LETTER TO THE EDITOR

A simple derivation of quasi-crystalline spectra

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Abstract. A quasi-crystalline solid is an *aperiodic* array of atoms which gives rise to Bragg-like diffraction peaks. Using the projection method for generating such structures, we provide a simple derivation of this paradoxical behaviour.

An exciting new development in condensed matter physics is the discovery of quasi-crystalline structures, i.e., aperiodic arrays of atoms which nevertheless give rise to Bragg-like peaks in diffraction patterns. On the experimental front, these patterns were observed by Shechtman *et al* (1984). On the theoretical side, Levine and Steinhardt (1984) found a structure with a Fourier transform fitting the observed pattern. In addition, these authors pointed out that this structure is not just an isolated case, but belongs to a new class of ordered structures which they termed 'quasi-crystals'. The construction for these structures given by Levine and Steinhardt (1984) does not display any clear 'hidden symmetry', so that the emergence of delta functions in the Fourier transform seems somehow mysterious. By contrast, an elegant method for generating these structures, which manifestly displays a 'hidden symmetry', was reported by Elser (1985). This symmetry in turn allows a simple way of obtaining the spectrum of the quasi-crystalline structure, showing transparently how delta functions appear. Following a brief description of a quasi-crystalline solid, we then show how to obtain the spectrum of a one-dimensional quasi-crystalline array and finally we give some observations on generalisations and applications.

A quasi-crystalline structure may be mathematically represented as a sum of delta functions located at a discrete set of points in D-dimensional space, distributed neither randomly nor periodically. For $D = 2$, for example, these points may be located at vertices of a Penrose tiling pattern (Penrose 1974, Gardner 1977). In general dimensions, such a set of points, called a Penrose lattice by Levine and Steinhardt (1984), are generated by a complex set of rules. They represent generalisations of vertices in the Penrose tiles. The clearest way to obtain a quasi-crystalline array of points is provided by Elser (1985), briefly described here.

Consider a regular periodic lattice of points in N-dimensional hyperspace with a D-dimensional $(D < N)$ hypersurface embedded at a generic orientation with respect to the lattice. Take the set of points which lie within a certain distance, w, from the hypersurface and project them onto it. The resultant is a generalised Penrose lattice in D dimensions. For the sake of simplicity, we restrict ourselves for the moment to

a square ($N = 2$) lattice with a line embedded, so that we deal with a quasi-crystalline 'string'.

To be specific, we choose a line passing through a point on the square lattice with unit spacing. Let the angle between this line and the x axis be θ. Label this line the ξ axis, the orthogonal one the η axis, and the point is the origin in both the xy and $\xi\eta$ coordinate systems (see figure 1). If $\tau = \cot \theta$ is not rational, then, there will not be another point on this line and the projected set of points will clearly be arranged aperiodically. (Technically, such an array is intimately related to quasi-periodic and almost-periodic functions. See, e.g., Katznelson (1976) and Avron and Simon (1981).)
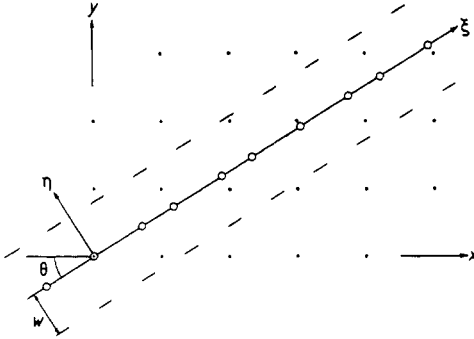

Figure. 1. The quasi-crystalline string of atoms are located at the open circles. They are images of the square lattice points, lying within the broken lines, projected onto the ξ axis.

The square lattice of points may then be represented by the (generalised) function $U_0(x, y)$:

$$U_0(x, y) = \sum \delta(x - j)\delta(y - l)/(4\pi^2), \tag{1}$$

where the sum is over all integers (j, l). The projection described above amounts to nothing more than
  (a) writing U_0 in terms of ξ and η,
  (b) multiplying by $R(\eta) \equiv \{1 \text{ if } |\eta| < w \text{ and } 0 \text{ otherwise}\}$ and
  (c) integrating over η.
Thus, the function

$$Q(\xi) = \int d\eta \, R(\eta) U(\xi, \eta) \tag{2}$$

where $U(\xi, \eta) \equiv U_0(\xi \cos \theta - \eta \sin \theta, \xi \sin \theta + \eta \cos \theta)$, is an implicit way of writing a sum of δ functions located at a set of 'quasi-crystalline' points $\{x_n\}$.

If a string of atoms were placed at these points and a diffraction pattern taken from this array, the intensity of the pattern is related simply to the Fourier transform of $Q(\xi)$. This we denote by $F(k)$, which is $\sum_n \exp(ikx_n)$. If we were to find an explicit form for $\{x_n\}$ first and then do the sum over n, we would face a non-trivial task. However, to take the transform of Q in its implicit form (2.2) is exceedingly simple. Recall that, if f is the Fourier transform of g, then $\int g$ is just $f(0)$. Thus, $Q(\xi)$ may be regarded as the transform (with respect to η only) of the function $R(\eta)U(\xi, \eta)$, evaluated at zero. Since we have a product, the Fourier transform is a convolution.

Writing $S(p)$ and $M(\xi, p)$ as the transforms of R and U respectively, we have

$$Q(\xi) = \int dp\, S(-p) M(\xi, p). \tag{3}$$

However, we are interested not so much in $Q(\xi)$ as its transform $F(k)$, which is

$$F(k) = \int dp\, S(-p) V(k, p), \tag{4}$$

where $V(k, p)$ is the (complete, 2D) Fourier transform of U.

To continue, we simply note that U is just U_0 rotated, i.e., a sum over δ functions at rotated points. Therefore, V is also nothing more than a sum over 'rotated' δ functions, i.e.,

$$V(k, p) = \sum \delta[k - 2\pi(n \cos\theta + m \sin\theta)]\delta[p - 2\pi(-n \sin\theta + m \cos\theta)]. \tag{5}$$

With the explicit $S(p) = 2w[\sin(pw)/pw]$, we have

$$F(k) = \sum S[2\pi(-n \sin\theta + m \cos\theta)]\delta[k - 2\pi(n \cos\theta + m \sin\theta)], \tag{6}$$

which displays clearly the spectrum as a sum of densely distributed δ functions, with appropriate weights.

Equation (2.6) also shows that the location of the δ's are those from projection of the complete square but rotated lattice in k-space, with lesser strengths for the more distant lattice points. Thus, given a finite sensitivity in the measuring instrument, only δ's with a large enough weight will be detected. The result, regardless of the sensitivity of the instrument, is a discrete spectrum which will resemble Bragg-like peaks. The 'hidden symmetry' we have referred to is just the periodicity in the higher dimension, which is responsible for the δ's in k-space.

We point out here that the quasi-crystalline string in the Elser (1985) construction corresponds to the special case $w = \frac{1}{2}(\cos\theta + \sin\theta)$, so that:

(1) it consists of an aperiodic sequence of long and short intervals, $\cos\theta$ and $\sin\theta$ respectively (for $|\theta| < \frac{1}{4}\pi$, as in figure 1);

(2) the ratio of the intervals is τ;

(3) the ratio of the number of long intervals to short is also τ;

(4) the average distance between atoms is $\Delta \equiv 1/(\cos\theta + \sin\theta)$; and

(5) τ is not restricted to be the solution to a quadratic equation, as in the $k = 2$ case of Levine and Steinhardt (1984).

Finally, note that, in general, centring R about any other quantity ζ leads to phases for each δ in the spectrum $F(k)$. The effect is not observable in the intensities; neither is the effect of translating $Q(\eta)$. Clearly, if ζ is a (rational) fraction of Δ, then the line will pass through another lattice point, so that translations for R are identical to those for Q.

As it stands, the example given above belongs to the class of structures known as incommensurate crystals or modulated crystals (Janssen *et al* 1983), with the displacement from periodicity being modulated by an incommensurate wavevector. Significantly, these authors have introduced the language of superspace (the D-dimensional one) and studied cases for $D \leq 6$. Others (Weigel *et al* 1984, Veysseyre *et al* 1984) have investigated point groups in superspace. What seems novel here is the appearance of five-fold symmetries, like Penrose tiles in $N = 2$ and icosahedral symmetry in $N = 3$. Thus it is crucially important to examine the subgroups of these

high-dimensional point groups. An appropriate term for those subgroups which are not crystal point groups may be 'quasi-crystalline point groups'. Undoubtedly, many authors are scrutinising this issue.

Symmetry considerations aside, generalisations of the procedure described earlier to higher dimensions in both D and N are immediate. One possible approach is to regard a quasi-crystalline solid as the Radon transform (Radon 1917, Gel'fand *et al* 1966) of RU_0. The special case where ϕ, the angle in the transform, is the same as θ has already been given. Relaxing this constraint would allow the construction of more general quasi-periodic structures. To illustrate, consider $\phi = 0$, which corresponds to a set of δ's located at *periodic* points but with one of two weights, distributed along the string in a quasi-periodic fashion. The connection to the Radon transform, a powerful tool of analysis, should help further studies in the properties of quasi-crystals. Other generalisations might consider imbeddings in a variety of periodic systems in D dimensions besides hypercubic lattices. Another way to obtain higher-dimensional structures is to 'stack' lower-dimensional ones, in a periodic or quasi-periodic way. Of course, it is possible that all these structures are already included in the most general N and D case. Investigations are under way.

A seemingly unlimited variety of quasi-crystals may be generated by choosing other apodisation functions (also known as filter functions or characteristic functions) R. Clearly, the $(D - N)$-dimensional volume of R determines the average number density of the quasi-crystal. So, even for fixed density, it can take on different shapes for $D > N + 1$. For example, the construction in Elser (1985) for the $N = 3$, $D = 6$ structure with icosahedral symmetry employs an R in the form of a triacontahedron. Are there any other interesting quasi-crystalline structures with other R's?

The apodisation functions mentioned here all have sharp edges and are independent of the N $\{\xi\}$ variables. Elser (1985) pointed out that both restrictions can be relaxed. Physically, this corresponds to including random deviations from a 'perfect' quasi-crystal. This may be achieved by 'summing over' an ensemble of apodisation functions which deviate slightly from the 'flat' one, leading to an effective R which is still independent of $\{\xi\}$ but which does not have sharp edges. The consequences are measurable since the intensities of the diffraction peaks are related to (the transform of) R.

All the quasi-crystalline structures described so far are Euclidean, in that they arise from embedding (extrinsically) flat N-dimensional spaces and projecting. It is natural to examine the structures that would arise from curved objects. A singularly interesting one may be the result of projecting points onto a cone. In this a possible way to understand 'point defects' in a quasi-crystal? The formalism presented earlier is well suited for studying these structures in that they are obtained simply by choosing different R's.

Turning to the applications, we first remark that, since a quasi-crystalline structure is the Radon transform of very simple functions, the numerous and diverse applications (Deans 1983) of the transform should be explored for possible uses.

Besides the exploitation of Radon transforms, other applications of this embedding scheme come to mind. One example is the study of routes to chaos, via imbeddings in larger and larger D. For the special $D = 2$ case, we had only two intervals between atoms. In general, there are at least D intervals. So, one picture of a system becoming chaotic is the appearance of many distinct intervals. A model for that would be a string (or higher N-dimensional objects) 'moving' into higher dimensions. Again, one could impose true randomness by considering a distribution of R's at any stage.

Another fertile field is critical phenomena on a quasi-crystalline lattice. In a real space renormalisation group (see, e.g., Burkhardt and van Leeuwen 1982) approach, one typically studies systems on periodic lattices and thins out degrees of freedom so that the new lattice is a larger version of the old one. Generally applicable methods involve discrete changes in length-scale so that the lattice structure is preserved. By abandoning periodicity in favour of quasi-crystalline lattices, one can change length-scales by infinitesimal amounts. This is easily achieved by changing the volume of R, which controls the density of lattice points.

Finally, we provide an example of applying the study of quasi-crystalline structures outside physics. It is well known that a truely unbreakable code would involve total randomness. However, the disadvantage lies in having to carry a rather bulky key for decoding. Quasi-periodic sequences could be of some advantage here. One imagines generating a sequence consisting of, say, 26 symbols with approximately equal frequency via a string in $D = 26$. Then the key could be as small as 25 numbers, representing the hyperspherical angles associated with this line. Note that, since tangents of angles exist in rational fractions of a degree or a radian, quasi-periodicity is achieved without the use of irrational numbers in the key.

It is reasonable to believe that, besides the obvious avenues of investigating electronic and elastic properties in quasi-crystals, many new directions of research will follow these discoveries.

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