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CURVED SPACE MODEL FOR AMORPHOUS STRUCTURES AND ITS RELATION WITH
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Résumé - Le modèle d'espace courbe permet une description systématique de l'ordre dans les structures amorphes. Nous présentons ici un calcul analytique des fonctions d'interférence des modèles parfaits.

Abstract - The curved space model provides a new systematic approach of the order in amorphous structures. We present here some analytical calculations of the interference function for ideal models.

I - INTRODUCTION

The structure of condensed matter results from the competition between local interactions, topologic and geometric rules imposed by the space filling requirement. It is thus an exception when there is no discrepancy between the two types of rules. In other cases, the structure results from a compromise. To analyze this problem, as a first step, we do not impose to Euclidean space. So we let the space in which the structure is growing, be defined by a propagation, from point to point, of local geometrical properties. One of these fundamental properties is the curvature of the space. If we consider a structure in which there is no change in the local interaction (no change in the chemical composition), the curvature must be constant and consequently the underlying space can be spherical, hyperbolic or eventually euclidean. In the first part of this paper, the curved space approach is detailed using the example of the tetrahedral packing which can be a model for pure amorphous metals. Metallic atoms are considered to interact with a spherical potential and consequently to pack together like spheres. Locally, spheres have their centres on tetrahedron vertices. But on a large scale, there are some difficulties, due to the impossibility to fill Euclidean space with regular tetrahedra. Only if the space is given a curvature, can it then be tiled by regular tetrahedra. The complete description of the structure is achieved if a network of defects can be introduced in order to cancel the curvature. In the second part, we present some analytical calculations of interference and radial distribution functions of the ideal models described in curved space. Indeed it is possible to define an equivalent to the $S(q)$ function characterizing amorphous structures. This has already been presented by D. Nelson and M. Widom, using hyperspherical harmonics defined in the 4D space (1). Our description gives similar results but is obtained with a simpler formalism comparable to Debye analysis.

II - SPHERE PACKING AND THE (3.3.5) POLYTOPE

If one tries to pack spheres in a dense way by a discrete aggregation process, one easily finds that the regular tetrahedron (where a sphere is placed at the tetrahedron vertices) is the best solution with $N = 4$ spheres. 5 tetrahedra can share a common edge, but a void necessarily remains between two triangular faces (fig. 1b). This is due to the fact that the tetrahedron dihedral angle ($\sim 70^\circ$) is not a submultiple of 2π . This misfit angle manifests itself when one tries to propagate the tetrahedral local configuration and completely surrounds a given vertex. An imperfect icosahedron is then obtained (fig. 1c). Note that amorphous metal structure is well described by the so-called pseudo icosahedral ("compact or polytetrahedral") models. It is desirable to define an ideal model in which the space can be perfectly tiled

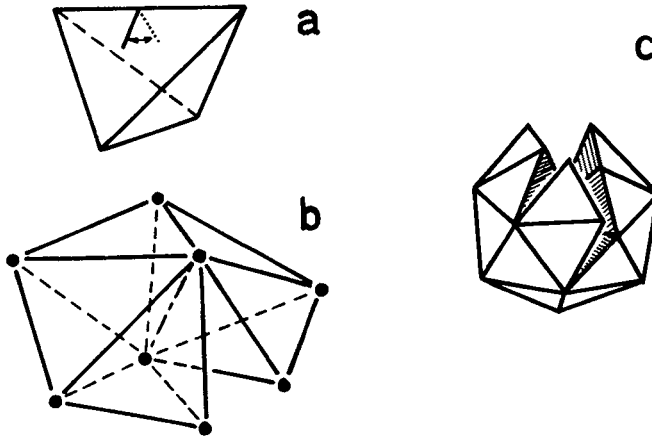


Fig. 1. : The dihedral angle of a tetrahedron (a) is not a submultiple of 2π . Five tetrahedra with a common edge leave a small unfilled space (b). An imperfect icosahedron (c) with a misfit between dashed faces.

by tetrahedra. This is achieved using an S^3 spherical space. This hypersphere can be embedded in the 4D euclidean space with equation :

$$x_1^2 + x_2^2 + x_3^2 + x_4^2 = R^2$$

Note that only 3 of the 4 coordinates are independent, S^3 being a 3D (curved) manifold. In term of the tetrahedron edge length, the radius of curvature is $R = (1 + \sqrt{5})/2$. The perfect tetrahedral packing on S^3 is called a "polytope" (the analogue of a polyhedron in a higher dimension). This polytope is a finite structure (S^3 is finite) and contains 120 vertices. Exactly 5 tetrahedra share a common edge and each vertex has 12 neighbours in a perfect icosahedral configuration. This polytope is called $\{3,3,5\}$ using the standard Schläffli notation and is well described by Coxeter. We now proceed to present its structure as simply as possible. One can use a 2D analogy. Suppose one tries to represent, on the euclidean plane, geometrical configurations belonging to the surface of a sphere. The simplest way to do it, is to generate orthogonal mapping. If the plane is tangent to the north pole, the set of parallels (in the geographical sense) is mapped into a bundle of concentric circles. As long as the mapped region remains small (in any case restricted to the northern hemisphere), the configuration on the plane is a rather faithful image of the geometry on the sphere. In the case of a hypersphere S^3 orthogonally mapped on a tangent hyperplane at the "north" pole, one gets a bundle of concentric spheres centered at the pole. So if the polytope is oriented in such a way that one vertex coincides with the pole, the set of successive coordination shells are recovered after the mapping. (2).

III - THE $\{3,3,5\}$ RADIAL DISTRIBUTION FUNCTION

We have to determine the number of $\{3,3,5\}$ vertices at the distance (arc length) r or a given vertex. The description of the $\{3,3,5\}$ polytope shell by shell (fig. 2) surrounding the pole leads to this function $N(r)$. This function is a set of delta functions, as the polytope is a perfect regular structure. The table I shows these numbers depending on the angular distance ψ . ($r = R \cdot \psi$ where R is the 3. sphere radius).

Fig. 2. : Representation in lower dimension of the different shells in the 3D space. In 3D space, the successive shells are :

- an icosahedron (coordination polyhedron)
- a dodecahedron (2nd neighbours of the vertex on the pole)
- an icosahedron, but two vertices of this icosahedron are not first neighbour
- an icosidodecahedron in equatorial position on the 3D sphere

The same polyhedra occur in reverse order in the opposite hemi (hyper) sphere.

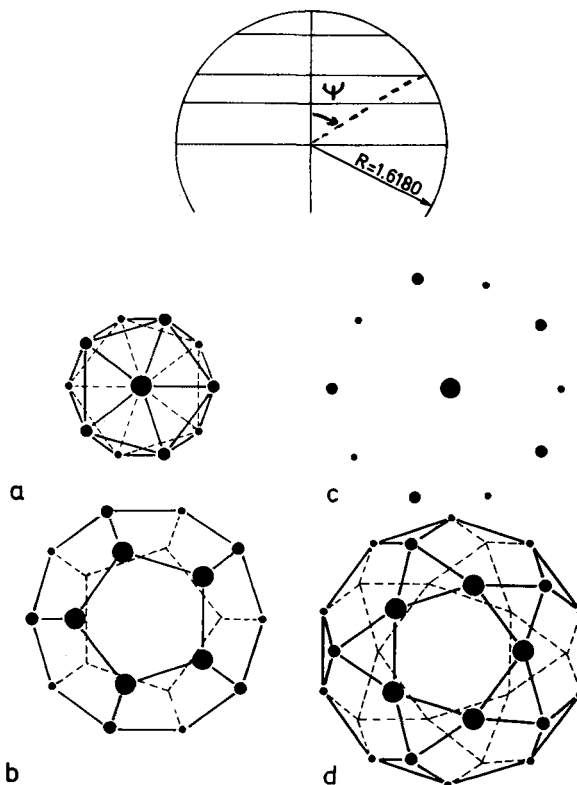


TABLE I

$\psi_i =$	0	$\pi/5$	$\pi/3$	$2\pi/5$	$\pi/2$	$3\pi/5$	$2\pi/5$	$4\pi/5$	π
N_i	1	12	20	12	30	12	20	12	1

The radial distribution function for a model obtained by mapping the {3,3,5} polytope on euclidean space corresponds to a distortion of the $N(r)$ function which can be described by a broadening of the delta functions (3).

In Euclidean space a radial distribution function $G(r)$ oscillates around a parabolic function $g_o(r) = 4\pi r^2 \rho_o$ where ρ_o is the atomic density. In spherical space, the equivalent function $g_o(\psi) = 4\pi R^2 \rho_o \sin^2 \psi$ which is the same function for small ψ , but decrease down to zero for $\psi = \pi$ (the opposite pole to the origin). The ψ variation domain is $[0, \pi]$, but it seems accurate to extend the function from $-\infty$ to $+\infty$ in order to have a periodic function. We repeat with the same periodicity the function $N(\psi)$. This function is the radial distribution of a chain of polytope glued poles to poles in a linear way. The distance is measured on geodesic lines (great circles) from on pole.

IV - THE STRUCTURE FACTOR OF POLYTOPES

The structure factor in Euclidean space is defined by

$$S(K) = \int_0^{\infty} 4\pi r^2 \rho(r) \frac{\sin(K.r)}{K.r} dr$$

it is an analysis in terms of spherical wave amplitude $\sin(K.r)/(K.r)$. In a space with positive curvature concentric spherical waves have an amplitude $\sin(\ell.\psi)/(\ell.\sin \psi)$. As the spherical space is finite, there are only a discrete set of concentric waves : the number ℓ is an integer. In order to have the same notation as in ref (1) we call this number $n + 1$. This can also be justified by comparison of the value of $\sin(K r)/K r$ for $K \rightarrow 0$ and the value of $\sin([n+1]\psi)/([n+1]\sin\psi)$ for $n = 0$ which are both equal to unity. In order to introduce a structure factor for the polytope we make the association of variables in spherical space and in Euclidean space.

$$4 \pi \sin^2 \psi \sim 4 \pi r^2 \quad (\text{we suppose } R = 1)$$

$$N(\psi) \sim G(r)$$

$$n + 1 \sim K \quad (\text{the modulus of the reciprocal space vector}). \text{ Note that Nelson et al. identify } K \text{ to } \frac{1}{\sqrt{n(n+2)}}$$

We can define the atomic density

$$\rho(r) = \frac{G(r)}{4\pi r^2} \text{ in euclidean space}$$

$$\text{and } \eta(\psi) = \frac{N(\psi)}{4\pi \sin^2 \psi} \text{ in spherical space}$$

Now the structure factor in spherical space is defined using these analogies.

$$S_n = \int_0^\pi 4\pi \sin^2 \psi \cdot \eta(\psi) \cdot \frac{\sin((n+1).\psi)}{(n+1) \cdot \sin \psi} d\psi$$

$$\text{compare to } S(K) = \int_0^r 4\pi r^2 \cdot \rho(r) \cdot \frac{\sin(K.r)}{K.r} dr$$

If we introduce a mean density η_0 (ρ_0 in euclidean space), it follows :

$$S_n = \int_0^\pi 4\pi \sin^2 \psi \cdot (\eta(\psi) - \eta_0) \cdot \frac{\sin((n+1).\psi)}{(n+1) \cdot \sin \psi} d\psi + t$$

$t = 0$ for all n , but $t = N_0$ for $n = 0$ (N_0 is the total number of vertices on the polytope).

$$S_n = \frac{4\pi \eta_0}{(n+1)} \int_0^\pi \sin \psi \cdot \left(\frac{\eta(\psi)}{\eta_0} - 1 \right) \sin(n+1) \cdot \psi d\psi$$

$$\text{compare to } S(K) = \frac{4\pi \rho_0}{K} \int_0^\infty r \left(\frac{\rho(r)}{\rho_0} - 1 \right) \sin(K.r) dr$$

S_n is related to the Fourier component of the periodic function $\sin \psi \cdot \left(\frac{\eta(\psi)}{\eta_0} - 1 \right)$.

The function $\eta(\psi)$ is a set of delta functions and consequently :

$$S_n = \frac{1}{n+1} \sum_i N_i \frac{\sin[(n+1).\psi_i]}{\sin \psi_i}$$

where N_i and ψ_i are in table I for the {3,3,5} polytope.

V - THE STRUCTURE FACTOR OF THE {3,3,5} POLYTOPE

Due to the symmetrical repartition of shells relative to the Great Sphere ($\psi = \frac{\pi}{2}$) only S_n with even n are different from zero. For $n < 60$, $S_n \neq 0$ for $n = 0, 12, 20, 24, 30, 32, 36, 40, 42, 44, 48, 50, 52, 54, 56$. For $n > 60$, $S_n \neq 0$ for all even n .

The S_n values are given by :

$$(n+1)S_n = 120 \times \text{integer} \left(\frac{n}{60} \right) + S_{n \text{ mod } 60} \text{ showing that } (n+1)S_n \text{ is the sum of a periodic function defined by } S_n = 120 \text{ for } n = 0, 12, 20, \dots, 56 \text{ and a staircase function } 120 \times \text{integer} (n/60).$$

If we suppress the scattering due to the point at origin, as is usually done in conventional non-crystalline diffraction studies a reduced structure factor $s_n = (n+1) (S_n - 1)$ can be defined. It is associated to the usual function

$i(K) = K.[I(K) - 1]$. The s_n function (fig. 3) oscillates around zero and can be compared to the amorphous metal structure factor.

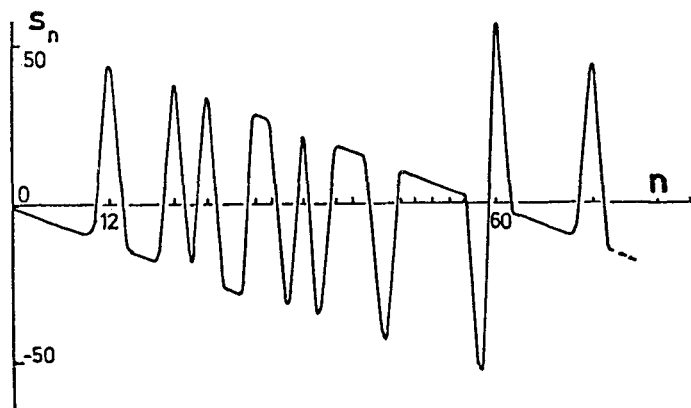


Fig. 3. : The s_n function is slightly broadened (on three points). It is a periodic function of period $n = 60$. It can be compared to currently obtained interference functions with metallic amorphous alloys.

In a first approximation it can be supposed that disclinations mix different s_n function of different internal scale. In a polytope containing a network of disclination which is obtained by the iterative inflation method (4), involving a scaling factor close to 3, the s_n function presents peaks at $n = 12, 20, 24, 30, \dots$ but also at $n = 36, 60, 72, 90, \dots$. Peaks of this last family are stronger than in the first one.

In a continuous limit the "structure factor" of a disclinated polytope can be written :

$$\sigma(n) = \int_0^{\infty} A(\alpha) S(\alpha n) d\alpha$$

The $A(\alpha)$ function is characteristic of the disclination distribution. At present, we do not know how to solve it analytically, but computer studies will give informations in order to understand this problem.

Studies of experimental results using this formalism will also be a good approach to understand disordered structures.

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