

## Interpenetration and segregation of interacting polymer chains in a solution: Exact results on fractal lattices

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A model of two interacting self-attracting, self-avoiding walks is proposed to study the critical behavior of two interacting chemically different linear polymers in a solution that may have different qualities for different chains. We solve the model exactly on truncated  $n$ -simplex lattices for  $4 \leq n \leq 6$  using the real-space renormalization-group transformation. Depending upon the solvent quality, the temperature, and the attractive interactions between interchain and intrachain monomers, the configuration of either segregation or interpenetration or zipping of chains may arise. It is shown that these configurations correspond to different fixed points of the renormalized-group transformation. The value of the contact exponent is calculated exactly at the tricritical points corresponding to the segregation-interpenetration and the interpenetration-zipped-state chain transitions. The phase boundaries of these states are shown on a plane of fugacity weight attached with a zipped step (i.e., a step in which both walks move side by side) and the Boltzmann factor associated with the attraction between unlike monomers. The phase diagram is shown to have different universality domains of critical behavior.

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### I. INTRODUCTION

The physical properties observable on a polymer chain length are calculated as statistical averages over all possible configurations of the polymers, and these configurations are obtained by mapping the polymer chain onto a walk embedded in an appropriate lattice [1–3]. Depending upon the physical situations, appropriate geometrical restrictions are imposed on these walks. For example, a model of a self-avoiding walk (SAW) simulates a polymer chain in a good solvent, while the model of a self-attracting self-avoiding walk (SASAW) represents a polymer chain in a poor solvent that can undergo a collapse transition when the chain contracts from an extended state to a globule state when the temperature is lowered [4–6].

Quantities of interest in the case of the one-chain problem are the number of distinct walks  $C_N$  of  $N$  steps starting at the same origin, the number of closed loops  $P_N$  of  $N$  steps, and the mean radius of gyration  $\langle R^2 \rangle^{1/2}$ . All these configurational properties of a polymer on a lattice are deduced from the generating function  $G(x, T) = \sum_{N,R} \Omega(N, R) x^N u^R$ , where  $\Omega(N, R)$  are the number of different configurations per site of a polymer chain having  $N$  monomers,  $R$  the number of nearest neighbors,  $x$  is the fugacity associated with each step of the walk, and the interaction strength  $u$  is related to the temperature by  $u = \exp(E/k_\beta T)$ . Here  $E (> 0)$  and  $k_\beta$  denote the attractive energy associated with a pair of nearest-neighbor bonds, and the Boltzmann constant, respectively.

An analogy with the power-law behavior of functions such as susceptibility, specific heat, and correlation

length of a critical phenomena allows one to study the issue of universality in polymer statistics in a renormalized-group (RG) framework. To perform the real-space renormalization-group (RSRG) transformations on polymer chains, here one studies the characteristic quantities describing the lattice walk change upon repeated length rescaling of the system. When these quantities remain invariant the chain is said to be “self-similar” at all length scales, and this is a “fixed point” of the rescaling transformation. The invariance of the correlation length is either zero or infinity, and the latter point corresponds to criticality [7].

Despite the apparent simplicity of the procedure outlined above, very few exact results have been found for the Euclidean lattices. This has motivated recent studies on fractal lattices where the asymptotic properties of the polymer chain are obtained by using exact recursion relations. The fractal lattices, like the truncated  $n$ -simplex lattices [8] or the Sierpinski gasket [9], are defined recursively, and, by splitting the generating function in finite subsets of partial contributions, it is possible to write a closed set of recursion equations in terms of a finite number of coupling constants [10]. The variables in these equations are just the partial generating functions corresponding to different polymer configurations for a given size of the fractal lattice. Linearizing the recursion near the nontrivial fixed points, the fixed points reached by the system depending on the initial conditions, one can find the eigenvalues of the transformation matrix which give the characteristic exponents of the system. Exact results have been reported for several lattice models viz., SAW’s, SASAW’s, TSAW’s (true self-avoiding walks), trails and their silhouettes, etc. for a single chain on fractal lattices [4–6, 9–13]. The usefulness of some of these models has also recently been shown for the problem of simultaneous surface adsorption and collapsed transition of a linear po-

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lymer chain [14–16]. Aside from being interesting in their own right, these results are often in qualitative (some times even quantitative) agreement with their counterparts for standard Euclidean lattices.

In this paper we show that the model of a SASAW can be extended to study the critical behavior of two chemically different interacting polymer chains in a solution which may have different qualities for different chains. Depending upon the solvent quality and the attractive interactions between interchain and intrachain monomers, many different situations may arise. In the renormalization-group formulation of this paper they correspond to different fixed points. We show that the system is in either of the three states described below.

(1) The system may be in a state in which the two polymers intermingled with each other in such a way that they cannot be told apart; i.e., a polymer chain cannot be distinguished from the other chain. This state is referred to as the interpenetrated state.

(2) When the attractive interaction between unlike monomers (i.e., the chemically different monomers of the two polymers) reaches a certain (critical) value, the two chains may be zipped together in such a way that they lie side by side. It is tempting to think that this configuration, in some approximate sense, is similar to the formation of double stranded DNA. Here we refer to it as a zipped state.

(3) If the quality of the solvent is such that one or both polymers are in a compact globule state, the two chains at high temperatures are separated from each other without any overlap. We refer to this as a segregated state.

By varying the temperature or tuning the interactions, we may transform the system from a zipped state to a state of interpenetration or to a state of segregation. The transition point corresponds to a tricritical point, and in its proximity a crossover regime may be observed. In the asymptotic limit the mean number of monomers  $M$  in contact with each other at the tricritical point is assumed to behave as

$$M \propto N^y, \quad (1.1)$$

where  $N$  is the total number of monomers in a chain and  $y$  is the contact exponent. In the asymptotic limit, the number of monomers in a chain tends to infinity.

The paper is organized as follows: In Sec. II we describe the model of two chemically different interacting polymer chains each described by a SASAW on a fractal lattice, and outline the general formulation of this problem. The details of the calculation of the fixed points and the value of the crossover exponent at the tricritical point corresponding to different configurations of the two-chain system using RSRG transformation on 4-, 5-, and 6-simplex lattices are given in Secs. III, IV, and V, respectively. The paper ends with a discussion given in Sec. VI.

## II. TWO INTERACTING POLYMER CHAINS ON A TRUNCATED $n$ -SIMPLEX LATTICE

In order to describe the critical behavior of a system of two chemically different homopolymers, denoted here as

$P_1$  and  $P_2$ , in a solution which may have different qualities for different polymer chains, we consider a model of two interacting SASAW's (hereinafter, for brevity sake, we refer to this model as a model of two interacting walks or TIW's) on a family of truncated  $n$ -simplex lattices. This model takes into account the physical condition that the interactions between monomers is repulsive at a short distance and attractive at a "large" distance. The non-crossing constraint represents the repulsion, and the attraction between monomers occupying the neighboring lattice sites is due to the attractive part of the interactions. The strength of these interactions depends on the solvent and the chemical nature of the monomers.

The truncated  $n$ -simplex lattice is defined recursively [8,12]. The graph of the zeroth-order truncated  $n$ -simplex lattice is a complete graph on  $(n+1)$  points. The graph of the  $(r+1)$ th-order lattice is obtained by replacing each of the vertices by the  $r$ th-order graph by a complete graph on  $n+1$  points. Each of the resulting  $n$  points is connected to one of the lines leading to the original vertex. The fractal and spectral dimensions of this lattice, respectively, are defined as

$$d_F = \frac{\ln n}{\ln 2}, \quad \tilde{d} = \frac{2 \ln n}{\ln(n+2)}. \quad (2.1)$$

The lattice with  $n \geq 3$  are of particular interest as they provide a family of fractals in which  $d_F$  can be varied to a wide range by changing  $n$  from 3 to  $\infty$ , while keeping  $\tilde{d}$  almost constant.

The end-to-end distance exponent for a random walk (RW) on fractals belonging to the universality class of the truncated  $n$ -simplex lattice is found to be [17]

$$\nu_R = \frac{\tilde{d}}{2d_F}. \quad (2.2)$$

The subscript  $R$  used here emphasizes the fact that the value is for a RW. Note that the result of Eq. (2.2) implies that the fractal dimensionality of a RW on the  $n$ -simplex lattice is

$$d_R = \frac{1}{\nu_R} = \frac{2d_F}{\tilde{d}}. \quad (2.3)$$

This leads to a condition

$$d_F \geq 2d_R$$

or, from Eq. (2.3),

$$\tilde{d} \geq 4 \quad (2.4)$$

for the repulsions between the monomers to be irrelevant.

It is worth noting that the condition of Eq. (2.4) depends only on the spectral dimension of the lattice and does not involve the fractal dimension. Since this condition is never satisfied [see Eq. (2.1)], there is no upper critical dimension for the  $n$ -simplex lattices.

For a single chain the models of SAW's and SASAW's have been solved exactly for  $3 \leq n \leq 6$  using the RSRG transformations [5,6,10,12]. The critical exponents  $\nu$ ,  $\alpha$ , and  $\gamma$  which are referred to as the exponents of the radius of gyration (or end-to-end distance), specific heat,

and susceptibility, respectively, have been calculated. For the model of the SASAW the collapse transition ( $\theta$  transition) is found to occur on 4- and 6-simplex lattices but not on 3- and 5-simplex lattices. Depending upon the temperature a polymer chain can, therefore, be either in a swollen state or in a collapsed compact globule phase, or at the tricritical point of the  $\theta$  transition on 4- and 6-simplex lattices. However, on 3- and 5-simplex lattices a polymer chain always remains in the swollen state.

The topological structure of a 3-simplex lattice is such that it cannot have two SAW's on it. In a previous paper [18] we considered a model in which we allowed the different walks to cross each other at most once on any

$$G(P_1, P_2, x_3, u_3) = \sum_{\text{all distinct walks}} (x_1^{N_1} u_1^{R_1})(x_2^{N_2} u_2^{R_2})(x_3^{N_3} u_3^{R_3}), \quad (2.5)$$

where  $N_1$  ( $N_2$ ) is the number of steps (monomers) in the polymer  $P_1$  ( $P_2$ ), and  $x_1$  ( $x_2$ ) denotes the fugacity weight attached to each step of polymer  $P_1$  ( $P_2$ ). Here  $u_1$  ( $u_2$ ) represents the Boltzmann factor associated with the attractive interaction between monomers of polymer  $P_1$  ( $P_2$ ).  $x_3$  and  $u_3$  denote, respectively, the fugacity weight attached with zipped walks (i.e., a step in which both walks move side by side) and the Boltzmann factor associated with the attractive interaction between monomers of  $P_1$  and  $P_2$ .  $N_3$  is the total number of zipped steps (i.e., the number of steps in which both walks move together).  $R_1$  ( $R_2$ ) is the number of monomers of the chain  $P_1$  ( $P_2$ ) occupying the nearest-neighboring sites of the lattice, and  $R_3$  is the number of monomers of different chains occupying the nearest-neighbor lattice points.

Since in all calculations reported below we take chains  $P_1$  and  $P_2$  in a given configuration, the variables ( $x_1, u_1$ ) and ( $x_2, u_2$ ) are therefore taken to be known. They are found from the study of a single chain [5,6] on a given lattice. Further, as is obvious from the figures given below (see, for example, Figs. 1 and 7) we choose

$$x_3 = \sqrt{x_1 x_2}. \quad (2.6)$$

Therefore, Eq. (2.5) involves only  $u_3$  as an independent variable.

From the generating function we can calculate the average number of monomers of the two chains which are in contact (nearest neighbor) with each other from the relation

$$\langle R_3 \rangle = u_3 \frac{\partial \ln G}{\partial u_3}. \quad (2.7)$$

For a finitely ramified fractal lattice it is possible to write the relevant generating function in terms of a finite number of restricted partition functions [10,12]. These partition functions are defined recursively as a weighted sum over all configurations for a given stage of the iterative construction of the fractal lattice. The recursions express the restricted functions for the  $(r+1)$ th-order lattice in terms of those of the  $r$ th generation one. The variables in these equations are just the partial generating

lattice point, and a lattice bond may be occupied by a step of one or by both walks and calculated the contact exponent. However, due to these somewhat artificial conditions, the model may not correspond to any real physical system. In this paper we therefore consider the lattices with  $n=4, 5$ , and 6. These lattices can have two interacting SAW's or SASAW's without the walks crossing each other at any lattice point. Thus the short range interactions between the monomers of  $P_1$  and  $P_2$  as well as between the monomers in a chain are taken to be repulsive.

The generating function for the problem we want to study can be written as

functions corresponding to different polymer configurations for a given size of the fractal lattice. Linearizing the recursions near the nontrivial fixed points, the fixed point reached by the system depending on the initial conditions, we can find the eigenvalues of the transformation matrix which give the characteristic exponents of the system.

### III. THE TRUNCATED 4-SIMPLEX LATTICE

The basic geometrical unit of construction of a truncated 4-simplex lattice is a tetrahedron with four corner vertices and bonds between every pair of vertices [8]. Each vertex connected through a direct bond is termed a nearest neighbor. The value of  $d_F$ ,  $\bar{d}$ , and the connectivity constant (for a SAW)  $\mu$  are 2, 1.5474..., and

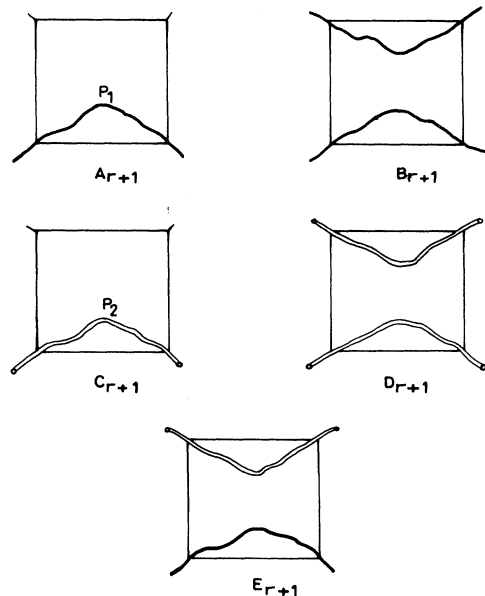


FIG. 1. Diagrams representing the four restricted generating functions for two chains [indicated by black ( $P_1$ ) and white ( $P_2$ ) lines] on a truncated 4-simplex lattice.

2.2866. . . , respectively.

The restricted generating functions of our interest are shown in Fig. 1, and their corresponding recursion relations are (see Ref. [8] for a single-chain problem)

$$A_{r+1} = A^2 + 2A^3 + 2A^4 + 4A^3B + 6A^2B^2, \quad (3.1)$$

$$B_{r+1} = A^4 + 4A^3B + 22B^4, \quad (3.2)$$

$$C_{r+1} = C^2 + 2C^3 + 2C^4 + 4C^3D + 6C^2D^2, \quad (3.3)$$

$$D_{r+1} = C^4 + 4C^3D + 22D^4, \quad (3.4)$$

$$E_{r+1} = A^2C^2 + 2ACE(A+C) + 2E^4 + 6E^2(B^2+D^2) + 4E^3(B+D). \quad (3.5)$$

Here and below we adopt a notational simplification in which we drop the index from the right-hand side of the recursion relations. In Fig. 2 we illustrate the configuration of order  $r$  which appears in Eq. (3.5). The starting values of these functions (see Fig. 3) are

$$A_1 = x_1^2 + 2x_1^3u_1 + 2x_1^4u_1^3, \quad (3.6)$$

$$B_1 = x_1^4u_1^4, \quad (3.7)$$

$$C_1 = x_2^2 + 2x_2^3u_2 + 2x_2^4u_2^3, \quad (3.8)$$

$$D_1 = x_2^4u_2^4, \quad (3.9)$$

$$E_1 = x_3^4u_3^4. \quad (3.10)$$

Note that indexes 1 and 2 on the right-hand side of Eqs. (3.6)–(3.9) correspond to chains  $P_1$  and  $P_2$ , respectively, and index 3 in Eq. (3.10) indicates the variables corresponding to interchain configurations, whereas index 1 on the left-hand side of these equations represents the order of the iteration.

We may note that the recursion relations for  $A$  and  $B$  (or  $C$  and  $D$ ) are independent of  $C$ ,  $D$ , and  $E$  (or  $A$ ,  $B$ , and  $E$ ). This is because we ignored, for simplification, the effect of one SAW on the self-avoidance of the other SAW. In a dilute solution, however, the size of a chain is

not expected to change due to interaction between monomers of different chains [19]. The effect of interactions between two polymers are taken through  $E$ . The critical behaviors of the individual chains remain as in the dilute case, and are given by the fixed points of Eqs. (3.1) and (3.2) and (3.3) and (3.4). The fixed point  $E^*$  of Eq. (3.5) contains information about the configurations of the two chains in an asymptotic limit.

The fixed points corresponding to different configurations of one polymer chain in the asymptotic limit are found by solving Eqs. (3.1) and (3.2) for chain  $P_1$  and Eqs. (3.3) and (3.4) for chain  $P_2$ . Since the two sets of equations are identical, they give identical fixed points. In Fig. 4 we plot the phase diagram, i.e., the self-attraction parameter  $u_1$  (or  $u_2$ ) as a function of the critical fugacity  $x_c$  for a single chain. The critical line representing a swollen state (indicated on the figure by  $S$ ) is separated from a critical line representing a compact globule state (shown by letter  $C$ ) by a tricritical ( $\theta$ ) point (shown by letter  $T$ ).

The state of a polymer chain depends on the quality of the solvent and on the temperature, and can therefore be in any one of the three states of the swollen compact globule, and at the  $\theta$  point described in the asymptotic limit by the fixed points  $(A^*, B^*)$  or  $(C^*, D^*) = (0.4294. . . , 0.04998. . .)$ ,  $(0, 22^{-1/3})$ , and  $(\frac{1}{3}, \frac{1}{3})$ , respectively [5]. The fixed point corresponding to the swollen state is reached for all values of  $u_1$  (or  $u_2$ )  $< u_c = 3.31607$  at  $x_1$  (or  $x_2$ )  $= x_c$ . The value of  $x_c$  is a function of the interaction  $u_1$  (or  $u_2$ ), which for the swollen state varies between 1 and 3.31607. . . . The end to end distance for a chain of  $N$  monomers in this state varies as  $N^\nu$  with  $\nu = 0.7294. . .$ . The fixed point corresponding to the compact globule state is reached for all values of  $u_1$  (or  $u_2$ )  $> u_c$  (i.e., at low temperatures) at  $x_1$  (or  $x_2$ )  $= x_c(u_i)$ , where  $i = 1$  or 2. In a compact globule state the polymer chain has a finite density of monomer per site when  $N \rightarrow \infty$ . At  $u_1$  (or  $u_2$ )  $= u_c = 3.39607$  and  $x_1$  (or  $x_2$ )  $= x_c(u_c) = 0.22913. . .$ , the  $\theta$ -point state is found.

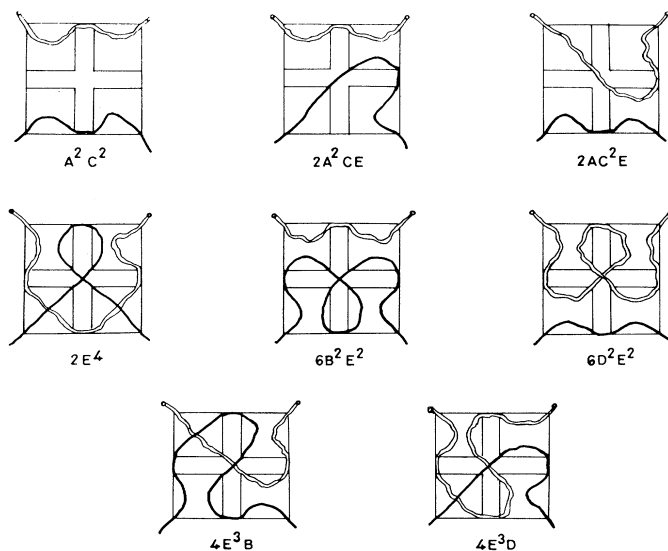


FIG. 2. Various possible configurations of  $r$ th order, which contribute to the recursion relation of  $E_{r+1}$ . The numerical factor of each term indicates the total number of different configurations of the type depicted by the corresponding figure.

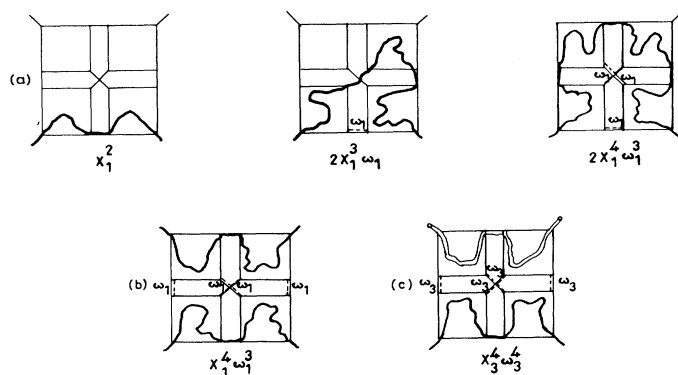


FIG. 3. Diagrams defining the initial weights for the SASAW on a 4-simplex lattice (a) for  $A_1$  ( $C_1$ ), (b) for  $B_1$  ( $D_1$ ), and (c) for  $E_1$ . The attractive interactions between neighboring sites are denoted by broken lines (for details, see text).

In a system of two chemically different polymer chains, we may have six independent combinations of the individual chains which we indicate by  $SS$ ,  $CC$ ,  $TT$ ,  $SC$ ,  $ST$ , and  $CT$ , where letters  $S$ ,  $C$ , and  $T$  stand for the swollen, compact globule, and  $\theta$ -point states. Using the fixed points ( $A^*$ ,  $B^*$ ) and ( $C^*$ ,  $D^*$ ) corresponding to these states of the polymers  $P_1$  and  $P_2$  we solve Eq. (3.5). This leads to two fixed points denoted as  $E_s^*$  and  $E_t^*$  for each combination of states of the individual chains (see Table I). We therefore have the following situations.

#### A. $SS$ state

(i) The fixed point ( $A^*=C^*$ ,  $B^*=D^*$ ,  $E_s^*$ ) = (0.4294 . . . , 0.04998 . . . , 0.04998 . . .) is reached for all values of  $u_3 < u_{3c}$  ( $x_3$ ) shown by line  $SS$  in Fig. 5. Line  $S$  which terminates at point  $T$  corresponds to the critical state of a linear polymer chain in a swollen state.

Since at this fixed point  $B^*=D^*=E_s^*$ , we conclude that when both polymers  $P_1$  and  $P_2$  are in a solvent which is good for both chains and the attraction between

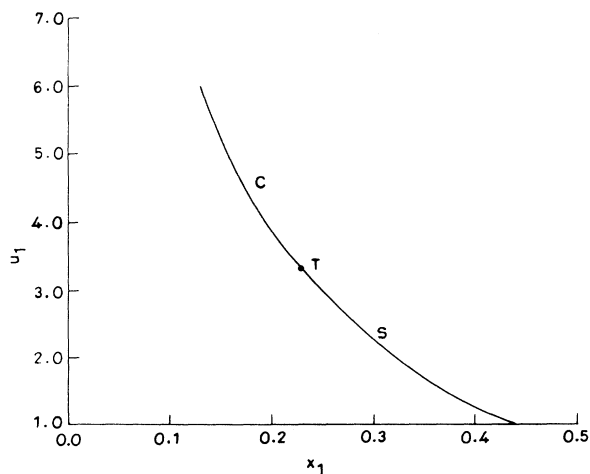


FIG. 4. The self-attraction parameter  $u_1$  as a function of critical fugacity  $x_1$  for a linear polymer chain on a 4-simplex lattice. The tricritical point which corresponds to  $u_1 = u_1^c = 3.31607$  and  $x_1 = x_1^c = 0.23308$  is shown by  $T$ .  $S$  and  $C$ , respectively, indicate the swollen and compact globule phases of the chain.

unlike monomers is less than a certain critical value, the polymer chains cannot be told apart; i.e., polymers  $P_1$  cannot be distinguished from polymers  $P_2$ . Therefore the two chains are in a state of interpenetration.

(ii) The fixed point ( $A^*=C^*$ ,  $B^*=D^*$ ) = (0.4294 . . . , 0.04998 . . . , 0.6125 . . .) is reached when  $u_3 = u_{3c}$  ( $x_3$ ). This is shown by line  $SS$  in Fig. 5. Linearization of Eqs. (3.1)–(3.5) about this point gives two distinct eigenvalues greater than 1:  $\lambda_s = 2.7965 . . .$ , which corresponds to the swollen state of a chain; and  $\lambda_t = 2.6420 . . .$ , which

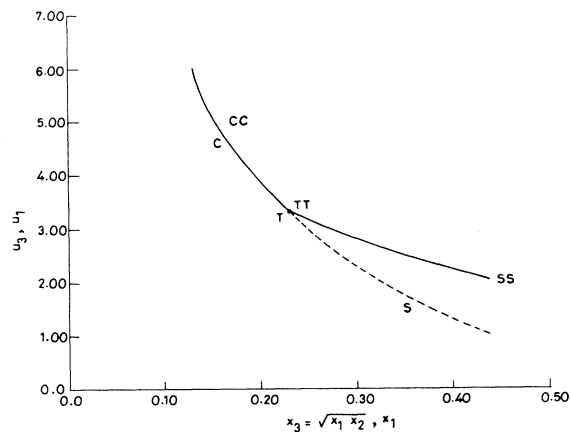


FIG. 5. The  $x_3$ ,  $u_3$  phase diagram of a system of two polymer chains in a nonselective solvent for a truncated 4-simplex lattice. Lines  $SS$  and  $CC$  represent the tricritical lines of the zipped state of two chains each in the swollen state and the interpenetration state of the chains each in a compact globule phase, respectively. Point  $TT$ , at which these lines meet, represents a transition point from a segregated to an interpenetrated state of the chains each at its  $\theta$  point. For a given value of  $x_3$  which corresponds to the swollen state of both chains, the chains are in interpenetrated state when the value of  $u_3$  is less than the value given by the line  $SS$ . For the value of  $x_3$  corresponding to the chains in their compact globule state or at their  $\theta$  points, the two chains are in a segregated state for all values of  $u_3$  less than the value given by line  $CC$  or point  $TT$ . We also show the  $x$ - $u$  phase diagram of a single chain for comparison's sake. Note that line  $CC$  overlaps with line  $C$ ,  $TT$  with point  $T$ . The meaning of the symbols  $S$ ,  $C$ , and  $T$  are explained in Fig. 4.

TABLE I. Values of fixed points, relevant eigenvalues, and the contact exponent  $y$  at the tricritical point for the 4-simplex lattice. The swollen, compact globule, and tricritical configuration of each chain is indicated by letters  $S$ ,  $C$ , and  $T$ , respectively.  $\lambda_1$  is the largest eigenvalue of the system, and  $\lambda_i$  refers to the two chain configuration point.

State of individual chains	$E_s^*$	$E_i^*$	$\lambda_i$	$\lambda_1$	$y$
$SS$	0.04998	0.6125	2.6420	2.7965	0.9447
$CC$	0.0	$22^{-1/3}$	2.5440	4.0	0.6735
$TT$	0.01484	1/3	2.2222	3.7037	0.6098
$SC$	0.0	0.4880	2.8520	4.0	0.7559
$ST$	0.02687	0.4294	2.8526	3.7037	0.6311
$TC$	0.0	0.3680	2.5740	4.0	0.6820

corresponds to the zipped state of the two chains. Therefore line  $SS$  is a tricritical line. Using these eigenvalues we calculate the value of the contact exponent from the relation [16]

$$y = \frac{\ln \lambda_i}{\ln \lambda_1} \quad (3.11)$$

and find its value equal to 0.9447. This value is close to unity and suggests that the two chains lie side by side.

#### B. $CC$ state

(iii) The fixed point  $(A^*=C^*, B^*=D^*, E_s^*)=(0, 22^{-1/3}, 0)$  corresponds to a segregated state in which the two chains, each in compact globule state, are separated from each other. This point is reached for all values of  $u_3(x_3) < \sqrt{u_1(x_{1c})u_2(x_{2c})}$ . For a given  $x_3 = \sqrt{x_{1c}x_{2c}}$ , all points below line  $CC$  in Fig. 5 correspond to this state of a two-chain system.

(iv) The fixed point  $(A^*=C^*, B^*=D^*, E_i^*)=(0, 22^{-1/3}, 22^{-1/3})$  is reached for  $u_3(x_3) = \sqrt{u_1(x_{1c})u_2(x_{2c})}$  and is shown by line  $CC$  in Fig. 5. Note that line  $CC$  coincides with the critical line of a single chain in a compact globule phase. This point corresponds to a state in which both chains intermingled with each other in such a way that they cannot be told apart. This is a state of interpenetration. The linearization of Eqs. (3.1)–(3.5) about this fixed point leads to two distinct eigenvalues which are greater than 1. The value  $\lambda_c=4.0$  corresponds to the compact globule state of a chain, and  $\lambda_1=2.5440$  corresponds to the interpenetration of chains. Since the point has two eigenvalues greater than unity, therefore line  $CC$ , which shows the value of  $u_{3c}$  as a function of  $x_3$  for which the fixed point is reached, is a tricritical line and represents the transition from a segregated state to an interpenetrated state. These eigenvalues lead to a value of  $y$  equal to 0.6735.

#### C. $TT$ state

(v) The fixed point  $(A^*=C^*, B^*=D^*, E_s^*)=(\frac{1}{3}, \frac{1}{3}, 0.01484)$  corresponds to a state when both chains are at their  $\theta$  point and are segregated from each other. This point is reached for all values of  $u_3 < u_c = 3.31607$ . . . at

$x_3 (=x_{1c}=x_{2c})=0.22913$  . . . .

(vi) The fixed point  $(A^*=C^*, B^*=D^*, E_i^*)=(\frac{1}{3}, \frac{1}{3}, \frac{1}{3})$  is reached for  $u_3=u_c=3.31607$  . . . at  $x_3=x_{1c}=x_{2c}=0.22913$  . . . , and is shown by point  $TT$  in Fig. 5. Note that point  $TT$  on the phase diagram also corresponds to the  $\theta$  point of a chain. This point represents a transition point from a segregated to an interpenetrated state. Linearization of relevant equations about this point yields three distinct eigenvalues greater than unity. This point is therefore a tetracritical point which connects two tricritical lines of zipped states of two chains each in the swollen state, and an interpenetrated state of two chains each in a compact globule state. The value of the contact exponent is found to be equal to  $y=0.6098$  . . . .

#### D. $SC$ state

(vii) The fixed point  $(A^*, B^*, C^*, D^*, E_s^*)=(0.4294, \dots, 0.04998, \dots, 0, 22^{-1/3}, 0)$  corresponds to a segregated state. The two chains, one in a swollen state and the other in a compact globule phase, are separated from each other without any overlap. This point is reached for all values of  $u_3(x_3) < u_{3c}(x_3)$ . The value of  $u_{3c}$  as a function of  $x_3$  is shown in Fig. 6 by line  $SC$ . For a given  $x_3$  the  $SC$  segregated state is found for all values of  $u_3$  lying below line  $SC$ .

(viii) The fixed point  $(A^*, B^*, C^*, D^*, E_s^*)=(0.4294, \dots, 0.04998, \dots, 0, 22^{-1/3}, 0.4880, \dots)$  at a given  $x_3$  is reached for  $u_3=u_{3c}(x_3)$ . The variation of  $u_{3c}$  as a function of  $x_3$  is shown by line  $SC$  in Fig. 6. This is a state in which a polymer chain in a swollen state wraps over a compact globule object of the other chain. This may be compared with adsorption of a linear polymer chain in a swollen state on a surface. Here the surface is provided by the compact globule shape of the other chain. The linearization of the relevant equations yield three distinct eigenvalues which are greater than 1. The contact exponent  $y$  is calculated using the largest eigenvalue, which in this case describes the compact globule state of a chain, and the eigenvalue which is found from the linearization of Eq. (3.5). These eigenvalues are  $\lambda_c=4.0$  and  $\lambda_i=2.8520$  . . . . The value of  $y$  is found to be 0.7559.

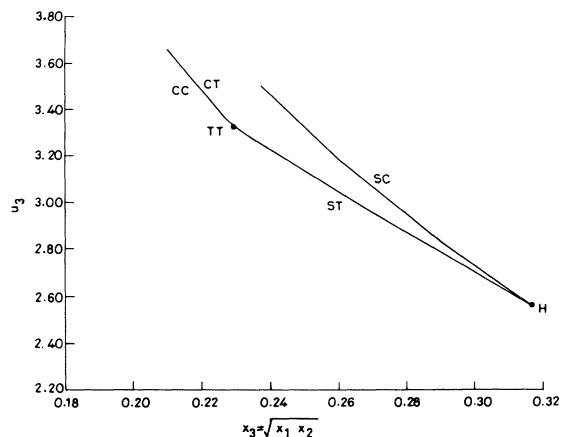


FIG. 6. The  $x_3, u_3$  phase diagram of a system of two chains in a selective solvent for a truncated 4-simplex lattice. Lines  $SC$  and  $ST$ , respectively, represent the interpenetration states of two chains when one chain is in a swollen state and the other is in compact globule phase or at its  $\theta$  point. Line  $CT$  corresponds to the configuration of interpenetration of the chains when one is in the compact globule phase and the other is at its  $\theta$  point. When the value of  $u_3$  is less than that given by corresponding lines, the two chains are segregated from each other. Note that the individual chains configurations remain intact whether they are segregated or intermingled.

#### E. $ST$ state

(ix) The fixed point  $(A^*, B^*, C^*, D^*, E_s^*) = (0.4294 \dots, 0.04998 \dots, \frac{1}{3}, \frac{1}{3}, 0.02687 \dots)$  is reached for all values of  $u_3$  less than the value given by line  $ST$  of Fig. 6. The two chains, one in a swollen state and the other at the  $\theta$  point, are segregated from each other without any overlap.

(x) The fixed point  $(A^*, B^*, C^*, D^*, E_i^*) = (0.4294 \dots, 0.04998 \dots, \frac{1}{3}, \frac{1}{3}, 0.4294)$  corresponds to a state in which both chains are interpenetrated with each other. This interpenetration is different from the case in which both chains are in an identical state. While in the latter case the two chains, being in identical states, intermingle in such a way that they cannot be told apart, in the  $ST$  state one chain is in a swollen state and the other oscillates between the swollen and compact globule phases at the  $\theta$  point. Because of the interaction between the two chains, their monomers come close to each other (given by configuration  $E$  in Fig. 1) while retaining their individual configurations. The value of the contact exponent is found to be equal to 0.6311...

#### F. $CT$ state

(xi) The fixed point  $(A^*, B^*, C^*, D^*, E_s^*) = (0, 22^{-1/3}, \frac{1}{3}, \frac{1}{3}, 0)$  corresponds to a state in which one chain is in the compact globule phase and the other chain is at its  $\theta$  point, and they are segregated from each other. The point is reached for all values of  $u_3$  less than the value shown by line  $CT$  in Fig. 6. Line  $CT$  coincides on the graph with line  $CC$  discussed above.

(xii) The fixed point  $(A^*, B^*, C^*, D^*, E_i^*) = (0, 22^{-1/3}, \frac{1}{3}, \frac{1}{3}, 0, 3.680 \dots)$  is reached when, at a given  $x_3$ , the values of  $u_3$  become equal to the value given by line  $CT$  in Fig. 6. The point represents a state in which both chains interpenetrate in a sense that one chain forms a compact globule phase and the other chain oscillates on the surface of it between swollen and compact globule phases in such a way that configuration  $E$  of Fig. 6 survives at all length scales. The value of the contact exponent is found to be equal to 0.6820.

Point  $H$  on Fig. 6, where lines  $SC$  and  $ST$  meet, has the values  $x_1 = 0.4373 \dots$ ,  $u_1 = 1.0$  and  $x_2 = 0.22913 \dots$ ,  $u_2 = 3.31607 \dots$ . Therefore at point  $H$ , one chain (say  $P_1$ ) is in a swollen state with no self-attraction, and chain  $P_2$  is at its  $\theta$  point. For line  $ST$ ,  $x_1$  and  $u_1$  are varied between 0.22913... and 0.4373... and 3.31607 and 1.0, respectively, while  $x_2$  and  $u_2$  are kept fixed at  $x_2 = 0.22913 \dots$  and  $u_2 = 3.31607$ . At point  $TT$ ,  $x_1 = x_2 = 0.22913$  and  $u_1 = u_2 = 3.31607$ . For line  $SC$ , while  $x_1$  and  $u_1$  have the same range of variation as that of  $ST$ ,  $x_2$  and  $u_2$  are varied between  $0.22913 \dots \leq x_2 \leq 0$  and  $3.31607 \leq u_2 \leq \infty$ .

#### IV. THE TRUNCATED 5-SIMPLEX LATTICE

The basic geometrical unit of the construction of a truncated 5-simplex lattice is a hypertetrahedron of five corner vertices and bonds between every pair of vertices, termed as nearest neighbor [12]. For this lattice the

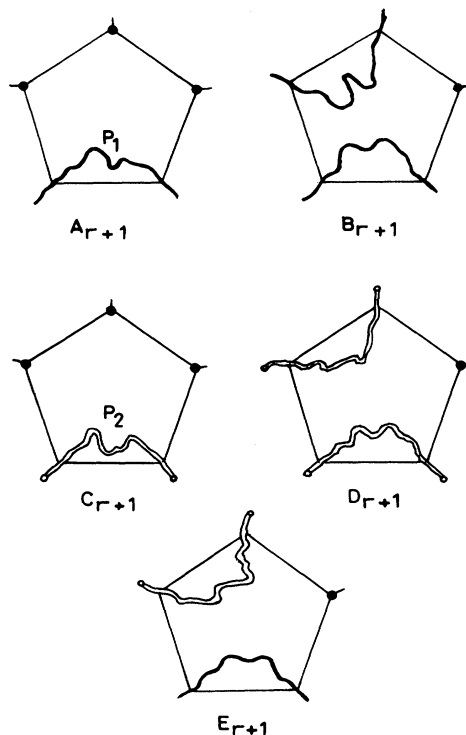


FIG. 7. Diagrams representing the five restricted generating functions for two interacting and chemically different chains on a truncated 5-simplex lattice. Other notations are the same as in Fig. 1.

values of  $d_r$ ,  $\bar{d}$ , and  $\mu$  are 2.3219, 1.6541, and 2.976 03, respectively. The restricted partition functions of our interest are shown in Fig. 7. The recursion relations for  $A_{r+1}$  and  $B_{r+1}$  are written as [6,12]

$$A_{r+1} = A^2 + 3A^3 + 6A^4 + 5A^5 + 18A^2B^2 + 96A^2B^3 + 18A^3B + 78A^3B^2 + 30A^4B + 132AB^4 + 132AB^5, \tag{4.1}$$

$$B_{r+1} = A^4 + 2A^5 + 13A^4B + 4A^3B + 32A^3B^2 + 88A^2B^3 + 22B^4 + 220AB^4 + 186B^5. \tag{4.2}$$

These relations correspond to polymer chain  $P_1$ , as shown in Fig. 7. A similar relation holds for  $P_2$  in terms of  $C$  and  $D$  (Fig. 7). The recursion relation for the partition function  $E_{r+1}$  can be expressed in terms of  $A_r$ ,  $B_r$ ,  $C_r$ ,  $D_r$ , and  $E_r$ . Thus

$$E_{r+1} = 6E^2D^2 + 42CE^2D^2 + 12C^2E^2D + 12ACED^2 + 2AC^2E + 32E^2D^3 + 2C^2E^2 + 8AC^2ED + 4AC^3E + A^2C^2 + A^2C^3 + 6B^2E^2 + 18B^2CE^2 + 42AB^2E^2 + 12AB^2CE + 30B^2E^3 + 4BE^3 + 16BCE^3 + 12ABCE^2 + 24ABE^3 + 20BE^4 + 32B^3E^2 + 16BE^3D + 8ABCE + 12A^2BE^2 + 16BE^3D + 8A^2BCE + 12A^2BE^2 + 4E^3D + 24CE^3D + 12ACE^2D + 2A^2CE + 14C^2E^3 + 30E^3D^2 + 6AC^2E^2 + 5A^2C^2E + 2E^4 + 10CE^4 + 8ACE^3 + 20E^4D + 6A^2CE^2 + 10AE^4 + 4A^2E^3 + 6E^5 + 16AE^3D + 4A^3CE + 2A^3E^2 + 18AE^2D^2 + A^3C^2. \tag{4.3}$$

The starting weights of these functions are

$$A_1 = x_1^4 + 3x_1^3u_1 + 6x_1^4u_1^3 + 6x_1^3u_1^6, \tag{4.4}$$

$$B_1 = x_1^4u_1^4 + 2x_1^5u_1^7, \tag{4.5}$$

$$C_1 = x_2^2 + 3x_2^3u_2 + 2x_2^4u_2^3 + 6x_2^5u_2^6, \tag{4.6}$$

$$D_1 = x_2^4u_2^4 + 2x_2^5u_2^7, \tag{4.7}$$

and

$$E_1 = x_3^4u_3^4 + x_3^5u_3^6u_1 + x_3^5u_3^6u_2. \tag{4.8}$$

The notational details are the same as in the case of the 4-simplex lattice.

As already mentioned, a polymer chain always remains in the swollen state for all values of intrachain (self-) attraction on a 5-simplex lattice. This state is characterized by the fixed point

$$A^*(=C^*)=0.3265\dots, \tag{4.9}$$

$$B^*(=D^*)=0.0279\dots$$

In this case we therefore have only one combination, i.e., SS, of chains. Using these values for  $A^*$ ,  $B^*$ ,  $C^*$ , and  $D^*$ , we have solved Eq. (4.3), which leads to the following fixed points:

$$E_s^*=0.0279\dots, \tag{4.10a}$$

$$E_i^*=0.2713\dots \tag{4.10b}$$

The fixed point  $E_s^*$  is found for all values of  $u_3 < u_{3c}$  ( $x_3$ ). At  $u_3 = u_{3c}$  ( $x_3$ ) we find the fixed point  $E_i^*$ . The linearization of Eq. (4.3) about the fixed point  $E_i^*$  gives the eigenvalue  $\lambda_i = 2.2401$ . This leads to the value of

$y = 0.7664\dots$

In Fig. 8 we plot  $u_3$  as a function of  $x_3 = \sqrt{x_{1c}x_{2c}}$ . The interpenetrated state is found for all values of  $u_3$  which lie below lines SS at a given  $x_3$ . When the value of  $u_3$  reaches a value given by line SS, the two chains are zipped together. Line S indicates the critical values of fugacity  $x_c$  and the self-attraction  $u_1$  (or  $u_2$ ) of a polymer chain.

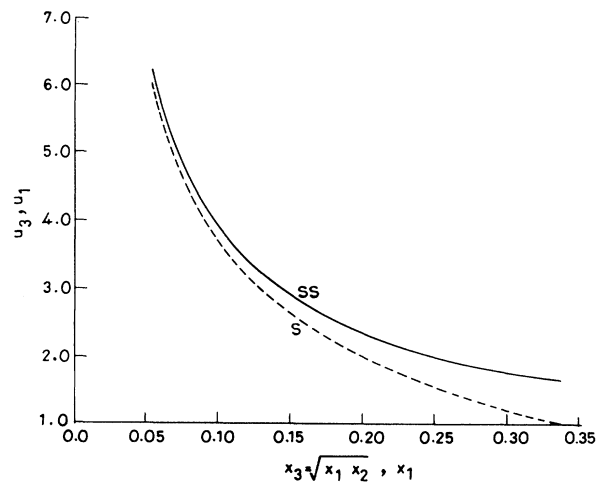


FIG. 8. The  $x_3$ ,  $u_3$  phase diagram representing the configurations of a system of two chains for a truncated 5-simplex lattice. Line SS, as in Fig. 5, represents the zipped state of the chains. For all values of  $u_3$  less than the value given by line SS, the two chains intermingle with each other. Line S represents the swollen state of a chain.



### V. THE TRUNCATED 6-SIMPLEX LATTICE

The basic geometrical unit of the construction of a truncated 6-simplex lattice is a hexagon with six-corner vertices and bonds between every pair of vertices [6,12].

For this lattice  $d_F = 2.5849 \dots$  and  $\bar{d} = 1.7233 \dots$ , respectively, and the connectivity (for a SAW)  $\mu = 3.6818 \dots$ . The restricted partition functions of our interest are shown in Fig. 9. The recursion relations for  $A_{r+1}$ ,  $B_{r+1}$ , and  $C_{r+1}$  are written as [6,12]

$$\begin{aligned} A_{r+1} = & A^2(1+36B^2+384B^3+5544B^4+4992B^2C)+4A^3(1+6B+78B^2+648B^3+432B^2C) \\ & +12A^4(1+10B+80B^3+10C^2+40BC)+24A^5(1+A+2C+9B) \\ & +24AB^3(1+22B+362B^2+636BC+472C^2)+48B^4(11B+137B^2+521C^2+428BC), \end{aligned} \quad (5.1)$$

$$\begin{aligned} B_{r+1} = & A^4(1+26B+144BC+324B^2+6A^2)+4A^5(1+4C+16B) \\ & +4A^2B^2(44B+905B^2+1272BC+708C^2)+4A^3B(1+16B+308B^2+208BC) \\ & +8AB^3(55B+822B^2+2140BC+2084C^2)+B^2(22B^2+372B^3+5440B^4+23520B^3C \\ & +48160B^2C^2+76800BC^3+94336C^4), \end{aligned} \quad (5.2)$$

$$\begin{aligned} C_{r+1} = & A^6+6A^5(C+2B)+4A^2B^2(9A^2+52AB)+18A^2B^3(159B+236C)+AB^4(2568B+6252C) \\ & +2940B^6+14448B^5C+43200B^4C^2+94336B^3C^3+541568C^6. \end{aligned} \quad (5.3)$$

These relations correspond to polymer chain  $P_1$  as shown in Fig. 9. A similar relation holds for  $P_2$  in terms of  $D$ ,  $E$ , and  $F$ , as shown in Fig. 9.

Since the recursion relations for the restricted partition functions  $G$ ,  $H$ , and  $I$  which correspond to configurations of two interacting chains are lengthy and run to several typed pages, we do not give them here. These relations can, however, be made available to the reader on request.

The starting weights of these functions are

$$A_1 = x_1^2 + 4x_1^3u_1 + 12x_1^4u_1^3 + 24x_1^5u_1^6 + 24x_1^6u_1^{10}, \quad (5.4)$$

$$B_1 = x_1^4u_1^4 + 4x_1^5u_1^7 + 6x_1^6u_1^{11}, \quad (5.5)$$

$$C_1 = x_1^6u_1^{12}. \quad (5.6)$$

The expressions for  $D_1$ ,  $E_1$ , and  $F_1$  are found from Eqs. (5.4)–(5.6) by replacing variables  $x_1$  and  $u_1$  by  $x_2$  and  $u_2$ , respectively. The starting weights for functions  $G_1$ ,  $H_1$ , and  $I_1$  are as follows:

$$\begin{aligned} G_1 = & x_3^4u_3^4 + 2x_3^5u_3^6(u_1+u_2) \\ & + 2x_3^6u_3^8(u_1^3+u_2^3+u_1u_2u_3), \end{aligned} \quad (5.7)$$

$$H_1 = x_3^6u_3^8u_1^4, \quad (5.8)$$

$$I_1 = x_3^6u_3^8u_2^4. \quad (5.9)$$

Notational details are the same as outlined in preceding sections.

The solutions of Eqs. (5.1)–(5.3) with starting weights of Eqs. (5.4)–(5.6) lead to three different positive real fixed points [6] with features given below.

(i) The fixed point  $(A^*, B^*, C^*) = (0.2623 \dots, 0.01758 \dots, 0.00070 \dots)$  corresponds to the swollen state of the chain with  $\nu = 0.5506 \dots$  and  $\alpha = 0.5767 \dots$ . For  $x_1 = x_c(u_1)$  this fixed point is reached for all  $u_1 < u_c = 4.9998 \dots$ .

(ii) The fixed point  $(A^*, B^*, C^*) = (0, 0, 0.07132 \dots)$  is reached for all  $u_1 > u_c$  (i.e., at low temperatures) and

$x_1 = x_c(u_1)$ . This phase has a finite density of monomers per site when  $N_1 \rightarrow \infty$ . This is a fixed point which describes the collapsed phase of the polymer.

(iii) The fixed point  $(A^*, B^*, C^*) = (0.1294 \dots, 0.09572 \dots, 0.05344 \dots)$  is obtained for  $u_1 = u_c$ . Since this point has two eigenvalues greater than 1, this point is identified as the tricritical point of the  $\theta$  transition.

The other two fixed points  $(A^*, B^*, C^*) = (0.2540 \dots, 0.02215 \dots, 0.07098 \dots)$  and  $(0.200 \dots, 0.0660 \dots, 0.666 \dots)$  cannot be reached starting with any suitable initial condition, so they are spurious [6].

Since the 6-simplex lattice exhibits a collapsed transition, there are six independent combinations of single-

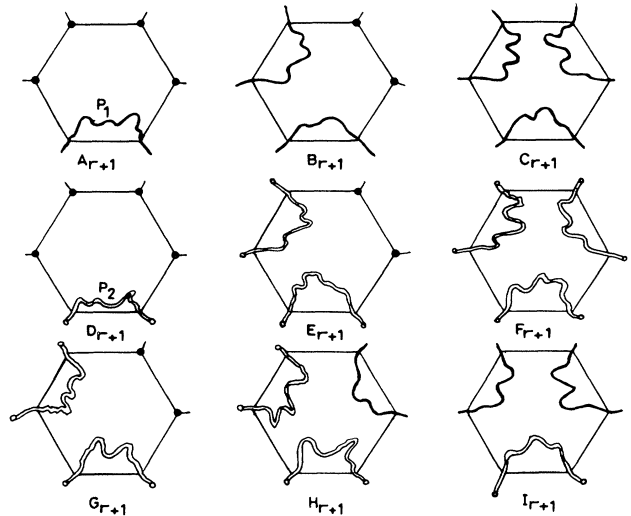


FIG. 9. Diagrams representing the nine generating functions for two interacting chains on a truncated 6-simplex lattice. Other notations are the same as in Fig. 1.

TABLE II. Values of fixed points  $G_S^*$ ,  $H_S^*$ ,  $I_S^*$ ,  $G_i^*$ ,  $H_i^*$ , and  $I_i^*$ . The labeling is the same as in Table I, but for 6-simplex.

State of individual chains	$G_S^*$	$H_S^*$	$I_S^*$	$G_i^*$	$H_i^*$	$I_i^*$	$\lambda$	$\lambda_1$	$y$
<i>SS</i>	0.0175	0.0007	0.0007	0.1406	0.0147	0.0147	2.2054	3.4965	0.6318
<i>CC</i>	0.0	0.0	0.0	0.0	0.0713	0.0713	4.1201	0.6318	0.7902
<i>TT</i>	$5.4 \times 10^{-4}$	$5.4 \times 10^{-4}$	$5.4 \times 10^{-4}$	0.0957	0.0535	0.0535	3.2225	5.4492	0.6902
<i>SC</i>	0.0	0.0	0.0	$6.75 \times 10^{-5}$	0.1049	0.0711	5.0529	6.0	0.9041
<i>ST</i>	0.0028	0.0003	0.0001	0.229	0.1047	0.0776	5.1434	5.4492	0.9659
<i>CT</i>	0.0	0.0	0.0	0.0060	0.0714	0.0806	4.4019	6.0	0.8271

chain states, similar to the case of 4-simplex lattice. In Table II we list the values of fixed points corresponding eigenvalues and contact exponents. With the initial conditions given by Eqs. (5.7)–(5.9), the two fixed points, namely  $(G^*, H^*, I^*) = (0.0651, 0.1023, 0.1023)$  and  $(0.0338, 0.1033, 0.11033)$ , belonging to *SS* configurations could not be reached.

Since the qualitative features of the phase diagrams are the same as that of the 4-simplex lattice discussed in Sec. III, we do not present them here.

## VI. DISCUSSION

We have examined the possibility of studying the critical behavior of two chemically different interacting linear homopolymer chains in their asymptotic limit dissolved in a solution which may have different qualities for different chains in a renormalization-group framework. The system is represented by two interacting SASAW's in such a way that (i) these walks do not cross each other at any lattice point, and a lattice bond can be occupied by at most a step of any one walk; and (ii) each pair of nearest-neighbor sites occupied by different walks interacts with a constant attraction. We call this model the model of two interacting SASAW's or, for brevity, a model of two interacting walks (TIW's).

The lattice model of a SASAW has been used to represent a linear polymer chain in a poor solvent that can undergo a collapse transition, where the chain contracts from an extended state to a compact globule state when the temperature is lowered [5,6]. Note that the model of a SAW is a critical  $O(q)$  model with a  $q \rightarrow 0$  component [1]. The SASAW changes the behavior of the phase transition of the SAW from second order to first order into a compact globule phase at a low temperature. At the intermediate temperature ( $\theta$  point), its behavior is described by a tricritical point of  $O(q)$ ,  $q \rightarrow 0$  spin model [1]. In this regime the upper critical dimension changes from 4 to 3 with the consequence that for  $d=3$ ,  $\nu$  is equal to  $\frac{1}{2}$  plus a logarithmic correction. The phase diagram for a SASAW model has been studied on both regular and fractal lattices [4–6,20].

The problem with which we have been concerned here is to predict the effect of attraction between unlike monomers on their configurations leading to interpenetration or zipping chains. In writing the recursion relations for the restricted partition functions of individual chains, we neglected the effect of one chain on the self-avoidance of the other chain; i.e., the critical behavior of a chain

remains unaffected by the presence of the other chain. This is a reasonable approximation in a dilute solution [19]. We solved exactly models of TIW's on truncated  $n$ -simplex lattices for  $4 \leq n \leq 6$  using real-space RG transformations.

When the solvent is good for both chains, and the attraction between unlike monomers is less than a certain critical value the magnitude of which depends on the critical fugacities of both chains, the system of two chains is found to be in a state of interpenetration. In this regime, therefore, the chemical difference between  $P_1$  and  $P_2$  chains completely drops out of the thermodynamics of the solution. A similar result was found by Joanny, Jeibler, and Ball [19] within the framework of the direct renormalization method introduced by des Cloizeaux [21]. Experimentally, however, one finds that the two chains have a tendency to segregate. Note that in actual experiment one never works exactly in the asymptotic limit. In dilute solution the degree of polymerization is never infinite. In terms of critical phenomena the segregation between  $P_1$  and  $P_2$  polymers is governed by a correction to scaling [19].

When the interaction between unlike monomers reaches a certain critical value the magnitude of which depends on the critical state of individual chains (shown by line *SS* in Figs. 6 and 8, respectively, for 4- and 5-simplex lattices), we find that the two chains form a zipped state in which the unlike monomers lie side by side.

For *TT* and *CC* states the chains are in a segregated state for  $u_3 \leq \sqrt{u_1(x_{1c})u_2(x_{2c})}$ . When  $u_3 = \sqrt{u_1(x_{1c})u_2(x_{2c})}$  the two chains form a state of interpenetration. In this state a polymer chain, say  $P_1$ , cannot be distinguished from  $P_2$ .

In the case of a selective solvent (i.e., when both chains are in different states) we find that they are segregated until  $u_3$  reaches a certain critical value. At the critical value of  $u_3$  the two chains become interpenetrated, but this interpenetration is different than the one found in the case of nonselective solvent. For example, for an *SC* state in which a chain (say  $P_1$ ) is in a swollen state and the other chain ( $P_2$ ) is in a compact globule phase, the interpenetrations means that chain  $P_1$ , while remaining in the swollen configuration, wraps around the compact globule configuration of chain  $P_2$ . This is similar to a surface adsorption transition.

In spite of the fact that the model developed here ignores the effect of one chain on the critical behavior of the other chain, it provides a qualitative description of

the phase diagram of a system of two polymer chains in a solution which may have different qualities for different chains. Our approach may serve as a starting point for more thorough investigations of segregation and entanglements in a real system.

#### ACKNOWLEDGMENTS

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