3.1 Publishable summary

3.1.1 Project description and objectives

With an increasing amount and percentage (currently 20%) of electrical energy being consumed by lighting there is a need to introduce energy efficient technologies to the consumer. Light emitting diodes (LEDs) based on indium aluminium gallium nitride (InAlGaN) materials can meet the requirements. Total energy savings can be a factor of two over fluorescent bulbs making it imperative to introduce this technology. The factors that need to be overcome in order to accelerate the widespread uptake of LEDs are in providing a high quality light with a colour balance from the luminaire which is attractive to the consumer while delivering the light at an acceptable cost, and by further improving the conversion efficiency.

While great inroads have been made, the LEDs currently on the market suffer from a reduction in their efficiency at higher currents, called droop, thus limiting the total amount of power from a single chip. Furthermore, the colour can change as a function of current. In addition, the white light is obtained using a down-converting phosphor leading to an energy loss whereas a combination of colours including direct yellow emission would be desirable for higher efficiency and colour control. A solution to these issues can be obtained by basing the LEDs on semi-polar planes where in addition to reduction of the polarization fields, high amounts of indium can be incorporated within the crystal. However, the necessary semi-polar substrates are difficult to engineer resulting in them being costly and having limited wafer size.

The ALIGHT project has assembled a consortium to investigate new approaches to large area, low defect density, semi-polar (11-22) substrate technologies based on structured r-plane sapphire and structured (113) silicon substrates. This is requiring fundamental modelling of the surface chemistry. Extensive work is taking place on growth processes along with detailed material characterisation. Using these template substrates highly efficient blue and yellow light emitters are being designed and the epitaxial processes developed. This work is supported by a thorough fundamental investigation of the composition dependent properties of InAlGaN materials.

The major material challenges are in the details of the patterning of the wafer structures, achieving growth process for the generation and coalescence of semi-polar planes while obtain low stacking fault density ($<10^5$/cm) and low dislocation density (10^6/cm^2). To address this, the influence of substrate fine orientation and growth parameters are being checked and their influence assessed by detailed X-ray measurements, luminescence and atomic scale imaging. Scaling of the substrates is being addressed by both Metal-Organic Vapour Phase Epitaxy (MOVPE) and Hydride VPE techniques. Quantum wells and dots are being utilised as the active light emitting material and the spatial distribution of emission is assessed by detailed microscopy. P-type doping of the structure and control of impurity incorporation are of critical importance. Finally fully packaged LEDs will be demonstrated.

3.1.2 Description of work and key results

Modelling of AlInN materials

$Al_{1-x}In_xN$ has a direct band gap that spans 6.2 to 0.7 eV making it an ideal candidate for visible LEDs. An accurate knowledge of the variation of the band gap with varying InN content x is required for designing devices. For most III-V alloy materials this is done with a parabolic approximation and a constant bowing parameter. However the AlInN literature values for this constant diverge widely with reported values from 2.5 eV, from measurements on high-x samples, up to 10 eV based on low x values. Recently, based on density functional theory (DFT) results, the

physical mechanisms underlying a breakdown in the assumption of a constant bowing parameter have been clarified. We have shown that cation-related localized states in the conduction band (CB) and the valence band (VB) lead to the breakdown of the virtual crystal approximation. These results support the assumption of a composition-*dependent* bowing parameter. It is important to note that when designing polarization-matched GaN quantum wells (QWs), using AlInN barriers, the evolution of the CB edge (CBE) and VB edge energies with InN content x is important, since this determines confinement energies for carriers.

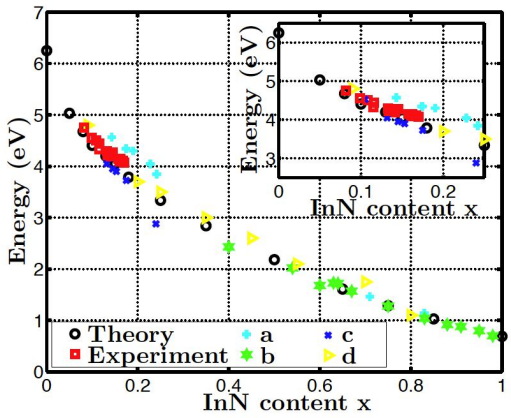

Fig.1. Comparison between the experimental and theoretical band gap of $Al_{1-x}In_xN$ as a function of x (from Appl. Phys. Express 6, 121001 2013).

Modelling of InGaN surfaces and alloy thermodynamics

The first step towards achieving a full control over the growth of semi-polar planes is to have a deep understanding of the relevant atomistic processes. The growth and the properties of AlGaInN QWs are an interplay between growth conditions, chemistry, and strain. In the first half period of ALIGHT project we have investigated the issues of bulk InGaN thermodynamics and the properties (energetics and electronic structure) of semi-polar as well as polar III-nitride surfaces. These calculations addressed the issues of spinodal decomposition, energetics of In incorporation, and surface morphologies during growth. We have addressed InGaN bulk thermodynamics by utilizing special quasi random structures and we have shown that spinodal decompositions is suppressed for coherent (i.e. pseudomorphic) growth on GaN. Our surface calculations revealed that semi-polar planes follow the general trends of III-nitride surfaces and they reconstruct following the electron counting rule under N rich conditions, while metal rich growth stabilizes metallic adlayers. The latter is advantageous for achieving smooth surface morphologies. However, for MOVPE growth In may incorporate at the surface only under extreme In rich and H poor conditions (i.e. low values of H partial pressure). Furthermore, we have shown that both chemistry and strain effects are equally important in In incorporation and provide a strong driving force for Indium surface segregation.

Development of semipolar GaN templates

Within ALIGHT two approaches are being developed to the creation of high quality large area semipolar GaN templates. The first is based on r-plane sapphire substrates which are patterned with trenches exposing a c-plane like side facet. Epitaxial growth is initiated in the inclined c-direction followed by coalescence. Both a low defect density and a smooth surface are required. We have conducted multiple structured experiments to optimise the substrate preparation and the growth conditions in order to improve the crystal quality by minimising the defect density and improving the surface smoothness. In particular, the integration of a SiN nanomask layer (Fig. 3 right) helped to block defects from extending to the semi-polar layer surface. This was proven by Synchrotron based X-ray diffraction, which confirmed a dramatic reduction of the basal plane stacking fault density down to $4x10^3$/cm.

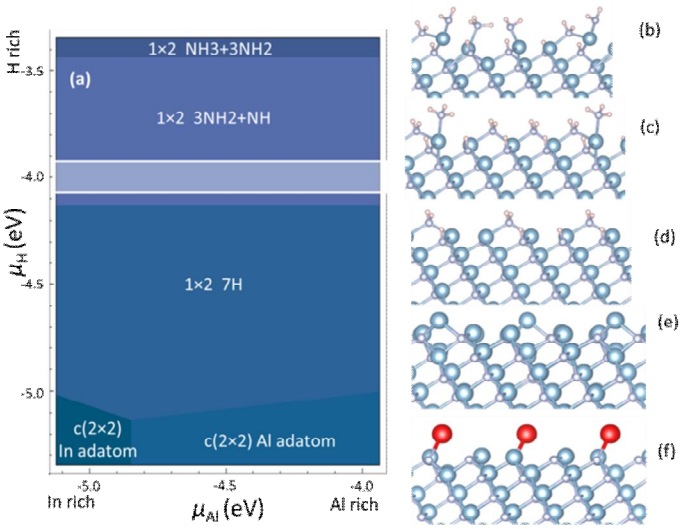

Fig. 2 (a) Phase diagram for the AlInN (11-22) surface in the presence of H, as a function of Al and H chemical potentials. Nitrogen rich conditions have been assumed for the value of the Nitrogen chemical potential. Only the lowest energy structure is indicated for various values of the Al and H chemical potentials. The shaded area indicates the MOCVD relevant conditions for the hydrogen chemical potential. (b), (c), (d), (e), and (f) Schematic representation of the 1×2 NH_2+3NH_2, 1×2 3NH_2+NH_2, 1×2 7H, c(1×2) Al adatom, and c(1×2) In adatom reconstructions respectively.

Such templates have been forwarded to the project partners for the subsequent deposition of AlGaInN quantum wells (see below). In order to obtain substantially thicker GaN layers, first hydride vapour phase epitaxy (HVPE) experiments have been carried out with a growth rate of 140 µm/h. 40 µm thick layers exhibit very promising characteristics, such as greatly improved surface flatness.

The second approach is a radical but potentially very low cost approach using silicon. Firstly, substrates with a particular offcut (311) have been prepared by structuring thereby exposing suitable crystal planes that allow subsequent initiation of GaN growth. This has been successfully implemented on a new reactor. Conditions necessary for defect control and layer coalescence have been investigated and a first demonstration of growth on a full 100mm diameter Si wafer has been made.

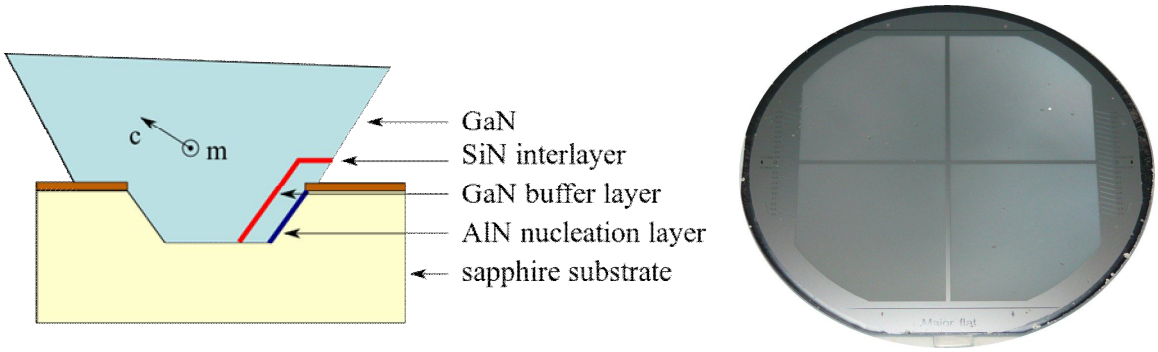

Fig.3. Schematic of the growth process for a single stripe of GaN material on a patterned sapphire substrate and, (right) photograph of a 100mm diameter S wafer with a layer of semipolar GaN grown.

Understanding the origin of surface hillocks in semi-polar templates by TEM

As mentioned above, HVPE is used to overgrow GaN on top of a MOVPE grown GaN layer initially prepared on pre-structured sapphire. However, a high density of faceted hillocks was observed by SEM in some regions on the sample surfaces (Figure 4a), which have been analysed in detail by TEM after focused ion beam (FIB) preparation. Figure 4b is a diffraction contrast TEM image from a FIB sample prepared from a hillock. Two boundary structures were observed (marked by black arrows). Convergent beam electron diffraction patterns (CBED) (Figure 4c) confirm that

the crystal structure inside the two boundaries has an inverse crystallographic polarity. The hillock is formed because the growth rate of the inversion domain is higher than that of the normal material. This direct correlation between a surface hillock and the formation of an inversion domain underneath is a vital piece of information for the grower to optimize the growth conditions aiming for a smooth template surface.

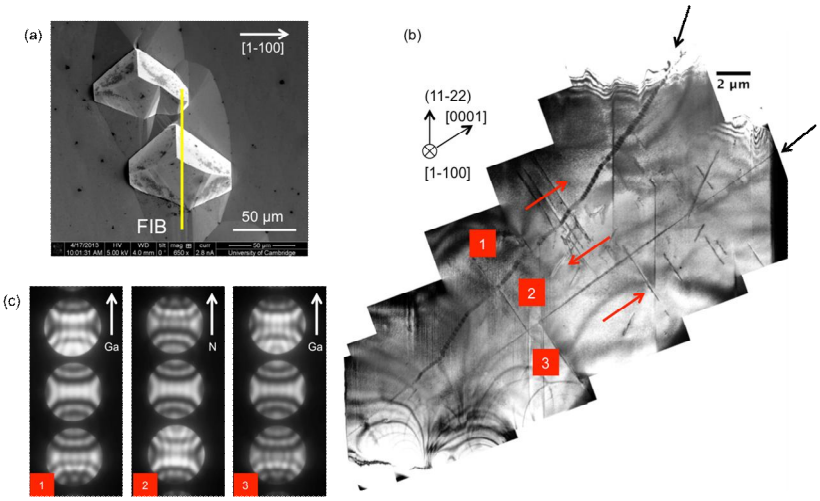

Fig. 4. (a) Faceted hillocks observed by SEM. (b) Two domain boundaries were revealed on a cross sectional sample prepared precisely through the core of a hillock, with an inversion in polarity confirmed by (c) CBED patterns acquired at regions 1, 2 and 3

Growth of semipolar quantum well on templates

In order to control the emission wavelength the indium content of the quantum wells needs to be accurately known. InGaN layers (~30 nm thick) were grown on semipolar (11-22) GaN templates with different growth temperatures. The indium content was estimated to be from 2% to 29%. Based on the growth and indium composition of InGaN layers, semipolar (11-22) InGaN single-quantum-well (SQW) structures were optimized with different thicknesses in order to achieve the highest luminescence efficiency in the blue-green-yellow spectral ranges. Figure 5 shows intense photoluminescence spectra measured at room temperature on three different InGaN SQW samples grown with different V/III ratios. Based on the standard method of comparing low and room temperature luminescence the highest internal quantum efficiency of the blue SQWs was estimated to be about 40%. Lastly, LEDs based on the SQW was successfully produced.

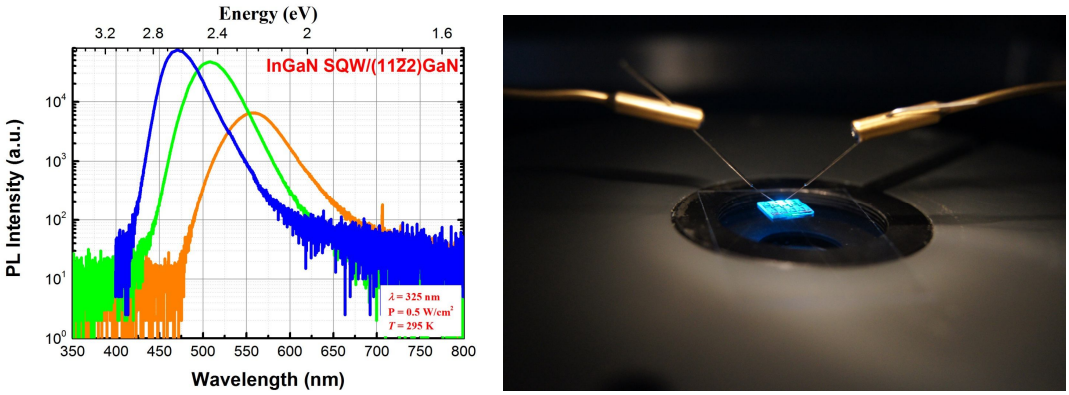

Fig.5. Room-temperature photoluminescence spectra of InGaN single-quantum-well emitting in the blue, green, and yellow spectrum range and light emission from single quantum well LED grown on ALIGHT semipolar template.

3.1.3 Expected final results and impact

We are making excellent progress towards our final goal of low defect density, large area, low-cost, semi-polar GaN templates and the realisation of high efficiency LEDs thereon. The technical work is training the personnel (students to professors) with enhanced expertise in this important industrial technology. The scientific reputation of collaborative European research is being enhanced. The work will be the basis of new materials and processes that can be taken on by European industry. This is an area where Europe is already strong with world-leading equipment manufacturers, substrate manufacturers and LED manufactures. The resulting products will transform lighting from that of fluorescent lamps to that of solid-state lighting resulting in higher quality, environmentally friendly mercury free, which will consume less energy.

http://www.alight-project.eu/