regions than others. The centroid of the structure-factor probability distribution is obtained, in this case, by taking the Fourier transform of the expected electron-density function. In other terms, each atom of a molecular replacement model would be smeared over its distribution of possible positions. It is assumed that there is a sufficient number of independent contributions to the difference in the structure factors, so that the central limit theorem applies and the probability distribution is a Gaussian about the centroid estimate.

For a model of a crystal structure, it is preferable to consider the average effect of a specific set of errors on a set of structure factors, in other words to consider the reciprocal-space vector as the random variable. The probability distributions underlying the differences between the model and the true structure enter through the frequencies of the errors over all the atoms. Essentially the same probability distributions of structure factors arise as in the previous case, because of the symmetry between real and reciprocal space in the Fourier transform.

Considered in terms of normalized structure factors, all sources of error have the same effect, which can be summarized in a single parameter, σ_E. This parameter plays the same role in the probability distributions as σ_A in the distributions of Srinivasan & Ramachandran (1965). Therefore, the methods suggested previously to estimate phase probabilities and to calculate electron-density maps (Read, 1986) are still valid. However, the interpretation of the parameter σ_E is different. In particular, the variation of σ_E with resolution cannot be attributed entirely to coordinate error. Methods such as the Luzzati (1952) plot and the σ_A plot (Read, 1986) to estimate coordinate error will therefore suffer from a number of sources of systematic error.

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On Inclined Cubic Sublattices of Cubic Lattices

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Abstract

Cubic sublattices of cubic lattices are described which share only some of the point-symmetry operations with the original lattices; the common operations form the point groups $\bar{3}m$, $\bar{3}$, $4/m$, $2/m$ or $\bar{1}$. Some properties of these sublattices, including the centred ones, are shown and tentative terminology, notation and classification are introduced. All the different types of inclined primitive cubic sublattices L_n for

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$n = 3, 5, \ldots, 31, 63, 81$ (n being the length of the unit cube edge) of L_1 are listed. Several remarks and hypotheses concerning theoretical crystallography and number theory are presented.

1. Introduction

For physical purposes, namely in treating the phonons in cubic crystals in terms of gradually enlarged unit cells (Frei & Deus, 1984; Frei & Polčík, 1984; Frei, Mandula & Slanina, 1985), cubic sublattices $\bar{L}$ of cubic lattices L have been examined, especially those with the maximal common point group $P^* = \bar{P} \cap P = m\bar{3}m$ (*i.e.* the *klassengleiche* sublattices). However, more general types of cubic sublattices have been found. Not being aware of any systematic treatment of this topic, which may be of interest in crystallography and number theory, the author presents here some facts and suggestions using tentative terminology and notation.

In the following, all the numbers dealt with are integers.

2. A simple example

$L_3(22\bar{1}; \bar{1}22; 2\bar{1}2)$ is an inclined cubic sublattice (ICS) of $L_1 = L_1(100; 010; 001)$ with $P^* = \bar{3}m$. Here $L_n(\mathbf{a}; \mathbf{b}; \mathbf{c})$ means a primitive cubic (p.c.) lattice with basis vectors of length n, the parameter a being omitted. Among all the ICS, L_3 has the smallest unit cell which can easily be imagined as embedded in L_1. Let us therefore point out some features of L_3; these will then be considered with L_n, $n > 3$.

(a) $3^2 = 2^2 + 2^2 + 1^2$ (not only, trivially, $3^2 + 0^2 + 0^2$). (b) $\mathbf{a} \perp \mathbf{b} \perp \mathbf{c}$. ($c$) The 'star' of $s = 24$ vectors $W\mathbf{a}$, $W \in P$, contains and is exhausted by bases of different ICS, say equivalent copies, all of which belong to the type $L_3(221)$ defined by the square decomposition of 3^2 given sub (a); actually, there are four such copies having no common basis vector. Note that, with $W \in m\bar{3}m$, the coordinates of $W(uvw)$ are all the permutations of $\pm u, \pm v, \pm w$. (d) The integers involved in the face diagonals as $\mathbf{d}' = 141$, $30\bar{3}$ *etc.* and in the body diagonals $\mathbf{d}'' = 333$, $\bar{1}5\bar{1}$ *etc.* of the unit cell of L_3 just correspond to all the following decompositions: $2n^2 = 18 = 4^2 + 1^2 + 1^2 = 3^2 + 3^2 + 0^2$, $3n^2 = 27 = 5^2 + 1^2 + 1^2 = 3^2 + 3^2 + 3^2$. ($e$) Finally, face-centred ICS such as $L'_3(141; 114; 411)$ *etc.* and body-centred ones such as $L''_3(\bar{1}5\bar{1}; \bar{1}\bar{1}5; 5\bar{1}\bar{1})$ *etc.* of L_1 are easily constructed using the diagonals $\mathbf{d}'$ and $\mathbf{d}''$ respectively. [The subscript 3 refers to the p.c. L_3; of course, we might also proceed by centring the unit cube of $L_6(44\bar{2}; \bar{2}44; 4\bar{2}4)$.] The corresponding copies of L_3, L'_3 and L''_3 have the same point groups $\bar{P}$ and P^*. Different copies of the centred ICS have in fact common basis vectors owing to the unequal numbers $s' = 24$ or 12 of the vectors in the 'stars' $\{W(141)\}$ or $\{W(30\bar{3})\}$ respectively, and similarly with $s'' = 24$ or 8.

3. Square decompositions and the ICS

To find further triplets of cubic basis vectors in L_1, we take $n = 2, 3, \ldots$, since for odd-dimensional spaces n must be an integer (Cassels, 1959), and look for all the p decomposition of n^2 into two or three squares of integers $u_k \geq v_k \geq w_k \geq 0$:

$$n^2 = u_k^2 + v_k^2 + w_k^2, \quad u_k, v_k > 0, \quad k = 1, \ldots, p(n). \tag{1}$$

Of course, from (1) it follows that

$$(hn)^2 = (hu_k)^2 + (hv_k)^2 + (hw_k)^2, \quad k = 1, \ldots, p(n) \tag{2}$$

so that any $L_n(\mathbf{a}, \mathbf{b}; \mathbf{c})$ gives rise to 'thinner' ICS $L_{hn}(h\mathbf{a}; h\mathbf{b}; h\mathbf{c}) \equiv hL_n(\mathbf{a}; \mathbf{b}; \mathbf{c})$, $h > 1$, but $p(hn) > p(n)$ is possible. For $m = h_1n_1 = h_2n_2 = \ldots$, those $q(m) < p(m)$ decompositions (1) of m^2 may be called essential which cannot be written in the form (2).

For $2n^2$ and $3n^2$ only decompositions into more than one square are possible, with k ranging up to $p'(n)$ and $p''(n)$ respectively; the quantities $q' < p'$ and $q'' < p''$ can be defined in analogy with the above consideration.

If $w = 0$ in (1) we obtain a Pythagorean case of ICS: $L_n(uv0; \bar{v}u0; 00n)$ with $P^* = 4/m$ and $\mathbf{c} \| 001$. Note that any two vectors uv, $\bar{v}u$ define an inclined, or rotated, square sublattice of $l_1(10; 01)$ owing to the even number of dimensions. [Enlarged cells based on similar two-dimensional square sublattices have been used in treating the disorder–order transition of the alloy $MoNi_4$ (Bärnighausen, 1980).]

4. On the ICS found

By inspection of the tabulated square decompositions of $s = 1, 2, \ldots, 1000$ (*International Tables for X-ray Crystallography*, 1959) and by calculation for n up to 83 the following analogues of (a) through (e) have been found: (A) At least one decomposition (1) exists for each $n > 1$, except for $n = 2^t$, $t = 1, \ldots, 6$, and we have $p(2^t n) = p(n)$ for t up to 4. (No restriction for t seems to be valid in both cases.) Less than $3p$ different integers appear in the decompositions (1) except for $n = 2^t.7$ where $p = 1$. Often $p > 1$: for $n = 33$, 65, 73, 79, $p = 10$; $p(57) = p(77) = 15$; $p \geq 20$ for 63 and 81, see Table 1. The more detailed study has then been restricted to $n = 3, 5, \ldots, 31$ ($< 1000^{1/2}$); 63, 81. It can be stated that (B) triplets of mutually orthogonal vectors always occur among the vectors $W(u_kv_kw_k)$, $W \in m\bar{3}m$, $1 \leq k \leq p(n)$ - see $n = 15$ and 25 in Table 1 - or $1 \leq k \leq q(n)$, but (C) the basis vectors may belong to 1, 2 or 3 different 'stars', apart from the Pythagorean cases mentioned in § 3. So we can distinguish the case I ICS with $P^* = \bar{3}$ (or $\bar{3}m$ if $u = v$), the case II with $P^* = 2/m$ as L_9 $(8\bar{4}\bar{1}; 14\bar{8}; 474)$ and case III with $P^* = \bar{1}$ only as L_{15}

Table 1. *Selected data on the ICS for* $n = 3, 5, \ldots, 31, 63, 81$

<table>
<tr><th>n</th><th>p</th><th>q</th><th>p′</th><th>p″</th><th>Cases</th><th>Type symbols of primitive ICS</th></tr>
<tr><td>3</td><td>1</td><td></td><td>2</td><td>2</td><td>I</td><td>(221)</td></tr>
<tr><td>5</td><td>1</td><td></td><td>3</td><td>2</td><td>P</td><td>(430)</td></tr>
<tr><td>7</td><td>1</td><td></td><td>3</td><td>2</td><td>I</td><td>(632)</td></tr>
<tr><td>9</td><td>3</td><td>2</td><td>4</td><td>4</td><td>II</td><td>(841|744)</td></tr>
<tr><td>11</td><td>2</td><td></td><td>4</td><td>4</td><td>II</td><td>(962|766)</td></tr>
<tr><td>13</td><td>2</td><td></td><td>5</td><td>3</td><td>P, I</td><td>(12, 5, 0), (12, 4, 3)</td></tr>
<tr><td>15</td><td>4</td><td>2</td><td>7</td><td>6</td><td>III</td><td>(14, 5, 2|11, 10, 2|10, 10, 5)</td></tr>
<tr><td>17</td><td>3</td><td></td><td>6</td><td>5</td><td>P, II</td><td>(15, 8, 0), (12, 9, 8|12, 12, 1)</td></tr>
<tr><td>19</td><td>3</td><td></td><td>6</td><td>5</td><td>II, I</td><td>(18, 6, 1|17, 6, 6), (15, 10, 6)</td></tr>
<tr><td>21</td><td>6</td><td>4</td><td>8</td><td>6</td><td>I, III</td><td>(20, 5, 4), (19, 8, 4|16, 13, 4|16, 11, 8)</td></tr>
<tr><td>23</td><td>3</td><td></td><td>7</td><td>5</td><td>III</td><td>(22, 6, 3|18, 14, 3|18, 13, 6)</td></tr>
<tr><td>25</td><td>4</td><td>3</td><td>10</td><td>7</td><td>P, III</td><td>(24, 7, 0), (20, 15, 0|20, 12, 9|16, 15, 12)</td></tr>
<tr><td>27</td><td>8</td><td>5</td><td>9</td><td>9</td><td>III,</td><td>(26, 7, 2|23, 14, 2|22, 14, 7),</td></tr>
<tr><td></td><td></td><td></td><td></td><td></td><td>II</td><td>(25, 10, 2|23, 10, 10)</td></tr>
<tr><td>29</td><td>4</td><td></td><td>9</td><td>6</td><td>III,</td><td>(24, 16, 3|24, 12, 11|21, 16, 12),</td></tr>
<tr><td></td><td></td><td></td><td></td><td></td><td>P</td><td>(21, 20, 0)</td></tr>
<tr><td>31</td><td>4</td><td></td><td>9</td><td>6</td><td>I, III</td><td>(30, 6, 5), (27, 14, 6|22, 21, 6|21, 18, 14)</td></tr>
<tr><td>63</td><td>20</td><td>12</td><td>22</td><td>17</td><td>III</td><td>(62, 11, 2|59, 22, 2|58, 22, 11),
(62, 10, 5|50, 38, 5|50, 37, 10),
⋮(53, 34, 2|46, 37, 22|43, 38, 26),
⋮(53, 26, 22|46, 43, 2|38, 37, 34)</td></tr>
<tr><td>81</td><td>22</td><td>14</td><td>23</td><td>23</td><td>III,</td><td>(79, 16, 8|64, 49, 8|64, 47, 16),
⋮(76, 28, 1|68, 41, 16|64, 44, 23),
⋮(76, 23, 16|68, 44, 1|64, 41, 28)
(65, 44, 20|56, 55, 20|55, 44, 40),</td></tr>
<tr><td></td><td></td><td></td><td></td><td></td><td>II</td><td>(56, 49, 32|56, 56, 17)</td></tr>
</table>

$(14, 5, 2; 2, -10, 11; 5, -10, -10)$. In the abbreviated type symbol for the case II ICS, the first triplet of integers is understood to appear in the coordinates of two basis vectors, see Table 1. Its last column shows that some 'stars' contain basis vectors of different ICS, *e.g.* of L_{15} and $5L_3$ (cases III and I, respectively) or of L_{25} and $5L_5$ (III, P). Some case III ICS exhibit nine different integers in the bases but for two different types simultaneously, as indicated by the vertical dotted lines. Finally, as examined in detail for all the L_n studied, the face and body diagonals of the unit cubes just correspond to all the decompositions of $2n^2$ and $3n^2$ respectively so that straightforward generalizations, (D) and (E), of (d) and (3) have been verified.

5. Further remarks and hypotheses

It is seen that the ICS differ from the coincidence-site lattices (see Vlachavas, 1985) but bear some resemblance to - or represent a special case of (Bärnighausen, 1980) - the derivative lattices (*International Tables for Crystallography*, 1987).

The subgroups of cubic translation groups form, under the condition $P^* = P$, a system of interconnected chains based on the relations $L_1', L_1'' \subset L_1$; $L_2 \subset L_1', L_1''$; $L_n \subset L_1$, n prime (Fuksa, 1986; Kopský, 1988*a*) but owing to the ICS the system of all the subgroups is much richer.

The properties of the ICS based on essential decompositions (1) can also be considered by reducing all of them to $(1/n)L_n$. Each unit cube with one vertex 000 has, then, further vertices with rational coordinates on the unit sphere and on the $2^{1/2}$ as well as $3^{1/2}$ spheres; some (finite?) part of the vertices lies in the points as for 111 (case I), further in the points as 001 and on the circular lines as $xy0$ and $xy1$, $x^2 + y^2 = 1$ (Pythagorean ICS) or as xyx, $2x^2 + y^2 = 1$ (case II) with additional axes and mirror planes due to the respective groups $\bar{P}$. From this it may be argued that $m\bar{3}m$ is not a maximal subgroup of the full orthogonal group as shown in Fig. 10.4.3 in Volume A of *International Tables for Crystallography* (1987) (Kopský, 1988*b*).

The ICS may have non-trivial implications in number theory. To the author's knowledge, the problem of the number of p.c. lattice points within a sphere of radius r (real number) has been dealt with involving decompositions of the form (1) while the existence of orthogonal bases for each $n, p(n) \geq 1$, and of all the appropriate diagonals seems to suggest interesting mathematical hypotheses.

No non-cubic inclined sublattices have been studied.

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An Envelope-Based Approach for Direct Phase Determination of Macromolecular Structures

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Abstract

An initial electron density distribution for a crystal structure may be directly derived from observed diffraction data by maximizing the product of the observed and calculated Patterson functions with respect to the electron density values within an envelope. This maximization problem may be formulated as an eigenvalue equation, in which potential electron density distributions are obtained as eigendensities (eigenvectors) of a symmetric matrix. Elements of this matrix depend only on the indices and intensities of the observed reflections, and on the coordinates of grid points inside the envelope. Eigendensities are calculated for a set of small envelopes (enclosing about 20% of the molecular volume) covering a unique region of the unit cell whose points are unrelated by space-group operations, origin shifts or changes in enantiomorph. On the basis of correlation coefficients between the observed and calculated values for both the Patterson function and structure-factor amplitudes, a small set of eigendensities are selected for combination into a final electron density distribution. This electron density distribution may be Fourier transformed to yield calculated structure factors. Test calculations on lysozyme indicate that phase errors of less than 60° may be obtained for strong low-resolution reflections by this procedure. An extension of this approach to handle crystal structures containing non-crystallographic symmetry is described.

Introduction

A molecular envelope divides the interior of macromolecular crystals into two regions of roughly equal volumes, consisting of either the molecule or the solvent. Owing to positional disorder of the solvent molecules, the electron density in the solvent region is approximately constant. Enforcement of this constraint on the solvent density ('solvent flattening') has provided a powerful phase refinement method, in both the presence (Rossmann, 1972) and absence (Wang, 1985) of non-crystallographic symmetry. In principle, knowledge of the molecular envelope also provides an approach to direct phase determination, but this has not yet been routinely achieved in practice. In this note, a constrained symmetric quadratic function of the electron densities of grid points within a molecular envelope is formulated that depends only on the indices and intensities of observed reflections. Electron density distributions derived from the maximization of this function may be used to directly obtain phase information.

Although crystallographic calculations may be formally accomplished in either real or reciprocal space, a real-space emphasis will be adopted in this work since the distinction between molecular and solvent regions has a particularly simple real-space interpretation. Assuming a molecular envelope has been defined (the determination of the envelope will be discussed more fully below), the problem to be addressed is to find density values for grid points within the envelope that are consistent with the observed Patterson function. This may be accomplished by finding electron density values that maximize Φ:

$$\Phi = \sum_k P_o(u_k)P_c(u_k) \qquad (1)$$

where Φ is the product of the multiply corrected observed and calculated Patterson functions, $P_o(u_k)$ and $P_c(u_k)$, respectively. The sum is over points at

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