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Competition between self-attraction and adsorption in branched polymers situated on a fractal lattice

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Abstract. We have studied the branched polymer model, situated on the Sierpinski gasket lattice, with competing monomer–monomer and monomer–surface interactions. We have determined critical properties of this model within an exact renormalization group approach. Our results reveal that the corresponding model system possesses a rich phase diagram that is typical for polymer surface physics.

Statistical properties of polymers in the vicinity of a surface have been intensively studied for both practical and theoretical reasons [1, 2]. Linear chain polymers have been considered in most of the respective theoretical studies [3]. Lam and Binder [4] were, to our knowledge, the first who studied adsorption of branched polymers at surfaces. Using the Monte Carlo and scaling approaches, they found that there is much similarity in the behaviour of linear and branched polymers in the presence of an adsorbing substratum. Branched polymers appear to be difficult to study theoretically and it is hardly possible to achieve a meaningful exact solution. In this paper, we study the standard lattice animal model of branched polymers situated on the Sierpinski gasket (SG) whose one boundary represents an attractive surface (see figure 1). Here, besides the surface–polymer interaction, we assume the presence of polymer self-attraction, in order to model the behaviour of a real polymer in a solvent. We have been able to solve this problem exactly within the renormalization group (RG) approach. In spite of the simplicity of the entire model, the results obtained demonstrate that the corresponding system displays rich critical behaviour that is typical for surface physics [5].

In order to explore the full phase diagram of the polymer system, we introduce the three Boltzmann factors— $w = \exp(-\epsilon_1/T)$, $v = \exp(-\epsilon_2/T)$ and $t = \exp(-\epsilon_3/T)$ —that correspond to the three interaction parameters ϵ_1 , ϵ_2 and ϵ_3 . The effect of a poor solvent is taken into account through the attractive interaction energy $\epsilon_1 < 0$ between two adjacent links (monomers) of a polymer configuration (see figure 1). Similarly, we associate the attractive energy $\epsilon_2 < 0$ with each monomer lying on the adsorbing surface. Finally, t refers to the monomer–surface interaction for those monomers that are present in the layer contiguous to the surface (see figure 1). The thermal and geometrical properties of the model system can be deduced from the generating function $G(x, w, v, t) = \sum \Omega(N, M, K, L)x^N w^M v^K t^L$, where $\Omega(N, M, K, L)$ is the number of all different configurations (per lattice site) having N monomers, M of which are the nearest neighbours that contribute to the self-interaction energy $M\epsilon_1$ of the system (see figure 1)

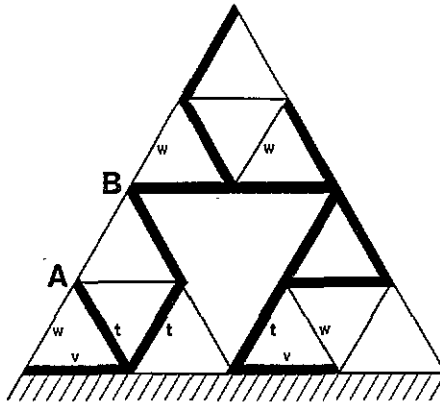


Figure 1. A schematic representation of a branched polymer configuration, situated on the SG fractal, in the vicinity of an adsorbing surface (the crosshatched strip). The monomers are depicted by the solid links, and their mutual interaction is weighted by the Boltzmann factor w . For simplicity, we restrict this interaction to links within the first-order triangles of the SG fractal. Accordingly, for instance, the AB monomer-monomer interaction is not present in our model. However, it turns that this restriction does not affect the scaling laws that describe geometry and thermal properties of the polymer system [8, 9]. The labels t and v denote the Boltzmann weighting factors of the monomer-surface interactions.

while K and L represent the number of monomers lying on the surface and the number of monomers lying in the layer next to the surface, respectively. Here x denotes the standard monomer fugacity parameter. As in the case of regular lattices, we assume that the function $G(x)$, for a given temperature T , has a power-law singularity $G(x) \sim (1 - \mu x)^{\theta-1}$, which sets in when x approaches the critical value $x_c = 1/\mu(T)$ from below. The critical exponent θ can take distinct values in different temperature regions, while the connectivity constant μ can be related to the polymer free energy per site through the relation $f(T) = -T \ln \mu(T)$.

It may be expected that the essential physics of the model described can be summarized by the phase diagram depicted in figure 2. Indeed, the physical behaviour is determined by the competition of the two interaction parameters w and v . When both the monomer-monomer and surface-monomer interactions are small ($w \sim 1$, $v \sim 1$), one can presume that the polymer resides in the extended states (random animal states). For relatively small values of v , increasing w should promote the appearance of collapsed branched polymer states (with a finite monomer density). On the other hand, increasing v , for a given w , should favour the binding of monomers to the surface (characterized by a finite fraction of adsorbed monomers, $M/N \neq 0$). The regions in the phase diagram, corresponding to the three different polymer states, should be separated by phase boundaries that can be assumed to meet at a multi-critical point (see figure 2). Our exact RG approach confirms the physical expectations arising from previous studies of linear polymers on fractal [6] and regular [7] lattices.

The generating function $G(x, w, v, t)$ can be expressed in terms of a finite number of restricted generating (partition) functions, whose definitions and recursion relations we are going to present in this paragraph. The schematic representations of the sixteen requisite restricted partition functions, that represent different polymer configurations, are given in figure 3. One should notice that the presence of the adsorbing wall necessitates the introduction of ten additional parameters (partition functions) in comparison with the case

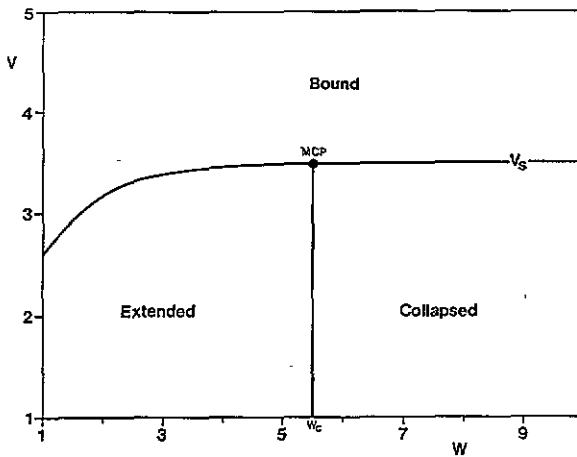


Figure 2. The phase diagram of a branched self-attracting polymer situated on the SG fractal, with one attractive boundary, in the space of the interaction parameters w and v . The two bulk phases (extended and collapsed) are separated from the bound-to-surface phase by the line $v = v_s$. This line merges with the extended-collapsed phase boundary ($w = w_c$) at the multi-critical point.

in which one studies only the polymer bulk properties [8]. To write down the recursion relations for different parameters one has to look for all possible ways in which a given polymer configuration of the $(r + 1)$ th-order fractal structure may be constructed from all possible polymer configurations that appear within the r th-order structure. Using the prime symbol as a superscript for the $(r + 1)$ th-order parameters and no indices for the r th-order parameters, we present the following set of recursion relations

$$A' = A(1 + 2B + 2B^2) + 2CB^2 + F(A^2 + B^2 + 2BD) \tag{1}$$

$$B' = B^2 + B^3 + BF(2A + 4C) + F^2(B + D) \tag{2}$$

$$C' = B^2(A + 3C) + F(2BD + 7C^2) + F^2(C + E) \tag{3}$$

$$D' = A^2(1 + 2B) + B(2D + 4AC + 3BD + 6C^2) + 2F(AD + BC + 2CD + BE) \tag{4}$$

$$E' = A^3 + 14C^3 + B(6AD + 12CD + 3BE) + F(3D^2 + 3C^2 + 12CE) \tag{5}$$

$$F' = F(3B^2 + 6CF + F^2) \tag{6}$$

$$A'_1 = A + 2BA_1 + 2BB_2A_2 + 2CB_2^2 + FB_2^2 + FA_1^2 + 2FB_2D_1 \tag{7}$$

$$A'_2 = A_2 + B_1A_2 + AB_2 + AB_1B_2 + BB_2(A_1 + C_1 + C_2 + F_1) + AA_2F_1 + DB_2F_1 + BF_1D_1 \tag{8}$$

$$B'_1 = B_1^2 + BB_2^2 + 2AB_1F_1 + 2BF_1(C_1 + C_2) + F_1^2(B + D) \tag{9}$$

$$B'_2 = BB_2(1 + B_1) + BF_1A_2 + FB_2(A_1 + C_1 + C_2) + 2CF_1B_2 + FF_1(B_2 + D_1) \tag{10}$$

$$C'_1 = AB_1^2 + CB_2^2 + 2B_1(BC_1 + DF_1) + FC_1^2 + CF_1(2C_2 + 4C_1 + F_1) + EF_1^2 \tag{11}$$

$$C'_2 = BB_2A_2 + 2BB_1C_2 + CB_2^2 + BF_1D_2 + FB_2D_1 + 4CF_1C_2 + F(2C_1C_2 + C_2^2 + F_1C_2 + F_1E_1) \quad (12)$$

$$D'_1 = (AA_2 + BD_1)(1 + B_1) + BA_1A_2 + DB_2 + BC_1A_2 + 2CB_2A_1 + BA_2C_2 + DB_1B_2 + BB_2D_2 + 2CC_1B_2 + 4CC_2B_2 + DA_2F_1 + FA_1D_1 + FC_2B_2 + CF_1B_2 + 2CF_1D_1 + FC_2D_1 + FC_1D_1 + EF_1B_2 + FE_1B_2 \quad (13)$$

$$D'_2 = A_2^2 + 2AA_2B_2 + 2B_1D_2 + 4AB_1C_2 + DB_2^2 + 2BB_2D_1 + 2BC_2^2 + 4BC_1C_2 + 2AF_1D_2 + 2BF_1C_2 + 4DF_1C_2 + 2BF_1E_1 \quad (14)$$

$$E'_1 = 4DB_1C_2 + 2BD_2(C_2 + C_1) + 4CB_2D_1 + 2BB_1E_1 + EB_2^2 + 2DF_1D_2 + FD_1^2 + 2CF_1C_2 + FC_2^2 + 4EF_1C_2 + 2CE_1F_1 + 2FE_1(C_1 + C_2) + 2CF_1E_1 + AA_2^2 + 6CC_2^2 + 8CC_1C_2 + 2BA_2D_1 + 2AB_1D_2 + 2DA_2B_2 \quad (15)$$

$$F'_1 = FB_2^2 + 2BB_1F_1 + 2FF_1(C_1 + C_2) + 2CF_1^2 + FF_1^2. \quad (16)$$

The above set of exact relations can be considered the RG transformations for the system under study. To perform the requisite numerical analysis, it must be supplemented by the appropriate initial conditions. In general, for this purpose we could adopt a set of sixteen independent parameters, with sixteen particular values, in order to explore the entire available phase space. However, to describe the simple adsorption of branched polymers, we find that the initial conditions (that correspond to the first-order SG fractal) can be expressed in terms of only four independent parameters

$$\begin{array}{llll} A = 1 & E = w^3 & B_1 = xv & D_1 = w \\ B = x & F = x^2(x + 3w) & B_2 = xt & D_2 = w \\ C = xw^2 & A_1 = 1 & C_1 = xvw^2 & E_1 = w^3 \\ D = w & A_2 = 1 & C_2 = xt w^2 & F_1 = x^2 t(xvt + tw + 2vw) \end{array} \quad (17)$$

where, for the sake of simplicity, we have omitted the superscript (1) associated with these first-order restricted generating functions. The above expressions spring from the possible polymer configurations which can be realized within the fractal unit triangle. For instance, $B_1 = xv$ is the weight of that part of a polymer branch that traverses a unit triangle along its edge that lies on the attractive impenetrable wall, whereas $B = x$ is the weight of the similar configuration within the bulk.

Numerical analysis of RG transformations (1)–(16) will reveal that there exist many fixed points that cannot be approached from the initial conditions (17), and, consequently, we have not found them to be relevant to the system studied (although, they might be of interest in the case of some similar model systems; for instance, one can choose initial conditions which would make it possible to reach fixed points that are relevant to the linear polymer problem). Besides, numerical analysis shows that it is necessary to assume that $t < 1$, since otherwise the polymer system always appears to be bound to the surface, as in the case of linear polymers [6]. This may be explained in terms of two competing factors (energy and entropy) in the corresponding free energy of adsorption. In a regular lattice, adsorption is accompanied with a gain in the internal energy and a loss in the configurational entropy of the polymer in the vicinity of the impenetrable wall. However, in the case of fractals

numerous impenetrable walls are present (due to holes of all sizes) in the bulk as well, and the corresponding loss in entropy is of the same order as the loss caused by the adsorbing boundary wall. This brings about the situation in which an attractive boundary wall makes the energy term dominant, and thereby the polymer chain appears to be always adsorbed. Under these circumstances, a simple way to promote the appearance of the desorbed state is to introduce the repulsive part ($\epsilon_3 > 0$, $t < 1$) of the monomer-wall interaction. In what follows we separate our discussion into three parts, according to the possible values of the monomer-monomer interaction parameter w (keeping $t < 1$ fixed).

(i) The first part of our discussion concerns the weak monomer-monomer interactions ($w \sim 1$), which corresponds to the swollen polymer phase [8]. To judge the adsorption state of the polymer, we need to know the ratio M/N , which can be expressed in terms of the critical fugacity $x_c = x_c(w, v, t)$. The requisite expression is of the form [6]

$$\frac{M}{N} = -\frac{v \, dx_c}{x_c \, dv} \tag{18}$$

This formula allows us to find M/N by calculating x_c , that is found as the maximal value of x below which iterations of (1)–(16) still converge. In this way, for small v , we have found that M/N vanishes, whereas when v increases we cross the boundary v_s above which the adsorption order parameter M/N takes finite values (see figure 2). For v slightly larger than v_s , our calculations confirm the following scaling law

$$M \sim N^\phi \tag{19}$$

where ϕ is the crossover exponent of the surface [3].

The critical properties of the polymers are described by three fixed points that correspond to the three consecutive cases of the surface-monomer interaction: $v < v_s$, $v = v_s$ and $v > v_s$. The iterations of the set (1)–(16), for $x = x_c(w, v, t)$, reveal that some of the restricted partition functions diverge. By exploring numerically the mode of their divergence (see, for example, [8]), we have found it convenient to introduce the change of variables

$$\begin{array}{llll} x_1 = AF & x_2 = B & x_3 = CF & x_4 = DF^2 \\ x_5 = EF^3 & x_6 = F & x_7 = B_1 & x_8 = B_2 \\ x_9 = F_1/F & x_{10} = A_1F & x_{11} = A_2F & x_{12} = C_1F \\ x_{13} = C_2F & x_{14} = D_1F^2 & x_{15} = D_2F^2 & x_{16} = E_1F^3 \end{array} \tag{20}$$

Of course, there are many other changes of variables that move from the divergence of the RG parameters. Without rewriting our exact RG transformations in terms of (20), we quote the appropriate findings. In the case of weak monomer-monomer interactions ($w < w_c$, $w_c = 5.485117$), for $v < v_s$ and for a fixed $t < 1$, the relevant fixed point has five coordinates different from zero ($x_1^* = 0.200870$, $x_2^* = 0.327604$, $x_3^* = 0.023100$, $x_4^* = 0.023238$, $x_5^* = 0.002198$), and eleven equal to zero. This fixed point describes the extended states of branched polymer, with the single relevant eigenvalue $\lambda^{(E)} = 2.630927$ and the concomitant critical exponent $\nu = \ln 2 / \ln \lambda^{(E)} = 0.716552$ for the gyration radius. These results are in agreement with those obtained in the study [8] of polymer bulk properties only.

When the surface-monomer interaction parameter v increases, with t kept fixed, the critical fugacity x_c does not change as long as v does not reach the phase boundary value

v_s that corresponds to a given w . At $v = v_s$, and $x = x_c$, we find the tricritical 'symmetric' fixed point: $x_1^* = x_{10}^* = x_{11}^* = 0.200\ 870$, $x_2^* = x_7^* = x_8^* = 0.327\ 604$, $x_3^* = x_{12}^* = x_{13}^* = 0.023\ 100$, $x_4^* = x_{14}^* = x_{15}^* = 0.023\ 238$, $x_5^* = x_{16}^* = 0.002\ 198$, $x_6^* = 0$, $x_9^* = 1$. The linearized RG transformations have two relevant eigenvalues at this fixed point. The larger eigenvalue $\lambda^{(E)}$ coincides with the one already found in the bulk case ($v < v_s$), and thereby the corresponding critical exponent ν remains unchanged. The smaller eigenvalue, $\lambda_s^{(E)} = 1.828\ 169$, determines (see, for example, [5]) the crossover exponent ϕ and the specific heat exponent α

$$\phi = \frac{\ln \lambda_s^{(E)}}{\ln \lambda^{(E)}} = 0.623\ 69 \quad \alpha = 2 - \frac{1}{\phi} = 0.396\ 64. \quad (21)$$

Interestingly enough, this result lies within the limits, found phenomenologically [6] for linear polymers on fractal lattices in the presence of an adsorbing surface, $1 - \nu(d_f - d_s) \leq \phi \leq d_s/d_f$, where d_f and d_s are, respectively, the fractal dimensions of the lattice and surface (in our case: $d_f = \ln 3/\ln 2$ and $d_s = 1$).

Finally, for $v > v_s$, the polymer is globally adsorbed and we can expect that its critical properties will be described by a self-avoiding walk on a line. Indeed, in this case, we find the fixed point with $x_7^* = 1$ and $x_i^* = 0$ ($i \neq 7$), which means that only the surface polymer correlation function is different from zero (see (9) and (20), and figure 3). Accordingly, there is only one relevant eigenvalue $\lambda^{(B)} = 2$, implying $\nu = 1$.

(ii) For $v < v_s$, increasing w brings about the appearance of the collapsed polymer phase beyond the line $w = w_c = 5.485117$ (see figure 2), which is characterized by a non-zero bulk monomer density. The critical properties of this phase are determined by the fixed point with coordinates $x_3^* = \sqrt{15}/30$, $x_5^* = 1/20$, and $x_i^* = 0$, for $i \neq 3, 5$. At this fixed point, there is only one relevant eigenvalue $\lambda^{(C)} = 3$, that leads to the critical exponent ν being equal to $1/d_f$, which is in agreement with [8]. In addition to the critical exponent ν , we quote here our result for the critical exponent for the generating function $G(x)$, $\theta = 1.535\ 026$. This ($\theta > 1$) implies that $G(x)$ is singular, but not divergent, for $x \rightarrow x_c$.

In the cases when w is fixed so that $w > w_c$, increasing ν leads the system to a line of points of continuous phase transitions, defined by $v = v_s$. For $v > v_s$, the system is in a bound state whose properties are of the type already discussed in this paper. However, at $v = v_s$, the behaviour of the system is governed by a new 'symmetric' fixed point, with seven non-zero coordinates $x_3^* = x_{12}^* = x_{13}^* = \sqrt{15}/30$, $x_5^* = x_{16}^* = 1/20$, $x_7^* = 1$, $x_9^* = 1$. The two largest eigenvalues at this fixed point are $\lambda^{(C)} = 3$ and $\lambda_s^{(C)} = 2$, so that the critical exponent of the gyration radius is the same as in the collapsed bulk phase $\nu = \ln 2/\ln 3 = 1/d_f$, whereas the critical exponent of the crossover ϕ , having the same value as ν , coincides with the upper bound suggested by Bouchaud and Vannimenus [6]. However, in contrast to the linear polymer case [6], we have found no segment of the line $v = v_s$ that corresponds to first-order phase transitions.

(iii) The final section of our discussion of the model phase diagram concerns the line $w = w_c$ and its merging with the line $v = v_s$. The two lines merge at the multi-critical point, located at $w = w_c$ and $v_s^{(c)} = 2.225\ 427$ (see figure 2). For $v < v_s^{(c)}$, the bulk collapse transition is controlled by the tricritical fixed point: $x_1^* = 0.059\ 164$, $x_2^* = 0.075\ 014$, $x_3^* = 0.125\ 260$, $x_4^* = 0.022\ 503$, $x_5^* = 0.047\ 351$ and $x_i^* = 0$ for $i \geq 6$. Its relevant eigenvalues are $\lambda_1^{(c)} = 2.991\ 818$ and $\lambda_2^{(c)} = 1.199\ 411$, which brings about a critical exponent of the gyration radius $\nu_c = 0.632\ 50$ and the corresponding crossover exponent $\phi_c = 0.165\ 92$, in agreement with the previous study for $\nu = 1$ [8]. Furthermore, we have calculated the

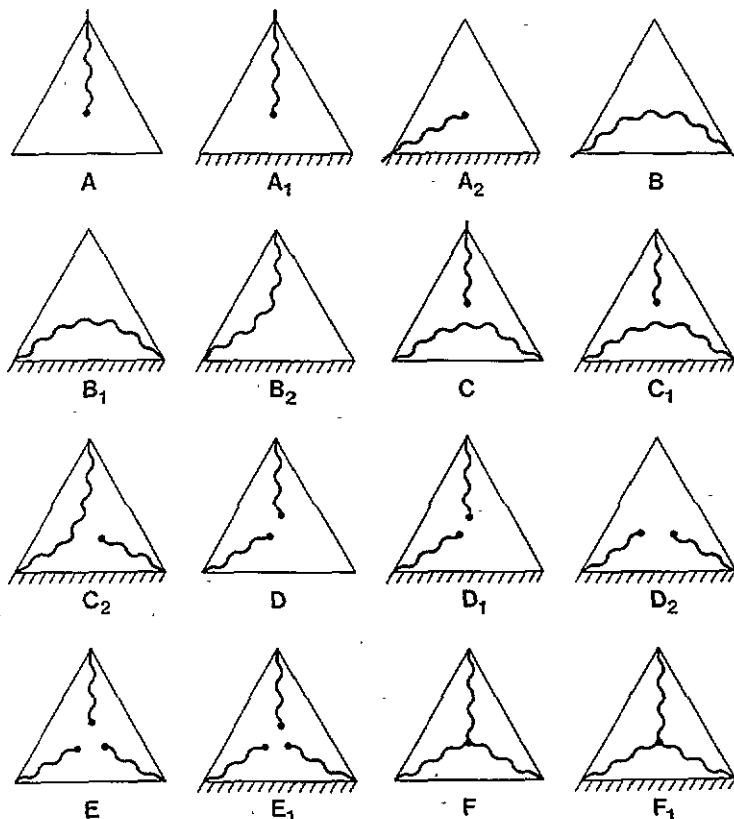


Figure 3. Diagrams representing the sixteen restricted partition functions used to describe the statistics of a branched polymer situated on the SG fractal in the vicinity of an adsorbing substratum. The triangles depict the SG r th-order triangle (the $r = 1$ order corresponds to the unit triangle, whereas the complete fractal lattice follows in the limit $r \rightarrow \infty$). The parameter F_1 , for instance, represents the polymer configuration in the case when all three triangle vertices are connected, in contrast to the situation described by E_1 when the vertices are occupied but not mutually connected. In order to clarify definitions of the restricted partition functions, we observe that the configuration depicted in figure 1 is of the $B_2^{(3)}$ type and its weight is given by $F^{(2)}(B_2^{(2)})^2$.

corresponding critical exponent of the generating function $\theta_c = 1.521799$, which, as in the collapsed phase case, implies that $G(x)$ is singular, but not divergent, for $x \rightarrow x_c$.

The final point of the phase diagram to be discussed is related to the special pair of values ($w = w_c$ and $v = v_s^{(c)}$) of the interaction parameters. For these parameters, the RG transformations iterate towards the symmetric 'adsorbed—collapsed' fixed point: $x_1^* = x_{10}^* = x_{11}^* = 0.059164$, $x_2^* = x_7^* = x_8^* = 0.075014$, $x_3^* = x_{12}^* = x_{13}^* = 0.125260$, $x_4^* = x_{14}^* = x_{15}^* = 0.022503$, $x_5^* = x_{16}^* = 0.047351$, $x_6^* = 0$ and $x_9^* = 1$. This is in contrast to the linear polymer case [6], where the corresponding 'adsorbed—collapsed' fixed point appears to be non-symmetrical. In our case, the linearized RG transformations have two relevant bulk eigenvalues $\lambda_1^{(\ominus)} = 2.991818$ and $\lambda_2^{(\ominus)} = 1.199411$, and one additional surface eigenvalue $\lambda_s^{(\ominus)} = 1.995542$. Thus, the critical exponent of the gyration ν , at this multi-critical fixed point, is, once again, determined by the largest bulk eigenvalue, that is $\nu = \nu_c = \ln 2 / \ln \lambda_1^{(\ominus)} = 0.63250$. We calculate the crossover exponent using the

standard formula [5], $\phi = \ln \lambda_s^{(\odot)} / \ln \lambda_1^{(\odot)}$, whereby we find $\phi = 0.630466$. This result we confirmed by direct numerical calculation of the adsorption order parameter which behaves according to the power law $M/N \sim (v_s - v)^{(1-\phi)/\phi}$. It is interesting to note that the latter method was the only one by which ϕ was estimated in the case of linear polymers, as the standard formula turned out to be inapplicable [6]. In addition, we observe that our result $\phi = 0.630466$ appears to be just slightly below the upper bound $1/d_f = 0.630930$ proposed in the same study of linear polymers [6].

In conclusion, we have presented an exact study of branched polymers, situated on a fractal lattice, with competing monomer–monomer and monomer–surface interactions, which caused the simultaneous appearance of collapse and adsorption transitions. Most of our findings are in qualitative agreement with results found in the case of linear polymers [6, 7]. In particular, the general structure of the branched-polymer phase diagram is similar to the corresponding structure obtained for linear polymers. However, there are still some differences. For example, in contrast to the linear polymer case [6, 7], we have found that the collapse transition does not noticeably lower the adsorption temperature (see figure 2). Furthermore, we found that the multi-critical fixed point is symmetric, whereas in the study of linear polymers [6] it was emphasized that the corresponding point should be asymmetric. The specific cause of the observed differences is not clear. Of course, if there was evidence that such differences do not exist in Euclidean lattices, we could ascribe them to the peculiarities of the underlying fractal lattices. However, we would like to speculate on the fact that, in our study, the polymer in the adsorbed state does not retain the topological structure it had in the bulk state, since we found that the bound phase is described by the fixed point for a one-dimensional self-avoiding walk. To vindicate this speculation, it would be desirable to perform a similar study for the three-dimensional Sierpinski gasket, in which case one could also expect a richer phase diagram.

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