DIMACS Technical Report 2000-35
November 2000

Close to close packing [1]

by

Oliver Penrose[2]
Mathematics, Heriot-Watt University, Edinburgh EH14 4AS, UK

and

George Stell[3]
Chemistry, SUNY, Stony Brook, NY, USA

Journal of Statistical Physics, 100, 89-95 (2000)

[1]We dedicate this work to the memory of Ann Stell, beloved friend of OP and beloved wife of GS.
[2]Supported by DIMACS grant NSF STC-91-19999
[3]Supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy.

ABSTRACT

For various lattice gas models with nearest neighbour exclusion (and, in one case, second-nearest neighbour exclusion as well), we obtain lower bounds on m, the average number of particles on the non-excluded lattice sites closest to a given particle. They are all of the form

$$m/m_{cp} \geq 1 - \text{ const.}(N_{cp}/N - 1)$$

where N is the number of occupied sites, m_{cp} is the coordination number, snd N_{cp} is the value of N at close packing. An analogous result exists for hard disks in the plane.

KEY WORDS: close packing, lattice models, inequalities, nearest-neighbour exclusion, hard disks.

1 Introduction

In a classic piece of work [3], L Fejes Tóth gave a proof that the highest-density packing of non-overlapping disks in a plane is, as one expects intuitively, the hexagonal close packing. His ideas were used by the present authors [2] obtain information about the packing of such disks at densities slightly below the close-packing density. Defining $m(r)$ to be the average number of disks whose centres lie within a distance r of a given disk, we showed that $m(r)$ satisfies an inequality

$$1 \geq \frac{m(r)}{6} \geq 1 - \frac{A/A_{cp} - 1}{(r^2/a^2) - 1} - \epsilon \text{ if } 1 < r/a < \frac{1}{2} \text{ cosec } \frac{\pi}{7} = 1.15 \ldots \tag{1}$$

Here a is the diameter of the disks, A is the area of the (hexagonal) region available to the disks, $A_{cp} = \frac{1}{2}\sqrt{3}Na^2 \approx 0.866Na^2$ is the value of A at hexagonal close packing, N is the number of disks, and ϵ denotes a correction term which is much smaller than the term shown if $A/a^2 \gg 1$ and $r/a - 1 \ll 1$.

This result can be used, for example, to obtain upper and lower bounds on the energy of a system of hard disks with a sufficiently short-range square-shoulder or square-well interaction

$$U(r) = \begin{cases} +\infty & (0 \leq r < a) \\ U_0 & (a \leq r < b) \\ 0 & (b \leq r) \end{cases} \tag{2}$$

where U_0 and b are constants, with $a < b < a$ cosec $(\pi/7)$. These bounds provide an upper bound on the error in the formula for the mean energy per particle given by thermodymamic perturbation theory; this upper bound is proportional to the deviation of the density from the close-packing density.

In view of the recent mathematical progress [1] on the three-dimensional analogue of Fejes Tóth's problem (often called Kepler's conjecture), one may hope that a three-dimensional analogue of the inequality (1) may some day be proven. In the present note, however, our ambitions are much more modest: to prove some results analogous to (1) for lattice gases with nearest-neighbour exclusion, both in two and three dimensions. We shall consider (though not in that order) a square lattice with nearest-neighbour exclusion, a triangular lattice with nearest-neighbour exclusion, a cubic lattice with nearest neighbour exclusion, and a cubic lattice with both nearest- and second-nearest neighbour exclusion. In all the cases considered the result we obtain can be written in the form

$$1 \geq \frac{m}{m_{cp}} \geq 1 - \text{ const. } \left(\frac{N_{cp}}{N} - 1\right) \tag{3}$$

where N is the number of particles (i.e. occupied lattice sites), m is the average number of particles at the non-excluded lattice sites nearest to a given particle, and N_{cp}, m_{cp} are the values of N, m at close packing. Except in the case of the triangular lattice, the constant is 1. Since the fractions A/A_{cp} in (1) and N_{cp}/N in (3) are equal in the thermodynamic limit, there is a close analogy between (1) and (3).

2 Plane square lattice

Consider a plane square lattice with L sites, N of which are occupied, subject to nearest-neighbour exclusion. We denote by m the average number of second-nearest neighbours per particle (the number of nearest neighbours is, of course, zero). To avoid edge effects we take the underlying space to be a torus and give the lattice an even number of sites along each axis, so that a perfect close-packing arrangement is possible.

Let p_0, p_1, p_2 denote, respectively, the number of plaquettes with $0, 1,$ and 2 corners occupied; because of the nearest-neighbour exclusion rule, p_2 is the number of plaquettes with two opposite corners occupied and the other two unoccupied. The number of second-nearest neighbour pairs is then

$$p_2 = \frac{1}{2} m N \tag{4}$$

by the definition of m

Since, on this lattice, the total number of plaquettes is equal to the number of sites we have

$$p_0 + p_1 + p_2 = L \tag{5}$$

Also, since each site meets four plaquettes, the total number of occupied corners of plaquettes is equal to four times the number of occupied sites, and hence

$$p_1 + 2p_2 = 4N \tag{6}$$

At close packing, all the plaquettes have two occupied corners, so that $p_0 = p_1 = 0, p_2 = L$. From the above equations, the values of N and m at close packing are

$$N_{cp} = \frac{1}{2} L \tag{7}$$

$$m_{cp} = 4 \tag{8}$$

Evidently m_{cp} is the coordination number of the lattice. From (4) and (6) we find

$$(4 - m)N = p_1 \tag{9}$$

and, from (7), (5) and (6),

$$N_{cp} - N = \frac{1}{2} L - N = \frac{1}{2} p_0 + \frac{1}{4} p_1 \tag{10}$$

so that

$$0 \le (4 - m)N \le 4(N_{cp} - N) \tag{11}$$

This gives, as our analogue of (1) for the square lattice with nearest-neighbour exclusion,

$$1 \ge \frac{m}{4} \ge 1 - (N_{cp}/N - 1) \tag{12}$$

3 Simple cubic lattice, nearest-neighbour exclusion

Now consider a simple cubic lattice with L sites, N of which are occupied, subject to nearest-neighbour exclusion. As before, we denote by m the average number of second-nearest neighbours per particle and take the underlying space to be a (three-dimensional) torus. In the close-packed arrangement, the occupied sites form a face-centred cubic lattice.

As before, let p_0, p_1, p_2 denote, respectively, the number of plaquettes with $0, 1,$ and 2 corners occupied; because of the nearest-neighbour exclusion rule, p_2 is the number of plaquettes with two opposite corners occupied and the other two unoccupied. The number of second-nearest neighbour pairs is again given by (4), but for this lattice, the total number of plaquettes is three times the number of sites so that

$$p_0 + p_1 + p_2 = 3L \tag{13}$$

Also, since each site meets 12 plaquettes, the analogue of (6) is

$$p_1 + 2p_2 = 12N \tag{14}$$

At close packing, all the plaquettes have two occupied corners, so that $p_0 = p_1 = 0, p_2 = L$. The values of N and m at close packing are

$$N_{cp} = \frac{1}{2}L \tag{15}$$

$$m_{cp} = 12 \tag{16}$$

From (4) and (14) we find

$$(12 - m)N = p_1 \tag{17}$$

and from (15), (13) and (14),

$$N_{cp} - N = \frac{1}{2}L - N = \frac{1}{6}p_0 + \frac{1}{12}p_1 \tag{18}$$

so that

$$0 \leq (12 - m)N \leq 12(N_{cp} - N) \tag{19}$$

This gives, as our analogue of (1) for the cubic lattice with nearest-neighbour exclusion,

$$1 \geq \frac{m}{12} \geq 1 - (N_{cp}/N - 1) \tag{20}$$

4 Simple cubic lattice, nearest- and second-nearest neighbour exclusion

A similar result also exists for the same lattice but with exclusion on both nearest and second-nearest neighbour sites. In this case the occupied sites in the closest-packed configuration form a body-centred cubic lattice

This time, instead of plaquettes, we consider the small cubes whose corners are nearest-neighbour lattice sites Let c_0, c_1, c_2 denote, respectively, the number of such cubes with $0, 1,$ and 2 corners occupied; because of the exclusion rule, c_2 is exactly the number of small cubes with two opposite corners occupied and the other two unoccupied. Let m' denote the average number of second-nearest neighbours of a given occupied site, so that the number of second-nearest neighbour pairs is given by

$$c_2 = \frac{1}{2}m'N \tag{21}$$

Since the total number of small cubes is equal to the number of sites we have

$$c_0 + c_1 + c_2 = L \tag{22}$$

Also, since each site meets 8 small cubes, the analogue of (6) is now

$$c_1 + 2c_2 = 8N \tag{23}$$

At close packing, all the small cubes have two occupied corners, so that $c_0 = c_1 = 0, c_2 = L$. From the above equations, the values of N and m at close packing are

$$N_{cp} = \frac{1}{4}L \tag{24}$$

$$m'_{cp} = 8 \tag{25}$$

From (21) and (23) we find

$$(8 - m')N = c_1, \tag{26}$$

and from (15), (13) and (14),

$$N_{cp} - N = \frac{1}{4}L - N = \frac{1}{4}c_0 + \frac{1}{8}c_1 \tag{27}$$

so that

$$0 \leq (8 - m')N \leq 8(N_{cp} - N). \tag{28}$$

This gives, as our analogue of (1) for the cubic lattice with nearest- and second-nearest-neighbour exclusion,

$$1 \geq \frac{m'}{8} \geq 1 - (N_{cp}/N - 1). \tag{29}$$

5 Triangular lattce

To obtain an inequality for the triangular lattice with nearest neighbour exclusion, a slightly more complicated argument is necessary. This time the plaquettes are triangles. Using the same notation p_0, p_1, p_2 as in section 2, we see that $p_2 = 0$ because of the exclusion rule, so that the analogues of (5), (7) and (6) are

$$p_0 + p_1 = 2L = 6N_{cp} \tag{30}$$
$$p_1 = 6N \tag{31}$$

from which it follows that

$$p_0 = 6(N_{cp} - N) \tag{32}$$

Let us define also s_0, s_1, s_2 as the number of second-nearest neighbour pairs of sites with, respectively, neither site occupied, one site occupied, or both occupied. They too satisfy relations analogous to (4), (5) and (6); the ones we shall need are

$$s_1 + 2s_2 = 6N \tag{33}$$
$$s_2 = \frac{1}{2}mN \tag{34}$$

from which it follows that

$$s_1 = (6 - m)N \tag{35}$$

For every second-nearest pair of sites, one of which is occupied and the other unoccupied, there is one empty plaquette and one singly occupied plaquette. Since each empty plaquette can belong in this way to at most three second-nearest pairs of this type, we have

$$s_1 \leq 3p_0 \tag{36}$$

From (31), (35) and (36) we find

$$(6 - m)N \leq 18(N_{cp} - N) \tag{37}$$

so that the analogue of (1) for the triangular lattice with nearest-neighbour exclusion is (since the m_{cp} for this lattice is 6)

$$1 \geq \frac{m}{6} \geq 1 - 3(N_{cp}/N - 1) \tag{38}$$

References

[1] Thomas C Hales, http://www.math.lsa.umich.edu/ hales/countdown/ , cited by Keith Devlin, http://www.maa.org/devlin/devlin_9_98.html

[2] G Stell and O Penrose, Phys Rev Lett **51**, 1397-1400 (1983) First-order thermodynamic perturbation theory is exact in the two-dimensional close-packing limit.

[3] L Fejes Tóth, *Regular figures* (MacMillan, New York, 1964) page 165.