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## Relaxation of Mackay Icosahedra

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### Abstract

Multilayer icosahedra, first introduced about twenty years ago by Mackay [*Acta Cryst.* (1962), **15**, 916–918], are no longer considered a geometrical curiosity, as small icosahedral particles have been observed in a great number of experiments. The hard-sphere models, previously considered, are not really suited to the study of physical properties because they fail to express the important stresses due to the icosahedral structure. Therefore, Mackay icosahedra, made of atoms interacting through a Lennard-Jones potential, were constructed and allowed to relax freely. Results of the calculation are given, consisting of a detailed description of relaxed icosahedra with up to nine layers, *i.e.* with up to almost 3000 atoms.

### I. Introduction

Mackay (1962) noted that a cuboctahedron formed of rigid rods can be transformed into an icosahedron, it being sufficient for this that one of the diagonals of each square face be contracted to the length of the edge of a primitive cuboctahedron, while at the same time the face is folded following the same diagonal so as to form two equilateral faces (Fig. 1). This transformation to an icosahedron is very simple to visualize for a cuboctahedron of 13 atoms containing a single layer of atoms surrounding a central one, and for the cuboctahedron of 55 atoms, which contains an additional layer of atoms, it is sufficient that two layers are deformed at the same time, each square (100) face being transformed into two equilateral (111) faces. Thus to each cuboctahedron

there corresponds an icosahedron presenting, on the one hand, the same number of layers and, on the other, in a layer of index  $n$ , the same number of atoms in the layer ( $10n + 2$ ) and on the corresponding edge ( $n + 1$ ). It is remarkable that the former packing is crystallographic while the latter, having fivefold symmetry, is not.

Mackay described the icosahedron as made up of 20 identical tetrahedra possessing a common vertex and connected with each other through adjacent faces, each of which forms twinning planes, the icosahedron and the tetrahedra from which it is composed possessing the same number of atoms on each edge. To a first approximation the tetrahedra represent a cubic close-packed structure having thus four (111) faces; in reality however this structure is slightly deformed, because the tetrahedra are not absolutely regular. In fact the three radial edges derived from the common vertex are equal, just as the three surface edges are, but the latter are longer than the former by approximately 5%. At the same time the three faces of the tetrahedra within the icosahedron are not precisely equilateral, the angle at the center being slightly enlarged and taking a value of  $63^\circ 26'$ .

Mackay calculated the effective density for the case of an icosahedron of hard spheres. The radial interatomic distances are thus put equal to the diameter of the spheres while the lateral distances are somewhat larger. Because of this, he found an effective density somewhat less than that of the face-centered cubic structure (0.69 as against 0.74). It therefore appeared to him 'somewhat unlikely that a large number of atoms would naturally adopt a multilayer icosahedral arrangement', all the less probable it would seem in view of the simplicity of the mechanism of transition to the face-centered cubic structure which he had clearly demonstrated.

These studies of models with pentagonal symmetry remained almost unnoticed until in 1966 (Ino, 1966; Gillet & Gillet, 1966; Allpress & Sanders, 1967) the discovery by electron microscopy of icosahedral particles in thin gold films caused a revival of interest. Numerous experimental confirmations have appeared subsequently in which the existence of such particles is

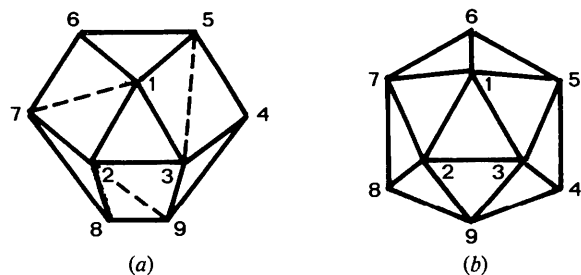


Fig. 1. Transformation of (a) a cuboctahedron into (b) an icosahedron.

attributed either to more favorable kinetic paths of growth (Gillet & Gillet, 1972) or to a greater thermodynamic stability (Farges, de Feraudy, Raoult & Torchet, 1977).

At the same time one knows that the configuration of a hard-sphere model can at most only approximately represent that of an isolated aggregate. On applying a more realistic potential between the atoms of such a model, for example the Lennard-Jones potential, and allowing it to relax under the forces thus introduced, the atoms are found to be displaced from their respective sites towards equilibrium positions which vary according to the site in question. If one considers a microcrystalline model such as that based on the cuboctahedron, then the relaxation affects mainly the surface and its effect on internal interatomic distances in the model is found to be relatively weak and to be localized sufficiently that one may justify the use of a hard-sphere model for, at least, an approximate description of the corresponding crystalline aggregate. By contrast, if one considers an icosahedral model, its geometry imposes forces sufficiently strong that their effect in its whole structure becomes appreciably more important than that due to its surface.

The aim of the present paper is not to discuss the stability of the icosahedral models but to show the importance of the geometrical effects to which they are subject after relaxation. This we shall demonstrate qualitatively in § II with the help of a macroscopic model, which will also bring out the necessity of using relaxed icosahedral configurations in the study of the properties of such aggregates. In § III, we elucidate the calculation method which we use to allow free relaxation of icosahedral configurations. In § IV, a precise description of the relaxed configurations of icosahedral structures of from one to nine layers (*i.e.* of up to almost 3000 atoms) is given.

## II. Macroscopic approach

### 1. Stresses in a continuous icosahedron

Let the icosahedron be considered as a solid composed of 20 adjacent irregular tetrahedra, the tangential edges of which are some 5% longer than the radial ones. Let us further imagine each tetrahedron to be sectioned into prismatic elements of height  $h$ , with equilateral bases parallel to the surface.

Consider now the two such elements belonging to two adjacent tetrahedra – having one lateral face in common (Fig. 2). The extension of the tangential edges ensures that each section exerts on this face a tensile stress  $s_1$ , perpendicular to the common tangential edge and parallel to their respective bases. The resultant  $S_1$  of these two equal stresses lies in the common lateral face and is perpendicular to the common edge. The

external section of any one of the 20 tetrahedra is thus subject to a stress  $T_1$  which is the resultant of the three stresses  $S_1$  operating on its three lateral faces. This stress  $T_1$ , uniformly distributed, is perpendicular to the base of the rigid section. Furthermore, the stress  $T_1$  is transmitted to the whole tetrahedron and consequently contributes by compression to the 5% extension of the following section, this second section exerting on its lateral faces a tensile stress  $s_2$ , somewhat weaker than  $s_1$ . The stress  $T_2$  which thereby results is thus added to the stress  $T_1$ , in such a way that the second element is subject to the total stress  $T = T_1 + T_2$ .

In fact, since they are transmitted to decreasing base surfaces, stresses like  $T$  grow, while at the same time the additional stress  $T_n$  produced by each successive element depends on the extension provoked by the stress  $T$  due to the elements which it supports. If, beyond a certain depth,  $T$  is such that it alone would cause an extension greater than 5%, the lateral faces become subject to compressive stresses  $s$  reducing  $T$  in order to maintain the icosahedral geometry. Nevertheless  $T$  continues to grow indefinitely as one approaches the center of the icosahedron, because it acts upon elements whose surfaces tend to zero. Consequently, the increase in density of the elements, supposedly of the same height  $h$ , may itself become more and more important, as one goes from the surface towards the center of the icosahedron.

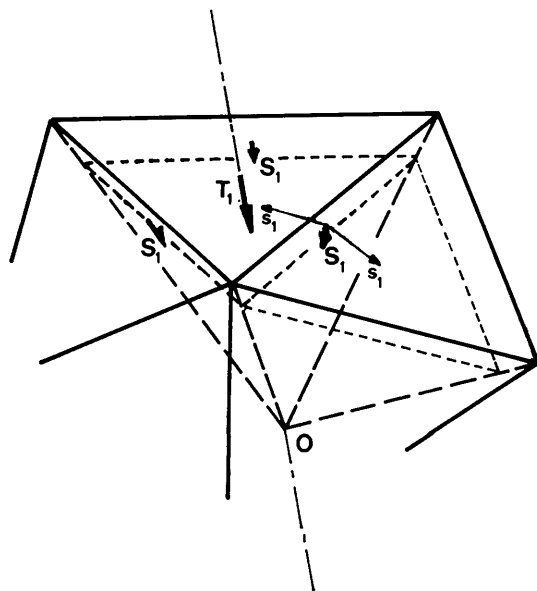


Fig. 2. Stresses exerted on the external section of one of the 20 tetrahedra forming a continuous icosahedron.  $S_1$  is the resultant of the two tensile stresses  $s_1$  exerted by adjacent sections on their lateral common face.  $T_1$  is the resultant of the three equivalent stresses  $S_1$ .

## 2. Form of the sections (Fig. 3)

It has been implicitly admitted in § 1 that the prismatic sections have an infinite rigidity and thus keep flat bases after relaxation. In fact, their rigidity being finite, the sections take up a certain convexity. On the other hand it is clear that the stresses are appreciably stronger in the region of the radial edges where five lateral faces coincide than in the center of the lateral faces. The tangential edges of each section consequently have a convexity which accentuates that of the sections themselves.

## III. Method of relaxation

Let us consider an icosahedron composed of  $N$  atoms and seek the configuration for which the total potential energy of interaction is a minimum when the pair potential between two atoms  $i$  and  $j$  a distance  $r_{ij}$  apart is given by the Lennard-Jones potential:  $V_{ij} = 4 (r_{ij}^{-12} - r_{ij}^{-6})$ .

To this end we allow the icosahedron to relax freely, each of its atoms being displaced under the action of the forces of each of its neighbors, to the point where the resultant of all such forces vanishes. Since the relaxation is static, and the initial configuration is compact, the structure remains icosahedral throughout the whole calculation. It is thus possible by taking account of the symmetry of this structure to reduce considerably the time of the calculation.

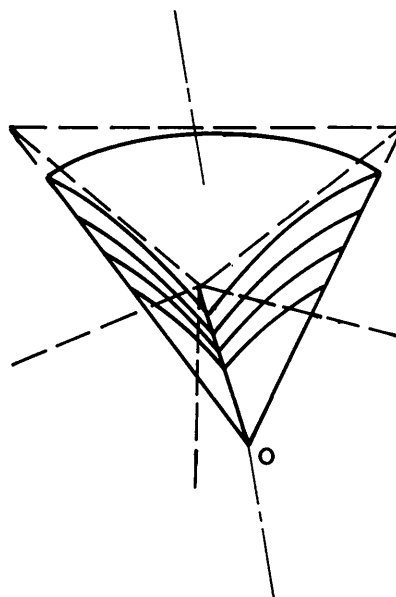


Fig. 3. Schematic form of the sections inside a tetrahedron, as a result of the stresses presented in Fig. 2. The convexity of a few sections has been suggested — and exaggerated for clearer presentation.

The calculation proceeds somewhat as follows.

(a) The initial configuration is taken to be a geometrically perfect icosahedron: its tangential distances between neighbors are equal and some 5% longer than their radial counterparts.

(b) The force  $\mathbf{F}_i$  exerted upon each atom  $i$  is calculated in magnitude and direction.

(c) We seek and find a new configuration in which the forces  $\mathbf{F}_i$  are lessened and the potential energy of the whole icosahedron is lower.

(d) We repeat the procedure displacing all atoms according to the new values of forces  $\mathbf{F}_i$  obtained in step c, and so on. In this way all the forces present in the icosahedron are progressively relaxed.

For each configuration the mean potential energy per atom is calculated according to

$$U = \frac{1}{N} \sum_i \sum_{j>i} V_{ij},$$

the mean force per atom according to

$$\bar{F} = \frac{1}{N} \sum_i ||\mathbf{F}_i||$$

and the standard deviation of this force  $\sigma_F = (\bar{F}^2 - \bar{F}^2)^{1/2}$  according to

$$\bar{F}^2 = \frac{1}{N} \sum_i ||\mathbf{F}_i||^2.$$

At the beginning of the relaxation calculation, the icosahedron would be far from its equilibrium configuration, the relative standard deviation  $\sigma_F/\bar{F}$  being quite large, of the order of one. In order to reduce the latter, we displaced the atoms proportionally to the square of the absolute value of the force exerted on them. In this way, it is possible to move those atoms more rapidly which are far from the equilibrium position, without the risk of displacing the ones which are relatively close. As soon as  $\sigma_F/\bar{F}$  becomes less than 15 or 20%, the displacement  $dr_i$  of atom  $i$  is taken to be proportional to the force  $\mathbf{F}_i$ ,  $dr_i = k\mathbf{F}_i$ .

The relaxation is held to be proceeding correctly if, from one configuration to the next, the three magnitudes  $U$ ,  $\bar{F}$  and  $\sigma_F$  simultaneously reduce in a monotonic fashion. Tests of  $\bar{F}$  and  $\sigma_F$  allow us to control the progress of the relaxation and if necessary modify its rate, raising or lowering the value of  $k$  throughout the calculation. For example, an increase of  $\sigma_F$  from one configuration to the next is an indication that the model has begun to deviate locally from equilibrium because  $k$  is too large.

Since the range of the potential extends over all relevant distances in the icosahedron, as the force on a particular atom diminishes, so the effect of its distant neighbors becomes correspondingly important. For example, it is impossible to relax freely an icosahedron

of more than three layers if one limits the range of the interaction potential to some three atomic diameters.

The question then arises as to how the calculation should be terminated. To this end, we observed the evolution of the various distance parameters with decrease in the forces  $\bar{F}$ . (Note by way of example the curve in Fig. 4, which shows how the radial distances of nearest neighbors,  $d_1$  at the center and  $d_9$  at the surface of a nine-layer icosahedron, vary linearly with  $\bar{F}$ , as one would expect in the neighborhood of an energy minimum.) Instead of keeping the extrapolated values for  $\bar{F}$  equal to zero for all distances present in the aggregate, we saved only those values obtained when  $\bar{F}$  had reduced to some  $10^{-14}$  N. Fig. 4 shows that if one takes for  $d_1$ , which is somewhat sensitive to  $\bar{F}$ , the value obtained for  $\bar{F}$  equal to  $2 \times 10^{-14}$  N instead of the extrapolated value, the resulting error is less than one part in  $10^{-4}$ .

With a relaxation program using the symmetry of the icosahedron, the time necessary for calculation was some two hours on the UNIVAC 1110 for relaxation of an icosahedron of nine layers ( $N = 2869$ ).

## IV. Results

### 1. Form of the faces

The coordinates of the atoms and the interatomic distances are given in reduced units (1 r.u. = 3.405 Å).

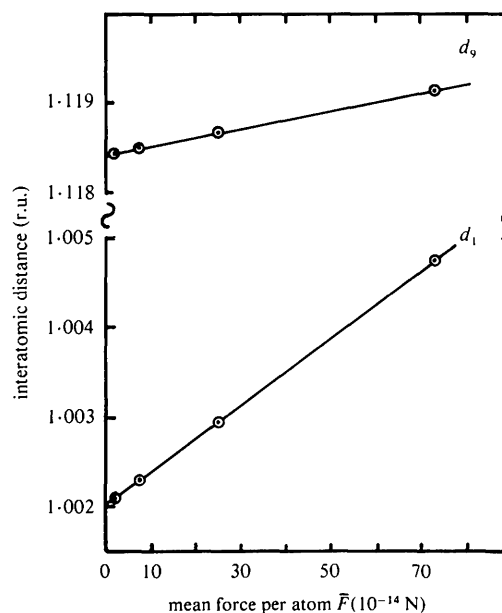


Fig. 4. Interatomic distances vs mean force per atom  $\bar{F}$  during the relaxation process. Complete relaxation corresponds to  $\bar{F} = 0$ .  $d_1$  and  $d_9$  are radial distances between nearest-neighbor atoms, at the center and at the surface respectively of the nine-layer icosahedron ( $N = 2869$ ). 1 r.u. = 3.405 Å.

The diagram in Fig. 5 shows the convex form of one of the twenty (111) faces at the surface of the six-layer icosahedron ( $N = 923$ ). The coordinates of atoms situated on an edge and on two perpendicular rows are reported in Fig. 6. The  $z$  axis originates from the atom situated at the centre  $O$  of the icosahedron, being normal to the given face, passes through the center of gravity of that face. The difference  $\delta z$  between the level at the center and that at a vertex is equal to 0.08, this representing some 9% of the mean distance separating the surface layer for  $n = 6$  from the immediately preceding layer  $n = 5$ .

In agreement with the form of this face the minimal interatomic distances ( $d = 1.115$  r.u.) are those which separate a vertex atom from its nearest neighbors, the maximal distances occurring at the center of the face ( $d = 1.125$  r.u.).

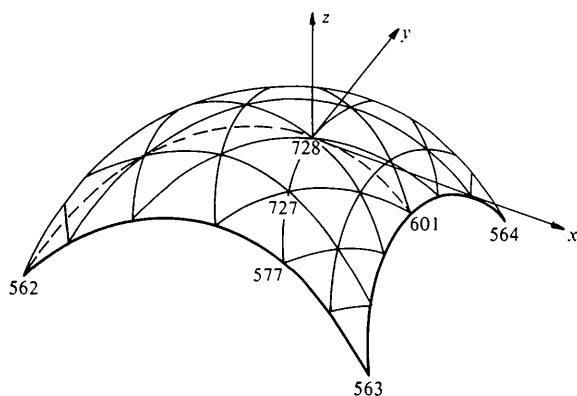


Fig. 5. Schematic view of one of the 20 faces at the surface of the six-layer icosahedron ( $N = 923$ ). Numbers refer to atom labeling in the relaxation calculation. The  $z$  axis originates from the icosahedron center and passes through the center of gravity of that face (atom labeled 728). The convexity has been exaggerated.

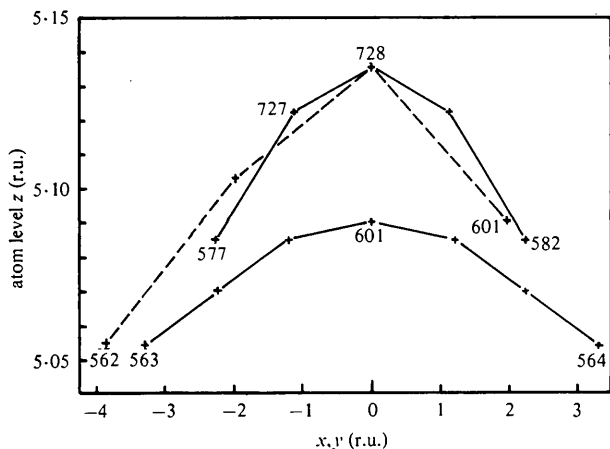


Fig. 6. Coordinates of several atoms situated on the face presented in Fig. 5. Two rows are parallel to the  $y$  axis (—), one to the  $x$  axis (---).

We should now examine the manner in which the atoms of a given layer approach the center  $O$  as the number of layers in the model increases. Let us consider a face belonging to the layer  $n = 3$ . In Fig. 7, we have given the coordinate of several atoms belonging to this face, four atoms from an edge and the three atoms of the parallel row, in the various models studied. In the three-layer model ( $N = 147$ ) these atoms are on the surface. One notices that as layers are added the face and thus the whole layer tends to approach the center. In Table 1 we have collected results concerning two of the atoms of the layer  $n = 3$  as extracted from Fig. 7: on the one hand the decrease  $dz/z$  of the level of atoms situated at the center of the face and at the vertex, when one adds successively one, two, three and then six layers, and on the other hand the difference  $\delta z$  between these levels in the different models.

We see that the addition of the first layers causes a stronger decrease in  $dz/z$  than the addition of those following. This shows the effect of the positive

Table 1. Compression of central and vertex atom in the  $n = 3$  layer according to the number of supported layers

Total number of layers	3	4	5	6	9
Number of supported layers	0	1	2	3	6
$100 \times \frac{dz}{z}$ (center of face)	—	-1.17	-0.75	-0.55	-1.16
$100 \times \frac{dz}{z}$ (vertex)	—	-1.51	-0.82	-0.58	-1.18
$\delta z = z_{\text{center}} - z_{\text{vertex}}$	0.037 <sub>3</sub>	0.045	0.048	0.048	0.048

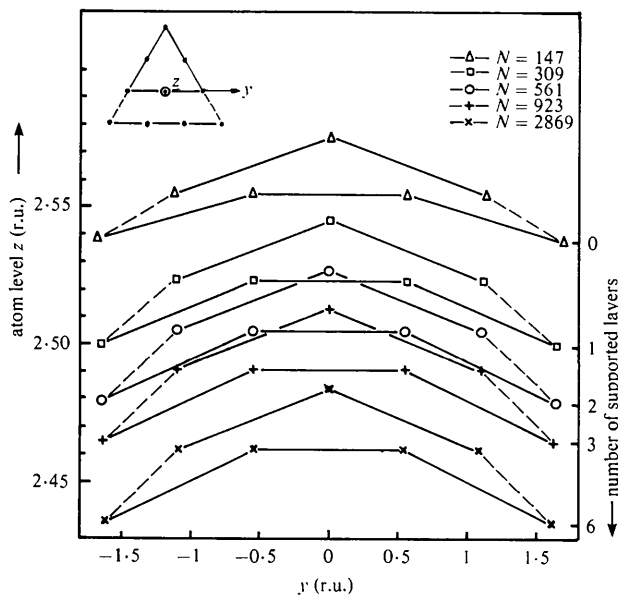


Fig. 7. Coordinates of seven atoms situated on a face belonging to the layer  $n = 3$ , as the number of supported layers is increased. Both rows are parallel to the  $y$  axis.

Table 2. Distances between the vertices of layers  $n - 1$  and  $n$  in the nine-layer icosahedron

Layers $n-n-1$	1-0	2-1	3-2	4-3	5-4	6-5	7-6	8-7	9-8
$d_n$ (r.u.)	1.0023	1.0232	1.0395	1.0513	1.0605	1.0684	1.0768	1.0885	1.1185
$100 \times \frac{d_n - d_{n-1}}{d_{n-1}}$		2.1	1.6	1.1	0.9	0.8	0.8	1.1	2.8

relaxation as it affects atoms when they belong to the surface layer, or, to a lesser extent, to the immediate subsurface layer. In both cases one notices that the level of the atom at a vertex experiences a more pronounced decrease than that of the atom at the center. Thus it comes about that the spread  $\delta z$  between these levels grows as one adds the two first layers and remains constant as the successive layers are added. One can therefore say that a face belonging to a layer of given index conserves the same convexity in all icosahedral models, once it is covered with three or more layers.

Let us now examine the variation of the difference in level  $\delta z$  in the different layers of the same icosahedron. Fig. 8 illustrates the case of icosahedra of six and nine layers. In the layers of index  $n$  where there is no atom at the center of the face ( $n \neq 3, 6, 9$ ), we have taken the level of the nearest atom to the center, which leads to an under-evaluation of  $\delta z$ . It is clear that  $\delta z$  grows linearly with  $n$  in the deeper layers of the icosahedra, a fact which demonstrates that the layers encase each other regularly.

Finally, we may remark that in accordance with the results described in the previous paragraph, the positive relaxation of the surface, affecting the vertices relatively more than the centers of faces, tends to diminish

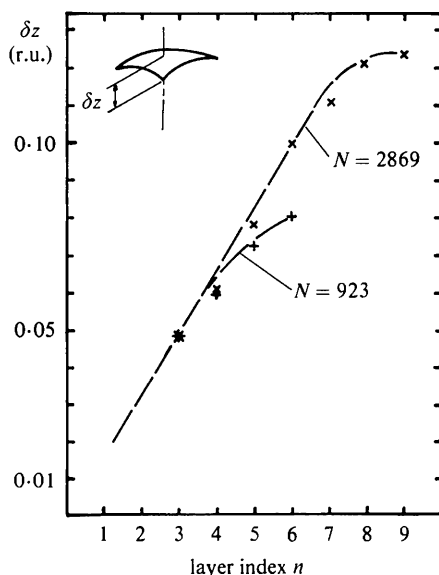


Fig. 8. Difference  $\delta z$  between the level of atoms situated at the center of a face and at a vertex vs layer index  $n$ . Results are given for the six-layer ( $N = 923$ ) and the nine-layer ( $N = 2869$ ) icosahedra.

$\delta z$ , and that the  $\delta z$  values corresponding to internal layers  $n = 3$  and  $n = 4$  are identical in the two icosahedra.

## 2. Radial distances

In each of the relaxed models, we calculated the distances between nearest-neighbor atoms on the fivefold symmetry axis, *i.e.* between the vertices of successive layers.

Table 2 gives the distances  $d_n$  between the vertices of layers  $n - 1$  and  $n$ , as well as the relative growth increments  $(d_n - d_{n-1})/d_{n-1}$ , in the case of the icosahedron of nine layers,  $N = 2869$ .

The decrease of  $d_n$  confirms that the effective atomic density grows as one approaches the center of the model. The relative growth varies as one moves along a radial axis. It increases in the passage through layers  $n = 7$  and  $n = 8$ , which reflects the positive relaxation of the vertex atoms situated on layer  $n = 8$  and on the surface layer  $n = 9$ . Above all, we notice that the relative growth increases as one moves away from the surface. Thus the effective atomic density grows more and more rapidly as one approaches the center of the model.

This radial compression is to be seen in all the models as is evident in Fig. 9, which gives the distances  $d_n$  in the various icosahedra. As abscissa, we have drawn the quantity  $N^{-1/3}$  which varies inversely as the mean diameter of the models, *i.e.* as the surface/volume ratio. One measure of the importance of this compression is in the observation that, once one moves away from the surface of the models, the distances  $d_n$  become less than 1.0917 r.u., this being the value between nearest-neighbor atoms calculated for the bulk LJ crystal (Lennard-Jones & Ingham, 1925). As the number of layers increases, so does the maximum distance  $d_n$ . This increase is a function linear in  $N^{-1/3}$  from the three-layer icosahedron and beyond (dotted line in Fig. 9), signifying that the compression in the surface layer varies inversely with the diameter of the surface layer. Lastly, we see that the distances between layers of the same index  $n$  decrease regularly as we increase the number of layers (continuous line in Fig. 9). In effect, each new external layer introduces an additional radial compression which is transmitted throughout the model. The growth in compression upon each layer depends, to first order, on its effective surface, a fact which implies the regular decrease in  $d_n$  observed.

### 3. Tangential distances

In each layer the tangential distances  $d_t$  between nearest-neighbor atoms were calculated in the neighborhood of the center of gravity of the faces. However, the configurations at the center of the faces are not strictly speaking identical, whatever the index  $n$  of the layer. When the center of gravity is occupied by an atom,  $d_t$  is the distance which separates this atom from one of its six nearest neighbors; when there is no atom at the center of gravity,  $d_t$  is the side of the triangle formed by the three central atoms. These distances are given in Fig. 10. We see that the distances  $d_t$  decrease whether, in a layer of a given index  $n$ , a supplementary layer is added, or, in the different layers of a model made up of  $N$  atoms, one is going towards the center of the model. Just as for the distances  $d_n$ , the distances  $d_t$  decrease at the same time as the compression of the layers increases.

In the surface layer (dotted line in Fig. 10) the distance  $d_t$  varies very little with the number of layers of the model. We may also note that  $d_t$  is slightly larger

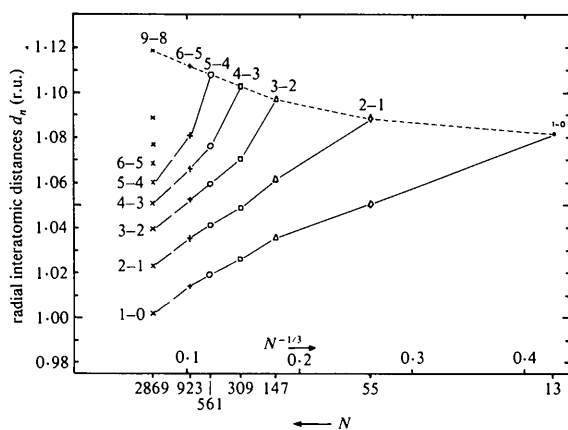


Fig. 9. Distances  $d_n$  between atoms situated on a five-fold axis, at vertices belonging to layers  $n$  and  $n-1$  ( $n=0$  refers to the central atom). Results are given for seven relaxed icosahedra. The quantity  $N^{-1/3}$  varies inversely as the mean diameter of the models.

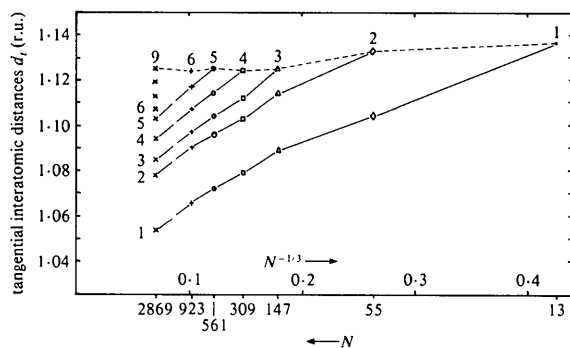


Fig. 10. Distances  $d_t$  between nearest-neighbor atoms situated in the neighborhood of the center of gravity of faces belonging to successive layers  $n$ .

than 1.125 r.u., the distance corresponding to the minimum LJ pair potential. This indicates that at the center of the faces which, as we have seen, are convex and where the interatomic distances are maximized, nearest-neighbor atoms are in a state of attraction.

Finally, we may note, in Fig. 10, that the distance  $d_t$  is distinctly smaller in the layer  $n=1$  than in the others, whatever the model. In this layer the faces comprise only three atoms and the distance  $d_t$  is that which separates two vertices. In this case we have therefore directly that  $d_t \sim 1.05 d_1$ ,  $d_1$  being the smallest radial distance  $d_n$  as seen in Fig. 9.

### V. Discussion

First of all, it seems clear from the preceding results that it is not necessary to make further calculations on larger Mackay icosahedra. In effect, we have seen in the relaxed configurations the regular compression for each layer as each further layer is added. With the potential used, there is nothing to prevent the building up of compression from layer to layer and one may predict the characteristics of models of more than nine layers without the necessity of long computer calculations, by extrapolating the diagrams, Figs. 9 and 10.

On the other hand, we may note that the macroscopic icosahedral model (§ II) gives a qualitative account of the results obtained with microscopic models, if one applies the special condition where each prismatic element is reduced to a flat section of type (111). In particular, the quantitative results strongly confirm the predicted compression as one approaches the center of a relaxed model: at the center of the nine-layer icosahedron, the radial interatomic distances are some 10% smaller than that in the bulk LJ crystal. This deformation is then much larger than the 1% normal displacement observed for the (111) surface of a relaxed LJ crystal (Allen & de Wette, 1969). Therefore, we may assert that the effects reported here originate in the icosahedral geometry and are not very sensitive to the pair potential used in the calculations.

Furthermore, rare-gas clusters with pentagonal symmetry are correctly described by the icosahedral model presented here, as expected by using a LJ potential. This model, with normal compression and regular change in interatomic distances is completely different from that made of 20 tetrahedral units with rhombohedral structure, which is used to describe gold particles (Yang, 1979). Then it would suggest that in metallic particles, the distortions which correspond to the 5% tangential extension and make one crystalline structure (f.c.c.) turn into another one (rhombohedral) provoke a radial compression possibly smaller than in rare-gas particles and in any case experimentally not detected.

Finally, whatever the number of atoms considered in the icosahedral model, the deformation caused by

relaxation is of appreciable importance to the point where one may expect a modification of certain physical quantities. It is thus indispensable to calculate such quantities, using relaxed model configurations rather than unrelaxed ones.

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## The SIR Program.

### II. The Evaluation and Use of Two-Phase Seminvariants

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#### Abstract

New faster and more accurate procedures for the evaluation of two-phase seminvariants by means of the representation theory are described. Some special relationships are also considered and specific methods for their estimation are proposed. The use of the generalized first representation is discussed and detailed examples are given. These ideas for the evaluation of two-phase seminvariants have been introduced in the SIR program and examples of actual applications are given.

#### 1. Introduction\*

Two-phase s.s.'s are those linear combinations of two phases,

$$\Phi = \varphi_u + \varphi_v, \quad (1)$$

\* Symbols and abbreviations are defined in the Appendix.

whose indices satisfy the condition  $\mathbf{h}_u + \mathbf{h}_v \equiv 0 \pmod{\omega_s}$  where  $\mathbf{h}_u$  and  $\mathbf{h}_v$  are the vectors seminvariantly associated with  $\mathbf{u}$  and  $\mathbf{v}$ , and  $\omega_s$  is the seminvariant modulus of the given space group.

The first representation (Giacovazzo, 1977) of a two-phase seminvariant of first rank is given by the set of quartet invariants of the form

$$\varphi_v + \varphi_{uR_p} + \varphi_{h_1R_p} - \varphi_{h_2R_p}, \quad (2a)$$

$$\varphi_v + \varphi_{uR_q} + \varphi_{h_1R_q} - \varphi_{h_2R_q}, \quad (2b)$$

where  $\mathbf{h}_1$  and  $\mathbf{h}_2$  are appropriate vectors which can vary over lines or planes of the reciprocal lattice; we will say that they belong to the sets  $\{\mathbf{h}_1\}$  and  $\{\mathbf{h}_2\}$ . The collection of diffraction magnitudes which are basis or cross terms of the quartets (2) constitutes the first phasing shell  $\{B\}_1$  of  $\Phi$ .

Two kinds of conditional probability distributions for  $\Phi$ , given all the magnitudes in  $\{B\}_1$ , were obtained by