Finite element analysis of the strain induced vertical ordering of islands and determination of compositional modifications in LPCVD-grown Ge$_x$Si$_{1-x}$–Si bilayers on Si(001)

K. TILLMANN*, W. JÄGER
Centrum für Materialanalytik, Universität Kiel, Kaiserstraße 2, D-24143 Kiel, Germany

B. RAHMATI†, H. TRINKAUS‡, L. VESCAN† and K. URBAN†
†Institut für Festkörperforschung and
‡Institut für Schicht- und Ionentechnik, Forschungszentrum Jülich GMBH, D-52425 Jülich, Germany

[Received 3 December 1998 and revised version accepted 12 April 1999]

ABSTRACT
The vertical ordering of Ge$_x$Si$_{1-x}$ islands along the growth direction during low pressure chemical vapour deposition of Ge$_x$Si$_{1-x}$–Si bilayers on Si[001] is studied by transmission electron microscopy and analysed quantitatively using finite element method simulations. The calculation of local strain fields and the determination of an island correlation function allows one to describe the observed ordering behaviour in terms of a strain induced preferential nucleus formation above existing islands. In addition the compositions of the Ge$_x$Si$_{1-x}$ layers are measured by a lattice fringe analysis of high-resolution transmission electron micrographs giving clear evidence for a substantial silicon germanium intermixing. The implications of these compositional modifications for the nucleation and ordering of islands are discussed indicating that islands are subjected to geometrical and compositional modifications during the growth process.

§1. INTRODUCTION
Over the past decade highly lattice parameter mismatched Ge$_x$Si$_{1-x}$–Si semiconductor heterostructures have become an object of intense research. As typical for semiconductor heteroepitaxy, the layers are characterized by a fairly low interface energy but a substantial lattice mismatch may be imposed by choosing a sufficiently large germanium content, which results in the Stranski–Krastanow (1939) growth mode. Under such conditions epitaxial growth starts with two-dimensional layer-by-layer growth which is followed by the formation of three-dimensional islands. The morphological transition is determined by the competition between an increased surface energy contribution for islands compared with a strictly
two-dimensional growth regime and an elastic relaxation achieved by the lateral bending of the islands’ free edges (Eaglesham and Cerullo 1990). Moreover, regularly ordered arrays of Ge$_x$Si$_{1-x}$ islands have been observed in periodic Ge$_x$Si$_{1-x}$-Si multilayers (Vescan et al. 1992, Carlino et al. 1996, Mateeva et al. 1997, Kienzle and Ernst 1998) and the lateral ordering of islands inside the growth plane has been attributed to anisotropic elastic material properties (Jäger 1994, Schukin et al. 1995) while the vertical ordering along the growth direction has been explained by a locally varying contribution of the elastic energy to the chemical potential at the growth surface (Christiansen et al. 1996, Rahmati et al. 1996). These stacked arrays of self-assembled epitaxial islands are regarded as a promising possibility for the formation of ensembles of quantum dots, whose electronic properties are determined by the island geometry, the layer composition and the mismatch induced internal elastic strains (Schittenhelm et al. 1995, Van De Walle 1995, Hartmann 1997).

In the present study we focus on highly lattice parameter mismatched Ge$_x$Si$_{1-x}$-Si bilayer structures with a nominal germanium content $x=1$ grown in the Stranski–Krastanow mode by low pressure chemical vapour deposition (LPCVD). By varying the thickness of a silicon interlayer separating two Ge$_x$Si$_{1-x}$ layers we examine the influence of buried Ge$_x$Si$_{1-x}$ islands on the spatial nucleation behaviour of subsequently grown Ge$_x$Si$_{1-x}$ islands on top of the interlayer. The general features of epitaxial growth concerned with the topographical arrangement of the islands are analysed by transmission electron microscopy (TEM) observations, while the composition of the Ge$_x$Si$_{1-x}$ layers will be assessed by a lattice parameter analysis on digitized micrographs obtained by high-resolution transmission electron microscopy (HRTEM) under experimentally well-defined conditions. While the experimentally observed vertical ordering of Ge$_x$Si$_{1-x}$ islands along the growth direction has been treated in terms of an analytical approximation in a previous investigation (Rahmati et al. 1996), in the present study finite element method (FEM) simulations, taking fully into account the layer morphology and the anisotropic elastic material properties of silicon and germanium, will be used to calculate the strain fields inside the heterostructures. These numerical data will be used to determine the spatially varying nucleation behaviour of islands by definition of an island correlation function dependent on the silicon interlayer thickness and to gain information on nucleus formation in the earliest stages of growth.

Section 2 gives a brief overview of the experimental techniques applied in this study. The results, as far as they are concerned with the layer morphology, are presented in § 3 and are discussed in the framework of an island nucleation model in § 4. Following a quantitative analysis of the layer composition by high-resolution electron microscopy and the discussion of the origins of compositional modifications in § 5, the most important results are summarized in § 6.

§ 2. Experimental setup

The heteroepitaxial growth of the Ge$_x$Si$_{1-x}$-Si bilayers was performed by LPCVD on Si(001) substrates at a constant deposition rate of about 0.3 nm s$^{-1}$ and a growth temperature of $T_S = 700^\circ$C. The epitaxy was carried out at 15 Pa total pressure in a load locked system. SiCl$_2$H$_2$ and GeH$_4$ were chosen as source gases while H$_2$ was used as the carrier gas. The structures consist of two Ge$_x$Si$_{1-x}$ layers separated by a silicon interlayer whose thickness $d_{Si}$ was varied between 40 nm and 300 nm. The Ge$_x$Si$_{1-x}$ layers were deposited under pure germanium conditions.
(x = 1) with a nominal thickness of $d_{Ge,nom} = 0.8$ nm each. Each heterostructure has 10 nm of silicon grown as a buffer layer and a silicon capping layer of 50 nm in thickness.

The morphology and the spatial arrangement of the Ge$_x$Si$_{1-x}$ islands inside the bilayers were analysed by conventional TEM of plan-view and cross-sectional samples using a JEOL 2000EX electron microscope. Specimens were prepared by mechanical polishing followed by Ar$^+$ ion milling at 4 keV with liquid nitrogen cooling until perforation and, subsequently, at 2 keV to minimize the thickness of an amorphous surface layer induced by the ion milling process. For the determination of the composition of the Ge$_x$Si$_{1-x}$ layers high-resolution micrographs were taken on negative plates with a JEOL 4000EX microscope operating at an acceleration voltage of 400 kV and characterized by a Scherzer resolution of 0.17 nm. The recorded images were digitized by a charge-coupled device camera system at a resolution of 1024 × 1024 picture elements. Subsequently, the images were analysed by an image processing routine (Tillmann et al. 1996) to extract local lattice parameter values from the high-resolution micrographs and to get an estimate of the real layer composition.

§3. Experimental Observations

The Ge$_x$Si$_{1-x}$–Si bilayers have been characterized by conventional transmission electron microscopy to gain information on the layer morphology and defect structure. Figure 1 gives a summary of representative bright-field TEM images of the bilayers for silicon interlayer thicknesses amounting to $d_{Si} = 40$ nm, 100 nm, 200 nm and 300 nm. Micrographs have been taken along the [110] projection of cross-sectional specimens. The images demonstrate that inside the Ge$_x$Si$_{1-x}$ layers of the heterostructures epitaxial islands have been formed on a two-dimensional wetting layer. All bilayers are characterized by completely coherent interfaces, i.e. misfit dislocations have not been nucleated. This is proved by plan-view images of the heterostructures showing neither dislocation contrast nor moiré fringes as illustrated by figure 2 in the case of an interlayer thickness $d_{Si} = 40$ nm. In this micrograph islands, which are randomly arranged inside the growth plane, are easily identified by their characteristic bright-dark contrast lobes along the direction of the imaging vector which result from small variations of the deviation parameter due to local crystal misorientations.

Independent of the thickness of the silicon interlayer $d_{Si}$ all islands, both of the first and the second Ge$_x$Si$_{1-x}$ layer, are characterized by a regular geometry. Based upon the analysis of a sufficiently large number of cross-sectional images the mean values of the lateral island extension ⟨l⟩ along the [110] direction and the island height ⟨h⟩ are summarized in table 1. For bilayer systems with a complete vertical ordering (cf. later this section) the value for the mean mutual island distance ⟨λ⟩ inside the growth plane was determined by the measurement of the island density N from plan-view images as $\langle \lambda \rangle = [1/(N)]^{1/2}$.

Moreover, figure 1 demonstrates that the variation of $d_{Si}$ in the range between 40 nm and 300 nm significantly influences the topographical arrangement of the Ge$_x$Si$_{1-x}$ islands. Below $d_{Si} = 100$ nm islands of the second Ge$_x$Si$_{1-x}$ layer have been formed above Ge$_x$Si$_{1-x}$ islands of the first layer. This vertical ordering decreases with increasing $d_{Si}$ values and results in an almost random arrangement at $d_{Si} = 300$ nm.
Figure 1. Vertical ordering behaviour of islands in the Ge$_x$Si$_{1-x}$–Si bilayers on Si(001) for silicon interlayer thicknesses (a) $d_{\text{Si}} = 40$ nm, (b) $d_{\text{Si}} = 100$ nm, (c) $d_{\text{Si}} = 200$ nm and (d) $d_{\text{Si}} = 300$ nm. The series of representative cross-sectional micrographs was taken along the [110] direction under bright-field conditions with an imaging vector $\mathbf{g} = (004)$ under which germanium-rich layers appear dark. An increased vertical ordering of the Ge$_x$Si$_{1-x}$ islands is observed with decreasing $d_{\text{Si}}$ values.
In order to characterize the observed ordering phenomenon in a more quantitative manner, we describe the vertical ordering of islands in terms of an island correlation function depending on the silicon interlayer thickness. To adapt the evaluation to our finite element simulations (later) we use a definition for the correlation function somewhat different from an earlier analytical approach (Rahmati et al. 1996).

We consider imaginary square columns above all buried islands of the first Ge$_x$Si$_{1-x}$ layer which define an array of squares in the [001] projection with an areal fraction amounting to $\lambda = \langle \lambda \rangle = (1/N)^{1/2}$, as obtained by the analysis of bright-field TEM micrographs.

By analysing cross-sectional micrographs we determine the fraction $P_{d Si}$ of all islands of the second Ge$_x$Si$_{1-x}$ layer whose centre of mass lies within this imaginary array of squares. Afterwards the data on $P_{d Si}$ are used to calculate a correlation function $C_{d Si}$ according to

$$C_{d Si} = P_{d Si} - \delta^2.$$  

(1)

This definition of a correlation function fulfills two reasonable boundary conditions, namely $C = 1$ in the case of complete vertical ordering ($P_{d Si} = 1$) and $C = 0$ when no ordering is found ($P_{d Si} = \delta^2$). The values of $C_{d Si}$ gained by analysing a large number of cross-sectional TEM images of the Ge$_x$Si$_{1-x}$–Si bilayers are summarized in Table 2. Strictly speaking, we would have to consider a potential source of error by applying this procedure since TEM is a two-dimensional projection technique that
may lead one to believe that uncorrelated islands being shifted along the direction of the electron beam appear vertically ordered. In the case of the investigated bilayer systems this source of error may be ruled out because the mean mutual island distance $h \approx 63 \text{ nm}$ distinctly exceeds typical TEM sample thickness values with an average value in the order of 100 nm. Additionally, the determined correlation values are in fair agreement with those values gained from tilt-experiments of plan-view samples (Rahmati 1996).

### § 4. VERTICAL SELF-ORGANIZATION OF ISLANDS

Under the chosen growth conditions the vertical ordering of islands inside the Ge$_x$Si$_{1-x}$-Si bilayers is decisively influenced by the thickness $d_{Si}$ of the silicon interlayer. In order to get a more quantitative understanding of the preferential vertical island ordering we discuss the experimental results in the framework of a nucleation model for islands of the second Ge$_x$Si$_{1-x}$ layer, which results in the determination of a physically motivated expression for the correlation function. To simplify matters we assume first of all pure germanium layers ($x = 1$) corresponding to the aspired stoichiometry. The implications of potential compositional modifications are considered in § 5 later.

We attribute the vertical island ordering to the elastic interaction between a newly formed island nucleus in the growing second germanium layer with adjacent islands in the buried germanium layer. Conspicuously, the mismatch induced strain fields of buried islands cause an inhomogeneously distributed effective lattice parameter misfit of the second germanium layer with respect to the silicon interlayer. The corresponding lowering of the elastic strain energy of an island nucleus moderates the activation barrier for the island formation itself. Accordingly, an enhanced nucleation probability for islands of the second layer is expected at positions of the maximum effective mismatch reduction. In the following, the quantification of this effect will be used to calculate the correlation function $C(d_{Si})$, which subsequently will be fitted to the experimental data obtained by transmission electron microscopy. This is performed by a three step procedure, starting with the determination of the strain distribution at the growth surface by applying finite element method simulations (§ 4.1), which is followed by the calculation of the correlation function $C(d_{Si})$ according to the following equation:

$$C(d_{Si}) = \frac{1}{2} \left( 1 + \frac{1}{1 + e^{-2d_{Si}/\lambda}} \right)$$

Table 2. Experimental values of the correlation function $C(d_{Si})$ according to equation (1) depending on the silicon interlayer thickness $d_{Si}$ as gained by the analysis of cross-sectional micrographs (see also figure 1). The accuracy of $C(d_{Si})$ is limited by the margins of error in the ($\ell$) and ($\lambda$) values according to table 1.

<table>
<thead>
<tr>
<th>$d_{Si}$ (nm)</th>
<th>$C(d_{Si})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>40</td>
<td>1.000 ± 0.000</td>
</tr>
<tr>
<td>100</td>
<td>0.980 ± 0.020</td>
</tr>
<tr>
<td>200</td>
<td>0.437 ± 0.112</td>
</tr>
<tr>
<td>300</td>
<td>0.131 ± 0.024</td>
</tr>
</tbody>
</table>

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function (§4.2) and the judgment of the reasonableness of determined fitting parameters (§4.3).

4.1. Strain distribution at the growth surface

To calculate the strain distributions near the growth surface directly after deposition of the silicon interlayer, three-dimensional FEM simulations taking fully into account the anisotropic elastic material properties, as well as the measured island shapes and sizes and the topographical arrangement according to table 1, were carried out by application of the ABAQUS finite element code (Hibbit, Karlsson & Sorensen Inc. 1997). Although the observed islands are of nanometre size we may apply these kinds of numerical simulations, which are normally used when dimensioning macroscopic objects and are based on continuum theory of elasticity, since the interatomic potential of small clusters will follow Hooke’s (1679) law for sufficiently small strain values. The geometry is meshed with three-dimensional 20-node cuboid finite elements, cf. figure 3. A pyramidal shape with quadratic basement is assumed for the germanium islands, whose base directions are supposed to be elongated parallel to the crystallographic \( h_{110} \) direction. Since we are interested in strain inhomogeneities due to buried islands and since for relevant silicon interlayer thicknesses the completely strained two-dimensional wetting layer will have only a negligible influence on the strain distribution of the surrounding silicon the absolute thickness of this layer is of minor importance and we choose a starting value of four monolayers for modelling purposes. Although the bilayer structures do not show a lateral ordering inside the \([001]\) growth plane we treat for simplification the in-plane island arrangement as periodical with a period length identical to the average mutual island distance \( \bar{\Delta i} \). Then, by reason of symmetry, only square column parts of the bilayer structures with a base area of \( \bar{\Delta i}^2 \) have to be modelled and appropriate boundary conditions can be applied to the FEM model by fixing all nodes on the free side faces of the model against displacements in the respective direction parallel to the surface normal. Additionally, all nodes at the bottom of the model are fixed against displacements along the \( z \) direction to avoid rigid body motion during processing.

For the numerical treatment we assume a nominal lattice mismatch of \( f = (a_{Ge} - d_{Si})/a_{Ge} = 0.04016 \), where \( a_{Si} = 0.543 \) nm and \( a_{Ge} = 0.566 \) nm denote the bulk lattice parameters of silicon and germanium. For the simulations second order elastic constants \( c_{11} \) have been chosen as \( c_{11, Si} = 165.8 \) GPa, \( c_{12, Si} = 63.9 \) GPa and \( c_{44, Si} = 79.6 \) GPa for silicon as well as \( c_{11, Ge} = 128.5 \) GPa, \( c_{12, Ge} = 48.3 \) GPa and \( c_{44, Ge} = 66.8 \) GPa for germanium according to Schulz and Blachnik (1982).

The contour plots in figure 3 show the in-plane strain component \( \varepsilon_{xx} \) along the \([110]\) direction as obtained from the simulations for two different interlayer thicknesses of \( d_{Si} = 50 \) nm and \( d_{Si} = 150 \) nm in the form of a cross-sectional (left) and a perspective (right) representation. The germanium island is assumed to be buried in the middle of the cube at the height of the two-dimensional germanium wetting layer which is represented by the blue contours. In the cross-sectional representations the darker red club-like contours around the germanium islands indicate that the surrounding silicon substrates and interlayers are strained in dilatation with a location dependent range of values between \( \varepsilon_{xx} = 0.000245 \approx f/200 \) and \( \varepsilon_{xx} = 0.0045 \approx f/10 \), while the blue contours indicate that the germanium islands and the two-dimensional layers are strained in compression in the order of the lattice parameter mismatch with \( \varepsilon_{xx} = 0.0337 \approx -0.8 f \). In contrast to \( d_{Si} = 50 \) nm, the
Figure 3. Contour plots of the in-plane strain distribution $\varepsilon_{xx}$ as gained by FEM simulations for an overgrown germanium layer assuming silicon interlayer thicknesses of $d_{Si} = 50\,\text{nm}$ and $d_{Si} = 150\,\text{nm}$. The numerical values of $\varepsilon_{xx}$ are depicted in the contour legend. The cross-sectional representations along the [110] projection (left in either case) illustrate that the mismatched buried germanium islands (triangular shape and blue contours) strain the surrounding silicon in dilatation (club-like darker-red contours) above and below the islands, while other parts of the silicon remain nearly unstrained (orange contours). The perspective representations (right in either case) reveal that buried islands cause significant displacements at the interlayer surface, which are magnified by a factor of 15 for visualization purposes, at $d_{Si} = 50\,\text{nm}$ but not at $d_{Si} = 150\,\text{nm}$.
darker red clubs representing a dilatation $\varepsilon_{xx} \geq f / 200$ do not reach the surface for $d_{Si} = 150$ nm, which results in a nearly homogeneous strain distribution at the growth surface for the subsequent deposition of the second germanium layer.

In order to calculate local nucleation probabilities for islands it is useful to consider the difference $\Delta \varepsilon$ of the mean strain values at the growth surface according to

$$\Delta \varepsilon(d_{Si}) = \langle \varepsilon_{Ib}(d_{Si}) \rangle - \langle \varepsilon_{Iay}(d_{Si}) \rangle,$$

where $\langle \varepsilon_{Ib} \rangle$ denotes the mean strain above a buried germanium island averaged over an area of $\langle l \rangle^2$, while $\langle \varepsilon_{Iay} \rangle$ represents the mean strain value at the growth surface above the two-dimensional wetting layer averaged over $\langle l \rangle^2$. The resulting behaviour of $\Delta \varepsilon$ is shown in figure 4 in dependence upon the thickness $d_{Si}$ of the silicon interlayer. The black dots represent $\Delta \varepsilon$ values as gained from the FEM simulations, while the solid curve results from a polynomial fit to these values. The difference of strains $\Delta \varepsilon$ decreases with increasing silicon interlayer thicknesses following a $d_{Si}^{-3}$ law in a first order approximation as expected from the analytical treatment of any long-range strain field governed by the elastic dipole contribution (Mura 1982). In the case of the lowest experimentally chosen silicon interlayer thickness amounting to $d_{Si} = 40$ nm the difference of the averaged strains is $\Delta \varepsilon(d_{Si} = 40$ nm $) = 0.0011 \approx f / 40$. Thus, the maximum fluctuation of the local strain at the growth surface is by more than one order of magnitude smaller than the nominal lattice parameter mismatch $f$.

4.2. Modelling of the correlation function

According to equation (1) the correlation function represents the normalized probability for island nucleation inside the second germanium layer above buried islands of the first germanium layer. From a thermodynamical point of view this nucleation probability is determined by the activation enthalpy of nucleation as well as the geometry of the heterostructures. Defining the geometrical parameter

$$\delta = \langle l \rangle / \langle \lambda \rangle$$

which represents the mean island extension-to-distance-ratio, the fraction of correlated islands $P(d_{Si})$ is given by the ratio of the nucleation probability above a buried island $P_{Ib}(d_{Si})$ and the probability for nucleation above the wetting layer $P_{Iay}(d_{Si})$:

$$P(d_{Si}) = \frac{P_{Ib}(d_{Si})}{P_{Iay}(d_{Si})}.$$
germanium island to the mean probability averaged over the total silicon interlayer surface, i.e.

$$P(d_{Si}) = \frac{\langle l \rangle^2 \langle J_{Is}(d_{Si}) \rangle}{\langle l \rangle^2 \langle J_{Is}(d_{Si}) \rangle + \langle (l)^2 - \langle l \rangle^2 \rangle \langle J_{Lay}(d_{Si}) \rangle} \left( 1 + \frac{1}{\langle (l)^2 - \langle l \rangle^2 \rangle \langle J_{Lay}(d_{Si}) \rangle / \langle J_{Is}(d_{Si}) \rangle} \right) \tag{4}$$

In this equation $\langle J_{Is}(d_{Si}) \rangle$ and $\langle J_{Lay}(d_{Si}) \rangle$ denote averaged island nucleation probability densities above the buried island and the two-dimensional wetting layer, which have to be weighted by the areal segments $\langle l \rangle^2$ and $\langle (l)^2 - \langle l \rangle^2 \rangle$ respectively to gain the fraction of correlated islands. Following Volmer and Weber (1926) we approximate $\langle J_{Is}(d_{Si}) \rangle$ and $\langle J_{Lay}(d_{Si}) \rangle$ by Boltzmann distributions according to

$$\langle J_{Is}(d_{Si}) \rangle \propto \exp\left[ -\langle A_{Is}(d_{Si}) \rangle / kT_S \right],$$
$$\langle J_{Lay}(d_{Si}) \rangle \propto \exp\left[ -\langle A_{Lay}(d_{Si}) \rangle / kT_S \right], \tag{5}$$

in which $\langle A_{Is}(d_{Si}) \rangle$ and $\langle A_{Lay}(d_{Si}) \rangle$ represent the maximum free enthalpy changes for island nucleus formation above buried islands and above the two-dimensional wetting layer, respectively.

Local changes in the free enthalpies for the formation of a completely strained planar island nucleus are averaged over the regions above the buried islands and the remaining regions, respectively. Assuming a two-dimensional, i.e. completely tetragonal distorted, island nucleus of a volume $V_{2D}$ the free enthalpy changes may be written as

$$\langle A_{Is}(d_{Si}) \rangle = A_0 + Y_{Ge} \langle f - \epsilon_{11}(d_{Si}) \rangle^2 V_{2D},$$
$$\langle A_{Lay}(d_{Si}) \rangle = A_0 + Y_{Ge} \langle f - \epsilon_{12}(d_{Si}) \rangle^2 V_{2D}, \tag{6}$$

where $Y_{Ge} = (c_{11,Ge} + 2c_{12,Ge})/c_{11,Ge}$ denotes the biaxial elastic modulus of the germanium epilayer. In these equations $A_0$ represents the change of the free enthalpy upon island formation during homoepitaxial growth whereas the second term takes the lattice mismatch and its change by a buried island of the first layer into account. According to equation (4) the fraction of correlated islands $P(d_{Si})$ is given by the ratio of the local island nucleation probabilities directly above the buried germanium islands and those of the remaining region. Therefore we do not have to reflect on the details of the nucleation process and the numerical value of $A_0$, but just have to consider the difference between $\langle A_{Is}(d_{Si}) \rangle$ and $\langle A_{Lay}(d_{Si}) \rangle$ according to equations (5) and (6).

By combining equations (1) up to (6) we obtain an expression for the correlation function $C(d_{Si})$, which depends only on material properties ($Y_{Ge}$, $f$), geometrical and growth parameters ($\delta$, $T_S$), the difference of strain values as obtained by FEM simulations ($\Delta \epsilon(d_{Si})$) and the volume of a newly formed island nucleus of the second germanium layer ($V_{2D}$) according to
Vertical ordering of islands in GeSi–Si bilayers on Si(001)

\[ C(d_{Si}) = \frac{\delta^2}{1-\delta^2} \left( \left\{ \delta^2 + (1-\delta^2) \exp \left[ -2 \frac{Y_{Ge} f V_{2D} \Delta_e(d_{Si})}{kT_S} \right] \right\} \right)^{-1} \]  

If the elastic interaction is sufficiently strong, equivalent to large \( \Delta_e(d_{Si}) \) values, then the numerator of the exponential function in equation (7) overweights the denominator and \( C = 1 \) is expected, i.e. the model predicts complete vertical ordering of the germanium islands. In the case of vanishing differences of the surface strains, equivalent to \( \Delta_e(d_{Si}) \to 0 \), the correlation function converges to \( C = 0 \), corresponding to a completely random island distribution. We emphasize that equation (7) may generally be applied to any lattice parameter mismatched bilayer system forming islands. When growth conditions and material properties are different from those chosen in this study geometrical parameters as well as the strain distribution in the heterostructures will however be affected and lead to different values of the correlation function.

As shown in § 4.1, \( \Delta_e(d_{Si}) \) may be obtained by FEM simulations. Hence, the nucleation volume may be used as a fitting parameter to adapt the correlation function to the experimental data, because \( V_{2D} \) is the only parameter which is not directly accessible by TEM investigations and FEM simulations. Based upon the numerical values of \( \Delta_e(d_{Si}) \) according to figure 4, the optimal adjustment of equation (7) to the experimental \( C(d_{Si}) \) values is achieved for \( V_{2D} = 7.0 \) nm\(^3\), equivalent to an island nucleus formed by approximately 320 germanium atoms, and represented by the solid black curve in figure 5. This result is in agreement with an island nucleus volume close to 200 germanium atoms as deduced from an analytical treatment (Rahmati et al. 1996) in which elastic isotropy, equal elastic constants of germanium and silicon, the dipole approximation for the elastic interaction, and a somewhat different definition of the correlation function have been used.

4.3. Three-dimensional versus two-dimensional island nucleus

So far we have assumed that island formation starts from a fully strained planar nucleus. Compared to two-dimensional islands, three-dimensional islands are elastically relaxed at least in part, i.e. their elastic strain energy is reduced. On the other hand, the surface energy contribution is increased. Therefore a three-dimensional

![Figure 5. Correlation function \( C(d_{Si}) \) according to equation (7) depending on the silicon interlayer thickness \( d_{Si} \). The curves bounding the grey shaded area are calculations based on germanium island nucleus volumes \( V_{2D} = 5.9 \) nm\(^3\), and \( V_{2D} = 9.2 \) nm\(^3\). The best fit to the experimental data (*) is found for \( V_{2D} = 7.0 \) nm\(^3\).](image)
island nucleus is inherently unstable below a certain critical volume from an energetic point of view. The condition for three-dimensional island formation is that the energy gain $\Delta E_{ela}$ due to the elastic relaxation has to balance the increased surface energy contribution $\Delta E_{surf}$ caused by island formation according to

$$\Delta E_{ela} + \Delta E_{surf} \leq 0.$$

(8)

In principle we would also have to include a small change in $\Delta E_{surf}$ due to the germanium–silicon interface energy which we neglect in the following. As also demonstrated in previous studies of the elastic relaxation of single layer structures by finite element method simulations (Vanderbilt and Wickham 1991, Christiansen et al. 1994, Tillmann et al. 1996) the elastic strain energy reduction may be described by an effectively reduced lattice mismatch $\langle f \rangle$ compared with the nominal misfit $f$ for completely strained germanium layers on silicon. For islands of pyramidal shape with a square basement and with a varying height-to-extension ratio $p = (h/l)$ the effective mismatch may be written as

$$\langle f \rangle = f(\phi(p)),$$

(9)

where the correction function $\phi(p)$ is obtained from the geometrical mean value of the in-plane strain components $\epsilon_{xx}$ and $\epsilon_{yy}$ of all island-related nodes $i \in \{1 ... n\}$ of the finite element mesh, i.e.

$$\phi(p) = [\phi_1(p) ... \phi_n(p)]^{1/n},$$

(10)

with

$$\phi_i(p) = [\epsilon_{xx}(p) \epsilon_{yy}(p)]^{1/2} / f.$$

(11)

By carrying out corresponding FEM simulations we obtain a discrete set of $\phi(p)$ values. The resulting values for the effective mismatch $\langle f \rangle$ are represented by the black dots in figure 6. These values are well approximated by an exponential function

$$\langle f \rangle = f \exp [-\eta p]$$

(12)

![Figure 6. Effective lattice mismatch $\langle f \rangle$ for pyramidal germanium islands on silicon depending on the height-to-extension ratio $p$. The calculated values (●) as gained by FEM simulations may be adjusted to an exponential function (—).](image-url)
which is given by the solid curve in the figure. For germanium islands on silicon the
decay parameter deduced from the fit amounts to $\eta = 4.452$. For $p \to 0$ equation (12)
yields an effective mismatch $\langle f \rangle (p \to 0) = f$, representing the case of a homogene-
ously strained two-dimensional layer.

The strain energy gain $\Delta E_{\text{ela}}$ for a newly formed three-dimensional nucleus of the
volume $V_{3D}$ is given by

$$\Delta E_{\text{ela}} = \rho_{\text{iso}} \left(1 - \frac{\langle f \rangle^2}{f_0^2}\right) V_{3D},$$  \hspace{1cm} (13)

with

$$\rho_{\text{iso}} = Y_{\text{Ge}} f^2$$  \hspace{1cm} (14)
denoting the strain energy density of a two-dimensional germanium layer on silicon
which is completely strained. Equation (13) is based on the assumption that the
modification of the lattice mismatch between the upper germanium layer and the
silicon interlayer due to the existence of buried germanium islands of the first layer is
negligible. This assumption is well-founded because the magnitude of the long-range
elastic strain field due to the buried germanium islands is more than one order of
magnitude smaller than $f$ even for the smallest silicon interlayer thickness investi-
gated as shown in §4.1.

The increase in the surface energy $\Delta E_{\text{surf}}$ due to three-dimensional island for-
mation is given by the product of the the specific surface energy of germanium $\gamma$ and
an increase of the surface area $\Delta F$ as compared to a two-dimensional layer. For
pyramidal islands with a square base considered here it can be shown from simple
geometrical considerations that $\Delta F$ is given by

$$\Delta F = (1 + 4p^2)^{1/2} - 1.$$

By combining equations (8) to (15) we obtain an estimate of the critical volume $V_{3D}$
for the formation of a three-dimensional island nucleus according to

$$V_{3D} = 9p \left[ \frac{1}{\rho_{\text{iso}} \left(1 - \exp \left[-2\eta p\right]\right)} \left(1 + 4p^2\right)^{1/2} - 1 \right]^3$$  \hspace{1cm} (16)

which for experimental relevant island height-to-extension ratios $p \leq 0.2$ may be
approximated by

$$V_{3D} = 9p \left[ \frac{2\gamma p}{\rho_{\text{iso}} \left(1 - \exp \left[-2\eta p\right]\right)} \right]^3.$$  \hspace{1cm} (17)

An isochor representation of the critical nucleus volume for germanium on silicon is
shown in figure 5 for a range of reasonable $\gamma$ and $p$ values. Among other parameters
the nucleus volume $V_{3D}$ depends on the height-to-extension ratio of the island itself.

For simplification we presume that the germanium islands are characterized by a
fixed height-to-extension parameter during the whole epitaxial growth process and choose
$\gamma = 13.57$ nm/140.67 nm = 1/10.37 as deduced from TEM
measurements at a later stage of growth. The nucleus volume $V_{3D}$ is also determined
by the specific surface energy $\gamma$ of the material forming the island nucleus. Because
reliable $\gamma$ values for LPCVD-grown germanium layers on silicon are lacking we take
values ranging between $\gamma = 1.3$ J m$^{-2}$ and $\gamma = 1.5$ J m$^{-2}$ which were deduced from
the analysis of the surface morphology of epitaxially grown silicon layers (Booker
and Joyce 1966) and from the equilibrium shape of small voids in ion-implanted silicon (Eaglesham et al. 1993). Based upon these \(p\) and \(\gamma\) values critical nucleus volumes are calculated that range between \(V_{3D} = 5.9\, \text{nm}^3\) for \(\gamma = 1.3\, \text{J/m}^2\) and \(V_{3D} = 9.2\, \text{nm}^3\) for \(\gamma = 1.5\, \text{J/m}^2\). This range of values is in agreement with \(V_{2D} = 7.0\, \text{nm}^3\) as obtained by fitting the correlation function according to equation (7) to the experimental data. However, equation (7) is based on the assumption of a completely strained two-dimensional island nucleus while equation (17) takes elastic relaxation as the origin of three-dimensional island formation into account. Therefore an exact quantitative agreement between \(V_{3D}\) and \(V_{2D}\) would only be fortuitous and we cannot decide if the critical nucleus is two- or three-dimensional.

We may however conclude that the calculated \(V_{3D}\) values are in agreement within an order of magnitude with the measured \(V_{2D}\) value obtained from the analysis of the topographical arrangement of islands inside the bilayers. By simply putting \(V_{2D}\) in equation (7) on a level with the numerical \(V_{3D}\) values calculated in this section the grey hatched area in figure 5 represents the whole range of calculated correlation functions. Also included in this range is the best fit to the experimental data gained for a two-dimensional nucleus volume amounting to \(V_{2D} = 7.0\, \text{nm}^3\). This value would be equivalent to a specific surface energy of \(\gamma = 1.37\, \text{J/m}^2\) when \(p = 1/10.37\) is presumed in equation (17) with \(V_{3D} = V_{2D}\). Therefore our FEM-based island nucleation model not only successfully describes the vertical ordering of islands depending on the silicon interlayer thickness but also results in a two-dimensional nucleus volume \(V_{2D}\) which is a quite reasonable fitting parameter. Our range of \(V_{3D}\) values is also consistent with the morphological phase diagrams on the Stranski–Krastanow growth calculated by Ratsch and Zangwill (1993), which delimit conditions under which a uniform-film-to-coherent island transformation may occur from an energetical point of view. Dependent on the applied specific surface energies these diagrams predict the formation of stable germanium islands on silicon.
for an island nucleus formed by 100 up to 600 atoms, i.e. for values being in fair agreement with the $V_{3D}$ values calculated in this study.

§ 5. Composition of the Layers

The coherency of the bilayers beyond the islands’ critical volume for defect formation as well as the miscibility of silicon and germanium in alloys throughout the complete concentration range motivates further investigations concerning the layer composition. If we consider pure germanium islands of pyramidal shape on silicon instead of the tile shaped geometry as discussed by Matthews (1972) and neglect any elastic relaxation we obtain a critical volume $V_c = 644 \text{ nm}^3$ for the nucleation of interfacial misfit dislocations. If elastic relaxation is taken into account by the effectively reduced misfit $\langle \rho \rangle (\rho = 1/10.37) \approx 0.65$ (compare figure 6) the critical volume amounts to $V_c = 975 \text{ nm}^3$, because of $V_c$ being inversely proportional to the effective lattice parameter mismatch. Compared to these values the experimentally observed mean island volume $\langle V \rangle = 894 \pm 76 \text{ nm}^3$ is distinctly larger. Therefore we may guess that the defect generation is kinetically blocked and/or that the composition of the islands differs from that of pure germanium since the critical volume increases with decreasing lattice parameter mismatch equivalent to a reduced germanium content.

Initially we may roughly estimate the average germanium content $\langle x_{Ge} \rangle$ of the Ge$_x$Si$_{1-x}$ layers from the nominal germanium layer thickness $d_{Ge,nom}$ and the layer geometry by simple geometrical considerations according to

$$\langle x_{Ge} \rangle = \frac{d_{Ge,nom}}{\langle l \rangle \langle h \rangle \langle \lambda \rangle}$$

in which $h_{2D}$ represents the thickness of the two-dimensional wetting layer, while $\langle l \rangle$, $\langle h \rangle$ and $\langle \lambda \rangle$ denote the average island length, height, and mean distance, whose numerical values have already been summarized in table 1. Since we cannot gain reliable numerical data on the thickness of the ultrathin two-dimensional Ge$_x$Si$_{1-x}$ layer by measurements on conventional micrographs because of the bending of lattice planes and Fresnel scattering due to the discontinuity in specimen potential which occurs at the interface, we may only calculate the average germanium content for different values of the wetting layer thickness $h_{2D}$ according to table 3. From these results, despite our limited knowledge on the real $h_{2D}$ value, we may conclude that the germanium content is distinctly lower compared to the nominal one but strongly depends on the exact value of $h_{2D}$.

5.1. Lattice fringe analysis of high-resolution micrographs

To get a more accurate estimate of the geometry and composition of the Ge$_x$Si$_{1-x}$ islands and the two-dimensional wetting layer between the islands a digital analysis of lattice fringe images was performed in order to extract local lattice parameters from high-resolution micrographs. The procedure (Tillmann et al. 1996) involves the detection of intensity maxima positions by a centre-of-mass analysis of contrast dots in unfiltered HRTEM images taken along the [110] direction. This analysis step is performed by standard peak finding routines available in the image processing package SEMPER by Saxton et al. (1979). Although high-resolution micrographs generally must not be interpreted in terms of a direct structure image and intensity maxima may not correspond to the actual positions of the
atomic columns, the distances between adjacent intensity maxima locally represent measured distances between atom columns in the absence of chemically sensitive contrast pattern variations. In our case, local (002)-plane distances $d_{002}^\ast$ measured along the growth direction $z$ are calculated from the two-dimensional array of intensity maxima positions. In order to reduce noise the plane distances are averaged inside each (002) plane of the digitized area at the expense of a loss of local information along the [110] direction. With respect to the well-known plane distance $d_{002}^{\text{Si}}$ of the silicon substrate the actual plane distance $d_{002}^\ast z$ of a Ge$_x$Si$_{1-x}$ layer depends on the mismatch $f(z)$ and the component $\varepsilon_{zz}(z)$ of the mismatch induced layer strain along the $z$ direction according to

$$d_{002}^\ast z = (1 + f(z))(1 + \varepsilon_{zz}(z))d_{002}^{\text{Si}} $$

The strain $\varepsilon_{zz}(z)$ of a misfitted layer generally amounts to $\varepsilon_{zz}(z, t \to \infty) = (2c_{12}/c_{11})f(z)$ for a TEM sample of infinite thickness $t$ and to $\varepsilon_{zz}(z, t \to 0) = (c_{12}/c_{11})f(z)$ when a vanishing specimen thickness parallel to the electron beam is considered, respectively, while intermediate values have to be used for experimentally relevant sample thicknesses. By application of Vegard’s law, i.e. $x_{\text{Ge}}(z) = f(z)\frac{x_{\text{Ge}}}{x_{\text{Si}}} + (1 - f(z))\frac{x_{\text{Si}}}{x_{\text{Si}}}$ we may however give a lower and an upper limit of the local germanium content $x_{\text{Ge}}^\ast(z)$ according to

$$x_{\text{Ge}}^\ast(z, t \to \infty) = \left(\frac{d_{002}^\ast}{d_{002}^{\text{Si}}} - 1\right) \frac{1}{1 + c_{12}/c_{11} f} \frac{1}{f},$$

$$x_{\text{Ge}}^\ast(z, t \to 0) = \left(\frac{d_{002}^\ast}{d_{002}^{\text{Si}}} - 1\right) \frac{1}{1 + 2c_{12}/c_{11} f} \frac{1}{f}$$

We emphasize that this method of quantitative compositional mapping by the direct measurement of lattice spacings from high-resolution micrographs is limited in accu-

<table>
<thead>
<tr>
<th>$h_{2D}$ (ML)</th>
<th>$(x_{\text{Ge}})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>0.533 ± 0.093</td>
</tr>
<tr>
<td>5</td>
<td>0.456 ± 0.009</td>
</tr>
<tr>
<td>6</td>
<td>0.390 ± 0.052</td>
</tr>
<tr>
<td>7</td>
<td>0.333 ± 0.041</td>
</tr>
<tr>
<td>8</td>
<td>0.281 ± 0.033</td>
</tr>
<tr>
<td>9</td>
<td>0.230 ± 0.028</td>
</tr>
</tbody>
</table>

Table 3. Calculated average germanium content $(x_{\text{Ge}})$ of the Ge$_x$Si$_{1-x}$ layers assuming different thicknesses $h_{2D}$ of the two-dimensional wetting layer according to equation (18). In the self-consistent approach $h_{2D}$ values are given in (002)-monolayer distance units (ML) of a silicon–germanium alloy with a germanium content amounting to $(x_{\text{Ge}})$. For the calculation, geometrical parameters according to table 1 have been used which determine the margins of error in the $(x_{\text{Ge}})$ values.
racy because of two effects. Firstly, the thin-foil relaxation of the HRTEM-specimen (Gibson and Treacy 1984) may lead to modified strain values of the sample and secondly the electron delocalization (Coene and Jansen 1992) during electron diffraction and the microscopic imaging process may cause an artificial blurring of the interface contrast leading to errors in the determination of the layer thickness in the order of one or two monolayers at each interface (Tillmann et al. 1999). However, the procedure may be applied to get a rough estimate on the average layer stoichiometry of germanium–silicon alloys as it can be shown by measurements on simulated images for a wide range of defocus and specimen thickness values.

5.2. Composition of the Ge\textsubscript{x}Si\textsubscript{1-x} layers

Figure 8 (a) shows a high-resolution micrograph of the two-dimensional germanium-rich wetting layer embedded in silicon and taken along the [110] projection in Vertical ordering of islands in GeSi–Si bilayers on Si(001)

Figure 8. (a) Cross-sectional HRTEM micrograph of the coherent interfaces of the two-dimensional germanium-rich wetting layer embedded in silicon taken along the [110] projection. Contrast across the interface arises from different scattering factors of silicon and germanium as well as strain induced local misorientations of the specimen. Numerical data (●) on the mean germanium content $x_{\text{Ge}}(z)$ across the interface is obtained by averaging local (002)-monolayer distances along the [110] direction. Positions along the growth directions are given in (002) monolayers along the $z$ axis. The black dots represent $x_{\text{Ge}}$ values deduced by assuming a specimen thickness $t = 0$ (upper curve) and $t \to \infty$ (lower curve). The two solid curves bounding the grey shaded area follow from an interpolation.
the case of a sample with $d_{Si} = 40 \text{ nm}$. Contrast pattern modifications between the wetting layer and the surrounding matrix arise due to different scattering properties of silicon and germanium as well as strain-induced local specimen misorientations. Contrast pattern shifts due to sample thickness variations are not observed in both, the silicon substrate and the silicon interlayer, indicating that the image is suitable for the previously described lattice-fringe analysis. A corresponding composition profile, as gained by employing the procedure according to § 5.1, is given in figure 8(b). Black dots are deduced from the measurement assuming $t = 0$ (upper profile) and $t \rightarrow \infty$ (lower profile), respectively, while the solid curves result from an interpolation between both types of dots and the grey shaded area between the two solid curves reflects the uncertainty in the determination of the composition profile resulting from the analysis of the high-resolution micrograph. If we associate all $x_{Ge}(z)$ values larger than the maximum value of fluctuation inside the silicon area of the plot to the $Ge_xSi_{1-x}$ layer, we gain a total wetting layer thickness of approximately nine (002) monolayers along the [001]-growth direction (interface positions labelled 'a' and 'b' in figure 8(b)). The composition profile $x_{Ge}(z)$ is characterized by a strong increase of the germanium content along the growth direction reaching a maximum value in the order of $x_{Ge}^{\text{max}} \approx 0.55$ and a distinctly weaker decay towards the silicon interlayer. The mean germanium content $\langle x_{Ge}\rangle$ of the two-dimensional layer, calculated as

$$\langle x_{Ge}\rangle = \int_{z=a}^{z=b} x_{Ge}(z)/(b-a)$$

yields $\langle x_{Ge}\rangle = 0.40 \pm 0.06$ wherein the uncertainty results from the unknown HRTEM sample thickness. This value indicates a considerable deviation of the layer composition compared to the deposition of pure germanium.

Figure 9(a) shows a high-resolution micrograph of the lower interface of a $Ge_xSi_{1-x}$ island and the adjacent silicon substrate in the case of a sample with a silicon interlayer thickness of $d_{Si} = 40 \text{ nm}$. Again, homogeneously distributed contrast pattern variations arise because of different scattering properties of silicon and germanium. By applying the image processing routine to the high-resolution micrograph we find an average germanium content of $\langle x_{Ge}\rangle = 0.43 \pm 0.05$, cf. figure 9(b). This value is of the same order of magnitude as gained for the two-dimensional wetting layer.

An average germanium content in the range between $0.34 \leq \langle x_{Ge}\rangle \leq 0.48$ deduced by the lattice parameter analysis of high-resolution micrographs is in agreement with the $\langle x_{Ge}\rangle$ values according to table 3 as calculated from the layer geometry and the nominal layer thickness assuming a wetting layer thickness $h_{2D}$ in the range between five and nine (002) monolayers. Corresponding $h_{2D}$ values are consistent with the compositional profile in figure 8(b) when we take into account the previously mentioned artificial blurring of the layer thickness of the order of one or two monolayers at both interfaces. The results also agree with photoluminescence measurements taken from identical samples yielding $\langle x_{Ge}\rangle \leq 0.50$ for the islands as well as for the two-dimensional wetting layer (Rahmati 1996, Hartmann 1997, Tillmann et al. 1997).
5.3. Origins of compositional modifications

During epitaxial growth of lattice parameter mismatched heterostructures surface diffusion of adatoms enables the evolution of morphological fluctuations, e.g. undulated surfaces or three-dimensional islands. From an energetic point of view this formation of non-planar surfaces results from a lowering of the total layer strain energy at the expense of an increasing total surface energy compared to the two-dimensional growth regime (Grinfeld and Srolovitz 1995). In detail the wavelength of the instability usually is long compared to the layer thickness and the growing layer has an apparent critical thickness due to a kinetic stabilization below which morphological fluctuations do not develop (Jesson et al. 1997). Nevertheless, a number of experiments have demonstrated that germanium layers tend to corrugate on silicon when growth conditions are properly chosen (Kuan and Iyer 1991, Cullis et al. 1992, Jesson et al. 1993). Moreover, compositional fluctuations preserving the amount of deposited material represent an alternative path of energy reduction by lowering the lattice parameter mismatch induced strains.

Figure 9. (a) Cross-sectional HRTEM micrograph of the coherent interface between the substrate and a buried island taken along the [110] projection. Contrast across the interface arises from different scattering factors of silicon and germanium as well as strain induced local misorientations of the specimen. (b) Numerical data (●) on the mean germanium content $x_{\text{Ge}}$ across the interface is obtained by averaging local (002)-monolayer distances along the [110] direction. Positions along the growth directions are given in (002) monolayers along the $z$ axis. The black dots represent $x_{\text{Ge}}$ values deduced by assuming a specimen thickness $t = 0$ (upper curve) and $t \rightarrow \infty$ (lower curve). The two solid curves bounding the grey shaded area follow from an interpolation.
In the case of the observed Ge₅₃Si₂₇–Si bilayer systems both strain relief mechanisms, island formation and intermixing, are present. The relief of strain energy by island formation has already been discussed in §4 and the significant deviations from the measured and the nominal germanium contents of both layer and island indicate that an alloy of silicon and germanium has formed during depositions of the heterostructures. This intermixing may be attributed to an exchange of lattice sites of silicon and germanium at already existing interfaces by interdiffusion and to a floating of germanium adatoms towards the free surface during epitaxial growth by segregation. Direct evidence for a substantial intermixing of silicon and germanium under comparable experimental conditions has been obtained from the analysis of layer compositions of short-period Ge₅₃Si₂₇–Si superlattices after annealing treatment between 600°C and 800°C (Stenkamp 1993, W. Jäger 1998, private communication). At least in the case of low-misfit systems segregation will be an important figure during LPCVD growth under conditions similar to those used in this study as shown by systematic parameter investigations (Walther et al. 1997b). In our case the asymmetric broadening of the two-dimensional wetting layer indicates that segregation will also contribute to the compositional intermixing.

Compositional modifications of germanium-rich layers and islands embedded in silicon have also been observed in a number of previous investigations focusing on heterostructures grown by LPCVD and MBE at similar growth temperatures, for both highly lattice mismatched Ge₅₃Si₂₇–Si heterostructures (Libera et al. 1993, Hovsepian et al. 1997) and low-misfit Ge₅₃Si₂₇–Si systems (Stenkamp and Jäger 1993, Carlino et al. 1996, Walther et al. 1996). In the case of undulated Ge₅₃Si₂₇ layers on silicon with a nominal germanium content of x = 0.20 recent investigations demonstrated a significant germanium enrichment Δx in ripple peaks and near the growth surface of ripple troughs at the expense of a reduced germanium content in other areas of the sample with a relative intermixing rate, amounting up to Δx/x = 0.35 (Walther et al. 1997a). Our results suggest that modifications are even more pronounced in the Stranski–Krastanow growth mode for deposition of pure germanium on silicon compared to the undulating growth regime and yielding −Δx/x values between 0.52 and 0.64 as gained in this study.

5.4. Impact of compositional modifications on the vertical ordering of islands

Concerning the vertical correlation of islands as modelled in §4 we would like to discuss the impact of compositional modifications on the island ordering since our numerical treatment of the layer strains assumed pure germanium layers embedded in silicon. The nucleation and ordering of islands at the growth surface is determined by inhomogeneities of strains at the growth surface, i.e. by the composition of the islands in the buried layer. As shown by our measurements these compositions deviate considerably from the nominal stoichiometry. Consequently, local strain values will moderate and result in different values of the correlation function and in an alteration of island nucleus volumes for reduced germanium contents.

Concerning the case of islands inside the bilayers characterized by a fixed geometry a decreased germanium content of the buried layer diminishes the strain inhomogeneity at the growth surface because of a reduced elastic deformation of the silicon interlayer by a buried island. Therefore the difference Δε of the mean strain values between the area above the buried island and above the wetting layer will be reduced by a factor ⟨x_{Ge}⟩, i.e. we have to replace Δε(d_{Si}) in equation (7) by
Moreover, the lattice mismatch $f$ of the newly formed nucleus also has to be replaced by $\langle x_{Ge} \rangle f$ in the correlation function if we think about the formation of the silicon germanium alloy already in the earliest stages of growth. Since all other parameters in equation (7), except for the nucleus volume $V_{2D}$, are either material properties or are determined by the layer geometry, we consequently have to use a $V_{2D}$ value increased by a factor of $\langle x_{Ge} \rangle^{-2}$ to adapt the correlation function $C(d_{Si})$ to the experimental data gained by transmission electron microscopy.

To reach conformity with the critical volume for three-dimensional island formation according to equation (17), which for an intermixed system predicts an increase of $V_{3D}$ by a factor of $\langle x_{Ge} \rangle^{-6}$ because of $g_{iso}$ following a $f^2$ law, we have to consider one or more of the following alternatives to slash an excessive increase in $V_{3D}$ values. At first we could assume a decreased specific surface energy $\gamma$ for germanium set against the values for silicon used in this study. A corresponding modification will abate the nucleus volume as can be seen from the contour representation in figure 7 by performing a cut along the $fi$ axis. This finding would be consistent with earlier results (Keck and van Horn 1953) demonstrating that the surface tension of liquid germanium is somewhat lower compared to that of silicon. Alternatively a lowered height-to-extension ratio for the newly formed nucleus compared to the $p$ values, depicted at a later stage of growth by cross-sectional micrographs, may be assumed, as again can be seen from figure 7 by performing a cut along the $p$ axis. This explanation would also be in agreement with results demonstrating that the strong compressive strains near the island edges prevent lateral adatom incorporation in germanium islands on silicon which results in a primarily vertical growth of islands during the later stages of epitaxial growth (Voigtländer and Zinner 1993, Dorsch et al. 1995). Moreover, we may think about the formation of the silicon germanium alloy at a later stage of growth and assume that the initial island nucleus still contains a significantly higher fraction of germanium than measured by the analysis of high-resolution micrographs. This assumption would once more emphasize that intermixing also originates from interdiffusion after the germanium layers have been overgrown with silicon.

However, since a wide range of reasonable $\gamma$, $p$ and $\langle x_{Ge} \rangle$ values for a diluted island nucleus may be used to gain a critical volume $V_{3D}$ scaling with a $\langle x_{Ge} \rangle^{-2}$ law we cannot quantify the contribution of all three alternatives by performing ex situ TEM experiments. Nonetheless, the basic predictions of the presented model describing the vertical ordering of larger islands are not altered by our limited knowledge about nucleus formation although slightly different values on the nucleus’ geometry and material properties are also consistent with our results. We may conclude by qualitative reasons that in the earliest stages of growth (i) the germanium content of a newly formed nucleus may be larger than those experimentally obtained later on by the digital analysis of HRTEM images of larger islands, (ii) the height-to-extension ratio of the islands increases during growth and/or (iii) that the specific surface energy of germanium is somewhat lower compared to silicon.

§ 6. CONCLUSIONS

The vertical self-organization of coherently strained islands in Ge$_{x}$Si$_{1-x}$–Si bilayers grown by low pressure chemical vapour deposition on Si(001) has been studied by transmission electron microscopy and analysed quantitatively by carrying out finite element method simulations. The ordering of islands is attributed to a long-range elastic interaction between buried islands and a newly formed nucleus growing
on top of a silicon interlayer. Under the chosen growth conditions the vertical correlation between islands decreases above an interlayer thickness of 276 nm due to a decrease of local strain fluctuations at the interlayer surface. An island nucleation model has been quantitatively evaluated by finite element simulations taking fully into account elastic anisotropy and the real layer morphology. An island correlation function $C(r_0)$ has been derived and by fitting $C(r_0)$ to experimental values the volume of a newly formed island nucleus is gained. Its value of 7.0 nm$^3$, corresponding to approximately 320 germanium atoms is in good agreement with energetic considerations on island formation.

A lattice parameter analysis of high-resolution micrographs shows that the deposition of pure germanium on silicon results in wetting layer and island compositions containing considerable fractions of silicon. This observation suggests that mixing processes leading to the formation of a silicon germanium alloy occur during epitaxial growth of the bilayer systems. The deviations between measured and nominal compositions have implications for the layer strain, the formation of islands and their ordering in bilayers, as described by the correlation function. If elastic strains of individual layers are moderated during deposition, or subsequently grown layers due to alloying the dilatation strains above buried islands will become smaller, hence reducing the interaction between buried and newly forming islands and resulting in slightly different nucleus volume values. Nevertheless, these compositional modifications do not affect the basic predictions concerning the vertical ordering of islands since necessary modifications of structural nucleus parameters during the earliest stages of growth can be reasonably motivated.

References

Vertical ordering of islands in GeSi–Si bilayers on Si(001)


