

served on our maps. Fig. 4 shows the variation of the 'satellite peak' separation with the amplitude of the displacement. Both modulation waves involved in the Patterson interaction have the same amplitude and phase. In Fig. 5, the maximum density in the central peak is shown for the Patterson interaction of two displacement modulation waves with equal phase but with different amplitudes. The amplitude of one wave is constant (0.55 Å); the amplitude of the other modulation wave varies from 0 to 0.6 Å.

Note added 29 August 1972. The least squares refinement of atomic coordinates based on measured 'a' reflections from this sample has been completed and published elsewhere. It produced coordinates that in some cases (Na/Ca cations) differed substantially from

those given by Phillips *et al.* (1971) but these differences do not effect the conclusions reached in this paper.

References

- BOWN, M. G. & GAY, P. (1958). *Z. Kristallogr.* **111**, 1–14.
 KOREKAWA, M. & JAGODZINSKI, H. (1967). *Schweiz. Miner. Petrogr. Mitt.* **47**, 269–278.
 MEGAW, H. D. (1956). *Acta Cryst.* **9**, 56–60.
 PHILLIPS, M. W., COLVILLE, A. A. & RIBBE, P. H. (1971). *Z. Kristallogr.* **133**, 43–65.
 TOMAN, K. & FRUEH, A. J. (1971). *Acta Cryst.* **B27**, 2182–2186.
 TOMAN, K. & FRUEH, A. J. (1972). *Acta Cryst.* **B28**, 1657–1662.
 TOMAN, K. & FRUEH, A. J. (1973). *Acta Cryst.* **A29**, 121–127.

Acta Cryst. (1973). **A29**, 133

The Phase Function: New Developments in the Symbolic-Addition Procedure

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A modification of the symbolic-addition procedure, based on the introduction of a 'phase function', is proposed. This function which determines the numerical values of the symbols, enables one to select rapidly the best solution from a large number of possible ones. Examples of non-centrosymmetric structures solved by this method are given here.

Introduction

It is well known that the most critical part of solving non-centrosymmetric structures by direct methods lies in the determination of a starting set of numerical phases. The number of such phases increases with the complexity of the structure (Germain & Woolfson, 1968).

Karle and Karle who have demonstrated the power of these methods have developed a successful procedure using symbols (Karle & Karle, 1966). The main difficulties in using this procedure are:

(a) in the first step, single indications of phases from equation (1) must be accepted

$$\varphi_{\mathbf{H}} \simeq \varphi_{\mathbf{K}} + \varphi_{\mathbf{H}-\mathbf{K}} \quad (\text{Cochran, 1955}), \quad (1)$$

(b) considerable care must be applied in the use of equation (2)

$$\varphi_{\mathbf{H}} \simeq \langle \varphi_{\mathbf{K}} + \varphi_{\mathbf{H}-\mathbf{K}} \rangle_{\mathbf{K}} \quad (\text{Karle \& Karle, 1966}) \quad (2)$$

(c) determination of the numerical values of the symbols.

Starting with numerical phases instead of symbols, the multisolution approach (Germain & Woolfson, 1968; Germain, Main & Woolfson, 1970, 1971) seems to be the most practical 'computer-based' method but is however limited in view of computational cost and, in unfavourable cases, of the number of Fourier syntheses to examine.

The need for a safe procedure to assign numerical values to the symbol used in the symbolic-addition method led us to the formulation of an appropriate test called 'the phase function' (Riche, 1970). We showed that the most probable combination of phases $\{\varphi_i\}$ belonging to a set of high $|E|$ values is given by the maximum of relation (6).

Later on, Schenk (1971) used some practical tests to select numerical values for the symbols. One of them (Q function) could be related to the phase function.

We recall briefly how we formulated relation (6) in § I. Use of the phase function and the change it produces in the symbolic-addition method is shown in § II. The practical procedure for phase determination is described in § III. In § IV results are discussed.

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I. Background

For simplicity, we assume that the space group is $P1$ and that all atoms are identical. The normalized structure factor E_H is then given by:

$$\mathbf{E}_H = \frac{1}{\sqrt{N}} \sum_{i=1}^N \exp(2\pi i \mathbf{H} \cdot \mathbf{r}_i),$$

\mathbf{H} : reciprocal lattice vector

\mathbf{r} : vector defining the position of the i th atom,

N : number of atoms in the unit cell.

The unitary structure factors are given by:

$$\mathbf{U}_H = \mathbf{E}_H / \sqrt{N}.$$

Let a set of reflexions of high $|E|$ values: $A = \{\varphi_i\}$ be given. Using the Sayre–Hughes formula (Sayre, 1952; Hughes, 1953) we can calculate the expected value V_H of E_H for a large number of reflexions belonging to a second set: B .

For $\mathbf{H} \in B$, V_H will be defined by:

$$\mathbf{V}_H = s_H \cdot \langle \mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i} \rangle_{\mathbf{K}_i} = \frac{s_H}{p_H} \sum_{i=1}^{p_H} \mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}. \quad (3)$$

In this expression $\mathbf{E}_{\mathbf{K}_i}$ and $\mathbf{E}_{\mathbf{H}-\mathbf{K}_i}$ belong to A ; p_H is the number of contributors ($\mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}$) and s_H is a scaling constant. We introduce for the i th contributor the notation:

$$\begin{aligned} v_{iH} &= \mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i} = a_i \exp(j\alpha_i) \\ a_i &= |\mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}| \\ \alpha_i &= \varphi_{\mathbf{K}_i} + \varphi_{\mathbf{H}-\mathbf{K}_i} \\ \delta_{iH} &= |\mathbf{V}_H - v_{iH}| \\ \Delta_{iH} &= |\mathbf{U}_H - v_{iH}|. \end{aligned}$$

These relations are illustrated in Fig. 1.

We wish to find the most probable combination of phases $\{\varphi_i\}$ through the determination of the most probable distribution of the vectors $\mathbf{E}_{\mathbf{K}_i} \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}$.

In the centrosymmetric case, the probability distribution of the product $\mathbf{E}_{\mathbf{K}} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}}$, for a given value of \mathbf{U}_H , was obtained by Cochran & Woolfson (1955). A similar distribution can be derived for a non-centrosymmetric space group: when $\mathbf{E}_{\mathbf{K}}$ and $\mathbf{E}_{\mathbf{H}-\mathbf{K}}$ have specified fixed values, it reduces to the well known circular normal distribution (Cochran, 1955).

We make the basic assumption that \mathbf{U}_H can be approximated in *phase* and in *modulus* by \mathbf{V}_H . Now considering the approximated distribution: $P(\delta_{iH}) \propto \exp(-\delta_{iH}^2)$, the most probable combination of all products $\mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}$,* and consequently the most probable combination of phases $\{\varphi_i\}$ can be obtained by a mean-square minimization of the expression:

$$S = \sum_{\mathbf{H} \in B} \sum_{i=1}^{p_H} \delta_{iH}^2. \quad (4)$$

S reduces to (Riche, 1970; 1972):

$$S = \sum_{\mathbf{H} \in B} \sum_{i=1}^{p_H} a_i^2 - \mathcal{F}(\varphi_1, \varphi_2, \dots, \varphi_n) \quad (5)$$

* It is assumed that the δ_{iH} 's are mutually independent.

$$\mathcal{F}(\varphi_1, \varphi_2, \dots, \varphi_n) = \sum_{\mathbf{H} \in B} \frac{G_H}{p_H} \left\{ \sum_{i=1}^{p_H} \sum_{j=1}^{p_H} a_i a_j \cos(\alpha_i - \alpha_j) \right\}_{\mathbf{H}} \quad (6)$$

and:

$$G_H = 2s_H - s_H^2 / \sqrt{N}. \quad (7)$$

S being always positive, it follows that: among all combinations of $\{\varphi_i\}$ the most probable are those that lead to the maximum of function (6).

When the modulus of \mathbf{V}_H is rescaled to the modulus of \mathbf{U}_H the scaling constant is independent of \mathbf{H} , and G_H can be ignored. Recently we introduced the generalized Sayre–Hughes formula (Hauptman, 1970):

$$\mathbf{E}_H \approx \frac{|\mathbf{E}_H| \langle \mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i} \rangle_{\mathbf{K}_i}}{\left\langle \frac{|\mathbf{E}_{\mathbf{K}_i} \cdot \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}|}{I_0(A_i)} \right\rangle_{\mathbf{K}_i}} \quad (8)$$

where $I_r(X)$ is a modified Bessel function of order r and:

$$A_i = \frac{2}{\sqrt{N}} |\mathbf{E}_H \mathbf{E}_{\mathbf{K}_i} \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}|.$$

In formula (7) the value of s_H is then given by:

$$s_H = \frac{|\mathbf{E}_H|}{\sqrt{N} \left\langle \frac{|\mathbf{E}_{\mathbf{K}_i} \mathbf{E}_{\mathbf{H}-\mathbf{K}_i}|}{I_0(A_i)} \right\rangle_{\mathbf{K}_i}}. \quad (9)$$

Relation (6), called the phase function, depends only on the phases of the A set.

It is important to note that \mathcal{F} is a 'structure-invariant quantity'. This means that \mathcal{F} is independent of the choice of origin and reference frame.

Universal values of \mathcal{F} are put on an 'absolute scale' by:

$$\tau = \frac{\mathcal{F}(\varphi_1, \varphi_2, \dots, \varphi_n)}{\sum_{\mathbf{H} \in B} \frac{G_H}{p_H} \left\{ \sum_{i=1}^{p_H} a_i a_j \right\}_{\mathbf{H}}} \quad (\tau \leq 1). \quad (10)$$

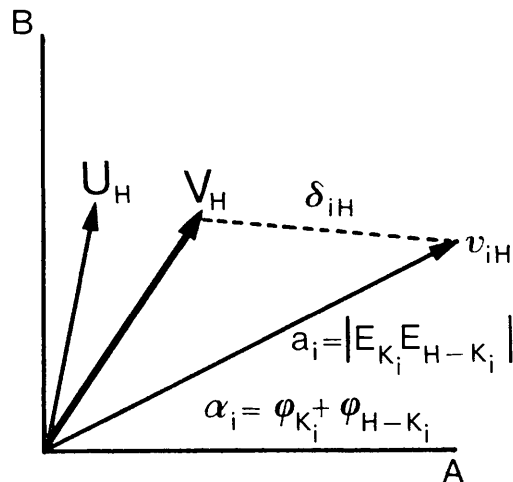


Fig. 1. Phase notation.

The value associated with a maximum is a measure of the internal consistency of the phase combination (Riche, 1971). Defining $\varphi_i \in A$ by:

$$\varphi_i = m_1\pi + \sum_{j=1}^r m_j x_j \quad (11)$$

where x_j are the r unknown phases of A , we obtained (Riche, 1972):

$$\mathcal{F}(\varphi_1, \varphi_2, \dots, \varphi_n) = \sum_k P_k \cdot \cos(M_1\pi + \sum_{j=1}^r M_j x_j) \quad (12)$$

Thus the phase function can be computed as an r -dimensional Fourier map and peaks in the map give the most probable values for the set of phases $\{\varphi_i\}$.

II. Introduction of the phase function in the symbolic-addition method

Maximizing relation (6) with a large number of unknowns in the A set would take too much computing time. Symbolic addition provides a useful way to increase the number of known phases in the A set starting from a small number of symbols. The application of \mathcal{F} is quite powerful in determining the numerical values of the unknown symbols. The reliability of the results depends mainly on the building-up of the A set and the introduction of \mathcal{F} in the symbolic-addition procedure leads to some modifications.

(1) Restriction of the 'symbolic addition'

Restriction of the use of equation (1) along with an increased number of symbols gives more reliable results (Riche, 1971).

Let us call Φ^0 the starting set, Φ^1 the set of phases generated by $\varphi_{\mathbf{H}}^1 = \varphi_{\mathbf{K}}^0 + \varphi_{\mathbf{H}-\mathbf{K}}^0$. This operation represents the first cycle of symbolic addition. Another cycle of symbolic addition can be performed, but through our personal experience, results are more reliable if we keep the number of symbolic-addition cycles to the lowest number possible: practically only one. Then $\{\Phi^0 + \Phi^1\}$ constitutes the A set for the computation of the phase function.

(2) Weighted symbolic addition

Generally the acceptance of a phase obtained through equation (1) is subject to the calculated variance associated with the triple product:

$$A = 2|\mathbf{E}_{\mathbf{H}}\mathbf{E}_{\mathbf{K}}\mathbf{E}_{\mathbf{H}-\mathbf{K}}|/VN \text{ (Karle \& Karle, 1966).}$$

This limits strongly the number of accepted phases. Without this restriction and starting from a set of symbolic phases Φ^0 , we can attain a large number of phases giving numerous but weakly correlated pieces of information.

By accepting all these phases and by giving them a 'weight', it is possible to include in the first cycle of symbolic addition all the information within reach. Let $w = 1$ be the weight of each reflection in the starting

set. We define:

$$w_{\mathbf{H}} = w_{\mathbf{K}}w_{\mathbf{H}-\mathbf{K}}w_r, \quad (13)$$

the weight of reflection \mathbf{H} obtained by (1). w_r reflects the reliability of equation (1) and is smaller than unity. The usual weighting scheme is: $w_r = \text{probability that}$

$$(\varphi_{\mathbf{K}} + \varphi_{\mathbf{H}-\mathbf{K}}) - 45^\circ < \varphi_{\mathbf{H}} < (\varphi_{\mathbf{K}} + \varphi_{\mathbf{H}-\mathbf{K}}) + 45^\circ.$$

(3) Introduction of multiplicity

Often, in the symbolic addition, a phase $\varphi_{\mathbf{H}}$ is given by two or more relationships leading, as below, to two or more symbolic expressions:

$$\begin{aligned} \varphi_{\mathbf{H}} &\simeq \varphi_{\mathbf{K}_1} + \varphi_{\mathbf{H}-\mathbf{K}_1} = a \\ \varphi_{\mathbf{H}} &\simeq \varphi_{\mathbf{K}_2} + \varphi_{\mathbf{H}-\mathbf{K}_2} = b. \end{aligned}$$

Without any information about the values of a and b we cannot use equation (2). If we choose one of these two values (the most probable one for instance) we lose the second one. A simple way to keep both of them through successive cycles of symbolic addition is to include this reflexion twice, using the two phases with a multiplicity factor which would be in the above example 0.5. More generally, in the cycle n of the symbolic addition, let a phase be given by r contributors $(\varphi_{\mathbf{K}} + \varphi_{\mathbf{H}-\mathbf{K}})_j (j=1, r)$. Consequently we shall include this reflexion r times in the cycle $n+1$. For the reflexions belonging to the starting set the multiplicity factor is unity. The reflexion number j will have then the phase $(\varphi_{\mathbf{K}} + \varphi_{\mathbf{H}-\mathbf{K}})_j$, the weight $(w_{\mathbf{K}}w_{\mathbf{H}-\mathbf{K}}w_r)_j$ and the multiplicity factor:

$$m_j = \frac{(m_{\mathbf{K}}m_{\mathbf{H}-\mathbf{K}})_j}{\sum_{j=1}^r (m_{\mathbf{K}}m_{\mathbf{H}-\mathbf{K}})_j} \quad (14)$$

(4) Weighted phase function

According to the above, it appears logical to include the notion of weight and multiplicity into the phase function. Each phase of the A set is characterized by $w_{\mathbf{H}}$ and $m_{\mathbf{H}}$. Let us define the weight of a contributor $(E_{\mathbf{K}}E_{\mathbf{H}-\mathbf{K}})_i$ to the Sayre-Hughes equation by:

$$\Omega_{i\mathbf{H}} = (w_{\mathbf{K}}w_{\mathbf{H}-\mathbf{K}}m_{\mathbf{K}}m_{\mathbf{H}-\mathbf{K}})_i \quad (15)$$

It can be shown (Riche, 1972) that relation (6) takes the following form:

$$\begin{aligned} \mathcal{F}(\varphi_1, \varphi_2, \dots, \varphi_n) \\ = \sum_{\mathbf{H} \in \mathbf{B}} \frac{G_{\mathbf{H}}}{c_{\mathbf{H}}} \left\{ \sum_{i=1}^{PH} \sum_{j=1}^{PH} \Omega_i \Omega_j a_i a_j \cos(\alpha_i - \alpha_j) \right\}_{\mathbf{H}} \end{aligned} \quad (16)$$

where:

$$c_{\mathbf{H}} = \sum_{i=1}^{PH} \Omega_{i\mathbf{H}} \quad (17)$$

represents a normalizing constant.

These definitions offer a direct way for programming symbolic addition. The final aim being to obtain, from the starting Φ^0 , the best set of phases A to compute

the phase function. This weighted symbolic addition is still limited to one or exceptionally two cycles.

III. Procedure

Choice of the starting set

This set of symbolic phases is built up in the usual way (Karle & Karle, 1966; Germain, Main & Woolfson, 1970), and must include origin-defining reflexions. In most cases nearly all these origin phases have numerical values but it is not always necessary as we shall see below.

Symbolic addition and computation of the phase function

Starting with a Φ^0 set, one cycle of weighted symbolic addition gives another set Φ^1 of reflexions. The computation of the weighted phase function follows immediately, all the phases $\{\Phi^0 + \Phi^1\}$ constituting the A set. Relation (6) is calculated for every combination of phases in an r -dimensional space Fourier map (r = number of symbols in Φ^0) with intervals of $\pi/4$ for the phase of a general reflexion.

Interpretation of the phase function

Emerging solutions. In this case each maximum of \mathcal{F} is well resolved. Through our personal experience their number rarely exceeds four.

Lack of resolution. In certain directions the maxima are not defined. By adding new symbols we improve the resolution and precision of the numerical values of the older symbols (Riche, 1972). In some cases it is easier to run a second cycle of symbolic addition before the computation of \mathcal{F} . This case arises especially in the monoclinic space group $P2_1$.

Tangent refinement. For each selected combination of phases given by \mathcal{F} a tangent refinement is performed. For the best refinements we calculate the E map.

IV. Results and discussion

A dozen unknown structures were solved in this manner (Table 1). We have selected two typical cases, each exhibiting special features.

(1) Determination of the structure of triphenylmethane

This compound (d , Table 1), crystallizes in space group $Pna2_1$ with two molecules in the asymmetric unit. The starting set is listed in Table 2.

Table 2. Starting set for triphenylmethane

h	k	l	E	φ	
15	9	0	3.73	0	
19	6	0	3.50	0	
3	2	1	2.17	0	
16	8	0	3.55	a	0 or π
3	4	3	2.51	b	0 to π
3	9	4	2.48	c	0 to 2π
16	1	4	2.67	d	0 to 2π
3	15	0	2.64	e	0 or π

The phase function calculated after two cycles of unweighted symbolic addition showed 8 maxima (Riche, 1972). 1280 combinations of phases were computed giving 2 numerical values for the symbols a and e (0 or π), 8 for c and d (0, $\pi/4$, ..., $7\pi/4$), and 5 for b (0, $\pi/4$, $\pi/2$, $3\pi/4$, π).

As the reference frame had not been fixed, maxima appear for the values 0 or π for the symbols b , c and d . To fix the reference frame it was enough to alter co-

Table 1. Unknown non-centrosymmetric structures solved using the phase function*

Compound	Formula	Space group	N/Z	Number of symbols in the starting set		Number of phase combinations in \mathcal{F}		Rank of the maximum in \mathcal{F} of the true solution
				Total	'Space'	calculated	selected	
a	$C_{13}H_{15}NO_2$	$P2_12_12_1$	16	4	2	256	4	1
b	$C_{22}H_{20}O_7$	$P2_12_12_1$	29	3	1	32	2	1
c	$C_{14}H_{14}O_4$	$P2_12_12_1$	18	5	3	1280	4	1
d	$C_{19}H_{15}$	$Pna2_1$	38	5	3	1280	8	1
e	$C_{31}H_{30}N_4O_2$ $2CH_3OH$	$P2_12_12_1$	41	5	3	2048	4	3
f	$C_9H_{10}N_2O_5$	$P2_12_12_1$	16	6	3	2560	4	1
g	$C_{22}H_{30}N_2O_4$	$P2_12_12_1$	28	4	4	2560	4	1
h	$C_{12}H_{26}O_4S_3$	$P2_1$	38	7	4	32768	4	4
i	$C_9H_{11}O_3$	$P2_1$	12	6	5	32768	4	1

- (a) Phyllochrysin. Riche (1970, 1972).
 (b) Isocollybolide. Pascard-Billy (1970, 1972).
 (c) Cyclohexane diepoxyde. Riche (1972).
 (d) Triphenylmethane. Riche (1972). Pascard-Billy (1972).
 (e) Isocinchophyllamine. Guilhem (To be published).
 (f) Cyclobutane uracyl. Pascard-Billy (1973).
 (g) Ervatamine. Riche (To be published).
 (h) 2-S-Ethyl-2-thio-D-mannose diethylthioacetal. Ducruix (To be published).
 (i) Phenyl hydracrylic acid. Cesario (To be published).

* 5 structures were solved in space group $P2_1/c$ and in space group $C2/c$: Cesario & Pascard-Billy (To be published): and De Sagey (To be published).

ordinates from the maximum in any of the three directions b , c or d . This was done by making $b = \pi/4$. The numerical values of c and d were thus determined. Tangent formula and E map were used in the usual way.

(2) *Determination of the structure of 2-S-ethyl-2-thio-D-mannose diethyldithioacetal*

This compound (h , Table 1) crystallizes in space group $P2_1$ with two molecules in the asymmetric unit; only $h0l$ reflexions with $h+l$ even were present in a list of E greater than 1.5. The starting set Φ^0 is listed in Table 3.

Table 3. *Starting set for 2-S-ethyl-2-thio-D-mannose diethyldithioacetal*

h	k	l	E	φ	
4	1	10	3.43	0	
-3	0	3	3.07	0	
-6	2	3	2.21	$\pi/4$	origin + enantiomorph
1	0	15	2.88	a	0 or π
-4	0	4	2.66	b	0 or π
-1	0	9	3.18	c	0 or π
-3	5	1	2.50	d	0 to 2π
6	4	2	3.08	e	0 to 2π
-8	3	1	2.40	f	0 to 2π
-1	4	9	2.82	g	0 to 2π

One cycle of weighted symbolic addition was run and after the examination of the phase function it appeared that a second cycle would be necessary. All the invariants involved in the first cycle were computed using the formula of Messenger & Tsoucaris (1972). One of these was eliminated because its computed value was greater than $\pi/4$, and a second cycle of weighted symbolic addition was run. The function gave four well resolved maxima. The corresponding solutions were refined by tangent formula and the structure appeared on one of the four E maps.

Considering now all these results we can point out the main features of the procedure:

(1) Starting with a large number of phase combinations it reduces the number of the possible solutions to a very small number. This leads to an even smaller number of E maps to be examined.

(2) Usually, the solution of the structure corresponded to the first maximum of \mathcal{F} .

The weighted symbolic addition and the weighted phase function are linked together in the same program, *DEVIN* written in Fortran IV. Computing time depends on the number of phase combinations we are

dealing with. For structure f in Table 1, the entire run (one cycle of symbolic addition and the computation of \mathcal{F}) takes 9 seconds on an I. B. M. 370/165. For structure i , it takes around 30 seconds. 4096 phase combinations were computed in the first case and 32768 in the second one.

Conclusion

We have shown that the phase function used in a modified symbolic-addition method gives the most probable numerical values of a set of symbolic phases. The principal advantages of this procedure are: its simplicity, the speed of the corresponding program and its efficiency.

As the symbols are more easily determined, we can increase their number with the consequence that the tangent refinement can be started with each phase determined with a lower variance. Solving structures of larger molecules would need a larger number of symbols. This extension of the symbolic addition method offers a simple way to manipulate many symbols to match the complexity of the structures.

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References

- COCHRAN, W. (1955). *Acta Cryst.* **8**, 473-478.
 COCHRAN, W. & WOOLFSON, M. M. (1955). *Acta Cryst.* **8**, 1-12.
 GERMAIN, G., MAIN, P. & WOOLFSON, M. M. (1970). *Acta Cryst.* **B26**, 274-285.
 GERMAIN, G., MAIN, P. & WOOLFSON, M. M. (1971). *Acta Cryst.* **A27**, 368-376.
 GERMAIN, G. & WOOLFSON, M. M. (1968). *Acta Cryst.* **B24**, 91-96.
 HAUPTMAN, H. (1970). Summer A.C.A. Meeting (U.S.A.), August 1972.
 HUGHES, E. W. (1953). *Acta Cryst.* **6**, 871.
 KARLE, J. & KARLE, I. L. (1966). *Acta Cryst.* **21**, 849-859.
 MESSAGER, J. C. & TSOUCARIS, G. (1972). *Acta Cryst.* **A28**, 482-484.
 PASCARD-BILLY, C. (1970). *Chem. Commun.* p. 1722.
 PASCARD-BILLY, C. (1972). *Acta Cryst.* **B28**, 331-337.
 PASCARD-BILLY, C. (1973). *Acta Cryst.* In the press.
 RICHE, C. (1970). *C. R. Acad. Sci. Paris*, **271**, 396-398.
 RICHE, C. (1971). *C. R. Acad. Sci. Paris*, **272**, 1422-1424.
 RICHE, C. (1972). Thesis, Paris. A.O. C.N.R.S.: 6513.
 SAYRE, D. (1952). *Acta Cryst.* **5**, 60-65.
 SCHENK, H. (1971). *Acta Cryst.* **B27**, 2037-2042.