4. Theoretical Monte Carlo study of the formation and evolution of defects in the homo-epitaxial growth of SiC

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Abstract. We use three dimensional kinetic Monte Carlo simulations on super-lattices to analyze the nucleation and evolution of several defects during the homoepitaxial growth of 4H silicon carbide. We show also that stacking faults can either extend throughout the entire epilayer (i.e. extended from the substrate up to the surface) or close in dislocation loops, in dependence of the surface kinetics, finding a close connection between the observed behaviors and surface instabilities (step bunching, step meandering). In the case of <11-20> misoriented 4H homoepitaxial growths, we find that all the stacking faults that generate intragap leves can be closed using far from equilibrium growth conditions, i.e. low temperature high growth rate depositions.

Introduction

Silicon carbide (SiC) is a IV-IV compound semiconductor material with a wide band gap, excellent electrical and mechanical properties. From a crystallographic point of view, it is the most famous material for showing
polytypism [1], that is the phenomenon of taking different crystal structures in one-dimensional variation with the same chemical composition. Indeed, the variation of occupation sites along the c-axis (along the Si-C bond) brings about as much as 200 different crystal structures named polytypes. The problem of the polytypes generation is, to a different extent, typical of all close packed crystalline structures grown on the <111> or (0001) surfaces but silicon carbide differs from the other crystals in that the different polytypes have very similar formation energies [2,3,4,5,6,7,8,9] so that it is easy, during the growth process, to have uncontrolled polytype mixing. In the Ramsdell notation, these different crystal structures are represented by the number of Si-C layers in the unit cell and the crystal system (C for cubic, H for hexagonal and R for rhombohedral). The structures of 3C-, 4H- and 6H-SiC, which are the most common polytypes, are shown in Fig.1.

Given the different electrical properties between the different polytypes, it becomes crucial, for device applications, to avoid uncontrolled polytype mixing during crystal growth. This control has been, nowadays, sufficiently reached and the potentials of the material are demonstrated by the production of several high-power devices such as Schottky diodes (which are now commercially

![Figure 1](image)

*Figure 1.* (top) Cross sectional representation of different polytypes. Refining the crystalline structure it is possible to represent each different polytype within the same super-lattice description. (bottom) top-view of the standard (left) and refined (right) crystalline structure.
available by several manufactures) and, more recently, power MOSFET. This improved control is mainly due to the introduction of the “step-controlled epitaxy” [10]. This methodology hinders the generation of other polytypes by creating a step-and-terrace morphology on the surface which forces the epitaxial grown layer to inherit the substrate polytype of the substrate. Indeed, during the growth, adsorbed species migrate on the terrace and are incorporated into the crystal at steps and/or kinks, where the surface potential is lower. At the steps the incorporation site is uniquely determined by the dangling bond configuration, and this fact forces the adsorbed specie to inherit the stacking order of the two layers forming the step and thus, of the substrate. However, there exists another competitive process: i.e. the nucleation on the terraces, which takes place when the excess of adatoms above the equilibrium concentration (also called supersaturation) is high enough. Supersaturation increases at higher deposition rates, therefore, imposing a low growth rate in the experimental setting is the usual empirical prescription to avoid the polycrystalline growth regime. The general assumption is that when the time interval between two successive deposition events in the same terrace area is much larger than the average time which the adatom spends in the terrace before encountering the step (diffusion limited regime), an optimal homoepitaxial growth should occurs. However, as we will show later, low growth rates are not always the best prescription of the growth of high quality, low-roughness, homoepitaxial films.

Several theoretical models can used to the study the homoepitaxial step-flow growth: analytical ones based on the theory of Burton, Cabrera and Frank (BCF) [11] for the dynamic of the step together with the Avrami nucleation theory [12] or numerical ones, either on-lattice (Kinetic Lattice Monte Carlo (KLMC) [13]) or off-lattice (ab-initio [14], molecular dynamics [15] and Monte Carlo [16]). Each method has its own advantages and limits: the BCF theory is based on a continuous description which is capable of easily accessing the time and length scale of the process (microns and seconds) but it misses the atomic scale description, on-lattice methods allow for an atomistic description but fails on describing the generation and evolution of defects because the kinetic particles can only occupy the characteristic lattice sites of the material in study. Finally, off-lattice methods can be successfully used for the study of defects but are unable to reach the time scale of the epitaxial process. More generally, using standard schemes, only particular topics of the growth kinetics can be investigated, since they all fail in describing the correlation between the concurrent aspects characterizing the micro-structural evolution of the system (island nucleation, correlation between the evolution of islands with different symmetry, correlation between
the island and step evolution, interaction between bulk and surface structures, etc.) on the correct time and length scale of the process.

To overcome these limits we developed an innovative method which allows in principle to access to the full complexity of the phenomenology in study. The method extends the concepts of the on-lattice Kinetic Monte Carlo growth simulation by thoroughly refining the lattice (i.e. using additional sites with respect to those of the reference lattice of the material in study [17]). This extension allows to describe all the 200 different polytypes within the same crystalline framework (see Fig.1), to study the generation and evolution of defects at the atomic level (see Fig.2A) and, on the same time, overcome the time scale limits typical of the off-lattice methods. The validity of this extension critically resides on the possibility to identify a limited set of metastable sites which have to be included in the new lattice (named super-lattice). In the case of SiC, this can be done by noting that the most common and stable defects are alterations of the stacking sequence along the [0001] direction (stacking faults, twins, polytype mixing) then, by including, in every [0001] layer, the possibility, for the kinetic particles, to occupy either one of the three possible site symmetries (A,B or C in the Zadhanov notation [15,18], see Fig.1, top) it has been possible to numerically study the behavior of these defects and their interaction with the surface morphology (i.e. with the steps). We called this method Kinetic superlattice Monte Carlo method (KslMC).

The chapter is organized as follows: the numerical methodology is presented in Par.II, in Par.III we discuss the possible surface alterations that, by modifying the step-and-terrace morphology, can change the nucleation or propagation of defects. In Par.VI we compare the Monte Carlo results, in terms of the growth rate limits (study of defects nucleation), with two analytical models and present a possible, controlled, generation of extended defects (3C islands) to generate a polytype conversion (from hexagonal to cubic one). In Par.V we study the propagation of staking faults (i.e. of extended defects) from the substrate into the epitaxial layer, during the growth process (study of defects evolution). Finally, in Par.VI we summarize all the presented results.

**Numerical algorithm**

The basic kinetic description of our KslMC code is similar to the standard on-lattice one [19,20] where the kinetics can be described as a stochastic sequence of elementary atomic displacement events for the Monte Carlo particles. In our case the events belong to the following elementary classes: adsorption from the gas phase, diffusion on the surface and desorption. These particles can represent either single atoms or clusters, such as the Si-C
dimers. We used the latter description (the kinetic particles represent single Si-C dimers oriented along the [0001] direction) because it is sufficient to describe all observed extended defects in the SiC hexagonal polytypes. The methodology can be easily extended (by splitting the kinetic particle into Si and C atoms) to describe single (Si or C) vacancies or stacking faults (SF) and antiphase domains (APD) in the cubic polytype [21].

The probability rule for a kinetic particle, residing at site \( i \), to move to a site \( j \), is based on the contribution of two kinds of bonds forming between the site \( i \) (\( j \)) and the neighbors. In our model up to 26 neighbors can in principle contribute to the adatom stability. The first neighbors (up to 8) contribute to “defective” bonds (see Fig.1b) and the related bonding energy \( E_{\text{defect}} \) (the energy which must be given to the system in order to break the bond) is negative, whereas the second order ones (up to 18) contribute to a “regular” bond and the related bonding energy \( E_{\text{regular}} = E_B \) is positive. The ratio \( R_s = E_{\text{regular}}/E_{\text{defect}} \) is an important parameter of the simulation which is directly related to the energy of the defects. Given that the effective value of \( R_s \) is unknown we have tested different values finding that the qualitative results are insensitive of the exact value of \( R_s \), so that, without loss of generality, we have set for all the presented simulations a value equal to \( R_s = -2 \).

Assuming that the bond strength does not depend on the bond network in a given configuration, we can have, in principle, two possible choices of \( P_{ij} \) which satisfy the detailed balance rule: the Bond-counting Rule (\( BcR \)) and the Kawasaki one (\( KR \)) [22]. Based on the results of ref. [22] we decided to use the \( BcR \) rule, so that the event probability \( P_i \) for each kinetic particle is set as:

\[
P_{ij} = P_i = \Theta_{\text{hop}} \exp \left(-\frac{E_{\text{def}} n'_i + E_{\text{reg}} n'^a_i + R_L E_{\text{LR}}}{k_B T} \right)
\]

where \( k_B \) is the Boltzmann constant, \( T \) is the system temperature, \( n_{ai} \) and \( n_{ri} \) are the number of regular and defective bonds for the initial configuration, \( E_B \) and \( E_{\text{defect}} \) are the associated bond strengths [22,23,24] and \( \Theta_{\text{hop}} \) is the hopping frequency pre-factor.

Following ref. 24 and 25, we include also a long-range interaction term (\( R_{LR} E_{LR} \)), which represents a beyond second-neighbors interaction along the [0001] direction that distinguishes particle energetic between cubic and hexagonal layers (indicated as \( C \) and \( H \) in Fig.1). \( E_{LR} \) represents the strength of this interaction whereas \( R_{LR} \) can be either 1 for cubic (C) or 0 for hexagonal (H) layers (Fig.1). This term generates an asymmetry of the surface diffusivity on the different surface layers during the crystal growth and a splitting of the step kinetics, this, as we will show later, has a critical impact on both the surface morphology (par.III) and defects behavior.
(par.IV). $E_{LR}$ has not been reliably computed nor measured, so we tested different values, in the range [0-0.12] eV (i.e. well below $E_{def}$ and $E_{reg}$).

Based on the experimental results on the growth rate limit of 4H-SiC [26,27,28] we used the following parameters for all the simulations considered:

$$E_{def} = 1\text{eV} \quad E_{reg} = 0.5\text{eV} \quad E_{LR} = [0,0.06] \text{eV} \quad T = [1500,1600] \text{C} \quad \Theta_{HOP} = 1.8 \times 10^{12} \text{ s}^{-1}$$

The initial substrate is made of a sequence of, equally distributed, monoatomic steps (except when the impact of a step bunched surface is considered, as in Par.IV), separated by terraces of width $T_w = h/tg \alpha$, where $h$ is the height of the step and $\alpha$ is the misorientation cut. Furthermore, in order to reduce finite size effects, we used boundary conditions along the growth plane, i.e. along the step direction (y) and perpendicularly to it (x). The most commonly used boundary conditions are the “Periodic Boundary Conditions” (PBCs). However this choice has strong limitations on this particular growth methodology because, using the standard PBCs, the growth is limited to the number of layers of the initial substrate given the fact that the growth on top of the hill cannot be considered part of the growth process being “step-free” (see Fig.2, bottom). Furthermore, during the simulation, the terrace width will “naturally” decrease generating a non-physical variation of the $T_w$ during the growth. Given these two important limitations we have decided to use what we called “Helicoidal Boundary Conditions (HBC)” [22]. This choice assumes the equivalence between the lowermost and uppermost terrace as it

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Figure 2. (top) View of a simulated growth, it is possible to identify: surface adatoms (black circles), island with same symmetry of the corresponding step (black arrows), islands with different symmetry, i.e. possible defects (yellow arrows) and roughening of the step (blue rectangle). bottom) Standard (A) and helicoidal periodic boundary conditions.
should occur in an ideal step-flow, this allows to growth any number of layers irrespective of the number of layers of the initial substrate, which can be kept to the minimum necessary [22].

**Alterations of the surface during epitaxial growth**

Before considering the problem of defect generation and propagation during the growth process we need to analyze the possible surface instabilities that, by altering the step-and-terrace surface structure, impact the behavior of the defects. In fact during the growth the step train can undergo several types of morphological changes that affect the quality on the growing film. Foremost, the formation of macro-steps with multiple bi-layer height (called Step Bunching, SB) and the roughening of the steps (called Local Step Bunching, LSB and step meandering SM in the case of incoherent and coherent roughening respectively, see Fig 3, top, are probably the most critical problems. This is because they induce i) a widening of the terrace widths between the macro-steps thus hindering the step-flow growth, and ii) a roughening of the surface, which can be critical during post-growth processes.

These surface instabilities can have several origins each one simultaneously

![Figure 3](image-url)

*Figure 3. Top*) Surface of a (A) 3C[1-10] and (B) 4H[1-100] homoepitaxial films. *Bottom*) (A) Direction of the bonds with respect to the crystallographic directions and (B) Differences in the stacking sequence of 3C and 4H for the different possible miscut directions.
and concurrently affecting the final morphology of the grown film. We have identified three different possible mechanisms: geometrical, energetic and kinetic one. and used the developed $Ks/MC$ code to independently study each origin and their influences on the different SiC polytypes.

**Geometrical origin**

The geometric surface instability is related to the crystallographic structure of the step-and-terrace surface for the different polytypes and the different miscut directions. Fig.3, bottom shows the Si to C bonds as seen from the [0001], or $<111>$, zone axis. Depending on the orientation of the miscut and on the polytype (cubic or hexagonal) each atom on the step can have either two bonds (either parallel, 3C[1-10], or misaligned, 4H[11-20] with respect to the steps) or one bond (3C[1-10]) or a periodic mix of both (all hexagonal polytypes with miscut along [1-100]).

This leads to different step kinetics and, because of this, different final step morphologies. Specifically for miscut towards the [1-10] ([1-110]) 3C surface we will have linear bunched steps (zig-zag ones) (Fig.4). For hexagonal polytypes we can have either straight bunched steps for miscut towards the [1-100] direction and zig-zag ones for miscut towards the [11-20] the standard one (Fig.4). These same features have been found in the simulated final morphologies [10,29,30,31,32,33]. Furthermore, we have compared the simulated surface roughness of the cubic 3C[-110] and hexagonal 4H[11-20] forms of SiC in the same deposition conditions.

**Figure 4.** Evolution of surface steps in cubic ($a$ and $b$) and hexagonal ($c$ and $d$) polytypes for two different miscut directions.
As can be seen from Fig. 5a, the surface roughness of 4H[11-20] is constantly higher than that of 3C[-110] for all deposition conditions. This shows that if 3C-SiC substrates were available, i.e. if homoepitaxial growth of 3C-SiC were possible, then cubic polytype would allow to give lower roughness compared with the hexagonal ones.

![Figure 5. a) Simulated effect of polytype and deposition rate on the surface roughness evaluated by root mean square (RMS) value of the the height. b) Simulated effect of the ES barrier on the surface roughness. The step bunching mechanisms increases for increasing ES barriers.](image)

**Energetic origin**

In addition to the geometrical origin, which affects the 3C<1-10> and the 4H<1-100> SB can also originate from both energetic and kinetic mechanisms. Indeed, in the framework of continuous-BCF models, it has been demonstrated [34] that an asymmetry of the step impinging coefficient (or, equivalently, of the energy barrier) of adatoms arriving to the two sides of the steps (i.e. from the lower and upper terrace) can induce an instability in the train of steps and, thus, produce step bunching. This is the so called Ehrlich–Schwoebel barrier (ESB). We have studied the surface roughness for a fixed polytype (3C[1-10], to suppress geometrical SB) and deposition conditions, and varying the ESB in the range of [0,0.1] eV. Indeed, this barrier can have a crucial impact on the grown film, not only by increasing the surface roughness (Fig. 5b) but also decreasing the maximum growth rate at which high-quality crystals can be grown (not shown).

We tested also the impact of the long-range term $E_{LR}$, which is critical to describe the surface of 4H[11-20] mis oriented films. As stated in the previous section, 4H[11-20] is not affected by geometrical SB. Yet, SB is commonly seen in homoepitaxial films grown in this miscut direction. A possible explanation for this discrepancy is that, in the case of the [11-20] miscut direction the origin of SB is not the crystallographic step structure but rather...
the diffusion coefficients of adatoms on the different terraces (cubic and hexagonal ones (Fig.1)). In order to verify this hypothesis we have investigated the surface morphology of a 4H[11-20], 4° off grown film as function of the long-range parameter $E_{LR}$ [35]. To this end, following ref.[36], we have analyzed the terrace width distribution ($TWD$) of simulated surfaces after homoepitaxial growths on [11-20] misoriented substrates varying the $E_{LR}$ parameter. The $TWD$ will be single peaked, with the peak located at the initial terrace width, if no step bunching is present [36] whereas step bunching will lead to a reorganization of the surface with a splitting of the distribution: one peak located at small terrace widths, associated to the bunched regions, and one located at large terrace widths (larger than the initial one). As can be seen in Fig.4, the surface morphology, indeed, presents step bunching only when $E_{LR}>0$ thus, verifying the influence of the long-range interaction on the step bunching instability.

**Figure 6.** Pre- and post- growth terrace width distribution (in units of the lattice parameter) as function of the long range interaction $E_{LR}$. A double peak distribution (present for $E_{LR}>0$) is indicative of step bunching (as in ref.[36]).

**Kinetic origin**

The final source of surface instabilities can be kinetic and, more specifically, it can be connected to the interaction of the train of steps with defects on the terraces such as dislocations, downfall or two dimensional defect, either generated during the growth or originating from the substrate. In the $KslMC$ simulations we can observe island nucleation and, consequently, we can investigate the interaction between the defects (islands) and the steps. We find that the defects can reduce the local step velocity inducing step bunching and, in the worse cases, leading to micropipes and carrots. Specifically, in our simulations we have observed that, if the step reaches an island having the same symmetry, a fast reconfiguration of the island particles to the step structure occurs. This increases the lateral roughness of the step potentially leading to LSB (see Fig.3, top). Instead, when the step reaches an island of complementary symmetry two scenarios are possible: either the step imposes a modification on the island type to
reproduce its stacking sequence, i.e. “healing” the defect, or, if the island is too big and consequently too stable, it wraps around it. In the latter case the islands can potentially initiate the vertical growth of a local fault in the stacking sequence (stacking fault generation) and, eventually, generate an extended polycrystalline structure. In both scenarios the encounter strongly modifies the effective step kinetics causing a deformation of the step which, in turn, favors the creation of other defects in the surface.

This mechanism is critical in the chase of large misorientation angles to correctly describe the step-flow to 2D-nucleation transition, as we will show in the next paragraph.

**Defect nucleation**

The principal feature of our KslMC methodology is that it allows to obtain a direct evidence of the failure of the step-flow growth condition in terms of crystal quality or, equivalently, defects nucleation. Indeed, since the step flow growth controls the nucleation and the growth of islands with complementary symmetry with respect to that of the substrate which would lead to the formation of polycrystalline structures (i.e. defects), the mono-to-polycrystalline transition corresponds to the step-flow to two dimensional nucleation transition. In order to coherently identify the transition, we need to define a critical islands density \( c_{\text{defect}} \) above which the proliferation of defects will be irreversible. We found that the critical deposition condition \( Gr_{\text{lim}} \)
(KMC) is not significantly affected by the chosen value of $c_{\text{defect}}$, as long as this value is larger than the fluctuations (without loss of generality we have used $c_{\text{defect}} > 0.02$ [22]). The $Gr_{\text{lim}}$ (KMC) values obtained, showed in Fig.1, as a function of the misorientation cut for a given choice of the energetic parameter can be used to calibrate and test the continuous BCF-like models. We used the value of $Gr_{\text{lim}}$ (KMC) at $\alpha = 1.3^\circ$ degree of 4H[11-20] to calibrate and compare the results of the atomistic approach with the predictions of two types of Burton-Cabrera-Frank (BCF) continuous models to highlight the concurrent mechanisms described by the KslMC with respect to the simpler description given by the continuous models. Furthermore, we applied the KslMC methodology to investigate the possibility to recover polytype conversion, from the hexagonal to cubic polytype (defect engineering).

**Continuous models**

In the continuous models of step flow growth atoms are uniformly deposited with a deposition flux $F_{\text{dep}}$ on a stepped crystal substrate. The steps velocity is assumed sufficiently slow so that, the steps, terminating the terrace at $\pm Tw/2$, can be considered stationary (i.e. no convection term [37], due to fast step motion, is included). It can be easily demonstrated that, consistently to these hypotheses, the steady state adatom density profile can only be effected by the presence of mobile clusters. Considering this result we have generalized the standard BCF model by means of the inclusion of the formation/dissolution of mobile dimers, i.e. assuming zero diffusivity for any cluster larger than a dimer. Following Refs. [38,39] we denote the adatom and dimer densities by $n(x)$ and $N(x)$ and the corresponding flows by $j(x)$ and $J(x)$. The stationary state of the growth can be described by the following set of coupled, non linear, differential equations [40]:

$$
\frac{\partial j(x)}{\partial x} = F - \frac{2z}{\tau_{\text{dep}}} n(x) - \frac{2\sigma}{\rho \tau_{\text{dif}}} n(x)^2 + \frac{2}{\tau_{\text{dif}}} e^{-E_a/k_B T} N(x)
$$

$$
\frac{\partial J(x)}{\partial x} = \frac{z}{\tau_{\text{dep}}} n(x) + \frac{\sigma}{\rho \tau_{\text{dif}}} n(x)^2 - \frac{1}{\tau_{\text{dif}}} e^{-E_a/k_B T} N(x)
$$

$$
\begin{align*}
J(x) &= -D_A \frac{\partial n(x)}{\partial x} \\
J(x) &= -D_D \frac{\partial N(x)}{\partial x}
\end{align*}
$$

where $z$ and $\rho$ are the surface parameters representing the number of neighbors and the surface density, $\sigma$ is the capture number i.e. the probability,
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for two neighbor adatoms, to form a dimer. \( \tau_{dep} = \rho / F \) and \( \tau_{dif}^{-1} = \theta_{hop} \exp(-3E_B / k_B T) \) are the characteristic times describing the deposition rate and the adatom surface diffusion respectively, \( 3E_B \) is the energy barrier for adatom diffusion, \( E_n \) is the dimer binding energy. The second and third terms of the right hand side of Eq.(2) represent the nucleation of the adatoms to form dimers, whereas the fourth term represents the increase of the adatom density due to the dissolution of the dimers. Evaporation of either adatoms or dimers is neglected. The last two equations represent the Fick’s law applied to the adatoms and dimers. Where \( D_A \) and \( D_D \) are the diffusion coefficients of adatoms and dimers respectively which are connected to the bond strength \( E_B \) to be consistent with the Monte Carlo simulations [40]. The parameter \( \gamma \in [0,1] \) incorporates the complex paths leading to dimer diffusion. The results are almost independent of this parameter, so we can set \( \gamma = 1 \). As already noted in the introduction, the continuous models have one unknown variable, the concentration \( n_{crit} \) (or the related \( Gr_{lim} \) value), above which two dimensional nucleation occurs, i.e. the SF-to-2DN will occur when \( n_{max} \), as determined by Eq.3.1, will be greater than \( n_{crit} \) a value which cannot be defined within the theory. We used the \( Ks/MC \) simulations, evidencing a sharp SF-to-2DN transition for large terrace widths, to set the critical conditions. Specifically, we used the \( Gr_{lim}(KMC) \) at the misorientation cut \( \alpha = 1.35^\circ \) (equivalent to a terrace widths of 40\( a \), where \( a \) is the inter-site lattice distance), see Fig.5 caption.

In Fig.8 we show the results of this comparison, we have included also the fit for the standard BCF model, i.e. without dimer formation, which can

![Figure 8. Effect of the misorientation angle on the growth rate limit.](image-url)
be obtained from Eq.(2), keeping only the first term on the right hand side of the equation. In the latter case Eq.3.1 can be solved analytically and we obtain the following equation defining the deposition limit $Gr_{lim}$:

$$Gr_{lim} = 4[Tw^2 + 2Tw/h](n_{crit}D_A)$$

As it can be seen in Fig.5, the agreement between the predicted deposition rate limits ($Gr_{lim}$) is limited only to small misorientation angles (below 3°) (for both continuous models considered), whereas, for larger angles, the two continuous theories predict un-physically large values of $Gr_{lim}$ (one order of magnitude higher than the experimental values of ~ 120 µm/h [41]).

A reason for this discrepancy is that, as shown in the previous paragraph, in the regime of large off-angle cuts, the surface instabilities play a central role on the on-set of two dimensional nucleation altering the standard description of the transition and rendering the BCF theories unable to correctly predict the growth rate limit, i.e., in this regime, the steps cannot be considered any more as straight and isolated objects. More generally, this hypothesis cannot be considered valid whenever the roughness of the steps (the root mean square value, RMS) becomes of the same order of magnitude of half the terrace width (i.e. when $R_{step} = (2\times RMS) / Tw \sim 1$). In order to validate this idea we have calculated this ratio in terms of the off-angle cut applying the KslMC simulations (Fig.9).

As we can see, the region of validity of the continuous models and the region for which $R_{Step} < 1$ completely overlap. This analysis demonstrates that, indeed, the region of validity of the BCF-like theories (as determined in Fig.1, i.e. below ~ 2.5 degrees) is the one for which $R_{Step} < 1$. This fact is also confirmed by the surface morphologies of grown crystals near the SF-to-2DN transition for different terrace widths. As it can be seen in Fig.3, for small terraces there is a strong step-to-step interaction which cannot be, inherently, modeled by means of the BCF-like description (which is based on ideally straight, isolated, stationary steps) independently of the nucleation theory used for the calibration. We have also compared the $Gr_{lim}$ value in terms of $J_A = E_D/k_BT$ or, equivalently, in terms of the system temperature $T$ or material property $E_B$. Specifically we have compared the $R_F = Gr_{lim} (2\alpha_0) / Gr_{lim} (\alpha_0)$ value for different values of $J_A$ fixing the misorientation cut to $\alpha_0 = 4^\circ$. Both the standard and the refined BCF theories predicts $R_F = (2\alpha_0 / \alpha_0) 2=4$, independently of $J_A$ and of the value of $\alpha_0$, whereas, in the case of the homoepitaxial growth of Silicon Carbide, grown through chemical vapor deposition, the experimental value is ~ 1.2 [41,42]. The inset of Fig.9 shows the KMC results. As it can be seen, in the case of the atomistic simulations,
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Figure 9. Left) Dependence of \( R_{\text{step}} = (2 \times \text{RMS}) / T_w \) in terms of the substrate geometry (off-angle). The region of validity of the BCF theory is shown. The simulation parameters are the same of Fig.5. The inset shows the dependence of \( R_F = F_{\text{lim}}(\alpha_0) / F_{\text{lim}}(\alpha_0) \) in terms of \( J_A = E_D / k_B T \) for \( \alpha = 4^\circ \). The BCF-limit (i.e. \( R_F = 4 \) can be obtained for low values of \( J_A \) and cuts with small misorientation angle (i.e. large terrace widths). Right) Snapshots of portions of grown surfaces for two different terrace widths close to the SF-to-2DN transition \((T_w/a = 20\text{ and } Gr = 66\mu m/h, \text{ upper panel and } T_w/a = 6\text{ and } Gr = 66\mu m/h \text{ lower one. The terrace with } (T_w) \text{ before the growth is shown.}

\( R_F(KMC) \) changes with \( J_A \) converging to the continuous result for higher temperature and “soft” semiconductors (small values of \( E_D \)). The value \( J_A^* \) at which \( R_F(KMC) \) depends on the misorientation cut \( \alpha \). More specifically \( J_A^* \) is an increasing function of \( \alpha \), and, as can be deduced from Fig.1, \( J_A^*(\alpha < 2^\circ) > 3.5^\circ \). Finally, we would also like to stress the fact the two continuous theories cannot fit the numerical data in the full range of off-cut angles independently of the diffusion coefficients of the adatoms and dimers or the system temperature. This shows that the origin of the discrepancy between the BCF and the experimental (and Monte Carlo) results does not depend on the inclusion of nucleation events within the continuous theories but, rather, on the specific evolution of the moving steps, which is beyond the capabilities of any 2D step-flow model.

Heteropolytypic growth

In the previous section we investigated the deposition conditions, in terms of growth rate, to hinder the nucleation of defects i.e. to avoid the polycrystalline regime. On the other hand, has been recently experimentally demonstrated [43,44,45,46,47] that it is possible to generate a controlled
nucleation of defects to induce polytype conversion, specifically to growth cubic SiC polytype on hexagonal substrates. However, despite extensive studies and experimental results, the exact mechanism leading to the complete and sudden formation of single domain 3C-SiC is not clearly understood. The general idea is that 3C growth starts with the nucleation of a more or less equal distribution of twinned and untwined (later called SiC-I and SiC-II, see Fig.1 and 10) domains at the β-/α-SiC interface (on the (0001) terraces), this random nucleation prevents a sharp interface and leads to low quality films (Fig.10). Small miscut angles favors 2D nucleation and are supposed to promote polytype transformation, for this reason most of the experimental results have been carried out on nominally on-axis substrates.

We used the KslMC code to investigate the how the substrate properties, specifically polytype (4H and 6H), initial surface morphology (with or without step bunching) and miscut angles, influence the initial nucleation and the subsequent evolution of the 3C–SiC islands. We found (Fig.11 and ref.[48]) that a proper surface preparation of misoriented 6H substrate, aimed to promote controlled step bunching, can lead to a stable configuration of only one exposed layer type (Cubic left or right, see Fig.1). This surface morphology favors only one type of 3C-SiC, leading to single domain 3C crystals. Furthermore, we found that extension of the 3C conversion region strongly depends on the misorientation of the substrate increasing for large miscut angles. This is an unexpected result, given the fact that misoriented substrates are generally used to promote the homopolytypical reproduction of the substrate. This behavior, which is very important from a technological point of view can be explained considering the density and distribution of nucleated islands for the different miscut angles and growth rate considered.

**Figure 10.** Hetero-polytypical growth of 3C on 6H polytype. The substrate is a 4° degree off <11-20> misorientated, silicon face, 6H polytype with ideal (step-bunch free) surface. Note the different 3C domains (called I and II) nucleated on the different terraces. Red arrows indicate the annihilation of a 3C-II domain in favour of 3C-II neighbouring ones.
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Figure 11. Different growth regimes (polycrystalline growth (Poly), 3C-SiC single domain growths (3C-G.) and step flow growth (S.F.) for different growth rates and miscut angles of the substrate. Long range interaction term varies between 0.06 eV and 0.12 eV.

Indeed, for small miscut angles, we have, in the 3C-G. is regime, a generation of large, stable, islands which compete with each other (through merging and Ostwald ripening) and with the steps. If the deposition rate is sufficiently low nucleation leads to well aligned 3C islands and to the generation of single domain 3C films. On the other hand, for large miscut angles, i.e. for a high density of surface steps, the distribution of nucleated islands, which are laterally confined by the steps, which acts as perfect sinks[49], will be mainly composed of many small, unstable, clusters with fast kinetics. These cluster will rapidly interact with each other and with the steps. This fast kinetics and competition leads to the generation of 3C films for a large range of growth rates.

The range of 3C growths corresponds to the defective step-flow or, equivalently, to the local step bunching regime. The extension of this regime, as shown in Par.II increases increasing the miscut angle, this explains the large process window for the large miscut angles. For these reasons, the use of a misoriented 6H-SiC substrates with a large ([4°-10°] degree off) miscut angles and a proper pre-growth process, that leads to a full step-bunched surface, can be the key for the growth of high quality, twin free, 3C-SiC films.

Defects propagation

In the previous paragraph we have analyzed the different growth mechanisms: pure step flow, defective step flow, polycrystalline and 3C-conversion on the basis of the deposition conditions and substrate properties. All these analysis focus on defects nucleation whereas, another important feature of our KslMC code, is the capability to selectively include defects directly into the substrate and analyze their behavior during the growth, i.e. to
Figure 12. Evolution of a SL-SSF defect. The different colors represent the three different particles sites: A, B and C. (top) the initial configuration of the substrate, Lbox and Limit are, respectively, the computational box and the initial defect extension along the [-12-10] direction, (bottom) the system after a short-time evolution. The presence of the defect is visible underneath the surface.

Figure 13. (top) Comparison between the crystallographic structure of a defect-free crystal (left) and a crystal with a single-layer stacking fault (right), as seen from the <1-100> zone axis. (bottom) Schematic evolution of the two defects as seen from the surface. The plus (+) and minus (-) signs indicate the kinetics of the steps on the basis of their local crystallographic structure. Specifically, the (+1,-1,+1,-1) and the (-1,+1,-1,+1) defects expose different step configurations at the surface. This fact will lead to different behaviors.
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Figure 14. Plan view of the growing defect for different growth rates. The dislocation cores tend to be aligned to the [11-20], equivalent, directions. We note that, at the slower growth rate of $Gr = 5 \mu m / h$ the defect never closes, i.e. the defect length $L_{def}$ exceeds the computational box.

analyze the mechanisms behind defect propagation rather than defect nucleation. This aspect is crucial because it is known that the substrate (either 4H or 6H) can be highly defective so that the limitations on the growth of high quality epilayers can reside on the propagation of the defects rather than their nucleation.

Using the KslMC code we have simulated the homoepitaxial growth of a 4H-SiC polytype with steps either perpendicular to the [11-20] and [1-100] directions (i.e. corresponding to miscuts towards the [11-20] direction, the standard one, and towards the [1-100] direction). In contrast with the previously presented results, we consider defective substrates containing staking faults (Fig.12) and evaluate their evolutions in terms of the growth conditions and staking fault type (see ref.[5] for a description of the main SFs observed in 4H epilayers). We find that, in dependence of the deposition conditions, the defects can either extend throughout the entire epilayer (i.e. extended from the substrate up to the surface) or close in dislocation loops. This different behavior can be explained in terms of a surface kinetic competition between the defect and the surrounding crystal: if the local growth rate of the defect is larger compared with that of the perfect crystal the defect will expands, otherwise it will closes. We begin by considering the $<1-100>$ miscut direction because, as shown in par.II, in this direction the exposed steps have a periodically sequence of different crystallographic
structures so it possible to clearly identify a correlation between the behavior of the defect and the, altered, step kinetics.

Evolution of stacking faults for <1-100> miscut direction

We start our analysis from a 4H-SiC, silicon face, 8° degree off <1-100> misoriented substrate containing one (+1,−1, +1,−1) stacking fault (Hagg’s notation [5], see Fig.2 top, left). This defect can be generated by two opposite shears of the crystal along the [1-100] shear direction. Only two, non-equivalent, SL-SSFs exist in the 4H polytype: the (+1,−1, +1,−1) and the (−1, +1,−1, +1), see Fig.13, top.

These defects can be referred as a “single layer Shockley-type stacking fault” (SL-SSF), having an extension of one single bi-layer along the direction normal to the basal plane ([0001]). In Fig.12 the final, i.e. post growth, morphology of the defect, as seen from the [0001] zone axis (i.e. in plan view), is shown. Note that the initial extension of the defect along the [-12-10] direction ($L_{init}$) and the misorientation cut of the substrate ($\alpha = 8^\circ$) are the same for all the simulations. Specifically, $L_{init} = L_{box}/2$ with this condition the extensions of the defective and non-defective regions ($L_{reg} = L_{box} - L_{init}$) are equal (see also Fig.10). Furthermore, the range of explored $Gr$ is below the step-flow to 2D island transition, i.e. below the growth rate limit $Gr_{lim} (\alpha = 8^\circ) \sim 120 \mu m / h$ [50]).

As can be seen in Fig.14, the length of the defect on the basal plane ($L_{def}$) decreases by increasing the growth rate ($Gr$), which means that the propagation of the defect can be hindered by increasing (not decreasing, as one would expect based on the intuitive idea that low deposition rates are the key for high quality films) the growth rate.

In Fig.15 we extend this analysis to study the impact of the miscut angle and temperature on the propagation of the SL-SSF, finding that the extension of the defect along the [0001] direction ($L_{0001} = L_{def} \times \sin(\alpha)$), which is directly related to the density of the defect on the film surface, increases for increasing miscut and temperatures.

These unexpected results can be interpreted in terms of a surface kinetic competition. That is, if the kinetic of the defect, i.e. its efficiency in capturing adatoms on the surface, is higher with respect to that of the surrounding steps the defect will tend to expand during the growth, otherwise it will shrink and, for long enough times, it will close into a dislocation loop (see Fig.14, bottom). The ratio between the lateral effective velocity of the defect and the lateral velocity of the surrounding steps, $R = \frac{v_{defect}}{v_{step}}$, specifies the evolution of the defect during the growth. In general, $R$ depends on the
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Figure 15. Defect extension along the [0001] direction ($L_{[0001]}$) as function of the different deposition parameters.

crystallographic structure of the exposed surface and on the deposition conditions. For the specific defect considered, (+1,−1, +1,−1), we obtain $R \leq 1$, for $Gr \geq 20 \mu m/h$.

To verify the close connection between surface kinetic and defect evolution we studied the other SL-SSF in 4H, i.e. the (−1, +1,−1, +1) SF (see Fig.13, top, right). This defect has the same bulk structure of the (+1,−1, +1,−1) and, thus, the same bulk formation energy ($\Delta H \sim 30 mJ/m^2[2]$) but exposes an opposite surface structure and, because of this, if the surface kinetics competition mechanisms is correct, we can expect to have an opposite behavior with respect the (−1, +1,−1, +1) stacking fault. The results of the Monte Carlo simulations, in terms of the defect morphology (Fig.17) and the $L_{def}(Gr,T)$ dependence (Fig.16), agree very well with this scenario.

The connection between $L_{def}$ (or, equivalently $L_{[0001]}$) and the SF-to-2DN transition can be strengthened introducing an effective “order parameter” $\theta = (Gr_{lim} − Gr) / Gr_{lim}$ and assuming a functional dependence for $L_{def} \approx L_{def}(\theta)$ close to the transition. Then, we can expand $L_{def}(Gr, \alpha, T)$ in terms of the order parameter and obtain [51]:
Figure 16. Defect extension along the [0001] direction as function of the growth rate and temperature of the (-1,+1,-1,+1) defect. (left) $L_{[0001]}$ for different growth rates, with $T=1800^\circ$C and $\alpha=8^\circ$. The temperature parameter used to study the growth dependency has been increased to overcome the limits of the computation box used (i.e. to allow for defect closure). (right) $L_{[0001]}$ for different temperature with $\alpha=8^\circ$ and $Gr=50\mu$m/h. Red lines indicate simulations for which the defect has exceeded the computational box.

Figure 17. Plan view of the two single layer stacking faults for the same growth condition. As can be seen, for the same deposition conditions, (top) (+1,−1, +1,−1) is a closing defect whereas (bottom) (−1, +1,−1, +1) is an expanding one.

\[
\delta L_{\text{def}} / \delta \alpha = (\delta L_{\text{def}} / \delta \theta)(\delta \theta / \delta Gr_{\text{lim}})(\delta Gr_{\text{lim}} / \delta \alpha) = \left[ (> 0)(Gr / Gr_{\text{lim}}^2 > 0)(> 0) \right] > 0
\]
\[
\delta L_{\text{def}} / \delta T = (\delta L_{\text{def}} / \delta \theta)(\delta \theta / \delta Gr_{\text{lim}})(\delta Gr_{\text{lim}} / \delta T) = \left[ (> 0)(Gr / Gr_{\text{lim}}^2 > 0)(> 0) \right] > 0
\]

The inequalities: $\delta Gr_{\text{lim}} / \delta \alpha > 0$ and $\delta Gr_{\text{lim}} / \delta T > 0$ are related to the properties SF-to-2DN transition with respect to the growth parameters and have been proven both numerically [51] and experimentally[52].
These relations show that the defects, close to the SF-to-2DN transition can be divided into two classes: **class I** defects will tend to close near the transition (i.e. for out-of-equilibrium conditions: low temperature and high growth rates), whereas **class II** defects will tend to close away from it. The only basic assumption is that the defect induces a local different growth kinetics. In the specific case of the two SL-SSFs the different growth kinetics can be attributed to the different crystallographic structures of the exposed steps [53]. In the next section we will show that, in the <11-20> miscut direction the long range term $E_{LR}$ is responsible for the splitting of the kinetics of the defect with respect to the defect-free crystal.

Finally, it is worth noting that, in the case of hetero-epitaxial growths of 3C silicon carbide on (111) silicon both twins and stacking faults will have different crystallographic structures, i.e. different kinetics and will either tend to expand or close during hetero-epitaxial growths. Furthermore, their kinetics, being related to the local crystallographic structure, will be reversed when switching the growing surface from the (abc) growth plane to the (-a-b-c) plane. This is the basic mechanism for stacking faults reduction within the “switch-back epitaxy” methodology [54].

**Evolution of stacking faults <11-20> miscut direction**

In the <11-20> miscut direction, as presented in par.II the exposed steps have all the same crystallographic structure so that, to correctly describe the, experimentally observed, roughening of the surface we included the long range interaction term $E_{LR}$ which is responsible for a splitting of the adatom surface kinetics, and as a consequence of the step kinetics. This splitting of the step kinetics, influencing also the surface roughening, is also the key for the evolution of the stacking faults in this miscut direction.

In order to study their behavior we considered several types of SFs: (4,4) (6,2) (7,1) and (10,2) [2,3,4,5]. All SF types considered can be referred as “cubic inclusions” [5], i.e. they all expose a larger number of cubic steps on the surface compared to that present in the perfect crystal. As noted before, the crystallographic structure of the steps for miscut along the [11-20] direction is always the same so that the steps have a tendency to swing, generating a zig-zag feature (see Fig.4d) but they do not lead to a step bunched surface unless the long range interaction term $E_{LR}$ is considered.

In Fig.18 the size (number of particles) of the defect during the growth is shown for different values of the $E_{LR}$ parameter in the case of the (4,4) stacking fault. As it can be seen, the defect initially propagates during the film growth (the defect extension increases with the film thickness) and then, suddenly, closes into a dislocation loop. This behaviour changes depending
on the strength of the long-range interaction parameter $E_{LR}$. This result can be ascribed to the lower diffusion coefficient for adatoms migrating on cubic layers with respect to hexagonal ones induced by a higher value of $E_{LR}$. Indeed, this lower diffusion decreases the kinetics of the steps belonging to cubic layers. Since the (4,4) SF generates, on the surface, a sequence of cubic layers [4,5], by increasing the $E_{LR}$ parameter we effectively decrease the kinetics of the defect with respect to the surrounding perfect crystal. This unbalanced kinetics leads to the closure of the defect during the growth process [55]. The same behaviour (not shown) has been observed also for the (6,2) (7,1) and (10,2) “in-grown” stacking faults. It is noteworthy that the considered SF closes even for a $E_{LR}=0$eV value. This is due to the specific growth conditions used for the simulations (high temperature, low growth rate) which increase the probability of defect closure, consistent with what experimentally found in ref. [51]. Considering the fact that the surface kinetics of all cubic inclusions ((1,3), (4,4), (6,2), (7,1) etc.) is governed by the single parameter $E_{LR}$, and that by increasing this parameter the kinetics of the cubic steps decreases with respect to that of the hexagonal ones (being $D_H/D_C = \exp(E_{LR}/k_B T)$) all the cubic stacking faults, in <11-20> misoriented films, will behave as classI defects. As a consequence, high growth rates and low temperatures can be considered the general prescriptions for the growth of high quality films, reducing all that stacking faults. These extended

![Figure 18. Evolution of a (4,4) SF as function of the grown film (number of layers) for different values of $E_{LR}$. The same qualitative behaviour has been found also for the (6,2), (7,1) and (10,2) SFs.](image-url)
defects, by generating intra-gap levels inside the 4H band structure, lead to carrier trapping and act as recombination or scattering centres degrading the overall transport properties of the films.

**Conclusions**

This work meets a requirement; i.e. the comprehensive extension of the kinetic simulations dedicated to epitaxial growth of close packed crystalline structures with an extensive applications to the 4H silicon carbide one. The motivation was the observation that, in spite of an accurate mapping of the energetic, conventional kinetics approaches can address only single or limited aspects of the complex phenomenology, characterizing these growth processes. This is a structural limit of other simulation approach and we can overcome it only releasing the restriction of using the conventional lattice of the material in study as the reference lattice of the stochastic method. Indeed, we have demonstrated that, for the case of hexagonal silicon carbide, we can formulate an efficient and reliable kinetic Monte Carlo on superlattices ($KsLMC$), by including in the, refined, lattice all the three category of highly symmetric sites (usually called A, B, C) which characterize the one dimensional stacking of the close packed lattice. Our code aims at investigating all the concurrent aspects characterizing the micro-structural evolution of the system (island nucleation, correlation between the evolution of islands with different symmetry, correlation between the island and step evolution, interaction between bulk and surface structures) on the

![Diagram](image)

**Figure 19.** Summary of the connections between deposition conditions growth mechanisms and defect typologies, as deduced by the $KsLMC$ simulations.
correct time scale of the process. Specifically, the impact of the islands nucleation, which preferentially occurs in system with large terraces in the high deposition regime, is correctly evaluated since islands of the two symmetries complementary to the terrace could nucleate and evolve. Islands with the same symmetry of the step do not alter substantially the step effective kinetics since they are easily incorporated by the step. The encounter between the step and islands with different symmetry strongly modifies the effective step kinetics, although the interaction between the two structures tends to change the symmetry of the adatom bounded to the island. Indeed, the encounter event could cause a strong deformation of the step (in the case of relatively small islands) or the stabilization of a region with complementary symmetry (in the case of large islands) on the exposed terrace and the consequent growth of a polycrystalline structure.

Besides islands nucleation we demonstrated that our approach allows also to consider the other mechanisms, namely the Global Step Bunching and the Local Step Bunching, which can lead to a progressive deterioration of the epitaxial quality of the growing films (eventually leading to a fault in the stacking order). The point like (LSB) and the extended (GSB) encounter between two or more adjacent steps show the proper morphological character and can be investigated in dependence of the substrate geometry and the deposition conditions. Their relevance is proper of the wide angle (small terrace) substrate geometry. However, maybe the more relevant feature of our code is the concurrent access to all the degradation mechanism of the stacking order so that the correlation between, island nucleation, LBS and GSB can be rightly investigated (e.g. GBS event could lead to the enlargement of some terraces boosting island nucleation, or nucleation events of small island can deform the steps, boosting LSB and so on). Finally, we can also characterize the generation of point like defects, denoting the Defective Epitaxial Growth Regime, which is a fundamental parameter for the quality of the experimentally grown epitaxial films. Fig.19 shows a summary of the simulation results.

References