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Invited

Real Time Monitoring of InAs Quantum Dot Formation

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1. Introduction

The one step formation of quantum dots in the Stranski-Krastanow-growth mode has been studied extensively [1,2,3], but a reliable production of high density, coherent quantum dot (QD) arrays for optoelectronic applications is still an open challenge. Therefore there is a high demand for a better understanding and control of the formation processes. In-situ analysis of the QD-formation can provide necessary information for this purpose. In the UHV-based growth methods this information can be partly gained by Reflection High Energy Electron Diffraction (RHEED) which allows to monitor the wetting layer growth and the growth mode transition, but does not give information on the subsequent evolution of the islands. By applying in-situ Reflectance Anisotropy Spectroscopy (RAS) and spectroscopic ellipsometry (SE) we are able to investigate all stages of QD-formation in real-time. In this paper we give an overview over our recent activities.

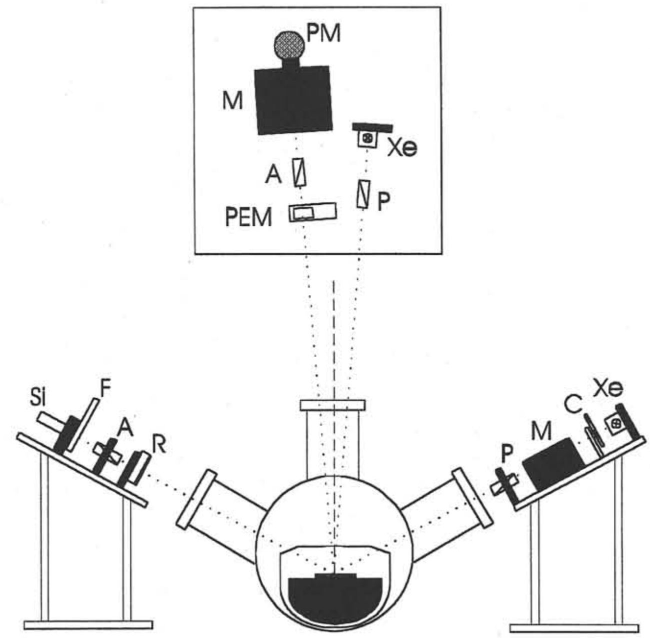

Fig. 1: Setup for real-time RAS and SE-measurements in MOVPE

2. Experimental

All measurements presented were performed in a low pressure, horizontal MOVPE-reactor. Both, RAS and SE are coupled to the MOVPE-process via purged, low-strain quartz windows (Fig. 1). With both optical systems we are able to take simultaneously spectra for photon energies from 1.5eV to 5.5eV and to perform timeresolved measurements with a resolution < 1s at fixed photon energies. The reflectance data shown are evaluated from ellipsometry measurement. Their main purpose is to indicate the scattering losses which reduce the reflectivity in case of 3-dimensional (3D) growth.

3. Results and Discussion

In previous experiments InAs-deposition was performed in submonolayer steps on GaAs(001) up to a coverage of 4ML. Spectra taken inbetween these steps showed strong changes in RAS at submonolayer coverages and a strong reduction of the effective dielectric function in SE when the transition from 2-dimensional (2D) to 3D-growth took place [4, 5]. From these spectral results it was decided to take the RAS-transients for real-time measurements at the characteristic As-dimer related energy of 2.6eV and the ellipsometry transients at 4.7eV, where the surface sensitivity is high. The interpretation of the spectra and the real-time transients is partly based on simultaneously performed RAS and RHEED-studies in CBE and MBE [6,7]. Fig. 2 summarizes the monitoring potential of the three quantities obtained on-line: the 2D growth of the wetting layer is seen by RAS, the onset of 3D-growth and QD development is especially observed with SE and the subsequent development of the islands into larger clusters is indicated by the reflectance.

The RAS signal change is caused by changes of the surface reconstruction. The signal increases linearly with the InAs-coverage. Saturation occurring for a total coverage of approximately 1.8 ML signalizes already the growth mode transition. The reduction of the RAS signal for larger coverages is then caused by a reduction in thickness of the InAs-wetting-layer since material is transferred from the layer to the islands.

Ellipsometry shows the strongest changes in the QD-formation region. There the effective medium includes also the gaseous medium between the islands which has a dielectric function of ≈ 1. To first order $<\varepsilon_2>$ turns out to be proportional to the amount of material that is forming the islands. Only weak changes are observed in the 2D growth mode since the dielectric function of InAs is slightly different from that of GaAs and for large cluster growth, because all light is scattered, SE becomes insensitive. On the other hand then the losses in the reflectance occurring through the scattered light might be used as a monitor when the islands grow beyond quantum size All three optical measurements can be used to optimize growth conditions.

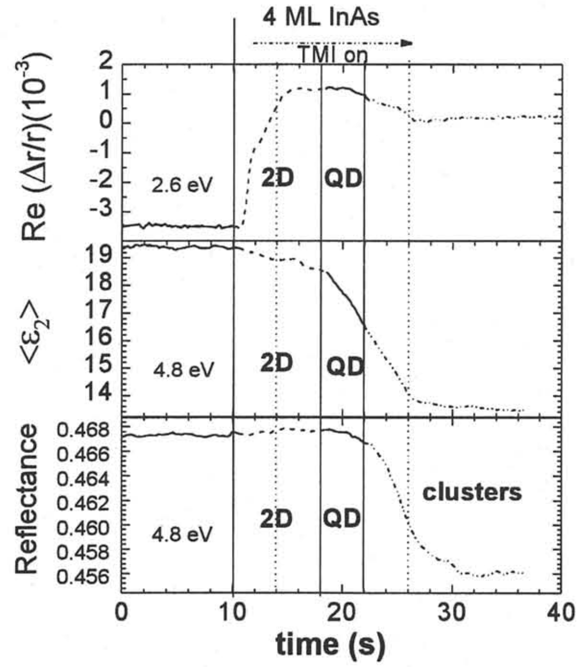

Fig. 2: RAS and SE transients and Reflectance data measured during the deposition of 4ML InAs/GaAs in MOVPE. All stages of QD-formation can be monitored.

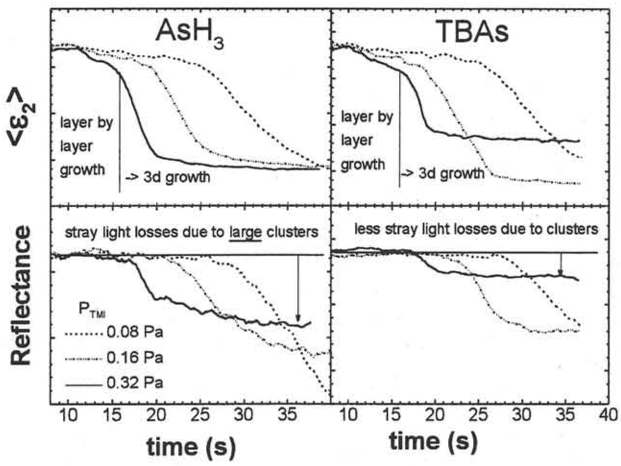

Fig. 3: SE- and Reflectance measurements during the deposition of 4 ML InAs using different As-precursors and deposition rates: the fewest reflectance losses were obtained under TBAs and for the highest growth rate (1ML/s)

In order to obtain small islands the effective dielectric function and the reflectance turn out to be especially useful since they signalize clearly the appearance of large clusters. Low InAs-deposition temperatures (T=450°C), high growth rates (>1ML/s), low V/III ratios and a low total pressure were found to give the lowest number of clusters and thus having the smallest islands (QDs) in highest density at the surface. These results were confirmed by ex-situ AFM and SEM-measurements.

A comparison of MBE, CBE and MOVPE growth with respect to the time necessary to reach surface equilibrium after deposition showed very fast processes in MOVPE in contrast to the UHV-based growth techniques [6]. The origin of this large difference (factor 10) is attributed to the H-radicals present at the surface in MOVPE. Using TMAs (Trimethylarsin) instead of Arsin and thus eliminating the As-H bonds resulted in slower ripening processes at the surface and a more UHV-like behaviour [8].

Moreover, the use of TBAs (Tertiarybutylarsin) instead of AsH_3 gave very promising results with respect to dot size and density. TBAs, due to the lower decomposition temperature, allows both low deposition temperatures (450°C) and low V/III-ratios. The number of As-H bonds is reduced, too. Thus an increased dot density and improved size uniformity was observed in AFM.

In Fig. 3 a comparison of transients taken under AsH_3 and TBAs is made. The reflectance data show less reduction in the case of of TBAs. AFM-pictures showed very high QD-densities (> 10^{11} dots/cm^2) indicating the high potential of TBAs as group V source for QD-formation in MOVPE.

4. Summary

RAS and SE are able to monitor and control the QD-formation in all growth environments. Using the reflectance data from SE-measurements a fast in-situ optimization of the growth conditions was possible. Applying alternative As-precursors for QD-deposition showed promising results.

Acknowledgments

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