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Direct Determination of Phases by the Use of Linear Equations between Structure Factors*

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From the Patterson function of a centrosymmetric structure it can be found that certain regions of the unit cell cannot contain any atoms. This information leads to the derivation of a function, the M function, which has the property that its product with the electron density is also the electron density. The convolution of the Fourier coefficients of the M function with the structure factors will therefore give the structure factors and this leads to linear equations of the form

$$F_h = \frac{1}{V} \sum_{h'=-\infty}^{h'=+\infty} X_{h'} F_{h-h'} \, .$$

There are at least as many equations as unknowns and, since the Fourier coefficients of the M function, the X's, can be evaluated, the equations can be solved to give information about the signs of the structure factors. The properties of the equations and their application to a hypothetical and a real structure are described. The lines along which further development may take place are given.

1. Theory

The basis of the method described in this paper will, for ease of illustration, be explained with reference to a one-dimensional example, but the arguments apply equally well in two or three dimensions. In Fig. 1, $\varrho(x)$ is a hypothetical electron-density distribution and $M(x)$ is any function which is unity wherever there is significant electron density in ϱ and takes the values zero or unity elsewhere as it pleases. For such a function M, the product of M and ϱ will be ϱ, *i.e.*

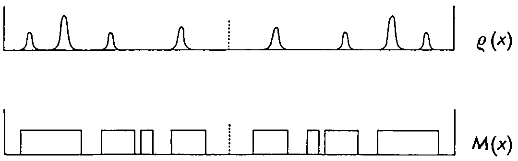

Fig. 1. One-dimensional electron-density distribution with a suitable M function.

$$\varrho(x) = M(x) . \varrho(x) \, . \tag{1}$$

Therefore, from the convolution theorem, the Fourier coefficients of ϱ, that is the structure factors, will be given by the convolution of the Fourier coefficients of ϱ and M, *i.e.*

$$F_h = \frac{1}{L} \sum_{h'=-\infty}^{h'=+\infty} X_{h'} F_{h-h'} \, , \tag{2}$$

where the F's are the structure factors and the X's are the Fourier coefficients of the M function. The length of the unit cell, L, may be replaced by the area, A, or the volume, V, in the two- or three-dimensional case. If the two functions ϱ and M are centrosymmetric as illustrated, then equation (2) represents a set of linear equations with at least as many equations as unknowns, although in practice there will be more equations than unknowns because of accidental absences. Thus, if a suitable M function can be found, the equations may be set up and solved, with a least-squares technique, to give the structure factors both in phase *and* magnitude!

A method which may nearly always be used to obtain an M function for a real structure is to derive it from the Patterson function as follows. In a centrosymmetric structure, with the origin placed on a centre of symmetry, a zero point in the Patterson function with a position vector $\mathbf{r}$ means there can be no significant electron density at a distance $\frac{1}{2}\mathbf{r}$ from the origin. Therefore, a function which is zero wherever there is insignificant Patterson density and unity elsewhere and reduced to half-scale will be the required M function.

2. The equations

Since the scale of M is half that of the Patterson function two unit cells of the M function will be required to cover one unit cell of the structure in one dimension. Regarding these two half-size cells as one of normal size results in the Fourier coefficients of the M function having even indices only. This will also be the case in two and three dimensions and it means that each equation can only contain structure factors belonging to the same parity group. A typical, though brief, set of equations linking structure factors belonging to the odd parity group of a one-dimensional structure is

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* See also p. 1056 of this issue.

$$\begin{aligned}(X_0-L+X_2)F_1+(X_2+X_4)F_3+(X_4+X_6)F_5&\\+(X_6+X_8)F_7&=0\\(X_2+X_4)F_1+(X_0-L+X_6)F_3+(X_2+X_8)F_5&\\+(X_4+X_{10})F_7&=0\\(X_4+X_6)F_1+(X_2+X_8)F_3+(X_0-L+X_{10})F_5&\\+(X_2+X_{12})F_7&=0\\(X_6+X_8)F_1+(X_4+X_{10})F_3+(X_2+X_{12})F_5&\\+(X_0-L+X_{14})F_7&=0\,.\end{aligned} \tag{3}$$

Normally the equations have a zero right-hand-side vector, but it is interesting to note that the equations relating structure factors with all even indices — the structure invariants — contain the zero order term F_0. Since the phase of this is known, terms containing F_0 will form the right-hand-side vector and the equations will be solved explicitly for the signs of the structure invariants. This means that for the structure invariant equations the F's will have to be on an absolute scale in order to calculate the magnitude of F_0, but this is not necessary for the other parity groups. In addition, for other parity groups there is no term of known sign, but multiplying each equation throughout by one of the unknown F's produces a squared term, the sign of which is known. The equations can then be solved explicitly for the product of the signs of two structure factors of the same parity group which again is a structure invariant. When a set of signs for each parity group has been obtained (explicitly for the structure invariant group and relatively for the others) the sign relationships inter-relating them can be used to form a consistent set of signs for the structure factors as a whole (Woolfson, 1957).

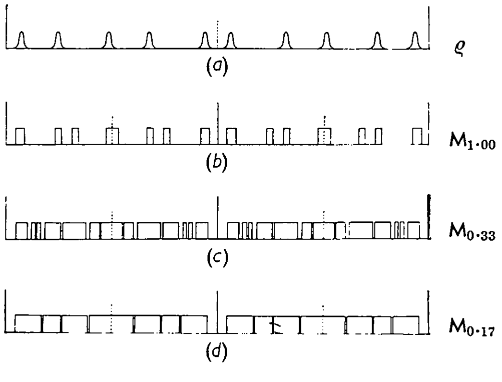

Fig. 2. (*a*) One-dimensional structure used in testing the properties of the equations. (*b*) 'Ideal' *M* function. (*c*) *M* function just capable of solving the structure. (*d*) *M* function obtainable from the Patterson function.

Equation (2) was tested with the structure factors and an M function derived from a hypothetical one-dimensional structure (Fig. 2(*a*) and (*b*)). The two sides of the equations were evaluated and compared and the comparison is shown in Table 1. The structure was centrosymmetric and contained five equal atoms in the asymmetric unit in a unit cell 100 Å long. The equations hold quite well and the agreement, expressed as a residual, is 1·2%. Most of the errors are probably due to termination of infinite series, since the structure factors were only calculated to the limit of copper $K\alpha$ radiation.

Table 1. *Comparison of left-hand sides and right-hand sides of equation* (2)

h	F_h	Σ	h	F_h	Σ	h	F_h	Σ
0	7·07	7·04	44	−1·56	−1·56	88	−0·31	−0·32
1	0·32	0·32	45	−0·16	−0·16	89	1·12	1·11
2	0·61	0·61	46	−3·25	−3·27	90	0·68	0·67
3	0·25	0·25	47	−0·30	−0·30	91	0·81	0·81
4	1·10	1·10	48	−1·04	−1·05	92	0·30	0·30
5	−1·49	−1·48	49	1·46	1·47	93	−0·57	−0·57
6	0·85	0·85	50	0·59	0·60	94	0·30	0·29
7	−0·69	−0·68	51	0·18	0·18	95	−0·04	−0·03
8	2·22	2·21	52	−2·44	−2·46	96	−0·48	−0·47
9	2·65	2·63	53	1·78	1·79	97	−0·29	−0·30
10	−3·22	−3·22	54	−0·56	−0·57	98	0·93	0·90
11	−1·90	−1·88	55	−0·61	−0·62	99	−0·36	−0·35
12	−0·85	−0·85	56	1·10	1·12	100	0·00	−0·01
13	−1·84	−1·83	57	2·44	2·47	101	−0·66	−0·65
14	−4·25	−4·24	58	1·33	1·34	102	−0·34	−0·32
15	1·10	1·10	59	1·09	1·11	103	−0·45	−0·42
16	−1·38	−1·37	60	0·02	0·01	104	−0·20	−0·20
17	2·14	2·12	61	−0·41	−0·41	105	−0·71	−0·69
18	−1·76	−1·77	62	1·66	1·67	106	0·40	0·37
19	−2·21	−2·21	63	−1·14	−1·15	107	0·31	0·30
20	−1·82	−1·82	64	0·19	0·19	108	−0·35	−0·33
21	1·40	1·38	65	1·33	1·35	109	−0·16	−0·14
22	−3·34	−3·33	66	1·72	1·73	110	−0·06	−0·05
23	0·39	0·38	67	−1·33	−1·34	111	−0·09	−0·07
24	3·22	3·22	68	0·32	0·32	112	−0·13	−0·13
25	1·79	1·77	69	−1·52	−1·55	113	0·19	0·16
26	1·38	1·38	70	−0·36	−0·37	114	0·02	0·02
27	0·10	0·09	71	−0·97	−0·99	115	0·78	0·69
28	0·18	0·19	72	−1·02	−1·03	116	0·08	0·08
29	0·68	0·67	73	−0·37	−0·39	117	−0·01	−0·01
30	1·81	1·81	74	1·26	1·28	118	0·04	0·05
31	−2·03	−2·03	75	−0·74	−0·75	119	0·31	0·27
32	4·10	4·11	76	−1·44	−1·45	120	−0·25	−0·21
33	2·65	2·66	77	0·04	0·03	121	0·19	0·14
34	1·02	1·02	78	−0·97	−0·97	122	0·14	0·13
35	−1·39	−1·39	79	−0·43	−0·43	123	0·25	0·19
36	1·11	1·12	80	−0·50	−0·49	124	0·19	0·15
37	−2·09	−2·09	81	0·52	0·52	125	−0·12	−0·10
38	0·16	0·16	82	0·52	0·52	126	−0·19	−0·13
39	−1·64	−1·63	83	1·47	1·49	127	0·01	0·02
40	−0·58	−0·58	84	−1·00	−0·99	128	−0·08	−0·07
41	1·86	1·88	85	0·39	0·39	129	−0·34	−0·26
42	0·10	0·10	86	0·41	0·41			
43	−2·83	−2·84	87	0·12	0·14			

RESIDUAL = 1·2%.

3. Solution of the equations

It seemed probable from the original tests of the equations that they could be solved to give a fairly reliable solution for both the magnitudes and phases of the structure factors, but the first attempts at obtaining such a solution indicated otherwise. The Fourier coefficients of an M function, derived from the Patterson function of the structure as described in § 1, were used to set up the equations, but the solutions bore little resemblance to the original structure factors. It was thought, however, that the more 'zero' as opposed to 'unity' there was in the M function, the better the solution of the equations would be. This is reasonable since it increases the magnitudes of the elements on the main diagonal of the left-hand-side matrix (see equation (3)) and makes the equations better conditioned. It can also be explained physically by the fact that this allows less space in the unit cell into which the atoms can be placed and so restricts to a greater degree the form the structure can take. In addition, the magnitudes of the structure factors were incorporated into the equations by writing $F_h=|F_h|S(h)$; the equations could then be solved for the S's, the values of which should be either $+1$ or -1.

With these ideas in mind, and using a knowledge of the structure, an 'ideal' M function was generated which contained the maximum amount of 'zero' compatible with the restrictions placed on the function. One such restriction was that it should have the same symmetry as that which could be obtained from the Patterson function, as shown in Fig. 2(*b*), since in any real case there would be such symmetry. When the equations set up with the use of the Fourier coefficients of this ideal M function and the magnitudes of the structure factors were solved, 36 out of the 63 signs determined were correct. This was obviously not very satisfactory, but it was noted that the signs of the S's corresponding to the largest structure factors were correct, with values of the order of ± 1, while the signs of the S's corresponding to structure factors of smaller magnitude were quite random, compared with the true signs, and with values ranging from -124 to $+17$. However, for these particular variables the equations were poorly conditioned because the coefficients of the equations were now multiplied by the magnitudes of the structure factors, making the coefficients for a particular S small when the magnitude of the corresponding structure factor was small. A solution in which all the S's were determined correctly may have been expected, since perfect data were used with an ideal M function. The fact that such a solution was not obtained in this case can only be due to the termination-of-series errors, apart from the poor conditioning mentioned earlier. Equation (2) represents an infinite number of equations with an infinite number of unknowns and these obviously cannot be handled in practice. The equations actually used are finite in number with a finite number of unknowns, and even with perfect data can be no more than an approximation to equation (2); it will be recalled that under these conditions the agreement between the two sides of equation (2), expressed as a residual, is 1·2%.

Some of the structure factors of low magnitude were made equal to zero by omitting the corresponding columns from the left-hand-side matrix, $S(h)=0$ being thought a better approximation to ± 1 than -124 or $+17$. This resulted in more equations than unknowns and when these were solved, by a least-squares method, the sign determination for the remaining structure factors was greatly improved. It was found empirically that neglecting all structure factors whose magnitudes were less than 1/10 of that of the largest gave the greatest likelihood of the solution being correct. When this was done, 36 out of 39 signs were correctly determined. The remainder were obtained by accepting the signs of the largest structure factors, which were the most likely to be correct, adding their contributions to the right-hand side of the equations and re-solving them, again neglecting the structure factors whose magnitudes were less than 1/10 of the maximum now present. By resolving the equations two or three times in this way all the signs were eventually determined correctly.

However, an ideal M function, which is unobtainable in practice, had been used for the sign determination, so more realistic functions were generated and used in turn in the equations to find out how much of the M function could be made equal to unity before the sign determination became unreliable. In the extreme case, where the M function is unity everywhere, the equations become $F_h=F_h$ and cannot be solved. It was convenient to define a parameter of the M function as $\varphi=l_0/l_i$, where l_0 is the length of the unit cell of the function equal to zero and l_i is the length of the unit cell equal to zero in the ideal case. Thus, in the ideal case, $\varphi=1{\cdot}00$ (Fig. 2(*b*)) and for an M function which is everywhere equal to unity, φ will be zero. With this nomenclature, it was found that the M function obtainable from the Patterson function for this particular structure corresponded to $\varphi=0{\cdot}17$ (Fig. 2(*d*)) and that the M function which was just capable of solving the structure, by the method described, had a value of φ equal to 0·33 (Fig. 2(*c*)). It should be noted that $M_{0\cdot33}$ is much nearer the practical than the ideal case, and it is rather surprising that the structure can be solved with the apparently small amount of information contained in this M function.

4. Further properties of the *M* function

So far only 'well-behaved' M functions have been mentioned, that is those which multiply all electron density by unity and have their zero points only where there is insignificant electron density. However, it is quite possible to generate an M function which multiplies a significant amount of electron density by zero, and it is important to see what effect this has on the sign-determining properties of the equations.

Since M functions with a low value of φ give poor sign determination, functions with a high φ value were chosen and modified to multiply various amounts of electron density by zero. It was found that sign determination deteriorated with the modified M functions, the amount of deterioration depending upon the value of φ and the way in which the electron density was multiplied by zero. With a value of φ equal to unity and electron density multiplied by zero in a symmetrical way, that is to multiply only the centres of the atoms by unity, the sign determination was very good, but when the atoms were multiplied by zero in an unsymmetrical way many wrong sign indications resulted. As soon as the value of φ was lowered the sign determination became very poor indeed, even when only a slight amount of electron density was multiplied by zero. It appears, therefore, that in the practical case the good sign-determining properties of the equations are critically dependent on all significant electron density being multiplied by unity in the M function.

This has direct bearing on the 'sharpening' of the Patterson function to increase the volume of insignificant density in order to improve the quality of the derived M function. It is well known that series termination errors can become quite significant when the structure factors are 'sharpened' and these give rise to spurious detail in a Fourier synthesis. As far as the M function is concerned this spurious detail, whether it be negative or positive, is real and therefore must be multiplied by unity as is the remainder of the electron density. Failure to do this greatly detracts from the ability of the M function to determine signs correctly. This has been borne out in practice and it is found that little is to be gained in the sharpening of the Patterson function. If more data were available, such as can be collected by using molybdenum as opposed to copper radiation, this would reduce the series-termination errors and it ought to be possible to sharpen the Patterson function a certain amount without serious consequences.

5. Practical results

Even though the method had failed to solve the one-dimensional structure, it was possible that an M function as good as $M_{0\cdot33}$, or better, could be obtained for a real structure in three dimensions. Two-dimensional work was out of the question because a two-dimensional Patterson function would not contain sufficient areas of negligible density. A known structure was therefore tackled for which three-dimensional data were available, namely tetraethyldiphosphine disulphide (Dutta & Woolfson, 1961). This was chosen because it contained only six atoms in the asymmetric unit and had a small unit cell. Even with this small cell, only reflexions out to a $\sin\theta$ value of 0·9 for copper $K\alpha$ radiation could be used in the sign determination because of the limited store in the Mercury computer used to perform the calculations.

The M function of the structure was obtained by the method described in § 2 and the equations for the structure invariants were set up and solved, using 111 reflexions out of the 149 which had been observed. The signs determined in this way were quite random as compared with the true signs, and the structure could not possibly have been solved. However, a parallel calculation was performed by using the moduli of the structure factors calculated from the known structure and the sign determination this time was very good, 57 signs being determined correctly out of 75. The calculations were stopped at this stage because, from experience, it was known that a result as good as this can easily be improved to give all the signs correctly.

The main difference between the two calculations was in the quality of the Patterson function. The experimental data with their random errors produced a Patterson function with a lot of background density and much spurious detail. This resulted in a poor M function with a φ value estimated at about 0·17, but with accurate data the background of the Patterson function was greatly reduced and an M function with an estimated φ value of about 0·48 was obtained. This is discussed further in § 7.

6. Weighted solution of the equations

It will be recalled that the equations give poor sign determination until the signs of the smaller structure factors are forced to be zero by omitting these variables from the left-hand-side matrix. A better way of approximating these signs to ± 1 is to add to the original equations further equations of the form

$$\omega S(h) = 0$$

and then to solve the resulting set by least squares. $S(h)$ is the sign of F_h and ω is a weighting factor inversely proportional to $|F_h|$. The constant of proportionality is defined as

$$W \cdot \frac{\text{r.m.s.}\,a_{rs}}{\text{r.m.s. } 1/|F_h|}$$

where r.m.s. $1/|F_h|$ is the root mean square of all the values of $1/|F_h|$, r.m.s. a_{rs} is the root mean square of the elements of the original matrix and W is an arbitrary constant. It has been found empirically that the best value for W is unity. One equation of this type is included for each variable and in this way the signs of the small structure factors which would normally tend to take any value between +100 and −100 are approximated to zero while the values the signs of large structure factors can take are almost unrestricted. The improvement in the sign determination is such that the one-dimensional structure may be solved by using an M function with a value of φ equal to 0·25.

7. Future developments

The Manchester University Mercury computer has been used to perform all the calculations and the backing store of this machine is limited to 10,752 words when Autocode is used. When the extra weighting equations just described are used this limits the number of reflexions which can be accommodated to 65. Even quite a simple structure will probably have more than 100 reflexions in each parity group in three-dimensional data, so work on actual structures will be held up until the new Atlas computer becomes fully available early in 1963. The computing times on Mercury tend to be rather long and it takes about 5 hours to carry out one of the three-dimensional calculations performed on tetraethyldiphosphine disulphide data; to solve this structure completely would take an estimated 20 hours of machine time. Calculations of this length are not really feasible on Mercury, so on this account also development will be held up until Atlas becomes available, but a number of ideas are waiting to be tried out when time and computer permit and are outlined below.

The weighting equations mentioned in § 6 serve to approximate the solutions of the equations to zero, this being nearer to ±1 than the extreme values for some S's obtained without the extra equations. A better approximation to ±1 would be achieved by using equations of the form

$$\omega(S^2(h)-1)=0\ .$$

This introduces quadratic equations which must be solved simultaneously with the original linear equations, and there exists a mathematical technique for doing this.

The sign determination with the experimental data for tetraethyldiphosphine disulphide failed because of the poor quality of the Patterson function. The present method of deriving the M function from the Patterson function is very much dependent on the background density of the latter. If a contour level is chosen such that any Patterson density below this is considered to be negligible, it is quite possible that some of the spurious detail can come above this contour and so detract from the quality of the M function. On the other hand, a contour level which is the height of the lowest genuine peak ought to be well above all background density. Using this contour to form the boundary between the 'zero' and 'unity' in the M function will produce an M function which will multiply a certain amount of electron density by zero because the Patterson peaks will be effectively narrowed. This can be avoided by making the M function unity within the necessary radius around all the volumes of 'unity' already present and the remainder of the M function will then be zero. This method of deriving the M function will be much less dependent on the background of the Patterson function and so less dependent on random errors in the intensity data.

A very useful prediction to make is the probability of the signs given by the equations being correct. This could well give an indication of the number of signs from a solution of the equations which can be accepted as being correct in order to add these terms to the right-hand side and redetermine the remainder. A method for doing this has been worked out in theory but has not yet been tried out in practice.

Even if these equations do not prove to be very powerful in determining signs directly from the Patterson function, they could still serve to determine the signs of the rest of the structure factors if once the phases of the structure invariants were known. A Fourier synthesis using the structure invariants alone contains the whole structure with extra translations added to the space group symmetry. An M function derived from such a Fourier synthesis would be of much better quality than one derived directly from the Patterson function, and the sign determination using this M function will be very good.

These equations can be put to further use as a test for sets of signs obtained by established direct methods. The exact form this test will take is not yet known, but many possibilities present themselves and these will be investigated in the course of time. The advantages of the proposed new test are that the test function will be completely independent of overlap and of different types of atom and that a one-dimensional summation only will be required as opposed to the two-dimensional summation necessary in the Z-test (Woolfson, 1958). The latter point is important because in the computer application of direct methods large numbers of plausible sets of signs may be generated which must be tested in the minimum amount of time. However, it remains to be seen whether this new test will be sufficiently discriminating against wrong sets of signs to allow the correct set to be identified.

8. Discussion

In § 2 it was mentioned that the equations are solved for structure invariants only, that is a structure factor whose indices are all even or the product of two structure factors belonging to the same parity group. This is in contrast with Harker & Kasper inequalities (Harker & Kasper, 1948), sign relationships (Sayre, 1952) and the method of Hauptman & Karle (1953), which in general give the sign of a product of three structure factors or of a single structure invariant. Only in a special application of Harker & Kasper inequalities is the sign of a product of two structure factors given and then it is always negative. It should be noted that apart from this one negative indication, inequality relationships can only be used to prove signs positive. In addition, sign relationships and the method of Hauptman & Karle (1953) can give strong indications that the sign of a structure invariant (either a single structure factor or the product of three related structure factors) is positive, but can only give very weak negative indications. On the other hand, the latest method of Hauptman & Karle (1962), which calculates phases directly from the Patterson function, can give negative sign indications as well as positive, and the method of sign determination described in this paper will determine the sign of a structure invariant, with equal facility and probability of it being correct, whether it be positive or negative.

From § 5 it appears that at the present stage of development simple centrosymmetric structures may be solved provided that accurate three-dimensional data are available. Using some of the ideas outlined in §§ 6 and 7 should improve the method and it remains to be seen whether it will turn out to be comparable to established direct methods in its power to solve structures. At the moment no predictions can be made about the method and it remains an interesting academic project which shows enough promise to make further research worth while. The limitations of the method are not yet precisely known, but obviously there will be a limit to the number of atoms in a unit cell of a structure whose Patterson function will contain a sufficient volume of negligible density. This limit is thought to be about 80 atoms.

As was mentioned previously, the use of this method is almost certain to be limited to three-dimensional work, but this should not be a great disadvantage in these days of automatic diffractometers and high-speed computers. Indeed, it is well suited to a completely automatic solution of a structure, since very few decisions must be made during the course of the calculations and these could easily be programmed in a computer.

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Diffraction by Atoms Distributed at Random at Lattice Sites

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A recurrence procedure is given for the calculation of the mean amplitude, its dispersion, and the mean intensity diffracted by a set of lattice sites occupied at random by atoms of various kinds. A three-dimensional lattice is treated when no correlations are present. An example (with no correlations) is given which pertains to Mossbauer scattering, and explicit formulae are given if there are only two types of atom with correlations.

1. Introduction

If a crystal contains atoms of iron of normal isotopic composition, the iron atoms will all be situated at lattice sites, but 2·2% of the sites will be occupied by ^{57}Fe atoms, distributed at random. These ^{57}Fe atoms diffract X-rays in exactly the same way as the other iron atoms except for wavelengths in the neighborhood of 0·86 Å. At about this wavelength X-rays can be resonantly scattered by the nuclei of the ^{57}Fe atoms as well as the atomic electrons. Since the phase and amplitude of the nuclear resonance scattering can be varied in a controlled way (Mossbauer effect), interference effects between scattering from the atomic electrons and the nuclei are to be expected, and these have very interesting implications for the phase problem. From the point of view of diffraction, however, the problems are: to calculate the average amplitude scattered from a set of atoms distributed at random at lattice sites, to see whether the distribution of amplitude is well defined about the average, and, if it is, to calculate the interference between this amplitude and the amplitude of the normal X-ray scattering from the atomic electrons. This will be discussed briefly in § 2.

In finding the solution to this problem, a simple recurrence procedure was found which could be used to obtain fairly complete statistical information about more general problems, and the purpose of this note is to apply the procedure to the case where lattice sites are occupied at random by several different types of atom. The results for the average scattered intensity seem to be known in the case where there are no correlations between atoms of various types. Nearest neighbor correlations have been considered by Hendricks & Teller (1942), Wilson (1942), and Jagodzinski (1954), but again only for the intensity.

Consider an ideal lattice of N sites which may be