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Structure factor of 1D systems (superlattices) based on two-letter substitution rules: I. δ (Bragg) peaks

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Abstract. The recent generalization to the case of arbitrary tile lengths and arbitrary scattering factors of the calculation of the structure factor of 1D substitutional systems is studied in detail. This method makes it easy to find all the peaks in the diffraction spectrum of a system. The well known periodic and quasiperiodic spectra with δ peaks at integer multiples of a single number and integer linear combinations of two incommensurate frequencies, respectively, were found to be the $l = 0$ subsets of two more general types of spectra, infinite-periodic (or limit-periodic) and infinite-quasiperiodic (or limit-quasiperiodic) characterized by rational numbers of the type m/n^l, $l = 0, \ldots, \infty$ in place of the above integers. Substitution rules that produce quasicrystalline quasiperiodic and infinite-quasiperiodic spectra give the same type of spectrum for all values of the ratio $\rho = \rho_a/\rho_b$ of the two tile lengths ρ_a and ρ_b. This is not the case for the other rules. Thus the same substitution rule (such as the copper-mean rule) can give an infinite-periodic spectrum for a single rational ratio $\rho = \rho_a/\rho_b$ of the two tile lengths ρ_a and ρ_b, a periodic-like spectrum for other rational ρ, and a spectrum in some aspects similar to that of a random system when ρ is an irrational number. On the other hand, a Thue–Morse system diffracts as a periodic crystal when $\rho \neq 1$ but has no non-trivial δ peaks when $\rho = 1$. Other Thue–Morse-like systems can have infinite-periodic spectra for all ρ.

1. Introduction

The discovery by Penrose [1] of the existence of non-periodic tilings with long-range order, and the first experimental indication by Shechtman *et al* [2] that such quasicrystals may be obtained under certain conditions in nature, stimulated extensive theoretical investigation of the properties of 1D, 2D and higher-dimensional deterministic aperiodic structures that are now believed to populate rather densely the previously unexplored territory between the periodic and random lattices. Though at least 2D models are needed to explain the properties of 'natural' quasicrystals, the study of 1D structures, besides being useful for testing of methods that can usually be applied to arbitrary-dimensional systems, is especially important in the light of the recently developed experimental techniques to manufacture arbitrarily ordered layered structures [3, 5], and to analyse them in terms of high-resolution x-ray diffraction spectra [4].

Two general methods are used to generate deterministic aperiodic lattices of various dimensions: (i) projection from a higher-dimensional periodic lattice [6, 7] (or its various higher-dimensional generalizations [8]), and (ii) repetitive use of substitution (inflation) rules

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starting with a simple seed [9]. While the former method can give only quasicrystals (defined by dense aperiodic arrays of Bragg peaks in their diffraction patterns), the latter method can also produce many interesting aperiodic systems that are not quasicrystals (e.g. Thue–Morse superlattices). Conversely, there exist projectional quasicrystals that cannot be generated by a single substitution rule. When calculating the structure factor of a substitutional system, it is easy to consider different scattering factors of the different building blocks as can be seen in this paper. On the other hand, the projectional quasicrystals studied so far were obtained from higher-dimensional periodic arrays of identical scatterers, and it is not obvious how to take into account different scattering factors of the two or more elementary building blocks in the projection (cut-and-project) method.

Here we deal only with 1D substitutional systems generated by substitution rules on an alphabet of ν letters [9] representing ν elementary building blocks (tiles) of the system. *By definition, the structure factor (Fourier transform) of 1D quasicrystals has δ-function (Bragg) peaks the positions of which cannot be expressed as integral multiples of a single spatial frequency (wavevector).* The first attempt to answer the question of which of the 1D substitutional systems are quasicrystals was made by Bombieri and Taylor [10], who conjectured that a sufficient condition for such a system to be a quasicrystal is that the Perron–Frobenius eigenvalue λ_1 of the substitution matrix be a Pisot–Vijayaraghavan (PV) number. As the entries of a substitution matrix are all integers, this is equivalent to requiring that $|\lambda_1| > 1$ and all other $|\lambda_j| < 1$. Other authors [3, 5, 11–19] studied in more detail several special systems, mainly with special values of the tile-length ratio ρ. In all these studies either the lengths of different tiles or the corresponding scattering factors differed but not both. Most of these studies were based on individually derived recursion relations for the structure factor of finite approximants of a single infinite chain. A unifying scheme readily applicable to all substitutional systems was developed by Kolář [20] using the matrix formulation [17] of the recursion relations, some elements of which were already present in [13] and recently in [19]. In this scheme, one calculates simultaneously the structure factors for a whole group of related infinite chains—the canonical chains [21, 22] generated by the given substitution rule from all possible single-letter seeds. This results in the recursion relations for the structure factor having a simple and transparent matrix form. The simultaneous treatment of all canonical chains proved to be very advantageous also for the calculation of the spectral properties of these systems as expressed in terms of transfer-matrix-trace maps [21, 22]. As in the case of the transfer-matrix traces [21], the structure factor of an arbitrary chain (obtained from an arbitrary seed) can be expressed in terms of the structure factors of the canonical chains. Luck *et al* [23] recently also studied the structure factor of a large class of substitutional systems along similar lines. See also the work of Combescure on the quantum autocorrelation function of the Hamiltonian of the kicked rotator [24].

Cheng and Savit [17] formulated their scheme only for two-letter 1D substitutional systems with *both tiles of equal length*, and applied it only to a few substitution rules. A lot of confusion still remained, e.g. as to the interpretation of the Bombieri and Taylor results—perhaps related to the problem of a suitable definition of a quasicrystal (cf [10, 13, 14, 16, 17, 20]). Recently, Kolář [20] obtained the positions of the δ peaks (i.e. the diffraction spots) for all two-letter substitutional systems with *arbitrary* tile lengths ρ_a and ρ_b and *arbitrary* scattering factors $s_a(\omega)$ and $s_b(\omega)$ of the two tiles. Based on the arrangement of the δ peaks, all two-letter substitutional systems fall roughly in the following classes: (i) periodic systems with δ peaks at integral multiples of a single number, (ii) 'classical' quasicrystals with δ peaks at integral linear combinations of two incommensurate spatial frequencies, (iii) infinite-periodic systems with δ peaks at certain rational multiples of a single spatial frequency, (iv) infinite-quasiperiodic systems with δ peaks at certain rational linear combinations of two incommensurate spatial frequencies, (v) aperiodic systems with periodic-like non-trivial δ-peak positions, and (vi) aperiodic systems with no non-trivial δ peaks. The most interesting situation (infinite-periodic or periodic-like spectra) can be found among the systems with an integer Perron–Frobenius eigenvalue λ_1, or when $|\lambda_2| > 1$ and the ratio $\rho = \rho_a/\rho_b$ is rational. Thus the class of systems with $\lambda_2 = 0$ includes all two-letter periodic systems and many of the new infinite-periodic systems. It also contains aperiodic systems with periodic-like spectra, such as a Thue–Morse (TM) chain with unequal tile lengths. The periodic-like spectra are even more abundant for $|\lambda_2| > 1$ than was reported in [20] (see section 5.2). Nevertheless, periodic-like spectra for $|\lambda_2| > 1$ do not represent a generic case because they are not robust under perturbation on ρ.

For structures other than the classical quasicrystals and infinite-quasiperiodic systems (i.e. when $|\lambda_2| \geqslant 1$), the presence of the δ peaks in the structure factor is generally not uniquely given by the used substitution rule; it also depends on ρ: for a rational ρ, the spectrum is infinite-periodic or periodic-like, while for an irrational ρ, there are no δ peaks except a single trivial one. For example, a copper-mean system ($\lambda_2 = -1$) has an infinite-periodic spectrum for a single rational value of ρ, can diffract as a periodic crystal for other rational ρ, and diffracts essentially as a random system for all irrational ρ. In the case of the TM substitution rule (which has the PV property) the situation is opposite in a sense: except for a special choice of the 'atomic' scattering factors there is always a periodic array of δ peaks in its structure factor. (This special choice of the atomic scattering factors, apparently of little physical significance, was used in the previous studies [12, 14, 17] of the TM chains, which led to occasional claims that a TM chain does not have δ peaks at all.) If the two tiles have different lengths, the periodic set of δ-peak positions is exactly the same as an (existing) periodic system, while it contains no non-trivial δ peaks if the tiles are of the same lengths (i.e. for a *single* value of ρ). However, the sets of all peaks, including those with scaling exponent smaller than 2 (which is the value for the δ peaks) are very similar in the TM and copper-mean systems as will be shown in a sequel to this paper. Hopefully these results can shed some light into the ongoing discussion [25, 26] on whether these systems are more random or more periodic than quasicrystals.

Luck *et al* [23] also got the infinite-quasiperiodic spectrum (they call it limit-quasiperiodic using the term of Gähler who is, as far as we know, the first who pointed out the existence of this kind of spectrum). They used $s_a = s_b = 1$ and mostly a single irrational value of ρ, and studied only the rules with irrational λ_1. Thus they were outside the region where infinite-periodic spectra can be found. They concentrated on the relation between classical quasicrystals and certain periodic structures (atomic surfaces) in a 2D superspace.

The rest of the paper is organized as follows. In section 2 we discuss the importance of canonical chains. In section 3 we summerize the properties of substitutional sequences and 1D systems based on such sequences. In section 4 we give all the details of the procedure for the structure factor calculation for arbitrary tile lengths. In section 5 we present a detailed derivation of the positions of the δ peaks and discuss some specific examples. In section 6 we summarize our results, and give some suggestions for future experiments. Some technical details are given in two appendices.

2. Canonical chains

Let us first define the notation. $\mathcal{A}$ is a set (alphabet) of ν letters a_i, $i = 1, \ldots, \nu$. $\mathcal{A}^k$ is the set of all words over $\mathcal{A}$ of length k, and $\mathcal{A}^* = \cup_{k=0}^{\infty}\mathcal{A}^k$. The length of a word $w \in \mathcal{A}^*$ is $|w|$, and its letters are $w[i]$; $i = 0, \ldots, |w| - 1$. The empty word is ε: $|\varepsilon| = 0$, $\varepsilon \in \mathcal{A}^0$.

a^k denotes k adjacent letters a, and $a^0 = \varepsilon$. A ν-letter substitution rule ξ is a map from $\mathcal{A}$ into $\mathcal{A}^*$. It is often written out as $a_i \mapsto \xi(a_i)$; $i = 1, \dots, \nu$, where

$$\xi(a_i) = a_{\gamma(0,i)} a_{\gamma(1,i)} \cdots a_{\gamma(k_i-1,i)} \in \mathcal{A}^* \qquad k_i = |\xi(a_i)| . \tag{1}$$

The mapping ξ can be extended in a natural way to words over $\mathcal{A}^*$. $N_{\chi\phi L}$ is the number of occurrences of letter χ in the word $\xi^L(\phi)$.

In physical applications, the ν letters a_i are assumed to represent ν different elementary building blocks of a one-dimensional chain or layered structure [9]. Each ξ produces ν canonical substitutional sequences (or chains) [21, 22] $\xi^\infty(a_i) = \lim_{L\to\infty} \xi^L(a_i)$, such that $\xi^0(a_i) \equiv a_i$, and $\xi^L(a_i) = \xi(\xi^{L-1}(a_i))$; $i = 1, \dots, \nu$. Some or all of $\xi^\infty(a_i)$ can be identical. $\xi^L(a_i)$ are called the Lth generation *canonical chains*. By induction, one can prove that (1) leads to the inflation (juxtaposition) rule

$$\xi^{L+1}(a_i) = \xi^L(a_{\gamma(0,i)}) \cdots \xi^L(a_{\gamma(k_i-1,i)}) . \tag{2}$$

For some ξ, the limits above need not exist in the strict sense for some or even all i because the corresponding $\xi^L(a_i)$ oscillate between successive approximations to infinite words that are the points of a periodic orbit of ξ. The respective $\xi^\infty(a_i)$ then denote these points (in some order). In this sense, for example, $(a \mapsto bab,\ b \mapsto a)$ gives $\{\xi^\infty(a_i)\} \equiv \{ababab\dots, babab a\dots\}$.

Canonical chains are simply the chains that are obtained from the simplest possible seeds—the letters of $\mathcal{A}$. They are useful for expressing the properties of chains C_L, C_∞ obtained from an arbitrarily long seed C_0: $C_L = \xi^L(C_0)$. Evidently

$$C_L = \xi^L(C_0[0])\xi^L(C_0[1])\xi^L(C_0[2]) \cdots = \xi^L(a_{i_0})\xi^L(a_{i_1})\xi^L(a_{i_2}) \cdots \tag{3}$$

where i_j is the index of the jth letter of C_0: $C_0[j] = a_{i_j}$. In [21] it was shown (at least for two letters) that the spectral properties of C_L as expressed in terms of traces of certain transfer matrices are uniquely determined by the canonical chains $\xi^L(a_i)$.

The trace of the transfer matrix associated with C_L is simply given by a polynomial in the traces of the transfer matrices associated with $\xi^L(a_i)$. Thus the trace associated with C_L is simply a 'slave' of a certain trace map given uniquely by $\xi^L(a_i)$. This trace map is not at all influenced by the trace associated with C_L, and is the same for all C_L. In the formulation of Bovier and Ghez [27], the seed C_0 is the only additional (composite) letter that must be added to the original alphabet $\mathcal{A}$ to get the trace map determining the spectral properties of C_L. This 'letter' is passive in the sense that the associated trace never occurs on the right-hand side of the equations of the extended trace map, which is just the trace map for all $\xi^L(a_i)$ supplemented with one passive equation for the trace associated with C_L. This additional equation is just the polynomial mentioned above.

A similar situation exists in the case of the structure factor. We show in section 4 (equation (20)) that the structure factor associated with C_L is a linear combination of the structure factors associated with $\xi^L(a_i)$. Structure factors of all $\xi^L(a_i)$ are determined by a certain 'structure factor map' that gives the structure factors of $\xi^L(a_i)$ in terms of those of $\xi^{L-1}(a_i)$. Generally, all the structure factors of $\xi^{L-1}(a_i)$ are needed to determine the structure factor of each $\xi^L(a_i)$.

Here we study the structure factor of non-periodic structures (canonical chains) constructed by arranging in a line the minimum of two building blocks using a two-letter substitution rule. The generalization of the whole procedure to the case of more building blocks ($\nu > 2$) is straightforward.

3. Two-letter substitution rules

Denoting the two letters a and b, $\mathcal{A} = \{a, b\}$, and a general two-letter substitution rule can be written in the form

$$\xi(a) = \alpha_{pq}(a, b) \qquad \xi(b) = \beta_{rs}(a, b) \tag{4}$$

where $\alpha_{pq}(a, b)$ $[\beta_{rs}(a, b)]$ denotes a string of total length $p + q$ $[r + s]$ consisting of a certain permutation of p $[r]$ letters a and q $[s]$ letters b. The corresponding canonical chains are, in this case, $\xi^L(a)$ and $\xi^L(b)$, and (2) can be rewritten as

$$\xi^{L+1}(a) = \alpha_{pq}(\xi^L(a), \xi^L(b)) \qquad \xi^{L+1}(b) = \beta_{rs}(\xi^L(a), \xi^L(b)) \qquad L > 0. \tag{5}$$

The primary objects of our investigation are infinite non-periodic self-similar chains with long-range order. To eliminate the cases which deviate most from this category, we consider only the *primitive substitution rules*, which means that $(p + s)\, q\, r \neq 0$. Then both $\xi^\infty(a)$ and $\xi^\infty(b)$ are infinite. However, some of the primitive ξ still give periodic or almost periodic chains.

The same substitution matrix $\mathcal{S} = \binom{p,q}{r,s}$ is shared by $\binom{p+q}{p}\binom{r+s}{r}$ different rules of type (4). All such rules give the same numbers $N_{\chi\phi L}$:

$$\mathcal{S}^L = \begin{pmatrix} N_{aaL} & N_{baL} \\ N_{abL} & N_{bbL} \end{pmatrix} \qquad L \geqslant 0. \tag{6}$$

The same recursion formula

$$N_{\chi\phi L} = (p + s)\, N_{\chi\phi L-1} + (qr - ps)\, N_{\chi\phi L-2} \tag{7}$$

holds for all $\chi, \phi \in \mathcal{A}$, and also for $|\xi^L(\phi)| = N_{a\phi L} + N_{b\phi L}$. Denoting $p + s = m_{\text{eff}}$ and $qr - ps = n_{\text{eff}}$, equation (7) acquires the same form as the formula for the generalized Fibonacci numbers [28], which is a special case of (7) with $p = m$, $q = n$, $r = 1$ and $s = 0$. Unlike n of a generalized Fibonacci substitution $[\xi(a) = a^m b^n,\ \xi(b) = a]$, n_{eff} can also assume negative values. The roots of the characteristic equation of the difference equation (7) are real numbers equal to the eigenvalues of the substitution matrix $\mathcal{S}$, $\lambda_1 = \frac{1}{2}\left(m_{\text{eff}} + \sqrt{m_{\text{eff}}^2 + 4n_{\text{eff}}}\right)$, $\lambda_2 = \frac{1}{2}\left(m_{\text{eff}} - \sqrt{m_{\text{eff}}^2 + 4n_{\text{eff}}}\right)$. λ_1 is a PV number if $|\lambda_2| < 1$. λ_1 is also called the 'mean' of the corresponding rule. Equation (7) can be rewritten as

$$N_{\chi\phi L} - \lambda_1 N_{\chi\phi L-1} = \lambda_2 (N_{\chi\phi L-1} - \lambda_1 N_{\chi\phi L-2}) \qquad \chi, \phi \in \mathcal{A}. \tag{8}$$

Thus, as $L \to \infty$, $|N_{\chi\phi L} - \lambda_1 N_{\chi\phi L-1}|$ goes to zero if $|\lambda_2| < 1$ (when λ_1 is a PV number), is constant if $|\lambda_2| = 1$, and diverges if $|\lambda_2| > 1$.

For primitive substitutions we have (cf (14) below)

$$\lim_{L\to\infty} \frac{N_{\chi\phi L+1}}{N_{\chi\phi L}} = \lambda_1 \qquad \chi, \phi \in \mathcal{A} \tag{9}$$

and

$$\lim_{L\to\infty} \frac{N_{aaL}}{N_{baL}} = \lim_{L\to\infty} \frac{N_{abL}}{N_{bbL}} = \tau \tag{10}$$

where

$$\tau = \frac{\lambda_1 - s}{q} = \frac{r}{\lambda_1 - p} = \frac{p - \lambda_2}{q} = \frac{r}{s - \lambda_2} > 0 \tag{11}$$

is also called the 'incommensurability' of the rule. Equation (11) gives

$$q\tau^2 = (p - s)\tau + r\,. \tag{12}$$

The right and left eigenvectors of $\mathcal{S}$, $\mathcal{S}\, e_i^{(r)} = \lambda_i\, e_i^{(r)}$ and $e_i^{(l)}\, \mathcal{S} = \lambda_i\, e_i^{(l)}$, $i = 1, 2$, are

$$e_1^{(r)} = \begin{pmatrix} q\tau \\ r \end{pmatrix} \qquad e_2^{(r)} = \begin{pmatrix} 1 \\ -\tau \end{pmatrix} \qquad e_1^{(l)} = (\tau, 1) \qquad e_2^{(l)} = (r - q\tau)\,.$$

Applying $\mathcal{S}^L$ as given by (6) to these eigenvectors, one can derive explicit formulae for all $N_{\chi\phi L}$ and various relations between them. For example, using the left eigenvectors gives

$$\begin{aligned} &\tau\, N_{aaL} + N_{abL} = \tau\, \lambda_1^L \qquad && \tau\, N_{baL} + N_{bbL} = \lambda_1^L \\ &r\, N_{aaL} - q\tau\, N_{abL} = r\, \lambda_2^L \qquad && r\, N_{baL} - q\tau\, N_{bbL} = -q\tau\, \lambda_2^L\,. \end{aligned} \tag{13}$$

The first two of these formulae give, for example,

$$(|\xi^L(a)| - \lambda_1\, |\xi^{L-1}(a)|)\, \tau + |\xi^L(b)| - \lambda_1\, |\xi^{L-1}(b)| = 0$$

(alternatively, this can be proven by induction starting from $L = 1$ and using (8)). All four then give

$$\begin{aligned} &N_{aaL} = \frac{q\tau^2\, \lambda_1^L + r\, \lambda_2^L}{q\tau^2 + r} \qquad && N_{baL} = \frac{q\tau\, (\lambda_1^L - \lambda_2^L)}{q\tau^2 + r} \\ &N_{abL} = \frac{r\tau\, (\lambda_1^L - \lambda_2^L)}{q\tau^2 + r} \qquad && N_{bbL} = \frac{r\, \lambda_1^L + q\tau^2\, \lambda_2^L}{q\tau^2 + r} \qquad r\, N_{baL} = q\, N_{abL}\,. \end{aligned} \tag{14}$$

Similarly, $\mathcal{S}^L\, e_2^{(r)} = \lambda_2^L\, e_2^{(r)}$ gives

$$N_{aaL} - \tau\, N_{baL} = \lambda_2^L \qquad N_{abL} - \tau\, N_{bbL} = -\tau\, \lambda_2^L \tag{15}$$

or

$$\tau\, (N_{aaL} - \tau\, N_{baL}) + (N_{abL} - \tau\, N_{bbL}) = 0$$

and for $|\lambda_2| < 1$ both terms in this sum go to zero independently as $L \to \infty$.

Non-primitive substitutions can also be of interest, for instance, if $q = 0$, then $N_{baL} = 0$, $N_{abL} = r\, (s^L - p^L)/(s - p)$, and $\tau = \lim_{L\to\infty}(N_{abL}/N_{bbL}) = r/(s - p)$ if $p < s$ and ∞ otherwise. If $r = 0$, then $N_{abL} = 0$, $N_{baL} = q\, (s^L - p^L)/(s - p)$, and $\tau = \lim_{L\to\infty}(N_{aaL}/N_{bbL}) = (p - s)/q$ if $p > s$ and zero otherwise. In both these cases, $N_{aaL} = p^L$, $N_{bbL} = s^L$, $\lambda_1 = \max(p, s)$ and $\lambda_2 = \min(p, s)$.

For the study of diffraction a rather simple model of the two building blocks of the 1D physical chains based on $\xi^L(a)$ and $\xi^L(b)$ will suffice. The building block associated with the letter ϕ is fully characterized by a length (tile) ρ_ϕ and an 'atomic-like' scattering factor $s_\phi(\omega)$ (see figure 1). Let us denote by $\mathcal{L}_{aL}$ and $\mathcal{L}_{bL}$ the total lengths of such Lth generation physical chains $\xi^L(a)$ and $\xi^L(b)$, respectively. Then

$$\boldsymbol{L}_L = \begin{pmatrix} \mathcal{L}_{aL} \\ \mathcal{L}_{bL} \end{pmatrix} = \mathcal{S}\, \boldsymbol{L}_{L-1} = \mathcal{S}^L \begin{pmatrix} \rho_a \\ \rho_b \end{pmatrix} \tag{16}$$

and for all primitive substitutions, we have

$$\lim_{L\to\infty} \frac{\mathcal{L}_{aL+1}}{\mathcal{L}_{aL}} = \lim_{L\to\infty} \frac{\mathcal{L}_{bL+1}}{\mathcal{L}_{bL}} = \lambda_1\,. \tag{17}$$

This means that both $\mathcal{L}_{aL}$ and $\mathcal{L}_{bL}$ scale with L as ${\lambda_1}^L$.

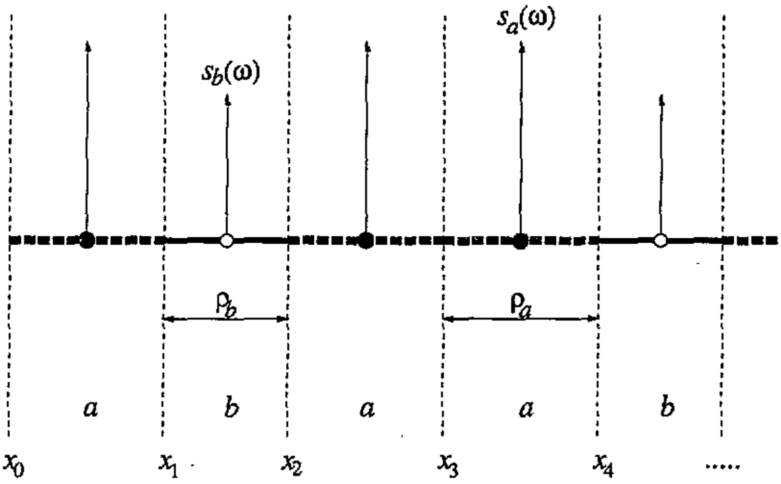

Figure 1. The two building blocks a and b are characterized by tiles of length ρ_a and ρ_b (e.g. in a superlattice equal to the thicknesses of two different layers the boundaries of which are indicated by thin dotted lines) and by wavevector dependent scattering factors $s_a(\omega)$ and $s_b(\omega)$, representing continuous or discrete distribution of suitable scatterers along the tiles.

4. Structure factor

The diffraction spectrum of a certain target is determined by the *Fourier transform* of the density of scatterers of the radiation used to probe the target, e.g. the electron density in the case of X-rays. This Fourier transform is usually called the *structure factor* of the target. The structure factor (or its x-component along the direction of the chains) of the two physical canonical chains $\xi^L(a)$ and $\xi^L(b)$ (linear arrays built according to ξ using two different building blocks as indicated in figure 1) is evidently

$$F_{\chi L}(\omega) = \sum_{n=0}^{|\xi^L(\chi)|-1} s_{\xi^L(\chi)[n]}(\omega)\, \mathrm{e}^{2\mathrm{i}\pi\omega x_n(\xi^L(\chi))} \qquad \chi \in \mathcal{A}. \tag{18}$$

Here $x_0(\xi^L(\chi)) = 0$, $x_{n+1}(\xi^L(\chi)) = x_n(\xi^L(\chi)) + \rho_{\xi^L(\chi)[n]}$ (cf figure 1), ω is equal to the x-component of the difference of the wavevectors of the scattered and incident waves,

$$s_\phi(\omega) = \int_0^{\rho_\phi} \sigma_\phi(x)\, \mathrm{e}^{2\mathrm{i}\pi\omega x}\, \mathrm{d}x$$

is the scattering factor (referred to the beginning of the block) of the building block ϕ, and $\sigma_\phi(x)$ is the density of the respective scatterers inside this block. For example, if $\sigma_\phi(x) = \sigma_\phi\, \delta(x - \rho_\phi/2)$ (a single scatterer at the centre of the tile), $s_\phi(\omega) = \sigma_\phi\, \mathrm{e}^{\mathrm{i}\pi\omega\rho_\phi}$. If $\sigma_\phi(x) = \text{const}$, then $s_\phi(\omega) = \sigma_\phi\, \mathrm{e}^{\mathrm{i}\pi\omega\rho_\phi} \sin(\pi\omega\rho_\phi)/\pi\omega$.

The structure factor of $\xi^\infty(\chi)$ can be obtained as the limit of the structure factors of the finite approximants $\xi^L(\chi)$: $F_{\chi\infty}(\omega) = \lim_{L\to\infty} F_{\chi L}(\omega)$. We then say that there is an $\alpha_\chi(\omega)$-peak in the structure factor of the infinite chain at an ω, if there exists a positive number $\alpha_\chi(\omega)$ such that $|F_{\chi L}(\omega)|^2$ scales as ${\mathcal{L}_{\chi L}}^{\alpha_\chi(\omega)}$. Then

$$\alpha_\chi(\omega) = 2 \lim_{L\to\infty} \frac{\ln|F_{\chi L}(\omega)|}{\ln \mathcal{L}_{\chi L}}.$$

A 2-peak is called the δ peak as it gives the Dirac δ-function in $|F_{\chi\infty}(\omega)|^2$. The intensity of an $\alpha_\chi(\omega)$-peak is defined as

$$I_\chi(\omega) = \lim_{L\to\infty} \frac{|F_{\chi L}(\omega)|^2}{\mathcal{L}_{\chi L}{}^{\alpha_\chi(\omega)}}. \tag{19}$$

Note that $\alpha_\chi(\omega) \leqslant 2$ for both $\chi \in \mathcal{A}$ because

$$\begin{aligned} |F_{\chi L}(\omega)| &\leqslant \sum_{n=0}^{|\xi^L(\chi)|-1} |s_{\xi^L(\chi)[n]}(\omega)| \\ &\leqslant \max\left(\frac{|s_a(\omega)|}{\rho_a}, \frac{|s_b(\omega)|}{\rho_b}\right) \sum_{n=0}^{|\xi^L(\chi)|-1} \rho_{\xi^L(\chi)[n]} = \max\left(\frac{|s_a(\omega)|}{\rho_a}, \frac{|s_b(\omega)|}{\rho_b}\right) \mathcal{L}_{\chi L}. \end{aligned}$$

Consequently, $I_\chi(\omega) \leqslant \max(|s_a(\omega)|/\rho_a, |s_b(\omega)|/\rho_b)$ when $\alpha_\chi(\omega) = 2$.

Evidently, the *structure factor* $\Phi_{C_0L}(\omega)$ of an arbitrary chain C_L of (3) can be written in terms of the structure factors of the canonical chains as

$$\Phi_{C_0L}(\omega) = \sum_{j=0}^{|C_0|-1} F_{C_0[j]L}(\omega) \exp\left(2i\pi\omega \sum_{k=0}^{j-1} \mathcal{L}_{C_0[k]L}\right) \tag{20}$$

where by convention, $\sum_{k=0}^{-1}$ gives a zero. (A similar formula holds for multi-letter rules.)

As in [17, 20], it is convenient to consider the two Fourier amplitudes $F_{aL}(\omega)$ and $F_{bL}(\omega)$ of (18) as the components of a column vector $\boldsymbol{F}_L(\omega)$. This makes it possible to write the recursion relation for the Fourier amplitudes as

$$\boldsymbol{F}_{L+1}(\omega) = M_L(\omega)\, \boldsymbol{F}_L(\omega) \tag{21}$$

where the 2×2 matrix $M_L(\omega)$ has components

$$[M_L(\omega)]_{\chi\phi} = \sum_{j=0}^{|\xi(\chi)|-1} \delta_{\xi(\chi)[j],\phi} \exp\left(2i\pi\omega \sum_{k=0}^{j-1} \mathcal{L}_{\xi(\chi)[k]L}\right) \qquad \chi, \phi \in \mathcal{A}. \tag{22}$$

This is a consequence of (20) as $F_{\chi\, L+1}(\omega) \equiv \Phi_{\xi(\chi)L}(\omega)$. Because of the Kronecker delta, the first sum in (22) runs only over those values of j for which the jth letter in the word $\xi(\chi)$ is the letter ϕ. For example, for the copper-mean rule, $\xi(a) = ab^2$, $\xi(b) = a$ [30, 31],

$$M_L(\omega) = \begin{pmatrix} 1 & e^{2i\pi\omega\mathcal{L}_{aL}}(1 + e^{2i\pi\omega\mathcal{L}_{bL}}) \\ 1 & 0 \end{pmatrix}. \tag{23}$$

As $F_{a0}(\omega) = s_a(\omega)$ and $F_{b0}(\omega) = s_b(\omega)$, (21) gives

$$\boldsymbol{F}_L(\omega) = \boldsymbol{T}_L(\omega)\, \boldsymbol{F}_0(\omega) \qquad \boldsymbol{F}_0(\omega) = \begin{pmatrix} s_a(\omega) \\ s_b(\omega) \end{pmatrix} \qquad \boldsymbol{T}_L(\omega) = \left[\prod_{l=L-1}^{0} M_l(\omega)\right]. \tag{24}$$

Assume that for a certain ω, $M_L(\omega)$ is independent of L. Let $\lambda_{\min}$ and $\lambda_{\max}$, $|\lambda_{\min}| \leqslant |\lambda_{\max}|$, be the two eigenvalues of this constant matrix, and $e_{\min}$ and $e_{\max}$ the corresponding right eigenvectors. One can express $\boldsymbol{F}_0(\omega)$ as

$$\boldsymbol{F}_0(\omega) = c_{\max}\, e_{\max} + c_{\min}\, e_{\min} \tag{25}$$

where $c_{\max}$ and $c_{\min}$ are some constants. From (24) one has

$$F_L(\omega) = \lambda_{\max}^L \, c_{\max} \, e_{\max} + \lambda_{\min}^L \, c_{\min} \, e_{\min} \,. \tag{26}$$

Assume now that for this value of ω, $|F_{\chi L}(\omega)|^2$ scale as $\mathcal{L}_{\chi L}{}^{\alpha_\chi(\omega)}$, i.e. as $\lambda_1{}^{\alpha_\chi(\omega)L}$. Comparing this assumption with (26) gives

$$\alpha_\chi(\omega) = 2\,\frac{\ln|\lambda_{\max}|}{\ln\lambda_1} \qquad \text{if} \qquad (e_{\max})_\chi \neq 0 \,. \tag{27}$$

This proves that at each ω for which $M_L(\omega)$ is independent of L, there is an $\alpha_\chi(\omega)$-peak with $\alpha_\chi(\omega)$ given by (27). If $c_{\max} = 0$ or $(e_{\max})_\chi = 0$, then (26) reduces to

$$F_{\chi L}(\omega) = \lambda_{\min}^L \, c_{\min} \, (e_{\min})_\chi$$

and $\lambda_{\max}$ in (27) must be substituted by $\lambda_{\min}$. For instance, for $(a \mapsto ab^2,\ b \mapsto bab)$ and $\rho_a = \rho_b = 1/(2\omega)$, $(e_{\max})_a = 0$. $c_{\max} = 0$ can occur only for a single value of $s_a(\omega)/s_b(\omega)$ (cf (24)). Provided that one can somehow realize physically this value of $s_a(\omega)/s_b(\omega)$, it would be possible to 'switch off' some δ peaks as is discussed in the next section.

Similarly, $M_L(\omega)$, periodic in L with period k also gives a peak with

$$\alpha_\chi(\omega) = \frac{2}{k}\,\frac{\ln|\lambda_{\max}|}{\ln\lambda_1} \qquad \text{if} \qquad (e_{\max})_\chi \neq 0 \tag{28}$$

where $\lambda_{\max}$ is now the larger-modulus eigenvalue of the matrix $\prod \lim_{l=k-1}^{0} M_l(\omega)$. If $c_{\max} = 0$ (for a certain single value of $s_a(\omega)/s_b(\omega)$), or $(e_{\max})_\chi = 0$, $\lambda_{\max}$ must again be replaced by the corresponding $\lambda_{\min}$.

As can be seen from (22), $M_L(\omega)$ has the same form for all L, so that one can write

$$M_L(\omega) = M(\boldsymbol{\Omega}_L) \qquad \text{where} \qquad \boldsymbol{\Omega}_L = \omega\, \boldsymbol{L}_L \,.$$

$M(\boldsymbol{\Omega})$ actually depends only on $\boldsymbol{\Omega}$ (mod1) (here mod1 is applied in each component of $\boldsymbol{\Omega}$). A constant matrix $M_L(\omega)$ corresponds to a 'fixed point modulo 1' $\boldsymbol{\Omega}_{L+1} \equiv \boldsymbol{\Omega}_L \pmod 1$ of the 2D map

$$\boldsymbol{\Omega}_{L+1} = \mathcal{S}\,\boldsymbol{\Omega}_L \pmod 1 \tag{29}$$

(see (16)). Also, for all values of ω for which $\boldsymbol{\Omega}_L$ eventually (for large L) ends up in this fixed point, (27) holds, and the same values of $\alpha_\chi(\omega)$ are obtained. A 'period k orbit modulo 1' $\boldsymbol{\Omega}_{L+k} \equiv \boldsymbol{\Omega}_L$ (mod1) of the map (29) gives a periodic sequence of $M_L(\omega)$, and thus a peak according to (28). Also, all the points $\boldsymbol{\Omega}_0$ (corresponding to different values of ω) that are ultimately attracted to this periodic orbit, give a peak with the same value of $\alpha_\chi(\omega)$. Thus, a single fixed point or periodic orbit modulo 1 of the map (29) may be responsible for peaks occurring at a generally infinite number of ω values having the same scaling exponent $\alpha_\chi(\omega)$.

Of primary importance are the δ peaks that correspond to the largest possible value of $\alpha_\chi(\omega) = 2$, and thus dominate the spectrum. To obtain this discrete part of the Fourier intensity measure of infinite systems, which is exactly given by such δ peaks, the following implication is useful (see appendix A for the proof):

$$\alpha_\chi(\omega) = 2 \qquad \text{for some} \qquad \chi \in \mathcal{A} \Longrightarrow \lim_{L\to\infty} \Omega_L = \boldsymbol{0} \pmod 1 \,. \tag{30}$$

Therefore, $\alpha_\chi(\omega) = 2$ can be achieved only through all the predecessors of $\mathbf{0}\,(\mathrm{mod}\,1)$, which is a fixed point modulo 1 of map (29). To obtain all the δ peaks, it is sufficient to find all the values of ω for which $\boldsymbol{\Omega}_L = \mathcal{S}^L\boldsymbol{\Omega}_0$ ends up in $\mathbf{0}\,(\mathrm{mod}\,1)$. Note that *this map is the same for all substitution rules with the same* $\mathcal{S}$, *for arbitrary tile lengths, and for arbitrary scattering factors* $s_\chi(\omega)$. Tile lengths appear only in the initial condition for $\boldsymbol{\Omega}_0$. Therefore, finding out all the points $\boldsymbol{\Omega}$ that are eventually attracted to $\mathbf{0}\,(\mathrm{mod}\,1)$ gives a superset of the sets that support the discrete part of the Fourier intensity measure for all substitutional systems with the same $\mathcal{S}$ and all possible tile lengths. As shown in the next section, this superset consists of the stable manifold (if there is one) of the $\mathbf{0}\,(\mathrm{mod}\,1)$ and of the direct predecessors of $\mathbf{0}\,(\mathrm{mod}\,1)$, and of the direct predecessors of all points in the stable manifold. For the given values of ρ_a and ρ_b, one has to select from this superset the subset of initial conditions that match $\boldsymbol{\Omega}_0$ for some value of ω. Finally, one has to verify for the given rule that for such a value of ω the matrix product in (24) does not vanish as a result of the orthogonality of some of its terms. This product then gives directly the intensity of the δ peak at ω. In summary, the type of the diffraction spectrum is generally determined by the substitution matrix (except for all integer λ_2), tile lengths determine the exact positions of various peaks, and the $s_\chi(\omega)$s can only influence the relative intensities of the peaks.

5. δ Peaks ($\alpha_\chi(\omega) = 2$)

In this section, we give a detailed derivation of the δ peaks for different classes of primitive two-letter substitution rules and for arbitrary tile lengths. Let us first establish the set of *trivial δ peaks*. These are the peaks that are present in every 1D system, including a random chain, with the given tile lengths ρ_a and ρ_b. They are given directly by all the points with integer coordinates in the $\boldsymbol{\Omega}$ space: $\boldsymbol{\Omega}_0 = \mathbf{0}\,(\mathrm{mod}\,1)$. For arbitrary ρ_a and ρ_b, there is always at least one such peak corresponding to $\omega = 0$. If $\rho = \rho_a/\rho_b$ is rational, there are infinitely many trivial peaks at $\omega_m = m/\rho_a$ for all such integer m for which there is an integer $n = m/\rho$. Their intensity depends on m only through $s_\chi(\omega)$. For example, for the $\xi^L(a)$ chain one gets, from (24) and (19),

$$I_a(\omega_m) = |\tau\, s_a(\omega_m) + s_b(\omega_m)|^2 \,/\, (\tau\rho_a + \rho_b)^2\,. \tag{31}$$

For a random chain, τ in this equation is equal to the limit value of N_a/N_b in agreement with (10).

If it is possible to choose $s_b(\omega_m) = -\tau\, s_a(\omega_m)$ in (31), one can switch off at least some of the trivial δ peaks. This corresponds to replacing $\lambda_{\max}$ in (27) by $\lambda_{\min}$. In [12, 14] this was achieved for the TM chains with $\rho_a = \rho_b$ by choosing $s_a(\omega) \equiv -s_b(\omega) \equiv 1$. Only then was it possible to claim that the structure factor of a TM chain does not contain any δ peaks, that it is purely singular continuous. The question remains whether such a special choice of ω-independent $s_\chi(\omega)$ has any physical meaning. Except for this very special situation, trivial peaks are present for any rational ρ. They exist whether λ_1 is a PV number or not.

Now let us turn our attention to the non-trivial peaks that are of actual importance for the classification of different structures. We want to find all solutions of (30). The trivial ones corresponding to the direct predecessors of $\mathbf{0}$ (mod 1) are given by

$$\boldsymbol{\Omega}_l = \mathcal{S}^l\,\boldsymbol{\Omega}_0 = \mathbf{0}\ \ (\mathrm{mod}\ 1)\,. \tag{32}$$

If $\det\mathcal{S} \neq 0$, which means $\lambda_2 \neq 0$, define

$$\begin{pmatrix}\mu(m,n,l)\\ \nu(m,n,l)\end{pmatrix} = (\det\mathcal{S})^l\,\mathcal{S}^{-l}\begin{pmatrix}m\\ n\end{pmatrix} \tag{33}$$

where m, n are arbitrary integers. From (6),

$$\begin{pmatrix}\mu(m,n,l)\\ \nu(m,n,l)\end{pmatrix} = \begin{pmatrix}mN_{bbl} - nN_{bal}\\ nN_{aal} - mN_{abl}\end{pmatrix}. \tag{34}$$

We claim that for all integers l,

$\boldsymbol{\Omega}_l = \mathbf{0}\,(\mathrm{mod}\,1) \iff \exists$ integer m, n such that

$$\omega = \frac{\mu(m,n,l)}{\rho_a(ps-qr)^l} \qquad \text{and} \qquad \rho = \frac{\rho_a}{\rho_b} = \frac{\mu(m,n,l)}{\nu(m,n,l)}. \tag{35}$$

The proof is evident. Note that all trivial δ peaks correspond to $l = 0$ in (35).

The remaining solutions of (30) can be obtained as follows. Let $\boldsymbol{L}_0 = \binom{\rho_a}{\rho_b} = c_1 e_1^{(r)} + c_2 e_2^{(r)}$, and

$$\boldsymbol{\Omega}_l = \mathcal{S}^l \boldsymbol{\Omega}_0 = \begin{pmatrix} d_1\lambda_1^l + d_2\lambda_2^l \\ e_1\lambda_1^l + e_2\lambda_2^l \end{pmatrix}. \tag{36}$$

Thus

$$\begin{gathered} c_1 = \frac{\tau\rho_a + \rho_b}{\tau(\lambda_1 - \lambda_2)} \qquad c_2 = \frac{r\rho_a - q\tau\rho_b}{\tau(\lambda_1 - \lambda_2)} \qquad d_1 = \omega q \tau c_1 \\ d_2 = \omega c_2 \qquad e_1 = \omega r c_1 \qquad e_2 = -\omega\tau c_2 . \end{gathered} \tag{37}$$

By (30), we have to find the values of these coefficients that give $\boldsymbol{\Omega}_l \to \mathbf{0}$ (mod 1). Since this means that both components of $\boldsymbol{\Omega}_l$ go to 0 (mod 1), we can use the following result. If d_1 and d_2 are real numbers, the two statements

(i) $d_1\lambda_1^l + d_2\lambda_2^l \longrightarrow 0(\mathrm{mod}\ 1)$

(ii) (1) if $0 < |\lambda_2| < 1$ then $\exists\, L \geqslant 0, m, n$ such that $d_1 = \dfrac{m + n\lambda_2}{\lambda_1^L(\lambda_1 - \lambda_2)}$ (38)

(2) if $|\lambda_2| \geqslant 1$ then $\exists\, L \geqslant 0$ such that $d_1\lambda_1^l + d_2\lambda_2^l = 0$ (mod 1) $\forall l \geqslant L$

(3) if $|\lambda_2| = 0$ then $\exists\, L \geqslant 0$ such that $d_1\lambda_1^l = 0$ (mod 1) $\forall l \geqslant L$

are equivalent. For proof, see appendix B.

5.1. Substitution rules with $0 < |\lambda_2| < 1$

This corresponds to $0 < n_{\mathrm{eff}} < 1 + m_{\mathrm{eff}}$, and if $m_{\mathrm{eff}} > 2$ also to $1 - m_{\mathrm{eff}} < n_{\mathrm{eff}} < 0$. We decompose (36) as follows

$$\boldsymbol{\Omega}_l = \begin{pmatrix} m_l \\ n_l \end{pmatrix} + \begin{pmatrix} \epsilon_l \\ \epsilon'_l \end{pmatrix}$$

where m_l, n_l are integers and ϵ_l, ϵ'_l tend to 0 as $l \to \infty$. Thus there exists an integer L such that

$$\begin{pmatrix} m_{l+1} \\ n_{l+1} \end{pmatrix} = \mathcal{S} \begin{pmatrix} m_l \\ n_l \end{pmatrix} \qquad \forall l \geqslant L .$$

Using (B4)

$$d_1 = \frac{m_{L+1} - m_L \lambda_2}{\lambda_1^L (\lambda_1 - \lambda_2)} = \frac{(p - \lambda_2) m_L + q n_L}{\lambda_1^L (\lambda_1 - \lambda_2)} = \frac{q (\tau m_L + n_L)}{\lambda_1^L (\lambda_1 - \lambda_2)}.$$

From (37),

$$\omega = \frac{1}{\lambda_1^L} \frac{\tau m_L + n_L}{\tau \rho_a + \rho_b}.$$

Thus every δ-peak position must satisfy this formula. One can easily verify that all

$$\omega = \frac{1}{\lambda_1^l} \frac{\tau \overline{m} + \overline{n}}{\tau \rho_a + \rho_b} \tag{39}$$

with arbitrary $l \geqslant 0, \overline{m}, \overline{n}$ give d_1 and e_1 satisfying (38)(ii), and thus represent the complete set of δ-peak positions.

Using (12), the equation

$$(\det \mathcal{S}) \begin{pmatrix} \overline{m} \\ \overline{n} \end{pmatrix} = \mathcal{S} \begin{pmatrix} m \\ n \end{pmatrix}$$

which always has integer solution for m, n, can be transformed into $(\det \mathcal{S})\,(\overline{m}\,\tau + \overline{n}) = (q\,\tau + s)(m\,\tau + n)$. Using this relation repeatedly l times, the set (39) can be shown to be equivalent to the set of

$$\omega_{lmn} = \frac{1}{(\det \mathcal{S})^l} \frac{\tau\, m + n}{\tau \rho_a + \rho_b} \tag{40}$$

with arbitrary $l \geqslant 0, m, n$. For a rational ρ, this set includes automatically all the peak positions given by (35).

The Fourier module given by (40) is a Z-module which, a priori, is infinite-dimensional. However, it is of rank 2 when $\det \mathcal{S} = \pm 1$, when it evidently reduces to the set

$$\omega_{mn} = \frac{m\tau + n}{\rho_a \tau + \rho_b}. \tag{41}$$

For $0 < |\lambda_2| < 1$, τ is always irrational, thus, according to (41), all the substitutional systems with $\det \mathcal{S} = \pm 1$ and arbitrary ρ_a and ρ_b are classical quasicrystals with δ-peak positions given by two incommensurate frequencies $1/(\rho_a \tau + \rho_b)$ and $\tau/(\rho_a \tau + \rho_b)$.

When $\det \mathcal{S} \neq \pm 1$, the more general set (40) has been tentatively called 'infinite-quasiperiodic' [20] or 'limit-quasiperiodic' [23]. It can be looked upon as the superposition of infinitely many quasiperiodic spectra (41) scaled by rational factors $(\det \mathcal{S})^{-l}$.

Note that

$$\boldsymbol{\Omega}_{(0mn),0} \equiv \begin{pmatrix} \rho_a\, \omega_{0mn} \\ \rho_b\, \omega_{0mn} \end{pmatrix} \equiv \begin{pmatrix} m \\ n \end{pmatrix} + \frac{n\,\rho_a - m\,\rho_b}{\rho_a\,\tau + \rho_b}\, e_2^{(r)}. \tag{42}$$

All the points $\boldsymbol{\Omega}_{(0mn),0}$ of (42) lie on a single straight line with the slope ρ_b/ρ_a, going through the origin: *they are intercepts of this line with all the lines along the direction of* $e_2^{(r)}$ passing through an integer grid point. This set of parallel lines constitutes the stable manifold of the fixed point $\mathbf{0}$ (mod 1) of the map (29). Thus the $l = 0$ subset of the set (40) of ω_{lmn} corresponds to all $\boldsymbol{\Omega}_0$ lying in this disconnected stable manifold for the given

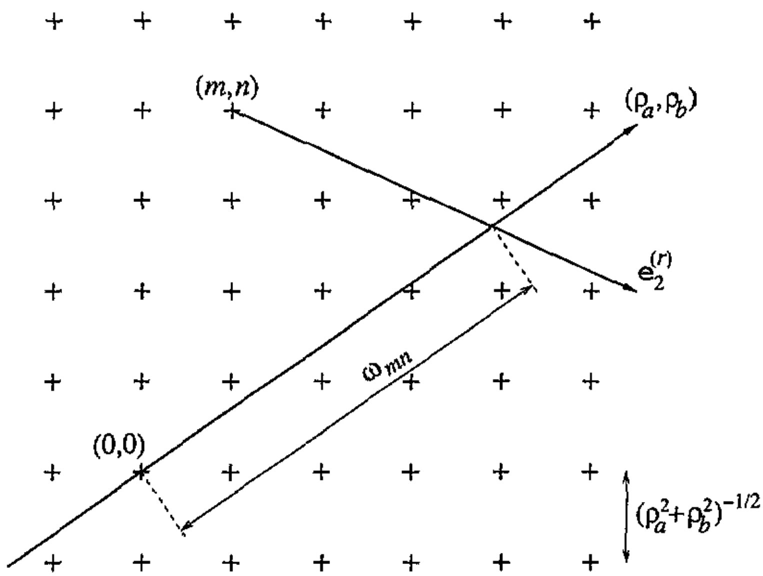

Figure 2. Classical quasicrystals ($0 < |\lambda_2| < 1$): the positions ω_{mn} of all δ peaks can be obtained by projection in the direction $e_2^{(r)} = (1, -\tau)$ of *all* points of a square lattice with the lattice constant $(\rho_a^2 + \rho_b^2)^{-\frac{1}{2}}$, onto the line with the slope ρ_b/ρ_a going through the origin.

ratio ρ. Similarly, one can show that all $\Omega_{(lmn),0}$ that correspond to all ω_{lmn} of (40) for an $l > 0$, are the lth generation direct predecessors of points in this stable manifold, i.e. $\mathcal{S}^l\,\Omega_{(lmn),0}$ are in the stable manifold. When $\det \mathcal{S} = \pm 1$, the map (29) is invertible, and each Ω point has only one distinct (mod 1) direct predecessor (i.e. all its 1st generation direct predecessors are equivalent mod 1). For a point in the stable manifold this means that its only distinct predecessor must again be in the stable manifold, and this corresponds to the reduction of the set (40) to (41).

The distance of $\Omega_{(0mn),0}$ from the origin is equal to $\omega_{mn}\,(\rho_a^2 + \rho_b^2)^{\frac{1}{2}}$. After scaling the whole Ω space by the factor $(\rho_a^2 + \rho_b^2)^{-\frac{1}{2}}$, the positions ω_{mn} of all δ peaks can be obtained directly by a non-orthogonal projection in the reciprocal superspace as depicted in figure 2. Modifying somewhat the standard direct-space projection method [7], the same positions of δ peaks as in (41) (or in the $l = 0$ subset of (40)) are obtained for all 1D quasicrystals generated by projecting orthogonally all the points of the 2D square lattice with lattice constant $(\rho_a^2 + \rho_b^2)^{\frac{1}{2}}$ contained in a strip of arbitrary width w about a line with the slope τ, onto another line with the slope ρ_a/ρ_b [20]. Note that the square lattices used in the two projections are mutually reciprocal. Also the slopes of lines involved in the two projections are mutually reciprocal or of opposite sign. Unlike the direct-space projection, the reciprocal-space projection of figure 2 is not orthogonal, and the source points are not contained within a strip of finite width. (Apparently, the same result in the direct space could also be obtained by projecting a rectangular lattice instead of the square one on the line with slope τ [3]).

Note also that all the Lth iterates $\Omega_{(0mn),L} = \mathcal{S}^L\,\Omega_{(0mn),0}$ lie on a single straight line with slope k_L passing through the origin:

$$k_{L+1} = \frac{r + s k_L}{p + q k_L} \qquad k_0 = \frac{\rho_b}{\rho_a}.$$

Also, all the points $\mathcal{S}^{L-l}\,\Omega_{(lmn),0}$, $L \geqslant l$ lie on the same line.

The values ω_{mn} of (41) are in agreement with the previous results for the golden-mean systems with $\tau = (1+\sqrt{5})/2$ [3, 16]. For $\rho_a = \rho_b = 1$, $\omega_{mn} = (m\tau + n)/(\tau+1) = \overline{m}\tau + \overline{n}$ [29]. For $\rho_a = \tau$, $\rho_b = 1$, $\omega_{mn} = (m\tau + n)/(\tau^2+1) = (\overline{m}\tau + \overline{n})/\sqrt{5}$ [11, 10].

5.2. Substitution rules with $|\lambda_2| > 1$

In this case, according to (38), δ-peaks can only correspond to the solutions of (32) which are given by (35). Evidently, non-trivial δ peaks exist only for rational ρ. For a given ρ (if not mentioned otherwise, in this subsection we will implicitly assume that ρ is rational) we define integers μ_0, ν_0 such that $\rho = \mu_0/\nu_0$ and $\gcd(\mu_0, \nu_0) = 1$. We want to find all m, n, l that satisfy (35). Because, in (35), $\mu(m,n,l)/\nu(m,n,l) = \rho$, there exists $k(m,n,l)$ such that $\mu(m,n,l) = k(m,n,l)\,\mu_0$ and $\nu(m,n,l) = k(m,n,l)\,\nu_0$.

First note that if (35) is satisfied for some m, n, l, it is also satisfied after replacing m, n, l with $\overline{m} = ms - nq$, $\overline{n} = np - mr$, and $l-1$ because

$$\mu(\overline{m}, \overline{n}, l-1) = \mu(m,n,l) \equiv k(m,n,l)\mu_0 \qquad \nu(\overline{m}, \overline{n}, l-1) = \nu(m,n,l) \equiv k(m,n,l)\nu_0$$

which follows from (34) and (6). This relation also gives

$$k(\overline{m}, \overline{n}, l-1) = k(m,n,l) \equiv k_0\,.$$

Inverting (33) gives

$$\begin{pmatrix} m \\ n \end{pmatrix} = \frac{k_0}{(\det \mathcal{S})^l}\,\mathcal{S}^l \begin{pmatrix} \mu_0 \\ \nu_0 \end{pmatrix} = \frac{k_0}{(\det \mathcal{S})^l} \begin{pmatrix} \mu_0\,N_{aal} + \nu_0\,N_{bal} \\ \mu_0\,N_{abl} + \nu_0\,N_{bbl} \end{pmatrix}. \tag{43}$$

Assume now that l in (35) can be extended to infinity for the given k_0. That would require that m, n as given by (43) be integer for arbitrary l. This could be satisfied only if $k_0(\mu_0\,N_{aal} + \nu_0\,N_{bal})$ and $k_0(\mu_0\,N_{abl} + \nu_0\,N_{bbl})$ are both divisible by $(\det \mathcal{S})^l = (\lambda_1\lambda_2)^l$ for arbitrary l. Using (14), one can see that this is not possible for any $|\lambda_2| > 1$. Therefore, for every k_0 there is a finite maximum value of l, denoted by $l_{\max}(k_0)$, satisfying (35). It is the largest value of l for which (43) gives m and n that are both integers. For the given ρ, the complete set of δ-peak positions as given by (35) is then

$$\omega = \frac{k_0\mu_0}{\rho_a(\det \mathcal{S})^l} \qquad l = 0, \ldots, l_{\max}(k_0) \tag{44}$$

where k_0 is arbitrary. Note that here l is no longer arbitrary as it was in (35), and that $k_0\mu_0$ can assume only values 'allowed' for the given ρ.

Let $D \equiv \det \mathcal{S}$ and $t \equiv p + s$. It is an obvious general property that $\gcd(k_0, D) = 1$ implies $l_{\max}(k_0) = l_{\max}(1)$. First assume that $\gcd(t, D) = 1$. Then

$$l_{\max}(k_0 D) = l_{\max}(k_0) + 1 \tag{45}$$

for arbitrary k_0. The proof is as follows. Let

$$\begin{pmatrix} m_0 \\ n_0 \end{pmatrix} \equiv \frac{k_0}{D^{l_{\max}(k_0)}}\,\mathcal{S}^{l_{\max}(k_0)} \begin{pmatrix} \mu_0 \\ \nu_0 \end{pmatrix}. \tag{46}$$

Since $(1/D)\,\mathcal{S}\binom{Dm_0}{Dn_0}$ is always an integer grid point, $l_{\max}(k_0 D) \geqslant l_{\max}(k_0) + 1$. The equality holds if and only if

$$\frac{D}{D^2}\,\mathcal{S}^2 \begin{pmatrix} m_0 \\ n_0 \end{pmatrix} = \frac{1}{D}\,\mathcal{S}^2 \begin{pmatrix} m_0 \\ n_0 \end{pmatrix}$$

is not an integer grid point (because then $l_{\max}(k_0)+1$ is the largest l for which m and n obtained from (43) when k_0 is replaced by $k_0 D$ are both integers). Using

$$\mathcal{S}^2 = -D\mathcal{I} + t\mathcal{S} \tag{47}$$

where $\mathcal{I}$ is the identity matrix (cf (7) for $L=2$), gives

$$\frac{1}{D}\mathcal{S}^2\begin{pmatrix} m_0 \\ n_0 \end{pmatrix} = \frac{t}{D}\mathcal{S}\begin{pmatrix} m_0 \\ n_0 \end{pmatrix} - \begin{pmatrix} m_0 \\ n_0 \end{pmatrix} \tag{48}$$

which has a non-integer component if and only if $(t/D)\,\mathcal{S}\binom{m_0}{n_0}$ has the same property. By definition of $l_{\max}(k_0)$, $(1/D)\,\mathcal{S}\binom{m_0}{n_0}$ is not an integer grid point. It cannot become integer when multiplied by t because $\gcd(t, D) = 1$. This proves (45). By induction on j, we get $l_{\max}(kD^j) = l_{\max}(1) + j$, where $\gcd(k, D) = 1$.

When D is a prime number, we thus already know $l_{\max}(k_0)$ for all k_0 and all ρ. Using this in (44), we can write the complete set of δ-peak positions as

$$\omega = \frac{k\,\mu_0}{\rho_a(ps-qr)^l} \qquad .\; l = 0, \ldots, l_{\max}(1)$$

where k is arbitrary. All these positions are the integral multiples of a single number—one can write them in the form

$$\omega_m = \frac{m}{\rho_0}\,. \tag{49}$$

Here

$$\rho_0 = \rho_a(ps-qr)^{l_{\max}(1)}/\mu_0$$

and m is an arbitrary integer. This set of peak positions is identical with that of a periodic system with lattice constant ρ_0, and we will call it *periodic-like*.

When D is not a prime, the set of δ-peak positions depends on the values of $l_{\max}(\kappa k_0)$, k_0 being arbitrary and κ an arbitrary factor of D. The spectrum may depend on ρ, and it can only be either periodic-like or infinite-periodic (see below) because all the δ-peak positions are mutually related by rational factors. For example, all substitution rules with integer eigenvalues that fall into the present category, give infinite-periodic spectra for a single ρ, and periodic-like spectra for all other rational ρ (see the next subsection).

Now let us turn to the case of $\gcd(D, t) \neq 1$. Assume first that D is prime. That means $\gcd(D, t) = D$, which implies $t = D$ because when $|\lambda_2| > 1$, $t \leqslant D$ or $t < -D$. Then (48) reduces to

$$\frac{1}{D}\mathcal{S}^2\begin{pmatrix} m_0 \\ n_0 \end{pmatrix} = \mathcal{S}\begin{pmatrix} m_0 \\ n_0 \end{pmatrix} - \begin{pmatrix} m_0 \\ n_0 \end{pmatrix}$$

which is always an integer grid point. Since

$$\frac{1}{D^2}\mathcal{S}^3\begin{pmatrix} m_0 \\ n_0 \end{pmatrix} = \frac{1}{D}\mathcal{S}^2\begin{pmatrix} m_0 \\ n_0 \end{pmatrix} - \frac{1}{D}\mathcal{S}\begin{pmatrix} m_0 \\ n_0 \end{pmatrix}$$

is not integer, $l_{\max}(k_0 D) = l_{\max}(k_0) + 2$. Thus, we again know $l_{\max}(k_0)$ for arbitrary k_0. Substituting these values into (44) makes it possible to write, for any rational ρ, the complete set of δ-peak positions as

$$\omega_{jk} = \frac{k\,\mu_0}{\rho_a D^j} \qquad j = 0, 1, \ldots, \infty \tag{50}$$

where k is arbitrary. This is an *infinite-periodic* spectrum, which can be considered to be the superposition of an infinite number of periodic spectra corresponding to lattice constants equal to $D^j \rho_0$, where $\rho_0 = \rho_a/\mu_0$. It is not a subset of integer multiples of any single number, thus it satisfies the definition of quasiperiodicity as given in the introduction. However, unlike in (39) and (41), there are no incommensurate frequencies involved. Nevertheless, both sets of (39) and (50) are equally dense. An open question remains whether systems with infinite-periodic spectrum with irrational λ_2 can also be called integer quasicrystals (as suggested in [20] for those with integer λ_2).

Finally, let us consider the case when $\gcd(D, t) = \kappa_1 > 1$ and D is not a prime number. Then there are integers κ_2, κ_3 such that $D = \kappa_1\kappa_2$ and $t = \kappa_1\kappa_3$, and (47) reads $\mathcal{S}^2 = \kappa_1(-\kappa_2\mathcal{I} + \kappa_3\mathcal{S})$. Therefore, for $\binom{m_0}{n_0}$ defined again by (46), $(\kappa_1\kappa_2^2/D^2)\,\mathcal{S}^2\binom{m_0}{n_0}$ is an integer grid point. This implies that $l_{\max}(\kappa_1\kappa_2^2 k_0) \geqslant l_{\max}(k_0) + 2$. Then the subset of δ-peak positions of (44) for all $k_0 = (\kappa_1\kappa_2^2)^j$ corresponding to a single value of $l = l_{\max}(1) + 2j \leqslant l_{\max}(k_0)$ is

$$\omega = \frac{\mu_0}{\rho_a D^{l_{\max}(1)} \kappa_1^j} \qquad j = 0, 1, \ldots, \infty . \tag{51}$$

This cannot be a subset of peak positions of a periodic-like spectrum. Since all the δ-peak positions are mutually related through rational factors, the full spectrum must be *infinite-periodic.*

Thus we have shown that when D is arbitrary and $\gcd(D, t) \neq 1$, the spectrum is infinite-periodic for all rational ρ. When $\gcd(D, t) = 1$ and D is prime, the spectrum is periodic-like for all ρ. When $\gcd(D, t) = 1$ and D is not prime, more study is needed. However, the type of spectrum—periodic-like or infinite-periodic—may be different for different values of ρ.

In addition to the periodic-like or infinite-periodic sets of δ peaks, a diffuse part of the structure factor will, also most probably, be present. However, in the limit of large l, for rational ρ, the dominant component in the diffraction spectra will be the above sets of δ peaks. For all irrational values of ρ, there is just a single trivial δ peak at $\omega = 0$, and the diffraction spectrum has a singular continuous and multifractal character (at least for $\xi(a) = a^3b$, $\xi(b) = b^2a$, as discussed in [19] for $s_a = s_b = 1$).

5.3. *Substitution rules with integer $\lambda_2 \neq 0$*

In this case, λ_1 and $q\tau$ are also integer. Except when $|\lambda_2| = 1$, this class of substitution rules is a subset of the previous one, and all the results of the previous subsection also apply fully here. The $|\lambda_2| = 1$ case differs from the $|\lambda_2| > 1$ case in that $l_{\max}(k_0)$ can be equal to infinity because $|D| = \lambda_1$.

For integer $|\lambda_2| \geqslant 1$, it is easy to find the spectrum when $\rho = q\tau/r$. Then $\Omega_0 = (\omega\rho_a/q\tau)\,e_1^{(r)}$ and $\Omega_l = (\omega\rho_a/q\tau)\,\lambda_1^l e_1^{(r)}$, and defining $\rho_0 = \rho_b \gcd(q\tau, r)/r$, we can write all the δ-peak positions as

$$\omega_{jm} = \frac{m}{\rho_0\,\lambda_1^j} \tag{52}$$

with arbitrary $j \geqslant 0$ and m. Therefore, for this special value of ρ, the spectrum is always *infinite-periodic.*

When $|\lambda_2| = 1$, and $\rho \neq q\tau/r$, the results of the previous subsection for $\gcd(t, d) = \gcd(\lambda_1 \pm 1, \pm\lambda_1) = \gcd(1, \lambda_1) = 1$ apply.

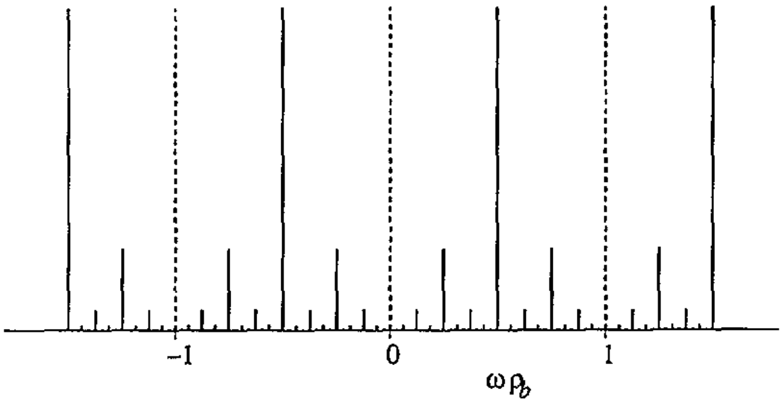

Figure 3. Relative intensities ($I_a(\omega)/|s_a(\omega)|^2$) of the δ peaks for a copper-mean system with $\rho_a = 2\rho_b$ and $s_b(\omega) = 0$. Trivial peaks are represented by dotted lines.

As an example, let us discuss in more detail the copper-mean rule ($a \mapsto ab^2$, $b \mapsto a$) [30, 31], for which $\lambda_1 = 2$, $\lambda_2 = -1$, $q = 2$, $r = 1$ and $\tau = 1$. A copper-mean system thus has the infinite-periodic spectrum (is an integer quasicrystal) for $\rho_a = 2\rho_b$ and it has the periodic-like spectrum for all other ρ. For $\rho = 2$, it has trivial δ peaks at $\omega_n = n/\rho_b$, and non-trivial ones at

$$\omega_{lk} = \frac{2k+1}{\rho_b\, 2^{l+1}} \qquad l \geqslant 0 \qquad k \text{ arbitrary}.$$

Using (24) and (23), one can show by induction on l that

$$\mathcal{T}_{l+1}(\omega_{lk}) = (-1)^l \begin{pmatrix} g_{lk} & -g_{lk} - g_{l+1,k} \\ g_{lk} & -g_{lk} - g_{l+1,k} \end{pmatrix}$$

where $g_{lk} = \exp[-\mathrm{i}(2k+1)\pi/2^{l-1}]$ (note that $g_{l+1,k}^2 = g_{lk}$). Thus

$$\mathcal{T}_L(\omega_{lk}) = (-1)^l\ \mathcal{S}^{L-l-1} \begin{pmatrix} g_{lk} & -g_{lk} - g_{l+1,k} \\ g_{lk} & -g_{lk} - g_{l+1,k} \end{pmatrix}$$

and

$$I_a(\omega_{lk}) = \frac{\left|g_{l+1,k}\left[s_a(\omega_{lk}) - s_b(\omega_{lk})\right] - s_b(\omega_{lk})\right|^2}{9 \times 4^l\, \rho_b^2} \qquad l \geqslant 0.$$

For the trivial peaks

$$I_a(\omega_n) = \frac{|s_a(\omega_{lk}) + s_b(\omega_{lk})|^2}{9\,\rho_b^2}.$$

The intensity $I_a(\omega_{lk})$ decreases quite quickly with l as shown in figure 3. One can expect that only the peaks corresponding to the values of l less than a certain threshold can be resolved in an experiment, and the rest simply contribute to the background diffuse scattering. Of course, a similar outcome can be expected in the case of the classical quasicrystals but here the surviving pattern would be more regular. Thus a rather interesting question is how the experimentally measured diffraction spectra of a copper-mean superlattice with $\rho_a = 2\rho_b$ would differ from those for rational $\rho \neq 2$, and from the spectra of periodic systems. A copper-mean superlattice is probably one of the best candidates to reveal such a difference as the above rate of the decrease of intensity with l may actually be one of the slowest among all integer quasicrystals.

5.4. Substitution rules with $\lambda_2 = 0$

In this case, $n_{\mathrm{eff}} = 0$ and thus the substitution matrix $\mathcal{S}$ is non-invertible. This makes the situation quite different from the case in subsection 5.1 although λ_1 is still a PV number. We have $\lambda_1 = p + s$ and $\mathcal{S}^l = (p+s)^{l-1}\,\mathcal{S}$, $l \geqslant 1$.

According to (38), δ peaks are given by (32) which now has solutions for any ratio ρ because of the singularity of $\mathcal{S}$ (because all integer grip points have infinitely many 1st generation predecessors). If, for integer x, y,

$$\begin{pmatrix} x \\ y \end{pmatrix} = \mathcal{S}^l \Omega_0 = \lambda_1^{l-1} \mathcal{S} \Omega_0 = \lambda_1^{l-1} (\tau\rho_a + \rho_b)\, \omega \begin{pmatrix} q \\ s \end{pmatrix}$$

then $x = qm/\gcd(q,s)$ and $y = sm/\gcd(q,s)$, m being an arbitrary integer. It gives non-trivial δ-peak positions at

$$\omega_{jm} = \frac{m}{\rho_0\, \lambda_1^j} \tag{53}$$

for arbitrary $j \geqslant 0$ and m not divisible by λ_1. It is the same type of the infinite-periodic spectrum as in (50), (51), and (52) but now $\rho_0 = \gcd(q,s)(\tau\rho_a + \rho_b) = \gcd(p,r)\,\rho_a + \gcd(q,s)\,\rho_b$. The main difference from the previous case is that now the new type of quasicrystals may exist for arbitrary real ρ_a/ρ_b, but note that here $\mathcal{L}_{aL}/\mathcal{L}_{bL}$ is always rational for $L \geqslant 1$. However, the set (53) represents only the largest possible set of δ peaks. Some of them need not materialize (have zero intensities) as a result of the orthogonality of some of the matrices in the product of (24). In fact, for periodic substitutional systems, which all have $n_{\mathrm{eff}} = 0$, all peaks with large j must be switched off to get a periodic set of peaks.

For example, a periodic chain generated by $\xi(a) = \xi(b) = ab$ has $\omega_{jm} = m/((\rho_a + \rho_b)2^j)$. Its $\boldsymbol{T}$ matrix reads

$$\boldsymbol{T}_L(\omega) = \begin{pmatrix} 1 & \exp(2\mathrm{i}\pi\omega\rho_a) \\ 1 & \exp(2\mathrm{i}\pi\omega\rho_a) \end{pmatrix} \prod_{k=L-1}^{1} [1 + \exp(\mathrm{i}\pi\omega 2^k(\rho_a + \rho_b))] \qquad L \geqslant 2\,.$$

Thus $\boldsymbol{T}_L(\omega_{jm}) = 0$ for $j \geqslant 0$, and the intensities are non-zero only for $j = 0$, for which

$$I_b^{\mathrm{per}}(\omega_{0m}) = I_a^{\mathrm{per}}(\omega_{0m}) = \frac{|s_a(\omega_{0m})|^2 + 2\mathrm{Re}[s_a^*(\omega_{0m})\, s_b(\omega_{0m})\, \mathrm{e}^{\mathrm{i}\varphi_m}] + |s_b(\omega_{0m})|^2}{(\rho_a + \rho_b)^2} \tag{54}$$

where $\varphi_m = 2\pi m\rho_a/(\rho_a + \rho_b)$.

A TM chain generated by $\xi(a) = ab$, $\xi(b) = ba$ has δ peaks at exactly the same positions but with modified intensities

$$I_b^{\mathrm{TM}}(\omega_{0m}) = I_a^{\mathrm{TM}}(\omega_{0m}) = I_a^{\mathrm{per}}(\omega_{0m})\ (1 + \cos\varphi_m)\,/\,2\,. \tag{55}$$

For large enough L, the δ peaks for $\rho_a \neq \rho_b$ must prevail over all other peaks with $\alpha_\chi(\omega) < 2$ that are responsible for the singular continuous part of the spectrum, and the diffraction spectrum of a TM chain will resemble that of the above periodic chain. For $\rho_a = \rho_b$, the peaks corresponding to $m = 2k - 1$ are switched off in the TM case because $1 + \cos\varphi_{2k-1} = 0$, which is the consequence of the orthogonality of some matrices in (24) as discussed for this special case already in [17]. Thus for $\rho_a = \rho_b$, a TM chain has only the trivial δ peaks (identical with those of a random chain). For $\rho_a \neq \rho_b$, it has a set of δ peaks in the same positions as the periodic chain, and the larger $\rho_a/\rho_b + \rho_b/\rho_a$ is, the closer the peak intensities approach those of the periodic chain. This can be understood in the light of the fact that when one tile is much larger than the other one, it fills most of the space in an 'almost periodic way'. The formula for the Fourier transform for arbitrary ω is

$$\begin{aligned} F_{\chi L}^{\mathrm{TM}}(\omega) = 2^{L-1}\,\mathrm{e}^{\mathrm{i}\pi\omega(\rho_a+\rho_b)(2^{L-1}-1)} \Bigg\{ C(\omega) \prod_{j=0}^{L-2} \cos[2^j\pi\omega(\rho_a+\rho_b)] \\ + (\delta_{\chi,b}-\delta_{\chi,a})\,(-\mathrm{i})^L\,S(\omega) \prod_{j=0}^{L-2} \sin[2^j\pi\omega(\rho_a+\rho_b)] \Bigg\} \end{aligned} \tag{56}$$

where $L \geqslant 2$ and

$$\begin{aligned} S(\omega) &= s_b(\omega)\,\mathrm{e}^{\mathrm{i}\pi\omega\rho_a}\sin(\pi\omega\rho_a) - s_a(\omega)\,\mathrm{e}^{\mathrm{i}\pi\omega\rho_b}\sin(\pi\omega\rho_b) \\ C(\omega) &= s_b(\omega)\,\mathrm{e}^{\mathrm{i}\pi\omega\rho_a}\cos(\pi\omega\rho_a) + s_a(\omega)\,\mathrm{e}^{\mathrm{i}\pi\omega\rho_b}\cos(\pi\omega\rho_b)\,. \end{aligned} \tag{57}$$

The diffraction spectra of some other TM-like systems are similar. For example for $\xi(a) = ab$, $\xi(b) = b^2a^2$, all the peaks of (53) with $l > 1$ do not materialize again. Unlike for the TM chain, for $\rho_a = \rho_b$ all the non-trivial peaks survive, and this chain diffracts as a periodic chain for all ratios ρ_a/ρ_b.

However, a different situation can be found for $\xi(a) = ab$, $\xi(b) = ab^2a$ (it has the same $\mathcal{S}$, but is in a different LI class to the former rule—see Table II of [21]). For the present rule, all the peaks of (53) seem to have non-zero intensity. Thus, this is another explicit example of an integer quasicrystal with δ peaks at infinitely many rational multiples of a single frequency. Formulae for arbitrary ω, ρ_a and ρ_b are quite complicated: for $L \geqslant 1$ one has

$$\begin{aligned} F_{aL}(\omega) &= (t_L + u_L)\,F_{a1} + (v_L + w_L)\,f \\ F_{bL}(\omega) &= F_{aL}(\omega) + E^{3^{L-1}}[\,(t_L + w_L)\,F_{a1} + (v_L + u_L)\,f\,]\,. \end{aligned}$$

Here

$$F_{a1} = s_a(\omega) + s_b(\omega)\,\mathrm{e}^{2\mathrm{i}\pi\omega\rho_a} \qquad f = s_b(\omega) + s_a(\omega)\,\mathrm{e}^{2\mathrm{i}\pi\omega\rho_a} \qquad E = \mathrm{e}^{2\mathrm{i}\pi\omega(\rho_a+\rho_b)}$$

$$t_1 = v_1 = w_1 = v_2 = 0 \qquad u_1 = t_2 = 1 \qquad u_2 = E \qquad w_2 = E^2\,.$$

For $L \geqslant 3$

$$t_L = \sum_{k=1}^{L-2} u_k E^{z_{kL}}\, Z_{kL} + u_{L-1} \qquad v_L = \sum_{k=1}^{L-2} w_k E^{z_{kL}}\, Z_{kL} + w_{L-1}$$

$$Y_j = 2\cos[2\pi\omega j(\rho_a+\rho_b)] \qquad z_{kL} = \sum_{l=k}^{L-2} 3^l \qquad Z_{kL} = \prod_{l=k}^{L-2}(1 + Y_{3^l})\,.$$

For $k \geqslant 1$

$$u_{2k+2} = E^{9^k}\,(u_{2k+1} + E^{9^k} w_{2k+1}) \qquad w_{2k+2} = E^{9^k}\,(w_{2k+1} + E^{9^k} u_{2k+1})$$

$$u_{2k+1} = E^{\zeta_k} \sum_{C_k^{(0)}} \left[\prod_{j=0}^{k-1} Y_{2^{c_j}9^j}\right] \qquad w_{2k+1} = E^{\zeta_k} \sum_{C_k^{(1)}} \left[\prod_{j=0}^{k-1} Y_{2^{c_j}9^j}\right] \qquad \zeta_k = 6\sum_{j=0}^{k-1} 9^j\,.$$

Here c_j; $j = 0, \ldots, k-1$ can assume two values, 0 and 1. Altogether, there are 2^k different ways to choose c_j. $\sum_{C_k^{(0)}}$ represents the sum over 2^{k-1} of these combinations for which $k + \sum_{j=0}^{k-1} c_j = 0 \pmod 2$. $\sum_{C_k^{(1)}}$ represents the sum over the other 2^{k-1} of these combinations for which $k + \sum_{j=0}^{k-1} c_j = 1 \pmod 2$. For example, $u_3 = E^6 Y_2$, $w_3 = E^6 Y_1$, $u_5 = E^{60}(Y_1 Y_9 + Y_2 Y_{18})$, $w_5 = E^{60}(Y_2 Y_9 + Y_1 Y_{18}), \ldots$.

From these formulae we have calculated the intensities of a few δ peaks for $\rho_a = \rho_b$ and $m = 1, 2$ in (53). In this case, $I_a(\omega_{lm}) = d_{lm}\,|s_a(\omega_{lm}) - s_b(\omega_{lm})|^2/(2\rho_a)^2$. For the first few values of l, $d_{01} = 0.\overline{1}$, $d_{11} = 0.\overline{037}$, $d_{21} = 2.323 \times 10^{-4}$, $d_{31} = 1.560 \times 10^{-7}$, $d_{41} = 1.159 \times 10^{-11}$, $d_{12} = 0.\overline{1}$, $d_{22} = 3.18 \times 10^{-3}$, $d_{32} = 8.660 \times 10^{-6}$ ($m = 2$ and $l = 0$ give a trivial peak with intensity $|s_a(\omega_{12}) + s_b(\omega_{12})|^2/(2\rho_a)^2$). Here the rate of decrease of intensity with l is even faster than that for the copper-mean superlattice discussed above.

6. Conclusions

In summary, the following classes of two-letter substitutional systems were found:

• Classical quasicrystals (two incommensurate spatial frequencies)	$0 < \lvert\lambda_2\rvert < 1,\ \lambda_1\lambda_2 = \pm 1$
• Infinite-quasiperiodic systems (rational multiples, two incommensurate frequencies)	$0 < \lvert\lambda_2\rvert < 1,\ \lambda_1\lambda_2 \neq \pm 1$
• Periodic systems (integer multiples of a single spatial frequency)	$\lambda_2 = 0$
• Infinite-periodic systems (integer quasi-crystals) (rational multiples of a single spatial frequency)	$\lvert\lambda_2\rvert > 1$, ρ_a/ρ_b rational (e.g. always when $\gcd(\det S, p+s) \neq 1$), or $\lambda_2 =$ integer $\neq 0$ and $\rho_a/\rho_b = q\tau/r$, or $\lambda_2 = 0$
• Systems with periodic-like arrays of non-trivial δ peaks	$\lvert\lambda_2\rvert \geqslant 1$ and ρ_a/ρ_b rational, or $\lambda_2 = 0$
• Systems with no nontrivial δ peaks	$\lvert\lambda_2\rvert \geqslant 1$ and ρ_a/ρ_b irrational, or $\lambda_2 = 0$.

The PV property ($|\lambda_2| < 1$) supplemented with the requirement that $\lambda_2 \neq 0$ is sufficient for obtaining a quasicrystal with either classical (quasiperiodic) or infinite quasiperiodic spectra. If one requires that a quasicrystal must involve some irrational (incommensurate) numbers, then there are no other 1D two-letter substitutional quasicrystals. However, if one uses a looser definition of quasicrystals as given in the introduction, which also occurs often in other literature (requiring a dense set of δ peaks at positions that cannot be expressed as integral multiples of a single number), then it is possible to claim that there are quasicrystals with infinite-periodic spectra also for $|\lambda_2| \geqslant 1$ for some or for all rational ρ, and for $\lambda_2 = 0$.

The presented scheme can be easily adapted to substitution rules with an arbitrary number of letters. Much more interesting would be its systematic application to higher-dimensional substitutional systems, which has so far been done only for a few systems, e.g. for the Penrose tiling [32]. In higher dimensions one has much less freedom in choosing different tile dimensions. For example, in parallelogram or parallelopiped tilings the dimensions of all the different tiles are identical. In analogy with the 1D results presented here, we can expect that the diffraction spectra of many such aperiodic deterministic structures with underlying periodic lattice will resemble in some way those of periodic crystals (have some higher-dimensional variants of periodic-like or infinite-periodic spectra). Could natural solids with such structures exist? Would it be possible to distinguish their diffraction spectra from truly periodic crystals when various defects and the Debye–Waller factor broaden all the peaks? Apparently the analysis of the diffuse background would be necessary. We think that a good test case to develop such analysis is provided by some of the artificial layered structures described by the formalism of this paper (see the discussion at the end of section 5.3). It might be interesting to try to do diffraction experiments on some of these superlattices that are either the integer quasicrystals or diffract as periodic lattices, to find out how their spectra differ from periodic superlattices made of the same building blocks.

We are also preparing a sequel (part II) to this paper dealing with non-δ peaks with $\alpha_\chi(\omega) < 2$ (the singular continuous component of the spectrum) originating from the periodic orbits of the map (29) with period larger than 1.

Acknowledgments

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Appendix A. The proof of equation (30)

We denote by $|M_L(\omega)|$ the matrix whose $\chi\phi$-entry is the modulus of the $\chi\phi$-entry of $M_L(\omega)$; $\chi, \phi \in \mathcal{A}$, and by $|\mathbf{x}|$ the vector whose χ-component is the modulus of the χ-component of the vector $\mathbf{x}$; $\chi \in \mathcal{A}$. From (22) we always have $|M_L(\omega)| \leqslant \mathcal{S}$ by which we mean that $[|M_L(\omega)|]_{\chi\phi} \leqslant \mathcal{S}_{\chi\phi}$ for all $\chi, \phi \in \mathcal{A}$. If for some (χ, ϕ), $[|M_L(\omega)|]_{\chi\phi} < \mathcal{S}_{\chi\phi}$, we write $|M_L(\omega)| < \mathcal{S}$.

We first prove that $|\lambda_{L\max}| \leqslant \lambda_1$, where $\lambda_{L\max}$ is the largest-modulus eigenvalue of $M_L(\omega)$: $e_1^{(l)}$ be the left eigenvector of $\mathcal{S}$ associated with λ_1 (cf section 3), and $e_{L\max}$ be a right eigenvector of $M_L(\omega)$ associated with $\lambda_{L\max}$; $e_1^{(l)}\mathcal{S} = \lambda_1 e_1^{(l)}$, $M_L(\omega)\, e_{L\max} = \lambda_{L\max}\, e_{L\max}$. Then

$$|\lambda_{L\max}|\, e_1^{(l)}\, |e_{L\max}| = e_1^{(l)}\, |M_L(\omega)\, e_{L\max}| \leqslant e_1^{(l)}\, |M_L(\omega)|\, |e_{L\max}| \leqslant e_1^{(l)}\, \mathcal{S}\, |e_{L\max}|$$

because both components of $e_1^{(l)}$ are positive. From this

$$|\lambda_{L\max}|\, e_1^{(l)}\, |e_{L\max}| \leqslant \lambda_1\, e_1^{(l)} |e_{L\max}|. \tag{A1}$$

Since $e_1^{(l)}\, |e_{L\max}| > 0$, (A1) implies $|\lambda_{L\max}| \leqslant \lambda_1$, which was to be proven. If $|M_L(\omega)| < \mathcal{S}$, one can show in the same way that $|\lambda_{L\max}| < \lambda_1$ because $\mathcal{S}$ has positive off-diagonal entries. As a consequence of this, we can say that if $|\lambda_{L\max}| = \lambda_1$ then $|M_L(\omega)| = \mathcal{S}$.

Now we prove that $|t_{L\max}| \approx \lambda_1^L \Longrightarrow \lim_{L\to\infty} |\lambda_{L\max}| = \lambda_1$, where $t_{L\max}$ is the largest-modulus eigenvalue of $T_L(\omega)$: from (24), $|T_L(\omega)| = \left|\prod_{l=L-1}^{0} M_l(\omega)\right| \leqslant \prod_{l=L-1}^{0} |M_l(\omega)| \leqslant \mathcal{S}^L$. Therefore, $|t_{L\max}| \leqslant \prod_{l=L-1}^{0} |\lambda_{l\max}(\omega)| \leqslant {\lambda_1}^L$. If $|t_{L\max}| \approx \lambda_1^L$ as $L \to \infty$, then $\prod_{l=L-1}^{0}(|\lambda_{l\max}|/\lambda_1) \to 1$ as $L \to \infty$, and since $|\lambda_{L\max}| \leqslant \lambda_1$ for all L, $|\lambda_{L\max}| \to \lambda_1$ as $L \to \infty$.

Next we prove that $\alpha_\chi(\omega) = 2 \Longrightarrow \lim_{L\to\infty} |M_L(\omega)| = \mathcal{S}$. Assume that the negation is true: $\alpha_\chi(\omega) = 2$ for some $\chi \in \mathcal{A}$ and $|M_L(\omega)|$ does not converge to $\mathcal{S}$. Since $D_L(\omega) \equiv \mathcal{S} - |M_L(\omega)|$ is a non-negative matrix, this means that $\exists\eta > 0\ \forall L_0\ \exists L > L_0$ such that at least one of the entries of $D_L(\omega)$ is greater than η. This can be written in a 'compact' way: $\exists\varepsilon > 0$ (ε now being a 2×2 matrix) $\forall L_0\ \exists L > L_0$ such that $D_L(\omega) \geqslant \varepsilon$. Then $(\mathcal{S}-\varepsilon) \geqslant |M_L(\omega)|$. Since $(\mathcal{S}-\varepsilon) < \mathcal{S}$, the larger eigenvalue λ_ε of $(\mathcal{S}-\varepsilon)$ is strictly less than λ_1 and goes to λ_1 when ε goes to $\mathbf{0}$, we have

$$\lambda_1 > \lambda_\varepsilon \geqslant \max[\,\text{eigenvalues}(|M_L(\omega)|)\,] \geqslant |\lambda_{L\,\max}|$$

which means that $|\lambda_{L\,\max}|$ cannot converge to λ_1, which is in contradiction with $\alpha_\chi(\omega) = 2$. Thus $\lim_{L\to\infty} |M_L(\omega)| = \mathcal{S}$. Note that the converse is false. For instance, in the golden-mean case, $(a \mapsto ab,\ b \mapsto a)$, $|M_L(\omega)| = \mathcal{S}\ \ \forall(L,\omega)$ but there exists ω such that $\alpha_\chi(\omega) \neq 2$ for a $\chi \in \mathcal{A}$.

Let us now consider a substitution such that for some $\chi, \phi \in \mathcal{A}$ and k an integer, $\xi^k(\chi)$ contains ϕ^2 (this is always satisfied except for certain periodic $\xi^\infty(\chi)$). If ξ is a primitive substitution, ξ^k is also primitive and gives the same structure factor. This allows us to work with ξ^k instead of ξ without any modification of the physical problem. Thus we can assume that $\xi(\chi)$ itself contains ϕ^2 (otherwise we replace ξ by ξ^k). Let k_0 be such that $\xi(\chi)[k_0] = \xi(\chi)[k_0+1] = \phi$, and let $\lim_{L\to\infty} |M_L(\omega)| = \mathcal{S}$. This convergence implies that for all k and k' such that $\xi(\chi)[k] = \xi(\chi)[k']$,

$$2\pi\omega \lim_{L\to\infty} \left\{ \sum_{j=0}^{k-1} \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\xi(\chi)[j]} - \sum_{j=0}^{k'-1} \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\xi(\chi)[j]} \right\} = 0 \pmod{2\pi} \tag{A2}$$

which gives

$$\lim_{L\to\infty} \omega \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\xi(\chi)[k_0]} \equiv \lim_{L\to\infty} \omega \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\phi} = 0 \pmod{1}. \tag{A3}$$

It remains to prove that the other component of Ω_L converges to $0\,(\mathrm{mod}\,1)$. We denote by ϕ' the other letter of $\mathcal{A}$. From (A3) we have

$$\begin{aligned} 0 \pmod{1} = \lim_{L\to\infty} \omega \left[\mathcal{S}^{L+1} \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\phi} &= \mathcal{S}_{\phi\phi} \lim_{L\to\infty} \omega \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\phi} \\ &+ \mathcal{S}_{\phi\phi'} \lim_{L\to\infty} \omega \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\phi'} = \mathcal{S}_{\phi\phi'} \lim_{L\to\infty} \omega \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\phi'}. \end{aligned}$$

Then, if $\mathcal{S}_{\phi\phi'} \neq 1$ (i.e. $\mathcal{S}_{\phi\phi'} > 1$ because it is an off-diagonal entry of $\mathcal{S}$), there exist k and k' such that $k < k'$, $\xi(\phi)[k] = \xi(\phi)[k'] = \phi'$, and for $k < j < k'$, $\xi(\phi)[j] = \phi$. Then from (A2),

$$\omega \lim_{L\to\infty} \left[\mathcal{S}^L \begin{pmatrix}\rho_a\\ \rho_b\end{pmatrix} \right]_{\phi'} = 0 \pmod{1}$$

which was to be proven. In the periodic case, a direct calculation shows that (30) holds, too.

Appendix B. The proof of (38)

B1. (i)$\Longrightarrow$(ii)

Let m_l be an integer such that $d_1\lambda_1^l + d_2\lambda_2^l = m_l + \epsilon_l$, where $0 \leqslant \epsilon_l \leqslant \frac{1}{2}$. We claim that

$$\exists L \qquad \text{such that} \qquad m_{l+2} - m_{\text{eff}}\, m_{l+1} - n_{\text{eff}}\, m_l = 0 \qquad \forall l \geqslant L \tag{B1}$$

and

$$m_l = d_1\,\lambda_1^l + \beta\,\lambda_2^l \qquad \forall l \geqslant L\,. \tag{B2}$$

Actually, $m_{l+2} - m_{\text{eff}}\, m_{l+1} - n_{\text{eff}}\, m_l = d_1\,\lambda_1^l\,(\lambda_1^2 - m_{\text{eff}}\lambda_1 - n_{\text{eff}}) + d_2\,\lambda_2^l\,(\lambda_2^2 - m_{\text{eff}}\lambda_2 - n_{\text{eff}}) + \epsilon_{l+2} - m_{\text{eff}}\,\epsilon_{l+1} - n_{\text{eff}}\,\epsilon_l$. The first two terms on the right-hand side are zero, and so the left-hand side is a sequence of integers converging to zero. This proves (B1). Clearly, there exists a couple α, β such that

$$m_L = \alpha\,\lambda_1^L + \beta\,\lambda_2^L \qquad m_{L+1} = \alpha\,\lambda_1^{L+1} + \beta\,\lambda_2^{L+1}\,. \tag{B3}$$

Using (B1), this implies

$$m_{L+2} = \alpha\,\lambda_1^L\,(m_{\text{eff}}\lambda_1 + n_{\text{eff}}) + \beta\,\lambda_2^L\,(m_{\text{eff}}\lambda_2 + n_{\text{eff}}) = \alpha\,\lambda_1^{L+2} + \beta\,\lambda_2^{L+2}$$

and (B3) will be proven once the equality $\alpha = d_1$ is established. By definition, $\epsilon_l = (d_1 - \alpha)\,\lambda_1^l + (d_2 - \beta)\,\lambda_2^L$ $\forall l \geqslant L$. Since $\lambda_1 > \max(1, |\lambda_2|)$, $d_1 - \alpha = \lim_{l\to\infty} \epsilon_l/\lambda_1^l = 0$. Finally, from (B3),

$$d_1 = \frac{m_{L+1} - m_L\lambda_2}{\lambda_1^L\,(\lambda_1 - \lambda_2)}\,. \tag{B4}$$

Thus (38)(ii)(1) is proven. Note that $\lambda_2 = 0$ implies that λ_1 is an integer, then (B4) implies (38)(ii)(3). If $|\lambda_2| < 1$, there is clearly no restriction on d_2 while for $|\lambda_2| \geqslant 1$, $\epsilon_l = (d_2 - \beta)\,\lambda_2^l$ does not converge to zero except when $\beta = d_2$. Thus $\epsilon_l = 0$ $\forall l \geqslant L$, and (38)(ii)(2) holds.

B2. (ii) $\Longrightarrow$ (i)

First note that the sequence of positive integers

$$f_{l+2} = m_{\text{eff}}\, f_{l+1} + n_{\text{eff}}\, f_l \qquad f_0 = 0 \qquad f_1 = 1$$

is of the Fibonacci type and

$$f_l = \frac{\lambda_1^l - \lambda_2^l}{\lambda_1 - \lambda_2}\,.$$

Thus

$$\begin{aligned} d_1\lambda_1^l + d_2\lambda_2^l &= (m + n\lambda_2)\,\frac{\lambda_1^{l-L}}{\lambda_1 - \lambda_2} - (m + n\lambda_1)\,\frac{\lambda_2^{l-L}}{\lambda_1 - \lambda_2} + \left(d_2 + \frac{m + n\lambda_1}{\lambda_2^L(\lambda_1 - \lambda_2)}\right)\lambda_2^l \\ &\leqslant m f_{l-L} - n\, n_{\text{eff}}\, f_{l-L-1} + \left(d_2 + \frac{m + n\lambda_1}{\lambda_2^L(\lambda_1 - \lambda_2)}\right)\lambda_2^l \end{aligned}$$

goes to zero (mod 1) when $l \to \infty$.

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