

Real–time control of quantum dot laser growth by reflectance anisotropy spectroscopy

U. W. Pohl¹⁾, I. Kaiander¹⁾, C. Kaspari¹⁾, S. Weeke¹⁾, R. L. Sellin¹⁾, J.–T. Zettler²⁾, D. Bimberg¹⁾, W. Richter¹⁾

1) Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany, 2) LayTec GmbH, Helmholtzstr. 13–14, 10623 Berlin, Germany

Introduction

The high complexity of advanced devices grown by metalorganic vapor phase epitaxy has recently stimulated an increasing demand for online diagnostic tools. In the gaseous ambient only optical techniques like reflectance anisotropy spectroscopy (RAS) provide the potential to monitor the actual temperature, growth rate, doping concentration and composition at the growing surface [1,2]. A particular challenge is the investigation of growth dynamics occurring on the subsecond time scale of seconds like the self–organized formation of quantum dots in the Stranski–Krastanow growth mode. Besides basic scientific interest such quantum dots are presently intensely studied for applications, e.g. due to a superior device performance of lasers [3,4]. To analyze the growth process of In(Ga)As quantum dots we developed a fast RAS system to simultaneously monitor the spectral response at different photon energies and report on first results. In addition, RAS spectra recorded during epitaxy of complete edge and surface emitting devices with active In(Ga)As quantum dot stacks are presented for the first time.

Experimental procedure

In(Ga)As/GaAs quantum dot samples were grown in two different MOVPE reactors equipped with low–strain quartz windows, using the alternative nonhydride precursors TBAs, TMGa, TMAI, TMIn for layer deposition and DETe, CBr₄ for doping. Basic studies on quantum dot formation were performed using RAS on non–rotating samples in an Aixtron–200 prototype reactor. To achieve a temporal resolution superior to scanning monochromator–based RAS systems with a recording time in the order of a minute per spectrum a new multichannel RAS spectrometer was developed. It employs a spectrograph with an eight–channel diode array instead of a monochromator with just one detector. The real part of RAS signal $\frac{\Delta r}{r} = 2 \frac{r_{[-110]} - r_{[110]}}{r_{[-110]} + r_{[110]}}$ is

processed by eight independent lock–in amplifiers and fed to a multichannel A/D–converter card. This gives the possibility to simultaneously record eight RAS transients at different photon energies between 1.5 and 5.3 eV with a temporal resolution of 100 ms. The FWHM of each diode element is 30 nm. A comparison of the response of the multichannel spectrometer with a spectrum recorded using a scanning RAS system is given in Fig. 1, proving the applicability of the fast RAS system. The spectral resolution is strongly reduced but still sufficient, the temporal resolution is improved by two orders of magnitude. Application studies, namely components of laser structures and complete lasers, were grown in an AIX200/4 reactor. Parameters for the growth of device components like claddings and Bragg mirrors, and the process of stacking quantum dot layers were monitored in–situ using a Laytec EpiRAS sensor. The response of this sensor yields the absolute value of the RAS signal.

Real–time response on quantum dot formation

First spectral real–time results obtained from InAs/GaAs quantum dot formation show a detailed complex response within the first few seconds after InAs deposition. Fig. 2 shows RAS transients simultaneously detected at three different photon energies for InAs island layer growth (2D wetting layer) and InAs QD growth (3D). The transformation of the 2–dimensional wetting layer into 3–dimensional quantum dots was previously identified by optical studies including single energy RAS transients [5]. Fig. 2 shows that the transitions change significantly with photon energy. Comparing QD growth to island layer growth, the minimum of the 1.5 eV transient is twice as deep and more pronounced in the case of QD growth. In the 2.4 eV and 4.1 eV transients an additional maximum appears at $t \sim 5$ sec. These measurements agree well with earlier work in the range of the previously studied energy, cf. Fig. 3.

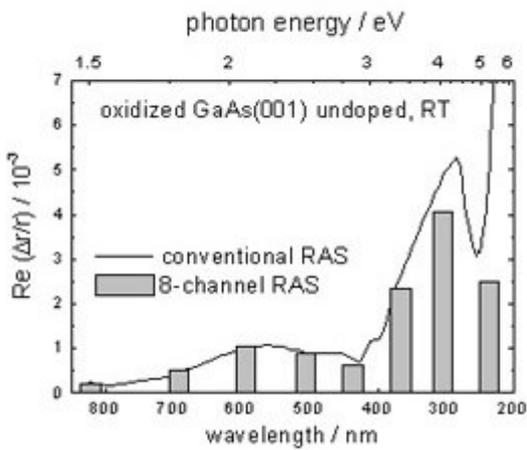


Fig. 1. RAS response of an oxidized GaAs (001) surface redorded with a conventional setup and the fast 8-channel system. The width of the bars corresponds to the spectral FWHM of the array elements.

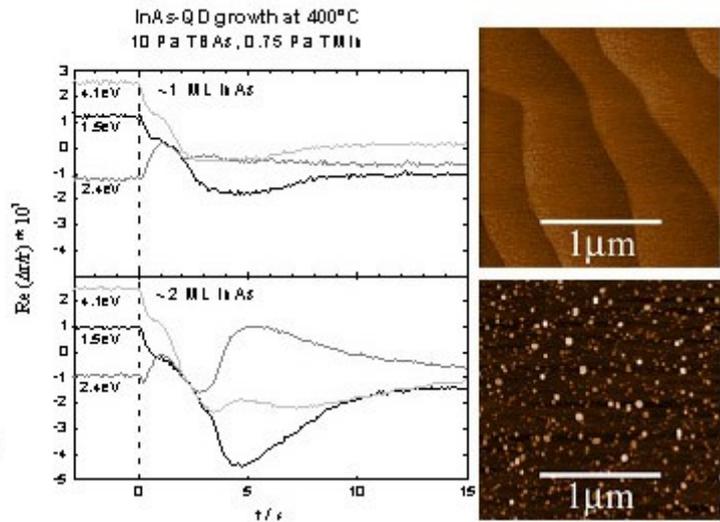


Fig. 2. RAS transients recorded with the multichannel setup at three different photon energies and the corresponding AFM images taken after growth. TMIIn was fed to the reactor at $t = 0$. The upper graph corresponds to island layer growth (wetting layer, ~ 1 ML InAs), the lower one to QD growth (~ 2 ML InAs).

In-situ monitoring of quantum dot laser growth

The core structure of an InGaAs quantum dot laser generally consists of a stack of In(Ga)As quantum dot layers which are separated by thin GaAs spacer layers. As a first step of in-situ controlled laser growth, different types of such dots, namely InAs/GaAs dots and InGaAs dots capped by either GaAs or InGaAs quantum wells for controlled emission wavelength tuning, were studied by RAS in the commercial setup. Fig. 4 shows transients of the RAS signal recorded at a single fixed photon energy of 2.6 eV. The data show a characteristic response on the successive process steps like In(Ga)As deposition, growth interruption to allow quantum dot formation, spacer deposition and flattening to obtain stacks with high structural and optical quality. The value of the RAS signal at the beginning of the transient refers to an As-stabilized GaAs surface at 500°C, the slight decrease prior to In(Ga)As deposition originates from a reduction of the As flux to the value required for the deposition. During In(Ga)As deposition the RAS signal drops to negative values, as indicated by a sharp dip in the plot of the absolute RAS signal at $t = 0$. The subsequently applied growth interruption is performed without precursor supply. During this period the RAS signal attains a constant value defined by the Ga composition. The nominal thickness of the In(0.5)Ga(0.5)As layer is higher than that of the InAs layer, the respective transient is thus shifted to later times. For the In(0.5)Ga(0.5)As quantum dots which were overgrown with a quantum well a longer growth interruption was applied. Growth is restarted at the end of the interruption (GRI) by deposition of the GaAs cap, spacer layer or InGaAs QW at the same low temperature used for quantum dot deposition. The RAS signal changes sign to positive values known for GaAs surfaces (dip to zero in Fig. 3). After growth of about 5 nm thick GaAs the deposition temperature is ramped up to 600°C in order to improve the quality of the GaAs matrix material surrounding the quantum dots. This ramping and a contingently applied second growth interruption at increased temperature were found to significantly improve the optical quality of the In(Ga)As/GaAs quantum dots and to form flat spacer surfaces for the deposition of a subsequent dot layer [6]. During the second GRI the As flux is ramped up for a subsequent growth of GaAs at 600°C with increased growth rate. In a laser structure the active quantum dot stack is cladded by doped GaAs layers and by AlGaAs layers. The RAS response on doping and composition is pronounced at high photon energies. Fig. 5 shows the RAS signal recorded at 3.9 eV, depending on the free carrier concentration of p- and n-type layers, doped using CBr₄ and DETe precursors, respectively.

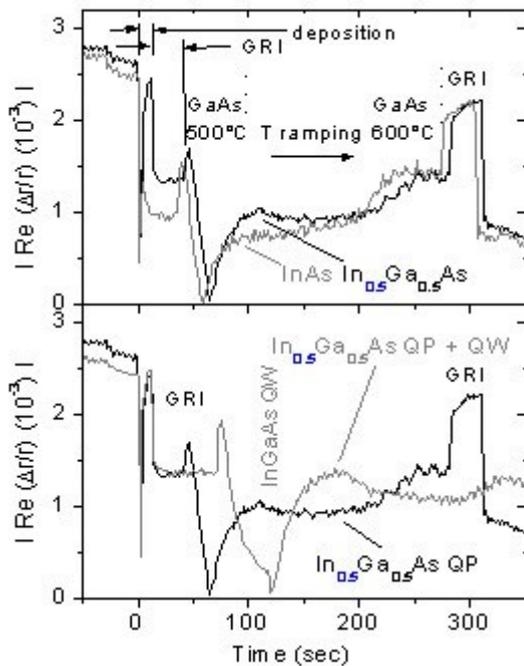


Fig. 4. Absolute value of the RAS transient, recorded at 2.6 eV during quantum dot growth. Black curves are transients of In(0.5)Ga(0.5)As quantum dots, upper grey curve refers to InAs dots, lower grey curve to In(0.5)Ga(0.5)As overgrown with an In(0.1)Ga(0.9)As quantum well. GRI denotes growth interruptions.

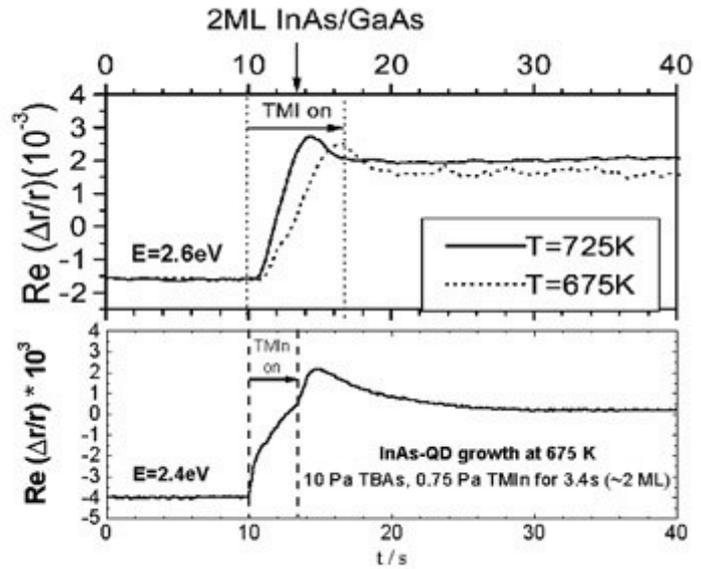


Fig. 3. RAS transients: comparison of previous work of Steimetz et al. [5] (upper graph, scanning RAS) to our experiments (below). Although the photon energies are not the same, the general shape of the 675K transients is similar.

The RAS spectra were obtained from a multiplayer sample with stepwise increased doping levels, separated by undoped GaAs spacers used as markers. The corresponding carrier concentrations were determined using Van-der-Pauw-Hall measurements of a set of 1 μm thick doped GaAs layers grown with the same precursor flows as used in the multiplayer sample. Fig. 5 shows that the optical response is particularly sensitive to n-type doping. The composition of the AlGaAs cladding layers was determined from reflectivity transients of an AlGaAs/GaAs multilayer test structure with 100 nm thick AlGaAs layers of different Al contents separated by GaAs layers. The Fabry-Pérot interference fringes and their first minima are used to evaluate the Al content, the growth rate and the optical constants, see Fig. 6. The growth parameters measured in situ were used to fabricate structures of both, edge and surface emitting lasers. The optical response on the growth of an edge emitting quantum dot laser with an active fivefold quantum dot stack is shown in Fig. 7. In this fingerprint features of the five quantum dots are clearly visible near 2.6 eV. The particular laser shown here has InGaAs quantum dots overgrown with InGaAs quantum wells as shown in Fig.4.

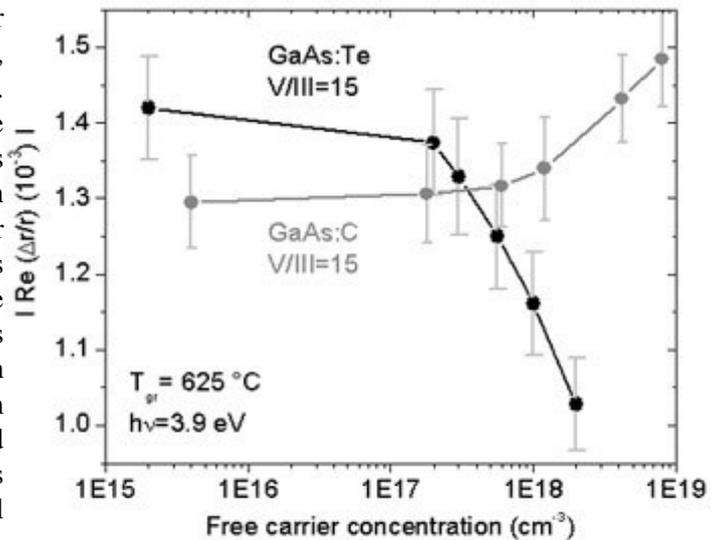


Fig. 5. Surface-sensitive RAS response on doping of GaAs recorded at 3.9 eV. Black: n-type doping with DETe, grey: p-type doping with CBr₄.

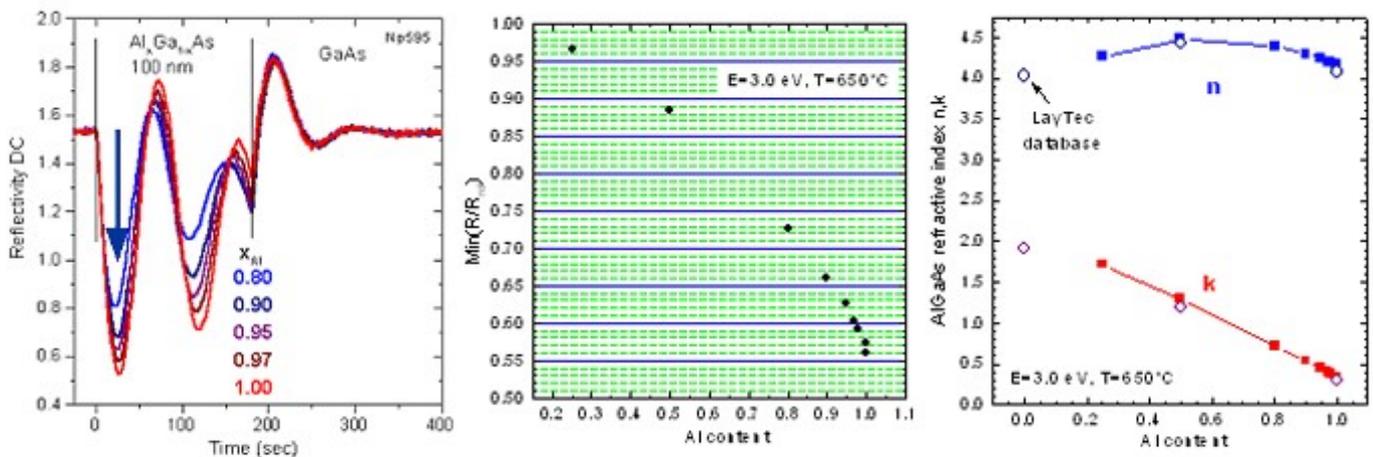


Fig. 6. Left: reflectance transients of an AlGaAs/GaAs multilayer structure with 100 nm thick AlGaAs layers of different Al contents. The onset of AlGaAs growth was shifted to $t = 0$ for all layers. Middle: Al content of the layers determined from the minimum marked by an arrow in the left figure. Right: Optical constants n and k , determined from the oscillations of the reflectivity transients.

The active quantum dot region is cladded by doped Al(0.6)Ga(0.4)As layers. The laser emits with up to 250 mW optical output in the lateral ground mode at 1156 nm. Signatures of doping appear in Fig. 7 in the high energy range near 3.9 eV. The RAS fingerprint shows that a significant part of the growth time is spent for the ramping of the temperature during quantum dot epitaxy, heating at the begin and cooling at the end of the run. Using this fingerprint, deviations from the intended growth recipe may easily be detected online.

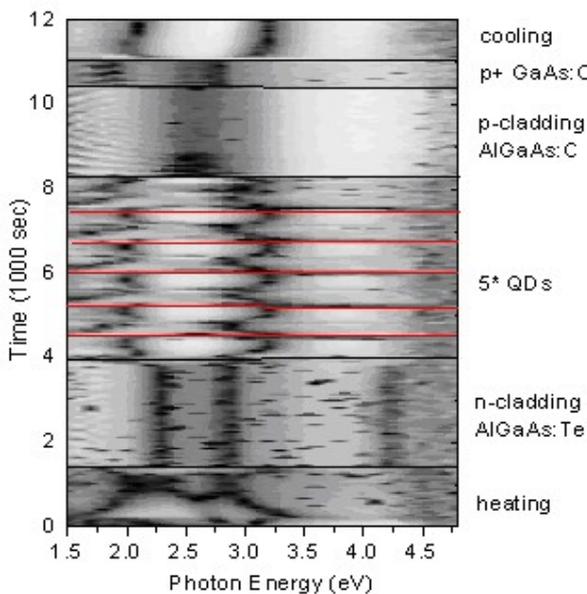


Fig. 7. Grey-scale coded RAS intensity of spectra continuously recorded during epitaxy of an edge emitting laser with a 5-fold quantum dot stack. The characteristic features of the quantum dots appear at 2.6 eV.

Conclusion

The spectral RAS response of the In(Ga)As/GaAs quantum dot formation could be studied online for the first time using a fast multichannel detection for simultaneous optical access at eight different photon energies. Furthermore, RAS fingerprints recorded during epitaxy of complete edge and surface emitting devices with active InGaAs/GaAs quantum dot stacks are presented for the first time.

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