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Least path criterion (LPC) for unique indexing in a two-dimensional decagonal quasilattice

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The least path criterion or least path length in the context of redundant basis vector systems is discussed and a mathematical proof is presented of the uniqueness of indices obtained by applying the least path criterion. Though the method has greater generality, this paper concentrates on the two-dimensional decagonal lattice. The order of redundancy is also discussed; this will help eventually to correlate with other redundant but desirable indexing sets.

1. Introduction

Following the discovery of a decagonal phase exhibiting two-dimensional quasiperiodicity as well as one-dimensional periodicity (Bendersky, 1985; Chattopadhyay *et al.*, 1985), an indexing scheme for the diffraction pattern of this phase was proposed by Koopmans *et al.* (1987) [an extension of the model proposed by Ho (1986)]. In these schemes, a pentagonal bipyramid of basis vectors is derived from a distorted icosahedral basis. Mandal & Lele (1991) and Prasad *et al.* (1997) employed a similar distorted icosahedral basis, derived from a six-dimensional orthogonal unit cell, where one basis vector is different in length from the others. Fitz Gerald *et al.* (1988) developed a completely different scheme in which the periodic and quasiperiodic basis vectors were kept separated: a planar pentagonal basis and a sixth vector along the periodic axis. By following the above scheme, Choy *et al.* (1988) simulated the diffraction patterns of the decagonal phase and indexed them accordingly. Aragon *et al.* (1990) also used the six basis vectors but with different scaling constant for indexing. However, the planar quasiperiodic basis vectors lead to non-unique indexing as the basis vectors are not linearly independent. Dalton *et al.* (1992) advocated an indexing scheme based on two appropriately distorted icosahedra rotated 36° about a common axis (*i.e.* periodic axis), giving a large index set with much more redundancy. Earlier, Mukhopadhyay *et al.* (1989) proposed a 'least path criterion' (LPC) in order to obtain unique indexing sets from the pentagonal basis by removing the redundancy of indices. A mathematical proof of the uniqueness was not established for any generalized set of indices by any mathematical proof. The aim of the present paper is to demonstrate a rapid method of identifying the index set of least path length and to present a mathematical proof of the uniqueness of index sets obtained in this way. We restrict ourselves here to the case of the two-dimensional decagonal quasilattice. The problem of non-uniqueness for the distorted icosahedral basis will be dealt with elsewhere.

It is known that the 2D decagonal quasilattice requires a minimum of four basis vectors for its unique indexing, but from the symmetry point of view five vectors are found to be useful. (The situation is analogous to the employment of three base vectors for the 2D hexagonal lattice.) Cervellino *et al.* (1998) used the four basis vectors and proposed a methodology for derivation of proper basis vectors after identifying the unit cell in a minimum higher-dimensional lattice through Patterson analysis. This approach identifies the correct scaling constant and uses minimum basis vectors but, as mentioned earlier, the symmetry relations among the various vectors are not obvious. In addition to the non-uniqueness of indices used for quasicrystal diffraction patterns owing to redundancy of the basis vectors, the self-similarity (scaling symmetry) of Bragg-peak positions also leads to the problem of selecting basis vectors with different length and orientation. As a result, the non-uniqueness hinders the comparison of diffraction results reported by different researchers. However, here we will address the problem of non-uniqueness due to the redundancy problem only. For consistency, the least path criterion (LPC) can be adopted. To the best of the authors' knowledge, no mathematical proof has hitherto been given for the uniqueness of LPC indices. For indexing schemes with redundancy, the useful concept of the order of the redundancy can be defined with respect to least path indices (for which the order of redundancy is zero). The examples of superfluous indices will be drawn from the literature and thus the importance of the least path criterion while indexing becomes apparent. While analysing the diffraction patterns of the decagonal quasilattice, one can find that the diffraction pattern exhibiting tenfold symmetry contains two important directions, which are designated as P and D. The P directions are along the pentagonal base vectors (and their negative counterparts). A D direction lies along the bisector of the angle between two consecutive P directions. The base vectors of a planar pentagonal basis (Fig. 1) satisfy

$$\mathbf{e}_1 + \mathbf{e}_2 + \mathbf{e}_3 + \mathbf{e}_4 + \mathbf{e}_5 = 0 \tag{1}$$

(*i.e.* the $\mathbf{e}_i$ are the position vectors of the vertices of a regular pentagon).

This linear dependence of the base vectors gives rise to a redundancy problem while assigning indices. To avoid the redundancy, one can take four basis vectors instead of five and assign the indices accordingly, by replacing one of the basis vectors:

$$\mathbf{e}_5 = -(\mathbf{e}_1 + \mathbf{e}_2 + \mathbf{e}_3 + \mathbf{e}_4). \tag{2}$$

Thus, for example, (00001) will be represented as $(\bar{1}\bar{1}\bar{1}\bar{1})$. More generally, the vector $(n_1, n_2, n_3, n_4, n_5)$ can be replaced by $(n_1 - n_5, n_2 - n_5, n_3 - n_5, n_4 - n_5)$ if one chooses to dispense with the fifth basis vector. However, the symmetry is obscured by this method and one cannot easily correlate with other symmetry-related vectors. Therefore, five basis vectors are found to be more useful. The redundancy problem may then be settled by following the least path criterion proposed by Mukhopadhyay *et al.* (1989).

2. Statement of the problem

Owing to redundancy in the basis, $(n_1, \ldots, n_5)$ and

$$(n_1 - k, n_2 - k, n_3 - k, n_4 - k, n_5 - k) \tag{3}$$

represent the same vector, whatever the value of k. The problem is how to select a unique index set from all these possibilities.

2.1. Least path criterion or least length criterion

Consider the vector $\mathbf{q}_0 = \sum n_i \bar{\mathbf{e}}_i$ (n_i integers; $i = 1, \ldots, 5$). The quintuplet for this vector can be changed by replacing any one of the integers, *i.e.*

$$\bar{\mathbf{q}}_i = \sum_{j=1}^{5} (n_j - n_i)\bar{\mathbf{e}}_j. \tag{4}$$

This gives six integer sets representing the same vector. Their path lengths are defined as

$$N_0 = \sum_{i=1}^{5} |n_i|, \quad N_j = \sum_{i=1}^{5} |n_i - n_j|, \quad j = 1, \ldots, 5. \tag{5}$$

Now, any N_j will be accepted if it is less than all other N_k ($k = 0, \ldots, 5$):

$$N_j \leq N_k, \quad k = 0, \ldots, 5. \tag{6}$$

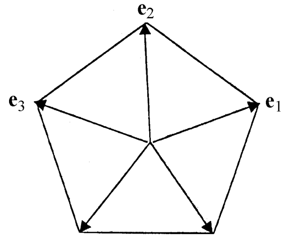

Figure 1
Pentagonal basis vectors.

This is the least path criterion (LPC) proposed by Mukhopadhyay *et al.* (1989). The criterion enables one to choose the indices unambiguously. We now state and prove a lemma that establishes the uniqueness of this choice and provides a fast method of immediately obtaining the index set of minimum path length from any given index set.

3. Lemma

The index set with the smallest path length

$$|n_1 - k| + \ldots + |n_5 - k| \tag{7}$$

is unique and is obtained when k is the index that comes in the middle when $n_1, \ldots, n_5$ are ordered numerically.

3.1. Proof

Let us assume that we have a numerically ordered integer set $n_1, \ldots, n_5$, *i.e.*

$$n_1 \leq n_2 \leq n_3 \leq n_4 \leq n_5. \tag{8}$$

Now select k to be the middle integer, n_3 in the present case. Note that choosing k in this way gives an index set with a zero, two non-negative and two non-positive integers:

$$(n_1 - n_3, n_2 - n_3, n_3 - n_3, n_4 - n_3, n_5 - n_3) = (-\alpha, -\beta, 0, \gamma, \delta), \tag{9}$$

where $\alpha, \beta, \gamma, \delta$ are non-negative integers. The path length is now

$$\ell = \alpha + \beta + \gamma + \delta. \tag{10}$$

Now we have to prove that

$$\ell_k = |k - \alpha| + |k - \beta| + |k| + |k + \gamma| + |k + \delta| > \ell, \tag{11}$$

for any non-zero k.

First, suppose that $k > 0$. Then $\ell_k = |k - \alpha| + |k - \beta| + 3k + \gamma + \delta$.

Case (i): Suppose $k - \alpha \leq 0$ and $k - \beta \leq 0$. Then, $\ell_k = \alpha - k + \beta - k + 3k + \gamma + \delta$ (because $|k - \alpha| = \alpha - k$ and $|k - \beta| = \beta - k$). We get

$$\ell_k \geq (k + \ell) > \ell.$$

Case (ii): Suppose $k - \alpha \leq 0$ but $k - \beta > 0$. Then $|k - \alpha| = \alpha - k$ but $|k - \beta| = k - \beta$ and we get

$$\begin{aligned} \ell_k &= \alpha - k + k - \beta + k + k + \alpha + k + \gamma \\ &= 3k - 2\beta + \ell \\ &= k + 2(k - \beta) + \ell > k + \ell > \ell. \end{aligned}$$

Case (iii): If $k - \alpha > 0$ and $k - \beta > 0$, then

$$\ell_k = 5k - \alpha - \beta + \gamma + \delta = 5k - 2\alpha - 2\beta + \ell.$$

But $\alpha \leq k$, $\beta \leq k$, so that $2(\alpha + \beta) \leq 4k$ and hence $5k - 2(\alpha + \beta) \geq k$. We again find that

$$\ell_k \geq (k + \ell) > \ell.$$

This completes the proof of the lemma for the cases where k is positive. Assuming, alternatively, that k is negative, the proof is obtained from the above by replacing α, β, γ, δ and k by δ, γ, β, α and $-k$, respectively.

Therefore we see that, for any index set $[n_1, \ldots, n_5]$, the equivalent index set with least path length is *unique*, and is given by (3), with k given by the middle term when the indices are numerically ordered. The index set obtained from the least path criterion can be taken as free from redundancy of any order.

3.2. Example

Let us take an example of an index set as $(11\bar{2}\bar{2}5)$ from which we can find $\ell_k = 11$. Therefore, we can easily find out the required integer to be subtracted: The numerically ordered set is $(\bar{2}\bar{2}115)$, $k = 1$ is the middle index. The unique LPC index set is therefore $(00\bar{3}\bar{3}4)$, $\ell = 10$. The original redundant set $(11\bar{2}\bar{2}5)$ can be defined as *redundant of the order* or the order of redundancy (OR) 1 (*i.e.* $|k| = 1$).

Now we can also discuss some examples from the literature. It has been noticed that redundant indices are often used in the literature. In some cases, it may have some advantages. In particular, Singh & Ranganathan (1996) and Ranganathan *et al.* (1994), following the Fitz Gerald choice of basis vectors, have used the redundant set for indexing the zone axis of decagonal quasicrystals. They demonstrated that with this redundant set one satisfies the zone-axis law, which is analogous to that of a crystalline lattice. For example, zone axes L and J (designated by them in the stereogram corresponding to the decagonal lattice), are indexed as $[3\bar{8}\bar{8}3101]$ and $[8\,\overline{21}\,\overline{21}\,8\,2\,6\,2]$, which are obviously redundant of order 3 and 8, respectively. By applying the LPC on the first five indices of the sets, one can easily find the corresponding sets as $[0\,\overline{11}\,\overline{11}\,0\,7\,1]$ and $[0\,\overline{29}\,\overline{29}\,0\,1\,8\,2]$, which are free from redundancy. The highest OR was found to be 11 after examining the table of zone axes worked out by Singh & Ranganathan (1996). It is important to note that the superfluous zone-axis law does not work after applying the LPC. We are currently studying this case and the findings will be reported elsewhere. Another important example we would like to discuss in connection with superfluous indices is that of Ryes-Gasga *et al.* (1992) who (following the five pentagonal and periodic basis vectors) have indexed two important reciprocal spots in electron diffraction patterns, which are $(10\bar{2}\bar{3}\bar{1}0)$ and $(21\bar{3}\bar{4}\bar{1}0)$, where the component along the sixth vector (periodic vector) is zero. By applying the LPC, one can obtain the non-redundant set as $(21\bar{1}\bar{2}00)$ and $(32\bar{2}\bar{3}00)$, respectively. By comparing the indices with those in Table 2, it is clear that these vectors belong to D-type vectors and they are related by τ scaling. Obviously, in this case the superfluous indices have obscured the symmetry relation, as the set was redundant of the order of 1 in both cases. Therefore, one can really see the advantages of LPC and the non-redundant set of indices.

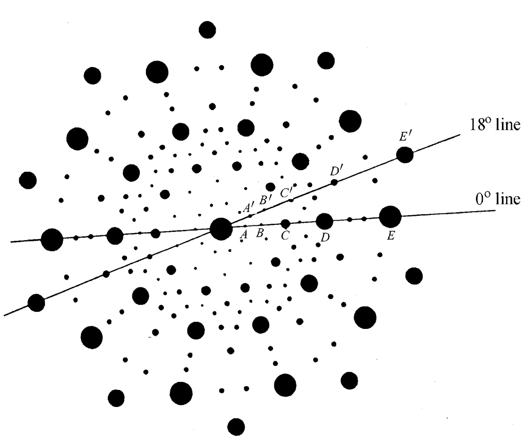

Figure 2
Computed diffraction patterns along a tenfold axis based on the Fourier transform of the orthogonal window function, which has been chosen as a decagon. The 0 and 18° lines corresponding to the P and D vectors are shown. The important vectors (A–E) and (A'–E') are indicated.

4. LPC for *P* and *D* directions

We have computed the diffraction patterns from the Fourier transform of decagonal quasilattices projected from five-dimensional space (Fig. 2). The details of the procedure have already been discussed earlier (Mukhopadhyay *et al.*, 1989). Here we want to show the important P and D types of reciprocal vectors in the computed patterns and their corresponding indices following the LPC.

The components of the five planar base vectors are given by

$$(\mathbf{e}_1 \quad \mathbf{e}_2 \quad \mathbf{e}_3 \quad \mathbf{e}_4 \quad \mathbf{e}_5) = \begin{pmatrix} 1 & -\sigma/2 & -\tau/2 & -\tau/2 & -\sigma/2 \\ 0 & \beta\tau/2 & \beta/2 & -\beta/2 & -\beta\tau/2 \end{pmatrix}, \tag{12}$$

where $\tau = (1 + 5^{1/2})/2$, $\sigma = (1 - 5^{1/2})/2$, $\beta = (3 - \tau)^{1/2}$.

They satisfy the identities

$$\mathbf{e}_1 = \tau(\mathbf{e}_2 + \mathbf{e}_5) = \sigma(\mathbf{e}_3 + \mathbf{e}_4) \tag{13}$$

$$\mathbf{e}_2 - \mathbf{e}_5 = \tau(\mathbf{e}_3 - \mathbf{e}_4) = \sigma(\mathbf{e}_4 + \mathbf{e}_5 - \mathbf{e}_2 - \mathbf{e}_3). \tag{14}$$

4.1. The *P* directions

Define the distance from the centre of the diffraction pattern to the spot labelled (10000) to be unity. According to (13), indices for other spots along this direction are linear combinations of (10000), (01001) and (00110); *i.e.* they are all of the form $(abccb)$. The distance from the centre to the spot labelled $(abccb)$ is

$$(a - b) + \tau(b - c).$$

Applying the least path criterion we get, from $(abccb)$, LPC index sets of the following forms, in which m and n have opposite signs:

$(0mnnm)$ if a is the 'middle index',
$(n0mm0)$ if b is the 'middle index',
$(mn00n)$ if c is the 'middle index'.

LPC indices for the other P directions are of course related to these by cyclic permutation.

Table 1
Indices of some important '*P*'-type vectors marked *A*–*E* along the 0° line in Fig. 2.

	...	...		...
	τ^{-3}	$(\bar{3}0\bar{2}\bar{2}0)$	$\rightarrow$	$(\bar{1}200\bar{2})$
	τ^{-2}	(20110)	$\rightarrow$	$(1\bar{1}00\bar{1})$
	τ^{-1}	$(\bar{1}0\bar{1}\bar{1}0)$	$\rightarrow$	(01001)
A	1	(10000)		
B	τ	$(00\bar{1}\bar{1}0)$		
C	τ^2	$(10\bar{1}\bar{1}0)$		
D	τ^3	$(10\bar{2}\bar{2}0)$		
E	τ^4	$(20\bar{3}\bar{3}0)$		
	...	...		...

Table 2
The indices of important '*D*'-type vectors, marked A'–E' along the 18° line in Fig. 2.

	...	...
	$\beta\tau^{-2}$	$(02\bar{1}1\bar{2})$
	$\beta\tau^{-1}$	$(0\bar{1}1\bar{1}1)$
A'	β	$(0100\bar{1})$
B'	$\beta\tau$	$(001\bar{1}0)$
C'	$\beta\tau^2$	$(011\bar{1}\bar{1})$
D'	$\beta\tau^3$	$(012\bar{2}\bar{1})$
E'	$\beta\tau^4$	$(023\bar{3}\bar{2})$
	...	...

Table 1 lists the LPC indices for the τ-inflation sequence along the *P* direction e_1, obtained from linear combinations of (10000) and $(00\bar{1}\bar{1}0)$ by a 'Fibonacci rule': each index set is a sum of the two sets preceding it in the list. The symbol $\rightarrow$ indicates the application of the LPC rule.

4.2. The *D* directions

Equation (14) implies that any spot along the direction perpendicular to e_1 can be indexed by a linear combination of $(0100\bar{1})$ and $(001\bar{1}0)$. These index sets thus have the characteristic pattern $(0ab\bar{b}\bar{a})$ (and index sets for the other *D* directions are related to these by cyclic permutation). Since the 'middle index' is 0, these index sets are already in the unique LPC form. The distance from the centre of the diffraction pattern to the spot labelled $(0ab\bar{b}\bar{a})$ is $\beta(b + a\tau)$. It can be seen that $(\bar{1}01\bar{1}1)$ is perpendicular to e_2. It labels the spot at a distance $\beta(\tau - 1) = \tau^{-1}\beta$. The τ-inflation sequence along the *D* direction perpendicular to e_1, obtained by the 'Fibonacci rule', is indicated in Table 2.

5. Experimental examples

By following the LPC, we have indexed important reflections observed by us in our experimental powder X-ray diffraction pattern obtained from an $Al_{65}Cu_{20}Co_{15}$ decagonal quasicrystal using Cu $K\alpha$ radiation (Fig. 3). These are also displayed in Table 3 along with their interplanar spacings and intensities. Some reflections, which are already marked in computed electron diffraction patterns (Fig. 2), are indicated in the table. It should be mentioned that in the present case the quasilattice (a_R) and the periodic lattice (c) constants, following the methodology proposed earlier by us (Mukhopadhyay *et al.*, 1989), are found to be 0.398 and 0.815 nm, respectively. These parameters are used for the present indexing scheme. It should be mentioned that the quasilattice constant determined by us is τ^3 times more than that of Tsai *et al.* (1989) and it fits reasonably well with the physical real-space parameter in 3D space. It is of interest to point out that Cervellino *et al.* (1998) have employed minimum 5D space to generate the quasilattice structure and indexed accordingly. However, as mentioned earlier, in the 5D space approach the symmetry relations among the various planes and directions are not at all obvious. In the present case, 6D space has been used to generate the structures and by the LPC the redundancy problem has also been eliminated in order to bring out the symmetry relations among planes and directions. It may be emphasized that, after removing the redundancy problem in the 6D space approach, both the 5D and 6D space approaches can be correlated and the equivalent indices can easily be obtained.

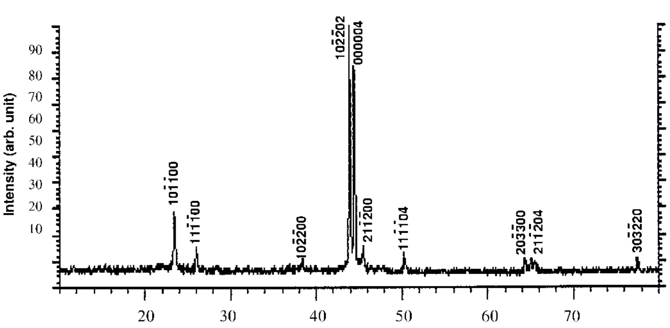

Figure 3
X-ray powder diffraction pattern (Cu $K\alpha$) obtained from the Al–Cu–Co decagonal phase, synthesized by slow cooling from the melt. All the reflections are indexed with a non-redundant set of indices after applying LPC.

6. A note on the hexagonal case

The case of *three* base vectors satisfying

$$\mathbf{e}_1 + \mathbf{e}_2 + \mathbf{e}_3 = 0 \tag{15}$$

is also not without interest, since it corresponds to the generally adopted basis for the hexagonal lattice (Frank, 1965). The standard prescription for eliminating the redundancy is to choose index sets that satisfy

$$n_1 + n_2 + n_3 = 0. \tag{16}$$

This gives fractional indices and so a scaling factor is introduced; for example, $[023] \rightarrow [-5/3, 1/3, 4/3] \rightarrow [\bar{5}14]$. The least path criterion gives $[023] \rightarrow [\bar{2}01]$. Clearly, the least path criterion is simpler to apply and provides index sets with smaller integers, but is unlikely to be adopted as an indexing scheme for hexagonal lattices – the prescription (16) is far too well established.

7. Concluding remarks

The lemma provides a simple and straightforward method of dealing with the redundancy problem in the indexing of

Table 3
Important reflections observed in the powder X-ray diffraction pattern obtained from Al–Cu–Co decagonal quasicrystalline phases; the indices are free from redundancy after applying LPC; the sixth indices are along the periodic direction of the decagonal phase (a_R = 0.398, c = 0.815 nm).

Sl No.	Indices ($n_1, \ldots, n_6$)	d (nm)	Intensity (I/I_{max})	Fig. 2 (related spots)
1	$10\bar{1}\bar{1}00$	0.3802	35	C
2	$11\bar{1}\bar{1}00$	0.3234	20	C'
3	$10\bar{2}\bar{2}00$	0.2349	12	D
4	$10\bar{2}\bar{2}02$	0.2062	100	
5	000004	0.2038	70	
6	$21\bar{1}\bar{2}00$	0.1998	23	D'
7	$11\bar{1}\bar{1}04$	0.1725	18	
8	$20\bar{3}\bar{3}00$	0.1452	10	E
9	$21\bar{1}\bar{2}04$	0.1427	12	
10	$30\bar{3}\bar{2}20$	0.1235	14	E'

diffraction patterns of decagonal quasicrystals. The proof is readily generalizable to the case of an arbitrary *odd* number of basis vectors satisfying a linear dependence relation analogous to (1). For an *even* number of base vectors, the uniqueness of the index set of least path length is lost. In these cases, there are two middle values when the indices are numerically ordered, say n_r and n_{r+1}, and the index sets with minimal path length are given by the values of k satisfying $n_r \leq k \leq n_{r+1}$. However, the case of five base vectors is of course the one of practical importance, since (1) corresponds to the Fitz Gerald choice of basis for decagonal quasicrystals.

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