

# Motion and Capture in the Presence of Cooperative Trap Interactions II: Exact Calculations for Perfect Absorbers in One Dimension<sup>1</sup>

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*Received October 18, 1983; revision received January 10, 1984*

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Exact calculations are presented for the survival fraction and other capture-related observables in a problem involving migration on a one-dimensional lattice containing perfectly absorbing traps whose placement within the crystal is determined by explicit interactions among the trap molecules. These interactions, which are treated exactly in a lattice-gas model, may be attractive, in which case they lead to trap cluster formation and a reduction of the trapping efficiency, or they may be repulsive thus leading to increased separation of the trap molecules and an enhancement of the trapping process.

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**KEY WORDS:** Master equation; trapping; Ising models; lattice gas; cooperative interactions.

## 1. INTRODUCTION

There has been considerable work done in recent years aimed at describing the trapping of particles which move on a crystalline lattice,<sup>(1-12)</sup> as in, e.g., experiments involving sensitized luminescence. In all this work it is generally assumed that the placement of traps in the crystal is random. We have recently begun to study the consequences of possible interactions among the traps, which would tend to affect their placement in the crystal and introduce correlations among the trap positions. We have introduced such interactions in a phenomenological manner,<sup>(13)</sup> and have also analyzed

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<sup>1</sup> Supported in part by the National Science Foundation under Grant Nos. DMR-8111434 and INT-8210098.

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them exactly<sup>(14)</sup> for a one-dimensional lattice-gas model. In these analyses the transport and capture processes are treated in a way which, although approximate,<sup>3</sup> is valid for arbitrary dimensionality, degree of transport coherence, and range of motion. It is, furthermore, capable of treating traps which may not be perfect absorbers. In the present paper, however, we restrict ourselves to perfectly absorbing traps and one-dimensional incoherent motion. As with randomly placed traps, a simplification occurs when this restriction is imposed, which makes it possible to obtain an *exact* solution to the dynamical problem.<sup>(11,12,15)</sup> We are able, therefore, to use the treatment of trap-trap interactions presented in Ref. 14, hereafter referred to as I, to study exactly the problem of particles migrating on a one-dimensional lattice in the presence of a *nonrandom* distribution of perfectly absorbing, deep traps.<sup>4</sup> The present paper thus complements the analyses presented in Refs. 13 and 14 and helps provide insight into the manner in which phenomena such as trap cluster formation can affect the proper interpretation of experimental results.

## 2. MODEL AND GENERAL SOLUTION

We assume that the transport particles in our model obey the following evolution equation for the probability  $P_m(t)$  that a particle occupies the  $m$ th site in the crystal at time  $t$ :

$$\frac{dP_m(t)}{dt} = -2FP_m(t) + F[P_{m+1}(t) + P_{m-1}(t)] - c_m P_m(t) \quad (2.1)$$

Here,  $F$  is the hopping rate from site  $m$  to its neighbors and the quantities  $c_m$  are random variables governing the decay of probability from the sites  $m$  due to the presence of traps. Since we treat the traps as perfect absorbers, the  $c_m$  take on the value zero when  $m$  is a host site and are infinite when it is a trap. In the random case these occur with weights  $(1 - \rho)$  and  $\rho$ , the host and trap concentrations, respectively. This result stems from the fact that the probability for a given site being occupied by a trap molecule is independent of the positions of the other traps. When interactions exist between trap molecules, however, the result is not valid. It is this case that we treat in this paper and in I.

The feature of the problem of motion and trapping on a one-dimensional lattice with perfectly absorbing traps which makes possible an

<sup>3</sup> Although approximate for general trap interactions, the theory developed in Ref. 14 is exact for *periodically* placed traps.

<sup>4</sup> We use the term "deep" to refer to the absence of detrapping rather than to the infinite rate associated with the trapping process itself (for which we reserve the term "perfectly absorbing"). This is in contrast to the usage of, e.g., Ref. 11.

exact solution<sup>(11,12,15)</sup> is that the traps divide the host sites into isolated clusters. Any particle starting within such a host cluster is cut off from the rest of the crystal. The survival fractions of host particles in the crystal may, therefore, be simply expressed as an average over host clusters, viz.

$$n(t) = \sum_{N=1}^{\infty} w_N n_N(t) \tag{2.2}$$

in which  $w_N$  is the probability that an arbitrary host site is part of a host cluster of size  $N$  and the quantity  $n_N(t)$  is the survival fraction for one such cluster with the initial condition that all sites of the cluster are populated equally with probability  $1/N$ . By a host cluster of size  $N$  we mean a group of  $N$  adjacent host sites with a trap at each end.

With perfectly absorbing traps, a cluster of  $N$  host sites with a trap at each end is dynamically equivalent to a *ring* containing one trap site and  $N$  host sites. The latter problem may be solved<sup>(4)</sup> straightforwardly through, e.g., the defect technique of Montroll<sup>(16)</sup> resulting in

$$\tilde{n}_N(\epsilon) = \frac{1}{\epsilon} \left[ 1 - \frac{1 - \epsilon \tilde{\psi}_0^{(N+1)}}{N \epsilon \tilde{\psi}_0^{(N+1)}} \right] \tag{2.3}$$

in which tildes denote Laplace transforms with Laplace variable  $\epsilon$ , and the quantity  $\psi_0^{(N)}(t)$  is, for a ring of  $N$  host sites containing no traps, the probability that a particle located initially at any site will be there at a time  $t$  later. Of the several known forms of this quantity we use the compact one,<sup>(5,13,17)</sup>

$$\epsilon \tilde{\psi}_0^{(N)}(\epsilon) = \frac{\tanh(\xi'/2)}{\tanh(N\xi'/2)} \tag{2.4}$$

in which  $\cosh \xi' = 1 + \epsilon/2F$ .

It is clear from (2.2) that, with  $n_N(t)$  given by (2.3) and (2.4), we require only the weights  $w_N$  in order to obtain an explicit solution for the survival fraction. The evaluation of these quantities requires a knowledge of the form of the trap-trap interaction. The description of this interaction in terms of a lattice-gas model and the resulting calculation of the  $w_N$  comprise the next section.

### 3. CLUSTER PROBABILITIES IN THE LATTICE-GAS MODEL

When no interactions exist among the trap molecules,  $w_N$  may be expressed as the product of the independent probabilities for the interior sites being occupied by host molecules, the probabilities for the two end sites being occupied by traps, and the number of distinct host clusters of

size  $N$  in which a specified host site may be located, i.e.,

$$w_N = N\rho^2(1 - \rho)^{N-1} \quad (3.1)$$

With (3.1), (2.3), and (2.4) substituted in (2.2) the result for the random distribution of traps is obtained, *viz.*

$$\tilde{n}(\epsilon) = \frac{1}{\epsilon} \left\{ 1 - \sum_{N=1}^{\infty} \rho^2(1 - \rho)^{N-1} \left[ \frac{\tanh[(N+1)\xi'/2] - \tanh(\xi'/2)}{\tanh(\xi'/2)} \right] \right\} \quad (3.2)$$

We show in Appendix A how this expression is equivalent to the result obtained earlier by Movaghar *et al.*<sup>(11)</sup> in their analysis of randomly placed traps.

In the presence of interactions among the trap molecules, (3.1) no longer gives the correct form for the cluster probabilities. These quantities must, therefore, be calculated from our model of trap-trap interactions. The approach that we adopt is detailed in I. It consists of using a lattice-gas model in which there is an interaction energy associated with every pair of trap molecules in the system, and of utilizing the well-known relationship between lattice gases and the Ising model. The interaction energy is infinite if two trap molecules occupy the same site, takes on the value  $-\Delta$  if they are nearest neighbors, and is zero otherwise. Clearly, positive values of  $\Delta$  correspond to attractive interactions and negative values correspond to repulsive ones. To obtain expressions for the trapping observables it is necessary to calculate  $n$ -point correlation functions in the trap system. While two-point correlations suffice in the treatment of I, the present analysis requires certain higher-order correlation functions.

In the one-dimensional lattice gas, a given configuration of trap and host sites can be represented by the set  $\{\sigma_1, \sigma_2, \sigma_3, \dots\}$ , where the  $\sigma_m$  are Ising model spin variables taking on the value 1 if site  $m$  is a trap site and  $-1$  if it is a host. The probability for a particular configuration to occur is given by  $Z^{-1} \exp[-\beta U(\sigma_1, \sigma_2, \dots)]$  in which  $\beta = 1/k_B T$ ,  $k_B$  is Boltzmann's constant,  $T$  is the appropriate temperature,  $U(\sigma_1, \sigma_2, \dots)$  is the energy associated with the configuration as determined by the interaction energy described earlier, and  $Z$  is the partition function for the system,

$$Z = \sum_{\{\sigma\}} \exp[-\beta U(\sigma_1, \sigma_2, \dots)] \quad (3.3)$$

If we let  $g_N$  denote the probability that  $N + 2$  adjacent sites in the crystal make up a host cluster of size  $N$ , then

$$g_N = \left\langle \left( \frac{\sigma_1 + 1}{2} \right) \left( \frac{|\sigma_2 - 1|}{2} \right) \left( \frac{|\sigma_3 - 1|}{2} \right) \dots \left( \frac{|\sigma_{N+1} - 1|}{2} \right) \left( \frac{\sigma_{N+2} + 1}{2} \right) \right\rangle \quad (3.4)$$

The angular brackets denote expectation values taken over the density matrix, i.e., if  $A$  is any function of the  $\sigma_m$  then

$$\langle A \rangle \equiv Z^{-1} \sum_{\{\sigma\}} A(\sigma_1, \sigma_2, \dots) \exp[-\beta U(\sigma_1, \sigma_2, \dots)] \quad (3.5)$$

In Appendix B it is shown that the expectation value (3.4) is given by

$$g_N = (1 - \rho)\rho^2(1 - x)^2[1 - \rho(1 - x)]^{N-1} \quad (3.6)$$

The quantity  $x$  has been introduced in I as

$$x = \frac{[1 - 4\rho(1 - \rho)(1 - e^{-\beta\Delta})]^{1/2} - 1}{[1 - 4\rho(1 - \rho)(1 - e^{-\beta\Delta})]^{1/2} + 1} \quad (3.7)$$

From the definitions of  $w_n$  and  $g_N$  it is clear that they are related through  $w_n = (1 - \rho)^{-1}Ng_N$  since  $(1 - \rho)$  is the probability that an arbitrary site is occupied by a host molecule. We have, therefore, for the weights appearing in (2.2),

$$w_N = N\rho^2(1 - x)^2[1 - \rho(1 - x)]^{N-1} \quad (3.8)$$

#### 4. RESULTS AND DISCUSSION

Equation (3.8) is remarkable in its simplicity. Comparison with (3.1) shows that the cluster probabilities in the presence of interactions among trap molecules may be obtained from those which apply when no interactions are present, simply by replacing the trap concentration  $\rho$  by an *effective* concentration  $\rho_e = \rho(1 - x)$ . Moreover, this clearly holds for *all* the relevant trapping observables as well, since we see from (2.2) that the  $\rho$  dependence of the survival fraction is contained entirely in the quantities  $w_N$ . After substitution into (2.2) we have, for the survival fraction in the presence of trap interactions,

$$\tilde{n}(\epsilon) = \frac{1}{\epsilon} \left\{ 1 - \sum_{N=1}^{\infty} \rho_e^2(1 - \rho_e)^{N-1} \left[ \frac{\tanh[(N+1)\xi'/2] - \tanh(\xi'/2)}{\tanh(\xi'/2)} \right] \right\} \quad (4.1)$$

The effective concentration  $\rho_e$  is plotted in Fig. 1 as a function of the actual trap concentration for different values of the trap interaction parameter  $E \equiv \exp(\beta\Delta)$ . Note that the effective concentration is increased when the trap interaction is repulsive ( $E < 1$ ) and is decreased when the interaction is attractive ( $E > 1$ ).

It should be clear from this discussion that the *form* of the decay of the survival fraction is unchanged in the presence of trap interactions, but

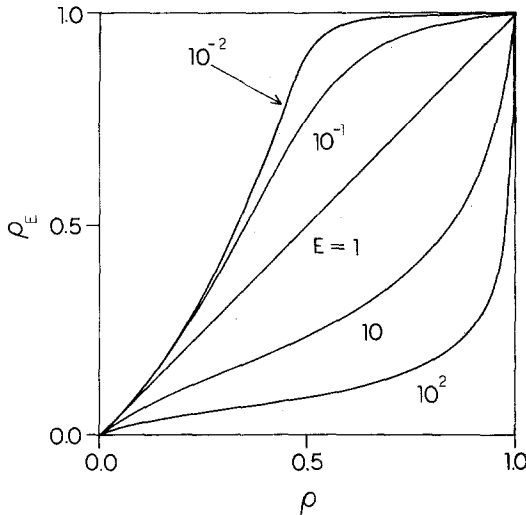


Fig. 1. Effective trap concentration  $\rho_e$  as a function of the actual trap concentration  $\rho$  with values of the trap interaction parameter  $E$  as indicated.

that a rescaling of the concentration occurs. Thus the result  $n(t) \sim \exp[-(t/\tau_0)^{1/3}]$  of Movaghar *et al.*,<sup>(11)</sup> where  $\tau_0$  is an appropriate characteristic time, continues to hold, but with a rescaled value of  $\tau_0$ . The rescaling enhances the trapping process when repulsive interactions are present ( $\rho_e$  is increased), and inhibits the trapping process when attractive interactions are present ( $\rho_e$  is decreased). This is qualitatively as one would expect since attractive interactions would lead to an increase in the average size of *trap* clusters. For perfectly absorbing traps no interior members of a trap cluster are ever involved in the trapping process. Hence the effective number of traps actually capable of trapping goes down as the average trap cluster size increases. It is also clear, therefore, why repulsive interactions increase the trapping efficiency: they lead to a decrease in the average size of trap clusters and an increase in the fraction of trap molecules which are *not* interior members of trap clusters. This change in the trapping efficiency with changing interaction strength has also been discussed in I.

In Figs. 2 and 3 this behavior is demonstrated in a numerical inversion<sup>(18)</sup> of (4.1) for two different trap concentrations. In Fig. 2  $\rho = 10^{-2}$  and in Fig. 3  $\rho = 0.5$ . The different curves in each figure correspond to different values of the trap molecule interaction parameter  $E$ , the curves with  $E = 1$  describing the case of no interactions studied previously.<sup>(11,12,15)</sup> For the trap concentration  $\rho = 0.5$  we note that as the interaction energy increases to positive infinity (the limit of very strong repulsive interactions)

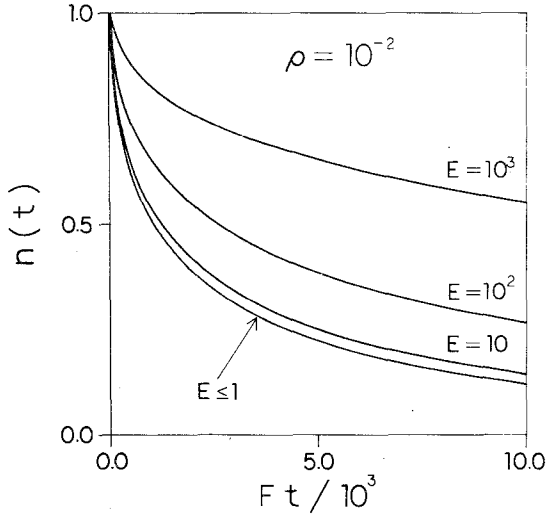


Fig. 2. Survival fraction  $n(t)$  as a function of the dimensionless time  $Ft$ , with trap concentration  $\rho = 10^{-2}$  and values of the trap interaction parameter  $E$  as indicated. Note that at this concentration the curves corresponding to values of  $E \leq 1$  (repulsive interactions) are indistinguishable.

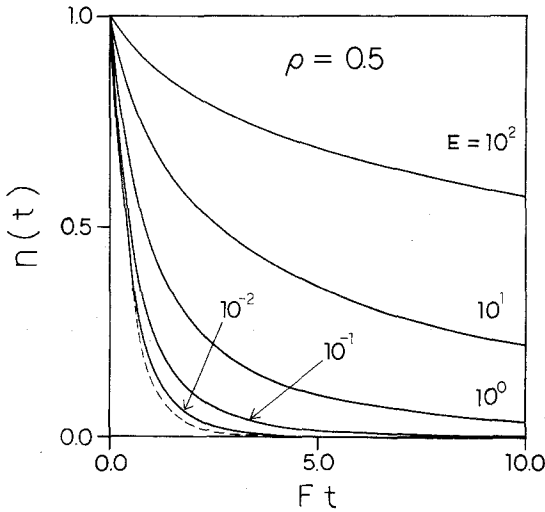


Fig. 3. Survival fraction of particles  $n(t)$  as a function of the dimensionless time  $Ft$ , with trap concentration  $\rho = 0.5$  and with values of the trap interaction parameter  $E$  as indicated. The dashed curve is exponential and corresponds to the limiting value  $E = 0$ .

the traps must occupy alternate positions within the lattice so as to avoid being nearest neighbors. When this happens the largest host cluster is of size one and the subsequent decay of the survival fraction is given by the exponential  $n(t) = \exp(-2Ft)$ , with each (isolated) host site decaying to the two traps which are its neighbors. The dashed curve in Fig. 3 represents this exponential decay and, indeed, one sees that as  $E$  approaches zero, the curves describing the host decay do approach the exponential limit.

In exciton trapping experiments, e.g., sensitized luminescence, the survival fraction is monitored through the detection of photons emitted by the hosts or the traps as they decay radiatively to the ground state. Radiative decay has not been included in (2.1) for reasons of simplicity and also to make the analysis relevant to particles other than Frenkel excitons. However, it has been shown elsewhere<sup>(4)</sup> that such effects may be accounted for by adding the term  $P_m(t)/\tau$  to the left-hand side of (2.1), by multiplying (2.2) by  $\exp(-t/\tau)$ , or by replacing  $\epsilon$  with  $\epsilon + 1/\tau$  in the corresponding transformed expressions. When this is done, the host and trap quantum yields  $\phi_H$  and  $\phi_G$ , defined to be that fraction of the initial excitation which decays radiatively from host and traps sites, respectively, can be obtained without inversion from (4.1) through the relation

$$\phi_H = \int_0^\infty dt \frac{n(t)}{\tau} e^{-t/\tau} = \frac{1}{\tau} \tilde{n}\left(\frac{1}{\tau}\right) \quad (4.2)$$

and by conservation of probability through  $\phi_G = 1 - \phi_H$ . Thus, for example, we have the following explicit expression for  $\phi_G$ :

$$\phi_G = -\rho_e + \rho_e^2 \sum_{N=1}^{\infty} \frac{(1 - \rho_e)^{N-1} \tanh[(N+1)\xi/2]}{\tanh(\xi/2)} \quad (4.3)$$

in which  $\cosh \xi = 1 + 1/2F\tau$ . In Fig. 4 we present plots of  $\phi_G$  as a function of  $\rho$  with the same values of the parameter  $E$  as in Fig. 1.

Although the analysis of I has a greater range of applicability than the present one because of its ability to treat arbitrary degree of coherence and range in the motion and arbitrary capture rates, it involves an approximation procedure. On the other hand, the theory presented here, although restricted to nearest-neighbor incoherent motion and to infinite capture rates  $c$ , involves no approximations. It is therefore of interest to test the validity of the limiting form of I which corresponds to the present analysis. To this end we present Fig. 5, which shows a comparison of the guest yield  $\phi_G$  as predicted by I and by the present treatment. The dashed curves in Fig. 5 correspond to the limit  $c \rightarrow \infty$  of Eqs. (1.6) and (3.22) of I. The solid curves correspond to Eq. (4.3) of the present paper. The values of the interaction parameter  $E$  are as indicated. We observe that the appropriate



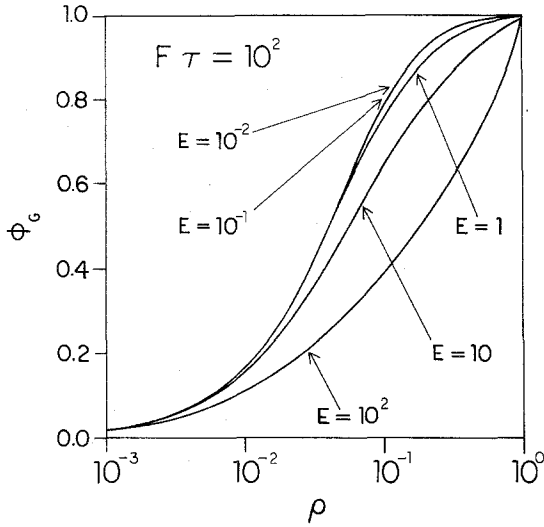


Fig. 4. Trap yield  $\phi_G$  plotted against trap concentration with values of the trap interaction parameter  $E$  as indicated. We have taken  $\tau = 10^2/F$ . Note that the curves for values of  $E \leq 0.1$  are indistinguishable for this choice of  $\tau$ .

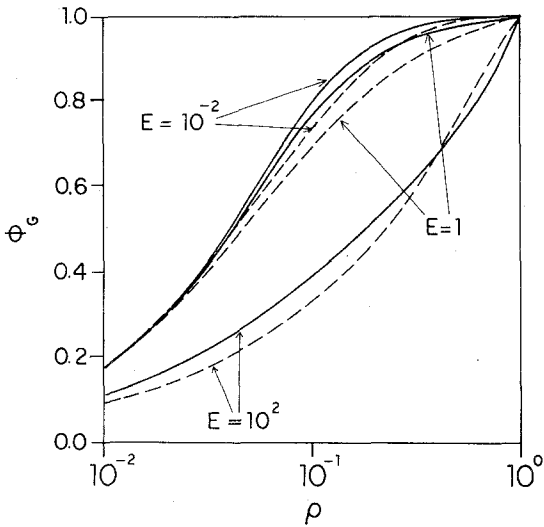


Fig. 5. Trap yield  $\phi_G$  vs. trap concentration  $\rho$  as predicted by the theory of I (dashed curves) and the theory of this paper (solid curves). Values of  $E$  are as indicated and  $\tau = 10^2/F$ .

limit of the theory contained in I is in reasonable quantitative and excellent qualitative agreement with the exact results presented here.

The purpose of this paper has been to present the result of exact calculations which include the effects of trap-trap interactions on the observables related to the migration and trapping of excitations on a one-dimensional lattice. We have shown how, for a simple lattice-gas model of these interactions, the total intermolecular interaction may be accounted for exactly by rescaling the trap concentration. It is hoped that calculations of the sort presented here and in I will be of use in determining the magnitude of effects such as trap cluster formation on capture phenomena.

## APPENDIX A

In this appendix we wish to demonstrate the equivalence of the form given in (3.2) for the survival fraction with that obtained by other authors.<sup>(11)</sup> To accomplish this we first introduce the quantity  $h(\epsilon) \equiv \exp(-\xi')$  and reexpress the hyperbolic functions of (2.4) in terms of sums of powers of  $h(\epsilon)$ . We obtain after some rearrangement

$$\tilde{n}(\epsilon) = \frac{1}{\epsilon} - \frac{2\rho^2}{\epsilon} \sum_{N=1}^{\infty} \frac{(1-\rho)^{N-1} h(h^N - 1)}{(1-h)(1+h^{N+1})} \quad (\text{A1})$$

Now, expanding the factor of  $(1+h^{N+1})$  which appears in the denominator of the summation in (A1) as a geometric series we obtain

$$\epsilon \tilde{n}(\epsilon) - 1 = \sum_{k=0}^{\infty} \sum_{N=1}^{\infty} \frac{2\rho^2 h^{k+1} (-1)^k}{1-h} (h^{N(k+1)} - h^{Nk})(1-\rho)^{N-1} \quad (\text{A2})$$

The sum over  $N$  may now easily be performed yielding

$$\epsilon \tilde{n}(\epsilon) - 1 = - \sum_{k=0}^{\infty} \frac{2\rho^2 h^{2k+1} (-1)^k}{[1 - (1-\rho)h^k][1 - (1-\rho)h^{k+1}]} \quad (\text{A3})$$

The summand may be reexpressed, however, by making use of the identity

$$\begin{aligned} & \frac{2h^{2k+1}}{[1 - (1-\rho)h^k][1 - (1-\rho)h^{k+1}]} \\ &= \left[ \frac{h^{k+1}}{1 - (1-\rho)h^{k+1}} - \frac{h^k}{1 - (1-\rho)h^k} \right]^2 \\ & \quad - \frac{h^{2k+2}}{[1 - (1-\rho)h^{k+1}]^2} - \frac{h^{2k}}{[1 - (1-\rho)h^k]^2} \end{aligned} \quad (\text{A4})$$

Notice that the last two terms on the right-hand side (A4) become identical

when  $k$  is replaced by  $k - 1$  in the first of them. Because of the oscillating factor of  $(-1)^k$  which multiplies them, these two terms will cancel each other when substituted into the sum (A3). The only contribution will come from the unpaired  $k = 0$  term, which has the value  $-1$ . Combining this with the first term on the right-hand side of (A4) we obtain after some straightforward algebra

$$\tilde{n}(\epsilon) = \sum_{k=0}^{\infty} \frac{\rho^2 h^{2k} (1-h)^2}{\epsilon [1 - (1-\rho)h^k]^2 [1 - (1-\rho)h^{k+1}]^2} \tag{A5}$$

which is identical to equation (6) of Ref. 11.

**APPENDIX B: EVALUATION OF CLUSTER PROBABILITIES**

The one-dimensional lattice gas is equivalent to a one-dimensional Ising model<sup>(19)</sup> with configuration energy  $U(\sigma_1, \sigma_2, \dots)$  given by

$$U(\sigma_1, \sigma_2, \dots) = -J \sum_{i=1}^n \sigma_i \sigma_{i+1} - B \sum_{i=1}^n \sigma_i \tag{B1}$$

where  $\sigma_{n+1} \equiv \sigma_1$ . The quantity  $J$  is related to  $\Delta$  by the relation  $4J = \Delta$  and for a large system ( $n \rightarrow \infty$ ) the magnetization  $M$  is<sup>(20)</sup>

$$M = \sinh \beta B [\sinh^2 \beta B + e^{-4\beta J}]^{1/2} \tag{B2}$$

The equivalence  $M = 2\rho - 1$  gives  $B$  as a function of  $\rho$  and  $\Delta$ , the variables of the lattice gas model. For the Ising model, the quantities  $g_N$  can be written

$$g_N = \frac{1}{\text{Tr} P} \text{Tr} \left\{ P^{n-N-1} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} P \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \left[ P \begin{pmatrix} e^{-2\beta B} & 0 \\ 0 & 0 \end{pmatrix} \right]^{n-1} P \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \right\} \tag{B3}$$

where Tr indicates a trace and the matrix  $P$  is given by

$$P = \begin{pmatrix} e^{\beta(J+B)} & e^{-\beta J} \\ e^{-\beta J} & e^{\beta(J-B)} \end{pmatrix} \tag{B4}$$

Equation (B3) can be evaluated exactly in the limit  $n \rightarrow \infty$  yielding

$$g_N = \frac{1}{8} (1 + M^2)(1 - M)(1 - x)^2 [1 - (1/2)(1 - x)(1 + M)]^{N-1} \tag{B5}$$

in which

$$x = \frac{[1 - (1 - M^2)(1 - e^{4\beta J})]^{1/2} - 1}{[1 - (1 - M^2)(1 - e^{4\beta J})]^{1/2} + 1} \tag{B6}$$

Substituting  $\Delta$  for  $4J$ , and  $2\rho - 1$  for  $M$  in (B5) and (B6) gives (3.6) and (3.7).

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