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Diffraction properties of one-dimensional finite size Fibonacci quasilattice

V S K BALAGURUSAMY, S BARANIDHARAN, E S R GOPAL*, and V SASISEKHARAN[†]

Department of Physics, [†]Molecular Biophysics Unit, Indian Institute of Science, Bangalore 560012, India

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Abstract. The diffraction patterns from Fibonacci quasilattices have been calculated. Finite-size effects are evaluated for weak and strong peaks. For a smaller number of scatterers (< 100) there are fluctuations in the intensities of weak and strong peaks. The fluctuations in weak peaks are greater than that in strong peaks. The fluctuations in intensities of weak and strong peaks near the origin are larger than in the corresponding cases of weak and strong peaks far away from the origin. Small shifts in peak-positions are unexpectedly found, the shifts being proportional to $N^{-3/2}$ for a large number of scatterers. The diffraction pattern of a one-dimensional crystal and random structure is compared with that of the Fibonacci quasilattice. The strong peaks observed in the diffraction pattern of 1-d crystal show negligible peak-shifts, they being comparable with computational errors even when the number of scatterers is as small as 5. The implications for analysing the experiments are briefly indicated.

Keywords. Quasilattice; diffraction; quasicrystal.

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1. Introduction

Soon after the discovery of rapidly quenched Al-Mn alloys showing the crystallographically forbidden 5-fold symmetry in the electron diffraction patterns (Shechtman *et al* 1984), quasiperiodic structures were proposed (Levine and Steinhardt 1984) as possible models for the atomic structure of these phases. The numerically computed diffraction patterns of these models show the features of the experimentally observed diffraction patterns. Quasilattices in one-, two- and three-dimensions can be obtained by projecting higher dimensional periodic lattices onto a lower dimensional space (Kramer and Neri 1984; Duneau and Katz 1985; Zia and Dallas 1985; Elser 1985, 1986; Gratias and Cahn 1986; Janssen 1986; Levine and Steinhardt 1986; Valsakumar and Vijaykumar 1986; Prince 1987). The method of generating quasilattices using a self-similarity principle is also known (Penrose 1974; Sasisekharan 1986). Quasilattices can also be obtained by the generalized dual method or multigrid method (de Bruijn 1981; Socolar *et al* 1985).

Long before the experimental observation of quasicrystals, tiling a plane aperiodically with two kinds of rhombuses preserving the 5-fold rotational symmetry had been

*To whom correspondence should be addressed

suggested (Penrose 1974). Starting with a pattern, having 5-fold rotational symmetry, consisting of these two kinds of rhombi an aperiodic tiling can be generated by following a rule while matching the rhombuses. The number of rhombi of each kind generated after n substitutions follows the Fibonacci sequence: 1, 1, 2, 3, 5, 8, 13, 21, 34, 55, 89, 144,... The ratio of the number of one kind of rhombi to the other converges to the golden mean, as the number of substitutions applied to generate the tiling tends to infinity.

Fibonacci superlattices have been grown experimentally using GaAs/AlAs layers (Todd *et al* 1986) and Nb/Cu layers (Hu *et al* 1986) and their diffraction properties have been studied. The present work is concerned with these situations.

2. Model calculations

The one-dimensional (1-d) Fibonacci quasilattice can be generated using two length-scales by the following substitution rule: L goes to LS, S goes to L. Thus we get the following sequence of L's and S's: S; L; LS; LSL; LSLLS; LSLLSLSL; LSLLSLSLLSLLS... The total number of L's and S's generated by the above substitution rule follows the Fibonacci sequence. The ratio of the number of L's to the number of S's converges to the golden mean $\tau = (1+\sqrt{5})/2$. The numbers forming the Fibonacci sequence can be generated from the recursion rule: $F_n = F_{n-1} + F_{n-2}$, $n > 2$ (with $F_1 = 1$, $F_2 = 1$) where F_n refers to the nth term of the sequence. The ratio F_n/F_{n-1} oscillates about the value of τ and converges to τ as n tends to infinity. The 1-d Fibonacci quasilattice is obtained by placing δ-function scatterers at the positions given by the formula:

$$x_n = A + n + (1/\tau)\,[B + n * (1/\tau)]$$

where x_n is the position of the nth scatterer, $A = 0{\cdot}0$ and $B = 1{\cdot}0$, [....] denotes the integer part. A segment of the 1-d quasilattice is shown in figure 1.

The diffraction pattern was numerically calculated by numerical Fourier transformation: $F(k) = \sum_n \exp(ikx_n)$. The diffraction pattern obtained from 610 scatterers by scanning the $x^*(x^* = k/2\pi)$ axis is shown in figure 2. The peaks with intensity less than 1% are not shown in the figure. The peaks could be indexed using the infinite-quasilattice formula: $k_{pq} = (2\pi/d)^*(p + q/\tau)$, $(x^*_{pq} = k_{pq}/2\pi)$ where p and q are integers and d is defined to be the average spacing between the scatterers. For the infinite Fibonacci quasilattice, $d = d_{\text{ideal}} = 1 + (1/\tau^2)$. For the finite Fibonacci quasi-

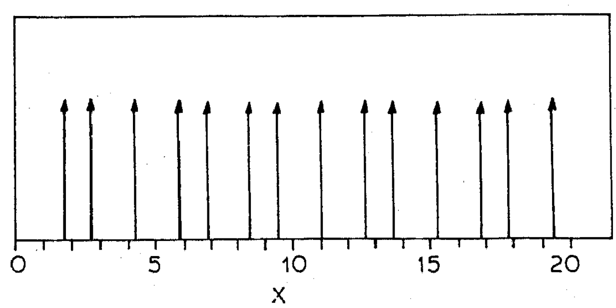

Figure 1. A segment of the one-dimensional Fibonacci quasilattice with unit δ-function scatterers.

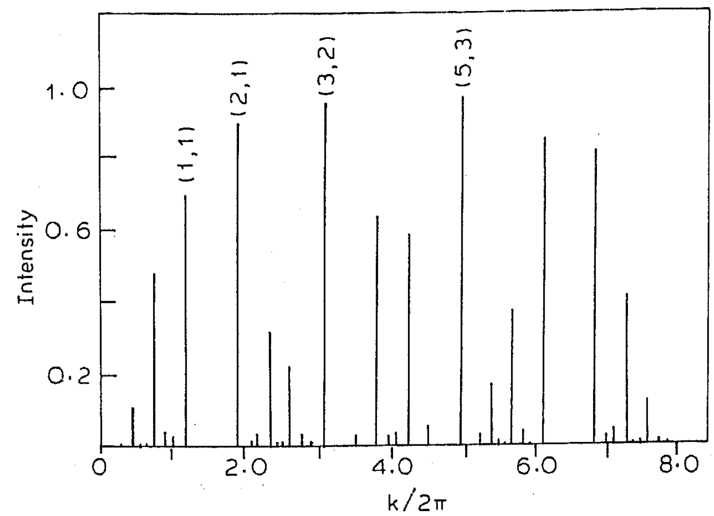

Figure 2. Diffraction from the Fibonacci quasilattice with 610 scatterers. Peaks with intensity less than 1% of the central peak ($x^*=0$) are not shown.

lattice $d=d_{\text{finite}}=(N_L\tau+N_S)/(N_L+N_S)$, where N_L=number of long intervals and N_S=number of short intervals. Some of the peaks indexed by the above formula can be seen in figure 2. When p and q are consecutive Fibonacci integers, it is found that they index the strong peaks. The weak peaks are found to be indexable when p and q are non-Fibonacci integers.

3. Results and discussion

In order to evaluate the finite size effects of the quasilattice on its diffracting property the ratio of intensity of peak to the square of number of scatterers (I/N^2) was calculated for a number of peaks (peaks near the origin and far away from the origin). It was calculated for different number of scatterers N. For a strong peak near the origin ($p=3$, $q=2$) the intensity has been followed as a function of the number of scatterers (figure 3). The fluctuations tend to settle down when the number of scatterers is greater than 50. Figure 4 shows the plot of I vs N^2 for the strong peak ($p=610$, $q=377$) far away from the origin. It can be seen that the fluctuations are within the computational error even for a small number of scatterers ($N<50$).

Figure 5 shows the plot of I vs N^2 for a weak peak ($p=2$, $q=2$) near the origin. The fluctuations tend to settle down only when the number of scatterers is greater than 800. Figure 6 shows the plot of I vs N^2 for a weak peak ($p=621$, $q=386$) far away from the origin. In this case also the intensity of the peak stabilizes around $N\sim 700$. In all cases, I/N^2 approaches constant values asymptotically.

Another interesting aspect of the diffraction from the quasilattice is the shifts in peak positions as a function of the number of scatterers. The peak-positions were found out by scanning the x^*-axis starting from the position of the peak to be expected for the diffraction from an infinite quasilattice and locating the position of the peak nearest to the peak-position for the infinite quasilattice. The peaks were found unexpectedly to shift and oscillate and finally converge to a mean value with increasing number of scatterers. For example, figure 7 shows the peak-shifts as a function of number of scatterers for a strong peak near the origin ($p=3$, $q=2$). The peak-shifts

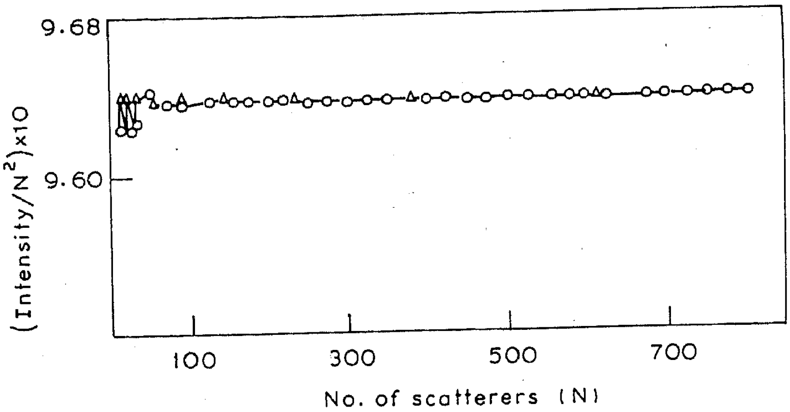

Figure 3. Plot of I/N^2 for the strong peak ($x^*_{p=3,q=2} \approx 3{\cdot}065$) nearer the origin. When the number of scatterers is around 50 the fluctuations settle down. Strong peak $p=3$, $q=2$, Circles and triangles represent non-Fibonacci and Fibonacci integers respectively.

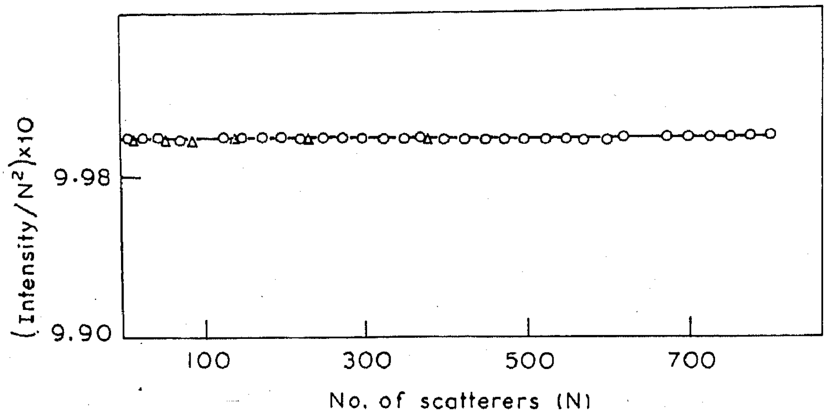

Figure 4. I/N^2 is plotted for the strong peak ($x^*_{p=610,q=377} \approx 610$) far away from the origin. Even when the number of scatterers is around 10 the fluctuations are within the computational error. Strong peak ($p=610$, $q=377$); Circles and triangles denote non-Fibonacci and Fibonacci integers respectively.

are within computational error when $N > 50$. Figure 8 is the plot of peak-shifts vs N for a strong peak ($p=610$, $q=377$) far away from the origin. The peak-shifts are negligible even for a very small number of scatterers. Figure 9 shows the peak-shifts for a weak peak ($p=2$, $q=2$) near the origin. The changes in the peak position are larger and become negligible when $N > 100$. Figure 10 shows the plot of peak-shifts vs N for the weak peak ($p=621$, $q=386$) far away from the origin. It is interesting to see that the peak-shifts are much larger and also persist till $N \sim 300$. For the weak-peaks discussed above (figures 9 and 10), the asymptotic behaviour of peak-shifts (figures 11a and 11b) seem to be almost proportional to $N^{-3/2}$.

The formula for indexing the peaks involves the d-spacing values and the d-spacing values change due to changes in N_L and N_S. The N_L and N_S values depend on the choice of the finite segment of the quasilattice. Hence the d-spacing values for various segments of the quasi-lattice containing the same number of scatterers have been calculated. In order to see if there is any correlation between the d-spacing values and the peak-position shifts, the diffraction from various segments containing the same number of scatterers has been calculated. Figure 12 shows the d-spacing changes

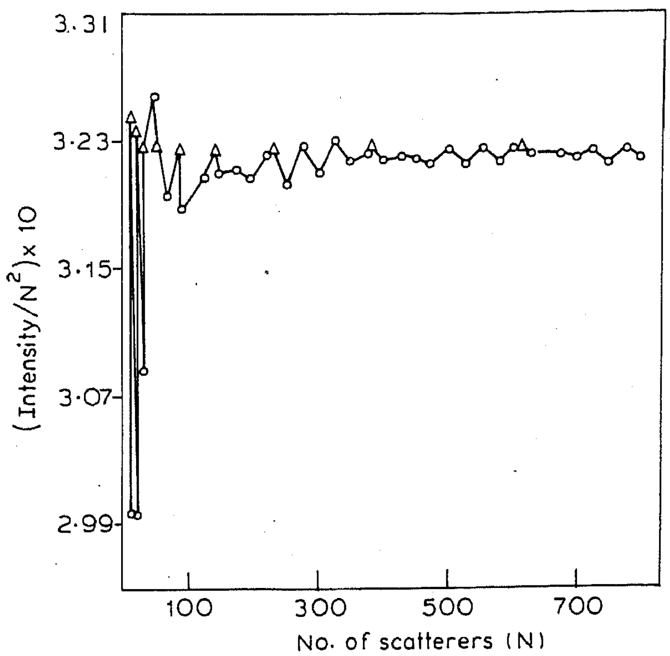

Figure 5. For the weak peak ($x^*_{p=2,q=2} \approx 2{\cdot}341$) nearer the origin, the fluctuations in I/N^2 are shown. The fluctuations tend to settle down when the number of scatterers is around 700. Weak peak ($p=2$, $q=2$).

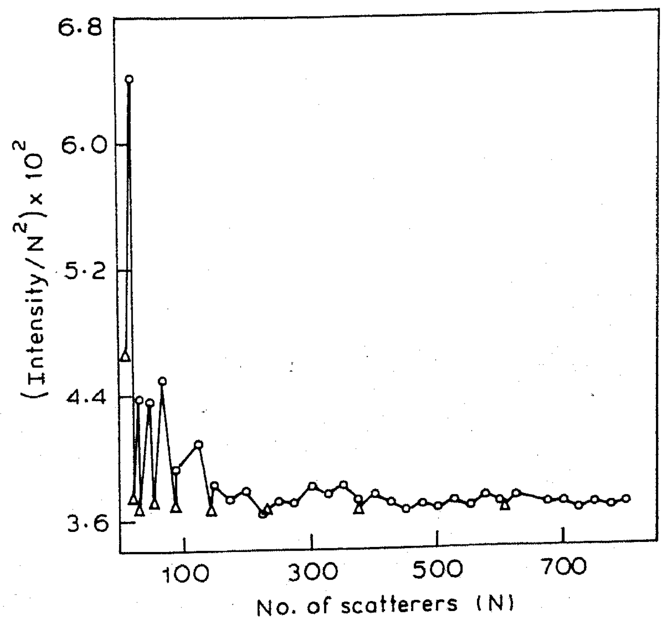

Figure 6. Plot of I/N^2 for the weak peak ($x^*_{p=621,q=386} \approx 621{\cdot}984$) far away from the origin. The fluctuations persist till $N \sim 700$. Weak peak ($p=621$, $q=386$).

for various segments with 20 scatterers along with the selected peak position shifts. Both the strong and weak peaks follow the same trend regarding the peak position shifts. It seems that there is a partial correlation of peak-shifts with the d-spacing changes. However even when the d-spacings are not changing, the peaks are found to shift in a few places. This implies the existence of contributions to the peak positions which are decided by the sequence of long and short intervals. The boundary effects

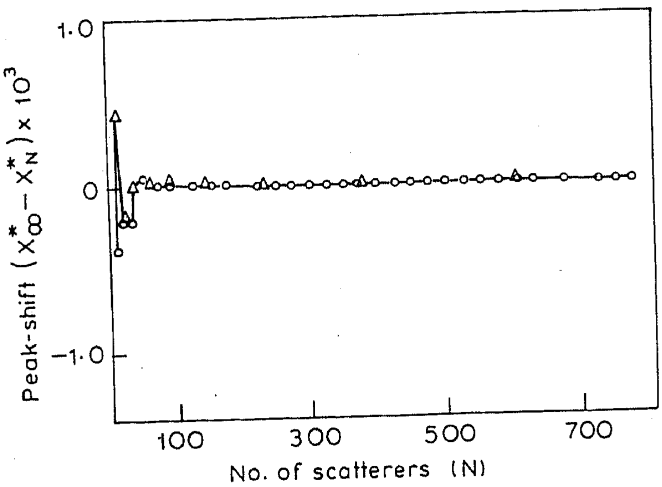

Figure 7. Shifts in peak-positions of the strong peak ($x^*_{p=3,q=2} \approx 3{\cdot}065$) nearer the origin. The peak-shifts are within the computational error when $N \sim 100$. Strong peak ($p=3$, $q=2$).

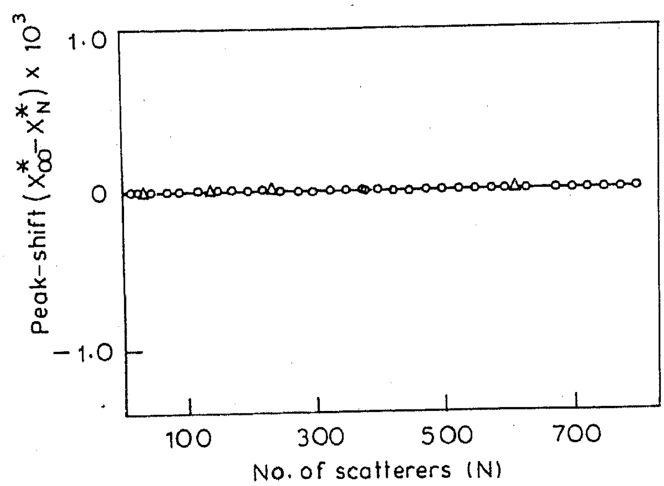

Figure 8. Shifts in peak-positions for the strong peak ($x^*_{p=610,q=377} \approx 610$) far away from the origin. Even when $N \approx 10$ the shifts are within the computational error. Strong peak ($p=610$, $q=377$).

clearly persist as the residual effect, besides the contribution from the d-spacings.

It is observed that the fluctuations in I/N^2 is larger for a weak peak than for a strong peak when both the peaks are either nearer the origin or far away from the origin. This is due to the fact that a strong peak corresponds to the constructive interference of the waves diffracted from a large number of scatterers. In the case of a weak peak, there is a partial cancellation of the many contributions. The fractional fluctuations are greater in this case of weak spots. When the size of the quasilattice changes, it may not affect the intensity of a strong peak as much as it does a weak peak.

It is found that for a weak peak or strong peak nearer the origin, the changes in peak intensities are larger than for a weak or strong peak far away from the origin, respectively. This can be understood in terms of the property of Fourier transforms. The changes in the values of the function at points far away from the origin in real space will be reflected more in the values of the transform of the function at points near the origin of the transformed space. This gives rise to the observed larger

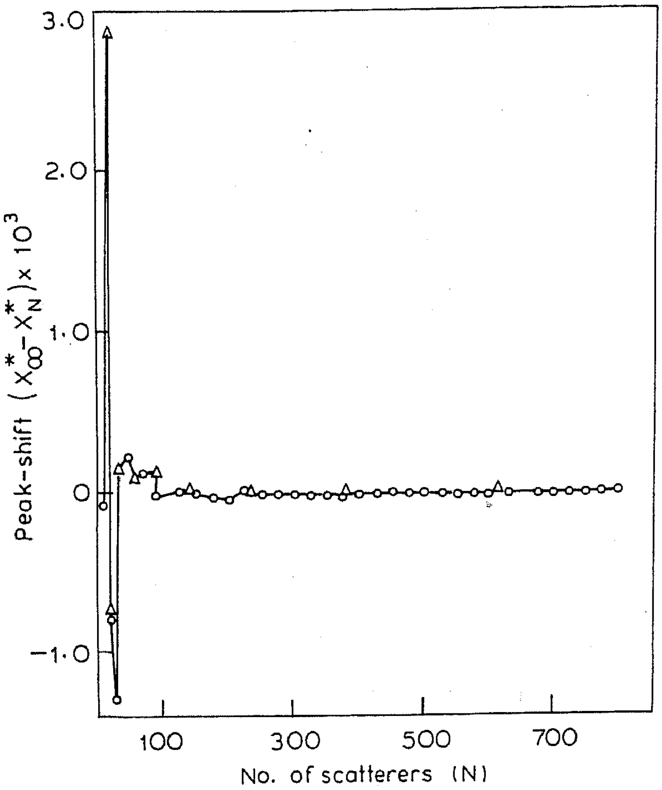

Figure 9. For the weak peak ($x^*_{p=2,q=2} \approx 2{\cdot}341$) nearer the origin, the shifts in peak-positions are shown. The peak-shifts are within the computational error when $N \sim 300$. Weak peak ($p=2$, $q=2$).

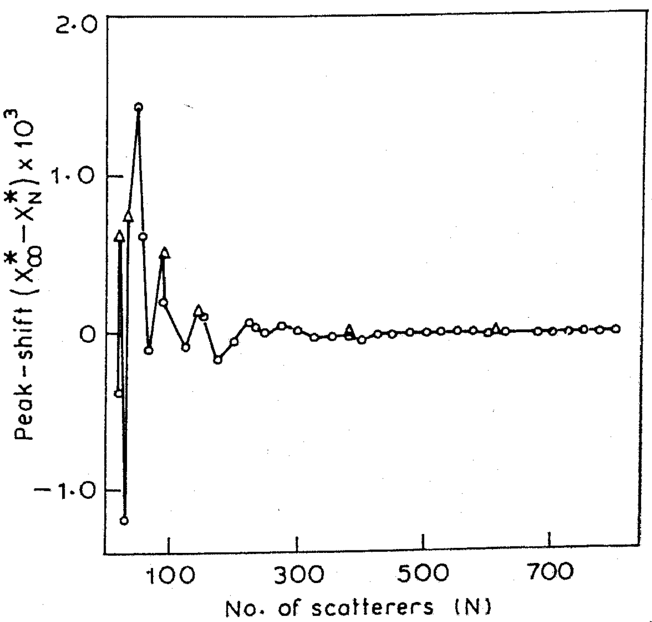

Figure 10. Peak-shifts in the weak peak ($x^*_{p=621,q=386} \approx 621{\cdot}984$) far away from the origin are shown. The peak-shifts tend to settle down when $N \sim 400$. Weak peak ($p=621$, $q=386$).

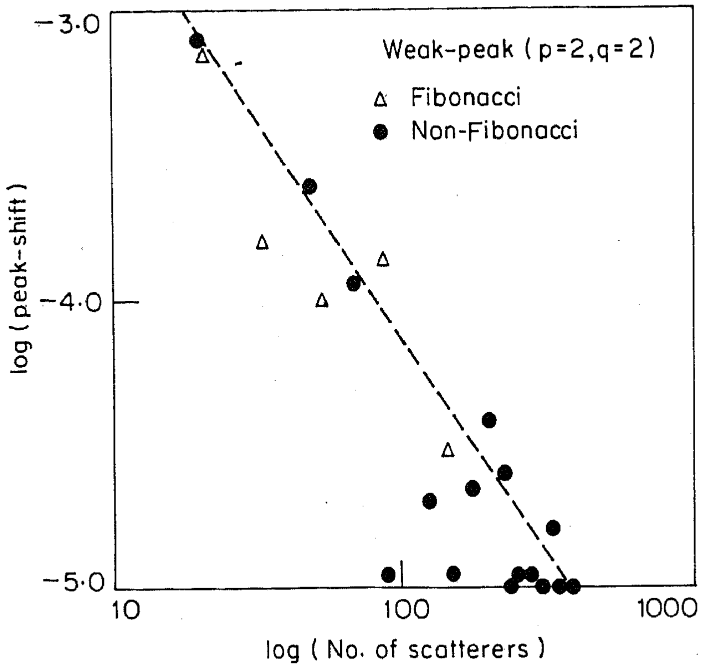

Figure 11a. Peak-shifts against number of scatterers is plotted in a log-log scale for the weak-peak ($p=2$, $q=2$). The asymptotic behaviour of the peak-shifts is shown by a dotted line. The peak-shift behaves almost like $N^{-3/2}$ for a large number of scatterers.

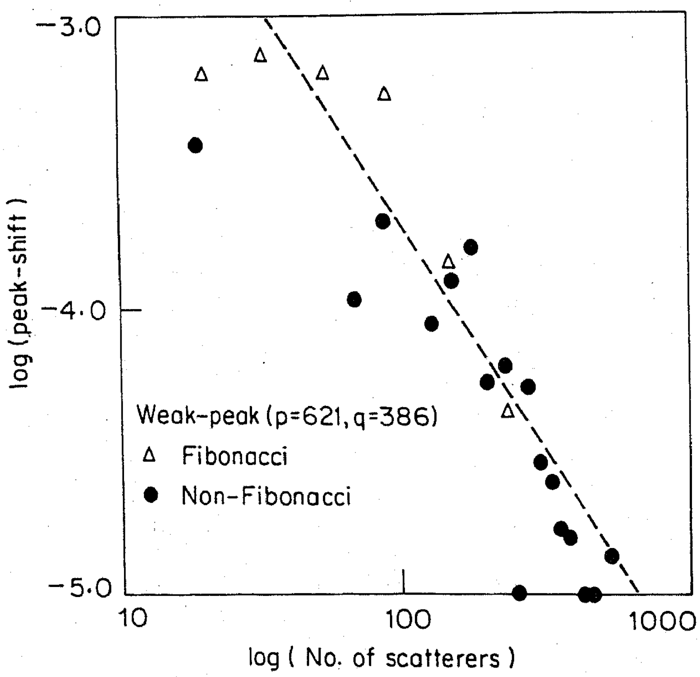

Figure 11b. Log-log plot of peak-shifts against number of scatterers for the weak peak far away from the origin ($p=621$, $q=386$). This also shows the similar asymptotic behaviour as the other weak peak.

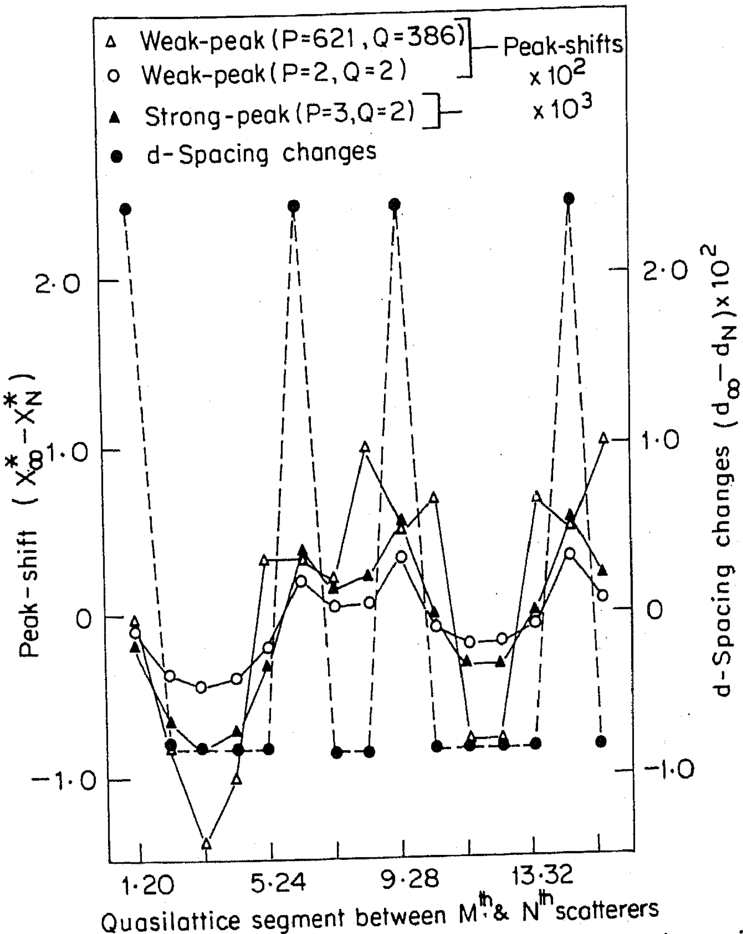

Figure 12. Peak-shifts corresponding to various segments of the quasilattice containing the same number of scatterers are shown. The dotted lines connect the d-spacings corresponding to various segments. The continuous lines connect the peak-shifts for various peaks.

peak-intensity variations nearer the origin when the size of the quasilattice and hence the boundary effects are changed.

The peak-positions of the diffraction from an infinite quasilattice have been worked out (Levine and Steinhardt 1986; Zia and Dallas 1986). But the peak-positions of the diffraction from a finite-size quasilattice will be different from that for the infinite quasilattice. The analytical methods of calculating the Fourier transforms of finite segments of Fibonacci quasilattices use the periodic boundary conditions of an infinite repetition of the finite segment. In the present case segments with a finite number of scatterers are discussed. This necessitates the use of numerical Fourier transformation to calculate the diffraction pattern. For the scattering from a truly finite number of points, the mean d-spacing and the boundary effects are noticeable.

In order to see whether similar effects are observable in the diffraction patterns of finite-size 1-d crystal, we calculated the Fourier transform of a 1-d crystal with interatomic spacings $1, \tau, 1, \tau, 1, \tau, \ldots$ (figure 13). We observed two kinds of peaks: strong and weak. Strong peaks occur at positions which are integral multiples of the inverse of the lattice spacing (Sommerfeld 1954). Weak peaks occur between the strong peaks. As the size of the crystal increases, the peak-width decreases. The strong peaks were found to have shifts in their position comparable with the computational error

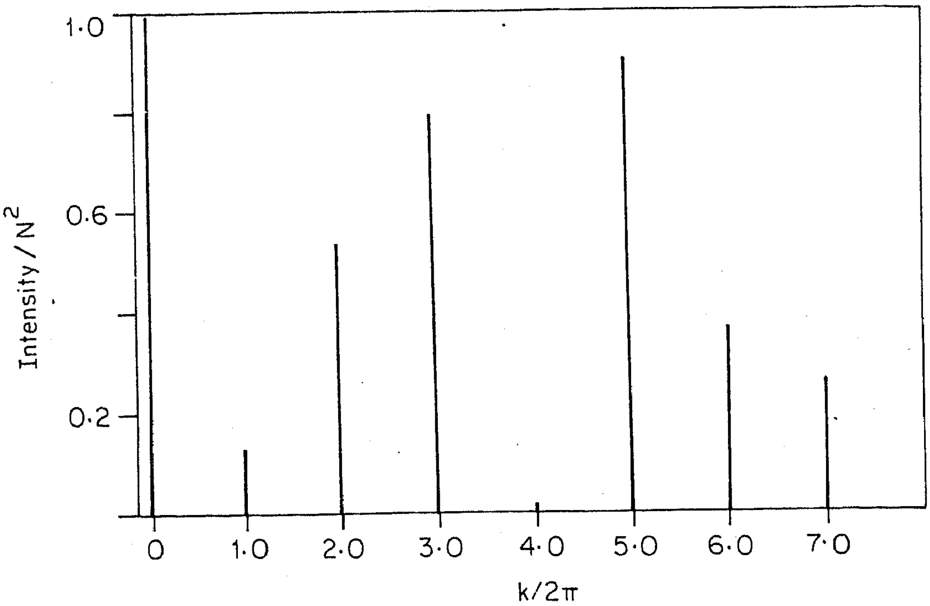

Figure 13. Diffraction pattern of 1-d crystal (1, τ, 1, τ,...) with 600 scatterers. The intensity of the strong peaks can be seen modulated in contrast to the diffraction from an undecorated lattice.

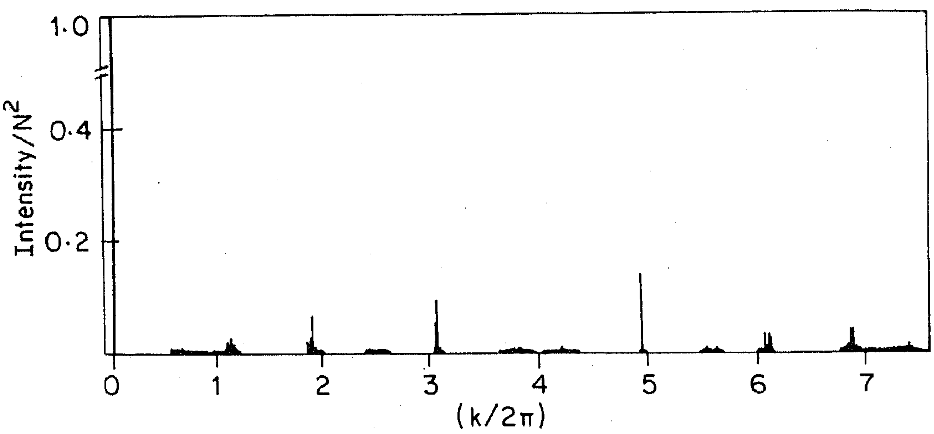

Figure 14. Diffraction from a random sequence of 1 and τ forming 600 scatterers. It shows clusters of peaks with large peak-widths.

even when the number of scatterers was as small as 5. The weak peaks were found to have shifts in their positions slightly more than that observed in the case of 1-d Fibonacci quasilattice.

In figure 14 the diffraction from a structure consisting of a random sequence of 1 and τ is shown. One can see clusters of peaks with large width compared to the 1-d Fibonacci quasilattice and also 1-d crystal. Only a few strong peaks are present when compared to the other two cases of ordered structures, within the same region of reciprocal space. The diffraction from a random structure with small number of scatterers shows many stronger peaks than from a large number of scatterers. When N is large, one tends to the situation of diffuse rings characteristic of glasses.

Recently, GaAs/AlAs and Nb/Cu Fibonacci superlattices have been grown by molecular beam epitaxial method by Todd *et al* (1986), Hu *et al* (1986), Gay and

Clemens (1987), and their diffraction properties have been studied using X-rays. The relevance of Fibonacci sequence to the 1-d quasicrystals (He *et al* 1988) discovered in Al-Ni-Si, Al-Cu-Mn and Al-Cu-Co has been pointed out (He *et al* 1988). Since peak-shifts are observed even when the number of scatterers is around 300, attempts to experimentally solve the sequencing of layers in one-dimensional quasicrystals, may have to take the peak-shifts into account. In fact, the finite segments of 1-d Fibonacci quasilattices have been used as the repeating unit in the structure factor calculations of vacancy ordered phases (Chattopadhyay *et al* 1987).

The distortions in the electron diffraction patterns of 3-d-quasicrystals have been explained only in terms of phason strain (Bancel and Heiney 1986; Budai *et al* 1987). Our calculations indicate that there will be considerable peak-shifts when the diffraction patterns of two different quasicrystalline grains of the same alloy or different regions of the same quasicrystalline grain are compared, even if the same amount of disorder is present. It has also been pointed out recently (Ma *et al* 1989), that ignoring the background in the diffraction pattern while calculating the Patterson function may lead to a shift in peak-positions and the disappearance or introduction of spurious peaks. It seems possible that peak-shifts can be observed experimentally at the resolution at which the X-ray diffraction experiments on Fibonacci superlattices have been performed by Terauchi *et al* (1988). The peak-positions and widths should change when the number of layers forming the Fibonacci superlattice is increased from a small value. Some attempts are being made to analytically find out the peak positions of the diffraction from finite size quasilattices.

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