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Atomic size effects on the stability of the 2D Penrose pattern

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Abstract. The atomic size conditions for stabilising the 2D Penrose pattern were studied. The 2D Penrose patterns decorated by two kinds of Lennard–Jones particles according to Socolar and Steinhardt were relaxed by molecular dynamics. From structural analysis by Voronoi polygons and calculations of kinematical diffraction patterns, it was found that the atomic radius ratio (r_a) had to be in the range 0.5177–0.7900 to keep the 2D Penrose pattern topologically stable. Interstitial and substitutional FCC(111) solid solutions were obtained as the stable atomic arrangements of the systems with $r_a = 0.2071$ and 1.000, respectively. The other systems with $r_a = 0.3107$ –0.4660 and 0.8000–0.9000 showed the twelve-fold and ten-fold symmetry in the diffraction patterns, respectively, indicating the existence of the other kind of aperiodic structure and the multiply twinned structure, respectively.

1. Introduction

The quasicrystal which was found by Shechtman *et al* (1984) has been recognised as a new phase. However, its mechanical and thermal stability is not clear yet. The Al–X (where X is a transition metal) quasicrystalline phases were produced by the melt spinning method similar to that used in producing amorphous materials and their stability was investigated experimentally by DSC measurements (Kimura *et al* 1985, 1986), DTA measurements (Dunlap and Dini 1986), in situ x-ray diffraction (Wang *et al* 1986) and in situ TEM observations (Follstaedt and Knapp 1986a, b). From these experiments, it has been concluded that the quasicrystal is a metastable phase with a free energy intermediate between those of crystal and amorphous phases. Recently, however, the millimeter size grain of the single icosahedral phase was found to be produced by the conventional casting method (Dubost *et al* 1986, Gayle 1987), which suggests the existence of an intrinsically stable quasicrystalline phase. X-ray diffraction (Kortan *et al* 1987), EXAFS (Ma *et al* 1987) and superconductivity (Jim *et al* 1987) of these samples were also studied. Therefore it is of great interest to clarify the conditions under which the Penrose pattern composed of different kinds of atoms could be stable. In this paper, the Penrose tiling is considered as a model of the quasicrystal structure (Ogawa 1985). The mechanical stability of quasicrystal structures has not been investigated very much so far; the phenomenological consideration of Landau free energy (Alexander and

McTague 1978, Bak 1985, Mermin and Troian 1985, Jaric 1985, Nelson and Sachdev 1985, Hornreich and Shtrikman 1986, Rokhsar and Sethna 1986), the structural relaxation by the conjugate gradient method (Levine and Steinhardt 1986) or the Monte Carlo method (Widom *et al* 1987), the electronic band theory (Voisin and Pasturel 1987) and quasicrystal growth simulations (Minchau *et al* 1987). These works suggest the possibility of the existence of stable quasicrystals but the conditions for their stability are not yet evident. In the present study, the stability of the Penrose patterns decorated with two kinds of atoms was investigated by molecular dynamics. Since the atomic size factor is considered to be an essential factor in the stability of quasicrystal structures, the atomic radius ratio of small to large particles was the only variable parameter taken into account in the present simulations.

Contrary to our assumption, there is another possibility; that the anisotropic potential, which can be derived from the electronic states in the material, plays an essential role in forming the quasicrystal structure. The anisotropic effect of the interacting potential may be important in the aggregation of nucleated icosahedral clusters in forming the ordered structure with quasiperiodicity. We did not study the role of the anisotropy of the atomic interaction nor the aggregation process of icosahedral (pentagonal in the 2D case) clusters. Simulation of nucleation and aggregation of clusters in quasicrystal formation is the subsequent problem.

For simplicity of the structural analysis, the simulation was performed for 2D systems. Therefore it is not completely clear what the relationship between our results and the actual 3D quasicrystal structure is. At this stage, we should be content to infer the situation in the 3D case according to the following assumption: the 2D quasicrystal structure can be understood as the assembly of the pentagon clusters and the icosahedral clusters in the 3D quasicrystal play the same role as the pentagon clusters in the 2D quasicrystal. However, the small monatomic clusters prefer to form icosahedral clusters in the 3D space than in the 2D space, so the details of the quasicrystal formation process cannot be known until more realistic 3D models are studied. Our simulation method, presented here, can be applied to such more complicated systems.

2. Methods of calculation

The first problem is how to decorate the 2D Penrose pattern with the two kinds of atoms. Of the many ways of decoration proposed so far, the Socolar–Steinhardt type (Socolar and Steinhardt 1986) shown in figure 1 was chosen. In this atomic arrangement, smaller atoms (A) are always surrounded by larger atoms (B) so as to make its coordination number 5. The interaction potentials were assumed to be of the Lennard-Jones type,

$$\varphi(r) = 4\varepsilon((\sigma/r)^{12} - (\sigma/r)^6). \quad (1)$$

Three kinds of interaction, A–A, B–B and A–B should be considered for two-component systems. Here we focus upon the parameter σ which dominates the atomic radius and fixed the other parameter ε through the three types of interaction. Taking into account the interactions up to the fifth neighbour, σ_{AA} and σ_{BB} were determined, in order to reproduce the lattice constants of pure systems of A and B in the FCC(111) structures,

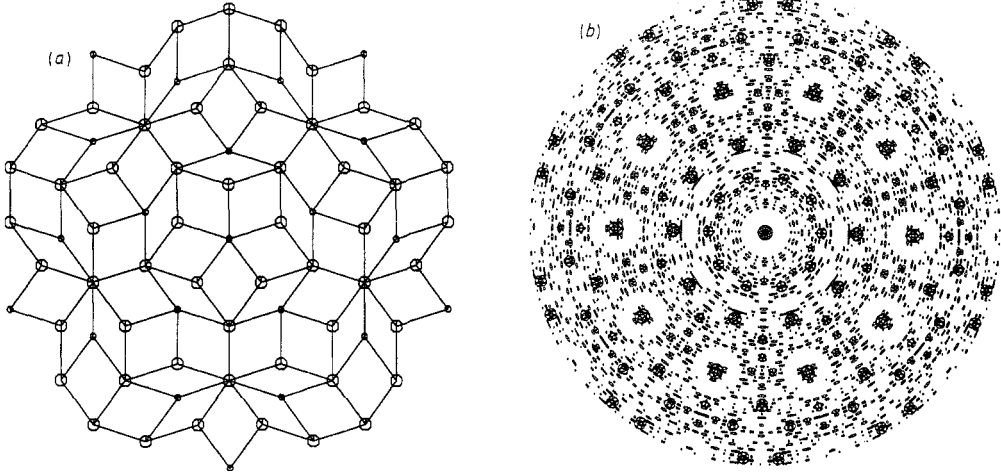


Figure 1. (a) Socolar–Steinhardt type decoration of the 2D Penrose pattern and (b) its diffraction pattern.

respectively. In this paper, we have taken the unit of distance as the lattice constant of B. Then σ_{BB} was determined as

$$\sigma_{BB} = 0.89933 \quad (2)$$

and is fixed throughout the present simulations. The only variable parameter σ_{AA} is expressed by the atomic radius ratio (r_a),

$$\sigma_{AA} = r_a \cdot \sigma_{BB}. \quad (3)$$

σ_{AB} was set simply by the following equation,

$$\sigma_{AB} = (\sigma_{AA} + \sigma_{BB})/2. \quad (4)$$

We determined the last parameter ε of the pure system B to reproduce the cohesive energy of aluminium, 3.39 (eV/atom),

$$\varepsilon = 1.0076 \text{ (eV)}. \quad (5)$$

We have also performed the whole simulation using several different values of ε and found that the simulated results such as stability conditions hardly depended on the parameter ε at all. If the locally densely packed structures are considered, the atomic radius ratio

$$r_{a5} = 1/\sin(\pi/5) - 1 = 0.7013 \quad (6)$$

gives the coordination number 5 and

$$r_{a4} = \sqrt{2} - 1 = 0.4142 \quad (7)$$

gives the coordination number 4. The 2D quasicrystal structure by Socolar–Steinhardt could be stable in the vicinity of r_{a5} in atomic radius ratio but elsewhere it would change into other stable phases such as crystal structures.

The 2D Penrose patterns composed of two kinds of atom with a variety of atomic ratios ($r_a = 0.2071\text{--}1.000$), were constructed and energetically relaxed by molecular

Table 1. The atomic radius ratios and the potential parameters for the simulated systems. $r_a = 0.4142, 0.7013$ and 1.000 are the appropriate values for sphere packings with coordination numbers 4, 5 and 6, respectively. The potential parameters for the B–B interaction, σ_{BB} and ϵ , were fixed as 0.89933 and 1.0076 eV, respectively.

r_a	σ_{AA}^6	r_a	σ_{AA}^6
0.2071	0.4175×10^{-4}	0.7013	0.6294×10^{-1}
0.3107	0.4756×10^{-3}	0.7900	0.1286
0.4142	0.2672×10^{-2}	0.8000	0.1387
0.4660	0.5417×10^{-2}	0.9000	0.2812
0.5177	0.1019×10^{-1}	1.0000	0.5291
0.5610	0.1650×10^{-1}		

dynamics. The relaxation time step was of the order of 10^{-15} s and the total potential and kinetic energies were monitored during the relaxations. We have studied the final atomistic structures by Voronoi polygon analysis and simulated their diffraction patterns by the kinematical theory,

$$I(\mathbf{q}) = \left| \sum_j f_j \exp(i\mathbf{q} \cdot \mathbf{r}_j) \right|^2 \quad (8)$$

where \mathbf{q} , \mathbf{r}_j and f_j are the wavenumber vector, the position and the atomic form factor of the j th atom, respectively. Here f_j has two kinds of values, f_a and f_b for atom A and atom B, respectively, and their ratio was assumed to be

$$f_a : f_b = 25 : 13 \quad (9)$$

which is the ratio of atomic numbers of manganese to aluminium. The system containing 76 atoms was mainly studied but in some cases a larger system with 476 atoms was also relaxed to check the effects of the boundary atoms on the simulated results.

3. Results and discussion

The atomic radius ratios and the potential parameters of the simulated systems are shown in table 1. Figure 2 shows the changes of atomic configurations and diffraction patterns for the systems with $r_a = 0.5610$. The system with $r_a = 0.5610$ was able to keep the structure with five-fold symmetry stable, although the tiles of the 2D Penrose pattern were fairly distorted. The diffraction patterns shown in figure 2(b) reflect this fact. Figure 3 shows the stable atomic configurations and their diffraction patterns for the systems with various atomic radius ratios; $r_a = 0.2071, 0.4142, 0.5177, 0.7013, 0.8000, 0.9000$ and 1.000 . The types of relaxed structure and the symmetry of their diffraction patterns are summarised in table 2. The system with $r_a = 0.2071$ corresponds to the interstitial solid solution of the FCC(111) structure and those with $r_a = 0.5177$ – 0.7900 the aperiodic structures with five-fold symmetry. The systems of $r_a = 0.8000$ – 0.9000 also gave the diffraction patterns with five-fold symmetry but the structures can be recognised as the multiply-twinned crystals from the atomic arrangements. The systems of $r_a = 0.3107$ – 0.4660 showed the twelve-fold symmetry in the diffraction patterns which can be related to the 2D quasilattice with both twelve-fold symmetry and self-similarity. This phase was found experimentally in the nickel–chromium fine particles (Ishimasa *et al* 1985). It

Table 2. The diagram of the relaxed structures with various atomic radius ratios.

r_a	Structure	Symmetry in the DP†
0.2071	FCC(111); interstitial solid solution	Six-fold symmetry
0.3107	Quasilattice with twelve-fold symmetry	Twelve-fold symmetry
0.4142		
0.4660	The 2D Penrose pattern	Ten-fold symmetry
0.5177		
0.5610		
0.7013		
0.7900	Multiply twinned crystal	Ten-fold symmetry‡
0.8000		
0.9000	FCC(111); substitutional solid solution	Six-fold symmetry
1.000		

† DP: diffraction pattern.

‡ Hardly noticeable.

should also be emphasised that the ternary fine particles of nickel–chromium–silicon show the eight-fold symmetry in the diffraction pattern (Wang *et al* 1987).

In the system of $r_a = 1.000$, i.e. the single particle system, the embryos of the FCC(111) crystal appeared immediately after relaxation and finally changed into a single crystal of FCC(111) structure. The detailed relaxation processes of this system, the structures and the phonon and electronic spectra have been reported in previous papers (Sasajima *et al* 1987, 1989).

The most favourable condition of atomic radius ratio for the stability of the 2D Penrose pattern was $r_a = 0.5610$ and that of the quasilattice with twelve-fold symmetry was $r_a = 0.4142$. These results suggest the existence of stable quasilattice structures with five-fold and twelve-fold symmetry when the two kinds of constituent atoms have the appropriate atomic radius ratio, $r_a = 0.5177$ – 0.7900 and 0.3107 – 0.4660 , respectively. The Voronoi polygons were calculated for each system. The examples of the Voronoi polygon partitions are shown in figure 4 and the fundamental properties are listed in table 3. It was confirmed that the 2D Penrose pattern did not show the structural change topologically if $r_a = 0.5177$ – 0.7013 . In the systems with $r_a = 0.7500$ – 0.7900 , the number of hexagons is much greater than in the Penrose pattern. Since the hexagons are almost pentagon shaped and the diffraction patterns showed ten-fold symmetry, these systems could be classified as an aperiodic structure with five-fold symmetry. Nevertheless it is open to question whether these structures were really ‘quasicrystals’ because they have neither the sharp peaks nor the self-similarity in the diffraction pattern. Although those structures were assumed to be assemblies of micro grains of quasicrystal phases, the irrational number ratio in the distances of the diffraction rings could not be found. This was probably due to the small system size or inappropriate ways of atomic decoration of the Penrose pattern. To check the effects of the system size, the Penrose pattern containing 476 atoms with $r_a = 0.5610$ was simulated. The atomic arrangement and its diffraction pattern are shown in figure 5. Compared to the 76 atom case, it was confirmed that the system size hardly changed the results. This is supported by the fact that the

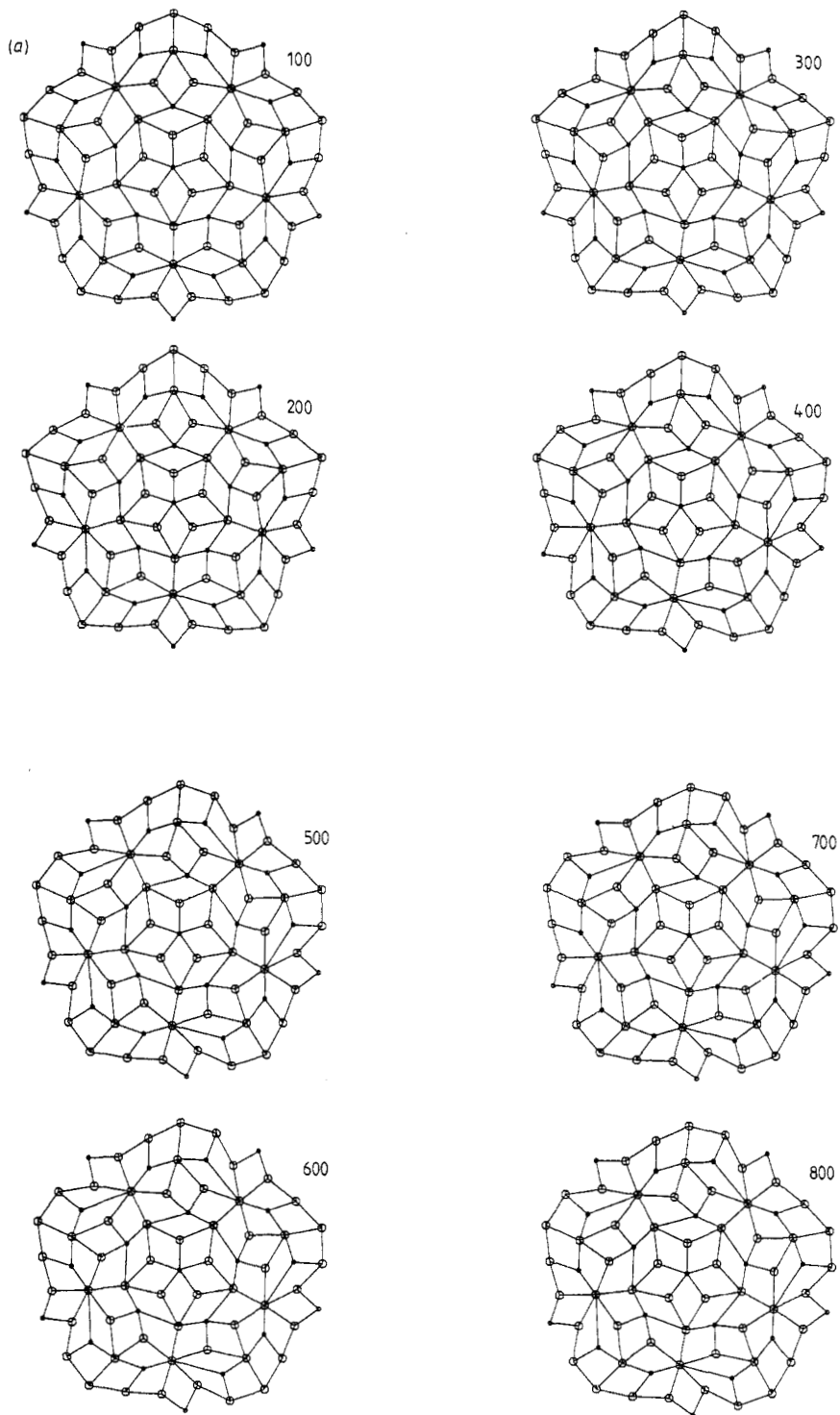
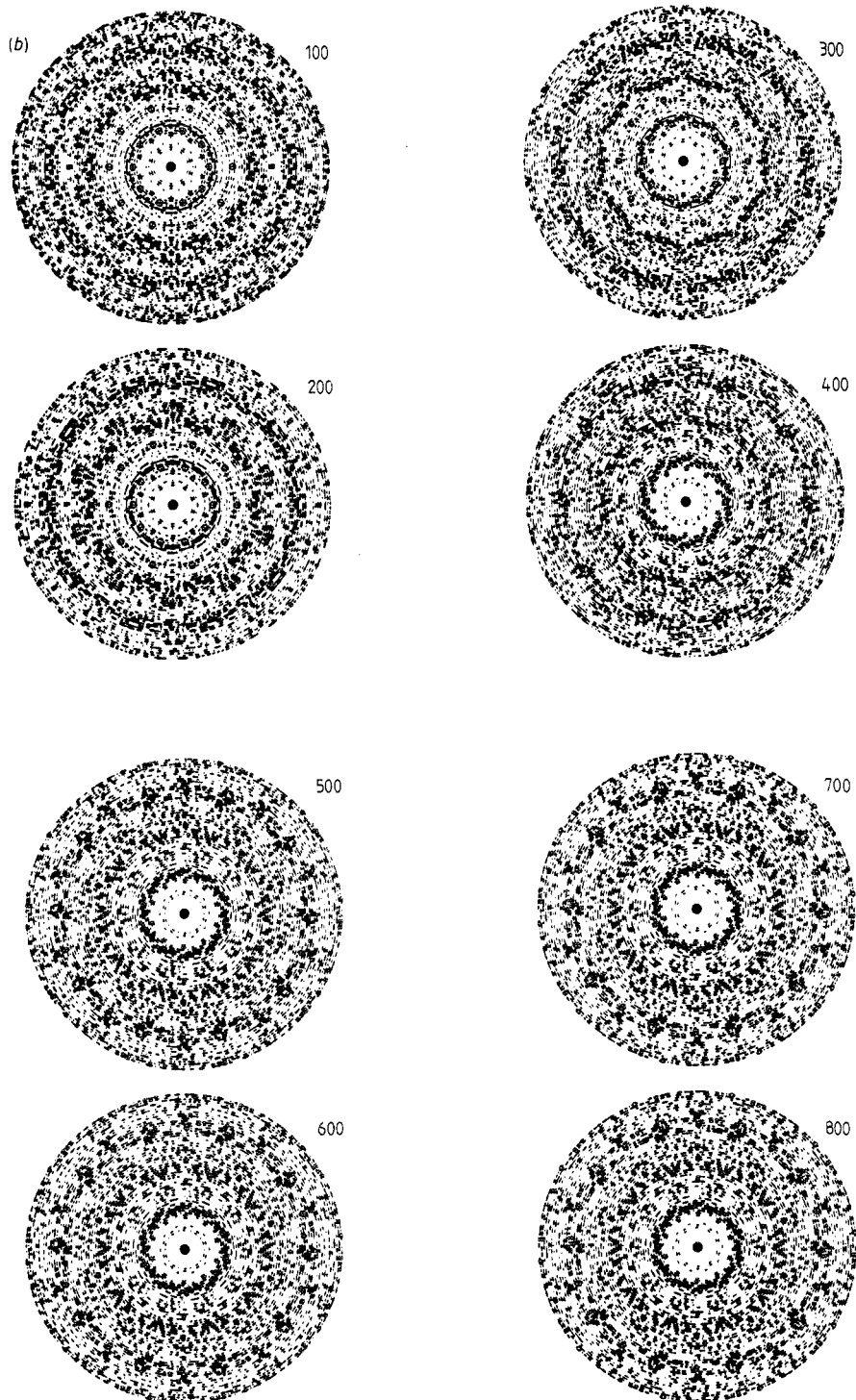


Figure 2. The changes of (a) the atomic configurations and (b) the diffraction patterns for the system of $r_a = 0.5610$. The inserted numbers represent the relaxation time steps.



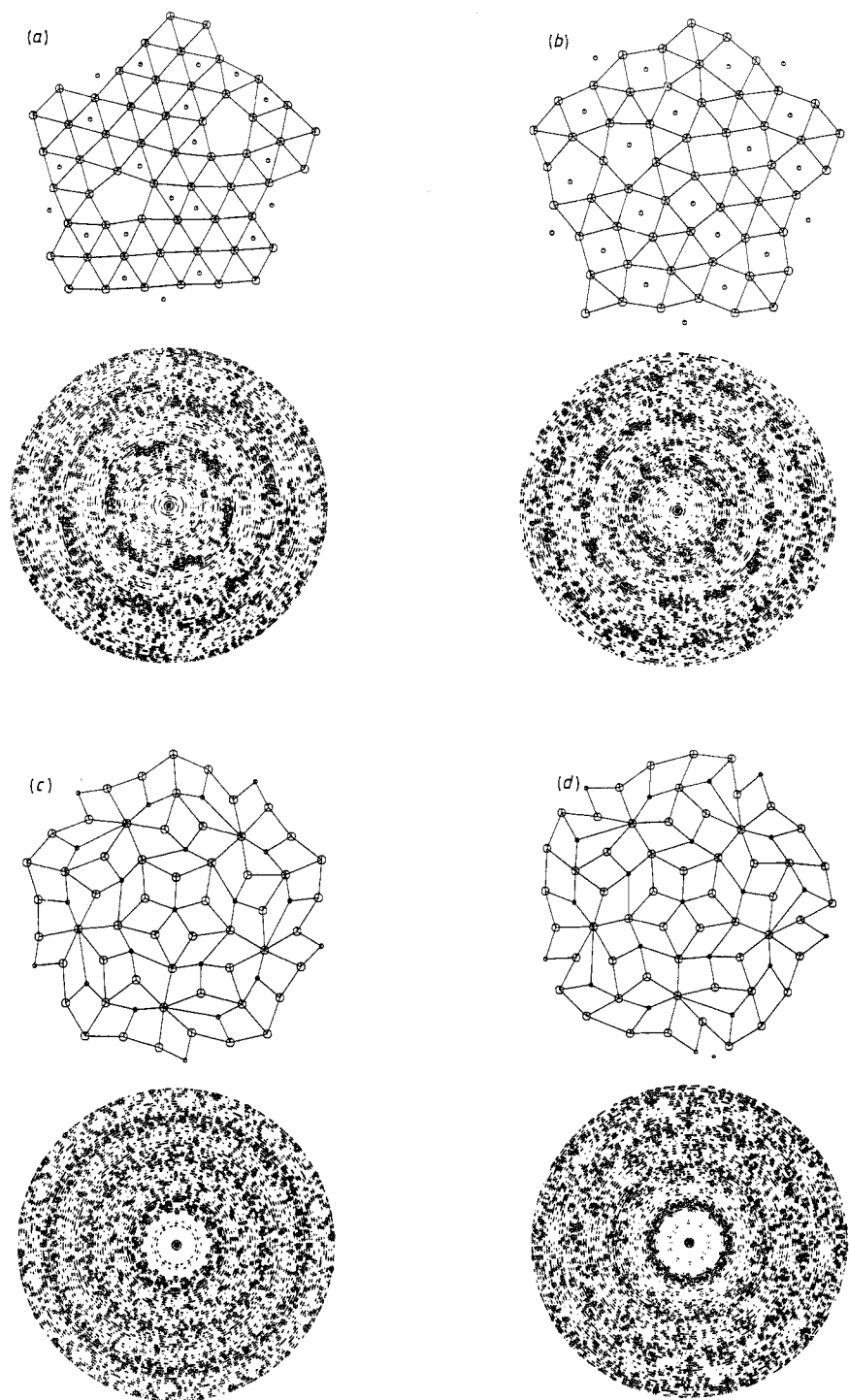
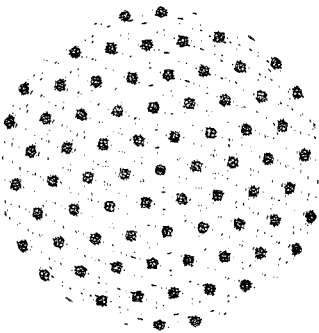
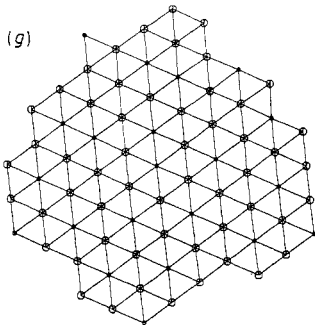
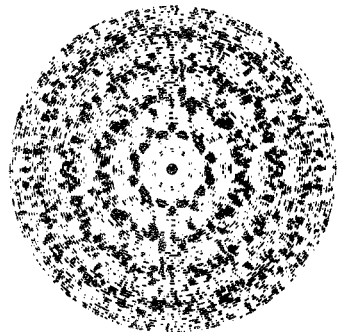
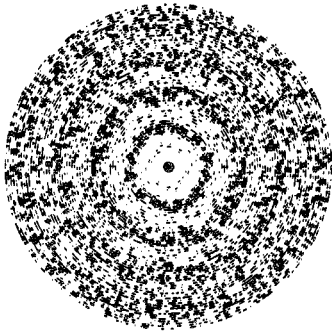
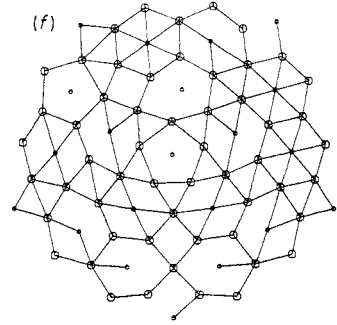
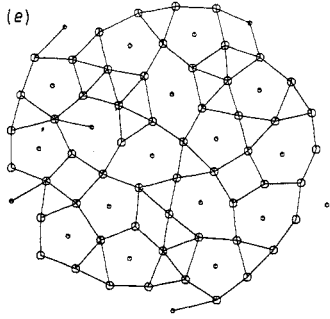


Figure 3. The relaxed atomic configurations and corresponding diffraction patterns for the systems of $r_a =$ (a) 0.2071; (b) 0.4142; (c) 0.5177; (d) 0.7013; (e) 0.8000; (f) 0.9000; (g) 1.000; for relaxation time step of 800.



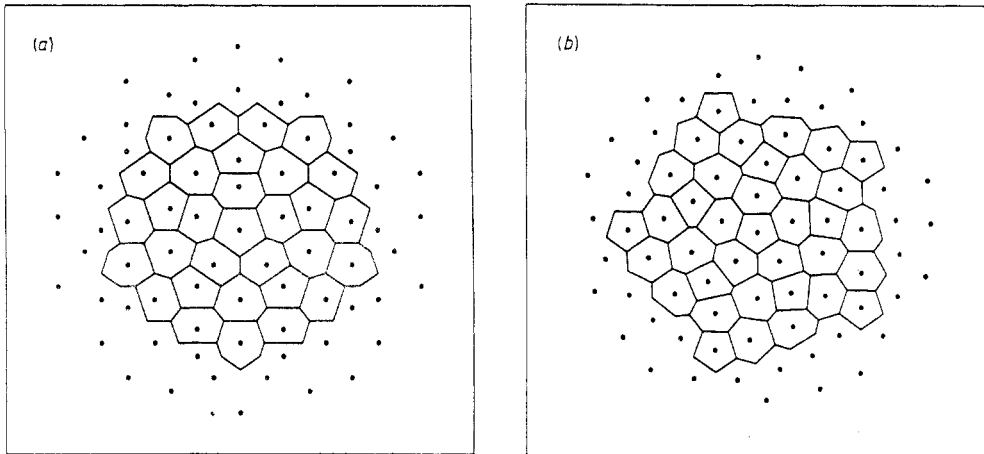


Figure 4. The Voronoi polygons of (a) the Penrose pattern and (b) the related structure of the system of $r_u = 0.5610$.

same structure in any region of the Penrose pattern can be found in the vicinity of that region. Contrary to the Penrose tiling model, the quasicrystal structure can be interpreted as the periodic structure with a large unit cell. Actually, the Penrose pattern is changed into the periodic pattern if its tiling is slightly changed. It is worth mentioning that the calculated Fourier transforms of these quasiperiodic and periodic patterns show almost the same results. The periodic structural model is not studied in our simulation. The calculated system with 76 atoms can be interpreted as the small nucleus formed in the melt at initial stage of quasicrystal formation. Thus its stability is at least a necessary condition to guarantee the stability of the 2D quasicrystal structure with the Socolar–Steinhardt type of decoration.

Table 3. The distribution of the Voronoi polygons for the relaxed structures.

Step	$\langle 3 \rangle$	$\langle 4 \rangle$	$\langle 5 \rangle$	$\langle 6 \rangle$	$\langle 7 \rangle$	$\langle 8 \rangle$
0	—	—	6	15	10	—
1.000	—	—	—	33	—	—
0.9000	—	—	2	29	1	—
0.8000	—	—	6	18	9	—
0.7900	—	—	2	23	6	—
0.7800	—	—	1	25	5	—
0.7700	—	—	1	25	5	—
0.7500	—	—	2	23	6	—
0.7013	—	—	9	15	10	—
0.5610	—	—	11	15	10	—
0.5177	—	—	11	15	10	—
0.4660	—	10	5	9	21	—
0.4142	—	10	2	12	7	—
0.3107	—	4	3	20	11	—
0.2071	—	6	4	11	10	—

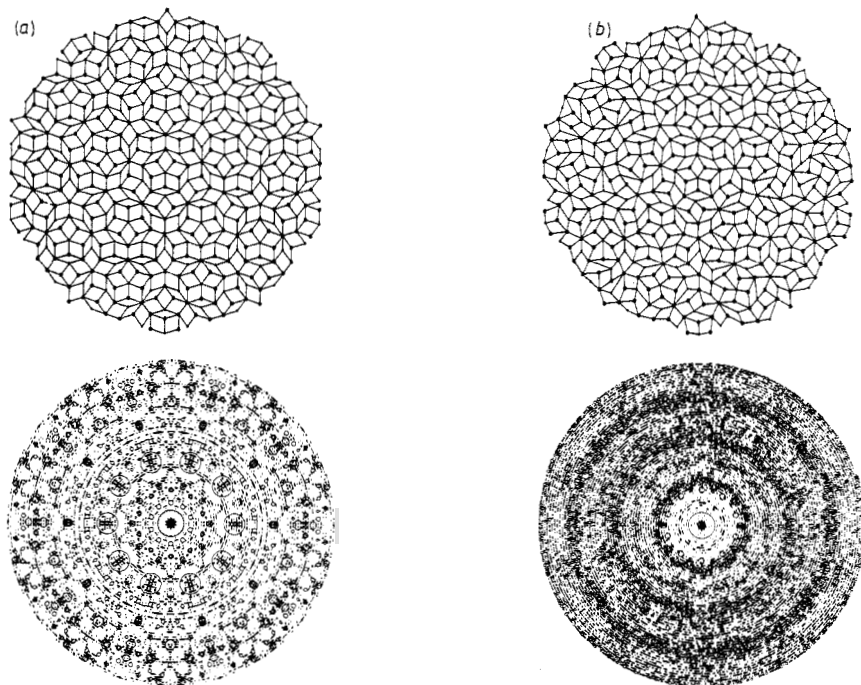


Figure 5. The atomic structures of the system of $r_a = 0.5610$ containing 476 atoms at (a) 0 step and (b) 800 step. Their diffraction patterns are shown below the structures.

To find the more stable quasicrystal structure, the other types of decoration of the Penrose pattern must be studied. From the viewpoint of the hard disc packings, the Henley type decoration (Henley 1986) is considered to be a good model for the stable quasicrystal. The results will be reported in a subsequent paper.

4. Conclusion

The stability of the 2D Penrose pattern in which the smaller atoms are always surrounded by the larger atoms with coordination number 5, i.e. the Steinhardt and Socolar model, were studied by molecular dynamics. The relaxed structures were analysed by Voronoi polygon partitions and using diffraction patterns. The range of values the atomic radius ratio needed to keep the quasilattice stable was revealed. The stable quasiperiodic structures with five-fold and twelve-fold symmetry could be realized if $r_a = 0.5177$ – 0.7900 and 0.3107 – 0.4660 , respectively. The final structures of the systems with $r_a = 0.2071$ and 1.000 were the interstitial and substitutional solid solutions of FCC(111), respectively. The systems with $r_a = 0.8000$ – 0.9000 were changed into the multiply twinned crystals such that they showed ten-fold symmetry in the diffraction patterns. The Penrose tiles of the system with $r_a = 0.5610$ was distorted least among the simulated systems.

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