Simultaneous observation of confined energy levels and multi-excitonic features in a quantum dot

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Abstract: We study the photoluminescence properties of GaAs/AlGaAs QD arrays grown by MBE on a patterned GaAs (311) substrate. At low excitation density, we observe sharp (<100 meV) excitonic recombination features, due to both the ground state and the exited energy levels of the individual dots. The excited levels are occupied due to the phonon bottleneck and allow to determine the dot shape. At higher excitation density, distinct bi-excitonic and multi-excitonic features appear which show a strong non-linear behavior.

Introduction

Usually, macro-photoluminescence (PL) spectra of quantum dot (QD) materials show strong inhomogeneous broadening due to the size distribution of the individual QDs [1]. Higher confined energy levels become visible [1] at high excitation density due to state-filling. Micro-PL measurements are employed to excite a limited number of dots, resulting in a PL spectrum with a number of sharp peaks, spread over the same spectral range as the macro-PL line. In such a situation, it is impossible to determine whether some of the sharp emission peaks belong to a single QD or whether all peaks are due to different dots. Only in the case when the micro-PL could be focused on a single QD [2], the spectra have been interpreted in terms of bi-exciton and multi-excitonic features.

We performed micro-PL measurements on atomic H assisted MBE-grown arrays of GaAs/$Al_{0.7}Ga_{0.3}As$ QDs on a patterned GaAs (311) substrate [3]. The QDs are aligned in 1-dimensional arrays along a [01-1] mesa edge, patterned on a GaAs substrate. A [01-1] oriented mesa on a (311)A substrate provides a fast growing sidewall, resulting in a [01-1] oriented GaAs/AlGaAs quantum wire in the absence of atomic hydrogen. In the presence of atomic hydrogen, these quantum wires are subdivided into an array of QDs due to an atomic hydrogen induced step bunching along the [-233] direction, i.e. perpendicular to the wire. The QD dimensions are approximately 40 to 50 nm squared and 6 nm high as determined by AFM measurements [3]. The QD density is $1.5 \cdot 10^5$ cm^{-1}.

A very special property of these QD arrays is their local homogeneity. In particular, a PL-linewidth of only 68 μeV is observed between 1.60 and 1.65 eV over spatial areas of up to 16 μm [3]. The PL of the hydrogen-induced corrugated quantum well is located around 1.78 eV.

Experimental results and discussion

The sample containing the arrays of QDs is cooled down to 5 K. An aspheric 2.0 mm focal length lens is mounted in the cryostat, directly in front of one of the QD arrays to

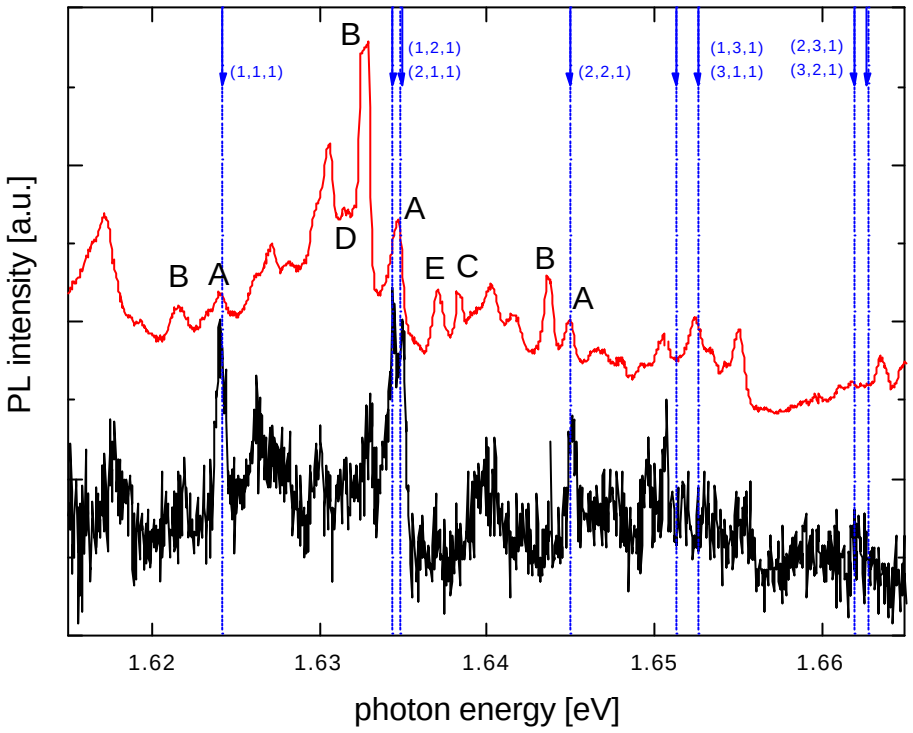

Figure 1: Micro-PL spectra of a QD array. The lower curve is at low excitation density (4W/cm^2) showing only single exciton transitions. The upper curve is at high excitation density (20W/cm^2), showing also bi-excitonic and multi-excitonic features labeled B-E. The vertical lines are fits to the low-excitation density spectrum using a particle-in-a-box model.

provide a laser spot size of 1.8 µm. The spot position can be precisely adjusted using external controls. For excitation we use a synchronously pumped dye laser at 2.12 eV, which is below the $Al_{0.7}Ga_{0.3}As$ band gap. The PL signal is dispersed by a Jobin-Yvon triple monochromator using either additive or subtractive mode. Finally, the PL is detected by a LN cooled 2-dimensional CCD detector. When using the CCD in imaging mode, we can image the PL spectra of an array of QDs over a distance of 80 µm.

The observed PL spectrum is shown in Fig. 1 for both low and high excitation. Particularly at high excitation, the observed PL spectrum is very rich. A closer examination of the excitation power dependence of the PL spectra reveals that most of the peaks increase super-linearly with excitation, pointing towards bi-excitonic and multi-excitonic features due to the interaction of multiple excitons in a single QD [2]. At low excitation power, the observed PL spectrum still consists of at least 10 features. The main question is however whether these peaks are due to dots of different size or whether these peaks have to be attributed to the excited state of an array of homogeneous dots. In the remainder of this paper, we will provide evidence that these peaks are at least partly due to the excited states of a homogenous array of dots.

The first piece of evidence is provided by the PL images along the array of dots on the CCD detector. As shown in Fig. 2, we typically observe all different peaks on a single position along the array of dots. All observed PL peaks thus originate from a ≈2µm long piece of the QD array.

Also by moving the sample up and down along the array of QDs a few microns, we don't observe the emerging and disappearing of PL peaks in a random way. This proves that the array of QDs does not consist of dots of randomly fluctuating size on a scale of several tens of microns. However, since the spatial resolution of our optical imaging system is limited, it is not impossible that the dots are inhomogeneous within the 2 μm laser spot. We however conclude that a random array of inhomogeneous dots is *not* observed at a length scale of 2-20 μm.

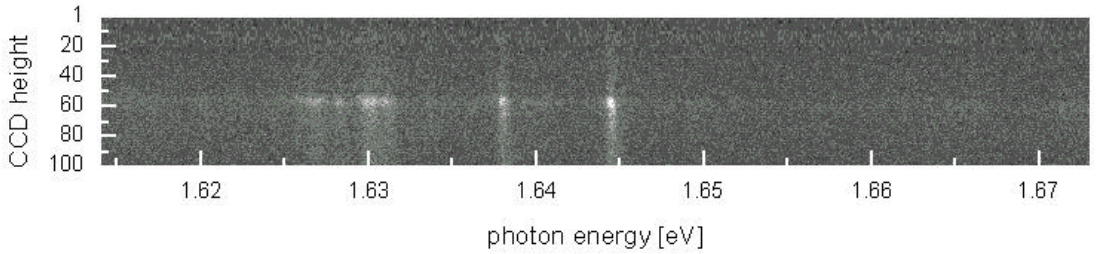

Fig. 2: CCD image of the QD array. The full height corresponds to 80 mm on the sample. The spectrum is dispersed along the horizontal axis.

Our second piece of evidence is that the spectral position of the PL peaks at low excitation density is not random. In Fig. 1 we show that most of the observed PL peaks can be fitted by a very simple particle-in-a-box model, occupied by a single exciton. For these fits, we use QD dimensions of 43.1 x 41.9 x 8.06 nm, which are very close to the QD dimensions observed in AFM [3]. In Fig. 1, we only plotted the calculated (n_e, m_e, 1) to (n_{hh}, m_{hh}, 1) transitions with the in-plane quantum numbers $n_e = n_{hh}$ and $m_e = m_{hh}$. The light-hole exciton transitions and e.g. (1,1,2) transitions due to excited states in the growth direction are at much higher energy. When fitting the (1,1,1), (1,2,1) and (2,1,1) lines, the model correctly predicts the (2,2,1) line and the spacing between the (1,3,1) and (3,1,1) lines. The error in the exact position of the latter two lines might be due to the simplicity of the particle-in-a-box model. It should be noted that the model does not include all peaks observed. Noteworthy exceptions are the lines at 1.617 and 1.626 eV. These two lines however have almost the same spacing as the 1.626 eV and 1.634 eV peaks, again suggesting that these peaks do not seem to have random spectral positions.

The occurrence of excited-state PL due to single excitons suggests the existence of a phonon bottleneck in our QDs. The existence of a phonon bottleneck is a controversial issue in literature, since one usually observes rapid relaxation towards the ground state of the QD. However, this rapid relaxation is usually observed either at higher carrier injection levels where carrier-carrier scattering and Auger processes become important, or in strained self-assembled QDs where the strain field induces a polarized exciton within the dot yielding an enhanced Fröhlich interaction [4]. Our GaAs/AlGaAs QDs are however completely strain-free, implying that a phonon bottleneck might very well be present at low excitation density.

We finally focus on the spectrum at higher excitation density. We observe the appearance of bi- and multi-excitonic QD states, which grow super linearly with excitation power,

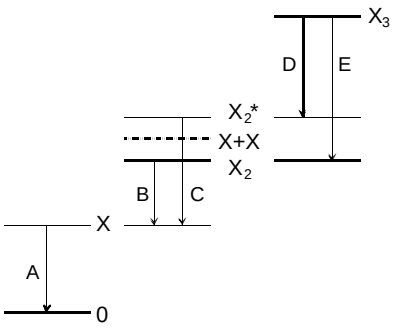

Figure 3: Level scheme for the bi-exciton X_2 to exciton X and the tri-exciton X_3 to bi-exciton transitions. X_2 is the excited bi-exciton with one exciton in the (1,1,1) ground level and a second exciton in the (2,1,1) or (1,2,1) excited state. The transitions A-E correspond to the PL spectrum in Fig. 1*

while the single exciton peaks (A) saturate. At 2.4 meV below the (1,1,1) peak, a second peak (B) appears in the high-excitation spectrum, which we attribute to the bi-exciton line with a bi-exciton binding energy of 2.4 meV. An even stronger bi-excitonic feature is observed below the (1,2,1)/(2,1,1) line. Here we find a bi-exciton binding energy of 2.0 meV. Multi-excitonic features appear when the dot is filled with three or more excitons. As shown in Fig. 3, one expects PL peaks corresponding to the relaxation of the tri-exciton X_3 to the bi-exciton X_2 or to the excited bi-exciton X_2* in which one of the excitons occupies an excited confinement level. The splitting between the two tri-exciton lines D-E is equal to the splitting of the two bi-exciton lines B-C. As shown in Fig. 1, we indeed observe a set of peaks around the (1,2,1)/(2,1,1) lines corresponding to the C-E lines in the multi-exciton level scheme, although an unambiguous classification of all peaks observed is difficult.

Conclusion

Micro-PL measurements on an array of homogeneous QDs show a set of one-exciton peaks at low excitation density that can be described with a particle-in-a-box model. At high excitation, distinct bi-exciton and tri-exciton peaks appear. The PL images show that all peaks observed originate from the same 2 μm diameter part of the sample, also suggesting that all peaks observed originate from an array of homogeneous QDs.

References

[1] S. Raymond, X. Guo, J.L. Merz and S. Fafard, "Excited-state radiative lifetimes in self-assembled quantum dots obtained by state-filling spectroscopy", Phys. Rev. B59, pp 7624, 1999; O. Stier, M. Grundmann and D. Bimberg, "Electronic and optical properties of strained quantum dots modeled by 8 band k.p the Phys. Rev. B59, pp 5688, 1999

[2] M. Bayer, T. Gutbrot, A. Forchel, V.D. Kulakovskii, A. Gorbunov, M. Michel, R. Steffen and K.H. Wang, "Exciton complexes in $In_xGa_{1-x}As$/GaAs quantum dots", Phys. Rev. B58, pp 4740, 1998

[3] R. Nötzel, Z. Niu, M. Ramsteiner, H.-P. Schönherr, A. Tranpert, L. Däweritz and K.H. Ploog, "Uniform quantum-dot arrays formed by natural self-faceting on patterned substrates", Nature 392, pp 56, 1998

[4] A.W.E. Minnaert, A. Yu. Silov, W. van der Vleuten, J.E.M. Haverkort and J.H. Wolter, "Fröhlich interaction in InAs/GaAs self-assembled quantum dots", Phys. Rev. B63, pp 75303, 2001