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MOVPE grown InGaAs quantum dots of high optical quality as seed layer for low-density InP quantum dots

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Abstract. To achieve a low density of optically active InP-quantum dots we used InGaAs islands embedded in GaAs as a seed layer. First, the structural InGaAs quantum dot properties and the influence of the annealing technique was investigated by atomic force microscope measurements. High-resolution micro-photoluminescence spectra reveal narrow photoluminescence lines, with linewidths down to 11 µeV and fine structure splittings of 25 µeV. Furthermore, using these InGaAs quantum dots as seed layer reduces the InP quantum dot density of optically active quantum dots drastically. InP quantum dot excitonic photoluminescence emission with a linewidth of 140 µeV has been observed.

In recent years, investigations on semiconductor quantum dots (QDs) have been motivated by their potential application as single photon sources and as quantum bits in the field of quantum information processing [1, 2, 3, 4]. For this purpose it is essential to optically or electrically address individual QDs. However, the growth of self-assembled QDs formed by Stranski-Krastanov growth mode [5], often leads to high QD densities using MOVPE. Therefore the samples have to be structured into mesas or shadow masks have to be applied to isolate an individual QD. A solution to overcome this problem should be the growth of self-assembled QDs exhibiting a low density. A narrow linewidth of QD PL emission makes them suitable for research, e.g. for single spin measurement [6], two photon interference [7] and single-photon emitters, respectively. As current single-photon detectors have their highest photon detection efficiency in the red spectral range it is also preferable to fabricate single QDs emitting at such wavelengths [8]. For that reason we focused on the growth of InP QDs embedded in GaInP barrier material that is lattice-matched to GaAs [9]. This material system can emit over a wide spectral range in the visible. The growth of InP QDs in GaInP on such substrates produces QD densities of around $10^{10}$ cm$^{-2}$ which is orders of magnitude too high to study a single QD on an unstructured sample even when using micro-photoluminescence ($\mu$-PL) techniques. To achieve a low density of optically active InP QDs we used InGaAs islands embedded in GaAs as a seed layer. Using the strain produced by the underlying InGaAs islands the InP QDs are expected to preferentially grow on top of the InGaAs seed QDs resulting in a low density of InP QD.

The sample structure was fabricated by MOVPE with standard sources at low pressure (100 mbar) on (100) GaAs substrates oriented $6^\circ$ toward the [111]A direction. Before depositing a GaAs buffer layer the sample is heated up to 750°C to desorb surface oxides and reach the
growth temperature for the following layers. The 50 nm thick Al$_{0.5}$Ga$_{0.5}$As layer prevents from carrier diffusion towards the substrate during optical measurements, due to the higher bandgap compared to the subsequently grown GaAs barrier. The layer of self-assembled InGaAs QDs was grown using the Stranski-Krastanov growth mode by depositing 2 monolayers (ML) of InAs at 520°C and a growth rate of 0.63 ML/s on top of a 134 nm thick GaAs barrier layer. After the deposition of the QDs a growth interruption of 60 s was applied to allow QD ripening. This step was followed by an additional deposition of 3 nm GaAs on top of the InAs QDs at 520°C to reduce the Indium desorption for the following steps. Additionally, a combined temperature ramping to 600°C and annealing step was applied. During the 15 min lasting annealing step Indium-Gallium intermixing took place and therewith a blueshift of the QD PL emission was induced. PL samples were capped with 30 nm GaAs. (Fig.1(a) lower part)

**Figure 1.** (a) Reference sample structure. (b) µPL emission of the InGaAs QDs. (c) Autocorrelation measurement $g^{(2)}(\tau)$ for the investigated sample, which is nearly background free and dips to a value of $g^{(2)}(\tau = 0) = 0.03$. The red line represents a fit to the experimental data. (d) Ensemble PL measurements for different InAs deposition amounts. (e) QD density as a function of the amount of deposited material. The red line is a guide to the eye. (f) shows a µ-PL spectra and the corresponding power dependence of the excitonic, biexcitonic and trionic labeled peaks.

To gain control of the InGaAs QD growth, the amount of deposited material was varied. Using AFM and scanning electron microscopy (SEM) measurements the structural QD properties were examined for not overgrown samples. The QD density can be controllably adjusted over a range of three orders of magnitude between $10^9$ cm$^{-2}$ and $10^6$ cm$^{-2}$. The average QD height ranges from 10.2 nm (4 ML) to 4.2 nm (2 ML). This very pronounced influence on the structural QD properties should also be reflected in the optical properties. The samples were investigated by ensemble PL measurements (Fig.1(d)). The spectra show distinct peaks at around 826 nm originating from the GaAs barrier and a wetting layer (WL) emission around 885 nm. The relatively broad emission at higher wavelength originates from the QD ensemble. The QD ensemble emission blueshifts by reducing the InAs amount, corresponding to smaller sized QDs. Furthermore, the decrease in the integrated PL intensity can be attributed to the reduced QD density.

In order to perform µ-PL measurements a standard resolution PL [10, 11] and high resolution photoluminescence (HRPL) setups are used which are described elsewhere [10]. Investigations
on two-photon correlations have been performed in terms of second-order $g^2(\tau)$ auto-correlation measurements on a Hanbury-Brown and Twiss (HBT)-type setup on a sample with 2 ML InAs deposition.

Due to the low QD density ($\sim 10^7 \text{ cm}^{-2}$) we were able to adress single QDs optically without any need for mask processes. A typical QD emission is displayed in detail in the $\mu$-PL spectra in Fig. 1(b). The emission energy is typically located around 920 nm. In Fig. 1(b) the signal to noise ratio of the PL peak at 916 nm is of about 120, signal to noise ratios of up to 140 were found. Since, there was no significant background observed, the use of the QDs as a single photon-source at high repetition rate is feasible. For single photon emission with high signal to noise ratios, no further background correction has to be applied. This is essential for high bit rate data communication.

In order to get a deeper insight into the emission properties of the grown QDs, we performed power-dependent $\mu$-PL measurements to identify the exciton complexes responsible for the observed lines. The spectrum is dominated by the peak at 908.2 nm which we attributed to an exciton (X) emission (Fig. 1(f)). A second peak is observed at 908.6 nm and becomes more pronounced for higher excitation energies (Bicexciton, (XX)). The third peak at 908.9 nm is of less intensity and maybe attributed to charged exciton emission. Many-body interactions like carrier exchange and Coulomb interactions give rise to a red shift of the charged excitonic states compared to the neutral states [1], therewith the redshifted, $X^*$ labeled PL peak is estimated to be a charged exciton. The assignment of X, XX and $X^*$-emission to the PL peaks is based on analysis of the different integrated PL intensities (Inset in Fig. Fig. 1(f)). We extracted a exponent $p_X$ of 1.1 for the excitonic PL peak and a value of $p_{XX}$=1.9 for the biexcitonic PL emission.

In order to probe the zero-dimensional state-densities of the investigated InAs QDs, we performed continuous-wave (cw) second-order auto-correlation measurements. The correlation function $g^2(\tau)$ is shown in Fig. 1(c). The $g^2(\tau)$ measurements were performed on an excitonic emission. For true single photon emitters a $g^2(\tau=0)$ value of zero is expected. The measurement yields a value of $g^2(\tau=0)=0.03$. Thus, the emitted photons depict a clear antibunching behavior. A deviation of $g^2(\tau=0)$ from zero takes place due to background. The nearly background free $g^2(\tau)$ and signal to noise ratios of up to 140 already indicate a good optical quality. In order to determine the intrinsic linewidth, HRPL measurements were accomplished. The linewidth of the PL peak was determined in terms of full width of half maximum of a fitted Lorentzian function. For an excitation power density of 0.7 W/cm$^2$ a FWHM of 11 $\mu$eV was found. To further resolve the fine structure splitting $\Delta E_{FS}$, HRPL measurements with a resolution of $\Delta E_{\text{res}}=0.3 \mu$eV have been performed. Under stepwise variation of the linear polarization detection angle a clear fine-structure splitting of $\Delta E_{\text{HRPL}}=25 \mu$eV becomes obvious.

In a next step InGaAs QDs were used as an seed for InP QDs. Therefore the sample structure was changed corresponding to Fig. 1(a) upper part. The InGaAs QDs were overgrown by 20 nm GaAs. A 10 nm lattice-matched Ga0.51In0.49P layer followed at 720°C. The self-assembled InP QDs were grown by depositing 2 ML of InP at 710°C and a growth rate of 1.05 ML/s. After a growth interruption of 20 s to ripen of the QDs a 30 nm cap of GaInP followed.

In Fig. 2(b) the AFM height topography of some InP QDs is shown. A density of approximately $4 \times 10^9 \text{ cm}^{-2}$ could be observed. This is more than two orders of magnitude higher than expected. Also the heights of the InP QDs are much larger (up to 40 nm) than the heights without a seed layer (around 2 nm [9]). Two different island types can be distinguished: small truncated pyramid structures and large dome shaped islands that show highly indexed crystal planes. The strain produced by the underlying InAs islands results in a different growth environment for the InP/GaInP QDs. $\mu$PL measurements of these InP QDs show that only a small percentage of the QDs are optically active. Therefore no structuring was needed to
investigate single QDs. A typical emission linewidth of 140 µeV could be measured (Fig. 2(a)).
Autocorrelation measurements reveal a \( g^2(\tau = 0) = 0.4 \) for the PL emission in Fig. 2(a). In
ensemble PL (2(c)) measurements the PL of the InGaAs seed layer (around 930 nm), the InP QDs (around 660 nm) as well as GaAs barrier are observed. Only the small QDs as shown in the AFM measurements seem to be optically active. The growth of InP QDs on such defective InAs clusters might induce also a defect in the overgrowing layer, and thus in the InP QDs on top. Therefore the large dome shaped InP islands seem to be incoherent and optically inactive. These structures may provide non-radiative decay channels to the carriers leading to decreased quantum efficiency. Only the small InP QDs which were grown on top of the small InAs islands or in between big InAs clusters are optically active.

In summary, we have shown that we are able to deposit low-density (In\(_x\)Ga\(_{1-x}\))As QDs by MOVPE using partial capping and annealing steps for narrow linewidth single dot emission. HRPL measurements show a FWHM of the excitonic QD emission of 11 µeV. Polarization dependent measurements revealed a fine structure splitting of 25 µeV. We proved the zero-dimensional state-density of the InAs islands by performing correlation measurements \((g^{(2)}(\tau = 0) = 0.03)\). The InGaAs QDs have been used as seed for InP QDs, resulting in a lower density of optically active InP QDs. This work was supported by the Deutsche Forschungsgemeinschaft (FOR730).

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