Nonlinear Behaviour of the Resonance Fluorescence from Excitons in Quantum Wells

G K G Burau¹, G Manzke¹, F Kieseling¹, H Stolz², D Reuter² and A Wieck²

¹Institut für Physik, Universität Rostock, D-18051 Rostock Germany
²Fachbereich Physik, Universität Bochum, D-44780 Bochum, Germany

E-mail: gerolf.bura@uni-rostock.de

Abstract. We examine the resonance fluorescence from localized excitons in GaAs/AlGaAs quantum wells with increasing excitation power and for different temperatures. Extending our experimental setup by a microscope objective with high numerical aperture in the cryostat, the detection of the emission from localized excitons is possible. We find a nonlinear behaviour of the emitted intensity with increasing laser power, which is explained as a transition of the emission from excitonic to electron-hole plasma states due to many-body effects between excited carriers. This is supported by our theoretical description based on the semiconductor Bloch equations.

1. Introduction
Resonance fluorescence has been established as a valuable tool for the detection of localized excitons in quantum wells (QW) [1,2,3], appearing due to fluctuations of the well sizes. In previous papers [2, 3] we have investigated spectra of the resonance fluorescence in GaAs/GaAlAs quantum wells (QWs) detected from a large area of the sample, averaging over many localized excitons. The reason was a limited spatial and spectral resolution and a rather large excitation spot. To investigate single localized exciton states we have developed a new experimental setup with high numerical aperture objectives inside the cryostat giving a spatial resolution of 650 nm at a wavelength of 812 nm of the exciting laser. This enables (i) the spectroscopy of single localized exciton states, and (ii) an increase of the excitation intensity in the laser spot by a factor of 50.

Our sample consists of a series of single QWs with varying well width [4]. The sample is excited by a single mode cw laser with a line width of 1 MHz resonantly to the exciton states. We investigate the behaviour of the resonance fluorescence of localized excitons with increasing laser power and for different temperatures in the cryostat. With our theoretical calculations based on the semiconductor Bloch equations we explain that the nonlinear behaviour of the intensity of emission with increasing excitation power is caused by many-body-effects between excited carriers.

2. Experimental Setup
Our sample is a high quality GaAs/AlGaAs heterostructure consisting of a series of QWs with different well sizes, ranging from 3.3 nm to 19.8 nm, which was used in earlier experiments [2, 3,4]. The sample is contained in a He-flow cryostat (“4π-microscopic-cryostat”) and cooled to temperatures from 4.5 K to 100 K. We have modified the cryostat to be able, to change both the position of the sample and a microscope objective inside the cryostat, in order to resolve the localized excitons and to increase the detected intensity of emission. In the experiments reported here resonantly excited the 19.8 nm QW with a tuneable single mode semiconductor cw laser (1 MHz line width). The laser beam was stretched with a beam expander and focused lateral on the QW below the Brewster angle, generating a small excitation spot with high intensity. In comparison to earlier experiments we could
enhance the intensity of the spot. The intensity of the excitation was reduced with a circular neutral density filter leaving the excitation area unchanged. In addition we use a chopper to prevent a local heating of the sample. The wavelength and the stabilisation of the single frequency laser (mode hopping) was controlled by a wavemeter including a spectrum analyser.

The secondary emission of the QW was collected with the microscope objective inside the cryostat having a high numerical aperture (NA 0.8), a small working distance and a magnification of 100. This collimated signal was alternatively either imaged onto a cooled CCD-camera with a pixel size of 6.45μm x 6.45μm, or detected by a spectrograph with a spectral resolution of 8 μeV.

3. Results and discussion

Figure 1 shows the spectral (left) and the spatial distribution of the emission for low excitation (laser power 100 μW). Due to fluctuations of the size of the quantum well the excitons feel a localisation potential [3]. Localized exciton states are clearly resolved in the middle and right part of the figure. The intensity of the emission directly reflects the oscillator strength the number of localized excitons. The left part of the figure shows the spectrally resolved intensity of the emission of a 10 μm broad slit, detected by the spectrometer.

![Figure 1. Spatial (right) and spectral (small bar left) distribution of the emission at resonant excitation 369.1423 THz of the 19.8 nm QW and a temperature of 5 K.](image)

For the further spectral analysis, we selected one localized state and tuned the laser power from 100 μW to 19 mW (Fig. 2). The sharp resonant Rayleigh peak is located at the maximum of the broader exciton emission (please note the logarithmic scale). The linewidth of the exciton is at low excitation about 40 μeV reduced by a factor of 4 in comparison to earlier experiments [2,3], where the emission is detected for a large area of the sample, comprising and averaging many of the different localized states, which are shown in the right part of Fig. 1.

At low laser power (up to 3 mW) we find a decrease of the Rayleigh peak with increasing excitation, while for higher laser power the excitonic groundstate vanishes and the Rayleigh peak is increasing again. The same qualitative behaviour is found for different localized exciton states and can be recovered quantitatively by repetition of the experiments, too. Furthermore, a weak shift of the exciton to higher energies can be observed, although the energy of the exciting laser is unchanged. This shift strongly depends of the temperature. The blue shift turns into a red shift, when the temperature is increased to 15 K. Supported by our theoretical treatment in the next section we explain the nonlinear behaviour of the emission by the breaking of the excitonic binding due to many-body effects between excited carriers, called Mott transition of excitons [7,8], which should appear for an excitation between 3 mW and 6 mW. For lower excitation the emission comes from excitons (bound e-h pair states), for higher excitation from the e-h plasma (scattering states of the e-h pairs).

To illustrate this, we have divided the intensity of the Rayleigh peak $I_R(\omega_L)$ by the intensity of the exciting laser $I_L(\omega_L)$, what should be a measure for the absorptivity $A(\omega_L)$ [5]. Figure 3 shows the
Figure 2. Spectrally resolved intensity of emission with increasing power of excitation for a localized exciton state.

density of excited carriers (left) and the absorptivity $A(\omega_L)$ in dependence of the power of excitation $I_L(\omega_L)$ (right). The excitation density is determined considering the integral intensity, the excitation area, the quantum well size, and the live time of the excitons. Due to the large absorptivity at low excitation the density of carriers strongly increases with increasing laser power. This turns into a weak increase for high excitation. Simultaneously, the absorptivity strongly decreases followed by a weak increase. We interpret this nonlinear behaviour of the emission (see next section) as the transition from bound e-h pairs (excitons) to non-bound e-h pairs (e-h plasma), called Mott transition.

Figure 3. Density of excited carriers (left) and absorptivity $A(\omega_L)$–$I_L(\omega_L)/I_L(\omega_L)$ (right) in dependence of the power of excitation.

4. Theoretical results and interpretation

In order to explain the nonlinear behaviour of the resonance fluorescence with increasing excitation we have calculated the changes of the susceptibility of the 19.8 nm quantum well by solution of the semiconductor Bloch equations [4,6]. Fig. 1 shows the imaginary part of the susceptibility determining the absorption spectrum for temperatures of $T = 5$ K / 15 K and different densities of carriers. Full lines show the spectra for localized excitons, dashed lines model spatially integrated spectra. The second ones correspond to those measured in earlier experiments [4] and were calculated with an inhomogeneous linewidth of $\gamma = 0.18$ meV. According to our new experiments for the localized excitons the linewidth is reduced by a factor of 4 (see section 3). The dynamical screening of the Coulomb interaction between carriers leads to a broadening and a shift of the exciton line with increasing excitation. The shift of the exciton line with increasing excitation to higher energies at $T = 5$ K transfers to lower energies for $T = 15$ K. This is in qualitative accordance with our experiments and was already discussed both experimentally and theoretically in [7] for ZnSe heterostructures. The
absolute values of the shifts are overestimated by our theoretical treatment for the higher densities and lower temperatures. This is due to the use of chemical potentials for ideal (non-correlated) particles in our calculations, which would be strongly decreased if many-body effects are taken into account [8,9].

The vertical black line marks the position of the exciting laser being fixed in the experiments. The cross points with the absorption curves (see arrows in the right figure) reflect the decrease of the absorption with increasing excitation and are related to the points given in the right part of Fig. 3. By further increase of the carrier density the exciton peak is broadened out and vanishes in the band edge. This is demonstrated in the inset, where the behaviour of the band edge and the vanishing of the 2s-exciton state with increasing excitation is better resolved in logarithmic plots.

Finally, fixing the energy and increasing the intensity of the laser we observe a transition of the resonance fluorescence from excitons (bound electron-hole pairs) to states of correlated carriers (continuum, non-bound electron-hole pairs), which is known as Mott transition of excitons [3, 4]. As for the excitonic ground state (1s-state) we have investigated the Mott transition of the 2s-state, too. This is possible only for those experiments, where localized excitons are resolved, while the 2s-features in the spatially integrated spectra are broadened out (see dashed lines in both insets).

**Figure 4.** Imaginary part of the susceptibility (theoretical calculation) of the 19.8 nm quantum well for different temperatures and carrier densities: $n = 10^{14}$ cm$^{-3}$ (corresponds to non-excited case) (a), $n = 10^{15}$ cm$^{-3}$ (b), $n = 5.10^{15}$ cm$^{-3}$ (c), $n = 10^{16}$ cm$^{-3}$ (d), and $n = 2.10^{16}$ cm$^{-3}$ (e). The energy scale is given as distance to the gap energy $E_g$ and in units of the binding energy in bulk GaAs ($E_{xb} = 4.2$ meV). Spectra for localized excitons are given with solid, spatially integrated spectra with dashed lines (see text).

5. Acknowledgements
We acknowledge financial support by the Deutsche Forschungsgemeinschaft through the research group FOR 485 “Quantum optics in Semiconductor Nanostructures” and the SFB 652 “Strong correlations and collective effects in radiation fields: Coulomb systems, clusters and particles”.

6. References
[1] Hegarty J, Sturge M D, Weisbuch C, Gossard A C and Wiegmann W 1982 *Phys. Rev. Lett.* 49 930
[2] Schwedt D, Nacke Ch, Stolz H, Eshlaghi S, Reuter D and Wieck A 2003 *Phys. Rev. B* 67 195303
[3] Schwedt D, Schwartz R, Stolz H, Reuter D and Wieck A 2006 *physica status solidi (c)* 3 2477
[4] Nacke Ch, Stolz H, Manzke G and Henneberger K 2002 *Eur. Phys. J. B* 30 303
[5] Richter F, Florian M and Henneberger K 2008 *Phys. Rev. B* 78 205114
[6] Manzke G and Henneberger K 2002 *phys.stat.sol. (b)* 234 233
[7] Manzke G, Peng Q Y, Henneberger K, Neukirch U, Hauke K, Wundke K, Gutowski J and Hommel D 1998 *Phys. Rev. Lett.* 80 4943
[8] Zimmermann R 1988 *Many-Particle Theory of Highly Excited Semiconductors* (Leipzig: Teubner)
[9] Ebeling W, Kraeft W D and Kremp D 1976 *Theory of Bound States and Ionization Equilibrium in Plasmas and Solids* (Berlin: Akademie–Verlag)