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Interstitial space in hard-sphere clusters* By E. N. Sickafus, *Scientific Research Staff, Ford Motor Company, Dearborn, Michigan* 48121, *U.S.A.* and Neil A. Mackie, *Physics Department, University of Denver, Denver, Colorado, U.S.A.*

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The interstitial space in a cluster of spheres is examined to determine the largest sphere that can be placed in its voids. A method is given for obtaining the interstitial sphere belonging to a group of arbitrarily arranged spheres by examining tetrahedral configurations. Given the coordinates of the centers and the radii of the spheres of a tetrahedral group, the coordinates and radius of the tetrahedral interstitial sphere can be found. The method can be applied to interstices of any coordination number. It is applicable to sphere packings with or without crystallographic symmetry.

The packing of hard spheres is often used as a model in analyses of crystalline structure, molecular structure, aeration beds, and other structures. Such models reveal an interesting interstitial space whose geometry is amenable to further sphere packing – and so on, ad infinitum. The interstitial space can be examined from tetrahedral groups of 'defining' spheres – spheres that bound the interstice.

In this note an exact solution is given for the problem of calculating the size and location of an interstial sphere that can be inscribed in a tetrahedral group of non-coplanar spheres.

In a recent paper Mackay (1973) presented a generalization of the problem to a four-space simplex in the form of a determinant. He suggests that an approximate solution to the interstitial-sphere radius can be obtained by an iterative solution (pivotal convergence) of his determinant. Some years ago we derived an exact solution for this problem which we have used in various analyses of crystallographic structures. We developed a generalized computer program for examining the 14 Bravais lattices and the 230 space groups and use a preferred-orientation solution for ease in desk-type calculations. Exact solutions are obtained for the radius R_c of an interstitial sphere and for the coordinates of its center (x_c,y_c,z_c). A brief summary of our analysis follows.

The analysis is based on finding the largest sphere that can be placed within a given region of interstitial space without overlapping the bounding spheres. In essence this is a three-dimensional formulation of the famous problem of Apollonius† (see for example, Courant & Robbins, 1941). However, in the case of finding a sphere that is tangent to four given spheres, there will not necessarily be a solution for every arbitrarily arranged group. The existence of an interstial-sphere solution for an arbitrary defining group is characterized by positive definite roots in the interstitial equation. In geometries where more than four spheres may be tangent simultaneously to their interstitial sphere, any four non-coplanar members of this group constitute a defining tetrahedral group. This point is a useful aid when examining higher-order coordination groups.

In a tetrahedral group of non-coplanar spheres there are four points of tangency to their common interstitial sphere (if one exists). These points lie on four lines joining the centers of the spheres in the tetrahedral group with the center of their common interstitial sphere. If the coordinates and radii of the defining group are given by x_i,y_i,z_i and R_i and corresponding values for the interstitial sphere are given by x_c,y_c,z_c, and R_c then the latter are defined by the simultaneous solution of four distance equations of the form:

$$(x_i-x_c)^2+(y_i-y_c)^2+(z_i-z_c)^2=(R_i+R_c)^2 \quad (1)$$

where $i=0,1,2,3$. The sum of the radii is used to define 'external' tangency, meaning that no overlap occurs between the interstitial sphere and the defining group.

The solutions to equations (1) can be expressed exactly as follows:

$$x_c=\frac{E}{D}+\frac{F}{D}R_c\,,\quad y_c=\frac{G}{D}+\frac{H}{D}R_c\,,$$
$$z_c=\frac{P}{D}+\frac{Q}{D}R_c\,, \quad (2)$$

and

$$R_c=\frac{U}{T}\left[1\pm\left(1-\frac{VT}{U^2}\right)^{1/2}\right],$$

where

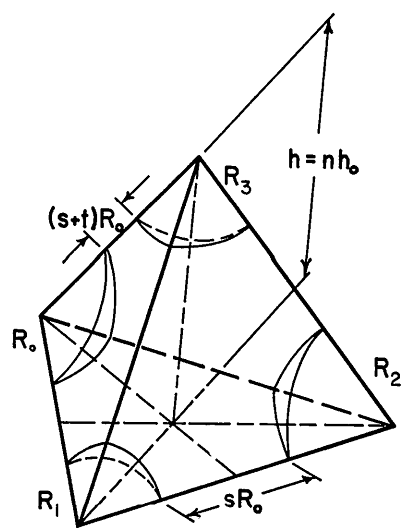

Fig. 1. The tetrahedron formed by the centres of the defining spheres in a configuration.

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† In the third century B.C., Apollonius of Perga posed the problem of finding the circle that is tangent to any given set of three (coplanar) circles.

$$T=\left(\frac{F}{D}\right)^2+\left(\frac{H}{D}\right)^2+\left(\frac{Q}{D}\right)^2-1\,,$$

$$U=\left(x_0-\frac{E}{D}\right)\frac{F}{D}+\left(y_0-\frac{G}{D}\right)\frac{H}{D}+\left(z_0-\frac{P}{D}\right)\frac{Q}{D}+R_0\,,$$

$$V=\left(x_0-\frac{E}{D}\right)^2+\left(y_0-\frac{G}{D}\right)^2+\left(z_0-\frac{P}{D}\right)^2-R_0^2\,,$$

and D, E, F, G, H, P, and Q are determinants.

The determinant D is defined as

$$D=\begin{vmatrix} x_0-x_1 & x_0-x_2 & x_0-x_3 \\ y_0-y_1 & y_0-y_2 & y_0-y_3 \\ z_0-z_1 & z_0-z_2 & z_0-z_3 \end{vmatrix}$$

from which the other determinates can be obtained. The absolute value of D is six times the volume of the tetrahedron whose vertices are located at the points whose coordinates are given in D (*e.g.*, Olmsted, 1947). A real solution to the interstitial problem requires that $D\neq 0$. To obtain the other determinants, first set

$$C_i^2=x_i^2+y_i^2+z_i^2 \quad (i=0,1,2,3)\,,$$

and

$$A_j=C_0^2-C_j^2+R_j^2-R_0^2 \quad (j=1,2,3)\,.$$

Then $2E$, $2G$, or $2P$ may be obtained by replacing respectively the first, second, or third rows of D with the row $[A,B,C]$, where $A\equiv A_1$, $B\equiv A_2$, and $C\equiv A_3$. Similarly, F, H, and Q are obtained using the row $[(R_1-R_0), (R_2-R_0), (R_3-R_0)]$ to replace respectively the first second, and third rows of D.

A useful simplification of the above equations results when a preferred orientation of the defining tetrahedron is selected, namely $R_0(0,0,0)$, $R_1(x_1,0,0)$, $R_2(x_2,y_2,0)$, and $R_3(x_3,y_3,z_3)$. When applied to a defining group of equal radii spheres ($R_0=R_1=R_2=R_3$), a common problem, we find that $F=H=Q=0$ so that $T=-1$, $U=R_0$, $D=-x_1y_2z_3$, and $V=(x_1/2)^2+(G/D)^2+(P/D)^2$ which is also $=x_c^2+y_c^2+z_c^2$. The interstitial sphere is characterized by $x_c=x_1/2$, $y_c=G/D$, $z_c=P/D$ and $R_c=\sqrt{V}-R_0$. A solution exists if $V>R_0^2$ which requires that $(x_1/2)^2+(G/D)^2+(P/D)^2>R_0^2$. Since $|x_1/2|$ is always greater than R_0 in hard-sphere packing *there always exists a uniquely defined interstitial sphere for a non-coplanar tetrahedral group of equal-radii spheres no matter how they may be arranged in space.*

Some useful results of the above analysis applied to a group of equal radii spheres are summarized below: Four parameters are used to specify the configuration of the defining group, see Fig. 1. In each configuration the base of the tetrahedron is equilateral. Relative to this base; (1) the altitude of the tetrahedron is $h=\eta h_0$, where h_0 is the altitude when the apex sphere (R_3) is in contact with the basal spheres, (2) the separation of the basal spheres is sR_0, and (3) the separation of the apex sphere from the basal spheres is $(s+t)R_0$. A fourth parameter is introduced when one sphere (the apex sphere) has its radius varied from that of $R_0=R_1=R_2$, that is, when $R_3=pR_0$. The first three parameters constitute 'packing constraints' for hard spheres if $s\geq 0$, $t\geq 0$, and $\eta\geq 1$. Radius ratios, R_c/R_0, were found as functions of these parameters for several cases:

(I) Variation of η with $R_0=R_1=R_2=R_3$, and $s=0$.

$$\frac{R_c}{R_0}=\sqrt{\tfrac{2}{3}}\eta+\tfrac{1}{3}\sqrt{\tfrac{3}{2}}\,\frac{1}{\eta}-1\,.$$

If $\eta\geq 7{\cdot}04$ then $R_c/R_0=0{\cdot}817\eta-1$ within a 1% error. The radius ratio is unity, $R_c/R_0=1$, when $\eta=\sqrt{\tfrac{3}{2}}+1$ or $\sim 2{\cdot}225$.

(II) Variation of s with $R_0=R_1=R_2=R_3$ and $t=0$

$$\frac{R_c}{R_0}=\tfrac{1}{2}\sqrt{\tfrac{3}{2}}S+\sqrt{\tfrac{3}{2}}-1\,.$$

The radius ratio is unity when $s=4\sqrt{\tfrac{2}{3}}-2$ or $\sim 1{\cdot}265$.

(III) Variation of t with $R_0=R_1=R_2=R_3$ and $S=0$

$$\frac{R_c}{R_0}=\frac{(2+t)^2}{2[(2+t)^2-\frac{4}{3}]^{1/2}}-1\,.$$

If $t\geq 6{\cdot}23$ then $R_c/R_0=t/2$ within a 1% error. The radius ratio is unity when $t=\sqrt{8}\sqrt{(1+\sqrt{\tfrac{2}{3}})}-2$ or $\sim 1{\cdot}812$.

(IV) Variation of s, t, and p where $R_0=R_1=R_c\neq R_3=pR_0$.

$$\frac{R_c}{R_0}=\frac{(1+s+t+p)^2-1+p^2-2p\sqrt{(1+s+t+p)^2-\frac{1}{3}(2+s)^2}}{2[1-pt\sqrt{(1+s+tsp)^2-\frac{1}{3}(2+s)^2}]}$$

and the variation of η with $s=0$ yields

$$\frac{R_c}{R_0}=\frac{1+3p^2+\eta^2[3(1+p)^2-4]-6p\eta\sqrt{(1+p)^2-\frac{4}{3}}}{6[1+p+\eta\sqrt{(1+p)^2-\frac{4}{3}}]}\,.$$

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