Dependence of indium incorporation upon the substrate misorientation during growth of $\text{In}_x\text{Ga}_{1-x}\text{As}$ by metalorganic vapour phase epitaxy

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Abstract

The incorporation of indium during growth of $\text{In}_x\text{Ga}_{1-x}\text{As}$ (with $x < 5 \times 10^{-3}$) by metalorganic vapour phase epitaxy on GaAs substrates with orientations vicinal to (100) has been studied as a function of the growth temperature, the input mole fraction of trimethylindium (TMI) and the misorientations of the substrates towards (011) and (011). The average apparent activation energy for indium incorporation is found to be 9.7 kcal mol$^{-1}$, which is explained by the presence of indium adducts in the gas phase at low temperatures which decompose at higher growth temperatures. The differences in indium concentrations as a function of the direction and the degree of the misorientation are explained by considering the incorporation processes at the group III sublattice sites at the steps, thereby taking into account the arsenic coverage of the surface. The behaviour of the indium incorporation as a function of the misorientation is described by diffusion of growth species towards the steps, according to the BCF theory, which is counteracted by preferential arsenic desorption from the step edges.

1. Introduction

The optimization of the metalorganic vapour phase epitaxy (MOVPE) process has offered the possibility of growing epitaxial layers of III–V semiconductor materials of high optical, electric and morphological quality. These layers have found their applications in the optoelectronic industry for the production of devices as lasers and photodetectors. In most of these devices, structures such as multiple quantum wells (MQW) and strained layer superlattices (SLS) are of great importance for good operating performance [1,2]. Since the dimensions of these structures are decreasing more and more, a good control of the growth process is needed, in order to obtain homogeneous compositions of ternary and quaternary compounds, homogeneous dopant concentrations in the layers and extremely sharp interfaces between two subsequent layers. In order to realize this, a thorough knowledge of the growth mechanisms of the MOVPE process is required.

In studies on growth of III–V compounds by MOVPE a lot of attention has been paid to optimize the deposition conditions in the reactor during the growth processes [3], whereas the effect of the orientation and the misorientation of the substrate surfaces...
on the composition and the quality of the grown epitaxial layers has been studied in less detail. It is known that the incorporation process at the surface is influenced by the degree of misorientation of the substrate, resulting in differences in the lateral growth rate and in the impurity concentration [4–8]. An overview of studies on orientation dependence of impurity incorporation during MOVPE growth of III–V compounds has been given by Kondo et al. [9]. In their paper a description is given of the incorporation processes on exactly oriented (h11)A and (h11)B surfaces, including (100), (111)A and (111)B. However, in order to obtain more insight into the incorporation processes at the steps present at the surface during growth, one should not only study the growth processes on exactly oriented substrates, but also on substrates with vicinal orientations. Recently, Anders et al. [7] have investigated the misorientation dependence of zinc incorporation in GaAs in more detail around the (100) orientation by misorienting the substrate surface towards (111)A and (111)B up to a maximum of 6°. It has been found that zinc incorporation at the group III sublattice sites is influenced by “step trapping” at small misorientations and the increase of available kink sites at higher misorientations.

In the present study the incorporation of the isoelectronic dopant indium during MOVPE of GaAs has been investigated as a function of the indium concentration in the gas phase, the growth temperature and the substrate misorientation. The influence of the different configurations at the steps at the surfaces on the incorporation of indium is discussed and compared with results of incorporation studies as described in the literature.

2. Experimental details

The incorporation processes at the steps during MOVPE have been investigated by growing In_xGa_1-xAs epitaxial layers on misoriented (100) semi-insulating GaAs substrates. The misorientation is chosen in the direction of the principal steps at the (100) surface, i.e. towards the [011] and the [01̅] directions. The step direction is defined as the step normal, parallel to the major flat surface. The following orientations have been used: (100), (100)2°(011), (100)2°(01̅), (100)4°(011), (100)4°(01̅), (100)6°(011) and (100)6°(01̅). The accuracy of the misorientation angle was better than 0.5°, as specified by the supplier.

On these substrates undoped In_xGa_1-xAs epitaxial layers have been grown by low-pressure MOVPE in a horizontal, radiation heated, cold-wall reactor. Trimethylgallium (TMG), trimethylindium (TMI) and arsine (AsH₃) have been used as source materials. The growth temperature was varied between 640 and 760°C. The reactor pressure was 20 mbar. The arsine partial pressure was 0.3 mbar. The ratio of the arsine (V) and the organometallic precursors TMG and TMI (III) partial pressures, the V/III ratio, at the entrance of the reactor was 125.

The substrates were placed in the reactor in the area where under the growth conditions as described above the temperature and the flow profiles are fully developed [10] and no return flows are present [11,12]. The vertical growth rate was between 0.4 and 0.6 nm/s, depending on temperature. The total thicknesses of the In_xGa_1-xAs layers, between 1.6 and 1.8 μm, were below the critical layer thickness for the formation of misfit dislocations [13], caused by the difference in lattice constant between the GaAs substrates and the In_xGa_1-xAs layers. In order to check the reproducibility, these experiments have been performed in duplicate.

The indium concentrations in the completely relaxed epitaxial layers were determined from photoluminescence (PL) spectra. The energy bandgap of the In_xGa_1-xAs layers is shifted with respect to that of GaAs by the indium concentration in GaAs in more detail around the (100) orientation by misorienting the substrate surface towards (111)A and (111)B up to a maximum of 6°. It has been found that zinc incorporation at the group III sublattice sites is influenced by “step trapping” at small misorientations and the increase of available kink sites at higher misorientations.

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the PL spectra of GaAs homoepitaxial layers [18]. The PL measurements were performed at 4.3 K with the sample in helium exchange gas. Optical excitation was provided by the 2.41 eV (514.5 nm) line from an Ar$^+$ laser. The luminescence was dispersed by a 0.6 m double monochromator and detected by a cooled photomultiplier tube with a GaAs photocathode. The energy resolution of these measurements was better than 0.1 meV around the spectral range of the bandgap of GaAs, corresponding to an accuracy of the determination of the indium content in the In$_{x}$Ga$_{1-x}$As layers of $1 \times 10^{-4}$.

3. Results

All the In$_{x}$Ga$_{1-x}$As layers showed a specular morphology: no cross-hatched patterns, indicating the presence of misfit dislocations, have been observed at the surface. From this it is concluded that the layers are totally unrelaxed. The photoluminescence spectra of these layers, of which a typical example is given in Fig. 1, showed a clear splitting of the exciton bound peaks and a relatively low band acceptor peak related to carbon impurity. This proves that the grown layers are of a high optoelectronic quality. The indium concentrations in the layers have been determined from the shift of the donor bound exciton (D$^0$X) peak with respect to that of GaAs.

The indium concentration, $x$, in the In$_{x}$Ga$_{1-x}$As layers, grown on (100)$^\circ$(011) and on (100)$^\circ$(011) substrates at 640°C, is shown in Fig. 2 as a function of the input mole fraction of TMI relative to the total mole fraction of group III species $x_G$. It is shown that the indium incorporation in the layers grown on the (100)$^\circ$(011) substrates is slightly, but significantly, higher than that in the layers grown on the (100)$^\circ$(011) substrates.

In order to study the temperature dependence of the indium incorporation a series of experiments has been performed at temperatures between 640 and 760°C. During these experiments all the other growth conditions were kept constant; the input mole fraction of TMI was $4.3 \times 10^{-7}$, as compared to the TMG input mole fraction of $1.25 \times 10^{-4}$. An Arrhenius plot of the indium incorporation for all the orientations used in this study is shown in Fig. 3. It appears that the indium incorporation during MOVPE of GaAs is enhanced with increasing temperature, resulting in a positive value of the apparent activation energy. When averaged over all the misorientations towards (011), including the exact (100) samples, a value of 9.7 kcal mol$^{-1}$ is found (Fig. 3a). The apparent activation energy for the samples misoriented 2$^\circ$ and 4$^\circ$ towards (011) (Fig. 3b) is somewhat smaller (7.2 and 8.5 kcal mol$^{-1}$ respectively), whereas for the samples grown on the (100)$^6^\circ$(011)-oriented substrates it is somewhat higher (11.4 kcal mol$^{-1}$). The indium distribution coefficient, i.e. the
ratio $x_g/x_s$, averaged over all the misorientations at a growth temperature of 640°C is 0.65, which is comparable to the value found by Kuo et al. [21], and increases to unity when the growth temperature is increased to 760°C.

The dependence of the indium incorporation during MOVPE growth of GaAs on the misorientation towards (011) and (01̅1) is shown in Fig. 4 for four different growth temperatures. In this figure the results of the (100) exact orientation are not shown, since the local variations in misorientation results in a large scattering in the incorporation data. The input TMI mole fraction in the gas phase was $4.3 \times 10^{-7}$ for all the experiments. It appeared from duplicate experiments that the indium concentrations can be reproduced within $2 \times 10^{-4}$. From this it is concluded that the deviations observed from these duplicate experiments are smaller than the differences in indium concentration determined in the layers with the various misorientations as shown in Fig. 4.

At all growth temperatures the indium concentration in the layers shows a minimum value at a misorientation of 4° towards (011), whereas it shows a maximum at a misorientation of 4° towards (01̅1). At a growth temperature of 640°C the indium concentration in the (100)2°(011)-oriented sample is higher than that in the (100)2°(01̅1)-oriented sample, which was also shown in Fig. 2. For higher growth temperatures, i.e. 680 and 720°C, these differences in the indium concentrations between the two different misorientations decrease. Finally, at a growth temperature of 760°C, the indium concentration in the (100)2°(011)-oriented sample is lower than that in the (100)2°(01̅1)-oriented sample, which is just the opposite of the situation at the lower growth temperatures.
4. Discussion

The indium incorporation experiments during growth of GaAs used in this study are performed in the temperature range in which the GaAs deposition is controlled by the diffusion of the gallium species through the gas phase [22], except for the growth at 760°C, which is in the transition zone between the diffusion controlled and the thermodynamic temperature range. The diffusion controlled growth makes the differences in the indium concentration observed in the layers smaller and therefore more difficult to interpret than in the case that the growth is limited by the incorporation at the steps of the species that are present at the substrate surface. However, as is observed from duplicate experiments and from the temperature series in Fig. 4, the differences in the indium concentrations between the various misorientations are reproducible and significant. Therefore, the interpretation of the observed differences in the indium concentrations requires a more detailed description of the incorporation processes at the two different step orientations on the (100) surface as a function of temperature and of the degree of misorientation.

4.1. Temperature behaviour of indium incorporation

The general trend in the growth of In$_x$Ga$_{1-x}$As found for all the misorientations studied is that the indium incorporation process shows a positive apparent activation energy with an average value of 9.7 kcal mol$^{-1}$. This trend can be explained by the presence of an adduct of TMI and AsH$_3$, which dominates at lower growth temperatures [3]. The presence of this adduct results in a reduced indium incorporation rate at lower growth temperatures since the diffusion flux of the adduct to the surface is smaller because of the smaller diffusion coefficient and the larger Soret effect. At higher temperatures the adduct becomes less stable and decomposes with an enthalpy of 10 kcal mol$^{-1}$, raising in this way the surface concentration of indium. The indium distribution coefficient averaged over all the misorientations increases from 0.65 to 1 when the growth temperature is increased from 640 to 760°C.

The positive value for the reaction enthalpy found in this study is in contrast to the results reported in the literature for the growth by MOVPE of In$_x$Ga$_{1-x}$As on InP [23] and of In$_x$Ga$_{1-x}$P on GaAs [24] with $x \approx 0.5$; in the latter studies it is found that increasing the temperature leads to a decrease in indium incorporation. In these cases this is explained by either a removing of indium from the gas phase by parasitic reactions, or by an enhanced desorption of indium from the surface [25]. It appears that in our case of very low indium input mole fractions the presence of the adduct in the gas phase has a more pronounced influence on the final indium concentrations in the layers than in the case of high input mole fractions.

4.2. Anisotropic indium incorporation at the steps

There are three factors which give rise to an anisotropic behaviour of the incorporation: the kink density at the step, the arsenic coverage of the step and the diffusion fluxes over the surface to the step. These effects will be treated first in this order.

4.2.1. Kink density

Under arsenic-rich conditions, which are normally used during low-pressure MOVPE, the GaAs(100) surface has a c(4 × 4) reconstruction because of dimer bond formation between neighbouring arsenic atoms, which cover the arsenic surface [26]. Due to this reconstruction the steps in the [011] and the [01\bar{1}] directions on the arsenic surface [26]. Due to this reconstruction the steps in the [011] and the [01\bar{1}] directions on the (100) surface show a different behaviour [5,27]. The step in the [011] direction, or A-step, is more undulated than the B-step in the [01\bar{1}] direction [28]. On an atomic scale, the [01\bar{1}] step is a stable step, consequently the [01\bar{1}] step front is atomically rough. At small misorientations, each group III species arriving at the step edge can form a triple bond at any step site and is directly incorporated. Desorption from this position is not possible. On the other hand, the [01\bar{1}] step is fully kinked, i.e. the [01\bar{1}] step front is atomically rough. At small misorientations, each group III species arriving at the step edge can form a triple bond at any step site and is directly incorporated. Desorption from this position is not possible. On the other hand, the [01\bar{1}] step is stable step, consequently the [01\bar{1}] step front is straight with an occasional kink site. Group III species arriving at this step can only adsorb by the formation of a single bond. For an adsorbed species the activation energy for diffusion is lower than the energy required for breaking an arsenic dimer bond at a step site to get incorporated [29], so diffusion along the step or to the surface is favoured above direct incorporation, involving the breaking of dimers. Incorporation of
group III species at the [011] step only takes place at the specific kink sites [30], where a double bond can be formed.

Although the [011] step at small orientations is stable and straight, this will no longer be true at high misorientation when due to local irregularities short range variations will lead to a roughening of the step. However, this will not lead to a higher kink density, as will be treated in the next section.

4.2.2. Arsenic coverage

An increase in temperature generally leads to a decrease in arsenic coverage of the (100) surface because of arsenic desorption [3] and, consequently, to a decrease in the reconstruction by the formation of arsenic dimer bonds. This also strongly affects the atomic arrangements at the steps. The desorption of arsenic species at the [011] steps, possessing only one back bond, is enhanced [31–33] as compared to arsenic species at the [011] steps, which have two back bonds. Since the [011] step is initially fully kinked, the arsenic desorption leads to a smaller number of kink sites at this step for group III growth species. At the [011] steps the concentration of incorporation sites for group III species, and therefore the lateral growth rate of GaAs, at small misorientations is hardly influenced by the arsenic partial pressure [5]. However, at larger misorientations especially the straight [011] step will become much rougher as discussed above. The consequence is that at larger misorientations the arsenic evaporation at the [011] step will also strongly increase, even at low temperatures. This will lead to a strong reduction of available kink sites.

In short, for small misorientations and arsenic-rich conditions the [011] step is rough and possesses a high kink density for group III species, whereas the [011] step is stable and the group III species have to diffuse along the step to the kink sites. At high temperatures the high kink density at the [011] step is strongly reduced, whereas the number of kink sites at the stable [011] step is hardly affected for small misorientations. The arsenic evaporation at the [011] step will only occur at high misorientations.

4.2.3. Diffusion fluxes

The incorporation process as a function of the misorientation has been described by Anders et al. [7] as a competition between diffusion fluxes of growth species towards the steps, based on the crystal growth theory of Burton, Cabrera and Frank (the BCF theory) [34]. It was shown for the incorporation of zinc during MOVPE of GaAs that in the case that the growth is determined by the adsorption/desorption equilibrium of one of the growth species at the steps, the incorporation is increased with increasing misorientation angle for that species which has the lowest binding energy with arsenic. The physical reason is that for low misorientations and large terrace lengths the species with the smallest binding energy has a higher chance to desorb. At higher misorientations the step distance becomes smaller than the surface diffusion length of the particular species, resulting in a higher incorporation rate. This theory can be adapted for the indium incorporation, thereby assuming that the surface concentration of the indium species, i.e. the species with the lowest binding energy, near the step is not zero. The surface concentration of adsorbed gallium species in the vicinity of the steps is assumed to be zero, which is the case for the diffusion controlled growth and not too high temperatures. For the fluxes of indium respectively gallium towards the steps we obtain

\[
r(\text{In}) = 2k_a c^0_{\text{In}} \lambda_{\text{In}} \sinh \left( \frac{y_0}{2 \lambda_{\text{In}}} \right) \left[ \cosh \left( \frac{y_0}{2 \lambda_{\text{In}}} \right) + \left( \frac{D_{\text{In}}}{k_{\text{inc}} c_{\text{In}}} \right) \sinh \left( \frac{y_0}{2 \lambda_{\text{In}}} \right) \right]^{-1}
\]

and

\[
r(\text{Ga}) = 2k_a c^0_{\text{Ga}} \lambda_{\text{Ga}} \tanh \left( \frac{y_0}{2 \lambda_{\text{Ga}}} \right),
\]

in which \( k_a \) and \( k_d \) are the rate constants for adsorption and desorption from the gas phase, \( k_{\text{inc}} \) is the rate constant for incorporation at the step, \( c^0 \) is the concentration at the middle of the terrace, \( y_0 \) is the width of the terrace, \( \lambda \) is the surface diffusion length before desorption, calculated by the relation \( \lambda = \sqrt{D_s \frac{y_0}{k_d}} \), where \( D_s \) is the surface diffusion coefficient, given by \( D_s = \frac{1}{4} \nu \tau \exp(-E_m/kT) \) and \( k_d = \nu \exp(E_b/kT) \), \( \nu = 10^{13} \text{ s}^{-1} \), \( \tau = 0.38 \text{ nm} \), which is the “jump” distance at the GaAs(100) surface and \( E_m \) is half the binding energy. Using \( E_\text{Ga}(\text{Ga}) = 39.4 \text{ kcal mol}^{-1} \) [7] and \( E_\text{In}(\text{Ga}) = 35.2 \text{ kcal mol}^{-1} \) [25,35], we obtain for the surface diffusion lengths of Ga and In at the temperatures of 640 and 760°C that \( \lambda_{\text{Ga}} \) is 44 and 24 nm, and \( \lambda_{\text{In}} \) is 25 and 14 nm,
respectively. The terrace length \( y_0 \) decreases from 8.6 to 2.8 nm upon increase of the substrate misorientation angle from \( 2^\circ \) to \( 6^\circ \). From this it can be shown [7] that the flux ratio \( r(\text{In})/[r(\text{In})+r(\text{Ga})] \) should increase with decreasing \( y_0 \), i.e. with increasing misorientation angle, the grown material should become more rich in indium.

We now return to the discussion of the experimental results. The differences in incorporation mechanisms at the [011] and the [01\{1\}] steps are reflected by the difference in incorporation behaviour of indium as a function of the misorientations towards (01\{1\}) and (01\{1\}) in Figs. 2 and 4. From the above discussion it follows that under arsenic-rich conditions, i.e. at high arsenic coverages of the steps, both group III species will be incorporated directly at the step sites of the [01\{1\}] steps, since this step is fully kinked. Desorption from this step only can take place at the terraces between the steps. The situation at the [01\{1\}] step is different. Here the group III atoms also might desorb from the step. As indium has a lower bond strength with arsenic than gallium [25,35], it will have a higher desorption probability than gallium as long as it is attached to this step. This higher desorption probability during the diffusion process from the surface along the step to a kink site results in a lower indium incorporation at the stable [01\{1\}] steps, compared to that at the kinked [01\{1\}] steps at substrates with the same degree of misorientation under the same growth conditions (Fig. 2). At higher temperatures the kink density at the [01\{1\}] steps is reduced. This leads to a lower increase in indium incorporation compared to that at the [01\{1\}] steps, where for small misorientations the arsenic concentration is less sensitive to the temperature. This behaviour is experimentally observed for the couple \((100)2^\circ(011)-(100)2^\circ(01\{1\})\) as can be seen in Fig. 4: initially at 640°C the indium concentration in the (100)2\(^\circ\)(011)-oriented layer is higher than that in the layer grown on (100)2\(^\circ\)(01\{1\}), slowly turning to the situation at 760°C, where the indium concentration in the (100)2\(^\circ\)(011) layer is lower than that in the (100)2\(^\circ\)(01\{1\}) layer.

An increase in indium incorporation is expected for misorientations larger than \( 2^\circ \), as is discussed above using the BCF theory [7]. The increase of the indium concentration observed for all growth temperatures upon increase of the misorientation from \( 2^\circ \) to \( 4^\circ \) towards (01\{1\}), indeed indicates that in this situation the indium incorporation during growth of GaAs proceeds via an adsorption/desorption equilibrium as described above (Fig. 4).

However, the flux model does not predict the decrease in indium incorporation observed at higher misorientations towards (01\{1\}). Although we have no strong evidence, we believe that this can be explained by the preferential arsenic evaporation from the step edges [31], which becomes apparent at misorientations of \( 6^\circ \) and higher. The roughening of the [01\{1\}] step, which is expected for high misorientations, will lead to a stronger arsenic evaporation and consequently less kink sites for group III species will be present. The higher step densities at higher misorientation angles require a higher arsenic partial pressure in order to maintain the full arsenic coverage of the steps as was assumed in the theory. Therefore, under the constant arsenic partial pressures in the present series the indium species arriving at the steps will find less kink sites, resulting in a decrease in indium concentration in the layer. Apparently, this is not counteracted by the increase in kink site concentration due to the higher step density.

In contrast to the incorporation at the [01\{1\}] steps, a decrease in the indium concentration is observed in the layers \( 4^\circ \) misoriented towards (01\{1\}). This illustrates the fact that here the incorporation is not determined by the adsorption/desorption equilibrium for indium following from the BCF theory as is the case at the [01\{1\}] steps. It was discussed above that the group III growth species are directly incorporated upon arrival at this step, meaning that the BCF-flux theory is not applicable at these low misorientations. Furthermore, as was observed from the temperature series of the \( 2^\circ \) samples the arsenic desorption from this step is much easier compared to the step in the [01\{1\}] direction. Consequently, here the effect of preferential desorption of arsenic from the step edges is already observed at low misorientation angles. Only at high misorientations can the competition between the fluxes of indium and gallium, as derived from the BCF theory, become clear, as so much arsenic has desorbed that also here the adsorption/desorption equilibrium of indium at the step can establish itself, raising in this way the indium concentration for all temperatures.
The effect of step bunching, which is only expected at the lower temperatures [28], is reflected in Fig. 4 by the fact that at the low growth temperatures the differences between the indium concentrations at the various misorientations are relatively small, whereas at the higher growth temperatures, in the absence of step bunching, the effect of the misorientations becomes more pronounced.

One might argue whether these results possibly could be explained because of an anisotropy in the surface diffusion coefficients of group III species on a GaAs substrate. The surface diffusion coefficient of gallium in the [011] direction has been reported to be higher than that in the [011] direction [36,37]. However, since for indium the same relation can be expected, in the case that the incorporation is surface diffusion controlled the indium concentration observed in the layers misoriented towards (011) and (011) would be equal. This is in contrast with the results shown in Figs. 2 and 4. In general the experimental observations are in agreement with the tendency found for the incorporation of group II elements during MOVPE of III–V materials, which preferentially takes place at the steps in the [011] direction [7,9].

5. Summary and conclusions

The dependence upon the misorientation of the indium incorporation process during the growth of In$_x$Ga$_{1-x}$As by MOVPE on (100) vicinal GaAs substrates has been studied. The increase in indium incorporation as a function of temperature has been explained by the presence of indium adducts in the gas phase at low temperatures, which decompose at high temperatures. The observed temperature behaviour of the indium concentrations in the In$_x$Ga$_{1-x}$As layers as a function of the direction of the misorientation has been explained on the bases of the different incorporation processes at the steps, thereby taking into account the arsenic coverage of the steps and the roughening of the steps at high misorientations. The larger sensitivity to the temperature of the desorption of arsenic from the [011] steps leads to a lower increase in indium incorporation in layers misoriented towards (011), compared to that in layers misoriented towards (011).

Finally, the indium incorporation as a function of the misorientation angle is described by a competition between the gallium and indium fluxes towards the steps according to the BCF theory. This process is counteracted by the preferential desorption of arsenic from the step edges, which reduces the kink site density. In the [011] direction the BCF-flux mechanism is active at low misorientations, what results in an initial increase in the indium concentration in the layers with increasing misorientation. At higher misorientation the strong arsenic evaporation reduces the indium incorporation. In the [011] direction the arsenic evaporation is already active at low misorientations, whereas at larger misorientations the BCF fluxes become dominant.

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References