Lonsdale (1947) ascribes the divergent-beam pattern to secondary extinction. The experiments seem to show that the Compton interference lines must also be due to secondary extinction. Diamonds showing the interference effect are probably perfect over very small regions. The interaction between the Compton radiation emitted from an atom and the surrounding atoms within a perfect region is thus small and probably without importance for the appearance of the interference line. The fact that perfect diamonds do not give interference diagrams is further evidence, that secondary extinction, and not the nature of the wave-field in a perfect crystal, is the cause of the interference pattern. Since the crystal must be large enough for the secondary extinction to be effective, it is understandable why small diamonds do not show interference lines.

When radiation is reflected in a certain direction from the lattice planes, the intensity of the incident beam is reduced. Reflection from the opposite side of the planes will in the same directions decrease and increase the intensity respectively. From energy considerations we would expect the sum of the excess and defect of intensity compared to the general background to be zero in the two directions. With [1$\bar{1}$0] vertical and [111] in the direction of the incident beam, three of the (111) lines pass close to the undeviated beam. The value of φ for Compton radiation in such a direction is close to zero, and considerably smaller than for the Compton radiation reflected in the same direction by the lattice planes. As expected, the lines appear dark. The corresponding light lines in the background are also seen.

Closing remarks

It has been shown that the interference pattern observed for diamond is due to Compton radiation emitted from the atoms within the crystal and reflected from the lattice planes. The fine structure of the lines is explained by assuming secondary extinction. Such interference diagrams may also be obtainable with other mosaic-type crystals. Like diamond, they should have low absorption and certain strongly reflecting planes.

The present work was carried out at the Polytechnic Institute of Brooklyn, and I am indebted to Prof. Fankuchen for use of the laboratory and X-ray equipment. My thanks are due to Prof. Ewald for suggesting the problem, for obtaining the necessary diamonds, through the courtesy of Dr Grenville-Wells, and for his advice during the investigation. My work in Brooklyn was made possible through a grant from the International Cooperation Administration, for which I want to express my gratitude.

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Acta Cryst. (1958). **11**, 4

The Utilization of Relationships between Sign Relationships

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(*Received* 24 *June* 1957)

It is shown that sign relationships are interrelated in such a way that the failure of one particular sign relationship may inevitably lead to the failure of a number of others. A method of utilizing these interrelationships is illustrated by the determination of signs for projections of the (known) structures of purpurogallin and α-glucose.

For the application of direct methods of sign determination it is often convenient to represent the unknown signs by alphabetical symbols. In this paper the nomenclature suggested by Woolfson (1957) will be followed, and two-dimensional reflexions (say of type *hk*0) are divided into four groups

(*a*) *h* even *k* even;
(*b*) *h* odd *k* odd;
(*c*) *h* odd *k* even;
(*d*) *h* even *k* odd.

The signs of the reflexions of group (*a*) are represented

by the symbols $a_1, a_2, \ldots$ etc. and similarly for the other groups, each symbol being taken as either $+1$ or -1.

Cochran & Douglas (1955) found it convenient to express sign relationships as equations of the form

$$pqr = s\,,$$

where p, q and r are the signs of three reflexions and the value of s is either $+1$ or -1, but more probably $+1$.

The signs of groups (a), (b), (c) and (d) can be interrelated in five ways, by relationships of type

$$\begin{aligned} a_r a_s a_t &= s_u\,, \\ a_r b_s b_t &= s_v\,, \\ a_r c_s c_t &= s_w\,, \\ a_r d_s d_t &= s_x\,, \\ b_r c_s d_t &= s_y\,. \end{aligned}$$

It is sometimes found that the relationships fall into sets such as

$$\begin{aligned} a_r a_s a_t &= s_1\,, & a_s b_s b_u &= s_3\,, \\ a_r b_s b_t &= s_2\,, & a_t b_t b_u &= s_4\,, \end{aligned}$$

from which it can be seen that

$$s_1 s_2 s_3 s_4 = +1\,. \qquad (1)$$

Such relationships between the symbols s are, in fact, found and used in the Cochran & Douglas method. From (1) it can be seen that if one of the sign relationships fails then at least one other must also fail; of the four values of s either four, two or none must be $+1$.

Let us now see how this sort of relationship may be utilized. As an example we shall take the $hk0$ projection of purpurogallin (Dunitz, 1952; Taylor, 1952) for which the two-dimensional space group is *pgg*. The 32 largest unitary structure factors (those for which $|U| \geq 0{\cdot}25$) are divided into four groups and sign symbols are allocated as shown below:

$$\begin{aligned} a_1 &= s(0, 4) & c_1 &= s(1, 12) \\ a_2 &= s(0, 16) & c_2 &= s(3, 8) \\ a_3 &= s(0, 24) & c_3 &= s(3, 20) \\ a_4 &= s(2, 20) & c_4 &= s(3, 24) \\ a_5 &= s(6, 14) & c_5 &= s(5, 14) \\ a_6 &= s(6, 18) & c_6 &= s(5, 18) \\ a_7 &= s(8, 4) & c_7 &= s(7, 6) \\ a_8 &= s(8, 6) & c_8 &= s(9, 4) \\ a_9 &= s(8, 10) & & \\ a_{10} &= s(10, 0) & d_1 &= s(2, 11) \\ a_{11} &= s(10, 8) & d_2 &= s(2, 21) \\ a_{12} &= s(4, 2) & d_3 &= s(2, 23) \\ & & d_4 &= s(4, 3) \\ b_1 &= s(3, 21) & d_5 &= s(6, 7) \\ b_2 &= s(7, 15) & d_6 &= s(8, 1) \\ b_3 &= s(9, 1) & d_7 &= s(8, 3) \\ b_4 &= s(11, 11) & d_8 &= s(8, 7) \end{aligned}$$

The sign relationships interrelating these reflexions are:

$$\begin{aligned} a_1 a_{12} a_{12} &= s_1 & -a_{11} c_1 c_8 &= s_{25} \\ a_1 a_5 a_6 &= s_2 & a_{12} c_1 c_5 &= s_{26} \\ a_1 a_8 a_9 &= s_3 & a_{12} c_2 c_7 &= s_{27} \\ a_2 a_{11} a_{11} &= s_4 & -a_1 d_6 d_7 &= s_{28} \\ a_2 a_8 a_9 &= s_5 & a_1 d_7 d_8 &= s_{29} \\ a_4 a_6 a_{12} &= s_6 & a_4 d_3 d_4 &= s_{30} \\ a_4 a_5 a_8 &= s_7 & a_5 d_1 d_4 &= s_{31} \\ a_7 a_{12} a_{12} &= s_8 & -a_5 d_1 d_7 &= s_{32} \\ a_9 b_1 b_4 &= s_9 & a_5 d_2 d_8 &= s_{33} \\ a_1 c_3 c_4 &= s_{10} & -a_6 d_2 d_4 &= s_{34} \\ a_1 c_5 c_6 &= s_{11} & a_6 d_2 d_7 &= s_{35} \\ -a_2 c_5 c_2 &= s_{12} & -a_6 d_1 d_8 &= s_{36} \\ -a_3 c_1 c_1 &= s_{13} & -a_7 d_1 d_5 &= s_{37} \\ -a_4 c_1 c_2 &= s_{14} & a_8 d_4 d_4 &= s_{38} \\ -a_4 c_5 c_7 &= s_{15} & -a_{11} d_1 d_7 &= s_{39} \\ -a_6 c_1 c_7 &= s_{16} & -a_{12} d_2 d_3 &= s_{40} \\ -a_8 c_4 c_6 &= s_{17} & a_{12} d_4 d_6 &= s_{41} \\ -a_8 c_3 c_5 &= s_{18} & -b_1 c_6 d_7 &= s_{42} \\ -a_8 c_2 c_5 &= s_{19} & -b_1 c_5 d_8 &= s_{43} \\ -a_8 c_1 c_7 &= s_{20} & -b_2 c_1 d_7 &= s_{44} \\ -a_9 c_4 c_5 &= s_{21} & -b_2 c_8 d_1 &= s_{45} \\ -a_9 c_2 c_6 &= s_{22} & -b_3 c_2 d_5 &= s_{46} \\ -a_{10} c_5 c_5 &= s_{23} & -b_4 c_6 d_5 &= s_{47} \\ -a_{10} c_6 c_6 &= s_{24} & b_4 c_2 d_7 &= s_{48} \end{aligned}$$

The negative signs in these equations arise owing to space-group considerations. The approximate theory of sign relationships (Cochran & Woolfson, 1955) applied to the data shows that only two or three of the values of s should be -1. Allowing the theory a margin of error we should not expect more than four of the sign relationships to fail. If these sign relationships are used to derive signs by the usual chain process then the early acceptance of a non-valid sign relationship has catastrophic effects on sign determination from that point on.

Now relationships are sought between the values of s, a procedure which, after some practice, can be carried out in a reasonably systematic way. A non-exhaustive selection of such relationships is given below:

$$\begin{aligned} s_{17} s_{19} s_{21} s_{22} &= +1 \quad (2) & s_{31} s_{33} s_{34} s_{36} &= +1 \quad (12) \\ s_{10} s_{11} s_{19} s_{22} &= +1 \quad (3) & s_{31} s_{32} s_{34} s_{35} &= +1 \quad (13) \\ s_3 s_{11} s_{19} s_{22} &= +1 \quad (4) & s_{11} s_{19} s_{42} s_{43} &= +1 \quad (14) \\ s_3 s_{10} s_{18} s_{21} &= +1 \quad (5) & s_{25} s_{39} s_{44} s_{45} &= +1 \quad (15) \\ s_6 s_{14} s_{16} s_{27} &= +1 \quad (6) & s_9 s_{22} s_{42} s_{48} &= +1 \quad (16) \\ s_6 s_{15} s_{16} s_{26} &= +1 \quad (7) & s_{14} s_{15} s_{19} s_{20} &= +1 \quad (17) \\ s_2 s_{29} s_{32} s_{36} &= +1 \quad (8) & s_{14} s_{15} s_{26} s_{27} &= +1 \quad (18) \\ s_2 s_{29} s_{33} s_{35} &= +1 \quad (9) & s_{19} s_{20} s_{26} s_{27} &= +1 \quad (19) \\ s_6 s_{30} s_{34} s_{40} &= +1 \quad (10) & s_2 s_{11} s_{31} s_{35} s_{42} s_{43} &= +1 \quad (20) \\ s_{32} s_{33} s_{35} s_{36} &= +1 \quad (11) & s_3 s_{19} s_{22} s_{29} s_{42} s_{43} &= +1 \quad (21) \end{aligned}$$

Many more such relationships may be found, particularly ones involving more than four values of s. Some of those listed above are interdependent: for example (19) may be inferred from (17) and (18).

Examination of this list of relationships shows that six of them, (2), (4), (14), (17), (19) and (21), involve s_{19}. These are written out in the following way:

$$s_{19}s_{22} \qquad s_{17}s_{21} = +1 \qquad (2)$$

$$s_{19}s_{22}s_{11} \qquad s_{3} = +1 \qquad (4)$$

$$s_{19} \quad s_{11}s_{42}s_{43} \qquad = +1 \qquad (14)$$

$$s_{19} \qquad s_{20}s_{14}s_{15} = +1 \qquad (17)$$

$$s_{19} \qquad s_{20}s_{26}s_{27} = +1 \qquad (19)$$

$$s_{19}s_{22} \quad s_{42}s_{43} \quad s_{29}s_{35} = +1 \qquad (21)$$

A study of these equations shows that if s_{19} equals -1 then at least four other s's must equal -1, for example s_{21}, s_3, s_{43} and s_{20}. The failure of these may lead to other failures and so on. Thus if the sign relationship corresponding to s_{19} fails then at least five relationships of the set s_1–s_{48} must fail. This is greater than the total number of failures expected and it is therefore almost certain that s_{19} equals $+1$. Since so few sign relationships are expected to fail in this case, it might be expected that any sign relationship will hold if its failure will lead to three or more failures in all. By this method of examination several of the sign relationships are found which are almost certain to hold. Only these are accepted in a chain process of sign determination; any others are used only to find single signs which are not in their turn used to find other signs. In this way the failure of one of the less certain sign relationships will not propagate itself into a large number of incorrect sign determinations.

For purpurogallin this process enabled enough sign relationships to be established as 'inviolable' to find eight possible sets of signs for 27 of the original 32 reflexions. Of these solutions one had only two incorrect signs, d_3 and a_{10}. The incorrect sign determination for d_3 was due to the failures of s_{30} and s_{40}, which occur in relationship (10). However, since s_{30} and s_{40} each require only one other failure (each other in this case) these relationships were not used as part of the chain process of sign determination. The other incorrect sign, that for the axial reflexion a_{10}, was indicated as negative from s_{23} and s_{24}.

This process has also been tested with the data for the $hk0$ projection of α-glucose (McDonald & Beevers, 1952). An interesting case arises here involving the signs

$$\begin{array}{ll} a_3 = s(6, 10) & c_2 = s(3, 12) \\ a_8 = s(12, 4) & c_6 = s(9, 8) \\ b_2 = s(3, 11) & d_2 = s(6, 9) \\ c_1 = s(3, 2) & d_3 = s(6, 13) \end{array}$$

These give rise to the following sign relationships:

$$\begin{array}{ll} b_2c_1d_3 = s_{12} & -a_8c_2c_6 = s_{21} \\ -a_3c_1c_6 = s_{16} & -a_3c_1c_2 = s_{22} \\ -b_2c_1d_2 = s_{20} & -a_8d_2d_3 = s_{23} \end{array}$$

from which it is found that

$$s_{12}s_{16}s_{20}s_{21}s_{22}s_{23} = -1 \, .$$

This means that at least one of the six sign relationships must fail. It may also be found that s_{12}, s_{16} and s_{23} are almost certaily equal to $+1$, which indicates that the failure must be one of the sign relationships corresponding to s_{20}, s_{21} or s_{22}. These three sign relationships are therefore avoided in the process of sign development.

In conclusion it must be pointed out that the examination of relationships between sign relationships can be of use only in those cases, such as purpurogallin and α-glucose, where sign relationships hold with high probability. When sign relationships are expected to break down more often it is less likely that the failure of any one sign relationship will lead to so many other failures that definite conclusions may be drawn.

I must express my gratitude to Professor H. Lipson and Dr C. A. Taylor, who suggested purpurogallin as a suitable test structure and provided the necessary data.

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