PHYSICAL REVIEW B **68**, 064111 (2003)

Diffraction patterns of stacked layer crystals

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(Received 23 September 2002; revised manuscript received 22 January 2003; published 26 August 2003)

The relation between diffraction and planar faulting is studied. The layer displacement probability correlation function is shown to be related to the Fourier coefficients of the decomposed diffraction pattern. Peak displacement can be considered as a consequence of the departure of the faulted structure from the original periodicity, while peak broadening is associated with the loss of correlation. Several definitions of distance are introduced to compare stacking sequence and measure their degree of randomness. A run-length encoding procedure is considered, well suited for the identification of mixed polytypes and as a measure of disorder. The problem of identification of faulting complexes is discussed in terms of the pair correlation function of the binary sequence representing the layer stacks.

DOI: 10.1103/PhysRevB.68.064111 PACS number(s): 61.72.Nn, 61.72.Dd, 61.10.Nz, 61.72.Bb

I. INTRODUCTION

Several theories and approaches have been put forward to characterize by x-ray diffraction the occurrence of polytypes and planar disorder. Early works by Landau[1] and Lifschitz[2] were perhaps the first attempts to give an account of the influence of planar disorder in the x-ray diffraction pattern. Works by Warren,[3] Hendricks and Teller,[4] Wilson,[5] Jagodzinski,[6] Dornberger-Schiff, and Farkas-Jahnke,[7,8] Lele[9] and Cowley *et al.*[10] among others, further developed the theory. Other significant references can be found in Welberry.[11] With the advent of faster computers, Monte Carlo methods and other computer procedures have been devised to simulate diffraction patterns of more complex structures with more complex faulting. Example of such approaches can be found in the work of Berliner and co-workers,[12–14] the software DIFFAX developed by Treacy *et al.*,[15] and the more recent works of Weiss and Capkova[16] and Ustinov.[17]

The most simple layer structure is the so called close packed structure where two layers cannot occur one over the other with no lateral displacement. The most simple stacking defect one can imagine is the random missing of one layer in a close packed structure, or the occurrence of an additional layer in the otherwise perfect sequence. Such defects have historically been called deformation faults. Twin or growth faults, on the other hand, are random reversions of the ordering sequence within the stack, while order and periodicity are maintained inside each block. When a high density of planar faulting occurs in the material, random noninteracting deformations or twin faults are too simple models to describe the occurring disorder.

In spite of the large amount of work in planar faulting accumulated over the years, the majority of the existing methods are rather limited in their applicability, and are of an indirect nature. Most of the approaches are valid only for simple structures and noninteracting defects and therefore fail to describe correctly the occurrence of heavy planar faulting. Heavy disorder is precisely the type expected to be found in a rearrangement of a layer sequence during a phase transformation between two polytpes or in structures with almost energetically equivalent polytypes. Even more ‘‘realistic’’ approaches such as Monte Carlo simulations depend on the ingenuity of the researcher to propose beforehand a planar faulting model for their simulation.

Usually all of these approaches rely on what Varn *et al.*[18] call the fault model, which considers an underlying perfect stacking sequence perturbed by several types of stacking disorder described by their probability of occurrence. In the case of heavy disorder it does not seem correct, from a physical point of view, to assume a particular stacking sequence as the underlying perfect sequence. At least in the case of structures with different coexisting polytypes this assumption is incorrect. This should also be the case when analyzing the phase transition between different polytypes belonging to the same polytypic family. During the transition the structure can be in a state made up of several stacking sequences where none predominates.

There have been more direct approaches to find a solution to the quantitative characterization of layer ordering from the diffraction pattern of a layered structure. Attempts were made by Zachariasen[19] and by Farkas-Jahnke[8] for the particular case of ZnS and related structures. Recently, another procedure which makes use use of a so called ϵ machine has been proposed.[18]

A direct solution of the diffraction pattern from a crystal structure with planar faulting has been reported by the authors.[20] The use of the term ‘‘direct solution’’ must be understood in the sense that quantitative information on layer ordering is derived directly from the kinematical equations of diffraction, avoiding the need of any prior assumption about the kind of stacking disorder occurring in the crystal and making no use of particular models of faulting with associated faulting probabilities. The solution gives the correlation functions describing the probability of finding two layers, Δ layers apart, and displaced with respect to each other by a given vector. In this sense the formalism is of general use for any layer structure as long as it is considered that the planar faulting does not alter the interlayer distance. The practical aspects of this direct method of analysis were fur-

0163-1829/2003/68(6)/064111(12)/$20.00

ther explored in Ref. 21, where a least-square procedure was described for dealing with the experimental diffraction patterns in order to obtain the correlation functions.

The fact that the solution is derived directly from the diffraction pattern permits the study, in a general framework, of the relation between the different features of the diffraction pattern and the disorder of the stacking sequence. As the correlation function contains the maximum information from a diffraction pattern, we should explore that information regarding the appearance of different polytypes and disorder.

In this work we will further study the relation between planar faulting and diffraction. The physical interpretation of the Fourier coefficients of the diffraction pattern will be studied. We will define distance measures between stacking sequences which will allow us to compare different stacking orderings and find measures of departure from periodicity. A run length encoding procedure will be described for the identification of polytypes and as a measure of the stacking sequence complexity. Finally, it will be shown how simple parameters characterizing the probability of occurrence of different types of faulting can be derived.

II. DIRECT SOLUTION OF THE CORRELATION FUNCTION FROM THE DIFFRACTION PATTERN OF A LAYERED STRUCTURE

In an earlier paper[20] it was found that the powder diffraction pattern of a layered crystal can be decomposed in a cosine series where the coefficients could be linearly related to the so called correlation probability function. We will start in this section by making a more general deduction for any diffraction pattern within the kinematical approximation, and prove that, in the general case, the diffraction pattern can be considered a Fourier series where the coefficients can have a direct physical interpretation. The relation with other approaches will be considered. Once we have derived the general relations we will then, in Sec. III, focus on the interpretation of the Fourier coefficients and the relation between the diffraction pattern features, the planar disorder and the Fourier coefficients.

Layer structures can be considered to be built up by translational equivalent layers. Every layer has associated the same bidimensional lattice $\boldsymbol{r}_{uv}=u\boldsymbol{a}+v\boldsymbol{b}$ where u and v are integers and $(\boldsymbol{a},\boldsymbol{b})$ define a primitive cell for the bidimensional lattice. A three-dimensional lattice is then build up by stacking the two-dimensional layers in a prescribed sequence along a third direction.

Any vector pointing to a lattice point in one of the bidimensional layers can be decomposed in two vectors, a vector $\boldsymbol{R}_w$ that goes from the origin of object space to the origin of the w layer in the crystal and a vector $\boldsymbol{r}_{uv}$ which points from the origin of the bidimensional layer to the corresponding lattice point (Fig. 1). Let $\mathcal{L}(\mathbf{r})$ represent the layer crystal of size $N_a\times N_b\times N_c$; then

$$\mathcal{L}(\boldsymbol{r})=\rho_m(\boldsymbol{r})\otimes\left(\sum_{u=-N_a/2}^{N_a/2}\sum_{v=-N_b/2}^{N_b/2}\delta(\boldsymbol{r}-\boldsymbol{r}_{uv})\right)\times\sum_{w=0}^{N_c-1}\delta(\boldsymbol{R}-\boldsymbol{R}_w),\tag{1}$$

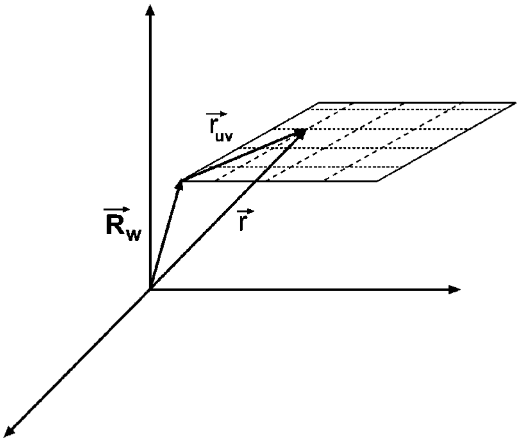

FIG. 1. Schematic representation of the bidimensional lattice of a layer with the lattice origin at $\boldsymbol{R}_w$. A object space vector $\boldsymbol{r}$, pointing to the (u,v) node in the layer, can be decomposed as the sum of the vector pointing to the layer lattice origin $\boldsymbol{R}_w$, and a vector contained in the layer $\boldsymbol{r}_{uv}$.

where $\otimes$ represents the convolution operator, δ represent the Dirac delta, and $\rho(\boldsymbol{r})$ the scatterer density.

The amplitude of a diffracted wave will be the Fourier transform of the density of scatterer in object space.[23] If $F_m(\boldsymbol{r}^*)$ is the Fourier transform of the scatterer density $\rho_m(\boldsymbol{r})$, the corresponding amplitude will be proportional to

$$F(\boldsymbol{r}^*)\propto F_m(\boldsymbol{r}^*)\left(\sum_{u=-N_a/2}^{N_a/2}\sum_{v=-N_b/2}^{N_b/2}\exp(2\pi i\boldsymbol{r}^*\cdot\boldsymbol{r}_{uv})\right)\times\sum_{w=0}^{N_c-1}\exp(2\pi i\boldsymbol{r}^*\cdot\boldsymbol{R}_w).\tag{2}$$

The corresponding diffracted intensity for N_a, $N_b\gg 1$ will then be proportional to

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

$\boldsymbol{r}^*=h\boldsymbol{a}^*+k\boldsymbol{b}^*+l\boldsymbol{c}^*$ being the reciprocal space vector.

Expression (3) is equivalent to Eq. (4) found in Ref. 12. The term inside the curly brackets in Eq. (3) is the contribution to the intensity of the layer arrangement and therefore it is the term we are interested in:

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

$\mathcal{Q}(\boldsymbol{r}^*)$ can be considered a interference function measurable from the intensity profiles of the diffraction pattern as explained in Refs. 20 and 21.

If we now take the origin of object space in a node of any of the layers, the vector $\boldsymbol{R}_w$ can be decomposed as the sum of two vectors, one along the stacking direction plus a vector parallel to the layer lattice,

$$R_w = s_w \boldsymbol{r}_{ab} + w\boldsymbol{c}, \tag{5}$$

for layer structures with constant vector displacement, s_w is an integer,[22] $\boldsymbol{r}_{ab}$ is the displacement vector parallel to the layer lattice, and $\boldsymbol{c}$ is a vector along the stacking direction with a length equal to the interlayer spacing. The stacking sequence will now be given by the sequence of s_w values corresponding to each w layer. If there are M possible s values then $M\boldsymbol{r}_{ab}$ will be a lattice vector of the layer, and Eq. (4) can be written as

$$\mathcal{Q}(\boldsymbol{r}^*) = 1 + \frac{2}{N_c} \sum_{\Delta=1}^{N_c-1} \sum_{s=0}^{M-1} N_s(\Delta) \times \cos[2\pi s(hx+ky) + 2\pi\Delta l], \tag{6}$$

where we have written $\boldsymbol{r}_{ab} = x\boldsymbol{a} + y\boldsymbol{b}$, and $N_s(\Delta)$ is the number of pair of layers, Δ layers apart, and laterally displaced one with respect to the other by $s\boldsymbol{r}_{ab}$ (Δ pairs).

If we multiply and divide by $N_c - \Delta$, the total number of Δ pairs, then we can write Eq. (6) in terms of the probability of finding a Δ pair with lateral displacement $s\boldsymbol{r}_{ab}$. Let us call such a probability $P_s(\Delta)$,

$$\mathcal{Q}(\boldsymbol{r}^*) = 1 + 2\sum_{\Delta=1}^{N_c-1} \left(1 - \frac{\Delta}{N_c}\right) \sum_{s=0}^{M-1} P_s(\Delta) \times \cos[2\pi s(hx+ky) + 2\pi\Delta l], \tag{7}$$

which can be rewritten as

$$\mathcal{Q}(\boldsymbol{r}^*) = 1 + 2\sum_{\Delta=1}^{N_c-1} A_\Delta \cos(2\pi\Delta l) + B_\Delta \sin(2\pi\Delta l), \tag{8}$$

where

$$A_\Delta = \left(1 - \frac{\Delta}{N_c}\right) \sum_{s=0}^{M-1} P_s(\Delta) \cos[2\pi s(hx+ky)] \tag{9}$$

$$B_\Delta = \left(1 - \frac{\Delta}{N_c}\right) \sum_{s=1}^{M-1} P_s(\Delta) \sin[2\pi s(hx+ky)]. \tag{10}$$

Equation (8) expresses $\mathcal{Q}(\boldsymbol{r}^*)$ as a Fourier series, from where

$$A_\Delta = \int_{-1/2}^{1/2} \mathcal{Q}(\boldsymbol{r}^*) \cos(2\pi\Delta l)\, dl, \qquad \Delta = 1,2,\ldots, \tag{11}$$

$$B_\Delta = \int_{-1/2}^{1/2} \mathcal{Q}(\boldsymbol{r}^*) \sin(2\pi\Delta l)\, dl, \qquad \Delta = 1,2,\ldots. \tag{12}$$

As explained in Ref. 20, in the case of a random powder sample, the probability $P_s(\Delta)$ will be equal to the probability $P_{M-s}(\Delta)$. This result indicates that the only difference between a $s\boldsymbol{r}_{ab}$ displacement in one grain and a $-s\boldsymbol{r}_{ab} = M - s\boldsymbol{r}_{ab}$ displacement in another grain is a rotation of π along an axis parallel to $\boldsymbol{c}$. Then for a random powder sample the B_Δ coefficients vanish and Eq. (8) reduces to

$$\mathcal{Q}(\boldsymbol{r}^*) = 1 + 2\sum_{\Delta=1}^{N_c-1} A_\Delta \cos(2\pi\Delta l), \tag{13}$$

with

$$A_\Delta = \left(1 - \frac{\Delta}{N_c}\right) \left\{ P_0(\Delta) + 2\sum_{s=1}^{(M-1)/2} P_s(\Delta) \times \cos[2\pi s(hx+ky)] \right\}, \quad M \text{odd} \tag{14}$$

$$A_\Delta = \left(1 - \frac{\Delta}{N_c}\right) \left\{ [P_0(\Delta) + P_{M/2}(\Delta)] + 2\sum_{s=1}^{(M/2)-1} P_s(\Delta) \cos[2\pi s(hx+ky)] \right\}, \quad M \text{even}. \tag{15}$$

The collapse of Eq. (7) to Eq. (13) for a powder sample can also be viewed as a result of $\mathcal{Q}(\boldsymbol{r}^*)$ being an even function of the reciprocal variable l. Conversely, in the case of a single crystal, an even function $\mathcal{Q}(\boldsymbol{r}^*)$ of the reciprocal variable l implies $P_s(\Delta) = P_{-s}(\Delta)$. Equation (14) reduces to expression (10) in Ref. 20 in the limit of an infinite crystal.

In the deduction of this result no assumption of a particular sequence of layers was necessary and therefore, the result is valid for any sequence and any density of stacking faults present on the layer arrangement. In the same sense as Varn *et al.*,[18] the formalism developed has to be considered an approach ‘‘beyond the fault model’’ as no underlying perfect sequence had to be defined.

Relation to other approaches

Another direct approach was that of Zachariasen.[19] The A_Δ coefficients in the present work can be seen to be equivalent to the $W^M_{H_1H_2}$ coefficients in the Zachariasen approach, yet he failed to consider the B_Δ coefficients and the conditions for nonaffected reflections are incorrectly stated.

In another formalism, use is made of the so called average phase factor.[17,24] If we start by writing the $\mathcal{Q}$ function from a layer structure as

$$\mathcal{Q}(\boldsymbol{r}^*) = \frac{1}{N_c} \sum_{w} \sum_{w'} e^{i\phi(\Delta)} e^{2\pi i \Delta l}, \tag{16}$$

where w and w' goes through all layers in the stack and $\Delta = w - w'$, $\phi(\Delta) = 2\pi(s_w - s_{w'})\boldsymbol{r}^* \cdot \boldsymbol{r}_{ab}$. Making the average of all Δ neighbor pairs, $\langle e^{i\phi(\Delta)} \rangle$, the double sum in Eq. (16) can be reduced to a single sum:

$$\mathcal{Q}(\boldsymbol{r}^*) = \frac{1}{N_c} \sum_{\Delta=-(N_c-1)}^{N_c-1} (N_c - |\Delta|) \langle e^{i\phi(\Delta)} \rangle e^{2\pi i \Delta l}. \tag{17}$$

Noting from the definition that $\phi(-\Delta)=-\phi(\Delta)$ then $\langle e^{i\phi(-\Delta)}\rangle=\langle e^{-i\phi(\Delta)}\rangle=\langle e^{i\phi(\Delta)}\rangle^*$ where * denotes complex conjugate. Equation (17) can now be rewritten as

$$\mathcal{Q}(\boldsymbol{r^*})=1+2\sum_{\Delta=1}^{N_c-1}\left(1-\frac{\Delta}{N_c}\right)\{\mathrm{Re}[\langle e^{i\phi(\Delta)}\rangle]\cos(2\pi\Delta l)-\mathrm{Im}[\langle e^{i\phi(\Delta)}\rangle]\sin(2\pi\Delta l)\} \tag{18}$$

where Re and Im stands for real part and imaginary part, respectively. A Comparison of Eq. (18) with Eq. (8) gives us

$$A_\Delta=\left(1-\frac{\Delta}{N_c}\right)\mathrm{Re}[\langle e^{i\phi(\Delta)}\rangle], \tag{19}$$

$$B_\Delta=\left(1-\frac{\Delta}{N_c}\right)\mathrm{Im}[\langle e^{i\phi(\Delta)}\rangle]. \tag{20}$$

A_Δ is therefore related to the real part of the average phase factor $\langle e^{i\phi(\Delta)}\rangle$, while B_Δ is proportional to the imaginary part. The imaginary part of the average phase factor for a powder sample vanishes.

Farkas-Jahnke and Dornberger-Schiff[8] used a direct approach for a polytype analysis which can be related to the one presented here. Their analysis was for the particular cases of ZnS, SiC, and similar structures, and therefore the above deduction can be considered in some sense a generalization of their results. An equivalent approach using the so called unitary structure factor was independently introduced by Tokonami.[25]

The unitary structure factor is defined by writing the structure factor of the unit cell in a perfect periodic layer crystal as

$$F(\boldsymbol{r^*})=F_0(\boldsymbol{r^*})S(\boldsymbol{r^*}), \tag{21}$$

where $F_0(\boldsymbol{r^*})$ denotes the structure factor of a structure having the same unit cell as the real structure but the scattering density distribution of only a single layer, and $S(\boldsymbol{r^*})$ is the unitary structure factor which is the Fourier transform of a periodic function with the same unit cell, characterizing the stacking of the layers.

Starting from Eq. (2) and writing $\boldsymbol{R}_w$ as in Eq. (5), we get

$$F(\boldsymbol{r^*})\propto F_m(\boldsymbol{r^*})\sum_{w=0}^{N_c-1}\exp[2\pi i s_w(hx+ky)+2\pi iwl], \tag{22}$$

where we have made use of $r_{ab}=x\boldsymbol{a}+y\boldsymbol{b}$ and $\boldsymbol{r^*}=h\boldsymbol{a^*}+k\boldsymbol{b^*}+l\boldsymbol{c^*}$. Due to the periodicity of the perfect structure $s_w=s_{w+D}$, where D is the number of layers building up a unit cell, and Eq. (22) will then be

$$F(\boldsymbol{r^*})\propto\frac{N_c}{D}F_m(\boldsymbol{r^*})\sum_{w=0}^{D-1}\exp[2\pi i s_w(hx+ky)+2\pi iwl] \tag{23}$$

for Dl of an integer value. Comparing Eq. (23) with Eq. (21),

$$F_0(\boldsymbol{r^*})=\frac{N_c}{D}F_m(\boldsymbol{r^*}) \tag{24}$$

and

$$S(\boldsymbol{r^*})=\sum_{w=0}^{D-1}\exp[2\pi i s_w(hx+ky)+2\pi iwl]. \tag{25}$$

Equation (25) is a straightforward generalization of the unit structure factor used by Dornberger-Schiff and Farkas-Jahnke.[7] From the above equation it also follows that

$$\mathcal{Q}(\boldsymbol{r^*})\propto S^2(\boldsymbol{r^*}). \tag{26}$$

III. FOURIER COEFFICIENTS

Equations (11) and (12) are central to the faulting problem considered here, which shows that the A_Δ and B_Δ coefficients can be obtained from the experimental diffraction data. The peak broadening and peak shift will have a direct influence over the values of both A_Δ and B_Δ.

On the other hand, in Eq. (8), A_Δ and B_Δ play similar roles as coefficients in a Fourier expansion. In what follows of this section we will make our analysis for the A_Δ coefficient and results will be also generally valid for the B_Δ coefficients.

In the limit of an infinite crystal we can write

$$A_\Delta=\sum_{s=0}^{M-1}P_s(\Delta)\cos[2\pi s(hx+ky)]. \tag{27}$$

If we determine A_Δ for as much (h,k) as unknown $P_s(\Delta)$ are, then Eq. (27) defines a linear set of equations solvable for $P_s(\Delta)$. $P_s(\Delta)$ are then obtainable from the experimental data through the A_Δ and B_Δ coefficients. From Eq. (27) it is clear that for $hx+ky=n$, where n is an integer value, A_Δ will not depend on the particular stacking arrangement, that is, the corresponding reflection in the x-ray diffraction pattern will not be affected by planar disorder.

Two types of stacks can be considered. In one case there is no restriction on the layer ordering and for the extreme case of a complete random sequence we will have $A_\Delta=0$ for all Δ values, $\mathcal{Q}(\boldsymbol{r^*})$ will be constant along a l row. All $P_s(\Delta)$ will have a value $1/M$, M being the total number of possible lateral displacements.

A second type of stacking is the close-packed structure where two adjacent layers cannot be found without a lateral displacement. The best known close-packed crystals are those with three possible displacements of the layers, usually described by the letters *A*, *B*, and *C*. One of the letters stands for a zero displacement, another for $\frac{1}{3}\boldsymbol{a}-\frac{1}{3}\boldsymbol{b}$, and the third one for $2(\frac{1}{3}\boldsymbol{a}-\frac{1}{3}\boldsymbol{b})$. Which letter corresponds to which displacement is irrelevant as long as consistency is kept. The close-packed condition will then mean the impossibility of finding the same letter consecutive times in the stacking sequence. In the case of a close-packed structure,

$$A_\Delta=P_0(\Delta)+[1-P_0(\Delta)]\cos\left[\frac{2\pi}{3}(h-k)\right] \tag{28}$$

and those reflections with $h-k=3n$ will not depend on the stacking sequence.

Here we are constrained by the fact that two nondisplaced layers cannot be found one over the other, which leads to the condition $P_0(1)=0$. In such a case, for a random sequence,

$$A_\Delta=\begin{cases}\cos\left[\frac{2}{3}\pi(h-k)\right], & \Delta=1\\ 1/3\left\{1+2\cos\left[\frac{2}{3}\pi(h-k)\right]\right\}, & \Delta\neq 1.\end{cases}\tag{29}$$

A_Δ will be constant for $\Delta>1$. For a close-packed structure with random sequence the $\mathcal{Q}(\boldsymbol{r}^{*})$ function for a faulted affected l row $(h-k\neq 3n)$ will be

$$\mathcal{Q}(l)=1-\left(1-\frac{1}{N_c-1}\right)\cos(2\pi l).\tag{30}$$

The peak profiles will be a cosine function with maximum at l integer values and a full width at half maximum value of $\frac{1}{2}$. The cosine profile for the random sequence given by Eq. (30) is a consequence of the nearest neighbor order resulting from the close-packed condition.

In the other limit of a perfect periodic crystal $P_s(\Delta)$ will have the value 1 for Δ a integer number of times the periodicity of the crystal along the stacking direction. The $P_s(\Delta)$ functions will be a periodic function with repeating length, Δ_p, the period of the crystal along the stacking direction, correspondingly the A_Δ will exhibit the same periodic behavior. In the case of a perfectly periodic close packed structures, the $P_0(\Delta)$ functions will also be constrain by the condition $P_0(\Delta_p-1)=0$.

For close packed structures Eq. (8) together with definition of the Fourier coefficients (9) and (10) will lead to the extinction condition for forbidden reflections l_0,

$$l_o=\text{noninteger},\quad h-k=3n,$$

$$\sum_{\Delta=1}^{\Delta_p}\{\sqrt{3}[P_1(\Delta)-P_2(\Delta)]\sin(2\pi\Delta l_o)+[3P_0(\Delta)-1]\cos(2\pi\Delta l_o)\}=0,\quad h-k\neq 3n.$$

Influence of layer disorder on the peak profile and displacement

A partly disordered crystal will be an intermediate case between the random sequence and a perfect periodic one. For small values of Δ, A_Δ will be almost equal to the values for the perfect periodic structure, but will tend to the value of the random sequence as the Δ value increases. This behavior in a partly disordered sequence is the result of a loss of correlation between the layer displacement for large values of Δ. We can define a layer correlation length Δ_c, the characteristic length of the system above which correlation between layers can be considered lost.[20] Knowing the stacking sequence up to Δ layers, we cannot predict in any sense how stacking sequence behaves beyond $\Delta+\Delta_c$.

Along the integration path in Eq. (11) several diffraction peaks could occur (e.g. in the close-packed rhombohedral structure, for $h-k\neq 3n$, two diffraction maxima occur), and the equation can be written as a sum of integrals over each diffraction peak profile,

$$A_\Delta=\sum_i\int_{-1/2}^{1/2}\mathcal{V}_i(l-l_{o_i})\cos(2\pi\Delta l)\,dl=\sum_i{}_iA_\Delta,\tag{31}$$

where $\mathcal{V}_i$ is the peak profile for the *i* reflection and l_{o_i} is the position of the maximum. Each integral in Eq. (31) can be written as

$${}_iA_\Delta=\cos(2\pi\Delta l_{o_i})\int_{-(1/2)-l_{o_i}}^{(1/2)-l_{o_i}}\mathcal{V}_i(l)\cos(2\pi\Delta l)\,dl-\sin(2\pi\Delta l_{o_i})\int_{-(1/2)-l_{o_i}}^{(1/2)-l_{o_i}}\mathcal{V}_i(l)\sin(2\pi\Delta l)\,dl.\tag{32}$$

The terms in front of the integrals will determine a periodic oscillating behavior in A_Δ. If the peak position can be described as a rational number $l_{oi}=p/q$ $(q\neq 1)$ then the period of the oscillation will be *q*. The integral terms, on the other hand, will give, for a Dirac delta function, a value of 1; the contribution of this profile to the A_Δ values will then consist only of the oscillating part extending to infinity with a periodicity of *q*. This case corresponds to the perfect periodic stacking.

If the peak profile is not a delta function but instead a planar faulted broadened peak, then the integral terms will give a damped oscillating function tending to zero as Δ increases. This behavior can be understood if we look at Fig. 2, where we have plotted the integrand in the first term of Eq. (32), and notice that the integral is nothing else but the area under the curve. The absolute area will remain constant for all Δ values, but, as Fig. 2 shows, for increasing Δ values the cosine term will have an increasing number of oscillations within the peak width and therefore the negative area contribution will tend to increase or decrease at the expense of the positive area contribution while the sum will tend to zero. The contribution of one peak profile to the A_Δ coefficients can be summarized as

$${}_iA_\Delta=\sum\text{``decay term''}\times\text{``oscillating term .''}\tag{33}$$

The decay term, which depends on the peak shape and width, will be the sole element responsible for determining the correlation length Δ_c of the stacking sequence. The oscillation term, on the other hand, will be reflected in the peak shift and will further affect the behavior of the A_Δ coefficients without further loss of correlation length in the stacking sequence.

The limiting case of a delta function peak profile corresponds to a perfect periodic sequence and the A_Δ coefficients will be an oscillating periodic factor, with periodicity depending only on the peak position. The correlation length in this case will be infinite. The peak shift in this case (without

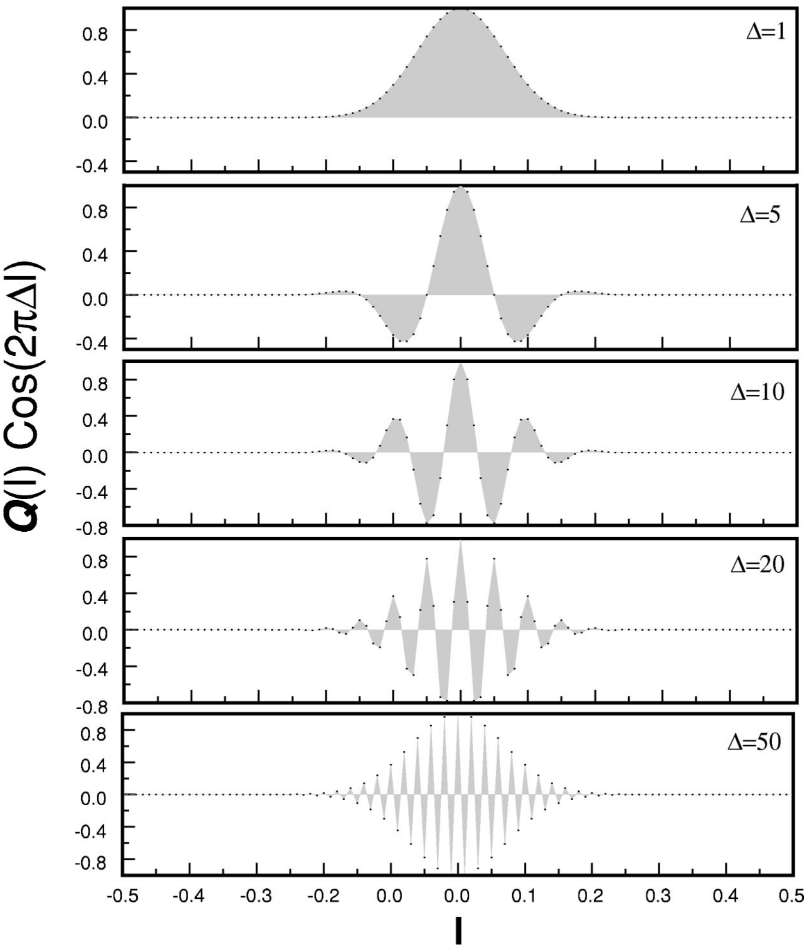

FIG. 2. Integrand of the first term of Eq. (32). The figure illustrates the fact that with increasing Δ value, the integral, which is the area under the curve, will tend to zero.

broadening) can be considered to reflect the departure of periodicity from the original sequence towards other periodic orderings.

Let us consider another limiting case, a peak profile given by a step function; for simplicity let us further assume that the function is symmetric in the integration interval and completely contained within it

$$\mathcal{Q}(l)=\begin{cases} f^{-1}, & -\dfrac{f}{2}\leqslant l\leqslant \dfrac{f}{2} \\ 0 & \text{otherwise.} \end{cases} \tag{34}$$

In this case the contribution of the peak profile to the A_Δ coefficient will be

$${}_{i}A_{\Delta}=\frac{\sin \pi\Delta f}{\pi\Delta f}. \tag{35}$$

The decaying term will be given by $1/\pi\Delta f$. When a correlation function follows a power law, the decrease of the correlation is always at the same rate and a characteristic length cannot be defined.

We can ask if there is a physically consistent peak profile which can lead to a decaying term proportional to Δ^{-n} with $n\neq 1$. It follows directly from Eq. (11) that the peak profile function following a $-l^{2p}$ $(p=1,2,3\ldots)$ law defined over its positive range will give a decaying term with the leading term following a Δ^{-2} power law regardless of the actual value of p. Both the step function and the power law peak profiles have a definite cutoff or total width value, and both give rise to a power law decaying term.

Real profiles are never cutoff functions but instead slowly decaying functions. In this case we have found that the decaying term in A_Δ will follow an exponential law for all the common profiles used in diffraction. For the Lorentz profile the decaying term follows an $\exp(-\Delta f\pi)$ law, with f being the full width at half maximum, while for the Gauss term the decaying law was found to be $\exp[-(\Delta f\pi)^2]$. Figure 3 shows a comparison of the decaying term for Lorentz, Gauss, and step profile functions. All profiles were taken with the same f value. A characteristic length can be defined as $\Delta_c=1/(f\pi)$. The power law decreasing function for the step profile decays slowly to zero with a constant decay rate. Voigt profile functions, which are usually considered better suited for describing diffraction peak profiles, are an intermediate function between the Lorentz and Gauss functions and therefore will introduce no new feature to the above analysis.

Two different characteristics can now be understood in the diffraction pattern of a planar faulted crystal. On the one hand, peak broadening is associated with the decaying term in A_Δ, and consequently with the loss of correlation in the layer stack due to the presence of random disorder. The peak shift, on the other hand, is related to the periodicity of the A_Δ

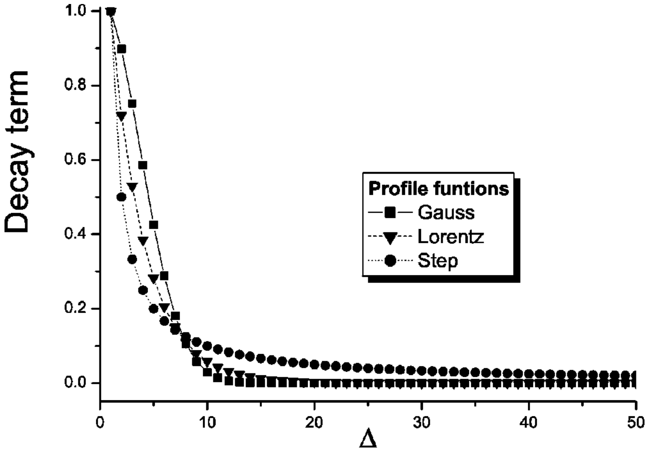

FIG. 3. Decay term for different diffraction peak profiles. The step function gives a constant decay rate characteristic of decay power laws, the other two profiles show different exponential decays, and a characteristic decay length can be defined.

coefficients $[P_s(\Delta)$ functions]. The peak shift will reflect that, as a result of the appearance of disorder, the stack of layers gradually loses its original periodicity and eventually can move nearer to another stacking arrangement.

The loss of correlation in the stack and the change of periodicity in the $P_s(\Delta)$ functions are to be considered two competing effects. It would make no sense to ascribe to a stacking sequence a periodic ordering of length larger than the correlation length. The peak shift and peak broadening do not occur independently, and while the peak shift can be thought of as a departure from the old periodic order, and therefore a possible new stacking order of the layers, the peak broadening can show that such an order is frustrated by the loss of the correlation length. The ‘‘new order’’ suggested by the peak shift is not real as long as the correlation length is smaller than the periodicity length of the ‘‘new’’ ordering. Only when the correlation length is larger than the periodicity of a possible ‘‘new periodic order’’ can we consider that such an order has emerged, and the stacking sequence, although still disordered, is now nearer to this new periodic stacking arrangement than to the ‘‘original’’ one.

A peak shift occurs as a continuous function of position, for small shifts of the peak maximum, if they can be given by a rational number, will give rise to long stacking periodicity orders which are frustrated by the underlying disorder, as discussed above. Further peak shifts can be taken as a strong indications that an increasing correlation between the faults is occurring, and that the ordering of the faulting is leading the stacking sequence to a new polytypic ordering. If disorder is introduced without further peak shifting, this can be taken as an indication that although a higher density of faulting is occurring in the crystal, this faulting is not accompanied by correlation between the faults, and the sequence is shifting towards a random distribution of layers.

The above discussion concentrated on the effect of planar disorder over one reflection in the diffraction pattern. In a complete analysis all the reflection contributions ${}_iA_\Delta$ have to be considered on the final Fourier coefficient A_Δ. If more than one peak is present in one periodic interval in l, then the periodic order is determined by the combined contribution of each peak position. The emergence of a new order, as explained above, is accompanied by the appearance of new reflections. The generalization is straightforward, yet several cases now appear for the resulting oscillating term.

If at least one of the peak positions is an irrational number, the ‘‘periodicity’’ is effectively infinite and the closer rational number which leads to a periodic length smaller than Δ_c is the one describing the underlying periodic order. If the peak positions for all peaks are rational numbers, one must still consider if the periodic length is larger or smaller than the correlation length. If the periodicity is larger than the correlation length, then the closer peak position which leads to a nonfrustrated periodic order is the one describing the underlying periodic order.

The oscillation term for the ‘‘perfect underlying structure’’ can be determined as described in the preceding paragraphs. Yet even in this situation two cases arise. One case corresponds to the existence of only one underlying polytype: the periodic correlation functions yields a stacking ordering which can be determined as explained, for the case of close packed structures, in Ref. 28. In the other case, several polytypes occur without correlation among them, in this case the periodic correlation functions calculated from all the peak contributions are not consistent with any layer arrangement of this periodic length.

An example: intermetallic rare earth-cobalt structures

$R_2\mathrm{Co}_{17}$ (R is a rare earth) alloys can be described as close packed layer structures which can be found in two crystallographic modifications, one described by a rhombohedral crystal system (for the lighter rare earths) and the other by an hexagonal crystal system (for heavier rare earths).[29] The rhombohedral stacking order corresponds to a sequence $ABCABCABC\ldots$ while the hexagonal sequence corresponds to a $ABABAB\ldots$ stacking order. The minimum lateral displacement vector between the layers is the same as in the fcc structure (for details, see Ref. 20).

Figure 4 shows the diffraction patterns corresponding to three typical rare earth compounds. For the Nd alloy the rhombohedral structure is clearly determined and indexed, while for the Lu alloy the hexagonal structure is the one present. The Dy alloy shows mixed reflections which could indicate the presence of a higher order polytype or a mixture of the hexagonal and rhombohedral stacking.

Figure 5 shows the fitted profiles for the $(02\,4/3+\delta l_1)$ for the Nd_2Co_{17} sample and the $(20\,3/2+\delta l_2)$ for the $\mathrm{Lu}_2\mathrm{Co}_{17}$ sample. Fitting was done using a least square procedure as explained in Ref. 21.

From the powder diffraction pattern of each sample the P_0 functions were determined and the correlation length calculated. Nd had a correlation length Δ_c of 134 layers, larger than the correlation length of 31 layers for Dy, and slightly smaller than Lu, which had a correlation length of 169 layers. The percent of hexagonality was also determined from each P_0 function, in this case the results where 1% for Nd, 45% for Dy, and 98% for Lu.

The diffraction pattern of the Dy alloy shows the simultaneous occurrence of two peaks at $(02\,4/3+\delta l_1)$ and

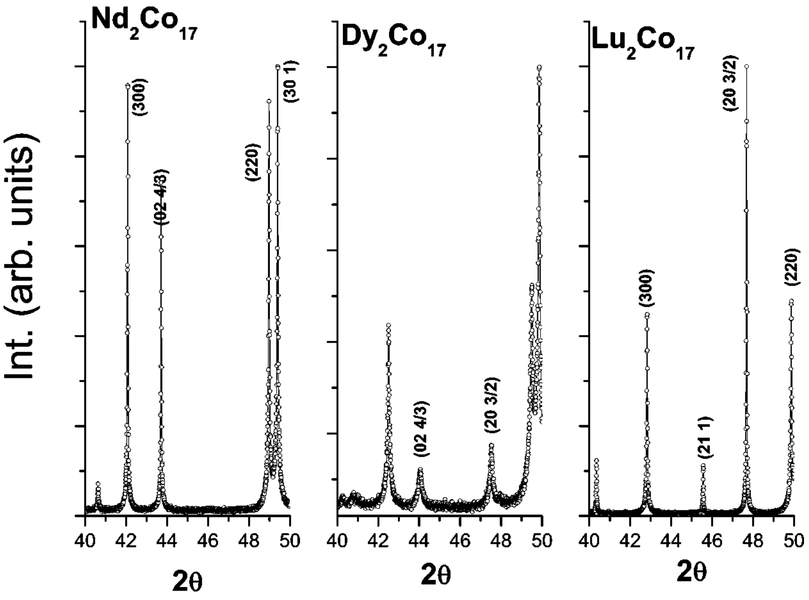

FIG. 4. Diffraction pattern of R_2Co_{17} alloys for R-Nd, Dy, and Lu. Nd and Lu diffraction peaks can be indexed as rhombohedral and hexagonal structures, respectively. The Dy diffraction pattern shows peaks belonging to both rhombohedral and hexagonal orderings.

$(20\,3/2+\delta l_2)$. The peak shifts were calculated as $\delta l_1=0.0007$ for $(02\,4/3+\delta l_1)$ and $\delta l_1=0.0009$ for $(20\,3/2+\delta l_2)$. From the peak shifts and the correlation length, the closer "ideal" underlying periodic order was found to be of periodic length 6. The corresponding combined P_0 correlation function for such ideal order is shown in Fig. 6 together with the faulted one.

The "ideal" correlation function of Fig. 6(b) is incompatible with any close packed stacking sequence of six layers repeating unit cell. The correlation function obtained can thus be explained as the mixture of two polytypes, namely the rhombohedral one, to which the $(02\,4/3+\delta l_1)$ peak belongs, and the hexagonal one, to which the $(20\,3/2+\delta l_2)$ reflection belongs. Dy alloy diffraction pattern then shows that, in this alloy, the hexagonal polytype nucleate randomly within the rhombohedral phase.

IV. QUANTITATIVE CHARACTERIZATION OF PLANAR DISORDER

A question arises as to what type of information can be derived from the probability correlation functions. It is already known that the percent of hexagonal environment can be directly determined as $P_0(2)$ while the relative cyclicity can be calculated as $P_1(1)-P_2(1)$.[26]

From the probability correlation functions representative layer sequences of length up to the correlation length could be calculated from a Markov process.[18] The possibility of obtaining such typical sequences allows the introduction of additional parameters for the characterization of disorder.

In this section, further parameters will be proposed for the characterization of disordered layered crystal, and measures of distance that could allow us to compare different sequence orders will be defined. As long as representative stacking

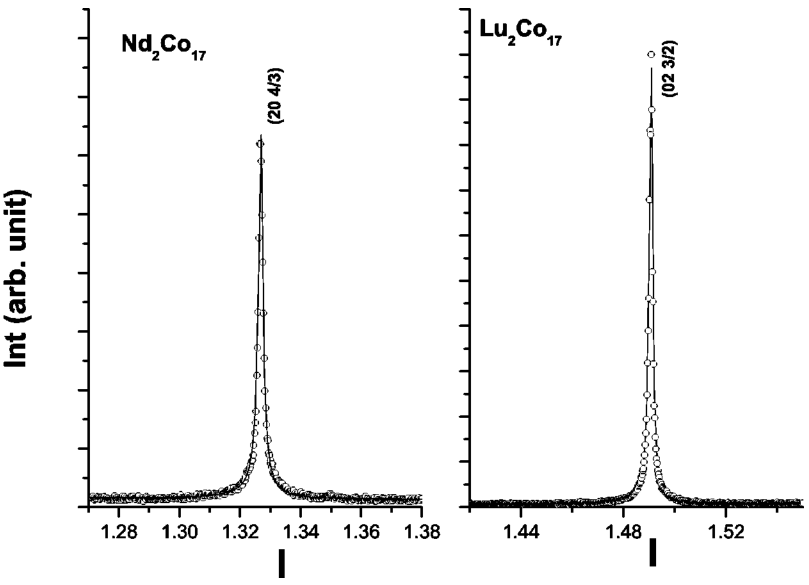

FIG. 5. Peak profile of the $(02\,4/3+\delta l_1)$ peak of the Nd_2Co_{17} sample and the $(20\,3/2+\delta l_2)$ reflection of the Lu_2Co_{17} sample. The peaks are given in terms of the reciprocal variable l. The peaks were fitted using a least square procedure as explained in Ref. 21.

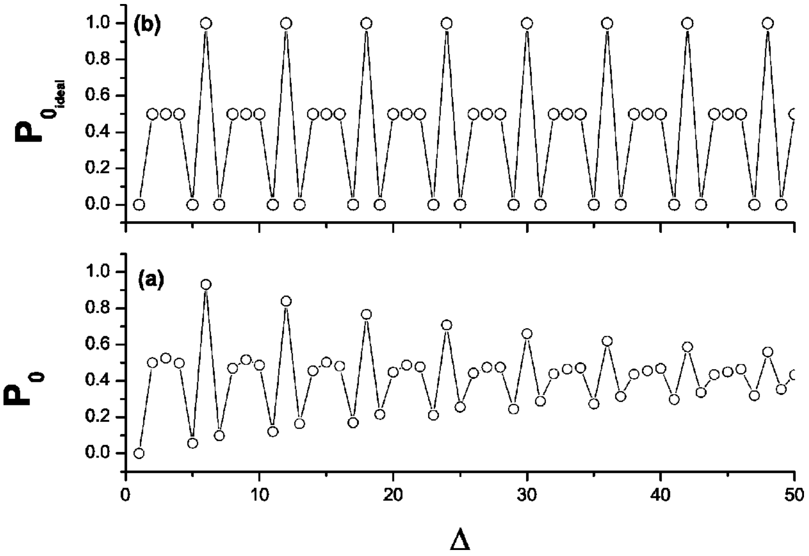

FIG. 6. (a) Calculated correlation function from the $(02\,4/3+\delta l_1)$ and the $(20\,3/2+\delta l_2)$ reflections for the Nd_2Co_{17} sample. The “ideal” underlying periodic correlation function is shown in (b).

sequences can be deduced from the correlation function, the so called Hamming distance and the run length encoding distance can be defined. The run length encoding procedure can also be used for the identification of polytypes in a sequence and as a measure of complexity or randomness of the disorder stack. Finally the identification of faulting complexes will be discussed.

For the discussion that follows it will be important to understand how a stacking sequence can be coded as string of numbers. We will limit our discussion, for the sake of clarity, to close-packed structures.

In a close-packed structure we can write any sequence as a binary string where every “1” stands for two consecutive layers with lateral displacement vector r_{ab} and 0 for a lateral displacement $-r_{ab}$. Any two symbols in the sequence $A \rightarrow B \rightarrow C \rightarrow A$ will be represented by a 1, while any two symbols in the sequence $A \rightarrow C \rightarrow B \rightarrow A$ will be represented by a 0 (e.g., a sequence $ABCABACBCACBA$ will be represented by 111100011000). This notation is equivalent to the Hagg notation[27] with “1” used instead of “+” and “0” instead of “-.” An extension to non-close-packed structures is straight forward if we consider a sequence of number in some other base larger than 2.

Measure of distance between two stacking sequences

For a straightforward definition of a “distance” measure between two stacking sequences the Δ_c correlation length can be used. It is clear that a more random sequence will have a shorter correlation length, and vice versa. The limitation with such a definition is that it will not allow a distinction between two different polytype faulted sequences as it will only measure the loss of correlation due to disorder.

It can be shown[28] that for the case of a close packed structure disordered by deformation and growth faults, the correlation length for low density of faulting can be given by

$$\Delta_c = -\frac{2}{\log(1-2\alpha+3\alpha^2-2\beta)}, \qquad (36)$$

where α and β are the deformation and twin faulting probability, respectively. Figure 7 shows the corresponding behavior of the Δ_c value with twin faulting probability. The correlation length falls rapidly for increasing faulting probability and then reduces it rate of decrease. For faulting densities beyond the applicability of Eq. (36), the correlation length should saturate as a result of the close-packed constraint. If we release the close-packed constraint, then Δ_c will decrease up to a theoretically limiting value of zero for a completely random sequence.

Correlation length gives an absolute measure of disorder, but fails to compare how close a particular disordered structure is to a predetermined perfect periodic sequence. Another disadvantage of using the correlation length as a measure of disorder is its nonlinear character, as seen in Fig. 7.

1. Hamming distance

In order to be able to compare two structures differing in their stacking order the concept of Hamming distance,[30]

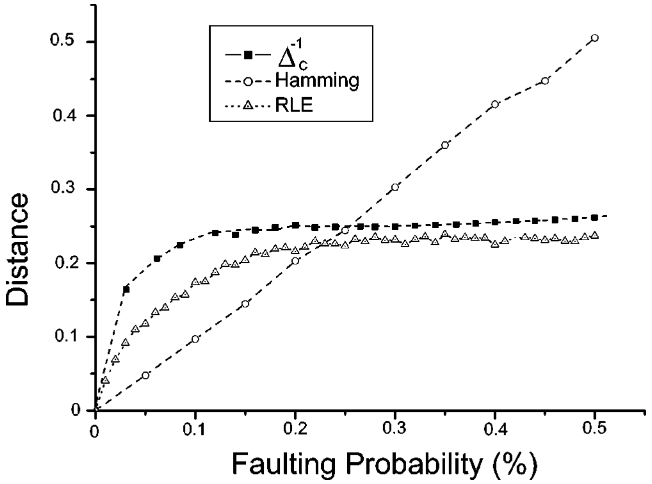

FIG. 7. Distance measures as a function of twin fault probability. The Hamming distance shows a linear behavior contrary to the correlation length distance and the RLE. See the text for details.

taken from information theory, can be used. The Hamming distance is given by the number of sites when two sequences differ from each other divided by the length of the sequence,

$$D_H(\{_j\alpha_i\})=\sum_i |_1\alpha_i - _2\alpha_i|/N, \tag{37}$$

where $_1\alpha_i$ denotes one sequence and $_2\alpha_i$ denotes the other.

In the case of the Hamming distance of a binary sequence, if a value d_h above 0.5 is obtained, then the value $1-D_h$ is taken, corresponding to the crystallographically equivalent bitwise negated sequence. The Hamming distance distinguishes between sequences belonging to different underlying polytypes, and will give a clearer distinction than the correlation length distance.

A conceptual difference between the Hamming distance and the Δ_c measure is that the latter gives an absolute measure of departure from disorder, while the Hamming distance, a relative measure, gives the mismatch between one sequence and a second sequence (in principle) arbitrarlly chosen. We could separate the correlation functions from the oscillating part and determine the closest perfect sequence, as described in Sec. III. In this case, for both the correlation distance and the Hamming distance, a value of zero represents nondeparture from the perfect periodic order. The Hamming distance definition used above is not limited to binary sequences; however, in the case of binary sequences, the Hamming distance can be obtained by adding the two sequence (mod 2) and dividing by the sequence length.

2. Run length encoding as a measure of disorder

We can consider the stacking sequence as a string of integer numbers given in some base (for the case of close packed structure a binary sequence suffices, as Sec. III showed) and try to find the most compact representation for the string. If we choose to ‘‘compact’’ the sequence using a run length encoding (RLE),[31] then the perfectly periodic sequence will correspond to a string of symbols representing the smallest repeating unit and the corresponding number of times the unit cell repeats itself in the complete sequence.

Consider, for example, in the case of a compact structure, the periodic sequence

$$110100110100110100110100110100110100110100110100110100.$$

The RLE code for this sequence will be

$$(2112)_9,$$

which consists of four symbols algorithmically representing that to reproduce the sequence, we need to repeat nine times a sequence formed by two 1’s, one 0, one 7, and two 0’s. With increasing size of the perfectly periodic sequence the RLE code will not increase but only the number of times the code is repeated. For the limit of an infinite periodic sequence the number of symbols in the RLE code divided by the length of the sequence will tend to zero.

On the other hand, if the sequence represents a disorder structure with a faulting probability larger than zero, the length of the RLE code will be larger than the corresponding RLE code for the perfect sequence. If we consider the same perfect sequence used above, but with a faulting probability of 0.2, we could get

$$110011011010111000010010011010011010111110011110010.$$

The RLE code for this sequence will be

$$(22)(21)_2(11)(34)(12)[(12)(21)]_2(11)(52)(42)(11),$$

which now consists of 22 symbols.

In this case the RLE code will increase with an increasing sequence length, and in the limit of an infinite disordered sequence the number of symbols in the RLE code divided by the length of the sequence will tend to a finite value larger than zero and only depending on the percentage of random disorder. Figure 8 represents the tree structure of the RLE code of a faulted sequence and the corresponding underlying faulted sequence. Except for the end branch node, each node gives the number of times their subnodes should be repeated. The end branch node gives the RLE symbols.

We can define the run length encoding distance as the number of symbols in the RLE code divided by the length of the sequence:

$$D_{rle}\{\alpha_i\})=\#[\mathrm{RLE}(\{\alpha_i\})]/N, \tag{38}$$

where $\#[\mathrm{RLE}(x)]$ stands for the number of symbols in the RLE code of x.

Again the RLE distance is not limited to a close-packed structure, and from the definition a generalization to other types of stacking sequence is straightforward. RLE encoding in the case of nonbinary sequence is a known procedure in image processing and frequently used in common image file formats.[32]

Figure 7 compares the three defined distance measures. As in the case of the Δ_c value, D_{rle} can be considered an absolute measure of departure from periodicity. Compared to the Hamming distance the D_{rle} value does not behave linearly. While the Hamming distance is measures just the degree of mismatch of the measured sequence with a predefined one, the D_{rle} distance will identify if the faulted stacks gets closer to another periodic order different from the

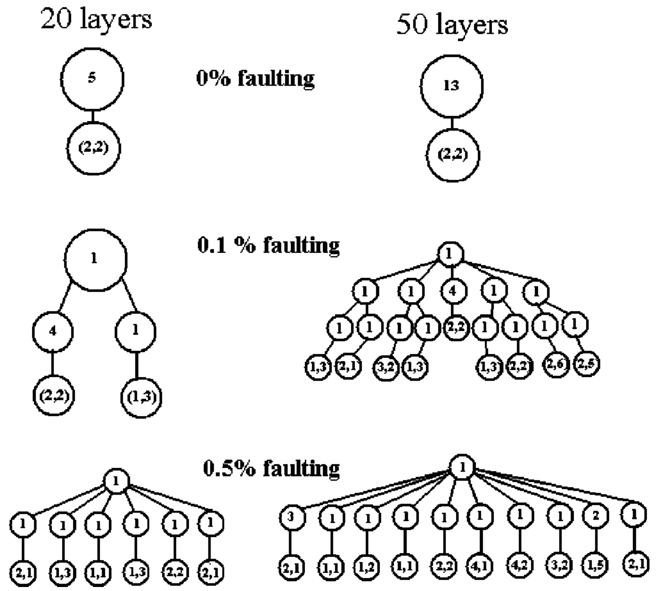

FIG. 8. Tree structure of the RLE code of faulted sequences with increasing fault probability. Except for the end branch node, each node gives the number of time their subnodes should be repeated. The end branch node gives the RLE symbols.

original one. The RLE coding, contrary to the use of the Δ_c measure, also carries information of the new periodicity which is being ‘‘born’’ as a result of the rearrangement of the layers through faulting (Fig. 8).

The RLE coding can also be used for the identification of repeating patterns in the sequence and their correlations. An occurring polytype β_i could be defined as a pattern repeating itself above some defined threshold. The RLE distance can be related to the so called algorithmic randomness.[32]

3. Identification of faulting complexes

The concept of a damage field[30] can be applied to the solved faulted and perfect sequences. The damage field is defined as the difference between the perfect sequence and the faulted sequence.

Consider the perfect sequence

11001100110011001100110011001100

and the faulted sequence

11011100010011001100011011001101.

The damage field between both sequences will then be

00010000100000000000101000000001,

where a 1 is found in every position the perfect and faulted sequences differ.

Faulting complexes will appear as patterns in the damage field. For example double faulting in two adjacent layers will appear as 11 patterns in the damage field. The identification of other faulting complexes present in the original sequence should be also straightforward.

V. CONCLUSIONS

It has been shown that, contrary to previous indirect procedures, the probability correlation functions of the staking arrangement in layer structures can be directly determined from the diffraction pattern. This determination follows from kinematical equations of diffraction, and in their calculation no assumption about the particular stacking order of the crystal or the density of faulting is needed; in this sense it can be considered as a method beyond the faulting model discussed by Varn *et al.*[18]

The obtained equations for the diffraction pattern allowed us to study, in a general framework, the effect of faulting in peak broadening and shift. Peak broadening is related to the lost of correlation length due to the random character of faulting, while a peak shift describes the departure of the ideal order from the perfect periodicity towards a possible new ordering. The competing effect between the departure from the ideal ordering and the correlation length has been discussed. No new order can be considered to occur if the periodicity length is larger than the correlation length. It has been shown how the probability correlation length can serve as a fingerprint of planar disorder and polytypism. The identification of polytypes and disorder has been exemplified with experimental data from rare earth-Co alloys.

Parameters for a quantitative characterization of disorder have been proposed, taking concepts from information theory and algorithmic complexity. It is shown how the RLE procedure can be useful for the identification of intermixed polytypes and as distance measure of disorder. The introduced parameters can serve as theoretical tools for a further discussion of phase transformations involving polytypism and planar disorder. Experimental work is underway in single crystal to apply the defined distance parameters to real structures.

ACKNOWLEDGMENTS

Thanks are owed to Professor P. Scardi and Dr. M. Leoni for helpful discussion. One of the authors (E.E.R.) acknowledges support from the Humboldt Foundation. Part of this project was carried out under TWAS research Grant No. 99-082 REG/PHYS/LA and under a Alma Mater grant from the University of Havana. The experimental diffraction work was carried out at the LNLS synchrotron facility in Campinas Brasil with financial support from CLAF and the LNLS.

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