

## Average Unit Cell of the Fibonacci Chain

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### Abstract

An average unit cell for the Fibonacci chain has been constructed in physical space. The positions of atoms have been replaced by a statistical probability function of atomic displacements from the nearest point of the reference lattice. An analytical expression for diffraction peak intensities has been derived.

### 1. Introduction

The diffraction pattern of any structure can be obtained by Fourier transformation of its atomic positions. For periodic structures, like single crystals, this transformation can be reduced to the relevant unit cell only, and the diffraction pattern consists of sharp Bragg peaks which coincide with the points of the appropriate reciprocal lattice. Such a concept of the reciprocal lattice is extremely powerful in diffraction analysis and reduces the problem of structure determination to finding atomic positions in the unit cell. However, for aperiodic structures, such a simple unit cell does not exist in physical space and the diffraction pattern is then an average over the whole structure. In the case of quasicrystals and modulated structures, one can recover periodicity of the structure going to higher dimensions (de Bruijn, 1981; Kalugin *et al.*, 1985; Kramer & Neri, 1984; Duneau & Katz, 1985; Elser, 1985; Jaric, 1986; Janssen, 1988; Jagodzinski, 1991; Hof, 1995, 1997; Senechal, 1997). In this approach, it is assumed that the structure is periodic in higher-dimensional space and the projection of a certain part of such an artificial lattice gives the real quasi-periodic structure ('cut and project method'). The periodicity of the higher-dimensional structure allows the conventional analysis of diffraction patterns, which is based on the idea of the reciprocal lattice (Steurer *et al.*, 1993). One can also perform analytical calculations in the perpendicular space. For a Fibonacci chain, a flat window function with a width  $h_0 = \tau^2$ , where  $\tau$  is an irrational number of about 1.618, gives the following expression for the intensity:

$$I = [\sin(w)/w]^2, \quad (1)$$

where

$$w = k^{\text{perp}} h_0 / 2 \quad (2)$$

and

$$\begin{aligned} k &= [2\pi/(\tau^2 + 1)](\alpha_1 \tau + \alpha_2) \\ k^{\text{perp}} &= [2\pi/(\tau^2 + 1)](-\alpha_1 + \alpha_2 \tau). \end{aligned} \quad (3)$$

$\alpha_1$  and  $\alpha_2$  are integers labelling a reciprocal-lattice point in quadratic higher-dimensional space. For the particular direction [1, 1] in two dimensions, the two indices  $\alpha_1$  and  $\alpha_2$  are equal and then (2) can be written as

$$w^{[1,1]} = k/2\tau. \quad (4)$$

Convoluting (4) and (1), one gets the analytical expression for the envelope function  $A$  in physical space (Fig. 1).

However, the physical-space analysis for quasicrystals should be sufficient to get similar results (see Mermin, 1992). Additionally, there are many 'defected' structures, like structures with random displacements of atoms, for which the higher-dimensional lattices do not exist and the appropriate analysis based on the reciprocal lattice is less useful.

In this paper, another approach to diffraction pattern calculations is applied (Wolny, 1992, 1993, 1998*a,b*; Wolny & Pytlik, 1993). In such an approach, a new concept of periodicity is used, which is quite different from the one discussed above. The periodicity has no relation to the structure itself, which can be either periodic or not, and it is simply related to the wavelength of the scattered wave vector. Such natural periodicity of the scattered waves defines the so-called reference lattice. It finally gives new expressions for the structure factor which are based on probability distributions of distances between the atomic positions and the 'points' of the reference lattice.

### 2. Structure factor

It has already been shown (Wolny, 1998*a*) that using the concept of the reference lattice, for  $L$  different types of atoms (with absolute values of scattering power equal to  $f_i$ ) arranged in a cluster of  $N$  atoms, one gets the following expression for the structure factor:

$$\begin{aligned}
 F(\mathbf{k}_0) &= \sum_{n=1}^N f_n \exp(i\mathbf{k}_0 \cdot \mathbf{r}_n) \\
 &= \sum_{n=1}^N f_n \exp(ik_0 u_n) \\
 &= N/L \sum_{l=1}^L f_l \int_{-\lambda/2}^{\lambda/2} P_l(u) \exp(ik_0 u) du, \quad (5)
 \end{aligned}$$

where  $P_l(u)$  is the probability distribution of distances  $u$  (the shortest distance between the atomic position and the appropriate 'point' of the reference lattice) for the  $l$ th type of atom.

Probability distributions  $P_l(u)$  are bounded to the range of  $u$  from  $-\lambda/2$  to  $\lambda/2$  ( $\lambda = 2\pi/k_0$ ) and define the average unit cell. The structure factor is simply calculated by the Fourier transform of such a unit cell. Unfortunately, this cell depends on the scattering vector and, in principle, for each scattering vector one gets another unit cell. However, for any periodic series of scattering vectors ( $k = mk_0, m = 1, 2, 3, 4$ ), the distributions are equivalent in a Fourier space which means that, from the point of view of diffraction analysis, the average unit cell given by an appropriate probability distribution  $P_l(u)$  describes also diffraction intensities for all higher harmonics of a given scattering vector.

Formula (5) describes the structure factor only for a periodic set of scattering vectors and it can be effectively used for analysis of any periodic set of diffraction peaks. For modulated structures (including quasicrystals), there are usually two periods,  $a, b$ , which can also be incommensurate. Using two reference lattices, the first one having periodicity  $a$  and the second one periodicity  $b$ , the structure factor for the sum of two scattering vectors  $k_0 = 2\pi/a, q_0 = 2\pi/b$  can be expressed by

$$\begin{aligned}
 F(k_0 + q_0) &= \sum_{n=1}^N f_n \exp[i(k_0 + q_0)x_n] \\
 &= \sum_{l=1}^L f_l \sum_{n=1}^{N_l} \exp[i(k_0 u_n^l + q_0 v_n^l)] \\
 &= \sum_{l=1}^L f_l \iint P_l(u, v) \exp[i(k_0 u + q_0 v)] du dv, \quad (6)
 \end{aligned}$$

where  $u$  and  $v$  are the shortest distances of the atomic position from the appropriate points of the two reference lattices and  $P(u, v)$  is the corresponding probability distribution, which simply stands for the unit cell. Although the unit cell [*i.e.*  $P(u, v)$ ] is determined for scattering vector  $(k_0 + q_0)$ , it can be also used for a sum of all higher harmonics of  $k_0$  and  $q_0$ , *i.e.*  $nk_0 + mq_0$ , where  $n, m$  are integers. This means that the average unit cell, calculated for the wave vectors of the main structure and its modulation, can be used to calculate the peak intensities of any of the main reflections and its satellites of arbitrary order. Using (6), it is possible to calculate the intensities of all peaks observed in the diffraction patterns.

### 3. Average unit cell

In this section, for simplicity we will suppose that all  $f_l$  are equal to unity, which is the case for the discussed Fibonacci chain. If the lengths of the two building elements of the Fibonacci chain are equal to 1 and  $\tau$ , the average distance is then equal to  $a = 1 + 1/\tau^2 \approx 1.382$ , and the corresponding scattering vector  $k_0 = 2\pi/a \approx 4.547$ . The same scattering centres have been placed at the join interface between consecutive bounds. From the numerical calculations, one gets that the probability distribution (and also the average unit cell) for  $k_0$  is a flat function bounded within  $\{-u_0 + u_0\}$ , where  $u_0 = 1/(2\tau)$ , as is shown in Fig. 2(a). The same function describes all higher harmonics of  $k_0$  ( $k = mk_0$ ) and its Fourier transform is:

$$F(k) = N \sin(ku_0)/ku_0, \quad (7)$$

which leads to the following expression for the envelope function connecting peak maxima of periodic series of diffraction peaks (dashed line A in Fig. 1):

$$I(k)/N^2 = [\sin(ku_0)/ku_0]^2. \quad (8)$$

Formula (8) is similar to (1) and (4) obtained from the higher-dimensional approach. There is also scaling observed for this structure, which results in the appearance of other peaks for  $k_1 = k_0\tau, k_2 = k_0\tau^2$ , and so on. As shown in Fig. 2(b), all the probability distributions for  $k_n = k_0\tau^n$  have a rectangular shape. Performing a Fourier transform (5), one immediately gets an expression for an appropriate envelope function

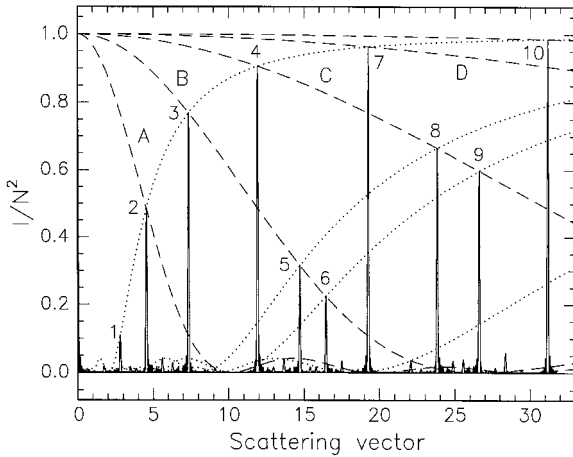


Fig. 1. The diffraction pattern of the Fibonacci chain. The dashed curves have been calculated for the rectangular probability distributions shown in Fig. 2. The dotted lines represent equation (9) and connect diffraction peaks ruled by an inverse Debye-Waller factor.

(e.g. dotted line going through peaks 1, 2, 3, 4, 7 and 10 in Fig. 1):

$$I/N^2 = [\sin(w)/w]^2, \quad \text{where } w = k_0^2 u_0/k. \quad (9)$$

Other envelope functions marked by other dotted lines in Fig. 1 can be obtained from (9) by replacing  $k_0$  by its higher harmonics, i.e.  $2k_0, 3k_0$ . Usually, such envelope functions are calculated in higher dimensions using a perpendicular-space window function. Here it is shown that all the calculations can also be performed in physical space only.

The Fibonacci chain is an example of an incommensurately modulated structure and there are infinitely many periodic series of diffraction peaks. One can call them main reflections or first-order satellites, second-order satellites, and so on. Using the two distances  $a$  and  $b = \tau a$ , the calculated probability distribution  $P(u, v)$  has a rectangular shape along the line given by the relation

$$v = -\tau^2 u - b/2 \quad (10)$$

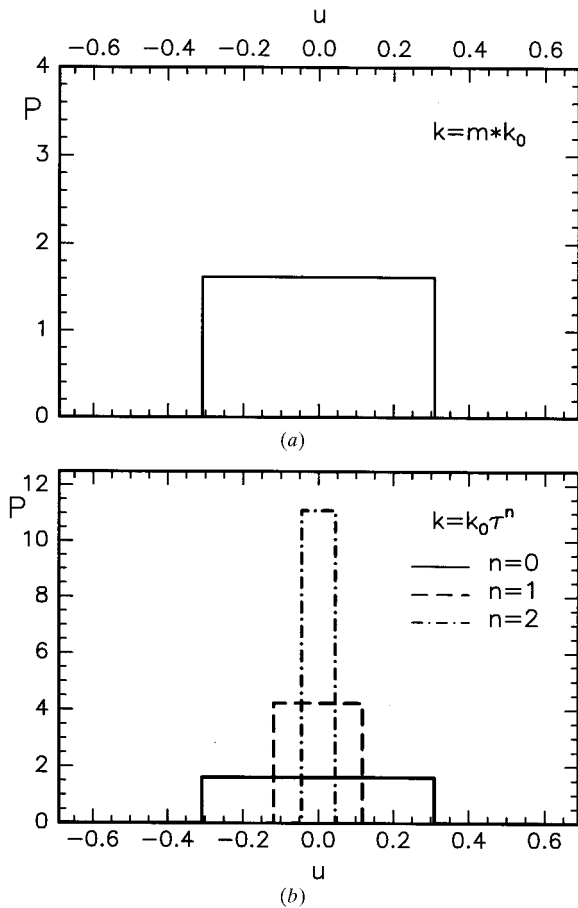


Fig. 2. Average unit cell for (a) a periodic series of scattering vectors ( $k = mk_0, k_0 = 2\pi/a \approx 4.547$ ), (b) scattering vectors given by the scaling relation.

(see also Fig. 3). In this parameter space, the unit cell is bounded to  $(-a/2, a/2)$  for  $u$  and  $(-b/2, b/2)$  for  $v$ . Performing the Fourier transform, one gets the following expression for the intensity of the diffraction peaks and their satellites:

$$I/N^2 = [\sin(w)/w]^2, \quad (11)$$

where

$$w = (nk_0 - \tau^2 mq_0)u_0 = k_0(n - m\tau)u_0 \quad (12)$$

and  $n$  and  $m$  are the indices of main reflection and its satellite;  $u_0 = 1/(2\tau)$ ,  $k_0 = 2\pi/a$ ,  $q_0 = 2\pi/b = k_0/\tau$ . Equations (11) and (12) can be used to calculate the intensity of any peak observed in the diffraction pattern for the Fibonacci chain. One can easily recognize that (12) is identical to (2) for the following substitutions of parameters:

$$\alpha_1 = n + m \quad \text{and} \quad \alpha_2 = n, \quad (13)$$

which means that these two different approaches to diffraction analysis of the Fibonacci chain are fully equivalent. Knowing that the peak position in the diffraction pattern is given by

$$k = nk_0 + mq_0 = k_0(n + m/\tau), \quad (14)$$

one can write (12) in the form

$$w = [k - (1 + \tau^2)mq_0]u_0 = (k - mq_1)u_0. \quad (15)$$

Expression (15), together with (11), has been used to calculate all the envelope functions shown in Fig. 4 and connecting peak maxima of the main reflections ( $m = 0$ ) (full line), and their satellites (up to third order, i.e. for  $m = 1, 2, 3$ ) (broken lines), respectively. The value of  $q_1$ , i.e.

$$q_1 = (1 + \tau^2)q_0 = 2\pi\tau \approx 10.17 \quad (16)$$

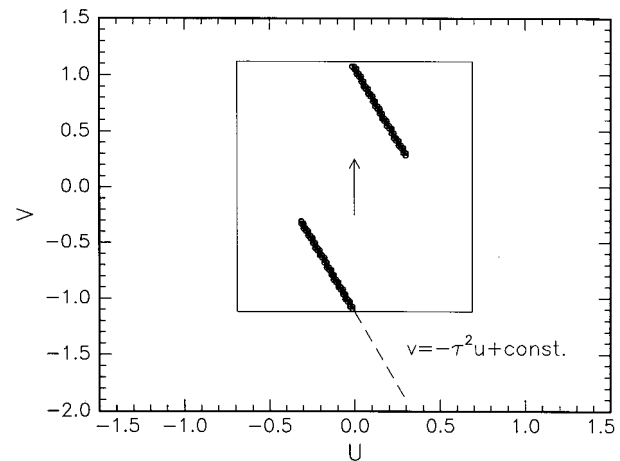


Fig. 3. Average unit cell in a parameter space. The marked rectangle represents the boundary of the discussed unit cell. The probability distribution  $P(u, v)$  is non-zero only along the marked line with a constant value equal to  $1/3^{1/2}$ .

represents the shifts of the envelope functions of the satellites for a given order and corresponds to the same value marked as  $k''$  (Figs. 2 and 4 in Wolny, 1998a) and obtained as an intersection of the physical space with rational direction [1, 1]. More generally speaking, the modulated-structure approach in higher dimensions is always connected with the analysis of the diffraction patterns along some rational direction. By using an appropriate labelling, like that given by (13), one can easily calculate all the diffraction peak intensities in an average unit-cell approach.

#### 4. Concluding remarks

It has been shown that for a Fibonacci chain any peak intensity can be calculated in physical space by the Fourier transform of an appropriate average unit cell. Such a unit cell depends on the scattering vector. The atomic positions of the crystallographic unit cell are replaced by probability distributions of atoms around the points of the reference lattice. For the Fibonacci chain, all of these distributions have a rectangular shape (see Fig. 2) and describe the appropriate average unit cells. Such a property of the average unit cell leads to a simple formula for envelope functions connecting peak intensities of periodic series of diffraction peaks (they are also called peaks ruled by a normal Debye–Waller factor, which is the first approximation to their intensities). In a similar way, one can obtain the envelope function going through the peak maxima of series of peaks described by scaling; their intensities increase with increasing value of the scattering vector and such an envelope function is ruled by an inverse Debye–Waller factor [this name comes from the approximation

which is limited to the second moment of the window function in perpendicular space of higher-dimensional analysis (Wolny, 1992)].

A very simple description of the diffraction pattern of the Fibonacci chain is obtained whenever this chain is regarded as a modulated structure. The diffraction pattern can then be calculated using only a single average unit cell (Fig. 3). The corresponding probability distribution has a rectangular shape along the line given by (10), which allows calculation of the main diffraction peaks and their satellites. The obtained analytical expression for diffraction intensities is similar to that obtained from higher-dimensional analysis. However, the great advantages of the presented approach compared with higher-dimensional analysis are the following: (i) all the calculation has been performed in physical space; (ii) any displacement of the atoms from their perfect positions can be easily incorporated in calculations by simple modification of the average unit cell and of the corresponding probability distribution  $P(u)$ . Such displacements in higher-dimensional analysis are limited only to calculations of the so-called phason contribution.

The presented approach can be easily applied to the analysis of any experimental data. For each direction, one should measure several diffraction intensities which can be used to fit an appropriate probability distribution. In the first approximation, such a distribution can be chosen as a one-parameter Gaussian distribution. For more complicated distributions, a cumulant expansion can be used which leads to a linear equation for some moments of such distribution (Wolny, 1993).

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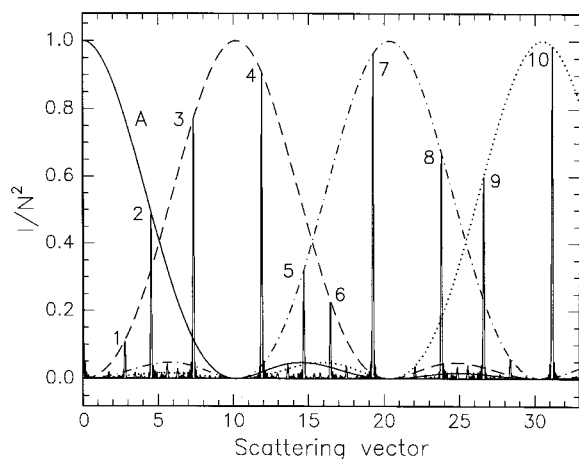


Fig. 4. Diffraction pattern of the Fibonacci chain. All the envelope functions have been calculated by using the average cell shown in Fig. 3. The solid line goes through the main reflections and the broken lines connect peak intensities of the first-, second- and third-order satellites, respectively.

#### References

- Brujin, N. G. de (1981). *Proc. K. Ned. Akad. Wet. Ser. A*, **84**, 39–66.
- Duneau, M. & Katz, A. (1985). *Phys. Rev. Lett.* **54**, 2688–2691.
- Elser, V. (1985). *Phys. Rev. B*, **32**, 4892–4898.
- Hof, A. (1995). *J. Phys. A*, **28**, 57–62.
- Hof, A. (1997). *The Mathematics of Long-Range Aperiodic Order*, edited by R. V. Moody. *NATO ASI Series, Series C: Mathematical and Physical Sciences*, Vol. 489, pp. 239–268.
- Jagodzinski, H. (1991). *Z. Kristallogr.* **196**, 1–19.
- Janssen, T. (1988). *Phys. Rep.* **168**, No. 2, 55–113.
- Jaric, M. V. (1986). *Phys. Rev. B*, **34**, 4685–4698.
- Kalugin, P. A., Kitaev, A. Yu. & Levitov, L. S. (1985). *J. Phys. Lett.* **46**, L601–L607.
- Kramer, P. & Neri, R. (1984). *Acta Cryst.* **A40**, 580–587.
- Mermin, N. D. (1992). *Phys. Rev. Lett.* **68**, 1172–1175.

- Senechal, M. (1997). *The Mathematics of Long-Range Aperiodic Order*, edited by R. V. Moody. *NATO ASI Series, Series C: Mathematical and Physical Sciences*, Vol. 489, pp. 521–548.
- Steurer, W., Haibach, T., Zhang, B., Kek, S. & Lück, R. (1993). *Acta Cryst.* **B49**, 661–675.
- Wolny, J. (1992). *Acta Cryst.* **A48**, 918–921.
- Wolny, J. (1993). *J. Phys. Condens. Matter*, **5**, 6663–6672.
- Wolny, J. (1998a). *Philos. Mag.* **A77**, 395–412.
- Wolny, J. (1998b). *Aperiodic '97*. In the press.
- Wolny, J. & Pytlik, L. (1993). *Phase Transit.* **43**, 67–72.