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A Landau's octagonal quasilattice

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Abstract. An Ammam-Beenker 2D octagonal quasilattice is derived from a Landau like method starting from an octagonal seed. The level of stability of this quasilattice, i.e. the level of difficulty in creating defects, is directly measured.

PACS. 61.44.Br Quasicrystals

1 Introduction

Metastable infinite clusters with icosahedral symmetry have been predicted to occur since 1982 [1] as a structure derived from the energy optimization [2] during a search for a material issued from an icosahedral seed. Soon after, in 1984, a first observation of icosahedral symmetry on a macroscopic sample of a rapidly solidified alloy AlMn [3] was done. Since the diffraction patterns of this sample were composed of nearly delta function peaks with non-periodical symmetry, the name quasicrystal was coined to such materials. Since that time numerous examples of other quasicrystals, i.e. materials with diffraction patterns composed of nearly delta function peaks with non periodical symmetry, have been observed. And among them, quasicrystals with 2D octagonal symmetry and periodicity in the transverse direction [4,5] were also observed. Thus the goal of the present paper is to find an octagonal structure by the optimization method used before for icosahedral symmetry [1,2].

Of course, observations of quasicrystals have always been closely linked with theoretical considerations. Different theoretical methods are available to build quasicrystalline structures. Firstly, geometrical methods of packing and covering [7] define quasicrystalline arrangements of convenient tiles. The cut and projection method starts from a 2D or 3D cut of a hypercubic space with a minimal thickness and projects it in a one-to-one correspondence onto physical 2D or 3D space [8]. This construction is linked with the algebraic properties of numbers which are the roots of the algebraic equations associated with the basic symmetries of the initial hyperspace [9].

These different theoretical methods have a more or less common underlying origin which is the energy minimization. The energy minimization for a system of particles linked by pair interactions leads to an optimal structure resulting from a superposition of density waves [2,10]. Then a discretization process such as the cut and projection method, can be used to define the quasilattice. Here we want to consider the discretization process used years ago to deduce an icosahedral seed [1]. Thus the goal of the present paper is to show that the principle of the energetic method leads to an octagonal tiling when starting from an octagonal seed.

In a first section the details of the energetic method are reported. The following section is devoted to the application of this method to the case of a sample invariant under eightfold symmetries. Conclusions are drawn from comparisons between possible defects and calculated diffraction patterns of these structures.

2 The energetic method

This method [2,10,11] is based upon the consideration of pair interactions defined by pair potentials $V_{ij}(\overrightarrow{r})$ between two atoms of species i and j which are located at a distance $\overrightarrow{r}$. Pair potentials are taken because of simplicity since known quasicrystals are metallic alloys. Thus the part of covalent binding can be neglected, as well as Fermi surface effects for these rather bad conductors. Introducing continuous densities for atomic species extends the set of considered structures and leads to the classic total static interaction energy E with the classical Bragg-Williams expression:

$$E = \frac{1}{2}\sum_{i,j}\int n_i(\overrightarrow{x})n_j(\overrightarrow{y})V_{ij}(\overrightarrow{x}-\overrightarrow{y})d\overrightarrow{x}\,d\overrightarrow{y}. \qquad (1)$$

Because of the convolution term, this interaction energy is easily translated in terms of Fourier transforms of atomic

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density and interaction potential:

$$E = \frac{1}{2}\sum_{i,j}\int n_i^*(\overrightarrow{p})n_j(\overrightarrow{p})V_{ij}(\overrightarrow{p})d\overrightarrow{p}. \quad (2)$$

This interaction energy is then optimized by considering a set of convenient infinitesimal variations of the specific optimal densities:

$$\delta n_i(\overrightarrow{p}) = C_i\delta(\overrightarrow{p} - \overrightarrow{p_0}) \quad (3)$$

where the Dirac delta-function is used, defining a wave-like variation of wavevector. Such variations do not keep constant the atomic density but are useful to define optimal configurations. When neglecting the boundary conditions, i.e. for an infinite sample, the deduced conditions for every species lead to the set of variational equations which form the linear system:

$$\sum_j n_j(\overrightarrow{p})V_{ij}(\overrightarrow{p}) = 0. \quad (4)$$

So, in order to obtain a non trivial solution of this linear system for the densities, the condition:

$$\det[V_{ij}(\overrightarrow{p})] = 0 \quad (5)$$

must be fulfilled for some wavevector $\overrightarrow{p_k}$.

Pair interactions are isotropic. Thus equation (5) simply defines the radial wavevector p_r and is indifferent to the wavevector direction. Apart from this directional degeneracy, equation (5) can also have several roots as it occurs actually for basic pair potentials [2]. Finally the directional degeneracy is lifted by the symmetry choice of the selected quasicrystalline structure in the practical calculation, equation (5) has a finite set of solutions.

Close to a single root $p_n\overrightarrow{u_m}$ where $\overrightarrow{u_m}$ is a unit vector, the equations (4) are easily linearized by developing the pair potentials $V_{ij}(\overrightarrow{p})$. One can deduce from equations (4) the set of approximate equations

$$\sum_j n_j(\overrightarrow{p})(A_{ij}\overrightarrow{p} + B_{ij}p_n\overrightarrow{u_m}) = 0 \quad (6)$$

with the final result after convenient linear operations using equation (5):

$$(\overrightarrow{p} - p_n\overrightarrow{u_m})\sum_j C_j n_j(\overrightarrow{p}) = 0. \quad (7)$$

Then with the assumption that all roots are simple ones, which is a weak constraint, leads to the solution for the optimal atomic densities $n_i(\overrightarrow{p})$ calculated as distribution functions $n_i(\overrightarrow{p}) = C_{n,m,i}\delta(\overrightarrow{p} - p_n\overrightarrow{u_m})$. Here the theorem which gives $C\delta(x) = T$ for the solution of the equation $xT = 0$ has been used [13]. Adding the contributions coming from all admissible roots, the resolution reads for every specific atomic density:

$$n_i(\overrightarrow{p}) = \sum_{n,m} C_{n,m,i}\delta(\overrightarrow{p} - p_n\overrightarrow{u_m}) \quad (8)$$

where a set of arbitrary unit vectors has been introduced. Thus the spatial density in real space is:

$$n_i(\overrightarrow{x}) = \sum_{n,m} C_{n,m,i}\exp[p_n\overrightarrow{u_m}\cdot\overrightarrow{x}]. \quad (9)$$

This set of equations for the species i gives the respective densities as a superposition of waves, as announced. A similar result was obtained from a direct analysis in real space, by means of a Taylor's expansion in the simple case of a single species [2]. Let us notice that a part A of an optimal structure contained within a box Δ has for density $n_{i,A}(\overrightarrow{x}) = n_i(\overrightarrow{x})Y(\Delta)$ where $Y(\Delta)$ is the step function of the domain Δ. The Fourier transform of this new density is deduced from the convolution:

$$F[n_{i,A}] = F[n_i] \circ F[Y_\Delta].$$

The delta peaks of the density $n_i(\overrightarrow{p})$ become smoothed with a width $\frac{\pi}{L}$ and a finite intensity for the density $n_{i,A}(\overrightarrow{p})$. The remark of the similarity between the Fourier transforms of the optimal structure and of a part of it enables us to deduce the basic wavevectors $p_n\overrightarrow{u_m}$ just from the Fourier transform of the density $n_{i,A}(\overrightarrow{x})$ of an optimal structure restricted within a box Δ. This definition of the basic wavevectors from a seed structure contained in a box is approximate because of the peak linewidth. And it does not define the phase and amplitude of the coefficients $C_{n,m,i}$ which can be selected either from a fit with experimental diffraction data or from simple arguments as done here. The great advantage of this method is that the knowledge of the detailed interactions within the sample is not required to obtain a structure. Since there are already tens of thousands of different quasicrystals with similar symmetries [12] this saves a considerable amount of work for a first approach. We call this method a Landau-like method by reference to numerous Landau's works and specially with helium rotons [6] because of the use of a variational method in the reciprocal space. The next point to understand is how the basic wavevectors of this seed do match themselves to define the structure.

3 2D quasicrystal construction with eightfold symmetry

The starting point is the definition of a simple set A, i.e. our seed cluster. Since eightfold symmetry is not compatible with crystalline symmetry while fourfold symmetry is, a reasonable definition of a cluster with eightfold symmetry introduces a cluster with fourfold symmetry, a centered square and duplicates this symmetry with two symmetric neighbors at each apex. The distance between neighbors is unity. This defines the set A as an octagon with a centered square inside as shown in Figure 1 which also indicates that two distinct initial squares lead to the same final octagon. The radius of the octagon is $L/2 = \sqrt{2+\sqrt{2}} = 1.85$. The wavevector density $n_{i,A}(\overrightarrow{p})$ of the set A is calculated on a polar lattice of 100×100 points restricted to a disk with a radius of a

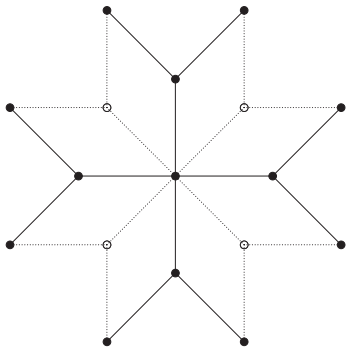


Fig. 1. The 12 sites cluster with eightfold symmetry, with unit length links. A variant of the central cluster is shown in dotted lines.

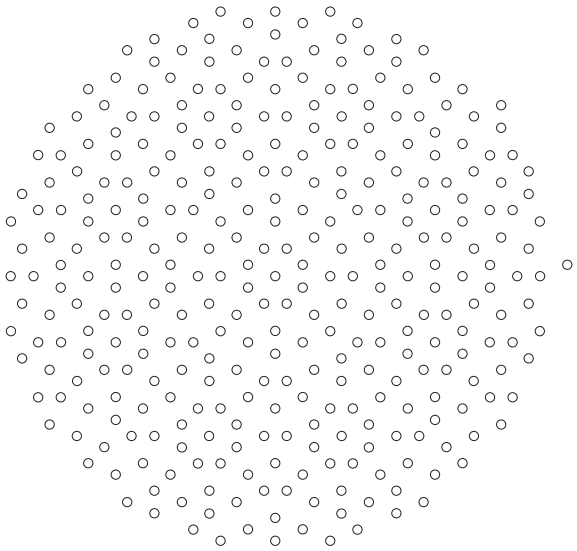


Fig. 2. The central part of the quasicrystalline structure with eightfold symmetry with $\{n_1 = 2, r_0 = 0.99\}$.

few 2π. From these points a steepest descent method is used to reach the maximum values of $n_{i,A}(\overrightarrow{p})$ and their location. Then the choice of a threshold $n_0 = 2$ enables us to restrict the set of maximums to eight, with octagonal invariance and $n'_0 = 5.73$, $p_0 = 7.039$. These results show both the imperfect interference since the maximal value of n is thirteen. The wavevector length is quite close to 2π, the geometrical value.

The quasicrystalline structure with eightfold symmetry in real space is then defined by the set of maxima of the atomic density defined in equation (9), where all coefficients are taken equal to unity for the sake of symmetry. Such a structure is characterized by the values $\{n_1, r_0\}$ where n_1 is the density threshold and r_0 is the minimal distance between admitted sites, a Delaunay's condition [14], which introduces the short range repulsive part of the interaction $V_{ij}(\overrightarrow{r})$. Obviously the value r_0 of the minimal distance must be chosen close to and smaller than unity in order to be consistent with the cluster construction, here $r_0 = 0.99$. The application of Delaunay's condition leads to select the highest maxima when two density maxima are closer than r_0. Of course each maximum is compared to all the previous ones. The structure deduced from a density value $n_1 = 2$ is shown in Figure 2 within a radius of 16.81 units. This structure extends to infinity the central octagon and its eightfold symmetry, and is practically independent of the choice of the density value which can be taken as high as four without any change, while the maximum value of $n(\overrightarrow{r} = 0)$ is nine since the null wavevector is also used as a basic vector. This relative independence of the structure upon the practical parameter choice is a strong proof for structural stability. Practically the atomic density values are different for the selected sites and are invariant under a central eightfold symmetry.

The distances between sites as they appear in a radial distribution function or in a pair distribution function define a discrete set as numerically observed in quasicrystalline structures [1]. Thus in order to compare the structure of Figure 2 with geometric tilings made of joint geometric figures, links between neighboring sites whose distance is close to a given value $d = \sqrt{2}$, $d' = \sqrt{2+\sqrt{2}}$, $d'' = \sqrt{2}\sqrt{2+\sqrt{2}}$ are drawn on the structure as shown in Figures 3. This enables us to show in Figure 3a with the pairing distance d that the central octagon is reproduced. Similar octagons and parts of octagons appear in the whole sample. In Figure 3b with the pairing distance d', i.e. the octagon radius, the whole quasilattice is defined by links. Only two tiles of side d are involved in this construction, namely a square and a rhombus with angles 45° and 135°, a Ammam-Beenker octagonal lattice. These tiles are obviously linked with the eightfold symmetry as already shown by other authors [16]. It must be noted that this tiling is perfect. In Figure 3c the resulting figure with the pairing distance d'' contains two entangled sets of regular percolating lines and a central independent octagon as well as isolated points. In Figures 2 and 3 the structure deduced from the initial external cluster seed of eight sites shown in Figure 1 is found. The process extends the basic structure of this initial seed up to infinity.

When using a modified process different defects can be produced. These local defects do not perturb the site network over more than a few distance units. As a matter of fact these defects also exhibit a local eightfold symmetry. The first defect is a symmetric variant of a small unit composed of two rhombi and one square, while the second defect is localized within an octagon made of two squares and four rhombi. In the last cases, several symmetric configurations can occur and a few ones are observed in less accurate numerical treatments. This defines some phasons of this structure [15]. In Figure 4 the structure with $\{n_1 = 4.5, r_0 = 0.99\}$ is reported with the pairing distance d'. There is evidence for a few local defects of various sizes. There are eight defects which are eight individual lacunas in a perfect sample of 5869 sites. In the linked lattice these defects define four darts and four stars with two sharp edges, while the other part is unmodified.

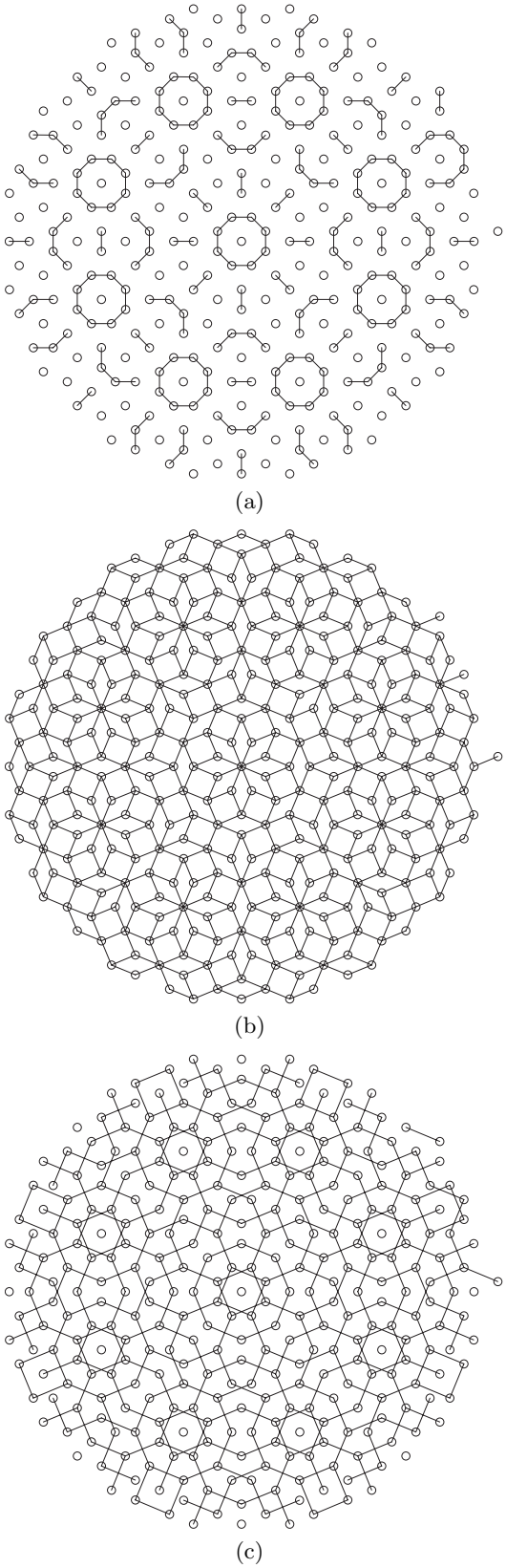


Fig. 3. (a) The same structure as in Figure 2 with links between pairs with $d = 1.4$. (b) The same structure as in Figure 2 with links between pairs with $d' = 1.85$. (c) The same structure as in Figure 2 with links between pairs with $d'' = 2.61$.

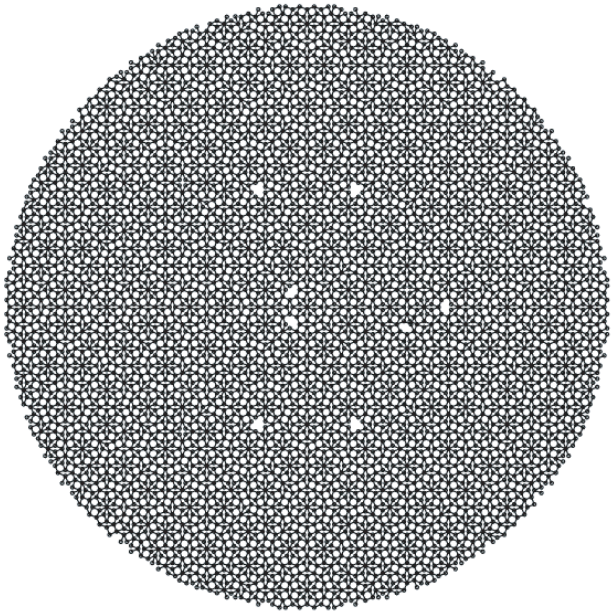


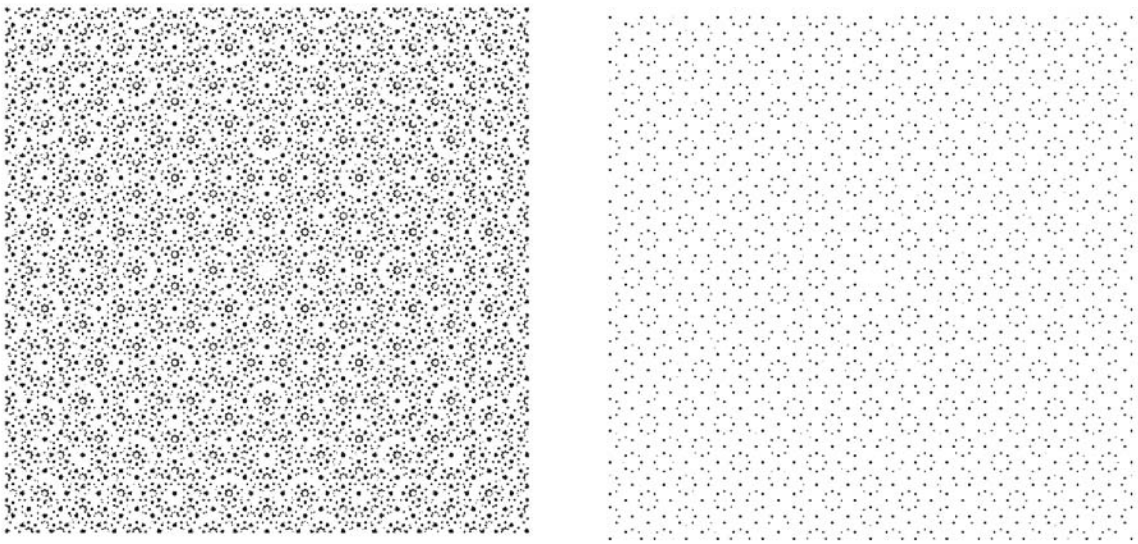
Fig. 4. The central part of the quasicrystalline structure with eightfold symmetry with $\{n_1 = 4.5, r_0 = 0.99\}$, i.e. 5861 sites with links between pairs with $d' = 1.85$.

Of course the structure deduced from $\{n_1 = 5, r_0 = 0.99\}$ or $\{n_1 = 6, r_0 = 0.99\}$ contains larger and more numerous defects.

The diffraction patterns associated with the structure of 353 sites shown in Figure 2 are reported respectively in Figure 5a when the amplitude threshold n_2 for the selection of a point is 50 and in Figure 5b when $n_2 = 100$. Both Figures exhibit a perfect eightfold symmetry, practically threshold independent, with an extreme accuracy. This diffraction pattern is quite resistant to the introduction of defects in the structure. As an obvious conclusion of the clear definition of this structure, its moderate richness in defects and its well defined diffraction pattern, the eightfold symmetry leads to a stable structure. This confirms the experimental observations [4,5].

4 Conclusive remarks

So the energetic method used here leads to an octagonal quasilattice when starting from an octagon seed made of thirteen atoms: an octagon, a square and a central atom. Moreover during the proper use of the energetic method, the quasilattice stability is evidenced since the resulting structure is unaffected by significant changes of the used parameters. Sites are seen to be non equivalent. And thus lacunas can be created, with evidence for different local environments. The generalization to other seed symmetries, practically sevenfold, elevenfold and thirteen has been developed with the introduction of new structures and arguments on their relative stability.



(a) (b)

Fig. 5. The diffraction patterns of the structure shown in Figure 2 with (a) $n_2 = 50$ and (b) $n_2 = 100$.

References

1. J.-C.S. Levy, D. Mercier, J. Appl. Phys. **53**, 291 (1982); D. Mercier, J.-C.S. Lévy, Phys. Rev. B **27**, 1293 (1983)
2. J.-C.S. Levy, Surf. Sci. **104**, 1 (1981)
3. D. Shechtman, I.A. Blech, D. Gratias, J.W. Cahn, Phys. Rev. Lett. **53**, 1951 (1984)
4. N. Wang, H. Chen, K.H. Kuo, Phys. Rev. Lett. **59**, 1010 (1987)
5. J.-C. Jiang, S. Hovmöller, X.-D. Zou, Phil. Mag. Lett. **71**, 123 (1995)
6. L.D. Landau, J. Phys. (USSR) **5**, 71 (1941); L.D. Landau, J. Phys. (USSR) **11**, 91 (1947)
7. B. Grunbaum, G.C. Shephard, *Tilings and Patterns* (Freeman Editions, New York, 1987)
8. N.G. de Bruijn, Nederl. Akad. Wetensch. Proc. Ser. A **43**, 39 (1981)
9. R.V. Moody, J. Patera, J. Phys. A **26**, 2829 (1993)
10. P.M. de Wolff, Acta Cryst. A **30**, 777 (1974)
11. J.-C.S. Lévy, *Nouvelles Structures de Matériaux* (Masson, Paris, 1993)
12. P.J. Lu, K. Deffeyes, P.J. Steinhardt, N. Yao, Phys. Rev. Lett. **87**, 275507 (2001)
13. L. Schwartz, *Théorie des Distributions* (Hermann, Paris, 1967)
14. B.N. Delaunay, N.P. Dolbilin, M.I. Shtogrin, R.V. Galiulin, Dokl. Akad. Nauk. **227**, 19 (1976)
15. D.M. Frenkel, C.L. Henley, E.D. Siggia, Phys. Rev. B **34**, 3649 (1986)
16. P.A. Kalugin, in *Beyond Quasicrystals*, edited by F. Axel, D. Gratias (Springer, Les Éditions de Physique, 1995)