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Lattice Gas Activity Series from Secular Equations

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The secular equations for finite tori of lattice sites are obtained by computer expansion of determinants for a hard-particle lattice gas. The secular equations yield all of the thermodynamic functions for finite systems and the beginning terms of activity expansions for all of the eigenvalues of the infinite system. The same secular equation that yields the low-density series for the equation of state of the infinite-circumference system also yields the beginning terms in the high-density expansion. As examples we treat two hard-particle lattice gases in two dimensions, the lattice gas with nearest-neighbor exclusion (which has a second-order transition), and the case of dimers (which is analytic all the way to close packing).

KEY WORDS: Dimer entropy; high- and low-density activity series; lattice gases; second-order transitions; secular equations; symmetry-reduced transfer matrices.

1. INTRODUCTION

The scaling of the behavior of finite systems to predict the behavior of infinite systems is an important theme in modern statistical mechanics.[1-3] Here we treat finite strips of lattice gases and extract the beginning terms in the activity series for the pressure for the infinite system. We have previously explored this approach and have shown how to extract series from transfer matrices.[4] Here we give a different, simpler method, namely the explicit generation of the secular equation for a finite strip of lattice gas. We will show that one can obtain the beginning coefficients in the equation of state of the infinite system in both the high- and low-density limits. In addition, we can obtain all of the other eigenvalues as activity series. Given

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the explicit secular equation, one can also obtain all of the thermodynamic properties without having to take numerical derivatives, the only numerical task being the extraction of the largest eigenvalue. We begin by treating the case of hard particles with nearest-neighbor exclusion on the plane-square lattice. This system exhibits an order–disorder critical point (second-order transition); at close packing the system must exist in one of two perfectly ordered arrangements. We will also treat the case of dimers on the same 2D lattice. In this case there is no ordering transition as one approaches close packing, and the limit of close packing is characterized by random packing (with a characteristic entropy per particle).

Consider a lattice gas in two dimensions. The grand partition function for an $M \times L$ strip of lattice sites (as illustrated for two alternate lattice orientations in Fig. 1) with periodic boundary conditions in both directions (producing a torus) is given exactly as the trace of a matrix product[4–6]

$$\Xi(L, M) = \mathrm{Tr}\, \mathbf{W}(M)^L = 1 + Q_1 z + Q_2 z^2 + \cdots \qquad (1.1)$$

where $\mathbf{W}$ is the appropriate transfer matrix that correlates the states of a given column of M sites with those of the neighboring column (or columns, depending on the range of the interaction between the particles). For hard-particle systems (the only interaction being excluded volume), which we will restrict ourselves to here, $\mathbf{W}$ is a function of the activity z of the

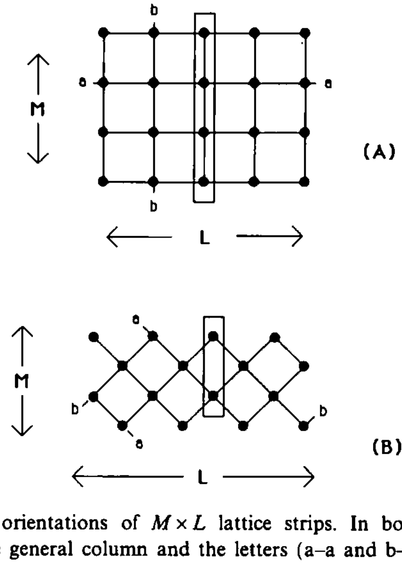

Fig. 1. Two different orientations of $M \times L$ lattice strips. In both, the square rectangle encloses the sites of the general column and the letters (a–a and b–b) indicate the nature of the periodic boundary conditions producing a torus. (A) The lattice axes are perpendicular to the axis of the torus. (B) The lattice axes are tilted by 45° to the axis of the torus.

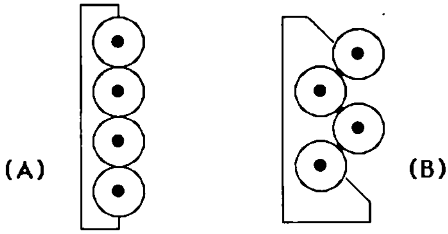

Fig. 2. Illustration of the fact that four particles result in an interaction going around the diameter of the torus in both lattice orientations (A) and (B) shown in Fig. 1. Since this interaction is impossible on the infinite square lattice, the two torii give the activity series of the infinite system only through three terms.

particles and the circumference M of the torus; the Q_n in (1.1) are the number of ways of placing n particles on a lattice of M sites.

We can express Ξ in terms of the eigenvalues Λ of the matrix $\mathbf{W}$

$$\Xi = \sum_{k=1}^{\sigma} \Lambda_k^L \tag{1.2}$$

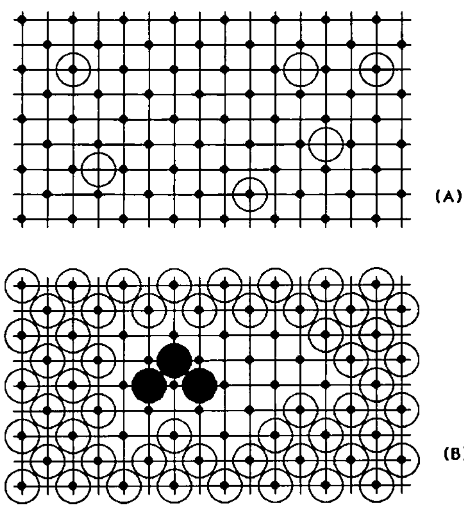

Fig. 3. Sample low- and high-density particle configurations for the hard-particle lattice gas with nearest-neighbor exclusion. The solid dots indicate one sublattice in each case. (A) Low density. Particles randomly occupy both sublattices with equal probability. (B) High density. At close-packing, particles are exclusively on one of the two sublattices. As particles are removed and pools of empty sites are formed, particles can "break off" from the main sublattice and "float" out onto the other sublattice (illustrated by the shaded particles).

where the number of eigenvalues σ depends on the size of the matrix **W**, which in turn depends on M, the circumference of the torus. In the limit of large L, Ξ will depend only on Λ_1, the largest eigenvalue:

$$\lim_{L \to \infty} L^{-1} \ln \Xi = \ln \Lambda_1 \tag{1.3}$$

Now Λ_1 is the eigenvalue per column of sites (as defined in Fig. 1). Defining the eigenvalue per lattice site as

$$\lambda_1 = \Lambda_1{}^{1/M} \tag{1.4}$$

then we have that (1.3) becomes

$$N^{-1} \ln \Xi = \ln \lambda_1 \tag{1.5}$$

where N is the total number of lattice sites (analog of the volume)

$$N = M \times L \tag{1.6}$$

Since $\ln \Xi = pN/kT$ (where p is the pressure and $1/kT$ has the usual meaning), one has

$$p/kT = N^{-1} \ln \Xi = \ln \lambda_1 = \sum_{n=1}^{\infty} b_n z^n \tag{1.7}$$

where the sum is the standard Mayer activity expansion for the pressure.

Thus knowledge of Λ_1 (or equivalently, λ_1) yields the equation of state. It is well known[4] that the coefficients b_n in (1.7) obtained from a matrix $\mathbf{W}(M)$ for a finite torus give (for the case of nearest-neighbor interactions) the exact b_n for the infinite system through $n \leqslant (M-1)$ for the arrangement in Fig. 1A and through $n \leqslant (2M-1)$ for arrangement in Fig. 1B (the coefficients are correct for n up to one minus the number of particles required to go around the torus).

There are many techniques known[4,7] for the calculation of the beginning b_n in (1.7) and hence for obtaining the activity expansion of Λ_1 (or λ_1). Here we give a method for calculating the activity expansion of the other eigenvalues. It is based on the fact that all of the eigenvalues can be calculated as activity series if one knows the appropriate secular equation

$$|\mathbf{W} - \Lambda \mathbf{I}| = 0 \tag{1.8}$$

where **I** is the identity matrix. As the circumference of the torus is increased, **W** becomes very large and it is difficult to obtain (1.8) as an explicit finite polynomial in powers of Λ and z. For hard-particle systems one can obtain the explicit form of (1.8) by using a computer to expand the determinant. We will use this approach to obtain the secular equations for the 2D lattice gas of hard particles with nearest-neighbor exclusion on the square lattice. We will then examine the limiting form for the next largest eigenvalue as M becomes large. The importance of knowledge of all of the eigenvalues in determining the size dependence, the interfacial tension, and the correlation length for the 2D Ising model is well known.[8-13] Knowledge of the next largest eigenvalue is of interest since in the 2D Ising model the critical point occurs when the largest eigenvalue and the next largest eigenvalue become equal[14] (degeneracy of the largest eigenvalue). We begin by reviewing the construction of the matrices **W**.

2. SYMMETRY-REDUCED TRANSFER MATRICES

As a specific example, we consider the 2D lattice gas with nearest-neighbor exclusion on the square lattice. For the correlation of columns of lattice sites with periodic boundary conditions (giving rings of sites) as illustrated in Fig. 1A, Runnels and Combs[6] have discussed the construction of the matrices **W** in detail. The size of **W** as a function of M (the number of lattice sites in a ring) is shown in Table I. Runnels and Combs have shown that one need not consider all rotations of the various ring configurations with respect to one another as separate matrix elements, but rather a matrix element contains the sum of all possible rotations of one member of the irreducible set or ring configurations with respect to another member. This is the symmetry-reduced transfer matrix.

Table I. Matrix Size as a Function of Torus Circumference M for the Two Lattice Orientations Shown in Fig. 1[a]

Orientation A		n through which b_n is *exact*	Orientation B		
M	Matrix size		M	Matrix size	$\sigma(M)$
4	3×3	3	2	3×3	2
6	5×5	5	3	4×4	3
8	8×8	7	4	6×6	4
10	14×14	9	5	8×8	5
12	26×26	11	6	13×13	7
14	49×49	13	7	18×18	9

[a] The quantity $\sigma(M)$ is the order of the minimum secular equation as discussed with respect to Eq. (2.4).

For the present model one can make a further reduction in matrix size by tilting the square lattice by 45° and redefining the columns as shown in Fig. 1B. In Fig. 1A, M particles are required to go around the torus, while in Fig. 1B, $2M$ particles are required (even though the number of sites in a volume is M). The size of **W** as a function of M for the tilted configuration is also shown in Table I. The matrix sizes for the two orientations of the lattice can be compared by observing the number of exact b_n that can be extracted from a given matrix ($n = M - 1$ for Fig. 1A and $n = 1M - 1$ for Fig. 1B); this is shown in Table I.

As an example, consider the case of $M = 3$ for the orientation in Fig. 1B. The irreducible set of ring configurations has four members (a "1" indicates an occupied lattice site, while a "0" an unoccupied site)

$$\begin{array}{cccc} \begin{matrix} & 0 & \\ 0 & & 0 \end{matrix} & \begin{matrix} & 0 & \\ 0 & & 1 \end{matrix} & \begin{matrix} & 0 & \\ 1 & & 1 \end{matrix} & \begin{matrix} & 1 & \\ 1 & & 1 \end{matrix} \\ (1) & (2) & (3) & (4) \end{array} \tag{2.1}$$

The 4×4 matrix **W** is then

$$\mathbf{W} = \begin{array}{c} \\ 1 \\ 2 \\ 3 \\ 4 \end{array} \begin{array}{c} \begin{array}{cccc} 1 & 2 & 3 & 4 \end{array} \\ \begin{pmatrix} 1 & 3z & 3z^2 & z^3 \\ 1 & z & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix} \end{array} \tag{2.2}$$

where the (i, j) element gives all rotations of ring j relative to ring i with the number of z factors reflecting the number of particles in ring j. The secular equation for this matrix is

$$\Lambda[\Lambda^3 - (1 + z)\Lambda^2 - (2z + 3z^2 + z^3)\Lambda + (3z^3 + z^4)] = 0 \tag{2.3}$$

One eigenvalue is zero and clearly all of the thermodynamic information about the system is contained in the cubic equation in square brackets; we will refer to the expression in brackets as the minimum secular equation. For the present example one observes that the bottom two rows of the matrix are identical and the matrix can be contracted to the following 3×3 matrix:

$$\mathbf{W} = \begin{array}{c} \\ 1 \\ 2 \\ 3/4 \end{array} \begin{array}{c} \begin{array}{ccc} 1 & 2 & 3/4 \end{array} \\ \begin{pmatrix} 1 & 3z & 3z^2 + z^3 \\ 1 & z & 0 \\ 1 & 0 & 0 \end{pmatrix} \end{array} \tag{2.4}$$

the secular equation of which is the cubic polynomial given in (2.3). The order of the minimum secular equation (for the orientation of Fig. 1B) as a function of M is shown in Table I; this is $\sigma(M)$ as used in (1.2).

3. COMPUTER GENERATION OF SECULAR EQUATIONS

Given $\mathbf{W}(z)$ as constructed in the previous section, one obtains the secular equation by expanding the determinant (1.8). This is easily accomplished using a computer. One programs the $\sigma!$ permutations and simply collects the number of terms corresponding to i factors z and j factors Λ. The secular equation is then the double, finite polynomial in z and Λ

$$\sum_{i,j} \gamma_{i,j} z^i \Lambda^j = 0 \tag{3.1}$$

The coefficients $\gamma_{i,j}$ are integers (not necessarily positive). One can alternatively write (3.1) in the form

$$\sum_{n=0}^{\sigma} C_n(z)\, \Lambda^{\sigma-n} = 0 \tag{3.2}$$

where $C_n(z)$ are finite polynomials in the activity (these polynomials are also functions of M).

For the lattice gas with nearest-neighbor exclusion on the square lattice we have obtained the following explicit minimum secular equations for $M = 2$–7 (for the orientation in Fig.1B):

$M = 2$:

$$\Lambda^2 - \Lambda - (2z + z^2) = 0 \tag{3.3a}$$

$M = 3$:

$$\Lambda^3 - (1 + z)\, \Lambda^2 - (2z + 3z^2 + z^3)\, \Lambda + (3z^3 + z^4) = 0 \tag{3.3b}$$

$M = 4$:

$$\Lambda^4 - (1 + 2z)\, \Lambda^3 - (2z + 6z^2 + 5z^3 + z^4)\, \Lambda^2 + (5z^3 + 8z^4 + 2z^5)\, \Lambda + (2z^5 + 4z^6 + z^7) = 0 \tag{3.3c}$$

$M = 5$:

$$\Lambda^5 - (1 + 3z + z^2)\, \Lambda^4 - (2z + 9z^2 + 12z^3 + 6z^4 + z^5)\, \Lambda^3 + (7z^3 + 26z^4 + 26z^5 + 9z^6 + z^7)\, \Lambda^2 + (-z^5 + 9z^6 + 15z^7 + 7z^8 + z^9)\, \Lambda - (5z^8 + 10z^9 + 6z^{10} + z^{11}) = 0 \tag{3.3d}$$

$M=6$:

$$\Lambda^7-(1+4z+3z^2)\Lambda^6-(2z+12z^2+23z^3+20z^4+8z^5+z^6)\Lambda^5$$
$$+(9z^3+55z^4+107z^5+80z^6+26z^7+3z^8)\Lambda^4$$
$$+(-6z^5-8z^6+35z^7+79z^8+58z^9+18z^{10}+2z^{11})\Lambda^3$$
$$-(2z^7+42z^8+140z^9+188z^{10}+120z^{11}+36z^{12}+4z^{13})\Lambda^2$$
$$-(8z^{10}+24z^{11}+16z^{12}$$
$$+13z^{13}+15z^{14}+7z^{15}+z^{16})\Lambda$$
$$+(4z^{13}+24z^{14}+41z^{15}+29z^{16}+9z^{17}+z^{18})=0 \quad (3.3e)$$

$M=7$:

$$\Lambda^9-(1+5z+6z^2+z^3)\Lambda^8-(2z+15z^2+38z^3+46z^4+30z^5+9z^6+z^7)\Lambda^7$$
$$+(11z^3+95z^4+288z^5+386z^5+253z^6+87z^8+15z^9+z^{10})\Lambda^6$$
$$+(-13z^5-76z^6-112z^7+62z^8+283z^9+261z^{10}$$
$$+113z^{11}+24z^{12}+2z^{13})\Lambda^5$$
$$-(z^7+88z^8+586z^9+1600z^{10}+2268z^{11}+1823z^{12}$$
$$+861z^{13}+235z^{14}+34z^{15}+2z^{16})\Lambda^4$$
$$+(-z^{10}-18z^{11}+66z^{12}+334z^{13}+425z^{14}+182z^{15}$$
$$-20z^{16}-39z^{17}-11z^{18}-z^{19})\Lambda^3$$
$$+(24z^{13}+207z^{14}+837z^{15}+1753z^{16}+2030z^{17}+1359z^{18}$$
$$+541z^{19}+128z^{20}+17z^{21}+z^{22})\Lambda^2$$
$$-(2z^{16}+86z^{17}+334z^{18}+538z^{19}+460z^{20}$$
$$+234z^{21}+73z^{22}+13z^{23}+z^{24})\Lambda$$
$$-(14z^{20}+91z^{21}+224z^{22}+275z^{23}+184z^{24}+68z^{25}+13z^{26}+z^{27})=0 \quad (3.3f)$$

4. ACTIVITY SERIES FOR THE EIGENVALUES

Writing the general Λ_k as an infinite series in the activity

$$\Lambda_k=A_{0k}+A_{1k}z+A_{2k}z^2+\cdots \quad (4.1)$$

we obtain that the secular equation (3.2) is a recursion relation for the coefficients A_{jk} for the σ eigenvalues. For the polynomials (3.3) the recursion process begins in general (for all M) as follows. The σ A_{0k} are given as solutions of

$$A_{0k}{}^{\sigma-1}(A_{0k}-1)=0 \tag{4.2}$$

One solution is $A_{01}=1$, while there are $\sigma-1$ solutions $A_{0k}=0$. For the $\sigma-1$ solutions with $A_{0k}=0$ one finds

$$A_{1k}{}^{\sigma-2}(A_{1k}+2)=0 \tag{4.3}$$

There is one solution $A_{1k}=-2$ and $\sigma-2$ other solutions $A_{1k}=0$.

For the other A_{jk} one finds more complicated polynomials. For example, for $A_{0k}=A_{1k}=0$ the equations for A_{2k} are

$$\begin{aligned}
&(M=3) && -2A_{2k}+3=0\\
&(M=4) && -2A_{2k}^2+5A_{2k}+2=0\\
&(M=5) && A_{2k}(-2A_{2k}^2+7A_{2k}-1)=0\\
&(M=6) && A_{2k}^2(-2A_{2k}^3+9A_{2k}^2-6A_{2k}-2)=0\\
&(M=7) && A_{2k}^4(-2A_{2k}^3+11A_{2k}^2-13A_{2k}-1)=0
\end{aligned} \tag{4.4}$$

For the case of $A_{0k}=A_{1k}=A_{2k}=0$, the equations for A_{3k} are

$$\begin{aligned}
&(M=5) && -A_{3k}-5=0\\
&(M=6) && -2A_{3k}^2-8A_{3k}+4=0\\
&(M=7) && A_{3k}(-A_{3k}^3-A_{3k}^2+24A_{3k}-2)=0
\end{aligned} \tag{4.5}$$

Finally, for $A_{0k}=A_{1k}=A_{2k}=A_{3k}=0$ one has for A_{4k}

$$(M=7) \qquad -A_{4k}-14=0 \tag{4.6}$$

From (4.2) one sees that one eigenvalue (the largest) begins with $A_{01}=1$; all the rest have $A_{0k}=0$ and thus the series for most of the eigenvalues begins with some integer power of the activity (and hence all the eigenvalues except the first go to zero as $z\to 0$).

As an example, the beginning terms in the eigenvalues for $M=2$ and $M=3$ are

$$(M=2) \qquad \begin{aligned}
A_1 &= 1+2z-3z^2+\cdots\\
A_2 &= \quad -2z+3z^2+\cdots
\end{aligned} \tag{4.7}$$

$$(M=3) \qquad \begin{aligned}
A_1 &= 1+3z-3z^2+\cdots\\
A_2 &= \quad -2z+1\tfrac{1}{2}z^2+\cdots\\
A_3 &= \qquad\quad 1\tfrac{1}{2}z^2+\cdots
\end{aligned} \tag{4.8}$$

For $M=7$ there are nine eigenvalues that have the following beginning form:

$$\begin{aligned}
\Lambda_1 &= 1 + \Lambda_{11} z + \cdots \\
\Lambda_2 &= \Lambda_{21} z + \cdots \\
\Lambda_3 &= \Lambda_{32} z^2 + \cdots \\
\Lambda_4 &= \Lambda_{42} z^2 + \cdots \\
\Lambda_5 &= \Lambda_{52} z^2 + \cdots \\
\Lambda_6 &= \Lambda_{63} z^3 + \cdots \\
\Lambda_7 &= \Lambda_{73} z^3 + \cdots \\
\Lambda_8 &= \Lambda_{83} z^3 + \cdots \\
\Lambda_9 &= \Lambda_{94} z^4 + \cdots
\end{aligned} \tag{4.9}$$

5. THE LARGEST EIGENVALUE

From (1.7) we have

$$\lambda_1 = \exp\left[\sum_{n=1}^{\infty} b_n z^n\right] = 1 + \sum_{n=1}^{\infty} a_n z^n \tag{5.1}$$

As we have seen in the previous section, only the expansion for the largest eigenvalue begins with one. For $n \leqslant 2M-1$ the coefficients b_n (or alternatively the a_n) in (5.1) are independent of M and are the exact coefficients for an infinite lattice. Thus from the secular equation for $M=7$ for the model of nearest-neighbor exclusion on the square lattice [see (3.3)] one obtains the exact b_n for the infinite lattice through $n=13$. The values so obtained are shown in Table II. The b_n for this model are known[7] through $n=15$ as a by-product of the series for the 2D Ising model, and the values given in Table II agree with these numbers.

As an illustration of the fact that one gets the b_n exact for $n \leqslant 2M-1$, we give the first four n_n for $M=2$ (in this case we can solve the quadratic secular equation and obtains the series as an expansion of the square-root term):

$$p/kT = z - 2\tfrac{1}{2}z^2 + 10\tfrac{1}{3}z^3 - 50\tfrac{3}{4}z^4 + \cdots \tag{5.2}$$

which on comparison with Table II is correct through b_3, but b_4 is slightly different.

Given the expansion for Λ_1 using (1.4) and (5.1), it is not possible to work backward and construct the secular equation. That is, the secular equation contains much more information than just the coefficients b_n.

Table II. The Coefficients b_n for the 2D Lattice Gas with Nearest-Neighbor Exclusion on the Square Lattice as Obtained from the Secular Equation (3.3f) for $M=7$[a]

n	b_n
1	1
2	−2(1/2)
3	10(1/3)
4	−52(1/4)
5	295(1/5)
6	−1789(5/6)
7	11,397(1/7)
8	−75,238(1/8)
9	510,609(4/9)
10	−3,541,971
11	25,009,987
12	−179,211,452(11/12)
13	1,300,139,553(1/13)

[a] The coefficients are exact for the infinite lattice.

6. HIGH-DENSITY SERIES

On a strip with $M \times L$ sites for the present model with nearest-neighbor exclusion one can place a maximum of $(M \times L)/2$ particles (every other site occupied). At the close-packed density the grand partition function is

$$\Xi = z^{ML/2} = z^{N/2} \tag{6.1}$$

Taking the limit of close-packed density of (6.1) as a reference, one can construct Ξ as a series in inverse powers of z, a factor z^{-1} reflecting the removal of a particle to produce a hole. One has [compare (1.1), which gives an expansion about the low-density limit]

$$\Xi = \exp\left(\frac{pN}{kT}\right) = z^{N/2}\left[1 + Q_1'\left(\frac{1}{z}\right) + Q_2'\left(\frac{1}{z}\right)^2 + \cdots\right] \tag{6.2}$$

where the Q_n' are the combinatorial factors giving the number of arrangements of $N/2-n$ particles on a lattice of N sites. For finite N the quantities Q_n' in (6.2) are finite numbers. Forming p/kT from (6.2), one finds

$$\frac{p}{kT} - \frac{1}{2}\ln z = \frac{1}{N}Q_1'\left(\frac{1}{z}\right) + \frac{1}{N}\left[Q_2 - \frac{1}{2}(Q_1')^2\right]\left(\frac{1}{z}\right)^2 + \cdots \equiv f(z) \tag{6.3}$$

Whether or not the series expansion of $f(z)$ in inverse integer powers of z exists depends on whether or not the limits such as

$$\lim_{N\to\infty} (1/N)\, Q_1' \tag{6.4}$$

are finite. If the quantity in (6.4) is not finite, then $f(z)$ is not analytic in $(1/z)$ about $1/z=0$ (the close-packed limit). For hard-particle lattice gases in one dimension, Lee and Yang[15] have given the general form of Q_n' for a particle that spans m lattice sites:

$$Q_n' = \frac{(N-mn+n)!}{(N-mn)!\, n!} \tag{6.5}$$

For the simplest case of $m=1$ [independent particles: $\Xi=(1+z)^N = 1+Nz+\cdots$] we have

$$Q_1' = N \tag{6.6}$$

and $Q_1'/N=1$ and hence the series is analytic in $1/z$. For $m=2$ (the case of nearest-neighbor exclusion in 1D) one has

$$Q_1' = \frac{N}{4} + \frac{N^2}{8} \tag{6.7}$$

In this case Q_1'/N is not finite as $N\to\infty$ and hence $f(z)$ is not analytic in $1/z$.

One can see what is going on in the above case by examining the secular equation for the case of $M=1$ in (3.3). To construct the high-density version of p/kT from the secular equations we recall that the eigenvalue is for a column of M sites (see Fig. 1B). To alter the secular equation to give a series relative to the close-packed limit of (6.1), we introduce the following scaled variable (at close-packing every other column will be occupied; hence there is an average of $M/2$ particles per column)

$$\Lambda_1' = z^{-M/2}\Lambda_1 \tag{6.8}$$

In the limit of large L, Ξ is given in terms of the largest eigenvalue Λ_1':

$$\Xi = z^{ML/2}(\Lambda_1')^L \tag{6.9}$$

where

$$\Lambda_1' = A_0 + A_{1/2}z^{-1/2} + A_1 z^{-1} + A_{3/2}z^{-3/2} + A_2 z^{-2} + \cdots \tag{6.10}$$

where we have indicated that it is possible that fractional inverse powers of z might arise. For the current model one has $A_0 = 1$ in all cases (this is not so for the case of dimers treated in Section 10).

The equation of state can then be written as[16]

$$\begin{aligned} p/kT &= \tfrac{1}{2}\ln z + M^{-1} \ln A_1' \\ &= \tfrac{1}{2}\ln z + \sum_{n=1}^{\infty} b_n' z^{-n} + \sum_{n=1}^{\infty} c_n z^{-n/2} \end{aligned} \tag{6.11}$$

where we have separately grouped the terms involving inverse integer multiples of z and those involving the square root of z. It is known[4,16] that in the limit $M \to \infty$ the function $f(z)$ of (6.3) is analytic in $1/z$, and hence in the limit of large M the coefficients c_n of (6.11) must go to zero.

For M odd, there will be square-root terms. The origin of the square-root terms for finite M is that when one has a torus, removal of M particles from a column can result in major rearrangements of the lattice (for example, movement of all the particles to the other sublattice), rearrangements that are impossible in the limit of $M \to \infty$. This is illustrated by returning to the case of $M = 1$. From (3.3) with the definition of (6.8) one finds

$$A_1' = \tfrac{1}{2}z^{-1/2} + (1 + \tfrac{1}{4}z^{-1})^{1/2} = 1 + \tfrac{1}{2}z^{-1/2} + \tfrac{1}{8}z^{-1} + \cdots \tag{6.12}$$

giving

$$p/kT = \tfrac{1}{2}\ln z + \tfrac{1}{2}z^{-1/2} + 0.0z^{-1} + \cdots \tag{6.13}$$

We illustrate below the high-density activity series for $M = 1$ through $M = 7$. We give the series up to the first term that departs from the series for the infinite system. The exact series for the infinite system, as obtained by other means,[4,16] is shown for comparison.

$$\begin{array}{lllllll}
(M=1) & p/kT = \tfrac{1}{2}\ln z + \tfrac{1}{2}z^{-1/2} + \cdots & & & & \\
(M=2) & p/kT = \tfrac{1}{2}\ln z + & + \tfrac{3}{4}z^{-1} + \cdots & & & \\
(M=3) & p/kT = \tfrac{1}{2}\ln z + & + \tfrac{1}{2}z^{-1} + \tfrac{1}{6}z^{-3/2} + \cdots & & & \\
(M=4) & p/kT = \tfrac{1}{2}\ln z + & + \tfrac{1}{2}z^{-1} + & - \tfrac{1}{8}z^{-2} + \cdots & & \\
(M=5) & p/kT = \tfrac{1}{2}\ln z + & + \tfrac{1}{2}z^{-1} + & - \tfrac{1}{4}z^{-2} + \tfrac{1}{10}z^{-5/2} + \cdots & & \\
(M=6) & p/kT = \tfrac{1}{2}\ln z + & + \tfrac{1}{2}z^{-1} + & - \tfrac{1}{4}z^{-2} + & + \tfrac{3}{4}z^{-3} + \cdots & \\
(M=7) & p/kT = \tfrac{1}{2}\ln z + & + \tfrac{1}{2}z^{-1} + & - \tfrac{1}{4}z^{-2} + & + \tfrac{2}{3}z^{-3} + \tfrac{1}{14}z^{-7/2} + \cdots & \\
(M=\infty) & p/kT = \tfrac{1}{2}\ln z + & + \tfrac{1}{2}z^{-1} + & - \tfrac{1}{4}z^{-2} + & + \tfrac{2}{3}z^{-3} + \cdots &
\end{array} \tag{6.14}$$

One notes that the terms involving the square root of z in the odd terms has the form as a function of M as follows:

$$\frac{1}{2M} z^{-M/2} \tag{6.15}$$

so the coefficient goes to zero at large M and the position of the term in the series also moves out to infinity.

From (6.14) one sees that the equations of state for finite tori slowly converge to the equation of state for the infinite-circumference system.

7. THE GENERAL FORM OF THE SECULAR EQUATION

We divide both sides of (3.2) by Λ^σ and obtain

$$\sum_{n=1}^{\sigma} C_n(M, z)/\Lambda^n = 1 \tag{7.1}$$

Using the secular equations given in (3.3), one has enough information to write the beginning terms of (7.1) as general functions of M. One finds the following general forms (valid for $M \geqslant 4$):

$$\begin{aligned} C_1 &= 1 + (M-2)\, z + (6 - 3\tfrac{1}{2}M + \tfrac{1}{2}M^2)\, z^2 + \cdots \\ C_2 &= 2z + 3(M-2)\, z^2 + (17 - 11M + 2M^2)\, z^3 + \cdots \\ C_3 &= (3 - 2M)\, z^3 + (-46 + 31\tfrac{1}{2}M - 5\tfrac{1}{2}M^2)\, z^4 + \cdots \\ C_4 &= (6 - 6M + M^2)\, z^5 + \cdots \end{aligned} \tag{7.2}$$

Using the general expansion of Λ_k given in (4.1), one obtains the following general expansions for Λ_1 and Λ_2:

$$\begin{aligned} \Lambda_1 &= 1 + Mz - (2\tfrac{1}{2}M - \tfrac{1}{2}M^2)\, z^2 + \cdots \\ \Lambda_2 &= -2z[1 - (3\tfrac{3}{4} - M)\, z + (19\tfrac{7}{16} - 6\tfrac{1}{4}M + \tfrac{1}{2}M^2)\, z^2 + \cdots] \end{aligned} \tag{7.3}$$

We define

$$\begin{aligned} \Lambda_1 &= \exp(M\Gamma_1) \\ \Lambda_2 &= -2z \exp(M\Gamma_2) \end{aligned} \tag{7.4}$$

Then one finds

$$\begin{aligned} \Gamma_1 &= z - 2\tfrac{1}{2}z^2 + \cdots \\ \Gamma_2 &= (z - 2\tfrac{1}{2}z^2 + \cdots) + M^{-1}(-3\tfrac{3}{4}z + 12\tfrac{13}{32}z^2 + \cdots) \end{aligned} \tag{7.5}$$

One notes that Γ_1 is independent of M and that Γ_2 has the following specific M dependence:

$$\Gamma_2 = \Gamma_1 + M^{-1}\gamma_2 \tag{7.6}$$

where

$$\gamma_2 = -3\tfrac{3}{4}z + 12\tfrac{13}{32}z^2 + \cdots \tag{7.7}$$

From (7.6) one has

$$\lim_{M\to\infty} \Gamma_1 = \Gamma_2 \tag{7.8}$$

One obtains the quantities Γ_1 and γ_2 independent of M for the infinite system.

8. CALCULATION OF THERMODYNAMIC FUNCTIONS

Given the secular equation for an $M \times L$ torus, one can evaluate exactly all of the thermodynamic functions in the limit $L \to \infty$, the only numerical task being the calculation of Λ_1, the largest eigenvalue (i.e., one must obtain the largest root of the secular equation). For convenience we write the secular equation (3.2) in the form

$$\sum_{n=0}^{\sigma} c_n(z)\, \Lambda^n = 0 \tag{8.1}$$

where

$$C_{\sigma-n} = c_n \tag{8.2}$$

We numerically calculate the largest root, Λ_1, of (8.1). Then by implicit differentiation of (8.)) we obtain the following relations for the derivatives of Λ_1. We define

$$\begin{aligned}
S_1 &= \sum c'_n \Lambda_1^n & S_5 &= \sum n(n-1)\, c_n \Lambda_1^{n-2} \\
S_2 &= \sum n c_n \Lambda_1^{n-1} & S_6 &= \sum c'''_n \Lambda_1^n \\
S_3 &= \sum c''_n \Lambda_1^n & S_7 &= \sum n c''_n \Lambda_1^{n-1} \\
S_4 &= \sum n c'_n \Lambda_1^{n-1} & S_8 &= \sum n(n-1)\, c'_n \Lambda_1^{n-2}
\end{aligned} \tag{8.3}$$

$$S_9 = \sum n(n-1)(n-2)\, c_n \Lambda_1^{n-3}$$

where the prime indicates differentiation with respect to z. The first three derivatives of A with respect to z are then given by

$$\begin{aligned}
\frac{\partial A_1}{\partial z} &= -\frac{S_1}{S_2} \\
\frac{\partial^2 A_1}{\partial z^2} &= -\frac{S_3 + 2S_4 + S_5}{S_2} \\
\frac{\partial^3 A_1}{\partial z^3} &= -\frac{3S_4 + 3S_5 + S_6 + 3S_7 + 3S_8 + S_9}{S_2}
\end{aligned} \tag{8.4}$$

The pressure, the density ρ, the isothermal compressibility K_T, modified compressibility X, and the derivative of X are then obtained using standard thermodynamic relations:

$$\begin{aligned}
\frac{p}{kT} &= M^{-1} \ln A_1 \\
\rho &= \frac{\partial (p/kT)}{\partial \ln z} = M^{-1} D_1 \\
K_T &= \frac{1}{\rho} \frac{\partial \rho}{\partial p} = \frac{X}{kT\rho^2} \\
X &= \frac{\partial \rho}{\partial \ln z} = (\rho - M\rho^2) + M^{-1} D_2 \\
\frac{\partial X}{\partial \ln z} &= X(1 - 2M\rho) + M^{-1}(2D_2 - D_1 D_2 + D_3)
\end{aligned} \tag{8.5}$$

where

$$\begin{aligned}
D_1 &= \left(\frac{z}{A_1}\right) \frac{\partial A_1}{\partial z} \\
D_2 &= \left(\frac{z^2}{A_1}\right) \frac{\partial^2 A_1}{\partial z^2} \\
D_3 &= \left(\frac{z^3}{A_1}\right) \frac{\partial^3 A_1}{\partial z^3}
\end{aligned} \tag{8.6}$$

Figure 4 shows the modified compressibility X as a function of ρ for various values of M calculated using the secular equations (3.3) for the model of nearest-neighbor exclusion and the procedure outlined above. For

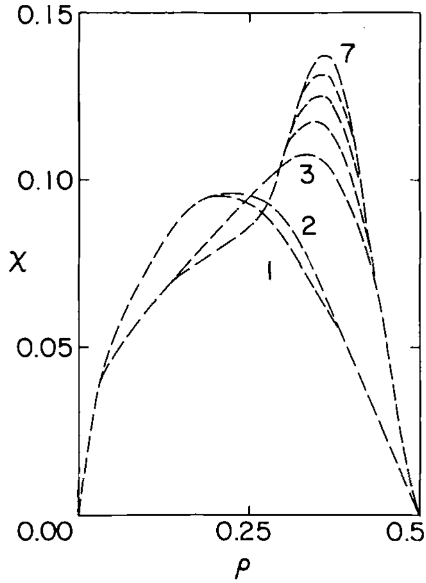

Fig. 4. The modified compressibility X as a function of density for $M \times \infty$ lattice strips. The compressibility is calculated using the exact secular equations of (3.3) and the appropriate relation given in (8.5). The numbers 2–7 refer to the M values for the 2D lattice gas with nearest-neighbor exclusion. The number 1 refers to the 1D lattice gas with nearest-neighbor exclusion; the secular equation for this system is given in (8.7). As $M \to \infty$ the maximum in X develops into the singularity of (8.8) at approximately $\rho_c = 0.37$.

comparison we also show $X(\rho)$ for the 1D lattice gas with nearest-neighbor exclusion. The secular equation for the 1D model is

$$\Lambda^2 - \Lambda - z = 0 \tag{8.7}$$

As shown previously by Runnels and Combs,[6] as M is increased, a sharp maximum in X develops near $\rho_c = 0.37$. In the limit $M \to \infty$, X has a singularity of the form[16,17]

$$X \sim -\ln(\rho - \rho_c) \tag{8.8}$$

This is a second-order transition that reflects the sublattice ordering that begins to set in at ρ_c as the density is increased.

9. THE RATIO Λ_2/Λ_1

We can factor out the largest eigenvalue in (1.2) and obtain

$$\Xi = \Lambda_1^L \left[1 + \left(\frac{\Lambda_2}{\Lambda_1} \right)^L + \cdots \right] \tag{9.1}$$

We use the general M dependence for Λ_1 and Λ_2 given in (7.3) and (7.4) and find that the ratio of the eigenvalues is independent of M

$$\Lambda_2/\Lambda_1 = -2z(1 - 3\tfrac{3}{4}z + 19\tfrac{7}{16}z^2 + \cdots) = \sum_{n=1}^{\infty} \alpha_n z^n \tag{9.2}$$

One finds that one obtains the α_n for the infinite system through $n \leqslant M-1$; this is illustrated in Table III. As discussed in the Introduction, the quantity L must be even. Thus even though, as seen from (9.2), the ratio of the eigenvalues is negative, it will be raised to an even power. Thus we introduce the positive quantity

$$r = -\Lambda_2/\Lambda_1 \tag{9.3}$$

Using the secular equation for $M=7$ of (3.3f), we thus can calculate the exact α_n for the infinite system through $n=6$. The series is (we quote the fractions to four significant figures)

$$r = 2z(1 - 3\tfrac{3}{4}z + 19\tfrac{7}{16}z^2 - 114.9062z^3 + 731.4180z^4 - 4882.9586z^5 + \cdots) \quad (M \geqslant 7) \tag{9.4}$$

The ratios of successive coefficients in (9.4) extrapolate smoothly to

Table III. The Coefficients in the Activity Expansion of Λ_2/Λ_1 as a Function of M, the Circumference of the Lattice Torus[a]

n	$M=2$	$M=3$	$M=4$	$M=5$	$M=6$	$M=7$
1	<u>1</u>	<u>1</u>	<u>1</u>	<u>1</u>	<u>1</u>	<u>1</u>
2	−3.5	<u>−3.75</u>	<u>−3.75</u>	<u>−3.75</u>	<u>−3.75</u>	<u>−3.75</u>
3	16	19(5/16)	<u>19(7/16)</u>	<u>19(7/16)</u>	<u>19(7/16)</u>	<u>19(7/16)</u>
4		−111.8125	−114.843	<u>−114.906</u>	<u>−114.906</u>	<u>−114.906</u>
5			729.293	731.387	<u>731.418</u>	<u>731.418</u>
6					−4889.443	<u>−4882.959</u>

[a] The quantities shown are the coefficients α_n of (9.2) divided by minus two. The underlined numbers are the limiting values for the infinite system.

$z = -0.12$, which is also[16] the radius of convergence of the pressure series (1.7).

We can express (9.3) as a density series using the expansion[16]

$$\rho = z - 5z^2 + 31z^3 - 209z^4 + 1476z^5 - 10739z^6 + \cdots \tag{9.5}$$

We obtain

$$r = 2\rho + 2\tfrac{1}{2}\rho^2 + 1\tfrac{7}{8}\rho^3 - 1.1875\rho^4 - 7.9141\rho^5 - 14.1484\rho^6 + \cdots \tag{9.6}$$

Both (9.4) and (9.6) give well-behaved series if they are inverted:

$$z = \tfrac{1}{2}r + \tfrac{15}{16}r^2 + (1.085938)\,r^3 + (0.8828092)\,r^4 + (0.4949532)\,r^5 + (0.1255641)\,r^6 + \cdots \tag{9.7}$$

$$\rho = \tfrac{1}{2}r - \tfrac{5}{16}r^2 + (0.2734375)\,r^3 - (0.2070313)\,r^4 + (0.1737067)\,r^5 - (0.165576)\,r^6 + \cdots \tag{9.8}$$

At $r = 1$, (3/4) and (4/3) Padé approximants give $z = 3.74$ and $z = 3.42$. At $r = 1$, (3/4) and (4/3) approximants give $\rho = 0.349$ and $\rho = 0.346$. These compare with the phase transition quantities[12,13] of $z = 3.75$ and $\rho = 0.37$.

10. DIMERS; SUMMARY

The combinatorics of placing dimers on a lattice is a classic problem in statistical mechanics; we consider the case of the plane-square lattice wrapped into an $M \times L$ torus as illustrated in Fig. 1B. The placing of a dimer on the torus is illustrated in Fig. 5A; each dimer has two possible orientations as illustrated in Fig. 5B. The construction of the truncated matrix for $M = 2$ is illustrated in Fig. 5C. Proceeding as before, we obtain the secular equations for $M = 1$ to $M = 5$:

$$(M=1)\quad \Lambda^2 - \Lambda - 2z = 0$$

$$(M=2)\quad \Lambda^3 - (1+2z)\,\Lambda^2 - (2z+2z^2)\,\Lambda + 4z^3 = 0$$

$$(M=3)\quad \Lambda^4 - (1+4z)\,\Lambda^3 - (2z+9z^2+8z^3)\,\Lambda^2 + (6z^3+8z^4)\,\Lambda + 12z^6 = 0$$

$$\begin{aligned}(M=4)\quad \Lambda^6 &- (1+6z+4z^2)\,\Lambda^5 - (2z+16z^2+32z^3+8z^4)\,\Lambda^4 \\ &+ (8z^3+50z^4+88z^5+40z^6)\,\Lambda^3 - (4z^5+4z^8)\,\Lambda^2 \\ &- (16z^8+24z^9+64z^{10})\,\Lambda + 32z^{12} = 0\end{aligned} \tag{10.1}$$

$$(M=5)\quad \Lambda^8-(1+8z+12z^2)\,\Lambda^7-(2z+23z^2+80z^3+90z^4+32z^5)\,\Lambda^6$$
$$+(10z^3+115z^4+430z^5+572z^6+184z^7)\,\Lambda^5$$
$$+(-10z^5-76z^6-160z^7+105z^8+576z^9+280z^{10})\,\Lambda^4$$
$$-(60z^8+446z^9+1340z^{10}+1672z^{11}+720z^{12})\,\Lambda^3$$
$$-(40z^{11}+100z^{12}+320z^{13}+329z^{14}+640z^{15})\,\Lambda^2$$
$$+(200z^{15}+320z^{16}+800z^{17})\,\Lambda+400z^{20}=0$$

As with the case of particles with nearest-neighbor exclusion considered previously, the number of dimers at close-packing is $N/2$ (where $N=M\times L$ is the total number of lattice sites). Unlike the previous problem where all of the particles must exist on one sublattice (illustrated in Fig. 3), there are many ways to arrange the dimers at close-packing (illustrated in Fig. 6A). If Φ is the number of arrangements of $N/2$ dimers on N sites at close-packing, one has for large N

$$\Phi=\phi^{N/2} \tag{10.2}$$

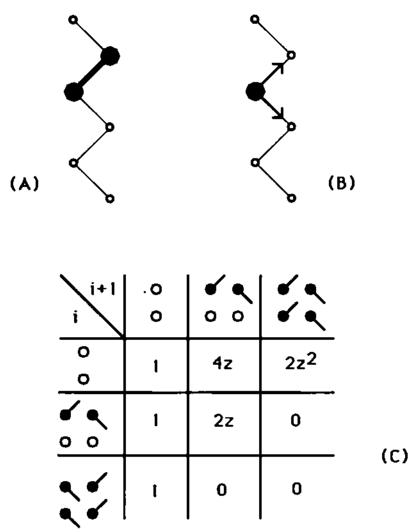

Fig. 5. Lattice gas of dimers. (A) The placement of a dimer on a lattice strip of type (B) illustrated in Fig. 1. (B) The two possible orientations of a dimer. (C) The transfer matrix for dimers for a lattice strip with $M=2$. The configurations illustrate the possible states of a column of two sites (a column as in Fig. 1B). The matrix elements are the contribution of column $i+1$ to the grand partition function.

where ϕ is the freedom per dimer at close-packing (one has $\phi = 2$ as $N \to 0$). The quantity ϕ is known exactly[18,19] and to five figures is given by

$$\phi = 1.7916 \tag{10.3}$$

It is known exactly that there is no singularity from zero to close-packed density.

The low-density activity series is known[21] through 15 terms; the series is also known[22] through 9 terms for the case of interacting dimers on the plane-square lattice. The secular equation for $M = 5$ gives the low-density activity series through $n = 9$ $(2M - 1)$ for the case of tori of infinite circumference. We note that the equilibrium activity series can also be used to treat the kinetics of the adsorption of dimers onto a 2D lattice.[23] The high-density activity series is of most interest and we turn to that now. We can write the grand partition function as a series in inverse powers of z relative to the close-packed state as follows [in analogy with (6.2)]:

$$\Xi = (z)^{N/2}\left[\left(\sum_{i=1}^{\Phi}\right) + \left(\sum_{i=1}^{\Phi}\sum_{j=1}^{N/2}\Gamma_{ij}\right)z^{-1} + \cdots\right] \tag{10.4}$$

The sum over i represents the sum over each of the Φ configurations of dimers at close-packing (as illustrated in Fig. 6A). For each of the i configurations we remove each of the $N/2$ dimers one at a time (the sum

(A)

(B)

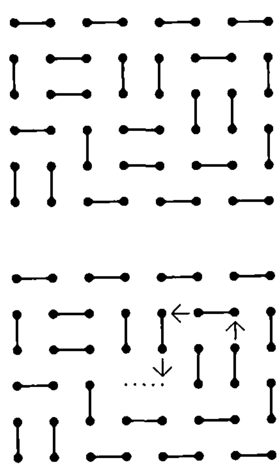

Fig. 6. (A) Close-packing of dimers on the plane-square lattice. There is no preferred orientation or sublattice structure. (B) The configuration shown in (A) with one dimer missing (dotted line). The arrows indicate a possible rearrangement of the remaining dimers made possible by the vacancy.

over j), giving a close-packed configuration with one dimer removed (illustrated in Fig. 6B). The quantity Γ_{ij} is the number of ways that one can rearrange the remaining particles when the jth particle is removed from the ith configuration. The factor z^{-1} indicates that one particle has been removed. Introducing the average Γ_{ij}

$$\langle \Gamma \rangle = \sum_{i=1}^{\Phi} \sum_{j=1}^{N/2} \Gamma_{ij} \Big/ \sum_{i=1}^{\Phi} \sum_{j=1}^{N/2} \tag{10.5}$$

one has [using (10.2) for Φ]

$$\Xi = (z\phi)^{N/2} \left[1 + \frac{N}{2} \langle \Gamma \rangle z^{-1} + \cdots \right] \tag{10.6}$$

The equation of state is then

$$\frac{p}{kT} = \frac{1}{2} \ln z + \frac{1}{2} \ln \phi + \frac{1}{2} \langle \Gamma \rangle z^{-1} + \cdots \tag{10.7}$$

As discussed in Section 6, if $\langle \Gamma \rangle \to \infty$ as $N \to \infty$, then $p/kT - \ln z/2$ is not analytic about $z^{-1} = 0$.

Our secular equations in (10.1) give us the following beginning terms for the equation of state of (10.7) for $M = 1$ to $M = 5$:

$$\begin{aligned}
&(M=1) \quad p/kT = 0.5 \ln z + 0.34657 + 0.35355 z^{-1/2} + 0.0 z^{-1} + \cdots \\
&(M=2) \quad p/kT = 0.5 \ln z + 0.34657 + 0.0 z^{-1/2} + 1.5 z^{-1} + \cdots \\
&(M=3) \quad p/kT = 0.5 \ln z + 0.29863 + 0.27217 z^{-1/2} + 0.51389 z^{-1} + \cdots \\
&(M=4) \quad p/kT = 0.5 \ln z + 0.30699 + 0.0 z^{-1/2} + 2.72084 z^{-1} + \cdots \\
&(M=5) \quad p/kT = 0.5 \ln z + 0.29415 + 0.24060 z^{-1/2} + 0.83766 z^{-1} + \cdots
\end{aligned} \tag{10.8}$$

First we note that there is an alteration in the character of the results between M odd and M even. The constant term should be asympototic to the value

$$\tfrac{1}{2} \ln \phi = \tfrac{1}{2} \ln(1.7916) = 0.29155 \tag{10.9}$$

and one sees that this is so.

The question of interest is whether as $M \to \infty$ the coefficients of $z^{-1/2}$ for M odd go to zero and whether simultaneously the coefficients of z^{-1} for M odd and M even approach the same finite limit. If this is so, then p/kT is analytic about the close-packed state in powers of z^{-1}. It is not impossible that this is so, but we clearly do not have enough data (M values) to support this interpretation.

It is of interest to calculate the entropy as a function of density from zero up to close-packing. We have the basic relation

$$\Xi = \lambda^N = \Omega(n^*, N)\, z^{n^*} \tag{10.10}$$

where N is the total number of sites ($N = M \times L$), λ is the largest eigenvalue per site ($= \Lambda_1^{1/M}$), and n^* is the number of particles on the lattice that corresponds to the maximum term in Ξ (the particle density is given by $\rho = n^*/N$). Using (10.10), one obtains

$$\frac{1}{N} \ln \Omega = \ln \lambda - \rho \ln z = \ln \omega \tag{10.11}$$

We obtain λ as the largest root of the secular equation and ρ from the appropriate derivative as outlined in Section 8. The quantity $\ln \omega$ defined in (10.11) is shown in Fig. 7 as a function of density for $M = 1$ and $M = 5$. In the limit $z \to \infty$ one has $\rho \to 1/2$ and

$$\Omega \to \phi^{N/2} \tag{10.12}$$

giving

$$\lim_{\rho \to \infty} \ln \omega = \tfrac{1}{2} \ln \phi \tag{10.13}$$

For the case of $M = 5$ this limit is very close to the value for the infinite system.

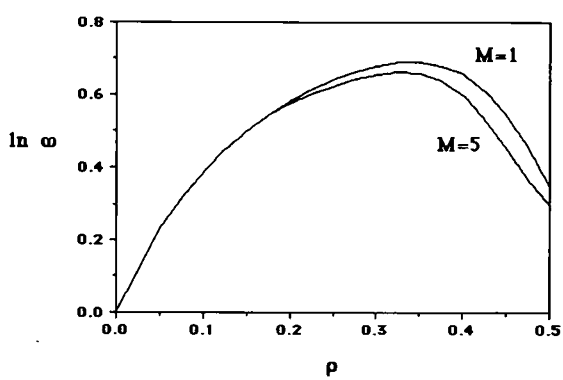

Fig. 7. The configurational entropy per lattice site S/Nk) for the dimer model as a function of density ρ for finite tori with $M = 1$ and $M = 5$. The close-packing limit for the case of $M = 5$ is indistinguishable on the scale shown from the value for the case of $M = \infty$.

In summary, we have shown that we can obtain the secular equations for lattice gas models on tori of finite circumference by computer expansion of the appropriate determinants constructed from symmetry-reduced matrices. The secular equations for finite tori yield the exact beginning coefficients in various thermodynamic series for the infinite system. The number of coefficients that can be obtained in this manner depends on the details of the system treated. In our example of the hard-particle lattice gas we easily obtained the first 13 exact vitial coefficients. The computer time required to expand the secular determinant goes up very fast with the size of the matrix and hence the number of coefficients that can be obtained in this manner in a reasonable amount of time is in the range of 15–18. In addition to giving a set of exact coefficients for low-and high-density expansions, the secular equation gives the exact thermodynamic behavior for the finite system.

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