# **Derivation of the proper basis of quasicrystals**

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A method based on the physical space Patterson (pair correlation) function is derived which allows the determination of the correct *n*-*D* Bravais lattice of quasicrystals. [Hereafter, we will abbreviate *n*-*D* for *n* dimensional. We will also call *perpendicular space* the (n-3)-*D* orthogonal space that is added to physical 3D space to form the *n*-*D embedding space*.] The optimum unit cell can be chosen and therefore the proper indexing of the diffraction pattern. The size of the integrated maxima of the Patterson function depends on their multiplicity and on their perpendicular space component. Lifting the positions of these maxima into *n*-*D* space allows the set of "quasilattice" vectors to be distinguished from the set of decoration vectors. This procedure leads to a unique *n*-*D* lattice. Taking advantage of scaling symmetries, the best choice of the *n*-*D* unit cell can be found. A detailed analysis of the decoration vectors reveals all possible positions of the hyperatoms therein. This powerful technique is illustrated on simulated data of a decorated Fibonacci chain and on experimental data of decagonal Al<sub>70.5</sub>Mn<sub>16.5</sub>Pd<sub>13</sub> quasicrystals. [S0163-1829(98)02918-X]

#### I. INTRODUCTION

Within the past 13 years structure analysis of quasicrystals has been improved drastically and many average structures have been solved.<sup>1</sup> However, up to now none of the existing structure solutions achieved the standards of conventional crystal structure determination. The reason is that quasicrystals are not periodic and they need to be described in an *n*-*D* space (n>3) to obtain periodicity. [In the case of quasicrystals the noncrystallographic symmetry group is not compatible with lattice periodicity in 3D space. However, it is possible to recover lattice periodicity in a *n*-*D* space (n>3) of which physical 3D space must be an invariant subspace.]

This has severe effects on the following steps of structure determination. The first step is the indexing of all Bragg peaks. For conventional crystals this step is straightforward and unambiguous while for quasicrystals it is not. The second is the reconstruction of the phases of the structure amplitudes and here the difficulty is comparable. In the last step, the three-dimensional (3D) structure and the atomic coordinates have to be found. For ordinary crystals this is done by a simple Fourier transform while quasicrystals require a complex procedure to derive the atomic surfaces in n-Dspace. [In the n-D description the atoms become hyperatoms. These are thought as the convolution of atomic physical space electron densities with a density function defined on (n-3)-D surfaces (atomic surfaces) parallel to perpendicular space and invariant under the space-group symmetry operations.

Most research has been focused on the second part. Several powerful structure solution techniques as well as refinement programs have been developed.<sup>2–6</sup> All promising techniques use the *n*-*D* embedding approach<sup>7,8</sup> to recover the phases. Up to now no unique method for choosing the *n*-*D* unit cell could be derived. The aim of this contribution is to present a method that allows the *n*-*D* Bravais lattice to be determined unambiguously and an optimum basis to be chosen.

Several methods for indexing the diffraction patterns of quasicrystals have been developed.<sup>9-11</sup> Most of them simply try to index strong diffraction spots with the lowest possible indexes. However, they all suffer from the problem of selfsimilarity (scaling symmetry) of the Bragg peak positions which is a common feature for diffraction patterns of quasicrystals. This is especially a problem in the case of superstructures<sup>12</sup> where in reciprocal space the same  $\mathbb{Z}$  module describes the Bragg peak positions. In fact, in contrast to 3D crystallography, the diffraction patterns of sub- and superstructures do not differ in the position of peaks but only in their intensity distribution. This constitutes a new problem that is not fully solvable based on a reciprocal space analysis only. Lançon et al., 13 have shown that misindexing the diffraction pattern, i.e., choosing a wrong unit cell, leads to bad partitioned atomic surfaces and a severely averaged structure in real space. To stress the problem it has to be recalled that for quasiperiodic structures Bragg peaks can be considered as projections onto reciprocal physical space of an n-D reciprocal lattice. Although the intensity distribution allows only a finite subset to be observable, Bragg peaks are a dense set in reciprocal physical space. For this reason as well as for the self-similarity, in physical reciprocal space scaling constitutes a symmetry group whose elements are the scaling factors  $\sigma^n$ ,  $n \in \mathbb{Z}$ , where, e.g., in decagonal or icosahedral quasicrystals  $\sigma$  is a given power of the golden mean  $\tau = (1$  $(+\sqrt{5})/2$ . In direct physical space the existance of smallest interatomic distances (i.e., of a smallest physical scale, the atomic scale) breaks this symmetry. The information on both the smallest interatomic distance as well as the smallest "quasilattice" vectors (unit tiles) will be included in the electron pair correlation function of the structure. This function is just the Fourier transform of the scattered x-ray intensity and crystallographers usually call it the Patterson function.

It will be shown how the different multiplicities of interatomic decoration vectors and quasilattice vectors allow the latter ones to be separated and the n-D lattice to be assigned uniquely. Consequently the Bravais lattice can be determined

11 223

within the chosen embedding (i.e., within the invariant subspace structure connected to the point group). Based on scaling symmetry one can select the optimum setting of the unit cell, and therefrom the proper indexing in reciprocal space can be determined. Finally, based on the n-D space group, all possible centers of the hyperatoms can be determined.<sup>5</sup> The technique will be demonstrated on a simulated Fibonacci chain with superstructure (as for superstructures indexing is most effectively troublesome<sup>12</sup>) and on real data of a decagonal quasicrystal.

## **II. BASIC CONCEPTS**

Quasicrystals can be described with few parameters using the *n*-*D* embedding method.<sup>7,8</sup> Within this approach the *d*-*D* (d < n) reciprocal space is considered to be a projection of an *n*-*D* lattice. Given a primitive lattice in *n*-*D* reciprocal space the choice of the unit cell and of the spanning vectors  $\underline{\mathbf{d}}_1^*, \ldots, \underline{\mathbf{d}}_n^*$  is not unique. Different choices can result also in different lattice parameters  $\underline{\mathbf{a}}_1^*, \ldots, \underline{\mathbf{a}}_n^*$  in physical reciprocal space. This is the reason for the well-known scaling symmetry concerning diffraction patterns of quasicrystals. While the projected reciprocal lattice is a dense set in physical space, a smallest distance and a "finest" tiling exist in direct space. So the scaling symmetry is only a semigroup based on a set of smallest unit tiles.

The Patterson function is the inverse Fourier transform of the scattered intensity. Provided that all observable Bragg peaks are included in the calculation, this function is totally *independent from indexing*. In fact, reciprocal space coordinates appear only as integration variables in the Fourier transform.

The Patterson function shows a peak for any interatomic vector in the structure weighted by its multiplicity and the product of the corresponding atomic cross sections. In ordinary crystals a family of equal Patterson peaks, which have the highest multiplicity and consequently are an infinite set of global maxima, is found at the origin and at any other lattice node. This allows the lattice to be easily identified.

In quasiperiodic structures, however, the situation is different. In the n-D description the Patterson function can be obtained as the convolution of all the hyperatoms. So it is represented by a density function defined on the convoluted atomic surfaces (hereafter hypervectors). Atomic surfaces are parallel to perpendicular space and consequently this holds for hypervectors as well. The Patterson density on a hypervector has its maximum in the center and shows a linear decay with increasing distance from the center. Physical space intersects some of the hypervectors. Let us take a hypervector. On any selected embedding basis allowed by the space group, the n-D coordinates of its center will be found to be  $\hat{\underline{\mathbf{r}}}_{C} = \hat{\underline{\mathbf{r}}}_{C}^{||} \oplus \hat{\underline{\mathbf{r}}}_{C}^{\perp}$ . Suppose it intersects physical space, the intersection point will be  $\hat{\mathbf{r}}_I = \hat{\mathbf{r}}_I^{||} \oplus \mathbf{0}^{\perp}$ . In physical space we will find a Patterson peak centered at  $\hat{\mathbf{\underline{r}}}_{C}^{||}$ , whose height will be given by the Patterson density on the hypervector evaluated at the distance vector  $-\hat{\mathbf{f}}_{C}^{\perp}$  from the center. All Patterson peaks can be thought of in the same way. The vectors  $-\hat{\mathbf{f}}_{\underline{C}}^{\perp}$  will always be different, due to the irrational slope of the crystal basis with respect to physical space. In particular,

consider the family of equal hypervectors (*lattice hypervectors*) centered at the origin and in every lattice node. Here we have the highest Patterson densities with global maxima in the center. Hence the 3D physical Patterson function will have only one global maximum (at the origin) while the "translation equivalent" peaks show different decays depending on the center-to-cutpoint  $-\hat{\mathbf{r}}_{C}^{\perp}$  distance. The same decay holds for peaks corresponding to the remaining (*decoration*) hypervectors.

On the other side, if we consider a large number of peaks we will have a sampling of the Patterson density on the hypervectors on a large number of points  $-\hat{\mathbf{r}}_{C}^{\perp}$ . Consequently, plotting the integrated maxima of the 3D Patterson function versus their perpendicular components  $\| - \hat{\mathbf{r}}_{C}^{\perp} \|$  will result in a set of smooth branches describing the radial Patterson density on the different hypervectors. They will be separated by a gap according to the respective multiplicities. The topmost branch (the branch containing the global Patterson maximum) is assigned to the lattice hypervectors of the n-D Patterson map. So we clearly identify a set of lattice nodes. Based on this subset of Patterson maxima the embedding matrix as well as the n-D metrics is derived. We call the embedding matrix the matrix transforming the vector components from the *n*-*D* lattice basis  $\underline{\mathbf{d}}_1, \ldots, \underline{\mathbf{d}}_n$  (usually called D basis) to another basis (V basis), which is obtained as an extension to the *n*-*D* space of a canonical (Cartesian) frame in physical space. Depending on the geometry the most convenient setting of the n-D unit cell can be chosen between the scaling equivalent ones. The optimum choice is the one with the perpendicular space projection of the n-D unit cell enveloping the origin peak. Hence, all Bragg reflections can be (re)indexed and n-D Patterson deconvolution techniques may be applied to locate all positions of the hyperatoms in the *n*-*D* unit cell.

## **III. DECORATED ONE-DIMENSIONAL TILINGS**

This section deals with a simple 1D quasiperiodic structure derived by decorating a Fibonacci chain. The decoration substructure is obtained by applying once the substitution rule (Ref. 13)  $L \rightarrow LLSL$  and  $S \rightarrow SLS$  on the Fibonacci chain and rescaling the lattice parameter by a factor  $\tau + 2 = \tau^2 + 1$ .

The Fibonacci chain can be embedded in 2D space. The chain is an irrational cut of a 2D square lattice with physical space. The hyperatoms have the shape of segments perpendicular to the cut. The cut axis and the perpendicular direction form a Cartesian frame. With respect to the associated canonical basis the lattice is spanned by the vectors

$$\underline{\mathbf{d}}_1 = \frac{a}{1+c^2+c^2\tau} \begin{pmatrix} 1\\ -c\tau \end{pmatrix}, \quad \underline{\mathbf{d}}_2 = \frac{a}{c^2+1+\tau} \begin{pmatrix} \tau\\ c \end{pmatrix}, \quad (1)$$

where c is an arbitrary factor for perpendicular space (hereafter c=1) and  $a/(\sqrt{2+\tau})$  is the lattice parameter. This setting defines the embedding matrix

$$T = \frac{1}{2+\tau} \begin{pmatrix} 1 & \tau \\ -\tau & 1 \end{pmatrix}, \tag{2}$$



FIG. 1. (a) The embedding 2D lattice (full lines) and the decoration sublattice (dotted lines) are shown. The horizontal and vertical axes are the parallel and perpendicular spaces, respectively. The hyperatoms are shown as vertical bars. (b) The corresponding Patterson function is shown. The contour lines show the decay of the hypervectors.

i.e., the vector components transformation from the  $\{\underline{\mathbf{d}}_1, \underline{\mathbf{d}}_2\}$  basis to the canonical one. For the decorated chain the 2D lattice contains five hyperatoms per unit cell which are related to a sublattice [Figs. 1(a) and 1(b)] which can be obtained from the main lattice by the transformation matrix

$$M^{-1} = \frac{1}{5} \begin{pmatrix} 3 & -1 \\ -1 & 2 \end{pmatrix}.$$
 (3)

The simulation of a diffraction experiment yields a diffraction pattern comparable with the one obtained from a not decorated Fibonacci chain (Fig. 2). Considering only the geometry of the diffraction pattern, i.e., the positions of the Bragg peaks, the two systems are not distinguishable. However, the scattered intensity distribution is quite different. The decorated chain shows a large number of weak and medium Bragg reflections.

For our simulation, we assumed realistic experimental conditions as given for a standard four-circle diffractometer (dynamical range of the detector 10<sup>6</sup>) giving 1311 observable unique reflections. For comparison with previous results,<sup>13</sup> a data set with an intensity cutoff at 6% of the strongest peak (dynamical range less than 10<sup>2</sup>, with only 12 observable reflections) was used as well. The results are completely equivalent, in spite of the severe truncation effects which do not affect the effectiveness of this technique.

In a first step the 1D Patterson function  $P(r^{||})$  was calculated (Fig. 3). The superscript || identifies the (single) physical space coordinate. The calculation was performed without

indexing the diffraction pattern. The scattered intensity  $I(q^{||})$  was sampled on a convenient grid of  $q^{||}$  (the transferred momentum wave vector) values. The inverse Fourier transform was calculated by numerical integration on this grid.



FIG. 2. Simulated diffraction pattern of the decorated Fibonacci chain assuming experimental conditions comparable with the fourcircle diffractometer at the beamline D3 HASYLAB (Ref. 18). The units for the horizontal axis are  $Å^{-1}$ . The cutoff threshold for the second simulation (Ref. 13) is shown.



FIG. 3. A section of the Patterson map resulting from our simulation of the decorated Fibonacci chain. In this simulation we suppose a dynamical range of  $10^6$  for intensity measures resulting in 1311 observable reflections. On top of the picture the upper line of segments marks the points of the lattice Z module, the lower line the points of the decoration Z module.

After this, all maxima of  $P(r^{||})$  were located and integrated. Given the quasiperiodicity of the structure, all the Patterson peaks are bound to stay on one or more conveniently scaled Fibonacci chains, depending on the decoration. In our case, as the decoration could be referred to a sublattice in the 2D embedding space, only one chain was sufficient. This chain can be described with a  $\mathbb{Z}$  module of rank two. If we denote with  $\underline{\mathbf{a}}_k^{||}$  ( $\underline{\mathbf{a}}_k^{\perp}$ ) the physical (perpendicular) space projections of  $\mathbf{d}_k$  the elements of this  $\mathbb{Z}$ module are  $\underline{\mathbf{u}}^{||} = a \sigma^{||} (n_1 \underline{\mathbf{a}}_1^{||} + n_2 \underline{\mathbf{a}}_2^{||})$ , with  $n_1, n_2 \in \mathbb{Z}$  and the *complementary*  $\mathbb{Z}$  module will be  $\underline{\mathbf{u}}^{\perp} = a \sigma^{\perp} (n_1 \underline{\mathbf{a}}_1^{\perp} + n_2 \underline{\mathbf{a}}_2^{\perp}).$  $[A \mathbb{Z} module is dense in space so it is necessary to bound the$ norm of the complementary  $\mathbb{Z}$  module, restricting to  $(n_1, n_2)$ couples such that  $\|\mathbf{u}^{\perp}\| < A$  for a conveniently large A.]  $\sigma^{\parallel}$ and  $\sigma^{\perp}$  are a convenient couple of scaling factors. The first  $\mathbb{Z}$ module is easily graphically matched with the existing Patterson map peaks (by adjusting the value of the product  $a\sigma^{||}$ , at this point a is supposed unknown). Taking advantage of the self-similarity of  $\mathbb{Z}$  modules a small number of trials is sufficient. In the case the decoration was not related to a sublattice, so matching all peaks with the same  $\mathbb{Z}$  module would not be possible. In this case, however, it is always possible to match at least a subset of the Patterson peaks, as the subset corresponding to the lattice hypervectors. The greater peak heights often allow this subset to be detected visually.

The next step is *lifting* all peak positions  $\underline{\mathbf{u}}^{\parallel}$  into the first unit cell, defined as  $C \equiv \{x_1\underline{\mathbf{d}}_1 + x_2\underline{\mathbf{d}}_2 | 0 \le x_1, x_2 \le 1\}$  in 2D embedding space. This is easier and numerically convenient for peaks which could be formerly matched by a  $\mathbb{Z}$  module as they can be immediately related to their respective hypervector center via 2D construction. Anyhow, it is always possible



FIG. 4. Plot of the integrated Patterson maxima versus the perpendicular space component of the centering  $\mathbb{Z}$  module. The topmost branch (asterisks) is easily identified. A complete separation from the others (diamonds) can be made by the analysis of coincidence between the lattice and decoration  $\mathbb{Z}$  modules or exploiting the results of the lifting.

to perform this task.<sup>5</sup> At the end we have a unit cell decorated with one or more hypervectors. In our case the decoration forms a sublattice so we could match all Patterson peaks with one  $\mathbb{Z}$  module. This results in lifting in the sublattice unit cell containing only one hypervector in the origin. If this sublattice was assumed as the proper lattice, the structure would be roughly averaged.

As a further step now we can plot the integrated Patterson peak values versus the respective perpendicular components. This plot (Fig. 4) shows several branches which can be clearly distinguished from each other. Based on concepts derived in Sec. II, all (and only) the maxima belonging to the topmost branch are to be considered quasilattice peaks (or belonging to lattice hypervectors).

The process of centering the peaks with the  $\mathbb{Z}$  module was repeated but now only on these quasilattice peaks, acting on the scaling factors couples  $(\sigma^{\parallel}, \sigma^{\perp})$ . Consequently, the proper embedding lattice was found. Due to the multiplicities of the "translation vectors" in physical space, one of the strongest Patterson peaks can be assigned to the edge length of an arbitrarily shaped unit tile. This will lead to the proper lattice parameter a. [These couples are not completely arbitrary. In fact, a couple  $(\sigma^{||}, \sigma^{\perp})$  defines a new lattice and the transformation  $M^{-1}$  between the old and new lattices. Selfsimilarity in physical space requires the new lattice to cover completely the old one or vice versa. Further restrictions in higher dimensional cases come from the necessity to preserve the invariant subspace structure induced by the space group. Hence the allowed couples form a discrete (countable) set and can be easily calculated. Then it is easy to identify the scaling couple such that only quasilattice peaks are matched.] The lifting of all peaks after setting this new



FIG. 5. Plot of the lifted Patterson maxima. The maxima lifted onto the hyperatom in the origin (squares) are in a one-to-one correspondence with the topmost branch of (Fig. 4). The others (diamonds) all belong only to the lower branches. This enforces the distinction between lattice translation vectors and decoration vectors.

basis shows the unit cell with the proper hypervector at the origin and four different decoration hypervectors (Fig. 5) centered at the points (0,0),  $\frac{1}{5}(2,1)$ ,  $\frac{1}{5}(1,3)$ ,  $\frac{1}{5}(3,4)$ ,  $\frac{1}{5}(4,2)$ , which can be easily assigned to the atomic positions [Fig. 1(a)]. For comparison the 2D Patterson map is also reported [Fig. 1(b)].

#### **IV. DECAGONAL QUASICRYSTALS**

This technique has been already tested on real diffraction data obtained from several different decagonal crystals, always with equivalent results. We have chosen decagonal Al<sub>70.5</sub>Mn<sub>16.5</sub>Pd<sub>13</sub> among these, and in this section we will present the concrete application of our technique on diffraction data of this alloy. The data have been collected at the synchrotron source HASYLAB. From the analysis of the diffraction pattern as well as the Bijvoet differences,<sup>14</sup> the crystal has been attributed the space group  $P10_5/mmc$ , and the Bragg reflexions have been initially indexed following the usual methods within a 5D embedding.<sup>15</sup> From these data, various large scale Patterson map sections (~200 Å×200 Å) orthogonal to the decagonal axis have been calculated.

For the sake of simplicity, the *z* physical coordinate (the decagonal axis) will be ignored in the following, as we focus on perpendicular planar sections. So our superspace is now reduced to four dimensions, while the physical space is 2D and the two added dimensions form the perpendicular space. The hyperlattice basis (or *D* basis) vectors  $\mathbf{d}_k$ ,  $k=1, \ldots 4$ , have nonzero projections in both physical and perpendicular



FIG. 6. A detail of the (xy0) Patterson map section of Al-Mn-Pd. On this scale the unit length corresponds to 3.851 Å. Levels of 6 and 37% of the origin peak height are contoured. Asterisks mark the points of the Z module (*I*). It can be noticed that it is not possible to center every peak with a Z module point. One of these peaks is marked with an arrow.

space. They are the direct sum of these projections,  $\underline{\mathbf{d}}_k = \underline{\mathbf{a}}_k^{||} \oplus \underline{\mathbf{a}}_k^{\perp}$ . With the physical space projections  $\underline{\mathbf{a}}_k^{||}$  we can build a  $\mathbb{Z}$  module  $\underline{\mathbf{u}}^{||} = \sum_{k=1}^4 n_k \underline{\mathbf{a}}_k^{||}$ ,  $n_k \in \mathbb{Z}$ . It is useful (see Sec. III) to select only the points  $\underline{\mathbf{u}}^{||}$  whose associated perpendicular space norm  $\|\underline{\mathbf{u}}^{\perp}\|$ ,  $\underline{\mathbf{u}}^{\perp} = \sum_{k=1}^4 n_k \underline{\mathbf{a}}_k^{\perp}$ , is less than the maximum perpendicular space extension of the hyperatoms. The restricted  $\mathbb{Z}$  module forms a pentagonal tiling in physical space and covers every quasilattice node without being dense.

In analogy with most decagonal quasicrystals the hyperatom centers are bound to stay on the positions  $\underline{\mathbf{c}}_q = q(\frac{1}{5}, \frac{1}{5}, \frac{1}{5}, \frac{1}{5})$ , for q = 0, ... 4, in the 4D unit cell. Adding the five  $\underline{\mathbf{c}}_q$  translations to each lattice point we obtain a sublattice<sup>16</sup> with a five times smaller cell volume. It is obtained applying to the former lattice the linear transformation

$$M^{-1} = \frac{1}{5} \begin{pmatrix} 2 & -1 & -4 & -2 \\ 2 & 4 & 1 & -2 \\ 2 & 4 & 6 & 3 \\ -3 & -1 & 1 & 3 \end{pmatrix}.$$
 (4)

From this sublattice we can also obtain a new restricted Z module, which results in it being identical to the former except for a 18° rotation and a scaling factor of  $(1 + \tau)/(\sqrt{2 + \tau})$  (see Sec. III). This can also be described in terms of a subtiling.<sup>15</sup> These two systems will be specified with (*I*) and (*II*), respectively.

In the first part of our research we have been superimposing the two  $\mathbb{Z}$  modules on the Patterson map sections. We observed three features. First (Fig. 6), the points of the  $\mathbb{Z}$ module (*I*) center many of the local maxima of the map but *not all of them*. This holds true also when applying any scal-



FIG. 7. Same as Fig. 6, but now asterisks mark the points of the  $\mathbb{Z}$  module (*II*). In this case, in contrast, every peak is centered by the  $\mathbb{Z}$  module.

ing factor  $\tau^n$ . In particular, the peaks corresponding to the nearest-neighbor Al-TM distance are not centered. This tells us that the lattice (*I*) is not the decoration sublattice. In this case every Patterson peak would be centered.

The natural progression was to try the  $\mathbb{Z}$  module (*II*). In this case *every Patterson peak is centered* (Fig. 7). This holds true in any of the constant-*z* layers. This confirms that also for this crystal the decoration constitutes a sublattice and that this sublattice is just lattice (*II*).

The third curious feature we observed (Fig. 8) was that, at



FIG. 8. A larger detail of the (xy0) Patterson map section. Dark gray areas indicate values over 25% of the origin peak height. The crosses mark the points of the Z module (*III*). The correspondence is almost one-to-one.



FIG. 9. The integrated peak values (vertical axis, % of the original peak height) of the z=0 Patterson map section are plotted as a function of their perpendicular space distance from physical space (horizontal axis, arbitrary scale). The subdivision in three branches is evident.

visual inspection, every Patterson peak higher than 25% of the original peak could be indexed with a third  $\mathbb{Z}$  module (*III*) obtained from (*I*) by multiplying the vectors  $\underline{\mathbf{a}}_{k}^{\parallel}$  by  $\tau^{3}$ and  $\underline{\mathbf{a}}_{k}^{\perp}$  by  $-\tau^{-3}$ . This is equivalent to applying to the 4D coordinates (*D* basis) the  $(S_{10})^{3}$  transformation, where  $S_{10}$  is the matrix<sup>17</sup> representing the operation of scaling by  $\tau$  in physical space (and by  $-1/\tau$  in perpendicular space). In the following we will refer to this operation simply as  $\tau$  scaling.

To substantiate this visual impression, we decided to plot the integrated peak values versus the relevant perpendicular space norm  $\|\mathbf{u}^{C}\|$ . This was done for every peak using the  $\mathbb{Z}$ module (II) and then transforming back into the setting (I). This resulted in a neatly three-branched plot (Fig. 9). The peaks belonging to the topmost branch were those and only those which were centered by the  $\mathbb{Z}$  module (III). The branches can be interpreted as the radial decay of the Patterson function on each different hypervector for increasing radius. The shape of the curves is smoothed by series truncation effects. As the Patterson function for lattice hypervectors must be higher than for decoration hypervectors, lattice (III) is assigned to the actual lattice. It is connected to lattice (I) by  $\tau^3$  scaling. Hence the two descriptions are not equivalent. If we scale the actual lattice by some powers of  $\tau^4$  ( $\tau^2$  for the Patterson density and centrosymmetric structures<sup>17</sup>), we obtain a completely equivalent de-



FIG. 10. (a) Lifted plot in the 4D unit cell (*I*). The section plane is  $(x_100x_40)$  in the usual 5D  $x_1, x_2, \ldots x_5$  notation for the coordinates in the canonical basis, where  $x_1 = x$ ,  $x_2 = y$ ,  $x_3 = z$  (here ignored) span physical space and  $x_4$ ,  $x_5$  span the perpendicular space. The main body diagonal of the cell is also plotted. The lifted peaks form five hypervectors centered on the fifths of the diagonal. Squares mark peaks belonging to the topmost branch of (Fig. 9). As expected they are all on the origin hypervector. Crosses and diamonds mark the lowest and the middle branch peaks, respectively. Each branch corresponds to a centrosymmetric pair of decoration hypervectors. (b) Same as in (a) but now the lifting is performed in system (*III*). The remarkable difference with the former plot is the interchange between the decoration hypervector pairs.



FIG. 11. Same as in Figs. 10(a) and 10(b) but now the lifting is performed in system (*IV*). This system is equivalent to system (*III*), just the general description is simplified here because the hypervectors do not exceed very much the cell boundaries (and as the hyper*atoms* are roughly half this size, they will not exceed it at all).

scription. Scaling by  $\tau^3$  implies a permutation of the hyperatoms on the  $\underline{\mathbf{c}}_q$  positions. This can influence the partitioning of the atomic surfaces.

Finally, to enforce our conclusions, we show the results of lifting the Patterson peaks into the hyperunit cell [Figs. 10(a) and 10(b)]. The lifting was performed in the (*II*) system using then the appropriate *D*-basis transformation to pass to the (*I*) or (*III*) system. Peaks belonging to different branches could be easily marked and followed in the lifting process. It results that (a) the peaks belonging to the topmost branch are always lifted into the hypervector at the origin of the cell, enforcing the conclusion that they belong to the lattice translation set; (b) the remaining two branches are brought, respectively, into the  $\mathbf{c}_1$ ,  $\mathbf{c}_4$  and into the  $\mathbf{c}_2$ ,  $\mathbf{c}_3$  positions on the main body diagonal. This correspondence is inverted in systems (*I*) and (*III*). Lifting in system (*II*) trivially brings every peak into the origin.

A fourth system was introduced (Fig. 11) for practical purposes. System (*IV*) is scaled by  $\tau^{-4}$  with respect to system (*III*). It is completely equivalent and has the advantage that according to the shape of the (*IV*) cell, the hyperatoms lay mostly inside. Therefore the necessity of recomposing parts of hyperatoms coming from different cells can be minimized. Consequently, the unit cell of system (*IV*) is allowed and can be considered to be the optimum cell setting. Based on this unit cell the diffraction pattern could be reindexed.

### V. DISCUSSION

It was our aim to show how the ambiguities in indexing quasicrystal diffraction patterns, which arise from the selfsimilarity between  $\tau$  scaled projected lattices (tilings), as well as with the projected decoration sublattice (subtiling), can be overcome by a careful inspection of the intensity distribution of the Patterson peaks. The method is based on the property of the Patterson function of being constituted of a set of sharp peaks corresponding to the interatomic vectors, which holds for periodic and quasiperiodic structures as well. Among them it is always possible to isolate a subset which is positioned on (quasi)lattice nodes and is characterized by the highest peak value distribution. This family can always be matched in physical direct space with a suited  $\mathbb{Z}$  module, which allows us to lift the quasilattice peaks into the n-D embedding space leading to a unique Bravais lattice. We would remark here an important feature of this technique, which is insensitive inside very broad limits to series truncation errors. On one side, these errors will modify the shape of the branches but the possibility of separation will not be affected. On the other side, the presence of a large amount of ripple will not hinder the procedure. It can always be separated after lifting the maxima. There could only be the necessity of a more sophisticated technique for locating and lifting the peaks with or without a matching  $\mathbb{Z}$  module as discussed later. Moreover, even the presence of high amounts of structural disorder and related high background has no serious consequences. Most Bragg peaks will remain observable and they contain all the information on the average (ordered) structure. Hence this technique can be effectively employed to detect the average structure.

We will try to focus here on two topics to underline the significance of the proposed technique and the directions of further developments. The first point is that the discussion has been limited to the important case (up to now the only case verified in our tests) in which the decoration hypervector centers are part of a sublattice. This gives us the possibility of indexing every Patterson peak with the same decoration  $\ensuremath{\mathbb{Z}}$  module. They can be lifted globally in a very simple and numerically convenient way. The radial profiles of the Patterson hypervectors can be compared and the correct n-Dlattice is eventually detected. When the set of all decoration hypervector centers do not form a sublattice this technique can be applied as well. It is now, obviously, no longer possible to index every Patterson peak with the same  $\mathbb{Z}$  module. If Patterson maps can be calculated with sufficient accuracy, however, it is always possible at least to index the "quasilattice" peaks. Besides, peaks can be found and integrated independently of a matching  $\mathbb{Z}$  module. Peak positions can also be lifted in the original unit cell without referring to a  $\mathbb{Z}$ module. Lifting only requires the knowledge of an embedding matrix which can be obtained in different ways. If the diffraction pattern has been completely indexed with the usual methods the related embedding matrix can be used for lifting. In the opposite case one can use any suitable embedding matrix between the ones compatible with the symmetryinduced invariant subspaces decomposition. Subsequently the plot of the radial profiles of the hypervectors will always allow any "trial" embedding to be checked and eventually corrected. If the quality of the data set is sufficiently good it is possible to detect correctly substructures as well as superstructures.

The third and most important point is the possibility of avoiding at all the first trial Bragg peaks indexing. The only strict requirements is the necessity to collect all observable Bragg peaks. With a four-circle diffractometer this may not be possible as the data collection process relies on a good estimate of possible peak positions. State-of-the-art area detectors allow, instead, all Bragg peaks to be found and to be located on any laboratory coordinate system. Based on these data a Patterson map can be always calculated. It has already been discussed how the problem of centering at least a subset of the Patterson peaks with a suitable  $\mathbb{Z}$  module, and/or to associate every peak with the relevant hypervector, can be managed. Once these steps have been accomplished we can start with our procedure to find the lattice. All these procedures presently appear complex and time consuming as determined by the available computer power. The advantages are the possibility of a more effective application of area detectors and also to always collect the most complete Bragg reflection set, including also satellite reflections or any Bragg peak whose indexing is troublesome, and then possibly to extract the related information which would otherwise be lost.

#### VI. SUMMARY

Using all observable Bragg peaks the Patterson function can be calculated. Embedding all peaks of the Patterson function in any suitable basis allowed by the *n-D* space group, the integrated peaks can be plotted versus their perpendicular space components. We have shown that the lattice peaks belong to the topmost branch showing a gap in the decoration peaks. Based on this diagram the *n-D* lattice of a decorated Fibonacci chain as well as the quasilattice of decagonal  $Al_{70.5}Mn_{16.5}Pd_{13}$  could be easily derived. This method also holds for all possible lattice arrangements even as sub- or superlattices. Using state-of-the-art area detectors this method may also be suitable as an *a priori* indexing procedure.

- <sup>1</sup>W. Steurer, *Physical Metallurgy*, edited by P.W. Cahn and P. Haasen (Elsevier, Amsterdam, 1996), pp. 463–467.
- <sup>2</sup>L. Elcoro, J.M. Perezmato, and G. Madariaga, Acta Crystallogr., Sect. A: Found. Crystallogr. **50**, 182 (1994).
- <sup>3</sup>Z. Fu, F. Li, and H. Fan, Z. Kristallogr. **206**, 57 (1993).
- <sup>4</sup>M. de Boissieu, P. Guyot, and M. Audier, *Lectures on Quasicrystals*, edited by F. Hippert and D. Gratias (Editions de Physique, Les Ulis, 1994), pp. 1–152.
- <sup>5</sup>T. Haibach and W. Steurer, Acta Crystallogr., Sect. A: Found. Crystallogr. **52**, 277 (1996).
- <sup>6</sup>J.-M. Dubois and Ch. Janot, J. Phys. (France) 48, 1981 (1987).
- <sup>7</sup>P. Bak, Phys. Rev. B **32**, 5764 (1985).
- <sup>8</sup>T. Janssen, Acta Crystallogr., Sect. A: Found. Crystallogr. 42, 261 (1986).
- <sup>9</sup>A. Singh and S. Ranaganathan, Philos. Mag. A 74, 821 (1996).
- <sup>10</sup>V. Elser, Phys. Rev. B **32**, 4892 (1985).

- <sup>11</sup>J.W. Cahn, D. Shechtman, and D. Gratias, J. Mater. Res. 1, 13 (1986).
- <sup>12</sup>S. Ritsch, C. Beeli, H.-U. Nissen, and R. Lück, Philos. Mag. A 71, 671 (1995).
- <sup>13</sup>F. Lançon, L. Billard, S. Burkov, and M. de Boissieu, J. Phys. I 4, 283 (1994).
- <sup>14</sup>T. Haibach, S. Kek, and W. Steurer, Z. Kristallogr. Suppl. **12**, 91 (1997).
- <sup>15</sup>W. Steurer, T. Haibach, B. Zhang, S. Kek, and R. Lück, Acta

Crystallogr., Sect. B: Struct. Sci. 49, 661 (1993).

- <sup>16</sup>M.A. Estermann, T. Haibach, and W. Steurer, Philos. Mag. Lett. 70, 379 (1994).
- <sup>17</sup> A. Janner, Acta Crystallogr., Sect. A: Found. Crystallogr. **48**, 884 (1992).
- <sup>18</sup>T. Haibach and W. Steurer, in *Proceedings of the International Conference on Aperiodic Crystals, Aperiodic '94*, edited by G. Chapuis and W. Paciorek (World Scientific, Singapore, 1995), pp. 399–403.