THERMODYNAMICS AND KINETICS OF NANOCLUSTER FORMATION ON SEMICONDUCTOR SURFACES: THE EXAMPLE OF Si GROWTH ON SiC(0001)

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Abstract

The molecular beam epitaxial growth of Si on SiC is investigated as a prototypical system for the formation of nanoclusters on semiconductor surfaces without interface intermixing effects. The investigation clearly indicates the presence of both equilibrium and non-equilibrium effects with regard to the island formation. The existence of an equilibrium wetting layer thickness d_{equ} *independent from deposition rate R and temperature T is shown from the wetting behaviour of Si on SiC(0001) associated with a change in the surface reconstruction. After 3D island formation* d_{equ} *is still present between the 3D-islands, what clearly confirms that the growth mode of Si is Stranski-Krastanov. The equilibrium wetting layer thickness is well described by equilibrium theory of heteroepitaxial growth considering strain as well as interface effects. At* $d > d_{equ}$ *and conditions that are close to the equilibrium surplus Si atoms always aggregate instantly into 3D islands. Under dynamical conditions of growth far from equilibrium, the range of 2D layer-by-layer growth can be extended to an excess wetting layer thickness* (d_{crit} - d_{equ})*. Investigation of* (d_{crit} - d_{equ}) *as function of R and T indicated a transition from thermodynamically to kinetically controlled regimes of 2D-3D transition by a change in deposition condition. Furthermore, the 2D-3D transition initially starts with the formation a larger 2D islands and the simultaneous formation of smaller 3D islands, which likely acts as precursors for the formation and growth of larger 3D islands. For the growth of islands during further deposition surface mass transport of adatoms is rate controlling. Post-deposition annealing of the highly stressed excess wetting layer* ($d > d_{equ}$) *gives rise to 3D island nucleation after deposition was stopped, what could be a long lasting process because adatoms stem mainly from the wetting layer decomposition.*

Keywords: *Stranski-Krastanov growth, molecular beam epitaxial growth, Silicon, nanocluster*

1. INTRODUCTION

The formation of nanocrystallites (dots) in a spontaneous process on semiconductor surfaces during epitaxial growth is still a subject of investigation [1,2]. This process is mostly based on a growth mode transition (Stranski-Krastanov growth (SK)) during heteroepitaxy. In the SK growth process layers grow initially layer-by-layer (2D). During the heteroepitaxial growth strain is accumulated into the layer and after a critical thickness (d_{crit}) is reached rapid formation of 3D-islands occurs as a mechanism to relax the strain. In many cases the growth mode transition is well described by equilibrium theories of heteroepitaxial growth, where the equilibrium thickness d_{equ} equals to d_{crit} for 2D-3D transition. However, kinetic limitations on island nucleation and adatom mobility can allow the wetting layer to become thicker as predicted for strain relaxation by islanding ($d_{crit} > d_{equ}$). This is reported especially for systems exhibiting larger lattice mismatch, such as Ge on Si [3], InAs on GaAs [4], InP on GaAs [5] or GaSb on GaAs [6]. Since kinetic limitation may result in different final island morphologies (islands size and spatial distribution) the investigation of the dynamic processes towards the clustered equilibrium structure is important.

In this work, the thermodynamics and kinetics of nanocluster formation during Stranski-Krastanov (SK) epitaxial growth of Si on SiC(0001) is reviewed and updated with respect to new consideration. This material system exhibits a large lattice mismatch of 20 %. Mostly systems exhibiting such strong mismatch are not considered so far for Stranski-Krastanov mode of growth and, therefore, investigated only to a small extend [7-10]. However, since there is no strain-driven intermixing effect as, for example, observed for Si/Ge [1,11] or III-V compounds [12], Si on SiC is a prototypical system for investigation of wetting behavior and the growth mode transition within a wide range of growth parameters, such as temperature (T) and deposition rate (R). This has already been demonstrated in earlier investigation for example with regard to the wetting behavior of Si on SiC(0001) [13]. Since the wetting behavior is important for the understanding of the thermodynamics of SK growth mode transition the results of these earlier investigations are summarized und included in this paper, too.

For high enough misfit Stranski–Krastanov growth mode transition can be described by equilibrium consideration but probably only for larger misfits [14]. It will demonstrate that this is also valid for the growth of Si on SiC(0001). Moreover, several kinetics mechanisms for wetting layer formation have been proposed. Snyder et al. [15] suggested a kinetically controlled critical thickness of the wetting layers. For this purpose, they segmented the 2D film into regions of lateral size equal to the surface diffusion length $L = \sqrt{D \cdot \tau}$, where D is the diffusion constant and τ the inverse of the deposition rate. By minimising the energy of the system, they found a metastable growth of 2D strained films for $d < d_{crit}$ and the formation of coherent islands for $d > d_{crit}$, with $d_{crit} \propto 1/L$. Consequently, in the thermodynamic limit where $L \rightarrow \infty$, d_{crit} will effectively approach zero [14]. This model was used for earlier studies of the Si growth on SiC(0001) [16]. However, meanwhile we know that this does not correspond exactly to what is found experimentally, since the equilibrium wetting layer is still visible after 3D island formation for the growth of Si on SiC(0001) even under near equilibrium condition (high T , low R) [13].

Another model by Dubrovskii et al. [17] is based on the assumption that the 2D-3D transition is a first-order phase transition. They showed that 3D islanding reduces the free energy of the hetero-system when the wetting layer thickness increases beyond its equilibrium value d_{equ}. 3D-island, as a first-order phase transformation, starts from the metastable strained film, and the new stable phase (the 3D-islands) nucleates by thermal fluctuations. The supersaturation needed for 3D island nucleation is the superstress stored by in the metastable layer ($d > d_{equ}$), which is defined in terms of $\zeta \equiv d/d_{equ} -1$ [18]. The “supersaturation” is a measurement of the wetting layers instability, which are consumed to form the 3D islands after transition. The time evolution of the superstress is thus driven by the consumption of atoms from the wetting layer and the impinging atoms onto the surface. It was thus possible to use the classical nucleation theory to calculate the time evolution of the island size distribution and the wetting layer thickness.

In the present study, kinetics of the growth mode transition is reinvestigated using data obtained previously [16] considering the superstress as the driving force for the 2D-3D transition. This reinvestigation likely results in a closer and more accurate insight into the SK growth of Si on SiC(0001). Further investigations are related to the stages of the 2D-3D transition, the subsequent 3D island growth during the further Si deposition and the post-growth evolution of the island on the metastable wetting layer after depositon was stopped.

Investigations were performed using reflection high-energy electron diffraction (RHEED) during molecular beam epitaxial growth of Si on SiC(0001). RHEED is one of the most powerful techniques for monitoring epitaxial growth of thin layers in vacuum. The feature of grazing incidence makes RHEED very sensitive to the growing surface with a resolution of up to submonolayers. The technique can reveal almost instantaneously changes in the coverage, surface structure and morphology. Furthermore, it is well established that the two-dimensional(2D) growth regime is associated with streaky RHEED patterns, whereas 3D growth is characterized by spotty patterns due to transmission diffraction through 3D islands. This makes it easy to investigate the transition towards island formation during the SK growth regime in situ and in real time by measuring intensity changes. The challenge is the interpretation of the results, which is quite complicated.

2. RESULTS AND DISCUSSION

2.1. Wetting layer formation and 2D-3D transition

Figure 1 presents the evolution of the RHEED specular (00)-beam intensity measured during the growth of Si on SiC(0001) at a temperature T = 920 K and using a depositon rate R = 0.04 monolayer(ML)/s. The intensity of the Si (222) bulk spot due to transmission diffraction from the 3D-Si islands is also shown. Two maxima in the specular beam intensity (or two oscillations) appear before Si bulk spots are visible in the RHEED pattern. The maxima correspond to about 1 and 1.4 ML Si coverage, where the coverage of 0 ML corresponds by definition to stoichiometric Si-SiC(0001). The oscillations of the (00)-beam intensity indicate a growth mode via 2D-nucleation. This implies that Si grows initially in 2D mode, i.e. it wets the SiC(0001) surface.

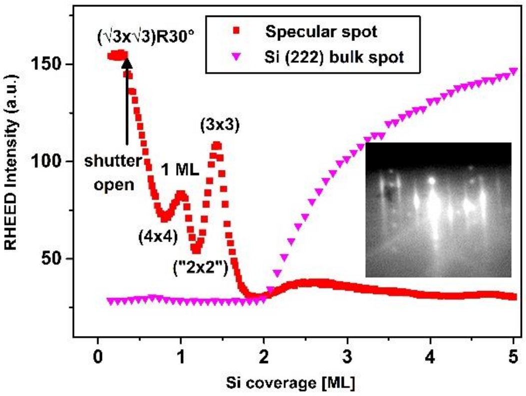


Fig. 1. RHEED specular beam and (222) Si bulk spot intensity during the growth of Si on SiC(0001). Inset shows RHEED diffraction pattern after Si island formation, exhibiting (3x3)-surface superstructure streaks and 3D-Si diffraction simultaneously.

Some surface superstructures have been observed in the RHEED pattern during the 2D growth, as indicated in Fig.1. The coverage corresponding to a given superstructure was determined by the appearance of the intensity maximum of fractional-order RHEED spots related to this superstructure during the Si deposition (not shown here). The growth was started on a SiC surface exhibiting a (√3x√3)R30° structure formed by an ordered 1/3 ML Si adlayer coverage, the superstructure transformed to (4x4) with a maximum intensity of the fractional-order RHEED spots at around 0.75 ML Si adlayer coverage.

During further increase in coverage, the superstructure changes to (2x2) with a maximum intensity at about 1.2 ML. In the diffraction pattern of the (2x2) structure several fine additional fractional-order Laue zones were observed indicating that this is not really a (2x2) periodicity; for that reason, we refer to this structure as (“2x2”). Based on low-energy electron diffraction and scanning tunnelling microscopy studies a structure model has been proposed corresponding to the Si coverage of 34/28 ML (1.21 ML) [19].

At the coverage of ca. 1.4 ML, corresponding to the second maximum of the (00)-spot intensity, there is also observed a maximum of fractional-order spot intensity related to a (3x3) surface structure. The full Si adlayer coverage on the Si-terminated SiC(0001) surface derived from the (3x3) structure model should be 13/9 ML (1.44 ML) [20,21], in excellent agreement with the Si coverage obtained experimentally. During further Si deposition, no other surface superstructure was observed. At a coverage of 1 ML corresponding to the first maximum in the RHEED specular beam intensity, the (4x4) and (“2x2”) coexist indicating that the new superstructure develops by ripening of larger (“2x2”) patches rather than by coalescence of smaller ones. The same can be said about the transition from (“2x2”) to (3x3). The appearance of superstructures at certain coverages was independent on R indicating that the stepwise Si saturation of the SiC(0001) Si-face was linear. However, far away from equilibrium, i.e. at higher R or lower T, the first oscillation of the RHEED specular beam intensity at 1 ML was not observed and, moreover, the (4x4) and (2x2) structures did not appear.

At Si coverage > 1.4 ML, surplus Si atoms always aggregate instantly into 3D-islands under equilibrium-like conditions (low R and high T). The inset of Fig. 1 shows the diffraction pattern after Si island formation exhibiting 3D transmission diffraction through Si islands. Moreover, diffraction from the flat (3x3) reconstructed SiC surface areas, which are not covered by Si islands, is also visible. This indicates clearly a Stranski-Krastanov-like growth mode of Si on SiC(0001). The (3x3) structure undergoes a reversible phase transition to a disordered (“1x1”) structure only ≈ 1300 K.[22] Therefore, it can be suggested, that 1.4 ML Si adlayer arranged in a (3x3) surface structure is thermodynamically stable on SiC(0001) at T < 1300 K and hence represents the equilibrium wetting layer thickness (d_{equ}).

Fig. 1 also indicates that Si can grow 2D at coverage larger than the equilibrium wetting layer thickness (hereinafter referred to as the critical wetting layer (d_{cri}) at which 2D-3D transition occurs); what is about 2.1 ML for the conditions in Fig.1. At lower T and higher R the metastable wetting layer could be even thicker. Fig.2 shows the RHEED intensity profiles obtained during Si deposition on the SiC(0001) covered initially by the (3x3) surface superstructure (1.4 ML Si). Two intensity maxima at a coverage corresponding to 2 and 3 ML are visible, before at ≈ 3.5 ML coverage 3D islanding becomes visible by a strong increase in Si bulk spot intensity. This clearly indicates that transition towards 3D islanding is significantly delayed at higher R and/or low T.

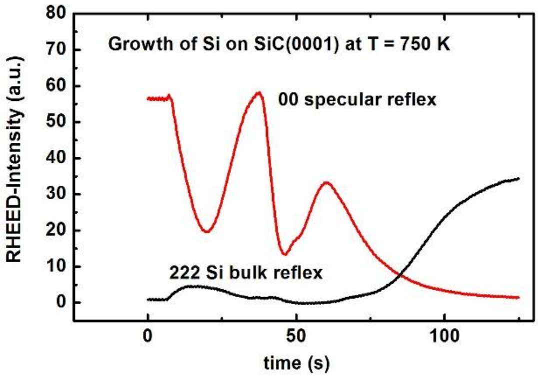


Fig. 2. RHEED intensity profiles of the (00) specular and the (222) Si bulk reflex obtained during epitaxial growth of Si on SiC at T=750 K and R=0.04 ML/s.

2.2. Thermodynamics of wetting layer stability

From the thermodynamic point of view, the obtained equilibrium wetting layer thickness of d_{equ} = 1.4 ML seems to be surprisingly large in terms of the large lattice misfit of 20 % between Si and SiC. In this case it should be possible to describe the growth mode transition by equilibrium consideration [14]. Usually, growth modes are solely classified based on thermodynamic consideration. In such consideration, a particular growth mode is determined by the relationship between the interfacial free energy between the growing layer and the substrate, the surface free energy of the layer and the surface free energy of the substrate. As predicted by E. Bauer [23], a necessary condition for wetting of the substrate by the layer material (2D layer-by-layer growth) is that the surface free energy σ_L of the layer plus the interfacial free energy σ_i is lower than the surface free energy σ_S of the substrate:

$$\Delta\sigma = \sigma_L + \sigma_{\mathrm{i}} - \sigma_S \leq 0 \tag{1}$$

Because the surface free energy of Si(111) ($\sigma_L = 1.02$ J/m^2) [24] is much smaller than that of SiC(0001) ($\sigma_S = 1.767$ J/m^2) [24], it can be suggested that Si on SiC(0001) grow in a layer-by-layer mode. Considering the interface energy, this trend will be even more pronounced. There is a significant energy gain ($\sigma_i = -1.722$ J/m^2) by the Si/SiC(0001) interface formation [25]. This agrees to the experimental results, where wetting of SiC(0001) by Si is clearly observed. Thereby, the Si-rich (3x3) surface structure exhibits the smallest surface energy [20]. However, further increase of Si layer thickness results in the transition to 3D island formation. In this context, we have to consider that the lattice constant of Si and SiC are different by about 20 %. Therefore, the contribution of strain energy has to be taken into account by adding an extra energy term to Equ. (1):

$$\sigma_{\mathrm{Str}} = \frac{E_{\mathrm{el}}}{A} = M \cdot \varepsilon^2 \cdot d \qquad (2).$$

where E_{el}/A is the elastic strain energy per unit area A stored in the thin 2D-layer; M the biaxial modulus and $\varepsilon = (a_S - a_L)/a_L$ the lattice mismatch, where a_S and a_L are the in-plane lattice parameters of the substrate and the layer, respectively. The Biaxial modulus in the {111} plane is given by:

$$M_{111} = \frac{E}{1-\upsilon} = \frac{6c_{44}(c_{11}+2c_{12})}{c_{11}+2c_{12}+4c_{44}} \qquad (3),$$

with E is the Young's modulus, ν is the Poisson's ratio and c_{ij} are the elastic constants. Thereby, the strain energy increases linearly with the number of deposited layers d, as long as the layers grow pseudomorphic:

$$\Delta\sigma = \sigma_L + \sigma_i - \sigma_s + \sigma_{Str}(d) \le 0 \qquad (4).$$

It is suggested that the elastic constants of thin adsorbate layer is the same as for the corresponding values of the bulk material. Moreover, since the elastic constants for Silicon only exhibits week T-dependence [26], biaxial modulus should be nearly constant for the whole T-range investigated. The same should be valid for ε with respect to the lattice constants. With the data of elastic constants given by Bradley at al. [27], the biaxial modulus for Si(111) is M = 229 GPa.

From the aforementioned data, we get a thickness-dependent normalized strain energy of $\sigma = 1.44 \cdot d$ J/m², where d is the layer thickness in monolayers (1 ML = $a_S(\mathrm{Si})/2\sqrt{3}$). The wetting layer thickness for 2D-3D transition can be estimated from the critical condition Δσ = 0 via equation 2 and 3:

$$d = \frac{\sigma_{\mathrm{SiC}} - \sigma_i - \sigma_{\mathrm{Si}}}{M \cdot \varepsilon^2} = \frac{[1.767 + 1.7622 - 1.02]\, J/m^2}{1.44\, \mathrm{J}/m^2} \approx 1.7\, ML \qquad (5).$$

The estimated value of 1.7 ML is close to the value of $d_{equ} \approx 1.4$ ML obtained experimentally under near equilibrium condition (higher T and low R).

However, at lower T and higher R, wetting layers with d ≈ 3.5 ML could be stabilized under dynamic conditions of growth. In this context, we have to consider that growth takes place under conditions of supersaturation, what could be of influence on the growth mode. Despite controversial discussion in literature [28], such analysis can be performed by a thermodynamic analysis within the framework of classical nucleation theory [29]. The free energy change in the formation of the critical nucleus is different in case of 2D and 3D islands with the same number of atoms. Therefore, to estimate the critical conditions for 2D-3D growth mode transition, we can compare the free energy of heterogeneous critical nucleus formation on (111) for the two growth modes. Considering that the growth takes place in a 2D mode, the Gibbs free energy $\Delta G_{2,c}$ of formation of a critical nucleus on a (111) or (0001) surface can be expressed in case of heterogeneous 2D-nucleation by:

$$\Delta G_{2,c} = \frac{b^4 \sigma_l^2}{\Delta\mu - b^2\left(\sqrt{3}/2\right) \cdot \left[\sigma_l + \sigma_i - \sigma_s + \sigma_{Str}(d)\right]} \qquad (6),$$

For heterogeneous 3D-nucleation, the Gibbs free energy $\Delta G_{3,c}$ of critical nucleus formation on the (111) or (0001) surface can be expressed by:

$$\Delta G_{3,c} = \frac{4\sqrt{3} b^4 \sigma_l^2 \mathrm{v}^2 \left[\sigma_l + \sigma_i - \sigma_s + \sigma_{Str}(d)\right]}{\Delta\mu^2} \qquad (7);$$

where b is the first neighbour distance corresponding to the in-plane lattice constant and v is the molecular volume. The value $\Delta\mu = kT\ln(p/p_0)$ is the bulk chemical potential change associated with the molecular change from the vapour to the condensed state, where k is Boltzmann's constant. The ratio p/p_0 corresponds to the so-called supersaturation ratio, where p and p_0 are the vapour pressures above the substrate (corresponding to the supply) and the equilibrium vapour pressure, respectively.

For the same number of atoms $\Delta G_{2,c}$ could be smaller than $\Delta G_{3,c}$ so that 2D nuclei are formed, provided $\Delta\mu$ is sufficiently high. The critical condition for 3D-2D transition can be estimated from $\Delta G_{2,c} = \Delta G_{3,c}$. That compares to the situation when the height of the 3D nucleus becomes equal to that of one monolayer [pseudo 2D growth]. The critical supersaturation at which this occurs is:

$$\Delta\mu_c \geq \sqrt{3}b^2\left[\sigma_L + \sigma_i - \sigma_s + \sigma_{Str}(d)\right] \tag{8.}$$

Therefore, the contribution of $\Delta\mu$ can be considered as follows:

$$\Delta\sigma = \sigma_{\mathrm{L}} + \sigma_{\mathrm{i}} - \sigma_{\mathrm{S}} + \sigma_{Str}(d) - \frac{\Delta\mu}{\sqrt{3}\mathrm{b}^2} \leq 0 \quad \leftarrow \text{2D - growth}$$

and

$$\boldsymbol{d \leq \frac{\sigma_{\mathrm{SiC}} + \frac{\Delta\mu}{\sqrt{3}b^2} - \sigma_i - \sigma_{\mathrm{Si}}}{M \cdot \varepsilon^2} \leftarrow \text{2D - growth}} \tag{9.}$$

That means $\Delta\mu$ could also contribute to the formation of the wetting layer. Since $\Delta\mu$ is mainly determined by the supersaturation, it can be adjusted by the deposition conditions (T and R).

In this study, the supersaturation is estimated using the data obtained for the critical flux R to stabilize the (3x3) surface structure without Si islanding on SiC(0001).[22] The estimated values for the excess wetting layer thickness (d_{crit} - d_{equ}) due to the supersaturation as function of T are included in Fig.4b together with the experimental data and data estimated using Arrhenius equation considering an activation energy of about 0.4 eV. Comparison with the experimental results indicates that supersaturation due to a change in temperature (equilibrium vapour pressure) cannot explain the delay in 2D-3D transition, in agreement to the earlier discussion [28]. Thereby, we have also to consider that the equilibrium vapour pressure of a laterally strained small crystal on a foreign substrate is higher than that of unstrained one due to its increased potential as a consequence of the elastic strain.[30] Therefore, the estimated value for d_{crit} represents only an upper limit. Furthermore, even an increase in Si deposition rate by a factor of 100 would also result only in a marginal increase in d ($\Delta d < 0.3$ ML).

2.3. Temperature and rate dependence of 2D-3D transition

At near equilibrium conditions ($T > 1000$ K, $R < 0.1$ ML/s) the 2D-3D transition takes place at Si coverage slightly above 1.4 ML coverage, where the (3x3) structure occurs. At lower T and higher R values the 2D-3D transition is kinetically delayed, which is already demonstrated in Fig.1 and 2. The increase in d_{crit} for the 2D-3D transition indicates that the wetting layer accumulates a lot of strain energy before strain relaxation via 3D islanding occurs.

In earlier investigation,[5,15] d_{crit} for the 2D-3D transition as function of the kinetic parameters R and T has been described using a power-law relation:

$$d \propto \left(\frac{\mathrm{R}}{\mathrm{D}}\right)^{\gamma} \tag{10,}$$

where $D = D_0\exp(-E_D/kT)$ is the surface diffusion constant and E_D is the energy barrier for surface diffusion. Values of γ between 0.12 and 0.5 are reported [5,15]. These values were derived with pure kinetic consideration. However, this is incorrect with respect to the growth of Si on SiC, where the wetting layer is still present after the island formation. Therefore, we analyzed the data for d_{crit} obtained in earlier investigations [16] with respect to the accumulated strain energy as function of the kinetic parameters T and R.

Fig. 3 shows the results for $(d_{crit} - d_{equ})$ as a function of R at 875 K. The investigation shows that at low rates the 2D-3D is nearly independent on R, whereas at higher R $(d_{crit} - d_{equ})$ increases linear with R. Such linear Rate-dependence was only reported so far in the context of RHEED investigations for the 2D-3D growth transition of InAs on GaAs [31].

Dubrovskii suggested a value of $(d_{crit} - d_{equ})$ nearly independent on R [32], what is found for low R in this study. They used the theory of first-order phase transition, where d_{crit} is a kinetic quantity controlled by the thermodynamics of the system and depends only slightly (logarithmically) on the kinetic parameter, such as deposition rate. Furthermore, the T-dependence is this case should be characterized by the relationship: $(d_{crit} – d_{equ}) \propto const/\sqrt{T}$ [32]. In Fig. 4a is plotted $(d_{crit} - d_{equ})$ as function of $1/\sqrt{T}$. At T > 850K the slope can be well fitted by a straight line, in agreement to the model by Dubrovskii et al. [32].

A deviation is observed for lower T. For comparison $(d_{crit} - d_{equ})$ as function of T is also shown in an Arrhenius plot in Fig. 4b. For T < 1000 K, the T-dependence (blue full line) is well described by the relationship: $(d – d_{equ}) \infty \exp(E/kT)$, where k is the Boltzmann constant and E is the energy for 2d-3D transition. The fitted line in Fig.4b corresponds to small activation energy of about 0.4 ± 0.05 eV, which compares to the energy barrier 0.4 - 0.5 eV calculated for the Si diffusion on the Si-rich SiC(0001) surface.[33]

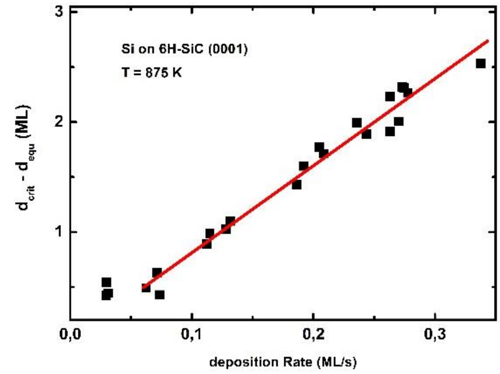


Fig.3. Excess wetting layer thickness for the 2D-3D-transition during growth of Si on SiC(0001) as function of deposition rate at T = 875 K.

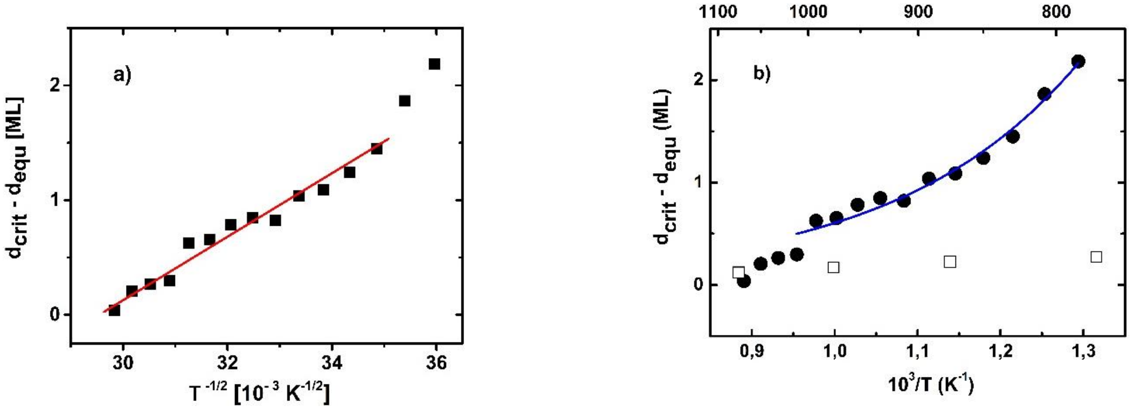


Fig. 4. Change in $(d_{crit} - d_{equ})$ for 2D- 3D transition for the growth of Si on SiC(0001) as function of T (R = 0.04 ML/s). a) using thermodynamic approach used by Dubrovskii [32]; b) using pure kinetic approach (Arrhenius plot), where the blue (full) line corresponds to an activation energy of about 0.4 ± 0.05 eV. The open symbols are the data of $(d_{crit} - d_{equ})$ calculated using equation (9) and an adsorption energy of E_a = 1.5 eV [22].

We see that depending on the used model deviations occur at low rates and high and low temperatures, respectively. It can be therefore suggested, that a change of the deposition condition results in a change from a thermodynamically (high T, low R) to kinetically (low T, high R) controlled regime of 2D-3D transition. The present investigations show that in case of the kinetically controlled regime $(d_{crit} - d_{equ})$ can be expressed a function of the kinetic parameters R and T by a linear relation: $(d_{crit} - d_{equ}) \propto (R/D)$.

Such linear dependence $(d_{crit} - d_{equ}) \propto (R/D)$ was not reported so far. However, it is interesting to note that for several hetero-systems, such as for the growth of InP on GaAs [5], InP on GaInP [34] and InAs on GaAs[35], the 3D-island density follows the same proportionality. This proportionality between island density and R/D is in agreement with the rate equation formalism for 3D island formation in SK growth process [Therefore, the linear dependence detected by RHEED could be related to the nucleation process. Here, we have to consider that the 2D-3D transition becomes visible in RHEED only if there is a significant number of 3D-islands nucleated. That happens at the moment when the wetting layer thickness has reached the critical value $(d_{crit} - d_{equ})$, where the 3D-island nucleation rate has an extremely sharp maximum [36]. The rate of island formation drops then rapidly during further deposition, what should result in nearly constant island density short after the 2D-3D transition. In the moment of the rapid 3D island formation, we see a strong increase in RHEED bulk spot intensity, which is usually interpreted as the 2D-3D transition.

At lower T or higher R the critical thickness could be larger. That could also result in a higher density of initially formed 3D islands. For example, such an increase in 3D island density by an increase in R and reduction in T was reported for the growth of InP on GaAs [37]. Therefore, it can be suggested that there is a direct correlation between $(d_{crit} - d_{equ})$ and the island number density during the 2D-3D transition. This could indicate that initially the growth contribution of wetting layer is small [5] during continuously supply and 2D-3D transition is due mainly due to agglomeration of adatoms from the incoming flux into 3D islands. This strongly supports kinetic models in which the first step, the nucleation, is dictated by the actual supersaturation at the surface (which is directly proportional to R). In other words, the initiation mechanism of the 2D-3D transition is nucleation, whereas the driving force for this transition is the strain in the 2D-layer. Therefore, transition towards 3D island formation detected by RHEED as function of R and T can be described by a classical nucleation process as suggested in earlier investigations [5]. In this process the maximum (saturation) 3D cluster density ρ as function of the deposition parameters for complete condensation can be described in the framework of standard rate equations theory by [38]:

$$\rho \propto \mathrm{R}^{\left(\frac{i}{i+2.5}\right)} \exp\left(\frac{E_i + iE_D}{(i+2.5)kT}\right) \qquad (11),$$

where i is the number of atoms forming the critical nucleus and E_i the energy barrier to form a critical nucleus. As seen from equation 11, a linear R-dependence of ρ provides that the number of atoms necessary to form a stable island is large ($i >> 1$). That agrees to earlier suggestions with respect to the Stranski-Krastanov growth [17,39,40] and corresponds to experimental results. For example, no islands smaller than 20 nm were observed by investigation of the growth of GaSb on GaAs even for lower T [41]. Moreover, as pointed out by Venables et al. [38] the large i value limit must eventually make a connection with the adsorbed state in which no cluster is really stable, so that E_i, the (free) energy difference between i atoms in the adsorbed state and in the islands, vanishes nearly in this limit [40]. A large critical nucleus can be expected based on a relatively small formation energy and, therefore, the island density should increase linearly with the deposition rate [40]. In this case the 3D island density only depends on the surface diffusion of adatoms. That is what we found for $(d_{crit} - d_{equ})$ in this study too. This indicates that the 2D-3D transition, which is driven by the stress in the extra wetting layer, is controlled by nucleation of 3D islands, where the activation energy to form a critical nucleus is mainly determined by the transport of adatoms (activation barrier for surface diffusion) to reach the necessary critical adatom density.

2.4. Kinetics of growth and post-growth evolution of islands

In further experiments, island growth during deposition was investigated and the post-growth island evolution after the deposition was stopped. The experiments were performed by depositing about 1 ML Si on the SiC(0001) surface covered by the (3x3) surface superstructure corresponding to a total Si coverage of about 2.4 ML. Subsequently, the deposition was halted at constant T. The RHEED intensity was measured for both the specular beam resulting from the two-dimensional diffraction of the SiC(0001) surface and the (222)-Si spot from the transmission diffraction through 3D Si islands. Fig. 5 shows the evolution of the (222) bulk spot intensity as a function of time for T in the range 525 °C – 775 °C during deposition and after the deposition was stopped. The increase in bulk spot intensity can be interpreted as the increase in Si amount bound in larger 3D islands and the change in intensity as function of time reflects the dynamics of 3D island formation and growth. Such interpretation is supported by earlier studies of the growth of InAs on GaAs, where a direct correlation of RHEED intensity change and the change of 3D island number density was found.[46]

At T > 600 °C, 3D-Si islands are rapidly formed already during the deposition, which is visible by the sudden increase in bulk spot intensity. Thereby, changes of the surface towards equilibrium much faster occur with an increase in T, indicating that kinetic processes are responsible for the island formation and growth. This indicates that beyond d_{crit} for islanding a fraction of the incident flux is immediately incorporated into islands, and this fraction increases with T. The critical thickness at which the 2D-3D transition occurred ranges from a total coverage of 1.65 ML for 775 °C to 2.1 ML for 600 °C, what corresponds to the data presented in Fig.3.

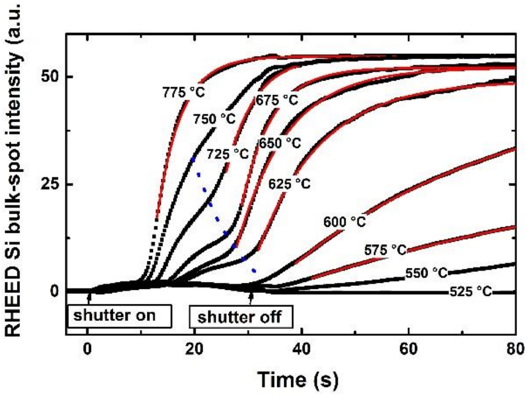


Fig. 5. RHEED data of the Si(222) bulk spot intensity due to transmission diffraction through 3D Si islands obtained during and after the deposition of ca 1 ML Si on (3x3) SiC(0001) at T between 525 °C and 775 °C (R = 0.04 ML/s). The solid (red) are the fitted results according to Equ. 12. The dotted (blue) line indicates a change in the intensity slope.

At lower T, Si bulk spots are visible only after a period where the deposition was already stopped. This indicates an increasing surface roughness due to subsequent development of 3D Si islands in a post-deposition evolution process. This is similar to observations made for 2D-3D growth mode transition of InAs on GaAs(001) [42] and Ge on Si(001) [43]. The island formation and growth is a relatively long lasting process at low T as seen from the slow increase in Si(222) bulk spot intensity.

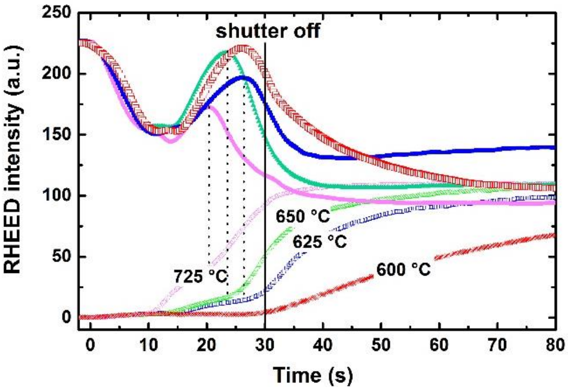


Fig. 6. RHEED data of the (00) specular and the (222) bulk spot intensity during and after deposition of ca 1 ML Si on (3x3) reconstructed SiC(0001) at 725 °C, 650 °C and 625 °C.

Furthermore, a change in the slope of RHEED bulk spot intensity is also seen towards a steep increase of intensity during the deposition at 750 °C ≤ T ≤ 625 °C, which is indicated by the dotted line in Fig.5. The steep increase is shifted towards lower coverage with an increase in T, accompanied by a faster and stronger increase in intensity. That means that the increase in RHEED bulk spot intensity results from different processes, an initially slower and a subsequent fast one.

When comparing with the RHEED specular reflected beam intensity, we see that the steep increase in 3D diffraction intensity happened just after the appearance of an intensity maximum in the RHEED specular beam intensity, as shown in Fig. 6. At T < 625 °C, the intensity maximum was always observed at a total coverage of ≈ 2 ML. This indicates an initially 2D layer-by-layer growth due to 2D island formation, where one period of oscillation corresponds to one monolayer. Therefore, the appearance of the intensity maximum indicates the formation of large flat areas on the surface. But, the intensity maximum is not associated with the completion of the second Si layer at higher T.

The change in the RHEED intensity slope indicates a change in surface features during the 2D-3D morphology transition due to a change in the growth kinetics. Investigations of 2D-3D transition of InAs on GaAs [44] and Ge on Si(001) [43] showed that during deposition first large 2D islands are formed, which are still part of the wetting layer. The same can be suggested for the growth of Si on SiC(0001) at least at medium T, where the formation of large 2D-islands is likely associated with the initial increase in RHEED specular beam intensity. However, an increase in Si bulk spot intensity is also simultaneously visible in our study. This can be explained as followed: The average surface roughness for kinetically-stabilized 2D layer growth is larger than for strain-free layer-by-layer growth. The explanation for this lies in the local interaction between 2D-islands. The strain field between the islands retards their merging thereby effectively increasing the nucleation rate for new islands on the surface. This leads to a steady-state evolution, which is in some measure layer-by-layer but with a rougher surface than found for comparable strain-free growth [45]. Furthermore, investigation of InAs on GaAs also showed that with coverage near to the critical wetting layer thickness the height of 2D islands increases local to form quasi 3D (Q3D) clusters, what is just after reaching a maximum in number density of 2D islands.[46]

The development of RHEED bulk intensity curves for the initial period in case of 3D layer formation during deposition and by post-deposition evolution looks similar. This suggests that the underlying mechanisms for the initial Q3D island formation and the subsequent increase in the volume of the 3D islands during post-deposition annealing are similar. That corresponds to investigations for the growth of InAs on GaAs.[42] There it was found , that the atoms for the Q3D clusters will come directly from the wetting layer. During further deposition, Q3D islands act then as precursors for the subsequent 3D island formation and growth. Similar, formation of metastable islands in the initial stage of 2D-3D transition was also reported for Ge on Si(001) [47] and CdTe on ZnTe.[48] Therefore, the results of the

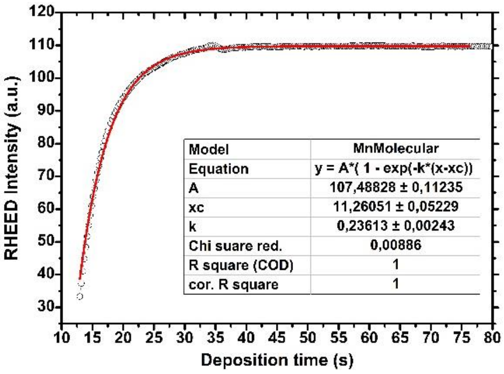
RHEED investigations for the growth of Si on SiC(0001) at T between 750 °C and 625 °C can be interpreted in a similar way.

To obtain insight into rate-controlling mechanisms of 3D island formation and growth, we have analysed quantitatively the dynamic processes leading to island growth from the data presented in Fig.5. For this analysis, we considered that the integral RHEED intensity *I* due to the diffraction from 3D-Si islands (bulk spot intensity) is directly proportional to the Si amount within the 3D-islands. This, for example, is supported by investigations of InAs island formation on GaAs, where RHEED intensity changes closely follow the island number density changes [44]. The evolution of RHEED bulk spot intensity during deposition and post-growth annealing, respectively, can be expressed by:

$$I = I_0\left(1 - \exp\left(-\frac{(t - t_c)}{\tau}\right)\right) \tag{12}$$,

where $\boldsymbol{I_0}$ corresponds to the saturation value of the RHEED bulk spot intensity, t_c is the critical time for the onset of 3D-islanding and τ is the time constant of the consumption of atoms from the wetting layer and the incoming flux during the dot formation and growth process.

In Fig. 7 is shown a typical fit of the time-dependent evolution of the (222) bulk spot intensity, which is where the data of time constant (τ) were extracted from. In the T-range between 750 °C and 625 °C, the time constant was obtained for the period after the change in the slope. The fitted results are shown by solid red lines in Fig. 5. The change of the intensity slopes with T indicates a strong T-dependence of the time constant, as shown in Fig. 8. In the range T > 625 °, the activation energy for the time constant is estimated to be about 0.55 eV. At T < 625 °C, the activation energy is much larger with about 2.5 eV, which indicates a change in the rate-controlling mechanisms during post-growth evolution.



Model	MnMolecular
Equation	y = A*(1 - exp(-k*(x-xc))
A	107,48828 ± 0,11235
xc	11,26051 ± 0,05229
k	0,23613 ± 0,00243
Chi suare red.	0,00886
R square (COD)	1
cor. R square	1

Fig. 7. Evolution RHEED of the RHEED (222) Si bulk spot intensity as function of time during Si deposition on SiC(0001) at 775 °C. Open scattered symbols are experimental data. The red solid line is the fitted line according to Equ. 12. The inset shows the data for the fitted line.

For discussion and interpretation of the obtained activation energies, we have to consider what is the main contribution to the growth of 3D islands during the deposition and after growth interruption. The 3D islands grow by the consumption of adatoms. During the deposition the main contribution comes from the impinging flux, whereas after growth interruption metastable islands and the wetting layer only act as sources for adatoms. Westwood et al. [49] et al. suggested from growth experiments of InAs on GaAs(001) that the largest fraction of the incident flux is immediately transferred into already formed islands. This fraction increases with T but is largely insensitive on the growth rate. They suggested that this is perhaps related to a requirement for a minimum cluster size for nucleation of the 3D-islands.

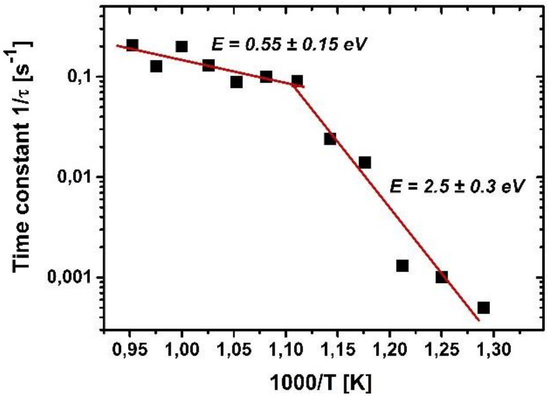


Fig. 8. Arrhenius plot of the reciprocal time constant obtained from the data shown in Fig.5. The slope of the line between 1050 K and 900 K corresponds to the activation energy of 0.55 ± 0.15 eV, whereas the slope at T < 900 K corresponds to the energy of 2.5 ± 0.3 eV.

On the other hand, it has been found that already during the deposition a significant mass transfer from metastable islands towards the 3D islands occurs.[44] Therefore, it can be suggested, that mainly two T-dependent mechanisms contribute to the activation energy for cluster growth: the surface diffusion with the activation energy for diffusion E_D and the adatom concentration with the energy for formation of free adatom E_a, i.e. the energy to remove an atom from already formed cluster or wetting layer onto the free surface. Therefore, the T-dependence of the time constant can be expressed by:

$$\frac{1}{\tau} \propto \exp\left(-\frac{E_D + E_a}{kT}\right) \qquad (13),$$

with E_a the adatom creation energy. The reciprocal time constant should be proportional to surface mass transport constant D·n, where n represents the adatom density on the surface.

The low activation energy at high T indicates that surface diffusion is mainly rate-controlling in the 3D island growth at higher T. However, mass transfer of adatoms released by decomposition of smaller metastable islands formed in the initial stage of 2D-3D transition could also contribute to the island growth. This contribution can be extracted by comparison of the obtained activation energy for the T-dependence of the 2D-3D transition (Fig.4b) with those obtained for the time constant. These activation energies differ only by 0.15 eV, what likely corresponds to the adatom formation energy and indicates only small activation energy of adatom formation. Indeed, the adatom formation energy can be as small as 0.35 eV [50]. A value of 0.16 eV was estimated for Ge on Si(001) to remove an atom e.g. from a king of a strained Ge island edge to the free surface.[51]

At lower T the obtained activation energy is quite large in comparison. Here, it can be suggested that the adatoms mainly stem from decomposition of the highly stained wetting layer. Since the initially wetting layer thickness (≈ 2.4 ML) is almost constant, the strain has always the same influence and the rate only depends on T. The excess strain energy in the wetting layer decreases as atoms detach from the wetting layer and the driving force for further release of atoms vanishes when the excess strain energy approaches zero. That was observed experimentally, after island formation the equilibrium wetting layer of 1.4 ML within an ordered (3x3) surface structure remained between the islands. It was proposed that the rate for wetting layer decomposition is proportional to the accumulated strain energy density [5]:

$$\frac{d\left(\mathrm{d} - \mathrm{d}_{\mathrm{equ}}\right)}{\mathrm{dt}} \propto (\mathrm{d} - \mathrm{d}_{\mathrm{equ}})\mathrm{M} \cdot \varepsilon^2 \cdot \exp\left(-\frac{\mathrm{E_b}}{\mathrm{kT}}\right) \qquad (14),$$

where E_b is closely related to the bond strength between the atoms of the wetting layer. The activation energy to transfer individual atoms out of the two-dimensional layer towards the free surface can be expected to be in the range of the Si-Si bond energy. From the activation energy of 2.5 eV and the energy barrier of surface diffusion (0.4 eV) we get bond strength of about 2.1 eV, which corresponds to the Si-Si bond energy (2.3 eV) considering some weakening of the bonds by strain.

3. SUMMARY AND CONCLUSION

In summary, the thermodynamics and kinetics of Stranski-Krastanov growth of Si on SiC(0001) is investigated in detail by RHEED measurements ranging from wetting behaviour to the 3D island formation and growth.

Investigations clearly demonstrate that Si effectively wets SiC(0001) up to an equilibrium layer thickness d_{equ}, where island formation can still be prevented. This layer thickness can be estimated using thermodynamic approach considering the Si/SiC surface and interface properties as well as strain. Increasing the layer thickness beyond d_{equ} results in the acculation of excess strain in the wetting layer up to an critical value. In this situation flat 2D-islands are not lorger stable and 3D-islands nucleated. The excess strain is the driving force for 2D-3D transition, which is initiated by 3D island nucleation. The nucleation can be kinetically delayed. The strength of this effect appears to depend on the kinetic parameters. At lower T and higher R, the critical excess wetting layer thickness for the 2D-3D transition as function of the kinetic parameters is found to be proportional to $(d_{crit} - d_{equ}) \propto R \cdot \exp(E/kT)$, where E corresponds to the energy barrier of surface diffusion. At higher T and low R, $(d_{crit} - d_{equ})$ deviates from this relationship, indicating a change in the wetting layer contribution to the 2D-3D transition from a kinetically to thermodynamically controlled regime by a change in the deposition condition. The critical excess thickness decrease with increasing T according to $(d_{crit} - d_{equ}) \propto \text{const}/\sqrt{T}$ and seems to be independent on R.

Furthermore, RHEED intensity measurements indicate that at higher temperature initially the 2D-3D transition starts with the formation larger 2D islands and the simultaneous formation of smaller 3D islands. The maximum in 2D island density shifts to higher coverage with a decrease in temperature. Just after the highest intensity for the 2D islands was obtained there is a sudden increase in diffraction from 3D islands. This can be interpreted as a sudden nucleation of a large number of 3D islands. The further increase in 3D diffraction intensity (corresponding to the island growth rate) during further deposition shows a strong dependence from the temperature. The activation energy of the 3D island growth rate indicates that the surface mass transport of adatoms towards the formed larger 3D is rate-controlling, where the adatoms mainly stem from the incoming flux and from decomposition of islands (smaller 3D and strained 2D islands).

Post-deposition annealing of highly stressed wetting layer ($d > d_{equ}$) also results in 2D-3D island transition, what could be a long lasting process at lower T. From the obtained higher activation energy it can be suggested, that mass transport of adatoms towards the formed larger 3D is also rate-controlling. However, since there is no flux, the adatoms are delivered from the decomposition of the wetting layer and of strained 2D islands.

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