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Thermodynamical Stability Analysis of a Model Quasicrystal

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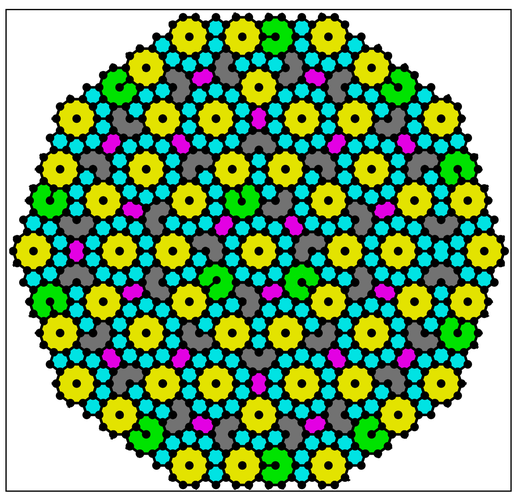
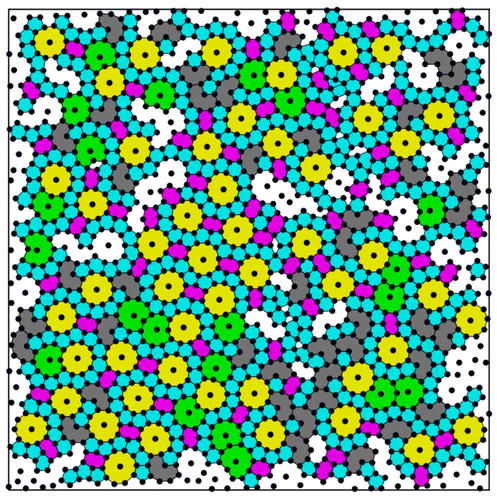
Introduction

- According to the random tiling hypothesis quasicrystals (QCs) are high temperature phases that stabilize due to large entropy [1].
- We prove the hypothesis by using the decagonal Tübingen tiling as a model QC.
- We apply exclusively geometric methods, namely the polar calculus.

The Tübingen tiling (TT)

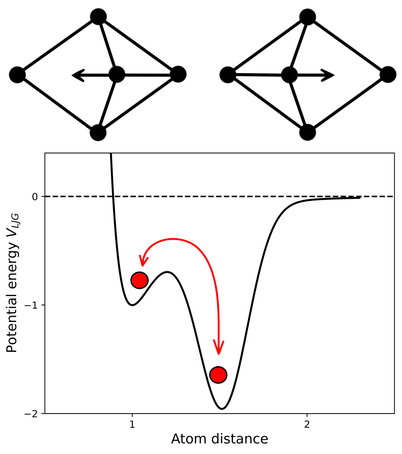


(a) The ideal TT, from mathematical construction. It has a generalized 10-fold rotational symmetry.

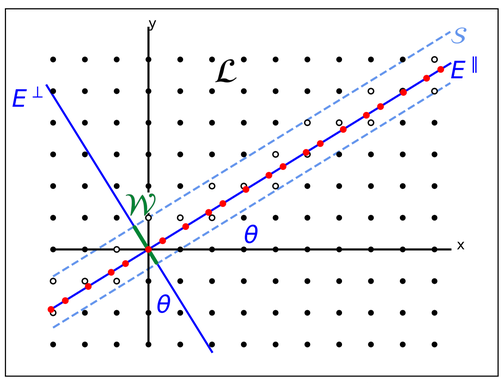


(b) A randomized version of the TT, from a molecular dynamics simulation [2].

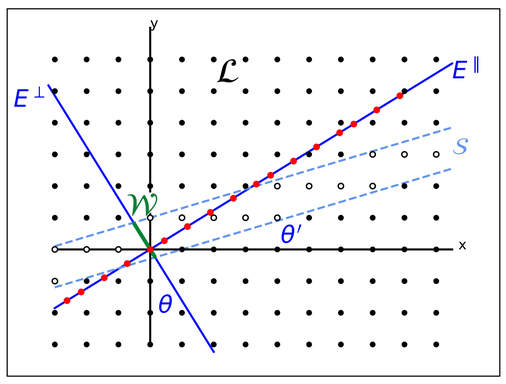
- A random tiling arises from the ideal TT through atom flips.
- Random flips do not change the generalized 10-fold rotational symmetry.
- A randomized TT arises also in a MD-simulation with a Lennard-Jones Gauß-potential.
- Many possible flip configurations lead to a large entropy.



Constructing crystals by cut and project scheme (CPS)



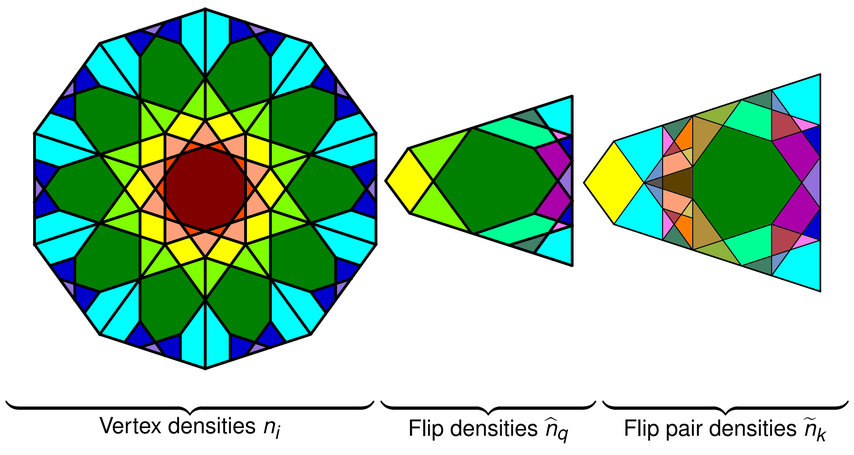
(a) Cut space $\mathcal{S}$ and projection space $E^\parallel$ have the same orientation.



(b) Cut space $\mathcal{S}$ and projection space $E^\parallel$ have different orientation.

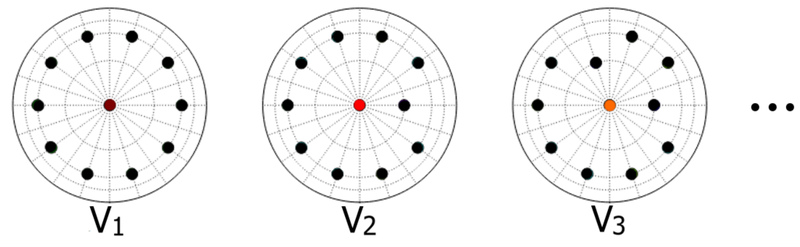
- 1D crystals can be constructed as the $E^\parallel$-projection of a 2D hyperlattice $\mathcal{L} \cap \mathcal{S}$.
- The structure of a projected 1D crystal is determined by *phason strain*: $\chi = \tan(\theta - \theta')$.
- The window $\mathcal{W}$ is the $E^\perp$-projection of $\mathcal{L} \cap \mathcal{S}$.
- Phason strain expresses itself in the tiling by flips.
- The CPS for the TT requires a 4D hyperlattice and yields two independent strain components: $\boldsymbol{\chi} = (\chi_1, \chi_2)$.

The dynamic polar calculus for vertices and flips

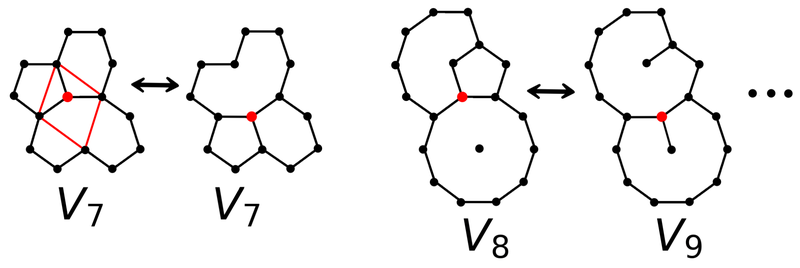


- The TT's window is a regular decagon that will be deformed by applying phason strain.
- Every atom configuration corresponds to a domain in $\mathcal{W}$.
- The density of such configurations is given by the domain's area.

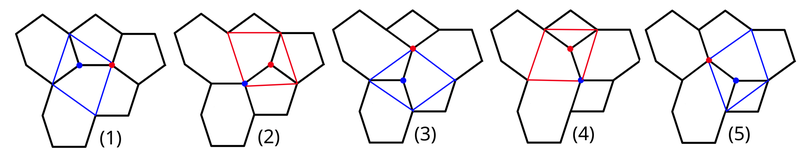
Atom environments and flip configurations in the TT for a cut-off radius $r_c = 2$



- The energy density of one specific tiling is determined by its atom environments V_i and their potential energies E_i.



- A fixed average phason strain χ corresponds to certain flip configurations and determines the free energy density $F(\chi, T)$.
- Asymmetric flips may increase or decrease the tiling energy depending on the energy difference of their two states, $\Delta E \lesseqgtr 0$.



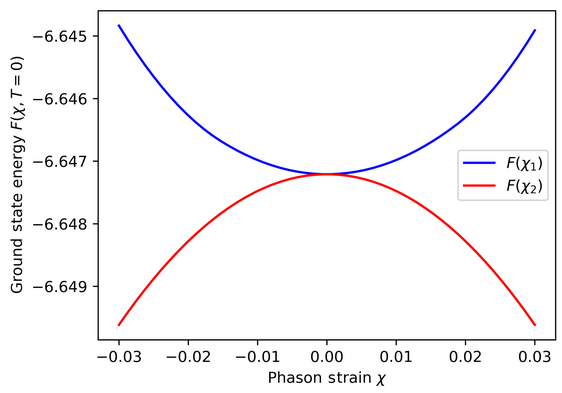
- The simplest model of correlated flips consists of pairs of coupled flips and has at most 5 non-degenerate states.

The free energy density

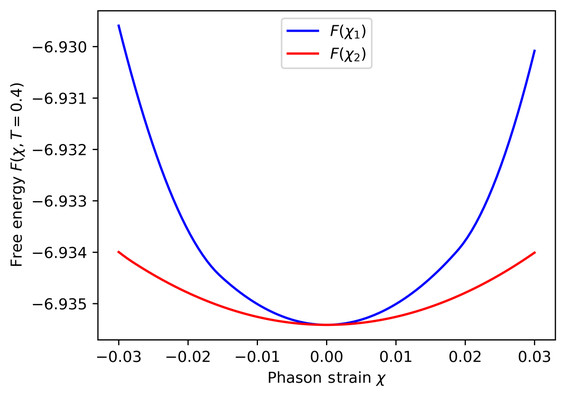
$$F(\chi, T) = \underbrace{\frac{1}{2} \sum_{\text{Vertices } i} n_i(\chi) E_i}_{\textit{projected tiling}} + \underbrace{\sum_{\text{Flips } q|\ \Delta E_q < 0} \widehat{n}_q(\chi) \Delta E_q}_{\textit{flip relaxation}} + \underbrace{\sum_{\text{Flip pairs } k} \widetilde{n}_k(\chi) \ln\Big(\sum_i \mathrm{e}^{-k_B \Delta E_k^{(i)}/T}\Big)}_{\textit{flip randomization}}$$

- n_i, $\widehat{n}_q$ and $\widetilde{n}_k$ are the densities of atom environments, flip configurations or flip pair configurations for a given average χ.
- **These quantities are calculated by a dynamic polar calculus.**

Free energy at low and high temperature

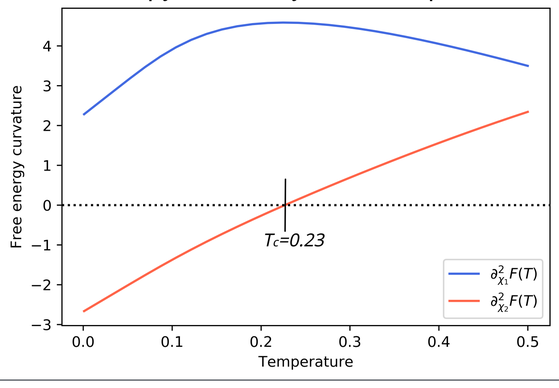


(a) Sattle point at $\chi = \mathbf{0}$ for $T = 0 \Rightarrow$ QC not stable.



(b) Minimum at $\chi = \mathbf{0}$ for $T > 0 \Rightarrow$ Stable QC.

- $F(\chi, T = 0)$ has saddle point at $\chi = \mathbf{0}$.
- $F(\chi, T)$ is minimized at $\chi = \mathbf{0}$ for $T > 0$ due to entropy caused by thermal flips.
- The phonon free energy is χ-independent [3].
- The free energy curvature $\partial^2_{\chi_2} F(\chi, T)|_{\chi=0}$ becomes positive above $T_c = 0.23$ and hence, the QC is stabilized.



References:

[1] Henley C. "Random Tiling Models". In: Quasicrystals - The State Of The Art. Singapore: World Scientific Publishing, 1991, pp. 429–524.

[2] Engel M. "Dynamics and Defects of Complex Crystals and Quasicrystals: Perspectives from Simple Model Systems". PhD thesis. Universität Stuttgart, 2008.

[3] Kiselev A. "Phasonen in quasikristallinen Strukturen des Lennard-Jones-Gauß-Systems". Diploma thesis. Universität Stuttgart, 2011.