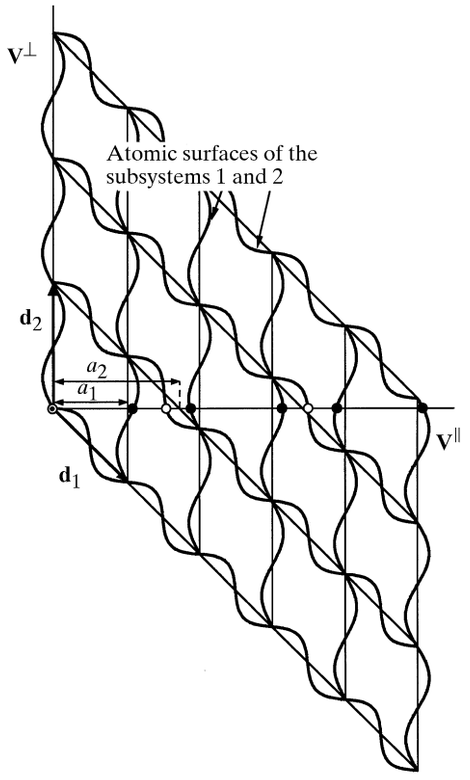

Fig. 4.6.2.5. 2D embedding of a 1D composite structure with mutual interaction of the subsystems causing modulations. Filled and empty circles represent the modulated substructures with periods a_1 and a_2 of the basic substructures, respectively. The atoms result from the parallel-space cut of the sinusoidal atomic surfaces running parallel to $\mathbf{d}_1$ and $\mathbf{d}_2$.

in both the 2D description $\mathbf{H} = h_1\mathbf{d}_1^* + h_2\mathbf{d}_2^*$ and in the 1D physical-space description with two parallel basis vectors $\mathbf{H}^{\parallel} = h_1\mathbf{a}_1^* + h_2\mathbf{a}_2^*$.

The reciprocal-lattice points $\mathbf{H} = h_1\mathbf{d}_1^*$ and $\mathbf{H} = h_2\mathbf{d}_2^*$, h_1, $h_2 \in \mathbb{Z}$, on the main axes $\mathbf{d}_1^*$ and $\mathbf{d}_2^*$ are the main reflections of the two substructures. All other reflections are referred to as satellite reflections. Their intensities differ from zero only in the case of modulated substructures. Each reflection of one subsystem coincides with exactly one reflection of the other subsystem.

4.6.2.4. 1D quasiperiodic structures

The Fibonacci sequence, the best investigated example of a 1D quasiperiodic structure, can be obtained from the substitution rule σ: S $\rightarrow$ L, L $\rightarrow$ LS, replacing the letter S by L and the letter L by the word LS (see *e.g.* Luck *et al.*, 1993). Applying the substitution matrix

$$S = \begin{pmatrix} 0 & 1 \\ 1 & 1 \end{pmatrix}$$

associated with σ, this rule can be written in the form

$$\begin{pmatrix} \mathrm{S} \\ \mathrm{L} \end{pmatrix} \rightarrow \begin{pmatrix} 0 & 1 \\ 1 & 1 \end{pmatrix}\begin{pmatrix} \mathrm{S} \\ \mathrm{L} \end{pmatrix} = \begin{pmatrix} \mathrm{L} \\ \mathrm{L+S} \end{pmatrix}.$$

S gives the sum of letters, L + S = S + L, and not their order. Consequently, the same substitution matrix can also be applied, for instance, to the substitution σ': S $\rightarrow$ L, L $\rightarrow$ SL. The repeated action of S on the alphabet $A = \{\mathrm{S}, \mathrm{L}\}$ yields the words $A_n = \sigma^n(\mathrm{S})$ and $B_n = \sigma^n(\mathrm{L}) = A_{n+1}$ as illustrated in Table 4.6.2.1. The frequencies of letters contained in the words A_n and B_n can

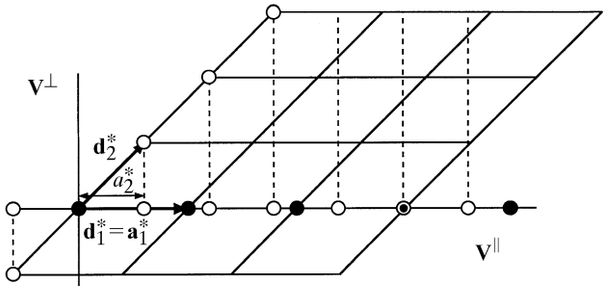

Fig. 4.6.2.6. Schematic representation of the reciprocal space of the embedded 1D composite structure depicted in Fig. 4.6.2.4. Filled and empty circles represent the reflections generated by the substructures with periods a_1 and a_2, respectively. The actual 1D diffraction pattern of the 1D CS results from a projection of the 2D reciprocal space onto the parallel space. The correspondence between 2D reciprocal-lattice positions and their projected images is indicated by dashed lines.

be calculated by applying the nth power of the transposed substitution matrix on the unit vector. From

$$\begin{pmatrix} \nu_{n+1}^A \\ \nu_{n+1}^B \end{pmatrix} = S^T \begin{pmatrix} \nu_n^A \\ \nu_n^B \end{pmatrix}$$

it follows that

$$\begin{pmatrix} \nu_n^A \\ \nu_n^B \end{pmatrix} = (S^T)^n \begin{pmatrix} 1 \\ 1 \end{pmatrix}.$$

In the case of the Fibonacci sequence, ν_n^B gives the total number of letters S and L, and ν_n^A the number of letters L.

An infinite Fibonacci sequence, *i.e.* a word B_n with $n \rightarrow \infty$, remains invariant under inflation (deflation). Inflation (deflation) means that the number of letters L, S increases (decreases) under the action of the (inverted) substitution matrix S. Inflation and deflation represent self-similarity (scaling) symmetry operations on the infinite Fibonacci sequence. A more detailed discussion of the scaling properties of the Fibonacci chain in direct and reciprocal space will be given later.

The Fibonacci numbers $F_n = F_{n-1} + F_{n-2}$ form a series with $\lim_{n\rightarrow\infty}(F_{n+1}/F_n) = \tau$ {the golden mean $\tau = [1 + (5)^{1/2}]/2 = 2\cos(\pi/5) = 1.618\ldots$}. The ratio of the frequencies of L and S

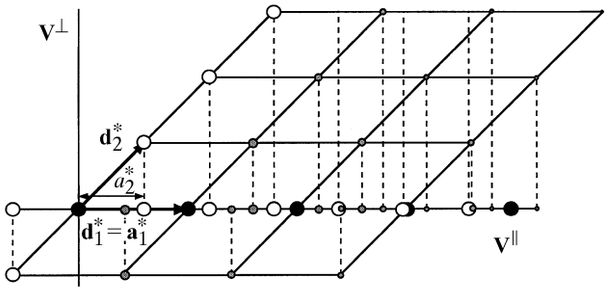

Fig. 4.6.2.7. Schematic representation of the reciprocal space of the embedded 1D composite structure depicted in Fig. 4.6.2.5. Filled and empty circles represent the main reflections of the two subsystems. The satellite reflections generated by the modulated substructures are shown as grey circles. The diameters of the circles are roughly proportional to the intensities of the reflections. The actual 1D diffraction pattern of the 1D CS results from a projection of the 2D reciprocal space onto the parallel space. The correspondence between 2D reciprocal-lattice positions and their projected images is indicated by dashed lines.

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Table 4.6.2.1. *Expansion of the Fibonacci sequence* $B_n = \sigma^n(\mathrm{L})$ *by repeated action of the substitution rule* σ: $\mathrm{S} \rightarrow \mathrm{L}$, $\mathrm{L} \rightarrow \mathrm{LS}$

ν_L, ν_S are the frequencies of the letters L and S in word B_n.

B_n	ν_L	ν_S	n
L	1	0	0
LS	1	1	1
LSL	2	1	2
LSLLS	3	2	3
LSLLSLSL	5	3	4
LSLLSLSLLSLLS	8	5	5
LSLLSLSLLSLLSLSLLSLSL	13	8	6
	⋮	⋮	⋮
	F_{n+1}	F_n	n

in the Fibonacci sequence converges to τ if the sequence goes to infinity. The continued fraction expansion of the golden mean τ,

$$\tau = 1 + \cfrac{1}{1 + \cfrac{1}{1 + \cfrac{1}{1 + \ldots}}},$$

contains only the number 1. This means that τ is the 'most irrational' number, *i.e.* the irrational number with the worst truncated continued fraction approximation to it. This might be one of the reasons for the stability of quasiperiodic systems, where τ plays a role. The strong irrationality may impede the lock-in into commensurate systems (*rational approximants*).

By associating intervals (*e.g.* atomic distances) with length ratio τ to 1 to the letters L and S, a quasiperiodic structure $s(\mathbf{r})$ (*Fibonacci chain*) can be obtained. The invariance of the ratio of lengths $\mathrm{L/S} = (\mathrm{L} + \mathrm{S})/\mathrm{L} = \tau$ is responsible for the invariance of the Fibonacci chain under scaling by a factor τ^n, $n \in \mathbb{Z}$. Owing to a minimum atomic distance S in real crystal structures, the full set of inverse symmetry operators τ^{-n} does not exist. Consequently, the set of scaling operators $s = \{\tau^0 = 1, \tau^1, \ldots\}$ forms only a semi-group, *i.e.* an associative groupoid. Groupoids are the most general algebraic sets satisfying only one of the group axioms: the associative law. The scaling properties of the Fibonacci sequence can be derived from the eigenvalues of the scaling matrix S. For this purpose the equation

$$\det|S - \lambda I| = 0$$

with eigenvalue λ and unit matrix I has to be solved. The evaluation of the determinant yields the characteristic polynomial

$$\lambda^2 - \lambda - 1 = 0,$$

yielding in turn the eigenvalues $\lambda_1 = [1 + (5)^{1/2}]/2 = \tau$, $\lambda_2 = [1 - (5)^{1/2}]/2 = -1/\tau$ and the eigenvectors

$$\mathbf{w}_1 = \begin{pmatrix} 1 \\ \tau \end{pmatrix}, \quad \mathbf{w}_2 = \begin{pmatrix} 1 \\ -1/\tau \end{pmatrix}.$$

Rewriting the eigenvalue equation gives for the first (*i.e.* the largest) eigenvalue

$$\begin{pmatrix} 0 & 1 \\ 1 & 1 \end{pmatrix}\begin{pmatrix} 1 \\ \tau \end{pmatrix} = \begin{pmatrix} \tau \\ 1 + \tau \end{pmatrix} = \begin{pmatrix} \tau \\ \tau^2 \end{pmatrix} = \tau\begin{pmatrix} 1 \\ \tau \end{pmatrix}.$$

Identifying the eigenvector

$$\begin{pmatrix} 1 \\ \tau \end{pmatrix}$$

with

$$\begin{pmatrix} \mathrm{S} \\ \mathrm{L} \end{pmatrix}$$

shows that an infinite Fibonacci sequence $s(\mathbf{r})$ remains invariant under scaling by a factor τ. This scaling operation maps each new lattice vector $\tau\mathbf{r}$ upon a vector $\mathbf{r}$ of the original lattice:

$$s(\tau\mathbf{r}) = s(\mathbf{r}).$$

Considering periodic lattices, these eigenvalues are integer numbers. For quasiperiodic 'lattices' (*quasilattices*) they always correspond to *algebraic numbers* (*Pisot numbers*). A Pisot number is the solution of a polynomial equation with integer coefficients. It is larger than one, whereas the modulus of its conjugate is smaller than unity: $\lambda_1 > 1$ and $|\lambda_2| < 1$ (Luck *et al.*, 1993). The total lengths l_n^A and l_n^B of the words A_n, B_n can be determined from the equations $l_n^A = \lambda_1^n l^A$ and $l_n^B = \lambda_1^n l^B$ with the eigenvalue λ_1. The left Perron–Frobenius eigenvector $\mathbf{w}_1$ of S, *i.e.* the left eigenvector associated with λ_1, determines the ratio S:L to 1:τ. The right Perron–Frobenius eigenvector $\mathbf{w}_1$ of S associated with λ_1 gives the relative frequencies, 1 and τ, for the letters S and L (for a definition of the Perron–Frobenius theorem see Luck *et al.*, 1993, and references therein).

The general case of an alphabet $A = \{\mathrm{L}_1 \ldots \mathrm{L}_k\}$ with k letters (intervals) L_i, of which at least two are on incommensurate length scales and which transform with the substitution matrix S,

$$\mathrm{L}_i' \rightarrow \sum_{j=1}^{k} S_{ij}\mathrm{L}_j,$$

can be treated analogously. S is a $k \times k$ matrix with non-negative integer coefficients. Its eigenvalues are solutions of a polynomial equation of rank k with integer coefficients (algebraic or Pisot numbers). The dimension n of the embedding space is generically equal to the number of letters (intervals) k involved by the substitution rule. From substitution rules, infinitely many different 1D quasiperiodic sequences can be generated. However, their atomic surfaces in the nD description are generically of fractal shape (see Section 4.6.2.5).

The quasiperiodic 1D density distribution $\rho(\mathbf{r})$ of the Fibonacci chain can be represented by the Fourier series

$$\rho(\mathbf{r}) = (1/V)\sum_{\mathbf{H}^\parallel} F(\mathbf{H}^\parallel)\exp(-2\pi i\mathbf{H}^\parallel \cdot \mathbf{r}),$$

with $\mathbf{H}^\parallel \in \mathbb{R}$ (the set of real numbers). The Fourier coefficients $F(\mathbf{H}^\parallel)$ form a Fourier module $M^* = \{\mathbf{H}^\parallel = \sum_{i=1}^{2} h_i\mathbf{a}_i^* | h_i \in \mathbb{Z}\}$ equivalent to a $\mathbb{Z}$ module of rank 2. Thus a periodic function in 2D space can be defined by

$$\rho(\mathbf{r}^\parallel, \mathbf{r}^\perp) = (1/V)\sum_{\mathbf{H}} F(\mathbf{H})\exp[-2\pi i(\mathbf{H}^\parallel \cdot \mathbf{r}^\parallel + \mathbf{H}^\perp \cdot \mathbf{r}^\perp)],$$

where $\mathbf{r} = (\mathbf{r}^\parallel, \mathbf{r}^\perp) \in \Sigma$ and $\mathbf{H} = (\mathbf{H}^\parallel, \mathbf{H}^\perp) \in \Sigma^*$ are, by construction, direct- and reciprocal-lattice vectors (Figs. 4.6.2.8 and 4.6.2.9):

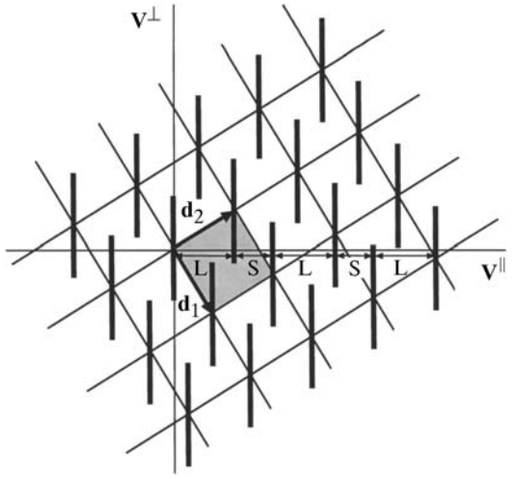

Fig. 4.6.2.8. 2D embedding of the Fibonacci chain. The short and long distances S and L, generated by the intersection of the atomic surfaces with the physical space $\mathbf{V}^\parallel$, are indicated. The atomic surfaces are represented by bars parallel to $\mathbf{V}^\perp$. Their lengths correspond to the projection of one unit cell (shaded) upon $\mathbf{V}^\perp$.

$$\mathbf{r} = n_1\mathbf{d}_1 + n_2\mathbf{d}_2, \text{ with } \mathbf{d}_1 = \frac{1}{a^*(2+\tau)}\begin{pmatrix}1\\-\tau\end{pmatrix}_V,$$

$$\mathbf{d}_2 = \frac{1}{a^*(2+\tau)}\begin{pmatrix}\tau\\1\end{pmatrix}_V;$$

$$\mathbf{H} = h_1\mathbf{d}_1^* + h_2\mathbf{d}_2^*, \text{ with } \mathbf{d}_1^* = a^*\begin{pmatrix}1\\-\tau\end{pmatrix}_V, \mathbf{d}_2^* = a^*\begin{pmatrix}\tau\\1\end{pmatrix}_V.$$

The 1D Fibonacci chain results from the cut of the parallel (physical) space with the 2D lattice Σ decorated with line elements for the *atomic surfaces* (*acceptance domains*). In this description, the atomic surfaces correspond simply to the projection of one 2D unit cell upon the perpendicular-space coordinate. This satisfies the condition that each unit cell contributes exactly to one point of the Fibonacci chain (*primitive unit cell*). The physical space $\mathbf{V}^\parallel$ is related to the eigenspace of the substitution matrix S associated with its eigenvalue $\lambda_1 = \tau$. The perpendicular space $\mathbf{V}^\perp$ corresponds to the eigenspace of the substitution matrix S associated with its eigenvalue $\lambda_2 = -1/\tau$. Thus, the physical space scales to powers of τ and the perpendicular space to powers of $-1/\tau$.

By block-diagonalization, the reducible substitution (scaling) matrix S can be decomposed into two non-equivalent irreducible representations. These can be assigned to the two 1D orthogonal subspaces $\mathbf{V}^\parallel$ and $\mathbf{V}^\perp$ forming the 2D embedding space $\mathbf{V} = \mathbf{V}^\parallel \oplus \mathbf{V}^\perp$. Thus, using $WSW^{-1} = S_V = S_V^\parallel \oplus S_V^\perp$, where

$$W = \begin{pmatrix}1 & \tau\\ -\tau & 1\end{pmatrix} = \begin{pmatrix}\mathbf{d}_1^* & \mathbf{d}_2^*\end{pmatrix},$$

one obtains

$$\begin{pmatrix}1 & \tau\\ -\tau & 1\end{pmatrix}\begin{pmatrix}0 & 1\\ 1 & 1\end{pmatrix}_D\begin{pmatrix}1 & \tau\\ -\tau & 1\end{pmatrix}^{-1} = \left(\begin{array}{c|c}\tau & 0\\ \hline 0 & -1/\tau\end{array}\right)_V = \left(\begin{array}{c|c}S^\parallel & 0\\ \hline 0 & S^\perp\end{array}\right)_V,$$

the scaling operations $S^\parallel$ and $S^\perp$ in parallel and in perpendicular space as indicated by the partition lines.

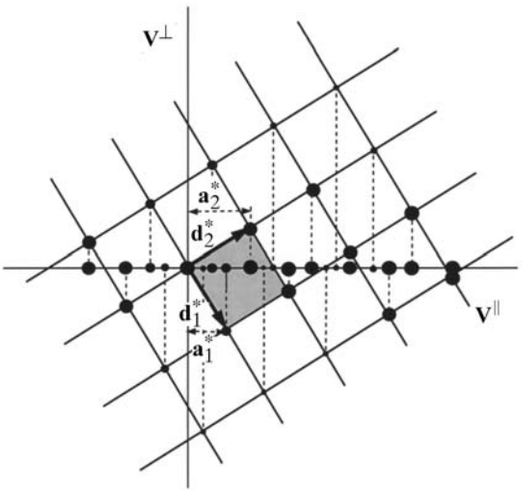

Fig. 4.6.2.9. Schematic representation of the reciprocal space of the embedded Fibonacci chain depicted in Fig. 4.6.2.8. The physical-space reciprocal basis $\mathbf{a}_1^*$ and $\mathbf{a}_2^*$ is marked. The diameters of the filled circles are roughly proportional to the reflection intensities. One 2D reciprocal-lattice unit cell is shadowed. The actual 1D diffraction pattern of the 1D Fibonacci chain results from a projection of the 2D reciprocal space onto the parallel space. The correspondence between 2D reciprocal-lattice positions and their projected images is indicated by dashed lines.

The metric tensors for the reciprocal and the direct 2D square lattices read

$$G^* = |a^*|^2(2+\tau)\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix} \text{ and } G = \frac{1}{|a^*|^2(2+\tau)}\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}.$$

The short distance S of the Fibonacci sequence is related to a^* by

$$\begin{aligned} S &= 1/[a^*(2+\tau)]\\ &= \min\left\{\left|\pi^\parallel(\mathbf{d}_i - \mathbf{d}_j)\right| \,\middle|\, \left|\pi^\perp(\mathbf{d}_i - \mathbf{d}_j)\right| < \Omega_{\rm AS} \wedge i, j \in \mathbb{Z}\right\}, \end{aligned}$$

with the projectors $\pi^\parallel$ and $\pi^\perp$ onto $\mathbf{V}^\parallel$ and $\mathbf{V}^\perp$. The *point density* $\rho_{\rm p}$ of the Fibonacci chain, *i.e.* the number of vertices per unit length, can be calculated using the formula

$$\rho_{\rm p} = \frac{\Omega_{\rm AS}}{\Omega_{\rm UC}} = \frac{(1+\tau)/[a^*(2+\tau)]}{1/[|a^*|^2(2+\tau)]} = a^*\tau^2,$$

where $\Omega_{\rm AS}$ and $\Omega_{\rm UC}$ are the areas of the atomic surface and of the 2D unit cell, respectively.

For an infinite Fibonacci sequence generated from the intervals S and L an average distance d can be calculated:

$$d = \lim_{n\to\infty}\frac{F_n\mathrm{S} + F_{n+1}\mathrm{L}}{F_n + F_{n+1}} = \lim_{n\to\infty}\frac{F_n(\mathrm{S}+\tau\mathrm{L})}{F_n(1+\tau)} = \frac{\mathrm{S}(1+\tau^2)}{(1+\tau)} = (3-\tau)\mathrm{S}.$$

Therefrom, the point density can also be calculated:

$$\rho_{\rm p} = 1/d = 1/[(3-\tau)\mathrm{S}] = [a^*(2+\tau)]/(3-\tau) = a^*\tau^2.$$

An approximant structure of the Fibonacci sequence with a unit cell containing m intervals L and n intervals S can be generated by shearing the 2D lattice Σ by the shear matrix S_m,

$$S_m = \frac{1}{2+\tau}\begin{pmatrix} \tau^2 + x\tau + 1 & -x \\ x\tau^2 & \tau^2 - x\tau + 1 \end{pmatrix}_D,$$

where $x = (n\tau - m)/(m\tau + n)$:

$$\begin{aligned}
\mathbf{d}_i' &= \sum_{j=1}^{2} S_{mij}\mathbf{d}_j; \\
\mathbf{d}_1' &= \frac{1}{2+\tau}\left[(\tau^2 + x\tau + 1)\mathbf{d}_1 - x\mathbf{d}_2\right] \\
&= \frac{1}{(2+\tau)a^*}\begin{pmatrix} 1 \\ -\tau - x \end{pmatrix}_V \\
&= \frac{1}{(2+\tau)a^*}\begin{pmatrix} 1 \\ -\dfrac{2n\tau + m\tau}{m\tau + n} \end{pmatrix}_V, \\
\mathbf{d}_2' &= \frac{1}{2+\tau}\left[x\tau^2\mathbf{d}_1 + (\tau^2 - x\tau + 1)\mathbf{d}_2\right] \\
&= \frac{1}{(2+\tau)a^*}\begin{pmatrix} \tau \\ -x\tau + 1 \end{pmatrix}_V \\
&= \frac{1}{(2+\tau)a^*}\begin{pmatrix} \tau \\ \dfrac{2m\tau - n\tau}{m\tau + n} \end{pmatrix}_V.
\end{aligned}$$

This shear matrix does not change the magnitudes of the intervals L and S. In reciprocal space the inverted and transposed shear matrix is applied on the reciprocal basis,

$$(S_m^{-1})^T = \frac{1}{2+\tau}\begin{pmatrix} \tau^2 - x\tau + 1 & -x\tau^2 \\ x & \tau^2 + x\tau + 1 \end{pmatrix}_D,$$

where $x = (n\tau - m)/(m\tau + n)$:

$$\begin{aligned}
\mathbf{d}_i^{*'} &= \sum_{j=1}^{2} (S_m^{-1})_{ij}^T \mathbf{d}_j^*; \\
\mathbf{d}_1^{*'} &= \frac{1}{2+\tau}\left[(\tau^2 - x\tau + 1)\mathbf{d}_1^* - x\tau^2\mathbf{d}_2^*\right] \\
&= a^*\begin{pmatrix} 1 - x\tau \\ -\tau \end{pmatrix}_V \\
&= a^*\begin{pmatrix} \dfrac{2m\tau - n\tau}{m\tau + n} \\ -\tau \end{pmatrix}_V, \\
\mathbf{d}_2^{*'} &= \frac{1}{2+\tau}\left[x\mathbf{d}_1^* + (\tau^2 + x\tau + 1)\mathbf{d}_2^*\right] \\
&= a^*\begin{pmatrix} \tau + x \\ 1 \end{pmatrix}_V \\
&= a^*\begin{pmatrix} \dfrac{2n\tau + m\tau}{m\tau + n} \\ 1 \end{pmatrix}_V.
\end{aligned}$$

The point $x_n(t)$ of the nth interval L or S of an infinite Fibonacci sequence is given by

$$x_n(t) = \{x_0 + n(3 - \tau) - (\tau - 1)[\mathrm{frac}(n\tau + t) - (1/2)]\}\mathrm{S},$$

where t is the phase of the modulation function $y(t) = (\tau - 1)[\mathrm{frac}(n\tau + t) - (1/2)]$ (Janssen, 1986). Thus, the Fibonacci sequence can also be dealt with as an incommensurately modulated structure. This is a consequence of the fact that for 1D structures only the crystallographic point symmetries 1 and $\bar{1}$ allow the existence of a periodic average structure.

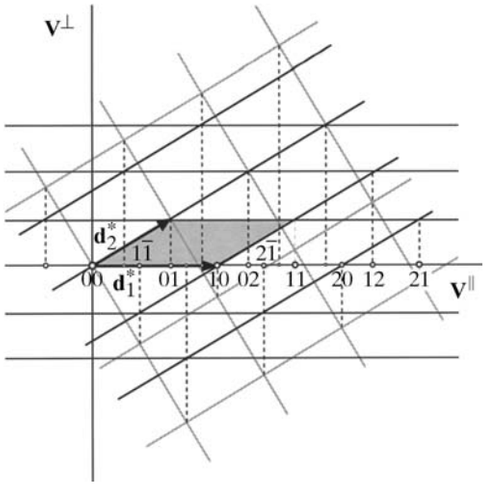

Fig. 4.6.2.10. Reciprocal space of the embedded Fibonacci chain as a modulated structure. Several main and satellite reflections are indexed. The square reciprocal lattice of the quasicrystal description illustrated in Fig. 4.6.2.9 is indicated by grey lines. The reflections located on $\mathbf{V}^\parallel$ can be considered to be projected either from the 2D square lattice of the embedding as for a QS or from the 2D oblique lattice of the embedding as for an IMS.

The embedding of the Fibonacci chain as an incommensurately modulated structure can be performed as follows:

(1) select a subset $\Lambda^* \subset M^*$ of strong reflections for main reflections $\mathbf{H} = h\mathbf{a}^*, h \in \mathbb{Z}$;

(2) define a satellite vector $\mathbf{q} = \alpha\mathbf{a}^*$ pointing from each main reflection to the next satellite reflection.

One possible way of indexing based on the same $\mathbf{a}^*$ as defined above is illustrated in Fig. 4.6.2.10. The scattering vector is given by $\mathbf{H}^\parallel = h(\tau + 1)\mathbf{a}^* + mq$, where $\mathbf{q} = \tau\mathbf{a}^*$, or, in the 2D representation, $\mathbf{H} = h_1\mathbf{d}_1^* + h_2\mathbf{d}_2^*$, where

$$\mathbf{d}_1^* = a^*\begin{pmatrix} 1 + \tau \\ 0 \end{pmatrix}_V$$

and

$$\mathbf{d}_2^* = a^*\begin{pmatrix} \tau \\ 1 \end{pmatrix}_V,$$

with the direct basis

$$\mathbf{d}_1 = \frac{1}{a^*(1+\tau)}\begin{pmatrix} 1 \\ -\tau \end{pmatrix}_V, \quad \mathbf{d}_2 = \frac{1}{a^*}\begin{pmatrix} 0 \\ 1 \end{pmatrix}_V.$$

The modulation function is saw-tooth-like (Fig. 4.6.2.11).

4.6.2.5. 1D structures with fractal atomic surfaces

A 1D structure with a *fractal atomic surface* (Hausdorff dimension 0.9157...) can be derived from the Fibonacci sequence by squaring its substitution matrix S:

references