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An analytical treatment of diffraction in quasiperiodic superlattices

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Abstract. We study the diffraction properties of a class of quasiperiodic superlattices described by the substitution rules $A \rightarrow A^pB$, $B \rightarrow A$ where p is a positive integer. These can be obtained by a projection method with a characteristic irrational σ, e.g., for the Fibonacci lattice ($p = 1$) $A \rightarrow AB$, $B \rightarrow A$, $\sigma = (1 + \sqrt{5})/2$. It is shown that the diffraction peak positions $K_{k,r}$ can be labeled by two integers k, r and are given by the expression $K_{k,r} = 2\pi\Lambda^{-1}r\sigma'^k$ where σ' are the so called precious means. It is shown that the Fibonacci lattice has the unique property that $\sigma = \sigma'$.

1. Introduction

In the last years there has been a large and growing interest in one-dimensional (1D) quasiperiodic systems. From the theoreticians standpoint the interest stems partly from the fact that although quasicrystals are perfectly ordered, the Bloch theorem is inapplicable since there is no translational symmetry. On the other hand, the wavefunctions are not all exponentially localised like in disordered 1D systems. Quasicrystals seem to be, in some sense, something intermediate between conventional crystals and disordered solids. Parallel to the theoretical development in the field of quasicrystals the advent of new experimental techniques (Shinjo and Takada 1987 and Chang and Giessen 1985) such as molecular-beam epitaxy (MBE) has made it possible to produce superlattices of extremely high quality. A superlattice is constructed by growing alternate layers of two different constituents A and B. A and B may for instance be n atomic layers of Mo and m atomic layers of V, respectively. The superlattice layers A and B are in general chosen to alternate periodically (Karkut *et al* 1985a, b and Terauchi *et al* 1985), but superlattices also provide an excellent method for realization of 1D quasiperiodicity. The samples grown will be quasiperiodic in the growth (z) direction and periodic in the xy plane. This was first achieved by Merlin *et al* (1985) who fabricated a sample grown by MBE of alternating layers of GaAs and AlAs arranged to form a Fibonacci sequence. The Fibonacci sequence can be described as the sequence obtained by starting with an A and repeated application of the substitution rules $A \rightarrow AB$, $B \rightarrow A$, i.e.,

$$A \rightarrow AB \rightarrow ABA \rightarrow ABAAB \rightarrow ABAABABA \rightarrow ABAABABAABAAB \rightarrow \dots$$

and the corresponding superlattice is obtained by attaching a basis to each A and B. The experiments done on quasiperiodic superlattices include diffraction (Merlin *et al* 1985,

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Karkut *et al* 1986 and Hu *et al* 1986), superconductivity (Karkut *et al* 1986) and Raman scattering (Merlin *et al* 1985), but so far only very few experiments have been performed on non-fibonaccian quasicrystals (Birch *et al* 1989). For the Fibonacci quasicrystal it is well known that the superlattice diffraction peaks can be labeled by two integers (k, r) such that the position of the peaks satisfy $K_{k,r} = 2\pi\Lambda^{-1}r\tau^k$, where $\tau = (1 + \sqrt{5})/2$ is the golden mean and Λ is an average lattice parameter. In this work we present a theoretical investigation of the superlattice diffraction properties of a class of quasicrystals generated by the substitution rule, $\mathrm{A} \rightarrow \mathrm{A}^p\mathrm{B}$, $\mathrm{B} \rightarrow \mathrm{A}$ and show that the diffraction peaks satisfy $K_{k,r} = 2\pi\Lambda^{-1}r\sigma'^k$ under certain specified conditions.

2. The superlattice

Consider the density distribution $\rho_\mathrm{S}(z)$, where S stands for superlattice,

$$\rho_\mathrm{S}(z) = \sum_{n=1}^{\infty} \delta(z - z_n) \qquad z_\mathrm{n} = \Lambda_\mathrm{B} n + (\Lambda_\mathrm{A} - \Lambda_\mathrm{B})\left[\frac{n}{\sigma}\right] \tag{1}$$

where [x] denotes the largest integer smaller than or equal to x and Λ_A and Λ_B are two different tile sizes. It is assumed that $\Lambda_\mathrm{A} \neq \Lambda_\mathrm{B}$. For σ rational equation (1) will describe a periodic density distribution, whereas an irrational σ will give rise to a quasiperiodic distribution of the tiles Λ_A and Λ_B. It has been shown previously (Lu and Birman 1986) that the Fourier transform of this distribution is given by

$$\mathcal{F}_\mathrm{S}(K) = \int \rho_\mathrm{S}(z) \exp(\mathrm{i}Kz)\,\mathrm{d}z = \sum_{m,n} \exp(-\mathrm{i}Z_{mn}) \frac{\sin(Z_{mn}/2)}{Z_{mn}/2} \delta(K\Lambda - K_{mn}\Lambda) \tag{2}$$

where

$$K_{mn} = \frac{2\pi}{\Lambda}\left(n + \frac{m}{\sigma}\right) \qquad Z_{mn} = \frac{2\pi}{\Lambda}(n(\Lambda_\mathrm{A} - \Lambda_\mathrm{B}) - m\Lambda_\mathrm{B}) \qquad \Lambda = \Lambda_\mathrm{B} + \frac{\Lambda_\mathrm{A} - \Lambda_\mathrm{B}}{\sigma} \tag{3}$$

and m, n are integers. The superlattice is constructed by introducing two densities $\rho_\mathrm{A}(z)$ and $\rho_\mathrm{B}(z)$ describing the two building blocks A and B and writing the total density, $\rho(z)$, of the superlattice as

$$\rho(z) = \begin{cases} \rho_\mathrm{A}(z) & z_{n+1} - z_n = \Lambda_\mathrm{A} \\ \rho_\mathrm{B}(z) & z_{n+1} - z_n = \Lambda_\mathrm{B} \end{cases} \qquad z_n \leq z < z_{n+1}. \tag{4}$$

For clarity, $\rho(z)$ is illustrated in figure 1. Now, dividing ρ_S into two parts ${}^\mathrm{A}\rho_\mathrm{S}$ and ${}^\mathrm{B}\rho_\mathrm{S}$

$${}^\mathrm{A}\rho_\mathrm{S} = \sum_{n, z_{n+1} - z_n = \Lambda_\mathrm{A}}^{\infty} \delta(z - z_n) \qquad {}^\mathrm{B}\rho_\mathrm{S} = \sum_{n, z_{n+1} - z_n = \Lambda_\mathrm{B}}^{\infty} \delta(z - z_n) \tag{5}$$

with z_n as in equation (1), we may write $\rho(z)$ as the sum of two convolutions

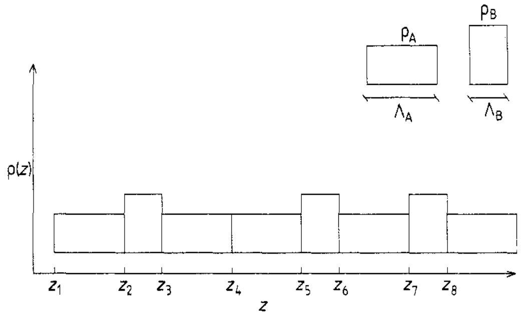

Figure 1. The superlattice density of equation (4) for the special case of a Fibonacci superlattice, $\sigma = (1 + \sqrt{5})/2$.

$$\rho(z) = \int \rho_{\mathrm{A}}(y)\,{}^{\mathrm{A}}\rho_{\mathrm{S}}(z-y)\,\mathrm{d}y + \int \rho_{\mathrm{B}}(y)\,{}^{\mathrm{B}}\rho_{\mathrm{S}}(z-y)\,\mathrm{d}y \tag{6}$$

also with z_n as in equation (1). Using the forms (5) for ${}^{\mathrm{A}}\rho_{\mathrm{S}}$ and ${}^{\mathrm{B}}\rho_{\mathrm{S}}$ one obtains

$$\rho(z) = \begin{cases} \displaystyle\sum_{n=1}^{\infty} \rho_{\mathrm{A}}(z - z_n) & z_n \le z < z_{n+1} \qquad z_{n+1} - z_n = \Lambda_{\mathrm{A}} \\ \displaystyle\sum_{n=1}^{\infty} \rho_{\mathrm{B}}(z - z_n) & z_n \le z < z_{n+1} \qquad z_{n+1} - z_n = \Lambda_{\mathrm{B}}. \end{cases} \tag{7}$$

Equation (6) suggests that we write the Fourier transform $\mathcal{F}(K)$ of the total density $\rho(z)$ as the product sum

$$\mathcal{F}(K) = \mathcal{F}_{\mathrm{A}}(K)\,{}^{\mathrm{A}}\mathcal{F}_{\mathrm{S}}(K) + \mathcal{F}_{\mathrm{B}}(K)\,{}^{\mathrm{B}}\mathcal{F}_{\mathrm{S}}(K) \tag{8}$$

where $\mathcal{F}_{\mathrm{A}}$, $\mathcal{F}_{\mathrm{B}}$, ${}^{\mathrm{A}}\mathcal{F}_{\mathrm{S}}$ and ${}^{\mathrm{B}}\mathcal{F}_{\mathrm{S}}$ are the Fourier transforms of ρ_{A}, ρ_{B}, ${}^{\mathrm{A}}\rho_{\mathrm{S}}$ and ${}^{\mathrm{B}}\rho_{\mathrm{S}}$, respectively. Now, the ${}^{\mathrm{A}}\rho_{\mathrm{S}}$ and ${}^{\mathrm{B}}\rho_{\mathrm{S}}$ describing the renormalised lattice of only A or B sites may also be written in the form (1) with renormalised parameters ${}^{\mathrm{A}}\Lambda$, ${}^{\mathrm{A}}\Lambda_{\mathrm{A}}$, ${}^{\mathrm{A}}\Lambda_{\mathrm{B}}$ and σ_{A} for ${}^{\mathrm{A}}\rho_{\mathrm{S}}$ and ${}^{\mathrm{B}}\Lambda$, ${}^{\mathrm{B}}\Lambda_{\mathrm{A}}$, ${}^{\mathrm{B}}\Lambda_{\mathrm{B}}$ and σ_{B} in the case of ${}^{\mathrm{B}}\rho_{\mathrm{S}}$.

3. The sublattices

In this section we write the sublattices ${}^{\mathrm{A}}\rho_{\mathrm{S}}$ and ${}^{\mathrm{B}}\rho_{\mathrm{S}}$ in the form of equation (1) and establish the values of the parameters ${}^{\mathrm{A,B}}\Lambda_{\mathrm{A,B}}$, $\sigma_{\mathrm{A,B}}$ and ${}^{\mathrm{A,B}}\Lambda$. We denote as previously the site n by A if $z_{n+1} - z_n = \Lambda_{\mathrm{A}}$ and by B if $z_{n+1} - z_n = \Lambda_{\mathrm{B}}$ and we let ${}^{\mathrm{A}}z_m({}^{\mathrm{B}}z_m)$ be the position of the mth A (B). We also define the number of A (B) sites in the original sequence of n sites as n_{A} (n_{B}). Note that we may without loss of generality assume $\sigma > 1$ since $[n/\sigma] = [n\{1/\sigma\} + n[1/\sigma]] = [n\{1/\sigma\}] + n[1/\sigma]$ with $\{x\}$ defined from $x = \{x\} + [x]$.

Then from equation (1) we have $n_A = [n/\sigma]$ and $n_B = n - n_A$. Then the position of the mth A site is given by

$$z_m^A = (m-1)\Lambda_A + n^B\Lambda_B \tag{9}$$

where n^B is the number of B sites before the mth A site. The number n^B is given by

$$n^B = p - [p/\sigma] \tag{10}$$

where p is the largest integer satisfying $[p/\sigma] = m - 1$. since we are interested only in irrational σ we may then put $p = [m\sigma]$. Inserting this into equations (9) and (10) we have

$$\begin{aligned} z_m^A &= (m-1)\Lambda_A + \Lambda_B([m\sigma] - m + 1) = m\Lambda_A + \Lambda_B[m(\sigma - 1)] + \Lambda_B - \Lambda_A \\ &= m\Lambda_A + \Lambda_B[m[\sigma - 1] + m\{\sigma - 1\}] + \Lambda_B - \Lambda_A \\ &= m(\Lambda_A + \Lambda_B[\sigma - 1]) + \Lambda_B[m\{\sigma - 1\}] + \Lambda_B - \Lambda_A. \end{aligned}$$

This can be written as

$$z_m^A = m\,{}^A\Lambda_B + ({}^A\Lambda_A - {}^A\Lambda_B)[m/\sigma_A] + \Lambda_B - \Lambda_A \tag{11}$$

where

$${}^A\Lambda_B = (\Lambda_A + \Lambda_B[\sigma - 1]) \qquad {}^A\Lambda_A - {}^A\Lambda_B = \Lambda_B \qquad \sigma_A = \{\sigma - 1\}^{-1}. \tag{12}$$

Similar considerations for the B sites lead to the following equation

$$z_m^B = m\,{}^B\Lambda_B + ({}^B\Lambda_A - {}^B\Lambda_B)[m/\sigma_B] \tag{13}$$

with

$${}^B\Lambda_B = (\Lambda_B + \Lambda_A[(\sigma - 1)^{-1}]) \qquad {}^B\Lambda_A - {}^B\Lambda_B = \Lambda_A \qquad \sigma_B = \{(\sigma - 1)^{-1}\}^{-1}. \tag{14}$$

With the correct choice of tile sizes, the peaks of ${}^A\mathscr{F}_S$ and ${}^B\mathscr{F}_S$ can be made to coincide provided σ_A and σ_B satisfy: $\sigma_A = \sigma_B = \sigma'$. From equations (12) and (14) we then have

$$\begin{aligned} &\{\sigma - 1\} = \{(\sigma - 1)^{-1}\} \Rightarrow (\sigma - 1)^{-1} = \sigma - 1 + p \Rightarrow \sigma \\ &\qquad = (2 - p + \sqrt{4 + p^2})/2 \quad p = 1, 2 \ldots . \end{aligned} \tag{15}$$

Notice that $\sigma(p = 1) = \tau = (1 + \sqrt{5})/2$ generating the familiar Fibonacci sequence satisfies $\sigma = \sigma_A = \sigma_B = \sigma'$. This is a unique property of the Fibonacci quasicrystal. It is also interesting to note that the $\sigma(p)$ satisfying equation (15) generates sequences also described by the substitution rules; $A \rightarrow A^pB$, $B \rightarrow A$. Before we proceed, and in order to simplify equations (12) and (14), it is useful to establish a few important relations for the numbers σ and $\sigma' = \sigma_A = \sigma_B$. From (12) and (15) we have, since $0 < \sigma - 1 < 1$

$$\sigma' = \{\sigma - 1\}^{-1} = (\sigma - 1)^{-1} = 2/(\sqrt{4 + p^2} - p) = (p + \sqrt{4 + p^2})/2 \tag{16}$$

and

$$[\sigma - 1] = 0 \Rightarrow [(\sigma - 1)^{-1}] = [\sigma'] = [\sigma - 1 + p] = [\sigma - 1] + p = p. \tag{17}$$

Following Holzer (1988) we call the irrationals σ' the 'precious means' and define the generalised Fibonacci numbers F_n such that $F_{n+1} = pF_n + F_{n-1}$ with $F_0 = 0$ and $F_1 = 1$. The following useful relation is then proved in the appendix.

$$\sigma'^n = F_n\sigma' + F_{n-1}. \tag{18}$$

If in the following we assume σ to be given by equation (15) we my, using equations (16)–(17), rewrite (12) and (14) as

$$^{\mathrm{A}}\Lambda_{\mathrm{B}} = \Lambda_{\mathrm{A}} \qquad ^{\mathrm{A}}\Lambda_{\mathrm{A}} - {}^{\mathrm{A}}\Lambda_{\mathrm{B}} = \Lambda_{\mathrm{B}} \qquad \sigma_{\mathrm{A}} = \sigma' \tag{19}$$

and

$$^{\mathrm{B}}\Lambda_{\mathrm{B}} = \Lambda_{\mathrm{B}} + p\Lambda_{\mathrm{A}} \qquad ^{\mathrm{B}}\Lambda_{\mathrm{A}} - {}^{\mathrm{B}}\Lambda_{\mathrm{B}} = \Lambda_{\mathrm{A}} \qquad \sigma_{\mathrm{B}} = \sigma'. \tag{20}$$

Equations (11), (13), (19) and (20) completely describe the sublattices consisting of the original A and B sites and thus the densities $^{\mathrm{A}}\rho_{\mathrm{S}}$ and $^{\mathrm{B}}\rho_{\mathrm{S}}$ are determined.

4. Tailoring the tiles

In this section we will describe how to tailor the tile sizes Λ_{A} and Λ_{B} in order to obtain a simple expression for the diffraction peak positions. Given that the ratio between the two tiles Λ_{A} and Λ_{B} is given by σ' and using equations (3), (19) and (20) we may write the quantities $^{\mathrm{A,B}}K_{mn}$, $^{\mathrm{A,B}}Z_{mn}$ and $^{\mathrm{A,B}}\Lambda$ for the sublattices as

$$^{\mathrm{A}}K_{mn} = (2\pi/{}^{\mathrm{A}}\Lambda)\,(n + (m/\sigma')) \qquad ^{\mathrm{A}}Z_{mn} = (2\pi/{}^{\mathrm{A}}\Lambda)\Lambda_{\mathrm{B}}(n - m\sigma')$$
$$^{\mathrm{A}}\Lambda = \Lambda_{\mathrm{B}}(\sigma' + 1/\sigma') \tag{21}$$

and

$$^{\mathrm{B}}K_{mn} = (2\pi/{}^{\mathrm{B}}\Lambda)\,(n + (m/\sigma')) \qquad ^{\mathrm{B}}Z_{mn} = (2\pi/{}^{\mathrm{B}}\Lambda)\Lambda_{\mathrm{B}}(n - m\sigma')\sigma'$$
$$^{\mathrm{B}}\Lambda = \Lambda_{\mathrm{B}}(\sigma^2 + 1) \tag{22}$$

where the last two relations in (22) follows from equation (18). Now, from (2) we see that large peaks will occur in $^{\mathrm{A,B}}\mathscr{F}_{\mathrm{S}}(K)$ for $K = K_{mn}$ such that $^{\mathrm{A,B}}Z_{mn} = 0$. Since σ' can be written as the simple continued fraction

$$\sigma' = p + \cfrac{1}{p + \cfrac{1}{p + \cfrac{1}{p + \cfrac{1}{p + \dots}}}} \tag{23}$$

the kth rational approximant to σ' is given by F_k/F_{k-1} and the condition $^{\mathrm{A,B}}Z_{mn} \simeq 0$ reduces to $n = rF_k$, $m = rF_{k-1}$, with integers r, k. Applying this to equations (21) and (22) and using equation (18) then leads to the following expression for the diffraction peak positions

$$^{\mathrm{A,B}}K_{kr} = (2\pi/{}^{\mathrm{A,B}}\Lambda) \quad (r/\sigma')(F_k\sigma' + F_{k-1}) = (2\pi/{}^{\mathrm{A,B}}\Lambda\sigma')r\sigma'^k \qquad r, k \in Z \tag{24}$$

Finally, noting that $^{\mathrm{B}}\Lambda = \sigma'\,{}^{\mathrm{A}}\Lambda$, we conclude that for $\Lambda_{\mathrm{A}}/\Lambda_{\mathrm{B}} = \sigma'$ $^{\mathrm{A}}\mathscr{F}_{\mathrm{S}}$ and $^{\mathrm{B}}\mathscr{F}_{\mathrm{S}}$ have peaks at *the same* K values $K_{k,r}$ given by

$$K_{k,r} = (2\pi/{}^{\mathrm{B}}\Lambda)r\sigma'^k \qquad r, k \in Z. \tag{25}$$

In summary, we have shown that quasicrystalline superlattices with the quasiperiodicity given by equation (1) with σ as in equation (15) have large diffraction peaks at wavevectors $K_{k,r}$ labeled by two integers (k, r). These quasicrystals can also be generated by a substitution rule; $A \rightarrow A^pB$, $B \rightarrow A$ the first $(p = 1)$ of which is the celebrated Fibonacci sequence. It is interesting to note that the Fibonacci sequence is indeed special since it is the only case for which $\sigma = \sigma'$.

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Appendix

The statement $\sigma'^n = F_n\sigma' + F_{n-1}$ (equation (18)) is easily proven by induction, as follows. Since $F_1 = 1$ and $F_0 = 0$, the statement is trivially true for $n = 1$. Also since

$$\sigma'^2 = ((p + \sqrt{4 + p^2})/2)^2 = p(p + \sqrt{4 + p^2})/2 + 1 = F_2\sigma' + F_1 \qquad \text{(A1)}$$

the statement holds for $n = 2$. Assuming equation (18) holds for some n and using (A1) we then have

$$\begin{aligned}\sigma'^{n+1}\ F_n\sigma'^2 + F_{n-1}\sigma' &= F_n(F_2\sigma' + F_1) + F_{n-1}\sigma' = (pF_n + F_{n-1})\sigma' + F_n \\ &= F_{n+1}\sigma' + F_n\end{aligned}$$

and equation (18) follows by induction.

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