Direct solution of the diffraction pattern of a crystal with planar faulting

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A direct formalism for the solution of the diffraction pattern from the faulted layer crystal is derived. The proposed method is not specific for any crystal structure. The solution avoids the need for specific planar faulting models and has a direct physical meaning. The correlation distribution function between lateral displaced layers can be directly obtained from the diffracted intensities. The solution was compared successfully to a Monte Carlo trial and error method for the fcc structure. The developed formalism was used to determine the layer-layer correlation distribution function of the Gd_2Co_{17} faulted layer structure.

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I. INTRODUCTION

It has long been known that the occurrence of planar faulting in layer crystals could have drastic effects on the materials properties and behavior. Stacking faults are usually presented by considering disorder in the stacking sequence along the [111] direction in face-centered-cubic (fcc) crystals or along the [001] stacking sequence in hexagonal compact (hcp) crystals. Yet stacking faults are found in a variety of materials used in a wide range of applications.

Studies of stacking fault defects comes from early works of Landau[1] and Lifschitz.[2] Hendricks and Teller[3] discussed planar faulting in fcc and hcp crystals using correlation probability matrices. Wilson[4] and Warren[5] used a different approach based on a difference equation, which became the method usually described in x-ray textbooks.[6,7] Jagodinski[8] further extended the difference equation method. Recently Velterop *et al.*[9] lifted some of the simplifying assumptions of the Warren formalism to extend its use to nonuniform fault probabilities and textured samples.

Most procedures for the analysis of planar faulted layer crystals have relied on particular models for the occurring faults in particular crystal systems (see for example Refs. 5,8, and 9). The most common model, used in fcc and hcp structures, is to assume the occurrence of simple planar faulting of two types, stacking faults and twinning, also known as deformation and growth faulting. Both type of planar faulting are described by their probability of occurrence α and β. The models also assume that individual planar fault occurs as independent events.

Deformation faults involve a ''jump'' in the otherwise perfect stacking sequence. The fault results in apparently missing layers in the sequence. A twin fault, on the other hand, involves a change in the stacking order, which in the case of the fcc structure results in a reversion of the stacking sequence.

The Warren[5] theory for stacking faults and twinning, although the most common used, is hard to follow, and the relation between measurable quantities from the diffraction pattern with α and β, was obtained for a particular crystal system (namely fcc, hcp, and bcc) after several simplifying assumptions. Velterop *et al.*[9] emphasizes in three assumptions made in Warren formalism that: (i) all components have equal integrated intensities, (ii) fault densities are very small, and (iii) fault densities are equal for all crystallites.

Other models rely on the Warren theory and add further modeling of the peaks for particular profile fitting functions.[10]

Berliner and co-workers, in a series of papers,[11–13] introduced a Monte Carlo method for simulation of the diffraction pattern from faulted structures. The method also relies on a particular model of planar disorder, and although easily extensible to other models, it still depends on the ingenuity of the researcher to propose an adequate model for the faulting. The formalism uses a trial and error procedure without clear matching criteria.

In this paper we present a direct solution for the layer-layer correlation probability distribution function from a powder diffraction pattern of a faulted crystal. The proposed formalism avoids the need for trial and error procedures and does not need to assume a particular model for the planar faulting. The paper is organized as follows. In Sec. II we develop the proposed formalism. The solution obtained will be compared to the Monte Carlo method proposed by Berliner *et al.*[11–13] for a fcc structure in Sec. III, and in Sec. IV the meaning and use of the obtained layer-layer correlation probability distribution will be discussed. Finally in Sec. V the same formalism will be used to determine the layer-layer correlation function for the Gd_2Co_{17} layer structure.

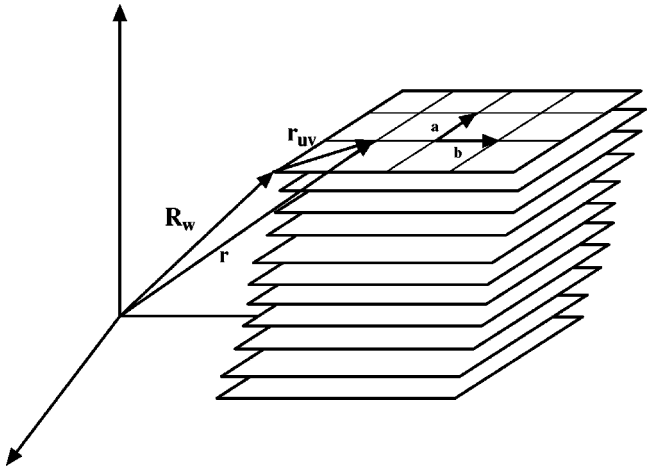

FIG. 1. Schematic representation of a layer crystal. A bidimensional periodic lattice can be associated with the layers. The quantity $(\boldsymbol{a},\boldsymbol{b})$ defines a unit cell within the layer lattice with $\rho(\boldsymbol{r})$ density of scatterers. A object space vector $\boldsymbol{r}$, pointing to the (u,v) node in the w layer, can be written as the sum of two vectors $\boldsymbol{r}=\boldsymbol{R}_w+\boldsymbol{r}_{uv}$.

II. DIFFRACTION IN A LAYERED CRYSTAL

A. General solution for a single crystal

A layer crystal is built by the assembly of identical layers perfectly periodic in two dimensions. To the perfectly periodic layer a two dimensional lattice can be associated with lattice vector $\boldsymbol{r}_{uv}=u\boldsymbol{a}+v\boldsymbol{b}$ where u,v are integers and $(\boldsymbol{a},\boldsymbol{b})$ define a primitive cell for the bidimentional lattice (Fig. 1). A three dimensional layer crystal of size $N_1\times N_2\times N_3$ can be represented by the sum of two vectors, a vector $\boldsymbol{R}_w$ that goes from the origin of object space to the w layer in the crystal and the vector $\boldsymbol{r}_{uv}$. Let $\mathcal{L}(\mathbf{r})$ represent the layer crystal, then we can write

$$\mathcal{L}(\boldsymbol{r})=\rho(\boldsymbol{r})\otimes\left(\sum_{u=-(N_1/2)}^{N_1/2}\sum_{v=-(N_2/2)}^{N_2/2}\delta(\boldsymbol{r}_{ab}-\boldsymbol{r}_{uv})\right)\times\sum_{w=0}^{N_3-1}\delta(\boldsymbol{R}-\boldsymbol{R}_w),\tag{1}$$

where $\otimes$ represents the convolution operator and $\boldsymbol{r}$, the object space vector, has been written as the sum of two vectors $\boldsymbol{r}=\boldsymbol{r}_{ab}+\boldsymbol{R}$, $\boldsymbol{r}_{ab}$ being parallel to the crystal layers. δ represent the Dirac delta and $\rho(\boldsymbol{r})$ the density of scatterers. The first term between parentheses describes the two dimensional lattice associated with the layers. In general Eq. (1) does not describe a three dimensional lattice, as $\boldsymbol{R}_w$ does not necessarily represent a lattice vector. According to kinematical diffraction theory, the amplitude of a diffracted wave, in units of the amplitude scattered by a single scattering center, will be the Fourier transform of the density of scatterers in object space.[7] If $F_m(\boldsymbol{r}^*)$ is the Fourier transform of the scatterers density $\rho_m(\boldsymbol{r})$ (the so called structure factor), the corresponding diffracted intensity for N_1,N_2 nodes will then be

$$I_c(\boldsymbol{r}^*)=F_m^2(\boldsymbol{r}^*)\left(\frac{\sin(N_1\pi h)}{\sin(\pi h)}\right)\left(\frac{\sin(N_2\pi k)}{\sin(\pi k)}\right)\times\sum_{w=0}^{N_3-1}\sum_{w'=0}^{N_3-1}\exp[2\pi i\boldsymbol{r}^*\cdot(\boldsymbol{R}_w-\boldsymbol{R}_{w'})],\tag{2}$$

$\boldsymbol{r}^*=h\boldsymbol{a}^*+k\boldsymbol{b}^*+l\boldsymbol{c}^*$ being a reciprocal space vector with reciprocal base vectors $(\boldsymbol{a}^*,\boldsymbol{b}^*,\boldsymbol{c}^*)$ dual to the object space base vectors $(\boldsymbol{a},\boldsymbol{b},\boldsymbol{c})$.

Interference effects can be described by the interference function[7]

$$\mathcal{F}(\boldsymbol{r}^*)=\frac{I_c(\boldsymbol{r}^*)}{NF_m^2(\boldsymbol{r}^*)}.$$

If we rearrange terms in Eq. (2) and call $\Delta=w-w'$, the interference function will be given for $N_1,N_2\gg 1$

$$\mathcal{F}(\boldsymbol{r}^*)=\frac{N_1N_2}{N_3}\left(\sum_{h_o=-\infty}^{\infty}\sum_{k_o=-\infty}^{\infty}\delta(h-h_o)\,\delta(k-k_o)\right)\times\left\{N_3+2\sum_{\Delta=1}^{N_3-2}\sum_{w=0}^{N_3-1-\Delta}\cos[2\pi\boldsymbol{r}^*\cdot(\boldsymbol{R}_w-\boldsymbol{R}_{w+\Delta})]\right\}.\tag{3}$$

The term inside the curly brackets in Eq. (3) is the contribution of the layer arrangement to the interference function. We will therefore, in what follows and for the remainder of the paper, only consider this term, which we will denote by $\mathcal{Q}(\boldsymbol{r}^*)$

$$\mathcal{Q}(\boldsymbol{r}^*)=1+\frac{2}{N_3}\sum_{\Delta=1}^{N_3-2}\sum_{w=0}^{N_3-1-\Delta}\cos[2\pi\boldsymbol{r}^*\cdot(\boldsymbol{R}_w-\boldsymbol{R}_{w+\Delta})].\tag{4}$$

If we take $F_m(\boldsymbol{r}^*)$ to be a slowly varying function of reciprocal coordinates, the intensity diffracted at $\boldsymbol{r}^*=h_o\boldsymbol{a}^*+k_o\boldsymbol{b}^*+l\boldsymbol{c}^*$, where h_o,k_o are integers, will be proportional to $\mathcal{Q}(\boldsymbol{r}^*)$ in the neighborhood of a integer l_0 value.

B. Diffraction of a layered crystal with constant lateral displacements

Let us consider a layer crystal where the displacement, perpendicular to the stacking direction, of any layer with respect to an arbitrary one chosen as the origin, is an integer multiple of a minimal displacement vector $\boldsymbol{r}_{w\Delta}=x\boldsymbol{a}+y\boldsymbol{b}$. The $\boldsymbol{R}_w$ vector which goes from the origin of the object space to the origin of the w layer can be decomposed in a lateral displacement vector parallel to $\boldsymbol{r}_{w\Delta}$ and a displacement vector along the stacking direction (Fig. 2)

$$\boldsymbol{R}_w=s\boldsymbol{r}_{w\Delta}+\frac{w\boldsymbol{c}}{n},$$

with s and w being integers, n the number of layers forming a unit cell, and taking $\boldsymbol{c}$ as the lattice vector along the stacking direction. If we take M to be the number of different

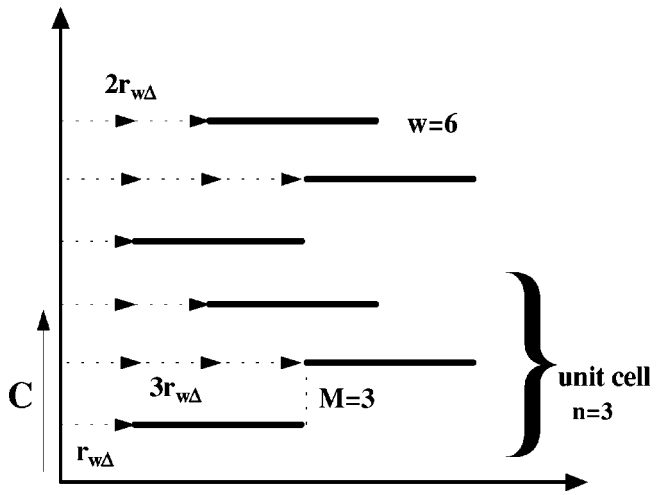

FIG. 2. Schematic representation of a layer crystal with constant lateral displacement shown perpendicular to the stacking direction. The position of each layer $\boldsymbol{R}_w$ can be described by the sum of a vector $w/n\boldsymbol{c}$ parallel to the stacking direction and an integer number of times a vector $\boldsymbol{r}_{w\Delta}$ parallel to the layer lattice. If M is the number of different lateral displacement then $M\boldsymbol{r}_{w\Delta}$ will be a translational vector of the bidimentional lattice. n is the number of layers that form a unit cell of height $|\boldsymbol{c}|$.

lateral displacement $s\boldsymbol{r}_{w\Delta}$ possible in the crystal, the expression Eq. (4) for $\mathcal{Q}(\boldsymbol{r}^*)$ can be written as

$$\mathcal{Q}(\boldsymbol{r}^*)=1+\frac{2}{N_3}\sum_{\Delta=1}^{N_3-1}\sum_{s=-(M-1)}^{M-1}N_s(\Delta)\times\cos\left[2\pi s(hx+ky)+2\pi\frac{\Delta l}{n}\right], \tag{5}$$

where $N_s(\Delta)$ is the number of pairs of layers, Δ layers apart (Δ pairs), with lateral displacement, one with respect to the other, of $s\boldsymbol{r}_{w\Delta}$. For a given value Δ, $N_3-\Delta$ will be the total number of Δ pairs for a crystal with N_3 layers height. $P_s(\Delta)=N_s(\Delta)/(N_3-\Delta)$ will then be the probability of finding in the crystal a Δ pair with lateral displacement $s\boldsymbol{r}_{w\Delta}$. Writing Eq. (5) in terms of $P_s(\Delta)$ and taking the limit for an infinite crystal along the stacking direction we arrive at the following expression for $\mathcal{Q}(\boldsymbol{r}^*)$

$$\mathcal{Q}(\boldsymbol{r}^*)=1+2\sum_{\Delta=1}^{\infty}\sum_{s=-(M-1)}^{M-1}P_s(\Delta)\times\cos\left[2\pi s(hx+ky)+2\pi\frac{\Delta l}{n}\right]. \tag{6}$$

From Eq. (6) it is clear that for reflections $\boldsymbol{r}^*=h\boldsymbol{a}^*+k\boldsymbol{b}^*+l\boldsymbol{c}^*$ such that

$$hx+ky=p \quad p=0,1,2,\ldots, \tag{7}$$

where p is an integer, the diffracted intensity does not depend on the particular stacking disorder.

From the bidimentional periodicity of the layers, $M\boldsymbol{r}_{w\Delta}$ is a translation vector of the associated lattice (Fig. 2) and Eq. (6) for $\mathcal{Q}(\boldsymbol{r}^*)$ reduces to

$$\begin{aligned}\mathcal{Q}(\boldsymbol{r}^*)=1+2\sum_{\Delta=1}^{\infty}\Bigg\{&P_0(\Delta)\cos\left(2\pi\frac{\Delta l}{n}\right)\\&+\sum_{s=1}^{(M-1)/2}(P_s(\Delta)+P_{M-s}(\Delta))\cos[2\pi s(hx+ky)]\\&\times\cos\left(2\pi\frac{\Delta l}{n}\right)+(P_{M-s}(\Delta)-P_s(\Delta))\\&\times\sin[2\pi s(hx+ky)]\sin\left(2\pi\frac{\Delta l}{n}\right)\Bigg\}\quad M\text{ odd},\end{aligned} \tag{8a}$$

$$\begin{aligned}\mathcal{Q}(\boldsymbol{r}^*)=1+2\sum_{\Delta=1}^{\infty}\Bigg\{&[P_0(\Delta)+P_{M/2}(\Delta)]\cos\left(2\pi\frac{\Delta l}{n}\right)\\&+\sum_{s=1}^{(M/2)-1}(P_s(\Delta)+P_{M-s}(\Delta))\\&\times\cos[2\pi s(hx+ky)]\cos\left(2\pi\frac{\Delta l}{n}\right)\\&+(P_{M-s}(\Delta)-P_s(\Delta))\\&\times\sin[2\pi s(hx+ky)]\sin\left(2\pi\frac{\Delta l}{n}\right)\Bigg\}\quad M\text{ even},\end{aligned} \tag{8b}$$

which describes the dependence of the diffracted intensity in the stacking order through the $P_s(\Delta)$.

C. The case of a powder sample

From the periodicity of the layer lattice, a displacement $(M-s)\boldsymbol{r}_{w\Delta}$ is crystallographically equivalent to a displacement $-s\boldsymbol{r}_{w\Delta}$. If we fix a coordinate system to a randomly oriented powder sample, a Δ pair with displacement $s\boldsymbol{r}_{w\Delta}$, from one crystal, is related to a Δ pair with displacement $-s\boldsymbol{r}_{w\Delta}$ from another crystal by a rigid motion. Therefore, the probability $P_s(\Delta)$ and $P_{M-s}(\Delta)$ in a powder sample should be the same,[14] and expression Eq. (8) for a powder sample can be written as

$$\mathcal{Q}(\boldsymbol{r}^*)=1+2\sum_{\Delta=1}^{\infty}G(\Delta)\cos\left(2\pi\frac{\Delta l}{n}\right), \tag{9}$$

with

$$G(\Delta)=P_0(\Delta)+2\sum_{s=1}^{(M-1)/2}P_s(\Delta)\cos[2\pi s(hx+ky)]\quad M\text{odd}, \tag{10a}$$

$$G(\Delta)=[P_0(\Delta)+P_{M/2}] + 2\sum_{s=1}^{(M/2)-1} P_s(\Delta)\cos[2\pi s(hx+ky)] \quad M \text{ even.} \tag{10b}$$

Equation (9) express the fact that the interference function $\mathcal{Q}(\boldsymbol{r}^*)$, which is proportional to the diffracted intensity, is a cosines series of $G(\Delta)$, and therefore $G(\Delta)$ can be obtained from $\mathcal{Q}(\boldsymbol{r}^*)$ as a cosine transform

$$G(\Delta)=\frac{1}{n}\int_{-(n/2)}^{n/2}\mathcal{Q}(\boldsymbol{r}^*)\cos\left(2\pi\frac{\Delta l}{n}\right)dl\Delta=1,2,\ldots. \tag{11}$$

The relation between $\mathcal{Q}(\boldsymbol{r}^*)$ and $G(\Delta)$ expressed by Eq. (11) shows the possibility of obtaining the P_s functions directly from the measured diffraction pattern. If we measure $\mathcal{Q}(\boldsymbol{r}^*)$ for as many (h,k) values as unknown P_s that we have, Eq. (10) together with Eq. (11) will lead to a linear set of equations which can be solved for each P_s. P_s is, at most, the maximum information you can get from a powder diffraction pattern regarding the stacking order of an ensemble of crystallites, as long as the crystallites can be considered individually to behave as infinite crystals. When the density of planar faulting is low the assumption of noninteracting occurrence of faults, such as those described by α and β, holds. Yet, if the crystal structure has heavy planar faulting, the stacking fault occurring in neighboring layers should interact and a description based solely in independent α and β probabilities is no longer valid. Velterop *et al.*[9] lifted the simplification made by Warren,[6] who omitted terms with α^2 and β^2, in order to use the same methods to higher planar faulting density. Yet, in their work, the description based on independent probabilities of simple planar faulting, described by α and β is kept, which still limits the validity of the method to not to high density of planar faulting. The use of Eq. (11) avoids the need of any prior assumption of the type of staking fault occurring in the crystals, and gives more complete information on the underlying stacking order from which usually used parameters can be obtained.

III. SIMULATIONS IN A FCC CRYSTAL

Berliner *et al.* proposed and successfully used a Monte Carlo trial and error method for the solution of layered crystal with stacking faults.[12] Here the method is briefly described. A particular model for occurrence of planar faulting is assumed for a layer crystal. For a fcc structure, the occurrence of single stacking faults in the otherwise perfect sequence is governed by the probability of occurrence α. The occurrence of twinning is described by the probability β. An ensemble of crystals are computer grown using pairs of random generated numbers: one for asserting the actual occurrence of a fault and a second one for discriminating between the two types of faults. The size of the crystal grown in this way was above 500 layers, where convergence of the results was found. The number of crystal grown in each trial was 10 000. The diffraction pattern of the ensemble of particles is then simulated and compared to the experimental pattern. Faulting probabilities, α and β, are changed until an appropriate match is found. In a fcc structure there are three positions for the layers usually called *A*, *B*, and *C*.[15] The position of the layers can be described by integer multiples of the vector

$$\boldsymbol{r}_{w\Delta}=\tfrac{1}{3}\boldsymbol{a}-\tfrac{1}{3}\boldsymbol{b}, \tag{12}$$

where the number of different displacement vectors can be 3 $(M=3,s=0(A),1(B),2(C))$. In such case we will be dealing with three probabilities P_0, P_1, and P_2. In a powder sample $P_1=P_2$, and therefore the following relation between probabilities will hold:

$$P_0=1-2P_1.$$

Taking into account the expressions for $G(\Delta)$ Eq. (10), for a fcc structure we will arrive at

$$G(\Delta)=P_0(\Delta)+(1-P_0(\Delta))\cos[2/3\pi(h-k)], \tag{13}$$

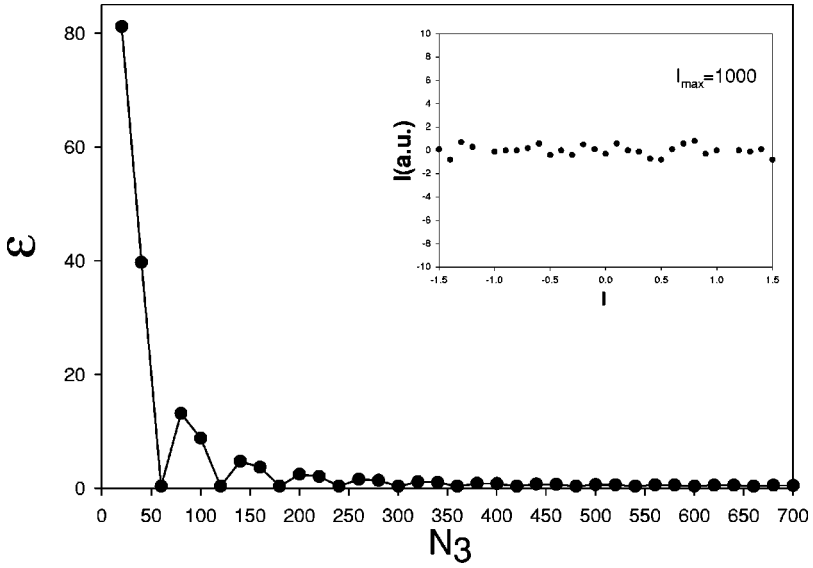

FIG. 3. Plot of ϵ as a figure of merit (see text) describing the convergence of the diffraction pattern from a N_3 layers height finite crystal to the diffraction pattern of an infinite crystal with increasing N_3. The inset plots the difference in the diffraction pattern from a 500 layer crystal and an infinite crystal. The maximum intensity value in each pattern was normalized to 1000 counts.

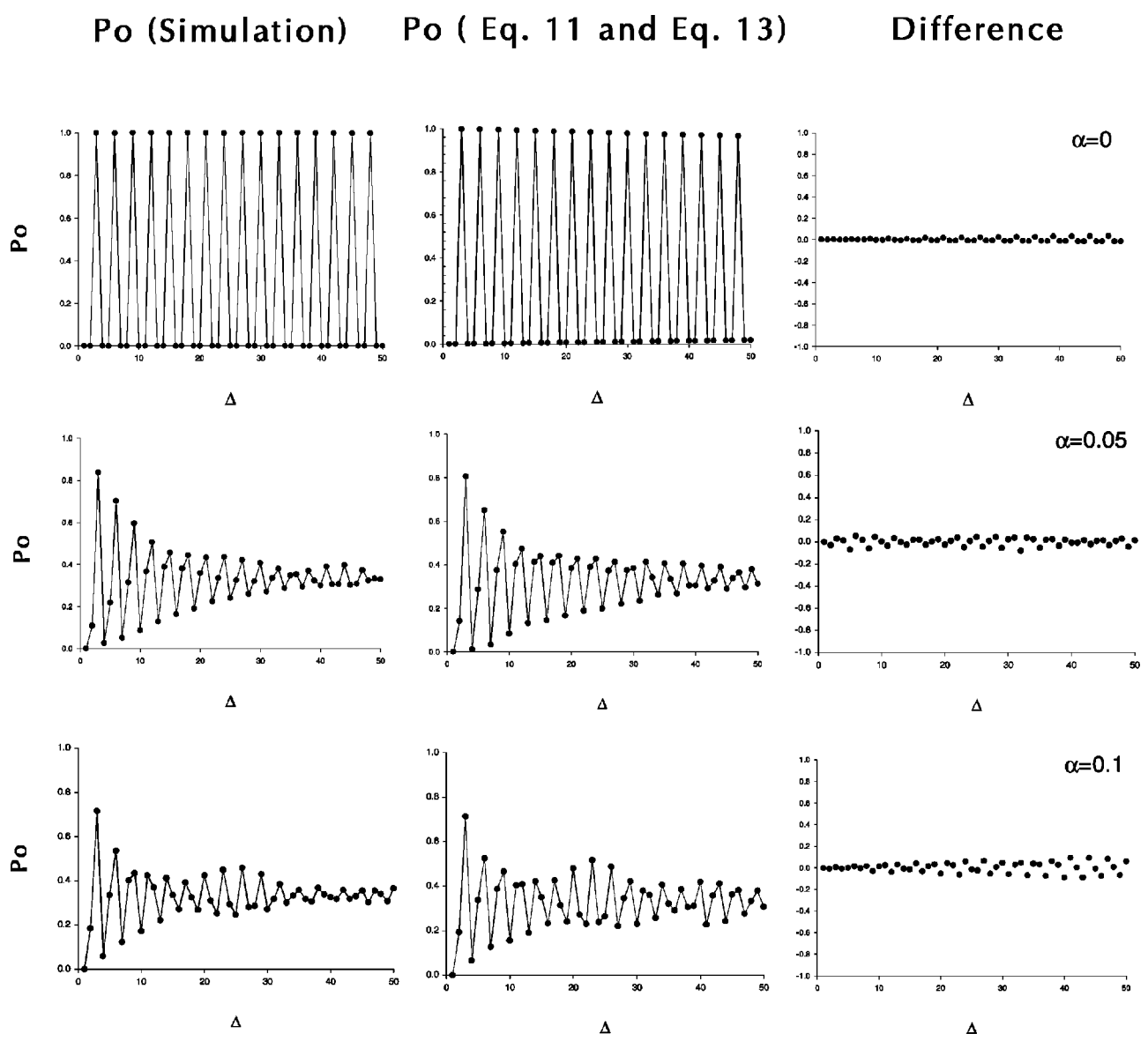

FIG. 4. Comparison between the P_0 function obtained from Monte Carlo simulation, and the P_0 function obtained by the use of Eq. (11) and Eq. (13) (see details in text). The third column plots the differences between both P_0 functions. Deformation faults were the only ones considered.

and the normalizing condition for the diffraction pattern in a close packed fcc structure can be written as

$$\cos\left(\frac{2}{3}\pi(h-k)\right)=\frac{1}{3}\int_{-3/2}^{3/2}\mathcal{Q}(\boldsymbol{r}^*)\cos\left(\frac{2\pi}{3}l\right)dl. \quad (14)$$

Let us define ε as the sum, over all values of l, of the absolute value of the difference between the diffraction pattern of an infinite crystal and the diffraction pattern of a finite size crystal

$$\varepsilon=\sum\ |\mathcal{Q}_\infty(l)-\mathcal{Q}_{N_3}(l)|. \quad (15)$$

The diffraction pattern of the finite crystal can be calculated using

$$\mathcal{Q}_{N_3}(\boldsymbol{r}^*)=1+\frac{2}{N_3}\sum_{\Delta=1}^{N_3-1}(N_3-\Delta)\Bigg(P_0(\Delta)+[1-P_0(\Delta)]\cos\left[\frac{2\pi}{3}(h-k)\right]\Bigg)\cos\left(\frac{2\pi}{3}\Delta l\right),$$

which is equivalent to the expression obtained in Ref. 12, while the $\mathcal{Q}_\infty$ can be calculated using expression (9) for an infinite crystal powder sample, using the appropriate parameters for a fcc crystal structure. As the number of layers N_3 in the finite size crystal increases, ε should tend to zero; this is precisely the behavior observed in Fig. 3. The inset of the figure shows the difference between the diffraction pattern of the infinite crystal and the 500 layer crystal, which shows that for such sizes the powder sample effectively behaves as an ensemble of infinite sized crystal.

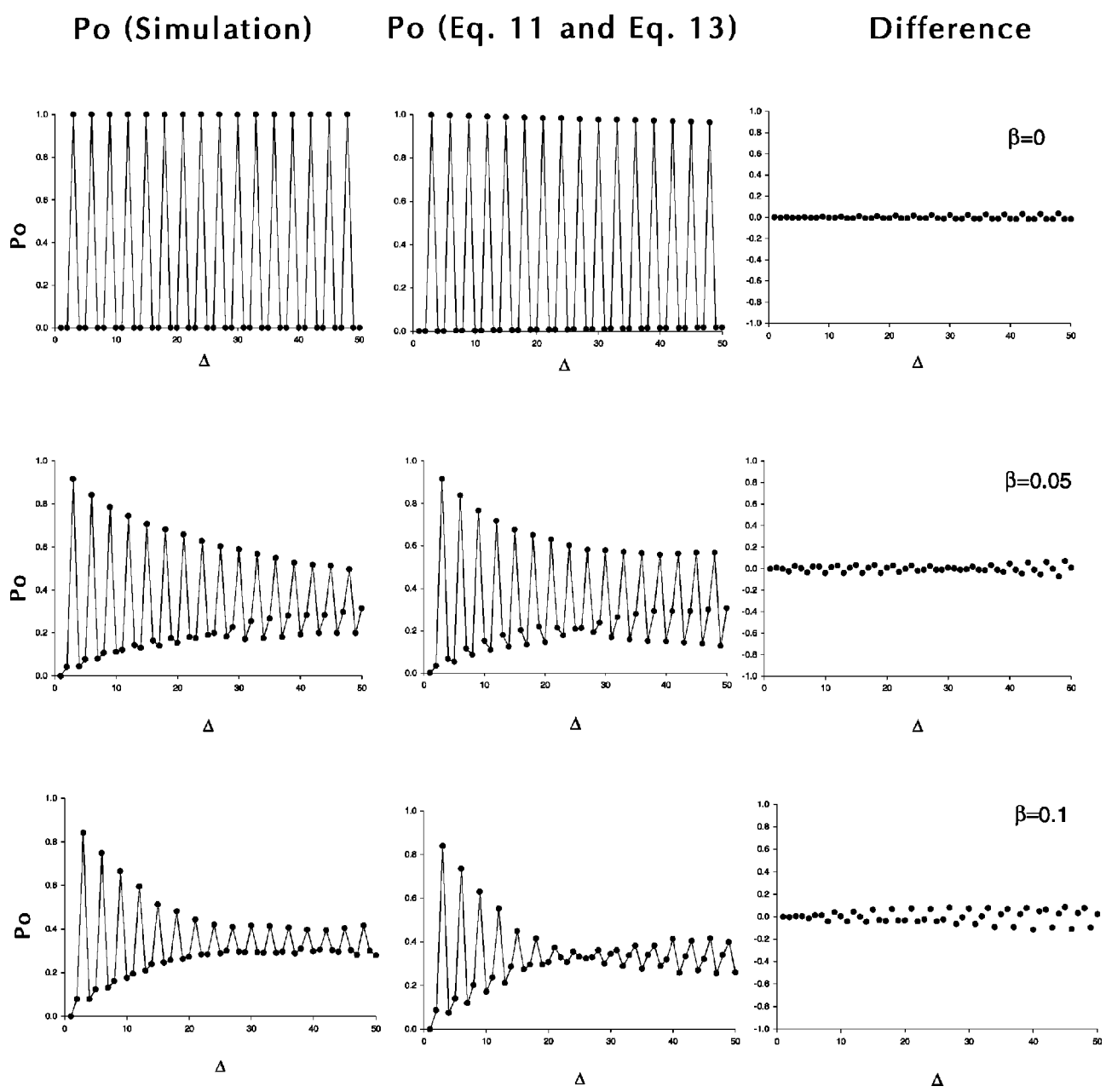

FIG. 5. Comparison between the P_0 function obtained from Monte Carlo simulation, and the P_0 function obtained by the use of Eqs. (11) and (13) (see details in text). The third column plots the differences between both P_0 functions. Twin faults were the only ones considered.

For a crystal with interlayer distance of around 0.2 nm (valid for some pure metals such as fcc iron or hcp cobalt) an effective ''infinite'' 500 layer crystal will be those with a length, along the stacking direction, of at least 0.1μm. In the case of some rare earth-transition metal binary alloys, where the interlayer distance is around 4 nm, the effective ''infinite'' crystal will be one with length of at least 2μm. From the above analysis we believe that the infinite crystal assumption is, for a wide range of cases, not a strong one. It should be noticed that for crystallite sizes below 100 nm, peak broadening due to small grain size is an additional factor affecting the peak profile.[6]

Following expressions (7) and (12), in a fcc structure the reflections affected by planar disorder along the [111] direction will be those with $h-k\neq 3p$, where p is an integer. For the perfect fcc stacking sequence we should expect ideally that the interference function $\mathcal{Q}(\boldsymbol{r}^*)$ would be a sequence of delta functions at the reciprocal lattice positions (h_o,k_o,l_o), where all three reciprocal coordinates are integers.[16] If we substitute this ''perfect'' $\mathcal{Q}(\boldsymbol{r}^*)$ function in Eq. (11) then the P_0 function will be $P_0(\Delta)=1$ if $\Delta=3p;0$ if $\Delta\neq 3p(p\in\mathcal{Z})$, which is the P_0 function we should expect for a $ABCABCABCABC\ldots$ perfect sequence as simple visual inspection of the sequence shows.

A series of α-faulted crystals were simulated using the Berliner[12] method, and diffraction patterns were calculated from such ensembles. The simulated diffraction patterns were then solved for P_0 using Eqs. (11) and (13), and the

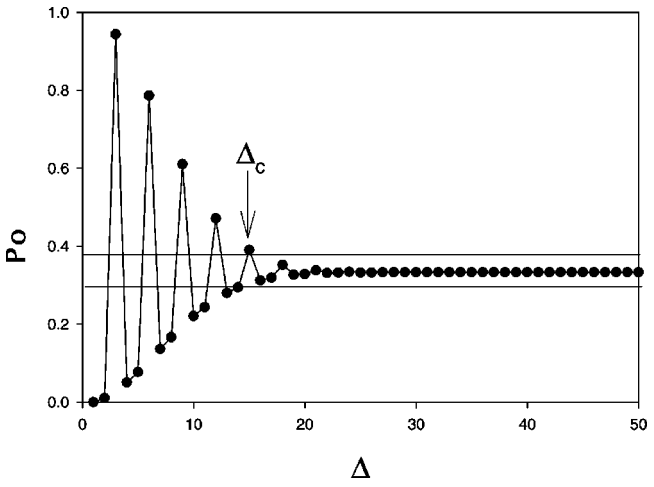

FIG. 6. Definition of the Δ_c parameter describing the distance above which all correlation between the displacement of Δ pairs is lost.

obtained values for P_0 were compared to the ones known for the simulated crystal. Figure 4 shows a comparison between the actual P_0 function and the calculated one for increasing values of the α parameter. The same procedure was followed but now with β-faulted crystal, Fig. 5 shows the corresponding results. A good match is found in both cases between the actual P_0 values and the calculated ones. Discrepancy between the simulated and calculated P_0 values increases with Δ. This can be explained as a result of numerical precision in the calculation. The discrete nature of the diffraction data does not allow the direct use of Eq. (11), but involves instead a discrete cosine transform of the same equation. The use of a discrete cosine transform imposes limiting conditions in the sampling interval of the diffraction data in order to obtain the P_0 function with enough accuracy up to a specified Δ value.

IV. THE P_s CORRELATION FUNCTION

The P_s functions have been usually overlooked in the treatment of planar faulting in layer crystal. The reason for this can be found in that up to now P_s has not been a function obtainable from measurable quantities from the diffraction pattern. P_s functions carry a lot more information than other parameters, such as α and β.

Long-range correlation between layers, and therefore long-range order, can be quantitatively asserted from the P_s function with the added benefit that no planar faulting model assumption is needed. Furthermore, the P_s function obtained is averaged over the hole ensemble of crystallites and therefore avoids the need for considering fault density equal for each crystallite.

Correlation between layers can be considered to be lost when the P_0 function stops oscillating and tends to a constant value of probability (in the case of a fcc structure this value should be 1/3). If we define Δ_c as the minimal Δ value for which the P_0 function has stopped oscillating, then we can define the maximum correlation distance between layers to be Δ_c and therefore the long-range order to be preserved up to this distance (Fig. 6). An appropriate window around the nonoscillating value of P_0 can be defined to fine tune the long-range order distance.

As stacking disorder increases, Δ_c should decrease. Figure 7 shows the loss of long-range order with increasing diffraction peak width at half maximum (FWHM) for a simulated pattern of a fcc structure with deformation fault disorder.

The P_s function can also be used to obtain the different average environments of the atoms in the structure and their probability of occurrence, which can be further used in calculation of other physical properties.

V. Gd_2Co_{17} LAYER STRUCTURE

R_2Co_{17} (R: rare earth) compounds have been studied intensively because of their favorable magnetic properties for permanent magnets applications (see, e.g., a review article by Strnat[17] and the papers cited therein). R_2Co_{17} alloys can be found in two crystallographic modifications, one described by a rhombohedral crystal system (Th_2Zn_{17}-type structure) and the other by an hexagonal crystal system (Th_2Ni_{17}-type structure).[18] Both structures differ in being different stacking sequences of identical layers.[19] The layer can be considered to be formed by two planes of atoms, a Co_9 bidimensional plane and a mixed R_2Co_8 plane. In the mixed plane a pair of Co atoms lie above and below the plane, which are usually referred to as the dumbbell site. The rhombohedral stacking order corresponds to a sequence $ABCABCABC\ldots$, while the hexagonal sequence corresponds to a $ABABAB\ldots$ stacking order (Fig. 8). The minimum lateral displacement vector between the layers is the same as in the fcc structure described by Eq. (12) (see Fig. 9).

It has been argued that fault and polytypes in the R_2Co_{17} compounds should be an important factor in the magnetic behavior of the alloy, especially on the magnetic anisotropy and magnetostriction.[19]

As we go from the lighter rare earth to the heavier ones, the structure at room temperature changes from rhombohedral to hexagonal, for the intermediate rare earth atoms heavy planar faulting has been observed.[20]

In order to quantify the occurrence of planar faulting in Gd_2Co_{17}, which lies at the intermediate range of the rare earth series, high resolution x-ray diffraction measurement were carried out at the LNLS synchrotron facility in Campinas, Brazil. The sample was obtained from 99.9 wt % purity starting material, which were melted several times to achieve homogeneity and further annealed at 1273 K for 2 weeks.

Diffraction patterns were recorded with a step size of 0.0066°, and the rhombohedral structure was indexed from the pattern. Reflections with $h-k\neq 3N$ showed broadening which we associate with planar faulting. Figure 10 shows the diffraction pattern around the (300) and (024) peaks. While the (300) reflections have a FWHM value of 0.06° the (024) reflection showed a FWHM value of 0.14°, which further affirms that the broadening seems to be associated with planar faulting. The diffraction pattern was deconvoluted for instrumental broadening using Si standard powder and converted from 2θ values to l values using the lattice parameters of Gd_2Co_{17}. The Gd_2Co_{17} lattice parameters were calculated

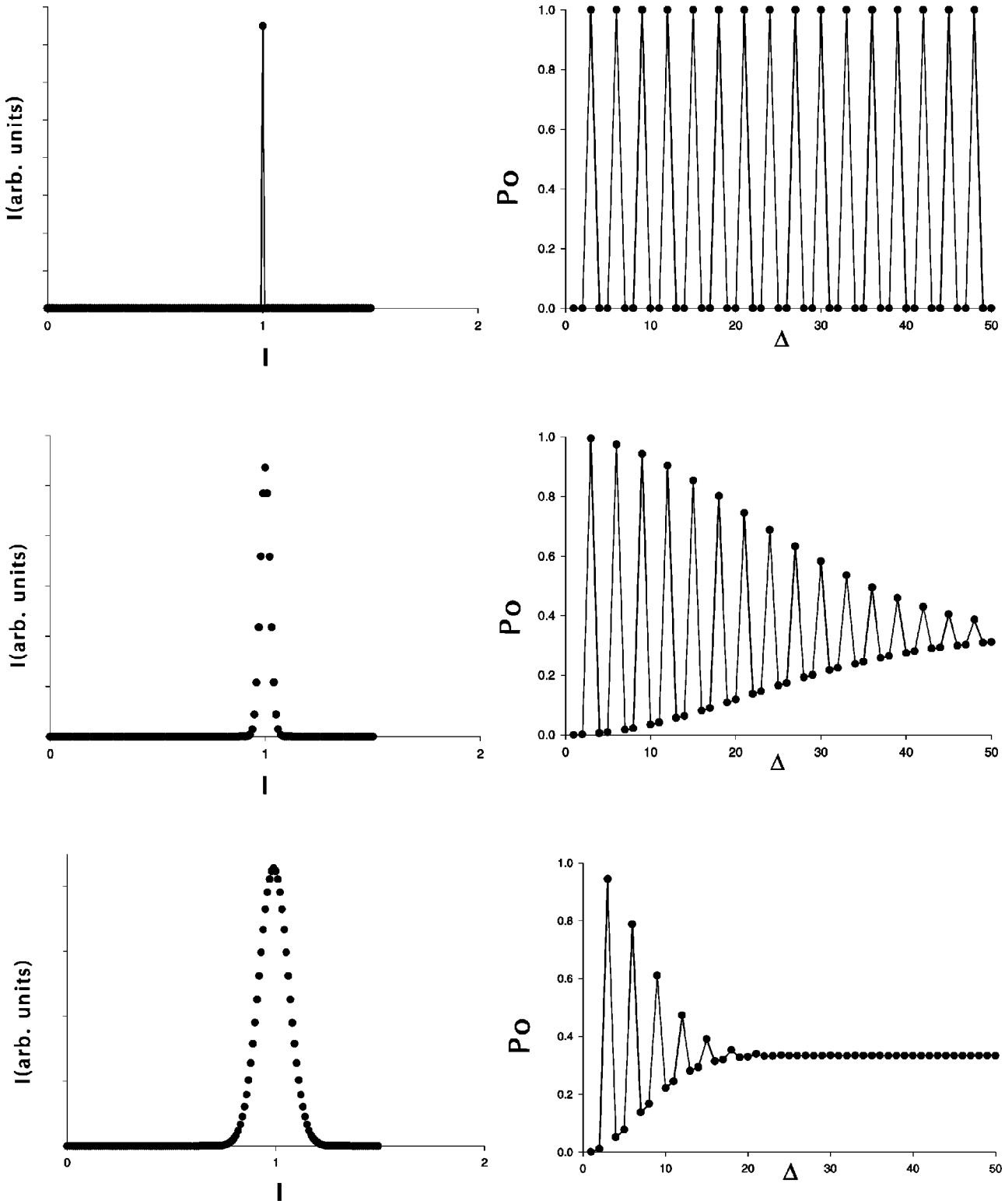

FIG. 7. Loss of long-range order, described by the $P_0(\Delta)$ distribution function, with increasing diffraction peak broadening for a simulated fcc crystal with deformation fault disorder.

from the diffraction pattern using a least square fit with an internal standard.[21] The interval in l values that resulted was 0.0009. From the diffraction pattern, the interference function was determined and further normalized. The P_0 function was determined from Eqs. (11) and (13). Linear fit for background subtraction was used. The obtained P_0 correlation function is shown in Fig. 11.

As expected the P_0 correlation function decreases with increasing delta value. The Δ_c value was determined to be 77 layers. No shift of the (024) peak could be determined within

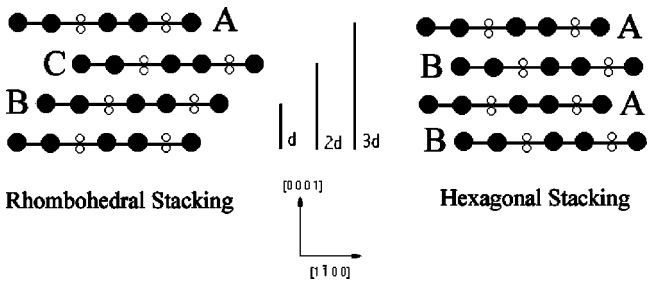

FIG. 8. The two stacking sequences in R_2Co_{17} alloys, which leads to an hexagonal and rhombohedral crystal system.

the precision of the measurement. The form of the P_0 correlation found does not resemble neither the α-faulted or the β-faulted correlation function as seen in Figs. 4 and 5. The Gd_2Co_{17} structure is intermediate between the rhombohedral and the hexagonal crystal structure; random individual planar faults as those described by α and β should not be the kind of faulting occurring in this alloy. Random individual faulting does not lead to a transformation between the two ordered layer stacking sequence. The P_0 correlation function determined shows modulation, which we think is an indication of complex planar faulting with the occurrence of combined stacking faults. This is a case where simpler analysis based on prior assumed models like those of α and β should fail.

The quality of the obtained P_0 function as shown in Fig. 11 can be seen as a result of the special care taken in the gathering and processing of the diffraction data. Usual consideration when using the Fourier transform in diffraction studies is valid for the proposed treatment. Special care has to be taken to include in the analysis of the reflections the long tails of the peak profile, in the Gd_2Co_{17} sample, and the well resolved character of the (024) reflection makes this treatment easier. Noise in the signal also affects the higher harmonics in the Fourier transform, which applies equally to

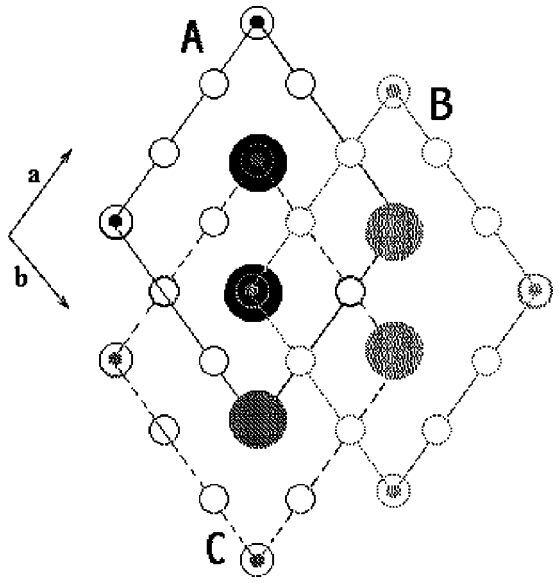

FIG. 9. Possible lateral positions of the mixed layers in R_2Co_{17} alloys.

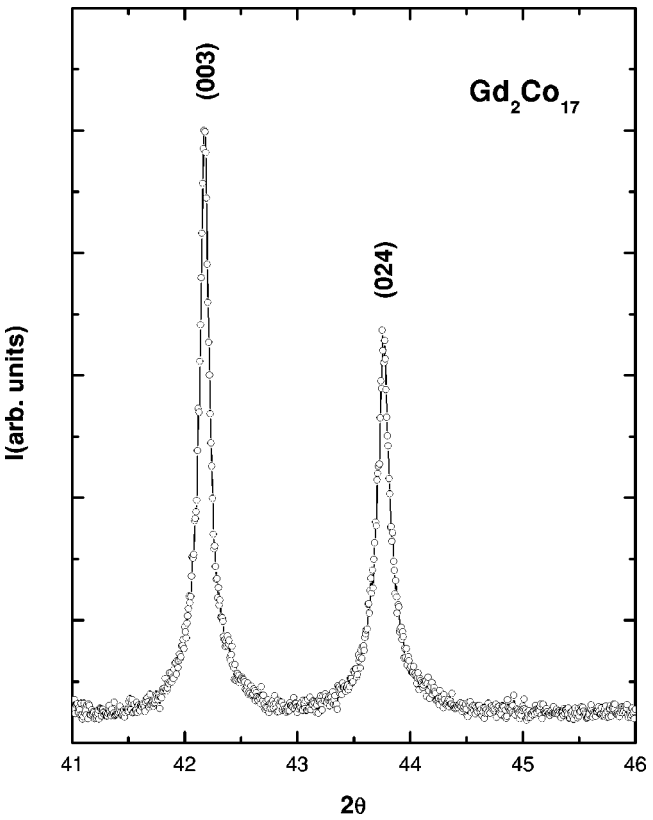

FIG. 10. Diffraction pattern of Gd_2Co_{17}.

our case; the high resolution and high signal-noise ratio was assured with the use of the synchrotron radiation. Fluctuation of the P_0 values are negligible until $\Delta = 90$ layers.

VI. CONCLUSION

A direct formalism for the solution of the diffraction pattern from the faulted layer crystal has been derived. The solution avoids the need for specific planar faulting models. The diffraction intensity from a powder sample was found to be the trigonometric series of a linear function of the $P_s(\Delta)$,

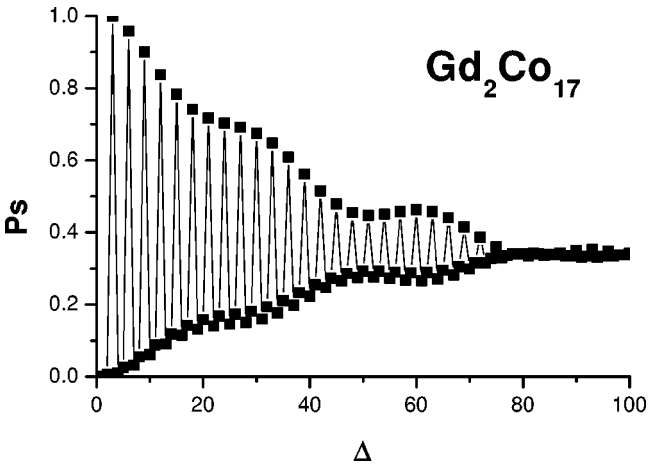

FIG. 11. P_0 correlation function of the Gd_2Co_{17} sample derived from the cosine transform of the diffraction pattern of Fig. 10.

the probability of finding two layers Δ layers apart and laterally displaced from each other a vector $s\mathbf{r}_{w\Delta}$. $P_s(\Delta)$ can be calculated trough the cosine transform of the diffracted intensity function. Crystals larger than 500 layers in height were found to behave like effective ''infinite'' crystals. The solution was successfully compared to a Monte Carlo trial and error method for the fcc structure. Long-range order and correlation can be easily derived from the P_s function. A Δ_c value can be defined above which all correlations between Δ pairs is lost. Relation between the usual parameters, α and β, and Δ_c can be found as well.

The formalism derived here was applied to the Gd_2Co_{17} structure where planar faulting was expected to occur. The delta value beyond which correlation between layers is lost was found to be 77 layers. The P_s function obtained showed negligible fluctuations up to $\Delta = 90$ layers. The kind of faulting that should be expected in this structure should not be described in terms of simple random individual faulting, yet our method can derive reliable structural information from the diffraction pattern.

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