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Simulation of the diffraction pattern of one dimensional quasicrystal

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Abstract

The effects of the variation of atomic spacing ratio of a one dimensional quasicrystal material are investigated. The work involves the use of the solid state simulation code, Laue written by Silsbee and Drager. We are able to observe the general features of the diffraction pattern by a quasicrystal. In addition, it has been found that each golden mean produces a unique diffraction pattern and that the lower the golden mean the better the diffraction pattern resembles that of a periodic chain. Also the intensity of the central peak was found to decrease as the golden mean increases. However the value of golden mean has no effect on the spacing between the Bragg planes.

Keywords: Quasicrystal

1.0 Introduction

The discovery of quasicrystal by Shechtman et al more than three decades ago [1] signaled the beginning of a remarkable scientific revolution [2], in which some of the most basic notions of condensed matter physics have undergone a thorough re-examination. Today, the science of quasicrystals, with its growing number of textbooks, is in its adolescence. Old paradigms are being carefully transformed into new ones [3]; definitions are being changed [4]; space-group theory has been generalized to quasicrystals using two alternative approaches [5], and even extended to treat novel long-range order possessing colour [6] or magnetic symmetry [7].

In a quasicrystal, the local arrangements of atoms are fixed, but each cell has a different configuration of cells nearby. Although the structures are strikingly similar to the quasiperiodic tiling invented by the mathematician, Roger Penrose and which Martin Gardner popularized in a 1977 Mathematical Games column in Scientific American, there was little in the crystallographic field to presage the experimental breakthrough[8]. Shechtman et al themselves did not immediately recognize the quasiperiodic structure in his sample, and was at first mystified by the diffraction pattern[9].

Diffraction is the primary technique for examining the structures of solid materials, and the consequence of aperiodicity on diffraction patterns provides a challenge and an opportunity to gain a deeper understanding of both diffraction and of quasicrystal structure [10-13]. Most common procedure for analyzing diffraction measurement from crystalline solid were originally developed from the assumption of a periodic structure unit. The definition of a crystal included the requirement of periodicity until 1992[14]. Aperiodicity and traditionally forbidden rotational symmetries introduce features in diffraction patterns that make interpretation by the traditional means more complex. Simulations can provide a means for building intuition of the effect of quasicrystalline order on diffraction patterns without resorting to a complicated analysis.

One of the most unexpected aspects of diffraction from quasicrystals is that they produce sharp, and discrete diffraction peaks [15]. A disruption of periodicity often leads to a (loss of definition) broadening of peaks, and increase in background intensity in the diffraction pattern. However, aperiodicity by itself does not cause a broadening of diffraction peaks. This can be demonstrated in one dimension by looking at the Fourier transform of the well-ordered but aperiodic Fibonacci array of one dimensional sequence of 'short' (S) and 'long' (L) atom generated by specific rules [16]. In the limit of an infinite number of atoms, the ratio of the number of L segment to S segment is given by the irrational number$\tau = \frac{(1+\sqrt{5}}{2}$, known as golden mean [17]. A Fourier transform of this array does not show the broad peaks that are

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normally associated with disordered structures. Unlike transform from periodic arrays, the transform from a Fibonacci array forms a dense set of sharp peaks. having different intensities. The positions of the peaks are related to each other by power of golden mean [17].

The history of quasicrystal begins with the 1984 paper [1] "Metallic phase with long-range orientation order and no translation symmetry" where D. shechtman et al demonstrated a clear diffraction pattern with a fivefold symmetry. The pattern was recorded from an Al-Mn alloy which has been rapidly cooled after melting. Ishimasa et al [19] reported twelve fold symmetry in Ni-Cr particles. Over the years, hundreds of quasicrystals with various compositions and different symmetries have been discovered. The first quasicrystalline materials were thermodynamically unstable-when heated. The first of many stable quasicrystals were discovered making it possible to produce large samples for study and opening the door to potential applications [3].

N. Ferralis, et al [20] investigate the diffraction from One- and two dimensional quasicrystalline gratings" on one dimensional quasicrystal using two different approaches (laser diffraction and calculated diffraction method using Fibonacci series). The research suggested that, laser diffraction allows an inductive approach for understanding complex aperiodic structure, and can provide educators with an innovative tool for introducing and extending the traditional concept of diffraction.

In 1985, Levine [21] conducted a theoretical work on the diffraction pattern for an ideal model of quasilattice in which identical atoms are placed at each point of the lattice, and obtained results that are in agreement with the observation earlier reported by Shechtmen, et al [1].

To generate a quasicrystal patterns, so called projection methods have been developed[22-24]. Projection methods are mathematical constructs that projects sections of hypercubic lattice onto lower dimensional spaces. The first such projection was given by de Bruijn [25], where he showed that the vertices of the two-dimensional penrose pattern of darts and kites can be generated by hypercubic lattice into a one-dimensional.

In this work, the effects of varying the atomic spacing ratio(golden mean) are investigated on one-dimensional quasicrystal material using the code 'Laue' written by Silsbee and Drager [18]. Laue is based on the projection technique, however, unlike traditional projection work, in which the emphasis is entirely on the diffraction pattern, the code can provide additional data that can be used to characterize the quasicrystal.

2.0 Diffraction Pattern from Quasicrystal

The diffraction patterns of the quasicrystal consist of a set of Bragg peaks that densely fill reciprocal space in an array with quasicrystal symmetry. Consider the case of one-Dimensional quasicrystal with atomic position of the N^{th} atom given by;

$$X_N = N + \alpha + \frac{1}{\tau}\left[\frac{N}{\tau} + \beta\right], \tag{1}$$

where τ = golden ratio ; α and β are arbitrary real numbers and $\left[\frac{N}{\tau} + \beta\right]$'s represent the greatest integer function. This particular example is central to study the pentagonal and icosahedral quasilattice. The atomic position of the 1D (Fibonacci) quasicrystal described by Eqn. (1) may be re-expressed as;

$$X_N = n\left(1 + \frac{1}{\tau^2}\right) + \beta\sqrt{5} + \left[-\frac{1}{\tau}\left\{\frac{n\left(1+\frac{1}{\tau^2}\right)+\beta\sqrt{5}}{\sqrt{5}}\right\} - \beta\tau + \alpha\right], \tag{2}$$

where $\left\{\frac{n\left(1+\frac{1}{\tau^2}\right)+\beta\sqrt{5}}{\sqrt{5}}\right\}$ signify the fractional part function and we have used the fact that $\left(1 + \frac{1}{\tau^2}\right) = \sqrt{5}$. An identity is $X = [X] + \{X\}$. The function $\{X\}$ is periodic in X with period 1. This expression is of the general form;

$$X_n = na + \varphi + F(na + \varphi), \tag{3}$$

where $F(x)$ is periodic in x with period $b, and\ \frac{a}{b}$ is irrational. Expressions of this variety arise in the study of Frenkel-Kontrova model[17], which describe a 1D in commensurate crystal. The Fourier transform of such a set of atomic positions consist of Bragg peaks at positions

$$K = \frac{2\pi M}{a} + \frac{2\pi N}{b},$$

where M and N are integers. This result may be obtained by expanding the exponential $e^{ikF(na+\varphi)}$appearing in the expression for the transform in a Fourier series of its own and employing the completeness relation for complex exponentials. For our case, this means that there will be peaks at

$$K_{pq} \equiv \frac{2\pi}{1+\frac{1}{\tau^2}}\left(p + \frac{q}{\tau}\right), \tag{4}$$

where p and q are integers. With this in mind, we will compute the diffraction pattern (i.e. Fourier transform of the 1D.

quasicrystal of Eqn. (2))

$$F_i(K) = \lim_{N\to\infty} \frac{1}{N} \sum e^{iKX_n}\,, \tag{5}$$

where we are summing over the N atomic position in the chain. First consider $F_i(K)$ for K of the form $K = K_{pq}$ as define in eqn. (4). Noting the identity $\left(1 + \frac{1}{\tau^2}\right) = \sqrt{5}$, the exponent in eqn. (5) is given by;

$$\begin{aligned} iK_{pq}X_n &= 2\pi i\left(pn + q\frac{n}{\tau}\right) + K_{pq}\left(\frac{\beta}{\tau} + \alpha - \frac{1}{\tau}\left\{\frac{n}{\tau} + \beta\right\}\right) \\ &= 2\pi i\left(pn + q\left[\frac{n}{\tau} + \beta\right]\right) + i\left(2\pi q - \frac{K_{pq}}{\tau}\right)\left\{\frac{n}{\tau} + \beta\right\} + iK_{pq}\alpha - i\left(2\pi q - \frac{K_{pq}}{\tau}\right)\beta \end{aligned} \tag{6}$$

The first term in the expression $2\pi i\left(pn + q\left[\frac{n}{\tau} + \beta\right]\right)$ is an integer times $2\pi i$ and therefore only yield a factor of unity upon exponentiation. The last two terms $iK_{pq}\alpha - i\left(2\pi q - \frac{K_{pq}}{\tau}\right)\beta$ are independent and contributes to the sum in an important fashion. Since $0 \le \left\{\frac{n}{\tau} + \beta\right\} \le 1$, the second term $i\left(2\pi q - \frac{K_{pq}}{\tau}\right)\left\{\frac{n}{\tau} + \beta\right\}$ lies between zero and iX, where $X \equiv 2\pi q - \frac{K_{pq}}{\tau}$ since τ is an irrational number, the value of the second term is uniformly and densely distributed in the interval (0, X), enabling us to approximate the sum in eqn. (5) by an integral;

$$F_i(K) = \frac{e^i}{X}\int_0^X e^{iy}dy = \frac{\sin\frac{X}{2}}{\frac{X}{2}}e^{i\gamma}, \tag{7}$$

where $\equiv\ K_{pq}\alpha - \left(2\pi q - \frac{K_{pq}}{\tau}\right)\beta$ and $\gamma \equiv\ + \frac{X}{2}$. Equation (7) is the value of $F_i(K)$ for the special values$K = K_{pq}$, which we argued, correspond to the position of bragg peaks. Thus we conclude that

$$F_i(X) = \sum_{pq}\left(\frac{\sin\frac{X}{2}}{\frac{X}{2}}e^{i\gamma}\right)\delta\left(K - K_{pq}\right). \tag{8}$$

Those familiar with the computation of the fourier transform of 1D quasicrystal via projection method. will recognize that the two methods agree exactly.

The brightest spots occur for those $K = K_{pq}$ where X is small. This occurs when $\frac{q}{p}$ is close to τ. It is well known that the best rotational approximants to τ occur when q and p are successive Fibonacci number F_n. This means that the sequence of most intense peaks corresponds to $(p, q) = (F_{n+1}, F_n)$[19].

3.0 Methodology

3.1 Laue Code

The code, "Laue" written by Silsbee and Drager [18] was employed in this work. Basically the code computes the electron density and diffraction pattern of a one-dimensional array of atoms. The one-dimensional crystal is represented by it is real-space electron density which is generated from superposition of atomic electron densities. The corresponding diffraction is then computed as the square of the Fourier transform of that electron density. To compute the diffraction pattern (Intensity) of the electron density, "Laue" uses a Fast Fourier Transform (FFT) routine. The intensities are normalized to give a height for the central peak of one for the monatomic quasicrystal.

The program has two main output windows for displaying the electron density and diffraction pattern respectively. The code has eight menus namely: Quit, Display, Configure, Presets, Help, Material, Modulation and Calculate. The material menu allows user to select the type of material simulated. The possible options are Monatomic crystal, Diatomic crystal, Single atom, Pair of atoms, Liquid and Quasicrystal. In this case of simulating quasicrystal, the program has the following variables: Lattice constant, Size of atom, the Spacing ratio and Atom shape. In this work the atomic shape is chosen to be Gaussian.

3.2 Procedure

The lattice constant, size of the atom were set to 4.0 A and 0.1 A respectively and the atomic potential was chosen to be Gaussian. The atomic spacing ratio was set to be 1.05 and then the simulation executed. The data for the diffraction pattern was then exported to excel for further analysis which includes among others, the comutation of the width of the diffraction pattern and the separation of the Bragg planes. This was repeated for atomic spacing ratio of 1.0, 1.2, 1.3, 1.4, 1.5, 1.6, 1.7, 1.8, 1.9 and 2.0.

4.0 Results And Discussion

4.1 Results

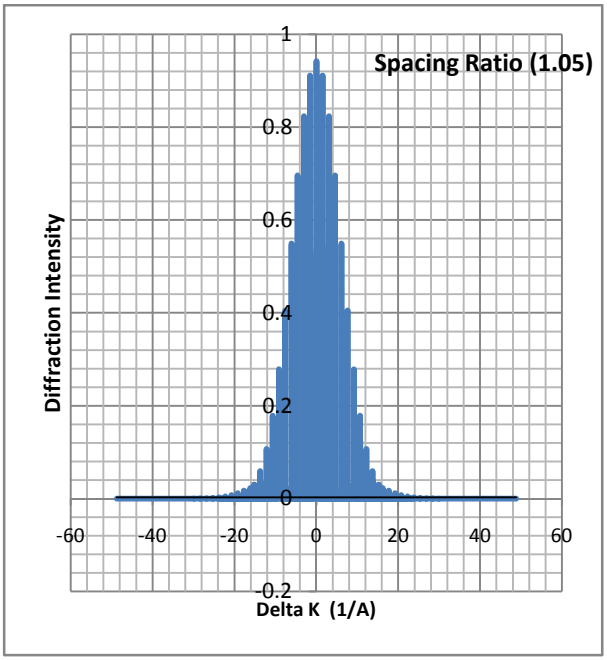

FIG. 1 The diffraction pattern of one dimensional quasicrystal quasicrystal at spacing ratio of 1.05

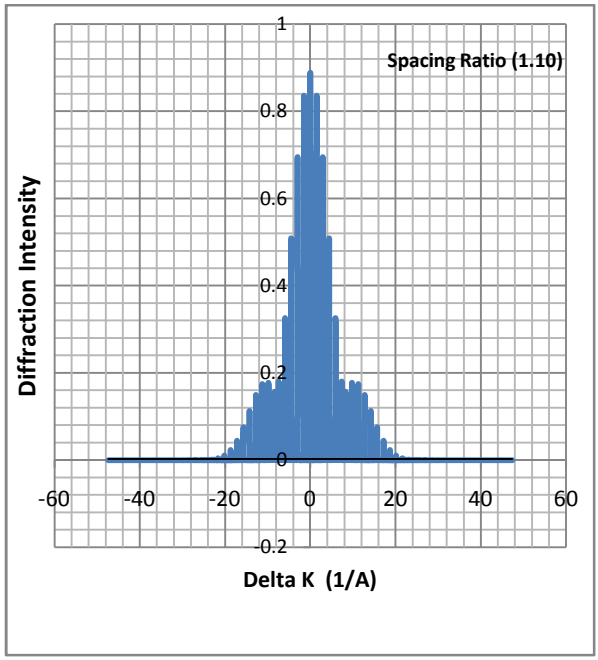

FIG. 2 The diffraction pattern of one dimensional quasicrystal at spacing ratio of 1.1

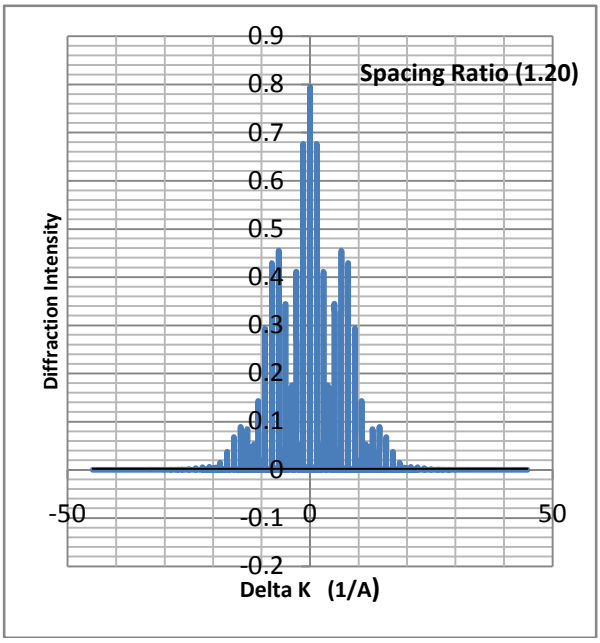

FIG. 3 The diffraction pattern of one dimensional quasicrystal quasicrystal at spacing ratio of 1.20

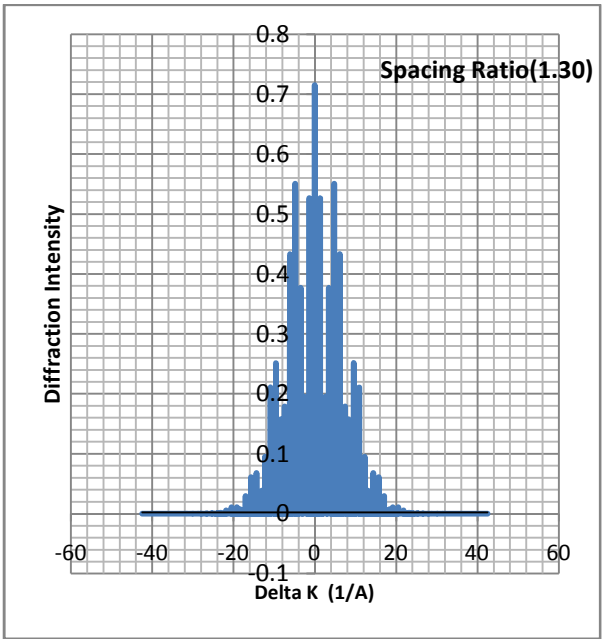

FIG. 4The diffraction pattern of one dimensional quasicrystal at spacing ratio of 1.30

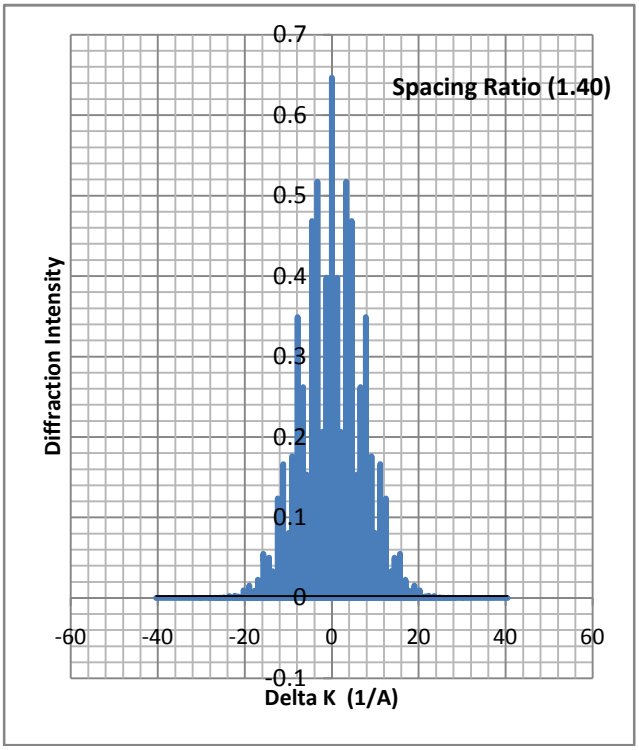

FIG. 5 The diffraction pattern of one dimensional quasicrystal quasicrystal at spacing ratio of 1.40

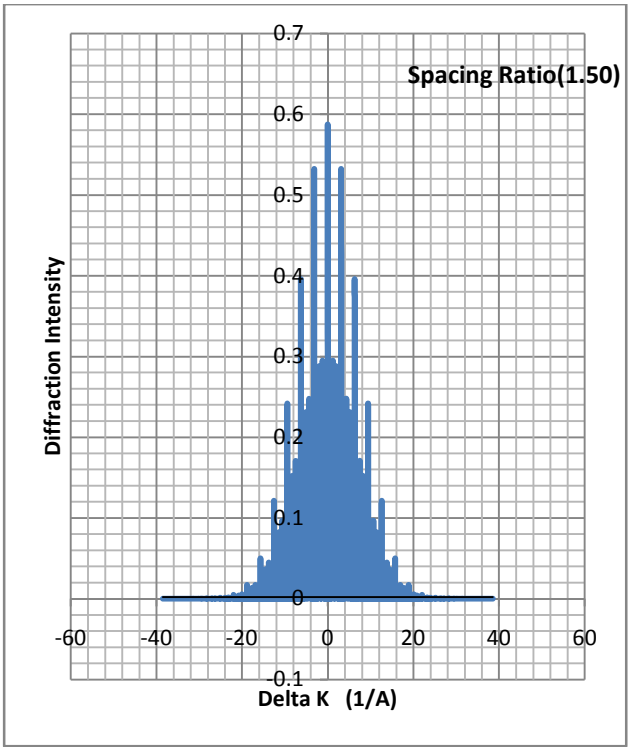

FIG. 6 The diffraction pattern of one dimensional quasicrystal at spacing ratio of 1.50

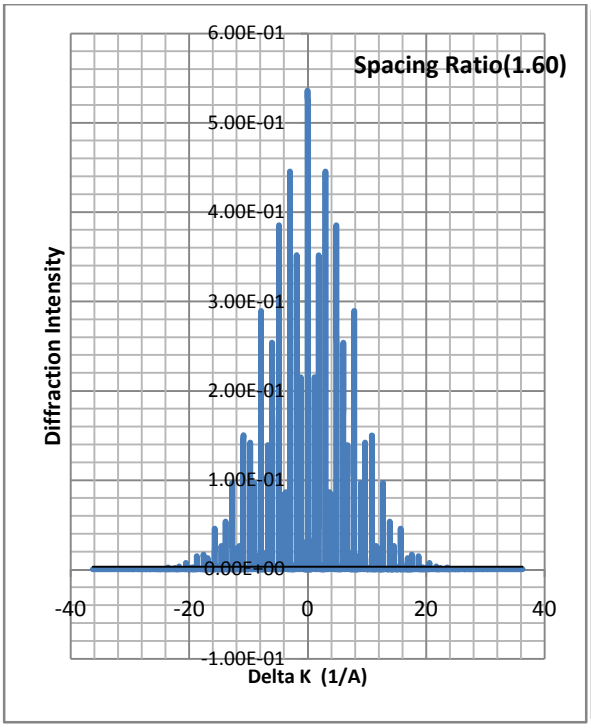

FIG. 7 The diffraction pattern of one dimensional quasicrystal quasicrystal at spacing ratio of 1.60

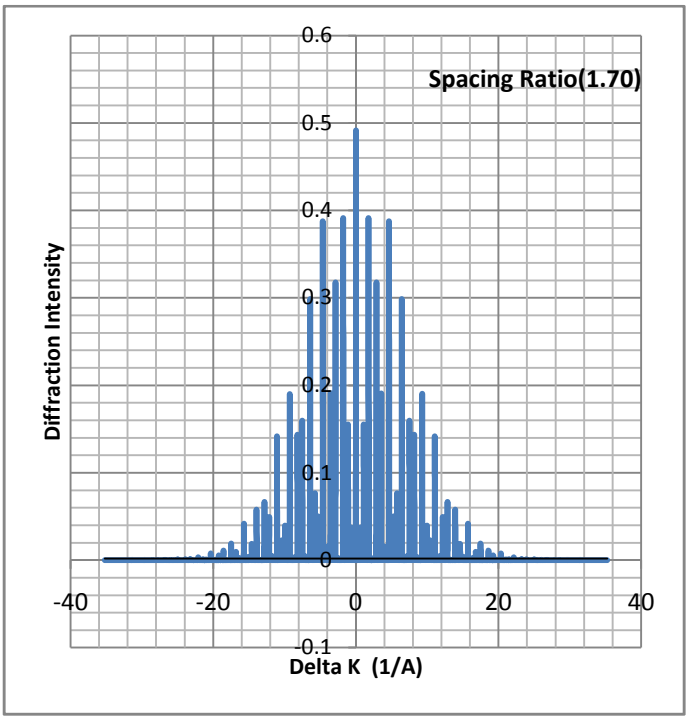

FIG. 8 The diffraction pattern of one dimensional quasicrystal at spacing ratio of 1.70

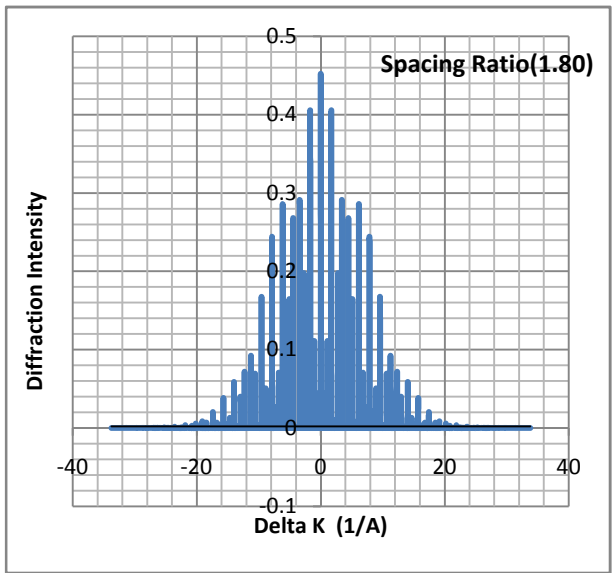

FIG. 9 The diffraction pattern of one dimensional quasicrystal quasicrystal at spacing ratio of 1.80

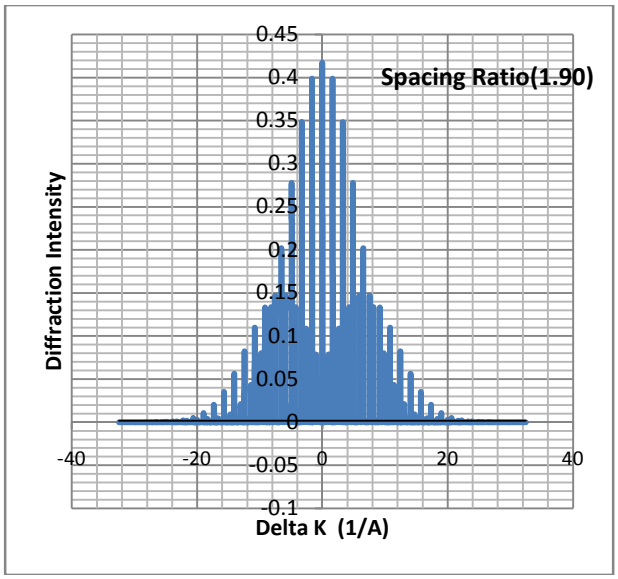

FIG. 10. The diffraction pattern of one dimensional at spacing ratio of 1.90

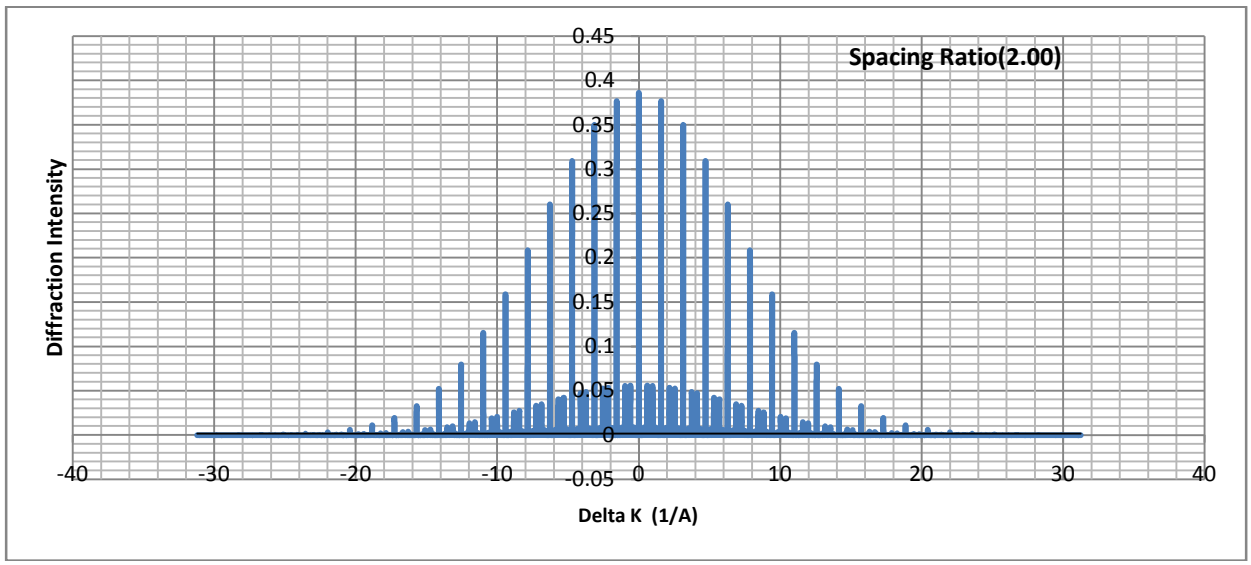

FIG. 11.0 The diffraction pattern of one dimensional quasicrystal at spacing ratio of 2.0

The diffraction patterns for the various golden means investigated are shown in figures 1-11. The following are observed:

i. Each diffraction pattern has sharp peaks, just like those of the periodic crystal, however, the peaks have sidebands and the sidebands have more sidebands. In general, there are more sidebsnds on the sidebands as you look with more sensitivity.

ii. The spatial distribution of the diffractions patterns are different, i.e each golden means produces a unique diffraction pattern. The lower the golden mean the better the diffraction pattern resembles that of the periodic monatomic chain.

iii. The intensity of the central peak decreases as the golden mean increases.

iv. The width of the diffraction pattern decreases as the lattice constant increases. Also the width decreases as the size of the atom is increase.

v. The variations of the separation between Bragg planes with scattering wave vector, ΔK are shown in table 1. It can be seen from the table that for a given golden mean,d_h decreases initially very fast with increasing scattering vector and latter becomes almost flat i.e. constant. Also as expected the variation of d_h with scattering vector is independent of golden mean.

Table 1: variation of the separation between Bragg planesd_h with scattering wave vector Δk

Golden mean					
1.05	$\Delta k\ (A^{-1})$	1.4740	3.2730	4.8480	6.4220
	$d_h\ (A)$	4.2627	1.9197	1.2960	0.9784
1.10	$\Delta k\ (A^{-1})$	1.6500	2.9610	4.4900	6.2380
	$d_h\ (A)$	3.8080	2.1220	1.3994	1.0072
1.20	$\Delta k\ (A^{-1})$	1.3540	6.5180	7.9650	9.2040
	$d_h\ (A)$	4.6405	0.9640	0.7889	0.6827
1.30	$\Delta k\ (A^{-1})$	1.4800	4.8110	6.3790	9.7100
	$d_h\ (A)$	4.2454	1.3060	0.9850	0.6471
1.40	$\Delta k\ (A^{-1})$	1.2210	3.2710	4.7620	8.1160
	$d_h\ (A)$	5.1459	1.9209	1.3194	0.7741
1.50	$\Delta k\ (A^{-1})$	3.1180	6.4930	9.6910	12.7100
	$d_h\ (A)$	2.0151	0.9677	0.6484	0.4943
1.60	$\Delta k\ (A^{-1})$	1.7910	2.9790	4.8460	7.9010
	$d_h\ (A)$	3.5082	2.1092	1.2965	0.7952
1.70	$\Delta k\ (A^{-1})$	1.8770	2.8520	4.8010	6.5880
	$d_h\ (A)$	3.3475	2.2030	1.3087	0.9537
1.80	$\Delta k\ (A^{-1})$	1.6440	3.3580	6.1630	8.0320
	$d_h\ (A)$	3.8219	1.8711	1.0195	0.7822
1.90	$\Delta k\ (A^{-1})$	1.5800	3.3760	5.0220	6.6690
	$d_h\ (A)$	3.9767	1.8611	1.2511	0.9421
2.0	$\Delta k\ (A^{-1})$	1.6640	3.2480	4.6880	6.4160
	$d_h\ (A)$	3.7759	1.9345	1.34030	0.9793

These observations are fundamentally the characteristic of the diffraction pattern of quasicrystals and are in agreement with the finding of other workers [1, 4, 21, 25].

4.2 Discussion

One of the most unexpected aspects of diffraction from quasicrystals is that they produce sharp, discrete diffraction peaks. A disruption of periodicity often leads to loss of definition i.e. broadening of peaks, increase in background intensity in the diffraction pattern. However, aperiodicity by itself does not cause a broadening of diffraction peaks. This can be demonstrated in one dimension by looking at the Fourier transform of the Fibonacci array. The Fourier transform of this array does not show the broad peaks that are normally associated with disordered structure. Unlike transforms from periodic arrays, the transform from Fibonacci array forms a dense set of peaks having different intensities. The positions of the peaks are related to each other by the powers of the golden mean[17]. Thus each golden mean produces a unique diffraction pattern as observed in this work.

As the lattice constant of a pure monatomic chai is increased, the width of the diffraction pattern increases and the intensity of the peak for a given scattering wave vector (ΔK) also increases [18]. Increasing the lattice constant decrease the primitive reciprocal lattice vector which in turn translate into decreasing the chances of destructive interference. Increasing the lattice constant of a pure monatomic chai is equivalent to increase the spacing between the atoms. Thus one

might wrongly conclude that as the golden mean of Fibonacci array is increased there will be an increase in the intensity of the diffraction peaks. However, we found in this work that intensity of the central peak decreases as the golden mean increases. In reality, the intensities of diffraction from crystal depend upon the relative positions of the atoms in the unit cell, as well as upon the atomic numbers of the atoms and the electron distribution within each of the atoms. It also be noted that, the spacing between the i^{th} and $(i+1)^{th}$ atom is given by the lattice constant if the i^{th} position in the Fibonacci chain is 0, otherwise the separation is given by the lattice constant times the spacing ratio. Thus the golden mean (spacing ratio) cannot be infinitely increased because any increase above a certain maximum will result in placing the i^{th}atom outside the i^{th}cell. Thus as the golden mean is increased (within the allowed range) the separation between the atoms in the neighboring cells decreases thereby resulting in the decrease in the intensity of the central peak as observe in this work.

5.0 Conclusion

The effect of the variation of atomic spacing ratio in a Fibonacci array has been investigated using the code 'Laue'. We are able to observe the basic features of the diffraction by a quasicrystal. Most importantly the following are established in this work:

i. Each golden mean produces a unique diffraction pattern and that the lower the golden mean the better the diffraction pattern resembles that of a periodic monatomic chain.
ii. The intensity of the central peak decreases as the golden mean increases
iii. The variation of the separation between Bragg planes is found to be independent of the golden mean

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