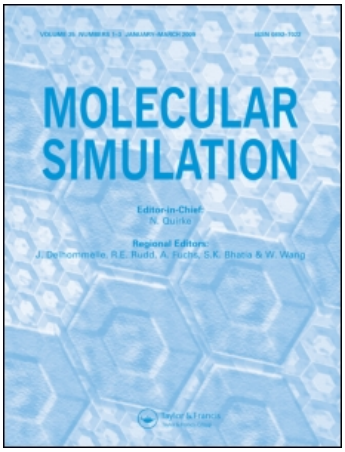

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Molecular Dynamics Simulation of Nucleation and Growth of a Binary Quasicrystal

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MOLECULAR DYNAMICS SIMULATION OF NUCLEATION AND GROWTH OF A BINARY QUASICRYSTAL

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The quasicrystal structure is considered to be a new type of ordered phase because its Fourier transform has Laue spots with icosahedral symmetry, which is inconsistent with crystal structure. Computer simulation of the formation process of a quasicrystal was performed by the molecular dynamics method. On the basis of the Strandburg type of quasicrystal model, we developed an algorithm of the formation process of binary quasicrystal reflecting the procedure as realistically as possible. The Fourier transform of some of the obtained structures has shown decagonal symmetry although the spots are rather diffused. It has been shown that the potential parameter and experimental condition should be limited to produce a perfect quasicrystal structure.

KEY WORDS: Quasicrystal, molecular dynamics, Penrose lattice, Strandburg-type model

INTRODUCTION

Quasicrystal is a new phase which has Laue spots with both fivefold symmetry and long-range order [1]. Since the discovery of quasicrystal by Shechtman *et al.*, many models and theories about the structures and origins of quasicrystal have been proposed so far [2]. We also have studied the growth process and stability of quasicrystals, using computer simulation [3-9]. The Monte Carlo method (MC) and molecular dynamics method (MD) provide powerful tools for computer experiments [10]. The Monte Carlo method provides samples representing phenomena of interest, statistically utilizing the random number generator. In the previous paper we studied the film growth process on the 2D Penrose pattern by the MC method [9]. This study has simulated the phenomena that particles migrate on the 2D quasicrystal lattice points and aggregate to form a quasicrystal structure. On the other hand, the molecular dynamics method (MD) makes it possible to study the dynamical properties of a system by numerical integration of classical or quantum mechanical equations of motion. Using this method, trajectories of particles in continuous space can be determined; therefore, we can obtain complementary information about the growth of quasicrystals from these two methods. The most important purpose of the present study is to clarify the growth of perfect quasicrystal by the molecular dynamics method.

STRUCTURAL MODEL AND METHOD OF CALCULATION

Model potential

In the present study, interaction between particles was assumed as the Lennard-Jones potential,

$$\psi_{ij}(r) = 4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r}\right)^{12} - \left(\frac{\sigma_{ij}}{r}\right)^{6}\right] \tag{1}$$

where r is the distance between particles, and ε_{ij} and σ_{ij} are parameters representing the strength of potential and atomic size, respectively. Subscripts i and j in this equation represent kinds of particles. We utilized this simple form of potentials in order to clarify the effect of binding energy and atomic size. In this study, the system is composed of two kinds of particles: large particles, represented as L, and small particles, represented as S. The potential parameters were determined so as to satisfy the following conditions.

(1) Tenfold symmetry of bonding around S-type and L-type particles should be preserved.
(2) Bonding energy between different species is larger than that between the atoms of the same kind.

Strandburg *et al.* proposed the following parameters appropriate for the above demands [11].

$$\begin{aligned} \sigma_{LS} &= 1.0 \\ \sigma_{LL} &= 2\sin(36°) = 1.176\ldots \\ \sigma_{SS} &= 2\sin(18°) = 0.618\ldots \\ \varepsilon_{LS} &= 1 \\ \varepsilon_{LS} &= \varepsilon_{SS} = \frac{1}{2} \end{aligned} \tag{2}$$

Figure 1 shows all of the local arrangements of the Strandburg-type model [11] of quasicrystals. In this study we examined another case such that

$$\varepsilon_{LS} = 2, \tag{3}$$

which is two times larger than that of the Strandburg model. Table 1 shows the Lennard-Jones potential parameter utilized in the present study. Hereafter we refer to the original Strandburg's parameter set as the S type and the modified version ($\varepsilon_{LS} = 2$) as the F type.

Principle of Molecular Dynamics Method

The molecular dynamics method can be defined as a method for calculating trajectories of particles deterministically by numerical integration of classical or quantum equations of motion. For accurate calculation, interaction potential should be calculated from the first-principle theory; however, this is practically impossible for many particle systems. On the other hand, approximation of empirical potential has

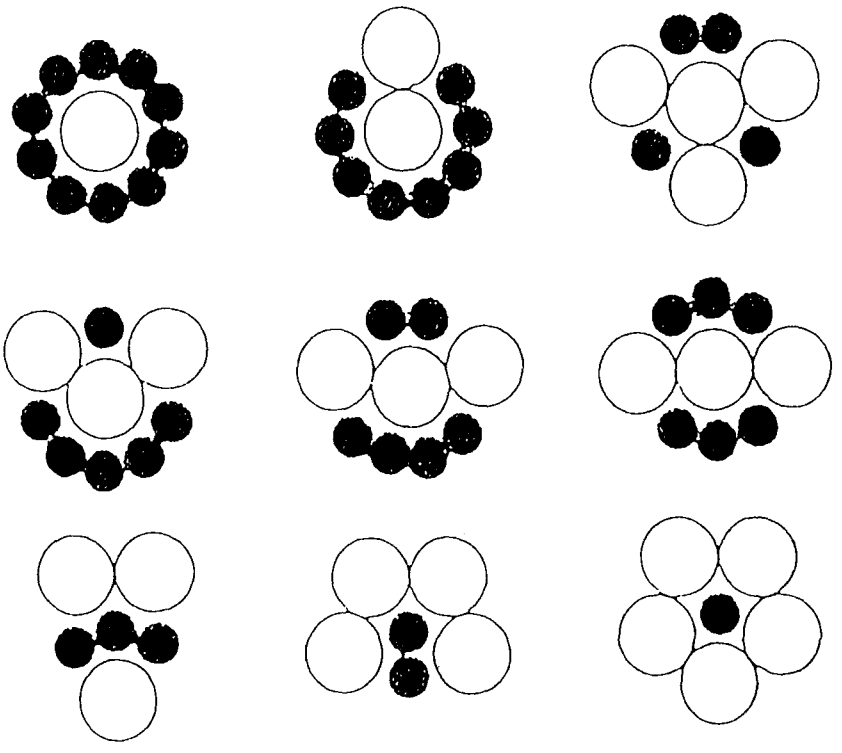

Figure 1 Complete set of low-energy local environments for Strandburg model [11].

Table 1 Lannard-Jones potential parameters ε_{LL}, ε_{LS} and ε_{SS} for the two types of Strandburg model. The parameters σ's are common for both systems; $\sigma_{LS} = 1.0$, $\sigma_{LL} = 2 \sin(36°) = 1.176$ and $\sigma_{SS} = 2 \sin(18°) = 0.618$.

Type Name / *Potential Prameter*	*Stype*	*Ftype*
ε_{LL}	0.5	0.5
ε_{SS}	0.5	0.5
ε_{LS}	1.0	2.0

been proven to be valid for solid and liquid states. In addition, two-body interaction appropriately describes the interaction in the homogeneous system, which can be considered to be isotropic. Among many functions that represent two-body potential, Lennard-Jones potential was utilized in this paper for its simplicity and popularity. The force acting on the i-th particle can be calculated as the sum of the two body force from all other particles as

$$F_i = \sum_j F_{ij} = \sum_j \nabla_i \phi(r_{ij}) \tag{4}$$

where ϕ is the two-body potential and r_{ij} *is the distance between the* i-th and j-th particles. The equation of motion of the i-th particle is

$$m_i\left(\frac{dv_i}{dt}\right) = F_i$$
$$V_i = \frac{dr_i}{dt}, \tag{5}$$

where m_i, v_i and r_i are the mass, velocity and position of the i-th particle, respectively. This differential equation cannot be solved analytically; therefore it should be replaced by a higher-order discretized equation and integrated numerically. One MD step, Δt, is the unit of the discretized time and determined to be on the order of one percent of the period of the lattice vibration. The discretization scheme we used here is according to Verlet [12]. Time and position are scaled here as

$$\left(\frac{m\sigma^2}{48\varepsilon}\right)^{\frac{1}{2}} \text{ and } \sigma\,, \tag{6}$$

respectively, where m is the mass of the particle, and ε and σ are the parameters of the Lennard-Jones potential. Time scale can be estimated as 3×10^{-13}s when the potential values for argon are inserted into the equation. In this study, simulation of nucleation and growth in a binary system has been performed under a constant temperature. Temperature of the system was evaluated by the equipartition law. The degree of freedom of an N-particle system in two dimensions is 2N-3, where -3 arises from the condition that translational movement is not allowed and that the temperature should be constant. Therefore the system temperature T is estimated from the following equation:

$$\left(\frac{1}{2}\right)k_B T = \left(\frac{1}{2N\text{-}3}\right)\sum_i \left(\frac{1}{2}\right)mv_i^2 \tag{7}$$

where k_B is Boltzmann constant. The temperature was kept constant at T_{ref} by scaling the velocity of every particle by the factor of

$$\beta = \left(\frac{T_{ref}}{T}\right)^{\frac{1}{2}}. \tag{8}$$

In the present study, the temperature is expressed by the dimensionless parameter T^*, calculated as

$$T^* = \frac{k_B T}{\varepsilon_{LS}} \tag{9}$$

where ε_{LS} is the strength of the potential between large (L) and small (S) atoms. The aim of this simulation is to study the growing process of an isolated grain of a quasicrystal. Therefore the boundary condition was set to be free and pressure was not controled because of isolation of the system.

Algorithm for Growth Simulation of a Binary System

To clarify the origin and the growth process of quasicrystal, it is necessary to study the microscopic motion of particles in the condensing process. One of the most effective methods for this purpose is the molecular dynamics method. The growth algorithm adopted here can be summarized as follows.

(1) Set a small particle at the origin as the seed.
(2) Create a large or small atom in the vicinity of the growing particle.
(3) Within a certain number of steps, calculate trajectories of the constituent atoms by means of molecular dynamics.
(4) Return to (2).

In our calculation, the experimental parameters are temperature T and growth interval J. Temperature was varied from 0.001 to 0.08 in dimensionless units, where 0.12 is the melting point of the two-dimensional monoatomic Lennard-Jones system. Growth interval J is set as 50, 500, 2500, or 10000 steps.

Fourier Transform

We calculated the Fourier transform of the final structure in order to calculate the symmetry and long-range order of the structure. The Fourier diffraction pattern corresponds to the electron diffraction pattern in the experiment. When r_j represents the location of the particles, we can calculate its Fourier transform by

$$|F(q)|^2 = |\sum_j e^{iq^* r_j}|^2, \tag{10}$$

where q is the wave vector. The Fourier spectrum is represented as the intensity map in the q-space, as shown in Figures 2–6(b). The area of the drawn circles was determined to be proportional to the intensity of the spectrum. The cutoff of the spectrum was 1% of the intensity of the incident beam.

RESULTS AND DISCUSSION

Table 2 represents system classification according to temperature and potential parameters. Figures 3 to 8 show the representative results of the simulation. The effects of the strength of interaction, temperature and growth interval on the final structure can be summarized as follows. First, we consider the strength of interaction. Local structures such as shown in Figure 1 should be numerous in the specimen if fivefold symmetry is constructed. However, the triangle clusters composed of the same kinds of atoms prevent the clusters in Figure 1 from nucleating in the system. Therefore in order to obtain defect-free quasicrystal, cluster formation of the same kinds of particles should be prevented and clusters composed of different particles should be produced preferably throughout the system. The results obtained here support this consideration because the tenfold symmetry of the structure is more evident for F-type systems than for S-type systems. In particular, comparison of

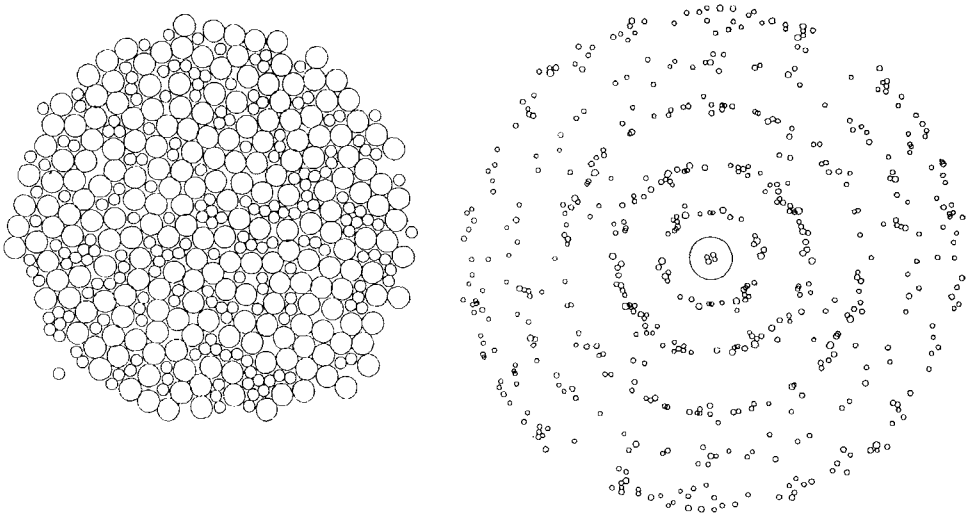

Figure 2 S01 type ($T = 0.001$, $\varepsilon_{LS} = 1.0$)
(a) The relaxed atomic structure at the 20,000th step and (b) its Fourier transform.

Figure 4 and Figure 5, shows that the stronger the interaction of different particles, the more evident the tenfold symmetry of the specimen. Furthermore we calculated the case of stronger interaction of different particles; however, effective atomic size ratios shifted from the Strandburg type and its Fourier transform was thus rather diffused. This finding indicates that the strength of interaction of different species should be two to four times larger than that of the same species, and that the conditions imposed on the atomic radius ratio are also important in producing perfect quasicrystal.

Secondly, we consider the system temperature. In the low-temperature region (see Figures 2 and 3), the final structure strongly depends on the initial structure because of the low mobility of particles. It is evident that the low mobility of particles prevents the formation of local structures of the Strandburg type. At high temperature (see Figures 6 and 7), and final structure contained many defects

Table 2 Experimental parameters for S and F type models. Growth interval is 500 MD step/atom for all systems.

System / *Temperature*	*Stype*	*Ftype*
0.01	S01	F01
0.1	S1	F1
0.2	S2	F2
0.4	S4	F4
0.6	S6	F6
0.8	S8	S8

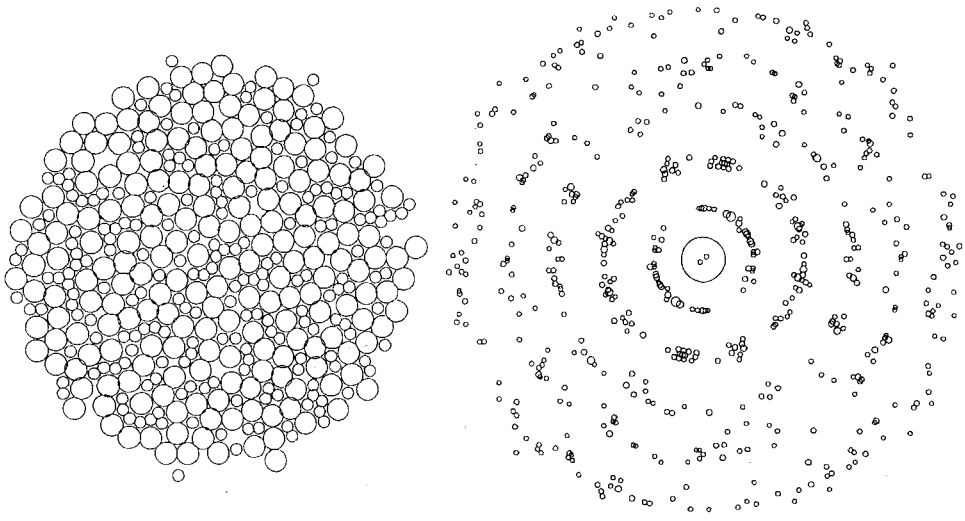

Figure 3 F01 type ($T = 0.001$, $\varepsilon_{LS} = 2.0$)
(a) The relaxed atomic structure at 20000 step and (b) its Fourier transform.

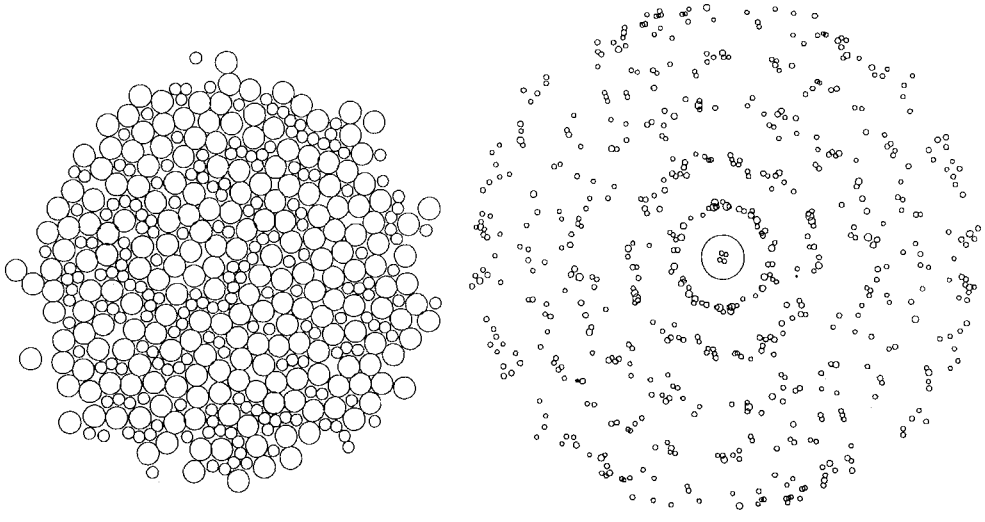

Figure 4 S2 type ($T = 0.02$, $\varepsilon_{LS} = 1.0$)
(a) The relaxed atomic structure at 20000 step and (b) its Fourier transform.

introduced according to the Boltzmann factor. Therefore, formation of quasicrystal is prevented both in the low- and high-temperature regions. Consequently, a moderate temperature (from 0.02 to 0.04) is necessary to produce defect-free quasicrystal. This temperature range is estimated to be about 8%–16% of the

melting point of the system, where we utilized the canonical Monte Carlo simulation to evaluate the melting point as 0.25. The "appropriate" temperature is rather low compared to that of crystalline growth. Consequently, the difficulty in producing perfect quasicrystal can be attributed to this result. But at higher system temperatures, the number of triangle clusters of the same particle decreases which prevents growth of quasicrystal. Considering the above result, an appropriate temperature gradient in the system is necessary for the growth of perfect quasicrystal.

The last parameter we should consider is the growth interval. It can be easily concluded that as the interval time increases, the formation of triangle cluster composed of same kind of atoms is more effectively prevented. In the present calculation the interval was increased to 10000 MD steps, but we could not obtain a sample with clear tenfold symmetry. Nevertheless, the diffraction spots are more evident than those of the short time interval, and the final structure is closer to the Strandburg-type configuration. A much longer interval should be examined to obtain defect-free quasicrystal.

In summary, a perfect quasicrystal structure could not be obtained in the present simulation, but, we succeeded in obtaining a structure which is similar to quasicrystal structure locally and of which the Fourier transform has tenfold symmetry, based on the nucleation and growth algorithm. The present simulation suggests that local rule of the formation process is sufficient to create a quasicrystal structure. Subsequent papers should be directed toward finding appropriate experimental conditions for the formation of a perfect quasicrystal structure.

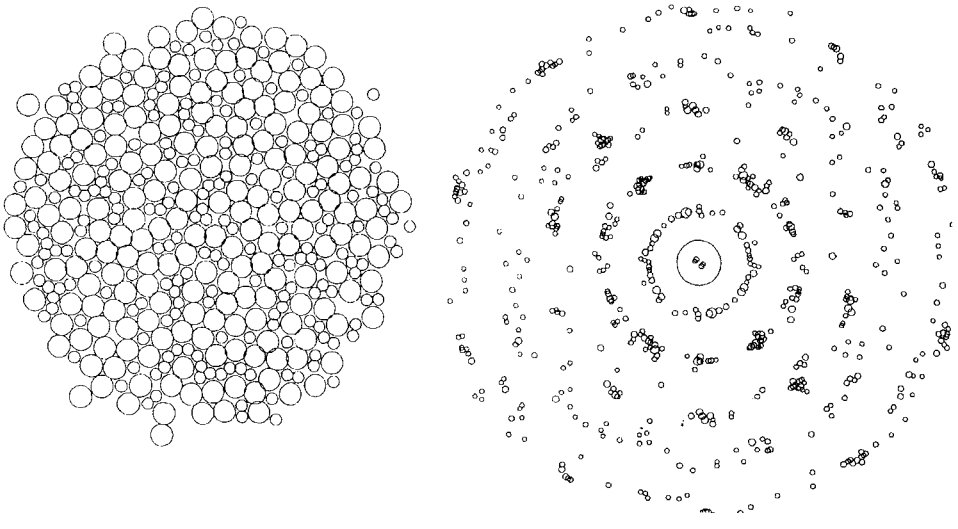

Figure 5 F2 type ($T = 0.02$, $\varepsilon_{LS} = 2.0$)
(a) The relaxed atomic structure at 20000 step and (b) its Fourier transform.

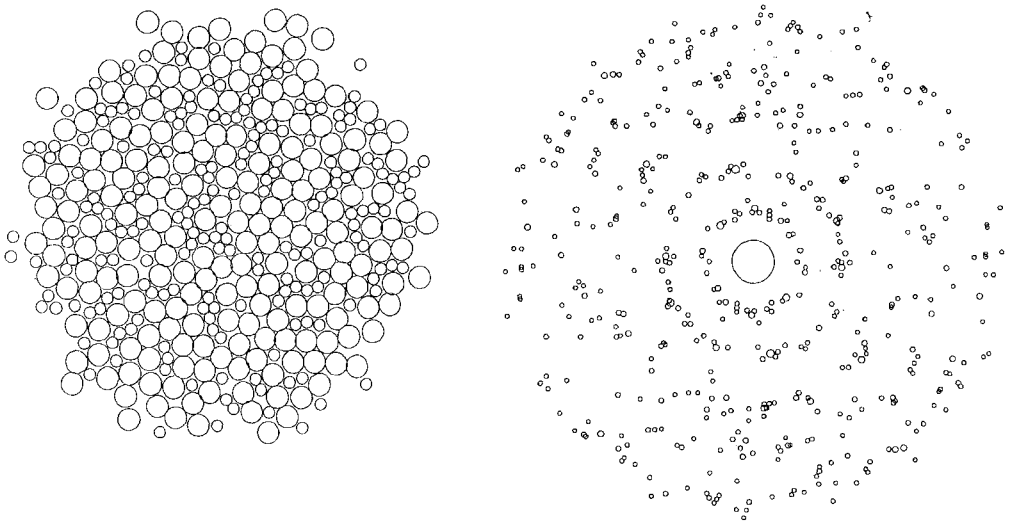

Figure 6 S8 type ($T = 0.08$, $\varepsilon_{LS} = 1.0$)
(a) The relaxed atomic structure at 20000 step and (b) its Fourier transform.

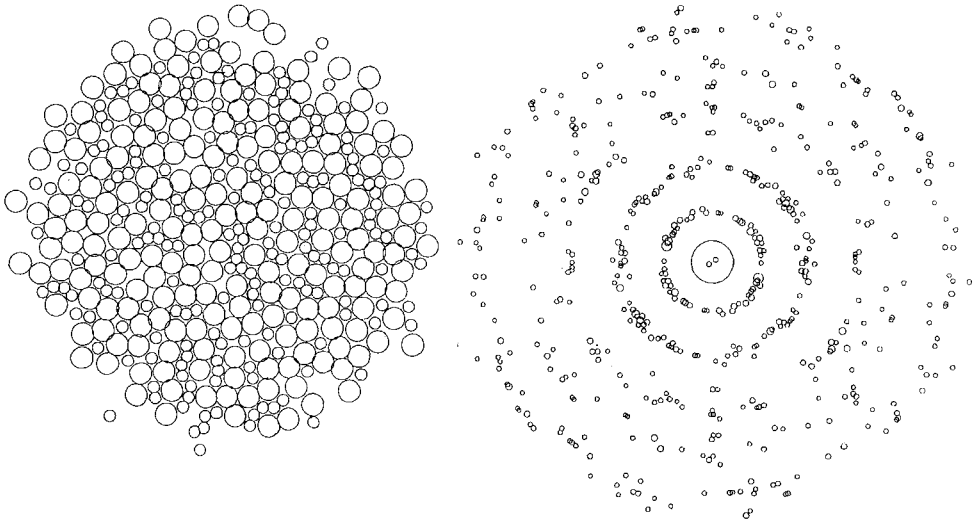

Figure 7 F8 type ($T = 0.08$, $\varepsilon_{LS} = 2.0$)
(a) The relaxed atomic structure at 20000 step and (b) its Fourier transform.

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