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DIFFUSE SCATTERING STUDIES AS A TOOL FOR CHARACTERIZING THE LOCAL ORDER STRUCTURE AND FOR OBTAINING PAIRWISE INTERACTION ENERGIES

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Abstract

Diffuse scattering is a mature method for characterizing the local order structure of alloy systems. An extension of such structural investigations, for alloys at equilibrium, allows one to obtain pairwise interaction energies. Having experimental pairwise interaction energies for the various coordination shells offers one the potential for more realistic Monte Carlo modelling of alloy systems as they relax toward equilibrium. The diffuse scattering method and the recovery of the interaction energies are reviewed and some preliminary results are used to demonstrate the kinetic Ising modelling technique.

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Introduction

The mere presence of a second species of atom requires that scattering from a solid solution produce a component of coherent diffuse scattering throughout reciprocal space, in addition to the fundamental Bragg reflections. This component of diffuse scattering is modulated by the way the atoms are dispersed on the lattice sites, and hence contains a wealth of information. An elegant theory has evolved which allows one to treat this problem quantitatively, within certain approximations, as have related techniques for visualizing and characterizing the real space atomic structure. More recently, it has been shown that pairwise interaction energies can be obtained from diffuse scattering studies, and these energies offer great promise in allowing one to do realistic kinetic Ising modelling to understand how, for example, supersaturated solid solutions decompose.

It is proposed first to present an outline summary of the state-of-art local order diffuse scattering theory. Next, procedures for modelling and characterizing the real space structure are considered. The method for extracting pairwise interaction energies from an extension of the diffuse scattering analysis is discussed, and an example of the use of such energies in kinetic Ising modelling is presented. This contribution is written in terms of x-ray experiments, since x-rays have been used for most local order diffuse scattering investigations.

Theoretical Development

Within the kinematic approximation of scattering theory, the coherent scattering from a binary alloy, with species A and B, is given in electron units by

$$I_{eu}(\vec{k}) = \sum_p \sum_q f_p f_q e^{i\vec{k}\bullet(\vec{R}_p - \vec{R}_q)}, \qquad (1)$$

where f_p and f_q are the atomic scattering factors of the atoms located at sites p and q, respectively, $\left(\vec{R}_p - \vec{R}_q\right)$ is the instantaneous interatomic vector and

$$\vec{k} = 2\pi\left(\frac{\hat{s} - \hat{s}_o}{\lambda}\right), \qquad (2)$$

where $\hat{s}_o$ and $\hat{s}$ are unit vectors in the directions of the incident and scattered beams, respectively, and λ is the x-ray wavelength.

The interatomic vector can be written as

$$\left(\vec{R}_p - \vec{R}_q\right) = \left\langle \vec{R}_p - \vec{R}_q \right\rangle + \left(\vec{\delta}_p - \vec{\delta}_q\right), \qquad (3)$$

where $\vec{\delta}_p$ and $\vec{\delta}_q$ are vector displacements from the average lattice sites. The $\langle\ \rangle$ brackets indicate an average over time and space. Thus

$$I_{eu}(\vec{k}) = \sum_p \sum_q f_p f_q e^{\left\langle i\vec{k}\bullet(\vec{\delta}_p - \vec{\delta}_q)\right\rangle} e^{\left\langle i\vec{k}\bullet(\vec{R}_p - \vec{R}_q)\right\rangle}. \qquad (4)$$

In essence, the problem in treating local order diffuse scattering is to evaluate the term $\left\langle f_p f_q e^{i\vec{k}\bullet(\vec{\delta}_p-\vec{\delta}_q)}\right\rangle$, taking into account all possible combinations of atom pairs; AA, AB, BA, and BB.

The modern theory and study of local atomic order diffuse scattering had their origins in the classical work by Cowley (1950) in which he set the displacements to zero. Experimental observations by Warren, Averbach and Roberts (1951), however, soon demonstrated the necessity to account for this atomic displacements effect, which tends to shift the local order diffuse maxima from positions of cosine symmetry.

Borie (1961) showed that a linear approximation of the exponentials containing the displacements allowed one to separate the local order and static atomic displacements components by making use of the fact that the various components of diffuse scattering have different symmetry in reciprocal space. This approach was extended to a quadratic approximation of the atomic displacements by Borie and Sparks (1971). Tibbals (1975) argued that the theory could be cast so as to allow inclusion of the reciprocal space variation of the atomic scattering factors, and this is included in the state-of-art formulation by Georgopolous and Cohen (1977), which is outlined here.

Quite generally, for a binary alloy one can write

$$\left\langle f_p f_q e^{i\vec{k}\bullet(\vec{\delta}_p-\vec{\delta}_q)}\right\rangle = X_A P_{pq}^{AA} f_A^2 \left\langle e^{i\vec{k}\bullet(\vec{\delta}_p^A-\vec{\delta}_q^A)}\right\rangle + X_A P_{pq}^{BA} f_A f_B \left\langle e^{i\vec{k}\bullet(\vec{\delta}_p^B-\vec{\delta}_q^A)}\right\rangle$$

$$+X_B P_{pq}^{AB} f_A f_B \left\langle e^{i\vec{k}\bullet(\vec{\delta}_p^A-\vec{\delta}_q^B)}\right\rangle + X_B P_{pq}^{BB} f_B^2 \left\langle e^{i\vec{k}\bullet(\vec{\delta}_p^B-\vec{\delta}_q^B)}\right\rangle, \quad (5)$$

where X_A and X_B are atom fractions of species A and B, respectively, and P_{pq}^{AB} is the conditional probability of finding an A atom at site p provided there is a B atom at site q. By invoking continuity arguments and introducing the Cowley-Warren (CW) order parameter (Cowley (1950)),

$$\alpha_{pq} = 1 - \frac{P_{pq}^{BA}}{X_B}, \quad (6)$$

eq. 5 reduces to

$$\left\langle f_p f_q e^{i\vec{k}\bullet(\vec{\delta}_p-\vec{\delta}_q)}\right\rangle = \left(X_A^2 + X_A X_B \alpha_{pq}\right) f_A^2 \left\langle e^{i\vec{k}\bullet(\vec{\delta}_p^A-\vec{\delta}_q^A)}\right\rangle$$

$$+2X_A X_B\left(1-\alpha_{pq}\right) f_A f_B \left\langle e^{\vec{k}\bullet(\vec{\delta}_p^B-\vec{\delta}_q^A)}\right\rangle$$

$$+\left(X_B^2 + X_A X_B \alpha_{pq}\right) f_B^2 \left\langle e^{\vec{k}\bullet(\vec{\delta}_p^B-\vec{\delta}_q^B)}\right\rangle. \tag{7}$$

If one makes series expansions of the exponentials and retains only quadratic and lower order terms, it follows that

$$I_{eu}(\vec{k}) = \sum_p\sum_q \left(X_A f_A + X_B f_B\right)^2 e^{i\vec{k}\bullet\vec{R}_{pq}}$$

$$+\sum_p\sum_q X_A X_B \left(f_A - f_B\right)^2 \alpha_{pq} e^{i\vec{k}\bullet\vec{R}_{pq}}$$

$$+\sum_p\sum_q \Big[(X_A^2 + X_A X_B \alpha_{pq}) f_A^2 \left\langle i\vec{k}\bullet(\vec{\delta}_p^A - \vec{\delta}_q^A)\right\rangle$$

$$+2X_A X_B (1-\alpha_{pq}) f_A f_B \left\langle i\vec{k}\bullet(\vec{\delta}_p^B - \vec{\delta}_q^A)\right\rangle$$

$$+(X_B^2 + X_A X_B \alpha_{pq}) f_B^2 \left\langle i\vec{k}\bullet(\vec{\delta}_p^B - \vec{\delta}_q^B)\right\rangle\Big] e^{i\vec{k}\bullet\vec{R}_{pq}}$$

$$-\sum_p\sum_q 1/2\Big[(X_A^2 + X_A X_B \alpha_{pq}) f_A^2 \left\langle\left[\vec{k}\bullet(\vec{\delta}_p^A - \vec{\delta}_q^A)\right]^2\right\rangle$$

$$+2X_A X_B (1-\alpha_{pq}) f_A f_B \left\langle\left[\vec{k}\bullet(\vec{\delta}_p^B - \vec{\delta}_q^A)\right]^2\right\rangle$$

$$+(X_B^2 + X_A X_B \alpha_{pq}) f_B^2 \left\langle\left[\vec{k}\bullet(\vec{\delta}_p^B - \vec{\delta}_q^B)\right]^2\right\rangle\Big] e^{i\vec{k}\bullet\vec{R}_{pq}}, \tag{8}$$

where $e^{i\vec{k}\bullet\vec{R}_{pq}}$ denotes $\left\langle e^{i\vec{k}\bullet\left(\vec{R}_p-\vec{R}_q\right)}\right\rangle$. The first double summation represents the fundamental Bragg reflections for the average lattice. The second summation is the order modulated Laue monotonic, the term of primary interest here. The third sum is the so-called first order atomic displacements, and it is purely static in nature. The final double summation is the second order atomic displacements and contains both static and dynamic contributions.

For alloys which exhibit only local ordering, it is convenient to replace the double summations by N times single sums over lattice sites, now specified by triplets of integers (lmn); N is the number of atoms irradiated. One can express the average interatomic vector as

$$\langle \vec{R}_{lmn} \rangle = l\frac{\vec{a}_1}{2} + m\frac{\vec{a}_2}{2} + n\frac{\vec{a}_3}{2}, \tag{9}$$

where $\frac{\vec{a}_1}{2}$, $\frac{\vec{a}_2}{2}$ and $\frac{\vec{a}_3}{2}$ are orthogonal vectors parallel to the cubic unit cell edges. The continuous variables in reciprocal space (h_1, h_2 and h_3) are given by

$$\left(\frac{\vec{s} - \vec{s}_o}{\lambda}\right) = h_1\vec{b}_1 + h_2\vec{b}_2 + h_3\vec{b}_3, \tag{10}$$

where $\vec{b}_1$, $\vec{b}_2$ and $\vec{b}_3$ are a set of vectors reciprocal to $\frac{\vec{a}_1}{2}$, $\frac{\vec{a}_2}{2}$ and $\frac{\vec{a}_3}{2}$. The coordinate scale used here is that conventionally employed in diffuse scattering work. Note that the 200 Bragg position becomes 1,0,0, etc., in this notation.

If, in addition to invoking the symmetry of the cubic lattice, one expresses the vector displacements in terms of components along the respective axes and simplifies the various expressions, the coherently scattered diffuse intensity which is observable becomes, in the quadratic approximation of the atomic displacements,

$$\begin{aligned}
\frac{I_D(h_1,h_2,h_3)}{NX_AX_B(f_A - f_B)^2} = \sum_l\sum_m\sum_n \alpha_{lmn} \cos 2\pi(h_1 l + h_2 m + h_3 n) \\
+ h_1\eta Q_x^{AA} + h_1\xi Q_x^{BB} + h_2\eta Q_y^{AA} \\
+ h_2\xi Q_y^{BB} + h_3\eta Q_z^{AA} + h_3\xi Q_z^{BB} \\
+ h_1^2\eta^2 R_x^{AA} + 2h_1^2\eta\xi R_x^{AB} + h_1^2\xi^2 R_x^{BB} \\
+ h_2^2\eta^2 R_y^{AA} + 2h_2^2\eta\xi R_y^{AB} + h_2^2\xi^2 R_y^{BB} \\
+ h_3^2\eta^2 R_z^{AA} + 2h_3^2\eta\xi R_z^{AB} + h_3^2\xi^2 R_z^{BB} \\
+ h_1h_2\eta^2 S_{xy}^{AA} + 2h_1h_2\eta\xi S_{xy}^{AB} + h_1h_2\xi^2 S_{xy}^{BB} \\
+ h_1h_3\eta^2 S_{xz}^{AA} + 2h_1h_3\eta\,\xi S_{xz}^{AB} + h_1h_3\xi^2 S_{xz}^{BB} \\
+ h_2h_3\eta^2 S_{yz}^{AA} + 2h_2h_3\eta\xi S_{yz}^{AB} + h_2h_3\xi^2 S_{yz}^{BB},
\end{aligned} \tag{11}$$

where $$\eta = \frac{f_A}{f_A - f_B} \tag{12}$$

and $$\xi = \frac{f_B}{f_A - f_B}. \tag{13}$$

The Q functions which describe the first order size effects component are of the form

$$Q_x^{AA} = -2\pi \sum_l \sum_m \sum_n \left(\frac{X_A}{X_B} + \alpha_{lmn}\right) \left\langle X^{AA} \right\rangle_{lmn} \sin 2\pi h_1 l \cos 2\pi h_2 m \cos 2\pi h_3 n, \qquad (14)$$

where $\left\langle X^{AA} \right\rangle_{lmn}$ is the average component of displacement of the A atom at site l,m,n.

The R and S functions describe the second order atomic displacements component of diffuse scattering and are of the form

$$R_X^{AA} = 4\pi^2 \sum_l \sum_m \sum_n \left(\frac{X_A}{X_B} + \alpha_{lmn}\right) \left\langle X_0^A X_{lmn}^A \right\rangle \cos 2\pi h_1 l \cos 2 h_2 m \cos 2\pi h_3 n \qquad (15)$$

and

$$S_{xy}^{AA} = 8\pi^2 \sum_l \sum_m \sum_n \left(\frac{X_A}{X_B} + \alpha_{lmm}\right) \left\langle X_0^A Y_{lmn}^A \right\rangle \sin 2\pi h_1 l \sin 2 h_2 m \cos 2\pi h_3 n. \qquad (16)$$

The first sum in eq. 11 contains the information about the local atomic ordering; it is a three-dimensional Fourier cosine series whose coefficients are the Cowley-Warren order parameters. The first term in this series is a measure of the integrated local order diffuse scattering, and, provided the data are normalized by the Laue monotonic unit, α_{000} should have the value of unity.

The coherent diffuse scattering thus consists of 25 components, each of which possesses distinct symmetry, and this fact permits the components to be separated. To effect the separation, intensity measurements are made at a set of points referred to as the "associated set". These associated points are selected according to crystallographic symmetry rules such that the corresponding 25 unknowns in eq. 11 have the same magnitude, but may differ in sign. An associated set is measured for each point in the required minimum volume for the local order component of diffuse scattering.

The theory outlined above has heretofore been used largely for studying short range ordering alloys (preference for unlike nearest neighbors); however, the theory is equally valid for alloy systems which undergo clustering (preference for like nearest neighbors), or even a combination of the two. If clustering occurs, local order diffuse scattering will be distributed near the fundamental Bragg positions; i.e., small angle scattering will be observed. In this case, a higher density of data points in the low-angle region will be required, and it would appear that the linear regression method of Williams (1976) is more appropriate than a direct Fourier transform to obtain the relevant order coefficients and size effects parameters.

Preliminary Data Treatment

In a given set of diffuse scattering measurements, there may be significant contributions from a number of sources other than those contained explicitly in the theory outlined in Section 2. These extraneous components must be either avoided experimentally or suitable corrections made on the diffuse scattering data before separation and analysis. Space constraints do not permit

detailed consideration here, but this information is available in the appendices of Schwartz and Cohen (1987). Nonetheless, a more-or-less listing of these corrections here appears to be desirable.

With conventional laboratory x-ray sources, detection system dead time losses for diffuse scattering measurements are usually not a major concern, but may be important at a synchrotron source. When important, dead time corrections should be made for all detectors involved in the experiment. Then, each primary count should be normalized to a constant number of monitor counts, since the incident flux generally is not constant.

Even for an electropolished sample, surface roughness can be a factor. The surface quality can be determined by inducing fluorescence in the sample and measuring this intensity as a function of scattering angle. If the surface is rough, the measured fluorescence will be less at small angles.

Air scattering can be greatly reduced by evacuating the sample area. Any remaining contribution can be determined by making a set of measurements with a lead trap in place of the sample.

If the detector has poor energy resolution, unwanted photon energies (such as fluorescence induced in the sample or scattering due to submultiple wavelengths present in the incident beam) must be measured, or estimated, and removed.

Although not a correction, the data need to be converted to electron units. This is usually done by measuring the Bragg scattering from a powder compact of aluminum; it is necessary, of course, to select a standard sample which does not have a preferred orientation, or texture.

Absorption effectively reduces the flux seen by atoms below the sample surface. Appropriate corrections are necessary, utilizing either tabulated absorption coefficients or experimentally determined values.

A sample dependent incoherent scattering process always accompanies the coherent scattering discussed in Section 2. This is the Compton scattering, and it has a slightly altered wavelength, which is dependent on the scattering angle. Tabulations of incoherent scattering cross sections are available which allow one to calculate and subtract this contribution. The Breit-Dirac recoil correction factor must also be applied here.

The tabulated atomic scattering factors are actually high frequency limits. For real systems, it is necessary to apply both real and imaginary dispersion corrections (Sasaki (1989)), commonly referred to as anomalous dispersion corrections. These become crucial for wavelengths near an absorption edge of an atomic species contained in the sample. Of course, this can sometimes be used to advantage in allowing one to enhance the scattering contrast. The scattering factor must also be corrected for a Debye-Waller temperature effect.

Finally, the quadratic approximation to atomic displacement discussed in Section 2 is deficient with regards to higher order thermal diffuse scattering (TDS). If atomic force constants are available from inelastic neutron scattering investigations, the higher order TDS can be calculated and subtracted. Otherwise, less satisfactory methods may be necessary, such as using a weighed average of the TDS from the pure constituents.

Real Space Modelling of the Local Order Structure

The Cowley-Warren order coefficients, which contain the information about how the lattice sites are occupied, are statistical in nature. This statistical definition made it possible for the elegant theory outlined above to be developed; however, it presents a challenge when one faces the task

of trying to visualize the real space structure of the alloy. To be sure, from a tabulation of the α_i it becomes readily apparent whether the system is short range ordered or clustered; a negative sign for α_1 signifies ordering and a positive sign signifies clustering. Of course, such qualitative information could have been inferred from a cursory inspection of the raw diffuse scattering profile. Although an exact solution is out of the question, it is highly desirable to be able to visualize the essential features of the real space atomic structure.

It was clear to Cowley (1950) from the beginning that the order parameters for the lower coordination shells constrain the values higher shells may have. Recognition of this fact led Gehlen and Cohen (1965) to generate a large computer model of the alloy, in agreement with the experimental CW order parameters. A number of such programs based upon this concept have since been written; however, regardless of whose program was used we choose to designate the modelled structure as the GC structure. How well the GC structure represents the real alloy has been considered by Cohen (1970). He argued that if the alloy is studied under conditions far from a critical point for phase separation, the excess specific heat should be small. Since the excess specific heat is proportional to the variance of the internal energy, a small excess specific heat signifies a small variance of the internal energy. Consequently, a limited number of possible atomic configurations was proposed.

While it is recognized that the GC structure is not unique, it is nonetheless our best presently available means of obtaining a reasonable, graphical representation of the local atomic structure from the statistical Cowley-Warren order coefficients. As a specific example of how the GC structure is generated, the program FCCFIT by Epperson (1979) is described briefly. A large, three-dimensional matrix of proper composition to represent the alloy is set up with 0's and 1's representing the major and minor atomic species, respectively. Periodic boundary conditions are used. A steering matrix consisting of a non-sequential array of the site indices specifies the order in which sites in the model are queried, and this scheme assures that each site has equal probability of being searched. A partial flow diagram of the modelling algorithm is shown in Fig. 1. In essence, the computer chooses a 0-1 pair at random and examines the environment around each site to determine if an atom exchange would result in a better fit with the experimental CW order parameters. If a better fit would result, the exchange is made and counters are upgraded, and the search-query-exchange process is repeated until pre-selected conditions are satisfied. Typically, a "close fit" is carried out, defined as fitting until further exchanges would not improve the agreement with the experimental CW coefficients. The

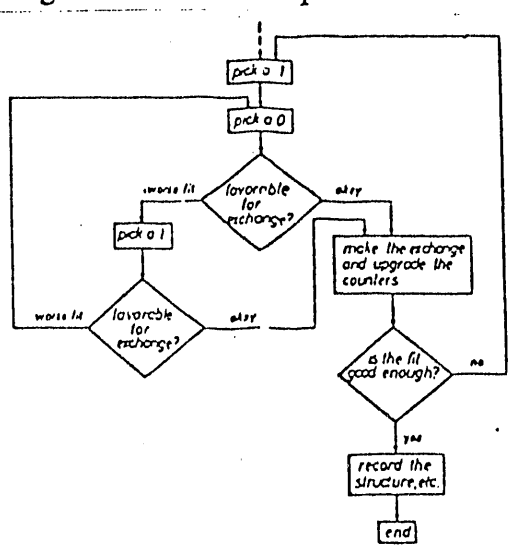

Fig. 1. Partial flow diagram used to generate the GC structure with the program FCCFIT by Epperson (1979).

experimental order coefficients thus effectively serve as boundary conditions for the modelling. A close fit to the first six α_i has been found to be generally practical, but it becomes more difficult with each additional shell, especially for systems with significant local atomic order. However, even after close fitting the first six α_i, statistically significant differences still exist between experimental and modelled order parameters for higher coordination shells. Presently, we have two models in use; an 8000 atom model for survey purposes and for short range ordering systems and another with 140,608 atoms for better statistics and for alloys which undergo clustering. Table 1 compares a set of experimental α_i with those modelled for a long time aged Cu-14.7$_6$ at. %Al alloy from Epperson, Fürnrohr and Ortiz (1978).

Table 1. Comparison of Experimental and Modelled (8000 atom) Cowley-Warren Order Coefficients for Cu-14.7$_6$ at % Al Aged 1580 hrs at 150°C, after Epperson, Fürnrohr and Ortiz (1978).

Coordination Shell (i)	Site Indices (lmn)	α_{lmn} Expt.	α_{lmn} Mod.
0	000	1.015	1.0000
1	110	-0.171	-0.1709
2	200	0.125	0.1245
3	211	0.042	0.0416
4	220	-0.004	-0.0043
5	310	-0.034	-0.0343
6	222	-0.032	-0.0324
7	321	0.003	0.0078
8	400	0.021	0.0305
9a	330	-0.008	0.0026
9b	411	0.005	0.0040
10	420	0.007	-0.0034

Characterization of the Modelled Structure

Having generated a real space model of the locally ordered structure from a set of CW order coefficients, the next task is to extract information about the structure. Provided there are sizeable clusters or ordered regions, it is feasible to characterize the GC structure by examining individually adjacent planes as has been done, for example, by Gragg and Cohen (1971), by Auvray, Georgopolous and Cohen (1981) and by Koo and Cohen (1989). This method is, at best, tedious and time consuming, and it would be easy to overlook small but significant features, especially if they are not confined to the cube planes; for example, consider the Cu-Al tetrahedra of Borie and Sparks (1964) which consist of five atoms and require examination of three adjacent cube planes. A method developed by Epperson (1979) makes extensive use of the computer in characterizing the GC structure. For a binary face centered cubic (fcc) alloy, there are

$$N_c = \prod_{i=o}^{12} \frac{12!}{i!(12-i)!} \tag{17}$$

possible arrangements of the atoms in the first coordination shell about a given atom. However, because of the symmetry of the cubic lattice these 4096 possibilities reduce to a more modest 144 crystallographically distinct first nearest neighbor (nn) configuration types. It is convenient to designate the sites in the first coordination shell about an arbitrary lattice site as indicated in Fig. 2.

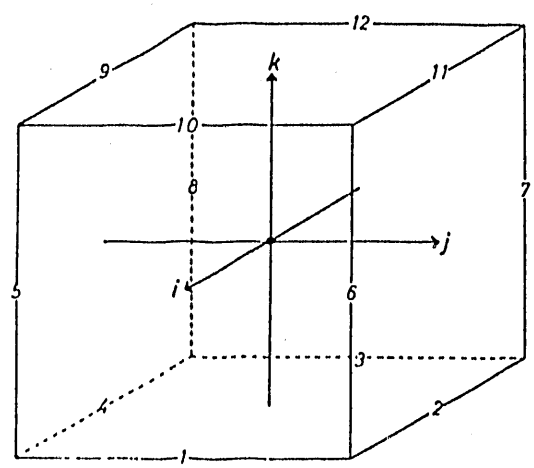

Fig. 2. Site designation for the nearest neighbor configurations in a binary face centered cubic alloy, after Epperson (1979).

A listing of the 144 distinct nn configurations is given in Table 2, along with the corresponding multiplicities. It is straightforward to create the complete catalogue of the 4096 nn configurations by applying, in succession, the 48 m3m point group symmetry operations to each of the 144 nn configuration types listed in Table 2, and by eliminating redundancies. This catalogue constitutes a look-up table in program NTKRKR and permits each site in the GC structure to be characterized in terms of its nearest neighbor environment.

The technique just described was first used to examine the reproducibility of the GC structure in terms of the number of coordination shells fitted. Close fit 8000 atom GC structures, as defined in the previous section, were constructed for a given set of α_i with program FCCFIT using, in succession, coordination shells 1-2, 1-3, 1-4, 1-5 and 1-6. One can define a mean relative deviation of the nn configuration concentration by

$$D_m = \frac{1}{N} \sum_{i=1}^{N} \left(\frac{2\sigma_i}{Ci^*} \right) \bullet 100, \tag{18}$$

where Ci^* is the concentration of configuration Ci (Table 2) and σ_i in the standard deviation computed from the six independent runs close fitted through a given coordination shell. The results are shown in Fig. 3.