Microstructure evolution in MOVPE–grown AlN/GaN superlattices with different periods

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Introduction

The (Al,Ga)N/GaN system has already shown excellent results for electrical device applications because of the large conduction band offset enabling the realisation of micro– and millimeter–wave power devices. In recent years mostly AlGaN/GaN heterostructures were explored in the compositional range x_{Al} <0.6, but only limited information on AlN/GaN quantum wells (QW) and superlattices (SL) grown with metalorganic vapour phase epitaxy (MOVPE) has been published because of several factors leading to difficulties in the crystal growth. The interface should be laterally smooth as well as the composition transition should be vertically abrupt because of the large impact of the interface roughness on the electron mobility of the two–dimensional electron gas (2DEG) forming at the heterojunction.

Growth related issues of prime importance are the difficulties to achieve the step flow growth mode of AlN similar to the GaN growth mode because of the reduced surface mobility of the AlN adsorbed species. As a result a strong tendency towards defect formation and transition into island growth mode and/or cracks formation is observed. The increased AlN roughness at the interface prevents the growth of constant period and regular thickness SLs. If the AlN and GaN layers within the SL are kept inferior to their critical thickness a balance of the 2D/3D growth could be achieved. Additionally, structural defects can be formed due to the alternating tensile/compressive strain within each period due to the lattice mismatch between AlN and GaN (~ 2.5 % misfit for the bulk lattice parameters). Also, defect formation might not exclusively originate from the strain, but threading dislocations that are formed during growth of the GaN base film due to the lattice mismatch to the sapphire substrate, could propagate through the SL layer to the sample surface.

In this work we report on microstructural studies of AlN/GaN SLs grown by MOVPE with different periods. The strain relaxation mechanism during growth in the different samples is addressed relative to the growth mode and the critical thickness for elastic/plastic relaxation occurrence.

Experimental

Crack–free SLs are grown by MOVPE and the details are published elsewhere [1,2]. A GaN base layer with a thickness of 2 µm underlying the SLs is grown on a (0001) sapphire substrate using a low–temperature AlN buffer. A series of samples (GaN)_{m}(AlN)_{n} (Table 1) is grown consisting of 10 period SLs with different well thickness, keeping the barrier–well thickness ratio constant n/m~3 and the same average AlN mole fraction of 0.24. The microstructural development in the SLs is studied by transmission electron microscopy (TEM) in low–magnification and high–resolution mode (HRTEM) using a Philips CM20 UT transmission electron microscope operated at 200 kV with a LaB_{6} filament. The samples were prepared in cross–section by a conventional procedure including mechanical polishing followed by low–angle and low–energy ion milling to electron transparency. The samples were viewed along the [11–20] zone axis and bright field images were taken.

E. Valcheva et al
Table 1. Parameters of the different layers in the studied samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>GaN base (µm)</th>
<th>GaN/AlN (Å)</th>
<th>number of periods</th>
<th>average AlN mole fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>SLa</td>
<td>2</td>
<td>7.3/22.8</td>
<td>10</td>
<td>0.24</td>
</tr>
<tr>
<td>SLb</td>
<td>2</td>
<td>28.6/88.6</td>
<td>10</td>
<td>0.24</td>
</tr>
</tbody>
</table>

Fig. 1. (a) Cross-sectional TEM image of sample SLb. (b) Magnified image of the SL region marked in (a) by white square showing cusp defects at the interfaces due to the growth of flat platelets of AlN. (c) Schematic picture of the modes of growth of AlN on GaN in tension (black arrows depict the cusps) and of GaN on AlN in compression.

Results

HRXRD (0002) 2θ/ω scans exhibit (not shown here) higher-order satellite peaks up to the third order and fringes between them, suggesting that the SLs are of good layer periodicity. The spacing of the satellite peaks indicates the periods of the superlattices to be consistent with the thicknesses determined by TEM.

The TEM imaging reveals a constant growth period and abrupt interfaces at the monolayer scale. However, defects are observed in some places causing different degree of disturbance of the regularity of the SL. Threading dislocations originating from the LT AlN buffer layer and propagating through the GaN base layer are seen to penetrate through the SLs (Fig. 1a). It seems that the GaN/AlN SL does not stop the propagation of most of them and could not be used to filter out dislocations.

E. Valcheva et al
Despite of the constant period the SLs grown with the large period (sample SLb) in particular areas (Fig. 1b) show apparent interdiffusion. The HRTEM images give evidence that the GaN and AlN sublayers are of irregular thickness, and cusps disturbing the smoothness of the interfaces are seen (Fig. 2). Their appearance might be due to the processes of strain relaxation by the growth of flat AlN platelets [2] on a thin wetting layer, as pointed out by white arrows in Fig. 1b. The reason could be the tendency of 2D growth of AlN on GaN being in tension as it is depicted schematically in Fig. 1c, while the GaN layers in compression grow 3D followed by island coalescence. The spacing left between the AlN platelets is overgrown with GaN thus forming the cusps. These defects are repeated in the upper layers and aligned along the (0001) direction.

Our observation is consistent with the growth of AlN layers with thickness beyond the critical thickness for pseudomorphic growth of strained layers, which is reported to be in the range 37–65 Å for the (AlN)$_m$/GaN$_n$ system with arbitrary $m$ and $n$ [3].

**Fig. 2.** (a) HREM image of the AlN/GaN interfaces and (b) magnified image of a cusp originating from the growth of AlN in tension.

**Fig. 3.** (a) HREM image of sample SLa showing (b) the 10 period SL with abrupt interfaces and (c) dark contrast areas looking like GaN quantum dots formed in the AlN matrix.

E. Valcheva et al
The SL with the small period (Fig. 3a, sample SLa) is coherently grown and strained, owing to the assumption that in SL structures with sufficiently thin AlN and/or GaN layers, the lattice constant mismatch is accommodated by internal strains rather than by the formation of misfit dislocations. Regions where the interfaces are abrupt on a monolayer scale are observed (Fig. 3b) as well as regions of fluctuating contrast that can be attributed to the presence of a dot–like structure (Fig. 3c). Vertical correlation of the dark GaN islands being typically of dimensions 3 nm base and 1.5 nm height, embedded in the AlN matrix, is also present. Accordingly, formation of GaN quantum dots (QD) in an AlN matrix appears because of the driving force of the strain field when the GaN layer thickness is inferior to the critical thickness for plastic relaxation. However, the optical emission properties need to be investigated before a conclusion of the appearance of QD structure could be made.

Because of the difference in the thicknesses of the wells and barriers of the strained short period AlN/GaN SLs here investigated, different modes of growth are observed relative to different strain relaxation mechanisms operative when the layer thicknesses are beyond the upper and lower bounds for elastic/plastic relaxation.

In conclusion, the structural properties of strained short period AlN/GaN SLs are investigated by means of electron microscopy, and the microstructural evolution with respect to the different relaxation mechanisms is investigated.


E. Valcheva et al