Acta Crystallographica Section A
Foundations of Crystallography
ISSN 0108-7673

Received 8 January 2008
Accepted 3 June 2008

Stacking and twin faults in close-packed crystal structures: exact description of random faulting statistics for the full range of faulting probabilities

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The classical model of independent random single deformation faults and twin faulting in face-centered-cubic and hexagonal close packing is revisited. The model is extended to account for the whole range of faulting probabilities. The faulting process resulting in the final stacking sequences is described by several equivalent computational models. The probability sequence tree is established. Random faulting is described as a finite-state automaton machine. An expression giving the percent of hexagonality from the faulting probabilities is derived. The average sizes of the cubic and hexagonal domains are given as a function of single deformation and twinning fault probabilities. An expression for the probability of finding a given sequence within the complete stacking arrangement is also derived. The probability $P_0(\Delta)$ of finding two layers of the same type Δ layers apart is derived. It is shown that previous generalizations did not account for all terms in the final probability expressions. The different behaviors of the $P_0(\Delta)$ functions are discussed.

1. Introduction

Planar faulting in close-packed structures has been studied for decades [see Welberry (1985) for a review of early work]. Independent random faulting is the simplest and most studied model of planar disorder in close-packed arrangements (Warren, 1969; Ustinov *et al.*, 2001). In this model, different types of defects have constant probabilities of occurrence which are independent from each other. The constant character of the faulting probabilities and their mutual independence implies that the stacking defects do not interact, forming a sort of one-dimensional 'ideal gas'. It is clear that such a model of non-interacting defects can only be valid in real materials for low densities of defects.

Close-packed three-dimensional structures are built from two-dimensional layers with hexagonal symmetry piled up one over the other. 'Close packed' implies that this type of structure attains the highest possible packing density (Conway & Sloane, 1991). The face-centered-cubic (f.c.c.) and the hexagonal close-packed (h.c.p.) structures are the two simplest and best-known close-packed structures realized by a large number of crystalline materials. In close-packed structures, two types of faults are usually considered: deformation faults, which are jogs in the otherwise perfect periodic sequence, and twin faults, which cause reversions in the stacking ordering. The probability for the occurrence of a deformation fault will be denoted by α, while the probability for the occurrence of a twin faulting will be denoted by β.

The perfectly ordered f.c.c. structure follows, along the hexagonal [001] direction, an $ABCABCAB\ldots$ or $ACBACBA\ldots$ sequence (Verma & Krishna, 1966). Each letter, A, B or C, represents a different lateral displacement of the layers perpendicular to the stacking direction. Instead of using the letters ABC to code the stacking arrangement, less redundant codings have been devised: the most common ones are the Hägg code and the HK coding. In the former, a pair of consecutive layers is given a plus (1) symbol if they form a 'forward' sequence AB, BC or CA, otherwise a minus sign (0) is given. The other common binary coding is to consider the nearest neighbors of any layer. If the layer above and below the layer concerned are of the same type (*e.g.* $A\underline{B}A$, $C\underline{A}C$, $B\underline{C}B$ *etc.*) a symbol **h** (0) is assigned, otherwise a letter **k** (1) is assigned.

Patterson (1952) approached single-stacking faulting in f.c.c. structures using a difference-equation procedure. Gevers (1954) further extended the theory by considering a four-layer interaction. This treatment followed previous work by Wilson (1942), Hendrick & Teller (1942) and Jagodzinski (1949*a*,*b*). Relevant contributions were also given by Kakinoki & Komura (1952). In Gevers' treatment, difference equations are derived which describe the relation between faulting probability parameters and the probability of finding a particular layer in the stack with a given displacement. Warren (1969) developed the theory for a low density of faulting and obtained an expression for the probability $P_0(\Delta)$ of finding two layers, Δ layers apart, with the same perpendicular

displacement (*i.e.* $A \ldots A$, $B \ldots B$, $C \ldots C$). $P_0(\Delta)$ is also known as the probability correlation function. The treatment by Warren has become the most used when dealing with faulting in close-packed structures. Pandey & Krishna (1977), analyzing SiC single crystals, solved Gevers' equations without a limitation on the density of faulting. Velterop *et al.* (2000) lifted some of the restrictions in Warren's treatment and considered the influence of texture and nonuniform faulting probabilities. Estevez-Rams, Leoni *et al.* (2003) demonstrated that the Warren treatment can be considered as a particular case of a more general proposition for the probability function $P_0(\Delta)$, from which the diffracted intensity can be calculated.

Recently, Tiwary & Pandey (2007*a*,*b*) have studied the decaying behavior of the $P_0(\Delta)$ functions, obtaining analytical expressions for the correlation length as a function of the deformation- and twin-fault probabilities.

Although the random model for faulting is of limited practical use, it may serve as a reference to which other models and experimental results can be compared. From such a comparison, information about the interaction of defects (*i.e.* deviations from a random distribution) may be obtained. It will also be shown in this work that the model leads to the definition of the so-called correlation lengths and domain sizes, which have a general validity beyond random faulting.

In this paper, the independent-probability random faulting model will be explored, while in a future paper the implications for the diffraction profiles for the whole range of faulting probabilities will be discussed. In the treatment that follows, any restriction in the faulting probability will be lifted, while, as in the existing restricted model, planar defects will be considered to occur on only one plane or, in other words, perpendicular to a unique crystallographic direction.

We start by describing the effect of deformation and twin faults in a close-packed arrangement. The transition tree of the stochastic process, allowing for any density of faulting, will be built. The first derivation of the transition tree for a single faulting model was probably given by Jagodzinski (1949*b*), who, starting from an unfaulted hexagonal close-packed structure, considered the probability of finding an *h* or *k* environment in the stacking arrangement. Gevers (1954) and Pandey & Krishna (1977) developed similar transition trees but in terms of faulting parameters. The approach used in the present paper for deriving the probability transition tree will follow a similar logic but will consider the possible simultaneous occurrence of deformation faults and twin faulting.

A finite-state automaton (FSA) describing the stacking process will be constructed. The description of the faulting as an FSA is due to the work done on faulting by Varn & Canright (2001), Varn (2001), Varn *et al.* (2002) and Varn & Crutchfield (2004), although their reconstruction process will not be followed. A nondeterministic stochastic machine will be used to derive an expression for the probability of finding a given sequence within the complete stacking arrangement in terms of the faulting probabilities α and β. Similarly, an analytical expression will be derived for the probability of finding within the crystal stack a face-centered-cubic domain with L number of layers. An analogous expression for finding a close-packed hexagonal domain within the crystal stack will also be given.

Finally, an expression for $P_0(\Delta)$ will be constructed. $P_0(\Delta)$ is essential in describing the effect of faulting on the diffraction pattern. A comparison with previous studies will explore the validity of the different approximations made. The probability correlation function shows three different behaviors depending on the faulting density. The loss of correlation as a result of the planar faulting defines the correlation length of the system.

2. The transition probability tree for independent random faulting

For the case of the f.c.c. stacking, a single deformation fault occurs everytime there is a jog in the perfect arrangement:

$$\ldots\ A\ B\ C\ A\ |\ C\ A\ B\ C\ A\ \ldots$$

A twin fault, on the other hand, reverses the original stacking order:

$$\ldots\ A\ B\ C\ A\ |\ C\ B\ A\ C\ B\ \ldots$$

The independent random faulting model considers the occurrence of any of the two defects to be independent events with a fixed probability which, following Warren (1969), will be denoted by α for deformation faults and β for twin faults.

For deformation faults the average distance between faults is $1/\alpha$ and, starting with a forward sequence, the probability that after an A (B, C) layer there will be an unfaulted B (C, A) layer will be given by $\overline{\alpha} = 1 - \alpha$, while the probability of a fault will be α. It is clear that $\alpha = \frac{1}{2}$ corresponds to the maximum state of disorder, while $\alpha = 1$ indicates that a completely ordered forward (backward) sequence reverses to a completely ordered backward (forward) sequence. Therefore, only values up to $\alpha = \frac{1}{2}$ need to be considered.

According to the above explanation, if in a forward sequence the $\Delta - 2$ layer is in the A position, there will be two transition paths to a Δ layer with A displacement, namely

$$A \overset{\overline{\alpha}}{\rightarrow} B \overset{\alpha}{\rightarrow} A,$$

$$A \overset{\alpha}{\rightarrow} C \overset{\overline{\alpha}}{\rightarrow} A.$$

The twin fault can be understood as a transition from a forward (backward) sequence to a backward (forward) sequence. If the sequence starts in a forward state, then the probability of finding after an A (B, C) layer a B (C, A) layer will be $\overline{\beta} = 1 - \beta$, while the opposite will have probability β. However, in this case, the occurrence of a twin fault reverses the stacking order and therefore the transition path of a three-layer sequence starting with a 'forward' A layer and ending with an A layer will be

$$A \overset{\overline{\beta}}{\rightarrow} B \overset{\beta}{\rightarrow} A,$$

$$A \overset{\beta}{\rightarrow} C \overset{\beta}{\rightarrow} A.$$

In the case of twin faulting a value $\beta = 1$ will denote a new stacking arrangement, the perfect h.c.p. stacking sequence.

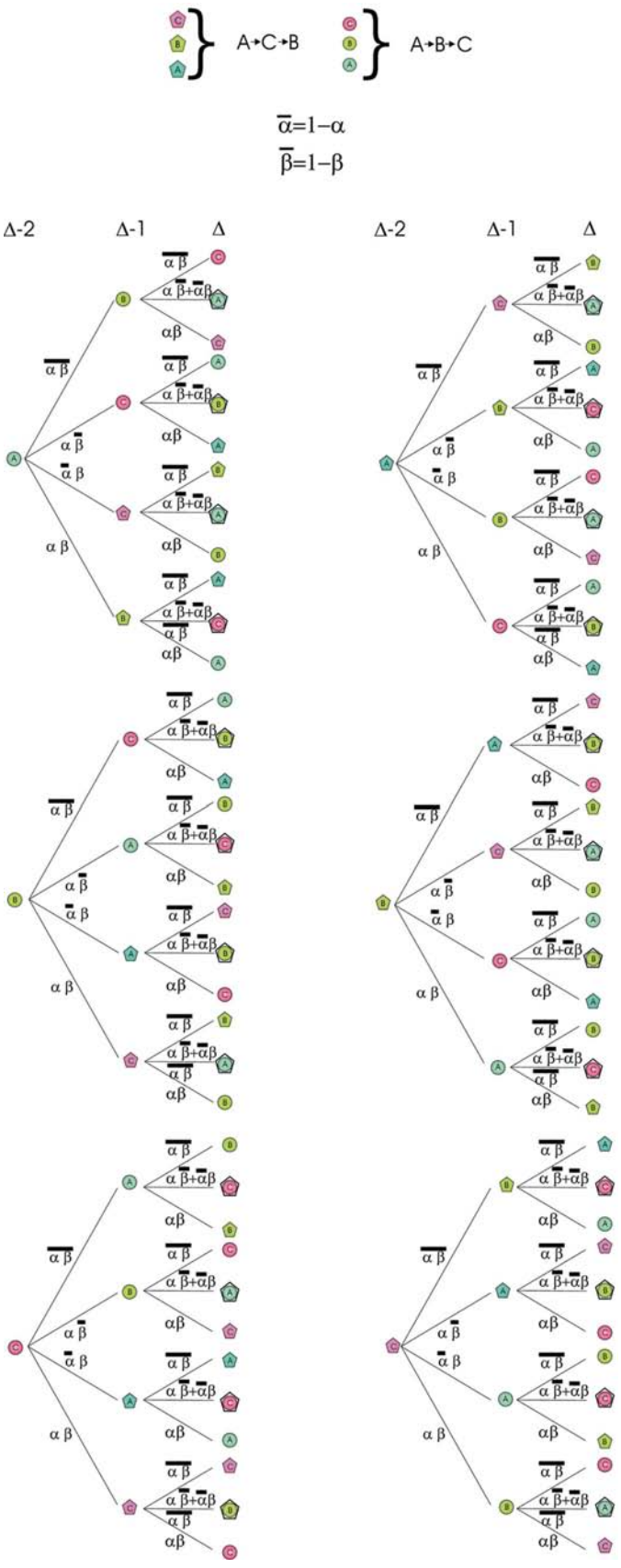


Figure 1
Probability tree for both deformation (α) and twin (β) faulting. Circles represent forward sequences and pentagons represent backward sequences.

This last fact introduces an important difference in the natures of the α and β faulting: While the first does not result in a change of stacking order, the latter does describe a reordering (reconstructive) transition, and must be considered in its whole range of values from 0 to 1.

Assuming a very low density of faulting, Warren (1969) considered that the transition paths between three layers could be taken separately for twin and deformation faults and the result added up as exclusive events. When the planar faulting density is larger than the values considered by Warren, the adding up of the two transition trees from deformation and twin faulting can not be done. Compared with the approximation for low faulting probabilities, now the possibility of the simultaneous occurrence of deformation and twin faulting ($\alpha\beta$) must be considered. The whole tree of transitions probabilities must be worked out (Gevers, 1954). Two cases must then be considered, in one case the $\Delta-2$ layer is in a 'forward' sequence, while in the second case the layer is in a 'backward' sequence. For example, the transition paths for a three-layer sequence starting at B and ending at A will be

$$B^{\mathrm{f}} \xrightarrow{\alpha\overline{\beta}} C^{\mathrm{f}} \xrightarrow{\overline{\alpha\beta}} A^{\mathrm{f}},$$
$$B^{\mathrm{b}} \xrightarrow{\alpha\overline{\beta}} C^{\mathrm{b}} \xrightarrow{\overline{\alpha}\beta+\alpha\overline{\beta}} A^{\mathrm{f,b}},$$
$$\xrightarrow{\overline{\alpha}\beta} C^{\mathrm{f}} \xrightarrow{\overline{\alpha\beta}} A^{\mathrm{f}},$$

where the superscripts denote whether the layer is in a 'forward' (f) or in a 'backward' (b) sequence. The same transition paths can be worked out for all possible combinations of starting and ending layers. The resulting transition probability tree is shown in Fig. 1. From this figure it can be seen that the transition probability tree repeats at each level. Thus, Fig. 1 is sufficient to represent the whole probabilistic information of the stacking process.

3. Independent random faulting as a finite automaton in the Hägg description

Starting from the Hägg code, one can give an equivalent description of the probability tree of the faulting process in terms of a finite-state automaton (FSA) or machine with an underlying Markov or stochastic process (Varn & Canright, 2001). A finite automaton in general will be defined by the automaton alphabet χ, which are the symbols emitted by the process, a set of states V and the transition probabilities between the states T (Varn, 2001). The finite automaton can be represented by a directed graph where each node represents a state and directed arcs between nodes denote the transitions from one state to another upon emitting a symbol. The starting state of an arc is called the source state of the transition, while the ending state of the arc is termed the destination state of the transition. Each arc will be labeled with an $s|p$ pair, where s is the emitted symbol and p the probability of the transition. For a Hägg code the alphabet set will be $\chi = \{1, 0\}$ representing the plus and minus sign, respectively.

A deformation fault in the f.c.c. structure will, in the Hägg code, reverse the sign at the position of the fault:

...	*A*	*B*	*C*	*A*	|	*C*	*A*	*B*	*C*	*A*	...
...	1	1	1	0	|	1	1	1	1	1	...

For a forward (backward) sequence a 1 (0) symbol is emitted with probability $\overline{\alpha}$ (α), and a 0 (1) symbol otherwise.

The corresponding deformation-fault finite-state automaton will be deterministic with two disconnected states describing the forward or the backward sequence. In the graph description of the automaton shown in Fig. 2(*a*), these two states are denoted by the letter 'f' for the forward sequence, and 'b' for the backward sequence. The state denoted by 's' is a ficticious starting state that chooses with equal probability an 'f' or a 'b' process. Once an 'f' or 'b' state is chosen, there is no way the deformation fault can drive the system to the other state. Each independent node is equivalent to the Bernoulli biased-coin process (Hopcroft & Ulman, 1979).

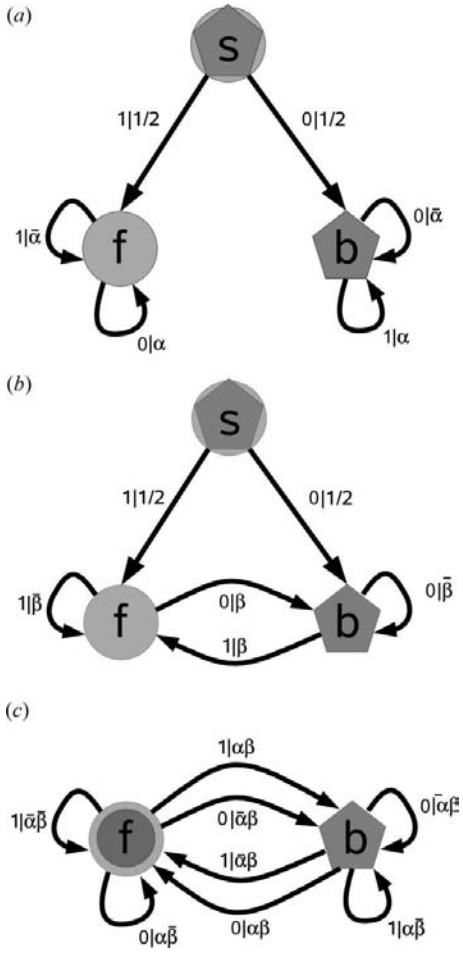


Figure 2
The Hägg random FSA with starting state 's' and two recurrent states 'f' and 'b' for the forward and backward sequence. The symbol $s|p$ represents a process that emits a symbol s with probability p. (*a*) The deformation faulting FSA is equivalent to a biased-coin or Bernoulli process with probabilities α for one event and $\overline{\alpha}$ for the complementary event. The two recurrent states are disconnected and once the process chooses between the 'forward' and the 'backward' state the system will stay in that state indefinitely. (*b*) The twin faulting FSA where the recurrent states are connected by the occurrence of twinning. (*c*) The combined deformation and twin nondeterministic FSA. The double circle in the forward state just signals that this is taken as the starting state. The nondeterministic character reflects that the emitted symbol does not determine unequivocally the next state of the process.

For twin faulting, if the sequence starts in a forward state, the Hägg code will consist of consecutive 1 symbols up to the first twin fault and 0 symbols from there on up to the next twin fault, where the sequence reverses again. The process is repeated on average every $1/\beta$ layers. For a single twin event a typical sequence will be

...	*A*	*B*	*C*	*A*	|	*C*	*B*	*A*	*C*	*B*	...
...	1	1	1	0	|	0	0	0	0	0	...

The finite automaton describing the faulted sequence will still have two recurrent states 'f' and 'b', but now the states are linked together by the twinning event which occurs with probability β. Fig. 2(*b*) shows the corresponding graph.

For combined deformation and twin faults, the Markov process will be more complicated. The finite automaton will again have the two 'f' and 'b' states. The transition between these two recurrent states will again be given by the event of a twin faulting; in this case, however, the twin faulting can occur as an isolated event $\overline{\alpha}\beta$ or as simultaneous event together with a deformation fault $\alpha\beta$. The complete automaton is depicted in Fig. 2(*c*), where the starting state has been omitted for simplicity and the forward state has been considered as the starting point of the process. Contrary to the single deformation and twin process, the combined automaton is no longer deterministic. This can be seen in the graph representation, noticing that each recurrent state has more than one source arc with the same symbol. The Markov transition matrix will describe the probability that a given event will result in a particular transition from one state to another state. For the finite state process being analyzed, there are two type of events and two states, and the corresponding transition matrices will be given by

$$T^{(0)} = \begin{pmatrix} \alpha\overline{\beta} & \overline{\alpha}\beta \\ \alpha\beta & \overline{\alpha}\overline{\beta} \end{pmatrix} \tag{1}$$

for events involving the emission of a 0 symbol and

$$T^{(1)} = \begin{pmatrix} \overline{\alpha}\overline{\beta} & \alpha\beta \\ \overline{\alpha}\beta & \alpha\overline{\beta} \end{pmatrix} \tag{2}$$

for events involving the emission of a 1 symbol.

Identifying the 'f' state with 1 and the 'b' state with 2, each entry $T_{ij}^{(s)}$ in a transition matrix represents the probability of making a transition from the i state to the j state upon emitting an s symbol. (In this case, for example, $T_{12}^{(1)}$ is the probability of making a transition from the 'f' state to the 'b' state while emitting a 1 symbol.) From the above matrices the following relation follows immediately:

$$T^{(0)} = \Upsilon \cdot T^{(1)} \cdot \Upsilon, \tag{3}$$

where Υ is the reversion operator

$$\Upsilon = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \quad (4)$$

The transition probability T between states without regard of the actual symbol output will be given by

$$T = T^{(0)} + T^{(1)} = \begin{pmatrix} \overline{\beta} & \beta \\ \beta & \overline{\beta} \end{pmatrix}. \quad (5)$$

The probability of being in an 'f' or a 'b' state will be $P(f) = P(b) = \frac{1}{2}$, independent of the position in the sequence.

Consider now a given stacking arrangement for the whole crystal and extract from the stacking sequence a particular subsequence of length L number of layers given by $w^L = d_1 d_2 d_3 \ldots d_L$. It can be shown (Hopcroft & Ulman, 1979) that for the type of finite automaton considered, the probability of finding a particular w^L sequence will be given by

$$P(w^L) = \tfrac{1}{2}\vec{1}^T \cdot T^{(d_1)} \cdot T^{(d_2)} \cdot T^{(d_3)} \cdot \ldots T^{(d_L)} \cdot \vec{1}, \quad (6)$$

where $\vec{1}^T = (11)$. (For example, the probability of finding in the sequence a 0 symbol followed by a 1 symbol will be $P(01) = \frac{1}{2}\vec{1}^T \cdot T^{(0)} \cdot T^{(1)} \cdot \vec{1}$.)

Consider now a w^L sequence starting with n_1 consecutive 1 symbols, followed by n_2 0 symbols, then n_3 1 symbols and so on, ending with n_k 0 symbols, then

$$P(w^L) = \tfrac{1}{2}\vec{1}^T \cdot \left\{(T^{(1)})^{n_1} \cdot \Upsilon\right\} \cdot \left\{(T^{(1)})^{n_2} \cdot \Upsilon\right\} \ldots \left\{(T^{(1)})^{n_k} \cdot \Upsilon\right\} \cdot \vec{1}. \quad (7)$$

Let $\tilde{w}^L$ be the sequence resulting from the transposition of 1 and 0 symbols in the original w^L sequence, then

$$P(\tilde{w}^L) = \tfrac{1}{2}\vec{1}^T \cdot \Upsilon \cdot \left\{(T^{(1)})^{n_1} \cdot \Upsilon\right\} \cdot \left\{(T^{(1)})^{n_2} \cdot \Upsilon\right\} \ldots (T^{(1)})^{n_k} \cdot \vec{1}. \quad (8)$$

Noting that $\vec{1}^T \cdot \Upsilon = \vec{1}^T$, equation (8) is identical to equation (7). Thus, $P(w^L) = P(\tilde{w}^L)$ [*e.g.* $P(01) = P(10)$] and equation (7) is a general expression for the probability of finding a subsequence w^L of any kind in the stacking sequence. If d^L is the sequence made by L consecutive d symbols one can notice that, as a particular case of equation (8), $P(1^L) = P(0^L)$, as should be expected, as both sequences represent the same crystal stacking arrangement, namely the f.c.c. structure.

For any sequence of length L number of layers, the FSA of Fig. 2 with $\alpha, \beta \neq 0$ will give a probability of occurrence larger than zero. This result has important implications for the use of the random FSA as a comparison for other types of faulting processes: *The random faulting model can generate any sequence of length L with probability larger than zero*.

The number of possible sequences of length L accepted by the FSA will be 2^L. Any other faulting process will generate a set of stacking arrangements, or sequence strings, which is a subset of the set of allowed sequence strings of the random FSA. How restrictive a particular model faulting is, in terms of the allowed sequences, with respect to the random faulting model can be quantified by exploring the subset strings spanned by the particular faulting model.

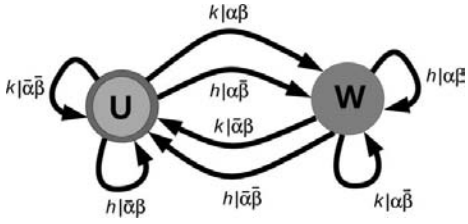


Figure 3
The nondeterministic HK random FSA with recurrent states 'U' and 'W'. The notation is the same as in Fig. 2.

4. Independent random faulting as a finite automaton in the HK description

In the Hägg notation, a twin fault results in a flip in the value of the sites starting from the position of the fault up to the end of the stacking sequence. The twin faulting viewed as an operator acting over the Hägg code has an infinite range. In the HK notation, on the other hand, a twin fault in a perfect f.c.c. structure just flips the value of one site in the code:

A	*B*	*C*	*A*	*B*	*C*	*A*	|	*C*	*B*	*A*	*C*	*B*
k	*k*	*k*	*k*	*k*	*k*	*h*	|	*k*	*k*	*k*	*k*	*k*

whereas a deformation fault changes the value at two neighboring sites:

A	*B*	*C*	*A*	*B*	*C*	*A*	|	*C*	*A*	*B*	*C*	*A*
k	*k*	*k*	*k*	*k*	*k*	*h*	|	*h*	*k*	*k*	*k*	*k*

Hence, both types of faulting are local operators in the HK coding. An FSA can also be constructed for the HK representation of the stacking arrangement and it is shown in Fig. 3. The resulting stochastic machine is also nondeterministic. In the case of this FSA, the U and W states can not be identified with the forward and backward sequence; they are computational states with no physical meaning. The HK random FSA is similar to the Hägg stochastic machine but with a relabelling of the directed arcs. The transition matrices in this case are

$$T^{(k)} = \begin{pmatrix} \overline{\alpha}\overline{\beta} & \alpha\beta \\ \overline{\alpha}\beta & \alpha\overline{\beta} \end{pmatrix} \quad (9)$$

$$T^{(h)} = \begin{pmatrix} \overline{\alpha}\beta & \alpha\overline{\beta} \\ \overline{\alpha}\overline{\beta} & \alpha\beta \end{pmatrix} \quad (10)$$

and the relation between $T^{(h)}$ and $T^{(k)}$ follows:

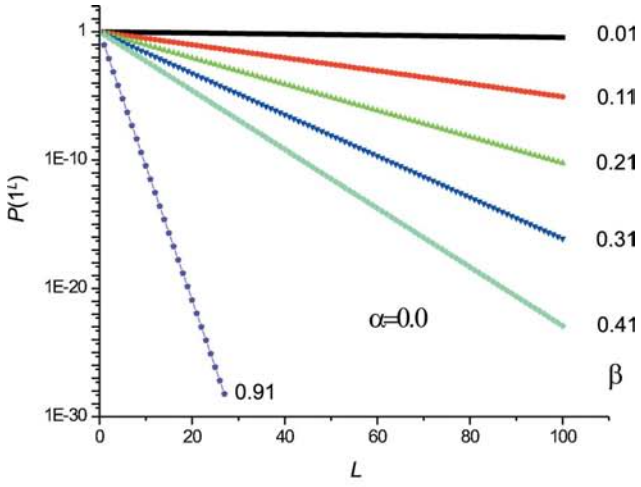
$$T^{(h)} = \Upsilon \cdot T^{(k)}. \quad (11)$$

The state transition matrix is

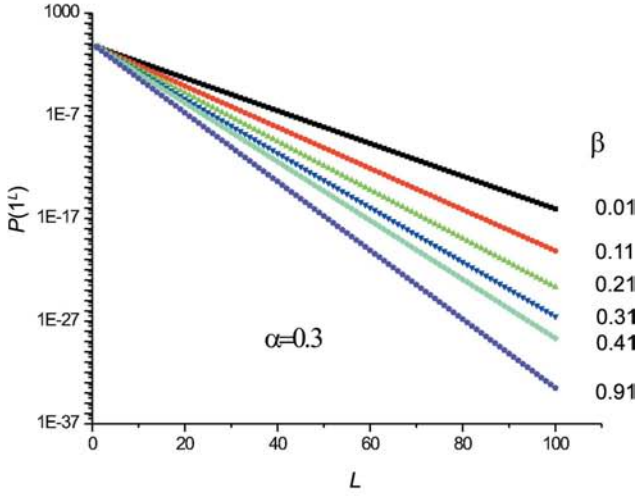
$$T = \begin{pmatrix} \overline{\alpha} & \alpha \\ \overline{\alpha} & \alpha \end{pmatrix}. \quad (12)$$

The probability of finding the FSA in the U state is $P(\mathrm{U}) = \overline{\alpha}$ and in the W state is $P(\mathrm{W}) = \alpha$, which indicates that the states U and W are related with the occurrence of deformation faulting. From the graph of the FSA shown in Fig. 3 one finds

$$\begin{aligned} P(h) &= P(\mathrm{U})(\overline{\alpha}\beta + \alpha\overline{\beta}) + P(\mathrm{W})(\alpha\beta + \overline{\alpha}\overline{\beta}) \\ &= \beta + 2\alpha\overline{\alpha}(1 - 2\beta), \end{aligned} \quad (13)$$



(*a*)



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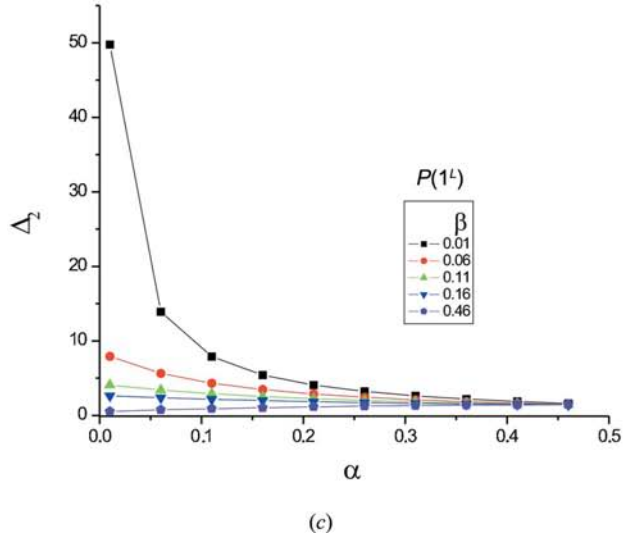

(*c*)

Figure 4
The probability of finding a k domain as a function of its size L number of layers for different β values. (*a*) $\alpha = 0$. (*b*) $\alpha = 0.3$. (*c*) The characteristic length of the decaying behavior of $P(k^L)$ as a function of α values for different β probabilities.

giving the probability of finding a close-packed hexagonal environment in the stack. The fraction of hexagonal sites increases linearly with β over its whole range of values [0, 1] and quadratically with α over the interval $[0, \frac{1}{2}]$. The probability of finding a cubic environment follows immediately from the relation $P(h) + P(k) = 1$. The expression for $P(h)$ and $P(k)$ relates the treatment of planar disorder, given by independent deformation and twin faults, with the probability of finding a hexagonal or cubic environment in the stacking arrangement, which are the parameters used in the early works of Jagodzinski.

Using a similar expression to equation (6), the probability of finding a k domain of length L in the stacking sequence will be

$$P(k^L) = (\overline{\alpha} \;\; \alpha) \cdot [T^{(k)}]^L \cdot \vec{1}. \tag{14}$$

If t_0 and t_1 are the eigenvalues of the $T^{(k)}$ matrix, and $X = (\vec{x}_0 \;\; \vec{x}_1)$ is the matrix of the corresponding eigenvectors, then

$$[T^{(k)}]^L \cdot X^T = X^T \cdot \begin{pmatrix} t_0 & 0 \\ 0 & t_1 \end{pmatrix}^L, \tag{15}$$

where X^T is the transpose matrix of X. Using equation (15) to find an explicit expression for $[T^{(k)}]^L$ and substituting into equation (14) leads to

$$P(k^L) = \frac{1}{2p}\left\{\left(\frac{\overline{\beta}+p}{2}\right)^L (p-q) + \left(\frac{\overline{\beta}-p}{2}\right)^L (p+q)\right\}, \tag{16}$$

where

$$p^2 = 1 + \beta^2 - 2P(h),$$
$$q = 2P(h) - (1+\beta).$$

The limiting values of the $P(k^L)$ function are given by

$$P(k^L) = \begin{cases} 2^{-L} & \alpha = \frac{1}{2} \\ 2^{-L} & \beta = \frac{1}{2} \\ \overline{\beta}^L & \alpha = 0, 1 \\ \overline{\alpha}^{L+1} + \alpha^{L+1} & \beta = 0. \end{cases}$$

Figs. 4(*a*),(*b*) show $P(k^L)$ as a function of L in a semilogarithmic plot for different values of α and β. Two characteristic lengths of the exponential decaying behavior of $P(k^L)$ can be obtained from equation (16):

$$\Delta_{k_1} = -\frac{1}{\log[(\overline{\beta}-p)/2]}, \tag{17}$$

$$\Delta_{k_2} = -\frac{1}{\log[(\overline{\beta}+p)/2]}. \tag{18}$$

Of the two lengths, Δ_{k_2} is the one determining the behavior of $P(k^L)$ and Fig. 4(*c*) shows Δ_{k_2} as a function of α for different β values. A similar analysis can be done for the h-domain blocks. In this case

$$P(h^L) = \frac{1}{2r}\left\{\left(\frac{\beta+r}{2}\right)^L (r-k) + \left(\frac{\beta-r}{2}\right)^L (r+k)\right\}, \tag{19}$$

where

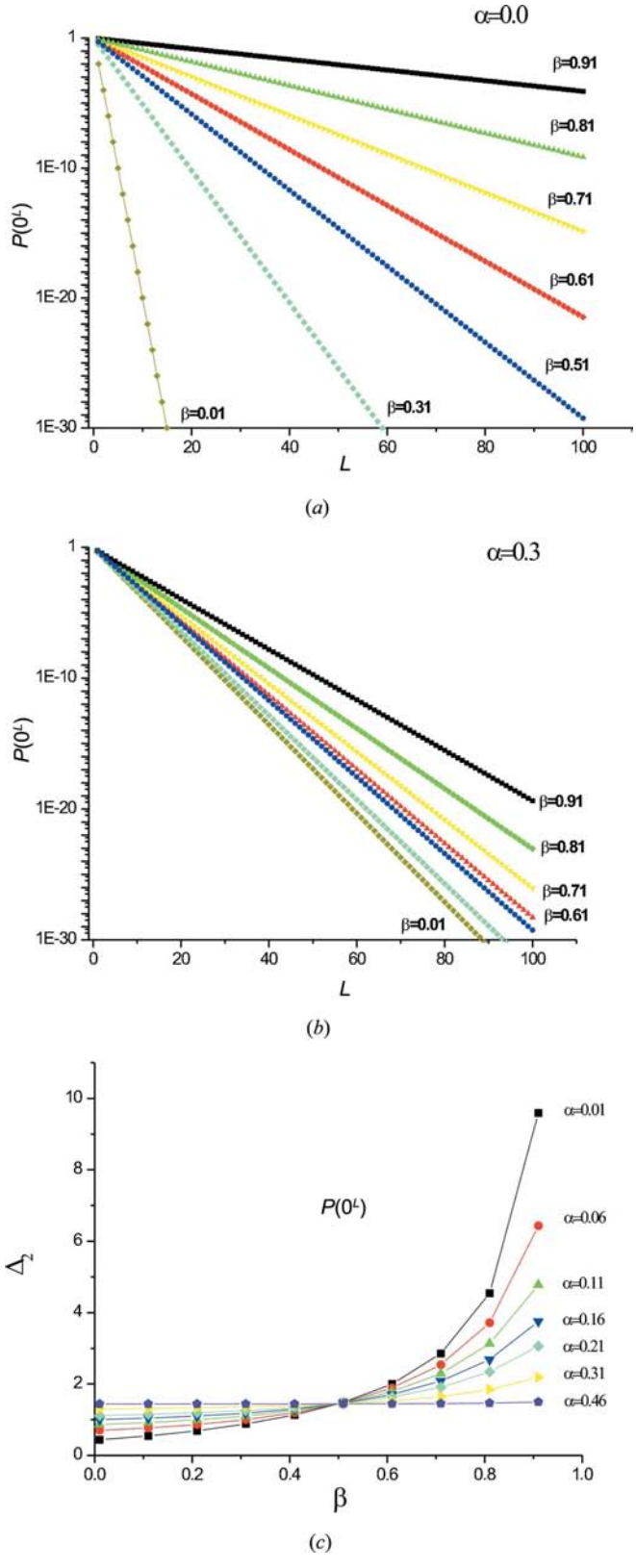

Figure 5
The probability of finding an h domain as a function of its size L number of layers for different β values. (*a*) $\alpha = 0$. (*b*) $\alpha = 0.3$ (the $\beta = 0.31$ and 0.41 labels have been omitted for reasons of space). (*c*) The characteristic length of the decaying behavior of $P(h^L)$ as a function of β values for different α probabilities.

$$r^2 = 1 - 2\beta + 2\beta^2 - p^2, \quad k = \beta - 2P(h).$$

The limiting values for the $P(h^L)$ function are

$$P(h^L) = \begin{cases} 2^{-L} & \alpha = \frac{1}{2} \\ 2^{-L} & \beta = \frac{1}{2} \\ \beta^L & \alpha = 0, 1 \\ \overline{\alpha}^{L+1} + \alpha^{L+1} & \beta = 1. \end{cases}$$

The exponential decaying behavior of $P(h^L)$ (Fig. 5*a*,*b*) allows the definition of two characteristic lengths:

$$\Delta_{h_1} = -\frac{1}{\log[(\beta - r)/2]}, \tag{20}$$

$$\Delta_{h_2} = -\frac{1}{\log[(\beta + r)/2]}. \tag{21}$$

The dominant characteristic length Δ_{h_2} is shown as a function of β for different α values in Fig. 5(*c*). Equations (16) and (19) can be used to calculate the average block size for the k and h domains:

$$\langle L \rangle_h = \frac{\beta + 2\alpha\overline{\alpha}(1 - 2\beta)[\alpha\overline{\alpha}(1 - 2\beta) + 2]}{[\alpha\overline{\alpha}(1 - 2\beta) - \overline{\beta}]^2}, \tag{22}$$

$$\langle L \rangle_k = \frac{\overline{\beta} + 2\alpha\overline{\alpha}(1 - 2\beta)[\alpha\overline{\alpha}(1 - 2\beta) - 2]}{[\alpha\overline{\alpha}(1 - 2\beta) + \beta]^2}. \tag{23}$$

5. The probability correlation function

An expression for $P_0(\Delta)$ will now be derived. $P_0(\Delta)$ is the probability of finding two layers, Δ layers apart and of the same type (*e.g.* $A \ldots A$, $B \ldots B$, $C \ldots C$), $P_{\rm f}(\Delta)$ [$P_{\rm b}(\Delta)$] is the probability when the layers are of the type $A \ldots B$, $B \ldots C$, $C \ldots A$ ($A \ldots C$, $B \ldots A$, $C \ldots B$). The $P_i(\Delta)$ functions are essential in describing the effect of faulting on the diffraction pattern (Warren, 1969).

Returning to the Hägg random FSA of Fig. 2, the following equations can be derived:

$$P_0(\Delta) = 2P(01)P_0(\Delta - 2) + 2P(11)[P_{\rm f}(\Delta - 2) + P_{\rm b}(\Delta - 2)], \tag{24}$$

$$P_0(\Delta - 1) = \tfrac{1}{2}[P_{\rm b}(\Delta - 2) + P_{\rm f}(\Delta - 2)] \tag{25},$$

where use has been made of $P(\rm f) = P(\rm b) = \frac{1}{2}$. $P(01)$ can be calculated from equation (6). $2P(01)$ is the probability of finding a hexagonal environment in the stack given by equation (13). The following boundary conditions pertaining to equations (24) and (25) (see Warren, 1969) must be added:

$$P_0(0) = 1,$$
$$P_0(1) = 0.$$

The first condition is just the trivial case of considering the same layer two times, the second condition is a result of the close-packed restriction: two consecutive layers of the same type can not occur in the stacking sequence. The solution to equations (24) and (25) can be found by proposing

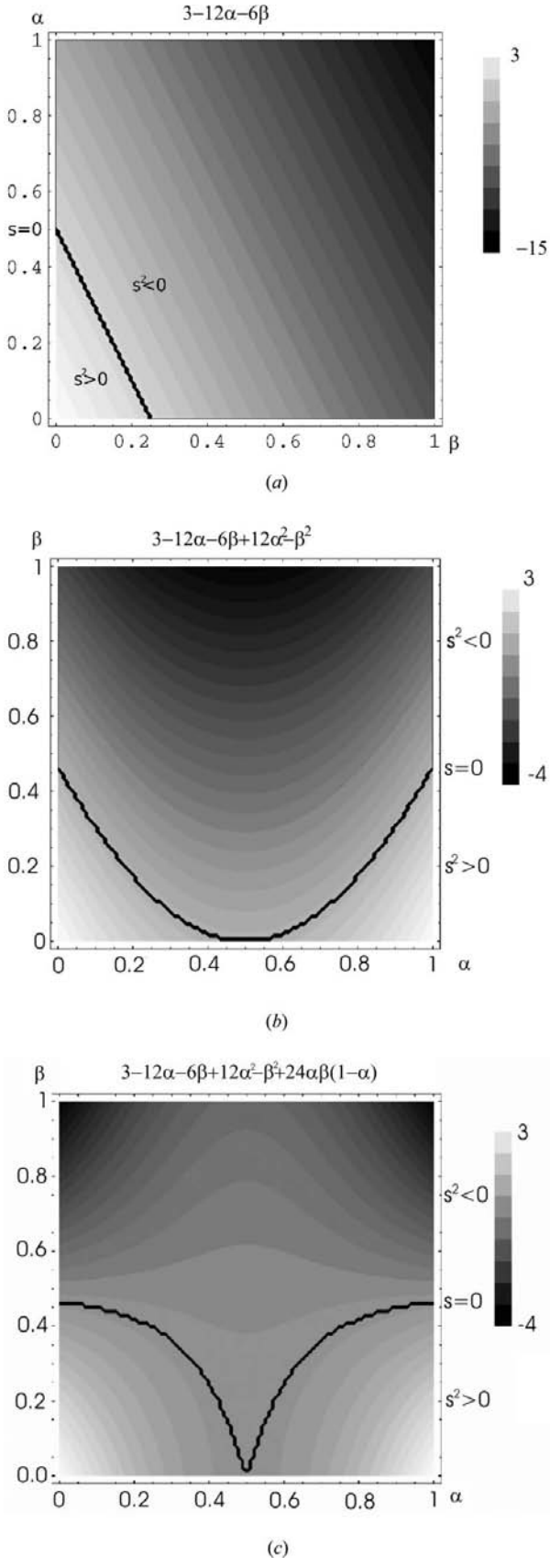

Figure 6
Density plot of s^2 for (*a*) the approximation according to Warren (1969), (*b*) the approximation according to Velterop *et al.* (2000) and (*c*) considering all terms.

$$P_0(\Delta) = a + bx^{\Delta}. \tag{26}$$

It is then found that

$$(3a-1)[1-\alpha+\alpha^2+\beta(2\alpha-2\alpha^2-1)] + bx^{\Delta-2}[(1-2\beta)(1+3\alpha^2-3\alpha)+(1-\beta)x+x^2] = 0.$$

This last equation has to be satisfied independently of the values of Δ, α and β, therefore each term alone has to be strictly zero. From this condition it follows that

$$3a-1=0,$$
$$(1-2\beta)(1+3\alpha^2-3\alpha)+(1-\beta)x+x^2=0.$$

From the first equation

$$a = \tfrac{1}{3} \tag{27}$$

and from the second quadratic equation

$$x = \tfrac{1}{2}[-(1/\beta) \pm is],$$

where

$$s^2 = (3-12\alpha-6\beta)+(12\alpha^2-\beta^2)+24\alpha\beta\overline{\alpha}. \tag{28}$$

Using the boundary conditions results in

$$P_0(\Delta) = \tfrac{1}{3}\left\{1+\left(1+\frac{i\beta}{s}\right)\left[\frac{-(1-\beta)+is}{2}\right]^{\Delta} + \left(1-\frac{i\beta}{s}\right)\left[\frac{-(1-\beta)-is}{2}\right]^{\Delta}\right\}. \tag{29}$$

In the classic treatment of Warren, only the first term on the right-hand of equation (28) is considered. Velterop *et al.* (2000) and Estevez-Rams, Leoni *et al.* (2003) took into account the quadratic terms in α and β but missed the mixed factor $24\alpha\beta\overline{\alpha}$, which gives a further quadratic contribution to the s value. Equation (28) is valid for the whole range of values for α and β. The additional third term in equation (28) accounts for the simultaneous occurrence of deformation and twin faults at higher faulting densities.

The expression for $P_0(\Delta)$ given by equation (29) can be described as an oscillating damped function.

In Tiwary & Pandey (2007*a*) an equivalent expression [equation (15) in their article] to equation (29) is made equal to a single exponential decaying function [equation (16) in their article], thus defining a single characteristic length for the decaying behavior. Care must be taken: to make a sum of two exponentials equal to a single exponential is in general not mathematically justified. We did not manage to reduce the sum of two exponentials to a single exponential. The particular cases treated by Tiwary & Pandey (2007*a*), though, are valid and similar to the results presented here.

Fig. 6 is a density plot of s^2 *versus* α, β for the three approximations. The linear Warren term strongly underestimates the region where $s^2 > 0$ and does not show the expected symmetric behavior of the system at $\alpha = \frac{1}{2}$. Velterop quadratic terms already correct the s^2 term showing the correct symmetry at $\alpha = \frac{1}{2}$ and improve the estimate of the region where $s^2 > 0$, especially at lower values of α, β.

Fig. 7 shows the error curves between the exact s^2 expression given by equation (28) and the Warren (*a*) and Velterop (*b*) approximations. The error curves have been plotted only for values where $s^2 > 0$, as this is the region where comparison makes sense. As can be seen, the Warren approach leads to gross errors even larger than 100% for the larger part of the probability region. Even for $\beta = 0$, the error is above 50% for β values slightly larger than 0.2 (Fig. 7*a*). The Velterop approach, where quadratic terms are taken into account, is an improvement over the Warren approach especially when one type of defect has a small probability. This behavior is expected as the mixed term in equation (28) will be nearly zero. However, for a mixed occurrence of defects, the error between the Velterop approach and the true s^2 value grows rapidly and for $\alpha = \beta > 0.15$ the error is already above 50% (Fig. 7*b*).

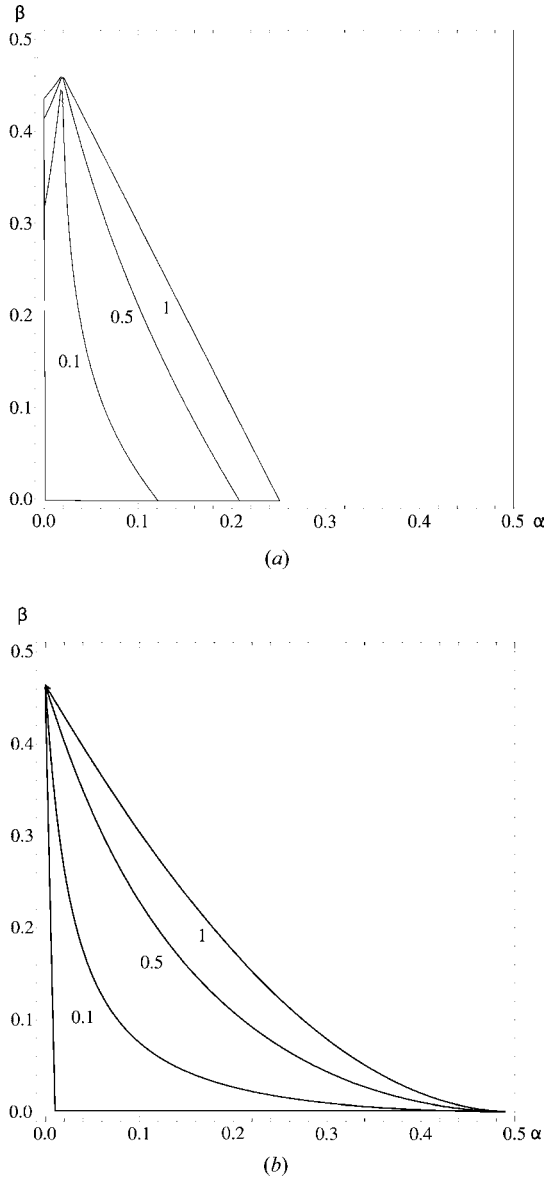

Figure 7
Relative error between the true s^2 function and the approximations due to (*a*) Warren (1969) and (*b*) Velterop *et al.* (2000). Any combination of defect probability will give an error above the nearest contour curve to the left and below the nearest contour curve to the right.

The solution given by equation (29) is valid when $s^2 \neq 0$. To find a solution for $s^2 = 0$ use still has to be made of equation (28) to derive a relation between α and β and deduce a new difference equation,

$$-(3-\beta)^2 + 12P_0(\Delta) + 12(1-\beta)P_0(\Delta-1) + 3(1-\beta)^2P_0(\Delta-2) = 0.$$

A trial solution of the form

$$P_0(\Delta) = a + bZ^\Delta + c\Delta Z^\Delta$$

can be proposed, and using the boundary conditions the following results:

$$P_0(\Delta) = \tfrac{1}{3}\left\{1 + 2(-1)^\Delta\left(\frac{1-\beta}{2}\right)^\Delta\left[1 + \left(\frac{\beta}{1-\beta}\right)\Delta\right]\right\}. \quad (30)$$

5.1. Case I: $s^2 > 0$

Following the approach of Warren (1969),

$$P_0(\Delta) = \tfrac{2}{3}(-1)^\Delta Z^\Delta[\cos\varphi\Delta + (\beta/s)\sin\varphi\Delta] + \tfrac{1}{3}, \quad (31)$$

with Z being a positive number,

$$Z = \frac{[(1-\beta)^2 + s^2]^{1/2}}{2}, \quad (32)$$

and

$$\tan\varphi = \frac{s}{1-\beta}. \quad (33)$$

Figs. 8(*a*)–(*d*) show the P_0 function for different values of α and β.

In equation (31) the Z^Δ term is a decreasing function of Δ ($Z < 1$ for all allowed values of α and β), while the remaining term corresponds to an oscillating function: equation (31) can thus be considered as a damped oscillating function.

The decaying term allows the definition of a characteristic length scale Δ_c of the system:

$$\Delta_c = -\frac{1}{\log Z}. \quad (34)$$

The occurrence of any kind of planar disorder results in the loss of correlation in the stacking arrangement: as a result of random events in the stacking sequence, the stack of layers loses its long-range 'memory'. The loss of long-range memory may be understood in the sense that Δ_c is related to the length of the system when from any position in the stacking arrangement the Δ value is sufficiently large that the type of layer that occurs can not be predicted with a probability larger than $\frac{1}{3}$ (Shrestha, 1996; Estevez-Rams, Aragon-Fernandez *et al.*, 2003).

Considering only one kind of defect, equation (34) reduces to

$$\Delta_c = -\frac{2}{\log[1-3\alpha(1-\alpha)]} \tag{35}$$

for deformation faults and to

$$\Delta_c = -\frac{2}{\log(1-2\beta)} \tag{36}$$

for twin faulting.

Equations (34), (35) and (36) are in accordance with similar results reported earlier (Estevez-Rams, Leoni *et al.*, 2003; Tiwary & Pandey, 2007*a*,*b*). The argument of the logarithmic function for the case of deformation faults grows faster than the same argument for twinning. Up to a common value of $\frac{1}{3}$, deformation faults will result in a smaller value of Δ_c and therefore larger disorder than twin faulting. For larger values of faulting, twin faults will result in larger disorder and Δ_c will drop abruptly to zero for $\beta = \frac{1}{2}$. A value of $\alpha = \frac{1}{2}$ will give $\Delta_c = \frac{1}{4}$.

The other term in (31),

$$(-1)^{\Delta}[\cos\varphi\Delta + (\beta/s)\sin\varphi\Delta], \tag{37}$$

describes an oscillating periodic function of Δ. The period of this term will be given by the argument φ. The interval of valid φ ranges from, including, zero to, excluding, $\pi/3$. If φ happens to be a rational multiple of π, $r\pi/q$, then the periodicity Δ_p of the oscillating function will be q if $r+q$ is even and $2q$ otherwise. In all other cases, no integer value of Δ will define a period. For $\alpha = \beta = 0$ the oscillation function has a period of three, corresponding to the perfect f.c.c. case. Thus, $s^2 > 0$ corresponds to the average f.c.c. disordered stacking arrangement as reported earlier by Tiwary & Pandey (2007*a*).

Combining equations (32), (33) and (34), the following expression can be obtained,

$$\beta = 1 - 2\exp(-1/\Delta_c)\cos\varphi, \tag{38}$$

which in turn allows the calculation of β from the parameters Δ_c and φ. Knowing β, then α can be calculated from Δ_c using equations (34) and (32).

As a correlation length and φ value can always be defined in any stacking disorder model, equations (38), (34) and (32) formally allow an α and a β value to be found for any stacking disorder, allowing comparison between different types of faulting.

5.2. Case II: $s^2 = 0$

According to equation (30)

$$Z_{s=0} = \overline{\beta}/2, \tag{39}$$

which allows the definition of a correlation length

$$\Delta_{c_{s=0}} = -\frac{1}{\log Z_{s=0}} \tag{40}$$

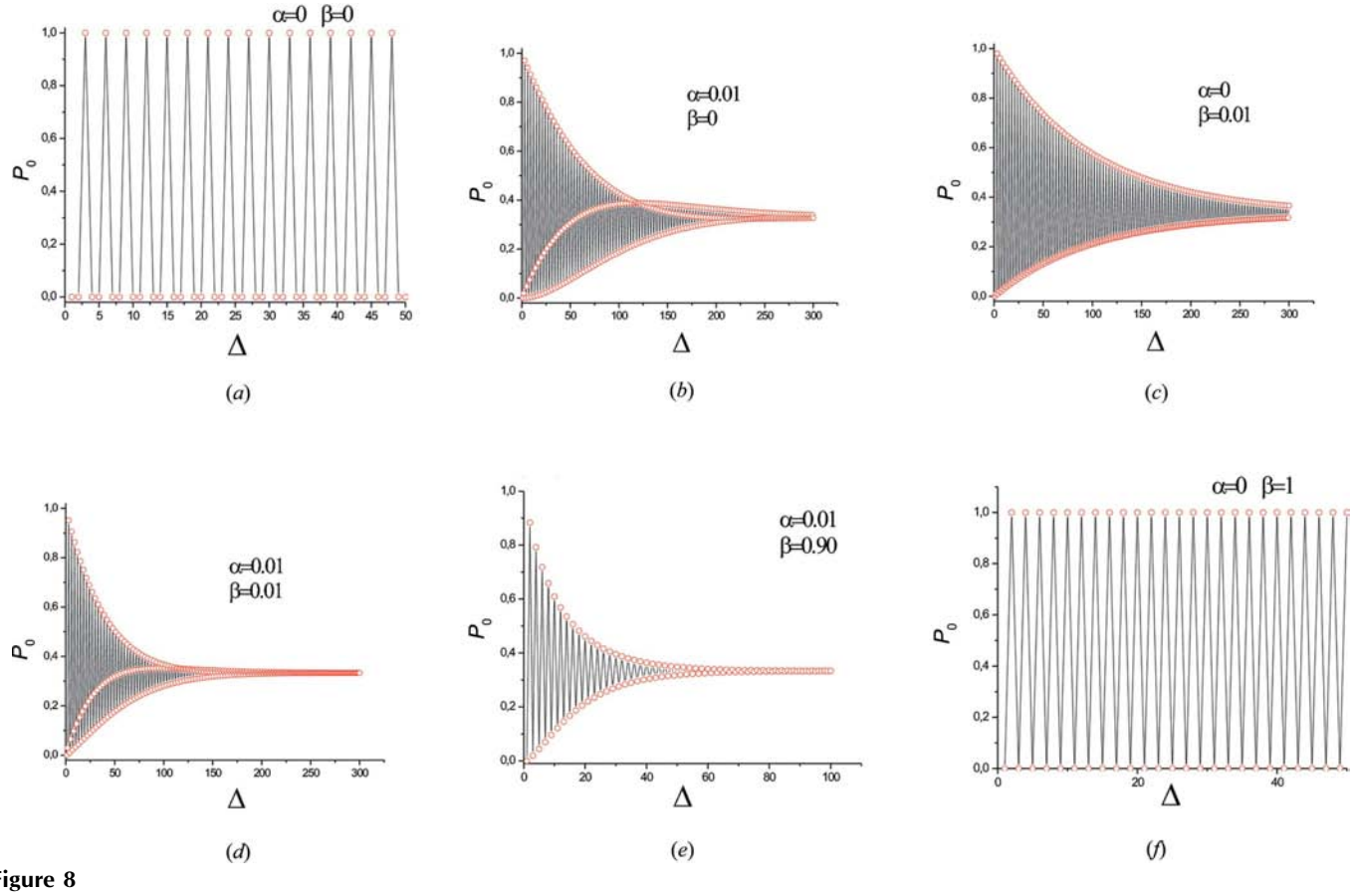

Figure 8
$P_0(\Delta)$ as a function of Δ for (*a*) $\alpha = 0, \beta = 0$; (*b*) $\alpha = 0.01, \beta = 0$; (*c*) $\alpha = 0, \beta = 0.01$; (*d*) $\alpha = 0.01, \beta = 0.01$; (*e*) $\alpha = 0.01, \beta = 0.90$; (*f*) $\alpha = 0, \beta = 1$. P_0 is defined for integer values of Δ; the lines are just a visual aid.

and

$$P_0 = \tfrac{1}{3}\left\{1 + 2(-1)^{\Delta}\left[1 + \left(\frac{\beta}{1-\beta}\right)\right]\exp(-\Delta/\Delta_{c_{s=0}})\right\}. \quad (41)$$

The P_0 function given by equation (41) has a different nature to that of (31). Even if an exponential decaying term with characteristic length $\Delta_{c_{s=0}}$ can be indicated, now the decaying function is multiplied by a linear function of Δ with a positive slope. The maximum value that can be attained by $\Delta_{c_{s=0}}$ is 1.44, indicating a very disordered stacking arrangement state, which is clearly seen in Fig. 9. The behavior of P_0 does not change significantly with β over all its range of allowed values. The nature of the disorder is the same for all values of α and β such that $s^2 = 0$.

The oscillating term is given by $(-1)^{\Delta}$, which has a periodicity of two for any value $s^2 = 0$; this behavior is completely different to that observed when $s^2 > 0$, where the periodicity changed with the faulting probability. Also, in all cases, the periodicity is already destroyed by the coherence length, which is smaller than two.

5.3. Case III: $s^2 < 0$

In this case

$$P_{\Delta} = \tfrac{1}{3}(-1)^{\Delta}\left[(1 + \beta/s')Z_1^{\Delta} + (1 - \beta/s')Z_2^{\Delta}\right], \quad (42)$$

where $s' = (-s^2)^{1/2}$ and

$$Z_1 = \frac{\overline{\beta} + s'}{2},$$
$$Z_2 = \frac{\overline{\beta} - s'}{2}.$$

Figs. 8(*e*), (*f*) show the P_0 function for different values of α and β. Both terms Z_1 and Z_2 have absolute values less than one and they determine the damping term. Z_1 is always a positive number, while Z_2 can take negative values for increasing α and β. In both cases the absolute values of Z_1 and Z_2 tend to one as $\alpha \to \frac{1}{2}$ and $\beta \to 1$. The term between square brackets is always positive.

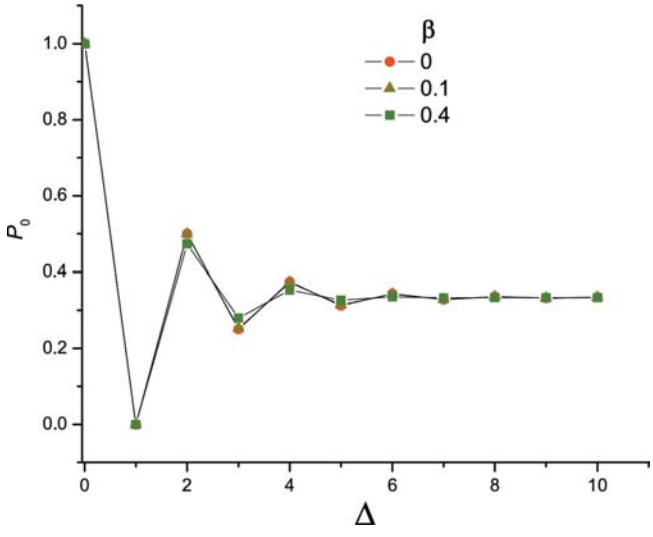

Figure 9
$P_0(\Delta)$ as a function of Δ for different β values and $s^2 = 0$.

The system shows two characteristic length scales,

$$\Delta_{c_1} = -\frac{1}{\log Z_1}$$
$$\Delta_{c_2} = -\frac{1}{\log |Z_2|}.$$

$|Z_2| > Z_1$ and therefore $\Delta_{c_1} > \Delta_{c_2}$.

The oscillating term is similar to the one found in the $s^2 = 0$ case. The oscillating term has a periodicity of two. Once the system has crossed the border from positive to negative s^2, the probability correlation function does not change the behavior of its oscillating term. Thus, $s^2 < 0$ corresponds to the average h.c.p. disordered stacking arrangement also as reported earlier by Tiwary & Pandey (2007*a*).

6. Conclusion

In this paper the independent random model of faulting for close-packed f.c.c. and h.c.p. structures has been analyzed over the whole range of deformation and twin fault probabilities. The defect mechanisms can be described as a probability tree or, equivalently, as a nondeterministic finite state automaton. It has been shown that in earlier treatments by Warren (1969) and Velterop *et al.* (2000) approximations neglect important terms in the layer correlation function. The FSA description allows the derivation of a closed expression for the probability of occurrence of h.c.p. or f.c.c. blocks of length L number of layers within the complete stacking arrangement. Closed mathematical expressions as a function of α and β were also found for the average f.c.c. and h.c.p. block length. The general equation governing the layer pair correlation function $P_0(\Delta)$ was found. For any defect density, the P_0 functions can be described as an oscillating term multiplied by a decaying term. For low faulting probability the oscillating term changes its periodicity towards larger potential polytypes with increasing defect density. This periodicity is destroyed by the prevailing disorder, which can be described by a corresponding characteristic length understood as the 'memory' length of the system. Above a certain boundary curve given by α and β, the underlying periodicity collapses to two layers, signaling the transition to a heavily disordered h.c.p. structure.

The approach used in this paper can be extended to the analysis of other types of independent or even correlated defects described by a finite number of occurrence probabilities.

We would like to thank one of the referees for many suggestions that resulted in a much improved article. One of the authors (EER) acknowledges support from the Humboldt Foundation through an AvH fellowship.

References

Conway, J. H. & Sloane, N. J. A. (1991). *Sphere Packing, Lattices and Groups*. Heidelberg: Springer.

Estevez-Rams, E., Aragon-Fernandez, B., Fuess, H. & Penton-Madrigal, A. (2003). *Phys. Rev. B*, **68**, 064111–064123.
Estevez-Rams, E., Leoni, M., Aragon-Fernandez, B. & Fuess, H. (2003). *Philos. Mag.* **83**, 4045–4057.
Gevers, R. (1954). *Acta Cryst.* **7**, 337–343.
Hendrick, S. B. & Teller, E. (1942). *J. Chem. Phys.* **10**, 147–167.
Hopcroft, J. E. & Ulman, J. D. (1979). *Introduction to Automata Theory, Languages and Computation*. New York: Addison-Wesley.
Jagodzinski, H. (1949*a*). *Acta Cryst.* **2**, 201–207.
Jagodzinski, H. (1949*b*). *Acta Cryst.* **2**, 208–214.
Kakinoki, J. & Komura, Y. (1952). *J. Phys. Soc. Jpn*, **7**, 30–36.
Pandey, D. & Krishna, P. (1977). *J. Phys. D: Appl. Phys.* **10**, 2057–2068.
Patterson, M. S. (1952). *J. Appl. Phys.* **23**, 805–811.
Shrestha, S. P. (1996). *Europhys. Lett.* **34**, 269–274.
Tiwary, P. & Pandey, D. (2007*a*). *Acta Cryst.* A**63**, 289–296.
Tiwary, P. & Pandey, D. (2007*b*). *Acta Cryst.* A**63**, 481–482.
Ustinov, A. I., Storchak, N. M. & Olikhovska, L. O. (2001). *Mater. Sci. Forum*, **378–381**, 154–159.
Varn, D. P. (2001). *Language Extraction from ZnS*. Thesis dissertation, The University of Tennessee, Knoxville, USA.
Varn, D. P. & Canright, G. S. (2001). *Acta Cryst.* A**57**, 4–19.
Varn, D. P., Canright, G. S. & Crutchfield, J. P. (2002). *Phys. Rev. B*, **66**, 174110–174113.
Varn, D. P. & Crutchfield, J. P. (2004). *Phys. Lett. A*, **324**, 299–317.
Velterop, L., Delhez, R., de Keijser, Th. H., Mittemeijer, E. J. & Reefman, D. (2000). *J. Appl. Cryst.* **33**, 296–306.
Verma, A. R. & Krishna, P. (1966). *Polymorphism and Polytypism in Crystals*. New York: Wiley.
Warren, B. E. (1969). *X-ray Diffraction*. New York: Addison-Wesley.
Welberry, T. R. (1985). *Rep. Prog. Phys.* **48**, 1543–1593.
Wilson, A. J. C. (1942). *Proc. R. Soc. London Ser. A*, **180**, 277–285.