Acta Crystallographica Section A
Foundations of Crystallography
ISSN 0108-7673

Received 7 July 2001
Accepted 5 October 2001

Algebraic determination of generating functions for coordination sequences in crystal structures

Jean-Guillaume Eon

Instituto de Química, Universidade Federal do Rio de Janeiro, A-632, Cidade Universitária, 21945-970 Ilha do Fundão, Rio de Janeiro, Brazil. Correspondence e-mail: jgeon@iq.ufrj.br

Given a connected crystalline structure, the set of paths and cycles of the quotient graph of its bond net is embedded into a commutative ring structure. Multiplication combines walks to build up geodesics of the net whereas addition stands symbolically for enumerating a collection of walks. Topological criteria are used to define zero divisors which enable the development of an algebraic generator into a combination of geodesics that are in a one-to-one correspondence with the vertices of the net. A ring mapping gives the generating function of the coordination sequence in the net.

1. Introduction

Coordination sequences in nets are defined as the sequences of numbers n_k of neighbors in the kth coordination shell of some fixed vertex of the graph. In a recent paper, Grosse-Kunstleve *et al.* (1996) showed that coordination sequences in zeolites and some other simple crystal structures can be described by generating functions $GF(x) = \sum_k n_k x^k$ which are rational fractions in the ring $\mathbf{R}[\mathbf{x}]$, that is, the generating function is of the form

$$GF(x) = P(x)/Q(x),$$

where $P(x)$ and $Q(x)$ are polynomials of the real variable x. Explicit functions were obtained by direct computation of the first terms of the series, assuming that extrapolation to the whole sequence was licit.

More recent works (Conway & Sloane, 1997; Bacher *et al.*, 1999) have shown that coordination sequences can be obtained rigorously in some simple root lattices. The argument used in these papers was of a geometrical nature; only partial results are available, however. The simple root lattice A_3, for instance, describes only one of the two vertices contained in the primitive cell of the diamond net.

The aim of this work is to report an algebraic method for calculation of generating functions. The analysis is performed in the framework of the quotient graph of the net as defined by Chung *et al.* (1984). First, the formalism is developed for minimal nets (Beukemann & Klee, 1992) through their integral embeddings (Eon, 1999); then it is extended to other cases.

2. Geodesics in integral embeddings

The nomenclature used throughout the paper is the same as that used by Eon (1999). For convenience, this section begins with a summing up of the main concepts leading to the definition of integral embeddings.

The quotient graph of a net is the finite graph G obtained by mapping translationally equivalent edges and translationally equivalent vertices of the net on some edge and vertex of G and respecting the adjacency relations. Let f be the corresponding mapping. The integral embedding is defined as a special embedding of the minimal net associated with the quotient graph. It is obtained by somehow reversing the mapping used for definition of the quotient graph. After the graph with q edges e_i ($i = 1, \ldots, q$) has been oriented, the embedding is generated by mapping the edges onto an orthonormal basis $\mathbf{e}_i$ ($i = 1, \ldots, q$) of the Euclidian space $\mathbf{E}^q$. Some point O of the embedding is chosen as the origin; the lines in the embedding are then equipollent to unit vectors of the basis, and the points $M = \sum_i t_i \mathbf{e}_i$ have integer coordinates.

Let g_M be a geodesic of the embedding, *i.e.* a shortest path, from the origin O to the point M, and $|g_M|$ be the length of the geodesic, that is, the number of lines in the respective path. The generating function of the coordination sequence associated with the origin can be obtained by summation of an infinite series over all the points of the embedding,

$$GF(x) = \sum_M x^{|g_M|}. \quad (1)$$

As a path, the geodesic is an alternate sequence of points P_i and lines l_i. In order to characterize geodesics from the viewpoint of the quotient graph, we define the walk $f(g_M)$ in G, corresponding to the alternate sequence of vertices $f(P_i)$ and (oriented) edges $f(l_i)$, and consider the mapping h of the geodesic into the 1-chains group of G defined by

$$h(g_M) = \sum_i f(l_i).$$

Clearly, translationally equivalent lines traversed in opposite directions along the geodesic will cancel out by pair in $h(g_M)$. On the other hand, since the lines l_i are equipollent to basis vectors of $\mathbf{E}^q$, the coefficient of the edge e_i after mapping by h is exactly equal to the coordinate of the point M along the axis associated with the vector $\mathbf{e}_i$, yielding

$$h(g_M) = \sum_i t_i e_i.$$

On geometrical grounds, the coordinate t_i corresponds clearly to the minimum number of times a line mapping the edge e_i of G must be crossed from O to M, so that

$$|f(g_M)| = |g_M| \geq \sum_i |t_i| \equiv |h(g_M)|,$$

where the summation index runs over all the edges of the graph, and the length of the 1-chain has been defined in the right-hand part of the equation. Let δ_M be the difference,

$$|g_M| = |f(g_M)| = |h(g_M)| + \delta_M.$$

The presence of this last term in the topological distance, if needed, means that some translationally equivalent lines must be crossed at least once in each direction along the geodesic. As an example, look at the geodesic g from O_1 to O_3 in the ReO_3 net represented in Fig. 1. Although not an integral embedding, this structure contains the adequate features for illustrating the discussion. The points P_i are all mapped on the same vertex M of the quotient graph drawn in Fig. 2; points O_1, O_2 and O_3 are mapped on vertices A, B and A, respectively. The lines l_1, l_2, l_3 and l_4 are mapped on the edges $-e_1$, e_3, $-e_4$ and e_1, respectively; the first and last lines of g, namely l_1 and l_4, are translationally equivalent but traversed in opposite directions. The previous definitions give

$$\begin{aligned} g &= [O_1\, l_1\, P_1\, l_2\, O_2\, l_3\, P_2\, l_4\, O_3], \\ f(g) &= [A - e_1 M e_3 B - e_4 M e_1 A], \\ h(g) &= e_3 - e_4, \\ \delta &= |f(g)| - |h(g)| = 4 - 2 = 2. \end{aligned}$$

In this case, the 1-chain $h(g)$ defines a cycle of length 2 which does not contain the origin of the walk $f(g)$. More generally, we call the *support* of the 1-chain, and by extension, of the geodesic, the directed subgraph consisting of the edges e_i corresponding to non-zero coordinates t_i together with the incident vertices, the orientation of the edge in the support being taken according to the sign of the coordinate. The support is not necessarily a connected graph. In the previous example, the support of g is the directed 2-cycle consisting of the two vertices M and B, and the oriented edges e_3 and $-e_4$.

Since the 1-chain $h(g_M)$ has been obtained from a walk in G between two of its vertices, it can be decomposed, generally not in a unique way, into a path p between these two vertices (unless they are translationally equivalent) and a set of possibly disjoint and n_k times repeated cycles C_k of G covering the respective support, and respecting the orientation of its edges,

$$h(g_M) = p + \sum_k n_k C_k. \tag{2}$$

We will call *g-chains* such 1-chains of G mapped by a geodesic.

Reciprocally, let a be any 1-chain of the above form, *i.e.* obtained by adding a path and some cycles of G in which each edge is always traversed in the same direction. By construction of integral embeddings, the 1-chain a defines the coordinates t_i of some point M in the embedding and satisfies

$$a = \sum_i t_i e_i = h(g_M)$$

for any geodesic g_M; a is thus a g-chain.

Let S be the support of a, and (S) be the smallest connected subgraph of G containing S. It is clear that one can build a walk in (S) between the extremities of the chosen path by using the 1-chain a together with the edges of (S) which do not belong to S, each being traversed in both opposite directions. The resulting walk maps a geodesic g_M in the embedding, since, by construction, it is not possible to find a shorter path. The value of δ_M for the respective geodesic can thus be obtained directly from the quotient graph as equal to the minimum number of edges that must be added to turn the chain into a connected walk starting from the vertex mapped by the origin. It must be noticed that this value depends only on the support of the geodesic, and not on the chosen point M.

3. The chains ring

Our main concern in this and the following sections will be to generate exactly one g-chain per point of the integral embedding, by composing paths and cycles of the quotient

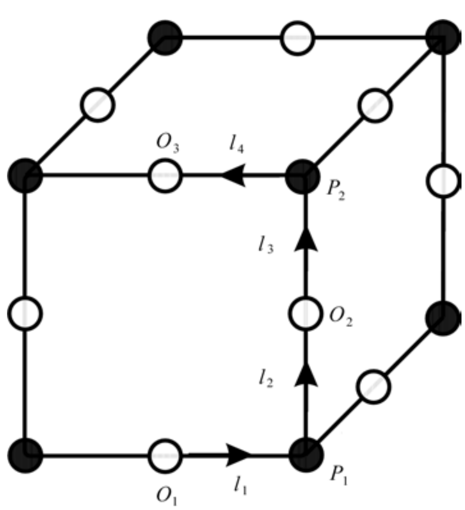

Figure 1
ReO_3 net with a geodesic between O_1 and O_3 marked by arrows.

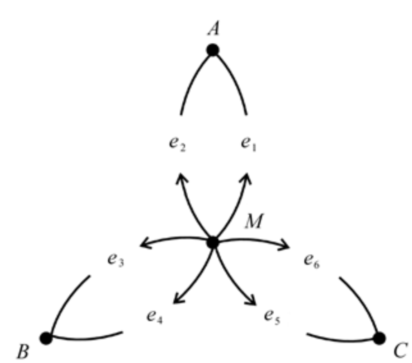

Figure 2
Labelled quotient graph of the ReO_3 net.

graph. This can be performed by embedding g-chains in a commutative ring structure.

We recall that a ring is defined in elementary algebra (Lang, 1995) as a set $\mathbf{R}$, together with two binary operations, addition and multiplication, which satisfy the following conditions:

(i) ($\mathbf{R}$, +) is an abelian group;

(ii) multiplication is associative and owns a unit in $\mathbf{R}$;

(iii) multiplication is distributive relative to addition.

As it is more convenient in the following to use an exponential notation instead of an additive one, we introduce formally a new set of elements, called *x-chains*, that are in a one-to-one correspondence with g-chains,

$$\mathbf{x}^G = \{x^a | a : g\text{-chain of } G\}.$$

Next, we consider the set $\mathbf{Z}[\mathbf{x}^G]$ consisting of all possible, finite or infinite, formal linear combinations of x-chains with integer coefficients. Alternatively, an element of $\mathbf{Z}[\mathbf{x}^G]$ may be thought of as a mapping of the points of the embedding on the set of integers, *i.e.* a subset of weighted points. Elements of this set will be referred to as *chains patterns*. Two especially important elements are the zero combination, denoted 0, for which the coefficient of each x-chain is null, and the unit combination, denoted 1, corresponding to the null g-chain ($1 \equiv x^0$). The sum of two elements in $\mathbf{Z}[\mathbf{x}^G]$ is naturally defined as the combination obtained after adding the coefficients of the respective x-chains,

$$\sum_a m_a x^a + \sum_a n_a x^a = \sum_a (m_a + n_a) x^a \quad (m_a, n_a \in \mathbf{Z}).$$

It is immediately seen from this definition that ($\mathbf{Z}[\mathbf{x}^G]$, +) is an abelian group with the zero combination as an additive unit.

Multiplication in $\mathbf{Z}[\mathbf{x}^G]$ is presently defined to allow the unique combination of paths and cycles into g-chains. Let a and b be two g-chains, one of which at least is only composed of cycles of G. The product in $\mathbf{Z}[\mathbf{x}^G]$ of x^a and x^b is defined by comparison of the supports of the g-chains. We will say that two g-chains a and b of G have *compatible supports* S_a and S_b whenever the edges which are common to both digraphs have the same orientation; it is clear then that $a + b$ is a g-chain, the support of which, S_{a+b}, is the union of both supports, *i.e.* the smallest digraph containing both supports S_a and S_b, and moreover satisfies a condition of additivity of lengths,

$$S_{a+b} = S_a \cup S_b,$$
$$|a + b| = |a| + |b|.$$

The product of the two x-chains, denoted with the asterisk symbol, is the x-chain defined by the exponential property,

$$x^a * x^b = x^{a+b} = x^{b+a}.$$

In this case, the additivity condition shows that the product of x-chains amounts to composing a geodesic by putting two geodesics together, without unnecessary wandering about the lines of the embedding. Otherwise, when two g-chains have incompatible supports, or when both of them contain a path contribution, we set the product to be null, which discards the combination. The product of any two chains patterns is formally defined by setting bilinearity of the product

$$mx^a * nx^b = mnx^{a+b},$$

and distributivity of multiplication relative to addition. This product is clearly commutative.

Associativity of multiplication will be verified in the case of three x-chains, two of them at least being composed only of cycles; the supports S_a, S_b and S_c are first assumed to be compatible by pairs. The property is a straightforward consequence of associativity of the union of compatible supports and associativity of 1-chains addition,

$$\begin{aligned} S_{a+(b+c)} &= S_a \cup S_{b+c} = S_a \cup (S_b \cup S_c) = (S_a \cup S_b) \cup S_c \\ &= S_{(a+b)+c}, \\ x^a * (x^b * x^c) &= x^a * (x^{b+c}) = x^{a+(b+c)} = x^{(a+b)+c} \\ &= (x^{a+b}) * x^c = (x^a * x^b) * x^c. \end{aligned}$$

If the supports are not compatible by pairs, or at least two x-chains contain a path contribution, it is easily verified that the ternary product is null, whatever the calculation sequence may be. Thus, multiplication of x-chains is associative. Extension to the product of chains patterns is a natural consequence of bilinearity, distributivity of multiplication over addition and associativity of x-chains multiplication.

By definition, the support of the null chain is empty and, as such, it is compatible with the support of any other g-chain. The unit combination is thus the unit for multiplication,

$$\begin{aligned} 1 * \sum_a m_a x^a &= x^0 * \sum_a m_a x^a = \sum_a m_a (x^0 * x^a) = \sum_a m_a x^{a+0} \\ &= \sum_a m_a x^a. \end{aligned}$$

This shows that ($\mathbf{Z}[\mathbf{x}^G]$, +, $*$) is a ring.

The main property of this structure is the existence of zero divisors. No cycle of G, for example, can be walked over in both orientations in a geodesic. Since the respective supports are not compatible, this observation translates now to the ring formalism,

$$x^a * x^{-a} = 0. \quad (3)$$

It is clear then that the chains patterns ring is not a structural extension of the known 1-chains group of graph theory. The exponential notation helps to avoid confusion.

We will now illustrate the whole method of determination of generating functions in the simplest case where G has a single vertex and one loop a. The integral embedding corresponds to a linear lattice with one point and one line per unit cell, with divalent coordination. Following methods used in combinatorial analysis (Jordan, 1958), we define the generator F in $\mathbf{Z}[\mathbf{x}^G]$,

$$F = \left(1 + \sum_{n>0} x^{na}\right) * \left(1 + \sum_{m>0} x^{-ma}\right).$$

This product describes every possible choice of n loops run in the positive direction with m loops run in the opposite direction. The expression is developed with the help of (3), in particular, to obtain

$$F = 1 + \sum_{n>0} x^{na} + \sum_{m>0} x^{-ma}.$$

It appears that F corresponds to the combination of all x-chains of G with unit coefficient. The generator is mapped on the generating function of the coordination sequence, $GF(x)$, of the linear embedding by substituting the symbol of the loops in x-chains by their length; here $|a| = |-a| = 1$. Remembering that, in $\mathbf{R}[\mathbf{x}]$,

$$\sum_{n\geq 0} x^n = 1/(1-x),$$

we have

$$\begin{aligned} GF(x) &= 1 + \sum_{n>0} x^n + \sum_{m>0} x^m \\ &= 1 + 2x/(1-x) \\ &= (1+x)/(1-x). \end{aligned}$$

4. The generator inverse

More generally, for a graph G with a set of paths p_j from some vertex chosen as an origin to all other vertices and a set of oriented cycles C_k, the generator of x-chains in $\mathbf{Z}[\mathbf{x}^G]$ is given by

$$F = \left(1 + \sum_j x^{p_j}\right) * \prod_k \left(1 + \sum_{n>0} x^{nC_k}\right).$$

In order to avoid manipulating infinite sums, we observe that each factor of F owns an inverse in $\mathbf{Z}[\mathbf{x}^G]$, since we have

$$\begin{aligned} \left(1 - x^{C_k}\right) * \left(1 + \sum_{n>0} x^{nC_k}\right) &= 1, \\ \left(1 - \sum_j x^{p_j}\right) * \left(1 + \sum_j x^{p_j}\right) &= 1. \end{aligned}$$

This allows the inverse F^{-1} of the generator to be defined,

$$F^{-1} = \left(1 - \sum_j x^{p_j}\right) * \prod_k (1 - x^{C_k}). \quad (4)$$

From comparison of the expressions of the generator and its inverse, it follows that inversion is truly a practical device which allows one to work on finite products during the calculation. Infinite summations on cycles are turned on again at the end of the development by back inversion of the result.

5. The graphite net

Geodesics between two vertices of a net are generally not unique, and branching off appears along the path. However, it is always possible to privilege some branches and set the other ones to zero. This is an arbitrary choice that no general law can express; it needs only to be coherent. The concept is best illustrated by calculating the generating function for the coordination sequence of the graphite net, whose quotient graph $G = K_2^3$ is represented in Fig. 3. As was discussed by Eon (1999), the graphite structure is the orthogonal projection of the integral embedding of the minimal net associated with the graph K_2^3. The projection is along the co-cycle space, onto the cycle space, and does not affect topological properties. In particular, the coordination sequences are the same in both embeddings. This allows calculations to be performed in the integral embedding.

If we choose vertex A as the origin, the three different paths are simply the three edges e_1, e_2 and e_3 of the graph K_2^3. The six oriented cycles are given by

$$a_{ij} = e_i - e_j, \qquad i \neq j, \quad i,j = 1, \ldots, 3.$$

The multiplication laws in $\mathbf{Z}[\mathbf{x}^G]$ give

$$\begin{aligned} x^{a_{ij}} x^{a_{jk}} &= x^{a_{ji}} x^{a_{kj}} = 0, \\ x^{e_j} x^{a_{ij}} &= 0, \end{aligned}$$

where the asterisk symbol for multiplication was omitted for practical calculations.

The inverse of the generator is thus

$$\begin{aligned} F^{-1} &= (1 - x^{e_1} - x^{e_2} - x^{e_3})(1 - x^{a_{12}} - x^{a_{21}})(1 - x^{a_{23}} - x^{a_{32}}) \\ &\quad \times (1 - x^{a_{31}} - x^{a_{13}}) \\ &= 1 - \sum_i x^{e_i} - \sum_{i,j} x^{a_{ij}} \\ &\quad + \sum_{ij} x^{e_i} x^{a_{ij}} + \sum_{ijk} x^{e_i} x^{a_{jk}} + \sum_{ijk} x^{a_{ij}} x^{a_{ik}} + \sum_{ijk} x^{a_{ij}} x^{a_{kj}} \\ &\quad - \sum_{ijk} x^{e_i} x^{a_{ij}} x^{a_{ik}} - \sum_{ijk} x^{e_i} x^{a_{ij}} x^{a_{kj}}. \end{aligned}$$

It appears now that not all the terms of this development do represent different g-chains. For instance, $x^{e_1} x^{a_{23}}$ and $x^{e_2} x^{a_{13}}$ map two geodesics between the origin and the point represented by the chain $e_1 + e_2 - e_3$.

Let us call these *parallel geodesics* and write

$$e_1 + a_{23} \simeq e_2 + a_{13}.$$

In order to avoid such repetitions, redundant products are set to zero:

$$x^{e_1} x^{a_{32}} = x^{e_2} x^{a_{13}} = x^{e_3} x^{a_{21}} = 0.$$

It must be kept in mind that whole sets of geodesics, such as those represented by the chains pattern

$$\sum_{n>0} x^{e_2} x^{na_{13}},$$

are then lost when inverting F^{-1} back to the generator. There is certainly no loss of points of the embedding, however, since these same points are mapped by the equivalent non-null chains pattern,

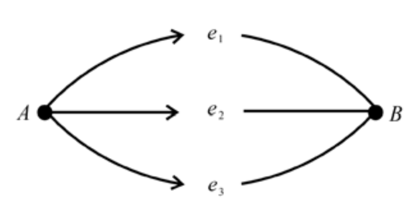

Figure 3
Labelled quotient graph of the graphite net.

$$\sum_{n>0} x^{e_1} x^{(n-1)a_{13}} x^{a_{23}}.$$

Consequently, of course, products such as $x^{e_1}x^{a_{12}}x^{a_{32}}$ have to be cancelled out to obtain the final development of F^{-1}. This amounts to avoid branching along the privileged geodesic.

Back inversion and mapping on the generating function of the coordination sequence of the graphite net are advantageously performed in a single step. It is sufficient, notwithstanding changes in sign, to substitute x-cycles by the sum of geometric series in the expression of the generator inverse. For example, the chains patterns

$$1, \quad x^{e_1}+x^{e_2}+x^{e_3}, \quad x^{a_{13}} \quad \text{and} \quad x^{e_1}x^{a_{12}}x^{a_{13}}$$

are mapped, respectively, in $\mathbf{R}[\mathbf{x}]$ upon

$$1, \quad 3x, \quad x^2/(1-x^2) \quad \text{and} \quad x[x^2/(1-x^2)]^2,$$

where account has been taken of the lengths

$$|e_i| = 1 \quad \text{and} \quad |a_{ij}| = 2.$$

Addition of all corresponding terms from the generator inverse gives the generating function

$$GF(x) = 1 + 3x + 6x^2/(1-x^2) + 6x^4/(1-x^2)^2 + 9x^3/(1-x^2) + 6x^5/(1-x^2)^2,$$

which simplifies to

$$GF(x) = (1+x+x^2)/(1-x)^2.$$

6. The ReO_3 net

The next example illustrates the calculation method in case the support does not define a connected walk. The task we propose is to obtain the coordination sequence of the oxygen atom, say O_1, in the ReO_3 structure, already described in Figs. 1 and 2.

The three independent cycles of the quotient graph are defined as

$$a = e_1 - e_2,$$
$$b = e_3 - e_4,$$
$$c = e_5 - e_6.$$

The cycle part of the generator inverse is given by the product

$$F_c^{-1} \equiv (1 - x^a - x^{-a})(1 - x^b - x^{-b})(1 - x^c - x^{-c}).$$

It is clear that any cycle combination which does not contain one of the two cycles a or $-a$ cannot map a connected walk between two oxygen atoms belonging to the same point lattice A. To this end, one must adjoin one more edge, say e_1, that has to be crossed once in both directions, so that $\delta_A = 2$ for the respective g-chains.

Accordingly, the cycle part of the generator inverse is divided into contributions from the supports defining connected and unconnected walks, respectively,

$$F_c^{-1} = (-x^a - x^{-a})(1 - x^b - x^{-b})(1 - x^c - x^{-c}) + (1 - x^b - x^{-b})(1 - x^c - x^{-c}).$$

The first term does not need to be developed since all products correspond to compatible supports. Noting that the cycles have a topological length of 2, we obtain the fraction

$$[2x^2/(1-x^2)][1 + 2x^2/(1-x^2)]^2. \tag{5}$$

The second term has to be developed and each non-trivial product corrected for connectivity by x^{δ_A}, which yields

$$1 + 4x^4/(1-x^2) + 4x^6/(1-x^2)^2. \tag{6}$$

Two paths ($p_1 = -e_1$ and $p_2 = -e_2$) lead from vertex A to M and eight equivalent paths, such as $p_3 = e_3 - e_1$, lead from A to the other two oxygen vertices. There is no parallel geodesic in this case so that the calculation can be performed separately for each path, only taking account of compatibility conditions.

The respective parts of the generator inverse are now written

$$x^{-e_1}F_c^{-1} = x^{-e_1}(1 + x^{-a})(1 + x^b + x^{-b})(1 - x^c - x^{-c}),$$
$$x^{e_3-e_1}F_c^{-1} = x^{e_3-e_1}(1 + x^{-a})(1 + x^b)(1 - x^c - x^{-c}).$$

After weighting by the number of paths, these products are mapped upon the following functions in $\mathbf{R}[\mathbf{x}]$,

$$2x[1 + x^2/(1-x^2)][1 + 2x^2/(1-x^2)]^2, \tag{7}$$
$$8x^2[1 + x^2/(1-x^2)]^2[1 + 2x^2/(1-x^2)]. \tag{8}$$

Summing up all contributions from cycles and paths from (5) to (8) yields the generator function

$$GF(x) = (1 + 2x + 7x^2 + 4x^3 + 19x^4 + 2x^5 - 3x^6)/(1-x^2)^3.$$

7. The common case

The examples that have been dealt with in the above sections, as well as any other integral embedding, are particular in the sense that all cycles of the quotient graph map a non-null vector of the translation group in the corresponding crystal structure. They are n-dimensional structures, with as many dimensions as there are independent cycles in the quotient graph. In the common case, some cycles of the quotient graph define rings, in the topological sense, of the crystal structure. This will affect the calculation of coordination sequences in two correlated aspects.

First, the x-chain that corresponds to the ring must obviously be set equal to zero. Then, as a consequence of the presence of rings, short cuts will be created in the net, and new zero divisors have to be introduced in the algebraic structure for eliminating the longest paths that are no longer geodesics. In the case of rings with an even number of edges, both halves of the ring also become parallel geodesics between two opposite points. The following section gives a simple illustration of these concepts.

8. The $(9^3, 3.9^2)$ net

The title net, drawn in Fig. 4, is obtained by projection of the (10, 3a) net (Wells, 1977) along any rotation axis of order three. The two nets have the same quotient graph K_4 represented in Fig. 5.

The cycles of K_4 can be named after the region they enclose in the plane of the figure; their positive orientation is defined counterclockwise. This gives the seven cycles, a, b, c, ab, ac, bc and abc, to which we must adjoin the opposite ones. We have, for example,

$$\begin{aligned} a &= e_1 + e_4 - e_2, \\ ab &= e_1 + e_4 + e_5 - e_3, \\ abc &= e_4 + e_5 + e_6. \end{aligned}$$

In all, there are eight cycles of length 3 and six of length 4. The cycle abc is chosen as the ring which defines the $(9^3, 3.9^2)$ net as it has been represented in the figure. We calculate the generating function for the coordination sequence of points A in this net.

To begin with, we note that $-e_6$ is a short-cut to the path $e_4 + e_5$ (we note $e_4 + e_5 > -e_6$), so that any x-chain corresponding to a g-chain containing both edges, such as x^{ab}, must be set equal to zero. This occurs similarly for the other cycles of length 4, leaving only the set S of the three cycles a, b, c and their opposites.

The multiplication laws apply to S as follows:

$$x^a x^b = x^b x^c = x^a x^c = x^{-a} x^{-b} = x^{-b} x^{-c} = x^{-a} x^{-c} = 0,$$

and restrain the development of the cycle part of the generator inverse to the six cycles of S and the six double products for each cycle and the opposite of the other two,

$$\begin{aligned} F_c^{-1} &\equiv (1 - x^a - x^{-a})(1 - x^b - x^{-b})(1 - x^c - x^{-c}) \\ &= 1 - \sum_{u \in S} x^u + \sum_{(u,v) \in S^2,\, u \neq v} x^u x^{-v}. \end{aligned}$$

Since all cycles of S contain the vertex A, there is no need for a connectivity correction ($\delta = 0$) and the following sum is mapped in $\mathbf{R}[\mathbf{x}]$:

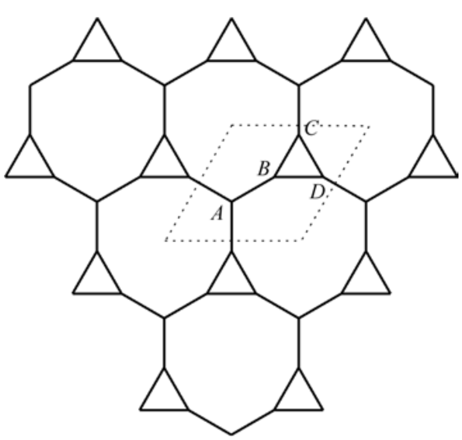

Figure 4
The $(9^3, 3.9^2)$ net.

$$1 + 6x^3/(1 - x^3) + 6x^6/(1 - x^3)^2. \tag{9}$$

One path of length 1, $p_1 = e_1$ and two paths of length 2, $p_2 = e_2 - e_4$ and $p_3 = e_3 + e_6$, lead from vertex A to vertex B. Paths of length 3, such as $e_2 + e_5 + e_6$, have short cuts among paths of length 2 (p_2 in this case), and need not be considered.

The contribution to the generator inverse from the first path is written as

$$\begin{aligned} x^{e_1} F_c^{-1} &= x^{e_1} \left[1 - \sum_{u \in S} x^u + \sum_{(u,v) \in S^2,\, u \neq v} x^u x^{-v} \right] \\ &= x^{e_1} [1 - x^a - x^b - x^{-b} - x^{-c} + x^a x^{-b} \\ &\quad + x^b x^{-c} + x^a x^{-c}], \end{aligned}$$

which is mapped in $\mathbf{R}[\mathbf{x}]$ on

$$x + 4x^4/(1 - x^3) + 3x^7/(1 - x^3)^2. \tag{10}$$

Although the other two paths, p_2 and p_3, are symmetrical relatively to the automorphisms group of K_4 that leave the ring invariant, their contributions to the generator inverse must be dealt with one by one in order to account for parallel geodesic. We have, for p_2,

$$x^{e_2 - e_4} [1 - x^{-a} - x^b - x^c - x^{-c} + x^b x^{-c} + x^c x^{-a} + x^{-a} x^b].$$

However, the walk $p_2 - c$ is not a geodesic since $e_1 + b$ is a short cut,

$$\begin{aligned} p_2 - c = (e_2 - e_4) - (e_6 + e_3 - e_1) &> e_2 + e_5 - e_3 + e_1 \\ &= b + e_1. \end{aligned}$$

We must set

$$x^{p_2} x^{-c} = 0$$

and consequently

$$x^{p_2} x^{-c} x^b = 0.$$

The contribution of the path reduces to

$$x^{e_2 - e_4} [1 - x^{-a} - x^b - x^c + x^c x^{-a} + x^{-a} x^b].$$

For p_3, we have

$$x^{e_3 + e_6} [1 - x^a - x^{-a} - x^{-b} - x^c + x^a x^{-b} + x^c x^{-a} + x^c x^{-b}].$$

It can be seen as above that

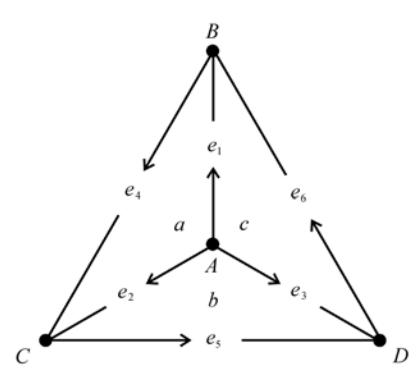

Figure 5
Labelled quotient graph of the $(9^3, 3.9^2)$ net.

$$p_3 + a > e_1 - b,$$
$$p_3 - a \simeq c + p_2.$$

The contribution of this path is thus reduced by the presence of both short cut and parallelism to

$$x^{e_3+e_6}[1 - x^{-b} - x^c + x^c x^{-b}].$$

The contributions from both paths of length 2 are mapped in $\mathbf{R}[\mathbf{x}]$ on the function

$$2x^2 + 5x^5/(1 - x^3) + 3x^8/(1 - x^3)^2. \quad (11)$$

The generating function for the $(9^3, 3.9^2)$ net is given by adding expressions (9)–(11):

$$GF(x) = (1 + 3x + 6x^2 + 4x^3 + 6x^4 + 3x^5 + x^6)/(1 - x^3)^2.$$

9. Summary

A characteristic ring structure has been associated with each net, allowing the formal definition of an algebraic generator of the geodesics. The correct definition of zero divisors in the ring is the key for determining the coordination sequences of the net; it allows the development of the generator into an expression that is a one-to-one mapping of the vertices of the net by geodesics. Zero divisors are obtained by applying three basic topological properties of the chains, which were described as compatibility, parallelism and short cuts. Although very simple examples have been analyzed in this work, it should be clear that parallelism brings up some arbitrary choices and needs to be examined with special care. Branching appears whenever the support of a *g*-chain can be decomposed in several ways into a path and some cycles of the quotient graph. All respective products but one should be set to zero. In order to keep internal coherence, however, the analysis of parallelism should be carried by a growing number of cycles.

The following steps are then suggested as a basis for an algorithm to find the generating function for the coordination sequence of a given crystal structure:

(i) Determine the quotient graph of the net, identifying all the cycles that correspond to topological rings of the structure; write short-cut relationships.

(ii) Write the generator inverse, discarding the paths and cycles that do not represent geodesics or become parallel geodesics because of short-cuts.

(iii) Define zero divisors applying compatibility conditions. Develop accordingly the cycle part of the generator inverse while keeping register of the points the corresponding geodesics reach; points already attained define parallel geodesics and new zero divisors must be stored. Complete the development of the generator inverse by including the path part and following the same rule to define parallel geodesics.

(iv) Map each term of the development on the corresponding fraction in the ring of polynomials, correcting for connectivity after analysis of the respective support, and sum up to obtain the generating function.

The author is grateful to Professor W. E. Klee (Karlsruhe, Germany) who first introduced him to the question of coordination sequences. This work was supported by CNPq, Conselho Nacional de Desenvolvimento Científico e Tecnológico of Brazil.

References

Bacher, R., De la Harpe, P. & Venkov, B. (1999). *Ann. Inst. Fourier*, **49**(3), 727–762.

Beukemann, A. & Klee, W. E. (1992). *Z. Kristallogr.* **201**, 37–51.

Chung, S. J., Hahn, Th. & Klee, W. E. (1984). *Acta Cryst.* A**40**, 42–50.

Conway, J. H. & Sloane, N. J. A. (1997). *Proc. R. Soc. London Ser. A*, **453**, 2369–2389.

Eon, J.-G. (1999). *J. Solid State Chem.* **147**, 429–437.

Grosse-Kunstleve, R. W., Brunner, G. O. & Sloane, N. J. A. (1996). *Acta Cryst.* A**52**, 879–889.

Jordan, J. (1958). *An Introduction to Combinatorial Analysis.* New York: Wiley.

Lang, S. (1995). *Algebra.* Reading, MA: Addison-Wesley.

Wells, A. F. (1977). *Three-Dimensional Nets and Polyhedra.* New York: Wiley.