# Soft Quasicrystals:
# From competing symmetries
# to additional degrees of freedom

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# Contents

# Abstract

Quasicrystals are structures with long range orientational order but no translational symmetry. They are not only fascinating due to their aesthetic appearance that is mainly related to the rotational symmetries that can only occur in quasicrystals but not in periodic crystals, but also due to additional degrees of freedom - called phasons - that in the limit of long wavelengths do not increase the free energy and correspond to complex correlated rearrangements of the particles in the quasicrystal.

In the articles that are part of this habilitation thesis we have studied the properties of induced or intrinsic colloidal quasicrystals - usually in two dimensions - with various rotational symmetries. Different simulation methods ranging from Brownian Dynamics simulations to Event-Chain Monte Carlo algorithms as well as theoretical approaches given by the Fundamental Measure Theory of hard disks or a modified Phase Field Crystal Model for quasicrystals are employed. Furthermore, we consider both, quasicrystals that are stabilized due to isotropic interactions with incommensurate length scales as well as patchy particles where certain binding angles are preferred.

A common topic of most of the articles is the role of the additional degrees of freedom: It is either explored how particles rearrange due to given phasonic displacements or determined which phasonic excitations occur due to thermal fluctuations, during the melting, or the growth.

For example, our works on the growth process demonstrate the importance of phasons. We find that during the growth of a quasicrystal phasonic flips often are built in that later might be repaired due to phasonic rearrangements. Similarly, the strain and stress that occurs when two quasicrystals that are grown from different seed meet can be relaxed by phasonic rearrangements thus some situations avert the formation of a domain border that for periodic crystals is unavoidable. Some of our predictions have been confirmed in experiments in the meantime.

Quasicrystalline structures that occur in soft matter systems do not only represent a nice new class of ordered soft materials, but they can be employed as model systems to obtain a deeper understanding of the special and sometimes unique features that accompany aperiodic order like the additional degrees of freedom that are in the focus of this thesis.

# Zusammenfassung

Quasikristalle sind Stukturen mit langreichweitiger Ordnung und keiner Translationssymmetrie. Sie beeindrucken nicht nur durch ihr kunstvolles Aussehen, welches hauptsächlich mit dem Auftreten von Rotationssymmetrien zusammenhängt, die in periodischen Kristallen nicht erlaub wären. Sie bestechen auch mit zusätzlichen Freiheitsgraden, bekannt als Phasonen, welche als Anregung im Grenzfall langer Wellenlängen nicht die Freie Energie ändern und als komplexe, korelierte Teichenumordnungen in Erscheinung treten.

In den Artikeln, die Teil dieser Habilitationschrift sind, werden - zumeist in zwei Dimensionen - die Eigenschaften von induzierten und intrinsischen kolloidalen Quasikristallen mit unterschiedlichen Rotationssymmetrien untersucht. Es werden unterschiedliche Simulationsmethoden, von Brownschen Dynamik-Simulationen bis hin zu Ereignisketten-Monte-Carlo-Simulationen, und zusätzlich theoretische Rechnungen mittel der sogenannten Fundamental Measure-Theorie oder von Phasenfeld-Kristall-Modellen verwendet. Dabei werden sowohl Quasikristalle betrachtet, die durch isotrope Wechselwirkungen mit mehreren inkommensurablen Längenskalen stabilisiert werden, also auch Quasikristalle, die man für sogenannte Patchy-Kolloide, also Kolloide mit bevorzugten Bindungsrichtungen erhält.

Ein sich durch die meisten Artikeln ziehendes Thema ist die Bedeutung der zusätzlichen Freiheitsgrade: So wird untersucht, wie genau sich Teilchen bei einer gegebenen phasonischen Verschiebung umordnen, oder es wird bestimmt, welche phasonischen Anregungen durch thermische Fluktuationen, durch Schmelzen oder beim Wachsen angeregt werden.

Beispielsweise zeigen wir in unseren Arbeiten zum Wachstumsprozess von Quasikristallen wie wichtig Phasonen sind: Wir beobachten, dass phasonische Anregungen während des Wachstumsprozesses eingebaut werden und dass diese dann später durch phasonische Umordnungen wieder entfernt und somit die falschen Teilchenpositionen repariert werden können. In ähnlicher Weise können Verzerrungen und und damit einhergehende Verspannungen, die entstehen wenn zwei wachsende Quasikristalle aufeinander treffen, häufig durch phasonische Umordnungen abgebaut werden. In vielen Situationen kann somit die Bildung einer Domänengrenze verhindert werden, auch wenn in einer entsprechenden Situation bei periodischen Kristallen eine Domänengrenze unvermeidlich wäre.

Quasikritalline Strukturen in Sytemen der weichen Materie sind nicht nur Teil einer interessanten neuen Klasse von geordneten weichen Materialien, sondern sie können auch verwendet werden, um ein tieferes Verständnis der speziellen und manchmal einzigartigen Eigenschaften aperiodisch geordneter Systeme zu erlangen wie es in dieser Habilitationsschrift beispielweise für die zusätzlichen Freiheitsgrade geschieht.

# Chapter 1

# Background: Additional degrees of freedom in quasicrystals

The main focus of this short introduction are the additional degrees of freedom, called phasons, that occur in quasicrystals and that are important for most of the articles that are collected in this thesis. Note that the fascinating history of the discovery, the occurrence of quasicrystalline order in various systems, and a lot of interesting properties of quasicrystals besides phasons are not discussed in this introduction as these topics are covered in many publications and also in my PhD thesis [1].

I want to start with a short comment on the definition of the terms *crystal* and *quasicrystal*: Today the definition of a *crystal* includes "any solid having an essentially discrete diffraction diagram" [2] and therefore crystals can be periodic or aperiodic. In the later case they are usually called *quasicrystals*. A discussion of the definition and possible issues related to it can be found in [3].

Following the idea of Levine et al. [4] that was already worked out shortly after the discovery of quasicrystals was published [5, 6], we expand the density field of a crystalline structure in terms of Fourier modes

$$\rho\left(\vec{r}\right) = \sum_{\vec{G}_j} \rho_{\vec{G}_j} e^{i\vec{G}_j \cdot \vec{r}}, \tag{1.1}$$

where the sum is over lattice vectors $\vec{G}_j$ that are suitable to describe the symmetry of the structure, e.g., for a crystal with $N$-fold symmetry in two dimensions one might choose

$$\vec{G}_j = (\cos[2\pi j/N], \sin[2\pi j/N]) \tag{1.2}$$

with $j = 0, ..., N - 1$.

As a side note, the minimum number of vectors that are needed to obtain all lattice vectors as linear combinations is called the *rank $R$* of a crystal. For a periodic crystal the rank $R$ equals the number of dimensions $d$ while for aperiodic structures $R > d$. For example, in two dimensions the periodic crystals with 3-, 4-, or 6-fold rotational symmetry posses rank $R = 2$. The rank of the most common quasicrystals with 5-, 8-, 10-, and

12-fold rotational symmetry is $R = 4$. In general, for $N$ lattice vectors given by Eq. (1.2), the rank is $R = 2\varphi(N)$ where $\varphi(N)$ is Euler's totient function. A detailed introduction into the mathematics of aperiodic structures can, e.g., be found in the books by Baake and Grimm [7,8].

To continue to follow the idea of Levine et al. [4], we note that the Fourier components can be written in the form

$$\rho_{\vec{G}_j} = \left|\rho_{\vec{G}_j}\right| e^{i\phi_j}, \tag{1.3}$$

such that for each lattice vector a phase $\phi_j$ can be introduced. Note that for lattice vectors that are not linear independent, the phases are also related. As there is a set of $R$ linear independent lattice vectors, the phases can be written as function of a global phase $\gamma$ and $R$ additional independent variables. In case the phases are chosen as in

$$\phi_j = \vec{G}_j \cdot \vec{u} \tag{1.4}$$

they lead to a displacement of the system by a vector $\vec{u}$. Therefore, of the $R$ independent variables $d$ can be chosen by the components of the vector $\vec{u}$ and there $R - d$ degrees of freedom remain. While for periodic crystals $R = d$ such that there are no additional degrees of freedom, for quasicrystals $R > d$.

In general, the vector composed of the phases $\vec{\phi} = (\phi_1, \phi_2, ...\phi_N)$ can be written as

$$\vec{\phi} = \mathbf{M}\vec{v}, \tag{1.5}$$

where $\vec{v}$ is a vector with $R$ components. As explained before, the first $d$ components of $\vec{v}$ are usually chosen to be the components of the displacement vector $\vec{u}$, i.e., $\vec{v} = (u_1, ..., u_d, w_1, ..., w_{R-d})$ where $w_1$ to $w_{R-d}$ denote the additional degrees of freedom. The first column of the matrix $\mathbf{M}$ is given by the first components of the lattice vectors, the second column by the second component etc.. The remaining $R - d$ columns usually are chosen such, that all columns of $\mathbf{M}$ are perpendicular to each other. Then $w_1$ to $w_{R-d}$ lead to changes of the phases and thus the density field $\rho(\vec{r})$ that are independent of the displacements according to $\vec{u}$ and if seen as part of an $R$-dimensional space also perpendicular to displacements along the components of $\vec{u}$ which correspond to *phonons* in the long wavelength limit. The additional degrees of freedom $w_1$ to $w_{R-d}$ usually are called *phasons*. Note that all components of $\vec{v}$ might be chosen as position-depended fields such that phononic or phasonic strain or phonons and phasons with finite wavelength can be introduced. In the following we will shortly discuss some of the properties of the phasons for the example of a quasicrystal with 5- or 10-fold rotational symmetry.

We now consider the case of a decagonal symmetry that can be described by lattice vectors

$$\vec{G}_j = (\cos[2\pi j/5], \sin[2\pi j/5]) \tag{1.6}$$

for $j = 1, 2, 3, 4$, or 5. The phononic displacement field $\vec{u}(\vec{r})$ and the phasonic displacement field $\vec{w}(\vec{r})$ can be chosen such that the phases are [4]

$$\phi_j(\vec{r}) = \vec{G}_j \cdot \vec{u}(\vec{r}) + \vec{G}_{3j \bmod 5}\vec{w}(\vec{r}) + \gamma, \tag{1.7}$$

where $3j \bmod 5$ is the remainder of a division of $3J$ by 5 such that the phasonic displacements are indeed perpendicular in the sense explained in the last paragraph. Note that in contrast to phononic or phasonic displacements in the long wavelength limit the global phase $\gamma$ changes the *local isomorphism* class which. A *local isomorphism* class is defined such that all arbitrary large but finite patches of a tilings given by a structures within a class are contained somewhere in the other tilings that are in the same class [9,10]. In other words, the global phase $\gamma$ changes the type of the quasicrystal while global phononic or phasonic displacements lead to patterns that correspond or at least are arbitrarily close to a displaced version of the original pattern. In case of continuous quasicrystalline patterns arbitrarily close means that a displacement can be found such that the integral over the differences are smaller than any arbitrarily chosen value.

The free energy can be expanded with respect to the Fourier modes. As a consequence, one can show that the free energy does not change if the global phononic or phasonic displacement is applied [4,11]. On the other hand, local phononic or phasonic displacements lead to an increase of the free energy. Up to second order the increase of the free energy can be written as function of the phononic and phasonic strain [4].

While phononic excitations lead to diffusive as well as propagating modes, it is argued that phasonic modes are always diffusive [11].

In this thesis we present studies of how particles are actually displaced if the phasonic displacement field changes [B1-B4,B6]. In addition to the case of decagonal quasicrstals [B1] that also was considered here, we also explain the phasonic modes in the other two dimensional quasicrystals with rank $R = 4$ [B3] and for quasicrystals with 14-fold rotational symmetry as an example of a rank $R = 6$-quasicrystals where 4 independent phasonic displacement directions exist [B4].

Note that in most of the other works that are part of this thesis the additional degrees of freedom somehow affect the properties of the system, e.g., the stability of a quasicrystals [B5,D1], the ordering on substrates [C1], and the growth of quasicrystals [E2-E5]. Therefore, the phasonic degrees of freedom are probably the most important reason why some properties of quasicrystals reported in the articles differ from what is expected in periodic crystals.

# Chapter 2

# Methods

In this chapter the methods that are used in the articles in this thesis are shortly summarized.

## 2.1 Brownian Dynamics Simulations

In Brownian dynamics simulations the overdamped Langevin equation is numerically integrated. The Langevin equation is a stochastic differential equation that can be obtained from Newton's equation. Including inertia the Langevin equation is [12] (a nice overview of Langevin dynamics can, e.g., be found in [13])

$$m\frac{\partial}{\partial t}\mathbf{v} + \gamma\mathbf{v} = \mathbf{F}_{int} + \mathbf{F}_{ext} + \mathbf{F}_{therm} \tag{2.1}$$

where $\mathbf{v}$ is the velocity of a particle, $m$ is its mass, and $\gamma$ the friction constant, e.g., $\gamma = 6\pi R\eta$ for a sphere with radius $R$ in a viscous medium with viscosity $\eta$. $\mathbf{F}_{int}$ denotes the total force due to internal interactions, $\mathbf{F}_{ext}$ the one due to external forces, and $\mathbf{F}_{therm}$ should effectively describe all forces due to collisions with particles of the surrounding media that move on time-scales that are much shorter than the time-scale that is of interest to describe the motion of the (much larger) particle described by the Langevin equation. Usually $\mathbf{F}_{therm}$ is realized by random kicks that are chosen such, that the first and second moments of the components $F_{therm,j}$ fulfill the following relations:

$$\langle F_{therm,j}(t)\rangle = 0, \tag{2.2}$$
$$\langle F_{therm,j}(t)F_{therm,k}(t')\rangle = 2k_BT\gamma\delta_{jk}\delta(t-t'), \tag{2.3}$$

i.e., on average the thermal force is zero and kicks in different directions or at different times are uncorrelated. Furthermore, the mean squared magnitude of a kick is chosen such that in the case of no additional internal or external forces the mean squared velocity in $n$ dimensions is

$$\langle\mathbf{v}(t)^2\rangle = \frac{nk_BT}{m}, \tag{2.4}$$

which is in agreement with the equipartition theorem. Note that the mean square displacement is

$$\left\langle (\mathbf{r}(t) - \mathbf{r}(0))^2 \right\rangle = 2nDt \qquad (2.5)$$

with the diffusion constant

$$D = \frac{k_B T}{\gamma}. \qquad (2.6)$$

This relation is the well-known Stokes-Einstein relation.

In the case of small particles and for times larger than $m/\gamma$ the inertial term $m\frac{\partial}{\partial t}\mathbf{v}(t)$ can be neglected. This leads to the so-called overdamped Langevin equation

$$\gamma\mathbf{v} = \mathbf{F}_{int} + \mathbf{F}_{ext} + \mathbf{F}_{therm} \qquad (2.7)$$

that is usually used in Brownian dynamics simulation by discretizing it and integrating it using basic Euler integration. The thermal force is given by random numbers that are chosen such that the condition (2.2) and (2.3) hold.

Brownian dynamics simulations are used to simulate the overdamped motion of particles in solution.

## 2.2 Metropolis Monte Carlo Simulations

Monte-Carlo algorithms usually are used to obtain statistical properties of a system. In the standard implementation introduced by Metropolis et al. [14] the canonical ensemble is considered where the probability of single configurations is given by the Boltzmann factor. The goal of such Monte Carlo simulations is to sample over enough states with the correct probabilities such that mean values of observables can be determined.

The configurations that are considered are determined randomly. Usually this is achieved by taking a previous configuration, choosing one particle randomly and displacing it randomly. The new configuration is accepted if the energy $E_{new}$ is smaller than the energy of the old configuration $E_{old}$ or in case the energy is increased if a randomly chosen number is smaller than

$$\exp\left(-\beta[E_{new} - E_{old}]\right) \qquad (2.8)$$

with $\beta = 1/(k_B T)$.

It can be shown that the Metropolis algorithm obeys detailed balance such that the configurations a sampled according to the Boltzmann distribution of the canonical ensemble in equilibrium. For more background on the Metropolis algorithm and the most common variations, see also the textbooks [15, 16].

Note that detailed balance is sufficient for reaching thermal equilibrium but it is not required as we will see in the next section where an alternative Monte Carlo approach is introduced.

## 2.3   Event-Chain Monte Carlo Simulations

While in most simulations based on the Metropolois algorithm single particles are displaced in each step, it is also possible to displace or rotate clusters of particles. If neighboring particles are displaced as well, this can even be done in a way that hardly any or no Monte-Carlo step will be rejected [17–19].

Another rejection-free Monte Carlo method was realized by considering event chains [20]. In case of a hard spheres system first a direction and a length is randomly selected then a random particle is displaced in the chosen direction until it hits another particle. Upon collision the first particle stops and the collision partner is newly selected and displaced along the chosen direction until another collision occurs. The process stops if the total discplacement of the selected particles reaches the previously chosen length. Note that no displacement is rejected. While this event chain approach violates detailed balance it can me shown that global balance is obbeyed and that as a consequence the algorithm converges against equilibrium if the system is ergodic [21,22]. The convergence usually is much faster that for the conventional Metropolis algorithm [23].

An event-chain approach with rejection-free implementation can also be introduced for soft particles [24,25] and in the article on the melting of quasicrystals [D1] that is part of this thesis, we have used the method for an even more complex interaction potentials that might include attractions.

## 2.4   Phase Field Crystal Model

Phase Field Crystal (PFC) models based on the expansion of a free energy functional with respect to a density-like field and its gradient [26,27]. This approach is motivated by the model used by Swift and Hohenberg [28] and similar to the free energy expansion by Alexander and McTague [29] and especially variations thereof as in [30]. In case of two lengths as used in the articles in this theses the method is equivalent (up to density conservation during minimization) to the model that has been introduced by Lifshitz and Petrich to describe Faraday waive patterns [31].

The standard PFC free energy that can explain the formation of a periodic hexagonal crystal (in case of a 2D system) is given by [26,27]

$$F[\psi(\vec{r})] = \int d\vec{r} \left[ \frac{1}{2} \psi(\vec{r}) \left\{ -\epsilon + \left( k^2 + \nabla^2 \right)^2 \right\} \psi(\vec{r}) + \frac{1}{4} \psi(\vec{r})^4 \right], \qquad (2.9)$$

where $\epsilon$ is a control parameter that roughly can be interpreted as a temperature and $2\pi/k$ gives the length scale of the structure that should form. Furthermore the mean density $\overline{\psi}$ can be used as control parameter that even is fixed if density-conserved dynamics is used as with with the overdamped diffusion-like equation [26]

$$\frac{\partial \psi(\vec{r}, t)}{\partial t} = \nabla^2 \left[ \frac{\delta F[\psi(\vec{r}, t)]}{\delta \psi(\vec{r}, t)} \right]. \qquad (2.10)$$

In order to obtain quasicrystalline patterns, two or more length scales can be introduced in the free energy functional, i.e.,

$$F[\psi(\vec{r})] = \int d\vec{r} \left[ \frac{1}{2}\psi(\vec{r}) \left\{ -\epsilon + \prod_{j=1}^{m} \left( k_j^2 + \nabla^2 \right)^2 \right\} \psi(\vec{r}) + \frac{1}{4}\psi(\vec{r})^4 \right], \qquad (2.11)$$

where $2\pi/k_1$, $2\pi/k_2$, ..., $2\pi/k_m$ are $m$ length scales.

This free energy will be used in the articles in this thesis. Note that such a similar free energy previously has been studied in [32] and in the framework of the Lifshitz-Petrich model in [31] where often non-conserved dynamics has been studied. The Lifshitz-Petrich model for quasicrystals patterns has also been studied, e.g., in [33–35]. Finally, a quasicrystal obtained by a PFC approach in three dimensions has been presented in [36].

Beyond the scope of this thesis, we have introduced a PFC model for active particles with inertia [37,38]. Furthermore, we have developed a PFC model of patchy particles [39].

The PFC free energy expansion can be derived from microscopic theories like density functional theories [40,41] that in modified versions can also lead to quasicrystalline patterns [42,43]. However, in this thesis we do not want to discuss density functional theories for particles with complex pair interactions. However, in the next section a special classical density functional theory for hard spheres or disks is introduced that we have used to study substrate-induced structures.

## 2.5  Fundamental Measure Theory

A very successful classical density functional theory for hard spheres was proposed by Rosenfeld [44] and is known as Fundamental Measure Theory. The excess part of the free energy functional, i.e., the part that goes beyond the ideal gas free energy, can be rewritten by decomposing the Mayer-$f$-function (here for hard spheres, where the spheres of species $j$ have radius $R_j$)

$$f_{jk}(r) = exp(-\beta V_{jk}(r)) - 1 = -\Theta\left( R_j + R_k - r \right) \qquad (2.12)$$

in terms of so-called weight functions

$$-f_{jk}(r) = \Theta(R_j + R_k - r) = \omega_j^{(3)} \otimes \omega_k^{(0)} + \omega_j^{(0)} \otimes \omega_k^{(3)} + \omega_j^{(2)} \otimes \omega_k^{(1)} + \omega_j^{(1)} \otimes \omega_k^{(2)} + \vec{\omega}_j^{(2)} \otimes \vec{\omega}_k^{(1)} + \vec{\omega}_j^{(1)} \otimes \vec{\omega}_k^{(2)},$$
$$(2.13)$$

where $\otimes$ denotes cross correlation given by $f \otimes g(r_j - r_k) = \int d^3r' \, f(r' - r_j) \, g(f' - r_k)$, The weight functions are given by

$$\omega_i^{(3)}(r) = \theta(R_i - r),$$
$$\omega_i^{(2)}(r) = \delta(R_i - r),$$
$$\omega_i^{(1)}(r) = \frac{1}{4\pi R_i}\omega_i^{(2)}(r)(r) = \frac{1}{4\pi R_i}\delta(R_i - r),$$
$$\omega_i^{(0)}(r)(r) = \frac{1}{4\pi R_i^2}\omega_i^{(2)}(r)(r) = \frac{1}{4\pi R_i^2}\delta(R_i - r),$$
$$\vec{\omega}_i^{(2)}(r)(r) = \frac{\vec{r}}{r}\delta(R_i - r),$$
$$\vec{\omega}_i^{(1)}(r)(r) = \frac{1}{4\pi R_i}\vec{\omega}_i^{(2)}(r) = \frac{1}{4\pi R_i}\frac{\vec{r}}{r}\delta(R_i - r), \qquad (2.14)$$

and can be seen as functions that measure geometric features, i.e., $\omega_j^{(0)}$, $\omega_j^{(1)}$, $\omega_j^{(2)}$, $\omega_j^{(3)}$ denote scalar properties like the surface area or the volume and for the volume and the vector weight functions $\vec{\omega}_j^{(1)}$ and $\vec{\omega}_j^{(2)}$ similar vector properties (for details, cf. [44]).

Then weighted density functions are introduced:

$$n^{(\nu)}(\vec{r}) = \sum_{i=1}^{m} \int d\vec{r}' \rho_i(\vec{r}')\omega_i^{(\nu)}(\vec{r} - \vec{r}') \qquad (2.15)$$

Finally, the excess free energy density functional $F_{exc}$ is expressed in terms of such weighted density functions

$$\beta F_{exc}[\{\rho_i\}] = \int d\vec{r}\Phi(\{n_\nu(\vec{r})\}), \qquad (2.16)$$

where

$$\Phi(\{n_\nu(\vec{r})\}) = f_1(n_3)n_0 + f_2(n_3)n_1 n_2 + f_3(n_3)\vec{n}_1 \cdot \vec{n}_2 + f_4(n_3)n_2^3 + f_5(n_3)n_2\vec{n}_2 \cdot \vec{n}_2. \quad (2.17)$$

If the the prefectors $f_j(n_3)$ are chosen such that the pressure at low densities can be correctly obtained as it is done by Rosenfeld in [44] the approach cannot explain the liquid to solid transition. Therefore, modifications have been introduced, e.g., [45–47]). For example, tensor weights can be introduced or the the prefactors can be chosen such, that the equation of state is well described. One of the best approaches up to today is the so-called White Bear II version [47]. This model has been successfully used to predict the properties of the solid fluid interface in coexistence [48, 49]. Furthermore, we have calculated higher order correlations of particles close to a wall and have compared them to Brownian Dynamics simulations [50, 51].

An interesting extension that will not be discussed here is a Fundamental Measure Theory for convex non-spherical particles [52, 53].

In this thesis a two-dimensional version of the Fundamental Measure Theory is used as proposed in [54]. Note that in two dimensions there is no exact decomposition of the Mayer-$f$-function, but only an approximate one (motivated in [54] with the Gauss-Bonnet theorem):

$$-f_{ij}(r) \approx \omega_i^{(2)} \otimes \omega_i^{(0)} + \omega_i^{(0)} \otimes \omega_i^{(2)} + C_0 \omega_i^{(1)} \otimes \omega_i^{(1)} + C_1 \vec{\omega}_i^{(1)} \otimes \vec{\omega}_i^{(1)} + C_2 \hat{\omega}_i^{(1)} \otimes \hat{\omega}_i^{(1)} + ... \quad (2.18)$$

with

$$\begin{aligned}
\omega_i^{(2)}(r) &= \theta(R_i - r), \\
\omega_i^{(1)}(r) &= \delta(R_i - r), \\
\omega_i^{(0)}(r) &= \frac{1}{2\pi R_i} \omega_i^{(1)}(r) = \frac{1}{2\pi R_i} \delta(R_i - r), \\
\vec{\omega}_i^{(1)}(r) &= \frac{\vec{r}}{r} \omega_i^{(1)}(r) = \frac{\vec{r}}{r} \delta(R_i - r), \\
\hat{\omega}_i^{(1)}(r) &= \frac{\vec{r}\vec{r}}{r^2} \omega_i^{(1)}(r) = \frac{\vec{r}\vec{r}}{r^2} \delta(R_i - r).
\end{aligned} \quad (2.19)$$

The weighted density functions can be introduced in the same way as in 3D and the excess free energy is given by

$$\Phi = -n_0 \ln(1 - n_2) + \frac{1}{4\pi(1 - n_2)} \left[ \tilde{C}_0 (n_1)^2 + \tilde{C}_1 \vec{n}_1 \cdot \vec{n}_1 + \tilde{C}_2 \mathrm{Tr}(\hat{\mathbf{n}}_1^2) \right]. \quad (2.20)$$

where suitable choices for the prefactors are [54]:

$$\tilde{C}_0 = \frac{b+2}{3}, \qquad \tilde{C}_1 = \frac{b-4}{3}, \text{ and } \tilde{C}_2 = \frac{2-2b}{3}, \quad (2.21)$$

where $b = \frac{11}{4}$ leads to a good approximation of the Mayer-$f$-function [54]. This two dimensional excess free energy is used in the articles that are included in this thesis.

Similar to dynamical PFC models a density conserved overdamped dynamics can be considered for density functional theories like the Fundamental Measure Theory (for a review see, e.g., [55]). For the applications that are presented in the articles in this thesis, we have also written a short review [56] that also discusses our result concerning hard spheres on periodic substrates [57] or the study of growing crystals from two seeds [58].

# Chapter 3

# Short Overview of the articles in this thesis

The articles listed in the following are included in full text in this thesis. Note that only articles that fit into the framework of this thesis are considered. Some additional articles that are somehow slightly related are not included, e.g., the work on paths in a quasicrystalline Lorenz gas [59], on the ordering or dynamics on incommensurate periodic substrates [56, 57, 60], on phases due to competing interactions [61], or on the growth process from multiple seeds [58]. Furthermore, articles like [30, 62–64] that have already been discussed in my PhD thesis [1] are not part of this thesis. A complete list of my publications can be found via the following link:
`theorie1.physik.uni-erlangen.de/people/mschmiedeberg/publications.html`

I also want to mention that several theses in the field of quasicrystals have been written under my supervision. This includes PhD theses [65, 66], Master theses [67–70], and Bachelor theses [71–77]. Some of the articles considered here have originated in the work on one of these theses. A list of all theses in my group (not necessarily related to quasicrystals) can be found via the following link:
`theorie1.physik.uni-erlangen.de/people/mschmiedeberg/theses.html`

## 3.1   A: Articles on Quasicrystalline Patterns

First, some articles related to laser fields with quasicrystalline symmetry are considered. Such laser fields had already been used before to study laser induced colloidal structures.

[A1] J. Mikhael, M. Schmiedeberg, S. Rausch, J. Roth, H. Stark, and C. Bechinger, *Proliferation of anomalous symmetries in colloidal monolayers subjected to quasiperiodic light fields*, PNAS **107**, 7214 (2010)

[A2] M. Schmiedeberg and H. Stark, *Comparing light-induced colloidal quasicrystals*

*with different rotational symmetries*, J. Phys.: Condens. Matter **24**, 284101 (2012)

[A3] F. Rühle, M. Sandbrink, H. Stark, and M. Schmiedeberg, *Effective substrate potentials with quasicrystalline symmetry depend on the size of the adsorbed particles*, Eur. Phys. J. E **38**, 54 (2015)

## 3.2   B: Articles on Phasons

To identify how particles in a quasicrystal move or even jump due to a change in the phases related to the additional degrees of freedom, also called phasons, we have exemplary studied how colloidal particles on quasicrystalline substrates are displaced in case of a phasonic drift.

[B1] J.A. Kromer, M. Schmiedeberg, J. Roth, and H. Stark, *What Phasons Look Like: Particle Trajectories in a Quasicrystalline Potential*, Phys. Rev. Lett. **108**, 218301 (2012)

[B2] J.A. Kromer, M. Schmiedeberg, J. Roth, and H. Stark, *Phason-induced dynamics of colloidal particles on quasicrystalline substrates*, Eur. Phys. J. E **36**, 25 (2013)

[B3] M. Sandbrink and M. Schmiedeberg, *Trajectories of colloidal particles in laser fields with eight-, ten-, or twelve-fold symmetry and phasonic drift*, in *Aperiodic Crystals*, edited by S. Schmid, R.L. Withers, and R. Lifshitz, (Springer, Berlin) (2013)

[B4] M. Martinsons, M. Sandbrink, and M. Schmiedeberg, *Colloidal Trajectories in Two-Dimensional Light-Induced Quasicrystals with 14-Fold Symmetry due to Phasonic Drifts*, Acta Physica Polonica A **126**, 568 (2014)

[B5] M. Sandbrink, J. Roth, and M. Schmiedeberg, *Comment on "Quantum Quasicrystals of Spin-Orbit-Coupled Dipolar Bosons"*, Phys. Rev. Lett. **113**, 079601 (2014)

[B6] M. Martinsons and M. Schmiedeberg, *Stability of particles in two-dimensional quasicrystals against phasonic perturbations*, J. Phys.: Conf. Ser. **1458**, 012019 (2020)

## 3.3   C: Articles on Competing Symmetries

As quasicrystalline patterns intrinsically are incommensurate to periodic structures, we previously had studied what structures are formed by colloids that want to arrange in a periodic way when they are placed on a quasicrystalline substrate. Here we present additional works along this line. Note that the focus is on quasicrystalline substrates and therefore works where only incommensurate periodic structures are considered are not included in this thesis.

[C1] M. Schmiedeberg, J. Mikhael, S. Rausch, J. Roth, L. Helden, C. Bechinger, and H. Stark, *Archimedean-like colloidal tilings on substrates with decagonal and tetradecagonal symmetry*, Eur. Phys. J. E **32**, 25 (2010)

[C2] T. Neuhaus, M. Schmiedeberg, and H. Löwen, *Compatibility waves drive crystal growth on patterned substrates*, New Journal of Physics **15**, 073013 (2013)

## 3.4 D: Articles on the Stability and Melting of Soft Quasicrystals

In this section we discuss both quasicrystals stabilized to two incommensurate length scales in the (isotropic) interaction potential and quasicrystals that occur due to patches on the colloidal particles. We study the stability or the onset of melting in such structure. The additional degrees of freedom often are of great importance for these processes.

[D1] M. Martinsons, J. Hielscher, S.C. Kapfer, and M. Schmiedeberg, *Event-chain Monte Carlo simulations of the liquid to solid transition of two-dimensional decagonal colloidal quasicrystals*, J. Phys.: Condens. Matter **31**, 475103 (2019)

[D2] J. Hielscher, M. Martinsons, M. Schmiedeberg, and S.C. Kapfer, *Detection of phonon and phason modes in intrinsic colloidal quasicrystals by reconstructing their structure in hyperspace*, J. Phys.: Condens. Matter **29**, 094002 (2017)

[D3] J. Hielscher, M. Martinsons, M. Schmiedeberg, and S.C. Kapfer, *Phasonic Diffusion and Self-confinement of Decagonal Quasicrystals in Hyperspace*, J. Phys.: Conf. Ser. **1458**, 012018 (2020)

[D4] A. Gemeinhardt, M. Martinsons, and M. Schmiedeberg, *Stabilizing quasicrystals composed of patchy colloids by narrowing the patch width*, EPL **126**, 38001 (2019)

## 3.5 E: Articles on the Growth of Soft Quasicrystals

In the final chapter, we study the growth of soft quasicrystals. We find that the phasonic degrees of freedom have an important impact on the growth, either due to defects or phasonic excitations that might occur in the grown structures or for the repair mechanism after the growth where strain is reduced by phasonic rearrangements.

[E1] M. Sandbrink and M. Schmiedeberg, *Course of dislocation lines in templated three-dimensional colloidal quasicrystals*, Phys. Rev. B **90**, 064108 (2014)

[E2] C.V. Achim, M. Schmiedeberg, and H. Löwen, *Growth Modes of Quasicrystals*, Phys. Rev. Lett. **112**, 255501 (2014)

[E3] M. Martinsons and M. Schmiedeberg, *Growth of two-dimensionalcolloidal quasicrystals*, J. Phys.: Condens. Matter **30**, 255403 (2018)

[E4] A. Gemeinhardt, M. Martinsons, and M. Schmiedeberg, *Growth of two-dimensional dodecagonal colloidal quasicrystals: Particles with isotropic pair interactions with two length scales vs. patchy colloids with preferred binding angles*, Eur. Phys. J. E **41**, 126 (2018)

[E5] M. Schmiedeberg, C.V. Achim, J. Hielscher, S.C. Kapfer, and H. Löwen, *Dislocation-free growth of quasicrystals from two seeds due to additional phasonic degrees of freedom*, Phys. Rev. E **96**, 012602 (2017)

# Chapter 4

# Quasicrystalline Laser Fields (related to Articles A)

In this and the following sections, the main results of the articles are put into the overall context of the thesis. Note that it is not the purpose of these sections to summarize all results as this is already done in the abstracts of the articles. The main focus of these sections lies on the connections between the articles and the overall picture of what has been achieved in our research.

The articles that are grouped in part A closely follow the research presented in my PhD-thesis [1] where I have studied colloidal dynamics and ordering in laser fields with quasicrystalline symmetry. Interfering lasers can be used to create laser fields that act as a substrate to colloidal particles and can be described by the following potential [62, 78]

$$V\left(\vec{r}\right) = -\frac{V_0}{N^2} \sum_{j=0}^{N} \sum_{k=0}^{N} \cos\left[\left(\vec{G}_j - \vec{G}_k\right) \cdot \vec{r} + \phi_j - \phi_k\right], \tag{4.1}$$

where $N$ gives the number of laser beams, $V_0$ is the maximum strength of the potential, the vectors $\vec{G}_j$ are given as in (1.2), and $\phi_j$ are phases that can be used to built-in phononic or phasonic displacements as explained in Chapter 1.

Such interference patterns have been successfully used to induce quasicrystalline order to colloidal particles in experiments (see, e.g., [79, 80]). Note that similar laser fields can be used as traps for cold atoms to induce quasicrystalline symmetry [81, 82].

After we had studied the dynamics of a colloidal particles on a quasicrystalline substrate [62] and for multiple colloids the orderings that occurs due to the competition between triangular structures that are preferred by colloids and the quasicrystalline laser field [63], we investigated what rotational symmetries can be induced best with laser fields [A1,A2] and how the size of the colloids influences the patterns [A3]. Questions related to competing symmetries were further explored in the articles presented in part C.

In [A1] we compare results from experiments and from simulations concerning colloidal ordering in laser fields that are obtained as interference patterns of 5 or 7 laser beams. The laser field created by 5 beams is typical for rank $R = 4$ quasicrystals, while the pattern

obtained with 7 beams possesses rank $R = 6$. These patterns are used to find out how efficient quasicrystalline symmetry can be induced depending on the rank. In [A2] extended simulations for other numbers of beams are presented proving that indeed the rank is the quantity that matters, i.e., patterns with the same rank behave similar concerning how colloidal structures with quasicrystalline symmetry are obtained.

Both in [A1] and [A2] we are able to identify the local symmetry centers - sometimes also termed flowers - as the parts of the substrate where the colloids adopt the quasicrystalline order first. The differences that are observed for patterns with different rank originate in the fact that there are less local symmetry centers in quasicrystals with larger rank. In summary, we find that the larger the rank the smaller is the number of local symmetry centers and the harder it is to induce colloidal quasicrystals, i.e., the potential strength has to be larger to observe quasicrystalline structures at all.

Our results suggest that local symmetry centers are important for the formation of quasicrystals and are in agreement with the observation that quasicrystals with smaller rank seem to be occur more likely than quasicrystals with large rank and that it is a hard task to obtain quasicrystals with rank $R \leq 6$ (in two dimensions or consisting of such layers) [83]. Note that even for the rank $R = 4$-quasicrystals there are differences concerning how easy they can be obtained (see, e.g., for extended calculations with the Lifshitz-Petrich approach: [35]). Furthermore, in soft matter systems 12-fold structures seem to occur more often than other rank $R = 4$-symmetries and 18-fold symmetries (with rank $R = 6$) are possible as well though usually not as perfect structures [84, 85].

In [A3] another question is studied: We explore how the size of a colloid affects the actual structure of induced quasicrystals given that the forces exerted by the laser beams not only depend on the laser intensity (or its gradient) in the center of a colloid but that all parts of the laser field that go through the particle matter. As a consequence, for a given laser fields different structures can be induced depending on the diameter of the colloids. In [A3] we show that these structures belong to different local isomorphism classes, i.e., they posses the same rotational symmetry but nevertheless repesent significantly different structures. Note that the term *local isomorphism class* is explained in chapter 1.

Therefore, laser fields can be used as a model system to understand the differences between quasicrystals with different rotational symmetries or different local isomorphism classes.

# Chapter 5

# Additional Degrees of Freedom in Quasicrystals (related to Articles B)

In the articles that are included in part B the properties of the additional degrees of freedom - also termed phasons - are studied. Usually the laser fields in two dimensions that are used to induce quasicrystalline colloidal structures (see articles of part A) are considered. As explained in the last chapter, such laser fields correspond to a substrate with a potential given in (4.1). The phases $\phi_j(\vec{r})$ can be chosen depending on a global phase $\gamma$, a phasonic displacement field $\vec{u}(\vec{r})$, and $R/2 - 1$ phasonic displacement fields $\vec{w}_1(\vec{r})$, $\vec{w}_2(\vec{r})$, ..., $\vec{w}_{R/2-1}(\vec{r})$ (corresponding to $R - 2$ phasonic components where $R$ is the rank) in the same way as outlined in chapter 1.

Different quasicrystals are considered in the articles: In [B1,B2] a quasicrystal with 10-fold rotational symmetry that is obtained with an interference pattern of 5 laser beams is considered. In [B3] we study the other rotational symmetries that posses rank $R = 4$, namely also the patterns that are induced by 8 or 12 beams. Finally, in [B4] a quasicrystal that is obtained with 7 laser beams and thus possesses rank $R = 6$ is considered. In all articles [B1-B4] we explore how colloidal particles move in the laser fields in case a global phasonic displacement is changed at a constant rate in time. These colloidal trajectories illustrate what kind of particle rearrangements correspond to phasonic excitations.

A special focus in [B1,B3,B4] lies on the point where particles jump from one position to another one due to a infinitesimal increase of one phasonic component. Such jumps obviously do not occur if components of the phononic displacement are changed and thus are a specific feature of phasons in quasicrystals.

While [B1,B3,B4] deal with idealized trajectories at zero temperature, we study the colloidal structures and motion at a finite temperature in [B2] with Brownian dynamics simulations.

[B5] is a comment on a letter by Gopalakrishnan, Martin, and Demler [86] on excitations in a quantum quasicrystals consisting of coupled bosons. Due to a small energy gap these excitations were incorrectly called phasons and it was claimed that there are more phasons in a quantum crystal than in a classical one. In our comment [B5] we explain that the phasonic degrees of freedom are a consequence of the symmetry when expanding

a density field (no matter whether it is classical or quantum). There is no difference between the number of phasonic modes in a classical system and phasonic modes in a quantum system. The excitations reported in [86] can also be found in classical systems. We show that they change the local isomorphism class and thus cannot be phasons (that conserve the local isomorphism class as explained in chapter 1). Note that another comment on [86] was written by Ron Lifshitz [87] where the same conclusions are reached thought a different approach via a free energy expansion is presented. The comments can be seen as a clarification of how phasonic excitations differ from other excitations and that the concept of phasons - and as a consequence the results of the other articles in part B - can be easily transferred from classical model systems to other systems like quantum quasicrystals.

The methods that we use in [B1,B3,B4] can also be employed to predict which particles in a quasicrystal will jump first upon a change of the phasonic displacement field [B6]. It is possible to determine the stability of certain parts of a quasicrystal that consists of particles that are likely to jump to another location upon a small thermal activation and particles that hardly move like the particles in the local symmetry centers. Therefore, local symmetry centers again turn out to be essential to stabilize the structure of a quasicrystal in support what we already have found out in [A1,A2].

The properties of the phasonic jumps that we have investigated in the articles of part B are important for the stability of quasicrystals (see articles of part D) and to understand the rearrangements during the growth of a quasicrystal (see articles of part E).

# Chapter 6

# Competing Symmetries (related to Articles C)

Charge-stabilized colloidal particles with purely repulsive pair interactions usually want to organize in a trangular crystal in two dimension. When subjected to a substrate that is incompatible to such a triangular structure it is a natural question to ask which structure will prevail. And sometimes it is not that one of structures (i.e., the triangular preferred by the colloids or the substrate structure) just wins the competition, but new structures might arise. As a quasicrystalline substrate always is incommensurate to the (periodic) triangular structure of the colloids and as even the rotational symmetries can be incompatible, the competition arising due to colloidal particles on a quasicrystalline order is non-trivial. In my PhD-thesis [1] I had studied the phases that arise for colloidal particles on a substrate with 10-fold rotational symmetry [64]. In addition to triangular and decagonal phases, we also report a phase with 20-fold rotational symmetry as well as a phase that is close to an Archimedean tiling. The later had already been observed in experiments [80].

A substrate consisting of attracting lines can be used as well and an incommensurate situation can be achieved by either putting the lines at distances according to a one-dimensional quasicrystal [30].

After my PhD I extended the study of structures due to incompatible substrates. For example, in case of the system with attractive lines one can varying the number density in the lines and consider the case where particle cannot jump from one line into another one [60]. Interestingly the energy cost of a defects does not vanish continuously if the system is tuned from a disordered state into the ordered, triangular state.

And there are many examples with competing symmetries. For example, we published results on the competition between incommensurate periodic structures. For example, if hard disks that want to order in a triangular way are put onto a square lattice, we discovered rhombic ordering [57] both in simulations and by using Fundamental Measure Theory.

In the articles that are included as part C in this thesis, [C1] and [C2], a quasicrystalline substrate is considered (at least in a part of the article).

In [C1] we explore the Archimedean-like tiling phases that occur on quasicrystalline

substrates with different symmetries. Both simulation results as well as results from experiments by the group of Clemens Bechinger (then Universität Stuttgart, now Universität Konstanz) are presented. Within specific density ranges we were able to observe these phases for various rotational symmetries. The Archimedean-like tiling consist of rows of squares and rows of triangles. While in a perfect Archimedean tiling one square row and one row of triangles would alternate, we find an aperiodic sequence of the rows reflecting the quasi-periodicity of the substrate. Even phasonic rearrangements of these rows can be observed that are similar to phasonic flips in one-dimensional quasicrystals.

Another type of competition is studied in [C2] where we use Dynamical Fundamental Measure Theory to determine how a triangular crystal composed of hard disks growth on an incommensurate substrate. We find that first positions are fixed that are compatible to both the substrate and the triagular crystal. Filling up the gaps in between these positions then occurs at a later time.

The works that employ Fundamental Measure Theory are also included in our short review of this method for systems on substrates [56].

# Chapter 7

# Stability and Melting of Quasicrystals (related to Articles D)

In the articles of part D quasicrystals that are stable due to the interactions between the constituent particles are considered. In [D1-D3] the quasicrystal is stabilized by a pair interaction potential that was introduced in [88] and possesses two incommensurate length scales. In contrast, the pair interaction potential of the particles in [D4] supports only one length directly. However, we consider preferred binding angles such that the interaction potential in [D4] can be seen as model for patchy colloids in two dimensions with symmetrically distributed patches (motivated by [89, 90]).

The focus of the articles in part D is to understand the stability given that there are important differences between aperiodic and periodic structures like the additional degrees of freedom.

In [D1] we employ Event-Chain Monte Carlo simulations to study the liquid to solid transition of a decagonal quasicrystal. For periodic crystals, melting in two dimensions can occur via an intermediate phase called the hexatic phase [91–94] and a similar intermediate phase has been predicted for quasicrystals [95–98]. According to this so-called Kosterlitz-Thouless-Halperin-Nelson-Young-theory (KTHNY) during the melting process first dislocation pairs are created that then dissociate at the solid to hexatic transition. At the hexatic to fluid transition the then isolated dislocations - correspond to pairs of disclinations - dissociate into isolated disclinations. While the description of defects and the elastic properties slightly differ between periodic and aperiodic structures, the general melting mechanism in principle might be the same in quasicrystals and periodic crystals [95–98]. However, in [D1] the KTHNY-mechanism is not observed for the melting of a quasicrystal, but instead a first order transition where melting occurs in one step is reported. However, we find a metastable intermediate phase that is due to the very slow relaxation process of phasonic excitations and that effectively possesses long-ranged orientational order despite exponentially decaying positional order. Therefore, the additional degrees of freedom are important for the melting properties.

In [D2] we describe how we reconstructed phasonic modes in an intrinsic quasicrystals and in [D3] simulations in hyperspace are used to study the diffusion in a quasicrystal due

to phasonic flips.

Finally, as mentioned above, in [D4] interactions with preferred bond directions that mimic patchy colloids in two dimensions are used to obtain meta-stable or stable quasicrystals. While structures with 12-fold rotational symmetry can be easily stabilized even for particles with 5 symmetrically arranged patches per particle (as had already been reported in [89, 90]), quasicrystals with 8- or 10-fold rotational symmetry require narrow patch widths to be stabilized.

# Chapter 8

# Growth of Soft Quasicrystals (related to Articles E)

In part E articles are presented where the growth of quasicrystals is studied.

In [E1] colloids are deposited layer by layer on a quasicrystalline substrate. We find that the quasicrystalline order can prevail for a substantial number of layers where the best results are obtained for substrates with 8-fold rotational symmetry. When we build in dislocations in the substrate we can follow dislocation lines in three dimensions and study the bending of dislocation line, their forking, and the annihilation of lines with opposite Burgers vectors. Note that dislocations in quasicrystals are accompanied with a phononic as well as a phasonic strain. Therefore, dislocations in quasicrystals are usually characterized by a Burgers vector whose number of components corresponds to the rank of the quasicrystal. Nevertheless, as we point out in [E1] many properties of the dislocation lines are similar to the properties of dislocation lines in periodic structures.

In [E2] a Phase Field Crystal approach (see also Sec. 2.4) is used to study the growth of a two-dimesnional quasicrystal from a seed. We find that close to the triple point where the fluid, the triangular, and the the quascrystalline phase coexist the grown quasicrystal is perfect, i.e., there are neither dislocations nor phasonic flips nor other defects or excitations. In contrast, if a quasicrystal is grown for parameters that are further away from the triple point, a structure is grown that does possess any dilocations. However, during the growth process a lot of phasonic flips are built in. Therefore, while the long-ranged order of the grown quasicrystal is perfect, locally there are a lot of rearrangements. In the case of a decagonal quasicrystal we even observe a two-stage growth process where first Archimedean-like tiling structures form (similar to what is described in [C1]) that later arrange into a full decagonal structure. This two-stage process is similar to what later has been observed in experiments [99].

In [E3] and [E4] we confirm our findings of [E2] with Brownian Dynamics simulations. For both pair interactions with two incommensurate length scales (as also used in [D1-D3]) and for interactions with preferred bond directions mimicking the interactions between patchy colloids [cf. [D4]) we find that the best quasicrystals are obtained for temperatures close to the solid to fluid transition. In contrast, for smaller temperatures phasonic flips

are built in during the growth and these local excitations cannot be repaired afterwards due to the too low temperature.

Finally, by using the Phase Field Crystal model, we study the growth of two quasicrystals from two seeds in [E5] and compare it to the growth of two periodic crystals. Note that we have studied a similar set-up for periodic crystals with Fundamental Measure Theory in [58]. While in case of the growth of periodic crystals a domain border forms when the two crystals meets as long as not both the distance between the seed centers is a multiple of the lattice constant and the orientation between the seed only differs by a small angle. In contrast, when two growing quasicrystals meet domain borders are observed less frequently. The reason is that phasonic rearrangements in one of the quasicrystals might occur such that its structure suits better to the other quasicrystal. While there might occur a large phasonic strain between when the two quasicrystals meet, but the additional degrees of freedom usually help to relax this strain. Recently, our findings have been confirmed in simulations and experiments [100].

In conclusions, the additional degrees of freedom and thus many of the results that have been presented in the previous parts of this thesis are important for the growth of quasicrystals. We have predicted the consequences of phasonic flips that are built in during the growth and of phasonic rearrangements that might occur as a repair mechanism. Some of our predictions have been confirmed by experiments in the meantime (e.g., [99, 100]).

# Danksagung

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# Links to the Articles

## Article [A1]

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