Acta Cryst. (1998). A**54**, 199–202

Generation of Quasiperiodic Structures by Amassment Centres in Multilattices

R. Bartusch and U. Schmidt*

Department of Physics, Martin-Luther-University, 06120 Halle, Germany. E-mail: schmidt@physik.uni-halle.de

(*Received 27 February 1997; accepted 24 September 1997*)

Abstract

In this paper, a general method for generating quasiperiodic lattices is given. It is based on point multilattices which may be transformed into quasilattices in a new manner. A rounding operation allows the conversion of amassments of points in the multilattice into single points of a perfect quasicrystalline structure. Several examples are given.

1. Introduction

Intensive investigations of quasiperiodic structures began with the discovery of the icosahedral phase in rapidly solidified AlMn (Shechtman *et al.*, 1984). It is still a matter of debate which models provide the most appropriate description of quasicrystalline structures. Following the methods of crystallography, the atomic structure of a quasicrystal is described with a quasiperiodic lattice which is decorated with clusters. A popular method for generating quasiperiodic lattices and atomic structures is the cut and projection of structures that are periodic in higher-dimensional spaces. This method was successfully applied to icosahedral quasicrystals of AlCuFe type (Janot, 1993; Le Lann & Devaud 1995). Besides such perfect quasiperiodic structures, random tiling models are used (Elser, 1996). Another technique is called the generalized dual or multigrid method. The generation of the Penrose tiling and its extension to the three-dimensional Penrose tiling is determined also by this technique (de Bruin, 1981; Kramer & Neri, 1984). The success of three-dimensional Penrose tiling for icosahedral quasicrystals was only qualitative. The tiling could not explain the real density of quasicrystals. The distances between the scatterers in this tiling are about 1–2 nm. A decoration of this tiling with clusters connected by linking atoms or with two or more clusters provides an acceptable solution in this case.

The several methods to obtain quasicrystals can be classified into four categories: the projection method and the section method, which do use higher-dimensional spaces; the inflation–deflation method and the dual method, which do not use higher-dimensional spaces explicitly (Yamamoto, 1996).

A general method, which is based on point multilattices and a three-dimensional construction principle, is presented in the following. It is necessary to decorate the structures obtained by the presented method with icosahedral clusters.

2. The method

The Bravais lattices are determined by all possible periodic arrays of points in space. A multilattice or N-lattice is defined by a set of N equal or different Bravais lattices. A star of $3N$ vectors in space (or $2N$ in the plane) describes the orientation of the N lattices amongst themselves, which is characterized by a point group. Now it is useful to determine one common point of the N lattices, which is called singularity. It is possible that the multilattice contains an infinite number of common points. In this case, all common points are arranged periodically.

A simple heuristic example explains the principle of the method in one dimension. Two linear lattices are given by

$$\begin{aligned} x_n &= 2n \\ y_n &= (5^{1/2}-1)n, \quad n \in N. \end{aligned} \tag{1}$$

This multilattice (2-lattice) of x_n and y_n has a singularity at zero. It is the only common point. But there are also many places where the distance between neighbouring x_i and y_i is shorter than half of the minimal neighbour distance of the two lattices $[|x_i - y_i| < \frac{1}{2}(5^{1/2}-1)]$. Such places are called amassments and the mean value of such neighbouring points is an amassment centre. All amassment centres represent an infinite quasiperiodic sequence of points. These points form a quasiperiodic chain of the two segments $L = 5^{1/2}$ and $S = \tau$: $LSLSLLS\ldots$ [τ is the golden mean, $\tau = \frac{1}{2}(5^{1/2}+1)$].

In an N-lattice, every point of the ith lattice is defined by the vector

$$\mathbf{r}_i = n_{i1}\mathbf{x}_{i1} + n_{i2}\mathbf{x}_{i2} + n_{i3}\mathbf{x}_{i3} = \mathbf{r}_i(n_{i1}, n_{i2}, n_{i3}), \tag{2}$$

where $\mathbf{x}_{ij}$ are fundamental vectors and n_{ij} are integers. It is reasonable to choose the singularity at the origin. The orientational arrangement of the N-lattices (and their type) determines the density of amassments in the environment of the singularity and is associated with the symmetry properties of the generating structure of amassment centres. It is important to obtain structures with a high density of points and an infinite expansion.

Thus, a wide range of possibilities can be found for choosing N-lattices. An amassment always contains N points (exactly one of every lattice) and the amassment centre is characterized by the vector

$$\mathbf{h} = (1/N)\sum_{k=1}^{N} \mathbf{r}_k \tag{3}$$

and the condition

$$\max\{|\mathbf{r}_1 - \mathbf{h}|, |\mathbf{r}_2 - \mathbf{h}|, \ldots, |\mathbf{r}_N - \mathbf{h}|\} < d/4, \tag{4}$$

where d is the minimal neighbour distance of the N-lattices and $\max\{\ldots\}$ means the maximal value of the set in curly brackets. From (4), it follows that

$$u = (1/N)\sum_{k=1}^{N} |\mathbf{r}_k - \mathbf{h}| < d/4. \tag{5}$$

u is called the radius of amassment.

Passing through all points of any lattice (the ith lattice) and checking whether the point belongs to an amassment or not yields the places of the amassments. Any vector of the ith lattice [equation (2)] may be expressed by fundamental vectors of the jth lattice:

$$\mathbf{r}_i(n_{i1}, n_{i2}, n_{i3}) = \mathbf{r}_i(n'_{j1}, n'_{j2}, n'_{j3}) = n'_{j1}\mathbf{x}_{j1} + n'_{j2}\mathbf{x}_{j2} + n'_{j3}\mathbf{x}_{j3}. \tag{6}$$

In (6), the n'_{j1}, n'_{j2}, n'_{j3} are real numbers. They may be rounded to integers:

$$\text{round}(n'_{jk}) = n_{jk}. \tag{7}$$

This operation yields the neighbouring point (or the associating vector) in the jth lattice. It may be expressed by the rounding operator R:

$$R\{\mathbf{r}_i(n'_{j1}, n'_{j2}, n'_{j3})\} = \mathbf{r}_j(n_{j1}, n_{j2}, n_{j3}). \tag{8}$$

The choice of the ith lattice is insignificant – it may be the first. The vector $\mathbf{h}$ is now expressed as

$$\mathbf{h} = (1/N)\left[\mathbf{r}_1(n_{11}, n_{12}, n_{13}) + \sum_{k=2}^{N} R\{\mathbf{r}_i(n'_{k1}, n'_{k2}, n'_{k3})\}\right], \tag{9}$$

as follows from (3) with the limitations (4) and (5) being valid. The limit can be substituted by any value a less than $d/4$. The resultant structure is expressed by $\{\mathbf{h}\}_a$. Its density of points is lower than in $\{\mathbf{h}\}_{d/4}$. Investigations revealed an interesting property: the structure $\{\mathbf{h}\}_a$ equals the structure $\{\mathbf{h}\}_{a'}$ if $a' = a/k$. They differ only in scale. k is called the inflation factor and $k > 1$. $\{\mathbf{h}\}_a$ contains all points of $\{\mathbf{h}\}_{a'}$. This property may be interpreted as the inflation rule.

Fig. 1 shows a quasiperiodic structure with a 12-fold symmetry axis in the centre. It was obtained by a multilattice of three square lattices. They are arranged along the axes of symmetry of an equilateral triangle. The side length of the squares is 1. It is the minimal neighbouring

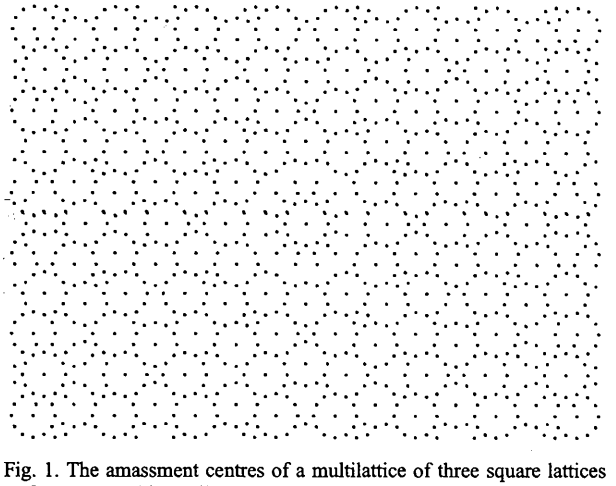

Fig. 1. The amassment centres of a multilattice of three square lattices form a 12-fold quasilattice.

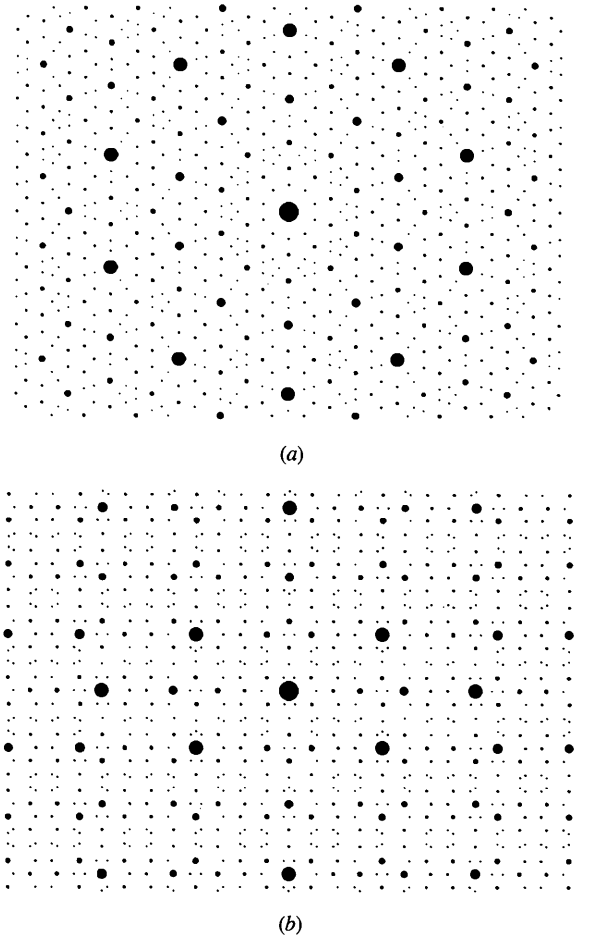

Fig. 2. Simulated fivefold (*a*) and twofold (*b*) diffraction patterns for a quasilattice of the AlMnSi type.

distance in the three lattices and $\frac{1}{4}$ is the limit in relations (4) or (5). However, the limit 0.23 was used. The inflation factor is $1/2\sin(\pi/12) \approx 1.93$.

3. Generation of icosahedral quasicrystals of AlMnSi type

It is not difficult to produce quasicrystalline phases in AlSiMn alloys by melt spinning and a following heat treatment. Our selected-area diffraction (SAD) analysis of melt-spinning ribbons of composition $Al_{77}Mn_{18}Si_5$ shows almost perfect icosahedral areas. This corresponds with the results of other authors, which are summarized in several publications (Kelton, 1993).

The simulated fivefold and twofold diffraction patterns are shown in Fig. 2. They represent the Fourier transformation of a structure, which was obtained by means of the method of amassment centres. A multilattice of five f.c.c. lattices was used. Every f.c.c. lattice was orientated along a triplet of three orthogonal twofold axes of an icosahedron. The length of the f.c.c. unit cube is 1, $d/4$ is about 0.177 and the inflation factor is τ^3 (arbitrarily chosen units). The reciprocal structure may be obtained with the same multilattice. Calculations revealed a simple relation between the limit a of the real structure $\{\mathbf{h}\}_a$ and the limit a_{rec} of the associated Fourier transformed structure $\{\mathbf{h}\}_{a_{\text{rec}}}$. The product aa_{rec} is constant and equals about $(0.09)^2$. Furthermore, for the intensities I:

$$I \propto 1/u^2. \tag{10}$$

u is called the radius of amassment [relation (5)] of the reciprocal structure. For these reasons, the patterns of Fig. 2 may be obtained with a limit of about 0.1008 and the area of the spots is proportional to $1/u^2$. The limit of the associated real structure is about 0.0804. Only discrete values of pair distances occur in this structure: 1.640 62, 1.894 43, 2.506 09, 2.679 13, 2.915 23, 3.065 25, 3.476 70 nm, ... There are many and diverse coordination types. Considering only the first five pair distances, about 500 types may be found. If the limit a increases over 0.082, the distances 0.688 19, 1.170 82, 3.141 56 nm, ... will appear additionally. This means that the density of points increases and the new scatterers cause extinctions in the diffraction pattern as well as a decrease of a_{rec}.

Most details of Fig. 2 correspond to our experimental SAD results. But the ranges of calculated real distances are rather wide: 1.640 62 → 1.10 nm, 2.915 23 → 1.96 nm, ... Therefore, the scatterers cannot be atoms. It is necessary to decorate the structure with icosahedral clusters. The Mackay icosahedron is the most suitable candidate in this case.

The possibility of stable clusters that can simulate different atoms in the Periodic Table and can serve as the building block for forming cluster materials is discussed by Khanna *et al.* (1995). The important role of sharing atoms by clusters for formation and stability of quasicrystals is demonstrated by Jeong & Steinhardt (1997) using a Penrose-like tiling that is constructed by a single prototile. The choice of basic atomic clusters by a shell-

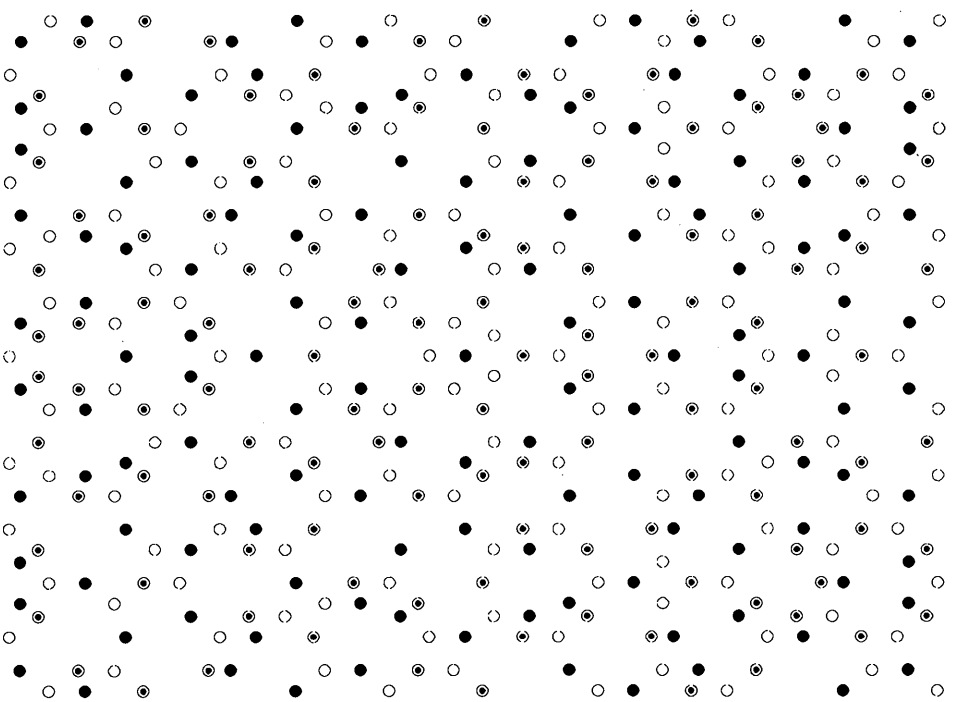

Fig. 3. First (●), second (○) and third (⊙) layers of a decagonal structure with a repetition unit of five equidistant layers.

by-shell algorithm and a demonstration of quasicrystalline growth using these clusters is reported by Borodin & Manichev (1996). Some experimental evidence was supplied by Ebert *et al.* (1996): they revealed the cluster structure of Czochralski-grown AlPdMn single quasicrystals by scanning tunnelling microscopy investigations. The surface is determined by clusters of various sizes which are formed by an elementary cluster of about 0.9 nm diameter. The pseudo-Mackay icosahedron is proposed here (Ebert *et al.*, 1996). The size of the observed elementary clusters is about of the order of the calculated distances between the scatterers in our model.

4. Generation of decagonal structures

The method of amassment centres provides many possibilities for modelling decagonal structures. Structures with up to seven different layers (repetition unit along the fivefold axis) besides structures with a quasiperiodic arrangement of the layers were found with this method up to now. One example will be given in the following.

The multilattice consists of five equal lattices. The unit cell of these lattices is an oblique rhombohedron. If the z axis is the fivefold one, the fundamental units of the first lattice are expressed by

$$\begin{pmatrix} 0.5 \\ (\tau/2)[1-1/(\tau+2)]^{1/2} \\ 1 \end{pmatrix},$$

$$\begin{pmatrix} -0.5 \\ (\tau/2)[1-1/(\tau+2)]^{1/2} \\ 1 \end{pmatrix}, \begin{pmatrix} 0 \\ 0 \\ \tau+2 \end{pmatrix}.$$

The fundamental units of the other four lattices will be obtained by rotating around the z axis (angle $2\pi/5$). The result is shown in Fig. 3. Five equidistant layers form the repetition unit of this structure. The fourth and fifth equal the third and second layers after rotation around π. The value 0.25 was chosen for the limit a. A rotation to the position of a fivefold axis other than the z axis of an imagined icosahedral structure reveals a pseudo-fivefold arrangement in this structure. In practice, the decagonal structures obtained by the method must be decorated with clusters. A pentagonal cluster-column model for AlNiCo quasicrystals is presented by Tsuda *et al.* (1996) and shows good agreement with experimentally observed HREM images.

5. Conclusions

The presented method offers the possibility of modelling experimentally observed quasicrystalline states. Solutions were also found for icosahedral phases of the AlCuFe type as well as octagonal quasicrystalline structures. In many cases, an additional decoration of the obtained structures will be required.

Only multilattices with a high density of amassments which fill the space homogeneously are important. It seems complicated to classify all structures that may be produced this way. There are good reasons for the assumption that for each structure the associated reciprocal structure also may be generated by means of this method.

References

Borodin, V. A. & Manichev, V. M. (1996). *Phys. Rev. B*, **54**, 15747–15753.
Bruijn, N. G. de (1981). *Math. Proc.* A**84**, 39–66.
Ebert, P., Feuerbacher, M., Tamura, N., Wollgarten, M. & Urban, K. (1996). *Phys. Rev. Lett.* **77**, 3827–3830.
Elser, V. (1996). *Philos. Mag.* B73, 641–656.
Janot, C. (1993). *J. Non-Cryst. Solids*, **156–158**, 852–864.
Jeong, H. & Steinhardt, P. J. (1997). *Phys. Rev. B*, **55**, 3520–3532.
Kelton, K. F. (1993). *Int. Mater. Rev.* **38**, 105–137.
Khanna, S. N., Janot, C. & Cyrot-Lackmann, F. (1995). *Clusters and Aggregates: Application to Quasicrystals*, pp. 401–407. Singapore: World Scientific.
Kramer, P. & Neri, R. (1984). *Acta Cryst.* A**40**, 580–587.
Le Lann, A. & Devaud, J. (1995). *J. Phys. (Paris) I*, **5**, 129–151.
Shechtman, D., Blech, I., Gratias, D. & Cahn, J. W. (1984). *Phys. Rev. Lett.* **53**, 1951–1953.
Tsuda, K., Nishida, Y., Saitoh, K., Tanaka, M., Tsai, A. P., Inoue, A. & Masumoto, T. (1996). *Philos. Mag.* A**74**, 697–708.
Yamamoto, A. (1996). *Acta Cryst.* A**52**, 509–560.