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Surface segregation in epitaxy of III–V compounds

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Abstract. Surface segregation in III–V compound heterostructures grown by Molecular Beam Epitaxy (MBE) and Metal-Organic Vapor Phase Epitaxy (MOVPE) is studied theoretically. The suggested model treats segregation as a transient process resulting in a delayed highly-volatile species incorporation into the crystal accompanied by its accumulation on the growth surface. Specific features of In segregation in InGaAs and InGaN are discussed with respect to control of the composition profile and efficiency of In incorporation into the crystal.

Introduction

Nowadays, principles of band gap engineering are widely employed for designing most advanced semiconductor devices. This requires to control heterostructure layer thicknesses, composition and doping profiles on the atomic scale. The growth techniques like MOVPE and MBE provide potential facilities for such a control. However, researchers are frequently face with natural physical limitations on abrupt interfaces, composition and dopant distributions. Among the limitation factors, surface segregation is one of the most important phenomena because of considerable effect on the composition profile in the grown III–V ternary compounds.

The earlier MBE studies revealed two principal experimental manifestations of segregation: (i) delayed incorporation of a highly-volatile species into the crystal at the epilayer growth onset, and (ii) penetration of the species into the cap layer after the species supply to the growth surface was terminated. A short review of the experimental observations is given in [1, 2]. The segregation depended significantly on temperature and, to a smaller extent, on the V/III ratio in the incident fluxes. Elastic strain in the epilayer due to lattice constant mismatch was found to be a crucial factor significantly enhancing segregation effects.

Recently we suggested a rate-equation model of surface segregation on ternary III–V compounds, providing excellent fitting of the literature data on MBE of InGaAs without any adjustable parameters [1, 2]. In this paper we apply the model to InGaN growth both by plasma enhanced MBE and ammonia MBE. The results are compared with those obtained for InGaAs. The segregation effect on In incorporation efficiency in MOVPE of InGaN is also discussed.

1. Control of composition profile in MBE of InGaAs

Detailed theoretical study of In segregation in MBE of InGaAs was reported in [1, 2]. Here we consider the ways to control the interface abruptness in a GaAs/InGaAs/GaAs heterostructure. The interface control utilizes the idea that In in the adsorption layer is nearly in equilibrium with the InGaAs crystal bulk, see, for instance, [2]. Indeed, Fig. 1 shows a nearly linear relationship between the In surface coverage (θ_{In}) and InGaAs composition

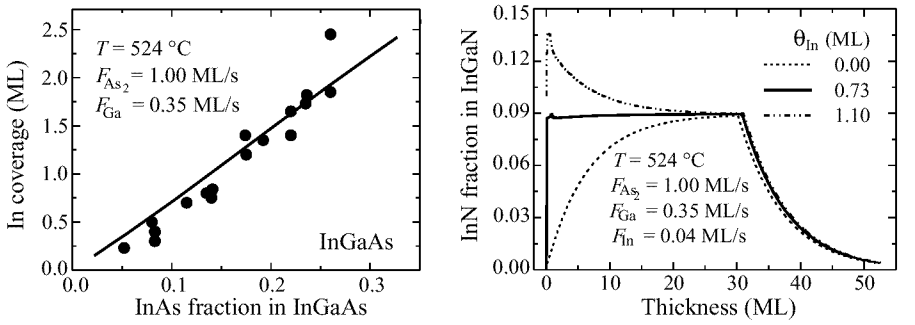

Fig. 1. In surface coverage against InGaAs composition measured in [3]; circles are experimental data, line is theoretical curve (left). InGaAs composition profile for the quantum wells grown with In pre-deposited on the growth surface (right).

(x) with a temperature-dependent slope, as measured in [3]. This enables one to introduce the segregation coefficient $\mathcal{S}(T) = \theta_{\text{In}}/x$, which is an evidence for the adsorption layer – crystal bulk near-equilibrium.

Normally, direct InGaAs/GaAs growth results in a delayed In incorporation into solid, see dashed line in Fig. 1 (right). Pre-deposition of In on the GaAs surface without As_4 flux produces In in the adsorption layer. If the In coverage is equal to an equilibrium value, no transient effect at the growth onset is observed [4, 5], see solid line in Fig. 1 (right). As a result, a perfectly abrupt front interface is formed. If the In coverage exceeds the equilibrium value, then inverse profiling of solid composition can be obtained, see dash-dotted line in Fig. 1 (right).

The back GaAs/InGaAs interface can be controlled by evaporation of In from the growth surface after the InGaAs epilayer has been grown. It must be done with the As_4 flux switched off to prevent growth surface from InAs island formation. Duration of this stage depends on the temperature-dependent rate of In desorption.

2. Indium segregation in MBE of InGaN

Specific feature of InGaN MBE is a relatively high growth temperature. This decreases the segregation length characterizing the transition layer thickness at the InGaN/GaN interface, see Fig. 2. However, the strain effect on surface segregation is rather strong due to a considerable lattice mismatch between the InGaN and underlying GaN. Therefore, In

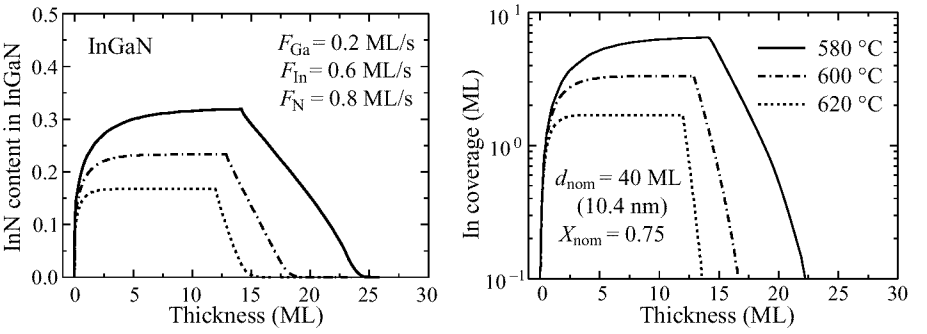

Fig. 2. InGaN composition profile (left) and In coverage (right) as a function of quantum well thickness computed for different temperatures of plasma-enhanced MBE.

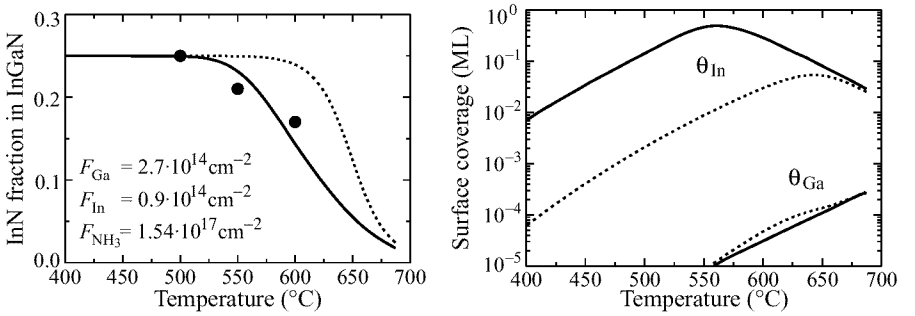

Fig. 3. Temperature dependence of InGaN composition (left) and In surface coverage (right) computed for ammonia MBE reported in [6].

accumulation on the growth surface is extremely high for InGaN growth (Fig. 2).

The effect of In accumulation is especially important for ammonia gas-source MBE. Figure 3 shows temperature dependence of the composition profile and In surface coverage calculated for the strained and relaxed InGaN/GaN heterostructures. The computations were carried out for the growth conditions reported in [6]. Comparison with the data [6] shows the theoretical predictions for the strained epilayers to be in good agreement with the observations. Up to 0.6 ML In is predicted to accumulate on the growth surface under the conditions used in [6].

It was found earlier that Ga accumulation on GaN(0001) surface resulted in NH_3 adsorption site blocking [7] and consequent growth rate reduction. If In behaves similarly to Ga, we can expect the termination of InGaN growth when In coverage exceeds unity. This occurs in a temperature gap with the width primarily dependent on the incident In flux. Normally, high growth temperatures are beneficial for InGaN improving their optical characteristics. However, In incorporation into the crystal is limited in the high-temperature range due to intensive In desorption. Computations show the InN content to be limited by 15–20% in that case.

3. Surface segregation in MOVPE of InGaN

Accumulation of surface In is also responsible for limited In incorporation efficiency in MOVPE of InGaN. Figure 4 displays the In coverage as a function of InGaN composition computed for MOVPE in a vertical rotating-disk reactor using N_2 as a carrier gas. One can see that the composition of the strained InGaN is limited by 30%. Use of H_2 as a carrier

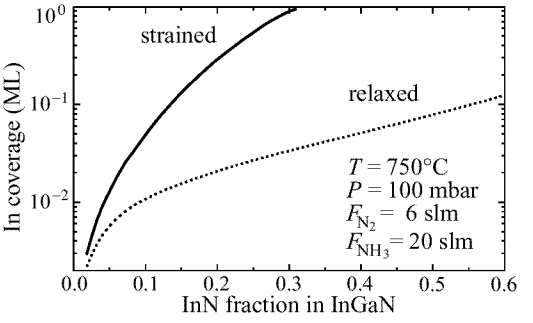

Fig. 4. In coverage as a function of InGaN composition computed for MOVPE in a vertical rotating-disk reactor.

gas results in enhancement of In segregation, especially at low temperatures. This is due to high reactivity of hydrogen on the nitride surface. In the case of the relaxed InGaN epilayers, no segregation effect on In incorporation is found.

Conclusions

In this paper various aspects of surface segregation have been discussed with the focus on InGaN growth by MBE and MOVPE. The segregation effects are found to be much more pronounced for InGaN/GaN structures than for InGaAs grown on GaAs. Modification of a heterostructure composition profile due to segregation is important for band-gap engineering of advanced semiconductor devices. The methods of interface control include pre-deposition of equilibrium amount of highly-volatile species on the growth surface before growth and evaporation of the species excess after the epilayer is formed.

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