

Diffraction from a quasi-crystalline chain

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Abstract. We present a general formalism for diffraction from a one-dimensional quasi-crystal with arbitrary length scales and sequences. The notion of sub-quasi-lattices is introduced and the effect of different basis on different sites is studied. The relevance of this work for the study of vibrational and electronic spectra of the chain is discussed.

Keywords. Diffraction; quasi-crystal; sub-quasi-lattices; vibrational spectra; electronic spectra.

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Recent observation by Shechtman *et al* (1984) of five fold electron diffraction pattern from rapidly cooled Al-14% Mn alloys has spurt widespread interest in the characterization and physical properties of this new phase of condensed matter. It is well known that five-fold rotational symmetry is inconsistent with translational invariance. Yet from the observation of sharp diffraction spots one should expect long-range-order, though the system is not periodic. A striking example of such structures is the Penrose tiling in two-dimensions (Gardner 1977). Guided by this observation, this new phase of Al-14% Mn is considered to be a generalization of the Penrose tiling to three dimensions (Levine and Steinhardt 1984). An elegant way of generating Penrose tiling in a d -dimensional space is to project a D ($> d$)-dimensional periodic lattice on a d -dimensional hyperplane (de Bruijn 1981; Duneau and Katz 1985). Following this procedure the diffraction pattern and intensities have been calculated (Elser 1985a) for two-, three- and five-fold patterns by taking a star of six vectors pointing towards the vertices of an icosahedron. These calculated diffraction patterns have striking similarity with those observed from Al-14% Mn alloys. But contrary to the calculations where identical atoms are supposed to populate every site, all quasi-crystals (studied experimentally) are binary or even multicomponent. Hence it is essential for a better understanding of the atomic distribution in this new phase, now known as quasi-crystals, to calculate the diffraction pattern with different atoms occupying different sites. Such a calculation is also a prerequisite for the analysis of the electronic and vibrational properties of quasi-crystals. We consider this problem in this paper and restrict ourselves to one dimension for simplicity.

The quasi-lattice structure studied in one dimension is the quasi-periodic sequence of two intervals of lengths d_1 and d_2 (Levine and Steinhardt 1984; Elser 1985a; Zia and Dallas 1985). In these studies the length ratio r ($= d_1/d_2$) and the relative concentration c (ratio of the number of intervals of lengths d_1 and d_2 respectively) are taken to be identical. In a physical context these two quantities could be different. We have

extended the existing mapping procedure to obtain $0 < r < \infty$ for any c which we take to be greater than one without any loss of generality. Furthermore, we introduce the concept of sub-quasi-lattices which, we hope, may prove useful in the characterization of higher dimensional quasi-crystals.

We start with a rectangular lattice* whose unit cell has sides a and b (see figure 1) such that

$$a = d_1 (1 + r\tau)^{1/2}/r, \quad b = d_1 [(1 + r\tau)/r\tau]^{1/2}, \quad \tau = c. \quad (1)$$

Now a line is drawn making an angle θ with the x -axis such that $\cot \theta = \sqrt{r\tau}$. We call this line as the ξ -axis and the perpendicular one as the η -axis. If the whole two dimensional lattice is projected on the ξ -axis, it will produce a dense distribution of points. In order to have non-vanishing distance between points, one must take a finite slit and project all lattice points within this slit on the ξ -axis. It is possible to have only two length scales by defining two slits of widths $W_\alpha = a \cos \theta$ and $W_\beta = b \sin \theta$. For

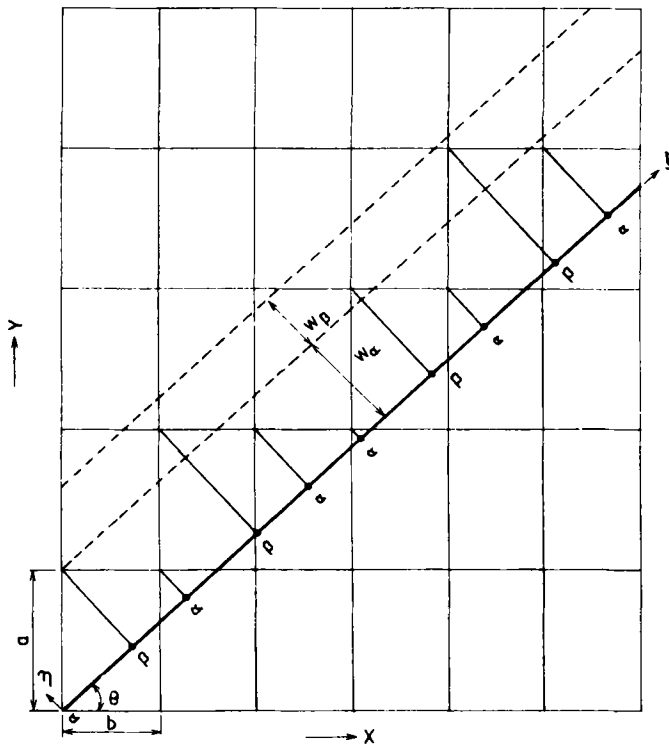


Figure 1. Projection of a part of the rectangular lattice with unit cell of sides a and b on a line (thick line). $W_\alpha = a \cos \theta$ and $W_\beta = b \sin \theta$ are the widths of the two slits measured along the η -axis. Projection of the points in the slit $0 \leq \eta < W_\alpha$ yields the α sub-quasi-lattice while those from the slit $W_\alpha \leq \eta < W_\alpha + W_\beta$ leads to the β sub-quasi-lattice. The specific picture is for $\tau = \tau_0$, the golden mean and $r = 4\tau_0/9$.

*This is not the only choice. One can start with other two-dimensional lattices such as the oblique one which leads to the same conclusion.

$0 \leq \eta < W_\alpha$, the sites in the two-dimensional lattice will be projected onto the end points of intervals of length d_1 while the end points of d_2 intervals can be obtained for $W_\alpha \leq \eta < W_\alpha + W_\beta$. We must stress that the sequence of α and β on the chain depends upon the value of τ alone. These two sets of projected points (called α and β in figure 1) again form two quasi-lattices. These two sub-quasi-lattices also have long and short intervals, but of different lengths. It is clear that these repeat in the same sequence as the original quasi-lattice. If r and τ are taken to be the golden mean $\tau_0 = (\sqrt{5} + 1)/2$, then the ratios of these two sub-quasi-lattice intervals are also equal to τ_0 . This is the self-similarity property of the one-dimensional quasi-lattice and the two sub-quasi-lattices correspond to inflation of the original one by τ_0 and τ_0^2 . In fact one can continue to subdivide the sub-quasi-lattices to obtain new ones. This process is likely to have advantage for the representation of multicomponent quasi-crystals. Also in general a quasi-lattice with n intervals can be generated by defining n appropriate slit functions. For example, with $a = b$ we can define three slits one over the other of widths $W_\alpha = a \cos \theta / \tau_0^2$, $W_\beta = a \cos \theta / \tau_0$ and $W_\gamma = a \cos \theta$ to obtain a quasi-periodic sequence of three intervals, with the relative number concentrations $N_\beta / N_\alpha = \tau_0$ and $N_\beta / N_\gamma = \tau_0^2$. The sequence of α , β and γ points generated in this way can be identified with the (111) planes of Ni, Al and vacancy respectively in the vacancy stabilised quasi-crystalline phase of Al-Ni alloys studied by Chattopadhyay *et al* (1985).[†]

We now proceed to calculate the diffraction pattern from a quasi-crystal with two different basis associated with the α and β sites. Let the positions of sites in the ξ -space be represented by ξ_j ($-\infty < j < \infty$, $j = \text{integer}$). Let the positions of atoms in the basis on α and β sub-quasi-lattices (with respect to ξ_j^α and ξ_j^β) be given by r_λ^α ($\lambda = 1, \dots, N$) and r_μ^β ($\mu = 1, \dots, M$), respectively. Then the scattered amplitude in a diffraction experiment is given by

$$F(k) = \sum_{j,\lambda} f_\lambda^\alpha \exp [ik(\xi_j^\alpha + r_\lambda^\alpha)] + \sum_{j,\mu} f_\mu^\beta \exp [ik(\xi_j^\beta + r_\mu^\beta)], \tag{2}$$

where f_λ^α is the scattering power of the λ th atom on the α sub-quasi-lattice. Using the geometrical structure factor $\tilde{f}^\alpha(k) = \sum_\lambda f_\lambda^\alpha \exp(ikr_\lambda^\alpha)$, we can rewrite $F(k)$ as

$$F(k) = \tilde{f}^\alpha(k) S^\alpha(k) + \tilde{f}^\beta(k) S^\beta(k), \tag{3}$$

where the partial structure factor $S^\alpha(k)$ is

$$S^\alpha(k) = \int_{-\infty}^{\infty} \exp(ik\xi) R^\alpha(\xi) d\xi. \tag{4}$$

Here $R^\alpha(\xi)$ represents the density of points along the ξ -axis corresponding to the α sub-quasi-lattice. We calculate $R^\alpha(\xi)$ following Zia and Dallas (1985). In the two-dimensional rectangular lattice the density of points in the (ξ, η) coordinate system is

[†] A ternary sequence ($\alpha\beta\alpha\gamma\alpha\beta\alpha\beta\alpha\gamma\alpha\beta\alpha\beta \dots$) can simply be obtained from a binary one ($\alpha\beta\alpha\alpha\beta\alpha\beta \dots$) by replacing α with $\alpha\beta$ and β with $\alpha\gamma$ in the latter sequence

given by

$$\rho(\xi, \eta) = \sum_{j,l} \delta(\xi - jb \cos \theta - la \sin \theta) \delta(\eta + jb \sin \theta - la \cos \theta), \tag{5}$$

where the sum is over all the integer values of j and l . The density of projected points can be obtained as

$$R^\alpha(\xi) = \int_{-\infty}^{\infty} d\eta P^\alpha(\eta) \rho(\xi, \eta),$$

and

$$R^\beta(\xi) = \int_{-\infty}^{\infty} d\eta P^\beta(\eta) \rho(\xi, \eta), \tag{6}$$

where $P(\eta)$ are indicator functions

$$P^\alpha(\eta) = \begin{cases} 1 & \text{for } 0 \leq \eta < W_\alpha, \\ 0 & \text{otherwise} \end{cases}$$

and

$$P^\beta(\eta) = \begin{cases} 1 & \text{for } W_\alpha \leq \eta < W_\alpha + W_\beta \\ 0 & \text{otherwise.} \end{cases} \tag{7}$$

Then the partial structure factors are derived using

$$S^\alpha(k) = \int_{-\infty}^{\infty} dp \tilde{P}^\alpha(-p) \tilde{\rho}(k, p),$$

and

$$S^\beta(k) = \int_{-\infty}^{\infty} dp \tilde{P}^\beta(-p) \tilde{\rho}(k, p), \tag{8}$$

where

$$\tilde{\rho}(k, p) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\xi d\eta \exp[i(k\xi + p\eta)] \rho(\xi, \eta),$$

$$\tilde{P}^\alpha(p) = \int_{-\infty}^{\infty} d\eta \exp(ip\eta) P^\alpha(\eta), \text{ etc.} \tag{9}$$

The results obtained are

$$S^\alpha(k) = \sum_{n,m} \exp(-i\phi_{nm}) \frac{\tau}{\tau d_1 + d_2} \frac{\sin \phi_{nm}}{\phi_{nm}} \delta(k - k_{nm}), \tag{10}$$

and

$$S^\beta(k) = \sum_{n,m} \exp[-i(2\phi_{nm} + \theta_{nm})] \frac{1}{\tau d_1 + d_2} \frac{\sin \theta_{nm}}{\theta_{nm}} \delta(k - k_{nm}),$$

where

$$\phi_{nm} = \frac{\pi \tau d_2}{\tau d_1 + d_2} (mr - n), \quad \theta_{nm} = \phi_{nm}/\tau,$$

and

$$k_{nm} = \frac{2\pi}{\tau d_1 + d_2} (m + n\tau). \quad (11)$$

Incidentally, $a = (\tau d_1 + d_2)/(1 + \tau)$ can be defined as the average quasi-lattice constant. Substituting (10) and (11) in (3), we get the required result. The point that emerges is that the peaks can be labelled to be at $k = 2\pi(m + n\tau)/(\tau d_1 + d_2)$. That is, the length ratio r only defines a length scale in k -space; the relative positions depend only on τ . This corroborates the claim by Merlin *et al* (1985). It is also clear that the fundamental nature of the diffraction pattern is unaltered even when we have two different basis associated with the sub-quasi-lattices. Only the intensities are modified, just like for the periodic case.

If r is a rational number N_1/N_2 (N_1, N_2 relatively prime) then from (3) and (10) it is clear that the intensities will be equal for $mN_1 = nN_2$ and will be given by $|(f^\alpha(k)\tau + \tilde{f}^\beta(k))/(\tau d_1 + d_2)|^2$. But within each such period, there will be a dense distribution of points. A very interesting example is the case when $r = 1$. In this case, we have a regular lattice. In the special case of $f_A^\alpha = f_B^\beta = f$, $F(k)$ reduces to the familiar case of a regular lattice,

$$F(k) = \sum_n \frac{f}{d_1} \delta\left(k - \frac{2\pi}{d_1} n\right), \quad (12)$$

as it should. However, if atoms A and B are placed on the α and β sites respectively, then in addition to peaks of equal intensities at $k = \text{integral multiples of } 2\pi/d_1$, as shown in figure 2(a) within each period there are peaks at k_{nm} given by (11). These extra peaks are equivalent to the occurrence of extra peaks in an ordered alloy. Accordingly we say that the quasi-period sequence has chemical-quasi long-range-order.

For the general case the diffracted intensities with $f_A^\alpha = f_B^\beta$ and $f_A^\alpha \neq f_B^\beta$ are shown in figures 2(b) and 2(c). As expected, for different values of f_A^α and f_B^β , the intensities of various peaks change drastically. We have taken f_A^α and f_B^β which roughly correspond to Al and Mn in the case of neutron diffraction. We shall therefore suggest neutron diffraction on these samples to get more information about the structure of quasi-crystals. Also shown in figure 2(d) are the diffraction intensities for the case when r and τ are rational numbers. For the chosen values, figures 2(c) and 2(d) seem almost identical. Therefore it will be useful to study the properties of long-period structures (see also Elser 1985b).

The chain having the chemical quasi-long-range-order can be used as a model to analyze the vibrational and electronic properties of quasi-crystals. To illustrate the idea we consider the stationary tight-binding model of electronic structure described by the equation

$$E\psi_n = \varepsilon_n\psi_n + t_{n,n+1}\psi_{n+1} + t_{n,n-1}\psi_{n-1}; \quad -\infty < n < \infty. \quad (13)$$

In (13) ε_n and ψ_n are respectively the site energy and the wave function on site n , and $t_{n,n+1}$ is the hopping integral from site n to $n+1$. We take $\{\varepsilon_n\}$ to be in a quasi-periodic sequence of two numbers ε_1 and ε_2 . Similarly $\{t_{n,n+1}\}$ is taken to be in a quasi-periodic sequence of two values t_1 and t_2 . Defining Fourier transforms of various quantities as

$$\tilde{\psi}(k) = \sum_n \exp(ikn)\psi_n, \quad \tilde{\varepsilon}(k) = \sum_n \exp(ikn)\varepsilon_n$$

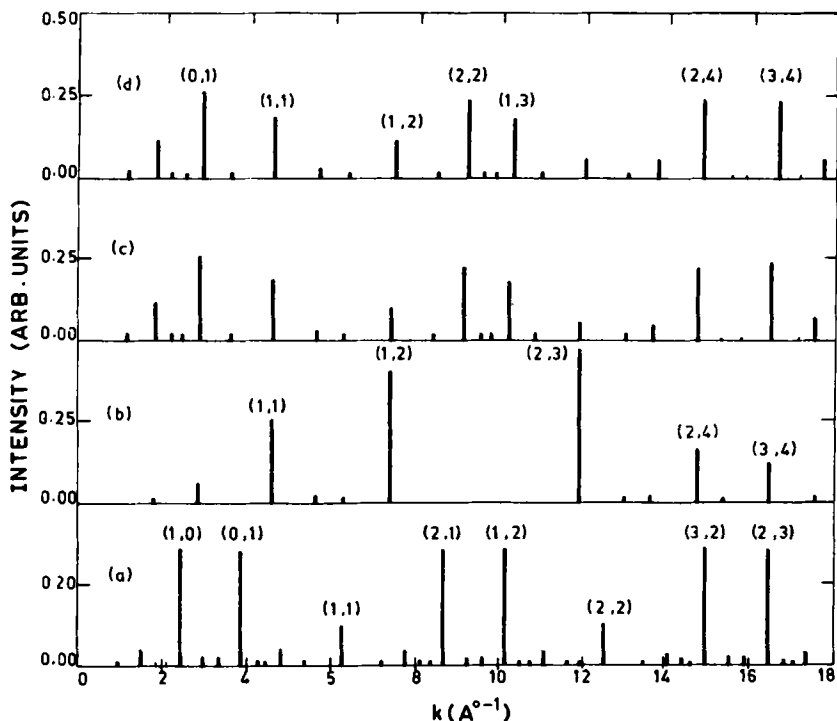


Figure 2. Plot of intensities versus wave vector k for a quasi-crystalline chain for the following choice of parameters: (a) $r = d_1/d_2 = 1.0$, $f_A^\alpha = 1.0$, $f_B^\beta = -0.8$ and $\tau = \tau_0$; (b) $r = \tau = \tau_0$, $f_A^\alpha = f_B^\beta = 1.0$; (c) $r = \tau = \tau_0$ and $f_A^\alpha = -f_B^\beta = 1.0$ and (d) $r = \tau = 8/5$ and $f_A^\alpha = -f_B^\beta = 1.0$. Peaks with indices (m, n) correspond to the wave vector $k_{mn} = (2\pi/\tau d_1 + d_2)(m + n\tau)$. Between any two peaks there exist an infinite number of other peaks which cannot be resolved in this figure. Peaks corresponding to $k = 0$ are not shown.

$$\tilde{T}(k) = \sum_n \exp(ikn) t_{n, n+1}, \quad (14)$$

(12) can be recast as

$$E \tilde{\psi}(k) = \int dk' [\tilde{\varepsilon}(k') + \{\exp(-ik) + \exp(i(k-k'))\} T(k')] \tilde{\psi}(k-k'). \quad (15)$$

To proceed further, we have to calculate the Fourier transform of a quasi-periodic function. If f_A^α and f_B^β are recognized as ε_1 and ε_2 or t_1 and t_2 , then from (3), the required Fourier transform is

$$F(k) = \sum_{n,m} \exp\left(-i \frac{\pi\tau(m-n)}{1+\tau}\right) \left[\frac{\tau f_A^\alpha}{1+\tau} \frac{\sin \frac{\pi\tau(m-n)}{1+\tau}}{\pi\tau(m-n)} \right. \\ \left. + \frac{f_B^\beta (-1)^{m-n}}{1+\tau} \frac{\sin \frac{\pi(m-n)}{1+\tau}}{\pi(m-n)} \right] \delta\left(k - \frac{2\pi(m+n\tau)}{(1+\tau)d_1}\right). \quad (16)$$

(15) together with (16) yields

$$[\lambda(k) - E] \tilde{\psi}(k) + \sum'_{n,m} U_{nm}(k) \tilde{\psi}(k - k_{nm}) = 0, \quad (17)$$

where $\lambda(k)$ and $U_{nm}(k)$ are functions of k depending parametrically on $\varepsilon_1, \varepsilon_2, t_1$ and t_2 . The prime on the summation in (17) denotes that $n = m = 0$ is excluded. This equation is analogous to the usual band structure equation in 1-D (Kittel 1971). It is now clear that at any $k = k_{nm}/2$ there should be a gap whose strength depends upon the intensity of peak in the diffraction pattern at k_{nm} , which is incorporated in $U_{nm}(k)$. Details of this investigation will be published separately.

In summary we have presented a general formulation for diffraction from a quasi-crystal where the length scales and the sequences can be arbitrary. The notion of sub-quasi-lattices is introduced and it can be used to characterize higher dimensional quasi-periodic structures (Kumar *et al* 1985). Here we recall that recently Kuriyama *et al* (1985) have analyzed the Al-Mn quasi-crystalline phase in terms of competition of two cubic cells with sides having ratio τ_0 . It may therefore be possible that the two cubes form two basis for the two sub-quasi-lattices. Finally, the results presented here can be carried over to the study of electronic and vibrational properties.

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