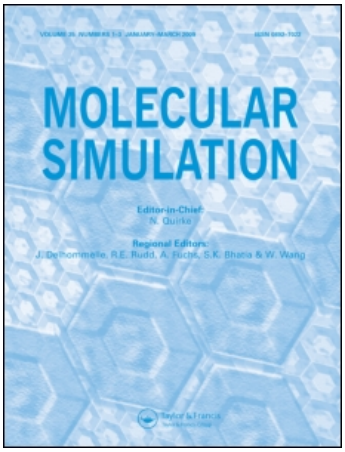

Molecular Simulation
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713644482

Computer Simulation of the Film Growth Process on the Two-Dimensional Penrose Pattern

To cite this Article (1994) 'Computer Simulation of the Film Growth Process on the Two-Dimensional Penrose Pattern', Molecular Simulation, 12: 3, 407 — 420
To link to this Article: DOI: 10.1080/08927029408023048
URL: http://dx.doi.org/10.1080/08927029408023048

Molecular Simulation, 1994, Vol. 12(3-6), pp. 407-420
Reprints available directly from the publisher
Photocopying permitted by license only

COMPUTER SIMULATION OF THE FILM GROWTH PROCESS ON THE TWO-DIMENSIONAL PENROSE PATTERN

YASUSHI SASAJIMA, HIDEKI TANAKA, KATSUMI ADACHI, MINORU ICHIMURA, MASANORI ITABA and SATORU OZAWA

Faculty of Engineering, Ibaraki University, 4-12-1 Nakanarusawa-cho, Hitachi Ibaraki 316, Japan

(Received January 1993, accepted May 1993)

We performed computer simulation of the film growth process on the two-dimensional (2D) Penrose pattern, which is considered a typical structural model of quasicrystal. The atomistic structure of the deposited atoms was obtained as a function of time under various conditions of atomic binding energy, temperature and deposition rate. The Monte Carlo method based upon the solid-on-solid model was utilized for the present calculation. We found a geometrical restriction on the growth front of grains in the 2D Penrose pattern, which is also expected for the growth of an actual quasicrystal.

KEY WORDS: Monte Carlo method, Penrose pattern, quasicrystal, solid-on-solid model, film growth process

INTRODUCTION

The diffraction pattern of crystals clearly shows translational symmetry, because of the periodicity which is one of the important characteristics of crystal structure. On the other hand, in the middle of the nineteenth century, it was proven that fivefold symmetry is inconsistent with translational symmetry and therefore it cannot appear in any crystal. In other words, no condensed phase could have both fivefold symmetry and long-range order. A new material with these properties, however, was discovered in 1984 [1] and was called "quasicrystal"; Shechtman *et al.* found that the electron diffraction pattern of a rapidly quenched Al-Mn alloy shows sharp Laue spots with icosahedral symmetry. This finding proved the existence of a new phase with long-range order and without translational symmetry. The atomic structure and properties of the new phase have been extensively studied because of its uniqueness [2]. After the discovery of Al-Mn quasicrystal, a variety of alloy systems were experimentally shown to have quasicrystalline states as a metastable or stable phase [3]. At the same time, the fundamental question "Why do quasicrystals appear in nature?" has been considered from many points of view. Many structural models of quasicrystal have been proposed so far. Among them, the quasilattice is generally accepted as the framework of quasicrystal [4]. The Penrose lattice is a typical example of a quasilattice [5]. It has fivefold symmetry and long-range order. In addition, it can be easily produced mathematically by means of a dual lattice [6] or projection method [7]. However, a

complete explanation of the growth process of quasicrystal has not yet been attained. In this work, we studied the film growth process on the 2D Penrose lattice, which is considered a typical structural model of quasicrystal. The purpose of the present simulation is to clarify the nucleation and growth process of the quasicrystal. It is desirable to reveal the origin of creation of quasicrystal without any artificial restriction. One possible goal is to simulate growth process of quasicrystal by molecular dynamics method, however, we found it is rather difficult because the simulated samples contained a lot of defects, strains and segregation. Therefore it is a good tactics to simulate the nucleation and growth process on a quasi-periodic substrate in order to get valuable information on the origin of quasicrystal.

METHODS OF CALCULATION

In this paper, the nucleation and growth process on a two-dimensional quasicrystal substrate is simulated and the effects of deposition rate, substrate temperature and potential parameters on the film structure are studied. A Penrose lattice was constructed as a simple model of a quasicrystal substrate and the growth process on the quasiperiodic lattice was simulated by means of the canonical Monte Carlo method [8]. In this model, a particle is only allowed to occupy the lattice sites as in the solid-on-solid model [9]; however, in the present simulation, the lattice sites constitute apexes of two-dimensional Penrose tiles which exhibit quasi-periodicity.

TWO-DIMENSIONAL PENROSE PATTERN

The two-dimensional (2D) Penrose pattern was composed of two rhombuses, shown in Figures 1(a) and 1(b). This pattern is constructed by filling the 2D space while conserving fivefold symmetry. When a new rhombus is added to the surface, it is required to satisfy the "matching rule": the arrows of the Penrose tiles shown in Figures 1(a) and 1(b) should coincide. Figure 1(c) represents the constructed Penrose lattice. This procedure had been developed by Penrose [10] before the discovery of quasicrystal. The characteristics of the 2D Penrose pattern are as follows.

(1) Self-similarity
(2) Aperiodicity
(3) Pentagonal symmetry

In Figure 2, the tiles along the directions of the fivefold symmetry are shaded. The pentagonal symmetry of the Penrose pattern can be easily seen from this figure. Pentagonal symmetry should be distinguished from fivefold symmetry; the fivefold symmetry shows perfect coincidence of structure with 72° rotation to origin, but pentagonal symmetry does not. Pentagonal symmetry means that the structure has many rhombuses which lie parallel to the fivefold directions. Pentagonal symmetry causes sharp Laue spots with fivefold symmetry.

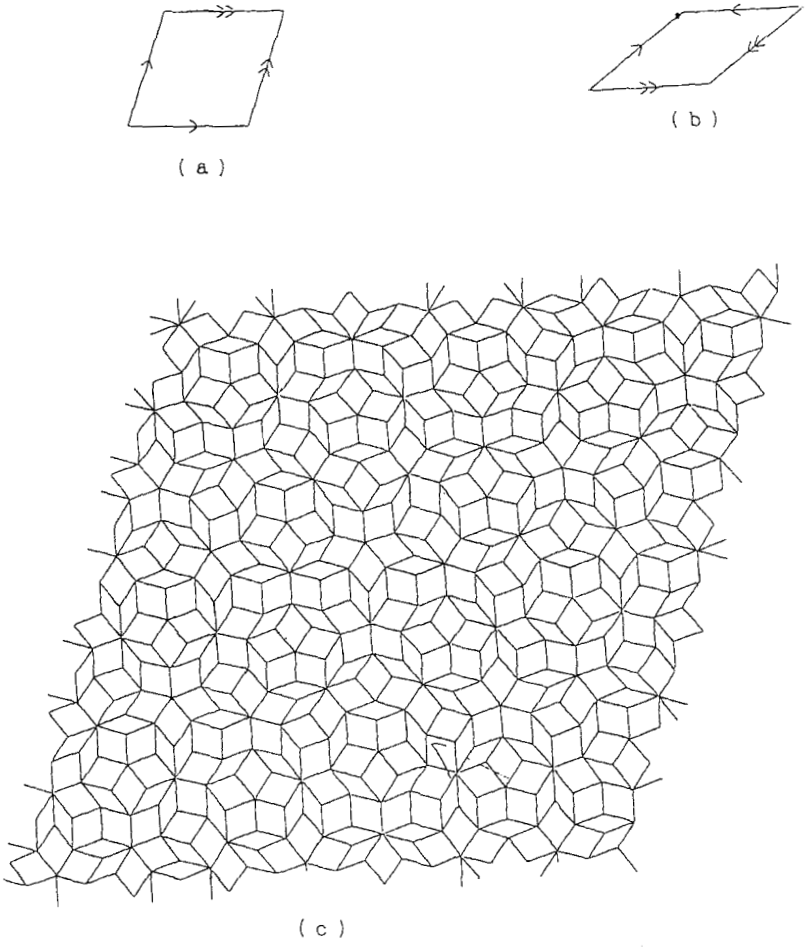

Figure 1 Penrose pattern.
(a) Fat rhombic unit of Penrose pattern.
(b) Skinny rhombic unit of Penrose pattern.
(c) Periodic Penrose lattice.

SIMULATION PROCEDURE

Deposition process of monoatomic system on the PPL

The simulation procedure of the film growth process on the quasicrystal surface is summarized as follows.

(1) Construct the 2D Penrose lattice with 521 sites. (We used periodic Penrose lattice (PPL) [11] in order to avoid the surface effect.)
(2) Deposit an atom onto one of the Penrose lattice sites. The lattice site is selected at random using a random number generator.

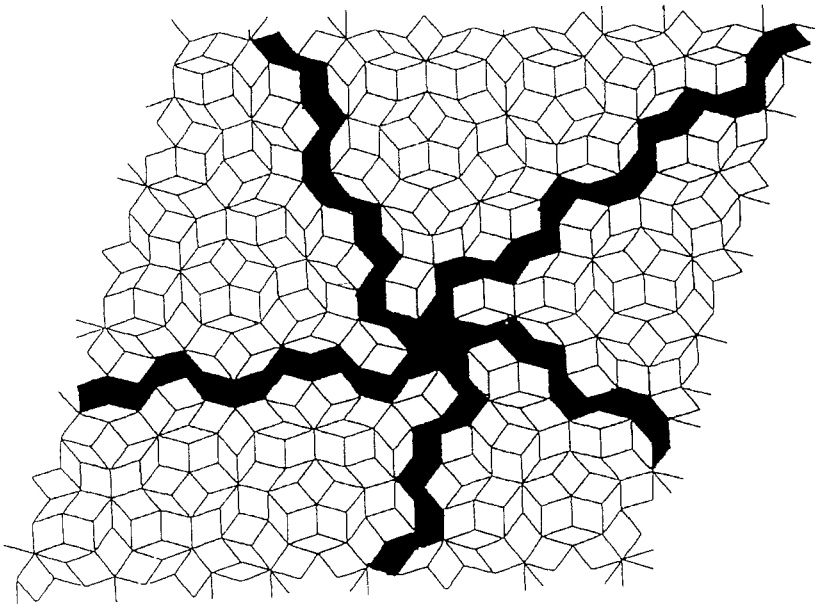

Figure 2 Periodic Penrose lattice showing the pentagonal symmetry.

(3) Move the deposited atoms on the lattice sites during a certain Monte Carlo step (MCS), where 1MCS is a computational unit of time used in Monte Carlo simulations. The migration of deposited atoms is determined stochastically by means of the Monte Carlo method at a constant temperature (i.e., canonical Monte Carlo method).
(4) Go back to (2) and repeat the steps until the number of atoms reaches 100.

The acceptance ratio of the migration of a particle was determined by Boltzmann's factor, $\exp(-\Delta E/k_B T)$, where ΔE is the change of the sum of the bond energy, k_B Boltzmann's constant and T the system temperature. The bond energy of a certain particle is calculated as the number of its nearest neighbors. Bond and diffusion pass of particles is restricted to the edges of rhombuses. The following should be noted concerning this model.

(1) Particles exist only on the sites of PPL.
(2) The present simulation does not include the interaction between film and substrate atoms. (The substrate structure still affects the migration of deposited atoms geometrically, i.e., diffusion pass of the particles is restricted to the edges of rhombuses.)
(3) The activation energy for thermal diffusion of a free particle is set to be 0 for computational efficiency.

A periodic boundary condition is imposed upon the PPL with 521 sites. We applied the same calculation to a PPL with 1351 sites, and confirmed that the finite size of the system does not significantly affect the results.

Equilibrium shape of grains on the PPL

In the second part of this paper, the equilibrium form of the nucleated grains on the PPL is also investigated. The simulation procedure is:

(1) Construct the 2D Penrose lattice with 1351 sites.
A grain composed of about 300–500 atoms is generated on the PPL. The initial shapes of the grains are square, rectangle and disk.
(2) Move the deposited atoms on the lattice sites for 120000 Monte Carlo steps (MCS), varying the system temperature.

Deposition process of binary Lennard-Jones system on the PPL

The last part of this paper is the simulation of formation of quasicrystal structure composed of binary Lennard-Jones system. We assumed Lennard-Jones potential as the interaction between atoms and used the potential parameter values proposed by Strandburg *et al.* [12] to produce stable quasicrystalline structure. Quasicrystal growth process has been simulated as creation and aggregation of atoms on the PPL, with the use of Monte Calro method varing the system temperature. The outline of the calculation procedure is the following way.

(1) Construct the periodic two-dimensional Penrose lattice composed of 1364 sites.
(2) Create a new atom every 100 MC steps up to the total number of 1200 atoms.

We determined the site to site movement of every atom proportional to the Boltzmann's factor $\exp(-\Delta E/k_B T)$, where ΔE is the difference of potential energy between the states before and after the trial movement. The location of deposited atoms is restricted to the lattice sites of the PPL. The interaction energy between constituent atoms was calculated assuming a simple form of Lennard-Jones potential in order to investigate the essential influence of atomic radius ratio and bond energies. In the simulation of the mono-atomic system, the system energy is evaluated by the number of bonds, while in that of the binary system, the system energy is calculated with the use of the Lennard-Jones potential. In the present simulation, the cut off of the potential was set to be the length of the edge of the Penrose tile, therefore short range interaction was assumed. In addition, in the present model the particles sit on the apexes of the Penrose tiles as similar to the SOS model. The above condition suggests that the obtained results will not be affected significantly by the specific potential function. Therefore we can conclude that the essential factors for the stability of the binary quasicrystal system are the strength of the potential (i.e., the bond energy) and the size of the particle. It has been proven that the quasicrystal structure proposed by Strandburg *et al.* is energetically metastable states, so we used the Strandburg's values of σ's and ε's. Table I shows the potential parameters at the present study. It should be noted that the equilibrium distance between A-B atoms is taken as the unit of distance, therefore the values as σ's are $2^{-1/6}$ times of the original values. Figure 3 shows the local structures of the two-component Lennard-Jones system of which potential values are tuned by Strandburg *et al.* They are also expected to be formed in quasicrystal structure. In other words, the parameter of Lennard-Jones potential has been determined to make the local structures shown in Figure 3 stable. On the two sorts of rhombus of Penrose pattern, the two kinds of atoms can be decorated

Table 1 Lennard-Jones potential parameters proposed by Strandburg *et al.* [12].

Type of bonding	σ	ε	*Equilibrium atomic distance*
A-A	1.0473	0.5	1.175
B-B	0.8909	1.0	1.0
C-C	0.5506	0.5	0.618

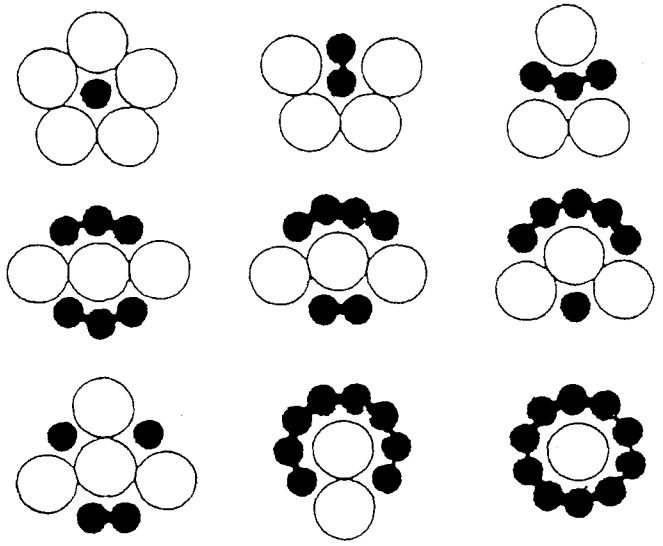

Figure 3 Local structure of the two-component Lennard-Jones system with the potential parameters proposed by Strandburg *et al.*

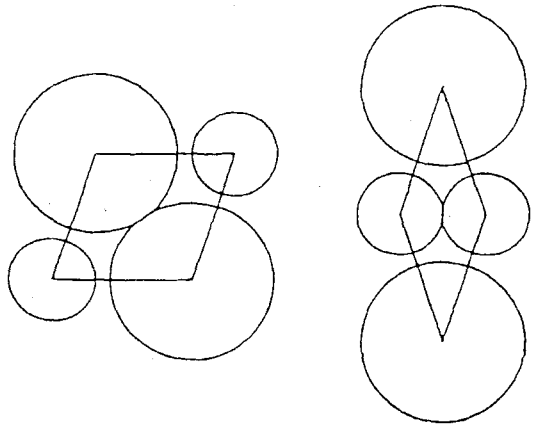

Figure 4 The atomic decoration of the two kinds of Penrose tiles.

as shown in Figure 4; On a fat rhombus, large atoms sit on the apexes of large angles and small atoms on the rest apexes, while on a skinny rhombus, vice versa. The two-dimensional Penrose pattern can be decorated by two kinds of atoms according to this rule. As the unit of temperature, we took a dimensionless parameter T^* which is calculated as

$$T^* = k_B T/\varepsilon_{AB}, \tag{1}$$

where ε_{AB} is the potential strength between A and B atoms. For convenience the results are presented as a function of TR which is the inverse of T^*.

RESULTS AND DISCUSSION

Deposition process of monoatomic system on the PPL

Three parameters, substrate temperature T, bond energy between film atoms E_{ff} and deposition rate J, are varied in the present calculation. The first two parameters appear in the following form,

$$\phi = E_{ff}/k_B T, \tag{2}$$

where ϕ is a dimensionless parameter. We carried out computer experiments of the deposition process of 100 film atoms on the PPL under the condition that

$$J = 2 \times 10^{-6} \sim 2 \times 10^{-3} (atoms/site \times MCS) \tag{3}$$

$$\phi = -1.0 \sim -9.0.$$

The minus sign in ϕ indicates the attractive interaction of the bonding between atoms. When J increases, a new particle arrives on the substrate before the previously deposited atoms are sufficiently relaxed. The smaller the value of ϕ, the higher the system temperature or the weaker the bond energy. Therefore $\phi = -1.0$ in the highest temperature case and $\phi = -9.0$ in the lowest temperature case. The high-temperature condition corresponds to the case of weak bond energy, as mentioned above. Let us discuss first the effect of the parameter ϕ on the film structure. Keeping rate J constant, the high-temperature (or weak-bond) condition converts the 2D system into a gaseous phase, the low-temperature (or strong bond) condition, a solidlike phase, and the intermediate temperature condition, a liquidlike phase. Figure 5 shows the relaxed atomic structures for (a) solidlike film ($\phi = -9.0$), (b) liquidlike film ($\phi = -3.0$) and (c) gaslike film ($\phi = -1.0$). In the liquidlike film, it was observed that a coalescent nucleus moves on the PPL. Secondly, the dependence of the film structure on the J can be stated as follows. At a high J, the shape of nuclei is not spherical, but has dendritic arms. In particular, in the low-temperature region, DLA (diffusion-limited aggregation) takes place. At a low J, the shape of nuclei is spherical, and some facetlike surfaces are observed. Figure 6 shows the structure of the deposited atoms for different J, the values of which are $2 \times 10^{-6} - 2 \times 10^{-3}$. The shape of nuclei seems to be determined by two competing effects. One is a surface-roughening effect due to deposition of atoms, and the other is a smoothing effect due to diffusion of peripheral atoms of nuclei. This suggests that the mobility of peripheral atoms of nuclei is strongly restricted by the PPL geometry [13]; therefore, the nucleation process and mobility of atoms on the PPL should be different from those on the

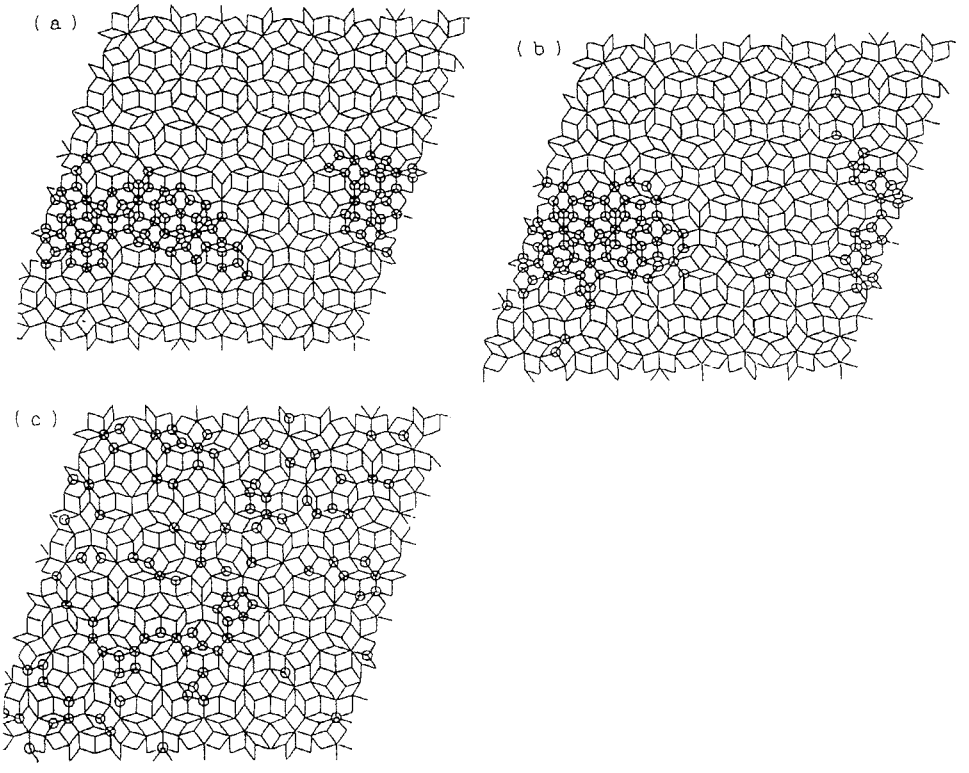

Figure 5 Relaxed atomic structures ($J = 2 \times 10^{-6}$).
(a) Solidlike film ($\phi = -9.0$).
(b) Liquidlike film ($\phi = -3.0$).
(c) Gaslike film ($\phi = -1.0$).

periodic lattice. The parameters J and ϕ are crucial for film growth on the PPL. Table II summarizes the present results; the symbol G represents gaslike film, L liquidlike film, S solidlike film, and D DLA-like film. The authors previously calculated the vacuum deposition process on a 1D single-crystalline substrate by means of the MC method and found that the deposited film shows crystalline growth, fractal-like growth or liquidlike behavior according to the experimental conditions [14]. The order estimation of the boundaries of J and ϕ between DLA-like, solidlike, liquidlike and gaslike films can be considered the same between 1D and 2D regular lattice systems, because the deposition process on a 1D regular lattice is a simplified model of that on one of the facets of a growing 2D grain. For the boundary state between solidlike film and DLA-like film in Figure l(b) in reference 14, the critical rate J_c and parameter ϕ can be estimated as $J_c \fallingdotseq 10^{-3}$ and $\phi \fallingdotseq -16$, while for the Penrose lattice, we can estimate $J_c \fallingdotseq 10^{-7}$ for the same parameter $\phi = -16$ from data extrapolation in Table II. It should be noted that the value of J_c is at least 10^3 times lower than that for the regular lattice. Therefore, we can conclude that the boundaries between the states of the films in

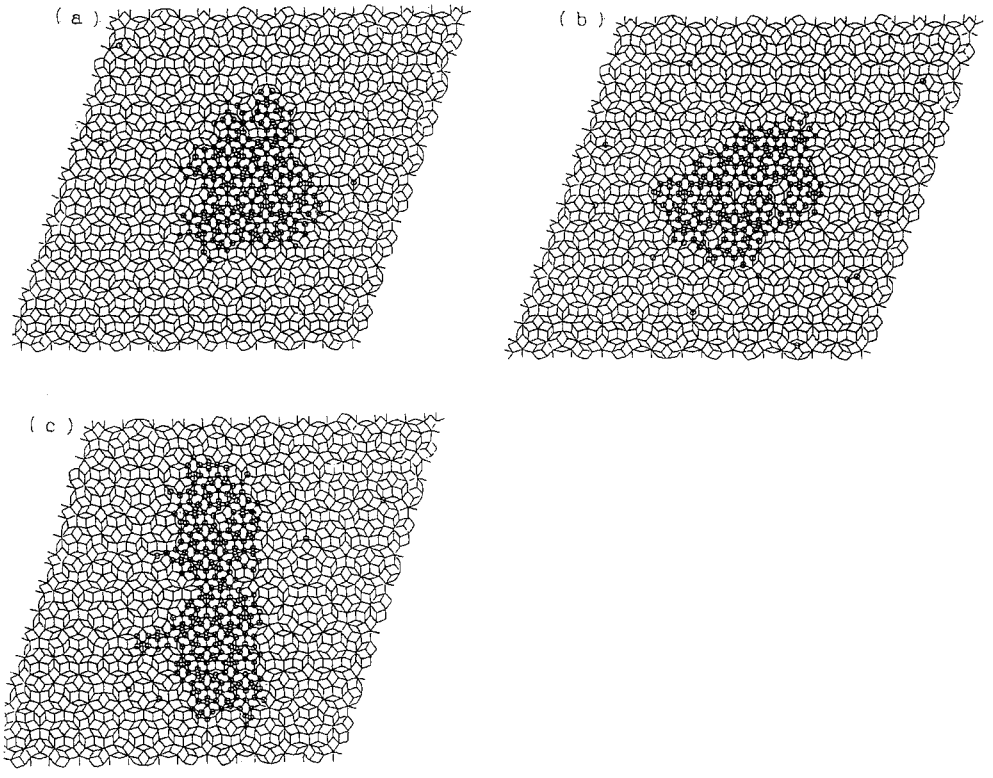

Figure 6 Relaxed atomic structures ($\phi = -5$).
(a) Solidlike film ($J = 2 \times 10^{-6}$).
(b) DLA-like film ($J = 2 \times 10^{-3}$).

Table 2 Dependence of film growth on J and ϕ: S: soloid like film, G: gaslike film, L: liquid like film and D: DLA-like film.

J \ ϕ	*−9*	*−7*	*−5*	*−3*	*−2*	*−1*
10^{-6}	S	S	S	L	G	G
10^{-5}	D	S/D	S	L	G	G
10^{-4}	D	D	D	L	G	G
10^{-3}	D	D	D	L	G	G

Table II will be shifted to the lower-left side because of the difference in mobilities of the deposited particles between the quasilattice and the regular lattice.

Equilibrium shape of grains on the PPL

The equilibrium shapes of the nucleated grains are observed to have corrugated facetlike surfaces for all the square-, rectangular- and disk-shaped grains under

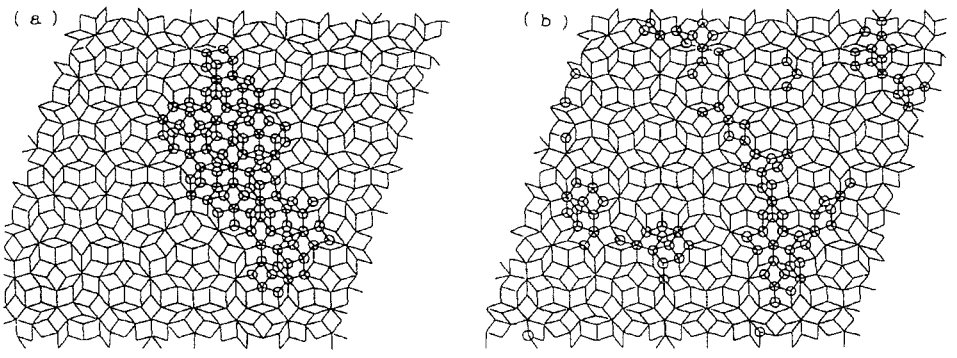

Figure 7 Relaxed atomic structures ($\phi = -3$) of the nuclei of which initial shapes were
(a) disk,
(b) square and
(c) rectangle.

$\phi = -3$. On the 2D regular lattice, nucleated grains are usually spherical so as to make surface energy minimum, while on the PPL, the mobility of the deposited particles is strictly limited by the geometry of the structure. Therefore, the initial structure strongly affects the results of our experiments. Figure 7 shows the relaxed atomic structures for the three types of grains.

Deposition process of binary Lennard-Jones system on the PPL

Figures 8 and 9 represent the atomic configurations at 60000 MCS (Monte Carlo steps) and 120000 MCS for TR = 1.75 and TR = 3.75, respectively. Table III shows the total number of the two kinds of rhombuses which forms local structures of quasicrystals. At TR = 1.75, which is the condition that the bonding energy of atomic interaction is relatively weak or system temperature is relatively high, atomic diffusion occurs easily and consequently, the deposited particles spread over the Penrose lattice at 60000 MCS as shown in Figure 8(a). As a result, local structure of quasicrystals is formed everywhere on the Penrose pattern in the final structure at 120000 MCS, however, segregation of the same kinds of atoms is more frequently occurs than formation of the local structure of quasicrystal, therefore we conclude that formation of quasicrystals cannot be observed under this condition. While at TR = 3.75, which is the condition that the strength of bond energies is relatively strong or system temperature is relatively low, the deposited atoms began to aggregate to form stable nuclei at 60000 MCS (see Figure 9(a)), and the nuclei continued to grow as increasing the coverage of the Penrose lattice. Therefore, in the final structure at 120000 MCS, it can be seen that clusters grew to form structure of quasicrystals more preferably than the former condition. However, segregation still exist at any place, then the perfect structure of quasicrystals was not be produced. Nevertheless, as can be seen from Table III the number of the rhombus which are the unit structure of quasicrystals increased with increasing TR. This fact shows that the structure of stable quasicrystals can be preferably formed as the deposited atoms are allowed to move and form stable clusters at relatively low

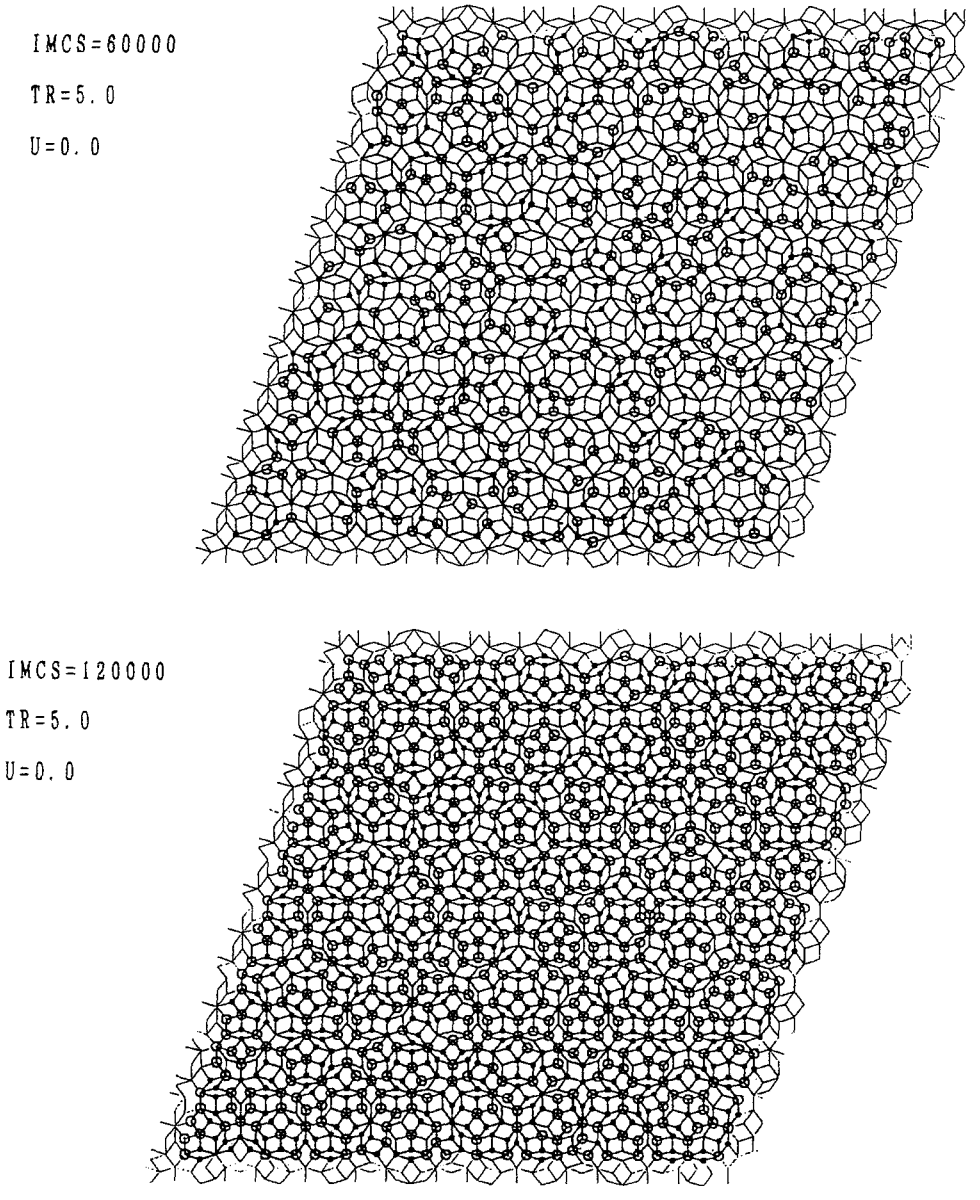

Figure 8 The atomic configuration of the binary Lennard-Jones system at (a) 60000 and (b) 120000 MCS. The inverse of the dimensionless system temperature (TR) is 1.75.

temperature condition. At the present study, the final coverage is more than 90% and deposition rate is rather high (7.33×10^{-6} atom/(site $\times$ MCS)), therefore formation of the stable clusters is prevented and segregation of the same kind of atoms preferably occurs.

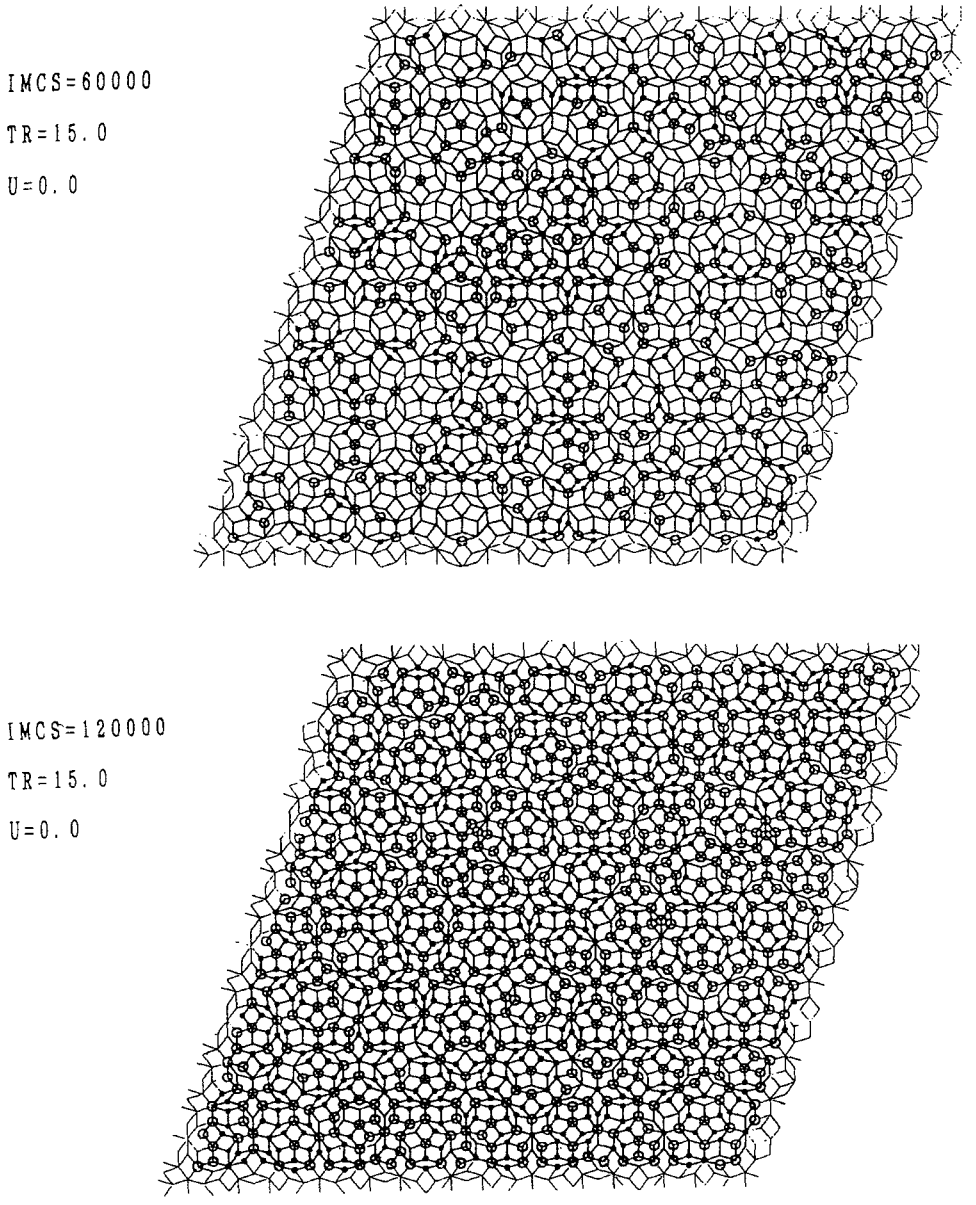

Figure 9 The atomic configuration of the binary Lennard-Jones system at (a) 60000 and (b) 120000 MCS. The inverse of the dimensionless system temperature (TR) is 3.75.

Table 3 The number of the produced Penrose tiles in the two component Lennard-Jones system.

Step / *TR*	*60000*	*120000*
1.25	13	244
3.75	64	337

CONCLUSIONS

Our simulation suggests that the mobility of peripheral atoms of nuclei on the PPL is strongly dependent upon the geometry of the substrate, compared to the case of the 2D regular lattice. The relaxed atomic structures show the strong influence of the shape of nuclei generated at early stages of the deposition process, and the surfaces of nuclei show wavy facets after relaxation. We conclude that the geometrical effect of PPL, i.e., aperiodicity, plays a crucial role in the growth of quasicrystal. This film growth process can generate quasicrystal more easily than the melt quenching or casting methods, because of the free particle diffusion on surfaces with fivefold symmetry. In the high-temperature region, particles move with high mobility and create the "wavy" surface. On the other hand, in the low-temperature region, a newly arriving particle attached to nuclei cannot move further; therefore DLA occurs. All peripheral atoms of nuclei on the PPL are strongly dependent upon the geometry of the aperiodic substrate. The present study has shown that realistic simulation of formation and growth process of quasicrystals is possible theoretically. The subsequent study will clarify the influence of system temperature, deposition rate and interaction potential more in detail and get clear perspective of the origin of the formation of quasicrystal structure.

Acknowledgements

The authors would like to acknowledge The Ministry of Education, Science and Culture for financial support under a Grant-in Aid for Scientific Research (Grant No. 63550471). The simulation was performed by HITAC 660H at the Information Processing Center of Ibaraki University and by HITAC 680H at the Computer Center of Tokyo University. Mr K. Sakayori and Mr H. Uemura are also to be acknowledged for their great help in developing computer program and performing preliminary calculations for the present study.

References

[1] D. Shechtman, I. Blech, D. Gratias and J.W. Cahn, "Metallic Phase with Long-Ranged Orientational Order and No Translational Symmetry", *Phys. Rev. Lett.*, **53**, 1951 (1984).
[2] P.J. Steinhardt and S. Ostlund, "The Physics of Quasicrystals", (World Scientific, Singapore, 1987).
[3] M.J. Yacaman, D. Romeu, V. Castano and A. Gomez, "Quasicrystals and Incommensurate Structures in Condensed Matter", (World Scientific, Singapore, 1990).
[4] D. Levine and P.J. Steinhardt, "Quasicrystals; A New Class of Ordered Structures", *Phys. Rev. Lett.*, **53**, 2477 (1984).
[5] A.L. Mackay, "Crystallography and the Penrose Pattern", *Physica A114*, 609 (1982).

[6] J.E.S. Socolar and P.J. Steinhardt, "Quasicrystals II: Unit-cell Cpnfigrations", *Phys. Rev. B*, **34**, 617 (1986).
[7] V. Elser, "The Diffraction Pattern of Projected Structures", *Acta Cryst. A*, **42**, 96 (1986).
[8] D.W. Heermann, "Computer Simulation Methods in Theoretical Physics." (Springer-Verlag, Berlin, 1990), 2nd ed, chapter 4.
[9] S. Ozawa and Y. Sasajima, "Computer experiments of initial growth kinetics of vacuum-deposited thin films", *Vacuum*, **41**, 1110 (1990).
[10] R. Penrose, "The Role of Aesthetics in Pure and Applied Mathematical Reseach", *Bull. Inst. Math. Appl.*, **10**, 266 (1974).
[11] M. Krajci and T. Fujiwara, "Strictly localized eigenstates on a three-dimensional Penrose lattice", *Phys. Rev. B*, **38**, 12903 (1988).
[12] M. Widom, K. J. Strandburg, R. H. Swendsen, "Quasicrystal equilibrium state", *Phys. Rev. dett.*, **58**, 706 (1987).
[13] Y. Sasajima, K. Sakayori, M. Ichimura and M. Imabayashi, "Monte Carlo simulation of Diffusion process on quasi-lattice", *Jpn. J. Appl. Phys.*, **31**, 1417 (1992).
[14] S. Ozawa, Y. Sasajima and T. Haseda, "Dynamics and Patterns in Complex Fluids", eds. A. Onuki and K. Kawasaki, (Springer-Verlag, Berlin, 1990), p. 198.