Confocal Micro-Photoluminescence and Micro-Photoluminescence Excitation Spectroscopy on Single Self Assembled InAs Quantum Dots

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Carrier dynamics in semiconductor quantum dots (QDs) have attracted much attention during the last decade, since a profound understanding of the underlying processes is essential for the development of novel optoelectronic devices. Furthermore, a direct addressing of the quantum dot states by means of optical coupling between electronic states will open the door for coherent manipulation of these states. However, most of the optical studies in QDs are carried out on dot ensembles because single dot spectroscopy needs high spatial resolution and appropriate dot densities in the range of approximately 10^8 dots/cm^2 or even less. While dot ensembles imply the advantage of a higher signal to noise ratio due to the higher amount of emitters, their spectroscopic signature suffers at the same time from ambiguities imposed by ensemble broadening and possible dot-dot interactions. Thus, for basic studies of optical QD properties it is desirable to have a tool to access and manipulate single QDs. In this contribution we present first results on our micro-photoluminescence (µPL) and µPL-excitation (µPLE) setup and demonstrate its abilities as a tool for nanophotonic studies on semiconductor QDs.

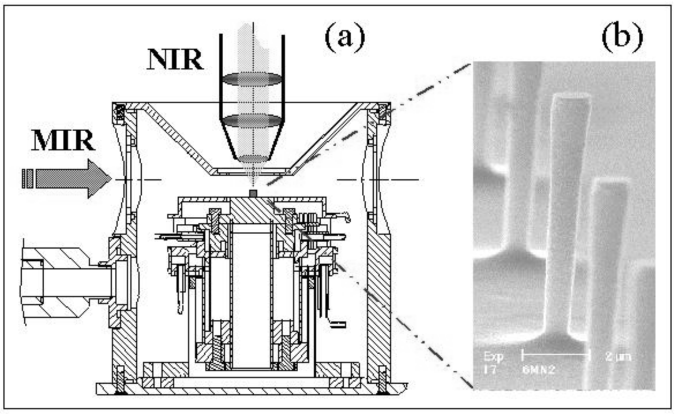

Fig.1: (a) Coupling scheme for the µPL and µPLE measurements. The MIR radiation is focussed by parabolic mirrors via the KBr side windows of the cryostat.
(b) Typical 1 µm diameter micropillars on which the single dot experiments are carried out.

The functional part of the experimental setup is sketched in Fig. 1 (a). The near infrared (NIR) excitation path as well as the PL detection path lead through a self-built confocal laser scanning microscope (LSM) with a lateral and axial resolution below 500 nm. The sample is placed in a low drift Oxford LHe flow cryostat and excited through the top window by a picosecond Ti:sapphire-laser at 745 nm with different excitation densities for the μPL measurements. For μPLE the light source is a cw Ti:sapphire-laser, which is continuously tunable between 700 nm and 1080 nm with a linewidth of less than 30 GHz. The μPL signal is filtered by appropriate long pass filters and dispersed in a 0.5 m spectrometer, which allows a spectral resolution better than 50 μeV with a nitrogen cooled CCD detector or for small dot ensembles the signal can be recorded by a nitrogen cooled InGaAs photodiode. The CCD limits the spectral region to 1100 nm, while the diode allows detecting signals up to 1500 nm. Depending on the wavelength and the polarization of the emission the spectral efficiency of the setup reaches 3 % to 10 % for low light applications with a high signal to noise ratio. Besides top coupling of light, the cryostat allows superimposing other light sources such as mid-infrared radiation (MIR) via the side windows in a cross correlation scheme.

For the samples a QD density of approximately 10^8 dots/cm^2 or below is required due to the lateral resolution of the confocal setup. We could obtain this density by a gradient molecular beam epitaxial (MBE) growth, where the rotation of the wafer is stopped while the InAs is deposited on the GaAs. In low density region the ground state emission of the dots varies between 1040 nm and 870 nm. In order to exclude signal contributions from neighboring dots the sample is processed into an array of micropillars as shown in Fig. 1 (b). These pillars, which are approximately 8 μm high and vary between 1 μm and 10 μm in diameter, allow the addressing of single QDs. Furthermore, they act as microantennas for the superimposed MIR radiation.

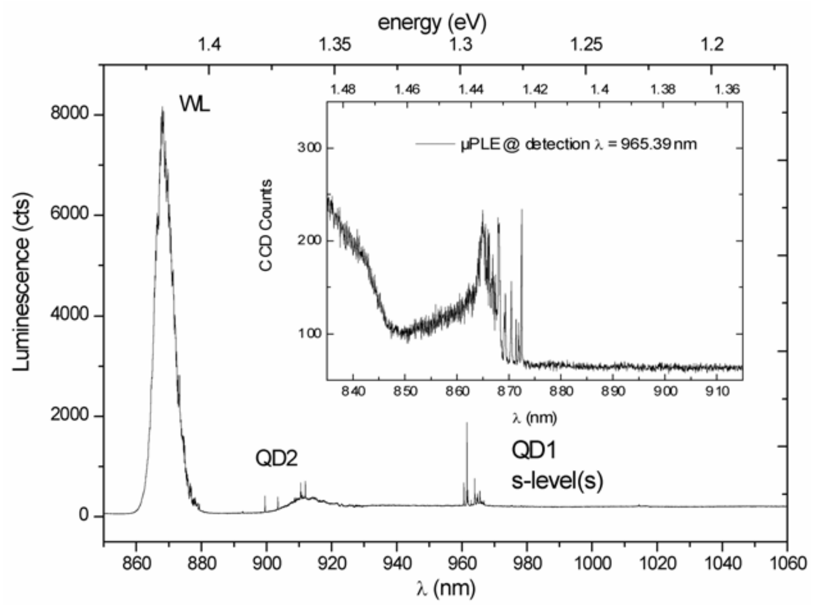

Fig. 2: μPL spectrum of 2 QDs within a 10 μm micropillar: The spectrum exhibits two groups of discrete lines, which correspond to the interband transitions of the excitons within the different QDs. The inset shows the μPLE spectrum of the 965.39 nm transition in QD1. Resonant states are found close to the broadened WL transition.

Figure 2 shows the high excitation μPL spectrum of a 10 μm micropillar which contains two quantum dots. The corresponding μPLE spectrum exhibits some sharp resonances around 873 nm very of the 965.39 nm 1-S exciton transition close to the wetting layer (WL), which is centered around 865 nm with and FWHM of 7 nm. This indicates that for the investigated dot the excited states can be found very close to the wetting layer states. The broadening of these excited states might be due to an interaction of discrete QD states with the broader two dimensional quantum well states of the WL.

In a second experiment we superimpose MIR radiation with the NIR excitation on a similar sample. An Ion-Optics glow bar generates the black body spectrum, which is used as THz/MIR source for the 10 meV to 900 meV radiation. This spectrum is coupled via two parabolic mirrors and through the KBr side window into the cryostat with a focal spot diameter of approximately 1 mm. The measurement in the lower energy level around 960 nm is performed in three steps: First the PL spectrum under NIR laser excitation is recorded with the CCD and labeled as NIR01. In the second step the MIR source is superimposed with the laser excitation and the second spectrum is recorded as MIR01. In a third step we use again only the laser excitation and label the spectrum NIR02. The whole procedure is than repeated for the spectra of the excited levels around 910 nm. Subtraction of the NIR01 spectra from the MIR01 spectra reflects the changes in the count rate for the different levels.

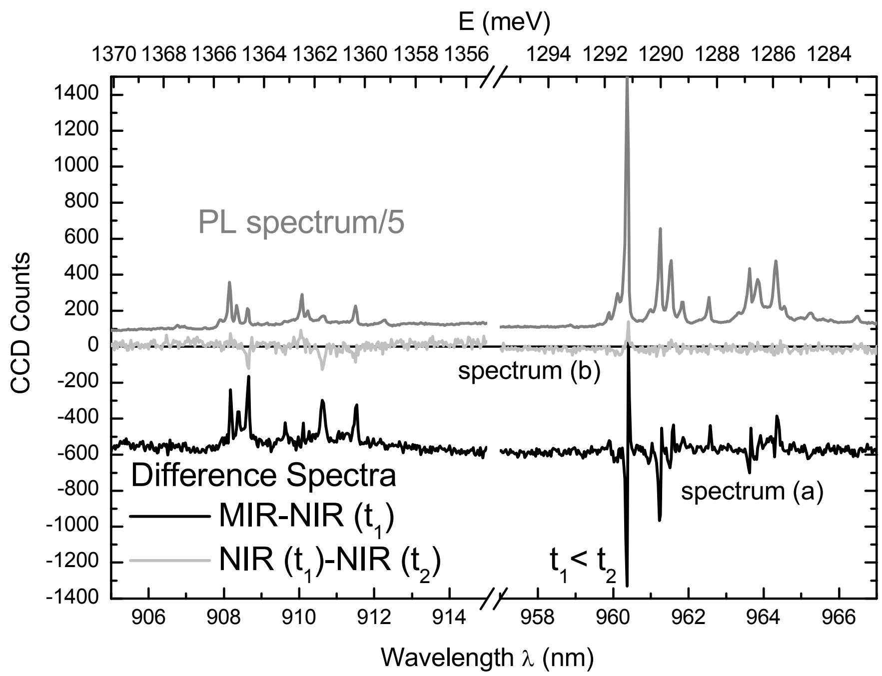

Fig. 3: Near infrared PL spectrum of the SQD at an excitation density of 576 W/cm^2 and corresponding difference spectra: In the case of the superimposed NIR, MIR excitation the level population changes are monitored via the NIR µPL signal. The count rate in the lower energy states is reduced, while an increase in the upper states can be measured.

Subtracting NIR02 from NIR01 (spectrum (b) in Fig. 3) gives a measure for drift in the whole system. The results in Fig. 3 show the impact of the MIR radiation on the count rate, while the NIR excitation density was kept constant at 1440 W/cm^2. In the lower energy levels of difference spectrum (a) in Fig. 3 the count rate is clearly reduced while in the higher levels, which are separated by approximately 75 meV from the lower levels, the luminescence is increased. This result strongly suggests that we observe a direct electron transfer from the lower electronic levels of the QD into higher levels. The drift during the experiment is negligible as indicated by spectrum (b). Because of the high cw excitation density, which nearly saturates the lower level PL, we assume that the underlying process for the PL changes results rather from a direct, optically induced transition into higher dot states than a mediated transition via WL or GaAs CB states [1]. The latter process would change the count rate in both levels into the same direction due to the very fast scattering mechanisms within the dot.

In conclusion, we could demonstrate that we built up a powerful microscopy setup, which provides us with the necessary tools for basic quantum dot studies. In first experiments we could already show a direct, optical induced electron transfer between intersublevels within a QD. In a new experiment we will extend the possibilities of the system by combining it with a cross correlation NIR pump, MIR probe setup. In this approach we want to further investigate the electron dynamics of QDs. The ensemble studies [2] will first be intensified on samples with reduced dot densities and finally in a second step single dot dynamics should be monitored by its NIR emission. Thus we can directly study the dynamical behavior of single quantum dots.

References

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[2] T. Müller, F.F. Schrey, G. Strasser, and K. Unterrainer, Appl. Phys. Lett. 83, 3572 (2003)