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Two-dimensional system with a quasi-crystalline ground state

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Résumé. — Nous étudions les empilements denses de particules avec des interactions qui favorisent la symétrie pentagonale. Ces structures peuvent être obtenues par décoration de pavages de Penrose ou directement par une méthode de coupe et projection. Nous prouvons qu'elles forment l'état fondamental dégénéré, dans le cas de potentiels de paire limités aux premiers voisins et pour des paramètres de forces d'interaction appartenant à un large domaine de valeurs. Des trempes à partir du liquide, simulées par dynamique moléculaire, conduisent soit à un verre, soit à un quasi-cristal, selon la vitesse de trempe.

Abstract. — We study dense packings of particles with interactions favouring pentagonal symmetry. Such structures can be obtained by decorations of Penrose tilings or directly with a cut-and-projection method. We prove that they form the degenerated ground-state in the case of pair potentials limited to first neighbours and for interaction strength parameters lying in broad ranges of values. Cooling runs from the liquid, simulated by molecular dynamics, produce either a glassy state or a quasi-crystalline state depending on the quenching rate.

1. Introduction.

The Penrose tilings [1-3] are two-dimensional analogs of the structures of quasi-crystalline materials. Thus the study of their properties has much interest in the field of aperiodic lattices [4-10]. On the other hand many physical properties depend on the atomic positions and not only on the underlying lattice. Mosseri and Sadoc [11] have proposed decorations which correspond to covalent coordinations. Henley [12] has built packings of disks placed on vertices of Penrose tilings and has studied the packing fraction and the coordination numbers. Levine and Steinhardt [5, 7] have described several atomic models. They have reported that a numerical relaxation can maintain orientational and translational order over the sample.

A decoration of the Penrose tilings has been proposed by Lançon *et al.* [13] (hereafter denoted as paper I) to produce an atomic model of a dense two-dimensional quasi-crystal. To study this two-component system, pair interactions have been introduced with peculiar bond lengths and bond strengths. A molecular dynamics simulation has shown that the quasi-crystal is stable over a broad range of temperature, it undergoes a first-order transition to the liquid phase and it is more stable than the amorphous phase.

The same atomic system has been recently used by Widom *et al.* [14] and Minchau *et al.* [15]. Widom *et al.* have shown that quasi-crystalline structures can be obtained with Monte-Carlo runs. They have also introduced another set of interactions to show that the quasi-crystal is a degenerated ground-state. We prove in this paper that this result is in fact true in a broad range of interaction strengths values.

Minchau *et al.* [15] have defined growth algorithms which stick particles onto an initial cluster. They have generated structures they analysed as alternations of strips of the Penrose rhombs.

Another quasi-crystal growth model has been proposed by Elser [16] : regular decagons are packed in the plane. The two kinds of particles defined in paper I can give a physical realization to these decagon packings : the large particles have to be set at the decagon centres and the small ones at the centres of the regular interstitial sites.

In this paper, we study how dense packings with pentagonal order can be obtained and we discuss their stability. We also describe molecular dynamics experiments : a quasi-crystalline or a glassy state can

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result from the quench of a liquid, depending on the quenching rate.

2. Binary tillings.

In paper I it has been shown that a two-component system having a pentagonal orientational order is stable with radially symmetric particle interactions. The bond lengths between particles are chosen so that

$$\begin{aligned} d_{LL}/d_{LS} &= 2 \sin (\pi/5) \\ d_{SS}/d_{LS} &= 2 \sin (\pi/10) \end{aligned} \tag{1}$$

where L and S denote large and small atoms respectively. Now we aim to discuss the geometrical properties of dense packings generated with these interactions. Let us note that these particles are not hard disks. Therefore we cannot use the packing fraction to measure the efficiency of the packing. Considering a polyatomic system defined by the values of given bond lengths $d_{\alpha\beta}$ for each type of pairs $\alpha - \beta$, our criterium for a close packing will be : all geometric neighbours (in the sense of Voronoï) — one of type α and the other of type β — must be at a distance equal to the bond length $d_{\alpha\beta}$. Geometrical neighbours are unambiguously defined by the partition of the plane into Voronoï polygons around each particle [17, 18] (also called Dirichlet regions [19] or Wigner-Seitz cells [20]). The network of the geometrical neighbouring pairs is formed by triangles which completely fill the plane [21]. Thus such a close packed structure is a tiling of triangles with edge lengths equal to d_{LL}, d_{LS} or d_{SS}. Because equilateral triangles formed by three similar atoms do not lead to pentagonal order, we do not consider tilings which contain such units. Therefore the only types of triangles are LLS and LSS. If we consider one of them, say LSS for instance, there is necessarily an L atom on the other side of the edge SS. Thus these structures correspond to tilings formed with the two Penrose rhombs shown in figure 1. The rhombs have the following vertex occupations : the vertices where the angle has a value which is an odd multiple of $\pi/5$ (namely $\pi/5$ and $3\,\pi/5$) are occupied by an L atom, whereas the vertices where this value is an even multiple of $\pi/5$ (namely $2\,\pi/5$ and $4\,\pi/5$) are occupied by an S atom. The edges of the rhombs are the LS pairs, and the LL and SS pairs from the short diagonal of the fat and the thin rhombs respectively.

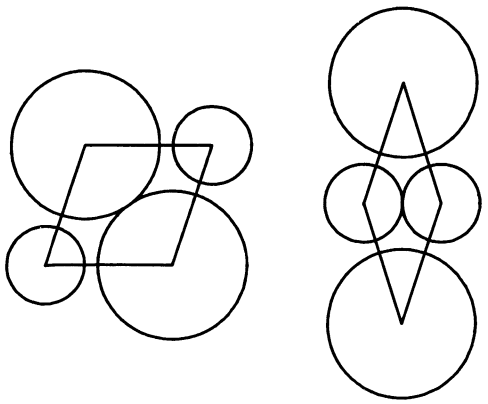

Fig. 1. — Decoration of the two Penrose rhombs in a binary tiling.

The definite vertex occupations are actually very simple matching rules for the packing of the rhombs. These matching rules are completely different from those of the original Penrose tiling and also from those derived by Kleman *et al.* [9] for generalized Penrose tilings. They do not assure translational order, but they imply the following striking property for the tiling : around any vertex the angles have values which are either all odd, or all even multiples of $\pi/5$. We shall call tilings with such a parity property binary tilings because they can naturally be decorated to produce a dense binary allow (one can also set spins alternatively up and down on the vertices). In the next section we shall see how to construct such binary tilings, and that they can be periodic, quasiperiodic or complex structures.

3. Quasi-crystalline packings.

3.1 DECORATION OF THE PENROSE CELLS. — In the same way as a decoration of a periodically repeated unit cell produces a crystal, a decoration of the two Penrose rhombs involved in the generalized Penrose tilings [6] produces a quasi-crystal. Such a decoration (Fig. 2) has been proposed in paper I.

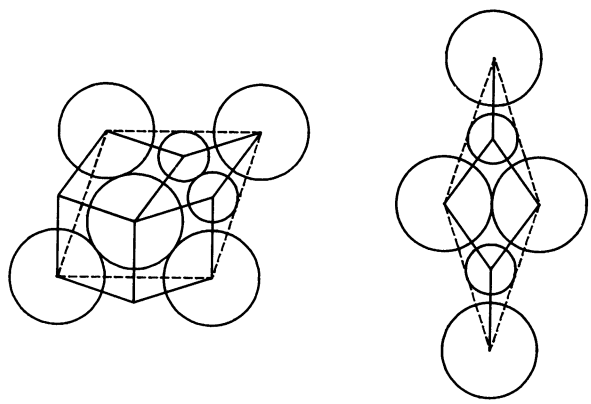

Fig. 2. — Decoration I of the two rhombic quasi-unit cells (dashed lines). The rhombs of the resulting binary tiling are drawn in solid lines.

It was noticed that there were two equivalent ways of decorating the fat rhomb, putting the interior L (large) atom near one or the other of the two acute angles. Using one or the other way randomly during tiling decorations, it happens that four L atoms may cluster together and induce an LL pair length not

equal to d_{LL}, in contradiction with our close packing rule of section 2.

In the case of a tiling being produced by the projection technique, we propose a rule which prevents such an event : following Duneau and Katz [6], we denote Δ the principal diagonal of the 5-dimensional cubic lattice. We consider for each fat rhomb and among the two vertices which are situated at an acute angle, the vertex which corresponds to the point in the perpendicular space which is the furthest from Δ. We put the interior L atom near this vertex. Geometrical considerations in the perpendicular space ensure that, when two fat rhombs have a common edge, the interior L atom of one is not in the neighbourhood of the interior L atom of the other, and therefore the forbidden configurations can never occur (see appendix B). The network of LS pairs in the decorated tiling produces a binary tiling shown in figure 3a.

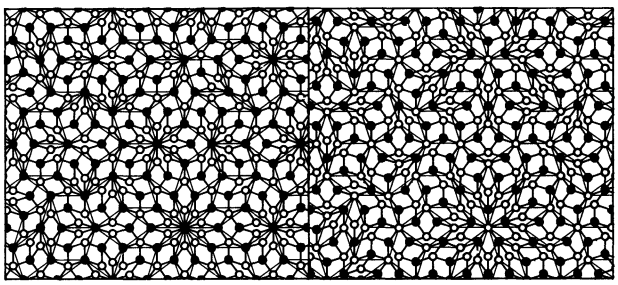

Fig. 3. — Binary tilings (thick lines) obtained from the same initial Penrose tiling (thin lines) by decorations number I (left) and number II (right). The two types of vertices are indicated as filled and empty disks.

Note that this binary tiling is quasi periodic but presents particular occurrences of the unit rhombs arrangements which are not those found in the original Penrose tiling, nor those found in the more general tilings obtained by translating the strip of the cut-and-projection method along Δ [6]. Thus it belongs to other local isomorphism classes (for a definition of this concept, see [3, 7, 8, 22]). Let us call this construction decoration number I. We shall now describe another way to get binary tilings.

3.2 DECORATION NUMBER II. — The Penrose rhombs can be decorated according to the scheme of figure 4. This decoration breaks the symmetry of both rhombs. Thus, to ensure a perfect binary tiling, the decorations for the differently oriented rhombs are deduced from each other by rotation of an angle which is a multiple of $2\pi/5$.

This decoration is possible from any tiling built with the Penrose rhombs : Penrose tilings, periodic tilings (and among them periodic approximants of Penrose tilings), binary tilings, ...

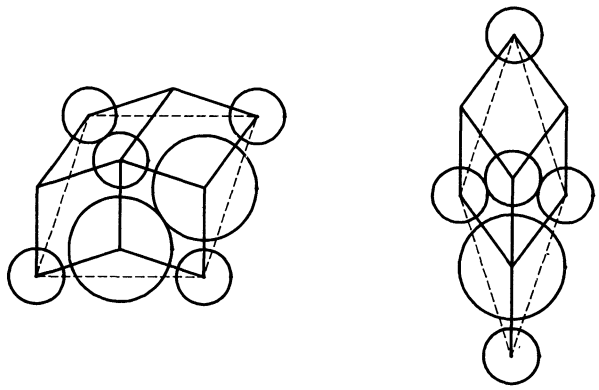

Fig. 4. — Decoration II of the two rhombic quasi unit cells (dashed lines). The rhombs of the resulting binary tiling are drawn in solid lines.

For instance two simple periodic binary tilings can be constructed, using one or the other of the two decorated Penrose rhombs as their unit cell. The crystal constructed with the fat rhomb has already been used in a previous molecular dynamics study [23].

If the initial tiling is quasi-periodic, the associated binary tiling is itself quasi-periodic and generally belongs to a different local isomorphism class of that associated by the decoration number I. Both are shown in figure 3. The tilings obtained by decoration number II can also be obtained by the cut-and-projection method from a 10-dimensional cubic lattice with the irrational orientation of the strip different from that of the tiling plane (see appendix A).

3.3 GEOMETRIC PROPERTIES. — If the numbers of fat and thin rhombs in the original tiling are respectively FA and TH, these numbers in the binary tilings obtained by both decorations are :

$$\begin{aligned} fa &= 3\,FA + TH \\ th &= FA + 2\,TH\,. \end{aligned} \tag{2}$$

The ratio of the sizes between the original and the new tiles is $2\sin(2\pi/5)$. The numbers of L and S atoms are $N_L = (3\,fa + th)/5$ and $N_S = (2\,fa + 4\,th)/5$ respectively. The binary tilings exist for $1/2 \leqslant N_L/N_S \leqslant 1$. In the case of Penrose tilings, $FA/TH = \tau$, the golden mean. Relations (2) show that $fa/th = \tau$ and $N_L/N_S = \tau/2$.

4. Energetic properties.

In order to study the physical properties of quasi-crystals, we must now introduce more precisely atomic interactions in our model. We assume pair potentials limited to first neighbours with depths ε_{LL}, ε_{LS}, ε_{SS} for the three different types of pairs. Let

N_{LL}, N_{LS}, N_{SS} be the numbers of these pairs. In the ground state, the energy per atom is

$$E = -(N_{LL}\,\varepsilon_{LL} + N_{LS}\,\varepsilon_{LS} + N_{SS}\,\varepsilon_{SS}) \times \times /(N_L + N_S) \quad (3)$$

where N_L and N_S are the numbers of L and S atoms respectively.

The candidates for the ground state structure are the close packed configurations, i.e., the triangular lattice of L-atoms, the triangular lattice of S-atoms and the binary tilings. Depending on concentration and on depths ε_{ij}, the ground state is one of these structures or a mixing of them.

In a triangular lattice, say of L-atoms, the numbers of pairs are : $N_{LL} = 3\,N_L$ and $N_{LS} = N_{SS} = 0$.

On the other hand, in the binary tilings, the numbers of pairs are related to the numbers of rhombs, and thus to N_L and N_S :

$$\begin{aligned} N_{LL} &= fa = 2\,N_L - N_S/2 \\ N_{LS} &= 2(fa + th) = 2(N_L + N_S) \\ N_{SS} &= th = -N_L + 3\,N_S/2\,. \end{aligned} \quad (4)$$

Therefore the energy is only dependent on the atomic concentration : all the binary tilings at a given concentration are members of a highly degenerate state. A similar conclusion has been drawn by Widom *et al.* [14] for a special choice of the potential depths $\varepsilon_{\alpha\beta}$, but actually is valid for any set of depths $\varepsilon_{\alpha\beta}$.

The phase diagram can be calculated as a function of the concentration and of the depths ε_{ij}. In the special case where $\varepsilon_{LL} = \varepsilon_{SS}$, the binary tilings, and among them the quasi-crystals, are the ground states when :

$$\varepsilon_{LS} > \varepsilon_{LL}\,. \quad (5)$$

If the concentration is out of the range of existence of binary tilings, the ground states are mixings of a binary tiling and a triangular crystal formed with the atoms in excess.

When $\varepsilon_{LL} > \varepsilon_{LS}$, the system is separated in two monoatomic phases.

For the molecular dynamics simulation, we have chosen :

$$\varepsilon_{LS} = 2\,\varepsilon_{LL} = 2\,\varepsilon_{SS} \quad (6)$$

as in paper I.

5. Molecular dynamics simulation.

5.1 INTRODUCTION. — We have shown that the binary tilings form the ground state when one chooses the potential parameters as given by equations (1) and (6). In paper I, it was shown by molecular dynamics that such a state is stable at low temperatures. This result is confirmed by Widom *et al.* [14] on quasi-crystalline structures obtained by Monte-Carlo coolings from random configurations. While Monte-Carlo and molecular dynamics techniques shall lead to the same thermodynamical quantities for equilibrium states, the former uses artificial processes, such as particle « flips », when the latter computes particle trajectories which correspond to a physical dynamics. A kinetic study, undertaken in paper I, has shown that a quench from the liquid state leads to a glassy state. We show in this section that this result is dependent on the quenching rate.

In order to analyse the quenched states, we compute their diffraction patterns (with the same atomic scattering factor for L and S atoms). The diffraction pattern for a perfect binary tiling structure is shown in figure 5 (*). Because the LS pairs do form the tiling, we focus on them. We consider the angles formed by an arbitrary vector and the vectors from L atoms to S first-neighbours, and compute the bond orientational distribution shown in figure 6 in the case of a quasi-crystal.

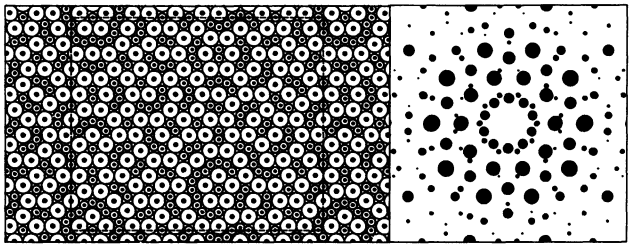

Fig. 5. — Model of a two dimensional quasi-crystal and its computed diffraction pattern (intensities above some given threshold are represented by the areas of the spots). The dotted line indicates the periodic boundaries used in the molecular dynamics calculations.

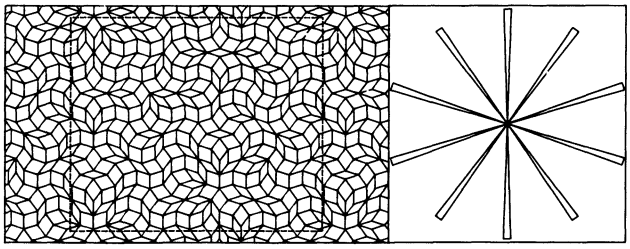

Fig. 6. — Left : Network of LS pairs between large and small neighbouring particles in the quasi-crystal model shown in figure 5. Right : Corresponding bond orientational distribution.

(*) Because we wish to use periodic boundary conditions, we consider periodic approximants to aperiodic tilings. They correspond to continued-fraction approximants to the golden mean τ [13].

5.2 METHOD. — We have employed the molecular dynamics technique with constrained equations of motions [24]. In this method, the temperature and pressure are constants of the motion [25, 26]. These equations can be generalized to produce a given variation $\dot{T}$ of temperature or a given variation $\dot{P}$ of pressure with time [27, 23]. Let $\mathbf{q}_i$ be the coordinates of atom i, $\mathbf{p}_i$ be the momentum of i divided by its mass m_i, $\mathbf{F}_i$ be the force acting on i, V be the volume occupied by the N particles, and n be the space dimension. The constrained equations of motions are :

$$\begin{aligned} \dot{\mathbf{q}}_i &= \mathbf{p}_i + \dot{\varepsilon}\mathbf{q}_i \\ \dot{\mathbf{p}}_i &= \mathbf{F}_i/m_i - \beta\mathbf{p}_i \\ \dot{V} &= nV\dot{\varepsilon}\,. \end{aligned} \tag{7}$$

For given derivatives of the temperature and of the pressure, $\dot{T}$ and $\dot{P}$, the damping coefficient β and the volume variation coefficient $\dot{\varepsilon}$, have the value :

$$\beta = \left[\sum_i (\mathbf{p}_i \cdot \mathbf{F}_i) - \frac{n}{2} Nk\dot{T}\right] \Big/ \left[\sum_i (m_i\, \mathbf{p}_i^2)\right] \tag{8}$$

$$\dot{\varepsilon} = \frac{nNk\dot{T} - n\dot{P}V - \frac{1}{2}\sum_{i,j\,(i\neq j)} \left[\left(\frac{\varphi'_{ij}}{q_{ij}} + \varphi''_{ij}\right)(\mathbf{q}_{ij}\cdot\mathbf{p}_{ij})\right]}{n^2PV + \frac{1}{2}\sum_{i,j\,(i\neq j)} \mathbf{q}_{ij}^2\left(\frac{\varphi'_{ij}}{q_{ij}} + \varphi''_{ij}\right)} \tag{9}$$

where φ_{ij} is the pair potential between the i and j atoms and is a function of their distance q_{ij}. We have chosen the modified Johnson potential [28] properly scaled for each of the three types of pairs. Let $r_0 = d_{LS}$ be the unit of length and let $\varepsilon_0 = \varepsilon_{LS}$ be the unit of energy. For simplicity, we let the mass of the L and S atoms be equal to the unit of mass m_0. With this set of units, the unit of time, t_0, is $(m_0\, r_0^2)^{1/2}$, the unit of (two-dimensional) pressure, P_0, is ε_0/r_0^2 and the unit of temperature, T_0, is ε_0/k. The numerical integration of the dynamical equations is done with a predictor-corrector algorithm [29], and with a time step equal to $0.001\, t_0$.

5.3 NUMERICAL EXPERIMENTS. — Our starting point is a liquid state obtained in paper I. It contains 152 L particles and 188 S particles equilibrated at the temperature $0.7\, T_0$ and the pressure P_0. We have carefully checked that all initial clusters of geometrical neighbours have been destroyed during this equilibrium run. In all the simulations the pressure has been kept constant at P_0. Because the derivative $\dot{T}$ of the temperature appears explicitly in equations (8) and (9), we can simulate cooling down linear with time. During a quench, the system goes through three temperature ranges : above the melting temperature T_m, the system is in thermodynamic equilibrium ; below the glass transition temperature T_g, the system is frozen because the diffusion coefficient is too low ; and thus it is during the supercooled liquid temperature range between T_m and T_g that more efficient atomic reorganizations are likely to occur. In this temperature range, the two simulations reported here have been made with quenching rates in the ratio $1 : 10^4$.

The values of T_m and T_g have been determined in paper I to be about $0.4\, T_0$ and $0.3\, T_0$ respectively. For the slow cooling, the quenching rate from the high temperature $0.7\, T_0$ down to T_m and from T_g down to the low temperature $0.01\, T_0$ was equal to $2 \times 10^{-3}\, T_0/t_0$. In the critical temperature range from T_m down to T_g, the quench was made with a rate equal to $2 \times 10^{-5}\, T_0/t_0$ and followed by an isothermal treatment during $260\, t_0$ before quenching to lower temperatures. For the fast quench the rate was constant and equal to $2 \times 10^{-1}\, T_0/t_0$.

The glass obtained with the higher cooling rate is shown in figure 7 ; its amorphous structure is confirmed by the diffuse rings in the diffraction pattern. With the lower cooling rate, the state obtained is shown in figure 8 : the diffraction spots indicate a pentagonal symmetry and the presence of translational order. This state corresponds to an imperfect quasi-crystalline structure. The potential energy of these configurations are respectively 14 % and 4 % above that of the ground state. The perfection of the packings can be estimated by looking at the Penrose rhombs present in the configurations. Figures 9 and 10 show the network of rhombs formed by the LS

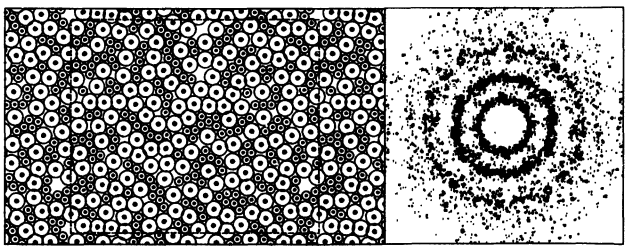

Fig. 7. — Two-dimensional glass obtained by cooling the liquid state and its diffraction pattern.

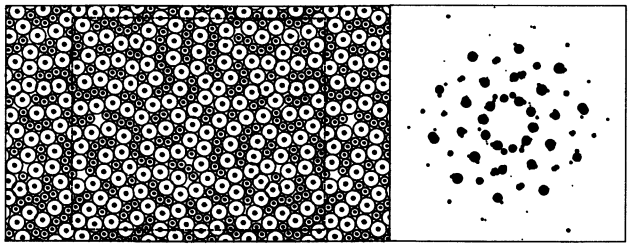

Fig. 8. — Imperfect quasi-crystal configuration obtained by cooling the liquid state and its diffraction pattern.

pairs. Their short diagonal must be an LL or an SS pair. Because the rhombs can be distorted, we consider as first neighbour pairs all those which contribute to the first peak of the radial distribution function. The quasi-crystalline configuration contains defects. Note that some of these maintain the pentagonal symmetry. The bond orientational distributions are also shown in figures 9 and 10. Because they are distributions of the angles of the LS pairs with a fixed direction, they do not correspond to a local orientational parameter and therefore no fivefold symmetry appears in the histogram of the glassy state (Fig. 9) ; on the other hand, the distribution of figure 10 shows that the pentagonal symmetry propagates through the whole configuration obtained with the lower cooling rate.

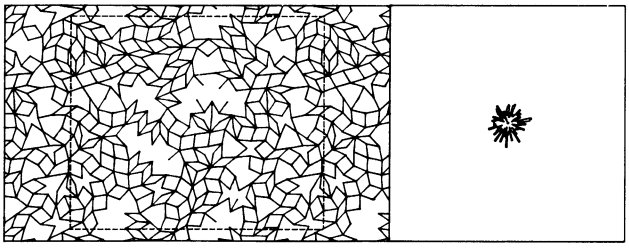

Fig. 9. — Network of LS pairs in the glass shown in figure 7 and the corresponding bond orientational distribution.

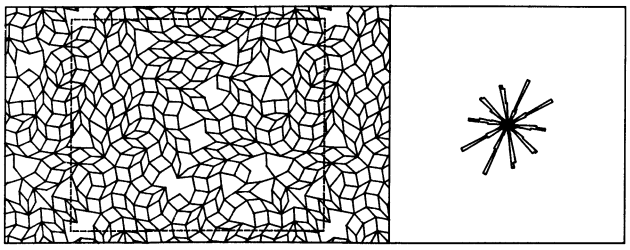

Fig. 10. — Network of LS pairs in the quasi-crystalline configuration shown in figure 8 and the corresponding bond orientational distribution.

6. Conclusion.

We have introduced two-dimensional quasi-crystalline tilings constructed with the Penrose rhombs and with special matching rules. These tilings belong to the general class of structures that we have called binary tilings because atoms with two different sizes setted at their vertices decorate them to form dense binary alloys. We have proved that pair potential interactions with given bond length values are sufficient to make them the ground state in a broad range of bond strength values. Molecular dynamics simulations show that the quasi-crystal or the glassy states are obtained by quenching the liquid state with different cooling rates.

One of us (F. L.) thanks D. Gratias and J. Toner for their stimulating lectures on quasi-crystals as well as helpful discussions at the « école de physique de la matière condensée » organized by C. Godrèche at Beg-Rohu, France, on September 1986.

Appendix A.

BINARY TILINGS CONSTRUCTED BY PROJECTION : From a Penrose tiling produced by the projection method from $\mathbb{Z}^5$ [6], we wish to construct a binary tiling by the same technique and with the following property : all the vertices of the initial Penrose tiling are vertices of the derived tiling. Let $C_n = \cos(n\pi/5)$ and $S_n = \sin(n\pi/5)$, and let $(\mathbf{e}_1, \ldots, \mathbf{e}_5)$ be the natural basis of $\mathbb{R}^5$. The tiling plane, E_T, is generated by the vectors $\mathbf{x}_1 = (C_{2i})_{i=0,4} = (1, C_2, C_4, C_4, C_2)$ and $\mathbf{x}_2 = (S_{2i})_{i=0,4} = (0, S_2, S_4, -S_4, -S_2)$. The vertices of a generalized Penrose tiling [6] are the projection onto E_T of the points of the cubic lattice $\mathbb{Z}^5$ which are contained in a strip S parallel to a plane $E_\parallel$. In this case the two planes E_T and $E_\parallel$ are the same. More precisely the strip is constructed by translating the unit 5D-cube by a vector $\mathbf{t}$ and then moving it along $E_\parallel$. Tilings, which are members of different local isomorphic classes, can be produced with suitable choices of vector $\mathbf{t}$.

Let $(\boldsymbol{\varepsilon}_1, \ldots, \boldsymbol{\varepsilon}_{10})$ be the natural basis of $\mathbb{R}^{10}$. We introduce the linear mapping, M, of $\mathbb{Z}^5$ into $\mathbb{Z}^{10}$ which transforms $\mathbf{e}_1$ into $M(\mathbf{e}_1) = \boldsymbol{\varepsilon}_8 + \boldsymbol{\varepsilon}_9$, $\mathbf{e}_2$ into $M(\mathbf{e}_2) = \boldsymbol{\varepsilon}_{10} + \boldsymbol{\varepsilon}_1$ and so on by circular permutation of the indices. Let E'_T be the plane of $\mathbb{R}^{10}$ generated by $\boldsymbol{\chi}_1 = (C_i)_{i=0,9} = (1, -C_4, C_2, -C_2, C_4, -1, C_4, -C_2, C_2, -C_4)$ and $\boldsymbol{\chi}_2 = (S_i)_{i=0,9} = (0, S_4, S_2, S_2, S_4, 0, -S_4, -S_2, -S_2, -S_4)$. If we identify E_T and E'_T, and by calculating the orthogonal projection of $\mathbf{e}_i$ and $M(\mathbf{e}_i)$ onto these planes, we can easily check that the projection of a point P is transformed into the projection of M(P) by a rotation of $-\pi/2$ and a scaling by a factor equal to $2\,S_2$. In particular this will be the case for the vertices of the initial Penrose tiling. Let $\mathbf{P}$ be a lattice point contained in strip S and thus which is projected at a vertex of the Penrose tiling. If vector $\mathbf{t}$

is the translation of the strip, **P** can be written in the form :

$$\mathbf{P} = \sum_{i=1}^{5} \lambda_i \mathbf{e}_i + \mathbf{t} + \mathbf{v}$$

where $\mathbf{v} \in E_\parallel$ and $\lambda_i \in [0, 1[$.
M(**P**) is equal to :

$$M(\mathbf{P}) = \sum_{i=1}^{10} \alpha_i \varepsilon_i + M(\mathbf{t}) + M(\mathbf{v})$$

where $\alpha_8 = \lambda_1$, $\alpha_9 = \lambda_1$ and so on with a circular permutation of the indices. Since the α_i lie within to the range $[0, 1[$, M(**P**) is contained in the strip S′ which is constructed by translating the unit 10-cube by a vector M(**t**) and then by moving it along $E'_\parallel = M(E_\parallel)$. Note that the plane $E'_\parallel$ which defines the strip is different from the tiling plane E'_T. In other words, all the points which are in strip S and are projected onto E_T at the Penrose tiling vertices, lead to points which are in strip S′ and which form, by projection onto E'_T, the same tiling (up to the $-\pi/2$ rotation and the inflation by a factor $2\,S_2$ seen above). But M transforms S only into a subset of S′. Hence the tiling obtained by the projection of the lattice points contained in strip S′ has new vertices which decorate the initial tiling as shown in figure 3b. Since Penrose tilings which are members of different local isomorphic classes correspond to different translations of the unit 5D-cube along its diagonal, they lead to binary tilings which correspond to different translations of the unit 10D-cube along its diagonal and thus which are themselves members of different local isomorphic classes.

Appendix B.

ANALYSIS OF DECORATION I : Because decoration I, shown in figure 2, breaks the fat rhomb symmetry, there are two equivalent ways of setting the interior L (large) particle. We show here that the rule given in section 3.1 is an unambiguous way to do it. We assume that the tiling to be decorated is obtained by the projection method from the cubic 5D-lattice and that the tiling plane, E_T, and the plane, $E_\parallel$, which fixes the orientation of the strip, are generated by the two vectors $\mathbf{x}_1 = (C_{2i})_{i=0,4}$ and $\mathbf{x}_2 = (S_{2i})_{i=0,4}$ (same notations as in appendix A). The perpendicular space, $E_\perp$, to plane $E_\parallel$ is a three-dimensional space generated by the vectors $\mathbf{x}_3 = (C_{4i})_{i=0,4}$, $\mathbf{x}_4 = (S_{4i})_{i=0,4}$ and $\mathbf{x}_5 = (1, 1, 1, 1, 1)$. Let $F_\perp$ be the plane generated by vectors $\mathbf{x}_3$ and $\mathbf{x}_4$, and Δ be the principal diagonal of the 5D-cube which defines the strip.

Penrose rhombs are the projections onto E_T of 2D-facets of the 5D-lattice, i.e., their edges are formed by two vectors $(\mathbf{e}_{i\parallel}, \mathbf{e}_{j\parallel})$ $(i \neq j)$, where $\mathbf{e}_{i\parallel}$ is the projection of the basis vector $\mathbf{e}_i$ onto $E_\parallel$. The fat and the thin rhombs are generated by vector pairs of type $(\mathbf{e}_{i\parallel}, \mathbf{e}_{i+1\parallel})$ and $(\mathbf{e}_{i\parallel}, \mathbf{e}_{i+2\parallel})$ respectively.

Consider a fat rhomb, for instance one whose corners are : M_0, $M_0 + \mathbf{e}_{1\parallel}$, $M_0 + \mathbf{e}_{2\parallel}$ and $M_1 = M_0 + \mathbf{e}_{1\parallel} + \mathbf{e}_{2\parallel}$. Points M_0 and M_1 are the two acute angle corners of the rhomb. Let $\mathbf{e}_{i\perp}$ be the projection of $\mathbf{e}_i$ onto $F_\perp$ (note that for our purpose we do not use this notation for the projection onto $E_\perp$). The above rhomb corners correspond respectively to the following points of $F_\perp$: N_0, $N_0 + \mathbf{e}_{1\perp}$, $N_0 + \mathbf{e}_{2\perp}$ and $N_1 = N_0 + \mathbf{e}_{1\perp} + \mathbf{e}_{2\perp}$, which form a thin rhomb.

The plane $F_\perp$ is divided into two half-planes : the location of the points N_0 where N_0 is nearer to Δ than N_1 and that where N_0 is further from Δ than N_1 (Fig. 11). The interior L atom is set near M_1 in the former case and near M_0 in the latter case (when N_0 lies on the boundary line, D, we arbitrary choose the first solution). This rule defines a unique way to decorate each fat rhomb.

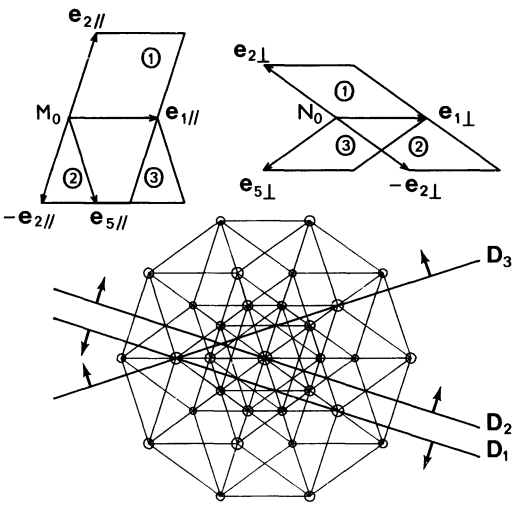

Fig. 11. — Above : rhombs number 1, 2 and 3 (left) whose common edge is M_0, $M_0 + \mathbf{e}_{1\parallel}$; and their corresponding rhombs in the perpendicular space $F_\perp$ (right). Below : projection onto the perpendicular space $F_\perp$ of the unit 5D-cube. The line D_i is the boundary line corresponding to rhomb number i : the arrows indicate the half-plane which is the locus of points N_0 such that the interior atom of rhomb number i is next to the edge M_0, $M_0 + \mathbf{e}_{1\parallel}$.

We now show that this procedure excludes the possibility to get two interior L atoms on each side of a common rhomb edge, i.e., they cannot be neighbours. Let for instance M_0 and $M_0 + \mathbf{e}_{1\parallel}$ be the extremities of the common edge. If one of the two rhombs, called rhomb 1, is constructed with M_0, $\mathbf{e}_{1\parallel}$ and $\mathbf{e}_{2\parallel}$ the other is constructed either with M_0, $\mathbf{e}_{1\parallel}$ and $-\mathbf{e}_{2\parallel}$ (rhomb 2) or with M_0, $\mathbf{e}_{1\parallel}$ and $\mathbf{e}_{5\parallel}$ (rhomb 3). The first situation never exists in the

tilings where $E_{\parallel}$ is defined as above, but it can occur in the periodic approximants. Let D_1, D_2 and D_3 be the boundary lines corresponding to the corner N_0 of the rhombs 1, 2 and 3 respectively (Fig. 11). Lines D_1 and D_2 are parallel. They divide $F_{\perp}$ in three regions in which, wherever N_0 lies, the interior L atoms are never next to the common edge at the same time.

Lines D_1 and D_3 divide $F_{\perp}$ in four regions. Only in the left quarter shown in figure 11 would both the interior L atoms of rhomb 1 and 3 be next to the corner M_0. But actually, this situation never occurs because, for these locations of N_0, the opposite corners would be outside of the unit cube projection onto $E_{\perp}$; i.e., they would correspond to points of the cubic lattice which should be outside of the strip. Therefore this quarter is a forbidden region for the local pattern formed by rhombs 1 and 3.

Thus decoration I always produces a close-packed structure as defined in section 2.

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