

Indexing the diffraction spectrum of a non-Pisot self-similar structure

C. Godrèche

Service de Physique de l'Etat Condensé, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette CEDEX, France.

J. M. Luck

Service de Physique Théorique, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette CEDEX, France

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We study the Fourier transform, or diffraction spectrum, of a self-similar structure in one dimension, generated by a substitution rule, which does not possess the Pisot property. Several consequences of this number-theoretical property are investigated. The structure exhibits unbounded density fluctuations with respect to its average lattice, which diverge as a power of the system size. The diffraction spectrum consists neither of Bragg peaks, nor of diffuse scattering; it is a singular continuous and multifractal measure. The Fourier transform exhibits *peaks* of singular scattering and is shown to possess scaling properties—in a weak sense—around those values of the wave vector. These local properties are analyzed in detail and put in perspective with open questions concerning the chaotic behavior of a generalized Arnold-Sinai cat map. We argue that the *peaks* of the Fourier intensity cannot be indexed by any simple scheme, such as a finite number of integers. In that respect, the present structure is therefore different from periodic or quasiperiodic structures and from most singular continuous cases studied previously.

I. INTRODUCTION

The aim of this paper is to present a descriptive investigation of the Fourier spectrum of a one-dimensional self-similar atomic structure generated by a substitution rule, in a generic case where the substitution does not possess the Pisot property, to be defined below. This work is motivated by our previous studies¹⁻⁵ of the diffraction spectra of various deterministic distributions of matter, with emphasis on the relationship between the nature of their Fourier spectra and the types of order present in those model structures.

Let us begin with a few definitions. Consider the substitution σ acting on an alphabet of two letters A and B according to the rules

$$\sigma: \begin{cases} A \rightarrow AAAB \\ B \rightarrow BBA \end{cases} \quad (1.1)$$

To this substitution we associate a matrix \underline{M} , whose columns give the numbers of letters A and B which occur in the transforms $\sigma(A)$ and $\sigma(B)$. This matrix reads therefore

$$\underline{M} = \begin{pmatrix} 3 & 1 \\ 1 & 2 \end{pmatrix}. \quad (1.2)$$

The characteristic polynomial of the matrix \underline{M} is $P(\lambda) = \det(\lambda \underline{1} - \underline{M}) = \lambda^2 - 5\lambda + 5$. The eigenvalues of \underline{M} are the roots of $P(\lambda)$, namely,

$$\begin{aligned} \lambda_1 &= 2 + \tau = \tau\sqrt{5} = 4 \sin^2(2\pi/5) \approx 3.61803, \\ \lambda_2 &= 3 - \tau = \sqrt{5}/\tau = 4 \sin^2(\pi/5) \approx 1.38197, \end{aligned} \quad (1.3)$$

where $\tau = (1 + \sqrt{5})/2$ denotes the golden mean.

Since we have $\lambda_1 > \lambda_2 > 1$, the substitution σ does not possess the Pisot⁶ property. This statement means the following. Consider an arbitrary substitution σ acting on p letters. The associated matrix \underline{M} has therefore p eigenvalues. The Perron-Frobenius theorem states that the largest eigenvalue λ_1 is real, positive, and larger than unity (see, e.g., Ref. 7). By definition, the substitution σ is said to have the Pisot-Vijayaraghavan (called PV or Pisot for short in the following) property if all other eigenvalues $\lambda_2, \dots, \lambda_p$ are smaller than unity in modulus. Let us assume furthermore that the characteristic polynomial $P(\lambda)$ is irreducible over the integers, i.e., cannot be written as the product of two polynomials of lower degrees with integer coefficients. Saying that the substitution σ has the Pisot property amounts then to saying that the largest eigenvalue λ_1 is a Pisot number. By definition, a Pisot number is an algebraic integer, namely, a solution of an irreducible polynomial equation of the form $P(\lambda) = \lambda^p + a_{p-1}\lambda^{p-1} + \dots + a_0 = 0$ (a_n being ordinary integers) such that λ_1 is real, larger than 1, and that all its algebraic conjugates, namely, the other solutions of $P(\lambda) = 0$, are smaller than 1 in modulus. For instance, τ is a Pisot number, whereas $2 + \tau$ is not.

Self-similar sequences and structures generated by substitution rules are not only fascinating mathematical objects. They are also naturally met in modeling actual structures. The most celebrated case is the Fibonacci chain, encountered in the study of quasicrystals. The associated substitution, recalled in Eq. (2.7), has the Pisot property. Indeed, the largest eigenvalue of the corresponding matrix \underline{M}_F is equal to τ , its conjugate being $-\tau^{-1}$. It is well known that the Fibonacci chain, obtained by associating long and short interatomic bonds to the letters A and B , is quasiperiodic. The usual way of realizing this property consists in making use of a geome-

trical construction, such as the cut and project method. It is also possible to extract information on the Fourier spectrum of the structure from the knowledge of the substitution matrix M_F alone. The connection between the arithmetical nature of a substitution and the presence of Bragg peaks in the corresponding Fourier transform has been pointed out by Bombieri and Taylor.⁸ The central point in this connection is a theorem due to Pisot,⁶ which states the following. If

$$x \theta^n \rightarrow 0 \pmod{1}, \quad (1.4)$$

for some real numbers x and θ , with θ larger than unity, then θ is a Pisot number. Furthermore, x has to belong to some \mathbb{Z} -module, i.e., to be a linear combination, with integer coefficients, of some elementary “frequencies” $\{\omega_k\}$, which are related to θ in a known fashion. From this theorem it is rather easy to deduce the Bombieri-Taylor criterion, namely, that the Fourier spectrum of a self-similar structure has generically an atomic component (Bragg peaks) if and only if the associated substitution matrix has the Pisot property.

In this paper we address a much more difficult problem, namely, that of describing the Fourier spectrum of an atomic structure generated by a non-Pisot substitution. For short, we have named such a structure a non-Pisot structure.³⁻⁵ We have already studied in full detail a non-Pisot structure, the circle structure,¹⁻³ and shown, in a convincing, albeit nonrigorous way, that its Fourier spectrum is purely singular continuous. This example is nevertheless special, inasmuch as the second largest eigenvalue of the substitution matrix is equal to -1 . The circle sequence is thus a marginal case, at the borderline between Pisot and non-Pisot structures. Another example deserves to be mentioned, namely, the Thue-Morse sequence, which is exceptional, since it corresponds to a Pisot case in the present classification, but exhibits a purely singular continuous Fourier intensity.⁹

The structure under consideration in the present paper, already defined in Eq. (1.1), is more generically non-Pisot than the examples recalled just above, since both eigenvalues given in Eq. (1.3) are strictly larger than unity. Let us stress at once the origin of the difficulty encountered in this problem: It is rooted in the state of ignorance on the mathematical question of how the sequence $\{x \theta^n \pmod{1}\}$ is distributed on the unit circle, for particular values of x , when θ is a non-Pisot algebraic number.

To conclude this Introduction, let us express our motivations for the choice of the substitution given in Eq. (1.1).

First, if one lists *all* the irreducible binary substitutions (acting on two letters), one hits upon that studied here if (i) one restricts consideration to non-Pisot cases (i.e., $\lambda_1 > |\lambda_2| > 1$), (ii) one requires a positive second eigenvalue λ_2 , and (iii) one requires the substitution to be minimal, i.e., the sum of the numbers of letters contained in the transforms of A and B , after one substitution step, to be minimal. In the present example, this sum is equal to 7.

If one relaxes condition (ii), one finds the following sub-

stitution:

$$\sigma: \begin{cases} A \rightarrow AB \\ B \rightarrow A A A \end{cases}, \quad (1.5)$$

with a sum of letters equal to 5. The associated eigenvalues read $\lambda_{1,2} = (1 \pm \sqrt{13})/2 \approx (2.30278, -1.30278)$. This example has been considered in Ref. 4.

Second, the structure generated by the substitution (1.1) is a one-dimensional analog of a fivefold non-Pisot tiling of the plane, remarkable for its simplicity. This tiling has been introduced by Lançon and Billard by iterating a decoration rule described in Ref. 10 (see also Refs. 5 and 11).

This paper is organized as follows. In Sec. II we define the structural model and study its properties in real space. We show in particular that the presence of a second eigenvalue larger than unity induces strongly diverging fluctuations of the structure in perpendicular space. In Sec. III we recall the relevant definitions concerning Fourier transforms and especially their measure-theoretical aspects. We consider the associated Hendricks-Teller model as an illustration of a random structure which exhibits “peaks” in reciprocal space. We then describe our numerical findings concerning various aspects of the Fourier spectrum of the structure under study, including the multifractal formalism, and a tentative local scaling analysis. Section IV presents a discussion, in which we question the possibility of indexing the peaks in reciprocal space, for a generic non-Pisot self-similar structural model.

The present study is mostly descriptive and observational. It is intended both for physicists as a warning of the possible intricacy of the diffraction spectra of simple structures which could be found in nature and for mathematicians as a concrete illustration of questions pertaining to open number-theoretical problems. We do not pretend to have solved such questions; we are nevertheless convinced of the interest in underlining both their physical context and their difficulty.

II. PROPERTIES OF THE STRUCTURE IN REAL SPACE

A. Definition of the structure

We consider the substitution σ which has been introduced in Eq. (1.1) and define the finite words $A_n = \sigma^n(A)$, $B_n = \sigma^n(B)$, obtained by acting repeatedly with the substitution σ on the letters A and B . These words obey the recursion formulas

$$A_{n+1} = A_n A_n A_n B_n, \quad (2.1a)$$

$$B_{n+1} = B_n B_n A_n. \quad (2.1b)$$

The words A_n converge to an infinite sequence $\sigma^\infty(A) = A A A B A A A B \dots$, which is left invariant by σ and is thus self-similar. The same would hold true for the words B_n . In the following, by the “infinite sequence” we always mean $\sigma^\infty(A)$.

The components of the normalized right eigenvector v

associated with the largest eigenvalue λ_1 , namely,

$$\mathbf{v} = \begin{pmatrix} \tau^{-1} \\ \tau^{-2} \end{pmatrix}, \quad (2.2)$$

give the frequencies of letters of each kind in the infinite sequence. In particular, the positivity of these weights is ensured by the Perron-Frobenius theorem.

To this abstract sequence we associate a structural model by replacing the letters A and B by long and short bonds of respective lengths l^A and l^B , which we place successively on a line, starting from an origin O . The choice of both bond lengths is *a priori* arbitrary. We have been led to make a particular choice according to the following argument. Let us define l_n^A (l_n^B) as the length of the finite structure associated with the word A_n (B_n). These lengths obey the linear recursion formulas

$$\begin{pmatrix} l_{n+1}^A \\ l_{n+1}^B \end{pmatrix} = \underline{M} \begin{pmatrix} l_n^A \\ l_n^B \end{pmatrix}, \quad (2.3)$$

which is a mere consequence of Eq. (2.1) and where the substitution matrix \underline{M} has been defined in Eq. (1.2). Thus, if we define x_n as the ratio l_n^A/l_n^B , the substitution induces a map T_σ on the x_n , namely,

$$x_{n+1} = T_\sigma(x_n) = \frac{3x_n + 1}{x_n + 2}. \quad (2.4)$$

When the generation label (number of substitution steps) n increases indefinitely, the ratios x_n converge toward the attractive fixed point of the map T_σ , which is $x = \tau$. We utilize beforehand this special value for the ratio of bond lengths and choose length units such that $l_0^A = l^A = \tau$ and $l_0^B = l^B = 1$.

The freedom of choosing the ratio of bond lengths is specific to one-dimensional structures. In the case of higher dimensions, namely, for tilings, the geometrical constraint of space filling determines the ratios of the areas of the different tiles.

The word lengths defined above can then be expressed as

$$l_n^A = \tau \lambda_1^n = (a_n + b_n)\tau + a_n, \quad l_n^B = \lambda_1^n = a_n\tau + b_n, \quad (2.5)$$

where a_n and b_n are integers, which obey the same recursion relations (2.3) as the word lengths themselves, with initial conditions $a_0 = 0$ and $b_0 = 1$. These integers have the following simple interpretation in terms of letter counting: The word A_n consists of $v_n^A = 2a_n + b_n$ letters, among which $(a_n + b_n)$ are A 's and a_n are B 's, whereas the word B_n consists of $v_n^B = a_n + b_n$ letters, among which a_n are A 's and b_n are B 's. The recursion relation (2.3) can be solved in closed form, allowing one to express the integers a_n, b_n in terms of the Fibonacci numbers F_p , defined by $F_p = F_{p-1} + F_{p-2}$, for $p \geq 2$, with $F_0 = 0$ and $F_1 = 1$. We have indeed

$$a_{2p} = 5^p F_{2p}, \quad a_{2p+1} = 5^p (F_{2p} + F_{2p+2}); \quad (2.6a)$$

$$b_{2p} = 5^p F_{2p-1}, \quad b_{2p+1} = 5^p (F_{2p-1} + F_{2p+1}). \quad (2.6b)$$

The presence of powers of 5 in these expressions origi-

nates in the fact that the matrix \underline{M} of the substitution σ and the matrix \underline{M}_F of the Fibonacci substitution

$$\sigma_F: \begin{cases} A \rightarrow AB \\ B \rightarrow A \end{cases} \quad (2.7)$$

are related by the identity $\underline{M}^2 = 5\underline{M}_F^2$. The integers a_n, b_n can also be expressed directly in terms of the eigenvalues of the matrix \underline{M} , namely,

$$a_n = (\lambda_1^n - \lambda_2^n) / \sqrt{5}, \quad b_n = \lambda_1^{n-1} + \lambda_2^{n-1}. \quad (2.8)$$

As a consequence of the above properties, the mean bond length (or average interatomic distance) a of the structural model reads

$$a = \lim_{n \rightarrow \infty} \frac{l_n^A}{v_n^A} = \lim_{n \rightarrow \infty} \frac{l_n^B}{v_n^B} = \tau^{-1} l^A + \tau^{-2} l^B = \lambda_2. \quad (2.9)$$

This expression may alternatively be deduced from Eq. (2.2).

B. Unbounded fluctuations

The non-Pisot nature of the substitution σ , i.e., the presence of a second eigenvalue larger than unity, has far-reaching consequences. One of them concerns the unboundedness of the fluctuations of the structure with respect to its average lattice.^{1,2}

The abscissa of the k th atom of the infinite structure $\sigma^\infty(A)$ reads

$$u(k) = v_1(k)\tau + v_2(k), \quad (2.10)$$

where $v_1(k)$ [$v_2(k) \equiv k - v_1(k)$] is the number of long (short) bonds among the first k ones.

Let us define the density fluctuation $\delta(k)$ as

$$\delta(k) = u(k) - ka. \quad (2.11)$$

This difference represents the fluctuation of the position of the k th atom around the average lattice of periodicity a . It has been shown rigorously in Ref. 12 that this fluctuation exhibits a strong power-law divergence with the distance k of the form

$$\delta(k) \approx k^\beta F(k), \quad (2.12)$$

where the fluctuation (or wandering) exponent β reads

$$\beta = \frac{\ln \lambda_2}{\ln \lambda_1} \approx 0.25157. \quad (2.13)$$

The amplitude F is a continuous but nowhere differentiable function, which is self-affine; i.e., one has

$$F(\lambda_1 x) = F(x). \quad (2.14)$$

Equivalently, $\delta(k)$ is asymptotically a self-similar function of the atomic label k of the form

$$\delta(k) \approx k^\beta G \left[\frac{\ln k}{\ln \lambda_1} \right]. \quad (2.15)$$

The amplitude $G(y)$ which enters the scaling law (2.15) is a periodic fractal function of its argument $y = \ln k / \ln \lambda_1$, with period unity.

Figure 1 shows the fluctuation $\delta(k)$ plotted against the atomic label k for the structure corresponding to the word A_6 , which is made of 2625 atoms.

The scaling behavior [Eqs. (2.12) and (2.15)] of the fluctuation $\delta(k)$ can be derived by the following heuristic argument. If we restrict the analysis to the words A_n and B_n , we have to consider the differences $\delta_n^A = l_n^A - v_n^A a$ and $\delta_n^B = l_n^B - v_n^B a$. The above results, especially (2.8), allow us to evaluate these quantities explicitly. We thus obtain

$$\delta_n^A = \tau^{-3} \lambda_2^n, \quad \delta_n^B = -\tau^{-2} \lambda_2^n. \quad (2.16)$$

Roughly speaking, we have the estimate $\delta(k) \sim \lambda_2^n$, with a number of atoms $k \sim \lambda_1^n$, whence the expression (2.13) of the wandering exponent β .

Let us now turn to the interpretation of the fluctuation $\delta(k)$ in the ‘‘hyperspace’’ representation of the one-dimensional structure considered so far. To do so, we associate with the letter A the vector \mathbf{e}_1 and with B the vector \mathbf{e}_2 , where $\{\mathbf{e}_1, \mathbf{e}_2\}$ form an orthonormal basis of unit vectors in the two-dimensional Euclidean plane. The structure is thus lifted up in the plane, and the sequence of long and short bonds is represented by an infinite staircase-shaped broken line in this plane. To the k th atom of the infinite structure corresponds thus an integer vector $\mathbf{V}(k)$, with the convention that $\mathbf{V}(0) = \mathbf{0}$. The end point of this vector fluctuates (wanders) around an average direction given by the vector \mathbf{v} , already defined in Eq. (2.2), which spans the ‘‘physical’’ space E_{\parallel} . One has indeed

$$\mathbf{V}(k) = v_1(k) \mathbf{e}_1 + v_2(k) \mathbf{e}_2 = k \mathbf{v} + (\mathbf{e}_1 - \mathbf{e}_2) \tau \delta(k), \quad (2.17)$$

where $v_1(k)$ and $v_2(k)$ have been introduced in Eq. (2.10).

The orthogonal projections of $\mathbf{V}(k)$ onto E_{\parallel} and E_{\perp} are proportional to the abscissa $u(k)$ and fluctuation $\delta(k)$,

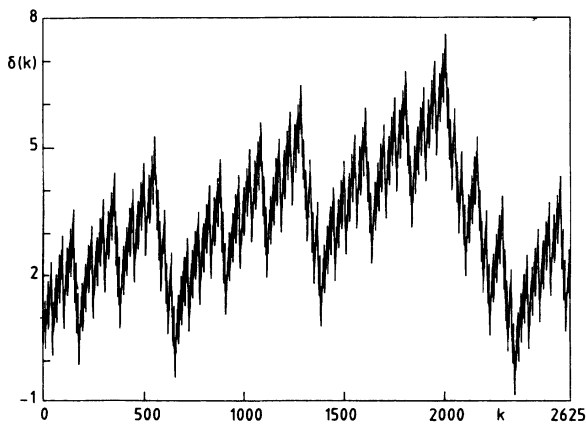


FIG. 1. Plot of the fluctuation $\delta(k)$ of the atomic abscissas of the structure with respect to its average lattice, against the atomic label k . The data correspond to the word A_6 ($v_6^A = 2625$ atoms).

respectively. The above results imply indeed with obvious notations,

$$V_{\parallel}(k) = \lambda_1^{-1/2} u(k), \quad V_{\perp}(k) = -\tau^2 \lambda_1^{-1/2} \delta(k). \quad (2.18)$$

The wandering of $\mathbf{V}(k)$ around its average direction obeys therefore the scaling law [Eqs. (2.12) and (2.15)]. In other words, the structure under consideration exhibits an unbounded fluctuation in E_{\perp} , which diverges as a power of its extension in physical space. This effect is a consequence of the non-Pisot character of the structure, which also explains the absence of Bragg peaks in its Fourier transform.

III. FOURIER SPECTRUM

A. General definitions

The diffraction spectrum of the structure defined above is the Fourier transform of some mass distribution living on the structure. This distribution is fully defined by associating a distribution of mass with each species of bonds. Let us choose for simplicity to put pointlike atoms at the end points of the bonds. The mass distribution on the structure reads then

$$\rho(x) = \sum_k \delta(x - u(k)), \quad (3.1)$$

where $u(k)$ is the abscissa of the k th atom of the infinite structure. Let us denote by $G_n^A(q)$ and $G_n^B(q)$ the Fourier amplitudes of the finite structures associated with the words A_n and B_n , respectively:

$$G_n^A(q) = \sum_{A_n} e^{-iqu(k)}, \quad G_n^B(q) = \sum_{B_n} e^{-iqu(k)}. \quad (3.2)$$

The corresponding structure factors, or intensities, are defined as

$$S_n^A(q) = \frac{1}{v_n^A} |G_n^A(q)|^2, \quad S_n^B(q) = \frac{1}{v_n^B} |G_n^B(q)|^2. \quad (3.3)$$

It will turn out that the intensities $S_n^A(q)$ and $S_n^B(q)$ do not exhibit simple convergence or scaling properties, as the generation index n becomes large, for any fixed value of the wave vector q . We will come back to this irregular behavior at length in the following.

In any case, from a rigorous viewpoint, the only well-defined concept attached to the Fourier spectrum of an infinite structure is its intensity measure or spectral measure. The distribution function of this measure, also called the integrated intensity, is defined as

$$H(q) = \lim_{n \rightarrow \infty} \int_0^q S_n^A(q') dq' = \lim_{n \rightarrow \infty} \int_0^q S_n^B(q') dq'. \quad (3.4)$$

This quantity is expected to be well behaved in any circumstance. In other words, the intensity, or structure factor, $S(q)$ of the infinite structure, defined formally by

$$dH(q) = S(q) dq, \quad (3.5)$$

has to be thought of as a generalized function or distribution. Furthermore, $dH(q)$ is best described within the framework of measure theory. Just as any positive mea-

sure, the intensity measure can be in general the superposition of an absolutely continuous (AC) component, a discrete component (Bragg peaks, i.e., δ functions), and a singular continuous (SC) component.

Without claiming too much mathematical rigor, we recall here that one may expect three possible kinds of *local* behavior of a diffraction spectrum, which are related to the measure-theoretic nature of the intensity measure. In the following general presentation, we use the notations $G_N(q)$ and $S_N(q)$ for the Fourier amplitude and intensity of a finite chain, using as a subscript the sample size N , i.e., the number of atoms (see Refs. 3–5).

1. Diffuse scattering

This situation corresponds to the structure factor $S(q)$ being a smooth function. This is, for instance, the case generically in amorphous structures, for which the Fourier amplitude $G_N(q)$ grows typically as $N^{1/2}$. The associated Fourier intensity measure is then AC.

2. Bragg peaks

These are values q_0 of q such that

$$G_N(q_0) \approx C(q_0)N, \quad (3.6)$$

$C(q_0)$ being some complex amplitude. $H(q)$ has then a discontinuity of strength $|C(q_0)|^2$ at $q=q_0$, and the structure factor $S(q)$ contains a δ function of the form $|C(q_0)|^2\delta(q-q_0)$. For periodic and quasiperiodic (almost-periodic) structures, the whole intensity is concentrated in Bragg peaks.

3. Singular scattering

Suppose that one has for some wave vector q_0 the power-law growth

$$G_N(q_0) \sim N^\gamma \quad (3.7)$$

with $\frac{1}{2} < \gamma < 1$. We have then $S_N(q_0) \sim N^{2\gamma-1}$. Let us assume furthermore that the intensity obeys the following scaling law for $q \rightarrow q_0$ and N large:

$$S_N(q) \approx N^{2\gamma-1} f(N(q-q_0)) \quad (3.8)$$

and that the scaling function $f(x)$ involved is regular enough. An integration then yields

$$|H(q_0+\epsilon) - H(q_0)| \sim |\epsilon|^\alpha, \quad (3.9)$$

with $\alpha = 2(1-\gamma)$. The local exponents α and γ depend on q_0 *a priori*. For $\frac{1}{2} < \gamma < 1$, we have $0 < \alpha < 1$. Thus the intensity $S(q_0)$, which is formally equal to the derivative of $H(q)$ at q_0 , is divergent, but “less infinite” than in the presence of a Bragg peak, which corresponds formally to the limiting case $\gamma = 1$, $\alpha = 0$.

It may also turn out that the Fourier amplitude $G_N(q)$ obeys no simple behavior at all, considered as a function of the sample size. We suspect that, in the case of SC Fourier transforms, generic values of the wave vector do fall into this last class. This statement is to be refined by

the statistical information provided by multifractal analysis, to be described later.

As a complement to the above discussion and especially in order to justify the scaling assumption (3.8) concerning the Fourier spectrum of finite structures, let us consider the simple example of a periodic structure with lattice parameter $a=1$. The structure factor of a system of N atoms reads

$$S_N(q) = \frac{1}{N} \left[\frac{\sin(Nq/2)}{\sin(q/2)} \right]^2. \quad (3.10)$$

The Fourier transform is made of Bragg peaks for the values $q_m = 2m\pi$ of the wave vector, where m is an arbitrary integer. In the neighborhood of each value q_m , we do observe a scaling law of the form (3.8), with $\gamma = 1$, namely,

$$S_N(q) \approx N f(N(q - q_m)),$$

with

$$f(x) = \left[\frac{\sin(x/2)}{x/2} \right]^2. \quad (3.11)$$

We note that the scaling function $f(x)$ is nothing but the normalized squared Fourier transform of a window function, corresponding to the finite size of the system. More generally, the function $f(x)$ of Eq. (3.11) is expected to describe the scaling behavior of the Fourier transform around any Bragg peak for periodic, quasiperiodic, and almost-periodic structures.

For generic values of the exponent γ of Eq. (3.7), the occurrence of scaling laws of the form (3.8), with more complicated functions $f(x)$, can be demonstrated, at least in some specific cases, either analytically, e.g., for the Thue-Morse sequence, or numerically, e.g., for the circle sequence.⁵

B. Recursion relations between Fourier amplitudes

Let us come back to the present example. The only tool at our disposal, in order to study the Fourier transform of the structure generated by the substitution σ , consists in the following linear recursion relations between the Fourier amplitudes $G_n^A(q)$ and $G_n^B(q)$:

$$G_{n+1}^A = (1 + e^{-iq_l^A} + e^{-2iq_l^A})G_n^A + e^{-3iq_l^A}G_n^B, \quad (3.12a)$$

$$G_{n+1}^B = (1 + e^{-iq_l^B})G_n^B + e^{-2iq_l^B}G_n^A, \quad (3.12b)$$

which can be easily derived from the relations (2.1), concerning the words A_n and B_n themselves. The initial conditions for the above recursion relations read

$$G_0^A = e^{-iq_l^A} = e^{-iq\tau}, \quad G_0^B = e^{-iq_l^B} = e^{-iq}, \quad (3.13)$$

and the lengths l_n^A and l_n^B have been given in Eq. (2.5).

Before we present our numerical results, let us note at once that the whole information is encoded in the asymptotic behavior of a product of noncommuting matrices

$$\underline{\Pi}_n = \underline{M}_{n-1} \cdots \underline{M}_1 \underline{M}_0. \quad (3.14)$$

Equations (3.12) and (3.13) imply indeed that the Fourier amplitudes are given by

$$\begin{pmatrix} G_n^A \\ G_n^B \end{pmatrix} = \underline{\Pi}_n \begin{pmatrix} e^{-iq\tau} \\ e^{-iq} \end{pmatrix}. \quad (3.15)$$

More explicitly, we have

$$\underline{M}_n = \begin{pmatrix} 1 + e^{i\phi_n^A} + e^{2i\phi_n^A} & e^{3i\phi_n^A} \\ e^{2i\phi_n^B} & 1 + e^{i\phi_n^B} \end{pmatrix}, \quad (3.16)$$

where the phases read $\phi_n^A = -ql_n^A$ and $\phi_n^B = -ql_n^B$. These quantities fulfill therefore the same linear recursion as (2.3), namely,

$$\begin{pmatrix} \phi_{n+1}^A \\ \phi_{n+1}^B \end{pmatrix} = \underline{M} \begin{pmatrix} \phi_n^A \\ \phi_n^B \end{pmatrix}. \quad (3.17)$$

Since these phases only enter Eq. (3.16) through complex exponentials, it suffices to consider them modulo 2π , i.e., to restrict their domain to the two-torus $T = [0, 2\pi]^2$, with periodic boundary conditions. The recursion relations (3.17) induce a map on T , which is a uniformly hyperbolic linear map, similar to the Arnold-Sinai cat maps,¹³ up to the noticeable difference that the present case is not area preserving ($\det \underline{M} \neq 1$).

The study of the local behavior of the Fourier intensity essentially amounts to investigating the orbits of the map (3.17) as a function of their initial point ($\phi_0^A = -q\tau$, $\phi_0^B = -q$). In spite of the existence of quite remarkable results,¹⁴ concerning especially number-theoretical aspects of the classification of the periodic orbits, a systematic study of the statistics of aperiodic orbits of the map (3.17) is still beyond the present scope of mathematics.

It is nevertheless possible to investigate, using the map (3.17), the possible presence of Bragg peaks in the Fourier spectrum, following the argument of Bombieri and Taylor:⁵ There is a Bragg peak in the Fourier spectrum for the value q_0 of the wave vector if and only if the sequences of phases ϕ_n^A and ϕ_n^B converge to zero (mod 2π) when $n \rightarrow \infty$. Indeed, the matrix product of Eq. (3.14) grows as the n th power of the leading eigenvalue λ_1 under those circumstances, hence building a Bragg peak. In the present case, the phases ϕ_n^A, ϕ_n^B converge to zero (mod 2π) if and only if they are exact multiples of 2π for n larger than some integer n_0 . This is equivalent to stating that the initial conditions of the recursion (3.17) be of the form

$$\phi_0^A = -ql^A = 2\pi m^A / 5^M, \quad \phi_0^B = -ql^B = 2\pi m^B / 5^M, \quad (3.18)$$

where M, m^A, m^B are arbitrary integers. The conditions (3.18) are never met with our choice $l^A/l^B = \tau$, except in the obvious case $q=0$. As a consequence, the Fourier transform of the model under study does not contain any Bragg peak, except for $q=0$. This result is in agreement with the general statements made in the Introduction

about the Pisot theorem, since $\lambda_1 = 2 + \tau$ is not a Pisot number.

In the following we will employ the recursion relations (3.12) in order to make an extensive numerical analysis of various aspects of the Fourier spectrum of the structure. We will henceforth use the notation

$$x = \frac{qa}{2\pi}. \quad (3.19)$$

C. Short-range order in the structure and the Hendricks-Teller model

Figure 2 shows a plot of the normalized amplitudes $G_n^A(q)/v_n^A$, corresponding to the finite structures associated with the words A_n , for $n=1-4$, against the reduced wave vector x defined in Eq. (3.19). A progressive, albeit irregular, emergence of sharper and sharper peaks at well-defined positions is clearly seen. For the time being, we employ the word "peaks" with its common meaning of "sharp maximum."

We are thus led to wonder why there should be peaks at all in the Fourier spectrum of the present structure. Before delving into a more quantitative study of the Fourier spectrum, it is therefore interesting to compare the deterministic structure under study, as well as its Fourier spectrum, to its random counterpart, namely, the amorphous binary structural model, obtained by putting at random along a line two types of bonds of lengths $l^A = \tau$ and $l^B = 1$, with respective probabilities $p^A = \tau^{-1}$ and $p^B = \tau^{-2}$. These probabilities are equal to the components of the normalized right Perron-Frobenius eigenvector \mathbf{v} given in Eq. (2.2).

This comparison will show how far from random the present structure is and, more precisely, which characteristics of its Fourier spectrum are induced by the short-range order of the structure, namely, the presence of only two interatomic distances, the values of these distances, and their relative weights.

The random structure considered just above is a special case of the so-called Hendricks-Teller (HT) model,¹⁵ defined by covering a line with bonds with independent random lengths l_k , drawn from some common distribution $\rho(l)dl$. It can be shown that the averaged structure factor of this model reads

$$S_{\text{HT}}(q) = \frac{1 - z\bar{z}}{(1-z)(1-\bar{z})}, \quad (3.20)$$

with

$$z = \int \rho(l)dl e^{iql}.$$

This absolutely continuous structure factor is an illustration of Sec. III A 1.

In the present case, the content in single bonds of the substitutional structure is identical to that of the binary distribution

$$\rho(l) = \tau^{-1}\delta(l-\tau) + \tau^{-2}\delta(l-1), \quad (3.21)$$

for which we get

$$S_{\text{HT}}(q) = \frac{\sin^2(q/2\tau)}{\tau^2 \sin^2(q\tau/2) + \tau \sin^2(q/2) - \sin^2(q/2\tau)} \quad (3.22)$$

This structure factor exhibits very sharp peaks for wave vectors such that both q and $q\tau$ are very close to integer multiples of 2π . Indeed, under such circumstances, all the atoms diffract almost coherently. The sharpest peaks occur for $q = q_n = 2\pi\tau^n/a$, with $a = \lambda_2$, as previously. For this particular sequence of peaks, an appropriate expansion of Eq. (3.22) around $q = q_n$ shows that the maximal intensity reads $S_{\text{HT}}(q_n) \approx (5/\pi^2)\tau^{2n-1}$ and that their line shape is a normalized Lorentzian with a half-width given by $\Gamma_n \approx (\pi/5)\tau^{1-2n}$. These asymptotic estimates, valid for large values of the index n , show that the structure factor contains arbitrarily sharp peaks, although it is evidently absolutely continuous. Wave vectors of the form $q = q_m + q_n$, etc., also correspond to sharp peaks.

This phenomenon is illustrated in Fig. 3, which shows the Fourier intensity $S_{\text{HT}}(q)$, given by Eq. (3.22), plotted against the variable x of Eq. (3.19).

D. Multifractal analysis

The next approach that we want to present consists in using multifractal analysis in reciprocal space along the very lines of Ref. 4, where the interested reader can find more details about the method.

Figure 4 shows a plot of the $f(\alpha)$ spectrum associated with wave vectors in the range $0 \leq q \leq 4\pi/a$, i.e., $0 \leq x \leq 2$. The plotted data correspond to the words A_8 and A_9 , which contain $v_8^A = 34\,375$ and $v_9^A = 124\,375$ atoms, respectively. The data have essentially already converged to a smooth limit $f(\alpha)$ curve, which is therefore a characteristic of the infinite structure.

We recall here briefly the physical meaning of the multifractal formalism of the $f(\alpha)$ spectrum and its most salient features. The quantity $f(\alpha)$ is commonly interpreted as being the dimension of the set of values of the wave vector q , around which the intensity measure scales as

$$|H(q \pm \epsilon) - H(q)| \sim \epsilon^\alpha \quad (\epsilon \rightarrow 0). \quad (3.23)$$

The following particular values deserve more interest. The exponent α_0 , corresponding to the top of the $f(\alpha)$

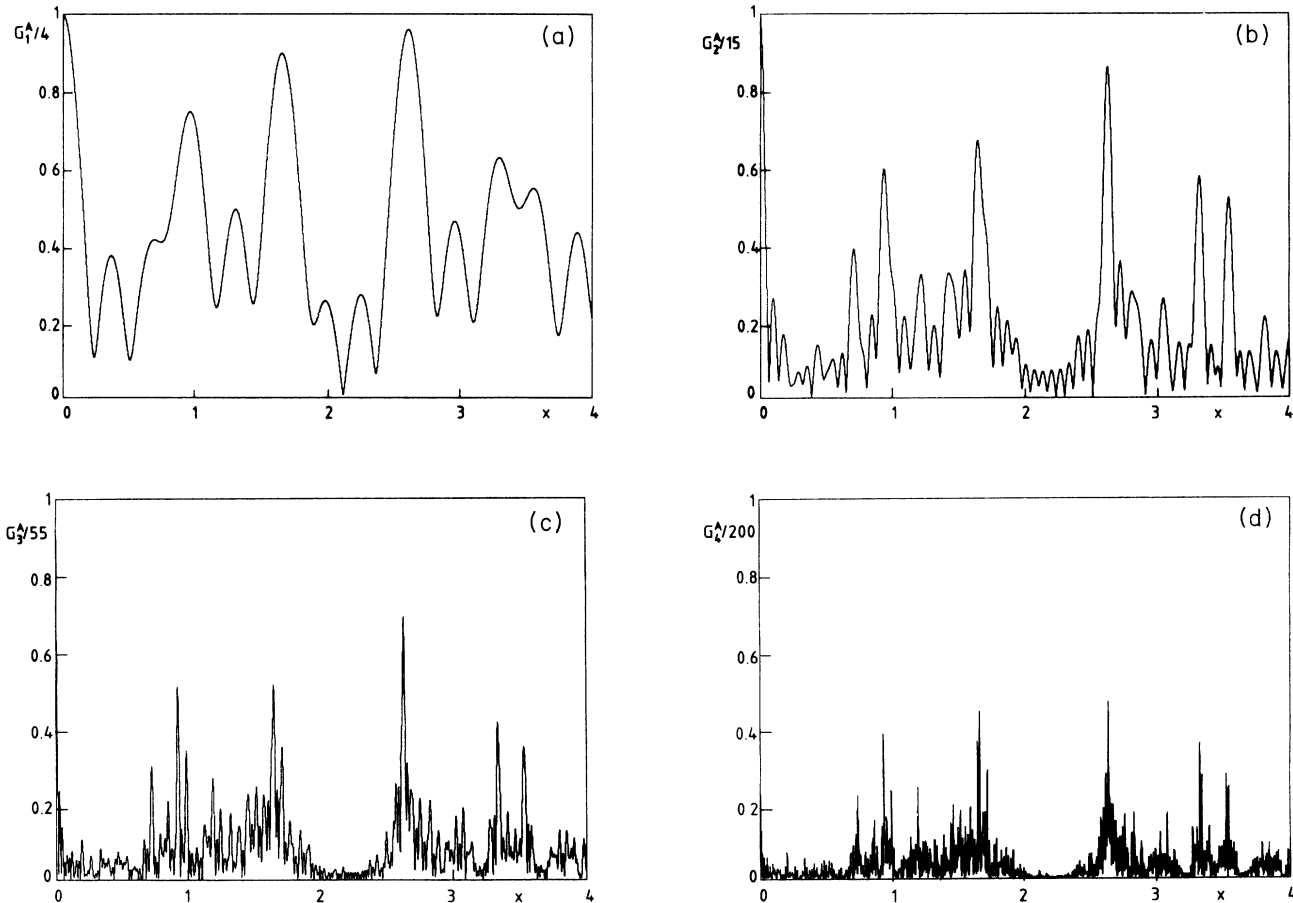


FIG. 2. Plot of the normalized Fourier amplitudes G_n^A/v_n^A , associated with the successive words A_n , against the reduced wave vector x . (a) $n=1$ ($v_1^A=4$), (b) $n=2$ ($v_2^A=15$), (c) $n=3$ ($v_3^A=55$), and (d) $n=4$ ($v_4^A=200$).

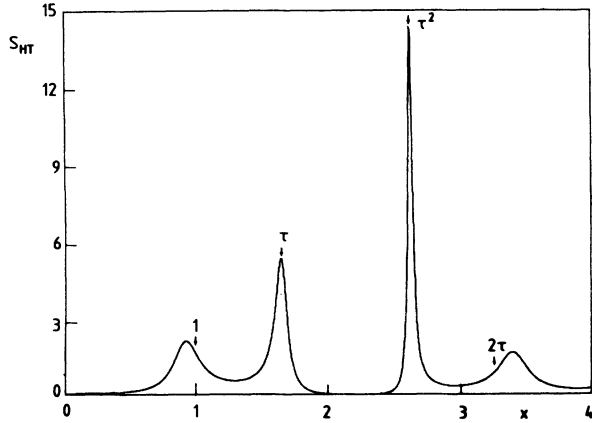


FIG. 3. Plot of the Fourier intensity S_{HT} of the random Hendricks-Teller model corresponding to the binary distribution (3.21) of bond lengths, against the reduced wave vector x . The indexation of the peaks is discussed in the text.

curve, characterizes to some extent the behavior of the intensity at a generic wave vector. We have $f(\alpha_0)=1$, the dimension of the support of the intensity measure, namely, the whole real q line. The values α_{\min} and α_{\max} are the extremal values of the local exponent α which occur with an appreciable weight. As a consequence of the above interpretation, the quantity $d_p=f(1)$ is the dimension of the set of wave vectors for which $\alpha=1$. It can be shown that d_p is also the dimension of the set of values of q for which the structure factor diverges ($\alpha \leq 1$). d_p can thus be interpreted as the *dimension of the peaks* of the diffraction spectrum. Finally, the value α_1 for which $f(\alpha)=\alpha$ is usually referred to as the (information) dimension of the intensity measure.

The characteristic values mentioned above can be determined in an accurate fashion by extrapolating the data presented in Fig. 4 and those corresponding to ear-

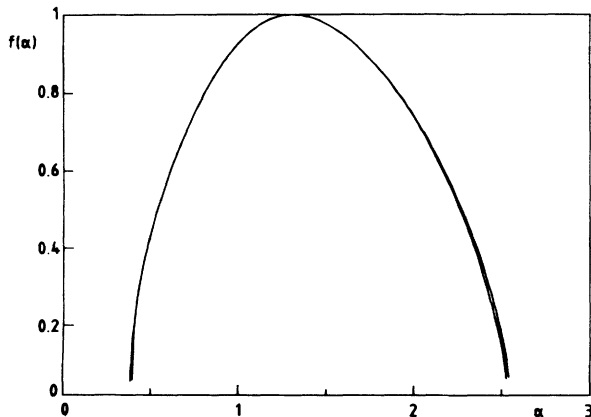


FIG. 4. Plot of the $f(\alpha)$ spectrum given by the multifractal analysis of the Fourier intensity discussed in the text, over the interval $0 \leq x \leq 2$.

lier generations. We thus obtain

$$\begin{aligned} \alpha_0 &\approx 1.29, \quad \alpha_{\min} \approx 0.38, \quad \alpha_{\max} \approx 2.55, \\ \alpha_1 &\approx 0.73, \quad d_p \approx 0.93. \end{aligned} \quad (3.24)$$

To conclude this analysis, we recall one of the main predictions of multifractal analysis in reciprocal space.⁴ Since α_0 is larger than unity, the Fourier intensity vanishes at a generic wave vector q , so that the intensity measure has no absolutely continuous component. Moreover the Bombieri-Taylor argument has already ruled out the presence of Bragg peaks, with the exception of a trivial one at $q=0$. We are thus left with the conclusion that the Fourier transform is singular continuous.

E. Toward a quantitative local analysis

This last step consists in looking for the genuine *peaks* of the diffraction spectrum, in the sense of values q_0 of the wave vector such that the scaling laws (3.7)–(3.9) of singular scattering hold true, at least in some weak sense.

The most direct way of searching peaks in the diffraction spectrum consists in looking for the values of q such that the partial Fourier intensities $S_n^A(q)$ are maximal. This tautological method suffers from an obvious resolution problem. Indeed, the intensities $S_n^A(q)$ exhibit many spiky, and apparently erratic, oscillations down to the scale of the inverse of the sample size, i.e., $\Delta x \sim \lambda_1^{-n}$, where x has been introduced in Eq. (3.19). The number of values of q to be explored, in order to find a maximum which can be trusted, grows therefore as $1/\Delta x$, i.e., exponentially, with the generation label n , so that it is hardly efficient above $n=10$ (A_{10} has 450 000 atoms).

The method can be improved by shrinking at each generation n the interval to be explored to a “reasonable” vicinity of the wave vector for which the intensity was maximal at the previous generation. There are nevertheless cases where the distance between “father-and-son” maxima of S_n^A and S_{n-1}^A is much larger (up to some 1000 times) than the expected scale Δx .

These anomalously large jumps between successive maxima are a consequence of the seemingly random nature of the oscillations of the partial intensities $S_n^A(q)$, which are clearly visible in Fig. 2. This complex behavior of the Fourier intensities originates itself in the chaotic, or intermittent, behavior of the aperiodic orbits of the map (3.17). Let us underline once more that, to the best of our knowledge, no satisfactory quantitative understanding of those orbits is available.

Using the above method as a mere numerical scheme, we have succeeded in locating a very distinct *peak*, the word being now taken in the sense of singular scattering, at the following value of the wave vector:

$$x_0 = \frac{q_0 a}{2\pi} \approx 0.917\,638\,024\,883\,44. \quad (3.25)$$

Figure 5 shows a logarithmic plot of the quantity v_n^A/S_n^A , against the generation label n , where the intensity is taken, for each n , at its maximum which is closest to q_0 . The data points exhibit irregular but weak fluctuations around a clear mean linear behavior. The slope s of the

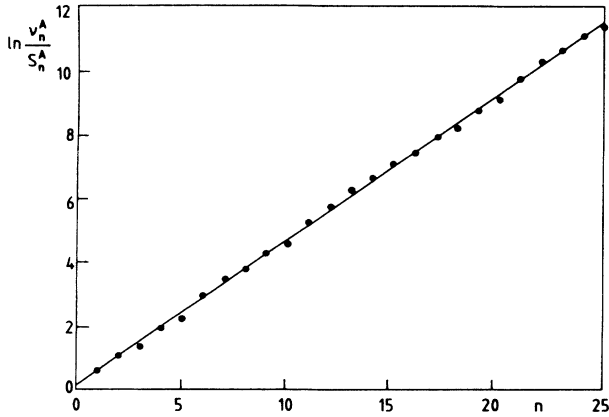


FIG. 5. Logarithmic plot of the ratios v_n^A/S_n^A , against the generation label n . The Fourier intensities are evaluated at their maximum closest to the wave vector q_0 , defined in Eq. (3.25). The straight line shows a least-squares fit of the data, which yields the value 0.823 for the mean local scaling exponent γ .

plot is related to the exponent γ of the power law (3.7) through $s = 2(1-\gamma)\ln\lambda_1$. A least-squares fit of the numerical data yields the slope $s \approx 0.455$, whence $\gamma \approx 0.823$. Taking for granted the identity (3.9) between the local exponents α and γ , we obtain the value $\alpha \approx 0.354$. We will come back to this result later.

Let us nevertheless emphasize that the nice and regular scaling properties described in Sec. III A 3 do not hold true, strictly speaking, in the present case. In particular, the intensities $S_n^A(q)$ definitely do not assume an asymptotic finite-size scaling law of the form (3.8) for x close to the selected x_0 , given in Eq. (3.25). In other words, the intensities of the successive generations have an intrinsically complex behavior. This is a manifestation of the chaotic effects mentioned above, which restrict the validity of the scaling laws (3.7)–(3.9). Let us therefore adopt the terminology of *mean local scaling* and call the values of γ and α determined above the *mean local exponents* at $q = q_0$.

We have confirmed the existence of a mean scaling behavior for the Fourier transform around q_0 by the following alternative approach, which consists in evaluating the spectral measure $H(q)$ of the infinite structure, in order to check the scaling law (3.9) and to measure the exponent α in a more direct way. It turns out that the integrals involved in Eq. (3.4) converge very nicely toward their limit, provided they are evaluated properly, in the sense of the above discussion about resolution, namely, with a variable integration step δx , such that the condition $\delta x \ll \lambda_1^{-n}$ is always fulfilled.

Figure 6 shows a log-log plot of $|\Delta H| = |H(x) - H(x_0)|$, against $|\Delta x| = |x - x_0|$, for both $x > x_0$ and $x < x_0$. We have performed a least-squares fit of the data, assuming that both data series follow power laws with a common exponent α and two different amplitudes A_{\pm} , i.e.,

$$|\Delta H| \approx A_{\pm} |\Delta x|^{\alpha} \text{ as } \Delta x \rightarrow 0^{\pm}. \quad (3.26)$$

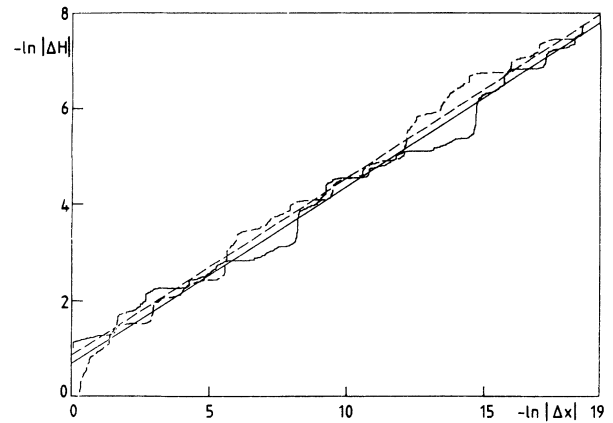


FIG. 6. Log-log plot of the differences of integrated intensity $|H - H_0|$, against $|x - x_0|$, for reduced wave vectors x close to the x_0 of Eq. (3.25). The straight lines are the result of a common least-squares fit of the data, according to Eq. (3.26). Solid curve and line, $x > x_0$; dashed curve and line, $x < x_0$.

The numerical fit yields $\alpha \approx 0.375$, $A_+ \approx 1.95$, and $A_- \approx 2.32$. The fluctuation of the data around the straight lines seem once more irregular, but rather weak.

To end up this tentative study, let us mention that a similar analysis has been performed around many other *peaks* in other ranges of values of the wave vector. The main lines of the picture remain unchanged; namely, a mean scaling behavior of the Fourier intensity is always observed and the outcomes of both numerical schemes described above are always consistent with the relation (3.9) between α and γ and yield everywhere the same value for the mean local scaling exponent, within numerical accuracy, say,

$$\alpha_{\text{loc}} = 0.36 \pm 0.02, \quad (3.27)$$

for the sake of definiteness.

IV. DISCUSSION

We have investigated a one-dimensional self-similar structural model generated by one of the simplest generic non-Pisot substitutions, aiming at understanding the nature of its Fourier transform. We have emphasized successively several remarkable consequences of the non-Pisot property, namely, that both eigenvalues of the substitution matrix \underline{M} are larger than unity.

The main outcomes of this study are the following. The fluctuations of the atomic abscissas with respect to the average lattice of the structure exhibit a strong divergence, growing as a power of the system size. The gross features of the Fourier spectrum resemble those of the binary Hendricks-Teller model of a random structure with the same content in interatomic bond lengths. The Fourier transform of the structure, more precisely its intensity measure, is singular continuous, since the Bombieri-Taylor argument rules out the existence of

Bragg peaks, except for a trivial one at the wave vector $q=0$, whereas multifractal analysis in reciprocal space shows that there is no intensity at a generic value of the wave vector (since $\alpha_0 \approx 1.29 > 1$) and that the set of *peaks* of the Fourier transform has a dimension $d_p \approx 0.93$.

The more difficult question of indexing the peaks of singular scattering was then addressed. We have presented a detailed numerical analysis of the local behavior of the diffraction spectrum, which suggests that there exists a *mean scaling behavior* around some particular values of the wave vector, in spite of apparently random variations of the partial intensities that are due to the chaotic behavior of the underlying generalized map (3.17).

For those values of the wave vector, such as, e.g., q_0 given in Eq. (3.25), the local scaling properties have been explored by looking at (i) the growth of the intensities of *finite* samples at $q=q_0$ as a function of the system size, yielding the exponent γ , and (ii) the behavior of the integrated intensity of the *infinite* system around $q=q_0$, yielding the exponent α . Both approaches are fully consistent with the scaling law (3.9) between the exponents α and γ and yield an accurate value for a unique *mean local scaling exponent* $\alpha_{loc} = 0.36 \pm 0.02$.

The above estimate for the local exponent α_{loc} coincides (within numerical accuracy) with the value of $\alpha_{min} \approx 0.38$, determined by multifractal analysis. Our opinion is that this empirical observation reflects more than a fortuitous coincidence.

Taking the equality $\alpha_{loc} = \alpha_{min}$ for granted, we are led to make the following speculations. According to the standard interpretation of the multifractal formalism, for any value of α in the open interval $]\alpha_{min}, \alpha_{max}[$, there is a set of positive dimension $f(\alpha)$, and hence a continuous (noncountable) infinity, of wave vectors which possess the local exponent α , at least in some weak sense. Even though multifractal analysis does not bring, strictly speaking, any precise information about the set of wave vectors with the minimal exponent α_{min} , we may suspect that this set is still *continuously infinite*.

The following scenario seems therefore plausible. The most pronounced *peaks* of the diffraction spectrum are all characterized by the scaling exponent α_{loc} of Eq. (3.27). They form a continuously infinite set and cannot therefore be indexed by any simple scheme, such as a finite set of integer indices. The value q_0 given in Eq. (3.25) and the other values of the wave vector at which we have performed a local scaling analysis are just a few members of

that set, which have been selected somehow at random by the procedure of Sec. III E, just because the intensity around these wave vectors is very high.

Let us add a few more arguments in favor of this possibility. First, it is worth noting that the value x_0 of Eq. (3.25), around which the singularity of the Fourier intensity is exceptionally large, is very close to a maximum of the structure factor of the Hendricks-Teller model. The presence of this maximum, which occurs for $x \approx 0.91639$, means that the short-range order of the structure is responsible for the Fourier intensity being large in this range of wave vectors. Second, we have had many unsuccessful attempts to find a simple indexing scheme for the value x_0 and for the location of other large peaks of singular scattering, either directly (e.g., by looking for regularities in various expansions of the number x_0 itself) or in terms of the map (3.17) (e.g., by looking for a system of the orbit corresponding to x_0 in terms of periodic orbits).

It is well known that, in the case of periodic crystals and quasiperiodic structures, such as incommensurates and quasicrystals, the Bragg peaks admit a simple indexing scheme in terms of integer indices. It is worth recalling here that most singular continuous diffraction spectra studied so far also share this property. This is especially the case with the circle³ and Thue-Morse sequences,¹⁶ for which the location of the most prominent peaks of singular scattering is known exactly. Let us finally note that, in all those instances, a simple indexing scheme always goes hand in hand with nice finite-size scaling laws of the form (3.8) for the partial Fourier amplitudes. Other such examples can be found in Ref. 5.

If all this holds true, generic non-Pisot self-similar structures bring, because of intrinsic chaotic effects, a contradiction to our physicists' intuition that the largest peaks in a diffraction spectrum should be related to the structure in some natural way and therefore possess a simple indexing scheme, such as e.g., by a finite set of integers, in the case of a simple deterministic structure.

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