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Kinematical Diffraction from Solid Solutions with Short Range Order and Size Effect

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A new approach to the kinematical theory has been developed for the case of binary alloy solid solutions with short-range ordering of the atoms and displacements of the atoms from the average lattice sites due to departures of the effective atomic radii from the average for the alloy. Both the pseudo-temperature factor on the Bragg reflexions and the diffuse scattering intensity are shown to depend on summations over higher-order correlation parameters, defined in terms of the probabilities that groups of three, four or more sites should be occupied in specific ways. Expressions involving these parameters, and the usual short-range order parameters, α_i, are derived with terms of up to the second order in the displacement parameters for the Huang scattering around the fundamental Bragg reflexions, for the short-range order diffuse peaks, and for additional diffuse scattering depending on higher-order correlation parameters only. Special cases of practical significance are explored, and some estimates are made of the relative magnitudes of the terms not included in previous treatments of this problem.

Introduction

The usual treatment of the diffraction of X-rays, or of electrons or neutrons, by binary alloy solid solutions involves the kinematical or single-scattering approximation. The complications resulting from dynamical interactions of diffracted beams are treated elsewhere (Fisher, 1965; Cowley 1966; Cowley & Murray, 1968). The initial kinematical treatments (see *e.g.* Cowley, 1950) involved the assumption that the atoms were placed on the lattice points of a periodic space lattice. The fundamental reflexions that are given by both the fully ordered and completely disordered states were then found to be independent of the state of order. The intensities of the diffuse scattering due to short-range order (s.r.o.), or of the superlattice peaks given by alloys with partial long-range order, could be expressed in terms of Fourier series with the Warren s.r.o. parameters, α_i, (Cowley, 1950, 1965) as coefficients.

The modulation of the diffuse scattering arising from the static displacements of atoms which result from the differences in atomic sizes was first observed by Roberts (1954). Warren, Averbach & Roberts (1951) modified the diffraction theory by the inclusion of size-effect coefficients, β_i, which were dependent on the α_i and contributed an anti-symmetrical part to the s.r.o. diffuse peaks.

Later Borie (1957, 1959) gave a more complete account of the size effect, based on a treatment due to Huang (1947) of scattering from atomic displacements associated with point defects. Borie predicted four effects; the reduction of the intensity of fundamental reflexions by the equivalent of a temperature factor, the broadening of outer s.r.o. diffuse peaks, size-effect modulation and displacement of the s.r.o. diffuse peaks, and a Huang diffuse scattering around the fundamental peaks. Related results have been reported by Krivoglaz (1958), Krivoglaz & Tikhonova (1960), and by Smirnov & Tikhonova (1960).

While Borie showed that his predictions were in fair agreement with experimental observations, his treatment was based on some rather severe approximations. In dealing with the Huang scattering for example, he dealt only with the limiting case of a small degree of s.r.o. While this proved sufficient for a first comparison with experiment, and established the general form of the diffuse scattering, it is probably an insufficient basis for the deriving of the more accurate values for s.r.o. and size-effect coefficients which may be required, for example, for comparisons with the theoretical results of Clapp & Moss (1966, 1968).

In discussing the limitations of his treatment, Borie (1957) points out that a more complete account of the scattering would involve correlation coefficients of higher order than the usual s.r.o. parameters which describe two-particle correlations only. Such higher-order coefficients would describe, for example, the probability of finding one type of atom at a site, **j**, given the occupancy of two sites, **m** and **n**. Coefficients of this sort have been discussed to some extent in relation to dynamical scattering effects in electron diffraction (Cowley, 1966; Cowley & Murray, 1968) but formal considerations of such coefficients have been initiated by Clapp (1967) and, on this basis, we attempt now a more complete formulation of the problem of diffraction from an alloy with short-range order and size effect.

A further complication to the diffraction problem in practice, which has not been included in the present treatment, is that the thermal vibrations of the atoms modifies the intensities of both the fundamental reflexions and the s.r.o. diffuse scattering (Walker & Keating, 1961) and also adds a background of thermal diffuse scattering. To the degree of approximation used by Walker & Keating, the modification of intensities may be included by combining a Debye–Waller factor with each atomic scattering factor.

Higher-order correlations and order parameters

For a binary, AB, alloy, with fractions m_{A} and m_{B} of A and B atoms, the Warren short range order parameters α_i relating to atomic sites **j** and **k** separated by a vector $\mathbf{r}_i = \mathbf{r}_k - \mathbf{r}_j$ may be defined in terms of the conditional probability $P(^{\mathrm{A}}_{\mathrm{j}}|^{\mathrm{A}}_{\mathrm{k}})$ that, given an A atom at **j**, there will be an A atom at **k**;

$$P(^{\mathrm{A}}_{\mathrm{j}}|^{\mathrm{A}}_{\mathrm{k}}) = m_{\mathrm{A}} + m_{\mathrm{B}}\alpha_i = m_{\mathrm{A}} + m_{\mathrm{B}}\alpha_{\mathrm{jk}} ,$$

or, in terms of the *a priori* probability of A atoms being on sites **j** and **k**,

$$P^{\mathrm{AA}}_{\mathrm{jk}} = m_{\mathrm{A}}^2 + m_{\mathrm{A}} m_{\mathrm{B}} \alpha_{\mathrm{jk}} = \langle \sigma^{\mathrm{A}}_{\mathrm{j}} \sigma^{\mathrm{A}}_{\mathrm{k}} \rangle , \tag{1}$$

where the brackets $\langle \ldots \rangle$ represent the ensemble average (Clapp & Moss, 1966) over the products of the quantities $\sigma^{\mathrm{A}}_{\mathrm{j}}, \sigma^{\mathrm{B}}_{\mathrm{j}}$ defined so that $\sigma^{\mathrm{A}}_{\mathrm{j}} = +1, \sigma^{\mathrm{B}}_{\mathrm{j}} = 0$ if there is an A atom at **j**, and $\sigma^{\mathrm{A}}_{\mathrm{j}} = 0, \sigma^{\mathrm{B}}_{\mathrm{j}} = +1$ if there is a B atom at **j**.

The correlation may also be expressed in terms of the Flinn parameters (Flinn, 1956), σ_{j}, defined by $\sigma_{\mathrm{j}} = m_{\mathrm{B}}$ for an A atom, and $\sigma_{\mathrm{j}} = -m_{\mathrm{A}}$ for a B atom at **j**.

Then

$$\sigma^{\mathrm{A}}_{\mathrm{j}} = \sigma_{\mathrm{j}} + m_{\mathrm{A}} ,$$

$$\sigma^{\mathrm{B}}_{\mathrm{j}} = m_{\mathrm{B}} - \sigma_{\mathrm{j}} ,$$

or

$$\sigma_{\mathrm{j}} = \sigma^{\mathrm{A}}_{\mathrm{j}} - m_{\mathrm{A}} = m_{\mathrm{B}} - \sigma^{\mathrm{B}}_{\mathrm{j}} . \tag{2}$$

Hence

$$\langle \sigma^{\mathrm{A}}_{\mathrm{j}} \sigma^{\mathrm{A}}_{\mathrm{k}} \rangle = \langle (\sigma_{\mathrm{j}} + m_{\mathrm{A}})(\sigma_{\mathrm{k}} + m_{\mathrm{A}}) \rangle = m_{\mathrm{A}}^2 + \langle \sigma_{\mathrm{j}} \sigma_{\mathrm{k}} \rangle , \tag{3}$$

and

$$\langle \sigma_{\mathrm{j}} \sigma_{\mathrm{k}} \rangle = m_{\mathrm{A}} m_{\mathrm{B}} \alpha_{\mathrm{jk}} , \tag{4}$$

since

$$\langle \sigma_{\mathrm{j}} \rangle = \langle \sigma_{\mathrm{k}} \rangle = 0 .$$

Similarly we may write, for the other pair correlations

$$\begin{aligned} P^{\mathrm{AB}}_{\mathrm{jk}} = P^{\mathrm{BA}}_{\mathrm{jk}} &= m_{\mathrm{A}} m_{\mathrm{B}} - m_{\mathrm{A}} m_{\mathrm{B}} \alpha_{\mathrm{jk}} = m_{\mathrm{A}} m_{\mathrm{B}} - \langle \sigma_{\mathrm{j}} \sigma_{\mathrm{k}} \rangle , \\ P^{\mathrm{BB}}_{\mathrm{jk}} &= m_{\mathrm{B}}^2 + m_{\mathrm{A}} m_{\mathrm{B}} \alpha_{\mathrm{jk}} = m_{\mathrm{B}}^2 + \langle \sigma_{\mathrm{j}} \sigma_{\mathrm{k}} \rangle . \end{aligned} \tag{5}$$

Definition of the higher-order correlation coefficients involved in the probabilities of occupation of three or more sites by specified types of atom then follows by a straightforward extension of these equations.

For example,

$$\begin{aligned} P^{\mathrm{AAA}}_{123} &= \langle \sigma^{\mathrm{A}}_1 \sigma^{\mathrm{A}}_2 \sigma^{\mathrm{A}}_3 \rangle = \langle (\sigma_1 + m_{\mathrm{A}})(\sigma_2 + m_{\mathrm{A}})(\sigma_3 + m_{\mathrm{A}}) \rangle \\ &= m_{\mathrm{A}}^3 + m_{\mathrm{A}} \{ \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_2 \sigma_3 \rangle + \langle \sigma_3 \sigma_1 \rangle \} \\ &\quad + \langle \sigma_1 \sigma_2 \sigma_3 \rangle . \end{aligned}$$

Such expressions may be compared with those given by Clapp (1967); they are identical with those given by him for the special case of 50:50 composition, with $m_{\mathrm{A}} = m_{\mathrm{B}} = \frac{1}{2}$, when all terms of odd power in σ_{j} vanish.

We may use the averages $\langle \sigma_1 \sigma_2 \sigma_3 \rangle$ as our correlation coefficients, or else, as in the case of pair correlations, introduce parameters such that the diffraction expressions are simplified: thus

$$\begin{gathered} \langle \sigma_1 \sigma_2 \rangle = m_{\mathrm{A}} m_{\mathrm{B}} \alpha_{12} , \\ \langle \sigma_1 \sigma_2 \sigma_3 \rangle = m_{\mathrm{A}} m_{\mathrm{B}} \tau_{123} , \\ \langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle = m_{\mathrm{A}}^2 m_{\mathrm{B}}^2 \varphi_{1234} \\ etc. \end{gathered} \tag{6}$$

Then

$$\begin{aligned} P^{\mathrm{AAA}}_{123} &= m_{\mathrm{A}}^3 + m_{\mathrm{A}}^2 m_{\mathrm{B}} (\alpha_{12} + \alpha_{23} + \alpha_{31}) + m_{\mathrm{A}} m_{\mathrm{B}} \tau_{123} , \\ P^{\mathrm{BBB}}_{123} &= m_{\mathrm{B}}^3 + m_{\mathrm{A}} m_{\mathrm{B}}^2 (\alpha_{12} + \alpha_{23} + \alpha_{31}) - m_{\mathrm{A}} m_{\mathrm{B}} \tau_{123} , \\ P^{\mathrm{AAB}}_{123} &= m_{\mathrm{A}}^2 m_{\mathrm{B}} + m_{\mathrm{A}} m_{\mathrm{B}}^2 (\alpha_{12}) - m_{\mathrm{A}}^2 m_{\mathrm{B}} (\alpha_{23} + \alpha_{31}) \\ &\quad - m_{\mathrm{A}} m_{\mathrm{B}} \tau_{123} , \\ &\quad etc. \end{aligned} \tag{7}$$

Similarly

$$\begin{aligned} P^{\mathrm{AAAA}}_{1234} &= m_{\mathrm{A}}^4 + m_{\mathrm{A}}^3 m_{\mathrm{B}} (\alpha_{12} + \alpha_{23} + \alpha_{34} + \alpha_{41} + \alpha_{13} + \alpha_{24}) \\ &\quad + m_{\mathrm{A}}^2 m_{\mathrm{B}} (\tau_{123} + \tau_{234} + \tau_{341} + \tau_{124}) + m_{\mathrm{A}}^2 m_{\mathrm{B}}^2 \varphi_{1234} , \\ P^{\mathrm{AAAB}}_{1234} &= m_{\mathrm{A}}^3 m_{\mathrm{B}} - m_{\mathrm{A}}^3 m_{\mathrm{B}} (\alpha_{14} + \alpha_{24} + \alpha_{34}) + m_{\mathrm{A}}^2 m_{\mathrm{B}}^2 (\alpha_{12} \\ &\quad + \alpha_{23} + \alpha_{31}) + m_{\mathrm{A}} m_{\mathrm{B}}^2 \tau_{123} - m_{\mathrm{A}}^2 m_{\mathrm{B}} (\tau_{124} + \tau_{234} \\ &\quad + \tau_{134}) - m_{\mathrm{A}}^2 m_{\mathrm{B}}^2 \varphi_{1234} , \\ &\quad etc. \end{aligned} \tag{8}$$

The higher-order correlation parameters introduced in this way have the desirable properties that they are zero for complete disorder, or when one (or more) of the sites considered is removed to infinity so that the occupancy of this site is unrelated to the occupancy of the remainder. For perfect order, the parameters tend to maximum values, either positive or negative. For example, for sites which are points of the same sublattice so that they will always be occupied by the same type of atom in the case of perfect order,

$$\langle \sigma_1 \sigma_2 \sigma_3 \rangle = m_{\mathrm{A}} (m_{\mathrm{B}})^3 + m_{\mathrm{B}} (-m_{\mathrm{A}})^3$$

and

$$\tau_{123}=m_{\rm B}-m_{\rm A}\,.$$

Similarly

$$\langle\sigma_1\sigma_2\sigma_3\sigma_4\rangle=m_{\rm A}m_{\rm B}^4+m_{\rm B}m_{\rm A}^4\,,$$

so that

$$\varphi_{1234}=\left(1+\frac{m_{\rm A}}{m_{\rm B}}+\frac{m_{\rm B}}{m_{\rm A}}\right)=\frac{1}{m_{\rm B}}+\frac{1}{m_{\rm A}}-1\,.$$

For the special cases that two or more of the sites considered coincide, these parameters reduce to combinations of lower-order parameters. For example,

$$\begin{aligned}\tau_{112}&=(m_{\rm B}-m_{\rm A})\alpha_{12}\,,\\ \tau_{111}&=(m_{\rm B}-m_{\rm A})\,,\\ \varphi_{1123}&=\alpha_{23}+\left(\frac{m_{\rm B}-m_{\rm A}}{m_{\rm A}m_{\rm B}}\right)\tau_{123}\,,\\ \varphi_{1112}&=\alpha_{12}\left(\frac{m_{\rm A}}{m_{\rm B}}+\frac{m_{\rm B}}{m_{\rm A}}-1\right).\end{aligned}\tag{9}$$

Some information on higher-order correlation probabilities has been derived by Gehlen & Cohen (1965), for the particular case of Cu_3Au above the critical temperature, from computer experiments. These authors generated a model of a sample of an alloy, containing either 4000 or 16,000 atoms, for given values of the first one, two or three short-range order parameters, by interchanging atoms progressively, starting from either a fully ordered or a random configuration. Irrespective of the starting point, they obtained consistent results for the probabilities that triplets or quadruplets of nearest neighbour atoms would have specified compositions, *i.e.* for the quantities $P_{123}^{\rm AAA}, P_{123}^{\rm AAB}$, *etc.* where the sites 1, 2 and 3 form an equilateral triangle of nearest neighbour sites, and $P_{1234}^{\rm AAAA}$ *etc.* If the values given for the triplet probabilities are inserted in the equations (7) we obtain the consistent result that, for $\alpha_1=-0{\cdot}195$, we obtain $\tau_{123}=-0{\cdot}072$. From the given nearest-neighbour probabilities it was also found possible to derive a consistent result for φ_{1234} from the equations (8).

These results of Gehlen & Cohen (1965) suggest that some at least, of the higher order correlation parameters may not be independent parameters, but may be completely determined by the s.r.o. parameters. The extent and form of this inter-dependence needs to be investigated.

Formulation of the diffraction problem

We assume that the displacements of atoms from their mean lattice positions are linear sums of displacements due to all other neighboring atoms. Thus, if the displacement of an A atom on site **j** due to an A atom on site **k** is written $\boldsymbol{\varepsilon}_{\bf kj}^{\rm AA}$, then the total displacement of the A atom at **j** is given by

$$(\boldsymbol{\varepsilon}_{\bf j})_{\rm A}=\Sigma'_{\bf k}\,(\sigma_{\bf k}^{\rm A}\boldsymbol{\varepsilon}_{\bf kj}^{\rm AA}+\sigma_{\bf k}^{\rm B}\,\boldsymbol{\varepsilon}_{\bf kj}^{\rm BA})\,.\tag{10}$$

The requirement that the average displacement for all A atoms from their mean lattice sites should be zero gives (*cf.* Borie, 1957)

$$m_{\rm A}\boldsymbol{\varepsilon}_{\bf kj}^{\rm AA}+m_{\rm B}\boldsymbol{\varepsilon}_{\bf kj}^{\rm BA}=0\,.\tag{11}$$

It is convenient to assume that $\boldsymbol{\varepsilon}_{\bf kj}^{\rm AA}=\boldsymbol{\varepsilon}_{\bf kj}^{\rm AB}\equiv\boldsymbol{\varepsilon}_{\bf kj}^{\rm A}$, *i.e.* that the displacement of a neighboring atom due to the excess or defect in size of a given atom is independent of its nature. If this assumption is not made, the calculation of diffraction intensities can be carried out in exactly the same way, but the resulting expressions are more complicated. It was considered that such additional complication is unwarranted at this stage.

It may be shown readily (see *e.g.* Guinier, 1963) that the total diffracted intensity in the kinematical approximation may be expressed as the sum of two parts: the sharp, Bragg reflexions given by the Fourier transform of the Patterson function for the average, periodic structure and the diffuse scattering, given by the Fourier transform of the Patterson function for the deviations from the average lattice (Cowley, 1965).

$$I_{\rm Bragg}\equiv I_{\rm B}=\Sigma_j\,\Sigma_k\,\bar{F}\bar{F}^*\exp\{2\pi i\mathbf{u}\,.\,(\mathbf{r}_j-\mathbf{r}_k)\}\tag{12}$$

$$=N\bar{F}\bar{F}^*\,\Sigma_l\exp\{2\pi i\mathbf{u}\,.\,\mathbf{r}_l\}\,,\tag{12a}$$

$$I_{\rm Diff}=\Sigma_{\bf j}\,\Sigma_{\bf k}\,\{\bar{F}-F_{\bf j}(\mathbf{u})\}\,\{\bar{F}^*-F_{\bf k}^*(\mathbf{u})\}\times\exp\{2\pi i\mathbf{u}\,.\,(\mathbf{r}_j-\mathbf{r}_k)\}\,,\tag{13}$$

where the usual constant terms, Lorentz and polarization factors have been omitted, **u** is a vector in reciprocal space, $\mathbf{r}_j$ and $\mathbf{r}_k$ are vectors from an arbitrary origin to the atoms at sites **j** and **k**, and $\mathbf{r}_l$ in (12*a*) is a space-lattice vector given by $\mathbf{r}_l=\mathbf{r}_j-\mathbf{r}_k$ for the averaged atoms at **j** and **k**.

Considering first the fundamental Bragg peaks, the average structure factor occurring in (12) may be written $\bar{F}=\bar{F}_{\rm A}+\bar{F}_{\rm B}$ where

$$\begin{aligned}\bar{F}_{\rm A}&=\langle\sigma_{\bf i}^{\rm A}F_{\rm A}(\mathbf{u})\exp\{2\pi i(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf i})\}\rangle\\ &=F_{\rm A}(\mathbf{u})[\langle\sigma_{\bf i}^{\rm A}\rangle-2\pi^2\,\Sigma_{\bf k}\,\Sigma_{\bf l}\,\{\langle\sigma_{\bf i}^{\rm A}\sigma_{\bf k}^{\rm A}\sigma_{\bf l}^{\rm A}\rangle\\ &\quad\times(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf ki}^{\rm A})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf li}^{\rm A})+\langle\sigma_{\bf i}^{\rm A}\sigma_{\bf k}^{\rm A}\sigma_{\bf l}^{\rm A}\rangle\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf ki}^{\rm A})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf li}^{\rm B})\\ &\quad+\langle\sigma_{\bf i}^{\rm A}\sigma_{\bf k}^{\rm B}\,\sigma_{\bf l}^{\rm A}\rangle\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf ki}^{\rm B})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf li}^{\rm A})\\ &\quad+\langle\sigma_{\bf i}^{\rm A}\sigma_{\bf k}^{\rm B}\,\sigma_{\bf l}^{\rm B}\rangle(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf ki}^{\rm B})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf li}^{\rm B})\}\\ &\quad+\frac{(2\pi)^4}{4!}\,\Sigma_{\bf k}\Sigma_{\bf l}\Sigma_{\bf m}\Sigma_{\bf n}\{\langle\sigma_{\bf i}^{\rm A}\sigma_{\bf k}^{\rm A}\sigma_{\bf l}^{\rm A}\sigma_{\bf m}^{\rm A}\sigma_{\bf n}^{\rm A}\rangle\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf ki}^{\rm A}\\ &\quad\times(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf li}^{\rm A})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf mi}^{\rm A})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf ni}^{\rm A})+\ldots\}+\ldots]\,.\end{aligned}$$

Terms of odd order in $(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\bf ki}^{\rm A})$ in this expansion are put equal to zero on the assumption that, as will be the case for the substances of simple structure usually considered, the environment of any site is statistically centrosymmetric.

Then, using the relationship (10) and the equations (7), we may write, up to the second order terms in $\boldsymbol{\varepsilon}_{\bf k}^{\rm A}$, with $\mathbf{i}=0$,

$$\begin{aligned}\bar{F}_{\mathrm{A}}=F_{\mathrm{A}}\Big[&m_{\mathrm{A}}-2\pi^2\,\Sigma_{\mathrm{k}}\Sigma_{\mathrm{l}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{l0}})\\&\times\left\{P^{\mathrm{AAA}}_{\mathrm{0kl}}-\frac{m_{\mathrm{A}}}{m_{\mathrm{B}}}(P^{\mathrm{AAB}}_{\mathrm{0kl}}+P^{\mathrm{ABA}}_{\mathrm{0kl}})+\frac{m^2_{\mathrm{A}}}{m^2_{\mathrm{B}}}P^{\mathrm{ABB}}_{\mathrm{0kl}}\right\}\\&+\ldots\Big]=m_{\mathrm{A}}F_{\mathrm{A}}[1-2\pi^2\,\Sigma_{\mathrm{k}}\Sigma_{\mathrm{l}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{l0}})\{\mu\alpha_{\mathrm{kl}}\\&\qquad+(1+\mu)\tau_{\mathrm{0kl}}\}+\ldots]\,,\end{aligned}\tag{14}$$

where $\mu=m_{\mathrm{A}}/m_{\mathrm{B}}$. Similarly

$$\begin{aligned}\bar{F}_{\mathrm{B}}=m_{\mathrm{B}}F_{\mathrm{B}}[1-2\pi^2\,\Sigma_{\mathrm{k}}\,\Sigma_{\mathrm{l}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{l0}})\,\{\mu\alpha_{\mathrm{kl}}\\-\mu(1+\mu)\tau_{\mathrm{0kl}}\}+\ldots]\,.\end{aligned}\tag{15}$$

Thus we may write

$$\bar{F}=m_{\mathrm{A}}F_{\mathrm{A}}E_{\mathrm{A}}+m_{\mathrm{B}}F_{\mathrm{B}}E_{\mathrm{B}}\,,$$

where E_{A} and E_{B} are the bracketed terms in (14) and (15) and may be considered as pseudo-temperature factors for A and B atoms respectively. Alternatively, we may combine (14) and (15) and, separating out the special cases, $\mathbf{k}=\mathbf{l}$, obtain the general expression for the Bragg peaks

$$\begin{aligned}I_{\mathrm{B}}=N\,\Sigma_i\exp\{2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}&\\\times\{(m_{\mathrm{A}}+F_{\mathrm{A}}+m_{\mathrm{B}}F_{\mathrm{B}})^2&[1-4\pi^2\mu\,\Sigma_k\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})^2\\-4\pi^2\mu\sum_{\mathrm{k}\neq\mathrm{l}}\sum&\alpha_{\mathrm{kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{l0}})+\ldots]\\-m_{\mathrm{A}}m_{\mathrm{B}}\Delta F(\mu F_{\mathrm{A}}+F_{\mathrm{B}})&\,[(1-\mu)\,\Sigma_{\mathrm{k}}\,\alpha_{\mathrm{0k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})^2\\+(1+\mu)\sum_{\mathrm{k}\neq\mathrm{l}}\sum&\tau_{\mathrm{0kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{l0}})+\ldots]\,.\end{aligned}\tag{16}$$

The first part of this expression applies a pseudo-temperature factor to the Bragg peak intensities calculated for no size effect, as envisaged by Borie. The second part adds a term proportional to $\Delta F=F_{\mathrm{A}}-F_{\mathrm{B}}$. It may noted that even the second-order term in this expansion depends on the third-order correlation parameters, τ_{0kl}. Further terms in the series will necessarily involve higher-order correlation parameters, although, in each case, the dependence will be only on summations of these parameters. It may be anticipated that these summations will give rise to appreciable contributions to the pseudo-temperature factors under particular circumstances. The contributions of τ_{0kl} terms will be zero for perfect order or complete disorder and also vanish for $m_{\mathrm{A}}=m_{\mathrm{B}}$ (Clapp, 1967). They will presumably be greatest for compositions well away from $m_{\mathrm{A}}=m_{\mathrm{B}}$ and for temperatures near the critical temperature.

For the calculation of diffuse scattering according to (13), it is convenient to define,

$$\begin{aligned}\bar{F}&=\bar{F}_{\mathrm{A}}+\bar{F}_{\mathrm{B}}\,,\\F^0_{\mathrm{i}}&=(\sigma^{\mathrm{A}}_{\mathrm{i}}F_{\mathrm{A}}+\sigma^{\mathrm{B}}_{\mathrm{i}}F_{\mathrm{B}})\,,\\F_i&=\sigma^{\mathrm{A}}_{\mathrm{i}}F_{\mathrm{A}}\exp\{2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_i\}+\sigma^{\mathrm{B}}_{\mathrm{i}}F_{\mathrm{B}}\exp\{2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}}\}\,.\end{aligned}$$

Then

$$\begin{aligned}I_{\mathrm{diff}}=\Sigma_{\mathrm{j}}\,\Sigma_{\mathrm{k}}\,[&(\bar{F}_{\mathrm{j}}-F^0_{\mathrm{j}})\,(\bar{F}_{\mathrm{k}}-F^0_{\mathrm{k}})+(\bar{F}_{\mathrm{j}}-F^0_{\mathrm{j}})\,(F^0_{\mathrm{k}}-F^*_{\mathrm{k}})\\&+(\bar{F}_{\mathrm{k}}-F^0_{\mathrm{k}})\,(F^0_{\mathrm{j}}-F_{\mathrm{j}})+(F^0_{\mathrm{j}}-F_{\mathrm{j}})\,(F^0_{\mathrm{k}}-F^*_{\mathrm{k}})]\\&\times\exp\{2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\,,\end{aligned}\tag{17}$$

and we may deal with the four terms involved separately. The first term of (17) by simple manipulation, taking $\mathbf{j}=\mathbf{0}$, $\mathbf{r}_j-\mathbf{r}_k=\mathbf{r}_i$ and averaging over all choices of origin $\mathbf{r}_0$, becomes

$$\begin{aligned}N\,\Sigma_{\mathrm{i}}\exp\{2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\,[\{m_{\mathrm{A}}F_{\mathrm{A}}(E_{\mathrm{A}}-1)\\+m_{\mathrm{B}}F_{\mathrm{B}}(E_{\mathrm{B}}-1)\}^2+m_{\mathrm{A}}m_{\mathrm{B}}\Delta F^2\alpha_{\mathrm{0i}}]\,.\end{aligned}\tag{18}$$

The fourth term of (17) may be written

$$\begin{aligned}N\,\Sigma_{\mathrm{i}}\,\langle\{\sigma^{\mathrm{A}}_0\sigma^{\mathrm{A}}_{\mathrm{i}}F^2_{\mathrm{A}}+(\sigma^{\mathrm{A}}_0\sigma^{\mathrm{B}}_{\mathrm{i}}+\sigma^{\mathrm{B}}_0\sigma^{\mathrm{A}}_{\mathrm{i}})F_{\mathrm{A}}F_{\mathrm{B}}+\sigma^{\mathrm{B}}_0\sigma^{\mathrm{B}}_{\mathrm{i}}F^2_{\mathrm{B}}\}\\\times[\exp\{2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_0\}-1]\,[\exp\{2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}}\}-1]\rangle\,.\end{aligned}$$

If we include only second order terms in $(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}})$, the product of the final exponential terms is approximately $4\pi^2(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_0)\,.\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}})$ where from (10), $\boldsymbol{\varepsilon}_{\mathrm{i}}=\Sigma_{\mathrm{k}}\,(\sigma^{\mathrm{A}}_{\mathrm{k}}\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{ki}}+\sigma^{\mathrm{B}}_{\mathrm{k}}\boldsymbol{\varepsilon}^{\mathrm{B}}_{\mathrm{ki}})$.

Then this term becomes

$$\begin{aligned}4\pi^2N\,\Sigma_{\mathrm{i}}\,\Sigma_{\mathrm{k}}\,\Sigma_{\mathrm{l}}\,[&\langle\sigma^{\mathrm{A}}_0\sigma^{\mathrm{A}}_{\mathrm{i}}\sigma^{\mathrm{A}}_{\mathrm{k}}\sigma^{\mathrm{A}}_{\mathrm{l}}\rangle F^2_{\mathrm{A}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{li}})\\&+\langle\sigma^{\mathrm{A}}_0\sigma^{\mathrm{B}}_{\mathrm{i}}\sigma^{\mathrm{A}}_{\mathrm{k}}\sigma^{\mathrm{A}}_{\mathrm{l}}\rangle F_{\mathrm{A}}F_{\mathrm{B}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{li}})\\&+\ldots\ldots]\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\,.\end{aligned}$$

Using the relationship (11), this may be written

$$\begin{aligned}4\pi^2N\,\Sigma_{\mathrm{i}}\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\,&\Sigma_{\mathrm{k}}\,\Sigma_{\mathrm{l}}\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{li}})\\\times[F^2_{\mathrm{A}}\{&P^{\mathrm{AAAA}}_{\mathrm{0ikl}}-\mu(P^{\mathrm{AAAB}}_{\mathrm{0ikl}}+P^{\mathrm{AABA}}_{\mathrm{0ikl}})+\mu^2P^{\mathrm{AABB}}_{\mathrm{0ikl}}\}\\+F_{\mathrm{A}}F_{\mathrm{B}}\{&(P^{\mathrm{ABAA}}_{\mathrm{0ikl}}+P^{\mathrm{BAAA}}_{\mathrm{0ikl}})-\mu(P^{\mathrm{ABAB}}_{\mathrm{0ikl}}+P^{\mathrm{BAAB}}_{\mathrm{0ikl}}\\+P^{\mathrm{ABBA}}_{\mathrm{0ikl}}&+P^{\mathrm{BABA}}_{\mathrm{0ikl}})+\mu^2(P^{\mathrm{ABBB}}_{\mathrm{0ikl}}+P^{\mathrm{BABB}}_{\mathrm{0ikl}})\}\\+F^2_{\mathrm{B}}\{&P^{\mathrm{BBAA}}_{\mathrm{0ikl}}-\mu(P^{\mathrm{BBAB}}_{\mathrm{0ikl}}+P^{\mathrm{BBBA}}_{\mathrm{0ikl}})+\mu^2P^{\mathrm{BBBB}}_{\mathrm{0ikl}}\}]\,.\end{aligned}\tag{19}$$

Then using (8), this reduces to

$$\begin{aligned}4\pi^2N\,\Sigma_{\mathrm{i}}&\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\\&\times m_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_{\mathrm{k}}\Sigma_{\mathrm{l}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{li}})\,[(\mu F_{\mathrm{A}}+F_{\mathrm{B}})^2\alpha_{\mathrm{kl}}\\&+(1+\mu)\,(\mu F_{\mathrm{A}}+F_{\mathrm{B}})\Delta F(\tau_{\mathrm{0kl}}+\tau_{\mathrm{ikl}})+\mu\Delta F^2\varphi_{\mathrm{0ikl}}]\,.\end{aligned}\tag{20}$$

The second and third terms of (17) are similar and may be treated together. The second may be written

$$\begin{aligned}N\,\Sigma_{\mathrm{i}}&\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\\&\times[\bar{F}\{F_{\mathrm{A}}\langle\sigma^{\mathrm{A}}_{\mathrm{i}}(1-\exp\{-2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}}\})\rangle\\&+F_{\mathrm{B}}\langle\sigma^{\mathrm{B}}_{\mathrm{i}}(1-\exp\{-2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}}\})\rangle\}\\&-\{\langle\sigma^{\mathrm{A}}_0\sigma^{\mathrm{A}}_{\mathrm{i}}(1-\exp\{-2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}}\})\rangle F^2_{\mathrm{A}}\\&+\langle(\sigma^{\mathrm{A}}_0\sigma^{\mathrm{B}}_{\mathrm{i}}+\sigma^{\mathrm{B}}_0\sigma^{\mathrm{A}}_{\mathrm{j}})\,(1-\exp\{-2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{j}}\})\rangle F_{\mathrm{A}}F_{\mathrm{B}}\\&+\langle\sigma^{\mathrm{B}}_0\sigma^{\mathrm{B}}_{\mathrm{i}}(1-\exp\{-2\pi i\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathrm{i}}\})\rangle F^2_{\mathrm{B}}\}]\\&=N\,\Sigma_i\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}[\bar{F}\{m_{\mathrm{A}}F_{\mathrm{A}}(1-E_{\mathrm{A}})\\&+m_{\mathrm{B}}F_{\mathrm{B}}(1-E_{\mathrm{B}})\}-F^2_{\mathrm{A}}\{2\pi i\,\Sigma_{\mathrm{k}}\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{yj}})\,(P^{\mathrm{AAA}}_{\mathrm{0ik}}\\&-\mu P^{\mathrm{AAB}}_{\mathrm{0ik}})+2\pi^2\,\Sigma_{\mathrm{k}}\,\Sigma_{\mathrm{l}}\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{ki}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{li}})\,[P^{\mathrm{AAAA}}_{\mathrm{0ikl}}\\&-\mu(P^{\mathrm{AAAB}}_{\mathrm{0ikl}}+P^{\mathrm{AABA}}_{\mathrm{0ikl}})+\mu^2P^{\mathrm{AABB}}_{\mathrm{0ikl}}]\}\\&-F_{\mathrm{A}}F_{\mathrm{B}}\{2\pi i\,\Sigma_{\mathrm{k}}\,\ldots\ldots\}-F^2_{\mathrm{B}}\{\ldots\}]\,.\end{aligned}$$

Combining this with the third term of (17) we obtain

$$\begin{aligned}N\,\Sigma_{\mathrm{i}}\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\,&[2\bar{F}\{m_{\mathrm{A}}F_{\mathrm{A}}(1-E_{\mathrm{A}})\\&+m_{\mathrm{B}}F_{\mathrm{B}}(1-E_{\mathrm{B}})\}+2\pi i\,\Sigma_{\mathrm{k}}\,\{\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathrm{kj}})\}\\&\times\{F^2_{\mathrm{A}}(P^{\mathrm{AAA}}_{\mathrm{0ik}}-\mu P^{\mathrm{AAB}}_{\mathrm{0ik}})+F_{\mathrm{A}}F_{\mathrm{B}}(P^{\mathrm{ABA}}_{\mathrm{0ik}}+P^{\mathrm{BAA}}_{\mathrm{0ik}}\end{aligned}$$

$$
\begin{aligned}
&-\mu P^{\mathrm{ABB}}_{\mathbf{0ik}}-\mu P^{\mathrm{BAB}}_{\mathbf{0ik}})+F^2_{\mathrm{B}}(P^{\mathrm{BBA}}_{\mathbf{0ik}}-\mu P^{\mathrm{BBB}}_{\mathbf{0ik}})\}\\
&-2\pi^2\,\Sigma_{\mathbf{k}}\,\Sigma_{\mathbf{l}}\,\{(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{l0}})\\
&+(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\}\,\{\ldots\}]\,, \qquad (21)
\end{aligned}
$$

where the content of the final bracket $\{\ldots\}$ is exactly the same as for the square bracket $[\ldots]$ of (19).

Adding all four terms of (17) we thus obtain

$$
\begin{aligned}
I_{\mathrm{Diff}}&=N\,\Sigma_{\mathbf{i}}\,\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\,[(m_{\mathrm{A}}F_{\mathrm{A}}+m_{\mathrm{B}}F_{\mathrm{B}})^2-\bar{F}^2]\\
&+Nm_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_i\,\exp\{-2\pi i\mathbf{u}\,.\,\mathbf{r}_i\}\\
&\times[\Delta F^2\alpha_{\mathbf{0i}}+2\pi i\,\Sigma_{\mathbf{k}}\,\{(\mu F_{\mathrm{A}}+F_{\mathrm{B}})\Delta F(\alpha_{\mathbf{ik}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\\
&-\alpha_{\mathbf{0k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}}))+(1+\mu)\Delta F^2\tau_{\mathbf{0ik}}[\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})]\}\\
&-2\pi^2\,\Sigma_{\mathbf{k}}\,\Sigma_{\mathbf{l}}\,\{(\mu F_{\mathrm{A}}+F_{\mathrm{B}})^2\alpha_{\mathbf{kl}}+(1+\mu)\,(\mu F_{\mathrm{A}}\\
&+F_{\mathrm{B}})\,.\,\Delta F(\tau_{\mathbf{0kl}}+\tau_{\mathbf{ikl}})+\mu\Delta F^2\varphi_{\mathbf{0ikl}}\}\\
&\times\{\mathbf{u}(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\}\,\{\mathbf{u}(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{l0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\}]\,. \qquad (22)
\end{aligned}
$$

The quantity $(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})$ which appears here represents the change in the interatomic vector from the origin to the atom at $\mathbf{r}_i$ due to an A atom at position $\mathbf{k}$.

This pleasantly compact form (22) of the result is unsatisfactory, however, because the second part contains some terms independent of $\mathbf{i}$ which cancel out with the first part, leaving no contribution to the sharp Bragg peaks, and also because the special terms $\mathbf{k}=\mathbf{0}$, $\mathbf{i}$ and $\mathbf{l}=\mathbf{0}$, $\mathbf{i}$ and $\mathbf{k}=\mathbf{l}$ have particular significance for the interpretation of the expressions in terms of diffuse scattering.

In separating out these special terms, we make use of relations such as (9) and also symmetry relationships such as

$$\Sigma_{\mathbf{k}}\,\alpha_{\mathbf{0k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})=\Sigma_{\mathbf{k}}\,\alpha_{\mathbf{ik}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})=0\,,$$

so that

$$\Sigma'_{\mathbf{k}}\,\alpha_{\mathbf{0k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})=-\Sigma'_{\mathbf{k}}\,\alpha_{\mathbf{ik}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})=-\alpha_{\mathbf{0i}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})\,.$$

Similarly

$$
\begin{aligned}
&\underset{\mathbf{k}\neq\mathbf{l}}{\Sigma'\Sigma'}\,\tau_{\mathbf{0kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\\
&\qquad=\underset{\mathbf{k}\neq\mathbf{l}}{\Sigma'\Sigma'}\,\tau_{\mathbf{ikl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{l0}})\,. \qquad (23)
\end{aligned}
$$

The primed summation sign $\Sigma'_{\mathbf{k}}$ denotes a summation over $\mathbf{k}$ excluding the special values $\mathbf{k}=\mathbf{0}$ and $\mathbf{k}=\mathbf{i}$.

The resulting expression, calculated for terms of up to the second order in $(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ij}})$, is somewhat cumbersome. We divide it into three parts: I_H, the Huang diffuse scattering, concentrated around the reciprocal lattice points; I_{sro}, the s.r.o. diffuse scattering, concentrated around the positions of superlattice reflections, and I_{HO}, the terms containing higher-order correlation parameters for which the distribution of diffuse scattering is not to be associated clearly with that of either of the previous terms. Thus

$$
\begin{aligned}
I_H&=Nm_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_{\mathbf{i}}\,\sin(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[4\pi\Delta F(\mu F_{\mathrm{A}}+F_{\mathrm{B}})\\
&\times\{(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})+\Sigma'_{\mathbf{k}}\,\alpha_{\mathbf{ik}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\}]\\
&+4\pi^2Nm_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_{\mathbf{i}}\,\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[\{(1-\mu)\Delta F(\mu F_{\mathrm{A}}\\
&+F_{\mathrm{B}})+\mu\Delta F^2\}\,\{-(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2+(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})\,\Sigma'_{\mathbf{k}}\,\alpha_{\mathbf{0k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\}\\
&+(\mu F_{\mathrm{A}}+F_{\mathrm{B}})^2\{\underset{\mathbf{k}\neq\mathbf{l}}{\Sigma'\Sigma'}\,\alpha_{\mathbf{kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\\
&+\Sigma'_{\mathbf{k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\}\\
&-\Delta F(\mu F_{\mathrm{A}}+F_{\mathrm{B}})\,\{(1-\mu)\,\Sigma'_{\mathbf{k}}\,\alpha_{\mathbf{0k}}[(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})^2\\
&-2(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})]\\
&+(1+\mu)\,\underset{k\neq l}{\Sigma'\Sigma'}\,\tau_{\mathbf{0kl}}[(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\\
&-2(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})]\}]\,. \qquad (24)
\end{aligned}
$$

Both the antisymmetrical and symmetrical parts of this expression are seen to increase with distance from the origin of reciprocal space, in the same way as thermal diffuse scattering, being zero around the incident beam direction.

$$
\begin{aligned}
I_{\mathrm{sro}}&=4\pi Nm_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_{\mathbf{i}}\,\sin(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\\
&\times[\Delta F(F_{\mathrm{A}}+\mu F_{\mathrm{B}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})\alpha_{\mathbf{0i}}]\\
&+Nm_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_{\mathbf{i}}\,\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[\Delta F^2\alpha_{\mathbf{0i}}\{1-2\pi^2\mu\,\Sigma'_{\mathbf{k}}\\
&\times[\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})]^2-4\pi^2(1-\mu)^2(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2\}\\
&+4\pi^2\mu(1+\mu)\,(F^2_{\mathrm{A}}+F^2_{\mathrm{B}})\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2\alpha_{\mathbf{0i}}]\,. \qquad (25)
\end{aligned}
$$

The first part is an antisymmetrical term added to the s.r.o. diffuse peak and resulting in the well-known displacement of the peak from the position of the superlattice reciprocal lattice position. The second part is the normal s.r.o. diffuse scattering modified by an effective temperature factor which will decrease the height of s.r.o. peaks and broaden them with increasing scattering angle. It may be noted that the effective temperature factor in this case is not the same as that for the Bragg peaks. There is also a weak, broadened peak which is dependent on the sum of the squares of the structure factors instead of on ΔF^2.

Finally,

$$
\begin{aligned}
I_{\mathrm{HO}}&=2\pi Nm_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_{\mathbf{i}}\,\sin(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\\
&\times[\mu(1+\mu)\Delta F^2\,\Sigma'_{\mathbf{k}}\,\tau_{\mathbf{0ik}}\{\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\}]\\
&+2\pi^2Nm_{\mathrm{A}}m_{\mathrm{B}}\,\Sigma_{\mathbf{i}}\,\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[2\{(1+\mu)\,(F_{\mathrm{A}}\\
&+\mu F_{\mathrm{B}})\Delta F+(1-\mu^2)\Delta F^2\}\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})\,\Sigma'_{\mathbf{k}}\,\tau_{\mathbf{0ik}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\\
&-(1-\mu^2)\Delta F^2\,\Sigma'_{\mathbf{k}}\,\tau_{\mathbf{0ik}}\{\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\}^2\\
&-\mu\Delta F^2\,\underset{\mathbf{k}\neq\mathbf{l}}{\Sigma'\Sigma'}\,\varphi_{\mathbf{0ikl}}\{\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\}\\
&\times\{\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{l0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\}]\,. \qquad (26)
\end{aligned}
$$

This term again contains components both symmetrical and anti-symmetrical about the reciprocal lattice points, but the particular importance of these components in practice is yet to be determined. It seems probable that they could be of appreciable magnitude for temperatures near the critical temperature.

Special cases

From the form of the expressions (24), (25), (26) it is clear that considerable simplifications will occur for some special cases. For example, if $\mu=1$, $m_{\mathrm{A}}=m_{\mathrm{B}}=\frac{1}{2}$, all the terms containing third-order parameters $\tau_{\mathbf{ijk}}$ as well as those containing $(1-\mu)$ will vanish.

Another case of special interest is that for which $F_A=F_B, \Delta F=0$. Then all antisymmetrical components vanish but some diffuse scattering remains around the reciprocal lattice points and s.r.o. peaks: thus

$$\begin{aligned}
I_H &= Nm_Am_B4\pi F_A^2(1+\mu)^2\,\Sigma_i\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\\
&\quad\times[\Sigma'_{\mathbf{k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\\
&\quad+\underset{k\neq l}{\Sigma'\Sigma'}\alpha_{\mathbf{kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})]\,,\\
I_{\mathrm{sro}} &= Nm_Am_B8\pi^2\mu(1\\
&\quad+\mu)F_A^2\,\Sigma_i\,\alpha_{\mathbf{0i}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,.
\end{aligned}\tag{27}$$

The higher order terms in I_{HO} are all zero.

To treat the case of diffraction from a relatively small number of substitutional impurity atoms or vacancies in a crystal, we may take the limiting case, $\mu\rightarrow0, m_A\rightarrow0, m_B\simeq1$, noting that from (11), $\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ij}}\gg\boldsymbol{\varepsilon}^{\mathrm{B}}_{\mathbf{ij}}$, so that all the atom displacements are those associated with the impurities.

For vacancies $F_A=0$, and we obtain

$$\begin{aligned}
I_H &= N4\pi m_AF_B^2\,\Sigma_{\mathbf{i}}\sin(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,\Sigma_{\mathbf{k}}\,\alpha_{\mathbf{ik}}(\mathbf{u}.\ \boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\\
&\quad+N4\pi^2m_AF_B^2\,\Sigma_{\mathbf{i}}\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[\Sigma_{\mathbf{k}}\,\alpha_{\mathbf{0k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\\
&\quad+\Sigma'_{\mathbf{k}}\Sigma'_{\mathbf{l}}\,\alpha_{\mathbf{kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\\
&\quad-\Sigma'_{\mathbf{k}}\Sigma'_{\mathbf{l}}\,\tau_{\mathbf{0kl}}\{(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})-2(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{li}})\}]\,,\\
I_{\mathrm{sro}} &= Nm_AF_B^2\,\Sigma_{\mathbf{i}}\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[\alpha_{\mathbf{0i}}-8\pi^2\alpha_{\mathbf{0i}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2]\,,\\
I_{\mathrm{HO}} &= Nm_A2\pi^2F_B^2\,\Sigma_{\mathbf{i}}\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[\Sigma'_{\mathbf{k}}\,\tau_{\mathbf{0ik}}\{2(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})\\
&\quad\times(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})-[\mathbf{u}\,.\,(\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}}-\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})]^2\}]\,.
\end{aligned}\tag{28}$$

Here the primes or $k\neq l$ restrictions have been removed from some summations for the sake of compactness. However in general one would expect the order parameters to tend to zero for this case, so that the $\mathbf{k}=\mathbf{0}$, $\mathbf{i}$ or $\mathbf{k}=\mathbf{l}$ terms may be the most important ones.

Finally, for any composition, in the limiting case of zero order, *i.e.* when $\alpha_{\mathbf{ij}}=0$ except $\alpha_{\mathbf{ii}}=1$ and all non-degenerate higher order parameters are zero, only the Huang scattering remains, apart from a uniform background. Then

$$\begin{aligned}
I_H &= Nm_Am_B4\pi\Delta F(\mu F_A+F_B)\,\Sigma_{\mathbf{i}}\,(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})\sin(2\pi\mathbf{u}\mathbf{r}_i)\\
&\quad+4\pi^2Nm_Am_B\,\Sigma_{\mathbf{i}}\cos(2\pi\mathbf{u}\,.\,\mathbf{r}_i)\,[(\mu F_A+F_B)^2\\
&\quad\times\Sigma'_{\mathbf{k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ki}})\\
&\quad-\{(1-\mu)\Delta F(\mu F_A+F_B)+\mu\Delta F^2\}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2]\,,\\
I_{\mathrm{sro}} &= Nm_Am_B\Delta F^2\,.
\end{aligned}\tag{29}$$

This is to be compared with the expressions for the disordered state obtained by Borie (1957, 1959), Huang (1947) and Cochran & Kartha (1956). In particular, it contains all the terms of equation (5) of Borie (1959) plus some smaller, additional terms, in $(\mathbf{u}\,.\,\boldsymbol{\varepsilon}_{\mathbf{i0}})^2$.

Discussion

The expressions for the diffracted intensities have been obtained here only as far as the second-order terms in the quantities, $(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})$ which are usually considered to be small. However, for large scattering angles, *i.e.* large values of $\mathbf{u}$, this approximation may be insufficient. The extension of the calculations to give higher-order terms is straightforward, although somewhat laborious, particularly if the form equivalent to (22) is expanded to give the special terms in detail, as in (24), (25) and (26).

It is clear that, for the fundamental Bragg reflexions, the next terms to be included will be those of fourth order and these will include summations over fifth-order correlation parameters, to be added to the pseudo-temperature factors of (14), (15) and (16). For the diffuse scattering, the inclusion of third-order terms will involve consideration of five-atom probability functions, leading to expressions containing fifth-order and some further fourth-order correlation parameters.

In the initial work on size effect by Warren, Averbach & Roberts (1951), the modification of the diffracted intensity was described by means of parameters $\beta_{\mathbf{i}}$ which, in the notation of the present paper, may be defined as

$$\beta^{\mathbf{i}}=\Delta F^{-1}\left(\frac{\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}}}{\mathbf{r}_i}\right)[(\mu F_A+F_B)+(F_A+\mu F_B)\alpha_{\mathbf{i0}}]\,.\tag{30}$$

Inclusion of these quantities as coefficients of a Fourier sine summation gives the first part of the sine summation of (24) and the sine summation of (25). Borie's (1957) expression for the pseudo-temperature factor, M', appears to include the single summations, $\Sigma_{\mathbf{k}}$, of (14), (15) and (16) but not the double summations, $\underset{\mathbf{k}\neq\mathbf{l}}{\Sigma\,\Sigma}$, and his expressions for the diffuse scattering are similarly limited.

The relative magnitudes of the various terms in (16) may be found approximately for the particular case of Cu_3Au at 450°C by using the experimental values of the α parameters determined by Moss (1964) and the value of τ_{123} for nearest neighbour triplets derived above from the results of the computer experiments of Gehlen & Cohen (1965). Assuming that $(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})\neq0$ and $\tau_{\mathbf{0kl}}\neq0$ only for nearest-neighbour atoms, the summations of (16) become

$$\Sigma_{\mathbf{k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})^2\simeq4(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2_{\max}\,,$$

$$\underset{\mathbf{k}\neq\mathbf{l}}{\Sigma\,\Sigma}\,\alpha_{\mathbf{kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{l0}})\simeq(-0{\cdot}53)4(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2_{\max}\,,$$

$$\Sigma_{\mathbf{k}}\,\alpha_{\mathbf{0k}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})^2\simeq(-0{\cdot}195)4(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2_{\max}\,,$$

and

$$\underset{\mathbf{k}\neq\mathbf{l}}{\Sigma\,\Sigma}\,\tau_{\mathbf{0kl}}(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{k0}})(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{l0}})\simeq(-0{\cdot}14)4(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})^2_{\max}\,,$$

where $(\mathbf{u}\,.\,\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}})_{\max}$ represents the value of the product for $\mathbf{u}$ parallel to $\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{i0}}$.

Inserting the appropriate values for m_A, m_B, F_A, F_B, we find that the four terms of (16) involving summations are approximately in the ratios of the numbers 1·0, −0·53, −0·14 and +0·20.

Although the significance of these values is severely limited by the approximations involved, they do serve to indicate that all the terms included in (16) may contribute appreciably to the value of the pseudo-temperature factor and none should be ignored.

In the above treatment we have avoided making an explicit assumption concerning the form of $\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ij}}$. It would be quite easy to substitute the specific form assumed by Borie and others, writing $\boldsymbol{\varepsilon}^{\mathrm{A}}_{\mathbf{ij}} = c_i \mathbf{r}_{\mathbf{ij}}/|\mathbf{r}_{\mathbf{ij}}|^3$. Then, following Borie, the summations over $\mathbf{k}$ and $\mathbf{l}$ not involving order parameters may be explicitly evaluated.

The task of sorting out in practice the relative importance of the various terms and the values of the correlation and displacement parameters involved in the expressions for the pseudo-temperature factor of Bragg peaks and the diffuse scattering is a difficult one. However considerable progress may be possible in particular cases if a series of measurements is made for different values of the adjustable parameters. The temperature may be varied. Variation of the composition may include use of the 50:50 composition, for which all odd-order correlation parameters vanish. Finally the scattering factors may be varied independently of all other parameters by choosing different radiations. For copper–gold alloys, for example, the condition $F_{\mathrm{A}} = F_{\mathrm{B}}$ is very nearly achieved in neutron diffraction and could be made exact by use of isotopically enriched elements. For this case it is interesting to note the prediction of equation (27) that a weak, broadened, short-range order peak should still appear. Measurement of such a peak would give values for the displacement parameters if the s.r.o. parameters were known, and *vice versa*.

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Elastic Coefficients in Crystals

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Jahn's method of reduction of a representation has been applied and extended to obtain the fourth order and the fifth order elastic coefficients for the 32 classes of crystals.

It is well known that the character method (Bhagavantam & Suryanarayana, 1949) could be successfully employed for deriving the number of independent constants in respect of the various physical properties for the 32 crystal classes. Following the suggestion of Chelam (1961), one of us (Krishnamurty, 1963) has applied the character method for obtaining the number of independent fourth-order elastic coefficients for the 32 crystallographic point groups.

In this note, it is proposed to apply and extend Jahn's (1949) method of reduction of a representation to derive the number of the fourth-order and the fifth-order elastic coefficients, which are symmetric in all the indices, for the 32 point groups. The results so ob-