be more pronounced if the subchain is located in the middle of the whole chain. Combining Monte Carlo experiments and perturbation results, Barrett[26] obtained approximate formulas for the subchain expansion factor.

An analytical expression for the swelling of subchains was derived by Duplantier[27] and shortly later confirmed by Schäfer and Baumgärtner.[28] According to their first-order ϵ-expansion ($\epsilon = 4 - d$) results, the mean square end-to-end distance $\langle R_{i,j}^2\rangle$ of the subchain between monomers i and j is given by

$$\langle R_{i,j}^{\ 2}\rangle = \langle R_{N=n}^{\ 2}\rangle\left(1 + \frac{\epsilon}{8}F(t,u)\right) \tag{2}$$

with

$$F(t,u) = \frac{1-t}{t}\left\{u \ln\left[1 + \frac{t}{(1-t)u}\right] + (1-u)\ln\left[1 + \frac{t}{(1-t)(1-u)}\right]\right\} - \frac{1}{2}(1-t) \tag{3}$$

and

$$t = \frac{n}{N} = \frac{|j-i|}{N} \qquad u = \frac{i}{(1-t)N}$$

$\langle R_{N=n}^2\rangle$ denotes the mean square end-to-end distance of an isolated chain of n segments (not to be confused with $\langle R_n^2\rangle$ which denotes in the following the mean square end-to-end distance of all subchains of length n). The variable t measures the length of the subchain and u measures the displacement of one extremity of the subchain with respect to the available displacement along the chain ($t, u \in [0, 1]$). In the excluded volume limit $\langle R_{N=n}^2\rangle$ is, according to eq 1, given by

$$\langle R_{N=n}^{\ 2}\rangle = \langle R_N^{\ 2}\rangle\, t^{2\nu} \tag{4}$$

Despite all of these efforts, additional work on this topic is needed. First of all very few data[23] exist in two dimensions, and from a theoretical point of view the two-dimensional case is also of great interest. Moreover a quantitative analysis of the average subchain expansion of all subchains of a given length has never been performed. Furthermore the function $F(t,u)$ was only tested for special cases of u in three dimensions.[28] Thus

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a comparison of $F(t,u)$ with simulation results over a wide range of u and t has not been carried out.

The intention of the present paper is to give a quantitative analysis of $F(t,u)$ with extensive simulation data in two and three dimensions. The outline of the paper is as follows. In the next section the simulation method will be described. The presentation of the results and a discussion will follow.

2. Method

For our calculations the bond-fluctuation model developed by Carmesin and Kremer[30] in two and three dimensions was used. On a lattice (simple square for $d = 2$ and simple cubic for $d = 3$) the monomers are represented by cubes of 2^d positions. The monomers are connected by a set of possible bond vectors (36 in two dimensions and 108 in three dimensions). The diffusion dynamics (in the free draining limit) is simulated by randomly chosen jumps of monomers in spatial directions. These jumps are accepted after checking for self-avoiding. Certain restrictions on the permitted bond vectors automatically lead to cut avoiding. This process fulfills excluded volume. No further interactions between the monomers were taken into account. Thus the simulation was athermal. For more details on the bond-fluctuation model we refer to the original papers.[30-33]

We studied single chains of $(N + 1) = 10, 15, 20, 30, 40, 50, 60, 70, 100$, and 200 monomers on lattices with periodic boundary conditions. In two dimensions the lattice extensions were 40×40 up to 600×600 and in three dimensions $20 \times 20 \times 20$ up to $110 \times 110 \times 110$. The dimensions of the lattices were large enough that interactions of the chain with itself through the periodic boundary conditions can be neglected. To obtain proper statistics, we performed 100 up to 1100 independent simulations. A single simulation was carried out as follows: After the chains were created, they were relaxed at least for 1 000 000 MCS. One MCS, one Monte Carlo step, is one attempted jump per monomer. Then the distances $R_{i,j}^2$ were measured in intervals of 500 MCS for a total amount of 3 000 000 MCS. The distances $R_{i,j}^2$ were averaged over time and over the independent simulations.

3. Results and Discussion

3.1. Average Expansion of Subchains.

The expressions eqs 2 and 3 are obtained in the excluded volume limit. Hence for a quantitative comparison of the simulation data with these expressions, one must verify whether this limit has been properly reached, i.e. whether eq 1 is fulfilled. Considering plots of the mean square end-to-end distance and mean square radius of gyration versus the chain length, we found that for $N > 20$ the simulations are well within the excluded volume limit. For such plots we refer to another paper.[34]

First we will discuss the average expansion factor α_n^2 of all subchains containing n segments, which is defined as

$$\alpha_n^{\ 2} = \frac{\langle R_n^{\ 2}\rangle}{\langle R_n^{\ 2}\rangle_0} \tag{5}$$

with

$$\langle R_n^{\ 2}\rangle = \frac{1}{N - n + 1}\sum_{i=0}^{N-n}\langle R_{i,i+n}^{\ 2}\rangle \tag{6}$$

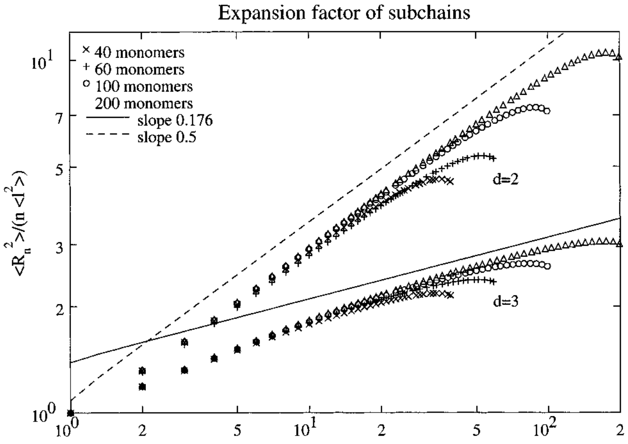

Figure 1. log–log plot of $\alpha_n^2 = \langle R_n^2\rangle/(n\langle l^2\rangle_0)$ versus n in two and three dimensions. The points represent the simulated data (40, 60, 100, and 200 monomers), and the lines have slopes of 0.5 ($d = 2$, dashed line) and 0.176 ($d = 3$, solid line), respectively.

and $\langle R_n^2\rangle_0$ analogous. For $\langle R_n^2\rangle_0$ we used $\langle R_n^2\rangle_0 = n\langle l^2\rangle_0$ where $\langle l^2\rangle_0$ is the mean square bond length in the bond-fluctuation model ($\langle l^2\rangle_0 \approx \langle l^2\rangle$).

Figure 1 shows α_n^2 versus n for chains with 40, 60, 100, and 200 monomers in two and three dimensions. For both dimensions the plot shows a concave downward shape and a maximum of α_n^2 for $n < N$. This maximum is more pronounced in two dimensions and is described by neither the original blob model[11-13] nor the modified blob models.[14-16] According to these models α_n^2 should behave as $\alpha_n^2 \sim n^\eta$ with an exponent η which is given by $\eta = 0$ for small n and by $\eta = 2\nu - 1$ for large n. Since in our athermal simulation the number of segments in a blob is about 1, an observation of a crossover between $\eta = 0$ and $\eta = 2\nu - 1$ is not expected even if the blob model is valid. Nevertheless on the basis of the blob models one expects a convex shape in contrast to the concave shape shown by the simulation results.[20]

Figure 1 also includes lines whose slopes are $2\nu - 1$, i.e. 0.5 for $d = 2$ and 0.176 for $d = 3$,[2,3] which represent the slopes in the blob models for large n. In two dimensions the slope 0.5 can be observed over a large n range, particularly in the case of long chains. In three dimensions only a short range with slope 0.176 is visible and it is not quite clear whether this range becomes larger with increasing N. The last observation is in agreement with the work of Mattice[22] who found no indication that the slope tends toward 0.2 as N becomes large.

In order to test whether the analytical expressions of Duplantier[27] and of Schäfer and Baumgärtner[28] are able to describe the data, we calculated $\langle R_n^2\rangle$ with eqs 2 and 3 by integrating over u. This yields together with eq 4

$$\langle R_n^{\ 2}\rangle = \langle R_N^{\ 2}\rangle t^{2\nu}\left(1 + \frac{\epsilon}{8}h(t)\right) \tag{7}$$

where

$$h(t) = \int_0^1 F(t,u)\,\mathrm{d}u = \frac{t+1}{2} + \frac{t-1}{t}\ln(1-t) + \frac{t}{1-t}\ln(t) \tag{8}$$

Since one does not expect a first-order ϵ-expansion result being able to describe the data quantitatively, we used a fit parameter a instead of $\epsilon/8$, i.e. $\langle R_n^2\rangle = \langle R_N^2\rangle t^{2\nu}(1 +$

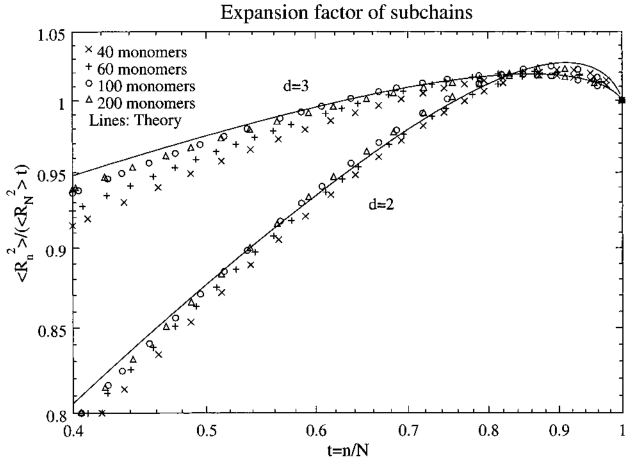

Figure 2. log−log plot of $\langle R_n^2\rangle/(\langle R_N^2\rangle t)$ versus $t = n/N$ in two and three dimensions. The points represent the simulated data (40, 60, 100, and 200 monomers), and the lines, $(1 + ah(t))t^{2\nu-1}$. $a = 0.33$ for $d = 2$ and $a = 0.152$ for $d = 3$.

$ah(t)$). Since the simulated chain lengths are in the excluded volume limit, the readjustment of $\epsilon/8$ by a may be said to be due to higher orders of ϵ and does not represent the introduction of a coupling constant (in the sense of ref 28), describing corrections to scaling.

In Figure 2 we plotted $\langle R_n^2\rangle/(\langle R_N^2\rangle t) = \alpha_n^2/N^{2\nu-1}$ versus $t = n/N$ in two and three dimensions. For large t all the data nearly coincide, as expected. The short chains (40 and 60 monomers) fall away from this scaling regime at intermediate t since the subchains become rather short and cannot follow the asymptotic behavior. Furthermore we find that eq 8 (solid lines) describes the data with $a = 0.33$ for $d = 2$ and $a = 0.152$ for $d = 3$. It should be stressed that the theory also predicts a maximum at $t < 1$. In three dimensions the result for $a = 0.152$ is closer to $\epsilon/8 = 0.125$ than in two dimensions where $a = 0.33$ in comparison to $\epsilon/8 = 0.25$. However, it is known that the ϵ-expansion results are better in three dimensions than in two dimensions (see for example ref 35). For $t < 0.4$ the differences between the theory and the data increase with decreasing t, because eqs 2 and 3 are valid for $n \to \infty$.

3.2. Position Dependence of the Subchain Expansion. To investigate the position dependence of the subchain expansion, we studied the quantity $\langle R_{i,i+n}^2\rangle/(\langle R_N^2\rangle t^{2\nu}) - 1$. If a subchain exhibits the same swelling as an isolated chain of the same number of bonds, this quantity will be zero. Using the analytical ϵ-expansion results eqs 2 and 3, this quantity is given by $aF(t,u)$ with a fit parameter a instead of $\epsilon/8$, as in the previous section.

In Figures 3 ($d = 2$) and 4 ($d = 3$) the quantity $\langle R_{i,i+n}^2\rangle/(\langle R_N^2\rangle t^{2\nu}) - 1$ plotted versus $u = i/(N - n)$ for various t. Since this quantity is sensitive to statistical errors and the analytical expression eq 3 holds only for large n, we plot only the data for chain length 200 which are based on 1100 independent simulation runs. For all subchains the figures show an excess swelling which increases with decreasing t. This means that the excess swelling for shorter subchains is larger than for longer ones. Clearly, if the subchain coincides with the whole chain ($n = N$), the excess swelling must vanish. Further, the figures show a strong dependence of the excess swelling on the subchain position in the chain. Hence the swelling is not uniform. The maximum swelling is reached if the subchain is located in the middle of the chain. A comparison of Figures 3 and 4 shows that the excess swelling in two dimensions is about twice as much as in three dimensions.

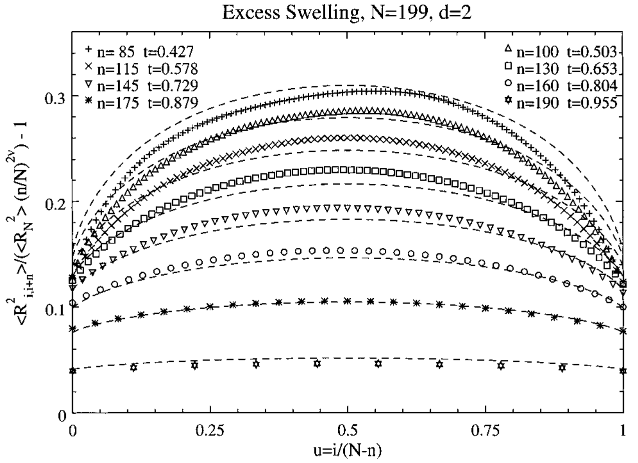

Figure 3. Plot of $\langle R_{i,i+n}^2\rangle/(\langle R_N^2\rangle t^{2\nu}) - 1$ versus $u = i/(N - n)$ in two dimensions. The points represent the simulated data of the 200 monomer chain for various t. The dashed lines represent the function $aF(t,u)$ with $a = 0.33$.

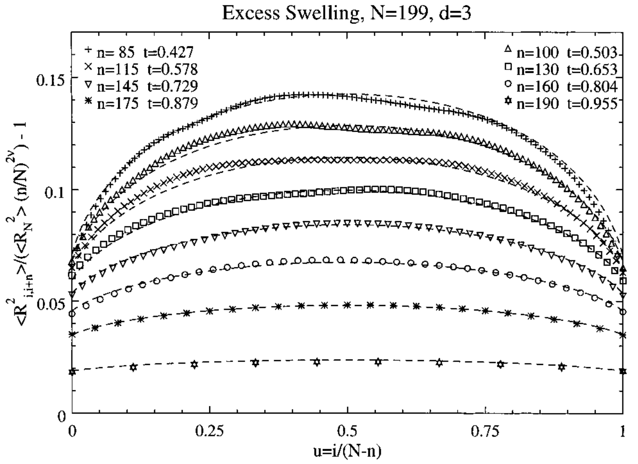

Figure 4. Plot of $\langle R_{i,i+n}^2\rangle/(\langle R_N^2\rangle t^{2\nu}) - 1$ versus $u = i/(N - n)$ in three dimensions. The points represent the simulated data of the 200 monomer chain for various t. The dashed lines represent the function $aF(t,u)$ with $a = 0.152$.

These facts may be understood as follows: A subchain feels the external segments of the rest of the chain which adds to its swelling. A subchain near the center of the chain is swollen by more external segments than the same subchain located near the ends of the chain. Clearly, the restrictions caused by excluded volume are stronger in two dimensions than in three dimensions. However, the exact mechanism of the excess swelling is more complicated. Schäfer and Krüger[29] found theoretically that the segment density in the center of a chain is reduced. This depression results from reduced fluctuations of the segment positions.

In order to compare these results with the theoretical results of Duplantier[27] and of Schäfer and Baumgärtner,[28] we also plot $aF(t,u)$ (dashed lines) in Figures 3 and 4. For the parameter a we use the values found in the previous section: $a = 0.33$ for $d = 2$ and $a = 0.152$ for $d = 3$. With these values the simulated data agree very well with eq 3 in three dimensions whereas in two dimensions some differences can be seen. As noted in the previous section, it is not surprising that a first-order ϵ-expansion result is a better quantitative description of the data in three dimensions than in two dimensions. In both dimensions the differences between

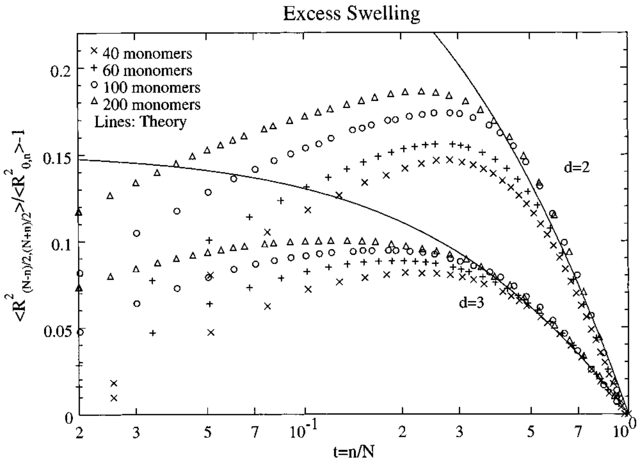

Figure 5. Single logarithmic plot of $\langle R_{(N-n)/2,(N+n)/2}{}^2\rangle/\langle R_{0,n}{}^2\rangle - 1$ versus $t = n/N$ in two and three dimensions. The points represent the simulated data (40, 60, 100, and 200 monomers), and the lines, $a(1 - t)/t \ln(1 + t)$. $a = 0.33$ for $d = 2$ and $a = 0.152$ for $d = 3$.

the data and eq 3 would increase if shorter subchains ($t < 0.4$) are considered. This is due to the fact that eq 3 is formulated for very long continuous chains. In the simulation short finite chains are not able to swell as in the asymptotic limit. In the extreme case of a single segment $n = 1$, the "subchain" cannot swell at all whereas eq 3 becomes large for $t \rightarrow 0$.

In order to perform a quantitative analysis of the influence of the chain length N, we studied the excess swelling of a subchain which belongs to the central part of the whole chain ($u = 0.5$) in relation to a subchain which constitutes one extremity of the whole chain ($u = 0$), i.e. the quantity $\langle R_{(N-n)/2,(N+n)/2}{}^2\rangle/\langle R_{0,n}{}^2\rangle - 1$. If a subchain in the middle and at the end of a chain would exhibit the same swelling, this quantity would be zero. Using eqs 2 and 3 it is given by

$$\frac{\langle R_{(N-n)/2,(N+n)/2}{}^2\rangle}{\langle R_{0,n}{}^2\rangle} - 1 = \frac{\epsilon}{8}[F(t,u{=}0.5) - F(t,u{=}0)] + 0(\epsilon^2) = \frac{\epsilon}{8}\frac{1-t}{t}\ln(1+t) \quad (9)$$

Figure 5 shows a plot of $\langle R_{(N-n)/2,(N+n)/2}{}^2\rangle/\langle R_{0,n}{}^2\rangle - 1$ versus $t = n/N$ for various chain lengths in two and three dimensions. The theoretical lines are given by eq 9. Again we used the fit parameter a instead of $\epsilon/8$. The data for all chain lengths nearly coincide for large n and are in agreement with eq 9. For small subchain lengths n the scaling breaks down and the data drop down to zero depending on N. This is due to the fact that in the simulation $\langle R_{(N-n)/2,(N+n)/2}{}^2\rangle/\langle R_{0,n}{}^2\rangle - 1$ must vanish in the limit of a single segment ($n = 1$) whereas the function $(1 - t)/t \ln(1 + t)$ monotonically increases with decreasing t (approaching 1 for $t \rightarrow 0$). Clearly, the data for the long chains must leave the theoretical curve at lower values of t than the data of shorter chains. Since for very small n the microstructure of the simulation model becomes noticeable, the breakdown of the scaling cannot be described by crossover considerations.[28]

3.3. Ratio of the Radius of Gyration and End-to-End Distance. Finally, we determined the ratio $\aleph$ of the mean square radius of gyration $\langle S_N{}^2\rangle$ and the mean square end-to-end distance $\langle R_N{}^2\rangle$ in the limit $N \rightarrow \infty$, i.e. $\aleph = \lim_{N\rightarrow\infty} 6\langle S_N{}^2\rangle/\langle R_N{}^2\rangle$. This ratio is influenced by the nonuniform swelling of subchains and depends only on the type of chains under consideration.[1] Using a plot of $6\langle S_N{}^2\rangle/\langle R_N{}^2\rangle$ versus $1/N$, we estimated $\aleph = 0.84 \pm 0.01$ for $d = 2$ and $\aleph = 0.95 \pm 0.01$ for $d = 3$ in agreement with the values reported in the literature (see for example ref 36 and references therein). The obtained values can be compared with results of a third-order ϵ-expansion:[1] $\aleph = 1 - 0.010417\epsilon - 0.030628\epsilon^2 - 0.007152\epsilon^3$. This formula gives $\aleph = 0.799$ in two dimensions and $\aleph = 0.952$ in three dimensions. As in the previous sections in two dimensions the deviation is larger.

4. Conclusion

Using the bond-fluctuation model, the expansion of the subchains of a single excluded volume chain was investigated with profound quantitative simulation data in two and three dimensions.

It was found that the average subchain expansion factor $\alpha_n{}^2$ becomes maximal for $n < N$. This maximum and the shape of the curves are seen in plots of $\alpha_n{}^2$ versus n and cannot be explained by the blob model[11-13] or by modified blob models.[14-16] In three dimensions these observations are in qualitative agreement with the works of Curro[20] and Mattice.[22] We showed that this maximum is even more pronounced in two dimensions. Furthermore, we found that the ϵ-expansion results, eqs 2 and 3 of Duplantier[27] and of Schäfer and Baumgärtner,[29] can be used to obtain a quantitative description of $\alpha_n{}^2$.

In accord with qualitative evidence of other investigations[21-24] we found a position dependence of the subchain swelling. Studying a wide range of subchain lengths and subchain positions in two and three dimensions results in a detailed picture of subchain swelling: The subchains are more swollen than an isolated chain containing the same number of segments. This excess swelling is most pronounced in the center of the chain. We demonstrated that the excess swelling in two dimensions is about twice as much as that in three dimensions.

Further, we showed in detail that the theory of Duplantier[27] and of Schäfer and Baumgärtner[28] is in good agreement with our data for the excess swelling. We showed that the position dependence is well described by the theory over a wide range of the parameters t and u, particularly in three dimensions. Moreover, we demonstrated the chain length dependence of the excess swelling in both two and three dimensions.

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