Which distributions of matter diffract? — Some answers

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1 Introduction

Diffraction experiments are important to determine the structure of a solid, even more so with the refined methods available today. Recent applications include aperiodic systems as well as systems with disorder.

Kinematic diffraction can be described and understood in terms of Fourier analysis. The diffraction image is related to the Fourier transform of the autocorrelation or Patterson function of the scattering obstacle, e.g., the electron density. The situation is well understood for (ideal) crystals, which show a complete lattice of periods, even though the corresponding inverse problem does not have a unique solution in general.

Beyond the periodic situation, firm results are sparse, and until recently, one did not even know which general distribution of scatterers would result in a pure point diffraction spectrum, i.e., in a diffraction image of pure Bragg peaks only. This review revolves around this question [1], and summarises the present state of affairs, with special emphasis on contributions obtained during the time of the DFG focus program on quasicrystals. We concentrate on the few results here that have been established *rigorously*, and indicate where further research is needed.

The article is organised as follows: In the first section, we state conditions (Theorem 1) under which a set of scatterers diffract, i.e., its diffraction spectrum consists of Bragg peaks only. Instead of looking at very general set of scatterers, we specialise in those sets that model physical structures (as weighted Dirac combs). Theorem 2 then states criteria under which these sets diffract. With this result, we explain in Sec. 3 how a cut and project scheme naturally appears. So, we obtain the associated internal space of model sets, like the Fibonacci or the rhombic Penrose tilings, through the information given by the autocorrelation. Model sets (or cut and project sets) are the most common models for aperiodic order (quasicrystals).

In Sec. 4 we investigate lattice substitution systems. We first explore the diffraction spectrum for scatterers distributed (aperiodic or even stochastic) over a lattice (Theorems 4 and 5), before turning to distributions obtained by substitutions. For this special case, we are again interested under which conditions they diffract (Theorem 6). As an application of a lattice substitution system as well as a model set with non-Euclidean internal space, we calculate, in Sec. 5, the diffraction spectrum of the paperfolding sequence.

Thereafter, we leave the area of deterministic point sets and turn to systems with disorder, i.e., to random tilings in Sec. 6. We carefully introduce the notion of a random tiling, before we state results about one dimensional binary random tilings in Theorem 8 and about the two dimensional Ising lattice gas in Theorem 9. We end this section by the example of a Fibonacci random tiling, where we are particularly interested in the role of the internal space of quasiperiodic random tilings for their diffraction spectrum (Theorems 10 and 11). We conclude our article with an outlook (Sec. 7) where we indicate future directions in diffraction theory.

2 Mathematical diffraction theory

The basic object of interest is a set of scatterers in a Euclidean[1] space $\mathbb{R}^d$, which we model by a *translation bounded complex measure*[2] ω. It describes the distribution of matter in a mathematically adequate way. To calculate the diffraction spectrum, we need the *autocorrelation measure* γ_ω attached to ω. The *diffraction spectrum* is then the Fourier transform $\hat{\gamma}_\omega$ of the autocorrelation measure. Here, $\hat{\gamma}_\omega(E)$ is the total intensity scattered into the volume element E, and thus describes the outcome of a diffraction experiment, compare [8]. It can uniquely be decomposed as $\hat{\gamma}_\omega = (\hat{\gamma}_\omega)_{\mathrm{pp}} + (\hat{\gamma}_\omega)_{\mathrm{sc}} + (\hat{\gamma}_\omega)_{\mathrm{ac}}$ by the Lebesgue decomposition theorem, see [4]. Here, $(\hat{\gamma}_\omega)_{\mathrm{pp}}$ is a *pure point measure*, which corresponds to the Bragg part of the diffraction spectrum, $(\hat{\gamma}_\omega)_{\mathrm{ac}}$ is *absolutely continuous* and $(\hat{\gamma}_\omega)_{\mathrm{sc}}$ *singular continuous* with respect to Lebesgue measure. The pure pointedness of the diffraction spectrum $\hat{\gamma}_\omega$ of ω is in question, i.e., whether the Lebesgue decomposition reduces to $\hat{\gamma}_\omega = (\hat{\gamma}_\omega)_{\mathrm{pp}}$ (then, the diffraction spectrum consists of Bragg peaks only).

We first fix an averaging sequence $\mathcal{A} = \{B_n \mid n \in \mathbb{N}\}$ of balls of radius n around 0, so we begin analysing the spectrum by looking at finite pieces (balls) of the structure in question. We define $\tilde{\omega}(f) = \overline{\omega(\tilde{f})}$, where $\tilde{f}(x) = \overline{f(-x)}$, and set $\omega_n = \omega|_{B_n}$ and $\tilde{\omega}_n = (\omega_n)\tilde{}$. Then, the measures

$$\gamma_\omega^{(n)} = \frac{\omega_n * \tilde{\omega}_n}{\mathrm{vol}(B_n)} \tag{1}$$

are well-defined, since they are (volume averaged) convolutions of two measures with compact support. The autocorrelation γ_ω of ω exists, if $(\gamma_\omega^{(n)})_{n\in\mathbb{N}}$ converges in the vague topology, compare [6], and is then, by construction, a positive definite[3] measure.

[1]Most results also apply to more general spaces, namely σ-compact locally compact Abelian groups, with Lebesgue measure replaced by Haar measure, etc., see [2].

[2]Frequently, we make use of the one-to-one correspondence between *measures* and regular *Borel measures* by the Riesz-Markov representation theorem, where a *measure* is a continuous linear functional on the space of compactly supported continuous functions on $\mathbb{R}^d$, while a *Borel measure* is defined on the Borel sets of $\mathbb{R}^d$. The *convolution* of two measures μ, ν is defined as $\mu * \nu(f) = \int_{\mathbb{R}^d \times \mathbb{R}^d} f(x+y)\, d\mu(x)\, d\nu(y)$ and is well-defined if at least one of them has compact support. A measure μ is *translation bounded* if, for all compact $K \subset \mathbb{R}^d$, $\sup_{t\in\mathbb{R}^d} |\mu|(t+K) \leq C_K < \infty$ for some constant C_K which only depends on K. Here, $|\mu|$ denotes the *total variation measure* (which is positive) and $t + K = \{t + x \mid x \in K\}$. See [2, 3, 4, 5, 6, 7] and references therein for details.

[3]A measure μ is *positive definite* iff $\mu(f * \tilde{f}) \geq 0$ for all complex-valued continuous functions with compact support, compare [3].

We say that a measure is *almost periodic* if the set of translates is *relatively compact* (i.e., its closure is compact). Of course, we have to fix a topology for this, and in our case we need the *strong* or *product topology* on the space of translation bounded complex measures (and not the vague topology), see [2, 5]. With this topology, we speak of *strong almost periodicity* to distinguish it from almost periodicity in other topologies. The key result reads:

Theorem 1 [2, 5] *The measure ω is pure point diffractive iff*

(i) γ_ω *exists.*

(ii) γ_ω *is strongly almost periodic.* □

Note that (i) is merely a convention, since one can always pick an appropriate subsequence of the averaging sequence $\mathcal{A}$ for which γ_ω exists.

So, given a measure ω, we have to check these two conditions to decide whether it is pure point diffractive. This can be done for many relevant examples. We would also like to solve the *homometry* or *inverse problem*, i.e., the question which measures ω account for a given diffraction spectrum $\hat{\gamma}$. This is a hard problem, because there is no inversion process of Eq. 1. In fact, rather different ω can have the same diffraction, see [9]. One also would like to understand the implications of a pure point spectrum. Here, we will not explore these last two questions further.

Instead, let us specialise on the situation of a countable set S of scatterers in $\mathbb{R}^d$ with (bounded) scattering strengths $v(x)$, $x \in S$. It can be represented as a complex Borel measure in the form of a *weighted Dirac comb*[4]

$$\omega = \sum_{x \in S} v(x)\, \delta_x ,$$

where δ_x is the unit point (or Dirac) measure located at x, i.e., $\delta_x(\varphi) = \varphi(x)$ for continuous functions φ. This way, atoms are modeled by their position and scattering strengths. Convolutions with more realistic profiles are not considered here, but can easily be treated by the convolution theorem, see [3].

Denote the set of "inter-atomic distances" by $\varDelta = S - S = \{x - y \mid x, y \in S\}$. Then we make the following three assumptions, see [2]:

(A 1) The measure ω is translation bounded, i.e, there exist constants C_K so that

$$\sup_{t \in \mathbb{R}^d} \sum_{x \in S \cap (t+K)} |v(x)| \;\leq C_K < \infty$$

for all compact sets $K \subset \mathbb{R}^d$.

(A 2) The *autocorrelation coefficients*

$$\eta(z) = \lim_{n\to\infty} \frac{1}{\mathrm{vol}(B_n)} \sum_{\substack{x,y \in S \cap B_n \\ x-y=z}} v(x)\, \overline{v(y)}$$

exist for all $z \in \varDelta$ (we set $\eta(z) = 0$ if $z \notin \varDelta$). Consequently (if also (A 3) holds), the autocorrelation measure $\gamma_\omega = \sum_{z \in \varDelta} \eta(z)\, \delta_z$ exists.

[4]For later reference, we write $\omega = A \cdot \delta_S$ if $v(x) = A$ for all $x \in S$.

(A 3) The set $\Delta^{\text{ess}} = \{z \in \Delta \mid \eta(z) \neq 0\} \subset \Delta$ is *uniformly discrete*, i.e., there is an $r > 0$ such that open balls of radius r centred at the points of Δ^{ess} are mutually disjoint.

The support of γ_ω (which is the support of $\eta(z)$) plays a special role, in particular the group L generated by it:

$$L = \langle \Delta^{\text{ess}} \rangle_{\mathbb{Z}} \subset \mathbb{R}^d \tag{2}$$

The point set S is *repetitive* if for any compact set $K \subset \mathbb{R}^d$, $\{t \in \mathbb{R}^d | S \cap K = (t+S) \cap K\}$ is relatively dense[5]; i.e., there exists a radius $R = R(K) > 0$ such that every open ball $B_R(y)$ contains at least one element of $t \in \mathbb{R}^d$ for which $S \cap K = (t+S) \cap K$. If S is repetitive, we have $\Delta = \Delta^{\text{ess}}$, hence $L = \langle \Delta \rangle_{\mathbb{Z}}$.

We define a *translation invariant pseudo-metric*[6] by

$$\varrho(s,t) = \left| 1 - \frac{\eta(s-t)}{\eta(0)} \right|^{\frac{1}{2}}. \tag{3}$$

If all weights $v(x)$ are non-negative, one has $\varrho(s,t) \leq 1$ for all $s,t \in \mathbb{R}^d$, but, in general, ϱ is bounded by $\sqrt{2}$. This pseudo-metric ϱ defines a *uniformity*, both on L and $\mathbb{R}^d$, see [2]. The induced topology is, in general, completely different from the usual Euclidean topology of $\mathbb{R}^d$. It is called the *autocorrelation topology*.

Next we define, for $\varepsilon > 0$, a set P_ε of ε*-almost periods* of the autocorrelation γ_ω through

$$P_\varepsilon = \{t \in \mathbb{R}^d \mid \varrho(t,0) < \varepsilon\}. \tag{4}$$

We clearly have the following inclusions for $\varepsilon < \varepsilon'$:

$$\{\text{periods of } \omega\} \quad \subset \quad P_\varepsilon \quad \subset \quad P_{\varepsilon'} \quad \subset \quad \mathbb{R}^d,$$

and furthermore $P_1 = \Delta^{\text{ess}}$ if all weights $v(x)$ are non-negative. Now, we are able to apply Theorem 1 to weighted Dirac combs.

Theorem 2 [2] *Let ω be a weighted Dirac comb that satisfies* (A 1), (A 2) *and* (A 3). *Then $\hat{\gamma}_\omega$ exists, and the following statements are equivalent:*

(i) *P_ε is relatively dense for all $\varepsilon > 0$.*

(ii) *γ_ω is norm almost periodic.*

(iii) *γ_ω is strongly almost periodic.*

(iv) *$\hat{\gamma}_\omega$ is pure point diffractive.* □

The *norm almost periodicity* in (ii) refers to the topology defined by the norm $||\omega||_K = \sup_{x \in \mathbb{R}^d} |\omega|(x+K)$ for some fixed compact K with nonempty interior, e.g., a closed unit ball. The concept of norm almost periodicity is independent of the choice of K.

This theorem applies to the diffraction from the visible lattice points, see [2, 10], but also to model sets which we will consider next.

[5] A set Q is *relatively dense* if there is a radius $R > 0$ such that every ball of radius R in $\mathbb{R}^d$ contains at least one point of Q.

[6] A *pseudo-metric* is a non-negative, symmetric function on $\mathbb{R}^d \times \mathbb{R}^d$ that satisfies the triangle inequality. Such a function ϱ is *translation invariant* if $\varrho(s+r, t+r) = \varrho(s,t)$ for all $r \in \mathbb{R}^d$.

3 Model sets

The well-known cut and project mechanism provides examples of weighted point sets which satisfy the assumptions (A 1), (A 2) and (A 3). But here, we start with a countable weighted set of scatterers in $\mathbb{R}^d$ for which the corresponding weighted Dirac comb fulfils Theorem 2 and is therefore pure point diffractive. This will lead us to a suitable cut and project scheme.

The constructive picture behind this is the following: On L of Eq. 2, we have a pseudo-metric ϱ, defined in Eq. 3, which in turn defines a uniformity, and then the *autocorrelation topology* on L. Inside L, the sets P_ε of Eq. 4 are the open balls of radius ε around 0 in the autocorrelation topology. The group L equipped with this topology admits a *(Hausdorff) completion*[7] H, which is a *locally compact Abelian group*[8]. This means that there is a continuous group homomorphism $\varphi\colon L \to H$ where $\varphi(L)$ is dense in H. The relative denseness of P_ε is crucial here, because for every set P_ε of L there is an open set $B(\varepsilon)$ of H so that $\varphi(P_\varepsilon) = \varphi(L) \cap B(\varepsilon)$ and $B(\varepsilon)$ has compact closure $\overline{B(\varepsilon)}$ for $0 < \varepsilon < 1$, giving the local compactness of H.

We obtain the following *cut and project scheme*, see [2]:

$$\begin{array}{ccccc} \mathbb{R}^d & \xleftarrow{\;\pi\;} & \mathbb{R}^d \times H & \xrightarrow{\;\pi_{\text{int}}\;} & H = \overline{\varphi(L)} \\ \cup & \nwarrow_{1\text{–}1} & \cup & \nearrow_{\text{dense}} & \\ L & \longleftrightarrow & \tilde{L} = \{(x, \varphi(x)) \mid x \in L\} & & \end{array} \qquad (5)$$

Here, $\tilde{L}$ is a *lattice* in $\mathbb{R}^d \times H$, i.e., $\tilde{L}$ is a closed subgroup so that the factor group $(\mathbb{R}^d \times H)/\tilde{L}$ is compact. The projection π_{int} is dense in *internal space* H and the projection π into *physical space* $\mathbb{R}^d$ is one-to-one on $\tilde{L}$.

The internal space H and the lattice $\tilde{L}$ both arise from the group L via the autocorrelation topology. In spite of its abstraction, the procedure gives back the familiar Euclidean internal spaces of the well-known examples (e.g. Fibonacci). In the case of the rhombic Penrose tilings it gives the minimal internal space possible for representing them in the cut and project formalism: $H = \mathbb{R}^2 \times (\mathbb{Z}/5\mathbb{Z})$, see [2]. For other tilings, the internal space may be p-adic, see Section 5 for an example. Note that the completion map φ is not one-to-one in general, its kernel is $\bigcap_{\varepsilon > 0} P_\varepsilon$, the group of statistical periods. For example, if S is a lattice with all weights equal, then $S = L$, $\varphi \equiv 0$, $H = 0$ and $\tilde{L} \cong L$, so the cut and project scheme collapses into triviality. In general, the internal space H ignores the periodic part of ω, for which no additional structure is required, and reflects only the aperiodic parts.

A set $\Lambda \subset \mathbb{R}^d$ is a *model set* for the cut and project scheme in Eq. 5, if there is a relatively compact set $W \subset H$ with non-empty interior and a $t \in \mathbb{R}^d$ such that

$$\Lambda = t + \Lambda(W) = t + \{x \in L \mid \varphi(x) \in W\}.$$

[7] H is a *completion* of L, if L has dense image in H and every Cauchy sequence in H has a limit in H, e.g., $\mathbb{R}$ is the completion of $\mathbb{Q}$. The completion is unique up to topological isomorphism.

[8] A topological space is called *locally compact* if each point is contained in a compact neighbourhood. If this space is an Abelian group, we speak of a locally compact Abelian group. Examples are Euclidean and p-adic spaces.

Note that, in the context of model sets, the map φ is often called the $\star$-map and denoted by $(\cdot)^\star$, i.e., one writes $x^\star = \varphi(x)$. A model set is always a *Delone set*, i.e., it is both relatively dense and uniformly discrete. A model set is called *regular* if the boundary of W has *Haar measure*[9] 0. Regular model sets are the most relevant model sets for the physical applications in the theory of quasicrystals. They also play a prominent role in the analysis of sequences with long-range (aperiodic) order, cf. [11] and references therein.

One of the cornerstones of the theory of model sets is:

Theorem 3 [6, 12] *Regular model sets are pure point diffractive.* □

Let us now go back to our discussion of diffraction in the context of the assumptions (A 1), (A 2) and (A 3). The pure point diffraction in Theorem 2 is intimately related to the cut and project scheme in Eq. 5. But it can happen that the set S itself is not a model set (e.g., as in the case of the visible lattice points), see [2]. So the question arises: Which pure point diffractive point sets *are* (regular) model sets? We have only partial progress on this.

Noting that $W_\varepsilon = \overline{\varphi(P_\varepsilon)}$ has non-empty interior for all $0 < \varepsilon < 1$, it follows form our above discussion that $P_\varepsilon \subset \Lambda(W_\varepsilon)$. Since L is countable, we even get that $P_\varepsilon = \Lambda(W_\varepsilon)$ can be violated for at most countably many values of ε, so that Δ^{ess} is the union of an ascending sequence of model sets. Furthermore we have

Proposition 1 [13] *Assume* (A 1), (A 2) *and* (A 3) *hold. Then* Δ^{ess} *is a model set.* □

This leaves open the question of whether or not Λ is a model set. Progress in this seems to depend on utilising the dynamical hull of Λ:

$$X(\Lambda) = \overline{\{\Lambda + t \mid t \in \mathbb{R}^d\}}, \tag{6}$$

where closure is taken with respect to the *local topology*[10]. Then $\big(X(\Lambda), \mathbb{R}^d\big)$ is a *dynamical system* under the obvious action of $\mathbb{R}^d$ on $X(\Lambda)$.

If Λ is a repetitive regular model set then it is known [12] that $X(\Lambda)$ is *strictly ergodic*, i.e., both minimal[11] and uniquely ergodic[11]. There is a conjecture to the effect that, conversely, any pure point diffractive *Meyer set*[12] of $\mathbb{R}^d$ for which $X(\Lambda)$ is strictly ergodic is in fact a model set.

4 Lattice substitution systems

An interesting class of point sets is formed by the subsets of a lattice. Even though they can be aperiodic or even stochastic, the underlying lattice Γ leaves its imprint, most notably in form of a periodicity of the diffraction, with the dual lattice Γ^* as lattice of periods.

[9] On every locally compact Abelian group there exists a unique (up to a multiplicative constant) translation invariant regular Borel measure. This is called the Haar measure, and is given by the Lebesgue measure on Euclidean space $\mathbb{R}^d$.

[10] Informally, two discrete and closed point sets are close in the local topology, if, after a small translation, these two coincide on a large compact region.

[11] A dynamical system (X, T) is *minimal* in case X has no proper closed T-invariant subsets. It is *uniquely ergodic* if there is only one T-invariant Borel probability measure on X.

[12] A Delone set S is a *Meyer set* iff the set of "inter-atomic distances" $\Delta = S - S$ is also a Delone set. Every model set is a Meyer set.

In a more general formulation, let $v\colon \Gamma \to \mathbb{C}$ be any bounded function, and consider the weighted Dirac comb

$$\omega = \sum_{x\in\Gamma} v(x)\, \delta_x .$$

This includes the previous case via $v = 1_S$, i.e., $v(x) = 1$ for $x \in S$ and 0 otherwise. Then, if γ_ω is any of its autocorrelations (e.g. as obtained along a suitable subsequence of averaging balls), we have the following result.

Theorem 4 [14] *Let Γ be a lattice*[13] *in $\mathbb{R}^d$ and ω a weighted Dirac comb on Γ with bounded complex weights. Let γ_ω be any of its autocorrelations, i.e., any of the limit points of the family $\{\gamma_\omega^{(n)} \mid n \in \mathbb{N}\}$. Then the following holds.*

(i) *The autocorrelation γ_ω can be represented as*

$$\gamma_\omega = \Theta \cdot \delta_\Gamma = \sum_{x\in\Gamma} \Theta(x)\, \delta_x ,$$

where $\Theta\colon \mathbb{R}^d \to \mathbb{C}$ is a bounded continuous positive definite function that interpolates the autocorrelation coefficients $\eta(x)$ as defined at $x \in \Gamma$. Moreover, there exists such a Θ which extends to an entire function $\Theta\colon \mathbb{C}^d \to \mathbb{C}$ with the additional growth restriction that there are constants $C, R \geq 0$ and $N \in \mathbb{Z}$ such that $|\Theta(z)| \leq C \cdot (1 + |z|)^N \cdot \exp(R\,|\mathrm{Im}(z)|)$ for all $z \in \mathbb{C}^d$.

(ii) *The diffraction spectrum $\hat{\gamma}_\omega$ of ω is a translation bounded positive measure that is periodic with the dual lattice*[14] *$\Gamma^* = \{k \in \mathbb{R}^d \mid \langle k, x\rangle \in \mathbb{Z} \text{ for all } x \in \Gamma\}$ as lattice of periods. Furthermore, $\hat{\gamma}_\omega$ has a representation as a convolution,*

$$\hat{\gamma}_\omega = \varrho * \delta_{\Gamma^*} ,$$

in which ϱ is a finite positive measure supported on a fundamental domain of Γ^* that is contained in the ball of radius R around the origin. $\square$

This reduces the analysis of the spectral type of $\hat{\gamma}_\omega$ to that of ϱ, which has compact support.

An interesting application concerns lattice subsets and their complements, which leads to the following result.

Theorem 5 [14] *Let Γ be a lattice*[13] *in $\mathbb{R}^d$, and let $S \subset \Gamma$ be a subset with existing (natural) autocorrelation coefficients $\eta_S(z) = \mathrm{dens}\,(S \cap (z + S))$. Then the following holds.*

(i) *The autocorrelation coefficients $\eta_{S^c}(z)$ of the complement set $S^c = \Gamma \setminus S$ also exist. They are $\eta_{S^c}(z) = 0$ for all $z \notin \Gamma$ and otherwise, for $z \in \Gamma$, satisfy the relation*

$$\eta_{S^c}(z) - \mathrm{dens}(S^c) = \eta_S(z) - \mathrm{dens}(S)$$

[13] These results can be generalised to lattice subsets in locally compact Abelian groups whose topology has a countable base, see [14] for details.

[14] The Euclidean scalar product is denoted by $\langle \cdot, \cdot \rangle$.

(ii) *If, in addition,* $\mathrm{dens}(S) = \mathrm{dens}(\varGamma)/2$*, the sets* S *and* S^c *are homometric.*

(iii) *The diffraction spectra of the sets* S *and* S^c *are related by*

$$\hat{\gamma}_{S^c} = \hat{\gamma}_S + (\mathrm{dens}(S^c) - \mathrm{dens}(S)) \cdot \mathrm{dens}(\varGamma)\, \delta_{\varGamma^*} .$$

In particular, $\hat{\gamma}_{S^c} = \hat{\gamma}_S$ *if* $\mathrm{dens}(S^c) = \mathrm{dens}(S)$.

(iv) *The diffraction measure* $\hat{\gamma}_{S^c}$ *is pure point iff* $\hat{\gamma}_S$ *is pure point.* □

As an immediate consequence of part (i), one can check that the two pseudometrics defined by S and S^c via Eq. 3 are scalar multiples of one another, hence define the same uniformity (as long as $\mathrm{dens}(S) \cdot \mathrm{dens}(S^c) > 0$).

Of considerable interest in this context are lattice subsets which are obtained by lattice substitution systems via *Delone multisets*[15]. They are the natural generalisation of one dimensional substitution rules with constant length, and it was recently possible [15, 16, 17] to find a complete generalisation of Dekking's coincidence criterion (see [18]) to this general situation. The result, stated below, is a circle of equivalences which directly puts pure pointedness and model sets on the same footing. Although one of the equivalent criteria is modular coincidence, we refer the reader to [17] for the rather technical definition, just pointing out that its primary virtue is that it is testable by a straightforward algorithm.

A *matrix function system* (MFS) on a lattice $\varGamma \subset \mathbb{R}^d$ is given by an $m \times m$-matrix $\varPhi = (\varPhi_{ij})$, where each $\varPhi_{ij}$ is a finite set (possibly empty) of mappings $\varGamma \to \varGamma$. Here, the mappings of $\varPhi$ are affine linear mappings, where the linear part has the form $x \mapsto Q\,x$ and is the *same* for all maps. We call Q the *inflation factor*. It is required to have all eigenvalues exceeding 1 in absolute value. Any MFS $\varPhi$ induces a mapping or *substitution* on $P(\varGamma)^m$, where $P(\varGamma)$ denotes the set of subsets of $\varGamma$:

$$\varPhi \begin{pmatrix} U_1 \\ \vdots \\ U_m \end{pmatrix} = \begin{pmatrix} \bigcup_{j=1}^{m} \bigcup_{f \in \varPhi_{1j}} f(U_j) \\ \vdots \\ \bigcup_{j=1}^{m} \bigcup_{f \in \varPhi_{mj}} f(U_j) \end{pmatrix} \tag{7}$$

We say that $\boldsymbol{U} = (U_1, \ldots, U_m)^t$ is a *fixed point* of $\varPhi$ if $\boldsymbol{U} = \varPhi \boldsymbol{U}$. Furthermore, we call $(\boldsymbol{U}, \varPhi)$ a *lattice substitution system* on $\varGamma$ if $\varPhi$ is an MFS on $\varGamma$, $\boldsymbol{U}$ is a fixed point of $\varPhi$, the U_i's are pairwise disjoint and all the unions in Eq. 7 are disjoint. A lattice substitution system is *primitive* if its corresponding *substitution matrix* $M = (\#\varPhi_{ij})_{1 \le i,j \le m}$ is primitive, i.e., if there is a $k \in \mathbb{N}$ such that M^k has positive entries only.

Let $\boldsymbol{U}$ be a lattice substitution system for the MFS $\varPhi$. We say that a $\boldsymbol{U}$-cluster $\boldsymbol{C} = \boldsymbol{U} \cap B_R(s) = (U_i \cap B_R(s))_{i \le m}$, defined by intersecting all the components of $\boldsymbol{U}$ with a

[15] A *multiset* in $\mathbb{R}^d$ is a subset $U_1 \times \cdots \times U_m \subset \mathbb{R}^d \times \cdots \times \mathbb{R}^d$ (m copies), where $U_i \subset \mathbb{R}^d$. We also write $\boldsymbol{U} = (U_1, \ldots, U_m)^t = (U_i)_{i \le m}$. We say that $(U_i)_{i \le m}$ is a *Delone multiset* in $\mathbb{R}^d$ if each U_i is Delone and the union $\bigcup_{i=1}^m U_i \subset \mathbb{R}^d$ is also Delone. It is convenient to think of a multiset as a set with types of colours (types of atoms), i being the colour of points in U_i.

ball of radius R around a point $s \in \mathbb{R}^d$, is *legal* if it lies in $\Phi^n(\boldsymbol{u})$ for some point $\boldsymbol{u}$ of $\boldsymbol{U}$. Furthermore, we define the symmetric difference of two multisets as symmetric difference of their corresponding components, i.e., $\boldsymbol{U} \triangle \boldsymbol{V} = (U_i \triangle V_i)_{i \le m} = ((U_i \setminus V_i) \cup (V_i \setminus U_i))_{i \le m}$. We also use the notation $S + T = \{x + y \mid x \in S,\ y \in T\}$ and $\boldsymbol{U} + S = (U_i + S)_{i \le m} = (\{x + y \mid x \in U_i,\ y \in S\})_{i \le m}$ for sets S, T and a multiset $\boldsymbol{U}$.

Theorem 6 [17] *Let $\boldsymbol{U}$ be a primitive lattice substitution system with expansive map Q for the lattice $\Gamma = \bigcup_{i \le m} U_i$ in $\mathbb{R}^d$ and suppose that every $\boldsymbol{U}$-cluster is legal. Let $\Gamma' = \Gamma_1 + \dots + \Gamma_m$, where $\Gamma_i = \langle \Lambda_i - \Lambda_i \rangle_{\mathbb{Z}}$. The following assertions are equivalent.*

(i) *$\boldsymbol{U}$ has pure point diffraction spectrum (meaning that each U_i has this property);*

(ii) *$\boldsymbol{U}$ has pure point dynamical spectrum (meaning that each U_i has this property);*

(iii) $\mathrm{dens}(\boldsymbol{U} \triangle (Q^n \alpha + \boldsymbol{U})) \overset{n\to\infty}{\longrightarrow} \boldsymbol{0} = (0)_{i \le m}$ *for all $\alpha \in \Gamma'$;*

(iv) *A modular coincidence relative to $Q^M \Gamma'$ occurs in Φ^M for some M;*

(v) *Each U_i is a regular model set, $i \le m$, for the cut and project scheme*

$$
\begin{array}{ccccc}
\mathbb{R}^d & \longleftarrow & \mathbb{R}^d \times \overline{\Gamma} & \longrightarrow & \overline{\Gamma} \\
 & & \cup & & \\
\Gamma & \longleftarrow & \tilde{\Gamma} & &
\end{array}
$$

□

Here, the *pure point dynamical spectrum* is defined as follows: For a dynamical system (X, T) (see Eq. 6) that has a (unique) invariant probability measure μ associated to it (which is the case for dynamical systems which arise from primitive substitutions), we have the Hilbert space $L^2(X, \mu)$ and the unitary operator B: $L^2(X, \mu) \to L^2(X, \mu),\ f \mapsto f \circ T$. If the eigenfunctions of B span $L^2(X, \mu)$, then we have a pure point dynamical spectrum. Note that the equivalence of (i) and (ii) can also be established in a more general setting, see [19].

The model set in this theorem is with respect to a very particular cut and project scheme. The key point is the internal group $\overline{\Gamma}$ which we now briefly explain. $\overline{\Gamma}$ is the Q-adic completion (in terms of a *profinite group*, see [20, 21])

$$\overline{\Gamma} = (\overline{\Gamma})_Q = \lim_{\leftarrow k} \Gamma / Q^k \Gamma' = \lim_{\leftarrow k} (\Gamma / \Gamma' \leftarrow \Gamma / Q\Gamma' \leftarrow \ldots \leftarrow \Gamma / Q^k \Gamma' \leftarrow \ldots)$$

of Γ, supplied with the usual topology of a profinite group (which makes it compact). We note that Γ embeds naturally into $\overline{\Gamma}$. Then, $\tilde{\Gamma}$ is the group $\{(t, t) \in \mathbb{R}^d \times \overline{\Gamma} \mid t \in \Gamma\}$.

This gives a satisfactory approach to those systems which *are* model sets, including an algorithm to test it. The latter, however, is of limited value to disprove the model set property, unless one can see that no coincidence can ever occur. For this situation, Frettlöh has recently proved several sufficient criteria. Although they are not exhaustive, they are easy to check and seem to cover many cases of relevance, see [22] for details.

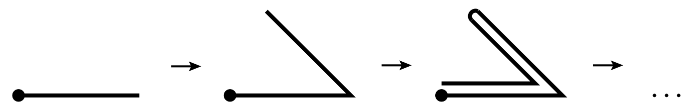
5 Paperfolding sequence as model set

To demonstrate the usefulness of the general setting, let us consider an explicit example with a p-adic internal space.

The so-called regular paperfolding sequence[16] starts as

$$11011001110010011101100011001001\ldots \tag{8}$$

and can be obtained by folding a sheet of paper repeatedly to the left, see [23]:



The sequence in Eq. 8 is obtained by unfolding the (infinite) stack and encoding a left (right) bend by 1 (0).

An alternative description employs two steps: The first determines the unique one-sided fixed point of the primitive 4-letter substitution of constant length[17]:

$$\sigma : \begin{array}{ccc} a & \to & ab \\ b & \to & cb \\ c & \to & ad \\ d & \to & cd, \end{array}$$

which starts as $abcbadcbabcdadcb\ldots$. The following second step maps a and b to 1 and c and d to 0, giving the sequence in Eq. 8.

Here, we change the point of view slightly, in that we consider two-sided (or bi-infinite) fixed points of σ, of which there are precisely two[18]:

$$\begin{array}{ccccccccc} b|a & \to & cb|ab & \to & adcb|abcb & \to & \ldots & \to & w_1 \\ d|a & \to & cd|ab & \to & adcd|abcb & \to & \ldots & \to & w_2 \end{array}$$

where $|$ denotes the seamline. Note that w_1 and w_2 differ only in the first position to the left of the seamline, otherwise they are equal.

Let us represent the letters with intervals of equal length 1, and points on their left end, of type a, b, c and d. Let Ω_a denote the a-points, etc. Then, the substitution together with the

[16] More generally, we can define a paperfolding sequence recursively: the sequence $\{a_1, a_2, \ldots\}$ is called a paperfolding sequence iff $a_1 = -a_3 = a_5 = -a_7 = \ldots$, and the remaining sequence $\{a_2, a_4, \ldots\}$ is a paper folding sequence. We obtain the regular paperfolding sequence for $a_n \in \{1, -1\}$ and writing 0 for -1.

[17] This is a primitive substitution of height 1 which has a coincidence (after two substitutions). Therefore it is pure point diffractive by a criterion of Dekking, see [18].

[18] The dynamical zeta function can be calculated with the method of Anderson and Putnam [24], and gives

$$\zeta(z) = \frac{1}{1 - 2z}.$$

From here, one sees that the number of fixed points of σ^m ($m \geq 1$) is 2^m, which shows that w_1 and w_2 are the only solutions of $\sigma(w) = w$ with w a bi-infinite word.

fixed point equation $\sigma(w) = w$ lead to the following system of equations:

$$\begin{array}{lcl} \Omega_a &=& 2\,\Omega_a \quad\cup\quad 2\,\Omega_c \\ \Omega_b &=& (2\,\Omega_a+1) \quad\cup\quad (2\,\Omega_b+1) \\ \Omega_c &=& 2\,\Omega_b \quad\cup\quad 2\,\Omega_d \\ \Omega_d &=& (2\,\Omega_c+1) \quad\cup\quad (2\,\Omega_d+1) \end{array}$$

where also $\Omega_a \cup \Omega_b \cup \Omega_c \cup \Omega_d = \mathbb{Z}$ by construction. With this, one quickly checks that the first and the third equation lead to the unique solution

$$\Omega_a = 4\,\mathbb{Z}, \qquad \Omega_c = 4\,\mathbb{Z} + 2$$

which reduces the other equations to

$$\begin{array}{lcl} \Omega_b &=& (8\,\mathbb{Z}+1) \quad\cup\quad (2\,\Omega_b+1) \\ \Omega_d &=& (8\,\mathbb{Z}+5) \quad\cup\quad (2\,\Omega_d+1). \end{array}$$

Since Ω_b and Ω_d are subsets of $\mathbb{Z}$, the general solution is

$$\begin{array}{lcl} \Omega_b &=& \bigcup\limits_{m\geq 1} 2^{m+2}\,\mathbb{Z} + 2^m - 1 \\ \Omega_d &=& \bigcup\limits_{m\geq 1} 2^{m+2}\,\mathbb{Z} + 3\cdot 2^m - 1, \end{array}$$

where the only remaining freedom consists in adding the singleton set $\{-1\}$ to either of them. This reflects the difference between the two fixed points, w_1 ($\{-1\}$ goes to Ω_b) and w_2 ($\{-1\}$ goes to Ω_d).

If one now follows the construction of a canonical cut and project scheme as derived in [2], one finds that the autocorrelation topology is the 2-adic topology, and this completes $\mathbb{Z}$ (the set of differences) to $\overline{\mathbb{Z}}_2$, the compact group of 2-adic integers. So we have the cut and project scheme

$$\begin{array}{ccccc} \mathbb{R} & \longleftarrow & \mathbb{R}\times\overline{\mathbb{Z}}_2 & \longrightarrow & \overline{\mathbb{Z}}_2 \\ & & \cup & & \\ & & \tilde{L} & & \end{array}$$

where $\tilde{L} = \{(m,m) \mid m \in \mathbb{Z}\}$ is a lattice. Our points are now model sets in this scheme. Defining the windows (as subsets of $\overline{\mathbb{Z}}_2$)

$$W_a = \overline{\Omega_a}, \quad W_b = \overline{\Omega_b}, \quad W_c = \overline{\Omega_c}, \quad W_d = \overline{\Omega_d},$$

one finds $W_a \cap W_b = \{-1\}$ and thus

$$\Omega_a = \Lambda(W_a), \quad \Omega_b = \Lambda(W_b), \quad \Omega_c = \Lambda(W_c), \quad \Omega_d = \Lambda(W_d \setminus \{-1\})$$

for w_1, while $\{-1\}$ moves from Ω_b to Ω_d for w_2.

As a regular model set, the paperfolding sequence is pure point diffractive[19]. More precisely, if $\omega = A\cdot\delta_{\Omega_a} + B\cdot\delta_{\Omega_b} + C\cdot\delta_{\Omega_c} + D\cdot\delta_{\Omega_d}$, the diffraction measure reads

$$\begin{aligned}\hat{\gamma}_\omega = &\left|\frac{A+B+C+D}{4}\right|^2 \delta_{\mathbb{Z}} \\ &+ \sum_{m \text{ odd}} \left[\left|\frac{A-B+C-D}{4}\right|^2 \delta_{\frac{m}{2}} + \left|\frac{A-C}{4}\right|^2 \delta_{\frac{m}{4}} + \sum_{r\geq 3}\left|\frac{B-D}{2^r}\right|^2 \delta_{\frac{m}{2^r}}\right]\end{aligned} \tag{9}$$

Finally, let us consider the binary reduction. One gets

$$\begin{aligned}\Omega_a \cup \Omega_b &= \bigcup_{m\geq 0} 2^{m+2}\,\mathbb{Z} + 2^m - 1 \\ \Omega_c \cup \Omega_d &= \bigcup_{m\geq 0} 2^{m+2}\,\mathbb{Z} + 3\cdot 2^m - 1\end{aligned}$$

plus $\{-1\}$ added to one of them. Clearly, these are again regular 2-adic model sets, and thus also pure point diffractive. To summarise:

Theorem 7 *The quaternary regular paperfolding sequences w_1 and w_2 are regular 2-adic model sets, with pure point diffraction spectrum as given in Eq.* 9*. Also, the binary reduction is a regular 2-adic model set, hence also pure point diffractive.* □

This structure is then inherited by the entire LI-class[20] of the paperfolding sequence. The members can be obtained via different folding sequences[21], see [23, 25] and references therein for details. Further examples along similar lines can be found in [2, 26, 27].

6 Systems with disorder

Here, we consider diffraction properties of stochastic point sets. Simple, well-understood examples are Bernoulli subsets of lattices or model sets [14, 28], and certain lattice gases, which can be analysed using elementary methods from stochastics. This approach has recently been generalised considerably [29] to cover stochastic selections from rather general Delone sets. The results prove the folklore claim that uncorrelated random removal of scatterers has two effects, namely reducing the overall intensity of the diffraction of the fully occupied set, without changing the relative intensities, and adding a white noise type constant diffuse background. The influence of disorder due to thermal fluctuations is discussed in [30].

[19] It is an example of a limit periodic system.

[20] Two structures Λ_1 and Λ_2 are *locally indistinguishable* (or *locally isomorphic* or *LI*) if each patch of Λ_1 (essentially, the intersection of Λ_1 with a compact set) is, up to translation, also a patch of Λ_2 and vice versa. The corresponding equivalence class is called *LI-class*.

[21] We get the paperfolding in Eq. 8 by setting $1 = a_1 = a_2 = a_4 = \ldots = a_{2^n} = \ldots$ (see Footnote 16), because we only fold in one direction. Using a different folding sequence, which corresponds to the a_{2^n} not being equal, we get a different paperfolding sequence, but this one is LI to the one in Eq. 8 and vice versa and therefore in the same LI-class. So, all such paperfolding sequences have the same diffraction spectrum, namely Eq. 9 in the binary reduction, i.e., $A = B$ and $C = D$.

A prominent class of stochastic point sets are random tilings [31, 32, 33]. Diffraction properties of these tilings are understood for systems without interaction, for one-dimensional Markov systems, and for product tilings [9, 34, 35]. Systems with interaction are generally difficult to analyse. Here, only few results are available for certain exactly solvable models from statistical mechanics with crystallographic symmetries, whose autocorrelation can be computed explicitly [9, 34, 35, 36]. Symmetries of a stochastic point set are understood to be symmetries on average. For a detailed discussion of this concept, see [32, 34].

Most examples of random tilings with quasicrystallographic symmetries are obtained from ideal quasicrystallographic tilings by relaxing the allowed local configurations. Since, as for ideal tilings, random tiling coordinates may be lifted into internal space via the $\star$-map, the random tiling ensemble possesses a so-called *height representation* which, in a way, can be understood as a description on the basis of a deviation from a model set. At present, there are no rigorous results concerning diffraction properties of random tilings with height representation in dimension $d \geq 2$. Henley [31] argues, using elasticity theory for the free energy of such an ensemble, that the discrete part of the diffraction spectrum only consists in the trivial Bragg peak at the origin in dimension $d \leq 2$, since the width of the distribution of scatterer positions in internal space diverges with the system size. In dimensions $d > 2$, the distribution width converges with the system size, implying a non-trivial discrete part in the diffraction spectrum. Even less is known about the nature of the continuous part in $d > 1$, though absolute continuity is expected. For a numerical investigation of diffraction properties of the randomized Ammann-Beenker tiling, see [35]. In what follows, we will focus on stochastic disorder in random tilings, mainly in the one-dimensional case, because the understanding of the higher dimensional situation is still rather incomplete.

The diffraction of 1D random tilings[22] has been investigated previously [34]. 1D binary random tilings have a non-trivial pure point part iff they have a rational interval length ratio $\alpha = u/v$.

Theorem 8 [34] *The natural density of Λ exists with probabilistic certainty and is given by $d = (pu + qv)^{-1}$. If $\omega = \delta_\Lambda = \sum_{x\in\Lambda} \delta_x$ denotes the corresponding stochastic Dirac comb, the autocorrelation γ_ω of ω also exists with probabilistic certainty and is a positive definite pure point measure. The diffraction spectrum consists, with probabilistic certainty, of a pure point (Bragg) part and an absolutely continuous part, so $\hat{\gamma}_\omega = (\hat{\gamma}_\omega)_{\mathrm{pp}} + (\hat{\gamma}_\omega)_{\mathrm{ac}}$.*

If $\alpha = u/v$, the pure point part is

$$(\hat{\gamma}_\omega)_{\mathrm{pp}} = d^2 \cdot \begin{cases} \delta_0 & \text{if } \alpha \notin \mathbb{Q} \\ \sum_{k\in(1/\xi)\mathbb{Z}} \delta_k & \text{if } \alpha \in \mathbb{Q} \end{cases}$$

where, if $\alpha \in \mathbb{Q}$, we set $\alpha = a/b$ with coprime $a, b \in \mathbb{Z}$ and define $\xi = u/a = v/b$. The absolutely continuous part $(\hat{\gamma}_\omega)_{\mathrm{ac}}$ can be represented by the continuous function

$$g(k) = \frac{d \cdot pq \, \sin^2(\pi k\,(u-v))}{p\,\sin^2(\pi k\, u) + q\,\sin^2(\pi k\, v) - pq\,\sin^2(\pi k\,(u-v))}$$

[22] A *(1D binary) random tiling* [31, 32] is a covering of the real line with two intervals of fixed lengths u and v without gaps or overlaps. Associated with each random tiling is the set Λ of left endpoint positions of its intervals. We call two random tilings *equivalent* if they are equal up to a translation. For each equivalence class, we choose a representative with $0 \in \Lambda$. The *random tiling ensemble* is the set of all non-equivalent random tilings.

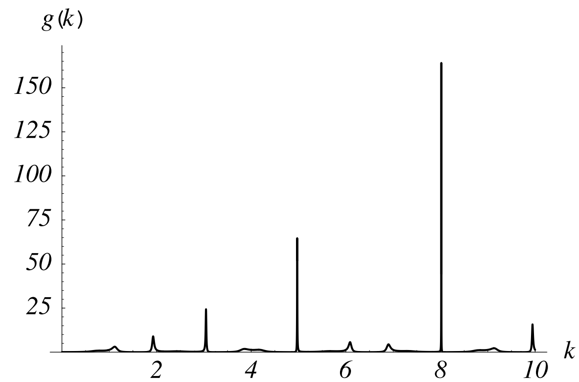


Figure 1: Absolutely continuous background of a Fibonacci random tiling.

which is well defined for $k\,(u-v) \notin \mathbb{Z}$. It has a smooth continuation to the excluded points. If α is irrational, this is $g(k) = 0$ for $k(u-v) \in \mathbb{Z}$ with $k \neq 0$ and

$$g(0) = \frac{d \cdot pq\,(u-v)^2}{p\,u^2 + q\,v^2 - pq\,(u-v)^2} = d\,\frac{pq\,(u-v)^2}{(p\,u + q\,v)^2}$$

For $\alpha = a/b \in \mathbb{Q}$ as above, it is $g(k) = 0$ for $k\,(u-v) \in \mathbb{Z}$, but $k\,u \notin \mathbb{Z}$ (or, equivalently, $k\,v \notin \mathbb{Z}$), and

$$g(k) = d\,\frac{pq\,(a-b)^2}{(p\,a + q\,b)^2}$$

for the case that also $k\,u \in \mathbb{Z}$. □

The most prominent 1D random tiling is the Fibonacci random tiling[23]. According to be above theorem, its diffraction spectrum consists with probabilistic certainty of a trivial Bragg peak at the origin and of an absolutely continuous background, see Fig. 1. The absolutely continuous background shows localised, bell-shaped needles of increasing height at sequences of points scaling with the golden ratio τ. This is reminiscent of the perfect Fibonacci tiling.

In dimensions $d \geq 2$, properties of the autocorrelation are known only for certain simple systems of statistical mechanics with crystallographic symmetries which can be interpreted in terms of dimer systems, see [34, 36, 37]. This includes the domino and the lozenge tiling, the Ising lattice gas, and others. Here, the asymptotic behaviour of the autocorrelation coefficients can be computed explicitly, leading to proofs of existence of an absolutely continuous part in addition to a pure point part.

[23]A *Fibonacci random tiling* is a random tiling with interval lengths $u = \tau = (1+\sqrt{5})/2$ and $v = 1$, with occupation probabilities $p = 1/\tau$ and $q = 1 - p = 1/\tau^2$ of the intervals (almost surely). Each interval endpoint of a representative of a Fibonacci random tiling belongs to the module $\mathbb{Z}[\tau] = \{m\tau + n \mid m, n \in \mathbb{Z}\}$. Every (ideal) Fibonacci tiling also appears as a Fibonacci random tiling.

Theorem 9 [34] *Away from the critical point, the diffraction spectrum of the Ising lattice gas almost surely exists, is $\mathbb{Z}^2$-periodic and consists of a pure point and an absolutely continuous part with continuous density. The pure point part reads*

1. $T > T_c : (\widehat{\gamma}_\omega)_{pp} = \frac{1}{4} \sum_{\boldsymbol{k} \in \mathbb{Z}^2} \delta_{\boldsymbol{k}}$
2. $T < T_c : (\widehat{\gamma}_\omega)_{pp} = \rho^2 \sum_{\boldsymbol{k} \in \mathbb{Z}^2} \delta_{\boldsymbol{k}},$

where the density ρ is the ensemble average of the number of scatterers per unit volume. At the critical point, the diffuse scattering diverges when approaching the lattice positions of the Bragg peaks.

We now analyse the effect of a cut and project setup on diffraction properties. For 1D random tilings, an embedding into $\mathbb{R}^2$ is given as follows. Each position $x \in \Lambda$ of an element of the random tiling ensemble may be written in the form $x = mu + nv$, where $m, n \in \mathbb{Z}$. If $\alpha = u/v \notin \mathbb{Q}$, the numbers m, n are uniquely determined. If $\alpha \in \mathbb{Q}$, uniqueness is achieved by parametrising $x = 0$ by $m = n = 0$ and incrementing (decrementing) m by addition of a u-interval to the right (left), and likewise with n for v-intervals. Identifying the unique coordinates m, n with points $(m, n) \in \mathbb{Z}^2$, we map a random tiling to a bi-infinite, directed walk on the edges of the square lattice. We call two walks equivalent if they are equal up to a translation. For each equivalence class of walks, we may choose a representative which passes through the origin in $\mathbb{Z}^2$. This establishes a one-to-one correspondence between non-equivalent random tilings and non-equivalent bi-infinite, directed walks.

Let us restrict to Fibonacci random tilings. Recall that the (ideal) Fibonacci tilings may be obtained within the cut and project setup by (orthogonal) projection of lattice points of a scaled copy of $\mathbb{Z}^2$ confined to a suitable strip onto the subspace with irrational slope $1/\tau$. This way, we obtain a one-to-one correspondence between (random) tiling coordinates in direct space and in internal space via the $\star$-map, given by $(m\tau + n)^\star = m\tau' + n$, where $\tau' = -1/\tau$ is the algebraic conjugate of τ. The value $x^\star$ of a tiling coordinate x is also called its *height*, and the above collection of direct and internal space together with the canonical projections is also called *height representation* of the Fibonacci random tiling ensemble.

In the following, we will consider the distribution of scatterer positions of Fibonacci random tilings in internal space. We restrict ourselves to patches of Fibonacci random tilings of M consecutive intervals on the positive half-axis, starting at $x = 0$. Following [35], we consider the occupation probability for the position of the rightmost interval. Since the random tiling patch is a Bernoulli system, the probability of the position being $x = m\tau + (M - m)$, or equivalently $x^\star = m\tau' + (M - m)$ in internal space, is given by

$$\tilde{\rho}(M, x^\star) = \binom{M}{m} p^m \, q^{M-m}.$$

According to the theorem of de Moivre-Laplace, the binomial distribution (with mean $\mu = Mp$ and variance $\sigma^2 = Mpq$) may be approximated by the Gaussian distribution for large fixed M, yielding

$$\rho(M, x^\star) = \sqrt{\frac{1}{\pi} \, \frac{\tau}{2M}} \, \exp\left[-\frac{\tau}{2M} \, x^{\star 2}\right].$$

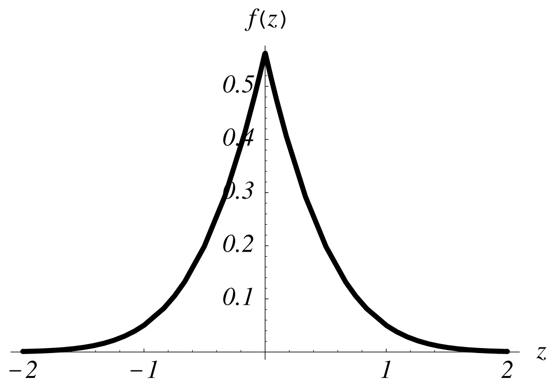


Figure 2: Distribution of scatterer positions in internal space for Fibonacci random tilings.

Note that in the limit $M \to \infty$, the admissible positions $x^\star$ lie dense in internal space. We fixed the normalisation such that the integral of the density over internal space equals unity. To obtain the distribution of all interval positions of random tiling patches of length N, we sum over all endpoint positions of patches with $n \leq N$ intervals and normalise[24],

$$\rho(x^\star) = \frac{1}{N} \sum_{n=1}^{N} \rho(n, x^\star) \simeq \frac{1}{N} \int_{n=0}^{N} \rho(n, x^\star) \, \mathrm{d}n. \tag{10}$$

In the second equation, we approximated the sum to leading order in N using the Euler-Maclaurin summation formula. This leads to the following result.

Theorem 10 *The distribution $\rho(x^\star)$ of scatterer positions of Fibonacci random tiling patches in internal space is, to leading order in the patch size N, given by*

$$\rho(x^\star) = \sqrt{\frac{\tau}{2N}}\, f\left(\sqrt{\frac{\tau}{2N}} \cdot x^\star\right), \qquad f(z) = 2\left(\frac{e^{-z^2}}{\sqrt{\pi}} - |z| \operatorname{erfc}\left(|z|\right)\right),$$

where $\operatorname{erfc}(x) = \frac{2}{\sqrt{\pi}} \int_x^\infty e^{-t^2} \mathrm{d}t$ denotes the complementary error function. □

The function $f(z)$ is shown in Fig. 2. Note that the width of the distribution grows with the system size as $\sqrt{N}$, but the distribution itself is not a Gaussian, as usually believed[25].

This result was derived for patches of N intervals, starting from the origin in positive direction. Since the result is symmetric in $x^\star$, it is also valid for tiling patches with N intervals, starting from the origin in negative direction, hence also for tiling patches with $2N$ intervals with N intervals on the negative and on the positive axis each.

[24] A more natural normalisation may be the density of points instead of unity, see below.

[25] For a comparison with numerical simulations of the Fibonacci random tiling ensemble and for the example of the two-dimensional Ammann-Beenker random tiling, see [35].

Within the cut and project scheme, the Dirac comb of a model set may be characterised as a sum of point scatterers over the projected lattice points

$$\omega = \sum_{x\in L} 1_{\Omega}(x^{\star})\, \delta_x ,$$

weighted by the characteristic function of the (compact) window in internal space. For diffraction of quasicrystallographic random tilings, the common approach [31] is to investigate properties of an averaged structure given by the Dirac comb weighted by the occurrence probability of each scatterer within the random tiling ensemble. The averaged distribution in internal space need not exist for the infinite tiling, as we showed in the previous section. It is generally believed [31] that it exists in dimensions $d > 2$, the case $d = 2$ being marginal with a logarithmic growth of the distribution width with the system size, leading to the statement that there is no non-trivial discrete component in the diffraction spectrum for $d \leq 2$ (for aperiodic systems).

As argued in Eq. 10, the averaged structure may be written in the form

$$\omega = \sum_{x\in L} \varphi(x^{\star})\, \delta_x , \tag{11}$$

where the support of φ is generally the whole internal space. Note that this object is generally ill-defined, since summation is over a dense set. In averaging, one loses information about correlations between scatterers, so that the analysis will at most yield information about the discrete part of the diffraction spectrum, but not about the continuous part, see the theorem below. We investigate under which conditions Eq. 11 defines a tempered distribution. To this end, we assume that φ vanishes sufficiently rapidly at infinity, which includes the Gaussian (and, in a certain sense, characteristic functions). Following [35], we consider the special situation of a Euclidean internal space $H = \mathbb{R}^m$ and assume that the canonical projections π and π_{int} are both dense and one-to-one. We denote by $\text{vol}(FD)$ the volume of a fundamental domain of the lattice $\tilde{L} \in \mathbb{R}^d \times H$ w.r.t. the product measure of the Lebesgue measures on $\mathbb{R}^d$ and on H. We denote the dual lattice of $\tilde{L}$ by $(\tilde{L})^* = \{\tilde{x} \in \mathbb{R}^d \times H \,|\, \langle \tilde{x}, \tilde{y}\rangle \in \mathbb{Z} \text{ for all } \tilde{y} \in \tilde{L}\}$, and its projection by $L^* = \pi((\tilde{L})^*)$.

Theorem 11 [38] *Assume $\varphi : \mathbb{R}^m \to \mathbb{C}$ continuous and $\lim_{y\to\infty} |y|^{m+1+\alpha}\, \varphi(y) = 0$ for some $\alpha > 0$. Then, the weighted Dirac comb in Eq.* 11 *is a translation bounded measure. It has the unique autocorrelation*

$$\gamma_\omega = \sum_{z\in L} \eta(z)\, \delta_z , \qquad \eta(z) = \frac{1}{\text{vol}(FD)} \int_{\mathbb{R}^m} \varphi(u)\, \overline{\varphi(u - z^{\star})}\, \mathrm{d}u,$$

being a translation bounded, positive definite pure point measure. Its Fourier transform is a positive pure point measure. If π and π_{int} are orthogonal projections, it is explicitly given by

$$\hat{\gamma}_\omega = \frac{1}{\text{vol}(FD)^2} \sum_{y\in L^*} |\hat{\varphi}(-y^{\star})|^2\, \delta_y ,$$

where $\hat{\varphi}$ denotes the Fourier transform of φ. □

A natural choice for the normalisation of the function φ arises form the observation that for Dirac combs satisfying the assumptions of the above theorem the density of points ρ exists,

$$\rho = \lim_{n\to\infty} \frac{1}{\mathrm{vol}(B_n)}\, \omega(B_n) = \frac{1}{\mathrm{vol}(FD)} \int_{\mathbb{R}^m} \varphi(u)\, \mathrm{d}u. \tag{12}$$

The above theorem sheds light onto the example of the Fibonacci random tiling. Here, the internal distribution may be described by a sequence of distributions of increasing width but constant mass. The limit of the corresponding sequence of measures will have a trivial discrete part and a continuous part, whose form may be compared to the above results.

7 Outlook

For our discussion of pure pointedness, we made substantial use of the uniform discreteness of $\varDelta^{\mathrm{ess}}$. This can certainly be relaxed, as Theorem 1 shows, but things become considerably more involved beyond this "barrier". This is also intimately related to stepping into the territory of mixed spectra, which seems particularly timely.

Pure pointedness of the diffraction is equivalent to strong almost periodicity of the autocorrelation. More generally, one can show that any autocorrelation γ_ω possesses a unique decomposition into a strongly almost periodic part and a weakly almost periodic part with zero volume average, compare [5]. So, we get

$$\gamma_\omega = (\gamma_\omega)_{\mathrm{sap}} + (\gamma_\omega)_{0\text{-}\mathrm{wap}}$$

where "weak" refers to the weak topology in relation to the strong (product) topology.

The Fourier transform of $(\gamma_\omega)_{\mathrm{sap}}$ is a pure point measure, while that of $(\gamma_\omega)_{0\text{-}\mathrm{wap}}$ is continuous [5], so that one has full control of this question on the level of the autocorrelation. Also, important issues of the diffraction of random tilings can be formulated and understood in this context, but most results are folklore, and still need to be proved. Furthermore, there is no such decomposition known that would allow a distinction of absolute versus singular continuity. It is highly desirable to improve this situation in the future.

Finally, even if all the spectral questions were settled, the big remaining question is how to characterise the homometry class, i.e., the class of measures with a given autocorrelation (and hence diffraction). This is part of the inverse problem, where results are rare at present. So, the question of the title should now be replaced by another one:

"Which distributions of matter are homometric?"

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