Layering: a novel real-space method for construction of quasicrystals from a quasi-unit cell and understanding phasons

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Despite the considerable advancements in the study of quasicrystals [1], developing an intuitive understanding of their structure remains a challenge for many researchers. While effective, methods based on the cut-and-project approach [2], often pose considerable obstacles for developing intuition due to their higher-dimensional nature. Although physical space methods for quasicrystal characterization exist (such as subdivision [3] or covering [4]), they intrinsically lack the ability to describe phasons.

To overcome these challenges, we present a novel real-space quasi-unit cell framework, which capitalizes on the self-similarity of quasiperiodic tilings to streamline their construction, description, comprehensibility, and analysis. Our framework utilizes a newly developed concept we call layering, which can predict and explain the origin of phason flips based solely on the structure of the quasi-unit cell. We show how the new framework applies to several popular 2-dimensional QC models (Penrose tilings, Ammann-Beenker tiling, etc.) and we showcase the validity of constructed tilings by comparing them to subdivision (deflation). By offering a more accessible and intuitive method, our layering framework promises to advance research and understanding in the field of quasicrystals and foster broader engagement from the scientific community.

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Study cases on the free energy calculations in 2D and 3D model quasicrystals

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Free energy calculations in quasicrystals are challenging because standard numerical methods consider only phonon contributions, but they struggle with phason contributions. These free energy contributions remain important to accurately obtain the thermodynamic stability of these quasicrystals relative to their approximant competitor phases. In this work, we advance free energy calculations of quasicrystals in two directions where we consider the phonon and phason contribution to free energy. First, we evaluate the effect of a periodic substrate potential on the stability of a dodecagonal quasicrystal in two dimensions (see Fig. 1). These simulations are inspired by experiments of thin film (Ba,Sc)TiO_3 quasicrystals and their approximants [1]. We find that the periodic substrate shifts the stability of complex dodecagonal quasicrystal approximants towards simplest ones, generating a "Devil's flower" pattern [2].

We also observe the spontaneous formation of modulated phases and a new hexagonal labyrinth-like quasicrystalline phase in simulation. Second, we investigate the stability of an icosahedral quasicrystal [3] as a function of phason strain and tile decoration density. We find that the icosahedral quasicrystal is metastable relative to its 1/1 approximant and spontaneously transforms into the 1/1 approximant by the continuous build-up of phason strain. The transformation is a highly complex, collective reorganization process involving all particles in the system. Our simulation results are assisted by novel bond-orientational order parameters that we developed to detect, quantify, and classify crystallographic order in particle simulation data.

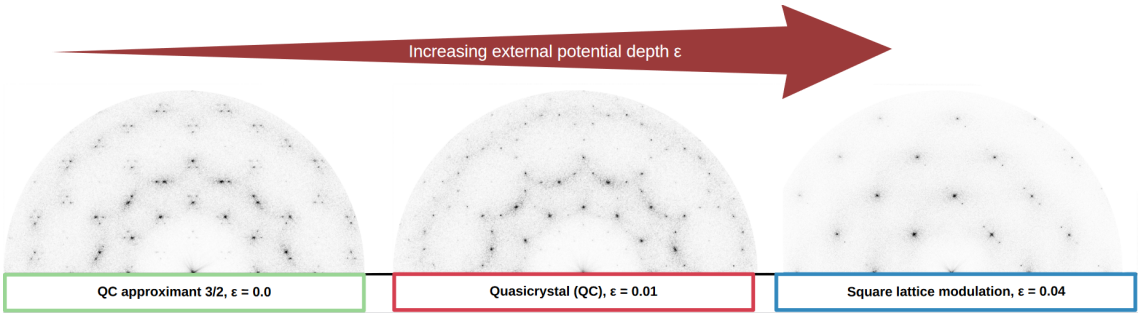

Fig.1: Diffraction patterns showing the evolution of phases as an external periodic potential depth ε is increased.

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Confirmation of the random tiling hypothesis by polar calculus

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The random tiling hypothesis, first proposed by Elser and Henley in the 1980s [1-3], states that quasicrystals are entropy-stabilized and, hence, are high temperature phases. Kiselev *et al.* [4] confirmed the hypothesis for a two-dimensional Tübingen triangle tiling which had been obtained in molecular dynamics simulations with a Lennard-Jones-Gauß potential [5]. By numerical simulations they investigated the temperature dependence of its two phason elastic constants λ_6 and λ_8, which are the second derivatives of the free energy $F(\chi_6, \chi_8, T)$ with respect to selected symmetrized phason strain modes χ_6 and χ_8. At $T = 0$, F showed a saddle point by descending along the χ_8 direction. Therefore, $\lambda_8 < 0$ characterized the quasicrystal's initial instability. The configurational entropy due to phason flips turned F upwards at higher temperatures, reversed the sign of λ_8 and lead to a stable quasicrystal. Kiselev *et al.* mimicked the phason flips by a system of uncorrelated Ising spins. We confirm this result by applying exclusively geometric methods providing insights beyond the numerics. Our method is the polar calculus, where the projection window W is divided into atomic domains C for each vertex environment. We increase the numbers of flip types and extend the calculus to a dynamic one by separating the window into areas P that characterize the different kinds of phason flips (Fig. 1). By phasonic deformation of the window, we can determine the types of flips and their frequency in dependence of phason strain, can perform energy relaxations by flips and compute the configurational entropy. Also, we introduce nearest neighbor correlations between flips and find that they constitute an important mechanism supporting the quasicrystal's stability.

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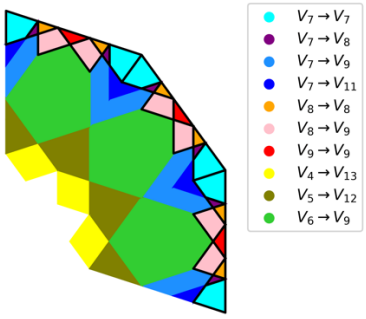

Fig. 1: Flip window in the ideal Tübingen triangle tiling for transitions between different vertices. The area outlined in black corresponds to the flips used by Kiselev *et al.*