Investigation of diffusion in AlAs/GaAs distributed Bragg reflectors using HAADF STEM imaging

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Abstract. In this contribution we have studied the diffusion of Al in AlAs/GaAs distributed Bragg-reflectors using the high angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) intensity. The measured intensity is normalized to the intensity of the incoming electron beam using a detector scan. The normalized intensity can be directly compared with a set of frozen lattice simulations yielding specimen thickness in regions with known composition or concentration in regions with known thickness. The thickness was evaluated both from GaAs and AlAs regions yielding that the specimen was about 15 nm thinner in AlAs regions due to oxidation. For the concentration evaluation the thickness was derived from GaAs regions and concentrations up to 1.2 were found due to the overestimated thickness. Concentration profiles were scaled down to 1.0 and fitted to the solution of Fick’s laws.

1. Introduction

Optoelectronic devices containing quantum dot structures have gained a lot of interest in recent years due to their improved and novel properties. A rather simple way to produce quantum dots is by exploiting the Stranski-Krastanow growth mode. However, such ensembles of quantum dots exhibit rather broad size and composition distributions. Using a rapid thermal annealing (RTA) treatment the optical properties, especially the energetic position of the ensemble of dots or of an isolated dot can be influenced [1, 2]. For fabrication of vertical-cavity surface-emitting lasers (VCSELs) the quantum dots have to be grown embedded between two distributed Bragg reflectors (DBRs). A subsequent RTA treatment influences the properties of the quantum dots, but may also influence the reflectivity of the DBRs due to diffusion. Most of the previous investigations have focused on the effect of RTA on the active layers (quantum dots or also quantum wells, see e.g. [3, 4, 5]).

In this contribution we investigated the diffusion of Al in AlAs/GaAs DBRs due to a thermal treatment at temperatures as high as 1100 ºC for 180 s by a quantitative analysis of the high
angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) intensity. Derived Al composition profiles were fitted by solving Fick’s laws.

2. Growth, annealing treatment and preparation of specimen
The 24-pair and 25-pair AlAs/GaAs DBRs were grown by solid source molecular beam epitaxy (SS-MBE) using valved phosphorous and arsenic cracking sources. The sample manipulator temperature was kept close to the typical GaAs growth setting yielding sample temperatures in the range between 580 and 600 °C. The growth rates for both AlAs and GaAs layers were about 1.0 µm/h. The 25-pair AlAs/GaAs DBR was treated by RTA at 1100 °C for 180 s. TEM specimens were prepared for a <110> zone axis orientation by cutting lamellas out of the wafer using a FEI dual beam focused ion beam (FIB) NOVA200. The lamellas were lifted out using a Kleindieck manipulator needle and fixed to a Cu ring. STEM HAADF images of the structures are depicted in Fig. 1. TEM specimens were directly transported from the FIB to the TEM or were stored in vacuum in order to prevent oxidation of the AlAs layers as much as possible. TEM investigations were carried out in a FEI Titan 80/300ST equipped with a corrector for spherical aberration of the objective lens, an EDAX EDX detector, a Fishione HAADF detector and a Gatan imaging filter (GIF).

3. Method for quantitative evaluation of HAADF intensity
Here we only briefly describe the basics of the quantitative evaluation technique, which is explained in more detail in Ref. [6]. Prior to the acquisition of the HAADF images an image of the detector is recorded by scanning the electron beam in image mode over the detector. In this way the intensity in the HAADF STEM images can be normalized to the incoming electron beam intensity and therefore can directly be compared with simulations.

A series of HAADF-STEM images were computed using the STEMsim software package [7] for concentrations from 0 to 100% and thicknesses of smaller than 350 nm. To take into account thermal diffuse scattering we used the “frozen lattice approximation”, in which it is assumed that all atoms vibrate independently. For each scan point in the HAADF images the intensity was averaged over 10 different “frozen lattice” configurations. Static atomic displacements [8] were not taken into account, since the ionic radii of Al and Ga are nearly the same (lattice parameter \( a_{\text{GaAs}}=0.5653 \) nm and \( a_{\text{AlAs}}=0.5661 \) nm). The HAADF STEM intensity as a function of thickness and composition is depicted in Fig. 2. It exhibits strong thickness dependency and a linear concentration dependency.

The specimen thickness can be determined from reference regions with known composition (e.g. a region in the GaAs substrate) by directly comparing experimental and computed HAADF-STEM intensities, both normalized to the incoming beam intensity. In regions with
unknown composition, the specimen thickness is interpolated. Then the intensity ratio is determined by normalizing to the reference regions. Finally, the composition is deduced by a comparison with the theoretical data.

4. Results and discussion

Fig. 1 shows overview images of the as-grown and the annealed samples. From a first visual inspection it becomes clear that the interfaces in the as-grown specimen are sharper than interfaces in the annealed one indicating that Al is diffusing out of the AlAs layers. In order to ensure that in the middle of the GaAs the Al concentration (and in the middle of AlAs the Ga concentration) is negligible we carried out energy dispersive X-ray spectroscopy (EDX) on several points lying on lines parallel to the GaAs (AlAs) layers. We found that the Ga (Al) concentration in the middle of the AlAs (GaAs) layer is below 1%. For further investigations we assumed that the Al concentration in the centre of the GaAs layer (Ga concentration in the AlAs layer) is zero.

In a further step we acquired HAADF STEM images and performed linescans in growth direction. We selected the GaAs substrate and regions in the centre of the GaAs layers for the determination of the specimen thickness (see sec. 3). For the lamella of the annealed specimen shown in Fig. 1 we found a specimen thickness ranging from 70 nm to about 260 nm. The specimen thickness was interpolated within the AlAs layers using a 3rd order polynomial. We also determined the specimen thickness from the centre of the AlAs layers yielding a range from 60 nm to 230 nm. The found thickness dependencies are depicted in Fig. 3. We found that the specimen is about $15\pm 4$ nm thinner in AlAs than in GaAs regions. This result was confirmed by thickness maps derived from energy filtered images. This also becomes obvious in Fig. 1, where at the edge of the lamella the GaAs layers reach further into the vacuum than the AlAs layers. Under the assumption of the specimen thickness derived from the GaAs regions we found Al concentrations up to 120%. For this purpose the reference data set shown in Fig. 2 was extrapolated linearly up to a concentration of 150%. The determination of these unphysical Al concentrations can be explained by the overestimation of specimen thickness in the AlAs regions. In the EDX experiments we found that the Ga-concentration with the AlAs layers was negligible and therefore, we rescaled the measured concentration profiles down to an average Al concentration of 100% in the centre of the AlAs layers.

Rescaled concentration profiles were fitted by solving Fick’s laws for square concentration profiles (see Fig. 4). For the as grown specimen only small deviations from a rectangular concentration profile are observed, which could be attributed to electron beam broadening or

Figure 2. Simulated intensity on the HAADF detector as function of Al concentration and specimen thickness.
segregation of Al during growth of the sample. Rosenauer et al. [9] found a low segregation ($R=0.5$) efficiency in this material, so that the main contribution should come from electron beam broadening. To estimate the broadening the profile was fitted with a diffusion profile yielding a diffusion length of about 1 nm. In the annealed specimen we found a diffusion length of about 10 nm, showing that the beam broadening should only influence the determination of the diffusion length in a minor way. The diffusion coefficients were found to be $1.93 \times 10^{-15}$ cm$^2$/s, respectively with a standard deviation of $0.25 \times 10^{-15}$ cm$^2$/s. Cai et al. [10] measured the diffusion coefficient at 900°C and found $2 \times 10^{-17}$ cm$^2$/s, which is about two orders of magnitude smaller than our value. However, assuming the activation energy of $3.5 \pm 0.1$ eV from Bracht et al. [11] one would expect a diffusion coefficient of $2.46 \times 10^{-15}$ cm$^2$/s at a temperature of 1100 °C, which is in agreement with our value.

![Figure 3](image3.png)  
**Figure 3.** Comparison of specimen thickness as a function of position on the TEM lamella. The specimen thickness was once evaluated from GaAs and from AlAs regions.

![Figure 4](image4.png)  
**Figure 4.** Comparison of concentration profiles measured in the as-grown and the annealed sample. Both profiles were fitted with a diffusion model.

5. Summary
In summary, we quantitatively evaluated Al concentrations from HAADF-STEM images of AlAs/GaAs DBR structures. The evaluation of as-grown and annealed specimens yielded a diffusion coefficient of $1.93 \times 10^{-15}$ cm$^2$/s for Al at 1100 °C.

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