total disorder (continuous diffuse lines instead of reflexions on X-ray diffraction photographs).

Moreover, we must pay attention to the fact that the different types of faults exert a similar influence on different points of the reciprocal lattice. Thus it is not possible to distinguish between some types of faults on the basis of the above parameters. One could try also to find expressions for measurable parameters describing lattice-point asymmetry and changes in the integrated intensity, as was done by Prasad & Lele (1971). However, these changes and peak asymmetry are usually too small to be estimated with sufficient accuracy. Thus peak shifts and half widths are recognized to be the best measures of faultiness. This was shown by Pandey & Krishna (1976) for the $6H(33)$ structure.

The limitations of our theory and inaccuracy in the results which follow from the assumption of small values of α_{jk} are the next problem for discussion. We will show that this assumption does not limit the generality of the above theory because only small values of α_{jk} have physical sense. In order to justify the above statement let us recall the definition of probability α_{jk}. It is equal to the ratio of the number of layers followed by faults of a particular type to the full number of layers in the examined sequence. For example, in the following sequence of an $8H(44)$ structure with stacking faults [(44334433444433444433344) - in Zhdanov symbols] we have $\alpha_{(33)} = 4/80 = 0{\cdot}05$. It is clear that consideration of these faults as the (33) type in $8H(44)$ structures makes sense only for $\alpha_{(33)} < 0{\cdot}1$. For $\alpha_{(33)} > 0{\cdot}1$ the frequency of the occurrence of faults of (33) type is so great that the Zhdanov symbols (33) must be united in groups and it is necessary to interpret this sequence as a $6H(33)$ structure with stacking faults of (4) type. For example, it is necessary to interpret the sequence (33433433433334) as a $6H(33)$ structure with $\alpha_{(4)} = 4/46$ but not as an $8H(44)$ structure with $\alpha_{(33)} = 5/46$. We expect that on X-ray diffraction photographs from the structure with this sequence the peak maxima will occur near the positions corresponding to those for a $6H(33)$ structure.

The assumption of a random distribution of single faults does not limit our theory either. In general, when this assumption is not fulfilled another polytypic structure is formed.

References

LELE, S. (1974a). *Acta Cryst.* A30, 689-693.

LELE, S. (1974b). *Acta Cryst.* A30, 693-698.

MICHALSKI, E. (1988). *Acta Cryst.* A44, 640-649.

MICHALSKI, E., DEMIANIUK, M., KACZMAREK, S. & ZHMIJA, J. (1981). *Proc. VI Int. Summer School on Lattice Defects in Crystals, Krynica, Poland,* 1980, pp. 300-309. Wrocław: Ossolineum.

PANDEY, D. & KRISHNA, P. (1976). *Acta Cryst.* A32, 488-492.

PRASAD, D. & LELE, S. (1971). *Acta Cryst.* A27, 54-64.

WARREN, B. E. (1959). *Prog. Met. Phys.* 8, 147-202.

Acta Cryst. (1988). A44, 657-661

Direct Methods: the Identification of Conditions Which Simplify the Generation of Inconsistent Quadrupoles

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Abstract

An algorithm is implemented to determine the form and phase shift for inconsistent type II quadrupoles for any space group having glide or screw-axis translations which are not a consequence of lattice centering. Cumulatively there are only six different Miller index restrictions and nine different phase shift forms common to all space groups of orthorhombic or lower symmetry. A similar analysis has been performed for a newly discovered type III class of quadrupoles. The configuration of the phase connections among the four triples of the type III quadrupole is different from the common configuration previously described for both normal (type I) and inconsistent (type II) quadrupoles. A knowledge of these constraint conditions for type II and III quadrupoles greatly simplifies a procedure for generating these relationships.

Introduction

A quadrupole has been defined as a relationship among four interdependent three-phase invariants,

$$\begin{aligned} \Phi_1 &= \varphi_{\mathbf{h}} - \varphi_{\mathbf{k}} + \varphi_{\mathbf{k}-\mathbf{h}} \\ \Phi_2 &= \varphi_{\mathbf{k}} - \varphi_{\mathbf{l}} + \varphi_{\mathbf{l}-\mathbf{k}} \\ \Phi_3 &= \varphi_{\mathbf{l}} - \varphi_{\mathbf{h}} + \varphi_{\mathbf{h}-\mathbf{l}} \\ \Phi_4 &= -\varphi_{\mathbf{k}-\mathbf{h}} - \varphi_{\mathbf{l}-\mathbf{k}} - \varphi_{\mathbf{h}-\mathbf{l}}, \end{aligned} \quad (1)$$

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such that the sum of phases, which occur as six Friedel-related pairs,

$$\Phi_1+\Phi_2+\Phi_3+\Phi_4=0. \tag{2}$$

An inconsistent quadrupole phase relationship (Viterbo & Woolfson, 1973),

$$\begin{aligned}\Phi_1&=\varphi_{\mathbf{h}}-\varphi_{\mathbf{k}}+\varphi_{\mathbf{k-h}}\\ \Phi_2&=\varphi_{\mathbf{k}}-\varphi_{\mathbf{l}}+\varphi_{\mathbf{l-k}}\\ \Phi_3&=\varphi_{\mathbf{l}}-\varphi_{\mathbf{h.R}_j}+\varphi_{\mathbf{h.R}_j-\mathbf{l}}\\ \Phi_4&=-\varphi_{(\mathbf{k-h}).\mathbf{R}_k}-\varphi_{(\mathbf{l-k}).\mathbf{R}_l}-\varphi_{\mathbf{h.R}_j-\mathbf{l}},\end{aligned} \tag{3}$$

can be formed in non-symmorphic space groups provided that conditions relating certain of the reflection indices can be satisfied, *i.e.*

$$(\mathbf{h}-\mathbf{k}).\mathbf{R}_k+(\mathbf{k}-\mathbf{l}).\mathbf{R}_l+\mathbf{l}-\mathbf{h}.\mathbf{R}_j=0, \tag{4}$$

and such that a non-zero phase invariant sum

$$\Phi_1+\Phi_2+\Phi_3+\Phi_4=2\pi[-\mathbf{h}.\mathbf{t}_j+(\mathbf{h}-\mathbf{k}).\mathbf{t}_k+(\mathbf{k}-\mathbf{l}).\mathbf{t}_l]\ (\text{modulo } 2\pi) \tag{5}$$

will result. The $\mathbf{R}_j$ are the inverse transforms of the rotation matrices for the various equipoints of the space group, the $\mathbf{t}_j$ are the associated translation vectors. Inconsistent quadrupoles have been referred to as type II quadrupoles (Cascarano, Giacovazzo, Camalli, Spagna, Burla, Nunzi & Polidori, 1984) in acknowledgement of the class of normal type I quadrupoles given as expression (1). Conditions which define these inconsistent quadrupole relationships in permissible space groups may be expressed in terms of a reduced set of any three of the six reflection vectors which themselves do not form a single three-phase invariant. Thus from (4) one obtains

$$\mathbf{h}.(\mathbf{R}_k-\mathbf{R}_j)+\mathbf{k}.(\mathbf{R}_l-\mathbf{R}_k)+\mathbf{l}.(\mathbf{I}-\mathbf{R}_l)=0, \tag{6}$$

which defines the constraints on the components of the three vectors which must be satisfied in order to form an inconsistent quadrupole.

One should be aware of a situation which may arise when zonal or axial reflections appear in such quadrupoles for non-centrosymmetric structures. For example, in space group $P2_12_12_1$

$$\begin{aligned}\Phi_1&=\varphi_{032}+\varphi_{\bar{1}\bar{6}\bar{2}}+\varphi_{130}\\ \Phi_2&=\varphi_{162}+\varphi_{\bar{1}\bar{4}0}+\varphi_{0\bar{2}\bar{2}}\\ \Phi_3&=\varphi_{140}+\varphi_{0\bar{3}\bar{2}}+\varphi_{\bar{1}\bar{1}2}\\ \Phi_4&=\varphi_{\bar{1}\bar{3}0}+\varphi_{022}+\varphi_{11\bar{2}}\\ \Phi_1&+\Phi_2+\Phi_3+\Phi_4=0\end{aligned} \tag{7}$$

defines a normal quadrupole relationship. However, because of the zeros, one can often perform Friedel-related operations (*e.g.* $h, k, l \rightarrow h, k, -l$) on several of the triple invariants,

$$\begin{aligned}-\Phi_3&=\varphi_{140}+\varphi_{0\bar{3}2}+\varphi_{\bar{1}\bar{1}\bar{2}}\\ -\Phi_4&=\varphi_{\bar{1}\bar{3}0}+\varphi_{02\bar{2}}+\varphi_{112}\end{aligned}$$

and observe that the same invariants also define a valid π-quadrupole,

$$\Phi_1+\Phi_2-\Phi_3-\Phi_4=\pi\ (\text{modulo } 2\pi). \tag{8}$$

Although (7) defines a normal quadrupole that suggests all four $|\Phi_i|$ may be clustered near 0°, condition (8) informs us that at least two of the $|\Phi_i|$ must be 45° or more in error. In such instances it is imperative to treat such ambidextrous quadrupoles as being inconsistent. A similar more trivial example exists when a normal quadrupole possesses only one Φ_i which is restricted, and to ±90°. Friedel inversion of this phase invariant ensures the formation of a π-quadrupole, and at least one of the three unrestricted $|\Phi_i|$ must be in error 30° or more. Conditions (5) and (6) are sufficient to identify unambiguously each of the above examples as an inconsistent quadrupole, regardless of the special nature of zonal reflections which make them ambidextrous.

We note that a new class of quadrupoles, referred to as type III, can be formed from a different configuration of six pairs of phases from four three-phase invariants, namely

$$\begin{aligned}\Phi_1&=\varphi_{\mathbf{h}}+\varphi_{\mathbf{k}}-\varphi_{\mathbf{h+k}}\\ \Phi_2&=-\varphi_{\mathbf{h}}-\varphi_{\mathbf{k.R}_j}+\varphi_{\mathbf{h+k.R}_j}\\ \Phi_3&=\varphi_{\mathbf{h+k}}-\varphi_{\mathbf{l}}-\varphi_{\mathbf{h+k-l}}\\ \Phi_4&=-\varphi_{\mathbf{h+k.R}_j}+\varphi_{\mathbf{l.R}_k}+\varphi_{(\mathbf{h+k-l}).\mathbf{R}_l},\end{aligned} \tag{9}$$

where the phases from each triple are linked to only two of the remaining three triples, rather than all three as was commonly noted for the type I and II relationships. It should be obvious that (9) cannot form a quadrupole in which all phase pairs are strictly Friedel related. Such a relationship would not contain four independent triples, but rather two independent pairs. It also follows that $\mathbf{l}$ cannot be a general vector, but must be constrained in order to satisfy the condition

$$(\mathbf{h}+\mathbf{k}).(\mathbf{R}_l-\mathbf{R}_j)+\mathbf{l}.(\mathbf{R}_k-\mathbf{R}_l)=0. \tag{10}$$

All such relationships are potentially inconsistent, as was noted for the type II quadrupoles, the phase shift for type III quadrupoles being

$$\Phi_1+\Phi_2+\Phi_3+\Phi_4=2\pi[-\mathbf{k}.\mathbf{t}_j+\mathbf{l}.\mathbf{t}_k+(\mathbf{h}+\mathbf{k}-\mathbf{l}).\mathbf{t}_l](\text{modulo } 2\pi). \tag{11}$$

An example of a type III quadrupole in space group $P2_12_12_1$ given as

$$\begin{aligned}\Phi_1 &= \varphi_{123} + \varphi_{336} + \varphi_{\bar{4}\bar{5}\bar{9}}\\ \Phi_2 &= \varphi_{\bar{1}\bar{2}\bar{3}} + \varphi_{\bar{3}36} + \varphi_{4\bar{1}\bar{3}}\\ \Phi_3 &= \varphi_{459} + \varphi_{\bar{6}\bar{3}\bar{6}} + \varphi_{2\bar{2}\bar{3}}\\ \Phi_4 &= \varphi_{\bar{4}13} + \varphi_{6\bar{3}\bar{6}} + \varphi_{\bar{2}23}\end{aligned} \qquad (12)$$

may be alternately written in configuration (9) with the reflections indexed as the vectors **l** and **h**+**k**+**l** interchanged to rearrange the last two three-phase invariants:

$$\begin{aligned}\Phi_3 &= \varphi_{459} + \varphi_{2\bar{2}\bar{3}} + \varphi_{\bar{6}\bar{3}\bar{6}}\\ \Phi_4 &= \varphi_{\bar{4}13} + \varphi_{\bar{2}23} + \varphi_{6\bar{3}\bar{6}},\end{aligned} \qquad (13)$$

such that the constraints (10) on the reflection indexed as the vector **l** are different. In the first example **l** (636) is restricted to have the same k and l indices as reflection **k** (336), while in the second case the reflection **l** ($\bar{2}23$) has the same k and l indices as reflection **h** (123). It is important to recognize that these differently arranged quadrupoles are identical, and the two distinct constraint conditions are redundant insofar as producing the same quadrupole is concerned, and it is immaterial which of the two is employed and whether reflection 636 or 223 is to be treated as the vector **l**. The reciprocal-lattice and phase-shift factors for any one of the two cases are related to those for the other case by interchanging the vectors **h** and **k** as they appear in the conditions.

Results

The conditions for type II quadrupoles imposed by equations (5) and (6) were explored systematically for all 230 space groups using the equivalent-positions-generating code described by Burzlaff & Hountas (1982) to obtain the rotation matrices and translation vectors required. The cumulative results for the numbers of distinct reflection index conditions (6) and translation components (5) for all space groups within each crystal class are given in Table 1. Seven of the 13 monoclinic space groups permit inconsistent quadrupoles, and these quadrupoles may be expressed in terms of two distinct reciprocal-lattice conditions and two different phase-shift forms. Of the 59 orthorhombic space groups 46 allow inconsistent quadrupoles; these may be expressed in terms of six distinct reciprocal-lattice conditions and nine different phase-shift forms. These six and nine orthorhombic conditions include those exhibited by monoclinic symmetries, such that these results are cumulative of the first 74 space groups in *International Tables for Crystallography* (1983). The situation becomes exceedingly more complex as daughter-related equivalent positions are introduced in higher-symmetry space groups. The first tetragonal space group that has inconsistent quadrupole relationships is $P4_1$, which has two parent-form $(x, y, z; -x, -y, \frac{1}{2}+z)$ and two daughter-form $(-y, x, \frac{1}{4}+z; y, -x, \frac{3}{4}+z)$ equivalent positions in the primitive reduced cell. These give rise to 43 distinct reciprocal-lattice conditions and 45 different phase-shift forms for forming inconsistent quadrupoles. The cumulative results for all tetragonal space groups are a staggering 705 distinct reciprocal-lattice conditions and 1601 different phase shifts, given the specific non-redundant form of the quadrupole defined by (3).

Table 1. *Cumulative number of reciprocal-lattice conditions and phase-shift type of type* II *quadrupoles for each crystal class*

Crystal class	Cumulative *HKL* conditions	Cumulative shift types
Monoclinic	2	2
Orthorhombic	6	9
Tetragonal	705	1601
Trigonal	686	45
Hexagonal	2897	303
Cubic	77093	14989

The trigonal, hexagonal and cubic space groups are seen to produce equally large numbers of ways of forming these relationships, such that it would be impossible to tabulate these various distinct conditions in the space provided in a paper of normal length. It should also be apparent that it would be self defeating to attempt to retrieve and utilize large numbers of conditions from such an unwieldy compilation if the intent is to incorporate these conditions into a procedure which would identify the inconsistent quadrupoles among a phasing set of triples. It would be best to generate these conditions from the space-group symbol, as was done in this investigation. Since the number of conditions for space groups of orthorhombic or lower symmetry is manageable, and since many organic and biomolecular compounds tend to crystallize in lower-symmetry habits, an exhaustive compilation is given for these space groups in Table 2.*

The six reciprocal-lattice conditions and nine phase shifts indicated are listed in Table 3. The $P2_12_12_1$ example given above conforms to the reciprocal-lattice condition A in Table 3 where $\mathbf{k} = -h_1, -k_1, -l_1$ (162), $\mathbf{l} = h_2, k_2, l_2$ (140), and $\mathbf{h} = h_3, k_3, l_3$ (032); $\mathbf{R}_k = \mathbf{I}$, $\mathbf{R}_j = \mathbf{R}_l$ $(h, k, l \to -h, k, -l)$. The phase shift given by entry 1 in the lower part of Table 3 requires $k_1 + k_2 + k_3$ to be odd if the quadrupole is to be in-

* Complete versions of Tables 2 to 5 have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 44862 (8 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. The excerpts of these tables used in the present paper are for the first 19 space groups, which include the $P2_12_12_1$ entries referred to in the text.

Table 2. *Condition and phase shift of type* II *quadrupole for non-symmorphic space groups of* $P2_12_12_1$ *or lower symmetry*

All symbols refer to Table 3.

Space group	Quadrupole
$P2_1$	$A1$
Pc	$B2$
Cc	$B2$
$P2_1/m$	$A1$
$P2/c$	$B2$
$P2_1/c$	$A1$ $B2$
$C2/c$	$B2$
$P222_1$	$C2$
$P2_12_12$	$A1$ $D3$
$P2_12_12_1$	$A1$ $C2$ $D3$

Table 3. *The Miller indices of this table are defined as* $\mathbf{h}=h_3, k_3, l_3$; $\mathbf{k}=-h_1, -k_1, -l_1$; $\mathbf{l}=h_2, k_2, l_2$ *referring to equation* (3)

Because there is no higher rotation operation than twofold and $\mathbf{R}_k=\mathbf{I}$ and $\mathbf{R}_j=\mathbf{R}_l$, for each case, only three diagonal elements of $\mathbf{R}_j$ and $\mathbf{R}_l$ are listed in the table.

(*a*) Symbols of quadrupole condition and type in Table 1.

Symbol	Condition	Operators
A	$h_1+h_2+h_3=0$ and $l_1+l_2+l_3=0$	-1 $+1$ -1
B	$k_1+k_2+k_3=0$	$+1$ -1 $+1$
C	$h_1+h_2+h_3=0$ and $k_1+k_2+k_3=0$	-1 -1 $+1$
D	$k_1+k_2+k_3=0$ and $l_1+l_2+l_3=0$	$+1$ -1 -1

(*b*) Symbols of quadrupole phase shift in Table 1.

Symbol	Phase shift
1	$\frac{1}{2}k_1+\frac{1}{2}k_2+\frac{1}{2}k_3$
2	$\frac{1}{2}l_1+\frac{1}{2}l_2+\frac{1}{2}l_3$
3	$\frac{1}{2}h_1+\frac{1}{2}h_2+\frac{1}{2}h_3$

consistent. Since condition A requires $h_1+h_2+h_3=0$ and $l_1+l_2+l_3=0$, it follows that the generative reflection $\mathbf{l}$ (140) must have the same h and l indices as $\mathbf{k}-\mathbf{h}$ (130), but have a permutable k index of opposite parity. Thus, given the generative reflection E_{140}, the quadrupole is completely defined, and one has only to determine whether E_{022} and E_{112} are also in the phasing set.

The conditions for type III quadrupoles given by (10) and (11) were explored systematically for all non-symmorphic space groups of orthorhombic or lower symmetry. Overall, these space groups give rise to a maximum of 24 distinct reciprocal-lattice conditions and 33 different phase-shift forms. This may be compared with the six reciprocal-lattice conditions and nine phase shifts required for the type II quadrupoles for these space groups. The 24 reciprocal-lattice conditions can be reduced to a non-redundant set of 12 conditions as was noted for the example (12, 13). Similarly 28 of the 33 phase-shift forms appear as redundant pairs and a reduced set of 14 plus 5 or 19 can be obtained.

Any two reduced sets of conditions, however, are not sufficient for defining all the space groups given in Table 4, as can be shown by the entries for the space groups $P2_1/m$, $P2/c$ and $P2_1/c$. Here the pertinent equivalent sets of conditions are ($A1$, $B2$), ($C3$, $D4$), ($E1$, $F2$) and ($E4$, $F3$). The isolation of a non-redundant set by selecting the conditions $A1$ and $C3$ in preference to $B2$ and $D4$ produces a conflict in that both $E1$ and $F3$ must be selected for their permitted phase shifts, in spite of the fact that the conditions E and F are a redundant pair. This conflict can be resolved by alternatively choosing $A1$ and $D4$ in preference to $B2$ and $C3$, and $E1$ and $E4$ in preference to $F2$ and $F3$, thus eliminating the symbols B, C, F, 2 and 3. Given that ($C6$, $D7$), ($Q3$, $R4$) and ($Q7$, $R6$) also appear as paired redundancies in Table 4, $C6$ and $Q3$ must be eliminated since the codes C and 3 have been rejected in favor of codes D and 4. It follows that neither $Q7$ or $R6$ can be represented by the reduced set of codes as both Q and 6 have been eliminated.

Table 4. *Conditions and phase shifts of type* III *quadrupoles for non-symmorphic space groups of* $P2_12_12_1$ *or lower symmetry*

All symbols refer to Table 5. Parentheses indicate the redundant equivalent pairs.

Space group	Type of quadrupoles
$P2_1$	$A1$ ($B2$)
Pc	$C3$ ($D4$)
Cc	$C3$ ($D4$)
$P2_1/m$	$A1$ ($B2$) $E1$ ($F2$)
$P2/c$	$C3$ ($D4$) $E4$ ($F3$)
$P2_1/c$	$A1$ ($B2$) $C3$ ($D4$) $E5$ ($F5$)
$C2/c$	$C3$ ($D4$) $E4$ ($F3$)
$P222_1$	$G4$ ($H3$)
$P2_12_12$	$A1$ ($B2$) $I6$ ($J7$)
$P2_12_12_1$	$A1$ ($B2$) $G4$ ($H3$) $I6$ ($J7$)

A non-redundant list of conditions may, however, be abstracted for any single space-group entry in Table 4, as it may be observed that no reciprocal-lattice condition is ever repeated more than once for any particular space group. Any non-redundant set of phase-shift conditions may be selected and there can be no conflict among the reciprocal-lattice conditions that are forced into this reduced set of conditions.

The $P2_12_12_1$ example (12) corresponds to condition $I6$ in Table 4 while the equivalent alternate form (13) corresponds to condition $J7$. In addition to the reciprocal-lattice constraints on the vector $\mathbf{l}$, Table 5 also defines $\mathbf{R}_j$ and $\mathbf{R}_k$ which guarantee closure for the quadrupole. An algorithm for identifying inconsistent type III quadrupoles is similar to that employed for type I and type II quadruples, insofar as the phases in the variant Φ_1 must be rotated in order to sample the three distinct phase pairs as the vectors $\mathbf{h}$ and $\mathbf{k}$. The procedure differs from other quadrupole-generating methods in that, given any pair $\mathbf{h}$ and $\mathbf{k}$, invariant Φ_2 is determined, as it does not involve the random vector $\mathbf{l}$ as it appears in either (1) or (3). It merely has to be determined whether

Table 5. *The Miller indices of this table are defined as* $\mathbf{h}=h_1, k_1, l_1$; $\mathbf{k}=h_2, k_2, l_2$; $\mathbf{l}=h_3, k_3, l_3$ *referring to equation* (9)

The three diagonal elements of $\mathbf{R}_j$ and $\mathbf{R}_k$ are listed in the table.

(*a*) Symbols of quadrupole condition and symmetry operators in Table 4.

Symbol	*HKL* condition	$\mathbf{R}_j$	$\mathbf{R}_k$
A	$h_2=h_3$ and $l_2=l_3$	−1 +1 −1	−1 +1 −1
B	$h_1=h_3$ and $l_1=l_3$	−1 +1 −1	+1 +1 +1
C	$k_2=k_3$	+1 −1 +1	+1 −1 +1
D	$k_1=k_3$	+1 −1 +1	+1 +1 +1
E	$h_2=h_3$ and $k_1=k_3$ and $l_2=l_3$	−1 −1 −1	−1 +1 −1
F	$h_1=h_3$ and $k_2=k_3$ and $l_1=l_3$	−1 −1 −1	+1 −1 +1
G	$h_1=h_3$ and $k_1=k_3$	−1 −1 +1	+1 +1 +1
H	$h_2=h_3$ and $k_2=k_3$	−1 −1 +1	−1 −1 +1
I	$k_2=k_3$ and $l_2=l_3$	+1 −1 −1	+1 −1 −1
J	$k_1=k_3$ and $l_1=l_3$	+1 −1 −1	+1 +1 +1

(*b*) Symbols of quadrupole phase shift in Table 4.

Symbol	Phase shift/360°
1	$\frac{1}{2}k_2+\frac{1}{2}k_3$
2	$\frac{1}{2}k_1+\frac{1}{2}k_3$
3	$\frac{1}{2}l_2+\frac{1}{2}l_3$
4	$\frac{1}{2}l_1+\frac{1}{2}l_3$
5	$\frac{1}{2}k_1+\frac{1}{2}k_2+\frac{1}{2}l_1+\frac{1}{2}l_2$
6	$\frac{1}{2}h_2+\frac{1}{2}h_3$
7	$\frac{1}{2}h_1+\frac{1}{2}h_3$

$\mathbf{h}+\mathbf{k}.\mathbf{R}_j$ corresponds to an E value in the phasing set. If so, the invariants Φ_3 and Φ_4 are constructed by searching the phase set for those few data which satisfy the constraints on the vector $\mathbf{l}$ and have a non-zero phase shift. Only the non-restricted components of the vector $\mathbf{l}$ need be permuted prior to finding whether $\mathbf{h}+\mathbf{k}+\mathbf{l}$ corresponds to a large E value employed in the phasing set. $\mathbf{R}_k$ is not required to identify the data which close the quadrupole, but merely establishes the signs on the Miller indices for the phases in Φ_4. The translational shifts $\mathbf{t}_j$, $\mathbf{t}_k$ and $\mathbf{t}_l$ required for the calculation of the phase shift (10) need not be given, as these values have been used to produce the explicit results in Table 5.

The conditions for generating type III quadrupoles seem slightly more numerous than those for type II quadrupoles, which cumulatively require only six different lattice conditions and nine phase shifts for all space groups of orthorhombic or lower symmetry. The space group *Pbca*, for example, requires the most sets of conditions for both type II and type III quadrupoles, *i.e.* 6 and 12 respectively. Higher-symmetry space groups generate a much larger set of reciprocal-lattice and phase-shift conditions. The first tetragonal space group that produces inconsistent quadrupoles is $P4_1$, there being 45 unique sets of conditions for type II *versus* 21 non-redundant sets of conditions for type III quadrupoles. Thus it cannot be concluded, as could be inferred from Table 1, that the number of type III conditions will equal or exceed those required for type II quadrupoles for higher-symmetry space groups.

Discussion

Whereas the question of identifying the conditions which give rise to inconsistent quadrupoles has been answered above as a search over three independent rotation matrices (6) or (10), the problem of generating the inconsistent quadrupoles common to a particular triple has been described as a time-consuming search involving four independent symmetry operations (Cascarano *et al.*, 1984). This in no way disputes the validity of the earlier work. In comparing expression (12) of Cascarano *et al.* (1984) with (3) or (9) in this work, it should be clear that four symmetry operations are involved in blindly generating these quadrupoles, but only three are required to define the conditions for their generation and establish that a search over a fourth operation is unnecessary. It may, moreover, be seen that the reciprocal-lattice conditions given in Table 3 further reduce the time for such a search since (*a*) the symmetry matrices are defined, and (*b*) the generative reflection $\mathbf{l}$ in expression (3) or (9) need not span the full list of strongest $|E|$ magnitudes, but only those vectors $\mathbf{l}$, if any, which are related to the indices of vectors $\mathbf{h}$ and $\mathbf{k}$ in a specific way. These relationships can often be established prior to performing the search. Once such an $E_\mathbf{l}$ has been found which produces a non-zero phase shift, one has only to discover whether $E_{\mathbf{l}-\mathbf{k}}$ and $E_{\mathbf{h}.\mathbf{R}_j-\mathbf{l}}$ are in the phase set. If both E values are large, the relationship is ensured and one need not scan and test the matrices $\mathbf{R_j}$, $\mathbf{R_k}$ and $\mathbf{R_l}$ to prove that such a relationship can be formed, as this information is given in Table 3. This algorithm would appear to be very efficient for non-symmorphic space groups of orthorhombic or lower symmetry. In higher-symmetry examples the number of distinct reciprocal-lattice conditions can often exceed the product of the number of symmetry operations and phased E values, such that it may be more efficient to use the more general scheme proposed by Cascarano *et al.* (1984).

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References

BURZLAFF, H. & HOUNTAS, A. (1982). *J. Appl. Cryst.* **15**, 464–467.

CASCARANO, G., GIACOVAZZO, C., CAMALLI, M., SPAGNA, R., BURLA, M. C., NUNZI, A. & POLIDORI, G. (1984). *Acta Cryst.* A**40**, 278–283.

International Tables for Crystallography (1983). Vol. A. Dordrecht: Reidel. (Present distributor Kluwer Academic Publishers, Dordrecht.)

VITERBO, D. & WOOLFSON, M. M. (1973). *Acta Cryst.* A**29**, 205–208.