THREE-DIMENSIONAL PACKING OF PERFECT TETRAHEDRA

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Abstract. We represent a novel geometric construction in 3D space—a *saturated polytetrahedron*, which is an infinite cluster of perfect face-adjacent tetrahedra where there are no faces to add further tetrahedra. A perfect tetrahedron is represented by a configuration of four identical balls. Therefore our polytetrahedron is a specific packing of hard spheres consisted of tetrahedral configurations, where all accessible pits between three spheres on faces of tetrahedra are occupied by spheres. We created these packings inside large spherical volumes of different radii (up to 60 diameters of hard spheres) and examined their structure. The saturated polytetrahedron is isotropic and homogeneous. Its packing fraction (fraction of volume of hard spheres) tends to 0.435 with growth of size. There is a pronounced "icosahedral short range order", however the long distance correlation between hard spheres is absent in our polytetrahedron. These features have an analogy with the structure of dense non-crystalline packings (simple liquids and glasses). "Polytetrahedral" nature of disordered packings of hard spheres was also discussed recently in [1].

Introduction

It is known the perfect tetrahedra cannot cover an Euclidean space without gaps. Dihedral angle of a tetrahedron ($\sim 70.5^\circ$) is an irrational part of the full angle. In a result, the face-adjacent tetrahedra cannot produce a close chain and any structural unit available for translation, Fig. 1. This geometrical feature of a tetrahedron results in important consequences for physics. It is a reason of the fact that simple liquids (like liquid metals), glasses, and dense disordered packings of spherical particles are relatively stable and widespread in spite of the

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crystalline structures are more preferable from point of view of the thermodynamics.

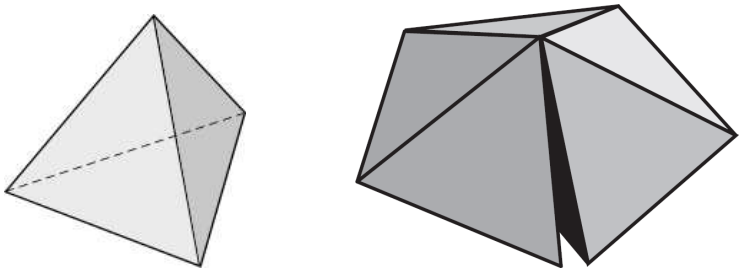


FIGURE 1. Perfect tetrahedra cannot cover space without gaps. Dihedral angle of the tetrahedron is an irrational part of the full angle ($\sim 70.5°$). "A ring" of five perfect tetrahedra around the common edge has a gap $\sim 7.5°$.

Perfect tetrahedral configuration of four identical balls is the densest local packing. Its packing fraction is ~ 0.7797 what is greater than the maximal packing fraction in 3D realizing in the densest crystalline packings (~ 0.7405), which contain also octahedral configurations with lower local density [2, 3]. Physicists know that atoms in matter tend to be arranged densely and, on the other hand, to fill space homogeneously as soon as possible. In the three dimensional space, however, there is a contradiction between these tendencies: the densest local configuration is a tetrahedron, but they cannot cover the space without gaps. The situation is different in the two-dimensional space where the densest local configuration of disks (regular triangle) is an element filling the plane completely. This means that the disordered phase is unstable in 2D because any cluster of regular triangles of disks is a fragment of the densest crystal structure on the plane.

Atoms in 3D can arise easily in tetrahedral configurations because they are preferable local structures. They form clusters with high local density, but the morphology of such aggregates is incompatible for lattices. In a result, the dense non-crystalline phase consists of regions with high local density (polytetrahedra) which are interlacing with the intervening medium of lower density [1, 4].

In spite of the polytetrahedral clusters play an important role in the structure of non-crystalline systems, their geometrical properties are little understood. What is well-known, the tetrahedra can form helices. The simplest one is Boerdijk helix [2] (in physics it is also known as "Bernal spiral"), which is a screw-axis pile of face-adjacent tetrahedra, Fig. 2(a). A numerous amount of other helices of different

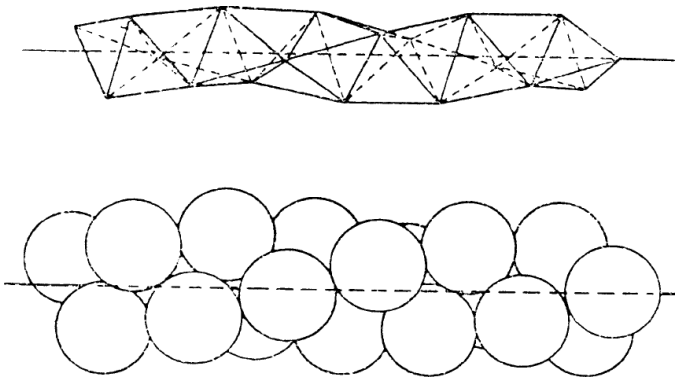
radii and pitches can be also constructed [5], Fig. 2(b). Different helix structures of tetrahedra were also found experimentally in crystal alloys of some metals. Fig. 2(c) demonstrates one of them in the gamma-brass structure [6]. However, tetrahedra can generate not only helices but also branched clusters. Indeed, a new chain of tetrahedra can be started from any open tetrahedral face on the surface of a helix.

For us, of interest are clusters growing extensively, i.e. which can be considered as a "bulk" system. Models of such structures can be easily constructed. Indeed, let us start from a perfect tetrahedral configuration of four balls, Fig. 3. To put a new ball in the pit between three balls on a face of the tetrahedron, we get an aggregate of two face-adjacent tetrahedra—triangular bipyramid. Then the next ball can be added to any face of this bipyramid to get a cluster of three tetrahedra. If we continue this procedure to add new balls to the accessible faces, we will get larger and more complex aggregates. Morphology of large clusters can be very different. Fig. 4 demonstrates three different realizations of a cluster of four tetrahedra. Variant 4(a) is a fragment of the Boerdijk helix. Cluster 4(b) corresponds to the ring in Fig. 1. It consists of four perfect tetrahedral configurations and the fifth one slightly distorted (one edge is greater then others on 8.8%). A gap can arise between the meeting faces where a new ball cannot be added. Cluster 4(c) looks like a star of tetrahedra. Clusters of five tetrahedra demonstrate a greater diversity. One can find, they have 6 different topological types, and some of them have a mirror twin. Obviously, the amount of different types grows drastically with size of clusters and becomes innumerable very soon.

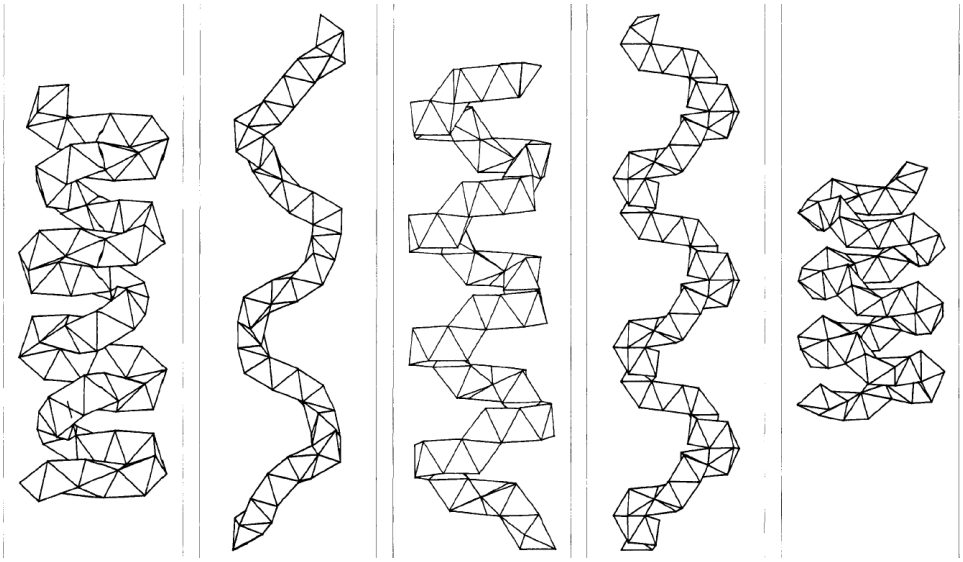
A polytetrahedral cluster can be as large as desired. At that, it may ramify and grow in any direction. To reduce a free space between branches, we should add balls to all accessible faces of the cluster. We will call such a cluster as *saturated polytetrahedron*. In this work, we generate such an object and study its structure.

1. Construction of the saturated polyhedron

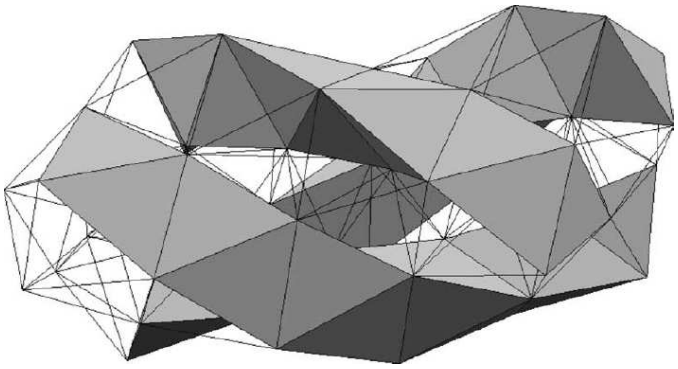
An initial tetrahedral configuration of four balls is placed inside a large sphere of radius R. Using the coordinates of the centers of three balls on a face of the tetrahedron, one can calculate the position of a new ball touching these three balls. If this new ball does not overlap any existing ball, then it can be added to the cluster. This process to add new balls continues. At the calculation, we control faces of the tetrahedra. If we added a ball to the given face, the face is considered as "closed". It is a common face for adjacent tetrahedra. If we



(a) Boerdijk helix, the picture taken from [2]



(b) Some helices from [5]



(c) Clusters obtained in gamma-brass structure [6]

FIGURE 2. Infinite clusters of perfect tetrahedra.

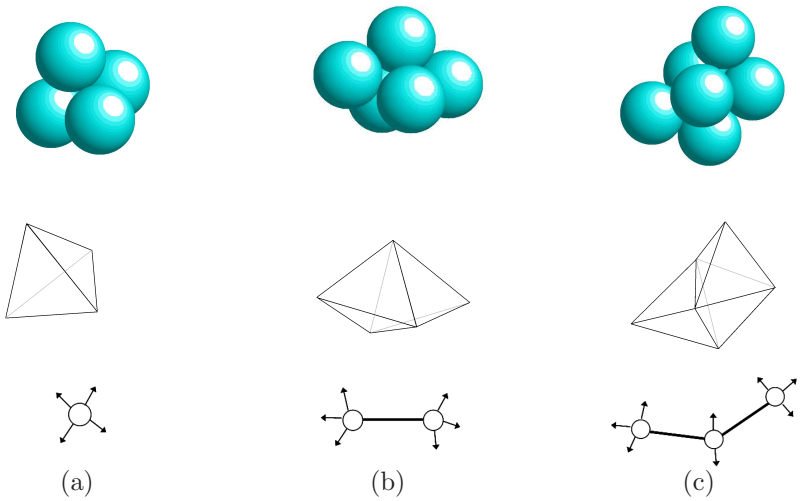


FIGURE 3. Clusters of tetrahedra. Perfect tetrahedron is the densest local configuration of four identical balls (a). Putting a new ball in a hole between three balls means the appearing of a face-adjacent tetrahedra. Cluster of two tetrahedra, trigonal bipyramid (b). Cluster of three tetrahedra (c). At the bottom: the topology of clusters is shown. Circles mean the centers of tetrahedra, segments show adjacency through the face, arrows mark "open" faces (a new tetrahedron (ball) can be added).

cannot add a ball to the given face (because of the overlapping of a new ball with an old one), we call it as "inaccessible". These faces form gaps between tetrahedra. At last, those faces where we can add a new ball are "open". For example, an initial tetrahedron has four open faces, Fig. 3(a). Cluster of four tetrahedra can have ten or eight open faces, see Fig. 4, clusters 4(a) and 4(b). The saturated polytetrahedron is created just after all open faces inside sphere R be exhausting. Faces which get out the surface of sphere R are not considered in our analysis.

We calculated different kinds of saturated polytetrahedra with the initial tetrahedron both at the center of the sphere R or away from the center. Also we calculated models starting from many separated tetrahedra distributed inside the sphere. In our calculations, we pick out open faces (to add a new ball) randomly. The largest model was constructed in sphere $R = 60$. It contains more then 750000 balls.

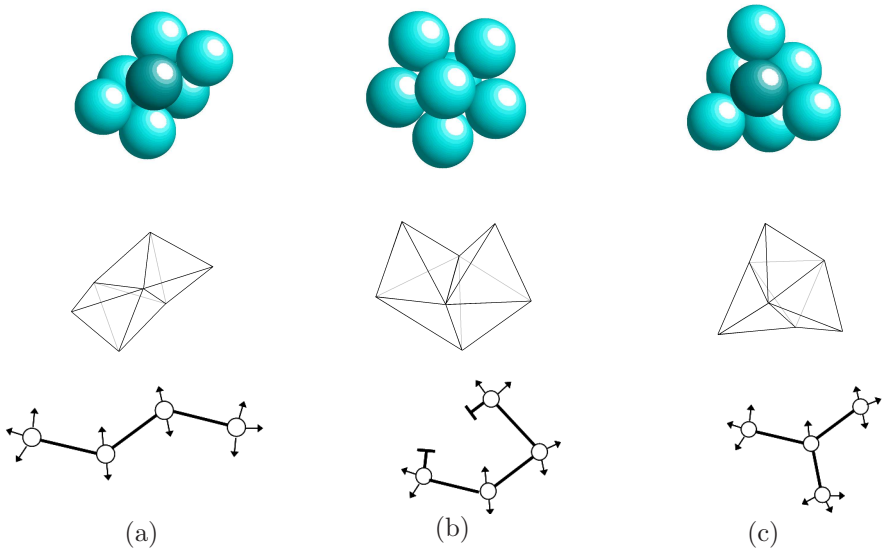


FIGURE 4. Clusters of four tetrahedra. See caption in Fig. 3. Obtuse arrows mark inaccessible faces (impossible to add a new tetrahedron (ball)).

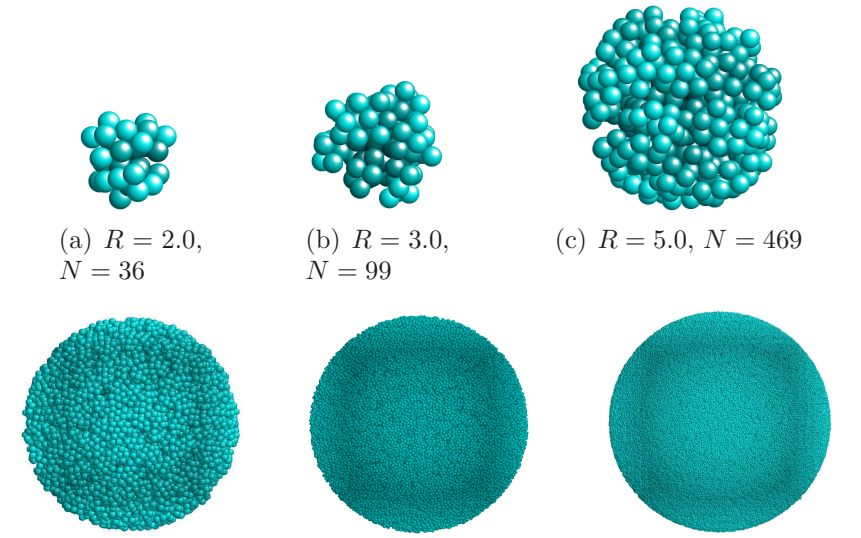
Fig. 5 demonstrates some models of a saturated polytetrahedron in spheres of different radii.

2. A saturated system of polytetrahedra

A saturated polytetrahedron constructed from one initial tetrahedron is a mathematical object. An ensemble of polytetrahedra growing from its own initial tetrahedron could be a more physical model. We also generated such models using different amounts of initial tetrahedra randomly distributed inside sphere R. The same algorithm to fill in open faces on all growing polytetrahedra was used. Preliminary analysis demonstrates that the structure of such "multi-polytetrahedral" systems is very similar to that of a single polytetrahedron. This means that the branches of different polytetrahedra interlace like branches on a single polytetrahedron. A model for the sphere of $R = 55$ shown in Fig. 5 has been made starting from 1000 initial tetrahedra.

3. Density of the saturated polytetrahedron

Large saturated polytetrahedron constructed according our algorithm is an isotropic and homogeneous system. At that it contains remarkable fluctuations of the density on the microlevel. Relatively



(a) $R = 2.0$, $N = 36$ (b) $R = 3.0$, $N = 99$ (c) $R = 5.0$, $N = 469$

(d) $R = 10$, $N = 3605$ (e) $R = 30$, $N = 95142$ (f) $R = 55$, $N = 576444$

FIGURE 5. Examples of saturated polytetrahedra inside a sphere of radius R. Diameter of hard balls is equal to 1. Value N means the total number of balls in a model. Initial tetrahedron had been located at the center of the sphere except the model of $R = 55$, where 1000 initial tetrahedra randomly distributed inside the sphere were used.

dense packing arises around the initial tetrahedron. As a rule, there are balls with 12 touching neighbors in this region, and the density of such local configurations is very high. Similar situations may arise also in different places of the polytetrahedron, however, the initial tetrahedron is preferred in this respect. It is a "root" from which the branches of the polytetrahedron start to grow. Loose regions are located between branches of the cluster. Fig. 6 demonstrates the behavior of the saturated polytetrahedron density as a function of the distance from the center of models. Packing fraction (fraction of volume occupied by balls) was calculated for sequential spherical layers. For a given layer, we take into account balls whose centers belong to the layer, calculate their volume, and divide it by the volume of the layer. Inaccuracy of this method is small and diminishes with increase in the radius of the layer. As it was be expected, the first layers are denser. However, the density tends to an asymptotic value 0.435 with growth of the radius of the model. As one can see, the total density of the saturated polytetrahedron is rather low. This value is less than the

density of the "loose random packing" (0.555) known in physics [7]. A real packing implies a "steady position" of all balls, what needs at least four touching neighbors for every ball. Our system is not a physical packing, some balls have only three neighbors (at ends of the branches).

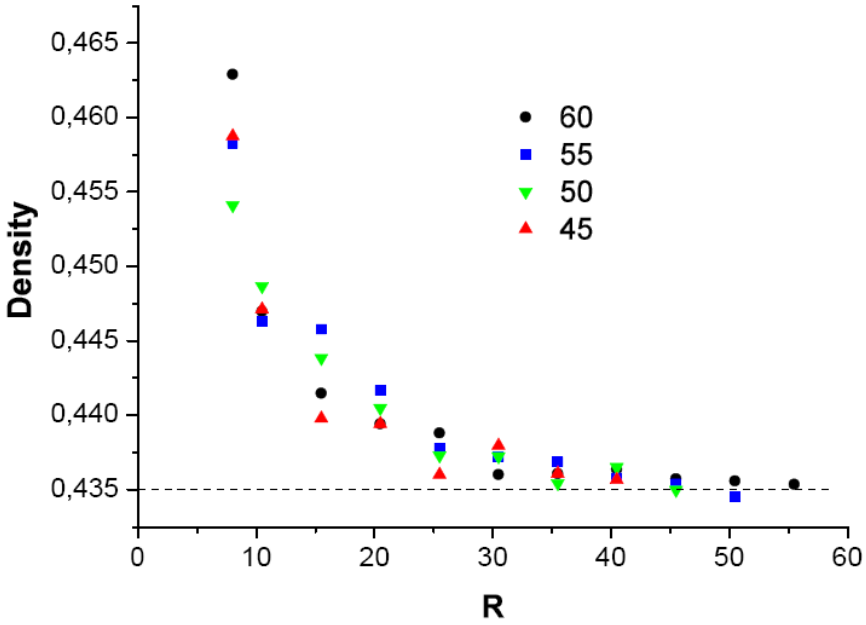


FIGURE 6. Packing fraction of the saturated polytetrahedron as a function of the distance from the center (calculated separately for spherical layers inside the models). Initial tetrahedron is located at the center of the models. Width of the layers is 5 diameters of balls. Different symbols correspond to models generated inside spheres of different radii R.

The spread of points in Fig. 6 illustrates the difference between independent realizations of the polytetrahedra. As was mentioned above, the structure depends on the succession to add new balls to the open faces. We used a random choice. Relatively large dispersion of points at smaller radii arises because of a smaller amount of balls in the nearby layers than that in the far layers. The fact that all our models tend to the same asymptotic density means that they are statistically equivalent and the saturated polytetrahedron is homogeneous in the limit.

Large voids are absent in the saturated polytetrahedron. It seems naturally because our algorithm is directed on the filling of holes. There is only a small fraction of voids where the diameter of an inscribed empty sphere is greater than the diameter of balls (an extra ball can be placed in such a void). However, there are no voids at all with the diameter of an inscribed sphere equal to or greater than 2. The cavities are located at places of "contact" of a few branches of the polytetrahedron. The faces of tetrahedra encompassing the void are oriented in such a manner that it is impossible to add a new ball, in spite of there is enough room: the inscribed sphere has diameter greater than 1. This analysis was made with help of Delaunay simplexes, which are a very efficient instrument for analysis of interatomic voids [8, 9].

4. Radial distribution function

Fig. 7 demonstrates the radial distribution function $D(r)$ for the saturated polytetrahedron. We recall that this function is a histogram of pair distances between balls and related to the pair correlation function $g(r)$ as $g(r) - 1 = D(r)/4\pi\rho r^2$, where ρ is the average density of the system.

Function $D(r)$ tends to the $4\pi\rho r^2$ behavior with increase in r for homogeneous systems in 3D. The saturated polytetrahedron also has this property. This fact confirms that we deal with a homogeneous (non-fractal) system. Indeed, one can think that our models can be fractals because of a cluster of perfect face-adjacent tetrahedra is a tree (from viewpoint of graph theory). The saturated polytetrahedron is homogeneous because of the requirement of the nonoverlapping of balls. This condition restricts the growth of many branches of the tree.

There is a structural "duality" of the saturated polytetrahedron. On the one hand, we see distinct short distances resulted by a finite set of local configurations of tetrahedra. On the other hand, the long-distance order is absent. This fact is not trivial and needs detailed specification. Long-distance correlations are usually related to a possibility to "predict" the position of atoms far from the given central atom. A lattice (system with a translation symmetry) is the known example of it. Having a unit cell and a rule for translation, one can pass on to any atom of the lattice. In the case of polytetrahedron, we have similar situation. Tetrahedral configuration is a unit cell, and the algorithm used for the "translation" over space is also well-defined (reflection on faces of tetrahedra). Thus, there is a possibility (at least theoretical)

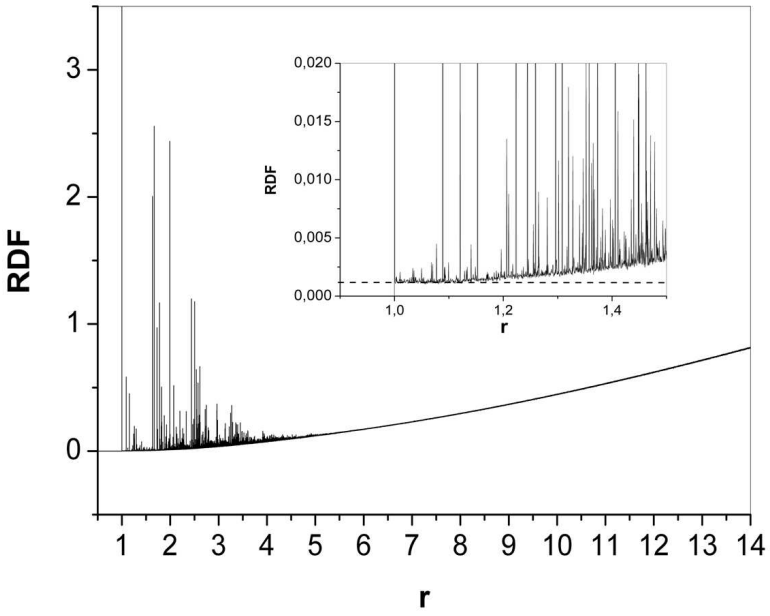


FIGURE 7. Radial distribution function of the saturated polytetrahedron. There is a well-defined "short-range order" and absence of correlation for large distances. In the insert: the same function on the expanded scale. Dashed line shows the presence of a small "base" of a continuous spectrum of distances.

to reconstruct the location of every ball against the central one. From this point of view, a "long-distance order" is an inherent property of the polytetrahedron. Therefore, a fact of the absence of long-distance correlation in our models is not trivial. A reason for this can be in the algorithm of the random filling of open faces. It must not be ruled out that in a case of another scenario, the long-distance order can appear in a saturated polytetrahedron.

The next non-trivial property of the saturated polytetrahedron is the presence of a "continuum" of distances. It is visible as a "base" on the radial distribution function, Fig. 7 (insert). Besides a discrete set of short distances, which is related to the finite number of possible local aggregations of tetrahedra, some balls can be located practically at any distance from each other. It is possible if they belong to different branches and the branches are long enough.

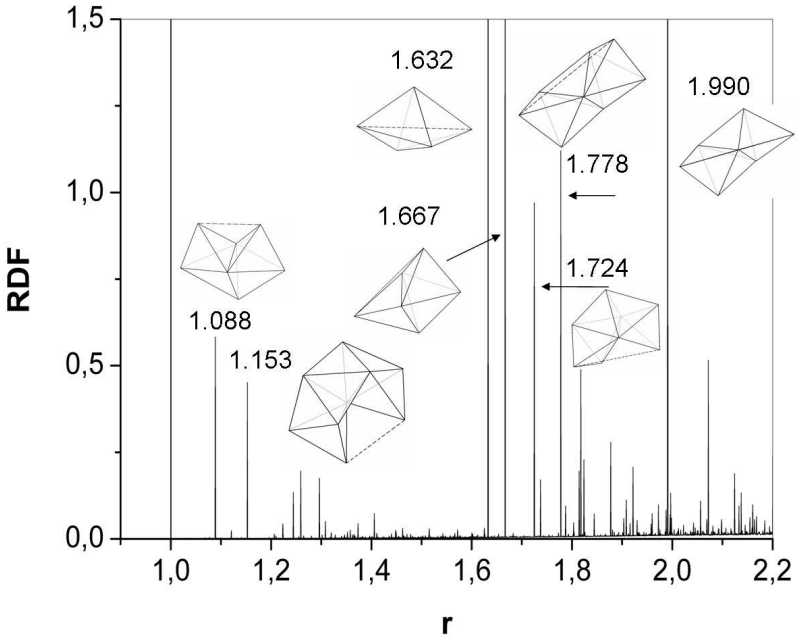


FIGURE 8. Structural interpretation of main peaks of the radial distribution function. Dashed lines on the clusters connect the centers of corresponding balls.

5. "ICOSAHEDRAL" SHORT-RANGE ORDER

It is not difficult to find structural units corresponding to the main peaks on the radial distribution function, Fig. 8. Obviously they are determined by the simplest clusters of tetrahedra. The most pronounced peaks at 1.632 and 1.667 belong to clusters of two and three tetrahedra; and the distance 1.990 is found in a lineal cluster of four tetrahedra. The distances 1.724 and 1.778 arise in five tetrahedra. The short distance 1.088 is a signature of the above-mentioned "ring" of tetrahedra. The next distinct distance 1.153 appears in a cluster of six tetrahedra, see Fig. 8. All discussed distances can be found in this aggregate, which can be considered as a half of "icosahedron". We understand the icosahedron as an aggregate of 13 balls representing a cluster of face-adjacent tetrahedra with common vertex, Fig. 9. Thus, one can say by following the physical terminology [10, 11] that the saturated polytetrahedron have an "icosahedral short-range order". On the other hand, the same pair distances can be found also in other fragments of the polytetrahedron, and there is no reason to appeal to icosahedron.

The saturated polyhedron is an independent geometrical substance, which has its own specific structure. From our point of view, it is more correctly to say the "polytetrahedral" order instead of "icosahedral" in concern to the structure of liquids and glasses.

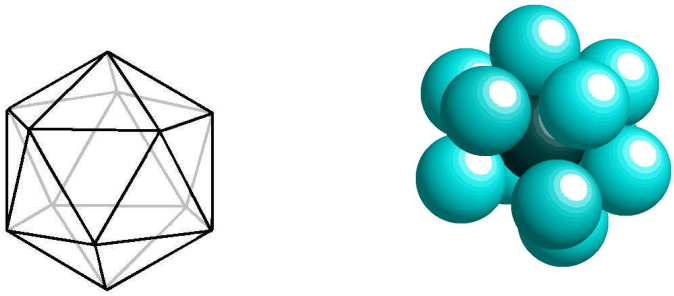


FIGURE 9. Icosahedron (left). "Icosahedral" configuration of balls (right)—a cluster of face-adjacent tetrahedra with common vertex. There are twelve neighbors in the shell around the central ball. There are gaps between some balls in the shell.

CONCLUSION

We have constructed a "packing" of identical hard spheres on the base of perfect face-adjacent tetrahedral configurations. During the construction, all open faces of the tetrahedra are closed by new adjacent tetrahedra. In a result, we obtain a "saturated polytetrahedron" where there are no faces of the tetrahedra to add new balls. The models were constructed inside large spheres; the largest one contains more then 750000 balls. It fills space homogeneously, and the average density (packing fraction) tends to 0.435 with growing size of the model. The structure of the saturated polytetrahedron demonstrates the features typical of dense disordered systems: there are "a short range order" and no long distance correlation. At that, the main peaks on the pair correlation function correspond to distances in the "icosahedral" aggregate of perfect tetrahedra. The saturated polytetrahedron is a new geometrical construction and needs further investigations.

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