TOPICAL REVIEW

Software package for structure analysis of quasicrystals*

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Abstract
Recently a software package for the structure analysis of quasicrystals has been released, giving a better environment for determining quasicrystal structures. Therefore we can analyze their structures if we know data collection and indexing methods and a theory of structure analysis. For the use of the package, the structure analysis method and several techniques used in the package are shortly reviewed. How to treat key information in the input files of the programs is described in detail.

Keywords: software, quasicrystals, structure analysis

1. Introduction
Quasicrystals are special ordered structures which have non-crystallographic point groups including 5, 8, 10 or 12-fold axis, which never appear in conventional crystals. Their diffraction patterns, however, consist of the aggregates of Bragg reflections as in crystals. In the diffraction patterns, 5, 8, 10 or 12-fold rotation symmetry is found corresponding to the symmetry of quasicrystals. Most quasicrystals found so far are binary or ternary alloys [1]. It has been known already in 1986, that is, 2 years after the discovery of the first quasicrystals, that such a structure is obtained from a periodic structure in a higher-dimensional space by taking a three-dimensional (3D) intersection passing through an arbitrary point [2]. Therefore, the role of the structure analysis is to determine such a higher-dimensional periodic structure. Single (quasi-)crystals with enough size necessitated for the single crystal diffraction method had not been obtained for several years from the discovery but in 1987, the first such crystals were obtained [3] and after that single crystals for many quasicrystals have become obtainable [1]. Their extensive analyses have however been performed after 1997. (For detailed review before 1997, see [4].)

There are two types in quasicrystals. One is called polygonal (dihedral) quasicrystals, which have one 8, 10 or 12-fold axis and is periodic along this axis. They are called octagonal, decagonal and dodecagonal quasicrystals. These structure takes an ordered structure (quasiperiodic structure) in a plane normal to such a periodic axis. Another one called an icosahedral quasicrystal has no period along any directions.

Soon after the discovery, they had been considered to consist of some kinds of atomic clusters. Direct observations of quasicrystals by high-resolution electron microscopy played an important role for this [5]. If we employ this idea, it is easily conceivable that the structure description becomes easy by considering the clusters and atoms linking them. Noting this point, a higher-dimensional cluster model has been introduced [6]. In this model, we describe the clusters distributing in 3D space as a periodic structure in a higher-dimensional space. A software package based on this description has been written by the author and been released recently [7] (http://quasi.nims.go.jp/). In this paper, we describe shortly an analysis based on this program package.

The contents of the paper can be classified into three categories. The sections 1–7 are devoted to a short review of the structure analysis of quasicrystals. The following 9 sections describe on the software package and several calculation techniques employed in the package. Finally,
recent development of the structure analysis of quasicrystals is shortly mentioned.

2. Diffraction data collections

For the diffraction data collections of quasicrystals, we can use an imaging plage (IP) Weissenberg camera or 4-circle diffractometers. One important point for using them, is that the diffraction pattern can not be indexed by 3 vectors but 5 vectors in polygonal quasicrystals or 6 vectors in icosahedral ones. Conventional programs attached in a 4-circle diffractometer do not take into account this, so that we have to use 3 vectors and 3 real numbers for indexing. (This is possible in a 4-circle diffractometer CAD4 of Bruker.) If it is impossible, we use fictitious large lattice constants so that 3 is possible in a 4-circle diffractometer CAD4 of Bruker.) If it have to use 3 vectors and 3 real numbers for indexing. (This circle diffractometer do not take into account this, so that we use an imaging plage (IP) Weissenberg camera or 4-circle diffractometers. One important point for using them, is that the structure description and the analysis using the package for simplicity.

In quasicrystals, sets of vectors which are transformed to each other by the similarity transformation correspond to those in different settings, so that they are equivalent. For instance, the similarity transformation matrix $S$ for unit vectors $d_j^*$ of a 5D decagonal lattice is given by

$$S = \begin{pmatrix} 0 & 1 & 0 & -1 & 0 \\ 0 & 1 & -1 & 0 \\ -1 & 1 & 1 & 0 & 0 \\ -1 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}. \tag{6}$$

Therefore, the unit vectors related with $d_j^* = \sum_{i=1}^{m} d_j^*(S^m)_{ji}$ are equivalent. Since the similarity transformation have to maintain the unit cell volume, the determinant of $S$ should be $\pm 1$. (In this case it is 1.) Consequently, its inverse matrix is also an integral matrix. The vectors $d_j^*$ which are transformed by $S^m$ are parallel to the original ones but have a $\tau^m$ times larger length in the external space, while in the internal space, their length is smaller by $\tau^{-m}$. From this fact, it is easily understood that the lattice constant $a*$ in the external space can not be determined uniquely. The ratio between equivalent lattice constants is called a similarity ratio. It is an irrational number. In decagonal lattices, it is the golden ratio $\tau = (1 + \sqrt{5})/2 \approx 1.618$. In icosahedral primitive lattices, it is given by $\tau^3$, while it is $\tau$ for an icosahedral face-centered lattice). In literatures, several different sets of unit vectors are used for the same structure, because of this reason.

When we measure diffraction intensities by a 4-circle diffractometer, we need to determine the crystal orientation after finding the appropriate strong reflections and generate indices by using equation (1). After that, we obtain the real number indices $k_1, k_2, k_3$ from equations (1)–(5) and measure only reflections with generated indices. Therefore, we need to generate indices with potentially strong intensity efficiently. In quasicrystals, the intensity depends strongly on the length of the internal space component of the diffraction vector $q^* = |\mathbf{q}^*|$. As a general trend, if it is larger, the intensity of the corresponding reflection becomes smaller. On the other hand, we need to generate (the positions of) all observable reflections within the largest observable diffraction vector in the external space, $q^* = |\mathbf{q}^*|$. In order to choose such reflections, we generate an enough number of reflections and choose one with the external and internal space component which are less than the specified $q^*_\text{max}$ and $q^*_\text{max}$. In a 4-circle diffractometer, we usually measure only independent reflections or a few sets of reflections equivalent to them, since the measurement takes long time. (For the data collection in Weissenberg method, see [8].) After the data collection,
the real number indices are transformed into integral indices. They are used in the structure analysis.

3. Space groups

Since a quasicrystal is described as a crystal (periodic structure) in an \( nD \) space, its symmetry can be specified by an \( nD \) space group. All such space groups have already been calculated [9, 10]. There are not so many space groups, because the quasicrystals have a high symmetry. In icosahedral quasicrystals, it is only 11. Among them, some space groups impose the appearance of extinction rules. As is similar to conventional crystals, a symmetry operation of an \( nD \) space group consists of the rotational and translational parts. The former can be given by an \( n \times n \) matrix while the latter, by an \( n \times 1 \) matrix. The rotation matrix can be defined by the transformation property of \( d^* \) and the translation vector is determined by the extinction rules [11]. The extinction rules are tabulated in literatures [9, 10], but there exist no tables of all the symmetry operators as in the International Tables for conventional crystals at the moment. We can calculate all the symmetry operators by giving the generators of a space group. All the programs included in the program package can be used by giving only the generators except for those of the translations in the input file. As a result, we can input only 3 or 4 symmetry operators. (For the generators see [41].) For the symbols of \( nD \) space groups, we have no standard ones, but IUCr has proposed their recommended symbols, although these symbols are not widely used yet [12]. For example, the diffraction patterns of the \( b\)-Ni phase in decagonal Al–Ni–Co quasicrystals show systematic extinction rules, which is consistent with the space group \( P10_5/mmc(5^{2}1\ mm) \).

4. Direct methods

As in a conventional crystal, we can apply the direct method to a quasicrystal in order to obtain an initial model for the structure refinement [13]. Thanks to this method, we can obtain electron density in the unit cell of an \( nD \) lattice. In the \( nD \) space, the electron density is periodic, from which we can derive the electron density at any point in the 3D (external) space. (In the program package, the program \texttt{lodemac} is available, which is an extension of the similar program applied to proteins to \( nD \) space.) From this electron density, we can calculate 2D or 3D intersections and can see them by a 2 or 3D visualization software.

In order to interpret the electron density, we need to know a characteristic feature of it in the \( nD \) space. An atom is not a point in the \( nD \) space but is continuously extended up to some extent along the \((n-3)D\) internal space. This region is called an occupation domain (OD hereafter). At first, we need to know its location and rough size and shape, since this determines the atom positions in the 3D space. For this purpose, it is convenient to draw a 2D map taking one axis in the external and the other axis along the internal space. Since quasicrystals are highly symmetric, we can obtain important information from the cut passing through a high symmetric point like the origin or body center in many cases.

![Figure 1](http://example.com/f1.png)

**Figure 1.** Fourier maps of the \( b\)-Ni phase in d-Al–Ni–Co quasicrystals [14]. (a) and (b) are the maps drawn by using structure factors obtained by the low-density elimination method while (c) and (d), by the structure refinement, where (a) and (c) are located at \((1, 1, 1, 5z)/5\) while (b) and (d) are at \((2, 2, 2, 5z)/5\) \((z = 1/4)\).

In the \( b\)-Ni phase, if we choose one axis in the external space normal to the 10-fold axis and the other one in the internal space, we will find the ODs centered at \(\pm(1, 1, 1, 5z)/5\) and \(\pm(2, 2, 2, 5z)/5\). Similarly for finding the ODs, we can also use the peak search of the electron density within the unit cell of the 5D space. (In this quasicrystal, there exists an inversion center at the origin, so that one position with the sign \(\pm\) is related to the other by the inversion center.) Therefore when we write a map in the 2D internal space passing through these points, we have the electron density as shown in figures 1(a) and (b). (Other two figures related with these two by the inversion will be abbreviated.) To draw these, the program \texttt{qcmem} is applicable. Such figures are obtainable almost automatically by the application of the direct method. By using such information, we construct an initial model which is necessitated in the structure refinement. It should be taken into account that the electron density maps obtained by the direct method may not be accurate enough, since it gives wrong phase for weak reflections in many cases. (It is noticeable that there exists difference between figures 1(a) and (b) obtained from the direct method and figures 1(c) and (d) after the refinement.) However, the electron density in figure 1(a) is higher than in figure 1(b), so that we can consider that the former corresponds to Ni or Co and the latter to Al.

5. Initial model building and higher-dimensional cluster models

The most difficult part in the structure determination of quasicrystals at the moment is the building of an initial model. In addition to the information obtained by the direct method,
we need to use the density and chemical composition of quasicrystals as additional important information. The lattice constant is not determined uniquely as mentioned above, but the point density of atoms (the number of atoms in a unit volume) can be determined uniquely. We need to adjust the size and shape of the area (volume in icosahedral ones) of the OD so as to give a correct density and chemical composition. In addition, in the case where the approximant crystals are formed, we use the information from them. This is because we can frequently obtain the information of clusters in quasicrystals. Furthermore, the density of the approximants is almost the same as that of quasicrystals. We assume for simplicity that the cluster structure is obtained from the structure analysis of the approximants. In addition, we suppose that the interpenetration of the clusters is not allowed.

In a higher-dimensional cluster model, we assume the arrangement of clusters in quasicrystals. That is, we usually assume that the cluster center is located at vertices of some quasiperiodic pattern. In the b-Ni phase, the OD shown in figure 2 was employed. (In this case, the inversion center is assumed, so that there are other two ODs which are related to these two by the inversion. We abbreviate the description about such ODs. In the following, replace ‘the OD 1 etc’ with ‘the OD 1 and the other OD related to it by the inversion etc’.)

In this model, we assume that two kinds of clusters are located at the vertices of a quasiperiodic pattern known as the Penrose pattern. This consists of two rhombi joined by solid lines seen in figure 3. As the two clusters, we consider those denoted by 1 and 2 in the same figure. After the determination of the ODs, we build a periodic structure in the nD space. After that we can calculate all the corresponding atom positions in the 3D space by taking a 3D intersection of the periodic structure. It is known that the vertices of the Penrose pattern are generated from the two small gray pentagons labeled by 1 and 5 in figures 2(a) and (b) [15]. qstrec is the program for calculating atom positions in 3D space for given ODs. The electron density in 3D space can be drawn by the program qcgen.

There exist two kinds of vertices around which there are 5 to 7 atoms and 10 atoms (denoted by 1 and 2 in figure 3). The former and latter comes from the ODs 1 and 5, respectively. We call clusters consisting of atoms around these cluster centers clusters 1 and 2. In figure 2, the large ODs consist of the pentagons similar to these but in some cases, they are partially overlapped. For example, the OD 6 is overlapped partially with the ODs 7 and 8. In figure 2, a part except for the central pentagons (ODs 1 and 5) generates peripheral atoms around the cluster center. In addition, a part shared by two ODs generates an atom position which are shared by two clusters. We can prove that an ODs, which generates an atom position far from the cluster center by a fixed distance in the external space, is obtained from the OD of the cluster center by shifting it by that distance along the external space. In the present case, such an OD is located at the peripheral of another large OD. As a result, depending on the kind of clusters, we may have a very complicated polygons with concave parts consisting of the aggregates of small ODs. This is a characteristic feature of quasicrystal structure models consisting of atom clusters. It is worth noting that even if the shape of the ODs is complicated, the principle leading to them is simple.

6. Closeness conditions

It is difficult to conceive that large holes are included in quasicrystals, since they are alloys. Therefore, we need to consider quasicrystals, which do not include such holes. It is also unrealistic that the structure shows an interatomic distance shorter than a value determined by the atomic radii. As a result, we need to consider a dense packing of atoms without including such shorter interatomic distances. Such a model imposes restrictions for the ODs.

Figure 2. ODs of the b-Ni phase in d-Al–Ni–Co quasicrystals. In (a), the OD is located at (1, 1, 1, 5z)/5, while in (b), it is at (2, 2, 2, 5z)/5 (z = 1/4).

Figure 3. Structure model of the b-Ni phase in d-Al–Ni–Co quasicrystals projected along the 10-fold axis. A black circle represents the transition metal (Ni,Co) and gray, Al. The boat-shaped region located at the outermost part in figure 2 (a part of OD 3) and whole area of figure 2(b) are assumed to be occupied by Al. On the other hand, the transition metal positions are generated by the large pentagonal region except for the boats in figure 2(a).
Let two ODs be located at \( x_1 \) and \( x_2 \). (Since the ODs extend in some extent in the internal space, it might be more accurate to say that they are passed through these points, but for simplicity, we say that they are located at these points.) In order to consider such restrictions, we have to know the external and internal space components of a point in the 5D space.

The unit vectors of the decagonal lattice, \( d_j \), \( (j = 1, 2, \ldots, 5) \) are given, from equations (2) and (3) and \( d_j' \cdot d_j = \delta_{jj}, \) by

\[
d_j = (2a/\sqrt{5})[(c_j - 1)a_1 + s_j a_2 + (c_{2j} - 1)a_4 + s_{2j} a_5], \quad (1 \leq j \leq 4),
\]

\[
d_5 = ca_3.
\]

The external and internal space components of the point \( x \) represented by coordinates \((x_1, x_2, x_3, x_4, x_5)\) can therefore be obtained from \( x = \sum_{j=1}^{5} x_j d_j \).

For the relation between the lattice constants in the direct and reciprocal space, we have relations \( a = 1/a^*, \ c = 1/c^* \) which are similar to the corresponding ones in conventional crystals, but it should be noted that in equation (2), the length in the external space of \( d_j' \ (j \leq 4) \) is not \( a^* \) but \( a^*/\sqrt{5} \).

Similarly, that of \( d_j \ (j \leq 4) \) is not \( a \) from equation (7).

We already mentioned that a 5D decagonal lattice with a lattice constant differ by the power of \( \tau \) is the same as the original one. We consider the case where we have a lattice with the lattice constant \( \tau a \) in the following. We write the unit vectors for this case as \( d_j^{'} \). Then it is well known that the transformation properties of the corresponding coordinates are transformed by the same rule as that of \( d_j^{'} \). Namely, \( x^*_i = \sum_{j=1}^{5} x_j \tau_i S_j \). The point represented by \((1, 1, 1, 1/5)\) is given by \((-2, 3, 3, -2, 5\tau)/5\) in the transformed coordinate system. This is equivalent to \((-2, 2, 2, -5\tau)/5\). Thus we should be careful when we compare results described in different coordinate systems.

Now we turn to the structure model of b-Ni again. As mentioned before there exist ODs at \( \pm(j, j, j, 1, 5\tau)/5 \) \((j = 1, 2)\) in this case. (Of course, there exist same ODs at every point which differs by a lattice vector.) The point \( x_1 = (1, 1, 1, 0, 0)/5 \) differs from \( x_2 = (2, 2, 2, 2, 0)/5 \) by \( 2a/\sqrt{5} \) both in the external and internal spaces. The point \( x_3 = (5, 1, 1, 5, 0)/5 \) far from the former by a lattice vector \((1, 0, 0, 1, 0)\) is at the distances of \( 2(2 - \tau^{-1})a/\sqrt{5} \) and \( 2(2 + \tau)a/\sqrt{5} \) in the external and internal spaces. That is, it is near in the external and far in the internal space. A quasicrystal structure is given by a 3D intersection passing through any point in the \( nD \) space, as mentioned above. Let the ODs at \( x_1 \) or \( x_3 \) overlap on the OD at \( x_2 \) when they are projected onto the internal space. Then if we consider the intersection which is passing through an overlapped region, we have an atom position with an interatomic distance of \( 2a/\sqrt{5} \) or \( 2(1 - \tau^{-1})a/\sqrt{5} \). Therefore if the former is allowed but the latter is not allowed as an interatomic distance, we can conclude that the ODs at \( x_2 \) and \( x_3 \) should not be overlapped. (Note that the OD at \( x_1 \) is distant from the OD at \( x_2 \) by \( 2(1 + \tau)a/\sqrt{5} \) in the internal space. This is larger than \( 2a/\sqrt{5} \)).

The point density of atoms is proportional to the area of the ODs, so that it can be understood that in order to obtain the densest structure without the latter interatomic distances, the ODs at \( x_2 \) and \( x_3 \) can be contact to each other without gaps. Figures 2(a) and (b) contact to each other under such a condition. Such conditions imposed to ODs are called closeness conditions.

7. Structure refinements

After the initial model building, we refine atom positions, temperature factors, and occupation probabilities by the least-squares method in order to obtain accurate distributions of atoms and their positions. In a higher-dimensional cluster model, we consider homogeneous parameters within a small ODs (in the case where the pentagons are subdivided by the overlapping, within its subdivided parts). That is, we assign same atomic shift, temperature factor and occupation probability for all points in each small OD. They are determined so that the difference between observed and calculated structure factors becomes minimum. (In this model, the structure factor can be calculated analytically, so that the calculation time is shortened [4].) However, the subdivision of the OD is not uniquely determined and we can divide them in several manners. As mentioned in the next section, exactly speaking, each atom environment can be all different from each other but if we consider only the nearest neighbor atoms, we have many atoms with a same local environment. However, the number of such atoms (positions) reduces when we take into account the difference of the environment in more distant atoms. Of course, if we consider the difference up to an infinite distance, no atoms are in the same local environment. Therefore as mentioned in the next section, the smallness of the subdivisions is different depending on the fact that up to what distance we take into account the difference of the environment.

In addition to these structural parameters, it is known that the phason temperature factor has to necessarily be introduced, as a parameter specific to quasicrystals. This is an overall temperature factor and it is common for all ODs. The cluster arrangement of real quasicrystals will be fluctuated from an ideal one which is assumed to be a simple quasiperiodic pattern. This is called the disorder due to the random phason. The ODs leading to such a disordered atom positions are given by an OD larger than in the ideal case. It is however not fully but statistically occupied. As a result total area (or volume in icosahedral quasicrystals) calculated by weighted integrals is the same as in the case where entire OD is fully occupied. In a structure analysis, the disorder by random phason is expressed by such a fading of the boundary of ODs. The effect of the fading to the structure factor is taken into account by the phason temperature factor. (This is similar to the fact that the fading of the atom position due to phonon is accounted for the temperature factor.) This is only one parameter but it strongly affects the diffraction intensity and without using it, we can not explain diffraction intensities of quasicrystals.

Another problem, which should be taken into account, is that one atom site is frequently occupied with two or more atoms, since quasicrystals are in many cases alloys.
shows the programs included in the program about 10%.

we use a synchrotron radiation, we have the R-factor of crystals.) Our experience shows however that even when reflections, we have the R-factor comparable with the value of the R-factor. (Of course, if we do not intend to use weak radiation, it becomes worse. This means that we can not measure many weak reflections by using synchrotron radiation, it becomes worse. This means that we can not expect the R-factor better (smaller) than the value for conventional crystals. In particular, if we refine quasicrystals, however, there are many weak reflections, so that many of their intensities are not so accurate enough. Therefore we can not expect the R-factor better (smaller) than the value for conventional crystals. In particular, if we do not intend to use weak reflections, we have the R-factor comparable with the value of crystals.) Our experience shows however that even when we use a synchrotron radiation, we have the R-factor of about 10%.

| program  | description                                      |
|----------|--------------------------------------------------|
| lodemac  | the direct method for aperiodic crystals          |
| qcdiff   | the structure refinement of quasicrystals        |
| qestrc   | the atom positions of quasicrystals in the external space |
| qcmem    | a drawing program for Fourier and MEM maps       |
| polygn   | a drawing program of ODs for polygonal quasicrystals |
| polyhyd  | a drawing program of ODs for icosahedral quasicrystals |
| pltint   | a program for drawing \( F_p-F_c, q^*F_p \) or \( q^*F_c \) diagrams |
| cluster  | a drawing program of clusters in quasicrystals    |
| extrhkl  | a program for extracting the most probable result from the output of lodemac |
| difftpn  | a program for simulating diffraction patterns of aperiodic crystals |
| lphason  | a program for calculating linear phason strain components of approximants |
| iatom    | a program extracting independent atom positions of crystalline approximants |

8. Software package for quasicrystals

The software package QUASI05.08 was released at the end of August 2005 which includes binary files of a set of programs for Linux, Windows and MacOS X operating systems. In the second release at the end of August 2007, other 3 programs (the last three in table 1) are added and the source codes of all the programs are included. This enable us to use the program package in any computers.

Table 1 shows the programs included in the program package QUASI07.08. All of them are written in Fortran 77 but their small fragments are written in C++. The programs are applicable to any quasicrystals known so far. In the programs, the icosaedral quasicrystals are specified by \( isys = 1 \) or 2, while the decagonal, octagonal and dodecagonal ones by \( isys = 3, 4 \) and 5, respectively. For \( isys = 1 \), one of the unit vectors, \( \mathbf{a}_3 \), of the external (and also \( \mathbf{a}_0 \) in the internal space) is taken along the 5-fold axis, while another one is along the 2-fold axis. On the other hand, when \( isys = 2 \) was used, the three unit vectors in the external space \( \mathbf{a}_1, \mathbf{a}_2 \) and \( \mathbf{a}_3 \) are all along the 2-fold axes. (See equations (37) and (60) in [4]. These two are equivalent but the former is convenient for considering a surface which is normal to the 5-fold axis, while the latter will be more convenient for calculating the cubic approximant structures from a 6D model of icosaedral quasicrystals.)

The essentially important programs are three: qcdiff, qestrc and lodemac as mentioned before. qcdiff is applicable to simultaneous use of several data sets. This allows the combined use of single crystal or powder x-ray and neutron data. The powder mode is convenient for the simulation of diffraction patterns for given \( nD \) structures. The program qestrc can calculate the coordinates of crystalline approximants which is derived from an \( nD \) model by introducing an appropriate linear phason strain, as mentioned later. The necessary linear phason strain is obtained by lphason. As mentioned later, qestrc outputs the Cartesian coordinates of the crystalline approximants and iatom extracts coordinates of their independent atoms.

In order to assist the model building and the understanding of refined structures, the package includes several other programs. The Fourier map is necessary for obtaining the electron density in an \( nD \) space or for its 2 or 3D intersections. They are calculated by qcmem. After getting rough...
electron density by the direct method using lodemac, we need to determine ODs for the structure refinement. In this process, we need to give the ODs for the cluster centers and the total ODs which are subdivided by them. In order to draw such ODs, polygon (in polygonal cases) and polyhd (in icosahedral cases) are available. They can draw a complicated ODs as in figure 2 or figures 3 and 4 in [16]. They output PostScript files, so that these can be modified by any other graphics editor of PostScript figures. The shading in figures 2–4 are obtained from the output files by such an editor. For icosahedral quasicrystals, the ODs are complicated polyhedra consisting of the ODs of the cluster centers. The program polyhd can be used for drawing such ODs. This produces a PostScript or VRML file and the shape of ODs can be visualized by an appropriate viewer of such files.

The program pltint is a simple program which plots $F_o-F_c$, $q^2-F_o$ and $q'-F_o$ diagrams, while diffptn is used for the simulation of diffraction patterns. The program extrhkl is a small program which extracts a specified solution by lodemac, since it gives multi solutions for each intensity data. The probable solutions are chosen among several trials by considering the R-factor and the ratio between maximum electron density. If the absolute value of the ratio is small and the R-factor is small, its solution is probable. (Note that minimum electron density is negative since the lodemac does not use the intensity for the reflection with $q = 0$, which gives the average electron density.)

9. Input format for ODs

All the programs are terminal-based, so that they have to be run in a terminal emulator. All of them but extrhkl read one or several input files. Among them one is regarded as the standard input file, in which file names of other files have to be specified if any. The standard input file is specified in the command line in Unix (Linux), Windows and MacOSX operating systems. Detailed data instructions for all programs are given in html files included in the package. The most complicated input data will be those for specifying ODs, which is explained below.

For programs qcdiff, qestr, polygon, polyhd, we need to input the detailed information of the ODs, which includes their shape, size and location in the $nD$ space. In general, a large OD in higher-dimensional cluster models has a complicated shape with concave parts. This consists of smaller one, which is related with the ODs generating the cluster centers. They are in many cases, polygons (or polyhedra) with high-symmetry. However, the subdivided ODs, which are given by the overlapped parts of the ODs for the cluster centers, is lower symmetric. We need to define a variety of such ODs. In the program, the symmetry operations are thoroughly used in order to reduce input data and to simplify the input file.

If we employ a higher-dimensional cluster model, similar ODs appear at many different positions as seen in figure 2. In order to specify them, therefore, we define once their common ODs and use them at different positions. In general, such ODs have symmetry less than the total symmetry of quasicrystals and therefore, we need to specify their orientations in addition to their shape and size. Noting that each (subdivided) OD is a polygon, this is further subdivided into triangles (for polygonal) or tetrahedra (for icosahedral quasicrystals). The four programs mentioned above read the shape of common ODs from a file with the extension ‘.pod’ (the ‘.pod’ files).

In this file, we define a common OD by specifying its corner vectors in $nD$ representations. The programs calculate their internal space components and use them. In decagonal cases, we use their 6D representation for convenience. The unit vectors $d_i$, reciprocal to those of the reciprocal lattice are given by equations (7) and (8). Instead of using these vectors, we use

$$\mathbf{d}_j = (2a/\sqrt{5})[c_j \mathbf{a}_1 + s_j \mathbf{a}_2 + c_{s_j} \mathbf{a}_4 + s_{s_j} \mathbf{a}_5], \quad (1 \leq j \leq 5), \quad (9)$$

although one of $d_i$ ($j = 1, 2, \ldots, 5$) is redundant. The coordinates with respect to $d_j^i$, $(x_1, x_2, x_3, x_4, x_5)$, are convenient to specify the shape of ODs in a decagonal quasicrystal. For example, the corner vectors of the pentagon with a radius $2a/\sqrt{5}$ are given by $(1, 0, 0, 0, 0, 0)^t$, $(0, 1, 0, 0, 0, 0)^t$, $(0, 0, 1, 0, 0, 0)^t$, $(0, 0, 0, 1, 0, 0)^t$ and $(0, 0, 0, 0, 1, 0)^t$, where the superscript $i$ denotes the internal space component of a 5D vector. On the other hand, their 5D coordinates with respect to $d_j$ ($j = 1, 2, \ldots, 5$) are $-(-4, 1, 1, 1, 0)^t/5$, $-(-1, -4, 1, 1, 0)^t/5$, $-(-1, 1, -4, 1, 0)^t/5$, $-(-1, 1, 1, -4, 0)^t/5$ and $-(-1, 1, 1, 1, 0)^t/5$. The program calculates from the 6D representation their internal space components, so that users can specify the 6D representation of corner vectors of the OD. Figure 4 shows an example of the input file for defining the OD shape.

Similarly, we use 7D coordinates for specifying the ODs of dodecagonal quasicrystals, which are those with respect to

$$\mathbf{d}_j = (2a/\sqrt{3})[c_j \mathbf{a}_1 + s_j \mathbf{a}_2 + c_{s_j} \mathbf{a}_4 + c_{s_j} \mathbf{a}_5], \quad (1 \leq j \leq 6), \quad (11)$$

where $c_j = \cos(2\pi j/12)$ and $s_j = \sin(2\pi j/12)$. For the position of the center of OD $x_0$ mentioned later, the 5D coordinates with respect to equations (7) and (8) or equation (34) in [4] are used in both cases. On the other hand, in octagonal and icosahedral quasicrystals, the same 5D or 6D coordinate system is used for specifying the shape of ODs, $x_0$ and $x'$.

In order to specify total ODs, we need to give all small ODs and their size and orientation. As mentioned above, the shape of all the ODs necessary for constructing the total ODs are defined in the ‘.pod’ file. On the other hand, the size and orientation is given in another file with the extension ‘.atm’. In this file, we choose the shape of the OD by the serial number of the OD, defined in the ‘.pod’ file and give the magnification (radius) of the OD. The orientation is specified by the symmetry operator number, which is generated by the generators in the ‘.pod’ file. (The programs also allow
The center of large ODs are, in many cases, at highly-symmetric special (Wyckoff) positions in the \( nD \) lattice. In the case of b-Ni, they are at \( \pm (j, i, j, i, 5\xi)/5 \) \((j = 1, 2; \xi = 1/4)\) as mentioned in section 2. Their site symmetry group is \( 5/mmm2(5^21m1) \) [12]. In polygonal quasicrystals, the OD is an object in the \( 2D \) internal space. They are restricted by its second part \( 5^21m1 \) in the parenthesis. (The mirror plane normal to the \( 5 \)-fold axis and 2-fold rotation axis do not restrict their shape. This is implied by 1 in this symbol.)

The center of the subdivided small ODs in figure 2 is in general apart from this center by a vector in the internal space. Therefore, we use two vectors, one of which, \( \mathbf{x}_0 \), specifies the center of the large ODs and the other vector, \( \mathbf{x}' \), in the internal space represents the shift from it. We specify a triangle, by two edge vectors \( \mathbf{e}_1 \) and \( \mathbf{e}_2 \) the origin of which is at \( \mathbf{x} = \mathbf{x}_0 + \mathbf{x}' \). Therefore the positions of the two corners are given by \( \mathbf{x} + \mathbf{e}_1 \) and \( \mathbf{x} + \mathbf{e}_2 \). (In icosahedral quasicrystals we need to use three vectors \( \mathbf{e}_1 \), \( \mathbf{e}_2 \) and \( \mathbf{e}_3 \) specifying three corners of a tetrahedron.) It should be noted that \( \mathbf{e}_1 \) (\( j = 1, 2 \)) are vectors in the internal space. For \( \mathbf{x}' \) and \( \mathbf{e}_j \), we can give \( nD \) vectors with an external space component but the program uses only their internal space component for the sake of convenience.

The shape of ODs is restricted by the site symmetry. When we consider an OD which includes a point with the highest symmetry within the OD, its shape has to maintain that symmetry. This means that the shape of the OD is restricted by the highest point symmetry within the OD. This restriction comes from the assumption used in the higher-dimensional cluster model, where the atom position, temperature factor, and occupation probability are assumed to be same within the subdivided OD. Then the restriction imposed to them is determined by the highest point symmetry, for which the largest restrictions are imposed.

According to this restriction, the shape of the OD located at the site symmetry group should be invariant under that group as in the ODs shown in figure 2, where the site symmetry group for the OD is \( 5/mmm2(5^21m1) \). Even under such a restriction, we can consider many possible ODs. The simplest one will be a convex polygon like a regular pentagon as in figure 2(b), but the symmetry restriction allows that the OD is a concave polygon or even a fractal. The programs included in the package do not expect a fractal OD, but assume a polygonal (or polyhedral) one. Therefore it can be decomposed into several convex polygons (or polyhedra in icosahedral cases). In particular, any polygonal (polyhedral) ODs can be divided into triangles (tetrahedra), even if it has a concave part as in figure 2(a). Therefore in the programs, the OD is specified by the corner vectors of triangles (tetrahedra).

It is evident that if we use the site symmetry, we can just specify the independent parts of ODs, since the other part can be derived from them by the symmetry operations. The independent parts of the OD in this case are the parts between occupation probability. Different from the conventional atom, however, the OD is neither a point nor a sphere in the \( nD \) space but extended in the internal space within the OD. This means that if the shape, size or orientation of the OD is different, its structure factor is different. (For the expression of the structure factor, see [4].)
the dotted lines on the mirrors denoted by \( m \) in figure 4. In the program, however, we sometimes specify larger parts, like shaded parts in the same figure, depending on structure models as mentioned below.

The programs calculate the site symmetry of \( \mathbf{x} = \mathbf{x}_0 + \mathbf{x}' \) and assume that (the shape of) an OD located at this position has this symmetry. This means that we need to chose a point with the highest point symmetry in the OD as the origin of the vectors \( \mathbf{e}_j \). Then the program can generate their equivalent ODs by using the coset representatives of the point group \( G \) of quasicrystals with respect to the site symmetry group \( G_s \).

Consider the shaded pentagonal ODs 1 and 5 located at the centers in two large ODs in figure 4. Both of them can be formed by 5 triangles surrounded by two of five lines (radial vectors) connecting the center and 5 corners and 1 of 5 edges of the pentagon. Therefore they can be specified by two radial vectors and 5-fold rotation operators. In these cases, \( \mathbf{x}_0 \) is \((1,1,1,5z)/4 \) or \((2,2,2,2,5z)/4 \) \((z = 1/4)\) and \( \mathbf{x}' \) should be the zero vector, because \( \mathbf{x}_0 \) is in the OD, and it has the highest point symmetry in this OD. Since the point group of \( \mathbf{x} = \mathbf{x}_0 + \mathbf{x}' \) is \( 5/mm2(5^21m1) \), such a small pentagon can also be specified by a smaller triangle defined by two dotted line and a half edge and the symmetry operators of this site symmetry group. Noting that the first part of the point group symbol shows the symmetry operation in the external space and the second part in the parentheses represent the corresponding symmetry operation in the internal space, the symbol indicates that the mirror which is normal to the 5-fold axis (the first \( m \) in the first part) leaves the OD invariant. (In such a case the occupation probability should be 1/2 in order to obtain the correct point density in \( \mathbf{q}_{\text{estcr}} \), since the multiplicity of the OD becomes 2. The same holds for the input data of \( \mathbf{q}_{\text{cdiff}} \).)

Now we consider the shaded pentagonal ODs 3 and 6. Their centers are on the mirror plane in the internal space. Therefore, the highest site symmetry of these ODs is \( mm2(1m1) \). It is convenient in this case to specify the corner vectors \( \mathbf{e}_j \) from the center of the pentagon similarly to the OD 5. Then \( \mathbf{x} = \mathbf{x}_0 + \mathbf{x}' \) is on the mirror. In this case, the program assumes an OD with the mirror symmetry as mentioned above. Therefore we need to specify the whole pentagon although its independent part is its half (squashed square bisected by the dotted line in figures 4(a) and (b)).

For all pentagons on the mirror, the coset representatives form a group \( 5(5^2) \). Therefore, other 4 pentagons (with the same numbers 3 or 6) around the large OD center are generated by the program. Similarly from the same reason, in order to specify the triangles 2 and 7, we need to specify the whole triangles, instead of their independent part. (Note that they are also on the mirror plane.) In this way, we can construct the whole ODs by 4 pentagons (ODs 1, 3, 5, 6) and two triangles (ODs 2 and 7). This is a simple subdivision of the ODs in \( \text{b-Ni} \), but it is not unique and different subdivisions are used in the real structure analysis [17]. The subdivision is closely related with the symmetry of the clusters generated by such a model as discussed in the next section.

10. Subdivisions of ODs and symmetry of clusters

Even if we assume the shape and size of large ODs from the electron density obtained from the direct method, we can consider many ways for their possible subdivisions as mentioned in the previous section. As an example, we compare the ODs shown in figures 4 and 5. In the former, all the small ODs are on the special positions, while in the latter, they are in a general position. That is, the special positions are on the boundary of ODs. Such a difference in the subdivision imposes a different restriction for the shift of ODs in parallel to the external space. This is very important in the structure analysis, since the shift of ODs causes the shift of atomic positions and the x-ray diffraction intensities are sensitive for such a small shift of atomic positions. The subdivision is directly related with the model, since possible shift of the subdivided ODs are highly restricted by the way of subdivisions.

In the higher-dimensional cluster model, the restriction imposed by the site symmetry is determined by the highest point symmetry in the ODs as mentioned before. Such a point is assumed to be given by \( \mathbf{x} = \mathbf{x}_0 + \mathbf{x}' \). Therefore if we consider that all the atoms in quasicrystals are on a general positions so that they can be shifted along any direction, \( \mathbf{x} \) for the corresponding OD should be at a general position. When \( \mathbf{x} \) is at a special position, this is achieved by shifting \( \mathbf{x}' \) by a small amount to a general position. For example, if we choose \( \mathbf{x}' \) for the corner of a triangle which lies on a mirror, we can shift it artificially along the direction normal to the mirror by small amount. Similarly, for the triangles at the center of the large ODs, like the triangle defined by two arrows in figure 4(d), \( \mathbf{x}' \) should not be the zero vector but be chosen so that they are neither on the 5-fold axis nor the mirror plane.

Because of the error in the floating point number in numerical computations, the programs assume that its deviation is the order of \( 10^{-5}a \), where \( a \) is a lattice constant. If the origins of all small triangles in figure 5 are chosen in such a way, the shifted ODs are enough to generate all the equivalent ones in contrast to figure 4, where non-independent parts have to be included. The programs output the site symmetry group \( G_s \) calculated from \( \mathbf{x} \) and the coset representatives. They are available to confirm the site symmetry and equivalent positions used in the calculations.

11. Shift of ODs in the external space

In the structure analysis, we refine the atom positions by shifting each small ODs in parallel to the external space. The allowed shifts are limited by the site symmetry as mentioned above. As in a conventional crystal, \( \mathbf{x} \) is on a rotation axis, their only allowed shift should be parallel to it. On the other hand, if it is on a mirror plane, the shift normal to it is prohibited. Since the rotation axis is not parallel to the unit vectors \( \mathbf{a}_j \) \((j = 1,2,3) \) in the external space in general, we specify their direction by three \( nD \) vectors \( \mathbf{x}'_j \) \((j = 1,2,3) \). Similarly to \( \mathbf{x}' \), we specify vectors with internal space components for \( \mathbf{x}'_j \). The programs project them onto the external space and normalize them so that the value of
The shift parameters $u_j$ ($j = 1, 2, 3$) along $x_j$ represents the shift in angstrom. The shift of an OD is therefore given by
\[ \sum_{j=1}^{3} u_j x_j'. \]
Some of $x_j'$ will be redundant for special positions, since only a shift along 1 or 2 directions is allowed, but even in such a case, we need to specify dummy vectors for them and can fix the corresponding shift in the structure refinement.

The dummy vectors can be the zero vector. These vectors are also used in the description of thermal ellipsoids when the anisotropic temperature factor is employed. In such a case, three non-zero vectors should be specified even when the ODs are at special positions and shift along some directions is not allowed. There are no restrictions for the direction of the dummy vectors, but it is convenient to choose orthogonal vectors, which are normal to a rotation axis or the normal of a mirror plane when the site symmetry includes such symmetry operations. Therefore the programs in the package do not use dummy vectors, but it is convenient to choose orthogonal vectors.

It is worth noting that since the ODs 2, 3, 6, and 7 in figure 4 are on one of the 2 mirrors passing through the center of ODs 1 and 5, they can only shift along the radial direction from the cluster center. Since they generate the first neighbor of the cluster center as stated in section 2, the cluster maintains the 5-fold symmetry for any allowed shift. For example, one of $x_j'$ of the OD 3 in figure 4 should be $(0, 0, 0, 0, 0, 0)$ while that of the OD 2 is $(1, 0, 0, 0, 0, 0')$, where we use the 6D representation for a 5D vector (see equation (9)).

12. Similarity transformation for 3D structures and nD Fourier maps

As is already mentioned in section 4, the use of Fourier maps is helpful in the model building in nD space. The fastest calculations of an nD Fourier map will be performed by the use of fast Fourier transformation. This sometimes necessitates huge computer memory, since the electron density is calculated at grid points in the unit cell of the nD space and the necessary number of grid points becomes huge. In particular, for icosahedral quasicrystals, it may...
be serious [18]. This number is however strongly depends on the choice of the unit vector \( \mathbf{d}_j \) as is similar to the choice of \( \mathbf{d}_j \) in the case of indexing. In order to include all the observed reflections in the calculations of Fourier maps, we need to use the number of divisions along the \( j \)-th direction more than \( 2h_{\text{max}} \), where \( h_{\text{max}} \) is the maximum index of the observed reflections for the direction of \( \mathbf{d}^* \).

Since, the maximum indices necessitated for indexing depend on the choice of the unit vectors of the reciprocal lattice, they depend on the choice of those for the direct lattice. In other words, we can use an appropriate unit vectors \( \mathbf{d}_j = \sum_{j=1}^{6} (S^{-1})_{ij} \mathbf{d}_i \) which are given by an appropriate order \( m \) of the similarity transformation. If the \( \Pi_{j=1}^{6} h_j \) is small, the necessary memory will be small. This is related with the fact that the diffraction intensity is small for the reflections with the diffraction vector which has a large internal space component. The maximum entropy method (MEM) and the low-density elimination method (LDEM) also use Fourier transformations repeatedly for calculating the electron density within the unit cell of the \( nD \) lattice. Therefore the same holds for MEM and LDEM.

The similarity transformations are also efficient for calculating all atom positions in the external space within some range. Provided that we cut an \( nD \) structure at the 3D hyper-plane passing through \( \mathbf{x}_i \) and the center of a large OD is at \( \mathbf{x}_i + \mathbf{x}_j \), if the internal space component of the vector \( \mathbf{x}_i + \mathbf{x}_j \) is larger than the maximum radius of that OD, the OD can not be cut by the 3D hyper-plane. Therefore, we do not need to consider the lattice points \( \mathbf{x}_i \) far from that distance in the internal space. The similarity transformation with an appropriate order \( m \) gives an efficient way to generate lattice vectors \( \mathbf{x}_i \) with small internal space components.

From this reason, in the program \texttt{qestrc}, \texttt{qcdiff}, \texttt{qcmem}, \texttt{lodemac}, we need to specify the order of the similarity transformation \( m \). In \texttt{qestrc}, \( m \) means the order of the similarity transformation for the unit vectors in the direct lattice: \( \mathbf{d}_j = \sum_{j=1}^{6} (S^m)_{ij} \mathbf{d}_i \), while for the latter three, it is the order for those in the reciprocal space: \( \mathbf{d}^*_j = \sum_{j=1}^{6} (S^m)_{ji} \).

We use such unit vectors only for generating indices or coordinates of grid points in a unit cell but after that the indices or coordinates are inversely transformed for obtaining those in the original coordinate system.

In the face-centered icosahedral lattice, the calculation of electron density for the whole volume of the conventional 6D unit cell is redundant, since we have 32 centering transformations. It is efficient to use the asymmetric unit defined by \( \mathbf{d}_j = \sum_{j=1}^{6} S_{ij} \mathbf{d}_i \) with

\[
S = \frac{1}{2} \begin{pmatrix}
0 & 1 & 0 & -1 & 0 & 0 \\
0 & 0 & 0 & 0 & -1 & 0 \\
1 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & -1 & 1 \\
0 & 0 & 0 & -1 & 0 & 0 \\
0 & 0 & 0 & -1 & 1 & 0
\end{pmatrix}.
\]

The determinant of \( S \) is \((1/32)\), implying that \( \mathbf{d}^*_j \) \( (j = 1, 2, \ldots, 6) \) span the same lattice. Therefore once we calculate the electron density at the grid points within this unit, we can calculate electron density at any other point by the interpolation. The coordinates with respect to these unit vectors, \( x_j \), corresponding to the original coordinates, \( x_j \), are related with \( x'_j = \sum_{j=1}^{6} (S^{-1})_{ij} x_j \), where the inverse matrix \( S^{-1} \) is given by

\[
S^{-1} = \begin{pmatrix}
0 & 0 & 2 & 1 & 1 & -1 \\
2 & 0 & 0 & -1 & 1 & 1 \\
0 & 2 & 0 & 1 & -1 & 1 \\
0 & 0 & 0 & -1 & 1 & 1 \\
0 & 0 & 0 & -1 & -1 & 1 \\
0 & 0 & 0 & 1 & -1 & 1
\end{pmatrix}.
\]

The use of the asymmetric unit can drastically reduce the computer memory necessary for the calculation of the electron density in face-centered icosahedral quasicrystals. Even when this is used the number of available grid points may be not enough for most computers, so that non-linear interpolation is used in the program \texttt{qcmem} in order to obtain smooth contours in a electron density map.

13. Constraint conditions

In the structure analysis, we need to restrain or constrain several parameters. The constraint conditions are written in a separate file, ‘.cnd’ file. In order to obtain physically reasonable occupation probabilities, temperature factors and chemical compositions, penalty functions are used [19]. Artificial constraints between several temperature factors or occupation probabilities have sometimes to be used for small ODs giving positions with similar local environments. If the area (or volume) of ODs are small, the refinement of independent temperature factors or occupation probabilities easily diverges, although the refinement of the atomic shift of such ODs is usually stable. In general, the correlations between temperature factors and occupation probabilities are strong. It will be the reason why the simultaneous refinement of the temperature factor and occupation probability is unstable. In order to avoid the instability in the least-squares refinement, the grouping of the temperature factors or occupation probabilities is efficient. In such a case, we can impose a constraint such that parameters in a group have a same value. Such a constraint is available in \texttt{qcdiff} and \texttt{qestrc}. For the use of the same constraint between them, the constraint condition is written in a file.

In the current version of \texttt{qcdiff} and \texttt{qestrc}, one line defines an independent refinable parameter. Therefore all the refinable parameters have to be defined in the ‘.cnd’ file. Each parameter can be specified by several numbers in a line. First number (after \( z= \)) is an independent parameter value. The second one (after \( \pm= \)) specifies the number of constrained parameters which is followed by the triplet of the OD number, the kind of parameters and the coefficient. The numbers 1 to 9 for the kind of parameters imply \( a_1, u_2, u_3, B, B_1, B_2, p, s_1, s_2 \), where \( B \) and \( B_1 \) are isotropic and pseudo-anisotropic temperature factors, respectively. \( B_2 \) stands for the phason temperature factor. The factor \( B_1 \) is applicable only to the polygonal (dihedral) quasicrystals. (See [19].) The numbers
13–18 represent the anisotropic temperature factors $U_{11}, U_{22}, U_{33}, U_{23}, U_{31}, U_{12}$. Note that the coordinate systems of these factors are different in various ODs in general as mentioned before.

The number 10–12 means the factor $v$ for $x_i$ given in the ‘.atm’ file and factors $r_{\text{min}}$ and $r_{\text{max}}$ for $e_i$ defined in ‘.pod’ files. The vector $x_i$ mentioned in section 10 is specified by $x_i^v$, the multiplication of a vector $x_i^v$ and the factor $v$, in qcdiff and qestrc etc for the shake of convenience. Usually, $r_{\text{min}} = 0$. Then $r_{\text{max}}e_i (i = 1, 2, \ldots, n)$ is used for the definition of a subdivided OD, provided that it is defined by $n$ vectors. If $r_{\text{min}}$ is non-zero, a part defined by $r_{\text{min}}e_i$ is removed. (Therefore there is an empty part near the origin of $e_i$.) These 3 parameters are fixed in a usual refinement in order to fix the size and location of ODs.

The value of each parameters specified by the serial OD number and the kind of parameters is given by the independent parameter value times the coefficient (the last number of the triplet). If $N$ parameters are constrained ($jz = N$), the triplet has to be repeated $N$ times. This expression can treat any kind of linear constraints. (But this might be too general and complicated.) It should be noted that in order to refine some parameter, it should appear in the ‘.cnd’ file. The refinement in each cycle can be controlled by the flags for these independent parameters.

### 14. Generation of indices

Usually, the reflections specified by $h_1, h_2, \ldots, h_n$ within $-h_{\text{max}} \leq h_i \leq h_{\text{max}}$ include many weak reflections which are unobserved. In addition, the number of reflections in this range rapidly increase with $h_{\text{max}}$. (Note that in icosahedral quasicrystals, it is $(2h_{\text{max}} + 1)^6$.) In order to index observed reflections, we need to generate lattice vectors with small internal space components efficiently as mentioned in section 2. In the icosahedral primitive lattice, the similarity ratio is $r^3$. On the other hand, in the face-centered icosahedral lattice, reflections with indices all even or all odd are the lattice points. In the former, we can not use $S$ or $S^2$ as a similarity transformation matrix, while in the latter, many vectors with integer indices are not allowed. Therefore we need to find an efficient way for generating indices. First we consider the face-centered lattice. Its indices can be generated efficiently by doubling all $h_i (i = 1, 2, \ldots, 6)$ within $-h_{\text{max}} \leq h_i \leq h_{\text{max}}$. Such indices fulfill the reflection condition. For each such reflection, we obtain another set of indices with $h_i$ all odd by adding $(-1,1,1,1,1,1)$. This also fulfills the condition. For such indices, we can apply any order of similarity transformation.

For the primitive lattice, we generate first the reflections for face-centered lattice by this method. After the similarity transformation, we can choose indices all even. Finally, we obtain the indices of the primitive lattice by halving such indices. In this method, we can apply any order of similarity transformation, since it is applied to the indices of face-centered lattice. In this case, a half of generated reflections is discarded but the merit of the similarity transformation exceeds this demerit.

### 15. Calculations of atom positions in 3D space

For generating the lattice point in the direct lattices of icosahedral quasicrystals, we can use a method similar to the method described above. The integral coordinates $l_i$ ($-l_{\text{max}} \leq l_i \leq l_{\text{max}}$) with respect to $d_i$ we apply similar method.

For the face-centered lattice, we have 32 centering translations. Instead of using such translations, we can choose coordinates, for which the parity of the sum $\sum_{i=1}^{6} l_i$ is even, and halve them. For such coordinates, we can apply any order of similarity transformation, since they represents the coordinates of the face-centered lattice. For the primitive lattice, we need to add (1/2,0,0,0,0,0) for the coordinates after the similarity transformation. After that they are doubled. For body centered lattice, one set of vectors are given by that of primitive lattice. Other set of vectors are obtained by adding them the centering translation $(-1,1,1,1,1,1)/2$.

Provided that all ODs in a unit cell (including equivalent ones generated from independent ODs) are given, the atom positions are calculated by qestrc. The ODs described in section 9 may not be efficient enough for the calculations of atom positions for complicated icosahedral quasicrystals. As a result, it may take long time. There are equivalent ODs, which are efficient for the calculations. Noting that the ODs located at some point $x$ is equivalent to those at the point different from it by a lattice vector $x_i$. When we choose $x_0 + x_i$ so that $x = x_0 + x_i - x_o$ has a minimum internal space component, all the ODs are located near the chosen 3D hyper-plane passing through $x_o$ in the internal space. In such a description, the lattice vectors for generating total 3D structures are only those with small internal space components. Such a lattice vector will be efficiently generated by the use of similarity transformation with large order.

### 16. Linear phason distortion and crystalline approximants

Since the program qestrc generates Cartesian coordinates in the external space when an $nD$ model is given, we can apply it to calculate crystalline approximant structures with any order. They are calculated by introducing an appropriate linear phason strain to the $nD$ model. The phason strain components can be calculated by $l$phason. This requires three unit vectors represented by $nD$ vectors, which should be on the external space under the linear phason strain. The program qestrc neglects the deformation of the ODs. This may cause some missing atoms or two atoms within a short distance which is related with the phason flip. This is usually too short as an interatomic distance. However, our experience seems to show that reasonable structures are obtained for observed crystalline approximants with larger lattice constants than 10 Å. For the necessary linear strain, refer to the literature [4]. An important point is that even if we fix the linear phason strain, different
3D intersections lead to different structures with different symmetries. In order to obtain approximants with a fixed space group, an appropriate intersection have to be used. The 3D symmetry for the approximants should be a subgroup of that of quasicrystals. The maximum point symmetry for obtained crystalline approximants agrees with the highest point symmetry in the intersection. Therefore, in order to obtain the approximate structure with high symmetry, we need to choose an intersection passing through an \( nD \) point with high symmetry. Usually, the lattice point of the \( nD \) lattice has a highest point symmetry. For example, in the case of icosahedral quasicrystals, cubic approximants obtained by the intersection passing through the origin or the body center will give the space group \( Pm\bar{3} \) or \( Im\bar{3} \) depending on the order of the approximants, since the group \( m\bar{3} \) is the subgroup of the corresponding site symmetry group \( m\bar{3}5 \) in the 6\( D \) space. On the other hand, we can show that the space group \( Pa\bar{3} \) or \( Im\bar{3} \) are obtained from the intersection passing through \((1,1,0,0,0,0,1/2)\).

After getting all the Cartesian coordinates within the unit cell of a crystalline approximant, the program \textit{iatom} can extract independent atoms under a given space group. In many cases, it will be helpful for checking the validity of obtained quasicrystal structures that we compare the real approximant structures with those derived from the final \( nD \) quasicrystal structure by introducing the linear phason strain.

17. Recent developments

In the structure shown in figure 3, if we consider only the cluster 2 consisting of up to the first nearest neighbor atoms (10 equidistant atoms from the center), most atoms are included in the cluster. In the cluster 1, 5 equidistant atoms and 2 atoms are located around the center. For convenience, we call only 5 equidistant atoms the first neighbor atoms, although the 2 atoms, shared with the first neighbor ones of the cluster 2, are the nearest neighbor of the cluster 1. This model corresponds to the situation in which we do not consider the ODs 2 in figure 4. In this case, the first neighbor atoms of the cluster 2 come from the ODs 3 and 6 while those of the cluster 1 from the ODs 7 in figure 2. On the other hand, the ODs 2 in figure 4 give the position of linking atoms. In addition, the overlap between the ODs 6 and 7 in figure 2 means that a part of the first neighbor atoms in the two clusters are shared. When we consider such small clusters, there exists no interpenetration of the clusters, except for the 2 nearest neighbor atoms of the cluster 1.

The clusters considered above are small but if we consider larger clusters, which are found in other quasicrystals, we will have many linking atoms among the gap (interstice) of the clusters. For linking atoms, it is difficult to consider a rule for putting them. If we allow the interpenetration of clusters, we may determine most atom positions and the number of interstice atoms will decrease. In fact, the idea in which quasicrystals consists of such interpenetrated clusters was introduced recently. It could reduce drastically the number of interstitial atoms [20].

In the following, therefore, we consider large clusters in the same structure, by extending the region of clusters, where the cluster consists of up to the second nearest neighbor atoms. In such a case, we will find an interpenetration of clusters.

If we apply this idea to the current model, it can be considered that the second nearest neighbor atoms of the cluster 1 are obtained from the ODs 2 and 8 in figure 2. Their most parts are overlapped with the ODs 3, 5 and 6 which are already considered. This corresponds the fact that the second nearest neighbor atoms are also, in most cases, the first nearest neighbor atoms or center of the cluster 2. Similarly, the second nearest neighbor atoms of the cluster 2 are given by ODs 4. In this model, we have no interstitial atoms like those considered above, since they agree with a part of second nearest neighbor atoms. Therefore we do not need to consider linking atoms. In this model, however, a heavy cluster interpenetration occurs.

In icosahedral Cd-Yb quasicrystals, this idea reduced the number of linking atoms down to 5% [20]. In such a model, many atoms are shared by more than two clusters and as a result, most parts of large ODs are divided by the ODs of the cluster centers into overlapped and unoverlapped parts. The subdivided ODs correspond to the atoms with similar local environment. In the same analysis, all the ODs were placed at general positions. This is similar to the subdivision shown in figure 5. As a result the cluster included in this structure has neither icosahedral nor any other high symmetry. Therefore we have one cluster at some position, other 119 clusters which have the same structure but with different orientation should appear in order to maintain total icosahedral symmetry, since the order of icosahedral group is 120. This is probable since if we consider the local environment of some cluster far from the cluster center, the site symmetry of the cluster center should be very low. In order to use such a model in the structure refinement, we need a large number of observed reflections. In this case, more than 5000 independent reflections were used, which were measured by using synchrotron radiation. It is noted that the use of good quality quasicrystals enabled us such an analysis.

The quasicrystals consisting of lower symmetric clusters seem to be a natural consequence as mentioned above. Recently it was demonstrated that the size effect in a fictitious quasicrystal also leads to such a model, where the ODs are subdivided into small fragments like ones shown in figure 5 [21].

18. Summary

We described an overview of the structure analysis of quasicrystals, using mainly the decagonal b-Ni quasicrystals as an example. The analysis is based on the assumption that they consist of similar clusters, which are situated at the vertices of assumed quasiperiodic patterns. The structures of the clusters are taken from those obtained by the analysis of crystalline approximants. The model building and the structure refinements using such information were described. In addition, the use of the program package for such an analysis, released recently, were shortly mentioned. The key input information is that for the shape, size and location of
the ODs included in a model used. The input format of such information employed in the program package was described in detail. In addition, the coordinate system for introducing the shift of atom positions in the external space was also mentioned. The efficient use of similarity transformations in the structure analysis of quasicrystals was discussed. Detailed structure analyses of quasicrystals performed so far are a few [17, 20, 22–26]. The author hopes that this package will be widely used and promote the structure analyses of quasicrystals.

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