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Decagonal and Dodecagonal Quasicrystals Obtained by Molecular Dynamics Simulations *

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Double-well potentials are used for molecular dynamics simulation in monatomic systems. The potentials change as their parameters are adjusted, resulting in different structures. Of particular interest, we obtain decagonal and dodecagonal quasicrystals by simulations. We also verify the results and explain the formation of quasicrystals from the perspective of potential energy.

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Quasicrystals are characterized by the absence of Bravais lattices and/or the observation of forbidden crystallographic symmetries,[1] such as five-, eight- and ten-fold rotational symmetries. In 1984, Shechtman *et al.*,[2] reported an icosahedral structure for the first time in Al-Mn alloy in a quick annealing experiment, and proved the existence of quasicrystal through the electron diffraction pattern. After that, more and more scientists began to focus on this intriguing field. Many other kinds of quasicrystals have been found, of which the typical two-dimensional ones are octagonal,[3] decagonal,[4] and dodecagonal quasicrystals.[5] Numerous works have been devoted to the structural properties of different quasiperiodic models.[6–12] In recent years, quasi-crystalline structures have also been found in organic matter[13] and soft matter.[14]

With the development of computational facilities, scientists can use computer simulation to study the formation of quasicrystal to supplement experimental results. They can observe how quasicrystal grows and clarify its mechanism. Molecular dynamics (MD) simulation is a valuable method to generate quasicrystal structures. Compared with real experiments, the system in MD simulation can be chosen freely according to the research objectives.

To simplify the physical mechanism, monatomic systems are often used in MD simulation. A large amount of identical particles are rearranged to form quasicrystal under an effective potential function. The interatomic potential function is a key factor in the computer simulation at the atomic scale. To obtain an ideal quasicrystal structure, one has to seek a reasonable potential function and apply it to describe the atomic interaction in the system. In previous studies, atoms rearranged under the Dzugutov potential and quasicrystal structures with 12- and 8-fold symmetries[15,16] were obtained. Engel *et al.* observed decagonal and dodecagonal quasicrystals[17] in two-dimensional systems through MD simulations using the Lennard–Jones–Gauss (LJG) potential. Later, the simplified oscillating pair potential was employed to generate an icosahedral quasicrystal in a three-dimensional system.[18] Furthermore, Zu *et al.* observed 8- and 12-fold symmetric quasicrystal structures in simulations for soft matter using a simple potential function without long-range interactions.[19]

Even though many results have demonstrated the existence of quasicrystals, there are abundant new structures with different prototiles and symmetries to be explored. For example, Engel and Trebin[20] showed a phase diagram of crystal and quasicrystal structures for different parameters in the LJG potential function, including such tiles as pentagon, hexagon, decagon, square and triangle, which are basic blocks of crystal or quasicrystal. Therefore, we believe that more than one kind of quasicrystal can be obtained under the LJG potential.

Compared with the LJG potential, the classical Lennard–Jones (LJ) potential with an energy minimum is often applied in simulating some crystal structures. However, for a quasicrystal, which possesses non-crystallographic symmetry, more than one interatomic distance will exist in the structure. Therefore, a Gaussian term is added between the distance of the energy minimum and the truncated distance, to obtain a potential with double wells, by considering the long-range interaction on the basis of the LJ potential. In the whole system, there are multiple metastable atomic positions, in which atoms can be constrained at the corresponding distances, thus it is possible to maintain the special structure of a quasicrystal.

In this work, we focus on simulating the two-dimensional quasicrystal systems based on the LJG potential by optimizing the parameters in the following expression

$$V(r) = \frac{1}{r^{12}} - \frac{2}{r^{6}} - \epsilon \exp\Big(- \frac{(r - r_0)^2}{2\sigma^2} \Big). \quad (1)$$

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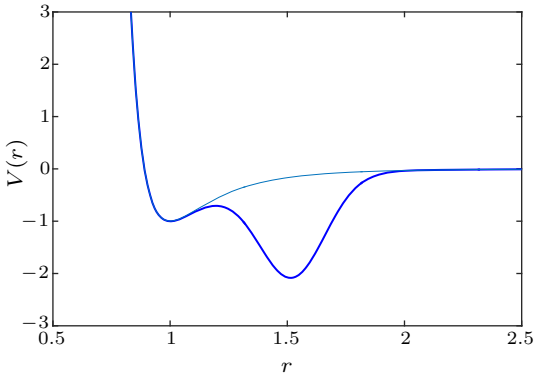

Fig. 1. LJG potential curve with $\epsilon = 0.5$, $r_0 = 1.50$, $\sigma^2 = 0.02$. The thin line is the original LG potential without the Gaussian term.

As shown in Fig. 1, there is only one potential well in the original LJ potential, and the second well occurs in the LJG potential when the Gaussian term is added. The potential around the first well represents the nearest-neighbor interaction, and the second potential well makes the main contribution to the next-nearest-neighbor interaction between atoms.

Using the LJG potential in Eq. (1) to simulate a two-dimensional monatomic system, the choice of the three parameters ϵ, r_0 and σ^2 is of central importance. We restrict the parameter space within $r_0 \in [1.3, 2.1]$, $\varepsilon \in [-0.6, -1.6]$ by fixing $\sigma^2 = 0.02$. The LJG potential function will change as we adjust the parameters to find a suitable pair potential to describe the atomic interactions of the quasicrystal system. After performing simulations in more than 800 samples with 1024 particles using different parameters, we have succeeded in obtaining decagonal and dodecagonal quasicrystal structures that are closer to the geometric models than those reported before,[17,21] and correspond to the dodecagonal quasicrystal found in experiments.[22]

In a two-dimensional plane, a monoatomic system consisting of 1024 identical atoms is built, in which the initial structure is a square lattice. In the NPT (number of atoms, pressure and temperature are constants) system, when we perform the MD simulation from the initial temperature $T = 0.5$ to the final temperature $T = 0.1$ over 1,000,000 steps, a decagonal quasicrystal structure is formed under the LJG potential with $r_0 = 1.52$ and $\varepsilon = -0.638$. When we perform the MD simulation over 500,000 steps from $T = 0.5$ to $T = 0.1$ with the same initial system, a dodecagonal quasicrystal structure is obtained under the LJG potential with $r_0 = 1.9$ and $\varepsilon = -0.866$.

The decagonal quasicrystal structure formed in our simulation is shown in Fig. 2(a), which consists of five tiles shown in Fig. 2(b) as given by Engel and Trebin.[20] The percentages of decagons and pentagons are higher in the structure obtained here, which means that they are closer to an ideal quasicrystal. The diffraction intensity is calculated by $I(\boldsymbol{K}) = \frac{1}{N}\left|\sum_{j=1}^{N} \exp(i\boldsymbol{K}\cdot\boldsymbol{r}_j)\right|^2$, where N is the total number of atoms, $\boldsymbol{K}$ is the reciprocal lattice vector, and $\boldsymbol{r}_j$ is the position vector of the jth atom. The diffraction pattern is presented in Fig. 2(c), where the 10-fold rotational symmetry is clearly seen.

The dodecagonal quasicrystal structure obtained in our simulation is shown in Fig. 3(a). As a comparison, the tiles proposed by Engel and Trebin[20] are also shown in Fig. 3(b). However, our structure consists of mainly squares and triangles. The corresponding diffraction patterns of Fig. 3(c) confirm the 12-fold rotational symmetry of the atomic structure.

To demonstrate the quasiperiodic tiles of the structures, we calculate the radial distribution function for the structures in Figs. 2(a) and 3(a). The results are shown in Figs. 4(a) and 4(c) for decagonal and dodecagonal quasicrystals, respectively.

The basic blocks that make up the decagonal structure are the regular pentagon and decagon as shown in Fig. 4(b). The nearest-neighbor distance is equal to the length between the adjacent vertices of a pentagon or a decagon, which corresponds to the first peak in the radial distribution. The next-nearest-neighbor distance as shown in Fig. 4(b) corresponds to the second peak in Fig. 4(a). The first peak is located at $r_1 = 0.936$ and the second peak at $r_2 = 1.532$. The ratio between the two distances is $r_1/r_2 = 1 : 1.630$, which is close to the ratio between two characteristic lengths in an ideal decagonal quasicrystal structure.[23]

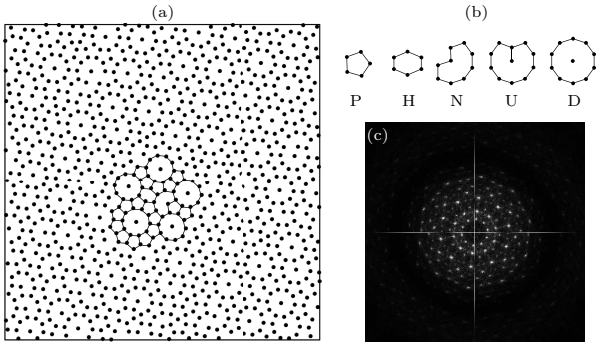

Fig. 2. (a) The decagonal quasicrystal structure obtained from the MD simulation. (b) The tiles proposed in Ref. [20]: (P) pentagon, (H) hexagon, (N) nonagon, (U) utile and (D) decagon. (c) The diffraction pattern.

For the dodecagonal quasicrystal in Fig. 3(a), the basic block is composed of squares and regular triangles with equal sides, with the two existing atomic distances being equal to the length of the side and the diagonal as shown in Fig. 4(d). In a single dodecagonal tile, there is also the third atomic distance being equal to the length from the central atom to the atom on the outside ring. Figure 4(c) is the radial distribution function of the simulation result, where the three peaks correspond to the three distances mentioned above. The first distance is 1, the second distance

is 1.440, and the third distance is 1.918, and their ratio is close to 1:1.414:1.932 in the ideal dodecagonal quasiperiodic tiling.[24]

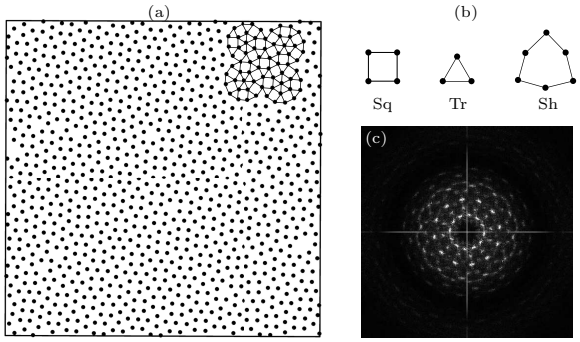

Fig. 3. (a) The dodecagonal quasicrystal structure obtained from the MD simulation. (b) The tiles proposed in Ref. [20]: (Sq) square, (Tr) triangle and (Sh) shield. (c) The diffraction pattern.

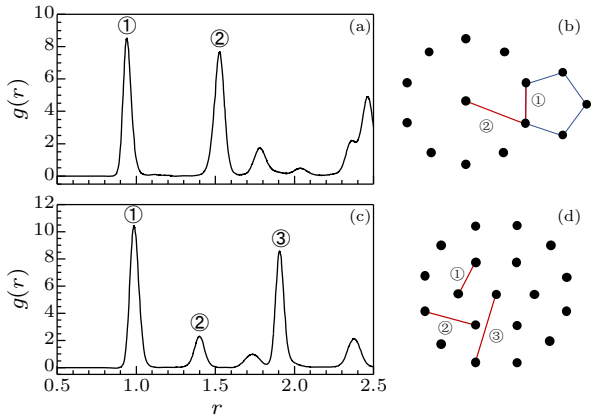

Fig. 4. (a) The radial distribution function of the decagonal quasicrystal and (b) the basic blocks of the structure. (c) The radial distribution function of dodecagonal quasicrystal and (d) the basic blocks of the structure.

To demonstrate the stability of the quasicrystals obtained in the simulation, we analyze the energy of the systems with the variation of the potential parameters when the locations of particles are fixed.[19,23]

Firstly, we calculate the energy $E = \sum_{i}^{N} \sum_{j}^{M_i} V_{ij}$ with different values of the parameter r_0 from 1.30 to 2.10 and with fixed depth of the second well by setting $\varepsilon = -0.638$. The results are shown in Fig. 5(a). In the decagonal structure of Fig. 2(a), the energy minimum is found when $r_0 = 1.52$. As a comparison, the energies in the square and hexagonal lattices are also calculated and their minima are found at $r_0 = 1.40$ and $r_0 = 1.73$, respectively.

Next, the system energy is calculated in the dodecagonal structure of Fig. 3(a). The parameter $\varepsilon = -0.866$ remains unchanged but the central position r of the Gaussian term varies between 1.3 and 2.1. The energy minimum of the dodecagonal quasicrystal is found at $r_0 = 1.90$ as shown in Fig. 5(b). The similar calculations in the hexagonal and square lattices and their energy minimum positions are $r_0 = 1.40$ and $r_0 = 1.77$, respectively.

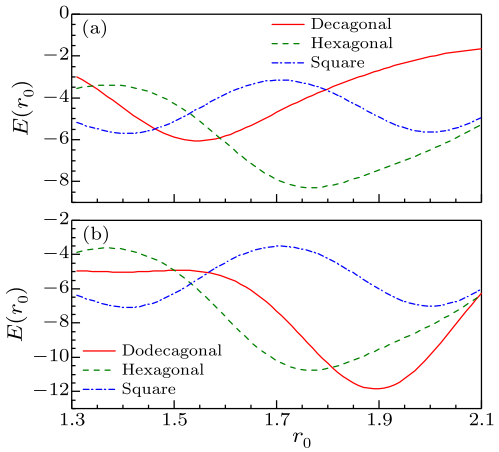

Fig. 5. (a) The system energy calculated in three fixed lattices by setting $\varepsilon = -0.638$ with various r_0 from 1.30 to 2.10. The solid, dashed and dashed-dotted lines are respectively the results for decagonal, hexagonal and square lattices. The minimum energy value occurs at $r = 1.52$ for the decagonal lattice. (b) The similar calculations in dodecagonal, hexagonal and square lattices are represented by the solid, dashed and dashed-dotted lines, respectively. For the fixed parameter $\varepsilon = -0.866$, the minimum energy position is found at $r_0 = 1.90$ for the dodecagonal lattice.

The above results on the energy minima strongly support the decagonal and dodecagonal quasicrystal structures that we have obtained under the LJG potential. On the other hand, it also suggests that when we try to construct a specific structure using a certain form of potential function, the feasibility may be evaluated by comparing the energy with different competing factors and finding a set of parameters favorable to the formation of the desired structure.

In summary, we have obtained significant decagonal and dodecagonal quasicrystals under LJG potentials by MD simulations. For the decagonal quasicrystal structure, we adjust the LJG potential function by changing the second potential well, and determine the position of the second potential well by observing the basic blocks in the simulation results. Then the depth of the potential well is changed and the simulation conditions are optimized to obtain the ideal decagonal quasicrystal structure, which has the lowest energy. We also confirm the results from the perspective of system potential energy. Following the same idea, we have also obtained a new dodecagonal quasicrystal structure, which corresponds to an observation in real experiments and which has not been reported by MD simulation before.

The present work confirms the validity that atoms can be rearranged to form different quasicrystals under the LJG potential function. Under such a simple double-well function, other kinds of quasicrystal struc-

tures may be discovered. It is worthwhile to further study whether other multi-well potentials can be applied to MD simulation to obtain different quasicrystal structures. Moreover, many other factors that are responsible for the formation of quasicrystal structure should also be considered, such as the depth of wells, temperature, pressure and cooling rate.[25]

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