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Critical exponents for simple non-uniform polymer networks

Dongming Zhao and Turab Lookman

Department of Applied Mathematics, University of Western Ontario, London, Canada
N6G 5B7

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Abstract. We study a number of non-uniform specified topologies and show rigorously that for certain topologies with cut edges, the critical exponent γ_t is in agreement with a conjecture given by Gaunt *et al* and that the exponent $\nu_t = \nu$, the exponent for self-avoiding walks. We also find that the scaling relations $\gamma_t - \gamma_t^1$ and $\gamma_t - \gamma_t^{11}$ are the same as for self-avoiding walks, previously conjectured only for uniform networks. By assigning an interaction energy to a nearest neighbour contact, we prove that the collapse transition for these topologies is the same as that for self-avoiding walks.

1. Introduction

The influence of topology on the critical exponent γ_t of polymer networks with the self-avoiding constraint has been previously investigated (Gaunt *et al* 1984b, Duplantier 1986). The number of configurations, G_N , of a network with a specified topology is expected to vary asymptotically as $e^{\kappa N} N^{\gamma_t - 1}$, where κ is the connective constant for self-avoiding walks and N is the total length. The results of scaling and renormalization theory for the critical exponent γ_t (Duplantier 1986, Duplantier and Saleur 1986) hold for the uniform or 'pseudo' uniform network for which the number of monomers, n in each of the self-avoiding chains is expected to be of $O(n)$. For the non-uniform case, in which finite chain lengths are allowed, Gaunt *et al* (1984b) have conjectured the form $\gamma_t = \gamma + b - 1$ for a network with b cut edges. (A cut edge of a graph is one which, if deleted, disconnects the graph). The conjecture is based on heuristic arguments and bounds and exact enumeration data for some simple topologies. In this work, we calculate rigorously critical exponents for a number of simple non-uniform networks given in figure 1. For the networks with cut edges (figures 1(a)–1(d)), we show that the subdominant exponent γ_t satisfies the conjecture $\gamma_t = \gamma + b - 1$. We also consider the exponent ν_t , characterizing the divergence of the mean-square end-to-end distance of a chain within these networks, and prove that it is equal to ν , the self-avoiding walk exponent. Similar results for the subdominant exponents hold for the half space problem if these topologies have their initial or both initial and terminal vertices attached to the surface. Consequently, we find that the scaling relations involving the bulk exponents and surface exponents are the same as for SAWs, previously conjectured only for uniform networks (Duplantier 1989, De'Bell, Lookman and Whittington 1990, De'Bell and Lookman 1993). By assigning an interaction energy to a nearest neighbour contact, we prove that the collapse transition for these topologies is the same as that for SAWs. For the figure-eight network (figure 1(e)), we show that the subdominant exponent ϵ is equal to

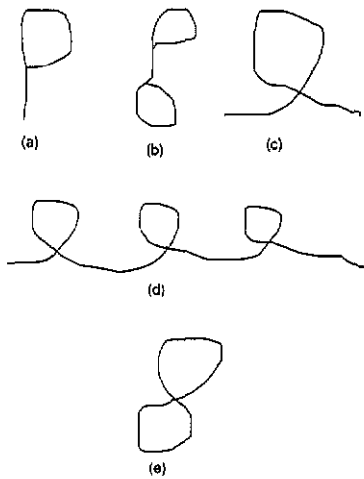


Figure 1. Examples of simple topologies: (a) tadpole, (b) dumbbell, (c) twin-tailed tadpole, (d) c twin-tailed tadpole, (e) figure-eight.

α , the exponent for polygons and that the collapse transition is the same as that for polygons.

The study of such networks is also related to the problem of lattice trails which are random walks on a lattice in which the edges are not allowed to overlap (Malakis 1975). Our results show that for trails in the honeycomb and Lavé lattices, in addition to the subdominant exponent γ (Guttmann 1985a), the exponent ν is also the same as that for SAWs. For trails in a hypercubic lattice, we show that for any trail with c cycles, which we term a c -trail, $\gamma_c = \gamma + c$ and $\nu_c = \nu$. These results for c -trails are analogues of the results $\theta_c = \theta_0 - c$ (Soteris and Whittington 1988) and $\nu_c = \nu_0$ (Zhao, Wu and Lookman 1992) for bond c -animals, where θ_c is the subdominant exponent and ν_c is the exponent for the mean-square radius of gyration for the c -animals; θ_0 and ν_0 are the corresponding exponents for bond trees.

In section 2 we consider a pattern theorem due to Kesten (1963) and obtain a corollary that we will use to investigate the specified topologies in section 3. In section 4, we discuss trails and c -trails. All of the models considered are embedded in the d -dimensional hypercubic lattice in which a vertex is a point in d -dimensional Euclidean space with integer coordinates $x = (x_1, \dots, x_d)$. The notations e_1, e_2, \dots, e_d will be used for the unit vectors.

2. Pattern theorem

For self-avoiding walks, a *pattern* P is a prescribed finite step self-avoiding walk and is said to *occur* in a self-avoiding walk W if it is part of W . It is said that P *occurs* r *times* in W if P appears at r distinct steps of W , and the vertices of two copies of P are disjoint or the terminal vertex of one is the initial vertex of another. P is called a *Kesten pattern* if there exists an m -step walk W where P appears more than twice in W .

Theorem 1 (Kesten 1963). If P is any Kesten pattern and $c_n(\varepsilon, \leq)$ is the number of n -step self-avoiding walks in which P occurs at most εn times, then there exists a

positive number $\varepsilon_0 > 0$ such that

$$\limsup_{n \rightarrow \infty} \frac{1}{n} \ln c_n(\varepsilon_0, \leq) < \kappa \quad (1)$$

where κ is the connective constant of self-avoiding walks.

A corollary which we need from the theorem is given as follows:

Corollary 1. Let P be a Kesten pattern. There exists a positive number ε_0 such that if $c_n(\varepsilon_0, >)$ is the number of n -step self-avoiding walks in which the pattern P appears more than $\varepsilon_0 n$ times, then

$$\lim_{n \rightarrow \infty} \frac{c_n(\varepsilon_0, >)}{c_n} = 1. \quad (2)$$

Proof. For any Kesten pattern P , from the theorem, there exist positive numbers $\varepsilon_0 > 0$, $\delta > 0$ and an integer of $N(\delta)$ such that for any $n > N(\delta)$, we have

$$0 < c_n(\varepsilon_0, \leq) < e^{(\kappa - \delta)n}. \quad (3)$$

Since

$$\frac{c_n(\varepsilon_0, >)}{c_n} = 1 - \frac{c_n(\varepsilon_0, \leq)}{c_n}. \quad (4)$$

By using (1), we obtain

$$1 - \frac{e^{(\kappa - \delta)n}}{c_n} < \frac{c_n(\varepsilon_0, >)}{c_n} < 1. \quad (5)$$

Taking limits yields

$$1 \leq \liminf_{n \rightarrow \infty} \frac{c_n(\varepsilon_0, >)}{c_n} \leq \limsup_{n \rightarrow \infty} \frac{c_n(\varepsilon_0, >)}{c_n} \leq 1. \quad (6)$$

This corollary can also be extended to other problems such as n -step self-avoiding polygons and n -step self-avoiding walks with certain geometrical constraints, provided that the connective constant is the same as that for self-avoiding walks.

3. Critical exponents for simple networks

3.1. Critical exponents in the bulk

We first consider tadpoles ($b = 1$). Let $c_n(r)$ and $t_n(r)$ be the number of n -step self-avoiding walks and tadpoles respectively with end-to-end distance r along the chain. If an edge incident at the branch point of degree 3 is deleted from the loop of a tadpole, we obtain an $(n - 1)$ -step SAW with end-to-end distance $r + \delta_0$ with $|\delta_0| \leq 1$. Therefore

$$t_n(r) \leq c_{n-1}(r + \delta_0) \quad (7)$$

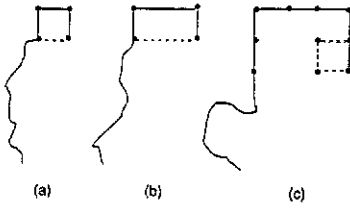


Figure 2. Conversion of a self-avoiding walk into a tadpole by adding a finite number of edges such as, one edge (a), two edges (b) and three edges (c).

and

$$t_n \leq c_{n-1}. \tag{8}$$

For an upper bound to the number of configurations for a tadpole, we note that at most 3 edges are needed to construct a tadpole from a SAW (figure 2). Thus

$$c_n(r) \leq t_{n+1}(r + \delta_1) + t_{n+2}(r + \delta_2) + t_{n+3}(r + \delta_3) \tag{9}$$

in which $-1 \leq \delta_1 \leq 1$, $-1 \leq \delta_2 \leq 2$ and $-1 \leq \delta_3 \leq 1$. Summing over r yields

$$c_n \leq 3t_{n+3}. \tag{10}$$

From equations (8) and (10), we have that

$$\lim_{n \rightarrow \infty} \frac{1}{n} \ln t_n = \lim_{n \rightarrow \infty} \frac{1}{n} \ln c_n = \kappa \tag{11}$$

and

$$\gamma_t = \gamma \tag{12}$$

results previously obtained by Guttman and Whittington (1978).

Since as $n \rightarrow \infty$, the mean-square end-to-end distance $\langle R_n^2 \rangle = \sum_r r^2 c_n(r) / c_n \sim n^{2\nu}$, we have that

$$\begin{aligned} & \frac{\sum_r r^2 t_{n+1}(r - \delta_0)}{3t_{n+3}} \\ & \leq \frac{\sum_r r^2 c_n(r)}{c_n} \\ & \leq \frac{\sum_r r^2 [t_{n+1}(r + \delta_1) + t_{n+2}(r + \delta_2)t_{n+3}(r + \delta_3)]}{t_{n+1}}. \end{aligned} \tag{13}$$

Using $1/9 \leq r^2 / (r + \delta_i)^2 \leq 4$, $i = 0, \dots, 3$, we obtain

$$\begin{aligned} & \frac{\sum_{r-\delta_0} (r - \delta_0)^2 t_{n+1}(r - \delta_0)}{27t_{n+3}} \\ & \leq \frac{\sum_r r^2 c_n(r)}{c_n} \leq \frac{4 \sum_{r+\delta_1} (r + \delta_1)^2 t_{n+1}(r + \delta_1)}{t_{n+1}} \\ & \quad + \frac{4 \sum_{r+\delta_2} (r + \delta_2)^2 t_{n+2}(r + \delta_2)}{t_{n+1}} \\ & \quad + \frac{4 \sum_{r+\delta_3} (r + \delta_3)^2 t_{n+3}(r + \delta_3)}{t_{n+1}} \end{aligned} \tag{14}$$

which implies that $\nu_t = \nu$.

The same arguments can be used to prove that the exponents γ_t and ν_t for dumbbells ($b = 1$) are the same as that for SAWs. However, for the upper bound of the dumbbell, care is needed to ensure that the edges added to form a loop at the end vertex of the tadpole do not interfere with the other loop of the tadpole. No assumptions are required for the leading behaviour (Guttmann and Whittington 1978).

Now we consider twin-tailed tadpoles ($b = 2$). All arguments and results for twin-tailed tadpoles can be extended to c twin-tailed tadpoles ($b = c + 1$) (figure 1(d)) in a straightforward manner. Let $s_n(r)$ be the number of n -step twin-tailed tadpoles with end-to-end distance r . Following the arguments of Gaunt *et al* (1984b), one can show that

$$s_n(r) \leq (n - 2)c_{n-2}(r) + nc_n(r) + nt_n(r) \leq 2nc_n(r) + nt_n(r). \tag{15}$$

From (7), we obtain

$$s_n(r) \leq 2nc_n(r) + nc_{n-1}(r + \delta_0). \tag{16}$$

and

$$s_n = \sum_r s_n(r) \leq \sum_r [2nc_n(r) + nc_{n-1}(r + \delta_0)] \leq 3nc_n. \tag{17}$$

We define a pattern P by

$$P = \{x + e_d, x + 2e_d, x + 2e_d + e_{d-1}, x + 2e_d + 2e_{d-1}, x + e_d + 2e_{d-1}, x + e_d + e_{d-1}, x + e_{d-1}x + 2e_{d-1}, x + 3e_{d-1}\} \tag{18}$$

(figure 3(a)). P is a finite self-avoiding walk and can occur more than twice in a SAW, for instance, it occurs three times in the SAW $W = PPP$ (figure 3(b)). Therefore, P is a *Kesten pattern*. (Note that such a pattern cannot occur in a neighbour avoiding walk. Thus, we see that μ , the connective constant for neighbouring avoiding walks is less than κ , previously conjectured by Gaunt *et al* 1984a). For such a pattern, by deleting the edges $[x + e_d, x + 2e_d]$ and $[x + 2e_d, x + 2e_2 + e_{d-1}]$ and adding the edges $[x + e_d, e_d + e_{d-1}]$ and $[x + e_d + e_{d-1}, x + e_d + 2e_{d-1}]$, we convert it into a finite twin-tailed tadpole (figure 3(c)). If W is an n -step self-avoiding walk in which the pattern P occurs more than ϵn times for some $\epsilon > 0$, we have at least $\binom{\epsilon n}{1} = \epsilon n$ ways to select one of them to convert W into an n -step twin-tailed tadpole. Under such a transformation, the positions of the initial and terminal vertices are not changed. Therefore, denoting by $c_n(\epsilon, >, r)$ the number of SAWs which have end-to-end distance r and on which P occurs more than ϵn times, we have

$$s_n(r) \geq (\epsilon n)c_n(\epsilon, >, r) \tag{19}$$

and

$$s_n = \sum_r s_n(r) \geq (\epsilon n) \sum_r c_n(\epsilon, >, r) = (\epsilon n)c_n(\epsilon, >). \tag{20}$$

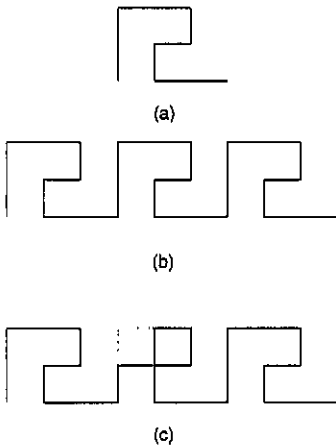


Figure 3. (a) the pattern P given by (18), (b) a walk with the pattern P occurring three times, (c) conversion of the walk in (b) into a twin-tailed tadpole at one pattern.

From (15), (16), (18) and (19), the mean-square end-to-end distance $\langle R_n^2 \rangle$ for twin-tailed tadpoles satisfies

$$\frac{(\varepsilon n) \sum_r r^2 c_n(\varepsilon, > r)}{3nc_n} \leq \frac{\sum_r r^2 s_n(r)}{s_n} \leq \frac{n \sum_r r^2 [2c_n(r) + c_{n-1}(r + \delta_0)]}{(\varepsilon n)c_n(\varepsilon, >)} \tag{21}$$

From theorem 1 and corollary 1, there exists $\varepsilon_0 > 0$ such that

$$\lim_{n \rightarrow \infty} \frac{c_n(\varepsilon_0, >)}{c_n} = 1 \tag{22}$$

and

$$0 \leq \lim_{n \rightarrow \infty} \frac{\sum_r r^2 c_n(\varepsilon_0, \leq, r)}{c_n} \leq \lim_{n \rightarrow \infty} \frac{n^2 c_n(\varepsilon_0, \leq)}{c_n} = 0. \tag{23}$$

Therefore, from (19) and (21), we obtain that for sufficiently large n

$$s_n \geq (\varepsilon_0 n) c_n \tag{24}$$

which, combining with (16) yields

$$\gamma_t = \gamma + 1. \tag{25}$$

From (23), (24) and that $r^2/(r + \delta_0)^2 \leq 4$, we obtain that for sufficiently large n , equation (22) becomes

$$\begin{aligned} \frac{(\varepsilon_0 n) \sum_r r^2 c_n(r)}{3nc_n} &\leq \frac{\sum_r r^2 s_n(r)}{s_n} \\ &\leq \frac{n[2 \sum_r r^2 c_n(r) + 4 \sum_{r+\delta_0} (r + \delta_0)^2 c_{n-1}(r + \delta_0)]}{(\varepsilon_0 n)c_n} \end{aligned} \tag{26}$$

which implies that the exponent ν characterizing the divergence of the mean-square end-to-end distance $\langle R_n^2 \rangle$, is the same for both self-avoiding walks and twin-tailed tadpoles.

If the terminal vertex of a twin-tailed tadpole coincides with its initial vertex, a figure-eight topology is obtained. Let p_n and e_n be the number of n -step polygons and figure eights respectively. We have that

$$\lim_{n \rightarrow \infty} \frac{1}{n} \ln p_n = \lim_{n \rightarrow \infty} \frac{1}{n} \ln e_n = \kappa. \quad (27)$$

We also assume the asymptotic forms

$$p_n \sim n^{\alpha-2} e^{n\kappa} \quad e_n \sim n^{\epsilon-1} e^{n\kappa}. \quad (28)$$

By applying the pattern theorem and the corollary to polygons on which the pattern P occurs more than $\epsilon'_0 n$ times for some $\epsilon'_0 > 0$, we obtain

$$e_n \geq (\epsilon'_0 n) p_n. \quad (29)$$

Guttmann and Whittington (1978) have shown that

$$e_n \leq n p_n. \quad (30)$$

These two inequalities yield

$$\epsilon = \alpha. \quad (31)$$

3.2. Critical exponents in half space

Let s_n^I and s_n^{II} be the number of n -step twin-tailed tadpoles which are confined to one side of an impenetrable surface $x_1 = 0$ and are attached to the surface by the initial vertex and by both the initial and terminal vertices respectively. The connective constant for SAWs is still κ in the half space, and the pattern P of figure 2(a) still occurs in SAWs restricted in this way. The procedure for the bounds can then be followed without change from the bulk case leading to

$$\lim_{n \rightarrow \infty} \frac{1}{n} \ln s_n^I = \lim_{n \rightarrow \infty} \frac{1}{n} \ln s_n^{II} = \kappa \quad (32)$$

and

$$\gamma_t^I = \gamma^I + 1 \quad \gamma_t^{II} = \gamma^{II} + 1 \quad (33)$$

where γ^I and γ^{II} are the exponents for the corresponding self-avoiding walks respectively. Combining equations (24), (25) and (32), we obtain

$$\gamma_t + \nu - 2\gamma_t^I + \gamma_t^{II} = \gamma + \nu - 2\gamma^I + \gamma^{II} = 0 \quad (34)$$

$$\gamma_t - \gamma_t^I = \gamma - \gamma^I \quad (35)$$

$$\gamma_t - \gamma_t^{II} = \gamma - \gamma^{II}. \quad (36)$$

These results can also be obtained for tadpoles and dumbbells. Thus, these simple non-uniform topologies satisfy the scaling forms for *uniform* networks and support the conjecture that $\gamma(G) - \gamma^I(G)$ and $\gamma(G) - \gamma^{II}(G)$ are independent of G , the topology of the network (Duplantier 1989, De'Bell *et al* 1990, De'Bell and Lookman 1993).

3.3. The collapse transition

The interaction of polymer networks with an adsorption surface (Zhao and Lookman 1991, Soteris 1992) shows that the adsorption transition is independent of the topology. However, few results exist on the influence of topology on the collapse transition, where an interaction energy is assigned to any two vertices in the network forming a nearest neighbour contact. (An ϵ expansion study is given by Duplantier 1989). More or less trivial results are that tadpoles and dumbbells have the same collapse transition as do self-avoiding walks, which are obtained directly from the arguments in subsection 3.1. The same result can also be obtained for twin-tailed tadpoles as follows:

We define the partition functions

$$C_n(\alpha) = \sum_m c_{n,m} e^{m\alpha} \quad S_n(\alpha) = \sum_m s_{n,m} e^{m\alpha} \quad (37)$$

where $c_{n,m}$ and $s_{n,m}$ are the number of n -step self-avoiding walks and twin-tailed tadpoles with m nearest neighbour contacts respectively and α is the interaction energy between any two vertices forming a nearest neighbour contact.

We note that the procedure of Gaunt *et al* (1984b) to convert a twin-tailed tadpole into a SAW results in only a finite change in the number of nearest neighbour contacts. Therefore, we obtain for the upper bound

$$S_n(\alpha) \leq 3n f(\alpha) C_n(\alpha) \quad (38)$$

in which the function $f(\alpha)$ is independent of n . On the other hand, by adding a 4-cycle or 5-tadpole at the top vertex of a SAW, we convert the SAW into a twin-tailed tadpole, which gives

$$C_n \leq g(\alpha) S_{n+5}(\alpha). \quad (39)$$

From these two equations, one can conclude that twin-tailed tadpoles have the same collapse transition as do SAWs, and

$$\gamma(\alpha) \leq \gamma_t(\alpha) \leq \gamma(\alpha) + 1. \quad (40)$$

However, we believe that

$$\gamma_t(\alpha) = \gamma(\alpha) + 1. \quad (41)$$

Similarly, one can show that the collapse transition for the figure-eight is the same as for the polygon.

4. Lattice trails

The problem of lattice trails, introduced by Malakis (1975), is an interesting generalization of the self-avoiding walk. Of interest is the question of whether trails and SAWs belong to the same universality class (Guttmann 1985a, b, Lim and Meirovitch 1989). On the honeycomb and Lavé lattices, which have coordination number 3, trails consist only of SAWs and tadpoles (Guttmann 1985a). We note that

the method we have used for the tadpoles and dumbbells for γ and ν also works for these two lattices. Therefore, we have shown that in addition to γ (Guttmann 1985a), the exponent ν for trails is also the same as for SAWs. Moreover, the collapse transition induced by the nearest neighbour interaction is also the same as for SAWs.

For lattices with coordination number greater than 3, analogous to lattice c -animals (Whittington *et al* 1983), we consider lattice c -trails (trails with c cycles) as a model to study the crossover from SAWs to trails. Thus, tadpoles and twin-tailed tadpoles are 1-trails, whereas dumbbells and figure-eights are 2-trails. Let $s_n(c)$ be the number of n -step c -trails. Its lower bound is that for the c twin-tailed tadpoles, for which the subdominant exponent is $\gamma + c$. The upper bound is obtained by essentially following the procedure of Gaunt *et al* (1984b) to decrease successively the degree of the branch points until they are one or two, which yield $s_n(c) \leq (3n)^c c_n$. Thus, for the c -trails, $\gamma_c = \gamma + c$. Similarly, one can show that $\nu_c = \nu$ and that the scaling relations (34)–(36) also hold for the c -trails. The same arguments for a lower bound for c -trails leads to the result that, there exists some $\varepsilon_0 \geq 0$, such that

$$T_n = \sum_c s_n(c) \geq \sum_{c=0}^{\varepsilon_0 n} s_n(c) \geq \sum_{c=0}^{\varepsilon_0 n} \binom{\varepsilon_0 n}{c} c_n = 2^{\varepsilon_0 n} c_n \quad (42)$$

where T_n is the number of n -step trails. This implies that the connective constant for trails is greater than κ , previously shown by Guttmann (1985b) by other means.

5. Summary

By studying specified topologies such as tadpoles, dumbbells and twin-tailed tadpoles, we may conclude that adding a loop to one end of a self-avoiding chain (that may be part of a network) leaves the exponent γ unchanged. If the loop is added at a vertex of degree two, γ for the chain or network is increased by one. For polygons, adding a loop at any vertex increases $\alpha - 1$ by one. We have also shown that for tadpoles, dumbbells and twin-tailed tadpoles the exponent ν and the collapse transition with nearest neighbour interaction are also the same as SAWs. Moreover, we note that the relations $\gamma_t - \gamma_t^1$ and $\gamma_t - \gamma_t^{11}$ for these non-uniform topologies have the same values as SAWs, a conjecture previously made only for uniform networks.

Acknowledgments

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