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On certain properties of high-resolution x-ray diffraction spectra of finite-size generalized Rudin–Shapiro multilayer heterostructures

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Abstract. We present a theoretical and numerical study of certain properties of x-ray diffraction of finite-size Rudin–Shapiro and generalized Rudin–Shapiro multilayer heterostructures. They are compared with spectra obtained in similar conditions from other deterministically disordered multilayer systems, in particular Thue–Morse heterostructures.

1. Introduction

In recent years, following the discovery of quasicrystals [1], the importance of deterministic structures having controlled aperiodic disorder described by sequences generated by substitutions on a finite alphabet (often a two-letter alphabet) or finite automata [2–4], has been increasingly recognized.

Mathematical studies in 2D have now already begun [5, 6] and as such models in 1D have been widely studied, they even have inspired quite a few experiments on model systems, in particular on multilayer superlattices made by molecular beam epitaxy (MBE). These superlattices have two kinds of layers arranged according to the Fibonacci [7–13] and the Thue–Morse sequences [12, 14], both of which are two-letter sequences.

Recently finite-size multilayer superlattices arranged according to the Thue–Morse sequence have been investigated by high-resolution x-ray diffraction [14] with the finding that the singular continuous nature of the infinite lattice diffraction pattern (i.e. the Fourier transform of the Thue–Morse sequence) has observable consequences for finite samples. Among these consequences are the indexing, in this case, of the diffraction peaks by the rationals $(2k+1)/[3.2^n]$, k and n being integers, with high accuracy, and the description of peak height evolution with sample size and wavevector by a measurable exponent $\alpha_n(q)$. These results emphasize for the first time the importance of the nature of the measure associated with the Fourier

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transform of the sequence generating the aperiodic deterministic disorder under study, as defined by the Lebesgue decomposition theorem

$$\mu = \mu_{\mathrm{AT}} + \mu_{\mathrm{SC}} + \mu_{\mathrm{AC}}$$

μ_{AT}, μ_{SC} and μ_{AC} being respectively the associated atomic, singular continuous and absolutely continuous (with respect to the Lebesgue measure) primitive components into which any measure μ can be uniquely decomposed.

The Fibonacci sequence Fourier transform has a purely atomic measure with a countable number of peaks, the Thue–Morse sequence Fourier transform is also a pure case, but of a singular continuous measure. We shall now be interested in the Rudin–Shapiro and the generalized Rudin–Shapiro sequences, and their Fourier transforms, which in the infinite limit are a pure case of the third type of primitive measure component, the absolutely continuous one. This together with the fact that a random sequence also has a purely AC Fourier transform (e.g. 'white noise') led us to raise, we believe for the first time in the study of the crystallography of disordered systems, the question of the possible existence of criteria allowing specific distinction between random and deterministic disorder having an AC Fourier transform by means of theoretical as well as of experimental studies of the high-resolution x-ray diffraction spectra of appropriate systems, model systems to begin with. We recall that in previous work [14], devoted to Thue–Morse disorder, one of us already brought the question of the specificity of the relationship between the deterministic generating sequence and the resulting x-ray diffraction multilayer spectra. Finally, we note that besides purely mathematical studies, [4, 15–17], some of the properties associated with the Rudin–Shapiro sequence have been described before [18, 19]. Our conclusion can be compared with part of the discussion in [20] based on the study of a different problem.

2. The Rudin–Shapiro and generalized Rudin–Shapiro sequences: definitions and properties

2.1. The '$\sqrt{N}$ property'

Let a sequence of $+1$ and -1, $\{\epsilon_N(j)\}$ be of length N. The intensity—square modulus—of its Fourier transform defined as usual is

$$|\tilde{\epsilon}_N(q)|^2 = \frac{1}{N}\left|\sum_{j=0}^{N-1} \epsilon_N(j)\mathrm{e}^{2\mathrm{i}\pi jq}\right|^2 . \tag{1}$$

Then trivially for every q in $[0, 1]$

$$1 = \int_0^1 \mathrm{d}q\,|\tilde{\epsilon}_N(q)|^2 \leqslant \sup_{q\in[0,1]} |\tilde{\epsilon}_N(q)|^2 \leqslant N. \tag{2}$$

A sequence is said to have the '$\sqrt{N}$ property' if

$$\sup_{q\in[0,1]}\left|\sum_{j=0}^{N-1} \epsilon_N(j)\mathrm{e}^{2\mathrm{i}\pi jq}\right| \leqslant K\sqrt{N}. \tag{3}$$

The sequence of +1 and −1 constructed by Shapiro [15], and then Rudin [16] described below, has this property which is shared by the 'generalized Rudin–Shapiro sequences' studied in [21] (where it is also proven that for such an infinite sequence, to have a bounded Fourier transform intensity, or equivalently the '$\sqrt{N}$ property', entails that its spectral measure is absolutely continuous (AC) with respect to the Lebesgue measure).

As a consequence, spectra having an associated atomic (AT) measure (e.g. Fibonacci or paperfolding) or a singular continuous (SC) measure (e.g. Thue–Morse) do not have a bounded intensity (we have defined and measured the specific exponents $\alpha_n(q)$ in the Thue–Morse case in [14]), nor do they have the '$\sqrt{N}$ property'.

2.2. *The Rudin–Shapiro sequence*

The Rudin–Shapiro sequence has been constructed independently by Shapiro [15] and Rudin [16] and can be defined in four different yet equivalent ways [4, 17, 18].

(i) u_n counts the parity of the number of times the sequence 11 occurs in the binary expansion of n (with possible overlap) [17]. Let

$$n = \sum_{q=0}^{\infty} \beta_n(q) 2^q. \tag{4}$$

With $u_0 = 0$, and $u_n \epsilon \{0, 1\}$

$$u_n = \sum_q \beta_n(q)\beta_n(q+1) \qquad \text{and} \qquad \begin{cases} u_{2n} = u_n \\ u_{4n+1} = u_n \\ u_{4n+3} = 1 - u_{2n+1}. \end{cases} \tag{5}$$

(ii) As the image t of the fixed point of a two-substitution σ on a four-letter alphabet $\{A, B, C, D\}$

$$\begin{cases} \sigma(A) = AB \\ \sigma(B) = AC \\ \sigma(C) = DB \\ \sigma(D) = DC. \end{cases} \tag{6}$$

With initial condition A, we obtain, by iterating σ:

$$\begin{array}{l} A \\ AB \\ ABAC \\ ABACABDB \\ ABACABDBABACDCAC. \end{array}$$

Note that $\sigma^n(A)$ has length 2^n. With t a projection on the alphabet $\{0, 1\}$ defined by

$$\begin{array}{l} t(A) = 0 \\ t(B) = 0 \\ t(C) = 1 \\ t(D) = 1 \end{array} \tag{7}$$

we finally have

0
00
0001
00010010
0001001000011101

and some simple algebra shows that the obtained sequence is identical with the above recursively defined sequence.

(iii) The Rudin–Shapiro sequence can also be generated by a two-automaton (see for example, [4]).

(iv) Consider the following double sequence of recurrent polynomials $P_n(X)$ and $Q_n(X)$ of degree $2^n - 1$, with coefficients ± 1:

$$\begin{cases} P_{n+1}(X) = P_n(X) + X^{2^n} Q_n(X) \\ Q_{n+1}(X) = P_n(X) - X^{2^n} Q_n(X) \end{cases} \tag{8}$$

with $P_0(X) = Q_0(X) = 1$. Then

$$P_1(X) = 1 + X \qquad Q_1(X) = 1 - X$$
$$P_2(X) = 1 + X + X^2 - X^3 \qquad Q_2(X) = 1 + X - X^2 + X^3$$

etc. We can write

$$P_n(X) = \sum_{j=0}^{2^n-1} \epsilon_n(j) X^j . \tag{9}$$

Then it can easily be seen that $\epsilon_n = (-1)^{u_n}$ with u_n defined in equation (5). ϵ_n is the Rudin–Shapiro sequence on the alphabet $\{-1, +1\}$. When $X = e^{2\pi i q}$, $P_n(e^{2\pi i q})$ is the Fourier transform of the Rudin–Shapiro sequence of length 2^n as indicated below.

2.3. Generalized Rudin–Shapiro sequences

Generalized Rudin–Shapiro sequences are defined ([21], see also [22]) by an extension of the Brillhart–Carlitz definition [17] as the parity of the number of times the sequence, $1 * 1$, $1 * * 1$, etc. occurs in the binary expansion of n with possible overlapping, where the star $*$ stands for 0 or 1. We shall be particularly interested in the simplest case, $1 * 1$, which yields for the first few iterations:

0000010100110110 . . . .

This entails the following definitions for z_n on the alphabet $\{0, 1\}$, with $z_0 = 0$:

$$\begin{cases} z_{2n} = z_n \\ z_{8n+1} = z_n \\ z_{8n+3} = z_{4n+1} \\ z_{8n+5} = 1 - z_{2n+1} \\ z_{8n+7} = 1 - z_{4n+3} . \end{cases} \tag{10}$$

If

$$\eta_n = (-1)^{z_n} \tag{11}$$

then η_n is a sequence on the alphabet $\{-1, +1\}$ with $\eta_0 = 1$

$$\begin{aligned} \eta_{2n} &= \eta_n \\ \eta_{8n+1} &= \eta_n \\ \eta_{8n+3} &= \eta_{4n+1} \\ \eta_{8n+5} &= -\eta_{2n+1} \\ \eta_{8n+7} &= -\eta_{4n+3}. \end{aligned} \tag{12}$$

3. Fourier transforms of the Rudin–Shapiro and the generalized Rudin–Shapiro sequences

3.1. The Rudin–Shapiro sequence

For a chain of length N, the Fourier transform is defined as usual by

$$\tilde{\epsilon}_N(q) = \frac{1}{\sqrt{N}} \sum_{j=0}^{N-1} \epsilon_N(j) \mathrm{e}^{2\pi \mathrm{i} j q}. \tag{13}$$

Then, obviously

$$(1/2^{n/2}) P_n(\mathrm{e}^{2\pi \mathrm{i} q}) = \tilde{\epsilon}_{2^n}(q) \tag{14}$$

and the intensity of the Fourier transform is then

$$|\tilde{\epsilon}_{2^n}(q)|^2 = (1/2^n) |P_n(\mathrm{e}^{2\pi \mathrm{i} q})|^2. \tag{15}$$

Then, taking into account the fact that $|P_n(X)|^2 + |Q_n(X)|^2 = 2^{n+1}$ where $X = \mathrm{e}^{2\pi \mathrm{i} q} = \mathrm{e}^{\mathrm{i}\alpha}$

$$\begin{aligned} |P_{n+1}(X)|^2 &= |P_n(X)|^2 + |Q_n(X)|^2 + P_n(X)\overline{Q_n(X)X^{2^n}} + \overline{P_n(X)}Q_n(X)X^{2^n} \\ &= 2^{n+1} + P_n(X)\overline{Q_n(X)X^{2^n}} + \overline{P_n(X)}Q_n(X)X^{2^n} \end{aligned} \tag{16}$$

and in an analogous way:

$$|Q_{n+1}(X)|^2 = 2^{n+1} - P_n(X)\overline{Q_n(X)X^{2^n}} - \overline{P_n(X)}Q_n(X)X^{2^n} \tag{17}$$

so that

$$|P_{n+1}(X)|^2 - |Q_{n+1}(X)|^2 = 2[P_n(X)\overline{Q_n(X)X^{2^n}} + \overline{P_n(X)}Q_n(X)X^{2^n}]. \tag{18}$$

Similarly

$$P_n(X)\overline{Q_n(X)X^{2^n}} + \overline{P_n(X)}Q_n(X)X^{2^n}$$
$$= 2\cos(2^n\alpha)\left[|P_{n-1}(X)|^2 - |Q_{n-1}(X)|^2\right] + 2\mathrm{i}\sin(2^n\alpha)$$
$$\times \left[P_{n-1}(X)\overline{Q_{n-1}(X)X^{2^{n-1}}} - \overline{P_{n-1}(X)}Q_{n-1}(X)X^{2^{n-1}}\right] \tag{19}$$

and

$$P_n(X)\overline{Q_n(X)X^{2^n}} - \overline{P_n(X)}Q_n(X)X^{2^n} = -2\mathrm{i}\sin(2^n\alpha)\left[|P_{n-1}(X)|^2\right.$$
$$\left.-|Q_{n-1}(X)|^2\right] - 2\cos(2^n\alpha)\left[P_{n-1}(X)\overline{Q_{n-1}(X)X^{2^{n-1}}}\right.$$
$$\left.-\overline{P_{n-1}(X)}Q_{n-1}(X)X^{2^{n-1}}\right]. \tag{20}$$

Let us then define

$$\begin{aligned}
u_n(X) &= |P_n(X)|^2 - |Q_n(X)|^2 \\
v_n(X) &= -\mathrm{i}\left[P_n(X)\overline{Q_n(X)X^{2^n}} - \overline{P_n(X)}Q_n(X)X^{2^n}\right] \\
&= 2\mathrm{Im}\left[P_n(X)\overline{Q_n(X)X^{2^n}}\right] \\
w_n(X) &= P_n(X)\overline{Q_n(X)X^{2^n}} + \overline{P_n(X)}Q_n(X)X^{2^n} \\
&= 2\mathrm{Re}\left[P_n(X)\overline{Q_n(X)X^{2^n}}\right].
\end{aligned} \tag{21}$$

Then

$$\begin{aligned}
u_n &= 2w_{n-1} \\
v_n &= -2\sin(2^n\alpha)u_{n-1} - 2\cos(2^n\alpha)v_{n-1} \\
w_n &= 2\cos(2^n\alpha)u_{n-1} - 2\sin(2^n\alpha)v_{n-1}
\end{aligned} \tag{22}$$

which can be rewritten in the following matrix form:

$$\begin{bmatrix} u_n \\ v_n \\ w_n \end{bmatrix} = 2\begin{bmatrix} 0 & 0 & 1 \\ -\sin(2^n\alpha) & -\cos(2^n\alpha) & 0 \\ \cos(2^n\alpha) & -\sin(2^n\alpha) & 0 \end{bmatrix}\begin{bmatrix} u_{n-1} \\ v_{n-1} \\ w_{n-1} \end{bmatrix} \tag{23}$$

and finally

$$\begin{bmatrix} u_n \\ v_n \\ w_n \end{bmatrix} = 2^n\prod_{j=1}^{n}\begin{bmatrix} 0 & 0 & 1 \\ -\sin(2^j\alpha) & -\cos(2^j\alpha) & 0 \\ \cos(2^j\alpha) & -\sin(2^j\alpha) & 0 \end{bmatrix}\begin{bmatrix} u_0 \\ v_0 \\ w_0 \end{bmatrix} \tag{24}$$

with initial conditions corresponding to $P_0(X) = Q_0(X) = 1$, that is $u_0 = 0$, $v_0 = -2\sin(2\pi q)$, $w_0 = 2\cos(2\pi q)$, which yields in turn $u_1 = 4\cos(2\pi q)$, $v_1 = -4\sin(2\pi q)\cos(4\pi q)$, $w_1 = -4\sin(2\pi q)\sin(4\pi q)$, and so on.

From equations [15] and [16]

$$|\tilde{\epsilon}_{2^n}(q)|^2 = (1/2^n)|P_n(e^{2\pi i q})|^2 = (1/2^n)(2^n + w_{n-1}(q)) = 1 + w_{n-1}(q)/2^n \quad (25)$$

is the intensity of the Fourier transform of a Rudin–Shapiro chain of length 2^n. This intensity is known to go to a finite constant when n goes to infinity, in accordance with the absolutely continuous character of the measure. An analogous calculation has been previously published in [19].

An alternative way of calculating this Fourier transform is as follows.

Let the two-vector U_n be defined as

$$U_n = \begin{bmatrix} \epsilon_n \\ \epsilon_{2n+1} \end{bmatrix}. \quad (26)$$

Then the following 2×2 matrices A_0 and A_1 are naturally defined:

$$\begin{aligned} U_{2n} &= \begin{bmatrix} \epsilon_n \\ \epsilon_n \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 1 & 0 \end{bmatrix} U_n = A_0 U_n \\ U_{2n+1} &= \begin{bmatrix} \epsilon_{2n+1} \\ -\epsilon_{2n+1} \end{bmatrix} = \begin{bmatrix} 0 & 1 \\ 0 & -1 \end{bmatrix} U_n = A_1 U_n. \end{aligned} \quad (27)$$

Then with

$$F(n,q) = \sum_{j=0}^{2^n-1} U_j e^{2\pi j q} \quad (28)$$

it can easily be seen that F verifies the simple equation

$$F(n+1,q) = A_0 F(n,2q) + e^{2\pi i q} A_1 F(n,2q) \quad (29)$$

or, if $M(q) = A_0 + e^{2\pi i q} A_1$,

$$F(n+1,q) = M(q)F(n,2q) \quad (30)$$

which recursively yields

$$\begin{aligned} F(n,q) &= M(q)F(n-1,2q) = M(q)M(2q)F(n-2,4q) \\ &= M(q)M(2q)\ldots M(2^{n-1}q)F(0,2^n q). \end{aligned} \quad (31)$$

Since for all q, $F(0,2^n q) = U_0 = [{}^1_1]$ for the Rudin–Shapiro sequence, $(u_1 = u_0 = 1)$, $\tilde{\epsilon}_{2^n}(q)$ is found as the first component of $F(n,q)$.

This formalism has the advantage that it extends naturally to generalized Rudin–Shapiro sequences.

3.2. Generalized Rudin–Shapiro sequences

Let

$$Z_n = \begin{bmatrix} \eta_n \\ \eta_{2n+1} \\ \eta_{4n+1} \\ \eta_{4n+3} \end{bmatrix} . \tag{32}$$

Then the following relationships hold:

$$\begin{aligned} Z_{2n} &= A_0 Z_n \\ Z_{2n+1} &= A_1 Z_n \end{aligned} \tag{33}$$

with

$$A_0 = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{bmatrix} \qquad \text{and} \qquad A_1 = \begin{bmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & -1 \end{bmatrix} . \tag{34}$$

If we define

$$G(n,q) = \sum_{j=0}^{2^n-1} Z_j e^{2\pi j q} \tag{35}$$

we are led as previously to

$$G(n,q) = M(q)M(2q)M(4q)\ldots M(2^{n-1}q)G(0,2^n q) \tag{36}$$

with

$$G(0,2^n q) = \begin{bmatrix} 1 \\ 1 \\ 1 \\ 1 \end{bmatrix} .$$

M is defined in an identical way as before:

$$M(q) = A_0 + e^{2\pi i q} A_1$$

but M is now a 4×4 matrix.

$\tilde{\eta}_{2^n}(q)$ is now found as the first component of $G(n,q)$.

4. Results and discussion of the spectra

We now proceed to discuss some of the consequences of these properties on the x-ray diffraction profiles of finite-size model systems, which we will compare to the Thue–Morse studies.

We assume the model system is in all cases a finite-size multilayer heterostructure made of two different kinds of plane layers deposited by MBE according to the Thue–Morse, Rudin–Shapiro and generalized Rudin–Shapiro sequences. The system is investigated with high-resolution x-ray diffraction, the experimental conditions being those of [14]. Then, besides the Fourier transform of the finite deterministic generating sequence which is in some sense the backbone of the x-ray diffraction profile, various other ingredients possibly contribute to the signal actually obtained, among which are:

(i) a background level coming from the diffraction profile of the substrate on which the deposition is being made;

(ii) a contribution from the entrance slit width for the beam;

(iii) a contribution from the detector slit width, the effect of which can be easily simulated numerically, by simply averaging the Fourier transform intensity over the slit width in q. Examples are shown below.

Comparison of figures 1(*a*) and 2(*a*) shows the striking differences exhibited by the computed spectra. The Thue–Morse situation clearly appears to be on the crystal or quasicrystal side with well defined peaks, stable under size change and unbounded intensity; while the Rudin–Shapiro case has, on the contrary, no peak stability under size changes, bounded intensity (in connection with the absolutely continuous character of the underlying measure), which obviously goes to a finite constant in the infinite limit. These characteristics are also shared by the spectra of the generalized Rudin–Shapiro sequence described above (see figure 3). They are not fundamentally altered when a detector slit width is simulated in the calculation, producing a certain deformation of the signal (in particular line broadening) (see figures 1(*b*), 1(*c*) and 2(*c*)).

An important common feature of Thue–Morse and Rudin–Shapiro situations is the existence of a fixed point for the change $q \to 2q$ which transposes in q-space the variations of the sequence length.

The Thue–Morse intensity which has been known for quite a long time (quoted, for example, in [14, 23], see also references therein)

$$I_n(q) = 2^{2^n} \prod_{j=0}^{n-1} \sin^2(2^j \pi q) \tag{37}$$

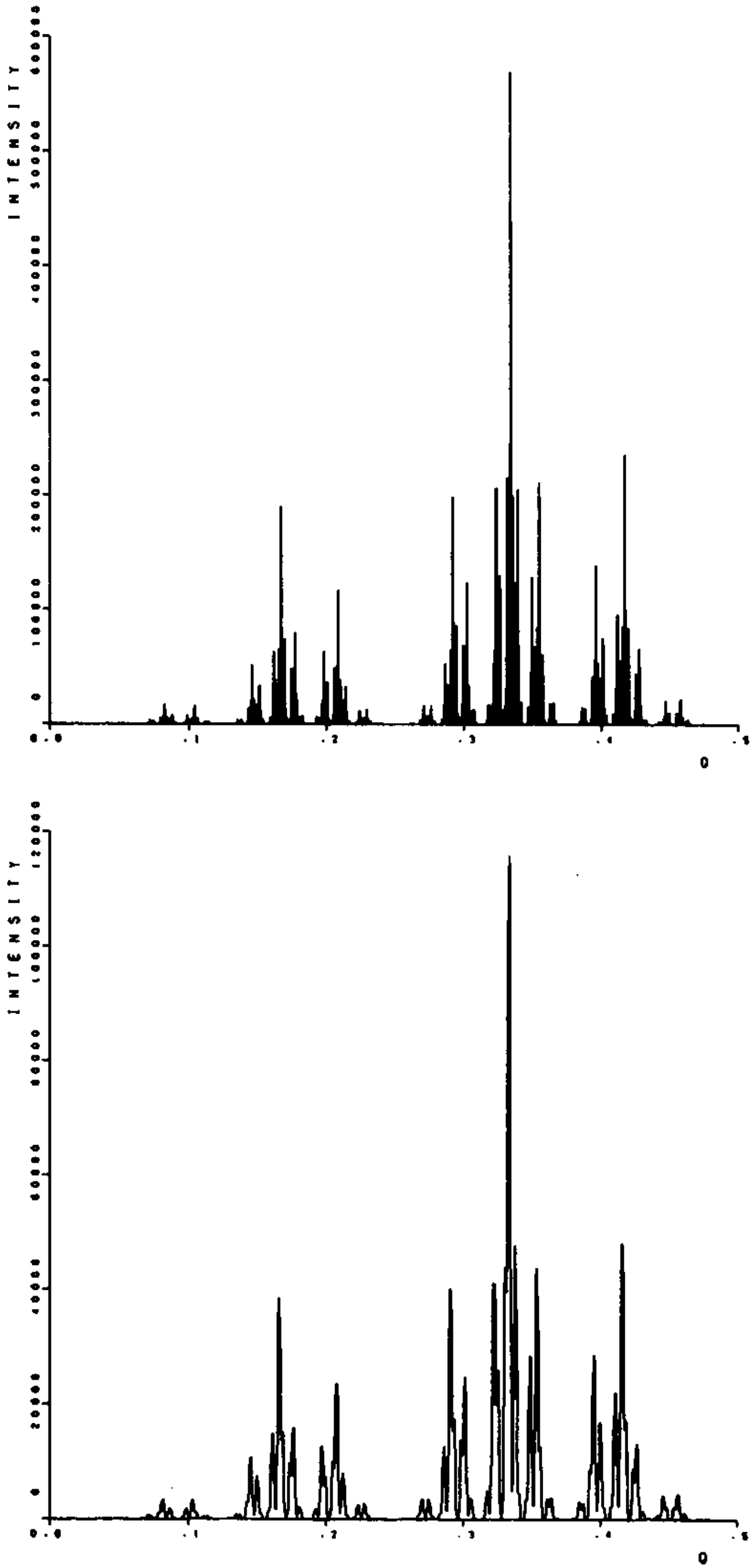
indicates the privileged role of the value $q = \frac{1}{3}$ while equations (23) and (24) show $q = 0$ modulo 2π to be the fixed point for the Rudin–Shapiro situation.

In the Thue–Morse case, when $q = \frac{1}{3}$ or when by multiplying q by a certain finite power of two one arrives at the value $\frac{1}{3}$ for q (i.e. when $q = (2k+1)/[3.2^p]$) there is an accumulation of intensity. Therefore we see that this fixed point plays a crucial role in the definition of the x-ray spectrum.

In the Rudin–Shapiro situation, when $q = 0$ modulo 2π or becomes 0 at step n after multiplication by a finite number of factors 2, i.e. $q = (2k+1)/2^p$, equation (23)

(a)

(b)



(*c*)

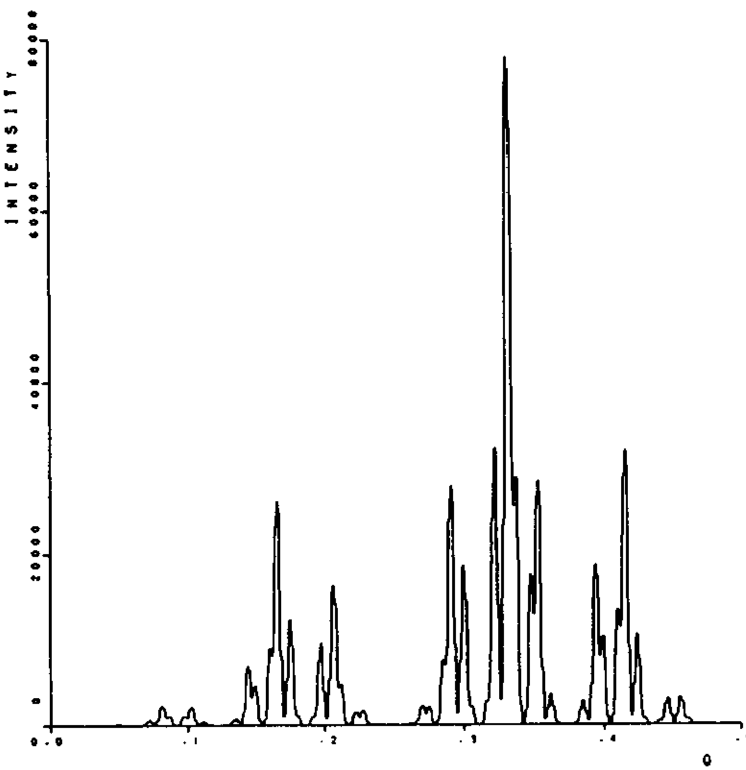


Figure 1. (*a*) The calculated intensity of the Fourier transform of the abstract Thue–Morse sequence of length $2^{12} = 4096$ (see text). The number of points in q is 16384. The intensity is unbounded. (*b*) The same length as (*a*) with a detector slit width of 64 in q ($\sim 4\%$). (*c*) The same length as (*a*) with a detector slit width of 128 in q ($\sim 8\%$).

yields

$$\begin{bmatrix} u_n \\ v_n \\ w_n \end{bmatrix} = 2 \begin{bmatrix} 0 & 0 & 1 \\ 0 & -1 & 0 \\ 1 & 0 & 0 \end{bmatrix} \begin{bmatrix} u_{n-1} \\ v_{n-1} \\ w_{n-1} \end{bmatrix} \tag{38}$$

or

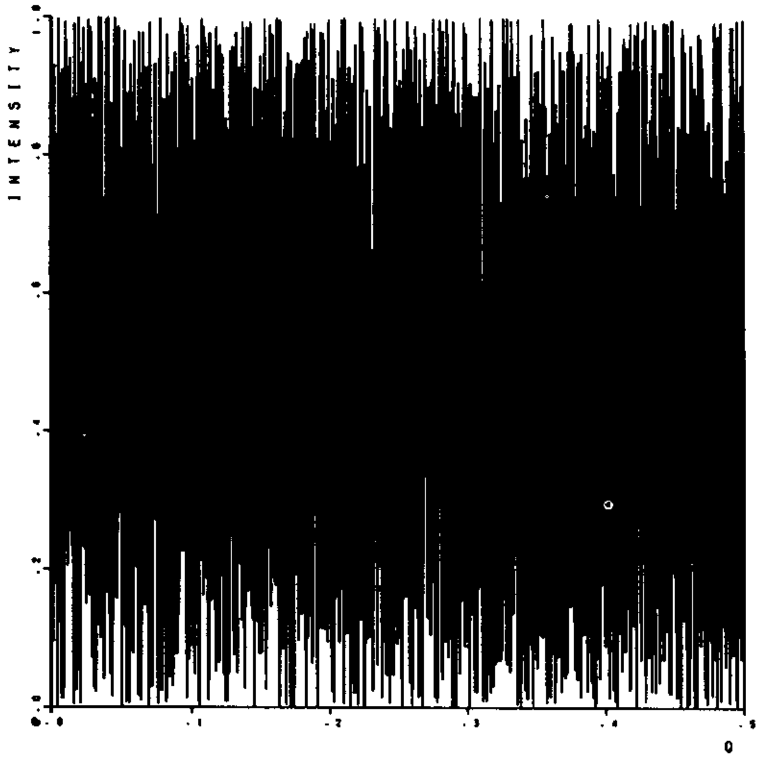
$$u_n = 2w_{n-1}$$
$$v_n = -2v_{n-1}$$
$$w_n = 2u_{n-1}$$

that is

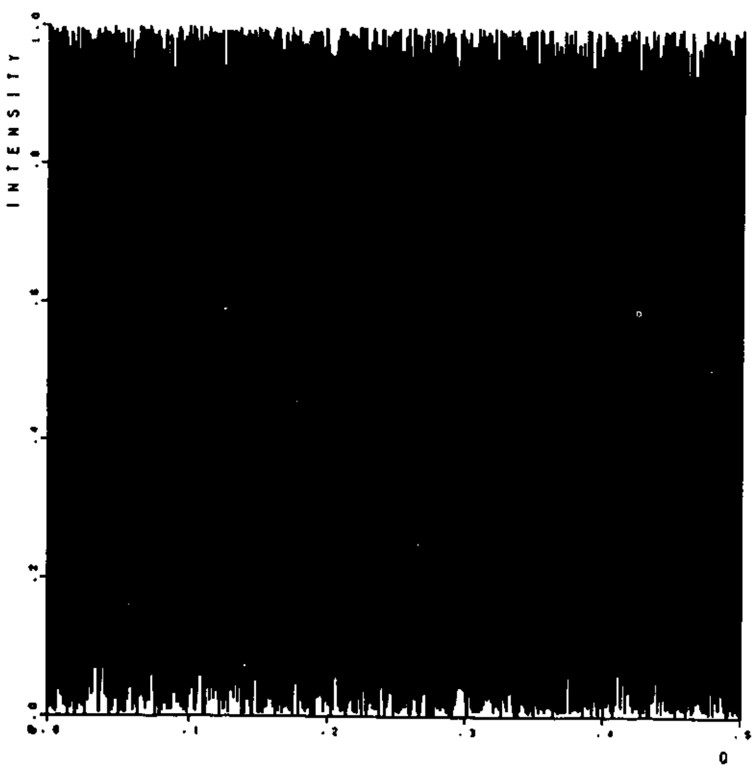
$$w_{n+1} = 4w_{n-1}$$

and the intensity then remains constant. However, this constant is *not* necessarily a maximum of the x-ray spectrum, where the peaks are not stable upon size changes.

(*a*)



(*b*)



(*c*)

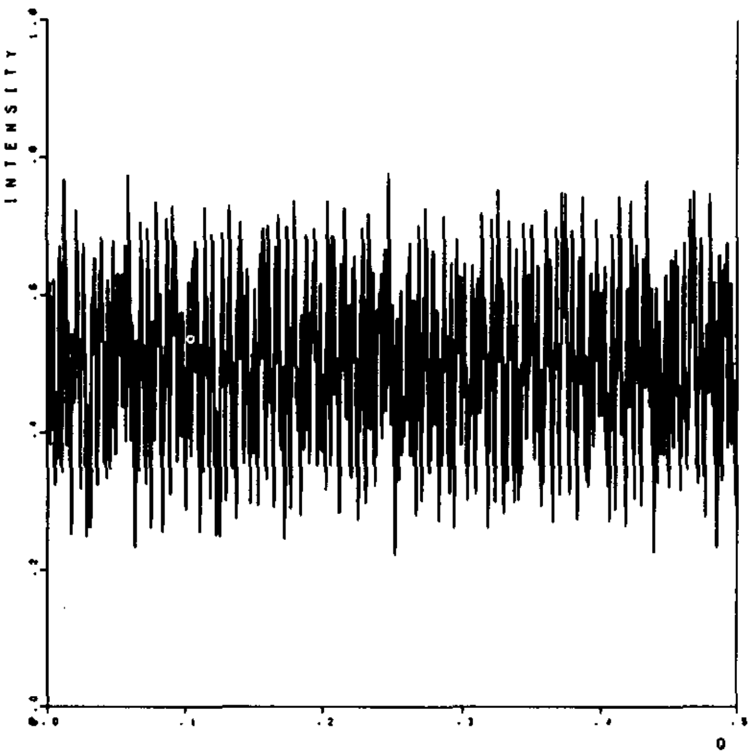


Figure 2. (*a*) The calculated intensity of the Fourier transform of the abstract Rudin–Shapiro sequence of length $2^{10} = 1024$ (see text). The number of points in q is 16884. The intensity is bounded and normalized. (*b*) The same as (*a*) with $2^{12} = 4096$. (*c*) The same length as (*a*) with a detector slit width of 64 in q ($\sim 4\%$).

5. Conclusion

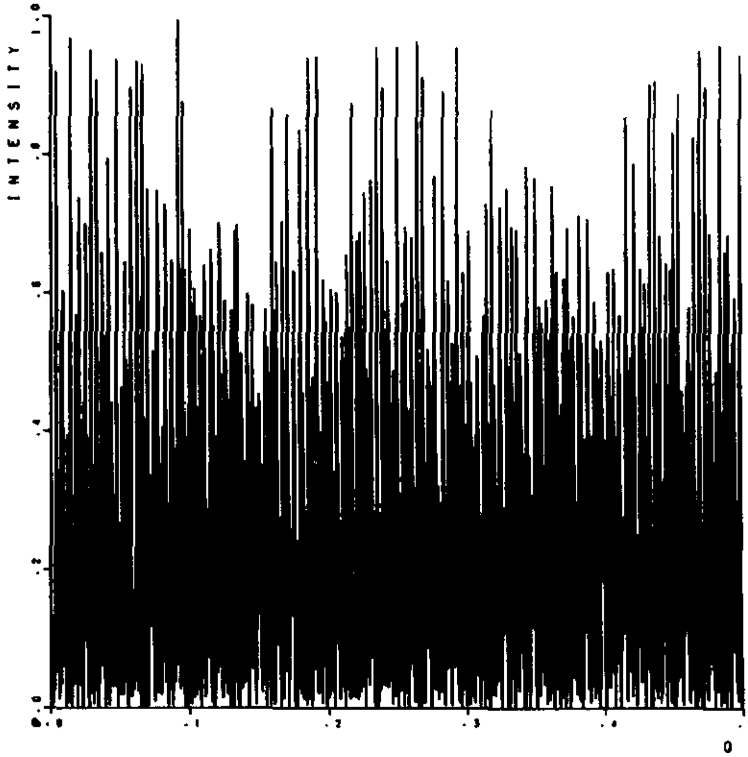
We studied theoretically and numerically the intensity of the Fourier transform of finite-size Rudin–Shapiro and generalized Rudin–Shapiro sequences, which is bounded in accordance with the '$\sqrt{N}$ property' of these sequences. Comparison with the Thue–Morse situation previously studied confirms that deterministic disorder generated by such sequences seems to share—at least from the measure-theoretic point of view—many properties of random disorder.

We are thus led to introduce a tentative classification for the x-ray diffraction spectra of these aperiodic deterministic finite-size multilayer heterostructures, based on the AT, SC, AC nature of the measure associated with the Fourier transform of the generating sequence, and the behaviour of the corresponding intensity:

(i) 'crystalline type' spectra would come from sequences having AT or SC associated measures. The peak positions are stable upon size change, they are well indexed by one or more integers (rationals) in convenient reduced units, their intensity is not bounded, and can be described by some scaling law;

(ii) 'random type' spectra would come from sequences having an AC associated

(*a*)



(*b*)

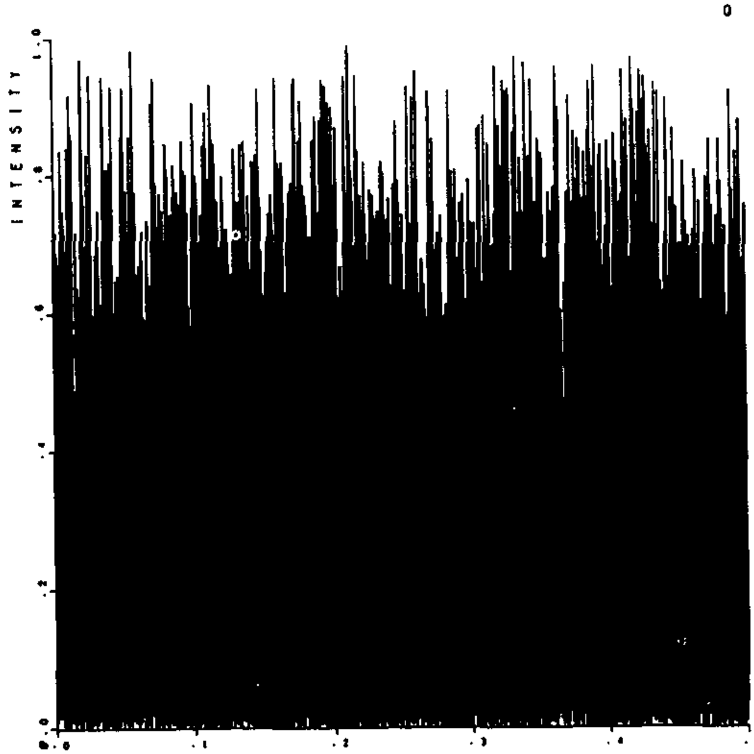


Figure 3. The calculated intensity of the Fourier transform of the generalized Rudin–Shapiro sequence defined in the text (equation (10)). The number of points in q is 16384. The intensity is bounded and normalized. The sequence length is $2^{10} = 1024$ (*a*) and $2^{12} = 4096$ (*b*).

measure with bounded intensities, random or deterministic as well. Peak positions show no stability upon size change and there is no simple description of intensity dependence on wavevector or size.

This novel classification, which relies in particular on measure–theoretic properties and is now defined only in 1D, could lay a basis for an extension of the concepts and methods of the crystallography of disordered systems.

Acknowledgments

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Note added in proof. Following the comment of our second referee, we recall the definition of the entropy S of an infinite sequence built on a two-letter alphabet:

$$S = \lim_{n\to\infty} \frac{1}{n} \log_2 p(n)$$

where $p(n)$ is the number of blocks of length n in the infinite sequence. Obviously

$$0 \leqslant p(n) \leqslant 2^n$$

and for automatic sequences (like the Thue–Morse and the Rudin–Shapiro sequences)

$$p(n) \leqslant Cn$$

with C a constant, hence

$$S = 0$$

in complete agreement with their completely deterministic character [24].

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