Dynamics of particle flips in two-dimensional quasicrystals

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The dynamics of quasicrystals is more complicated than the dynamics of periodic solids and difficult to study in experiments. Here, we investigate a decagonal and a dodecagonal quasicrystal using molecular dynamics simulations of the Lennard-Jones-Gauss interaction system. We observe that the short time dynamics is dominated by stochastic particle motion, so-called phason flips, which can be either single-particle jumps or correlated ring-like multi-particle moves. Over long times, the flip mechanism is efficient in reordering the quasicrystals and can generate diffusion. The temperature dependence of diffusion is described by an Arrhenius law. We also study the spatial distribution and correlation of mobile particles by analyzing the dynamic propensity.

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I. INTRODUCTION

Quasicrystals are long-range ordered structures without periodicity. They are traditionally found in metallic alloys [1] and recently also in other materials [2–4]. The dynamics of quasicrystals is characterized by two elementary excitations, phonons and phasons [5]. As in periodic solids, phonon modes correspond to the oscillatory motion of atoms around their equilibrium positions. In contrast, phasons describe local rearrangements of atoms, which are present due to the absence of periodic order. Possible rearrangements are restricted by the internal geometry of the quasicrystal. On the continuum level, phason modes are a concept of the hydrodynamic theory of quasicrystals [5}; in contrast to phonon modes, they are diffusive in nature and can only be excited internally via phason-phonon interactions. Long wave length phason fluctuations are present in thermodynamic equilibrium as evidenced by diffuse neutron scattering [9].

On the atomistic scale, the elementary process of a phason mode is a phason flip, which is the individual motion of a single atom or the correlated multi-atom motion of a few atoms. A phason flip transforms one local configuration into a similar, energetically nearly degenerate one by overcoming an energy barrier. The concept of phason flips plays an important role in explaining enhanced atomic diffusion [10, 11], dislocation motion [12], and structural phase transitions [13, 14] in quasicrystals. To analyze the flips on a microscopic level, various experimental works have been carried out. With the use of transmission electron microscopy images collective phason flips were observed in-situ as rearrangements of atomic clusters [15]. Indirect evidence for phason flips was obtained from the observation of phason walls traced out by dislocations [16]. Furthermore, quasi-elastic neutron scattering is believed to contain information about phason flips [17], although the contribution of the flips is hard to extract because it is hidden in the elastic peak. Despite the effort, phason modes and phason flips remain difficult to study in experiment and little is known about the underlying processes on an atomistic level.

Numerical studies of quasicrystal formation and dynamics were performed using model potentials such as the binary Lennard-Jones (LJ) potential in two dimensions [18] and three dimensions [19], and the one-component Dzugutov potential [20] among other systems [21]. In the binary LJ system, the ratio of the particle radii are chosen to favor decagonal (2D) and icosahedral (3D) order. Similarly, the potential of the Dzugutov system is characterized by two competing length scales. Distances around the potential maximum are strongly disfavored. By positioning the maximum at the characteristic interparticle distances of the triangular lattice or fcc/hcp, these simple close-packed lattices are destabilized and the particles are forced to find other, more complex configurations. Although quasicrystals have now been observed in several model systems as thermodynamically stable phases [21], the energetic ground state of all
systems studied in simulation so far is crystalline. Examples are two-dimensional monodisperse systems with a square potential [22] or the binary LJ system [23]. A quasicrystalline structure exists in these systems only at finite temperature and is therefore stabilized entropically.

Oscillatory interaction potentials are often used for quasicrystal simulations, because effective pair potentials for metals, which are by far the most important materials class for quasicrystals, are best described by a strongly repulsive part and a decaying oscillatory (Friedel, [24]) term. Classical pair potentials fitted to reproduce ab-initio forces typically show an oscillatory behavior [25, 26]. To investigate the influence of competing energy minima in the potential, Engel and Trebin introduced the Lennard-Jones-Gauss (LJG) system [27], which consists of identical particles interacting with a parameterized double-well pair potential. Its phase diagram as a function of the potential shape is complicated and includes at least two entropically stabilized quasicrystalline phases together with around a dozen of crystalline periodic phases [21, 28]. The LJG system therefore constitutes a simple model system for studying quasicrystals in simulation.

The aim of the present paper is to investigate phason flips using molecular dynamics simulations. In particular, we are interested in the basic mechanism on the particle level and the temperature dependence of the dynamics. Our model system consists of identical particles interacting in two dimensions with the LJG pair potential. The paper is organized as follows: Sec. II introduces the LJG system and numerical simulation methods. In Sec. III we examine the formation and thermodynamics of a decagonal and a dodecagonal quasicrystals. In Sec. IV we investigate individual and collective particle flips and discuss their role for diffusion. In Sec. V we study the spatial distribution of mobile particles from the dynamic propensity. Sec. VI concludes with a brief summary.

II. MODEL SYSTEM AND METHODS

A. Lennard-Jones-Gauss potential

The system under investigation consists of identical particles interacting with the LJG potential [27]

\[ V(r) = \varepsilon_0 \left\{ \left( \frac{r_0}{r} \right)^{12} - 2 \left( \frac{r_0}{r} \right)^6 - \varepsilon \exp \left( -\frac{(r-r_G)^2}{2\sigma^2} \right) \right\}, \]

where \( \varepsilon_0 \) is the energy unit and \( r_0 \) the length unit. \( r_G \), \( \varepsilon \), and \( \sigma \) are three potential parameters. The first two terms in Eq. (1) are the well-known LJ potential that remains fixed. The third term is a Gaussian well (\( \varepsilon > 0 \)), whose center, depth, and width are \( r_G \), \( \varepsilon \), and \( \sigma \), respectively. Depending on the choice of parameters, the LJG potential can be either a double-well (Fig. 1, top left) or single well with shoulder for small \( r_0 \) (Fig. 3, top left).

B. Molecular dynamics simulation

Molecular dynamics simulations are carried out with a system of 1024 particles using periodic boundary conditions. We solve the equations of motion with the leapfrog algorithm in the NPT ensemble employing a Nose-Hoover thermostat for temperature control and an Andersen barostat for pressure control. Throughout the paper the pressure is fixed at \( P = 0 \), which means we relax the boundaries such that the potential energy remains minimal. The particles are arranged randomly in the initial configuration with their velocities chosen according to a Maxwell-Boltzmann distribution. Simulation units are dimensionless: the length unit is \( r_0 \), the temperature unit is \( \varepsilon_0/k_B \), and the time unit is \( \tau = \sqrt{m r_0^2/\varepsilon_0} \). Here, \( k_B \) is the Boltzmann constant and \( m \) the particle mass. A single molecular dynamics time step is equal to 0.01\( \tau \). The potential cutoff is set to \( r_{cut}/r_0 = 2.5 \). Beyond \( r_{cut} \) the LJG potential is essentially zero for the potential parameters under investigation.

C. Correlation functions

The mobility of the \( j \)-th particle with trajectory \( \mathbf{r}_j(t) \) is given by the dynamic propensity [29, 30]

\[ \phi_j(t) = \langle (\Delta \mathbf{r}_j)^2 \rangle_{ic} = \langle (\mathbf{r}_j(t) - \mathbf{r}_j(0))^2 \rangle_{ic}. \]

(2)

The angle brackets indicate the average over an isoconfigurational ensemble, i.e. the simulation repeatedly starts from the same particle configuration but with momenta randomly assigned from the Maxwell-Boltzmann distribution. By averaging over the particles, we obtain the mean square displacement

\[ \langle r^2(t) \rangle = \frac{1}{N} \sum_{j=1}^{N} \langle (\mathbf{r}_j(t) - \mathbf{r}_j(0))^2 \rangle, \]

(3)

which is related to the diffusivity \( D \) of a \( d \)-dimensional system by the Einstein equation

\[ D = \frac{1}{2d} \lim_{t \to \infty} \frac{\langle r^2(t) \rangle}{t}. \]

(4)

The radial distribution function \( g(r) = \rho(r)/\rho \) is the average density at distance \( r \) divided by the global average density, where

\[ \rho(r) = \frac{1}{N} \sum_{j=1}^{N} \langle \delta(\mathbf{r} - \mathbf{r}_j(0)) \rangle, \]

(5)

is the density distribution. The distance and direction of the motion of an individual particle are measured by the van Hove autocorrelation function [33]

\[ G_a(r, t) = \frac{1}{N} \sum_{j=1}^{N} \langle \delta(\mathbf{r} - \mathbf{r}_j(t) + \mathbf{r}_j(0)) \rangle, \]

(6)
which has a peak at \( r = 0 \) and decays outwards. The static structure factor

\[
S(q) = \frac{1}{N} \left\langle \left| \sum_{j=1}^{N} \exp(-i\mathbf{q} \cdot \mathbf{r}_j(t)) \right|^2 \right\rangle_t \tag{7}
\]

is the usual non-energy resolved diffraction image \( S(q) = |\rho(q)|^2 \) as measured in diffraction experiments to determine crystal structure and symmetry.

D. Local structure analysis

In the theory of quasicrystals the crystal structure is described as a (decorated) tiling \([5]\). Each tile represents a certain local configuration. Due to the aperiodic order of quasicrystals, there is only a finite number of different tiles up to translation. Two-dimensional tilings are especially easy to visualize, because the tiles are polygons. In the quasicrystals found in the LJG system, a simple tiling is defined by the network of nearest neighbors: two particles are linked (i.e. nearest neighbors), if their distance is within the first peak of the radial distribution function. Individual tiles are identified by finding closed paths in the neighbor network. Examples for tile types that will be relevant in the following are triangles, squares, and pentagons. Polygonal tiles can be either empty (three to six vertices) or contain a single particle like for example the decagon discussed below. In general, tiles can also be concave or have irregular shape.

III. A DECAGONAL AND A DODECAGONAL QUASICRYSTAL

We study two sets of potential parameters that stabilize a decagonal and a dodecagonal quasicrystal in thermodynamic equilibrium. With the parameters

\[ r_G = 1.52, \; \varepsilon = 1.8, \; \text{and} \; \sigma^2 = 0.02, \tag{8} \]

a phase transition from a decagonal quasicrystal to a liquid is found at \( T_{M1} = 0.56 \pm 0.02 \) (Fig. 1). The transition is unusual, because (i) it is clearly first order and therefore does not resemble the melting transition of the hexagonal crystal in the 2D LJ system, which is characterized by the existence of an intermediate hexatic phase with unpinned disclination pairs \([31]\). No analog of a hexatic phase and no unpinned disclination are found in the decagonal quasicrystal before melting. (ii) The transition is accompanied by negative thermal expansion (Fig. 1 bottom left). During crystallization, the system expands by 3%. A similar behavior is known for example in water. As expected, the compressibility of the crystal is much lower than the compressibility of the liquid.

The reason for the negative expansion is the open structure of the decagonal quasicrystal (Fig. 2 left). Especially decagon tiles (a central particle surrounded by a ring of ten particles) are less dense than the average density of the liquid. Note also that the decagon number varies with temperature even within the stability region of the quasicrystal. The reason for this behavior is not the presence of structural defects (e.g. vacancies), but a change in the tile occurrence ratio.

The static structure factor (Fig. 2 right) shows the ten-fold symmetry of the quasicrystal. Strong diffuse scattering surrounds sharp peaks. According to the theorem by Mermin and Wagner \([32]\), Bragg peaks are not possible in two dimensions, but only algebraic divergencies. A close analysis of the figure reveals that the peaks (especially the inner weak peaks) are not aligned perfectly. The reason is the presence of residual phason strain in the system that has not relaxed completely over the finite simulation time. When comparing inde-
FIG. 3: (Top left) The Lennard-Jones-Gauss potential for $r_G = 1.42$, $\varepsilon = 1.8$, and $\sigma^2 = 0.042$ is a single well with shoulder. (Top right) The system exhibits two first order phase transitions with (Bottom left) positive thermal expansion and no thermal expansion, respectively. (Bottom right) While the quasicrystal (QC) and the liquid (L) have triangular, square-like, and pentagonal local configurations, no triangles or pentagons are present in the square phase (Sq).

A dodecagonal quasicrystal is found for the parameters

$$r_G = 1.42, \varepsilon = 1.8, \text{ and } \sigma^2 = 0.042. \tag{9}$$

The potential energy and the specific volume as a function of temperature (Fig. 3) indicate two phase transitions, the first from the decagonal quasicrystal to a crystalline square phase at $T_C = 0.35 \pm 0.01$ and the second from the square phase to a liquid at $T_{M2} = 0.40 \pm 0.02$. Hysteresis appears in the potential energy for both phase transitions. Contrary to the decagonal quasicrystal, the transition from the dodecagonal phase to the square crystal involves (conventional) positive thermal expansion. Neither expansion nor compression is observed for the square-liquid transition.

Only three tile types appear in the dodecagonal quasicrystal (Fig. 4): triangles, squares, and pentagons. Most pentagons are surrounded by a ring of twelve particles, but the five-fold symmetry of the pentagons breaks the twelve-fold symmetry. The square crystal (Fig. 4 bottom) has no triangles or pentagons. As will be shown below, particles are highly mobile in the square phase. This explains the appearance of a small number of defects (pentagons) found in the figure, which are present in thermodynamic equilibrium and help to stabilize the square phase (see the discussion in Sec. V).

The static structure factors for the dodecagonal quasicrystal and the square phase show twelve-fold and four-fold symmetry, respectively (Fig. 4 right). Rings of diffuse scattering are observed in the dodecagonal phase. As in the decagonal phase, the weak peaks are not well aligned, which is a hint for the presence of phason strain. The square crystal also shows pronounced diffuse scattering in the background, but in contrast to the quasicrystal the scattering appears in streaks instead of rings. The presence of such strong diffuse scattering in the square crystal is unusual and indicates a large local mobility of the particles in the square phase.

IV. STATISTICS OF PARTICLE FLIPS

In this section, we show that the elementary processes for displacive particle dynamics are single-particle flips in case of the decagonal quasicrystal and multi-particle flips in case of the dodecagonal quasicrystal. A sequence of flips is needed for collective motion as necessary for diffusion.

A. Individual flips

The average single-particle dynamics in a solid is measured by the van Hove autocorrelation function $G_\alpha(r, t)$. 

![Image](image-url)
Particles do not move far over short times, and the dominant contribution to $G_a$ comes from thermal motion around the local potential energy minima closest to the particle position. The oscillatory motion around the local equilibrium positions results in a peak at $r = 0$. In case of a harmonic well or at low temperatures, the peak has Gaussian shape.

The van Hove autocorrelation function $G_a(r, t)$, $r = (x, y)$ is shown in Fig. 5. We fix the time interval to $t = 100$, which is two to three orders of magnitude larger than the typical time for oscillation in a local minimum ($\Delta t = 0.1$ to 1.0). $G_a$ is non-isotropic for both quasicrystals and deviates from the expected behavior for a crystalline solid. In the case of the decagonal phase, rings of local maxima with outwards decreasing heights surround a central peak. The central peak of the decagonal quasicrystal decays exponentially and is significantly broader than what would be expected from pure oscillatory motion. This indicates that particles are highly mobile and phonons are not the only mechanism of particle dynamics.

To investigate the unusually high particle mobility in the quasicrystals, we study trajectories of particles belonging to characteristic high-symmetry clusters. A decagon cluster (Fig. 6, left) consists of a single central particle surrounded by a ring of ten particles. In the figure, one of the particles (‘8’) moves away from its position at a ring vertex to a position in the interior of the ring where it remains for the rest of the trajectory. The time for the switch of position is short, circa $\Delta t = 0.3$, which is in the order of a single phonon oscillation. We can say that the particle ‘jumped’ to its new position. The motion is an example of a single-particle flip.

The dodecagon cluster (Fig. 6, right) consists of five central particles surrounded by a ring of twelve particles. As shown by the trajectories of the particles, the elementary dynamical process is the correlated rotation of the central particles called a multi-particle flip. The rotary motion is not as well-defined as the single-particle flip. Since the five-fold center breaks the symmetry of the twelve-fold ring, the multi-particle flip has a lower energy barrier than the single-particle flip in the decagonal phase.

We can determine the flip distances and flip directions of the decagonal phase from the peaks positions in Fig. 5. There is only a single flip distance: $\Delta r \approx 0.6$. In an ideal tiling of edge length 1, the flip distance would be $r^{-1} \approx 0.62$ with the golden mean $r = (\sqrt{5} + 1)/2$. The sequence of arrows in Fig. 5 (left) indicate consecutive flips: An example of a single flip is the jump to the end of the first arrow, an example of two second flip is the jump to the end of the second arrow, etc. Up to four consecutive flips are observed during the observation time of $\Delta t = 10$. As expected, the probability for consecutive flips decays rapidly with the number of flips.

What determines which of the particles is going to flip next? Besides geometric restrictions, which can be understood from the underlying tiling [33], the height of the energy barrier plays an important role. Equilibrium positions with higher potential energies will in general be less favorable. Potential energy histograms for each of the eleven particles of the decagonal cluster are shown in Fig. 7. The central particle has by far the lowest energy with $E_{\text{center}} \approx -9$. The particles on the decagon rings have energies $E_j \approx -6$ for $j = 3, 4, 5, 6, 10$, $E_j \approx -5$ for $j = 1, 2, 7, 9$, and $E_j \approx -4$ for $j = 8$. The reason for the appearance of three energies is the different local configuration of the particle on the outside of the ring. The particle with highest potential energy is the one that is observed to flip in Fig. 6 (left).
range $t < a$ low potential energy position (Fig. 7). In the time a total time of $t = 10$. Particle in Fig. 6(left). Its trajectory is recorded over in Fig. 9. The particle under investigation is the ‘center’ (single arrow) and a string-like chain of four consecutive flipped (four arrows in the center) can be seen. Dashed lines serve as a guide for the occurrence. The displacement of the flipping atoms is indicated by arrows. Black lines if they are neighbors, i.e. if their distance is small enough, then they can induce a collective reorganization of the quasicrystal and lead to particle motion over long distances and eventually diffusion. An example for consecutive flips in the decagonal phase is shown in Fig. 8. Two particles (red points) are connected by black lines if they are neighbors, i.e. if their distance corresponds to the first peak in the radial distribution function. The left panel is the initial configuration, and the right figure the final configuration after the flips have occurred. The displacement of the flipping atoms is indicated by arrows. Dashed lines serve as a guide for the eyes to identify the particle motion. Five individual flips (single arrow) and a string-like chain of four consecutive flips (four arrows in the center) can be seen.

The long-time dynamics of a single particle is analyzed in Fig. 9. The particle under investigation is the ‘center’ particle in Fig. 8(left). Its trajectory is recorded over a total time of $t = 10^5$. At the beginning, the particle remains stable for some time, because it starts at a low potential energy position (Fig. 7). In the time range $t < 10^4$, few back-and-forth jumps to neighboring positions are observed. Only later, the particle starts moving further away. Equilibrium positions frequently form pentagons on the intermediate time scale. The time for a switch from one pentagon to another is on the order of $\Delta t = 10^4$ at the temperature under investigation ($T = 0.5$).

We can define an effective free energy $F(x, y) = -k_B T \ln Z(x, y)$ for a particle positioned at $(x, y)$. The restricted partition function $Z(x, y)$ is obtained by averaging over a constraint phase space, where the selected particle is tagged at $(x, y)$ and the others are allowed to perform only phonon motion but no additional flips. $F(x, y)$ can then be interpreted as the free energy landscape for this particle $[34]$. In simulation, the effective free energy can be determined in a first approximation from the probability density $P(x, y)$ of the particle position:

$$F(x, y) = -k_B T \log P(x, y). \quad (10)$$

Fig. 9(right) shows the intensity map of $F(x, y)$ for the particle on the left side. The particle favors staying at equilibriums positions arranged on vertices of pentagons. The particle dynamics can be understood as a flip motion among the basins of the free energy landscape.

C. Diffusion

The diffusivity $D(T)$ of the quasicrystals can be determined from the average slope of the mean square displacement $\langle r^2(t) \rangle$. In the case of the decagonal quasicrystal, we also measure the flip frequency as a function of temperature by counting the number of flips during the simulation.

We find that the diffusivity and the flip frequency decreases rapidly over many orders of magnitude during lowering of temperature (Fig. 10). In order to investigate whether individual flips and diffusion are activated
processes, we fit the curves by an Arrhenius law
\[ D(T) = D_0 \exp(-\Delta E/k_B T). \] (11)
The fit, indicated by blue lines in Fig. 10, works well confirming that flips and diffusion are indeed dominated by energy barriers. The slopes for the curves correspond to activation energies \(\Delta E\). Activation energies for diffusion in the decagonal quasicrystal (‘10’), the dodecagonal quasicrystal (‘12’), the square crystal (‘4’) and for flips in the decagonal quasicrystal (‘f’) are:
\[
\begin{align*}
\Delta E_{10} &= 5.46(14), \\
\Delta E_f &= 3.11(10), \\
\Delta E_4 &= 1.26(18), \\
\Delta E_{12} &= 1.54(7).
\end{align*}
\] (12-15)
The small deviations from the Arrhenius law observed in Fig. 10 for low and high temperatures can be a result of the change of barrier heights with temperature, which we expect to happen due to particle interactions during a flip. For example, if a particle wants to squeeze through two other particles to achieve a flip, then it is possible that the latter particles are pushed to the sides to make it easier for the former particle to pass through. Cooperative effects like this one will change with temperature, which alters the effective barrier heights. Furthermore, the change of the tile occurrence ratio with temperature can modify the flip type and therefore energy barrier height distribution. Finally, defect tiles and vacancies can be present at higher temperature and constitute a competing mechanism for diffusion, which can lead to a deviation from the ideal Arrhenius law. As can be seen in Fig. 10 (bottom), the effective energy barrier heights increase with increasing temperature for the decagonal system, while they decrease with increasing temperature for the dodecagonal system.

The activation energy \(E_f\) calculated for the flip frequency in the decagonal phase is lower than the activation energy \(E_{10}\) calculated for the diffusivity. Since diffusion takes place by successive flips, there has to be a distribution of energy barrier heights for individual flips. In general, lower barriers lead to faster movement. The highest barrier that has to be overcome for long-range particle motion to occur constitutes the bottleneck for diffusion.

Variations of barrier heights can be seen indirectly in Fig. 9. It is observed that the particle motion along the vertices of pentagons (‘intra-pentagon flip’) is fast and therefore corresponds to relatively low barriers. On the other hand, the transition from one pentagon to another one (‘extra-pentagon flip’) is found to be much slower and corresponds to relatively high barriers. The ratio of times for the two types of motions is approximated by \(t_{ep}/t_{ip} = \exp(\Delta E_{10} - \Delta E_f)/k_B T\). For \(T = 0.5\), \(t_{ep}/t_{ip} \approx 100\). This agrees well with what is typically observed in simulation. In other words, for every 100 intra-pentagon flips there is about one extra-pentagon flip, lowering the diffusivity compared to what would be expected from the flip frequency alone. Extra-pentagon flips are the bottleneck for long-range diffusion in the decagonal quasicrystal.

The energy barrier for diffusion in the dodecagonal quasicrystal is lower than the energy barrier for diffusion in the decagonal quasicrystal. This means that the dodecagonal quasicrystal reorganized more easily. The single-well nature of the LJG potential for the dodecagonal system helps to facilitate flips and explains the low energy barriers. In contrast, the intermediate bump in the LJG potential for the decagonal system leads to higher energy barriers.

As we will see below, the reason for the fast diffusion in the square crystal is of different nature; it is connected to the formation and propagation of local defects. Such defects are easily possible due to the softness of the interaction potential.

Note that both quasicrystals are stabilized entropically only above a certain critical temperature; at lower temperatures they transform into periodic approximants, i.e. periodic crystals with a similar local structure as the quasicrystals. The decagonal quasicrystal is unstable relative to the approximant Xi for \(T \leq 0.37\); and the dodecagonal quasicrystal is unstable relative to the approximant Pen 2 for \(T \leq 0.25\). In our simulations, periodic boundary conditions suppress the transforma-

![Figure 10: The diffusivity and the flip frequency of the decagonal quasicrystal (Left) and the diffusivity of the dodecagonal quasicrystal (Right) as a function of temperature (Top). (Bottom) The temperature dependence can be fitted with an Arrhenius law as indicated by blue lines. At low temperatures and in the absence of finite-size effects, the quasicrystals transform into the periodic approximants Xi and Pen 2.](image)
tions from quasicrystals to approximants. Nevertheless, we do observe that the dynamics slows down in the regions where the approximants are stable (most prominently for the dodecagonal quasicrystal) as visible by the deviation from the Arrhenius law at low temperatures in Fig. 10. Here, the quasicrystals are unstable.

V. DYNAMIC PROPENSITY

To investigate the distribution of mobile particles, the temperature dependence of the dynamic propensity $\phi_j(t)$ is studied. Since we are interested in short time motion, we fix $t = 10$ in the following. The isoconfigurational average (see Eq. (2)) is taken over 1000 different initial velocities. Fig. 11 illustrates the spatial distribution of $\phi_j(t)$ at $T = 0.5$ in the decagonal phase. We observe that most of the particles have low mobility. Particles with high mobility appear predominantly at the edges of decagon clusters. From the propensity of the green, yellow, and red particles ($\phi_j > 0.09$) we estimate that these particles move in average over a distance of $\Delta r > 0.3$, which means they perform flips with a probability of more than 50%.

Next, we consider the propensity in the case of the dodecagonal system, in particular close to the quasicrystal-square crystal transition at $T_C = 0.35$ and close to the melting transition at $T_{M2} = 0.4$. Fig. 12 shows the spatial distribution of propensity at the temperatures $T = 0.3$, 0.35, 0.36, and 0.38. In the dodecagonal phase ($T = 0.3$), the five central particles of dodecagonal clusters often exhibit high mobility. When the temperature is increased ($T = 0.35$), mobility increases and larger areas are observed to have high propensity. In the square phase ($T = 0.36$), defect sites of high mobility are pentagonal rings. As the temperature is increased ($T = 0.38$), the defect domains grow.

To analyze the size of the mobile regions, we calculate the participation ratio (PR) of the propensity for the dodecagonal system. The participation ratio is defined by

$$\text{PR} = \frac{1}{N} \left( \frac{\sum_{j=1}^{N} \phi_j}{N} \right)^2.$$

Note that the participation ratio satisfies $\text{PR} \approx 1$ for extended states and $\text{PR} \ll 1$ for localized states. Fig. 13 shows the temperature dependence for $0.3 < T < 0.4$. We can identify three regimes: For $0.39 < T$ (liquid), the participation ratio is close to unity, which indicates that mobile particles are distributed homogeneously in the sample. The participation ratio for $0.35 < T < 0.39$ (square crystal) is lower than that for $T \leq 0.35$. This is a characteristic of localized defects. The higher participation ratio for $T < 0.35$ (quasicrystal) indicates that mobile particles are again distributed regularly in real space.

To measure the correlation of mobile particles, we study the Fourier transformed propensity distribution

$$S_{pro}(q) = \frac{1}{N^T} \left| \sum_{\phi_j > \phi} \phi_j \exp(iq \cdot x_j) \right|^2,$$

where the sum is over particles with propensity higher than the average value $\phi = \sum_j \phi_j / N$ and $N^T$ is the num-

FIG. 11: Spatial distribution of the propensity $\phi_j(t)$, $t = 10$ for the decagonal phase at $T = 0.5$. The centers of the circles are atomic positions, and the color represents propensity. Red means high propensity, and dark blue low propensity.

FIG. 12: Spatial distribution of the propensity $\phi_j(t)$, $t = 10$ for the dodecagonal phase at $T = 0.5$. The centers of the circles are atomic positions, and the color represents propensity. The decagonal quasicrystal at $T = 0.3$ (top left) and $T = 0.35$ (top right), and the square crystal at $T = 0.36$ (bottom left) and $T = 0.38$ (bottom right) is shown. The dynamical correlation length $\xi$ is given below the subfigures.
The dynamical correlation length is a measure for the size of mobile regions, where part of this work was conducted. The work of T.O. was supported in part by a Grant-in-Aid for Scientific Research (C) 19540405 from the Japanese Ministry for Education, Culture, Sports, Science, and Technology (MEXT). T.O. was supported in part by a Grant-in-Aid for Scientific Research (C) 19540405 from the Japanese Ministry for Education, Culture, Sports, Science, and Technology (MEXT). The dynamics of individual particles is characterized by local oscillatory motion (phonon dynamics) and discrete particle jumps (phonon flips), which are activated by thermal motion. We found that an elementary flip is a single-particle jump for the decagonal quasicrystal and a correlated ring-like multi-particle motion for the dodecagonal quasicrystal. Due to the high structural complexity, particles in the quasicrystals have various local environments. Phason flips occur preferentially for those particles with potential energies higher than the average. Over longer times, successive jumps form a sequence of flips and particles start to diffuse through the system. The temperature dependence of the diffusivity is well described with an Arrhenius law, which suggests that the diffusion mechanism is a conventional activated process. The dynamic propensity measures the distribution of particle mobilities in the system. For the decagonal quasicrystal, mobile particles are isolated, while for the dodecagonal quasicrystal, pentagonal rings constitute the dynamically active sites.

The dodecagonal quasicrystal transforms into a periodic square crystal at increased temperatures. This is surprising at first, because quasicrystals are assumed to have high entropy and therefore should be increasingly preferred at higher temperatures. However, in the case of the square phase, the lack of flip entropy is compensated by the possibility of pentagonal structural defects, which are present in thermodynamic equilibrium and add to the configurational entropy. In fact, the mobility of the square phase turns out to be higher than the mobility in the quasicrystal as confirmed by the calculation of the participation ratio of the dynamic propensity.

It is illustrative to compare the dynamics of quasicrystals with the particle motion observed during the transition from a supercooled liquid to a glass. In general, the relaxation to the glassy state does not occur homogeneously, but heterogeneously over temporal and spatial ranges, which is called dynamical heterogeneity. On a local scale, phason flips strongly resemble the slow relaxation process found in glassy materials. In fact, a long-lived glassy state can be formed by the LJG system and flipping motions among local free energy minima have been observed therein as the slow β process.

VI. CONCLUSIONS

We have investigated the particle dynamics of one-component quasicrystals in two dimensions. The isotropic LJG pair potential is used as a simple model system. A decagonal quasicrystal and a dodecagonal quasicrystal are thermodynamically stabilized for two sets of parameters. The growth of the quasicrystal phases from the melt occurs via a first order phase transition with negative thermal expansion for the decagonal quasicrystal and positive thermal expansion for the dodecagonal quasicrystal. The static structure factors show a significant amount of diffuse scattering due to high particle mobility.

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