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

By A. M. Cormack

Physics Department, Tufts University, Medford, Mass., U.S.A.

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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 occupied at random by atoms of several kinds. There will be a large number of possible configurations of the atoms in the sites. Let the amplitude diffracted from the ith configuration be z_i and let the probability of the occurrence of this configuration be p_i. The p_i could be calculated explicitly by the binomial distribution, but this is unnecessary. We define the mean amplitude Z_N, the mean intensity, I_N, and the dispersion of the amplitude about its mean value, σ_N, by the equations

$$Z_N = \sum_i p_i z_i \,. \tag{1}$$

$$I_N = \sum_i p_i |z_i|^2 \,. \tag{2}$$

$$\sigma_N^2 = \sum_i p_i |z_i - Z_N|^2 \,. \tag{3}$$

It is easily seen that these are connected by the equation

$$\sigma_N^2 = I_N - |Z_N|^2. \tag{4}$$

If one is dealing with a single crystal, the configuration of atoms is unique and so is the diffraction pattern. Under these circumstances, σ_N has its usual statistical significance, namely, it gives information about the probability that the amplitude diffracted by the sample will differ by a prescribed amount from the amplitude diffracted by an average crystal. But if the sample contains many crystals with different configurations, the 'diffuse' scattering will be represented by σ_N.

In § 2 these quantities will be calculated for one- and three-dimensional lattices under the assumption that the probability that an atom of a particular type occupies a lattice site is independent of the types of atoms in neighboring sites. In § 3 correlations between nearest neighbors will be discussed for the one-dimensional case only. We shall speak of 'an atom of type k with atomic scattering factor F_k'. Clearly, for 'atom' one may read unit cell or layer in the three- and one-dimensional cases. F_k will be permitted to be complex because individual atomic scattering factors may be complex or because atoms may be displaced from lattice sites, but such displacements must be independent of the site and depend only on the type of atom.

2. No correlations

(a) The one-dimensional lattice

Let there be N lattice sites situated at the points $n\mathbf{a}\,(n=0, 1, 2, \ldots, N-1)$, and let the incident and diffracted radiation be described by the propagation vectors $\mathbf{k}_0$ and $\mathbf{k}$ respectively, where $k=k_0=2\pi/\lambda$. Then the amplitude diffracted by an atom of type k at $n\mathbf{a}$ will be $F_k \exp[i(\mathbf{k}-\mathbf{k}_0).n\mathbf{a}]$ and, if we write $(\mathbf{k}-\mathbf{k}_0).\mathbf{a}=\varphi$, this becomes $F_k \exp(in\varphi)$. Let w_k be the probability that an atom of type k occupies a site and suppose that this is independent of the site and of the atoms in neighboring sites. If a site must be occupied by one of the possible types of atoms $\sum_k w_k = 1$. Vacant sites can be included by considering them to be occupied by atoms with $F=0$.

Suppose that the first n sites are occupied and let z_i be the amplitude scattered by the ith configuration, which has the probability p_i. Then the probability that an atom of type k will occur in the $(n+1)$th site is w_k and the amplitude scattered by this new configuration will be $z_i + F_k \exp(in\varphi)$. Hence, the average scattered amplitude for a lattice with $n+1$ sites filled, Z_{n+1}, is given by

$$\begin{aligned} Z_{n+1} &= \sum_i \sum_k p_i w_k (z_i + F_k \exp[in\varphi]) \\ &= \sum_i p_i z_i \sum_k w_k + \exp[in\varphi] \sum_k w_k F_k \sum_i p_i \,. \end{aligned} \tag{5}$$

$$\sum_i p_i z_i = Z_n, \quad \sum_i p_i = \sum_k w_k = 1 \,,$$

and if we write the average scattered amplitude from a single site as Z, where

$$Z = \sum_k w_k F_k \tag{6}$$

equation (5) becomes the recurrence relation

$$Z_{n+1} = Z_n + \exp[in\varphi] Z \,. \tag{7}$$

Since $Z_1 \equiv Z$, equation (7) yields for the whole lattice:

$$\begin{aligned} Z_N &= \sum_{n=0}^{N-1} \exp[in\varphi] Z \\ &= Z \exp(i[(N-1)\varphi/2]) \frac{\sin(N\varphi/2)}{\sin(\varphi/2)} \,. \end{aligned} \tag{8}$$

The mean amplitude is thus the same as the amplitude from a lattice of identical atoms with an atomic scattering factor equal to the average atomic scattering factor Z. The exponential in equation (8) can be removed by shifting the origin of the coordinates to the center of the lattice. It has been assumed that the sites are filled seriatim. This has been done only to simplify the notation, for equation (7) would still be true if Z_n referred to the average amplitude from any occupied sites and Z_{n+1} referred to the average amplitude from these sites together with the $(n+1)$th site. This point will be referred to in connection with the three-dimensional lattice.

It is useful to introduce the average intensity, I, and the dispersion of the amplitude, σ, for an isolated site:

$$I \equiv I_1 = \sum_k w_k |F_k|^2 \,, \tag{9}$$

and, using equation (4),

$$\sigma^2 \equiv \sigma_1^2 = \sum_k w_k |F_k|^2 - |\sum_k w_k F_k|^2. \tag{10}$$

The calculation of I_n follows similar lines. From the defining equation (2), and using the argument above,

$$I_{n+1} = \sum_i \sum_k p_i w_k |z_i + F_k \exp[in\varphi]|^2.$$

Using equations (2), (9), (1) and (7), this becomes

$$I_{n+1}=I_n+I_1+2Rl\,\{\exp[-in\varphi]Z_nZ_1^*\}\,,$$

which, by equation (8), becomes

$$I_{n+1}=I_n+I_1+2|Z|^2Rl\,\{\exp[-in\varphi]\sum_{l=0}^{n-1}\exp[il\varphi]\}\,.$$

Since $I_1=I$, this recurrence relation yields

$$\begin{aligned}I_N &= NI+2|Z|^2Rl\sum_{l=1}^{N-1}(N-l)\exp[-il\varphi]\\ &= NI-N|Z|^2+|Z|^2\frac{\sin^2(N\varphi/2)}{\sin^2(\varphi/2)}\,.\end{aligned}$$

Hence, by equation (10),

$$I_N=N\sigma^2+|Z|^2\frac{\sin^2(N\varphi/2)}{\sin^2(\varphi/2)}\,, \tag{11}$$

and by equations (4) and (8)

$$\sigma_N^2=N\sigma^2. \tag{12}$$

If it is necessary to use the distribution function of the amplitude, care must be exercised with σ_N. In the complex plane the points z_i are distributed about Z_N and σ_N is an exact measure of their spread. But the shape of the distribution of the z_i about Z_N depends on the angle which Z_N makes with the real axis. For example, if the F_k are real, at a diffraction maximum ($\varphi=2m\pi$), the z_i are all real and the distribution of the amplitude is confined to the real axis. If Z_N does not lie on the real axis, the distribution of the z_i does not in general have any simple symmetry about Z_N and in cases like this more accurate measures of the dispersions should be introduced, for example, the dispersions along the real and imaginary axes, and these can be found in the same way as σ_N.

(*b*) *The three-dimensional lattice*

The treatment here is almost exactly the same as for the one-dimensional lattice. Let the lattice sites be at $\mathbf{r}=h\mathbf{a}+k\mathbf{b}+l\mathbf{c}$ where $h=0, 1, 2, \ldots, \mathrm{N}_1-1$; $k=0, 1, 2, \ldots, N_2-1$; $l=0, 1, 2, \ldots, \mathrm{N}_3-1$, and let $(\mathbf{k}-\mathbf{k}_0).\mathbf{a}=\alpha$, $(\mathbf{k}-\mathbf{k}_0).\mathbf{b}=\beta$, $(\mathbf{k}-\mathbf{k}_0).\mathbf{c}=\gamma$. Let p_i and z_i have the same meanings as before and let Z, I, and σ, Z_n, I_n and σ_n^2 be given by equations (6), (9), (10), (1), (2), and (3) respectively, and let n stand for any n occupied sites. Suppose that the site (h, k, l) is not one of these n sites and let the $(n+1)$th atom be added at (h, k, l). Then, as before,

$$\begin{aligned}Z_{n+1} &= \sum_i\sum_m p_iw_m(z_i+F_m\exp[i(h\alpha+k\beta+l\gamma)])\\ &= Z_n+Z\exp[i(h\alpha+k\beta+l\gamma)]\,.\end{aligned} \tag{13}$$

Thus the atom at the site h, k, l contributes $Z\exp[i(h\alpha+k\beta+l\gamma)]$ to the sum so

$$\begin{aligned}Z_N &= Z\sum_h\sum_k\sum_l\exp[i(h\alpha+k\beta+l\gamma)]\\ &= Z\exp[i[(N_1-1)\alpha+(N_2-1)\beta+(N_3-1)\gamma]/2]\\ &\quad\times\frac{\sin(N_1\alpha/2)\sin(N_2\beta/2)\sin(N_3\gamma/2)}{\sin(\alpha/2)\sin(\beta/2)\sin(\gamma/2)}\,.\end{aligned} \tag{14}$$

I_N and σ_N are derived in the same way as in the one-dimensional case and are given by

$$I_N=N\sigma^2+|Z|^2\frac{\sin^2(N_1\alpha/2)\sin^2(N_2\beta/2)\sin^2(N_3\gamma/2)}{\sin^2(\alpha/2)\sin^2(\beta/2)\sin^2(\gamma/2)} \tag{15}$$

$$\sigma_N^2=N\sigma^2 \tag{16}$$

where N stands for $N_1N_2N_3$, the number of sites in the lattice.

Thus, both the one- and three-dimensional lattices behave as though they were occupied by atoms with an average scattering amplitude Z, except for the intensity which contains the term $N\sigma^2$ which can represent diffuse scattering.

(*c*) *A special case: Mossbauer scattering*

As a special case consider a lattice in which sites are occupied by one type of atom with a probability w and are otherwise unoccupied, *i.e.* the probability of a site being unoccupied is $1-w$. Then we have

$$Z=pF,\ I=p|F|^2,\ \sigma^2=p(1-p)|F|^2, \tag{17}$$

and

$$\begin{aligned}I_N &= N_1N_2N_3\sigma^2+p^2|F|^2\\ &\quad\times\frac{\sin^2(N_1\alpha/2)\sin^2(N_2\beta/2)\sin^2(N_3\gamma/2)}{\sin^2(\alpha/2)\sin^2(\beta/2)\sin^2(\gamma/2)}\,.\end{aligned} \tag{18}$$

At a diffraction peak ($\sin(\alpha/2)=\sin(\beta/2)=\sin(\gamma/2)=0$), the second term in equation (18) has the value $p^2|F|^2(N_1N_2N_3)^2$, and the first has the value $N_1N_2N_3p(1-p)|F|^2$. Hence the statistical fluctuations or diffuse scattering (represented by $N_1N_2N_3\sigma^2$) will be unimportant provided

$$(N_1N_2N_3)^2p^2|F|^2 \gg N_1N_2N_3p(1-p)|F|^2$$

that is, provided

$$N_1N_2N_3+1 \gg 1/p\,. \tag{19}$$

In even a small crystal, equation (19) is very well fulfilled for any reasonable value of p, so we can conclude that within the diffraction peak the average scattering from a set of randomly occupied lattice sites will be very nearly the same, both in amplitude and intensity, as the scattering from the same lattice with all the sites occupied by atoms with an atomic scattering factor pF.

In the case of scattering from a lattice of a single chemical element of which only one isotope with fractional abundance p undergoes Mossbauer scattering, the ordinary X-ray scattering may be described by an atomic scattering factor F_X, and the Mossbauer scattering by a scattering factor F_M. Within a diffraction peak, the resultant scattering will be described by ascribing to each atom a scattering factor F_X+pF_M and calculating the intensity accordingly.

3. Correlation in a one-dimensional lattice

Consider all configurations of atoms in the first n sites. Let $s_{ir}^{(n)}$ and $z_{ir}^{(n)}$ be respectively the probability of the rth configuration of those which have an atom of type i in the nth site, and the amplitude scattered by that configuration. Then Z_n, I_n, and σ_n are defined as before:

$$Z_n = \sum_i \sum_r s_{ir}^{(n)} z_{ir}^{(n)} . \tag{21}$$

$$I_n = \sum_i \sum_r s_{ir}^{(n)} |z_{ir}^{(n)}|^2 . \tag{22}$$

$$\sigma_n^2 = \sum_i \sum_r s_{ir}^{(n)} |z_{ir}^{(n)} - Z_n|^2 . \tag{23}$$

Let $w_i^{(n)}$ be the probability that an atom of type i occurs at the nth site so that

$$\sum_r s_{ir}^{(n)} = w_i^{(n)} . \tag{24}$$

Let p_{ik} be the probability that an atom of type i in any site will be followed by an atom of type k in the next site. The p_{ik} therefore give the correlations between the nearest neighbors. The relation between the $w_i^{(n+1)}$ and $w_i^{(n)}$ is

$$w_k^{(n+1)} = \sum_i w_i^{(n)} p_{ik} \tag{25}$$

and this can be compactly written by letting the $w_i^{(n)}$ be the components of a row matrix $\mathbf{w}^{(n)}$ and the p_{ik} be the components of a matrix P. With this notation, equation (25) becomes

$$\mathbf{w}^{(n+1)} = \mathbf{w}^{(n)} P \tag{26}$$

and successive applications of this equation yield

$$\mathbf{w}^{(n)} = \mathbf{w}^{(1)} P^{n-1} \tag{27}$$

where $\mathbf{w}^{(1)}$ gives the probabilities for the first site.

If the rth configuration of those which end in an atom of type i is followed by an atom of type k in the $(n+1)$th site, the diffracted amplitude will be $z_{ir}^{(n)} + F_k \exp(in\varphi)$ and the probability of the new configuration will be $s_{ir}^{(n)} p_{ik}$, hence the mean scattered amplitude for $n+1$ sites will be

$$\begin{aligned} Z_{n+1} &= \sum_i \sum_k \sum_r s_{ir}^{(n)} p_{ik} (z_{ir}^{(n)} + F_k \exp[in\varphi]) \\ &= Z_n + \exp[in\varphi] \sum_k F_k \sum_i p_{ik} \sum_r s_{ir}^{(n)} \\ &= Z_n + \exp[in\varphi] \sum_k F_k \sum_i w_i^{(n)} p_{ik} \end{aligned}$$

which can be written

$$Z_{n+1} = Z_n + \exp[in\varphi] \mathbf{w}^{(n)} P \mathbf{F} \tag{28}$$

where F is a column matrix with components $F_1, F_2, \ldots, F_k, \ldots$. By (27) this becomes

$$Z_{n+1} = Z_n + \exp[in\varphi] \mathbf{w}^{(1)} P^n \mathbf{F} \tag{29}$$

hence, since

$$Z_1 = \sum_k w_k^{(1)} F_k = \mathbf{w}^{(1)} \mathbf{F},$$

$$Z_n = \mathbf{w}^{(1)} \sum_{l=0}^{n-1} \exp[il\varphi] P^l \mathbf{F}. \tag{30}$$

This shows that in principle the effect of the probability distribution of atoms in the first site persists throughout the lattice. The series of matrices occurring in equation (30) can be summed but this does not seem worth while. If the effect of the distribution in the first site makes itself felt throughout the lattice, the statistics will be applicable only to an examination of the diffraction patterns from a large number of different crystals with different initial distributions (*e.g.* caused by different conditions of growth). This would seem to be the hard way to investigate the initial distributions. The more interesting case is that in which the effect of the initial distribution disappears and an equilibrium distribution is established. This will happen in any case unless some of the $w_i^{(1)}$ are zero and some of the p_{ik} are inordinately small. If the effect of the initial distribution disappears rapidly, we might as well assume that $w_i^{(1)}$ is the equilibrium distribution, and this will be assumed in what follows.

Let $\mathbf{w}$ be the equilibrium distribution and assume that no element of P is either zero or one, so that any atom may be followed by any other atom. Under these conditions, it can be shown (Gantmacher, 1959) that: (i) the limit $\lim_{n\to\infty} \mathbf{w}^{(1)} P^n$ exists and is equal to $\mathbf{w}$ and is independent of $\mathbf{w}^{(1)}$, (ii) that $\mathbf{w}$ is the eigenvector of P belonging to the simple eigenvalue one, that is

$$\mathbf{w} = \mathbf{w} P \tag{31}$$

and hence for any n

$$\mathbf{w} = \mathbf{w} P^n . \tag{32}$$

Equation (31) enables one to calculate $\mathbf{w}$ from P. Equation (29) may be rewritten

$$Z_{n+1} = Z_n + \exp[in\varphi] Z \tag{33}$$

where Z is given by equation (6), and equation (30) becomes

$$Z_N = Z \exp[i(N-1)\varphi/2] \frac{\sin(N\varphi/2)}{\sin(\varphi/2)} . \tag{34}$$

This is the same as equation (8), so correlations have no effect on the mean amplitude.

It is useful to introduce the partial average amplitudes $\bar{z}_i^{(n)}$ defined by

$$w_i^{(n)} \bar{z}_i^{(n)} = \sum_r s_{ir}^{(n)} z_{ir}^{(n)} . \tag{35}$$

In terms of these equation (33) becomes

$$\sum_i w_i^{(n+1)}\bar{z}_i^{(n+1)} = \sum_i w_i^{(n)}\bar{z}_i^{(n)} + \exp\,[in\varphi] \sum_i w_i F_i$$

but since $w_i^{(n+1)} = w_i^{(n)} = w_i$ this becomes

$$\sum_i w_i(\bar{z}_i^{(n+1)} - \bar{z}_i^{(n)} - \exp\,[in\varphi]F_i) = 0$$

so

$$\bar{z}_i^{(n+1)} = \bar{z}_i^{(n)} + \exp\,[in\varphi]F_i\,,$$

and since $z_i^{(1)} = F_i$,

$$\bar{z}_i^{(n)} = F_i \sum_{l=0}^{n-1} \exp\,[il\varphi]\,. \tag{36}$$

By the argument used before,

$$\begin{aligned} I_{n+1} &= \sum_i \sum_r \sum_k s_{ir}^{(n)} p_{ik} |z_{ir}^{(n)} + F_k \exp\,[in\varphi]|^2 \\ &= I_n + I + 2Rl\,\{\exp\,[-in\varphi] \sum_k F_k^* \sum_i p_{ik} \sum_r s_{ir}^{(n)} z_{ir}^{(n)} \end{aligned}$$

where I is given by equation (9). By equations (35) and (36) this becomes

$$\begin{aligned} I_{n+1} &= I_n + I + 2Rl \\ &\times \{\exp\,[-in\varphi] \sum_{l=0}^{n-1} \exp\,[il\varphi] \sum_k F_k^* \sum_i w_i F_i p_{ik}. \end{aligned} \tag{37}$$

Let

$$\mathscr{F} = \sum_i \sum_k w_i F_i p_{ik} F_k^*\,. \tag{38}$$

Then, since $I_1 = I$, equation (37) may be used to give

$$\begin{aligned} I_N &= N[I - Rl\{\mathscr{F}(1 + i \cot\,(\varphi/2))\}] \\ &\quad + \frac{\sin\,(N\varphi/2)}{\sin^2\,(\varphi/2)} Rl\{i\mathscr{F} \exp\,[-in\varphi/2]\}\,. \end{aligned} \tag{39}$$

Again, σ_N^2 can be found from equations (3), (34), and (39). If there are no correlations, $p_{ik} = w_k$, and $\mathscr{F} = |Z|^2$, so equation (39) reduces to equation (11).

Case of two types of atom

In this, the simplest case, explicit formulae may readily be written down, principally due to the fact that even if F_1 and F_2 are complex, $\mathscr{F}$ must be real, which is not in general true if more types of atom are considered. The most general correlation matrix can be written

$$P = \begin{vmatrix} (1-\mathrm{a}) & a \\ b & (1-b) \end{vmatrix} \tag{40}$$

and the solution to equation (31) is

$$w_1 = \frac{b}{a+b}\,, \quad w_2 = \frac{a}{a+b}\,. \tag{41}$$

If there are no correlations, $p_{ik} = w_k$, which implies $a+b = 1$ in this case. Thus the ratio b/a determines the relative probabilities of the two kinds of atom, and the departure of $a+b$ from unity determines the amount of correlation. The expression for $\mathscr{F}$ is

$$\begin{aligned} \mathscr{F} = \frac{1}{(a+b)} [b(1-a)|F_1|^2 + a(1-b)|F_2|^2 \\ + 2abRl\{F_1F_2^*\}\,. \end{aligned} \tag{42}$$

The values of Z_N, I_N, and σ_N^2 are

$$Z_N = Z \exp\,[i(N-1)\varphi/2] \frac{\sin\,(N\varphi/2)}{\sin\,(\varphi/2)}\,, \tag{43}$$

where Z is given by equation (6),

$$I_N = N[I - \mathscr{F}] + \mathscr{F} \frac{\sin^2\,(N\varphi/2)}{\sin^2\,(\varphi/2)}\,, \tag{44}$$

where I is given by equation (9), and

$$\sigma_N^2 = N[I - \mathscr{F}] + [\mathscr{F} - |Z|^2] \frac{\sin^2\,(N\varphi/2)}{\sin^2\,(\varphi/2)}\,. \tag{45}$$

Hendricks & Teller (1942) considered a special case of this. They took F_1 and F_2 to be real and assumed that $w_1 = w_2 = \frac{1}{2}$, so that they had only one disposable constant which represented the amount of correlation. Equation (44) reduces to their result under these conditions, provided N is made infinite to correspond with the lattice which they considered.

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