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Scaling of Spatial Correlations in Cooperative Sequential Adsorption with Clustering

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We examine solvable cooperative sequential adsorption models on a linear lattice where adsorption rates produce strong clustering or island formation. We show that the spatial pair correlations in this regime assume a scaled form for separations comparable to a characteristic length (which diverges in the strong clustering limit). This scaled form is also determined directly from consideration of appropriate solvable continuum grain growth models.

KEY WORDS: Coorperative sequential adsorption; nearest-neighbor exclusion; clustering; pair correlations.

Random and cooperative sequential adsorption (RSA and CSA) on lattices provide examples of nontrivial far-from-equilibrium processes where exact analysis is possible (at least in 1D).(1,2) Not only is the adsorption kinetics accessible, but so are the spatial correlations, which exhibit an intrinsically nonequilibrium form.(3,4) Explicit expressions for the pair correlations are available for 1D random dimer filling(4,5) and for 1D RSA (of monomers) with nearest-neighbor exclusion,(6) which reveal superexponential asymptotic decay. It has also been demonstrated that much more general 1D cooperative adsorption processes display this type of decay(4) and this feature is expected in all dimensions.(2,7)

Here we focus on CSA processes where adsorption near previously filled sites is strongly enhanced, and the associated clustering produces a characteristic or correlation length l_c of many lattice constants. Specifically we show that the pair correlations adopt a scaled form for separations

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$l = O(l_c)$, only crossing over to superexponential decay for $l \gg l_c$. This scaled form is determined explicitly for two lattice models noting that these reduce to solvable grain growth models in the strong clustering limit.

Below we let P_m (with $m \geqslant 1$) denote the probability of finding a string of m empty sites (so $\theta = 1 - P_1$ gives the fraction of filled sites or coverage) and $P_{m,n}(l) = P_{n,m}(l)$ that of finding strings of m and n empty sites ($m, n \geqslant 1$) separated by l lattice vectors [so, e.g., $P_{m,n}(0) = P_{m+n-1}$ and $P_{m,n}(1) = P_{m+n}$]. The two-point correlations are then given by $C(l) \equiv P_{1,1}(l) - (P_1)^2$. Thus $C(0) = \theta - \theta^2$ and $C(l) \to 0$ as $l \to \infty$. Below, ○ and ● represent an empty and a filled site, respectively. Finally, a denotes the lattice constant.

MODEL A

First consider CSA of monomers at empty sites on a linear lattice with rates k_i for sites with i occupied nearest neighbors (NN).[2-5,8] Thus k_0, k_1, and k_2 represent rates for island birth, growth, and coalescence, respectively (see Fig. 1). This model could describe autocatalytic or zipping reactions on polymer chains, or the kinetics of first-order transitions.[2] The typical size of a cluster of filled sites and also the correlation length, at a fixed coverage, will clearly diverge with increasing ratio of growth to birth rates, $r = k_1/k_0$.

Since the analysis of this model is described elsewhere,[2-4,8] we only sketch the key features in the text, relegating further details and some new observations to Appendix A. Empty pairs of sites have a ○○-Markov shielding property here.[2] This allows one to obtain a closed set of equations for P_1 and P_2 which determine the kinetics.[8] Further utilization of the shielding property shows that the $P_{2,2}(l)$ satisfy a closed set of equations allowing immediate determination. The $P_{2,1}(l)$ couple to themselves and $P_{2,2}(l)$ and thus can be determined next. The $P_{1,1}(l)$ couple to themselves and to $P_{2,1}(l)$ and $P_{2,2}(l)$, so are determined last. The latter are of primary interest since they determine the $C(l)$. Although the $P_{2,2}(l)$ may be expressed in terms of hypergeometric functions,[4] the additional steps

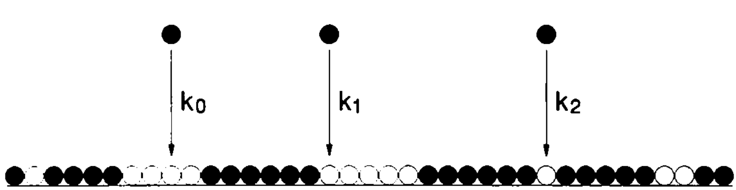

Fig. 1. Schematic for the deposition rates in model A.

required to obtain an expression for the $C(l)$ are very cumbersome (with the exception noted below). However, effectively exact $C(l)$ obtained by numerical integration of the rate equations are shown in Fig. 2.

Our key observation is that, in the large-r limit, the above lattice model coincides with a continuum model where grains are nucleated on the line at rate I per unit length (corresponding to k_0/a), and thereafter expand at constant velocity V (corresponding to $k_1 a$). Note that the mapping is independent of k_2, since coalescence is rare in the strong clustering limit. This continuum model was studied in D dimensions independently by Kolmogorov,[9] Johnson and Mehl[10] (JM), and Avrami.[11] For this process in $D=1$ the characteristic time $\tau=(IV)^{-1/2}$ (corresponding to $r^{-1/2}/k_0$) scales as the time needed to fill a fixed fraction of an initially empty line. The characteristic length $\xi=(V/I)^{1/2}$ (corresponding to $r^{1/2}a$) scales like the typical grain size, at fixed coverage.

Exact analysis of the continuum model[9-12] follows from the idea that for a point on the line to be *un*transformed at time t, no nucleation event can have occurred in the space-time "causal cone" (for velocity V) extending back from this point. It follows that the transformed (or filled) fraction of an initially empty line is given by $\theta^{\mathrm{JM}}=1-\exp[-(t/\tau)^2]$. The two-point correlations for separation x (corresponding to la) can also be determined, by an extension of this idea, as[12] $C^{\mathrm{JM}}(x,t)=F(x/\xi, t/\tau)$, where

$$\begin{aligned} F(y,s) &= \exp(-2s^2)\,\{\exp[(2s-y)^2/4]-1\} && \text{if} \quad y<2s \\ &= 0 && \text{if} \quad y\geqslant 2s \end{aligned} \tag{1}$$

Note that $C^{\mathrm{JM}}(0,t)=\theta^{\mathrm{JM}}(1-\theta^{\mathrm{JM}})$ and that the $C^{\mathrm{JM}}(x,t)$ are identically zero for separations large enough that the causal cones for the two points do not overlap.

As an immediate consequence of (1), it follows that the lattice correlations exhibit the scaling form

$$C(l)\sim F(r^{-1/2}l, r^{1/2}k_0 t) \qquad \text{as} \quad r\to\infty \tag{2}$$

Thus the correlation length for the lattice process (corresponding to ξ/a) is given by $l_c=r^{1/2}$. In Fig. 2 we choose $k_1=rk_0$, $k_2=(2r-1)k_0$, and show the convergence as $r\to\infty$ of the lattice model correlations to this nonzero scaled form for $l/l_c<2t/\tau$, for various coverages. For $l/l_c\geqslant 2t/\tau$, $C(l)$ approaches zero as $r\to\infty$ (for fixed l/l_c) as required, and decays super-exponentially as $l\to\infty$ (for fixed r).[2-4]

Of course the scaling form (2) for the correlations can be obtained from the exact solution of the lattice problem. We make two observations on this point. In the $r\to\infty$ strong clustering limit, *with θ fixed*, the

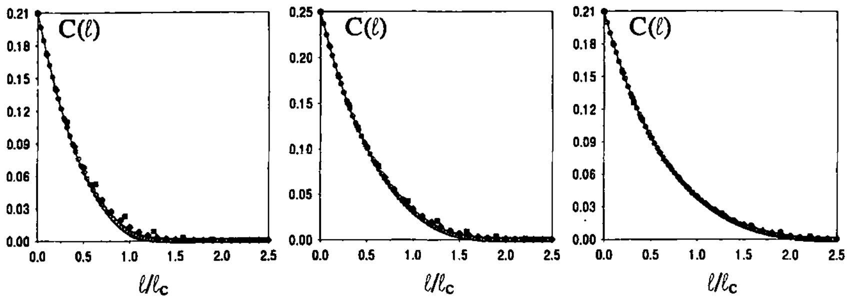

Fig. 2. The pair correlation $C(l)$ versus the scaled separation $l/l_c = r^{-1/2}l$ for model A with the rates $k_0 = 1$, $k_1 = r$, and $k_2 = 2r - 1$, where $r = 10$ (squares), 10^2 (diamonds), and 10^3 (circles). Here $\theta = 0.\ 3ML$ (left), $0.5ML$ (center), and $0.7ML$ (right). The solid line is the $r \to \infty$ limit.

probabilities of finding an empty site and an empty pair coincide (in other words, the probability of finding a filled neighbor of an empty site vanishes), so $P_{2,2}(l) - (P_2)^2$ and $C(l)$ must coincide [to within $O(r^{-1/2})$]. Indeed an asymptotic analysis of the hypergeometric expression for the former[4] does demonstrate convergence to the scaled form (1). Another useful observation draws on a result of Mityushin (see ref. 13 and Appendix A): if the k_i form an arithmetic progression, i.e., $k_1 = rk_0$ and $k_2 = (2r-1)\,k_0$, then only a single empty site is required to shield. While this yields somewhat simplified kinetics (as discussed in detail in ref. 3), the most dramatic and as yet unexploited simplification is seen in the determination of the correlations, which allows us to confirm directly the scaling form (1). See Appendix B.

MODEL B

Next we consider a model for CSA of monomers with NN exclusion on a linear lattice, where the adsorption rates k_i *now* depend on the number i of filled *second* NN sites.[14-16] Here k_0, k_1, k_2 correspond to the birth, growth, and coalescence of double-spaced islands of filled sites, respectively (see Fig. 3). Note that islands have a degeneracy of two, to which we can assign labels or phases α and β, say (see Fig. 3). Island coalescence as a result of two growing islands impinging is only possible if two islands are in phase. For example, after two β-phase islands impinge, the center empty site in ··○●○●○○○●○●○·· fills at rate k_2, thus forming a larger β-phase island. When out-of-phase islands (an α-phase and a β-phase island) meet, a permanent domain or antiphase boundary is formed, as in ··○●○●○○●○●○··. Thus the final jammed state of this model is nontrivial.

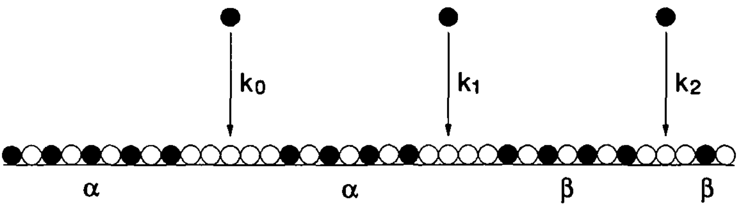

Fig. 3. Schematic for the deposition rates in model B. Island phases α and β are indicated.

This model mimics precursor-mediated island-forming chemisorption, where the adlayer typically has a superlattice structure.[16] Again the correlation length is determined by the ratio of island growth to birth rate, $r = k_1/k_0$. Exact analysis follows from a shielding property of quartets of empty sites.[14,15] Determination of the pair correlations $C(l)$ is more complicated than in model A only in detail, since now $P_{m,n}(l)$ with $m, n \leqslant 4$ must be considered.[4] It is clear that here the $C(l)$ will alternate in sign just as for an equilibrium lattice-gas model with NN repulsive interactions (or as in the antiferromagnetic Ising model).

For large r, behavior should again be described by a continuum model where grains are nucleated randomly at constant rate I (corresponding to k_0/a) and thereafter spread at constant velocity V (corresponding to $2k_1 a$). The difference from model A is that, upon nucleation, each grain is randomly assigned one of two phases, α or β, say (corresponding to the two phases of the double-spaced islands in the lattice model). Upon meeting, grains of like phase merge and those of opposite phase form a permanent domain boundary. The spatial correlations for this "two-state" Johnson–Mehl model[12,16] can also be determined exactly.[12] Here one starts by specifying pair probabilities $P^{\mathrm{JM}}_{\gamma,\delta}(x, t)$ for two points of various phases γ, $\delta = \alpha$ or β separated by distance x (corresponding to la) at time t. By symmetry, one has $P^{\mathrm{JM}}_{\alpha,\alpha} = P^{\mathrm{JM}}_{\beta,\beta}$ and $P^{\mathrm{JM}}_{\alpha,\beta} = P^{\mathrm{JM}}_{\beta,\alpha}$. These quantities are more complicated than the pair probabilities $P^{\mathrm{JM}}(x, t)$ for the standard JM model, but are related by

$$P^{\mathrm{JM}}(x, t) = P^{\mathrm{JM}}_{\alpha,\alpha}(x, t) + P^{\mathrm{JM}}_{\beta,\beta}(x, t) + P^{\mathrm{JM}}_{\alpha,\beta}(x, t) + P^{\mathrm{JM}}_{\beta,\alpha}(x, t) \tag{3}$$

Associated pair correlations are defined by $C^{\mathrm{JM}}_{\gamma,\delta} \equiv P^{\mathrm{JM}}_{\gamma,\delta} - P^{\mathrm{JM}}_{\gamma} P^{\mathrm{JM}}_{\delta}$ (so $C^{\mathrm{JM}}_{\alpha,\alpha} = C^{\mathrm{JM}}_{\beta,\beta}$ and $C^{\mathrm{JM}}_{\alpha,\beta} = C^{\mathrm{JM}}_{\beta,\alpha}$), where $P^{\mathrm{JM}}_{\gamma} = P^{\mathrm{JM}}_{\delta} = \{1 - \exp[-(t/\tau)^2]\}/2 = \phi$, say, with $\tau = (IV)^{-1/2}$ as above. Since $P^{\mathrm{JM}}(x, \infty) = 1$ at $t = \infty$, one can further show the "antisymmetry" property $C^{\mathrm{JM}}_{\alpha,\alpha} = C^{\mathrm{JM}}_{\beta,\beta} = -C^{\mathrm{JM}}_{\alpha,\beta} = -C^{\mathrm{JM}}_{\beta,\alpha}$ at $t = \infty$, a result that we shall exploit below. After reduction of the appropriate results in ref. 12, one obtains

$$\begin{aligned} C^{\mathrm{JM}}_{\alpha,\alpha}(x, t) &= [F(x/\xi, t/\tau) + G(x/\xi, t/\tau)]/4 \equiv C_e(x/\xi, t/\tau)/2 \\ C^{\mathrm{JM}}_{\alpha,\beta}(x, t) &= [F(x/\xi, t/\tau) - G(x/\xi, t/\tau)]/4 \equiv C_o(x/\xi, t/\tau)/2 \end{aligned} \tag{4}$$

with F defined as in model A, $\xi = (V/I)^{1/2}$ as before, and

$$\begin{aligned} G(y, s) &= 2\phi \qquad \text{if} \quad y = 0 \\ &= \left(\int_{y/2}^{y} du \int_{y-u}^{u} dw + \int_{y}^{s} du \int_{u-y}^{u} dw \right) e^{-S(y,u,w)} \\ &\quad + 2 \int_{y}^{s} du\,(u-y) \exp(-u^2) \qquad \text{if} \quad 0 < y < s \\ &= \int_{y/2}^{s} du \int_{y-u}^{u} dw\, e^{-S(y,u,w)} \qquad \text{if} \quad s < y < 2s \\ &= 0 \qquad \text{if} \qquad y \geqslant s \end{aligned} \tag{5}$$

where $S(y, u, w) = (3u^2 + 3w^2 - 2uw + 2yu + 2yw - y^2)/4$. The function G can be easily evaluated numerically. Finally note that $C^{\mathrm{JM}}_{\alpha,\alpha} + C^{\mathrm{JM}}_{\beta,\beta} + C^{\mathrm{JM}}_{\alpha,\beta} + C^{\mathrm{JM}}_{\beta,\alpha}$ reduces to C^{JM} in (1) as required.

Specific connection with the lattice model is more complicated here. One must first note that having two sites filled with l *even* (*odd*) in the lattice model corresponds to having two points covered by the *same* (*different*) phase in the continuum model. Thus, since $C_e = C_{\alpha\alpha} + C_{\beta\beta}$ and $C_o = C_{\alpha\beta} + C_{\beta\alpha}$, one concludes that, as $r \to \infty$, the lattice model correlations scale as

$$\begin{aligned} C(l) &\sim C_e((2r)^{-1/2} l, (2r)^{1/2} k_0 t) \qquad \text{for } l \text{ even} \\ &\sim C_o((2r)^{-1/2} l, (2r)^{1/2} k_0 t) \qquad \text{for } l \text{ odd} \end{aligned} \tag{6}$$

noting that the lattice coverage θ corresponds to ϕ (recalling that the islands are double-spaced). Again the choice of k_2 does not affect behavior

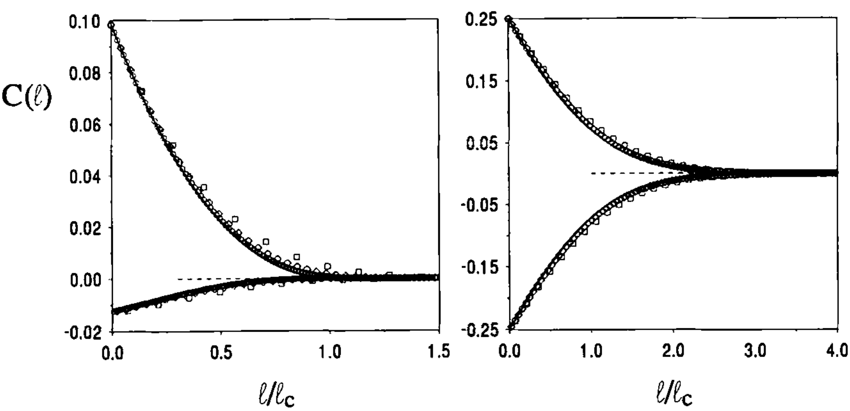

Fig. 4. The pair correlation $C(l)$ versus the scaled separations $l/l_c = (2r)^{-1/2} l$ for model B with the rates $k_0 = 1$, $k_1 = r$, and $k_2 = 2r - 1$, where $r = 10^2$ (squares), 10^3 (diamonds), and 10^4 (circles), at $0.11 ML$ (left) and at saturation (right). The solid line is the $r \to \infty$ limit.

as $r \to \infty$, where island coalescence is rare. The scaled forms on the RHS of (5) satisfy $C_e(l=0)=\theta(1-\theta)$, $C_o(l=0)=-\theta^2$, and $C_e(t=\infty)=-C_o(t=\infty)$ for all l, using the above "antisymmetry" property.

In Fig. 4 we show the convergence as $r \to \infty$ of the lattice model correlations (obtained from numerical integration of the rate equations, starting with an empty lattice) to the corresponding JM form at two different coverages. We have used a "Mityushin rate choice," $k_1 = rk_0$ and $k_2 = (2r-1)k_0$, for which only triplets of empty sites are required for shielding (see Appendix A). The observation that this choice of adsorption rates simplifies the shielding requirements (for model B) is new.

In summary, we have obtained the analytic scaling forms for the pair correlations in exactly solvable CSA models in the limit of strong clustering. This was achieved via consideration of appropriate continuum grain growth models. One might naturally extend this analysis to consider solvable 1D CSA models where islands have every Mth site filled (here we only treat $M=1$ and 2). Consideration of appropriate M-state JM models[12] reveals that the $C(l)$ adopt one (positive) scaling form for $l=0 \pmod{M}$ and another (negative) form for all other $l \neq 0 \pmod{M}$. There is natural interest in higher dimensions where the CSA models are not solvable, but the scaling form of the correlations can be determined exactly by virtue of the solvability of generalized JM models in all dimensions. The JM models must be "tuned" to incorporate appropriate island shape and possibly nonconstant expansion velocity. For 2D models of interest in chemisorption, simulation studies have revealed scaling behavior entirely analogous to the corresponding solvable 1D models discussed above.[16]

APPENDIX A: EMPTY-SITE SHIELDING, HIERARCHICAL TRUNCATION, AND THE "MITYUSHIN-TYPE" REDUCTION

Consider CSA models of monomer adsorption on a linear lattice with range $M-1$ exclusion, and where the adsorption rates k_i with $i=0$, 1, and 2 depend on the number i of filled Mth NN sites. It is well known[2,14] that strings of $2M$ consecutive empty sites can shield, allowing exact truncation of rate equations. One can further show that in fact only $2M-1$ empty sites are required for shielding when one chooses a "Mityushin-type" set of rates, forming an arithmetic progression.[3] One first considers the rate equations for the probabilities P_m with $m \geqslant 2M-1$:

[3] If the deposition rates for configurations of *one* filled Mth NN site are k_1 and k_1', say, for deposition at the right or left of the filled site, then the condition $k_0 - k_1 - k_1' + k_2 = 0$ produces a Mityushin-type reduction of shielding requirements.

$$-dP_{2M-1}/dt=(k_2-2k_1)\,P_{2M-1}-2(k_0-2k_1+k_2)\,P_{2M}$$
$$+(k_0-2k_1+k_2)\,P_{2M+1}$$
$$+2\sum_{j=0}^{M-1}\left[(k_0-k_1)\,P_{(2M-1)+j+1}+k_1P_{(2M-1)+j}\right] \quad (A.1)$$

and

$$-dP_{m\geqslant 2M}/dt=[m-2M]\,k_0P_m+2\sum_{j=0}^{M-1}\left[(k_0-k_1)\,P_{m+j+1}+k_1P_{m+j}\right] \quad (A.2)$$

Clearly, (A.1) reduces to the form of (A.2) for the Mityushin rates, and the "shielding identity" $P_{m+1}=QP_m$ with $Q=\exp(-k_0t)$, consistent with (A.2) for $m\geqslant 2M$, is then also satisfied for $m=2M-1$.

The above analysis does not guarantee that the shielding reduction extends to the probabilities $P_{m,n}(l)=P_{n,m}(l)$. Next we explicitly verify that indeed such an extension holds for $M=1$, and comment on the $M>1$ cases.

For $M=1$, one can use empty-doublet shielding to write any $P_{m,n}(l)$, with $l\geqslant 2$, in terms of $P_{1,1}(l)$, $P_{2,1}(l)$, and $P_{2,2}(l)$ only.[3,4] The latter satisfy a closed set of rate equations[3,4]

$$-dP_{1,1}(l)/dt=2\{k_2P_{1,1}(l)+(k_1-k_2)\,P_{2,1}(l)$$
$$+[k_1-k_2+(k_0-2k_1+k_2)\,Q]\,P_{2,1}(l-1)\} \quad (A.3)$$
$$-dP_{2,1}(l)/dt=[(2k_1+k_2)+(k_0-k_1)Q]\,P_{2,1}(l)$$
$$+(k_0-k_1)\,QP_{2,1}(l-1)+(k_1-k_2)\,P_{2,2}(l)$$
$$+[(k_1-k_2)+(k_0-2k_1+k_2)Q]\,P_{2,2}(l-1) \quad (A.4)$$

and

$$-dP_{2,2}(l)/dt=2\{[2k_1+(k_0-k_1)Q]\,P_{2,2}(l)+(k_0-k_1)\,QP_{2,2}(l-1)\} \quad (A.5)$$

For the Mityushin rate choice, terms containing the combination $k_0-2k_1+k_2$ drop out, and the coefficients k_0-k_1 and k_1-k_2 are identical. It is then a trivial matter to show that (A.3)–(A.5) are consistent with the identities $P_{2,2}(l)=Q^2P_{1,1}(l)$ and $P_{2,1}(l)=QP_{1,1}(l)$. Substitution of these identities into (A.3) immediately yields a closed set of equations for $P_{1,1}(l)$.

For $M>1$, the treatment is entirely analogous. One again proves consistency of the shielding property of $(2M-1)$-tuplets of empty sites with the identities $P_{m,n}(l)=Q^{m-(2M-1)}P_{2M-1,n}(l)$ for $m \geqslant 2M-1$ (and any $n \geqslant 1$) and $P_{m,n}(l)=Q^{n-(2M-1)}P_{m,2M-1}(l)$ for $n \geqslant 2M-1$ (and any $m \geqslant 1$).

APPENDIX B: TWO-POINT CORRELATIONS IN MODEL A WITH MITYUSHIN RATES

Here one can solve the closed set of rate equations for the $P_{1,1}(l)$ derived in Appendix A, using a standard generating function technique.[4] Alternatively, one can note that here $P_{2,2}(l)=Q^2P_{1,1}(l)$, so an expression for $P_{1,1}(l)$ follows directly from that obtained previously for $P_{2,2}(l)$ for general rate choices[4] (which is independent of k_2). Using either approach to determine $C(l)$, one obtains (for an initially empty lattice)

$$C(l=0)=Q^{2r-1}e^{(2r-2)(1-Q)}$$

$$C(l=1)=QC(l=0)$$

$$\begin{aligned}C(l \geqslant 2)=&\,Q^{(4r-2)}e^{(2r-2)(1-Q)}[(2r-2)^{l-1}(1-Q)^{l-1}/(l-2)!]\\&\times\left\{\int_0^1 dp\,[1-(1-Q)p]^{2-2r}(1-p)^{l-2}\right.\\&\left.-\int_0^1 dp\,e^{(2r-2)(1-Q)p}(1-p)^{l-2}\right\}\end{aligned} \tag{B.1}$$

Asymptotic analysis of (B.1), e.g., via steepest descent, confirms reduction to the scaled form in Eq. (1) for $l \simeq l_c = r^{1/2}$. For analysis of the $l \to \infty$ behavior at fixed r it is more convenient to expand $C(l \geqslant 2)$ as

$$\begin{aligned}&Q^{2-4r}e^{(2-2r)(1-Q)}[(2r-2)^{1-l}(1-Q)^{1-l}(l-2)!]\,C(l \geqslant 2)\\&=\sum_{k=0}^{\infty}[(2-2r)!//(2-2r-k)!k!-(2-2r)^k/k!]\\&\quad\times(Q-1)^k\int_0^1 dp\,p^k(1-p)^{l-2}\\&=(l-2)!\sum_{k=0}^{\infty}[(2-2r)!/(2-2r-k)!-(2-2r)^k]\\&\quad\times[(Q-1)^k/(l+k-1)!]\end{aligned} \tag{B.2}$$

using $\int_0^1 dp\, p^k(1-p)^{l-2} = k!(l-2)!/(l+k-1)!$. The term $k=2$ leads the sum giving

$$C(l) \sim Q^{4r-2} e^{(2r-2)(1-Q)} (2r-2)^l (1-Q)^{l+1}/(l+1)! \qquad \text{as} \quad l \to \infty \tag{B.3}$$

It is interesting to note[2] also that, for the Mityushin rate choice, the *filled*-cluster size distribution can be completely determined in terms of P_1 and $P_{1,1}(l)$ as a consequence of the shielding property of a single empty site.

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REFERENCES

1. M. C. Bartelt and V. Privman, *Int. J. Mod. Phys. B* **5**:2883 (1991).
2. J. W. Evans, *Rev. Mod. Phys.* **65**:1281 (1993).
3. N. A. Plate, A. D. Litmanovich, O. V. Noah, A. L. Toam, and N. B. Vasileyev, *J. Polymer Sci.* **12**:2165 (1974).
4. J. W. Evans, D. R. Burgess, and D. K. Hoffman, *J. Math. Phys.* **25**:3051 (1984).
5. N. O. Wolf, Ph.D. Thesis, Iowa State University (1979).
6. F. B. Pedersen and P. C. Hemmer, *J. Chem. Phys.* **98**:2279 (1993); C. Monthus and H. J. Hilhorst, *Physica A* **175**:263 (1991); J. W. Evans, *Surface Sci.* **215**:319 (1989).
7. T. M. Liggett, *Interacting Particle Systems* (Springer, Berlin, 1985).
8. J. B. Keller, *J. Chem. Phys.* **38**:325 (1963).
9. A. N. Kolmogorov, *Bull. Acad. Sci. USSR* **3**:355 (1937).
10. W. A. Johnson and R. F. Mehl, *Trans. AIME* **135**:416 (1939).
11. M. Avrami, *J. Chem. Phys.* **7**:1103 (1939); **8**:212 (1940); **9**:117 (1941).
12. S. Ohta, T. Ohta, and K. Kawasaki, *Physica A* **140**:478 (1987).
13. L. G. Mityushin, *Prob. Peredachi Inf.* **9**:81 (1973).
14. J. J. Gonzalez, P. C. Hemmer, and J. S. Høye, *Chem. Phys.* **3**:288 (1974).
15. J. W. Evans and D. R. Burgess, *J. Chem. Phys.* **79**:5023 (1983).
16. J. W. Evans, R. S. Nord, and J. A. Rabaey, *Phys. Rev. B* **37**:8598 (1988).