

# Diffraction analysis of decorated Fibonacci chains in the average unit-cell approach

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A novel approach to diffraction analysis of decorated quasicrystals is discussed. An average unit cell has been constructed for a decorated Fibonacci chain and used for analysis of its diffraction pattern. After some transformation of the scattering vectors, all the diffraction peaks are described by a single envelope function which is characteristic of a given decoration. It has been shown that by knowing several diffraction intensities, in a limited range of the scattering vector, it is possible to reconstruct the envelope function successfully and distinguish between different decorated structures.

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## 1. Introduction

The question ‘where are the atoms?’ for most quasicrystals is still unanswered. A very powerful method of solving the structures of quasicrystals, based on hyperspace crystallography, was formulated several years ago (Bak, 1985; Janssen, 1986; Gratias *et al.*, 1988*a,b*; Janot, de Boissieu *et al.*, 1989; Janot, Pannetier *et al.*, 1989; de Boissieu *et al.*, 1991; Cervellino *et al.*, 1998). Using such an approach, it was possible to solve, at least partially, many icosahedral (de Boissieu *et al.*, 1991; van Smaalen *et al.*, 1991; Yamamoto, 1992; Cornier-Quiquandon *et al.*, 1991; Boudard *et al.*, 1992) and decagonal (Steurer, 1990; Steurer & Kuo, 1990; Yamamoto *et al.*, 1990) phases. Several powerful structure-solution techniques as well as refinement programs have also been developed (Elcoro *et al.*, 1994; Fu *et al.*, 1993; Haibach & Steurer, 1996; Dubois & Janot, 1987). However, as has been pointed out by Lancon *et al.* (1994), misindexing the diffraction pattern, *i.e.* choosing a wrong unit cell, leads to badly partitioned atomic surfaces and a severely averaged structure in real space. Recently, Cervellino *et al.* (1998) have derived a method based on the physical-space Patterson function, which allows the determination of the correct  $n$ -D Bravais lattice of quasicrystals. As a result, the optimum unit cell can be chosen and a proper indexing scheme can be applied for the diffraction pattern.

In this paper, another approach to diffraction analysis of quasicrystals is used. The main idea of this approach is based on the concept of a reference lattice (Wolny, 1992, 1998*a,b*), which is a mathematically defined one-dimensional set of equidistant planes, perpendicular to the scattering vector  $\mathbf{k}_0$ , with periodicity related to the scattering vector as  $2\pi/k_0$ . Using such a reference lattice, one can calculate a probability distribution of atomic displacements from the nearest point of the reference lattice. Then, the Fourier transform of such a probability distribution gives values of form factors for a periodic set of scattering vectors given by  $k = mk_0$ , where  $m$  is an integer. Such a Fourier transform also defines a smooth

function, called the envelope function, connecting intensity maxima for periodic series of diffraction peaks.

All the calculations of the diffraction pattern are performed in the physical space and the envelope function can be obtained directly from the diffraction pattern. To get the envelope function from the experimental data, no Fourier transform of the diffraction pattern is required, which is very important in connection with the well known phase problem in crystallography. Knowledge of the envelope function gives several advantages. First, it reduces the influence of experimental errors on the final result of a structure refinement. It also allows correction of the intensities of some ‘badly measured diffraction peaks’. Finally, intensities of some unmeasured diffraction peaks can be easily completed – the envelope function is a smooth function of the scattering vector so the intensities of different reflections can be reliably predicted. For quasicrystals, for example, the envelope function can be well determined for many different reflections observed in their diffraction pattern, especially when reduced scattering vectors are used (for the definition of reduced scattering vector, see the text). Finally, by this procedure, one can almost uniquely define other unknown intensities lying in the neighbourhood of reduced scattering vectors for some measured reflections. On the other hand, the envelope functions look different for different decorations and such a property allows one to distinguish easily between different models.

## 2. Decorated chain

A simple 1D quasiperiodic structure derived by decorating a Fibonacci chain (Lancon *et al.*, 1994; Cervellino *et al.*, 1998) has been used to test the presented approach to diffraction analysis. The decorated tiling is obtained by applying once the substitution rules  $L \rightarrow LLSL$  and  $S \rightarrow SLS$  on the Fibonacci chain. The lengths of the two bounds have been chosen as

follows:  $L = \tau \approx 1.618$  and  $S = L/\tau = 1$ , where  $\tau$  is the golden mean. To calculate the diffraction pattern, it was assumed that the same fictitious atoms are placed at the ends of each bond, which leads to the mean average distance between atoms equal to  $a = 1 + 1/\tau^2 \approx 1.382$ , the same as for an undecorated Fibonacci chain. The calculated diffraction pattern, *i.e.* the normalized square of the absolute value of the structure factor (Fig. 1), is comparable with the one obtained from a nondecorated Fibonacci chain. The positions of the main Bragg peaks are the same; however, the scattering intensity distribution is different.

It is well known that the Fibonacci chain can be regarded as a modulated structure and the same holds for the decorated chain. Using two reference lattices (Wolny, 1998*a,b*), the first one having periodicity  $a$  and the second one periodicity  $b = \tau a$ , the structure factor for arbitrary integer combination of the two scattering vectors  $k_0 = 2\pi/a$ ,  $q_0 = 2\pi/b = k_0/\tau$  can be expressed by

$$F(nk_0 + mq_0) = \int_{-a/2}^{a/2} \int_{-b/2}^{b/2} P(u, v) \times \exp[i(nk_0u + mq_0v)] du dv, \quad (1)$$

where  $u$  and  $v$  are the shortest distances of the atomic positions from the appropriate points of the two reference lattices and  $P(u, v)$  is the corresponding probability distribution, which is the equivalent of an average unit cell. To calculate the whole diffraction pattern, one has to know the probability distribution in an average unit cell which is bounded by  $(-a/2, a/2)$  for  $u$  and  $(-b/2, b/2)$  for  $v$ . As has already been shown (Wolny, 1998*b*), such a probability distribution is nonzero only along the line given by

$$v = -\tau^2 u \quad (2)$$

(see also Fig. 2*a*). For the Fibonacci chain, the shape of the probability distribution is rectangular (Fig. 3), which leads to the following expression for the intensity of the diffraction peaks (*i.e.* main reflections and their satellites):

$$I/N^2 = [\sin(w)/w]^2, \quad (3)$$

where

$$w = (nk_0 - \tau^2 mq_0)/2\tau = k_0(n - m\tau)/2\tau \quad (4)$$

and  $n$  and  $m$  are indices of the main reflection and its satellites, respectively. Knowing the peak positions in the diffraction pattern, which are given by

$$k = nk_0 + mq_0 = k_0(n + m/\tau), \quad (5)$$

one can write (4) in the form

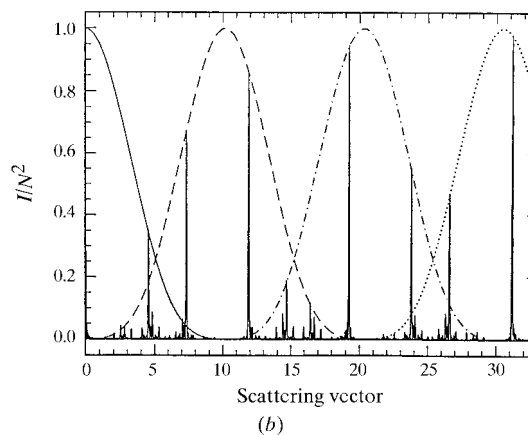
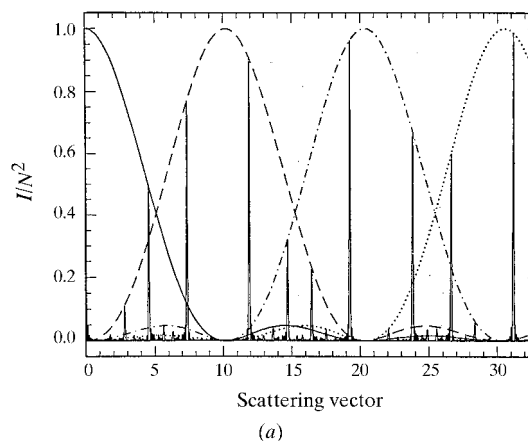
$$w = [k - (1 + \tau^2)mq_0]/2\tau. \quad (6)$$

Expression (6), together with (3), has been used to calculate all the envelope functions shown in Fig. 1(*a*). The envelope functions connect peak maxima of the main reflections, *i.e.* for  $m = 0$  (full line) and their satellites (up to third order, *i.e.* for  $m = 1, 2, 3$ ) (broken lines).

For the considered decorated structures, the probability distribution is nonzero along the same line as before (Fig. 2*b*), given by equation (2). However, the distribution changes from the rectangular shape for a Fibonacci chain to a step-like function for the decorated structure (Fig. 3). One can easily recognize the similarity of such a shape to the shape of the window function in perp-space (see also Fig. 6 of Lancon *et al.*, 1994). The main difference is, however, that the probability distribution is calculated only in real space, and no higher-dimensional analysis is required. For the probability distribution of a decorated structure (Fig. 3), the six different levels of probability are obtained and they are defined as follows:

$$\begin{aligned} P(u) &= 0 && \text{for } u \notin (u_1, u_{10}) \\ P(u) &= \tau/5 && \text{for } u \in (u_1, u_2) \quad \text{or } u \in (u_9, u_{10}) \\ P(u) &= 2\tau/5 && \text{for } u \in (u_2, u_3) \quad \text{or } u \in (u_8, u_9) \\ P(u) &= 3\tau/5 && \text{for } u \in (u_3, u_4) \quad \text{or } u \in (u_7, u_8) \\ P(u) &= 4\tau/5 && \text{for } u \in (u_4, u_5) \quad \text{or } u \in (u_6, u_7) \\ P(u) &= \tau && \text{for } u \in (u_5, u_6), \end{aligned} \quad (7)$$

where



**Figure 1** Diffraction pattern obtained for (*a*) the Fibonacci chain and (*b*) the decorated Fibonacci chain. The decorated structure is obtained by applying the substitution rules  $L \rightarrow LLSL$  and  $S \rightarrow SLS$  to the Fibonacci chain. All the diffraction-peak maxima are connected by the envelope functions calculated by Fourier transform of an appropriate probability distribution shown in Figs. 2 and 3. The solid line represents the envelope function for main reflections and the broken lines are for the first-, second- and third-order satellites, respectively.

$$\begin{aligned}
 u_1 &= 7 - 4\tau, & u_2 &= \tau - 1, & u_3 &= 12 - 7\tau, \\
 u_4 &= 4 - 2\tau, & u_5 &= 9 - 5\tau, & u_6 &= 6 - 3\tau, \\
 u_7 &= 2\tau - 2, & u_8 &= 11 - 6\tau, & u_9 &= 3 - \tau, \\
 u_{10} &= 8 - 4\tau.
 \end{aligned}
 \tag{8}$$

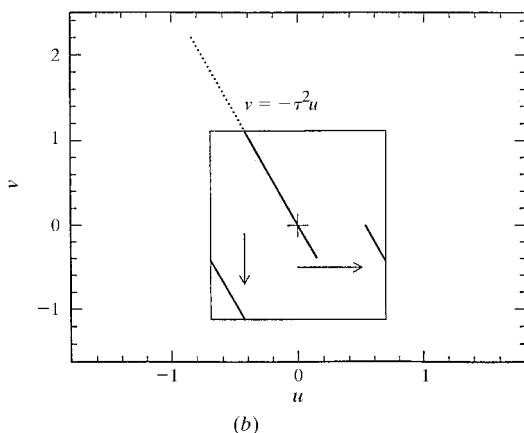
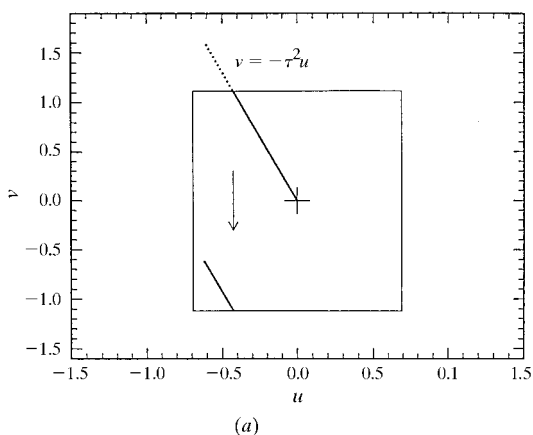
Fourier transform of the above distribution leads to the following expression for the envelope function connecting peak maxima of  $m$ th satellites:

$$\begin{aligned}
 I(k + mq_1)/N^2 &= (\tau^2/25 k^2) \\
 &\times \left( \left\{ \sum_{i=1}^5 [\cos(ku_{i+5}) - \cos(ku_i)] \right\}^2 \right. \\
 &\left. + \left\{ \sum_{i=1}^5 [\sin(ku_{i+5}) - \sin(ku_i)] \right\}^2 \right),
 \end{aligned}
 \tag{9}$$

where

$$q_1(1 + \tau^2)q_0 = 2\pi\tau \approx 10.17. \tag{10}$$

The envelope functions for a decorated structure are shown in Fig. 1(b). One can easily check that the obtained envelope functions properly describe intensities of all Bragg peaks (for



**Figure 2**  
The average unit cell for (a) the Fibonacci chain and (b) the decorated Fibonacci chain. The probability distributions of atomic displacements are zero almost everywhere except the line where  $v = -\tau^2 u$ . Owing to the boundary conditions, the distributions are shifted as indicated by the arrows.

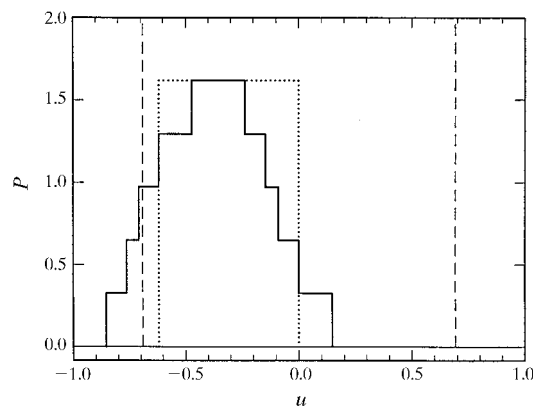
modulated structures, this should be understood as all the main reflections and their satellites) at positions given by (5).

### 3. How to distinguish between decorated structures

From the experimental point of view, one of the most important questions is the following: can the two structures (decorated and undecorated) be distinguished from each other by diffraction analysis? The answer is easy only for perfect structures with perfectly measured diffraction patterns. In reality, however, the structure is neither ideal (because of defects, static as well as dynamic) nor are the measurements free of experimental errors. Usual experimental conditions lead to diffraction patterns consisting of only a few well developed peaks in each direction. From these peaks, one has to judge the decoration, which, at least for quasicrystals, is a nontrivial task. The present approach to the diffraction pattern gives a very useful tool – the envelope function. After appropriate processing of the measured intensities, one can plot the peak intensities *versus* the reduced scattering vector, which is just the normal scattering vector modulo  $q_1$ . For such a plot (Fig. 4), all the experimental data should lie on a single curve describing an appropriate envelope function, which is, of course different for different decorations. Knowing the envelope function, one can easily test some models with different decorations and find the one that fits best to the experimental data. As the envelope function is a smooth function of the scattering vector, it can be determined much more accurately than the individual peaks. In many cases, the accuracy of the envelope function should be sufficient to distinguish between different decorations.

### 4. Concluding remarks

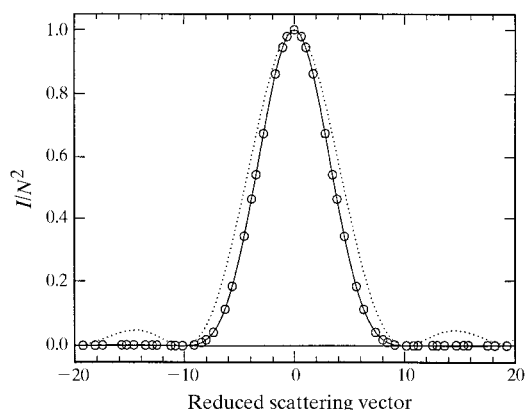
The diffraction pattern of a quasicrystalline structure can be effectively calculated in physical space using the concept of an average unit cell. In such a unit cell, the atoms are replaced by



**Figure 3**  
Probability distributions of atomic distances from the points of a reference lattice corresponding to a scattering vector equal to  $k_0 = 2\pi/a$ . The dotted line is for a Fibonacci chain, the solid line (the step-like function) is for a decorated Fibonacci chain. Dashed vertical lines describe the average unit-cell boundaries.

the probability distributions of atoms around the points of the reference lattice. Fourier transform of such a unit cell gives a set of envelope functions connecting peak maxima of the diffraction pattern. For a Fibonacci chain, the obtained probability distribution has a rectangular shape which leads to a simple expression for the envelope function of the main reflections and their satellites. For a decorated structure, the shapes of the probability distributions are more complex. In the case of the decoration discussed in this paper ( $L \rightarrow LLSL$  and  $S \rightarrow SLS$ ), a step-like function for the probability distribution was determined. Knowing this function, we have obtained and tested an analytical expression for the peak intensities. The obtained envelope function properly describes intensities of all main reflections and their satellites. It is also worth mentioning that the presented approach is not limited to structures for which a higher-dimensional representation is available (quasicrystals and modulated structures). More general structures like a Thue–Morse sequence with a singular-continuous diffraction pattern can be successfully analysed (Wolny, 1998c). Other random structures, like faulted lattices of ZnS (Farkas-Jahnke, 1998) can also be analysed with this approach, which will be published elsewhere.

Using the concept of the envelope function, a new method of elaboration of the experimental data has been suggested. All the measured intensities plotted *versus* reduced scattering vector lie on a single curve (*i.e.* along the envelope function),



**Figure 4** Peak intensities *versus* reduced scattering vector for a decorated Fibonacci chain (open circles). All the intensities lie on the envelope function (solid line). The respective envelope function for a Fibonacci chain is also marked as a dotted line.

which can be effectively used to distinguish between different decorated structures. Such an envelope function is determined directly from the experimental data and no Fourier analysis is required to obtain the curve.

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