From (36), (38) and (41), we get

$$\begin{aligned} K_0 &= 3(1+4\alpha)/4(1+\alpha)^2, \\ K_1 &= -3(1+2\alpha^2)/4(1+\alpha)^2, \\ K_2 &= 3(1-2\alpha)(1-2\alpha^2)/4(1+\alpha)^2. \end{aligned} \tag{42}$$

Following Gevers (1954) and Holloway (1969*a*), the diffracted intensity for the diffuse reflexions is given by

$$I_d(h_3) = K_0 + \left[2\sum_{j=0}^{3} A_j \cos(\pi j h_3) \Big/ \sum_{j=0}^{3} B_j \cos(\pi j h_3)\right], \tag{43}$$

where

$$\begin{aligned} A_0 &= b_2K_1 + b_1(K_2 + b_2K_1) - b_0^2K_0, \\ A_1 &= (1+b_1)K_1 + (b_2+b_0)(K_2+b_2K_1) - b_1b_0K_0, \\ A_2 &= b_0K_1 + K_2 + b_2K_1 - b_2b_0K_0, \\ A_3 &= -b_0K_0, \\ B_0 &= 1 + b_2^2 + b_1^2 + b_0^2, \\ B_1 &= 2(b_2 + b_2b_1 + b_1b_0), \\ B_2 &= 2(b_1 + b_2b_0), \\ B_3 &= 2b_0. \end{aligned} \tag{44}$$

Substituting from (40), (42) and (44) in (43), we have after simplification

$$\begin{aligned} I_d(h_3) = \{&3\alpha(1-\alpha)[2 - 3\alpha + 2\alpha^2 + (1+\alpha-2\alpha^2) \\ &\times \cos \pi h_3 + 2\alpha\cos^2 \pi h_3]\}/\{2(1+\alpha) \\ &\times [1 + \alpha^2 + 2\alpha\cos\pi h_3] \\ &\times [(1-\alpha)^2 + (1-\alpha^2) \\ &\times \cos\pi h_3 + 2\alpha\cos^2\pi h_3]\}, \end{aligned} \tag{45}$$

which is identical with the expression obtained by Lele, Anantharaman & Johnson (1967) and Holloway (1969*b*). It may be noted that except for the root with unit modulus and the corresponding integrated intensity, which could be obtained very simply, no other root or integrated-intensity values were necessary for the calculations.

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The Fragile Lattice Packings of Spheres in Four-Dimensional Space

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Abstract

In a fragile packing of spheres, the density of the packed spheres is minimized. There are exactly nine distinct indecomposable fragile lattice packings in four-dimensional space; they are described in terms of their associated quadratic forms.

The *three-dimensional* fragile lattice packings of spheres were determined by Fields (1980). There are only the simple cubic, simple hexagonal, body-centered cubic and body-centered tetragonal packings. Of these lattices, only the latter two are indecomposable: the simple cubic is the orthogonal sum of three one-dimensional lattices, and the simple hexagonal lattice is

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the orthogonal sum of a one-dimensional lattice and the two-dimensional stable lattice. In general, it is clear that any decomposable fragile lattice is the orthogonal sum of perfect and fragile lattices of lower dimension (*cf.* Fields, 1979). Our purpose here, therefore, is to determine the *indecomposable* fragile lattices in $\mathbf{R}^4$: we determine that there are exactly nine. Our method for finding them is based on the one used in the classification in $\mathbf{R}^3$. It depends on the following results about positive-definite quadratic forms:

(1) a form is fragile if and only if it is weakly eutactic and not perfect (Fields, 1979; *cf.* Lekkerkerker, 1969);

(2) any four linearly independent minimum vectors of a positive-definite quadratic form in four variables must generate the entire integer lattice, with one exception which occurs among the minimum vectors of one of the two perfect forms (Korkine & Zolotareff, 1877; *cf.* van der Waerden, 1956);

(3) a fragile form in n (=4) variables has n (=4) linearly independent minimum vectors (Fields, 1980);

(4) there is at most one fragile form possessing a given complete set of minimum vectors (Fields, 1979).

To find the fragile forms in four variables, therefore, it suffices to find those forms which are weakly eutactic with respect to their set S of minimum vectors, *i.e.* which satisfy

$$A^{-1} = \sum_{\mathbf{m} \in S} \rho_{\mathbf{m}} \, \mathbf{m}^t \, \mathbf{m},$$

where A is the matrix of our quadratic form, the minimum vectors $\mathbf{m}$ are represented as row matrices, and the $\rho_{\mathbf{m}}$ are some real numbers. Results (2) and (3) allow us to restrict our attention to sets of minimum vectors whose entries are either 0 or ± 1; there are 40 pairs of such non-zero vectors and their negatives. Results (2) and (3) also allow us to assume that the four unit vectors are among the minimum vectors of our fragile form, since we are interested in classifying these forms up to integral equivalence only.

Once the fragile forms have been found, sets of vectors which form bases of the fragile lattices can be determined *via* the classical method of finding the 'principal axes' of these forms (*cf.* Courant & Hilbert, 1937, pp. 23ff.). We give here the *Bravais type* of each lattice, corresponding to the general classification of four-dimensional lattices as given by Wondratschek, Bulow & Neubuser (1971) and Brown, Bulow, Neubuser, Wondratschek & Zassenhaus (1978). We also list for each lattice

(*a*) the order of its point group,

(*b*) the coordination number (CN),

(*c*) the sizes of the orbits of the point group acting on the set of nearest neighbors.

Determining the particular Bravais types was accomplished by computing the point group of each fragile lattice; recall that this group is isomorphic with the group of (4×4) matrices A with integral entries which satisfy

$$A^t B A = B,$$

where B is the corresponding fragile form. Determining the Bravais type was then easy whenever there was only one *indecomposable* Bravais lattice in the crystal system which the point group determined. In two of the crystal systems where there were more than one indecomposable Bravais lattice (system 25 for form III, and system 6 for forms V and VI), it became necessary to compute the orbits of minimum vectors and common eigenvectors to distinguish between the possible Bravais types.

Each fragile form listed below is integrally equivalent to a form of the same Bravais type as listed in Table 2*C* of Brown, Bulow, Neubuser, Wondratschek & Zassenhaus (1978). Once this identification is made, a primitive basis can be found *via* the centering matrix provided by Brown *et al.* (1978, Table 4*C*). However, since there is no direct procedure, short of a computer search, for finding the particular values of the parameters of the forms of Brown *et al.* (1978, Table 2*C*) which correspond to the forms on our list, we do not consider here the problem of finding primitive bases for the fragile lattices.

While the computation needed for the classification in $\mathbf{R}^3$ could be done by hand in a few hours, machine computation is absolutely essential in the four-dimensional case. More precisely, we programmed our computer to find indecomposable symmetric positive-definite matrices

$$A = \begin{matrix} 1 & a & b & c \\ & 1 & d & e \\ & & 1 & f \\ & & & 1 \end{matrix}$$

which satisfy the k linear equations

$$A[\mathbf{m}_1] = \ldots = A[\mathbf{m}_k] = 1$$

and the ten non-linear equations

$$A^{-1} = p_1 \, \mathbf{e}_1^t \, \mathbf{e}_1 + \ldots + p_4 \, \mathbf{e}_4^t \, \mathbf{e}_4 + q_1 \, \mathbf{m}_1^t \, \mathbf{m}_1 + \ldots + q_k \, \mathbf{m}_k^t \, \mathbf{m}_k,$$

where $p_i, q_j \in \mathbf{R}$, $\mathbf{e}_1, \ldots, \mathbf{e}_4$ are the four unit vectors, and $\mathbf{m}_1, \ldots, \mathbf{m}_k$ is an arbitrary subset of integral vectors whose entries are either 0 or ± 1. There are $10 + k$ unknowns: $a, \ldots, f, p_1, \ldots, p_4, q_1, \ldots, q_k$. The number k can be either 1, 2, 3, 4 or 5, depending on whether our lattice is to have 5, 4, 3, 2, or 1 degree of freedom [the only fragile lattice with six degrees of freedom (corresponding to the case $k = 0$) is the integer lattice, which is decomposable].

By reduction theory (*cf.* Lekkerkerker, 1969), any positive-definite solution A is integrally equivalent to a solution whose coefficients satisfy

$$0 \le a \le d \le f \le \tfrac{1}{2}, \qquad -\tfrac{1}{2} \le b,c,e \le \tfrac{1}{2},$$

so we may impose these additional constraints without loss of generality.

It was necessary to verify that for each set of vectors $\mathbf{m}_1, \ldots, \mathbf{m}_k$ for which our program yielded no solution, there was in fact no solution possible. It was also necessary to weed out those solutions which were perfect (one of the two perfect forms appeared as a solution), and those solutions for which the vectors $\mathbf{e}_1, \ldots, \mathbf{e}_4, \mathbf{m}_1, \ldots, \mathbf{m}_k$ were not minimum vectors. There were three such 'extraneous solutions' (listed below), for which these vectors turned out to be *secondary* minima. Finally, we had to determine which of our solutions were integrally equivalent.

In addition to the fragile forms themselves, we also list for each form

(*d*) its determinant (det),

(*e*) the minimum vectors $\mathbf{m}_1, \ldots, \mathbf{m}_k$,

(*f*) the values of the constants $p_1, \ldots, p_4, q_1, \ldots, q_k$.

The density of the corresponding lattice packing of spheres is given by *density* $= \pi^2/32\sqrt{\det}$. The numbers (*a*) (*b*) (*d*) and the sets of numbers (*c*) and (*f*) are invariant under integral transformations of the form (orthogonal transformations of the lattice). We would also like to point out the following:

(i) Two fragile forms, I and IX, are absolutely symmetric (*cf.* Fields, 1979). Form I is the analogue of the body-centered cubic form; it has 240 integral automorphs and is dual to (a scalar multiple of) the icosahedral stable form (*SN* centered). It also determines the least-dense indecomposable fragile lattice packing in $\mathbf{R}^4$. The di-isohexagonal othogonal form IX has 144 automorphs and is self dual (up to a scalar multiple). It determines the densest fragile lattice packing in $\mathbf{R}^4$. The two absolutely symmetric forms are the only indecomposable fragile forms which are strongly eutactic (*i.e.* for which $p_1 = \ldots = p_4 = q_1 = \ldots = q_k$). Both of the two stable lattice packings in $\mathbf{R}^4$ are denser than the densest fragile lattice packing.

(ii) Each fragile form in four variables has an automorphism group of order at least 16; *i.e.* each fragile lattice in $\mathbf{R}^4$ has a point group of order at least 16.

(iii) All but one of the fragile forms are eutactic (*i.e.* each p_i and q_j is strictly positive). The one exception is form III. Both it and form IV have the same number of minimum vectors and the same determinant, so that the packings they determine are equally dense, and the spheres in each packing have the same number of nearest neighbors; packing III exhibits much greater symmetry, however.

(iv) Two fragile forms, V and VII, have incommensurable coefficients. Their appearance came as a great surprise.

The indecomposable four-dimensional fragile forms and lattices

I. *Icosahedral*

Bravais type XXII/I
Order = 240
CN = 10
1 orbit of 10 vectors
det $= 5^3/2^8 = 0{\cdot}4883$
$\mathbf{m}_1 = (1,-1,1,-1)$
$p_1 = p_2 = p_3 = p_4 = q_1 = 4/5$

$$\begin{array}{cccc} 1 & \frac{1}{4} & -\frac{1}{4} & \frac{1}{4} \\ & 1 & \frac{1}{4} & -\frac{1}{4} \\ & & 1 & \frac{1}{4} \\ & & & 1 \end{array}$$

II. *Hexagonal orthogonal*

Bravais type XI/II
Order = 24
CN = 12
3 orbits: 6/4/2
det $= 2^2/3^2 = 0{\cdot}4444$
$\mathbf{m}_1 = (0,1,-1,1)$, $\mathbf{m}_2 = (1,-1,1,-1)$
$p_1 = 2/3, p_2 = p_3 = p_4 = 3/4, q_1 = 5/12, q_2 = 2/3$

$$\begin{array}{cccc} 1 & \frac{1}{6} & -\frac{1}{6} & \frac{1}{6} \\ & 1 & \frac{1}{3} & -\frac{1}{3} \\ & & 1 & \frac{1}{3} \\ & & & 1 \end{array}$$

III. *Cubic orthogonal*

Bravais type XVII/VI
Order = 96
CN = 14
2 orbits: 12/2
det $= 3^3/2^6 = 0{\cdot}421875$
$\mathbf{m}_1 = (0,0,1,-1)$, $\mathbf{m}_2 = (0,1,0,-1)$, $\mathbf{m}_3 = (1,0,0,-1)$
$p_1 = p_2 = p_3 = 2/3, p_4 = 0, q_1 = q_2 = q_3 = 2/3$

$$\begin{array}{cccc} 1 & \frac{1}{4} & \frac{1}{4} & \frac{1}{2} \\ & 1 & \frac{1}{4} & \frac{1}{2} \\ & & 1 & \frac{1}{2} \\ & & & 1 \end{array}$$

IV. *Tetragonal orthogonal*

Bravais type X/I
Order = 16
CN = 14
3 orbits: 8/4/2
det $= 3^3/2^6 = 0{\cdot}421875$
$\mathbf{m}_1 = (0,0,1,-1)$, $\mathbf{m}_2 = (0,1,-1,0)$, $\mathbf{m}_3 = (1,-1,1,0)$
$p_1 = p_2 = p_4 = 2/3, p_3 = 1/3, q_1 = q_3 = 2/3, q_2 = 1/3$

$$\begin{array}{cccc} 1 & \frac{1}{4} & -\frac{1}{4} & -\frac{1}{8} \\ & 1 & \frac{1}{2} & \frac{1}{4} \\ & & 1 & \frac{1}{2} \\ & & & 1 \end{array}$$

V. *Orthogonal*

Bravais type V/VIII
Order = 16
CN = 14
3 orbits: 8/4/2
det $= (35 + 13\sqrt{13})/216 = 0{\cdot}3790$

$$\begin{array}{cccc} 1 & a & (2a-1) & a \\ & 1 & a & \frac{1}{2} \\ & & 1 & a \\ & & & 1 \end{array}$$

$[a = (7 - \sqrt{13})/12]$

$\mathbf{m}_1 = (0,1,0,-1)$, $\mathbf{m}_2 = (1,0,1,-1)$, $\mathbf{m}_3 = (1,-1,1,0)$
$p_1 = p_4 = 3(1 - p_2)/2, p_2 = p_3 = q_2 = q_3$
$= (33 + 3\sqrt{13})/(35 + 13\sqrt{13}), q_1 = 1 - p_2$

VI. *Orthogonal*

Bravais type V/VIII
Order = 16
CN = 16
4 orbits: 8/4/2/2
det $= 3/2^3 = 0{\cdot}375$
$\mathbf{m}_1 = (0,0,1,-1)$, $\mathbf{m}_2 = (0,1,-1,0)$, $\mathbf{m}_3 = (0,1,-1,1)$, $\mathbf{m}_4 = (1,-1,1,-1)$
$p_1 = 2/3, p_2 = p_3 = p_4 = 1/2, q_1 = q_2 = 1/2,$
$q_3 = 1/6, q_4 = 2/3$

$$\begin{array}{cccc} 1 & \frac{1}{4} & 0 & \frac{1}{4} \\ & 1 & \frac{1}{2} & 0 \\ & & 1 & \frac{1}{2} \\ & & & 1 \end{array}$$

VII. *Tetragonal orthogonal*

Bravais type X/I
Order = 16
CN = 16
2 orbits: 8/8
det = $(6\sqrt{3}-9)/4 = 0{\cdot}3481$

$$\begin{array}{cccc} 1 & \frac{1}{2} & b & \frac{1}{2} \\ & 1 & \frac{1}{2} & b \\ & & 1 & \frac{1}{2} \\ & & & 1 \end{array}$$

$[b = (\sqrt{3}-1)/2]$

$\mathbf{m}_1 = (0,0,1,-1)$, $\mathbf{m}_2 = (0,1,-1,0)$, $\mathbf{m}_3 = (1,0,0,-1)$, $\mathbf{m}_4 = (1,-1,0,0)$

$p_1 = p_2 = p_3 = p_4 = (3\sqrt{3}-5)/(2\sqrt{3}-3)$,
$q_1 = q_2 = q_3 = q_4 = 1 - p_1$

VIII. *Hexagonal orthogonal*

Bravais type XI/II
Order = 24
CN = 18
2 orbits: 12/6
det = $1/3 = 0{\cdot}3333$

$$\begin{array}{cccc} 1 & \frac{1}{2} & \frac{1}{2} & \frac{1}{6} \\ & 1 & \frac{1}{2} & 0 \\ & & 1 & \frac{1}{2} \\ & & & 1 \end{array}$$

$\mathbf{m}_1 = (0,0,1,-1)$, $\mathbf{m}_2 = (0,1,-1,0)$, $\mathbf{m}_3 = (0,1,-1,1)$, $\mathbf{m}_4 = (1,0,-1,0)$, $\mathbf{m}_5 = (1,-1,0,0)$

$p_1 = p_4 = 1/2$, $p_2 = p_3 = 1/3$,
$q_1 = q_3 = q_4 = q_5 = 1/2$, $q_2 = 1/3$

IX. *Di-isohexagonal orthogonal*

Bravais type XXI/I
Order = 144
CN = 18
1 orbit: 18
det = $3^4/2^8 = 0{\cdot}3164$

$$\begin{array}{cccc} 1 & \frac{1}{4} & -\frac{1}{2} & -\frac{1}{2} \\ & 1 & \frac{1}{4} & -\frac{1}{2} \\ & & 1 & \frac{1}{4} \\ & & & 1 \end{array}$$

$\mathbf{m}_1 = (0,1,0,1)$, $\mathbf{m}_2 = (0,1,-1,1)$, $\mathbf{m}_3 = (1,0,0,1)$, $\mathbf{m}_4 = (1,0,1,0)$, $\mathbf{m}_5 = (1,-1,1,0)$

$p_1 = p_2 = p_3 = p_4 = q_1 = q_2 = q_3 = q_4 = q_5 = 4/9$

Final remarks

There were three other 'extraneous' positive-definite quadratic forms which were found by our algorithm. They have the following properties:

(1) some subset S of integral solutions $\mathbf{m}$ of the equation $A[\mathbf{m}] = 1$ contains four vectors which generate the entire integer lattice $\mathbf{Z}^4$;

(2) A is not perfect with respect to S: there exists a form $B \neq A$ such that $A[\mathbf{m}] = B[\mathbf{m}]$ for all $\mathbf{m} \in S$;

(3) A is weakly eutactic with respect to S:

$$A^{-1} = \sum_{\mathbf{m} \in S} p_{\mathbf{m}} \mathbf{m}^t \mathbf{m} \text{ for some } p_{\mathbf{m}} \in \mathbf{R}.$$

Each of these forms has two pairs of minimum vectors; the sets S contain certain *secondary* minima of these forms. The three forms are:

$$(a)\ \begin{array}{cccc} 1 & \frac{3}{8} & \frac{1}{2} & \frac{3}{8} \\ & 1 & \frac{3}{8} & -\frac{1}{2} \\ & & 1 & \frac{3}{8} \\ & & & 1 \end{array}; \qquad (b)\ \begin{array}{cccc} 1 & \frac{1}{2} & \frac{1}{4} & -\frac{1}{2} \\ & 1 & \frac{1}{2} & -\frac{1}{4} \\ & & 1 & \frac{1}{2} \\ & & & 1 \end{array}; \qquad (c)\ \begin{array}{cccc} 1 & a & b & -\frac{1}{2} \\ & 1 & \frac{1}{2} & e \\ & & 1 & \frac{1}{2} \\ & & & 1 \end{array},$$

$a = 1 + b + e = 0{\cdot}3892$
$e = (2b^2 + 7b + 2)/(6b + 3) = -0{\cdot}2571$
$b = x + \frac{1}{2}$, where $2x^3 - 12x - 9 = 0$
$= 2\sqrt{2} \cos [\frac{1}{3}(\cos^{-1} 9/8\sqrt{2}) + 240°] + \frac{1}{2}$
$= -0{\cdot}3537.$

The corresponding 'extraneous solution' in three dimensions is the I-centered tetragonal form

$$\begin{array}{ccc} 1 & \frac{1}{2} & \frac{3}{4} \\ & 1 & \frac{3}{4} \\ & & 1 \end{array}$$

which has four minima (CN = 4) and twelve secondary minima (*cf.* Fields, 1980, *Lemma* 3, Case IV).

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