Electronic properties of quasicrystals

Single electron properties of tight-binding quasiperiodic models

Nicolas Macé

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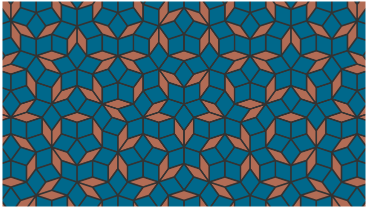

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Under the supervision of Anuradha Jagannathan
In collaboration with : Michel Duneau, Pavel Kalugin, Rémi Mosseri, Frédéric Piéchon.

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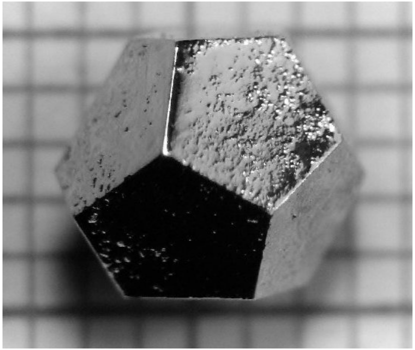
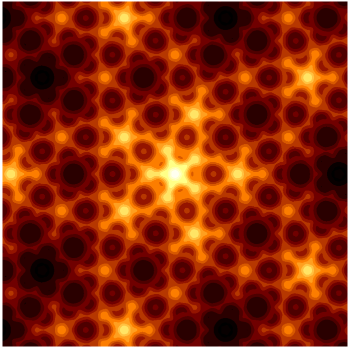


3D HoMgZn quasicrystalline sample
(doi:10.1038/nmat1244)

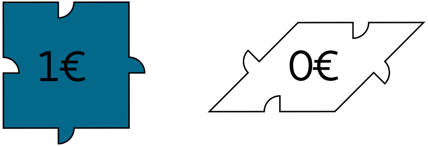


Groundstate electron density
(simple tight-binding model on a 2D tiling)

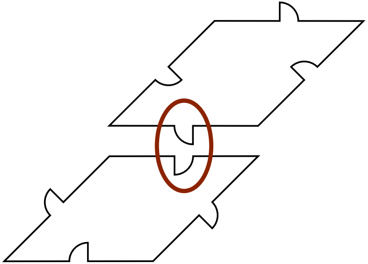
Quasicrystals : not periodic yet ordered structures (discovery : 1982).

Here : single electron properties of tight-binding quasiperiodic models.

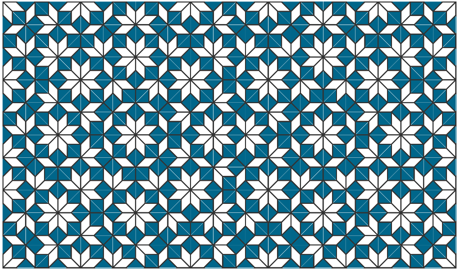
A quasiperiodic puzzle [Bédaride *et al.* 12]



Pay the squares, get the rhombuses for free !



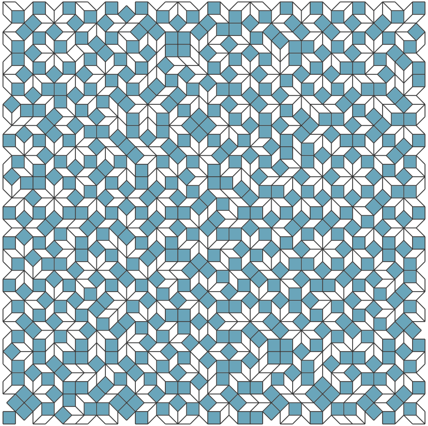
Forbidden configuration.



Patch of the Ammann-Beenker tiling.

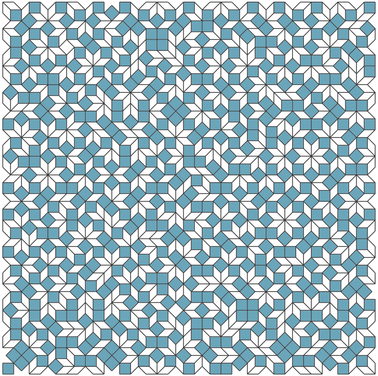
Periodic, quasiperiodic and random

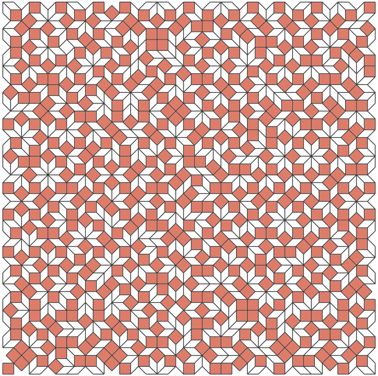
A random tiling :



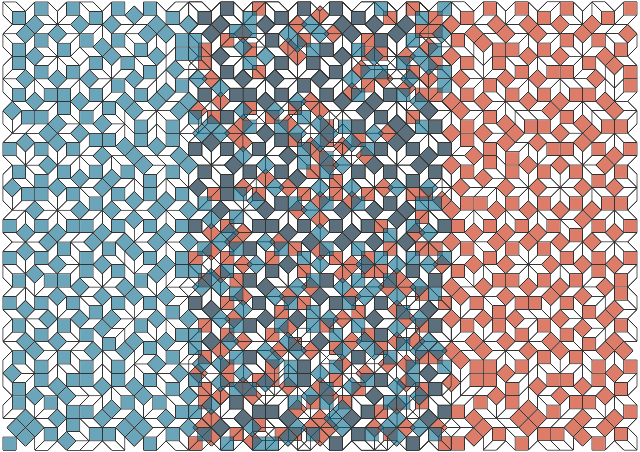
Periodic, quasiperiodic and random

Two copies of the tiling :



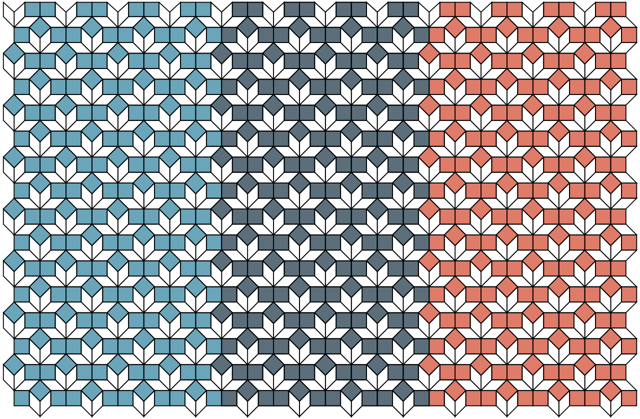


Periodic, quasiperiodic and random



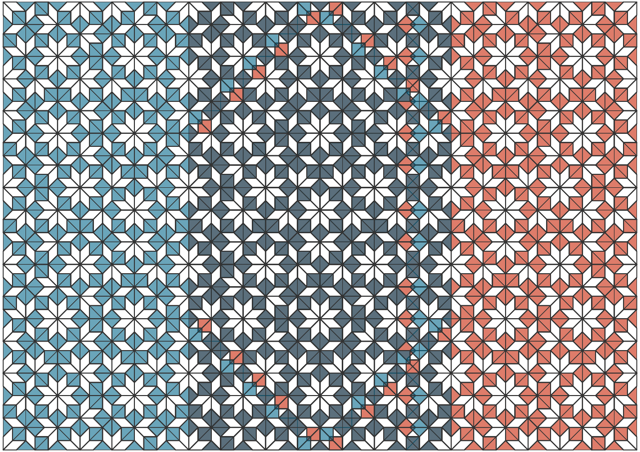
$\rightarrow$ no overlap $\rightarrow$ no order

Periodic, quasiperiodic and random



Perfect long range order : periodic

Periodic, quasiperiodic and random



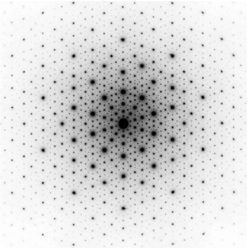
Long range order : quasiperiodic

(see Chap. 2 of [Grimm, Baake 13])

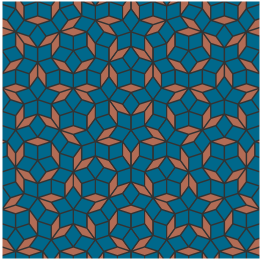
Quasicrystals

Quasicrystal $\rightarrow$ quasiperiodically arranged atoms :

- **aperiodicity**
- **long range order** (diffraction pattern exhibits sharp peaks).

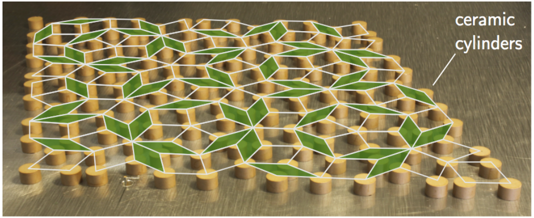


Diffraction pattern of a AlPdMn alloy (Conradin Beeli group)



A patch of the quasiperiodic Penrose tiling, used to model many quasicrystals.

Artificial quasiperiodic structures

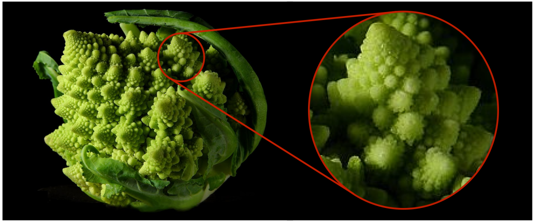


A network of dielectric resonators [Vignolo *et al.* 14]

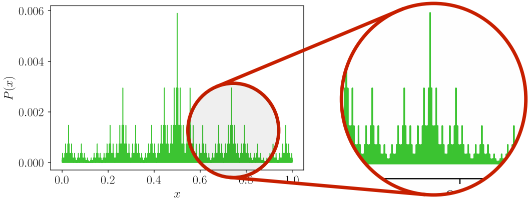
- Plasmons in semiconductor stacks [Merlin *et al.* 85]
- Microwaves in perforated metallic films [Matsui *et al.* 07]
- Microwaves in dielectric resonator networks [Vignolo *et al.* 14]
- Light solitons [Freedman *et al.* 07]
- Cold atoms in laser potentials [Guidoni *et al.* 97]
- Polaritons in wire cavities [Tanese *et al.* 14]

Fractals

Fractal : object invariant under rescaling



Romanesco broccoli (© Wikimedia commons)



Electronic density along a quasiperiodic chain

Quasicrystals *not* fractal...

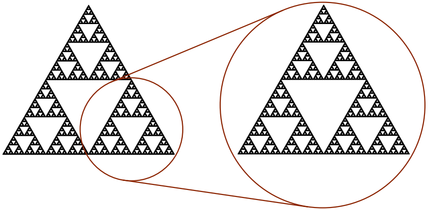
...but electrons on quasicrystals → fractal behavior

Goal :

Link the fractal behavior of the electrons to quasiperiodicity

Fractal dimensions

- $M(L) \propto L^d$ for a non-fractal d-dimensional object…What happens for a fractal one ?



A Sierpiński triangle

$$M(L) \sim L^{d_0}, \text{ with } d_0 = \log 3/\log 2$$

- d_0 is the Hausdorff fractal dimension
- $1 < d_0 \simeq 1.58 < 2$, signature of a fractal object
- Probe fractality of the q^{th} moment of a distribution $\rightarrow$ generalized fractal dimensions d_q.

Content

Constructing the Fibonacci chain

Fibonacci words

Substitution rule :

$$\mathrm{S} : \begin{cases} \mathrm{A} \to \mathrm{AB} \\ \mathrm{B} \to \mathrm{A}. \end{cases}$$

Generate a sequence of words C_l

$$C_{l+1} = \mathrm{S}(C_l)$$

$C_0 = \mathrm{B}$
$C_1 = S(C_0) = S(\mathrm{B}) = \mathrm{A}$

Properties of Fibonacci words

- Number of letters follows the Fibonacci sequence :
  $\#\mathrm{A}$ in $C_{1,2,3,4,\dots} = 1, 1, 2, 3, 5, \dots$
- $\#\mathrm{A}/\#\mathrm{B} \underset{l\to\infty}{\sim} \tau$, where
  $\tau = \frac{1+\sqrt{5}}{2} \simeq 1.61$ is the golden ratio.
- τ irrational $\to$ infinite word **aperiodic**

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$C_1 = A$
$C_2 = S(C_1) = S(A) = AB$

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$\ldots C_6 = \mathrm{ABAABABAABAAB}$

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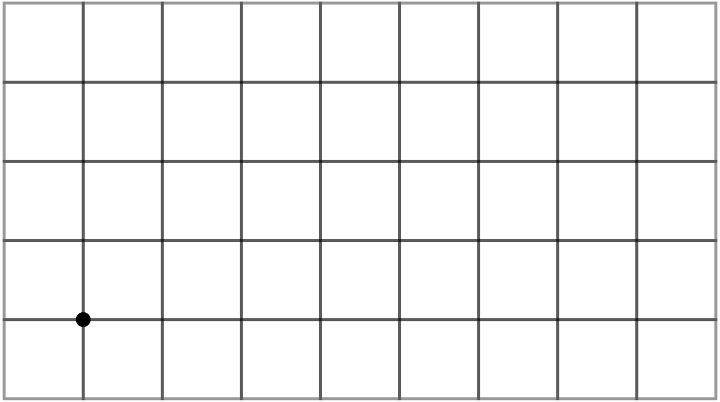
Fibonacci word from above

(Infinite) Fibonacci word : ABAABABAABAAB...

A $\leftrightarrow$ horizontal step, B $\leftrightarrow$ vertical step



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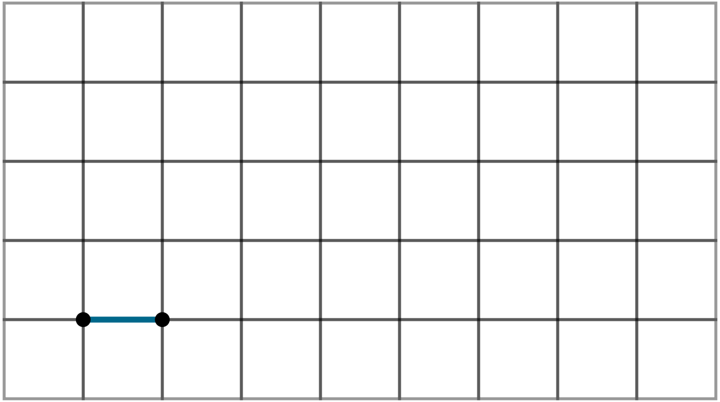

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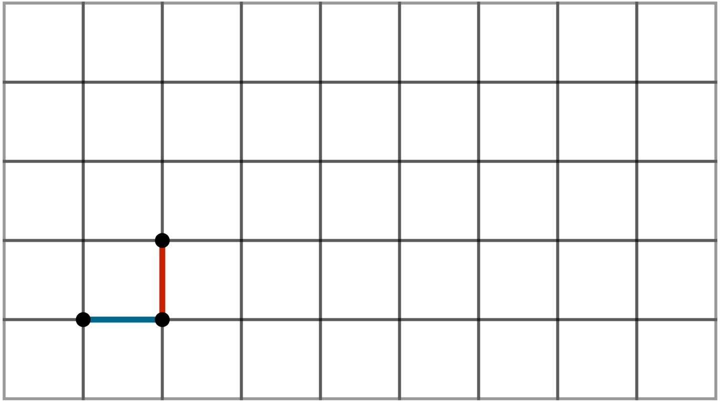

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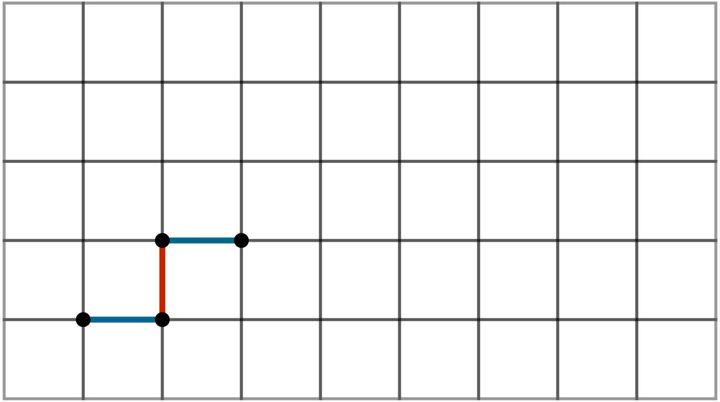

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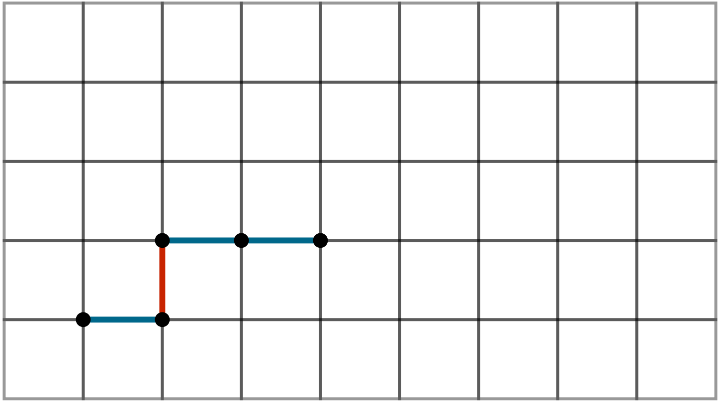

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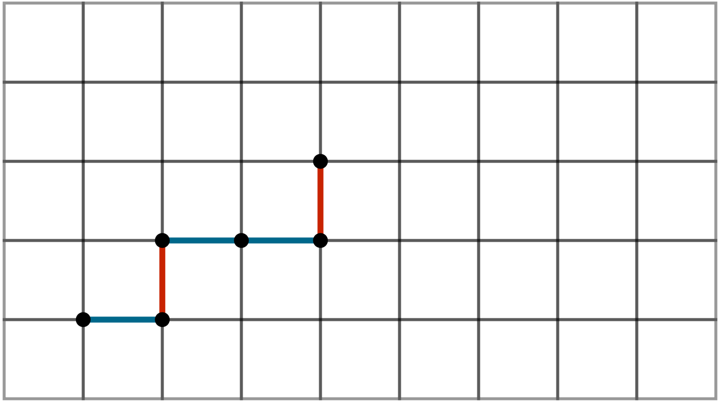

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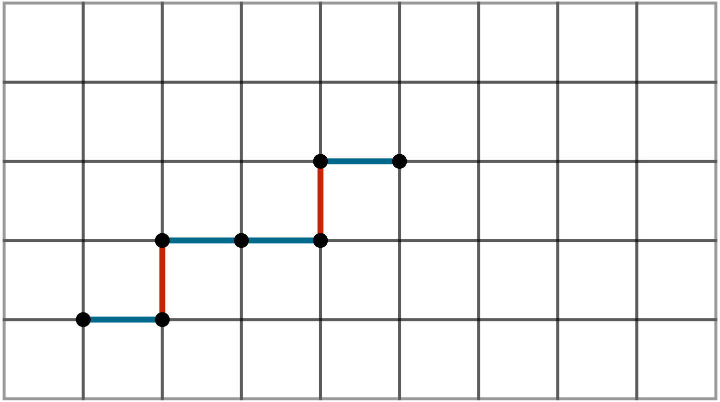

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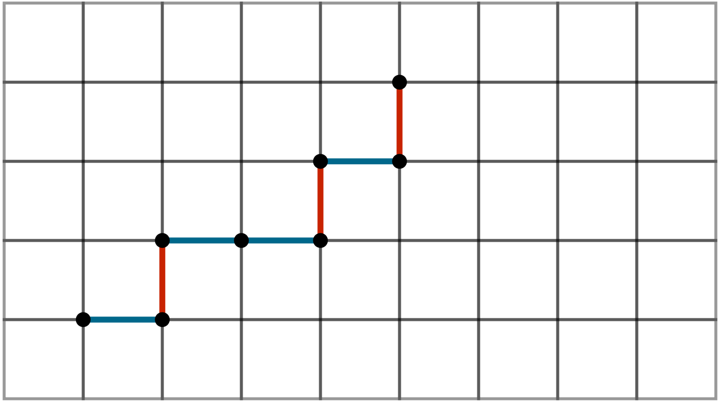

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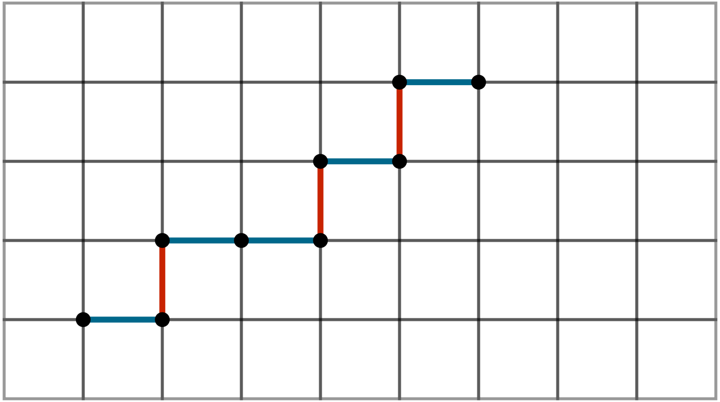

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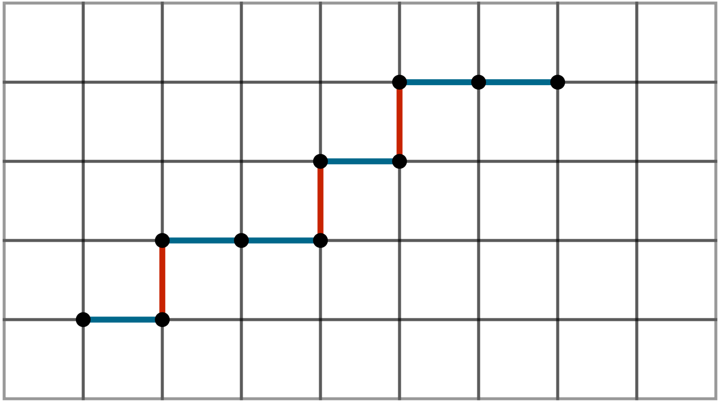

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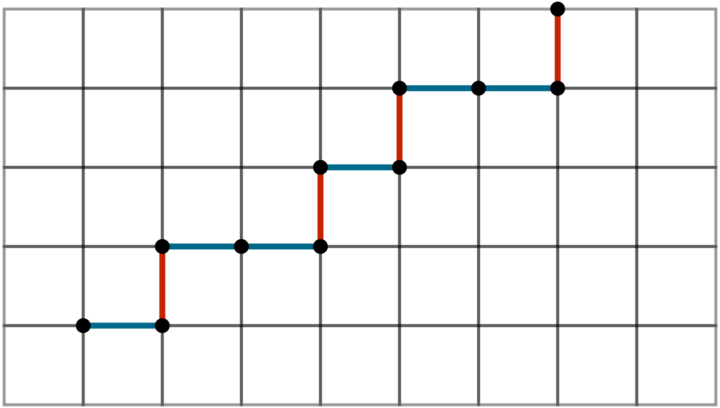

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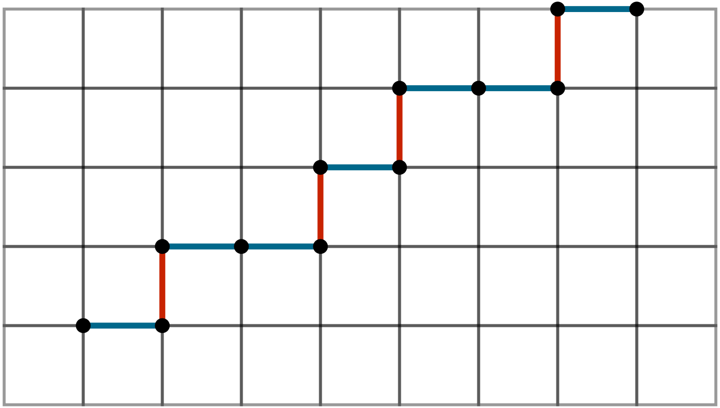

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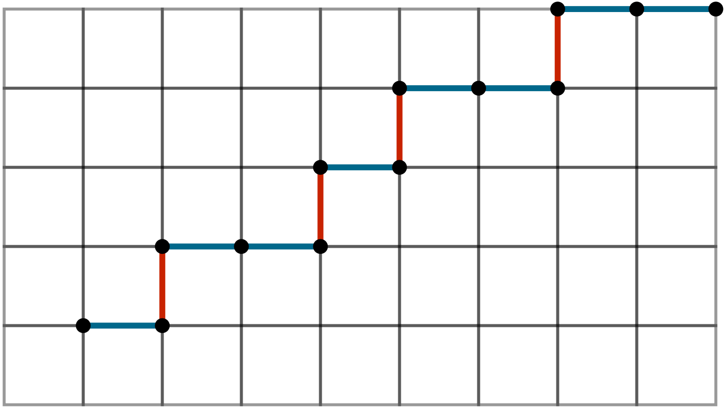
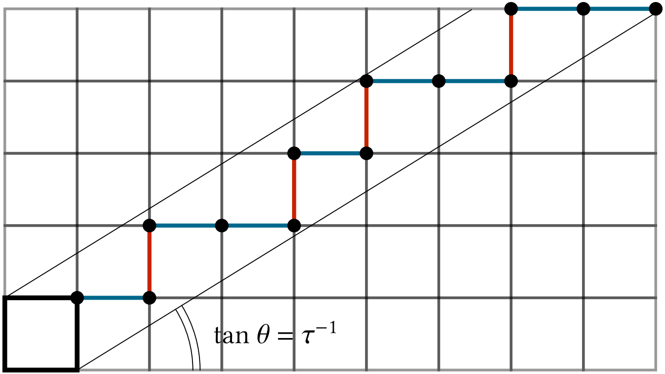


Quasiperiodicity of the Fibonacci word

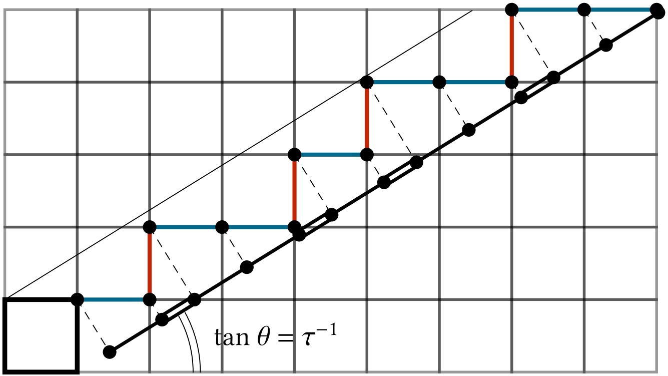


- average slope = inverse of the golden ratio ($\tau \simeq 1.6$)
- bounded fluctuations

$\to$ similar environments everywhere
$\to$ quasiperiodicity [Duneau, Katz 85]

Cut-and-project illustrated in the 1D case

C&P : general method to construct quasiperiodic tilings



The cut-and-project algorithm :

1. choose a hypercubic lattice (here $\mathbb{Z}^2$)
2. choose a "physical plane" $E_{||}$ (here a slope)
3. select points by translating the unit hypercube along $E_{||}$
4. project them onto $E_{||}$.

From letters to atoms

- The Fibonacci word : ABAABABA...
- The Fibonacci (tight-binding) chain of atoms :

Pure-hopping Hamiltonian :

$$\hat{H} = -\sum_m t_m \left|m-1\right\rangle \left\langle m\right| + \text{H.c}$$

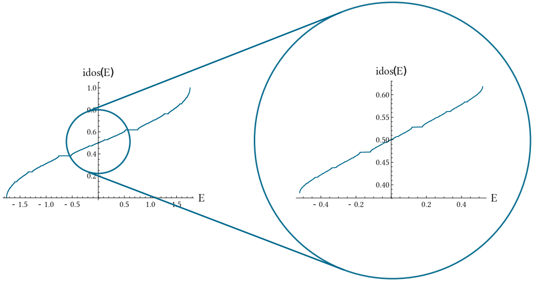
$\rightarrow \rho = t_A / t_B$ only free parameter.
Schrödinger equation for the eigenstate of energy E :

$$E\psi(m) = -t_m\psi(m-1) - t_{m+1}\psi(m+1)$$

Fibonacci spectrum and eigenstates

Numerical results

- Spectrum : fractal [Kohmoto *et al.* 83]

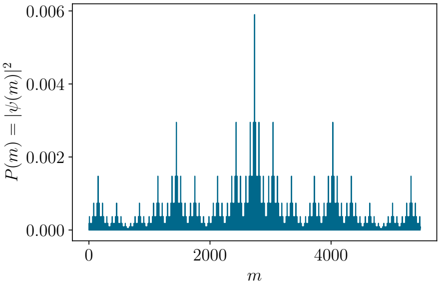


- Eigenstates : (almost all) fractal [Kohmoto *et al.* 83]

Analytical results

- Gap labeling [Bellissard 86]
- Fractal dimensions of the spectrum
  - $\rho = t_A/t_B \ll 1$: [Piéchon *et al.* 95]
  - $\rho \sim 1$: [Rüdinger, Sire 96]
- Fractal dimensions of the eigenstates, $\rho \ll 1$
  - leading order [Thiem, Schreiber 12]
  - next-to-leading order [Macé *et al.* 16]
- **exact description of the $E = 0$ state [Kohmoto *et al.* 87], [Macé *et al.* 17]**

THE BROCCOLI $E = 0$ STATE

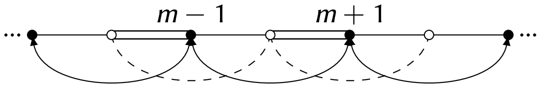


"Romanesco broccoli" fractal state at energy $E = 0$

- State at 0 energy verifies

$$t_m \psi(m-1) + t_{m+1}\psi(m+1) = 0$$

- Fibonacci chain decouples into two chains :



- Work on groups of two letters :
  - AB $\leftrightarrow$ R
  - BA $\leftrightarrow$ L
  - AA $\leftrightarrow$ U
  - BB : never occurs

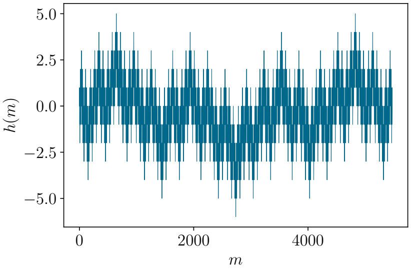
Structure of the broccoli

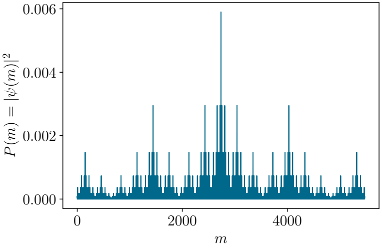
- Effective chain

$$\underbrace{AB}_{R}\,\underbrace{AA}_{U}\,\underbrace{BA}_{L}\,\underbrace{BA}_{L}\,\underbrace{AB}_{R}\,\underbrace{AA}_{U}\cdots$$

- **Arrow function** : $A(R) = +1$, $A(L) = -1$, $A(U) = 0$.
- **Height function** : $h(m) = \sum_{n \leq m} A_n$
- Let $\rho = t_B / t_A$.

$$\boxed{\psi(m) = (-1)^m \rho^{h(m)}}$$



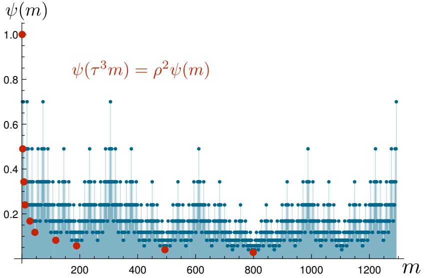


Geometry and eigenstate properties

- Arbitrary chain AAABBABB… (not necessarily Fibonacci)
- $E = 0$ state : $\psi(m) = (-1)^m \rho^{h(m)}$
- Geometry $\leftrightarrow$ $h(m)$ function

- Periodic chain : AAAAA…
  - Arrows $= 0 \implies |\psi(m)| = \text{cst}$
  - **extended state**
- Disordered chain : AAABBABB…
  - Random arrows $\implies h(m) \underset{m\to\infty}{\sim} \sqrt{m}$ $\implies |\psi(m)| \sim e^{-\sqrt{m}/\xi}$
  - **localized state**

- Quasiperiodic chain
  - Discrete scale invariance :



  - Local power-law behavior $|\psi(m)| \sim m^{-\alpha}$
  - Red points show such a local power-law, $\alpha = 2|\log \rho|/(3 \log \tau)$
  - **critical state**

Height distribution & multifractality

Partition function of the heights :

$$Z_L(\beta) = \sum_{m \in \mathcal{R}(L)} e^{-\beta h(m)}$$

Substitution $\to$ scaling law behavior :

$$Z_L(\beta) \underset{L\to\infty}{\sim} L^{\omega(\beta)}$$

with

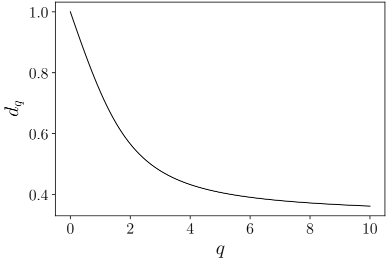
$$\omega(\beta) = \frac{\sinh^{-1}(1+\cosh(\beta))}{\log\left(2+\sqrt{5}\right)}$$

$\to$ access to the distribution of heights

Height grows slowly :

$$h_{\mathrm{typ}}(L) \sim \sqrt{\log L}$$

Fractal dimensions of the $E = 0$ state :



$0 < d_{q>0} < 1 \to$ **multifractal state**

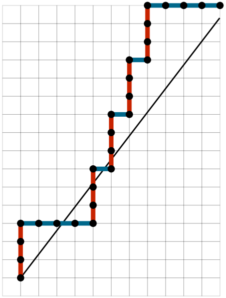
[Kohmoto *et al.* 87], [Macé *et al.* 17]

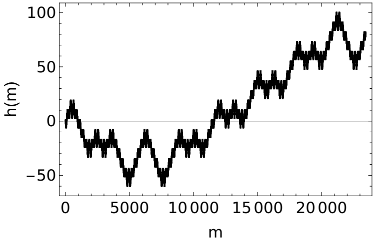
Beyond quasiperiodicity

B3 chain :

$$S : \begin{cases} A \to ABBB \\ B \to A. \end{cases}$$

Unbounded fluctuations (non-Pisot inflation factor)
$\to$ **not quasiperiodic** [Frank, Robinson 08]





Height : power-law growth [Dumont 89]

$$h(m) \sim m^{\alpha}$$

$$\alpha = \log 3 / \left(3 \log\left(\frac{1+\sqrt{13}}{2}\right)\right)$$

$\to$ **non-fractal, localized state**

$$|\psi(m)| \underset{m\to\infty}{\sim} e^{-m^{\alpha}/\xi}$$

Conclusions

$E = 0$ state of two-letters tight-binding chains :

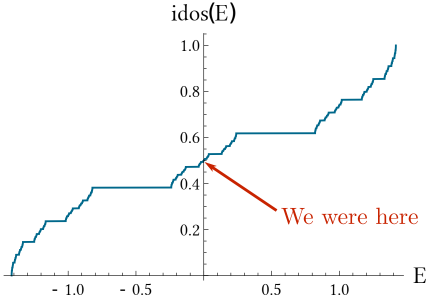
- Height field $h(m) \to \psi(m) = (-1)^m \rho^{h(m)}$

Effect of the (dis)order :

- **Periodic** $\to$ no height growth $\to$ **extended** state
- **Quasiperiodic** $\to$ slow height growth ($h(L) \sim \sqrt{\log L}$) $\to$ critical, **fractal** state
- **Random**/deterministic non-qp $\to$ fast height growth ($h(L) \sim L^\alpha$) $\to$ **localized** state

Renormalization group on the Fibonacci chain

Focused on the $E = 0$ state…



Integrated density of states (IDoS) of the Fibonacci chain

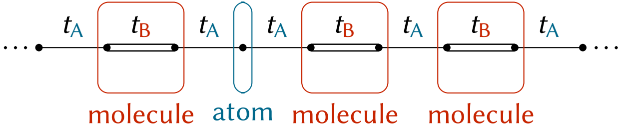
…what about the rest of the spectrum ?

Analytical results

- Gap labeling [Bellissard 86]
- Fractal dimensions of the spectrum
  - $\rho = t_A/t_B \ll 1$: [Piéchon *et al.* 95]
  - $\rho \sim 1$: [Rüdinger, Sire 96]
- Fractal dimensions of the eigenstates, $\rho \ll 1$
  - **leading order [Thiem, Schreiber 12]**
  - **higher order [Macé *et al.* 16]**
- exact description of the $E = 0$ state [Kohmoto *et al.* 87], [Macé *et al.* 17]

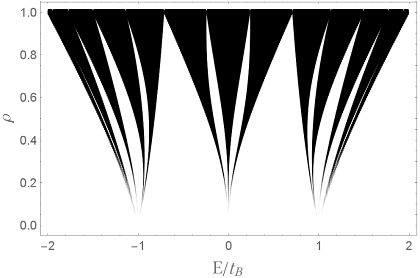
Atoms and molecules

Fibonacci chain in the limit $\rho = t_A/t_B = 0$:



$\rightarrow$ collection of decoupled atoms and diatomic molecules

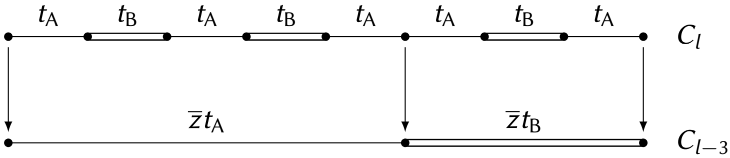
$\rho \neq 0,\ \rho \ll 1 \rightarrow$ lifted degeneracy, atomic and molecular energy clusters :



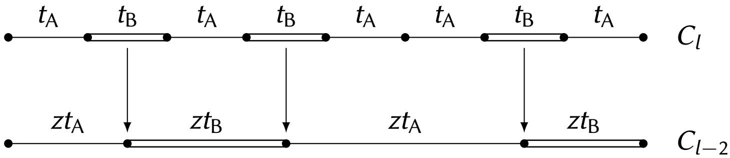
Renormalization

Substitution rule : $C_{l+1} = \mathrm{S}(C_l) \implies$ renormalization [Niu, Nori 86, Kalugin *et al.* 86]

- Atomic RG step (decimation of molecules)



- Molecular RG step (decimation of atoms)



In the limit $\rho \ll 1$, $z = \rho/2$, $\bar{z} = \rho^2$

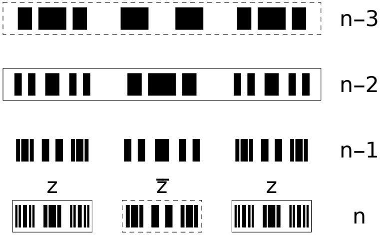
RG construction & renormalization paths

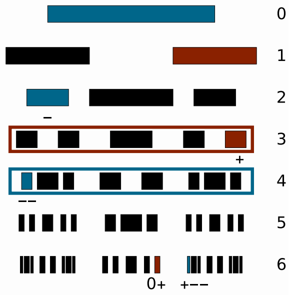
$$H_l = \underbrace{(zH_{l-2} - t_s)}_{\text{bonding levels}} + \underbrace{(\bar{z}H_{l-3})}_{\text{atomic levels}} + \underbrace{(zH_{l-2} + t_s)}_{\text{antibonding levels}} + \mathcal{O}(\rho^4)$$

→ recursive construction of the spectrum [Piéchon *et al.* 95]

Eigenstate ↔ renormalization path :

Energy spectrum

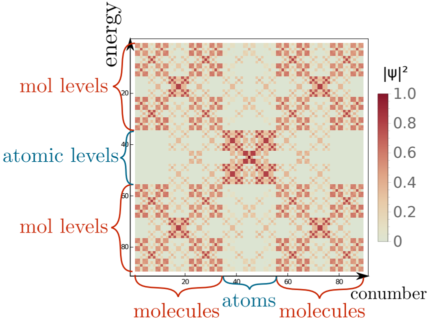




RG FOR THE EIGENSTATES

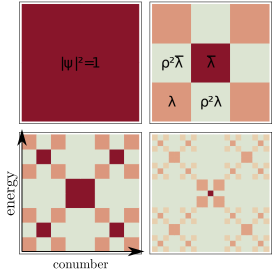
Reordering of the sites [Mosseri 88]

$\rightarrow$ self-similarity :



Fractal electronic density

RG reconstruction of the electronic density :



RG reconstructions for 4 different RG steps.

Renormalization factors :

atomic levels :

$$|\psi_m^{(l)}(E)|^2 = \bar{\lambda}|\psi_{m'}^{(l-3)}(E')|^2$$

molecular levels :

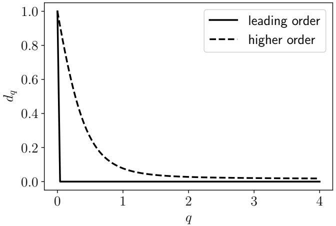
$$|\psi_m^{(l)}(E)|^2 = \lambda|\psi_{m'}^{(l-2)}(E')|^2$$

Fractality of the eigenstates

Leading order [Thiem, Schreiber 12]

$\overline{\lambda} = 1$

$\lambda = 1/2$



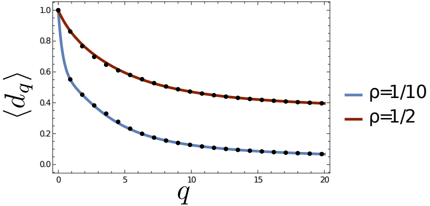
Fractal dimensions of the "broccoli state", $rho = 0.1$

$\rightarrow$ higher order : captures multifractality

Higher order [Macé *et al.* 16]

$\overline{\lambda} = \frac{1}{1+2\rho^2}$

$\lambda = \frac{1}{2+\rho^2}$



Fractal dimensions averaged over all states : numerics *vs* RG

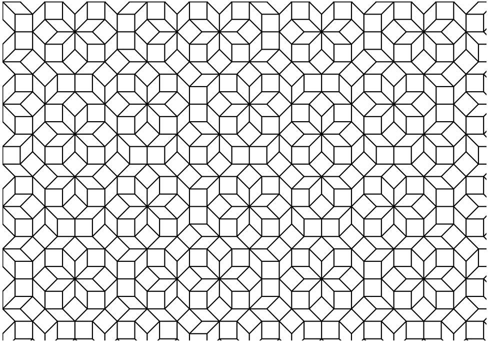
$\rightarrow$ higher order accurate for large ρ

Conclusions

Focus on the Fibonacci tight-binding chain, in the limit $\rho \ll 1$.

- Substitution rule $\rightarrow$ **scale invariance** $\rightarrow$ renormalization group
- RG $\rightarrow$ eigenstates all **multifractal**
- Higher order in ρ : captures this multifractality

Groundstate of the Ammann-Beenker tiling



A patch of the Ammann-Beenker tiling

Fractal states described by **height** functions ?

$$\psi(m) = e^{\kappa h(m)}$$

- RG description [Sire, Bellissard 90]
- Fractal spectrum [Sire 94]
- **Exact eigenstates [Sutherland 87], [Grimm, Repetowicz 98]**
- **Exact groundstate [Kalugin, Katz 14], [Macé *et al.* 17]**

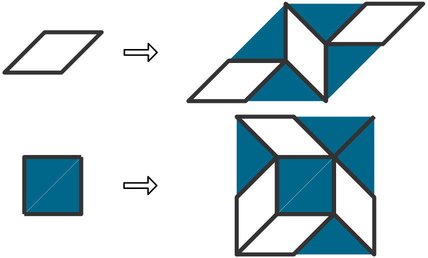
Pure hopping Hamiltonian :

$$\hat{H} = -t \sum_{\langle m,n \rangle} |m\rangle \langle n|$$

Quasiperiodicity encoded in adjacency

Looking for arrows

Like 1D chains, Ammann-Beenker has a substitution rule :

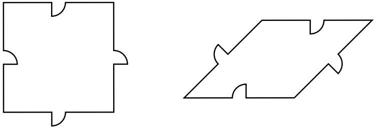


$\rightarrow$ scale invariance.

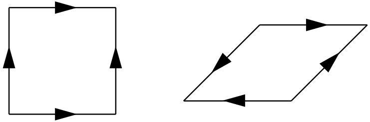
Height requires a field of arrows :

- invariant under substitution
- irrotational

Ammann-Beenker $\rightarrow$ notched tiles...



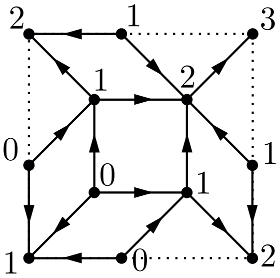
...exactly what we need !



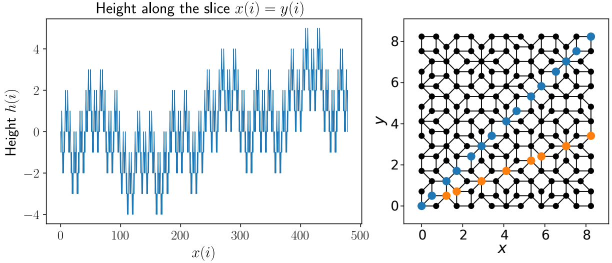
Height field :

$$h(m) = \sum_{0 \rightarrow m} \text{arrows}$$

Properties of the height field



The height field on a small patch of the tiling.



Height along a line, shown in blue on the tiling.

Partition function : $Z_L(\beta) \underset{L\to\infty}{\sim} L^{\omega(\beta)}$

$\to$ slow growth : $h_{\text{typ}}(L) \underset{L\to\infty}{\sim} \sqrt{\log L}$

$\to$ states $\psi(m) = e^{\kappa h(m)}$ fractal

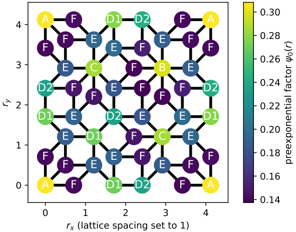
Groundstate

$$\hat{H} = -t \sum_{\langle m,n \rangle} |m\rangle \langle n|$$

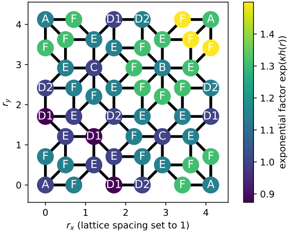
Conjecture [Kalugin, Katz 14] :

$$\psi_{\text{groundstate}}(m) = C(m) e^{\kappa h(m)}$$

$C(m)$: *local* function :

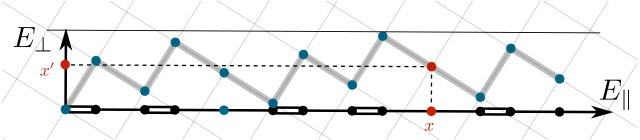


$e^{\kappa h(m)}$: non-local function :

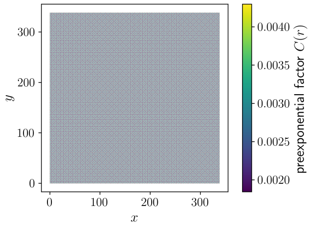


Testing the conjecture

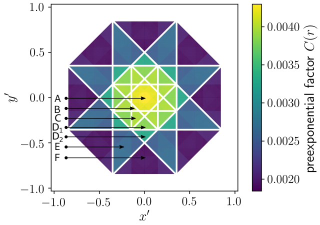
Cut-and-project $\rightarrow$ physical ($E_{||}$) and internal ($E_\perp$) space :



Internal space $\rightarrow$ sites classified by local environment $\rightarrow$ test the local nature of C :



C part in physical space (finite system of $\simeq 3 \times 10^5$ atoms)



The C part in internal space

Perturbing the Hamiltonian

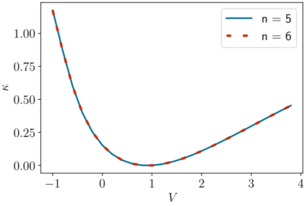
Adding an on-site potential :

$$\hat{H} = -t \sum_{\langle m,n \rangle} |m\rangle \langle n| + \sum_m V_m |m\rangle \langle m|$$

Laplacian-like [Sire, Bellissard 90] :

$V_m = V z_m$

$\to \psi(m) = C(m) e^{\kappa h(m)}$

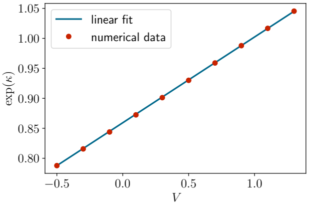


Prefactor κ as a function of V.

An "arbitrary" potential :

$V_m = V$ if m has 3 neighbors, else $V_m = 0$.

$\to \psi(m) = C(m) e^{\kappa h(m)}$



e^{κ} as a function of V.

Conclusions

Tight-binding models on a 2D quasiperiodic tiling

- Geometry (notches) $\rightarrow$ **height field** $h(m) \rightarrow \psi_{\text{groundstate}}(m) = C(m)e^{\kappa h(m)}$
- Slow height growth ($h(L) \sim \sqrt{\log L}$) $\rightarrow$ critical, **fractal** state

$\rightarrow$ Groundstate in 2D $\leftrightarrow$ $E = 0$ state in 1D

- Robust to symmetry-preserving on-site perturbations
- Same conclusions for the 10-fold symmetric Penrose tiling.

General conclusion

Simple tight-binding models on 1D and 2D quasiperiodic tilings

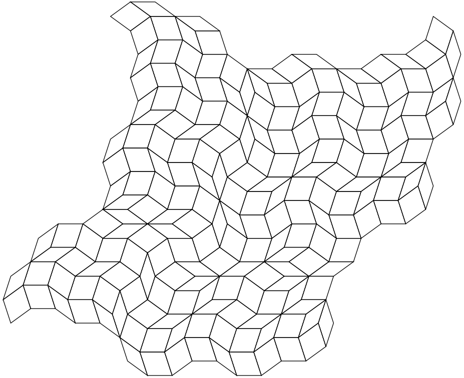
- Quasiperiodic structures : in between periodic and random, very close to periodic
- Critical, fractal eigenstates
- Consequence of quasiperiodic geometry

Perspectives

- Topology : the internal space degree of freedom can be used to probe topological properties, for the Fibonacci chain [Tanese *et al.* 14]. What about 2D quasicrystals ?
- Interactions : quasiperiodicity $\simeq$ deterministic disorder. Non-interacting eigenstates are not localized, yet many-body quasiperiodic systems seem to localize …how ?

Perspectives : exact eigenstates

Systematically study the exact eigenstates ($E = 0$ state in 1D, groundstate in 2D) for different *classes* [Julien 10] of aperiodic structures.



The binary tiling : a non-Pisot 2D tiling [Godrèche, Lançon 92]

Thanks to...

- Michel Duneau, Jean-Noël Fuchs, Pavel Kalugin, Rémi Mosseri, Frédéric Piéchon for fruitful discussions and collaborations,
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