Dislocation-free growth of quasicrystals from two seeds due to additional phasonic degrees of freedom

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We explore the growth of two-dimensional quasicrystals, i.e., aperiodic structures that possess long-range order, from two seeds at various distances and with different orientations by using dynamical phase-field crystal calculations. We compare the results to the growth of periodic crystals from two seeds. There, a domain border consisting of dislocations is observed in case of large distances between the seed and large angles between their orientation. Furthermore, a domain border is found if the seeds are placed at a distance that does not fit to the periodic lattice. In the case of the growth of quasicrystals, we only observe domain borders for large distances and different orientations. Note that all distances do inherently not match to a perfect domain wall-free quasicrystalline structure. Nevertheless, we find dislocation-free growth for all seeds at a small enough distance and for all seeds that approximately have the same orientation. In periodic structures, the stress that occurs due to incommensurate distances between the seeds results in phononic strain fields or, in the case of too large stresses, in dislocations. In contrast, in quasicrystals an additional phasonic strain field can occur and suppress dislocations. Phasons are additional degrees of freedom that are unique to quasicrystals. As a consequence, the additional phasonic strain field helps to distribute the stress and facilitates the growth of dislocation-free quasicrystals from multiple seeds. In contrast, in the periodic case the growth from multiple seeds most likely leads to a structure with multiple domains. Our work lays the theoretical foundations for growing perfect quasicrystals from different seeds and is therefore relevant for many applications.

I. INTRODUCTION

Structures with long-range order but no translational symmetry, also known as quasicrystals, have been observed in various metallic alloys (see, e.g., Refs. [1–3]) as well as in soft matter systems [4–8]. Quasicrystals can possess rotational symmetries that are usually not allowed to occur for periodic structures, though 5-, 8-, 10-, or 12-fold rotational symmetries seem to occur more often than other symmetries [9,10]. Furthermore, additional hydrodynamic modes, termed phasons, can occur in quasicrystals [11]. They correspond to complex correlated rearrangements of the particles [12–14]. Similar to phonons, exciting phasons do not cost any energy in the limit of long wavelengths. Furthermore, local phasonic excitations, also known as phasonic flips, lead to new growth modes where phasonic flips are built into the grown quasicrystalline structure, which nevertheless does still not possess any dislocation [15]. Note that in periodic crystals there is no corresponding counterpart for such a dislocation-free growth with built-in local phasonic flips. In experiments, the growth of a Al-Ni-Co decagonal quasicrystal was studied in Ref. [16], where frequent phasonic excitations are found at the growth front that usually relax later on. This is in qualitative agreement to what we observe for decagonal quasicrystals in Ref. [15]. Note that in soft matter systems, which we want to consider here, dodecagonal quasicrystals seem to occur more often than decagonal quasicrystals [17]. Experimentally, the growth of a dodecagonal quasicrystals has recently been studied in an oxide quasicrystal, which occurs as a wetting layer of $BaTiO_3$ on a Pt(111)-surface [18,19].

To explore the growth process of soft matter quasicrystals, we employ a so-called dynamical phase field crystal (PFC) model [20,21], which is based on an expansion of the free energy like a Swift-Hohenberg theory [22,23]. Furthermore, the PFC free energy can be derived from microscopic theories [24,25]. To describe periodic crystals, one characteristic length scale is employed in the free energy leading to a theory that can describe triangular, stripe, and fluid phases in two dimensions [20,21]. Quasicrystals are expected to occur in one-component soft-matter systems if the pair interaction contains at least two incommensurate characteristic length scales [15,26–34], which can be achieved in experiments [4]. In the framework of PFC or similar approaches, if a second length scale is added to the free-energy expansion, one finds quasicrystalline structures in two [15,26,27] or three dimensions [34]. Furthermore, dynamical PFC can be used to study the growth dynamics [15]. The dynamical PFC model that we employ here describes the overdamped dynamics as it usually occurs in soft matter systems.

Growth of crystals out of a melt normally occurs via nucleation [35,36]. Typically, there is not only a single nucleus initiating crystal growth but an ensemble of neighboring nuclei that all generate growing crystallites around them. These crystallites then meet. For the resulting final structure it is therefore essential to know how the growing crystallites merge. Here different scenarios are conceivable: either a large crystallite "eats up" a neighboring smaller one or incorporates it into its own structure, leaving behind a distortion, or the two neighboring crystallites both keep their structure and form a long-lasting grain boundary. In the latter case, the resulting

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crystalline texture is not monocrystalline. For quasicrystals, the same basic question is relevant, e.g., if these are grown from liquid metallic alloys [37,38]: Is it possible to grow perfect quasicrystals, i.e., structures without domain borders once several nucleation centers are present? For the case of a periodic crystal grown from two seeds, we observed [39] that if the orientation of the two seeds is similar, or they are close together, then the growth might result in a monocrystal, while if the seeds are far away from each other and rotated with respect to each other, then they grow into two stable crystals that form a domain boundary in between. Here we explore the growth of a quasicrystal from two seeds and explain that there is the possibility to excite phasonic strain, which suppresses the dislocations if the seeds are not too far apart from each other. As a consequence, there is no domain border in a quasicrystal for many cases where a domain border would form in the periodic case. This is important both from a fundamental point of view and for practical applications.

The article is organized as follows: We introduce the setup and explain the employed model system in Sec. II. In Sec. III, we first present results for the growth of a periodic crystal from two seeds before we explore the growth of a quasicrystal and analyze the phononic and phasonic strain fields. Finally, we conclude in Sec. IV.

II. MODEL AND SETUP

A. Dynamical phase-field crystal model

We employ a dynamical phase-field crystal (PFC) model, where a scalar density field $\psi(\vec{r},t)$ evolves following conserved dynamics [20], i.e., in reduced units,

$$\frac{\partial \psi(\vec{r},t)}{\partial t} = \nabla^2 \left[\frac{\delta F[\psi(\vec{r},t)]}{\delta \psi(\vec{r},t)} \right]. \quad (1)$$

As pointed out in the Introduction, Eq. (1) describes overdamped dynamics. Note that it has been shown in Ref. [40] that ballistic phonons cannot be correctly described by this approach. Therefore, in systems that are not overdamped, additional relaxation mechanisms might exist that are not considered in our present work. The free-energy functional $F[\psi(\vec{r})]$ is given by the following expansion that contains one or two incommensurate lengths scales [15,27]:

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

where ϵ can be interpreted as the mean-field temperature [20,21]. For a simple quasicrystal model, two length scales of the phase-field crystal model are chosen (i.e., $m = 2$) in terms of a unit length a such that $k_1 = 2\pi/a$ and $k_2 = 4\pi \cos(\pi/12)/a$. This leads to a stable quasicrystal with 12-fold symmetry [27,29]. In the periodic case, which we consider for comparison, $m = 1$ and $k_1 = 2\pi/a$. The remaining parameter of this model is the mean density $\bar{\psi}$ that is conserved under the dynamics of Eq. (1). The mean density is measured in reduced units relative to the reference

density, which is typically taken to be that of a unique reference point in the bulk phase diagram, here a triple point of three coexisting phases with the same triple density. Therefore, $\bar{\psi}$ can be negative, which means that the actual mean density is smaller than the triple density.

The static and dynamical properties of the 12-fold case were studied in Ref. [15]. At $\bar{\psi} = 0$ and $\epsilon = 0$, there is a triple point where the stable quasicrystalline phase with 12-fold rotational symmetry, triangular ordering, and the fluid phase meet. The properties of the static quasicrystalline phase as well as of the quasicrystal grown from a seed depend on the distance ϵ from the triple point. In the following, we will call the structures close to local symmetry centers flowers in accordance with Ref. [41]. For small ϵ, we observe that such flowers are surrounded by 12 density peaks of approximately similar height. Furthermore, the growth from a seed leads to perfect quasicrystals. However, for large ϵ, the density peaks in a flower usually have very different heights. In addition, when growing quasicrystals for large ϵ, many local phasonic flips are built into the structure, though there are no dislocations.

In the periodic case, we use $\epsilon = 0.018$ and the average density $\bar{\psi} = -0.0775$. The lowest free energy for these parameters is given by a triangular phase.

Concerning the numerical calculations, Eq. (1) with $F[\psi(\vec{r})]$ from Eq. (2) is solved using a semi-implicit spectral method [42–44], in which the linear term is treated implicitly and the nonlinear term explicitly. At each time t, we obtain the field at the next time $t + \Delta t$ using the following update rule:

$$\psi(\vec{k},t + \Delta t) = \frac{\psi(\vec{k},t) - \Delta t k^2 \mathcal{F}[\psi^3(\vec{r},t)]}{1 + \Delta t k^2 \left[-\epsilon + \prod_{j=1}^{m} \left(k_j^2 - k^2 \right)^2 \right]}, \quad (3)$$

where $\mathcal{F}$ denotes the Fourier transform. For the periodic case, we use square grids of sizes $N_x = N_y = 1920$ and discretization $\Delta x = \Delta y = 194a/N_x$. For the quasicrystals we use $N_x = N_y = 4096$ and $\Delta x = \Delta y = 252a/N_x$. The time step is $\Delta t = 0.05$ for all calculations. The starting configurations for both periodic systems and quasicrystals are circular cuts from the unrotated or rotated bulk structures (see next subsection for more details). All calculations were performed on NVIDIA graphics cards using CUDA [45] and cuFFT high perfomance library for the Fourier transforms [46].

B. Setup

For both the periodic and quasiperiodic case the initial setups contain two seeds of prescribed initial sizes at a distance d in x direction. The orientation between the two seed differs by an angle θ. We consider one large and one small seed. The smaller seed usually is only slightly larger than the critical seed size that is necessary to grow an ordered solid. Note that this critical seed size differs between the periodic and the aperiodic case and also depends on the parameter ϵ. Therefore, it does not make sense to quantitatively compare periodic and aperiodic systems with exactly the same parameters because their growth behavior as well as their free energy would be inherently different. Instead, we consider two different setups for the growth of a quasicrystal and compare it qualitatively to a similar system that leads to a periodic crystal.
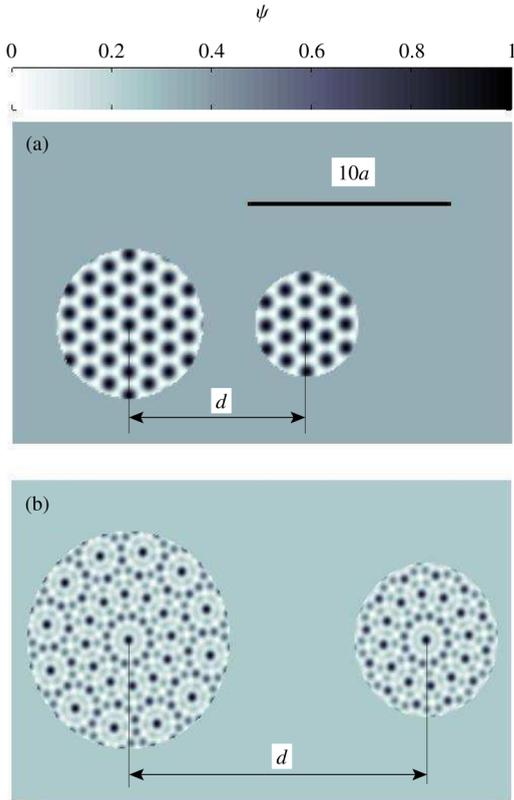
FIG. 1. Examples of initial configurations for (a) the periodic case, $\epsilon = 0.018$, $\bar{\psi} = -0.0775$ and (b) the quasiperiodic case $\epsilon = 0.25$, $\bar{\psi} = -0.315$. The periodic crystalline seeds are chosen such that they are symmetric around the center of the seed which contains a density maximum, while the quasicrystalline seeds contain the main symmetry center. The seeds are cut out from equilibrated solid configurations.

In the periodic case, the starting configuration is depicted in Fig. 1(a). The larger initial seed has radius $r_1 = 3.18a$ with 37 maxima and one of the main lattice directions is parallel to the y direction, while the small seed was fixed to $r_2 = 2.25a$ with 19 maxima and was rotated with the angle θ. For each angle the distance d between seeds is varied. A similar setup for the growth of periodic structures was studied by using density functional theory [39,47].

For the growth of a quasicrystal, the calculations were done for two sets of parameters that both lead to the formation of a stable quasicrystal. To be specific, we employ $\epsilon = 0.22$ or $\epsilon = 0.25$ and in both cases the average density $\bar{\psi} = -0.315$. Note that for $\epsilon = 0.22$ the system is closer to the triple point such that for $\epsilon = 0.25$ phasonic flips occur more often [15]. For $\epsilon = 0.22$ the large seed has the radius $r_1 = 12.30a$ and the small one $r_2 = 8.7a$. For $\epsilon = 0.25$, $r_1 = 5.09a$ and $r_2 = 3.6a$. As in the periodic case the large seed has one of the main lattice vectors parallel to the y axis, while the small seed is rotated by an angle θ [cf. Fig. 1(b)].

III. RESULTS

A. Growth of periodic crystals from two seeds

First, we present results for the periodic case. For large distances d between the seeds and sufficiently large θ, first two crystals grow independently. When the crystallization fronts meet, both of these crystals usually keep their orientation such that a metastable grain boundary consisting of dislocations occurs [shown in blue and with squares in Fig. 2(a), cf. snapshot in Fig. 2(e)]. For smaller distances d between the seeds and still sufficiently large θ, a defect-free crystal is formed [Fig. 2(d)]. Either, the larger seed takes over the smaller seed resulting in a strained crystal or the crystal grown from the smaller seed is rotated such that it fits the orientation of the large seed [marked red and with circles in Fig. 2(a)].

In case of small angles θ, it is easier to rotate the small crystal to fit the orientation of the large one. However, a domain border can still occur if the distance between the two seeds does not support the formation of one defect-free crystal, i.e., if d is an even multiple of the lattice constant a as in Fig. 2(b), a defect-free crystal is observed, but if d is an odd multiple of a as in Fig. 2(c), dislocations are formed. As a consequence, the regions of defect-free growth and the regions of growth with a domain border alternate in the diagram shown in Fig. 2(a) for small θ. Note that we never observe that dislocations in the domain border act as nucleation sites. In principle, this might occur in three dimensions.

In summary, in the periodic case the small crystal might adjust to the large crystal either by rotation or by absorbing the nonfitting ordering into a (phononic) strain field. However, such an adjustment, i.e., the formation of defect-free crystal, is only successful in the case of large angles and small distances, where the small seed still can be transformed quite easily or for small angles, if and only if the distance between the seeds is commensurate to a defect-free crystal. Note that we never observe the displacement of a whole crystal when two crystals meet at an incommensurate distance. In ballistic systems with only small or no viscous damping, such a displacement might be possible.

B. Growth of quasicrystals from two seeds

For the growth of a quasicrystal from two seeds, we consider two parameter sets to demonstrate that the quantitative behavior might depend on these parameters, while the general qualitative results do not. In Fig. 3(a), we show where the two systems that we study are located in the phase diagram. The type of long-distance behavior that we observe is indicated in Figs. 3(b) and 3(c). For both parameter sets, we find domain borders consisting of dislocations in case of large distances and large angles [indicated by squares and the blue color in Figs. 3(b) and 3(c)]. For small distances or small angles we discover the growth of a dislocation-free quasicrystal (marked by circles and red). Examples for the final density fields are shown in Figs. 3(d)–3(g).

Note that dislocations are not easily visible in quasicrystalline patterns. To analyze the density fields we have first determined the structure factor, filtered out two opposite first-order Bragg-peaks, and then applied an inverse Fourier transformation. In the resulting stripe pattern, dislocations can be identified [cf. Figs. 3(h)–3(k)]. More details on this method and its application are given in Refs. [48,49].

A difference between the systems with periodic and quasiperiodic structures can be found for small angles θ. In the periodic case the occurrence of domain borders depends
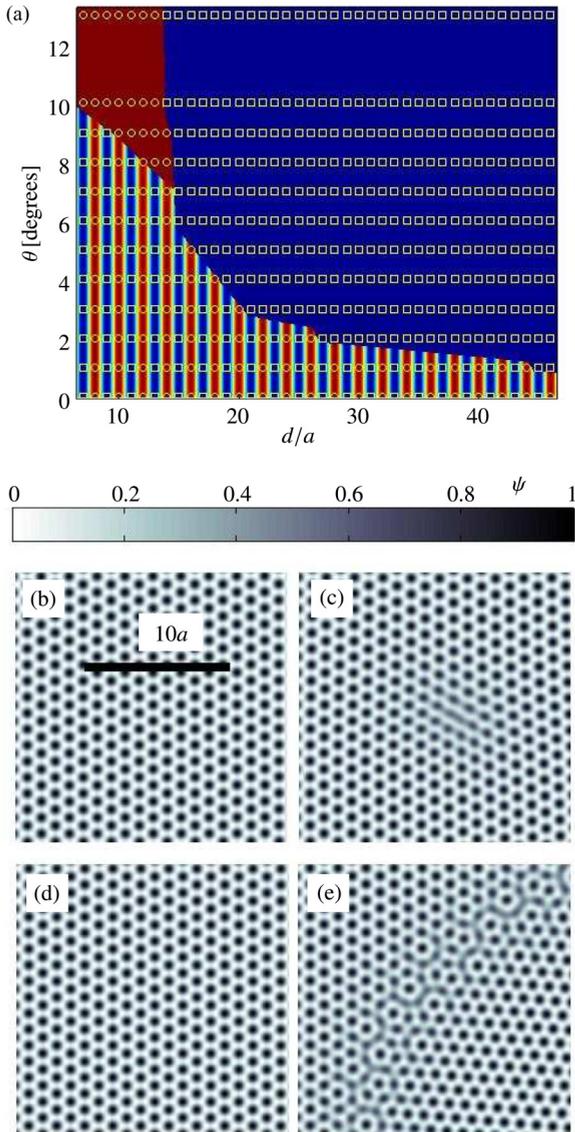
FIG. 2. Long-time situation of the growth started from two seeds at a distance d and orientations that differ by an angle θ for the periodic case. (a) Depending on d and θ, circles mark the points in the diagram where we observe defect-free crystals at late times. Squares denote cases where a domain border consisting of dislocations occurs. The red (light gray region with circles) and blue color (dark gray with squares) are used to highlight the parameter regions where defect-free final configurations or domain borders are found, respectively. (b–e) Density fields of the the grown structure at late times long after the two crystallites have been united. In (b, c) for a small angle, $\theta = 2°$, either the large seed incorporates the small seed if the seeds are at a commensurate distance, e.g., for $d = 16a$ as shown in (b) or both seeds are stable and an interface is formed in case of incommensurate distances, e.g., for $d = 17a$ shown in (c). For a large angle $\theta = 25°$, the large seed incorporates the small seed at small distances up to about $d = 13a$ shown in (d). For larger distances, e.g., $d = 14a$ (e), an interface occurs.

on whether the distance between the seeds is commensurate to a perfect triangular lattice or not. Note that in the case of quasicrystals the two seeds are always incommensurate, because the seeds have been constructed such that they

correspond to the local ordering around the unique perfect symmetry center, which in a perfect quasicrystal must occur only once. Despite this inherent misadjustment, we observe that a defect-free quasicrystal is always formed if the distance d between the seeds is sufficiently small or if the orientation of the seed is almost the same. Obviously there has to be a way how two quasicrystals can be strained such that they fit together without dislocations even though the two seeds are inherently nonfitting. As we will show in the next subsection, the dislocation-free connection of two quasicrystalline structures is facilitated by the possibility to use the additional phasonic degrees of freedom for an extra strain relaxation on top of the phononic strain.

For the growth from two seeds with a large difference in orientation on a first glance the results seem to be similar for the periodic and the quasiperiodic case. For large distances where both seeds could grow into stable structures before the ordered regions meet, we find the formation of a domain border while for small distances the small seed is taken over by the large one either by rotating the small seed or by straining both seed structures. In case strain structures are involved, the type and extent of strain might differ between the periodic and the aperiodic case as we will discuss in more detail in the next subsection. Since a quasicrystal has more degrees of freedom that help to distribute strain, we expect that a defect-free structure occurs more likely for quasicrystals than for periodic crystals. However, quantitative comparisons are not possible, because there is no natural choice of parameters for such a comparison. Already for the quasicrystalline case, the set of parameters influences the maximal angle or maximal distance that leads to defect-free growth as can be seen by comparing Figs. 3(b) and 3(c). The closer we are to the triple point, the larger the distance between quasicrystalline seeds can be without leading to a domain border [note the different scales for d in Figs. 3(b) and 3(c)].

An interesting detail for the growth of a quasicrystal can be seen in Fig. 3(b). For a constant distance around $35a$, we find a dislocation-free growth at small angles, while there is a domain border at intermediate angles. However, for large angles the growth again leads to a defect-free quasicrystal, i.e., we observe a *reentrant* behavior. The formation of a dislocation due to orientation mismatches occurs more likely for smaller than for larger angles θ.

C. Phononic and phasonic strain during the growth of a quasicrystal from two seeds

The formation of strain fields and as a consequence the mechanisms how two seeds can adjust to each other in the aperiodic case differ significantly from the periodic case. While in a periodic structure only phononic displacement fields are possible, additional degrees of freedom exist in quasicrystals [11–14], and in the following we show how they enable the growth of dislocation-free quasicrystals, even in the case the two original seeds are incommensurate.

In Fig. 4, we take a closer look into such a dislocation-free growth started from two quasicrystalline seeds. In Figs. 4(a)–4(c), we show the areas whose structures are dominated by one of the seeds. To be specific, the blue area (dark gray area on the left-hand side) indicates the region
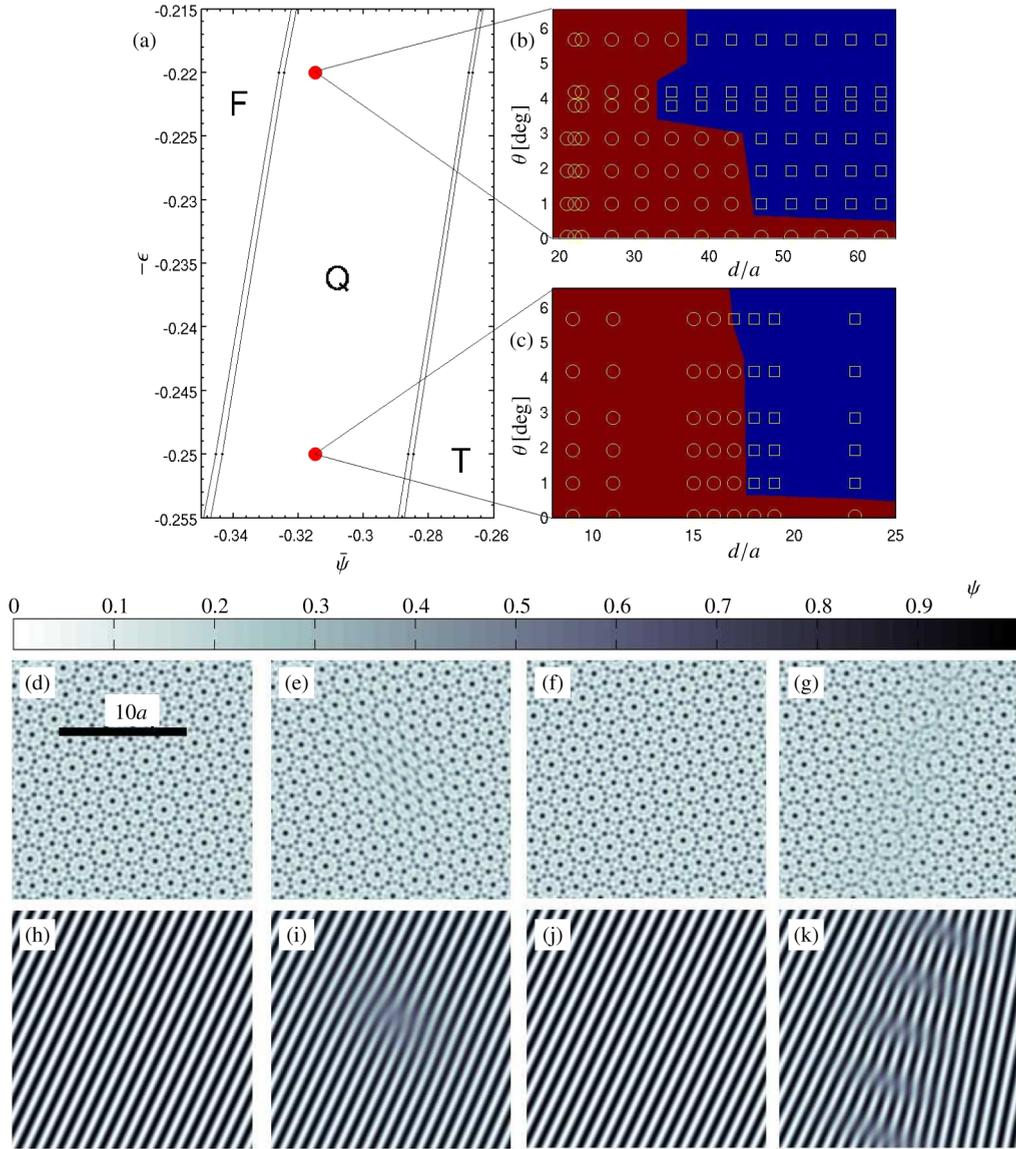
FIG. 3. (a) The phase diagram for the quasicrystal in the region of interest (for the complete diagram, see Ref. [15]). The fluid, quasicrystalline, and triangular phase are marked with the letters F, Q, and T, respectively. Coexistence regions are observed that occur between the two lines drawn at phase boundaries in the phase diagram. (b, c) Long-time situation of the structures grown from two seeds as a function of distance d and angle θ for $\epsilon = 0.22$ (b) or $\epsilon = 0.25$ (c). The circles and red (light gray) regions mark the cases with a dislocation-free quasicrystal at late times. The squares and blue (dark gray) regions denote where a metastable grain boundary is observed. (d–g) Density fields for $\epsilon = 0.22$ and (h–k) the corresponding filtered fields where dislocations become visible using the method explained in [48,49] for a very small angle $\theta = 0.093°$ (d, e, h, i) and a large angle $\theta = 13.625°$ (f, g, j, k). For small distances $d = 43a$ (d, h) or $d = 35a$ (f, j) no dislocations occur while for a larger distance $d = 47a$ (e, i) or $d = 39a$ (g, k) domain borders are observed.

where the density field corresponds to a quasicrystal that could be grown from the large, left seed alone. Correspondingly, in the regions colored in orange (light gray on the right-hand side) the structure fits to a quasicrystal grown from only the right seed. The coloring reveals that the quasicrystal grown from the left seed (blue, dark gray) stops the growth of the quasicrystal of the right seed (orange, light gray). The blue (dark gray) quasicrystal embraces the orange (light gray) one and will finally incorporate it.

To understand how the structure grown from the small seed is taken over by the structure grown from the large seed, we analyze the structure at time $t = 500$ in more detail, i.e., just

before the quasicrystal that originated from the small seed disappears. In Fig. 4(d), we show a zoom to Fig. 4(c) around the location of the small seed. Blue spots (dark gray on the left-hand side) denote points where the density field does not fit to the small seed while orange spots (light gray on the right-hand side) indicate that the density does not fit to the structure that one would expect from the large seed alone. Note that in the fluid on the right-hand side of Fig. 4(d), there are blue and orange (dark and light gray) spots, indicating that the fluid differs both from a quasicrystal grown from the large seed as well as from a quasicrystal grown from the small seed. Furthermore, while in the part that is dominated by
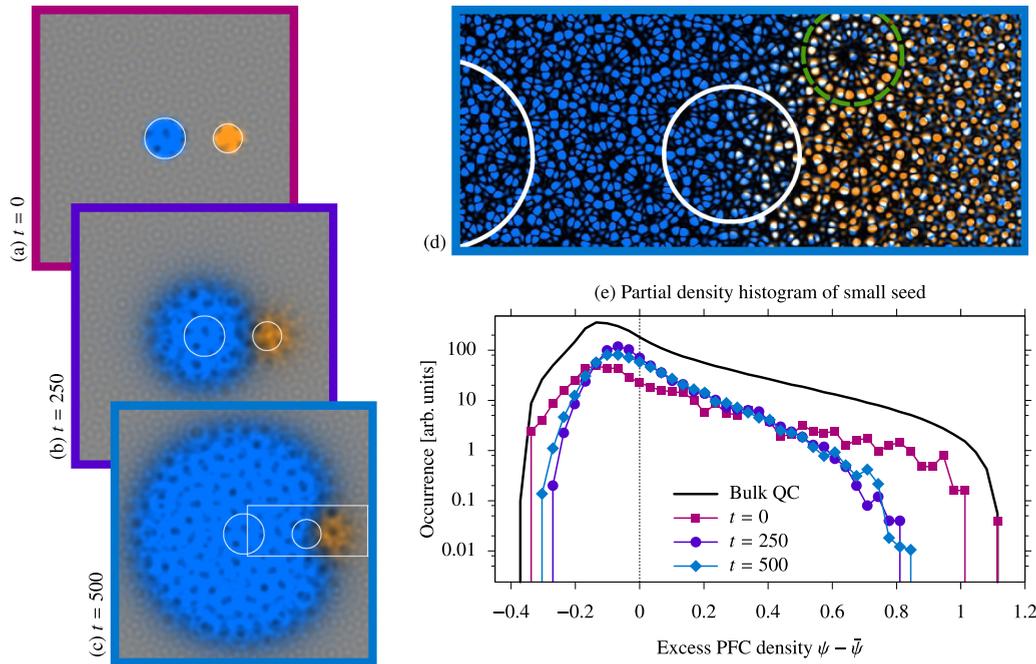
FIG. 4. Growth of a dislocation-free quasicrystal with $\epsilon = 0.25$ from two seeds that are rotated with respect to each other by $\theta = 13.625°$ and are placed at an incommensurate distance $d = 16a$. (a–c) Growth process that demonstrates how the quasicrystal that is grown from the small (right) seed is taken over by the structure that originated from the large (left) seed. The white solid circles denote the initial positions of the seeds. The orange (light gray) or blue (dark gray) areas on the right- or left-hand side of the figure indicate the region where the influence from the small or large seed dominates, respectively. The colors are determined by calculating the differences between the density field obtained from the growth from two seeds and the density field obtained by growing a quasicrystal from only the large (orange, light gray) or only the small seed (blue, dark gray). The resulting differences have been squared and smoothened by a Gaussian blur of width $\sigma = 4.1a$. Therefore, orange (light gray) or blue (dark gray) denotes the area where one mainly finds the structure that does not fit to the large or small seed, respectively. White, gray, and black areas outside of the grown structures originate from additive mixing of blue and orange, i.e., white denotes areas that do not fit to either seed while black areas fit to both. (d) Zoom view into the area marked by a white box in (c) (i.e., at time $t = 500$) in the same representation, except without blurring. Therefore, blue (dark gray) spots denote points that do not fit to the small seed and orange (light gray) spots indicate differences from a quasicrystal grown from only the large seed. (e) Histograms of density values that appear in the density fields in the regions that are dominated by the small seed [orange, light gray areas in (a–c)] for different times t. The black solid line indicates the distribution in a perfect quasicrystal (arbitrary scale). Note that for times $t \geqslant 750$, an orange (light gray) domain maintaining the orientation of the right seed is no longer detectable.

the large seed there are hardly any orange (light gray) spots, in the area that is mainly dominated by the small seed one can also find blue (dark gray) spots. Interestingly, blue (dark gray) spots appear at the centers of flowers while orange (light gray) spots close to these centers are already weak. Therefore, the most pronounced minima or maxima of the density field that are located in the centers of these flowers are taken over first. In the next step, the surroundings of the flowers have to be adjusted to the new structure. The flower marked with a green broken circle in Fig. 4(d) possesses blue as well as orange spots (dark and light gray spots) in its surrounding. The center of the flower already fits to the new (blue, dark gray) structure up to a small displacement, i.e., a small phononic strain. The surrounding of the flower corresponds to the new structure up to a phasonic flip as the ones that we have also observed in Ref. [15]. Therefore, the structure of the small seed is taken over by the structure of the large seed by transforming it into a structure with both phononic and phasonic strain that later is relaxed. Note that a strong phasonic strain causes orange and white spots but in case of the shown example, the phasonic strain that remains in the end is too small to be visible

[cf. left-hand side of Fig. 4(d)]. Especially, it is much smaller than the phasonic strain that can be excited thermally [50].

In Fig. 4(e), we plot the distribution of values of the density field at different times for the region that is dominated by the structure of the small seed, i.e., the region shown in orange (light gray) in Figs. 4(a)–4(c). For comparison, the distribution for a perfect quasicrystal is shown by a black line with an arbitrary overall normalization. The small seed at time $t = 0$ possesses the same distribution of density values as the perfect quasicrystal up to a rescaling factor and some noise due to the small size of the seed. At times $t = 250$ and $t = 500$ the largest as well as the smallest minima already have disappeared. Density values at intermediate values still occur and due to the growth of the quasicrystal might even occur more often than before. These findings confirm that the most pronounced maxima or minima that usually occur in the centers of flowers are lost first. The less pronounced minima and maxima can be built into the new structure corresponding to a quasicrystal with phasonic strain. Only later, the phasonic strain is relaxed and the remains of the structure of the small seed disappear.

IV. CONCLUSIONS

By using dynamical phase-field crystal models, we explored whether the growth of periodic or aperiodic structures from two seeds leads to defect-free orderings or whether a domain border is formed. For both a periodic crystal as well as for a quasicrystal, we systematically analyzed the impact of different distances between the seeds and different mismatches of the orientations of the seeds on final structure. In the case of significant differences in the orientations, we observe the formation of a rotated or strained defect-free periodic crystal or quasicrystal, if and only if the seeds are not too far apart in the beginning. Seeds that are at a large distance lead to the growth of two independent large structures that form a domain border when they get into contact.

For small differences between the orientations of the seeds, the behavior of periodic crystals significantly differs from the behavior of quasicrystals. In the case of periodic crystals, a defect-free structure only occurs if the two seeds are at a distance that is commensurate to a periodic lattice, while in the aperiodic case we always observe the formation of a dislocation-free quasicrystal, though the two quasicrystalline seeds are never perfectly commensurate at any distance.

A major difference in how two growing structures might adjust to each other is given by how stress can be distributed. While in periodic crystals the stress between two nonfitting structures only leads to a phononic strain field or dislocations, in the quasicrystalline case a phasonic strain field can be used to distribute the stress in addition. The phasonic strain field leads to dislocation-free quasicrystals, where local rearrangements have occurred that we observe as phasonic flips. Furthermore, the phasonic strain can be reduced or even resolved by local rearrangements.

Our results indicate that if a structure is grown from multiple random seeds, in the periodic case a lot of metastable domain borders will occur even if the seeds are close together, because many seeds that by chance have a similar orientation will not be at a commensurate distance. However, in the aperiodic case we expect that a dislocation-free quasicrystal can be grown if the seeds are sufficiently close together. The quasicrystal obtained from multiple seeds can differ from a quasicrystal that is grown from only one seed by a phasonic and phononic strain field that is associated with phasonic flips.

For applications, for which the growth of monocrystalline quasicrystals is crucial, our work indicates that the conditions to grow pure quasicrystals can be much milder than those needed to grow pure periodic crystals.

Future work should focus on more detailed models of quasicrystal growth. One option is to go for dynamical density functional theory [39,51], which is a more microscopic description for the particle interaction than the phase-field crystal model considered here. To obtain stable quasicrystals, one either needs a complex effective interaction in a one-component system or binary systems. However, we expect that the general idea that phasons provide additional efficient channels to anneal dislocations will still be valid in general. Finally, it would be interesting to study colloidal quasicrystals (see, e.g., Ref. [4]) on the particle-resolved level to check for the different growth scenarios predicted by our theory in an experiment. As has been documented in the periodic case, experiments on colloids can resolve the full dynamics of a growing crystal around a prescribed seed [52–55], such that the full evolving structure can be monitored. This should be applicable also to quasicrystals.

[1] D. Shechtman, I. Blech, D. Gratias, and J. W. Cahn, Phys. Rev. Lett. **53**, 1951 (1984).

[2] W. Steurer, Chem. Soc. Rev. **41**, 6719 (2012).

[3] E. Maciá, Rep. Prog. Phys. **69**, 397 (2006).

[4] S. Fischer, A. Exner, K. Zielske, J. Perlich, S. Deloudi, W. Steurer, P. Lindner, and S. Förster, Proc. Natl. Acad. Sci. USA **108**, 1810 (2011).

[5] X. Zeng, G. Ungar, Y. Liu, V. Percec, A. E. Dulcey, and J. K. Hobbs, Nature **428**, 157 (2004).

[6] X. Zeng, Curr. Opin. Colloid Interface Sci. **9**, 384 (2005).

[7] A. Takano, W. Kawashima, A. Noro, Y. Isono, N. Tanaka, T. Dotera, and Y. Matsushita, J. Polym. Sci. Polym. Phys. **43**, 2427 (2005).

[8] K. Hayashida, T. Dotera, A. Takano, and Y. Matsushita, Phys. Rev. Lett. **98**, 195502 (2007).

[9] J. Mikhael, M. Schmiedeberg, S. Rausch, J. Roth, H. Stark, and C. Bechinger, Proc. Natl. Acad. Sci. USA **107**, 7214 (2010).

[10] M. Schmiedeberg and H. Stark, J. Phys.: Condens. Matter **24**, 284101 (2012).

[11] J. E. S. Socolar, T. C. Lubensky, and P. J. Steinhardt, Phys. Rev. B **34**, 3345 (1986).

[12] J. A. Kromer, M. Schmiedeberg, J. Roth, and H. Stark, Phys. Rev. Lett. **108**, 218301 (2012).

[13] M. Sandbrink and M. Schmiedeberg, in *Aperiodic Crystals*, edited by S. Schmid, R. L. Withers, and R. Lifshitz (Springer, Berlin, 2013), pp. 261–267.

[14] M. Martinsons, M. Sandbrink, and M. Schmiedeberg, Acta Phys. Pol. A **126**, 568 (2014).

[15] C. V. Achim, M. Schmiedeberg, and H. Löwen, Phys. Rev. Lett. **112**, 255501 (2014).

[16] K. Nagao, T. Inuzuka, K. Nishimoto, and K. Edagawa, Phys. Rev. Lett. **115**, 075501 (2015).

[17] T. Dotera, Isr. J. Chem. **51**, 1197 (2011).

[18] S. Förster, K. Meinel, R. Hammer, M. Trautmann, and W. Widdra, Nature **502**, 215 (2013).

[19] S. Förster, J. I. Flege, E. M. Zollner, F. O. Schumann, R. Hammer, A. Bayat, K.-M. Schindler, J. Falta, and W. Widdra, Ann. Phys. **529**, 1600250 (2017).

[20] K. R. Elder, M. Katakowski, M. Haataja, and M. Grant, Phys. Rev. Lett. **88**, 245701 (2002).

[21] K. R. Elder and M. Grant, Phys. Rev. E **70**, 051605 (2004).

[22] J. B. Swift and P. C. Hohenberg, Phys. Rev. A **15**, 319 (1977).

[23] M. Cross and H. Greenside, *Pattern Formation and Dynamics in Nonequilibrium Systems* (Cambridge University Press, Cambridge, 2009), Chap. 5.

[24] S. v. Teeffelen, R. Backofen, A. Voigt, and H. Löwen, Phys. Rev. E **79**, 051404 (2009).

[25] K. R. Elder, N. Provatas, J. Berry, P. Stefanovic, and M. Grant, Phys. Rev. B **75**, 064107 (2007).

[26] R. Lifshitz and D. M. Petrich, Phys. Rev. Lett. **79**, 1261 (1997).

[27] J. Rottler, M. Greenwood, and B. Ziebarth, J. Phys.: Condens. Matter **24**, 135002 (2012).

[28] K. Barkan, H. Diamant, and R. Lifshitz, Phys. Rev. B **83**, 172201 (2011).

[29] K. Barkan, M. Engel, and R. Lifshitz, Phys. Rev. Lett. **113**, 098304 (2014).

[30] A. R. Denton and H. Löwen, Phys. Rev. Lett. **81**, 469 (1998).

[31] M. Engel and H.-R. Trebin, Phys. Rev. Lett. **98**, 225505 (2007).

[32] A. J. Archer, A. M. Rucklidge, and E. Knobloch, Phys. Rev. Lett. **111**, 165501 (2013).

[33] T. Dotera, T. Oshiro, and P. Ziherl, Nature **506**, 208 (2014).

[34] P. Subramanian, A. J. Archer, E. Knobloch, and A. M. Rucklidge, Phys. Rev. Lett. **117**, 075501 (2016).

[35] T. Palberg, J. Phys.: Condens. Matter **11**, R323 (1999).

[36] T. Schilling, H. J. Schöpe, M. Oettel, G. Opletal, and I. Snook, Phys. Rev. Lett. **105**, 025701 (2010).

[37] I. R. Fisher, Z. Islam, A. F. Panchula, K. O. Cheon, M. J. Kramer, P. C. Canfield, and A. I. Goldman, Philos. Mag. B **77**, 1601 (1998).

[38] T. J. Sato, H. Takakura, and A. P. Tsai, Jpn. J. Appl. Phys. **37**, 663 (1998).

[39] T. Neuhaus, M. Schmiedeberg, and H. Löwen, Phys. Rev. E **88**, 062316 (2013).

[40] V. Heinonen, C. V. Achim, and T. Ala-Nissila, Phys. Rev. E **93**, 053003 (2016).

[41] L. Zaidouny, T. Bohlein, R. Roth, and C. Bechinger, Soft Matter **9**, 9230 (2013).

[42] J. Zhu, L.-Q. Chen, J. Shen, and V. Tikare, Phys. Rev. E **60**, 3564 (1999).

[43] B. P. Vollmayr-Lee and A. D. Rutenberg, Phys. Rev. E **68**, 066703 (2003).

[44] G. Tegze, G. Bansel, G. I. Tóthb, T. Pusztai, Z. Fan, and L. Gránásy, J. Comput. Phys. **228**, 1612 (2009).

[45] J. Nickolls, I. Buck, M. Garland, and K. Skadron, ACM Queue **6**, 40 (2008).

[46] NVIDIA Corporation, *CUDA Fast Fourier Transform*, http://docs.nvidia.com/cuda/cufft.

[47] T. Neuhaus, A. Härtel, M. Marechal, M. Schmiedeberg, and H. Löwen, Eur. Phys. J. Special Topics **223**, 373 (2014).

[48] L. Korkidi, K. Barkan, and R. Lifshitz, in *Aperiodic Crystals*, edited by S. Schmid, R. L. Withers, and R. Lifshitz (Springer, Berlin, 2013), pp. 117–124.

[49] M. Sandbrink and M. Schmiedeberg, Phys. Rev. B **90**, 064108 (2014).

[50] J. Hielscher, M. Martinsons, M. Schmiedeberg, and S. Kapfer, J. Phys.: Condens. Matter **29**, 094002 (2017).

[51] S. v. Teeffelen, C. N. Likos, and H. Löwen, Phys. Rev. Lett. **100**, 108302 (2008).

[52] R. P. A. Dullens, D. G. A. L. Aarts, and W. K. Kegel, Phys. Rev. Lett. **97**, 228301 (2006).

[53] V. W. A. d. Villeneuve, R. P. A. Dullens, D. G. A. L. Aarts, E. Groeneveld, J. H. Scherff, W. K. Kegel, and H. N. W. Lekkerkerker, Science **309**, 1231 (2005).

[54] F. Ziese, G. Maret, and U. Gasser, J. Phys.: Condens. Matter **25**, 375105 (2013).

[55] K. Sandomirski, E. Allahyarov, S. U. Egelhaaf, and H. Löwen, Nat. Commun. **6**, 7110 (2015).