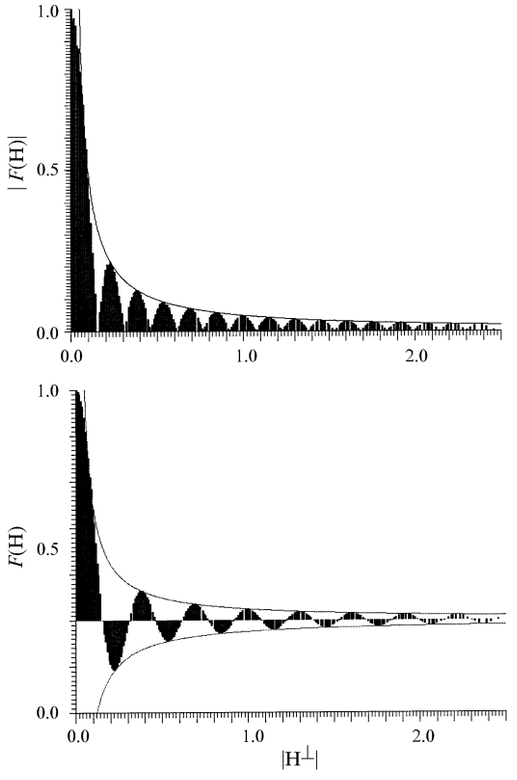


Fig. 4.6.3.6. The structure factors $F(\mathbf{H})$ (below) and their magnitudes $|F(\mathbf{H})|$ (above) of a Fibonacci chain decorated with equal point atoms are shown as a function of the perpendicular-space component $|\mathbf{H}^{\perp}|$ of the diffraction vector. The short distance in the Fibonacci chain is S = 2.5 Å, all structure factors within $0 \leq |\mathbf{H}| \leq 2.5$ Å$^{-1}$ have been calculated and normalized to $F(00) = 1$.

### 4.6.3.3.1.3. *Structure factor*

The structure factor of a periodic structure is defined as the Fourier transform of the density distribution $\rho(\mathbf{r})$ of its unit cell (UC):

$$F(\mathbf{H}) = \int_{\mathrm{UC}} \rho(\mathbf{r}) \exp(2\pi i \mathbf{H}\cdot\mathbf{r})\, \mathrm{d}\mathbf{r}.$$

The same is valid in the case of the $n$D description of a quasiperiodic structure. The parallel- and perpendicular-space components are orthogonal to each other and can be separated. In the case of the 1D Fibonacci sequence, the Fourier transform of the parallel-space component of the electron-density distribution of a single atom gives the usual atomic scattering factor $f(\mathbf{H}^{\parallel})$. Parallel to $x^{\perp}$, $\rho(\mathbf{r})$ adopts values $\neq 0$ only within the interval $-(1+\tau)/[2a^*(2+\tau)] \leq x^{\perp} \leq (1+\tau)/[2a^*(2+\tau)]$ and one obtains

$$F(\mathbf{H}) = f(\mathbf{H}^{\parallel})[a^*(2+\tau)]/(1+\tau) \times \int_{-(1+\tau)/[2a^*(2+\tau)]}^{+(1+\tau)/[2a^*(2+\tau)]} \exp(2\pi i \mathbf{H}^{\perp}\cdot x^{\perp})\, \mathrm{d}x^{\perp}.$$

The factor $[a^*(2+\tau)]/(1+\tau)$ results from the normalization of the structure factors to $F(\mathbf{0}) = f(0)$. With

$$\mathbf{H} = h_1\mathbf{d}_1^* + h_2\mathbf{d}_2^* + h_3\mathbf{d}_3^* + h_4\mathbf{d}_4^* = h_1 a_1^* \begin{pmatrix} 1 \\ -\tau \\ 0 \\ 0 \end{pmatrix} + h_2 a_1^* \begin{pmatrix} \tau \\ 1 \\ 0 \\ 0 \end{pmatrix} + h_3 a_3^* \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix} + h_4 a_4^* \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}$$

and $\mathbf{H}^{\perp} = a_1^*(-\tau h_1 + h_2)$ the integrand can be rewritten as

$$F(\mathbf{H}) = f(\mathbf{H}^{\parallel})[a^*(2+\tau)]/(1+\tau) \times \int_{-(1+\tau)/[2a^*(2+\tau)]}^{+(1+\tau)/[2a^*(2+\tau)]} \exp[2\pi i(-\tau h_1 + h_2)x^{\perp}]\, \mathrm{d}x^{\perp},$$

yielding

$$F(\mathbf{H}) = f(\mathbf{H}^{\parallel})(2+\tau)/[2\pi i(-\tau h_1 + h_2)(1+\tau)] \times \exp[2\pi i(-\tau h_1 + h_2)x^{\perp}]\big|_{-(1+\tau)/[2a^*(2+\tau)]}^{+(1+\tau)/[2a^*(2+\tau)]}.$$

Using $\sin x = (\mathrm{e}^{ix} - \mathrm{e}^{-ix})/2i$ gives

$$F(\mathbf{H}) = f(\mathbf{H}^{\parallel})(2+\tau)/[\pi(-\tau h_1 + h_2)(1+\tau)] \times \sin[\pi(1+\tau)(-\tau h_1 + h_2)]/(2+\tau).$$

Thus, the structure factor has the form of the function $\sin(x)/x$ with $x$ a perpendicular reciprocal-space coordinate. The upper and lower limiting curves of this function are given by the hyperbolae $\pm 1/x$ (Fig. 4.6.3.6). The continuous shape of $F(\mathbf{H})$ as a function of $\mathbf{H}^{\perp}$ allows the estimation of an overall temperature factor and atomic scattering factor for reflection-data normalization (compare Figs. 4.6.3.6 and 4.6.3.7).

In the case of a 3D crystal structure which is quasiperiodic in one direction, the structure factor can be written in the form

$$F(\mathbf{H}) = \sum_{k=1}^{n} \Big[ T_k(\mathbf{H}) f_k(\mathbf{H}^{\parallel}) g_k(\mathbf{H}^{\perp}) \exp(2\pi i \mathbf{H}\cdot\mathbf{r}_k) \Big].$$

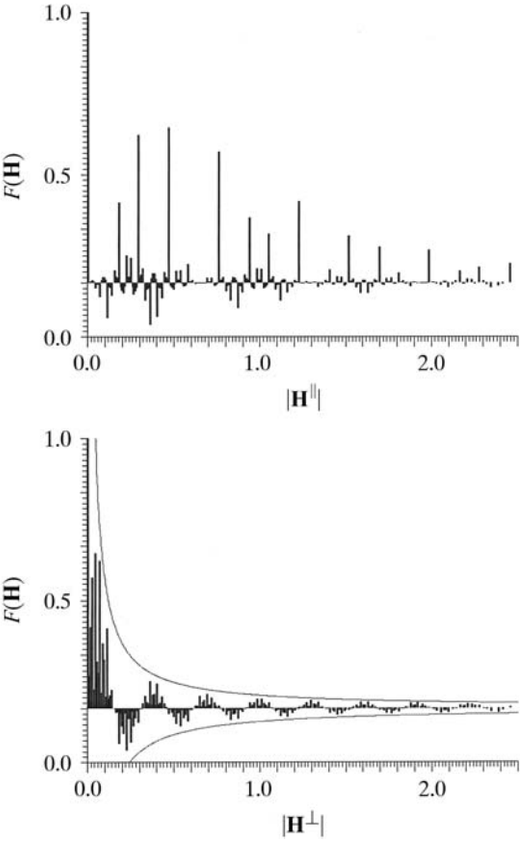


Fig. 4.6.3.7. The structure factors $F(\mathbf{H})$ of the Fibonacci chain decorated with aluminium atoms ($U_{\mathrm{overall}} = 0.005$ Å$^2$) as a function of the parallel (above) and the perpendicular (below) component of the diffraction vector. The short distance is S = 2.5 Å, all structure factors within $0 \leq |\mathbf{H}| \leq 2.5$ Å$^{-1}$ have been calculated and normalized to $F(00) = 1$.

The sum runs over all $n$ averaged hyperatoms in the 4D unit cell of the structure. The *geometric form factor* $g_k(\mathbf{H}^\perp)$ corresponds to the Fourier transform of the $k$th atomic surface,

$$g_k(\mathbf{H}^\perp) = (1/A^\perp_{\rm UC}) \int_{A_k} \exp(2\pi i \mathbf{H}^\perp \cdot \mathbf{r}^\perp)\, {\rm d}\mathbf{r}^\perp,$$

normalized to $A^\perp_{\rm UC}$, the area of the 2D unit cell projected upon $\mathbf{V}^\perp$, and $A_k$, the area of the $k$th atomic surface.

The atomic temperature factor $T_k(\mathbf{H})$ can also have perpendicular-space components. Assuming only harmonic (static or dynamic) displacements in parallel and perpendicular space one obtains, in analogy to the usual expression (Willis & Pryor, 1975),

$$\begin{aligned} T_k(\mathbf{H}) &= T_k(\mathbf{H}^\parallel, \mathbf{H}^\perp) \\ &= \exp(-2\pi^2 \mathbf{H}^{\parallel T} \langle \mathbf{u}_i^\parallel \mathbf{u}_j^{\parallel T} \rangle \mathbf{H}^\parallel) \exp(-2\pi^2 \mathbf{H}^{\perp T} \langle \mathbf{u}_i^\perp \mathbf{u}_j^{\perp T} \rangle \mathbf{H}^\perp), \end{aligned}$$

with

$$\langle \mathbf{u}_i^\parallel \mathbf{u}_j^{\parallel T} \rangle = \begin{pmatrix} \langle \mathbf{u}_1^{\parallel 2} \rangle & \langle \mathbf{u}_1^\parallel \cdot \mathbf{u}_2^{\parallel T} \rangle & \langle \mathbf{u}_1^\parallel \cdot \mathbf{u}_3^{\parallel T} \rangle \\ \langle \mathbf{u}_2^\parallel \cdot \mathbf{u}_1^{\parallel T} \rangle & \langle \mathbf{u}_2^{\parallel 2} \rangle & \langle \mathbf{u}_2^\parallel \cdot \mathbf{u}_3^{\parallel T} \rangle \\ \langle \mathbf{u}_3^\parallel \cdot \mathbf{u}_1^{\parallel T} \rangle & \langle \mathbf{u}_3^\parallel \cdot \mathbf{u}_2^{\parallel T} \rangle & \langle \mathbf{u}_3^{\parallel 2} \rangle \end{pmatrix}$$

$$\text{and } \langle \mathbf{u}_i^\perp \mathbf{u}_j^{\perp T} \rangle = \langle \mathbf{u}_4^\perp \rangle.$$

The elements of the type $\langle \mathbf{u}_i \cdot \mathbf{u}_j^T \rangle$ represent the average values of the atomic displacements along the $i$th axis times the displacement along the $j$th axis on the $V$ basis.

4.6.3.3.1.4. *Intensity statistics*

In the following, only the properties of the quasiperiodic component of the 3D structure, namely the Fourier module $M_1^*$, are discussed. The intensities $I(\mathbf{H})$ of the Fibonacci chain decorated with point atoms are only a function of the perpendicular-space component of the diffraction vector. $|F(\mathbf{H})|$ and $F(\mathbf{H})$ are illustrated in Figs. 4.6.3.5 and 4.6.3.6 as a function of $\mathbf{H}^\parallel$ and of $\mathbf{H}^\perp$. The distribution of $|F(\mathbf{H})|$ as a function of their frequencies clearly resembles a centric distribution, as can be expected from the centrosymmetric 2D sub-unit cell. The shape of the distribution function depends on the radius $H_{\rm max}$ of the limiting sphere in reciprocal space. The number of weak reflections increases with the square of $H_{\rm max}$, that of strong reflections only linearly (strong reflections always have small $\mathbf{H}^\perp$ components).

The weighted reciprocal space of the Fibonacci sequence contains an infinite number of Bragg reflections within a limited region of the physical space. Contrary to the diffraction pattern of a periodic structure consisting of point atoms on the lattice nodes, the Bragg reflections show intensities depending on the perpendicular-space components of their diffraction vectors.

The reciprocal space of a sequence generated from hyperatoms with fractally shaped atomic surfaces (squared Fibonacci sequence) is very similar to that of the Fibonacci sequence (Figs. 4.6.3.8 and 4.6.3.9). However, there are significantly more weak reflections in the diffraction pattern of the 'fractal' sequence, caused by the geometric form factor.

4.6.3.3.1.5. *Relationships between structure factors at symmetry-related points of the Fourier image*

The two possible point-symmetry groups in the 1D quasiperiodic case, $K^{\rm 1D} = 1$ and $K^{\rm 1D} = \bar{1}$, relate the structure factors to

$$\begin{aligned} 1 &: \quad F(\mathbf{H}) = -F(\bar{\mathbf{H}}), \\ \bar{1} &: \quad F(\mathbf{H}) = F(\bar{\mathbf{H}}). \end{aligned}$$

A 3D structure with 1D quasiperiodicity results from the stacking of atomic layers with distances following a quasiperiodic sequence.

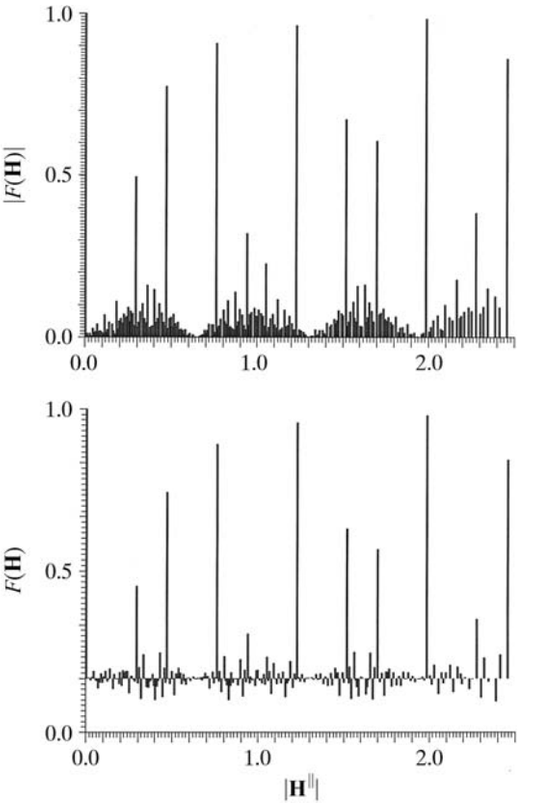


Fig. 4.6.3.8. The structure factors $F(\mathbf{H})$ (below) and their magnitudes $|F(\mathbf{H})|$ (above) of the squared Fibonacci chain decorated with equal point atoms are shown as a function of the parallel-space component $|\mathbf{H}^\parallel|$ of the diffraction vector. The short distance is $S = 2.5$ Å$^{-1}$, all structure factors within $0 \le |\mathbf{H}| \le 2.5$ Å$^{-1}$ have been calculated and normalized to $F(00) = 1$.

The point groups $K^{\rm 3D}$ describing the symmetry of such structures result from the direct product $K^{\rm 3D} = K^{\rm 2D} \otimes K^{\rm 1D}$. $K^{\rm 2D}$ corresponds to one of the ten crystallographic 2D point groups, $K^{\rm 1D}$ can be $\{1\}$ or $\{1, m\}$. Consequently, 18 3D point groups are possible.

Since 1D quasiperiodic sequences can be described generically as incommensurately modulated structures, their possible point and space groups are equivalent to a subset of the $(3+1)$D superspace groups for IMSs with satellite vectors of the type $(00\gamma)$, *i.e.* $\mathbf{q} = \gamma \mathbf{c}^*$, for the quasiperiodic direction [001] (Janssen *et al.*, 1999).

From the scaling properties of the Fibonacci sequence, some relationships between structure factors can be derived. Scaling the physical-space structure by a factor $\tau^n$, $n \in \mathbb{Z}$, corresponds to a scaling of the perpendicular space by the inverse factor $(-\tau)^{-n}$. For the scaling of the corresponding reciprocal subspaces, the inverse factors compared to the direct spaces have to be applied.

The set of vectors $\mathbf{r}$, defining the vertices of a Fibonacci sequence $s(\mathbf{r})$, multiplied by a factor $\tau$ coincides with a subset of the vectors defining the vertices of the original sequence (Fig. 4.6.3.10). The residual vertices correspond to a particular decoration of the scaled sequence, *i.e.* the sequence $\tau^2 s(\mathbf{r})$. The Fourier transform of the sequence $s(\mathbf{r})$ then can be written as the sum of the Fourier transforms of the sequences $\tau s(\mathbf{r})$ and $\tau^2 s(\mathbf{r})$;

$$\sum_k \exp(2\pi i \mathbf{H} \cdot \mathbf{r}_k) = \sum_k \exp(2\pi i \mathbf{H} \tau \mathbf{r}_k) + \sum_k \exp[2\pi i \mathbf{H}(\tau^2 \mathbf{r}_k + \tau)].$$

In terms of structure factors, this can be reformulated as

$$F(\mathbf{H}) = F(\tau \mathbf{H}) + \exp(2\pi i \tau \mathbf{H}) F(\tau^2 \mathbf{H}).$$

Hence, phases of structure factors that are related by scaling symmetry can be determined from each other.



references