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Fourth and Higher Order Inequalities

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An approach is made to the problems which arise in the application of inequalities of order four or more. The main features of the fourth-order case are examined in a way which lends itself to extension to orders exceeding four, and an example is given of the application of fourth-order inequalities to a small problem.

Introduction and notation

Karle & Hauptman (1950) showed that the necessary and sufficient condition that the electron density be nowhere negative is that all determinants of the form

$$\begin{vmatrix} F(\mathbf{o}) & F(\mathbf{h}_i) & F(\mathbf{h}_j) & \ldots F(\mathbf{h}_{n-1}) \\ F(\bar{\mathbf{h}}_i) & F(\mathbf{o}) & F(\mathbf{h}_j-\mathbf{h}_i) & \ldots F(\mathbf{h}_{n-1}-\mathbf{h}_i) \\ F(\bar{\mathbf{h}}_j) & F(\mathbf{h}_i-\mathbf{h}_j) & F(\mathbf{o}) & \ldots F(\mathbf{h}_{n-1}-\mathbf{h}_j) \\ \vdots & \vdots & \vdots & \vdots \\ F(\bar{\mathbf{h}}_{n-1}) & F(\mathbf{h}_i-\mathbf{h}_{n-1}) & F(\mathbf{h}_j-\mathbf{h}_{n-1}) & \ldots F(\mathbf{o}) \end{vmatrix} \quad (1)$$

shall be positive or zero. (Here $\mathbf{h}_i$ is any vector index and $\bar{\mathbf{h}}_i$ its inverse). These writers also showed that all possible inequalities based upon the positivity of ϱ are contained in these determinantal inequalities. As is well known, U may replace F in these determinants since the U's also have a positive transform, and this results in a strengthening of the inequalities. Following Kitaigorodski (1957, 1961) we call such determinants containing U's 'connecting determinants'.

Various writers, notably MacGillavry (1950) and Goedkoop (1950) have considered the systematic incorporation of crystallographic symmetry into such inequalities, whilst Bouman (1956) and Taguchi & Naya (1958) have shown that connecting determinants can be expressed in more compact form if the crystal is centrosymmetric and if the choice of indices $\mathbf{h}$ is such as to make the determinant symmetrical about both diagonals. They show, for example, that under a linear transformation the inequality

$$\begin{vmatrix} 1 & U(\mathbf{h}) & U(\mathbf{h}') & U(\mathbf{h}+\mathbf{h}') \\ U(\bar{\mathbf{h}}) & 1 & U(\mathbf{h}'-\mathbf{h}) & U(\mathbf{h}') \\ U(\bar{\mathbf{h}}') & U(\bar{\mathbf{h}}'-\bar{\mathbf{h}}) & 1 & U(\mathbf{h}) \\ U(\bar{\mathbf{h}}+\bar{\mathbf{h}}') & U(\bar{\mathbf{h}}') & U(\bar{\mathbf{h}}) & 1 \end{vmatrix} \geq 0 \quad (2)$$

is equivalent to the two inequalities

$$\begin{vmatrix} [1\pm U(\mathbf{h}+\mathbf{h}')] & [U(\mathbf{h})\pm U(\mathbf{h}')] \\ [U(\mathbf{h})\pm U(\mathbf{h}')] & [1\pm U(\mathbf{h}'-\mathbf{h})] \end{vmatrix} \geq 0 \quad (3)$$

recognizable as the 'sum and difference' inequalities in determinantal form. (This is also shown in the Appendix without the use of transformation ideas). In general a doubly symmetrical connecting determinant of order $2n$ (or $2n+1$) is reducible to a pair of determinantal inequalities of order n (or one of order n and one of order $n+1$), each element of which contains two U's instead of one (except for 1 row and 1 column of the larger determinant in the latter case).

In recent years the direct approach to crystal-structure determination has most often been made through probability methods, inequalities being too weak to solve any but fairly simple structures. Kitaigorodski (1957, 1961), however, has provided some stimulus to the study of inequalities of higher orders and has clearly shown that their strength increases as the order n of the determinant increases and should reach a strength comparable to the existing probability methods. These points are returned to in our discussion.

Von Eller (1955, 1960, 1961, 1962) has also made an extensive study of these inequalities, making use of the geometrical relationships which arise among the angles φ defined by $\varphi = \text{arc}\cos U$.

In the present paper we are concerned with the problems which arise in the application of inequalities of all orders by computer methods, with special attention to the fourth-order case, and with the objective of finding a unified approach.

Since the unknown quantities to be found are the signs of the U's it is apparent that the number of these which are involved in any given inequality is a more important criterion of the complexity of the inequality than is the order of the determinant employed. For this reason the compact form (3) has no advantage over the less compact form (2), and we formulate what follows in terms of the general form (1) treating (2) as a special case which is included in the general form.

In this paper we write

$$u_{ij} = |U(\mathbf{h}_i-\mathbf{h}_j)|,\; s_{ij} = U(\mathbf{h}_i-\mathbf{h}_j)/|U(\mathbf{h}_i-\mathbf{h}_j)|$$

and define $\mathbf{h}_0 = 0$. Thus s_{ij} is the complex conjugate of s_{ji}, $u_{ij} = u_{ji}$ and the connecting determinant of order n becomes

$$D_n = \begin{vmatrix} 1 & s_{i0}u_{i0} & s_{j0}u_{j0} & \dots & s_{(n-1)0}u_{(n-1)0} \\ s_{0i}u_{0i} & 1 & s_{ji}u_{ji} & \dots & s_{(n-1)i}u_{(n-1)i} \\ s_{0j}u_{0j} & s_{ij}u_{ij} & 1 & \dots & s_{(n-1)j}u_{(n-1)j} \\ \vdots & \vdots & \vdots & & \vdots \\ s_{0(n-1)}u_{0(n-1)} & s_{i(n-1)}u_{i(n-1)} & s_{j(n-1)}u_{j(n-1)} & \dots & 1 \end{vmatrix} \tag{4}$$

in which it is convenient to regard the $(n-1)$ off-diagonal elements on the top row or first column as being independently chosen, the remaining $\frac{1}{2}(n-1)(n-2)$ elements each side of the leading diagonal being dependent upon this choice and being termed *dependent elements*.

Now the expansion of a determinant of order n contains $n!$ *terms*, each of which is the product of n elements together with the appropriate sign. The n elements which occur in any term are such that no two are from the same row or the same column; thus in every term the subscript i occurs just once in the left-hand position and just once in the right-hand position. Thus the sum of the vector indices of the elements involved in any term is independent of $\mathbf{h}_i$. This is true for all i; it follows that the sum of the vector indices of the elements involved in each and every term is zero, *i.e.* every term is a structure invariant.*

Thus typical terms in the expansion of a fifth-order connecting determinant would have sign

$$(-1)^p s_{0i}s_{ik}s_{kj}s_{jl}s_{l0} \quad \text{or perhaps} \quad (-1)^p s_{0i}s_{kk}s_{ij}s_{ji}s_{l0}$$
$$\text{which is} \quad (-1)^p s_{0i}s_{ij}s_{jl}s_{l0}$$

where p is the total number of inversions of order among the subscripts (Muir & Metzler, 1960, p. 14). *Invariant* products of signs such as these will be called *tri-products, tetra-products, etc*, according to the number of non-trivial signs involved. Of such products the tri-product is the most useful since it is the simplest which is non-trivial and is already familiar in crystallography. A tri-product can be made up from any three different indices, such as $s_{ij}s_{jk}s_{ki}$; thus $n!/[3!(n-3)!]$ such tri-products may be formed for a determinant of order n. Tri-products which include the suffix zero will be called *primary tri-products* as these have special importance in what follows. Clearly, there is one primary tri-product for each *pair* of indices ij, such as $s_{0i}s_{ij}s_{j0}$, or one for each dependent element. Furthermore, since $s_{ji}s_{ij}=1$, any invariant sign product may be expressed as the product of a number of primary tri-products, as pointed out by Kitaigorodski, *e.g.*

$$s_{0i}s_{ik}s_{kj}s_{jl}s_{l0} = s_{0i}s_{ik}s_{k0}\,.\,s_{0k}s_{kj}s_{j0}\,.\,s_{0j}s_{jl}s_{l0}$$
$$s_{ik}s_{kj}s_{jl}s_{li} = s_{0i}s_{ik}s_{k0}\,.\,s_{0k}s_{kj}s_{j0}\,.\,s_{0j}s_{jl}s_{l0}\,.\,s_{0l}s_{li}s_{i0}\,.$$

For the purposes of evaluating the connecting determinant for a centrosymmetric structure we may regard each s_{ij} as an independent variable having the values ± 1, and therefore the primary tri-products may alternatively, and more conveniently, be regarded as independent variables. Since all invariant sign products may be expressed in terms of these it follows that a connecting determinant of order n has $2^{\frac{1}{2}(n-1)(n-2)}$ possible values, of which the negative ones correspond to disallowed sign combinations.

The remaining

$$\left[\frac{n!}{3!(n-3)!} - \tfrac{1}{2}(n-1)(n-2)\right] = \frac{(n-1)!}{6(n-4)!}$$

tri-products, such as $s_{ij}s_{jk}s_{ki}$, will be termed secondary tri-products. Information concerning these is just as valuable for the purposes of sign determination as is information concerning the primary tri-products; however they have the role of dependent variables.

If in (4) we multiply the ith row by s_{i0} and the ith column by s_{0i} then the value of the determinant is unchanged since it has been multiplied by $s_{i0}s_{0i}$. When all the rows and columns are so multiplied we have

$$D_n = \begin{vmatrix} 1 & u_{i0} & u_{j0} & \dots & u_{(n-1)0} \\ u_{0i} & 1 & s_{0j}s_{ji}s_{i0}u_{ji} & \dots & s_{0(n-1)}s_{(n-1)i}s_{i0}u_{(n-1)i} \\ u_{0j} & s_{0i}s_{ij}s_{j0}u_{ij} & 1 & \dots & s_{0(n-1)}s_{(n-1)j}s_{j0}u_{(n-1)j} \\ \vdots & \vdots & \vdots & & \vdots \\ u_{0(n-1)} & s_{0i}s_{i(n-1)}s_{(n-1)0}u_{i(n-1)} & s_{0j}s_{j(n-1)}s_{(n-1)0}u_{j(n-1)} & \dots & 1 \end{vmatrix}$$

in which the only sign products appearing are the $\frac{1}{2}(n-1)(n-2)$ primary tri-products which will henceforth be identified according to the scheme

$$\left|\begin{array}{cccccccc} \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & & \\ \cdot & \cdot & \alpha & \beta & \delta & \eta & & \\ \cdot & \bar{\alpha} & \cdot & \gamma & \varepsilon & \theta & & \\ \cdot & \bar{\beta} & \bar{\gamma} & \cdot & \zeta & \iota & \dots & \\ \cdot & \bar{\delta} & \bar{\varepsilon} & \bar{\zeta} & \cdot & \varkappa & & \\ \cdot & \bar{\eta} & \bar{\theta} & \bar{\iota} & \bar{\varkappa} & \cdot & & \\ & & \vdots & & & & & \end{array}\right.$$

* If $U(\mathbf{h})$ becomes $U'(\mathbf{h})$ when the crystallographic origin is shifted by $\delta\mathbf{r}$ then $U'(\mathbf{h}) = U(\mathbf{h}) \exp[2\pi i \delta\mathbf{r}\,.\,\mathbf{h}]$

$$\therefore\ \Pi U'(\mathbf{h}) = \exp[2\pi i \delta\mathbf{r}\,.\,\Sigma\mathbf{h}]\,\Pi U(\mathbf{h})$$

which is independent of the choice of origin if $\Sigma\mathbf{h}=0$.

in which the dots represent the independent and diagonal elements, *e.g.* $\alpha = s_{0j}s_{ji}s_{i0}$, and $\bar{\alpha}$ is the complex conjugate of α. In what follows we shall be concerned only with centrosymmetric structures, however, so that this distinction may be ignored.

The majority of these features have already been pointed out by Kitaigorodski (1957) but it should also be emphasized that the distinction drawn here between primary and secondary tri-products is a matter of choice (their roles may be interchanged by permuting rows and columns in D_n), the point being that only $\frac{1}{2}(n-1)(n-2)$ of the tri-products are independent and one must therefore choose which ones to regard as independent variables. Algebraically, we must expect all tri-products to have equivalent status and this is apparent in any expansion of D_n.

Preliminaries for the fourth-order case

The fourth-order connecting determinant may be expanded as

$$0 \le D_4 = \begin{vmatrix} 1 & s_{i0}u_{i0} & s_{j0}u_{j0} & s_{k0}u_{k0} \\ s_{0i}u_{0i} & 1 & s_{ji}u_{ji} & s_{ki}u_{ki} \\ s_{0j}u_{0j} & s_{ij}u_{ij} & 1 & s_{kj}u_{kj} \\ s_{0k}u_{0k} & s_{ik}u_{ik} & s_{jk}u_{jk} & 1 \end{vmatrix}$$

$$\begin{aligned} = 1 &- u_{i0}^2 - u_{j0}^2 - u_{k0}^2 - u_{ji}^2 - u_{ki}^2 - u_{kj}^2 \\ &+ u_{ji}^2 u_{k0}^2 + u_{ki}^2 u_{j0}^2 + u_{kj}^2 u_{i0}^2 \\ &+ 2\{\alpha u_{0j}u_{ji}u_{i0} + \beta u_{0k}u_{ki}u_{i0} + \gamma u_{0k}u_{kj}u_{j0} \\ &\quad + (\alpha\beta\gamma)u_{ij}u_{jk}u_{ki} \\ &\quad - (\beta\gamma)u_{0i}u_{ik}u_{kj}u_{j0} - (\gamma\alpha)u_{0i}u_{ij}u_{jk}u_{k0} \\ &\quad - (\alpha\beta)u_{0j}u_{ji}u_{ik}u_{k0}\} \end{aligned} \qquad (5)$$

in which the product of two primary tri-products is a tetra-product and the product $(\alpha\beta\gamma)$ is the secondary tri-product. The symbols α, β, *etc*, will always be enclosed in round brackets when their product is intended.

Suppose we set $\alpha = +$, $\beta = \gamma = -$ and calculate D_4 and find it negative, we may then make a *statement*, written $\langle\alpha|\beta\gamma\rangle$, which says

'If α is $+$ and β and γ $-$ then we get a violation'.

In this form the statement tells us what *must not be*. All remaining possibilities constitute what *may be*. However, we wish to find what *must be* and from the outset we phrase the statement positively, thus

'If α is $+$ then at least one of β and γ is $+$'

or

'If β and γ are $-$ then α is $-$'

or in general

'If every *item* on the left is $+$
then at least one item on the right is $+$'

or

'If every item on the right is $-$
then at least one item on the left is $-$'

in which an item is any invariant sign product (of whatever order), which appears in such a statement.*

Such a statement arising from a single test of D_n will be called a *primitive statement*. Evidently primitive statements are always conditional (except for $n=3$) and may be read in a variety of ways; any item may be transposed with change of sign, thus

$$\langle\alpha|\beta\gamma\rangle \equiv \langle\alpha(-\beta)|\gamma\rangle$$

so that any combination of items may be brought into the 'if' part, the remainder appearing in the 'then' part of the statement. Statements arising from a single determinant (*i.e.* a single choice for $\mathbf{h}_i$, $\mathbf{h}_j$, $\mathbf{h}_k$ *etc*) will be termed *associated statements*. There are eight primitive associated statements for the fourth-order case corresponding to the eight values of D_4:

$$\langle|\alpha\beta\gamma\rangle,\ \langle\alpha|\beta\gamma\rangle,\ \langle\beta|\gamma\alpha\rangle,\ \langle\gamma|\alpha\beta\rangle,$$
$$\langle\alpha\beta|\gamma\rangle,\ \langle\gamma\alpha|\beta\rangle,\ \langle\beta\gamma|\alpha\rangle,\ \langle\alpha\beta\gamma|\rangle$$

of which the first will be termed the *leading statement* and the last the *trailing statement*.

Examination of (5) shows that D_4 is smallest when $\alpha = \beta = \gamma = -$; therefore none of these statements can occur in the absence of the leading statement (this is general for all n). Further, they cannot all arise at once since no possible sign combination would then remain. These appear to be the sole constraints, so that $2^7 - 1 = 127$ combinations of primitive associated statements are possible for $n=4$.

Finally we define the length, l, of a statement as the number of items appearing in it.

It is now our objective to show, in as general a way as possible, how to reduce any combination of primitive associated statements to its most concise form, and especially to derive statements of length 1 which are unconditional.

Digestion of associated statements

I. *The digest regarded as the union of primitive statements*

We define the digest of any given statements as the shortest statement or statements which together completely contain the given statements. We consider first a pair of statements which differ only in respect of a single item which occurs on opposite sides of the given statements, *e.g.*

$$\langle\alpha\beta\gamma|\delta\varepsilon\zeta\rangle \quad \text{and} \quad \langle\alpha\beta|\gamma\delta\varepsilon\zeta\rangle$$

which may arise from a fifth-order inequality. If we rearrange these statements so that the unique item, γ, appears alone on the left, then they read

* If the 'if' part of a statement contains no item it is to be read as 'in any case ...', *e.g.* $\langle|\alpha\beta\gamma\rangle$ means 'in any case at least one of α, β, γ is positive, $\langle\alpha\beta\gamma|\rangle$ means 'at least one is negative'.

$$\langle\gamma|(-\alpha)(-\beta)\delta\varepsilon\zeta\rangle \text{ and } \langle(-\gamma)|(-\alpha)(-\beta)\delta\varepsilon\zeta\rangle;$$

thus whatever the sign of γ, at least one of the items on the right must be $+$, and the union, or digest, of the given statements is

$$\langle|(-\alpha)(-\beta)\delta\varepsilon\zeta\rangle \text{ usually written } \langle\alpha\beta|\delta\varepsilon\zeta\rangle.$$

This illustrates the principal rule for the combination of statements, which will be called the *cross-over rule*, that if two statements differ in this way then the digest is obtained from either statement by deleting the item which crosses over, and the digest is complete in the sense that it completely contains the truth of the given statements.

In order to broaden the applicability of the cross-over rule we consider next the *equivalence rule*. It has already been pointed out that by transposing and changing the sign of an item any combination of items may be brought into the left-hand part of the statement, which may be regarded as the 'if' part, so that any statement containing two or more items may be arranged to contain two items in the 'if' part. Suppose the 'if' part of a statement says 'If α and β are positive ...', then it is already known that $(\alpha\beta)$ is positive also, and of the three items α, β and $(\alpha\beta)$ any two will do to specify the condition. This leads to the general formulation of the equivalence rule as follows:

'Any item in a statement may be replaced by the product of itself with any second item in the statement, the product being transposed to the opposite partition if the second item occurs in the right-hand partition, and is not transposed if the second item occurs in the left-hand one'.

For example α may be replaced by $(\alpha\beta)$ in the following

$$\langle|\alpha\beta\gamma\rangle \equiv \langle(\alpha\beta)|\beta\gamma\rangle$$

and $(\alpha\beta)$ may again be replaced

$$\langle(\alpha\beta)|\beta\gamma\rangle \equiv \langle|(\alpha\beta\gamma)\beta\gamma\rangle;$$

or again

$$\langle\alpha|\beta\gamma\rangle \equiv \langle\alpha|(\alpha\beta)\gamma\rangle.$$

Now consider any two statements containing the same items, for example $\langle\alpha\boldsymbol{\beta}|\boldsymbol{\gamma\delta\varepsilon}\zeta\rangle$ and $\langle\alpha\boldsymbol{\gamma\delta\varepsilon}|\boldsymbol{\beta}\zeta\rangle$ in which the items which cross over are in bold type, and replace the first bold-faced item in the first statement by its product with the second, the second by its product with the third, and so on, and treat the second statement in the corresponding way, thus

$$\begin{aligned}\langle\alpha\boldsymbol{\beta}|\boldsymbol{\gamma\delta\varepsilon}\zeta\rangle &\equiv \langle\alpha|(\beta\gamma)\boldsymbol{\gamma\delta\varepsilon}\zeta\rangle \\ &\equiv \langle\alpha(\gamma\delta)|(\beta\gamma)\boldsymbol{\delta}\varepsilon\zeta\rangle \equiv \langle\alpha(\gamma\delta)(\delta\varepsilon)|(\beta\gamma)\boldsymbol{\varepsilon}\zeta\rangle\end{aligned}$$

and

$$\begin{aligned}\langle\alpha\boldsymbol{\gamma\delta\varepsilon}|\boldsymbol{\beta}\zeta\rangle &\equiv \langle\alpha\gamma\boldsymbol{\delta\varepsilon}|(\beta\gamma)\zeta\rangle \\ &\equiv \langle\alpha(\gamma\delta)\boldsymbol{\delta\varepsilon}|(\beta\gamma)\zeta\rangle \equiv \langle\alpha(\gamma\delta)(\delta\varepsilon)\boldsymbol{\varepsilon}|(\beta\gamma)\zeta\rangle\end{aligned}$$

then the two given statements have been converted to equivalent forms containing the same items, only one of which crosses over, so that the cross-over rule may be applied. Thus

$$\langle\alpha\beta|\gamma\delta\varepsilon\zeta\rangle.\langle\alpha\gamma\delta\varepsilon|\beta\zeta\rangle \supset \langle\alpha(\gamma\delta)(\delta\varepsilon)|(\beta\gamma)\zeta\rangle,$$

where $\supset$ denotes 'implies that'.

The process is evidently general; thus any two statements of length l containing the same items may be united to form a single statement of length $l-1$ which completely contains the given statements, and it follows from this that any statement of length l may be regarded as containing $2^{(l_0-l)}$ statements of length $l_0>l$. However, if more than two items cross over in the given statements then there is more than one way of expressing the digest, depending on the order in which the crossing items are replaced by products.

There is one further aspect of the problem which must be considered before the rules may be applied indiscriminately and this is the question of *completeness*. In the examples so far given, two statements have been reduced to one which completely contains the truth of the given statements, which may then be discarded, the digest statement alone being retained. Such digests are said to be *complete*. In the example

$$\langle\alpha|\beta\rangle.\langle\beta|\gamma\rangle \supset \langle\alpha|\gamma\rangle$$

the statement on the right follows from the given statements* but it does not completely contain them; it states that 'if α is $+$ then γ is $+$', which is true, but the information that β is also $+$ is lost if $\langle\alpha|\gamma\rangle$ is the only statement retained. The simplest approach to this problem is to enquire how many primitive associated statements are represented by the given statements and by the digest and to say that the latter shall not be less than the former, and if it is, then the shortest of the given statements must also be retained to make the digest complete, *i.e.*

$$\Sigma 2^{(l_p-l_g)} \leq \Sigma 2^{(l_p-l_d)} \tag{6}$$

in which the l_g are the lengths of the various given statements, l_d the lengths of the digest statements, and l_p the length of the primitive statements. If the equality in (6) is satisfied then the digest is said to be exact; if not then it is inexact in the sense that one or more of the primitive statements is contained more than once in the digest. In the following examples the statements on the left are given statements and the bold-faced statements are the ones required to form a complete digest in each case

$$\langle|\alpha\beta\gamma\rangle.\langle\alpha|\beta\gamma\rangle \supset \langle|\boldsymbol{\beta\gamma}\rangle \tag{7}$$

$$\langle|\alpha\beta\gamma\rangle.\ \boldsymbol{\langle\alpha|\beta\rangle} \supset \langle|\boldsymbol{\beta\gamma}\rangle \tag{8}$$

* This involves a simple extension of the cross-over rule, that when the given statements have one item which crosses over, this vanishes and each partition of the digest statement is the union of the corresponding partitions of the given statements.

$$\langle\boldsymbol{\alpha}|\boldsymbol{\beta}\rangle \,.\, \langle\boldsymbol{\beta}|\boldsymbol{\gamma}\rangle \supset \langle\alpha|\gamma\rangle\,. \tag{9}$$

Of these (7) is an example of an exact digest, (8) is inexact in that the primitive statement $\langle\alpha|\beta\gamma\rangle$ is contained twice within the bold-faced statements, and (9) is a case where no advantage in conciseness is obtained from uniting the given statements, two statements of length two being required however the result is expressed.

Evidently the left-hand side of (6) represents the number of primitive statements contained in the given statements provided that the former are all different. We now question whether (6) provides a valid test for completeness if this is not so. Suppose we have any two statements of length l_g as given statements and suppose that together they may be derived from three, not four, statements of length (l_g+1), then we may show that the two given statements cannot be united by the cross-over rule, so that the question of testing the completeness of the result does not arise. We illustrate a general case. Let the three statements of length (l_g+1) be $\langle|\alpha\beta\gamma\delta\rangle$, $\langle\alpha\beta|\gamma\delta\rangle$ and $\langle\alpha\gamma\delta|\beta\rangle$ and let the second of these be common to both the statements of length l_g, then

$$\left.\begin{array}{l} \langle|\alpha\beta\gamma\delta\rangle \equiv \langle(\alpha\beta)|\beta\gamma\delta\rangle \\ \langle\alpha\beta|\gamma\delta\rangle \equiv \langle(\alpha\beta)\beta|\gamma\delta\rangle \end{array}\right\} \begin{array}{l} \langle(\alpha\beta)|\gamma\delta\rangle \equiv \langle|(\alpha\beta\gamma)\gamma\delta\rangle \\ \qquad \equiv \langle(\gamma\delta)|(\alpha\beta\gamma)\delta\rangle \end{array} \tag{10}$$

$$\left.\begin{array}{l} \langle\alpha\beta|\gamma\delta\rangle \equiv \langle\alpha|(\beta\gamma)\gamma\delta\rangle \\ \quad \equiv \langle\alpha(\gamma\delta)|(\beta\gamma)\delta\rangle \\ \langle\alpha\gamma\delta|\beta\rangle \equiv \langle\alpha\gamma\delta|(\beta\gamma)\rangle \\ \quad \equiv \langle\alpha(\gamma\delta)\delta|(\beta\gamma)\rangle \end{array}\right\} \begin{array}{l} \langle\alpha(\gamma\delta)|(\beta\gamma)\rangle \\ \qquad \equiv \langle\alpha(\gamma\delta)|(\alpha\beta\gamma)\rangle \end{array} \tag{11}$$

in which $\equiv$ denotes application of the equivalence rule and $\}$ denotes union by the cross-over rule.

Thus any statement of length 3 from (10) and any of length 3 from (11) represent a typical pair of statements having a common statement of length 4 among their antecedents. Evidently they can be expressed so that the items they have in common, $(\gamma\delta)$ and $(\alpha\beta\gamma)$, both occur in the same partition with only one remaining item in each case, α and δ, which are different, so that there is no possibility of combining these statements further, except to form $\langle\alpha(\gamma\delta)|(\alpha\beta\gamma)\delta\rangle$ which is retrogressive, being equivalent to $\langle\alpha\beta|\gamma\delta\rangle$.

The three rules so far described are sufficient for the digestion of any combination of statements, and we conclude this section with some examples in which the primitive statements are listed one above the other, and bold face indicates the statements to be retained.

$$\text{(i)}\quad \left.\begin{array}{l} \left.\begin{array}{l}\langle|\alpha\beta\gamma\rangle \equiv \langle(\alpha\beta)|\beta\gamma\rangle \\ \langle\alpha\beta|\gamma\rangle \equiv \langle(\alpha\beta)\beta|\gamma\rangle\end{array}\right\} \langle(\alpha\beta)|\gamma\rangle \equiv \langle|(\boldsymbol{\alpha\beta\gamma})\boldsymbol{\gamma}\rangle \\ \langle\beta\gamma|\alpha\rangle \equiv \langle(\beta\gamma)\gamma|\alpha\rangle \equiv \langle\gamma|(\alpha\beta\gamma)\alpha\rangle \end{array}\right\} \langle|(\boldsymbol{\alpha\beta\gamma})\boldsymbol{\alpha}\rangle\,.$$

$$\text{(ii)}\quad \left.\begin{array}{l} \left.\begin{array}{l}\langle|\alpha\beta\gamma\rangle \\ \langle\alpha|\beta\gamma\rangle\end{array}\right\} \langle|\beta\gamma\rangle \\ \left.\begin{array}{l}\langle\beta|\gamma\alpha\rangle \\ \langle\alpha\beta|\gamma\rangle\end{array}\right\} \langle\beta|\gamma\rangle \end{array}\right\} \langle|\gamma\rangle$$

$$\text{(iii)}\quad \left.\begin{array}{l} \left.\begin{array}{l}\langle|\alpha\beta\gamma\rangle \equiv \langle(\beta\gamma)|\alpha\gamma\rangle \\ \langle\beta\gamma|\alpha\rangle \equiv \langle(\beta\gamma)\gamma|\alpha\rangle\end{array}\right\} \langle(\beta\gamma)|\alpha\rangle \equiv \langle|\alpha(\alpha\beta\gamma)\rangle \\ \left.\begin{array}{l}\langle\gamma\alpha|\beta\rangle \equiv \langle\alpha|(\beta\gamma)\beta\rangle \\ \langle\alpha\beta|\gamma\rangle \equiv \langle\alpha\beta|(\beta\gamma)\rangle\end{array}\right\} \langle\alpha|(\beta\gamma)\rangle \equiv \langle\alpha|(\alpha\beta\gamma)\rangle \end{array}\right\} \langle|(\boldsymbol{\alpha\beta\gamma})\rangle \tag{12}$$

$$\text{(iv)}\quad \left.\begin{array}{l} \left.\begin{array}{l} \left.\begin{array}{l}\langle|\alpha\beta\gamma\rangle \\ \langle\alpha|\beta\gamma\rangle\end{array}\right\} \langle|\boldsymbol{\beta\gamma}\rangle \equiv \langle(\beta\gamma)|\gamma\rangle \\ \langle\beta\gamma|\alpha\rangle \equiv \langle(\beta\gamma)\gamma|\alpha\rangle \end{array}\right\} \langle(\beta\gamma)|\alpha\rangle \equiv \langle|\alpha(\alpha\beta\gamma)\rangle \\ \left.\begin{array}{l}\langle\gamma\alpha|\beta\rangle \equiv \langle\alpha|(\beta\gamma)\beta\rangle \\ \langle\alpha\beta|\gamma\rangle \equiv \langle\alpha\beta|(\beta\gamma)\rangle\end{array}\right\} \langle\alpha|(\beta\gamma)\rangle \equiv \langle\alpha|(\alpha\beta\gamma)\rangle \end{array}\right\} \langle|(\boldsymbol{\alpha\beta\gamma})\rangle\,. \tag{13}$$

In (i), as in other examples, we prefer to express the digest in terms of $(\alpha\beta\gamma)$ rather than in terms of products such as $(\alpha\beta)$, since the former is a secondary tri-product and the latter a tetra-product.

In (iv) the statement $\langle|\beta\gamma\rangle$ is retained because it must be retained (to satisfy (6)) at the stage when the statement $\langle(\beta\gamma)|\alpha\rangle$ is obtained, and it is this latter statement which is then used further.

Finally, we remark that an alternative approach to the question of completeness does exist, and this is through the theory of information of Shannon (1948), according to which it may be shown that the information content of p statements of length l, containing the same items is given by

$$l-\log_2(2^l-p) \text{ bits}$$

on the assumption that the *a priori* probability of a statement of length 1 is $\frac{1}{2}$. On this basis the information contained in the five primitive statements given in (13) is $3-\log_2 3 = 1{\cdot}415$ bits; similarly that contained in $\langle|(\alpha\beta\gamma)\rangle$ is 1 bit and that in $\langle|\beta\gamma\rangle$ 0·415 bits. Thus the digest is complete, but the approach does not indicate that the digest given in (13) is inexact, the statement $\langle|\alpha\beta\gamma\rangle$ being contained twice. However, the relevance and implications of the theory of information have not yet been throughly studied.

II. *The digest regarded as the intersection of short statements*

In this section we consider a second approach to the general problem of forming the digest of a list of primitive statements which is complementary to the first and which has the merit of lending itself readily to machine computation. (Not all the features of this approach have yet been fully worked out however; see Discussion, § (iii)).

We consider the fourth-order case for which $l_p = 3$ and consider the primitive statements which are required to produce the statements $\langle|\beta\rangle$ and $\langle|\gamma\rangle$; these are

$$\langle|\alpha\beta\gamma\rangle\ \langle\alpha|\beta\gamma\rangle\ \langle\gamma|\alpha\beta\rangle \text{ and } \langle\gamma\alpha|\beta\rangle \text{ for } \langle|\beta\rangle$$

and

$$\langle|\alpha\beta\gamma\rangle\ \langle\alpha|\beta\gamma\rangle\ \langle\beta|\gamma\alpha\rangle \text{ and } \langle\alpha\beta|\gamma\rangle \text{ for } \langle|\gamma\rangle.$$

These groups of primitive statements will be termed the *specification* for $\langle|\beta\rangle$ and $\langle|\gamma\rangle$ respectively. Of these we note that the first two statements are common to both specifications, *i.e.* the *intersection* of the two specifications is $\langle|\alpha\beta\gamma\rangle.\langle\alpha|\beta\gamma\rangle$ and the digest of these is $\langle|\beta\gamma\rangle$. This result is quite general, that a statement of length l, (such as $\langle|\beta\gamma\rangle$), may be regarded as the intersection (of the specifications) of l statements of length 1, these latter having the property that their left-hand and right-hand partitions, when united (left with left and right with right), form the corresponding partitions of the longer statement. Fundamentally, the reason for this is that the specification for $\langle|\beta\rangle$, written S_β, consists of a list of all those primitive statements which have β on the right, S_γ is all those having γ on the right, whilst the specification for $\langle|\beta\gamma\rangle$ corresponds to all those statements having β and γ on the right, and these must obviously be the intersection of S_β and S_γ.

The relationship between this and the foregoing is most easily seen in terms of the diagram (Fig. 1(a)) which represents a rhombohedron having the eight primitive statements attached at its vertices. Every pair of primitive statements which are related by a single cross-over are then connected by edges of the rhombohedron; thus to every edge may be attached a statement of length 2 which is the digest of the two primitive statements thereby connected, and similarly to every face may be attached a statement of length 1, which is the digest of the four primitive statements at its corners. It is manifest that any statement such as $\langle|\gamma\alpha\rangle$ may be regarded as the union of the statements $\langle|\alpha\beta\gamma\rangle$ and $\langle\beta|\gamma\alpha\rangle$ which its edge connects in the diagram, or as the intersection of the faces $\langle|\alpha\rangle$ and $\langle|\gamma\rangle$ which meet at this edge. Similarly the statement $\langle\beta|\gamma\alpha\rangle$ occurs at the intersection of the three faces $\langle\beta|\rangle$, $\langle|\gamma\rangle$ and $\langle|\alpha\rangle$.

Fig. 1(b) shows an extension of this in which the four statements comprising the specification for $\langle|(\alpha\beta\gamma)\rangle$ (see (12) above) are joined to form a tetrahedron. This tetrahedron intersects the $\langle|\alpha\rangle$ face along the diagonal connecting $\langle|\alpha\beta\gamma\rangle$ and $\langle\beta\gamma|\alpha\rangle$ whose digest is $\langle|\alpha(\alpha\beta\gamma)\rangle$, *i.e.* the statement formed by uniting corresponding partitions of $\langle|\alpha\rangle$ and $\langle|(\alpha\beta\gamma)\rangle$.

Since every combination of primitive statements must include the leading statement, and since all primitive statements (except the trailing one which is

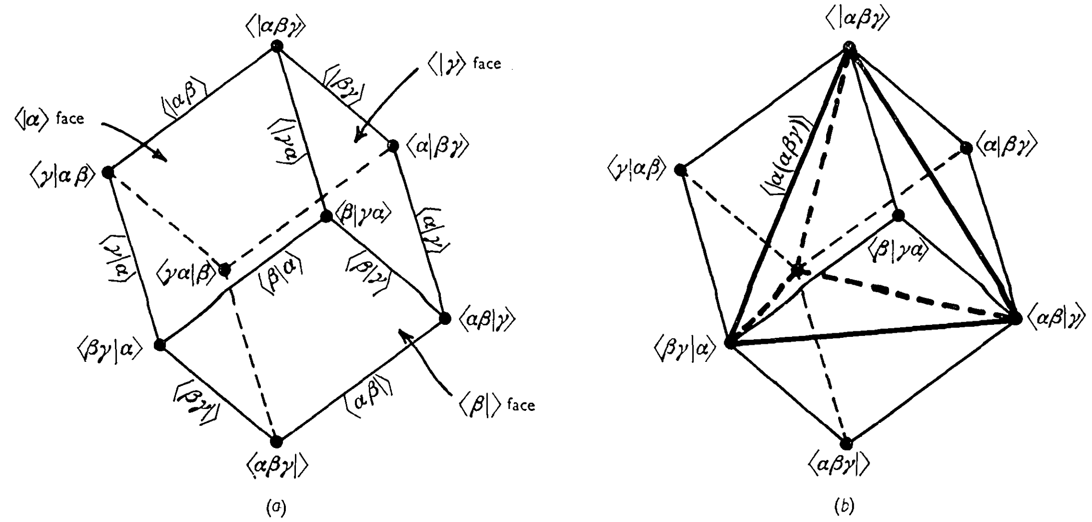

Fig. 1. A geometrical analogue showing the relationship between the eight primitive statements of the fourth order case, and various shorter statements. The arrows identify the three faces of the rhombohedron which are outlined entirely with full lines. For further explanation, see text.

Table 1. *A machine procedure for digestion*

		$\langle\vert\alpha\beta\gamma\rangle$	$\langle\alpha\vert\beta\gamma\rangle$	$\langle\beta\vert\gamma\alpha\rangle$	$\langle\gamma\vert\alpha\beta\rangle$	$\langle\alpha\beta\vert\gamma\rangle$	$\langle\gamma\alpha\vert\beta\rangle$	$\langle\beta\gamma\vert\alpha\rangle$	$\langle\alpha\beta\gamma\vert\rangle$	
S_α	=	1	0	1	1	0	0	1	0	= Specification for $\langle\vert\alpha\rangle$
S_β	=	1	1	0	1	0	1	0	0	= Specification for $\langle\vert\beta\rangle$
S_γ	=	1	1	1	0	1	0	0	0	= Specification for $\langle\vert\gamma\rangle$
$S_{(\alpha\beta\gamma)}$	=	1	0	0	0	1	1	1	0	= Specification for $\langle\vert(\alpha\beta\gamma)\rangle$
W	=	1	1	1	1	0	0	1	0	= Working list
C	=	1	1	1	1	1	1	1	1	= Check list, initial value
W & S_α	=	1	0	1	1	0	0	1	0	= S_α? Yes ∴ $\langle\vert\alpha\rangle$
$(-1-S)$	=	0	1	0	0	1	1	0	1	
$(-1-S)$ & C	=	0	1	0	0	1	1	0	1	= C', = C? No ∴ statement new, print
$W+C+1$	=	0	1	0	0	0	0	0	0	= 0? No ∴ digest not complete
W & S_β	=	1	1	0	1	0	0	0	0	= S_β? No ∴ $\langle\vert\beta\rangle$ not proved
$S_{\alpha\beta} = S_\alpha$ & S_β	=	1	0	0	1	0	0	0	0	= Specification for $\langle\vert\alpha\beta\rangle$
W & S	=	1	0	0	1	0	0	0	0	= $S_{\alpha\beta}$? Yes ∴ $\langle\vert\alpha\beta\rangle$
$(-1-S)$	=	0	1	1	0	1	1	1	1	
$(-1-S)$ & C	=	0	1	0	0	1	1	0	1	= C', = C? Yes ∴ statement not new, do not print
$S_{\beta\gamma} = S_\beta$ & S_γ	=	1	1	0	0	0	0	0	0	= Specification for $\langle\vert\beta\gamma\rangle$
W & S	=	1	1	0	0	0	0	0	0	= S? Yes ∴ $\langle\vert\beta\gamma\rangle$
$(-1-S)$	=	0	0	1	1	1	1	1	1	
$(-1-S)$ & C	=	0	0	0	0	1	1	0	1	= C', = C? No ∴ statement new, print
$W+C+1$	=	0	0	0	0	0	0	0	0	= 0? Yes ∴ digest complete

rare) are connected to it by lines which are the intersections of two of the four statements $\langle|\alpha\rangle$, $\langle|\beta\rangle$, $\langle|\gamma\rangle$ and $\langle|(\alpha\beta\gamma)\rangle$ it is clear that many of the 127 possible combinations can be expressed in terms of those alone, and we now describe a procedure which has been used for this purpose. The question of the adequacy of the procedure to deal with all possible situations will be returned to in our Discussion § (iii).

A machine procedure for digestion

A computer programme has been written in which successive determinants are set up and the eight values of each are calculated if, and only if, (i) at least one of the six u values involved is $\geq \frac{1}{3}$* and (ii) the value of D_4 is found to be negative when $\alpha=\beta=\gamma=-$. As each of the eight sign combinations is tried, a 1 is entered into a particular digit of an 8-bit fixed point binary number if D_4 is found to be negative. Thus a number is synthesized within the machine which indicates which primitive statements may be made; this number is termed the *working list* and is denoted by W.

For example,

	$\langle\vert\alpha\beta\gamma\rangle$	$\langle\alpha\vert\beta\gamma\rangle$	$\langle\beta\vert\gamma\alpha\rangle$	$\langle\gamma\vert\alpha\beta\rangle$	$\langle\alpha\beta\vert\gamma\rangle$	$\langle\gamma\alpha\vert\beta\rangle$	$\langle\beta\gamma\vert\alpha\rangle$	$\langle\alpha\beta\gamma\vert\rangle$
$W=$	1	1	1	1	0	0	1	0

implies the five primitive statements having 1's below them.

We also establish a *check list*, denoted C, which initially has a 1 in each of the eight positions. As each digest statement is formed zeros are planted in C in positions corresponding to the primitive statements contained in the statement formed, so that when the digest is complete $W+C=-1$ modulo 256 (*i.e.* $W+C$ has a 1 in every place). The procedure is illustrated in Table 1 in which the first four lines represent the specifications for the four statements shown; these are fixed point numbers and form part of the programme. The next two lines indicate starting conditions, and on the seventh line we collate W with the specification for $\langle|\alpha\rangle$, (*i.e.* form a number having a 1 wherever W and S_α have a 1, hence the symbol &). This is achieved with a single machine instruction on many computers. Now in this example W contains S_α ∴ W & $S_\alpha = S_\alpha$, if any digit in S_α were missing in W this would not be so; thus testing the equality of (W & S_α) with S_α is equivalent to testing for $\langle|\alpha\rangle$. Next we form $(-1-S)$ (modulo 256), S being the specification currently being considered. This number has a zero in the position corresponding to each of the primitive statements contained in $\langle|\alpha\rangle$, and 1's elsewhere. This is then collated with C to form a new check list C'; this new check list is compared with C and found to be different, *i.e.* some primitive statements not previously accounted for are accounted for by $\langle|\alpha\rangle$; $\langle|\alpha\rangle$ is therefore a new statement, not included in anything foregoing. $\langle|\alpha\rangle$ is therefore printed and C' replaces C (becomes C). Next we form the quantity $W+C+1$ and find it differs from zero; thus there is a digit or digits in W not yet accounted for and checked off in C; in this case the statement $\langle\alpha|\beta\gamma\rangle$ remains.

On finding $W+C+1 \neq 0$ the programme continues to cycle through the four given specifications, treating

* This condition is due to Kitaigorodski, see Discussion § (i).

each in the same way. W & S_β is the next to be formed, and this is found unequal to S_β; therefore the statement $\langle|\beta\rangle$ may not be made. Similarly with $\langle|\gamma\rangle$ and $\langle|(\alpha\beta\gamma)\rangle$.

When these four possibilities are exhausted the programme enters a double loop, so that it takes the four given specifications in pairs beginning with α and β. These two specifications are collated to form $S_{\alpha\beta}$, the intersection of S_α and S_β being the specification for $\langle|\alpha\beta\rangle$. The remaining steps are the same, $S_{\alpha\beta}$ being used as the current value of S, and this time we find that the statement $\langle|\alpha\beta\rangle$ may be made, but that $C'=C$, so that $\langle|\alpha\beta\rangle$ contains nothing new and is therefore not printed.

Eventually the cycle reaches $S_{\beta\gamma}$ and finds that $\langle|\beta\gamma\rangle$ is true and new and prints it, and this time $(-1-S)$ & C yields the complement of W and the digest is therefore complete.

Note that $\langle|\alpha\beta\gamma\rangle$ is contained twice in the digest, the digest being inexact, but the process is quite unaffected by the fact that this statement is checked off twice and the others only once. The programme may be entered at the double loop if W contains less than four digits, and a triple loop may be used to provide for the leading statement occurring alone. In practice this has been carried out on a machine using ten digit numbers; the only difference this makes is that C is initially -1 modulo 1024 and the spare digits in W and S are set to zero. The spare digits then have no effect on the process.

The doubly symmetrical case

The foregoing is principally concerned with the general fourth-order case in which three indices $\mathbf{h}_i$, $\mathbf{h}_j$ and $\mathbf{h}_k$ are independently chosen. If the choice is made such that $\mathbf{h}_i+\mathbf{h}_j=\mathbf{h}_k$ then special properties appear and the determinant takes the form (2). In this case evidently

$$\begin{aligned}\alpha &= s_{0j}s_{ji}s_{i0}\\ \beta &= s_{0k}s_{ki}s_{i0} = s_{0k}s_{j0}s_{i0}\\ \gamma &= s_{0k}s_{kj}s_{j0} = s_{0k}s_{i0}s_{j0} \equiv \beta\\ (\alpha\beta\gamma) &= s_{ji}s_{ki}s_{kj} = s_{ji}s_{j0}s_{i0} \equiv \alpha\end{aligned}$$

(centrosymmetric case).

It follows that we are not entitled to insert β and γ into the expression for D_4 with opposite signs, so that the statements $\langle\alpha\beta|\gamma\rangle$, $\langle\alpha\gamma|\beta\rangle$, $\langle\beta|\gamma\alpha\rangle$ and $\langle\gamma|\alpha\beta\rangle$ may never be made in this case even if testing for these leads to a negative value of (5). Furthermore, since β is always equal to γ and only those statements

Table 2. *The application of inequalities to a one-dimensional problem**

h	$\lvert U\rvert$	Sign symbol	3rd order	3rd order with 'sum and difference'	4th order un-conditional	4th order conditional					Correct solution	
1	0·177	*a*	*a*	*a*	*a*	*a*	*a*	*a*	*a*	*a*	*a*	$\langle\lvert n(aez)(dz)\rangle$
2	0·073	*b*		*b*	−	−	−	−	−	−	−	
3	0·088	*c*				(−*a*)			*c*		*a*	$\langle\lvert(ac)(cel)\rangle$
4	0·240	*d*		*d*	*d*	*d*	(+)	(−)	*d*	*d*	−	
5	0·068	*e*		*e*	*e*	*a*	*e*	−*a*	(*a*)	(−*a*)	−*a*	$\langle(ae)\lvert(cel)\rangle$
6	0·363	*f*	*f*	*ae*	*ae*	+	*ae*	−	+	−	−	
7	0·116	*g*		*be*	−*e*	−*a*	−*e*	*a*	−*a*	*a*	*a*	$\langle l\lvert(ac)\rangle$
8	0·320	*h*	*h*	*h*	−*ae*	−	−*ae*	+	−	+	+	
9	0·680	*i*	*i*	*abh*	*e*	*a*	*e*	−*a*	*a*	−*a*	−*a*	$\langle\lvert(cev)(ac)\rangle$
10	0·338	*j*	*ai*	*bh*	*ae*	+	*ae*	−	+	−	−	
11	0·137	*k*		*bdeh*	*ad*	*ad*	*a*	−*a*	*ad*	*ad*	−*a*	$\langle(em)\lvert d\rangle$
12	0·198	*l*				−		−	*ac*	*d*	−	
13	0·178	*m*						*a*			*a*	$\langle\lvert d(am)\rangle$
14	0·146	*n*									+	
15	0·775	*o*	*fi*	*beh*	*a*	*a*	*a*	*a*	*a*	*a*	*a*	$\langle d\lvert(as)\rangle$
16	0·370	*p*	*afi*	*abeh*	+	+	+	+	+	+	+	
17	0·395	*q*	*afh*	*eh*	−*a*	−*a*	−*a*	−*a*	−*a*	−*a*	−*a*	$\langle d\lvert(eu)\rangle$
18	0·240	*r*	+	+	+	+	+	+	+	+	+	
19	0·036	*s*					*a*				*a*	$\langle(mu)(dl)\lvert(elm)\rangle$
20	0·060	*t*									−	
21	0·098	*u*					*e*				*a*	$\langle\lvert(dl)(ae)\rangle$
22	0·031	*v*				−					−	
23	0·246	*w*		−*e*	−*e*	−*a*	−*e*	*a*	−*a*	*a*	*a*	
24	0·623	*x*	*f*	*ae*	*ae*	+	*ae*	−	+	−	−	
25	0·745	*y*	*af*	*e*	*e*	*a*	*e*	−*a*	*a*	−*a*	−*a*	
26	0·068	*z*									+	
Number of signs involved			11	17	17	20	19	19	19	18		
Number of independent signs			4	5	3	2	2	1	3	2		
Number of possibilities			8	16	4	2	2	1	4	2		

* *Note added in proof.* This hand-drawn table has since been checked by computer, use being made of the techniques described in the Discussion § (iv), and it has been found that the statement $\langle|d(am)\rangle$, though true, was not proved and was made in error. However, the additional statements $\langle l|(eu)\rangle$ and $\langle(aez)|(dz)\rangle$ may also be proved.

containing β and γ in the same partition can arise, it follows that γ may be ignored altogether, *i.e.*

$$\langle|\alpha\beta\gamma\rangle \equiv \langle|\alpha\beta\rangle$$

and we have in effect only four primitive statements in this case. These are $\langle|\alpha\beta\rangle$, $\langle\alpha|\beta\rangle$, $\langle\beta|\alpha\rangle$ and $\langle\alpha\beta|\rangle$. The simplest (if not the most efficient) way to deal with this situation in the digestion programme is to suppress the unwanted digits in S and W by collation with 11000011. By this procedure both the general and this special case can be handled by the same programme.

This special case is superior to the general case in that a statement with $l=1$ results from only two primitive statements. Thus $\langle|\alpha\rangle$ may be proved in this case where the general case may fail even if the u values are the same in both cases.

An example

The principles outlined in this paper have been applied to a synthetic one-dimensional problem which was created originally by Sayre (1952) for the purpose of testing his sign determining equation. The problem corresponds to a centrosymmetric structure containing eight equal atoms in a unit cell of 20 Å dimension, and the given data reach to the Cu $K\alpha$ limit. The results of this trial are shown in Table 2 where the sign of each U is denoted by a letter. When the third-order inequalities are applied exhaustively seven statements of the type $\langle|\alpha\rangle$ may be made, sufficient to eliminate the seven sign symbols j, o, p, q, r, x, y. At this stage eleven signs are inter-related, only four of which are independent.

In the next column we include the results of the application of fourth-order inequalities in which $\mathbf{h}_i+\mathbf{h}_j=\mathbf{h}_k$, so far as these result in statements of length one. At this stage 17 signs are inter-related, five of which are independent, and this situation represents the limit that may be reached by the application of conventional inequalities. The next column embodies all the statements of length one obtainable from the fourth-order inequalities; no further signs are involved, but the 17 are now expressible in terms of only three independent signs, and the entries in this column are unconditionally true.

There then remain eleven statements of lengths two or three which are written alongside the table in terms of the sign symbols remaining. These statements can only be used conditionally, and in the next five columns the bracketed signs have been chosen arbitrarily as a matter of trial and error. For example, if d is supposed negative, then on the basis of the seventh statement listed, the product (am) must be positive, or $m=a$. Furthermore, by the sixth statement the product (em) is negative, hence $e=-a$, and by the last statement $l=d=-$. Thus nineteen signs are now known on the basis of one supposition, the sign of a being immaterial. The correct solution is shown in the last column for comparison.

Discussion

In this discussion we comment on six features of the problem: (i) the extent of the information provided by the approach, (ii) the build-up of information from determinants of various orders, (iii) the capacity of the machine digestion process to deal with all situations which may arise, (iv) the utilization of information arising from different determinants, *i.e.* non-associated statements, (v) crystallographic symmetry, and (vi) the effects of experimental errors.

(i) *The extent of the information provided by the approach*

The results given in Table 2 have already shown what is generally true that the information derivable from fourth-order inequalities exceeds that available from conventional inequalities. The gain may seem marginal, but this is only because the territory of fourth-order inequalities is not virgin ground, and one might expect the gain of fifth-order inequalities over fourth-order to be comparable with the gain of fourth over third.

The writer has not made any statistical study of the amount of information that one may expect to arise when inequalities of order n are applied to a structure containing N atoms. Kitaigorodski (1957), however, has studied this problem in the simplified case in which all the u values appearing in a determinant are assumed equal; he then calculated D_n as a function of this single variable and the signs of the primary tri-products. He showed that unless

$$u \geq 1/(n-1) = \varepsilon$$

all combinations of signs will satisfy $D_n \geq 0$, and he terms ε the 'boundary' for inequalities of order n. Evidently, in a real case in which several different u values occur, at least one must exceed ε if the leading statement is to arise.

This gives the appearance that as n increases the amount of information obtainable will increase as ε decreases. As a matter of experience this is so, but one should not take ε to be a simple measure of the strength of an inequality because an inequality with a low boundary must yield more primitive statements than need one with a higher boundary in order to provide the same information. For example, third-order inequalities have boundary $\frac{1}{2}$ and one statement from such an inequality proves a sign product positive; fourth-order inequalities have boundary $\frac{1}{3}$ but must yield four primitive statements to prove a sign product positive. Kitaigorodski also shows that for this simplified problem the value of u that must be exceeded in order to prove all the primary tri-products positive also decreases as n increases, but is a comparatively slowly varying function of n. He also develops these ideas to the conclusion that inequalities alone should be capable of solving structures for which $\sqrt{\overline{u^2}} > \sim 0{\cdot}10$ to $0{\cdot}15$.

In this paper we comment on the experience gained with the example already given. The problem is clearly too small to enable one to draw precise conclusions concerning the strength of the method when applied to a crystallographic problem, but the behaviour of the method was as follows.

There are 169 distinct connecting determinants of order 3 which may be set up; of these seven yielded statements of length 1. There are 1378 distinct connecting determinants of order 4, of which 270 produced at least one primitive statement, a total of nearly 900 primitive statements being obtained. Many of the non-primitive statements so produced were repetitive; for example, the statement $\langle|(axy)\rangle$ was proved by 25 different fourth-order inequalities, in fact by every one which contains

$$\begin{vmatrix} 1 & U(1) & U(25) \\ U(\bar{1}) & 1 & U(24) \\ U(\overline{25}) & U(\overline{24}) & 1 \end{vmatrix}$$

as a minor. Every statement arising from the third order was obtained again from the fourth order. The trailing statement, which is required to prove a tri-product negative, occurred only once and yielded $\langle(awx)|\rangle$. Significantly, this arose from a determinant having $\mathbf{h}_i+\mathbf{h}_j=\mathbf{h}_k$. No contradictions arose.

(ii) *The build-up of information from determinants of various orders*

We have already seen that fourth-order inequalities regenerate information obtainable from the third-order level, and we now enquire whether inequalities of order n necessarily regenerate all the information obtainable from inequalities of order $m<n$.

In general

$$0 \le D_n = \frac{A_1A_2-B\bar{B}}{C} = \frac{A_1A_2-|B|^2}{C}$$

in which A_1 is a connecting determinant (a minor of D) of order $(n-1)$ obtainable from D_n by deleting the ith row and the ith column. A_2 is similarly obtained by deleting the jth row and jth column, B is obtained from D by deleting the ith row and jth column, and $\bar{B}$ is the complex conjugate of B obtained by deleting the jth row and ith column. C is obtained from D_n by deleting the ith and jth rows and columns, being of order $(n-2)$ (Muir & Metzler, 1960, § 149, p. 134). The correct set of signs must then satisfy the four requirements

$$D_n \ge 0,\ A_1 \ge 0,\ A_2 \ge 0,\ C \ge 0\,.$$

Evidently it is possible for a wrong set of signs to satisfy the first requirement by failing simultaneously to meet two of the remaining three requirements, *i.e.* tests of inequalities of orders $(n-1)$ and $(n-2)$ would yield information in such circumstances which the inequality of order n would fail to provide. If, however, a trial set of signs is known to satisfy the inequalities of orders $(n-1)$ and $(n-2)$ then the requirement $D_n \ge 0$ takes the form

$$A_1A_2 \ge |B|^2$$

which is clearly stronger than $A_1 \ge 0$ and $A_2 \ge 0$. Thus an inequality of order n adds to, but does not necessarily reproduce, the whole of the information obtainable from inequalities comprising its principal minors.

Taken collectively, however, the picture is rather different. We consider the case $n=4$ (*all* sign combinations then satisfy $C \ge 0$) and consider all D_4 for which $\mathbf{h}_i$ and $\mathbf{h}_j$ are constant and $\mathbf{h}_k$ varies from one to another. A_1 then depends only on α and is common to all these inequalities, A_2 depends only on $(\alpha\beta\gamma)$ and varies from one to another.* Suppose that A_1 is capable of proving $\langle|\alpha\rangle$ then D_4 will also prove it unless at least one of the four combinations of β and γ with α negative makes A_2 sufficiently negative to satisfy $A_1A_2 \ge |B|^2$. While this appears possible in any one instance it is virtually certain that it will not happen in every instance, *i.e.* for every $\mathbf{h}_k$. Thus collectively fourth-order inequalities may be relied on to reproduce all the information available from the third order although individually they may not.

For orders greater than four this appears to remain true, although the argument is complicated by the fact that a wrong sign combination may give $C<0$.

(iii) *The capacity of the machine digestion process to deal with all situations which may arise*

None of the specifications used in the machine digestion process has a 1 in the position corresponding to the trailing statement $\langle\alpha\beta\gamma|\rangle$. Therefore, if this statement should arise the process will fail as the digest could never be completed. There appear to be two ways in which this deficiency may be made up; both involve applying the method as it stands to completion and entering an extension of the digestion programme when it is found that the digest produced is still incomplete. These two extensions may be modelled as follows:

(*a*) The four specifications are replaced by their complements, whereupon they become specifications for the converse statements, and the programme is then re-entered, *i.e.* $S_\alpha=10110010$ is replaced by 01001101, the specification for $\langle\alpha|\rangle$, *etc.*

(*b*) The extension uses specifications for the three statements $\langle(\beta\gamma)|\rangle$, $\langle(\gamma\alpha)|\rangle$ and $\langle(\alpha\beta)|\rangle$ (*i.e.* 11000011,

* The first and last rows and columns of D_4 have here been selected for deletion.

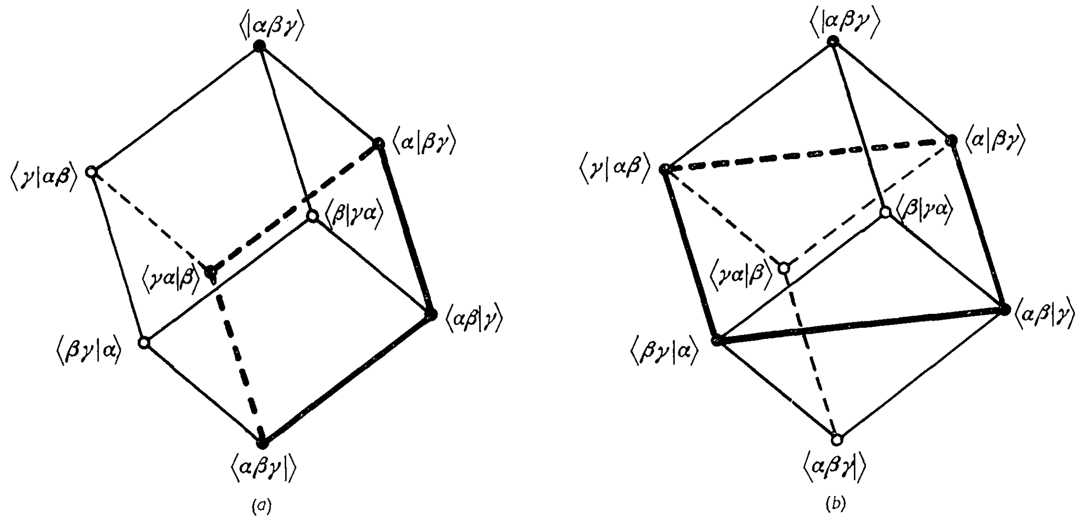

Fig. 2. Two special cases. For explanation, see text.

10100101 and 10011001*) and is otherwise similarly constructed.

(*b*) is thought to be superior to (*a*) as we may see on general grounds. By considering equation (5) in which each tri-product is multiplied by a positive numerical part and prefaced by a + sign we see that these may be demonstrably positive. The tetra-products are also multiplied by positive numerical parts and prefaced by − signs, so that we might expect to be able occasionally to prove a tetra-product negative; it therefore seems more sensible to search for negative tetra-products (scheme (*b*)) than negative tri-products (scheme (*a*)).

To compare these further we suppose that the situation is such that the statement $\langle\alpha|\rangle$ may be proved (in which case scheme (*a*) would appear preferable to (*b*)), then the situation is represented diagrammatically in Fig. 2(*a*) in which the five primitive statements which must then have occurred are marked with solid circles, the heavily outlined face being the $\langle\alpha|\rangle$ face. Algebraical studies have shown that if these five statements arise then it is likely that at least one of the remaining statements will also arise, although attempts to prove that this must happen have not so far been successful. If one of these does also arise then there are two consequences, (i) one of the three diagonal planes linking $\langle|\alpha\beta\gamma\rangle$ and $\langle\alpha\beta\gamma|\rangle$ will be completed, *i.e.* one of the statements $\langle(\beta\gamma)|\rangle$, $\langle(\gamma\alpha)|\rangle$ or $\langle(\alpha\beta)|\rangle$ can be made, and (ii) either one of the faces (for $\langle|\beta\rangle$ and $\langle|\gamma\rangle$) or the tetrahedron (for $\langle|(\alpha\beta\gamma)\rangle$) will also be completed, *i.e.* in such a case the digest need not explicitly contain $\langle\alpha|\rangle$ but could be expressed in a form such as $\langle|\beta\rangle.\langle(\alpha\beta)|\rangle$ in which case scheme (*b*) would serve as well as (*a*), the latter having lost its apparent advantage.

In the rare (if not impossible) event of a sixth primitive statement failing to arise, the process would still function but would fail to include a statement of length 1 in the digest, so that the fact that an unconditional statement could be made would be masked.

Another difficult circumstance is illustrated in Fig. 2(*b*). Again five primitive statements are supposed to have arisen, of which the four situated on the indicated diagonal plane are sufficient to yield $\langle|(\gamma\alpha)\rangle$. The machine digestion procedure outlined above would render this as

$$\langle|\alpha\beta\rangle.\langle|\beta\gamma\rangle.\langle|\alpha(\alpha\beta\gamma)\rangle.\langle|\gamma(\alpha\beta\gamma)\rangle$$

and again the unconditional statement would be obscured. However, if these five statements occur it appears likely (if not certain) that one of the remaining primitive statements (not $\langle\alpha\beta\gamma|\rangle$) will also occur; in which case we have the situation that $\langle|(\gamma\alpha)\rangle$ can only be proved in circumstances such that $\langle|\gamma\rangle$ and $\langle|\alpha\rangle$ or $\langle|\beta\rangle$ and $\langle|(\alpha\beta\gamma)\rangle$ may be proved, which the existing procedure can handle.

The whole question of the occurrence of these rare and difficult combinations and the technique for handling them should they arise is continually under review.

(iv) *The utilization of information arising from different determinants*

The foregoing has been principally concerned with the digestion of associated primitive statements. In this paragraph we outline a scheme which is cur-

* These specifications may be derived from S_α, S_β and S_γ by means of the non-equivalence function (see § (iv)). The specification for a statement involving a product such as $\langle|(\alpha\beta)\rangle$ is given by $S_{(\alpha\beta)}=(S_\alpha \not\equiv S_\beta)$, and the specification for $\langle(\alpha\beta)|\rangle$, written ${}_{(\alpha\beta)}S$, is the complement of this. Similarly $S_{(\alpha\beta\gamma)}=(S_\alpha \not\equiv S_\beta \not\equiv S_\gamma)$.

rently under development for the systematic application of the results to the individual signs s_{ij}. For this purpose we utilize the 'non-equivalence' function which is a feature of many computers, as it enables us to perform *algebraical* operations on signs and sign products.

Initially we allocate a sign symbol to each sign as in Table 2 and represent these in the machine according to the following scheme,

$$\begin{aligned} + &= 0000\ldots0000 \\ - &= 1000\ldots0000 \\ a &= 0100\ldots0000 \\ b &= 0010\ldots0000 \\ &\;\vdots \\ w &= 0000\ldots1000 \\ x &= 0000\ldots0100 \\ y &= 0000\ldots0010 \\ z &= 0000\ldots0001 \end{aligned}$$

the length of each 'word' being one greater than the number of independent signs to be determined, the first digit having the significance $-$, the second a, *etc.* The product of any number of such signs is then obtained using the non-equivalence order which takes two such 'words' as factors and places a zero in the result wherever the factors have the same digit, and a one wherever they differ. This is binary addition without carry.

Suppose a determinant yields $\langle|\alpha\rangle$ with $\alpha=(axy)$ then we form the sign symbol for α (using the non-equivalence order twice) and obtain

$$\alpha=0100\ldots0110=+\ .$$

Since α is positive we may multiply any sign symbol by α and use this to eliminate one such symbol.

We choose the highest in α for elimination—this is y, and then multiply any sign symbol containing y by α

$$\begin{aligned} y&=0000\ldots0010 \\ \alpha=axy&=0100\ldots0110 \\ \text{product}=\text{new } y&=0100\ldots0100=ax\,, \end{aligned}$$

thus y is replaced by ax. If then it is proved that the product (awx) is negative, so that

$$(-awx)=1100\ldots1100=+$$

we may use this to eliminate x, thus

$$\begin{aligned} x&=0000\ldots0100 \\ -awx&=1100\ldots1100 \\ \text{product}=\text{new } x&=1100\ldots1000=-aw \\ y&=0100\ldots0100 \\ -awx&=1100\ldots1100 \\ \text{product}=\text{new } y&=1000\ldots1000=-w\ . \end{aligned}$$

Thus a record may be kept in the machine equivalent to Table 2.

The same technique may also be used to test for relationships which may exist between α, β and γ before the values of D_4 are calculated. For example, in the special case having $\mathbf{h}_i+\mathbf{h}_j=\mathbf{h}_k$ the product $(\beta\gamma)$ may, by this means, be found to be positive at the outset, and could be used as the cue to apply the special procedure already described in this connection. This is probably the best way of handling this and other special cases, despite the fact that it may seem simpler to test for the relationship $\mathbf{h}_i+\mathbf{h}_j=\mathbf{h}_k$, because the identity $\beta\equiv\gamma$ may arise in two ways, either (i) because of such a vector relationship between the indices or (ii) because other inequalities previously employed have already established the fact,* and the second case will only be revealed by the use of this 'non-equivalence' technique.

(v) *Crystallographic symmetry*

It is well known that relationships among structure factors arising from space group symmetry, when allied to inequality relationships, yield inequalities of special form and utility characteristic of the space group involved. However, the approach being adopted is to utilize such crystallographic information at the outset by assigning sign symbols embodying such constraints. Equal structure factors may be given the same sign symbol, those which are equal but opposite in sign being given sign symbols which differ only in the first digit. All general inequalities then become special to the space group concerned and the procedures outlined in (iv) above take full account of such symmetry. Further, the special forms given by Goedkoop (1950) and MacGillavry (1950) including crystallographic symmetry occur spontaneously when appropriate values of $\mathbf{h}$ are chosen.

(vi) *The effects of experimental errors*

Erratic experimental data may cause trouble either by failing to produce statements which it ought to be possible to make, or by producing statements which it ought not to be possible to make. The second type of fault is much more serious as it may lead to contradictions. The question of the accuracy required in the data for the successful application of inequality methods has not yet been studied, though it is pretty clear that the requirements become more stringent as the order of inequality increases. Neither has the practical problem of dealing with inaccurate data been studied although three approaches to this question are in mind.

Firstly, since all inequalities depend on the constraint $\varrho\geq 0$ any apparent malfunction of the method must arise from the fact that the given (erratic) set of data gives regions of negative electron density even

* In this case the determinant would not necessarily be symmetrical about both diagonals, but it would call for the same treatment as those which are.

with the correct set of signs.* Such negative regions may be eliminated by adding a constant to ϱ, *i.e.* adding a constant to $F(000)$ and renormalizing. This provides a general slackening of the constraints which must remove any contradictions which may arise, but at the expense of failing to obtain some correct information that it ought to be possible to obtain with good data.

The second approach is to note with what frequency a given statement arises. It has already been pointed out that a number of different determinants may yield the same result, and if a particular sign product is proved positive many times and negative only a few then it would seem reasonable to accept it as positive. This approach would greatly complicate the machine programme, but it is a possibility.

The third possibility involves attaching a weight or credence to each statement derived from the value of D. Thus a statement arising from a determinant which is strongly negative would be given more credence than one rising from a determinant which is close to zero. In this way the true statement in a contradictory pair may be recognized by its greater weight. This again would greatly complicate the procedure and is regarded as a last resort.

From the practical standpoint it may turn out that the method requires data produced by counters or possibly photometry rather than eye-estimation.

APPENDIX

In this Appendix we show that the fourth order inequality having $\mathbf{h}_i+\mathbf{h}_j=\mathbf{h}_k$ is equivalent to the pair of sum and difference inequalities. We have

$$0 \le D = \begin{vmatrix} 1 & U(\mathbf{h}_i) & U(\mathbf{h}_j) & U(\mathbf{h}_i+\mathbf{h}_j) \\ U(\bar{\mathbf{h}}_i) & 1 & U(\mathbf{h}_j-\mathbf{h}_i) & U(\mathbf{h}_j) \\ U(\bar{\mathbf{h}}_j) & U(\mathbf{h}_i-\mathbf{h}_j) & 1 & U(\mathbf{h}_i) \\ U(\bar{\mathbf{h}}_i+\bar{\mathbf{h}}_j) & U(\bar{\mathbf{h}}_j) & U(\bar{\mathbf{h}}_i) & 1 \end{vmatrix}$$

which for brevity we write as

$$0 \le D = \begin{vmatrix} 1 & a & b & c \\ a & 1 & d & b \\ b & d & 1 & a \\ c & b & a & 1 \end{vmatrix} = \frac{\begin{vmatrix} 1 & a & b \\ a & 1 & d \\ b & d & 1 \end{vmatrix}^2 - \begin{vmatrix} a & b & c \\ 1 & d & b \\ d & 1 & a \end{vmatrix}^2}{\begin{vmatrix} 1 & d \\ d & 1 \end{vmatrix}} \equiv \frac{A^2-B^2}{1-d^2}$$

(Muir & Metzler, 1960, § 149, p. 134), A and B representing the two third order determinants, *i.e.*

$$0 \le \left(\frac{A+B}{1-d}\right)\left(\frac{A-B}{1+d}\right) .$$

Now the denominators are both positive, so that the quantities $(A+B)$ and $(A-B)$ must have the same sign. But A is itself a third-order connecting determinant, and must be positive, therefore of the quantities $(A+B)$ and $(A-B)$ at least one is positive, therefore both are positive, *i.e.*

$$\left(\frac{A+B}{1-d}\right) \ge 0$$

and

$$\left(\frac{A-B}{1+d}\right) \ge 0 .$$

Expanding, we find

$$A = 1+2abd-a^2-b^2-d^2$$
$$B = a^2d+b^2d+c-2ab-cd^2$$

$$A+B = 2abd-2ab-a^2+a^2d-b^2+b^2d+1-d^2+c-cd^2$$
$$= (1-d)[-2ab-a^2-b^2+(1+d)(1+c)]$$
$$\frac{A+B}{1-d} = [-(a+b)^2+(1+d)(1+c)] \ge 0$$

i.e. $[U(\mathbf{h}_i)+U(\mathbf{h}_j)]^2 \le [1+U(\mathbf{h}_j-\mathbf{h}_i)][1+U(\mathbf{h}_j+\mathbf{h}_i)]$.

Likewise

$\dfrac{A-B}{1+d} \ge 0$ yields

$$[U(\mathbf{h}_i)-U(\mathbf{h}_j)]^2 \le [1-U(\mathbf{h}_j-\mathbf{h}_i)][1-U(\mathbf{h}_j+\mathbf{h}_i)] .$$

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* Series termination effects may cause the transform of the U's to contain negative regions with the correct set of signs, but provided that u values lying outside the limiting sphere are not employed (treated as zero) in the inequalities this type of negativity in ϱ will do no harm.