

should correspond to the case of random loose packing of equal circles. The three-dimensional problem can be solved in a similar way. Figure 1 should be changed into the Gotoh and Finney spheres and tetrahedron, and show a hatched parallelepiped. The three vectors from the centre of the supported sphere to the centres of the supporting spheres will then mark the vertices of a spherical triangle on a sphere of radius  $2r$  (Fig. 1c). Since the probability of a configuration with  $b, c, A$  in the range  $dbdcA$  is proportional to

$$\sin b \sin c dbdcA,$$

using the formula

$$\cos a = \cos b \cos c + \sin b \sin c \cos A$$

one obtains

$$\sin b \sin c dbdcA \\ = (\sin a \sin b \sin c da db dc) / U$$

with  $U = (1 - \cos^2 a - \cos^2 b - \cos^2 c + 2 \cos a \cos b \cos c)^{1/2}$ . The volume of the hatched parallelepiped turns out to be  $8r^3 U$ .

The average volume of the parallelepiped is therefore (by analogy with equation (2))

$$V = \frac{8 \int_{\pi/3}^{\pi/2} \int_{\pi/3}^{\pi/2} \int_{\pi/3}^{\pi/2} \sin a \sin b \sin c da db dc}{\int_{\pi/3}^{\pi/2} \int_{\pi/3}^{\pi/2} \int_{\pi/3}^{\pi/2} \sin a \sin b \sin c da db dc / U} \quad (5)$$

With a simple change of variables ( $x = \cos a, y = \cos b, z = \cos c$ ) we get for the density  $d$  (by analogy with equation (3))

$$d = (4/3)\pi \int_0^{1/2} \int_0^{1/2} \int_0^{1/2} dx dy dz / (1 - x^2 - y^2 - z^2 + 2xyz)^{1/2} \quad (6)$$

With a numerical integration I found  $d \sim 0.596$ , in excellent agreement with the experimental random loose packing density.

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<sup>1</sup> Gotoh, K., and Finney, J. L., *Nature*, **252**, 202 (1974).

<sup>2</sup> Santaló, L. A., *Introduction to Integral Geometry* (Hermann, Paris, 1953).

GOTOH AND FINNEY REPLY—We are aware of the numerical closeness of  $2/\pi$  and 0.6366, but until now had no good nonnumerological reason for asserting its significance. We are grateful to Gamba for pointing out the possible use of integral geometry, and will follow up the

references quoted.

We stress the dangers of using two-dimensional analogues<sup>1</sup>, for the geometries of two and three dimensions are essentially different. The two-dimensional Ising problem has been solved, although the three-dimensional one has not<sup>2</sup>, and the third dimension changes the nature of percolation problems<sup>3</sup>. The average number of edges per Voronoi polygon can be shown to be exactly six for any two-dimensional array of points<sup>4</sup>, a fact which blurs the distinction between a two-dimensional "liquid" and crystal. The corresponding three-dimensional case is unsolved, and a discontinuous transition in the number of faces per polyhedron occurs on melting of a three-dimensional liquid<sup>5</sup>. Thus it is difficult to define states in two dimensions that may be equivalent to random loose or close packings in three dimensions<sup>6</sup>.

The simplicity of Gamba's explanation of random loose packing density is attractive, but we think it erroneous. In two dimensions, the addition of a fourth circle introduces excessive symmetry, and implicitly assumes that the covering can be considered equivalent to a covering of touching rhombuses. In a real two-dimensional non-crystalline

packing, in contrast to a dislocated solid, triangles and polygons occur. Similarly, in the extension to three dimensions, Gamba implicitly assumes that the ran-

dom loose packing can be replaced by a space-filling set of touching parallelepipeds. The parallelepiped is an octahedron with two tetrahedral caps, and immediately gives too many octahedra in comparison with the random packing. Moreover, they are distributed in too orderly a manner. The parallelepiped is bounded by six planar faces, each one being defined by four coplanar spheres. This does not fit with the reality of random packing: the extra freedom given by the third dimension leads to a low occurrence of such special arrangements, and a consequent reduction in the local symmetry within the aggregate.

The assumption of a uniform probability density for the angles  $a, b$  and  $c$  takes no account of the local geometrical restrictions on space occupation (for example, a peak near the icosahedral angle of  $65^\circ$ ?) although as we admitted in our original paper, we were unable to take adequate account of these ourselves. We tried to find an "average" density by the admittedly non-rigorous method of finding the "average" dimensions of an "average" tetrahedron. We tried to build in the condition that the tetrahedra, of which we try to consider the average, fill space. Thus an integral number meet at an edge, an integral number meet at a vertex, and each face is shared by two tetrahedra. In addition to these basic geometrical constraints, we also considered quasi-physical constraints, such that each sphere should make six contacts on average, and that there should be at least three contacts in any arbitrary hemisphere. Gamba, while he has taken plausible averages, does not take these or equivalent conditions into account. Moreover, by adding a similar unit to the original in a symmetrical manner to facilitate density measurement, he calculates an average over the unit cell of a primitive crystal lattice. It seems to us that his two-dimensional density is a weighted average over all possible unit cells from the square (density 0.785,  $a=90^\circ$ ) to the hexagonal (density 0.907,  $a=60^\circ$ ), whereas the three-dimensional value is an average over all possible primitive unit cells from the simple cubic (density 0.524,  $a=b=c=90^\circ$ ) to the hexagonal and face centred cubic (density 0.704). We do not think this represents random packing.

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<sup>1</sup> Finney, J. L., *Nature*, **242**, 398 (1973).

<sup>2</sup> Stanley, H. E., *Introduction to Phase Transitions and Critical Phenomena*, 17 (Oxford University Press, 1971).

<sup>3</sup> Polya, G., *Math. Ann.*, **84**, 149 (1921); Montroll, E. W., and Weiss, G. H., *J. math. Phys.*, **6**, 167 (1968).

<sup>4</sup> Smith, C. S., *Metal Interfaces*, 65 (*Am. Soc. Metals*, Cleveland, 1965).

<sup>5</sup> Finney, J. L., *Proc. R. Soc.*, **A319**, 479 (1970).

<sup>6</sup> Quickenden, J. I., and Tan, G. K., *J. Colloid Interface Sci.*, **48**, 382 (1974).

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