Nonlinear behavior of multi-excitonic recombinations in quantum dots.

F.P.J.de Groote, T.van Lippen, J.E.M.Haverkort, R.Nötzel, J.H.Wolter

COBRA Inter-University Research Institute, Eindhoven University of Technology, Physics Department
P.O. Box 513, NL-5600 MB Eindhoven, The Netherlands; E-mail: f.d.groote@phys.tue.nl

We studied quantum dot arrays using m-PL measurements. Clear multi-excitonic spectra are observed, which are attributed to homogeneous ensembles of QDs in the array. The spectra can be explained by applying a Hartree-Fock based model including the two-body Coulomb and exchange interactions. Eight lines in the m-PL spectrum are attributed to bi-exciton, excited bi-exciton and multi-exciton transitions. These lines show a clear nonlinear behavior since they only appear at high excitation power.

Introduction

Inhomogeneous broadening effects in self-assembled quantum dot (SAQD) samples tend to partially obscure multi-excitonic features [1]. One usually circumvents these problems by studying single QDs, e.g. by confocal photoluminescence (PL) microscopy [2]. In this report we take another approach by studying locally homogeneous QD arrays [3]. The PL linewidth of these dots is narrow enough to clearly resolve all multi-excitonic features which emerge at higher excitation density. The QDs within the arrays are in general rectangular of shape, lifting the in-plane excited state degeneracy. Moreover, as compared to SAQDs, our QDs are strictly strain-free, thus circumventing strain-induced carrier polarization effects [4] within the dot. In this paper, we will focus on the classification of the bi-excitonic and multi-excitonic features which show a nonlinear behavior as function of excitation density.

Experiment

The sample under study consists of atomic hydrogen assisted MBE grown arrays of $GaAs/Al_{0.7}Ga_{0.3}As$ QDs on a patterned GaAs (311)A substrate [3]. The QD dimensions are approximately 40 x 40 nm squared and 6 nm high as determined by AFM. The QD density is 1.5×10^{5} cm^{-1}. An aspherical lens with numerical aperture 0.5 is mounted inside the He-flow cryostat, 2 mm in front of the sample. It can be moved externally with a precision of 0.1 μm. The lens focuses the excitation beam down to a 1.8 μm diameter spot and also collects the PL. The excitation power density is varied between 10 and 10^{4} W/cm^{2} for measuring spectra in which the QDs are filled with either single or multiple excitons. Measurements are performed at 5K. The spectra are dispersed by a monochromator, followed by a cooled CCD detector. The overall system resolution is 80 μeV.

Experimental results

A typical example of the observed sharp multi-excitonic spectra is shown in Fig. 1. We obtain a number of strong indications for the local homogeneity of the QD-array:
(i) All peaks of a spectrum appear and disappear almost simultaneously when scanning the laser spot along the QD-array. *(ii)* The QD emission peaks show a small asymmetric broadening, as already reported by Nötzel [3], indicating small size variations between the

homogeneous QDs. *(iii)* Exciton lifetime measurements show that only the ground state exciton has the expected delayed risetime [5] at higher excitation density. *(iv)* None of the observed spectra show randomly distributed peaks along the wavelength axis; the position of different peaks is strongly correlated. On distances >10 μm along the QD array the PL spectra vary strongly, indicating that the QD dimensions are only locally homogeneous.

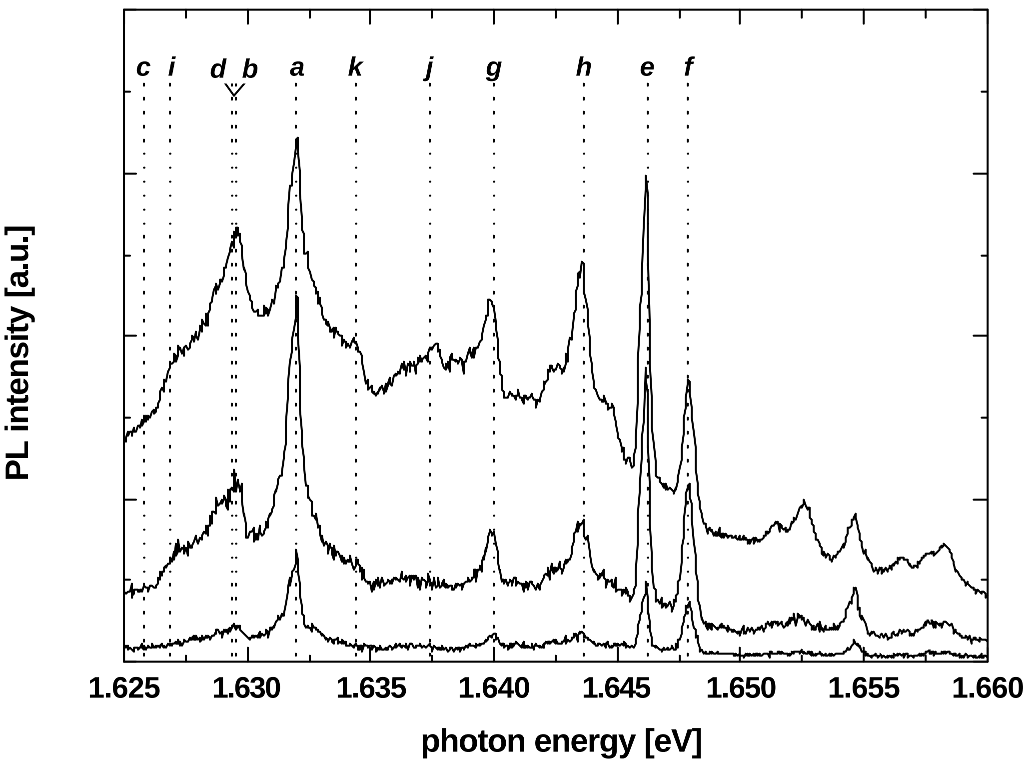

Fig. 1.: Three mPL spectra, taken at excitation densities of 16, 64 and 640 W/cm^2, respectively. The transitions labeled ‘a’, ‘e’ and ‘f’ emerge at low excitation and are identified as ground state (11) and excited state (12, 21) single-exciton recombinations. The bi-exciton transition is labeled ‘b’. The other peaks increase nonlinearly with excitation density. The dashed vertical lines are fits to the spectrum, see Table I.

For the spectrum displayed in Fig. 1 the (11) ground state single exciton (1X) is observed at 1.6320 eV. The bi-exciton (2X) is located at 1.6295 eV, corresponding to a 2X binding energy of 2.5 meV. Two additional sharp peaks at 1.6462 and 1.6479 eV, visible at low excitation density, are attributed to excited (12,21) single excitons. The choice for this assignment corresponds to a rectangular QD of $36.2 \times 38.9 \times 7.40$ nm^3, in agreement with AFM measurements [3]. Moreover, it allows fitting another 7 multi-exciton ($3X \rightarrow 2X$ and $4X \rightarrow 3X$) and excited bi-exciton features emerging at high excitation density, with only a small number of additional free parameters.

Theory

We treat the QD confinement levels with a simple particle-in-a-box model. The energy levels are denoted by ($n_x n_y$), with (11) the 1X ground state (in our QDs, $n_z = 1$). In the Hartree-

Fock (HF) approximation, the Coulomb (c) and exchange (x) corrections [6] to the single particle states are $V_{c,ij}^{p/q} = \langle \varphi_i^p \varphi_j^q V_{ij} \varphi_i^p \varphi_j^q \rangle$ and $V_{x,ij}^{p/q} = \langle \varphi_i^p \varphi_j^q V_{ij} \varphi_i^q \varphi_j^p \rangle$, with $V_{ij} = e^2 / 4\pi\varepsilon_0\varepsilon_r (\vec{r}_i - \vec{r}_j)$. Single particle wavefunctions are indicated by φ, with p and q the appropriate energy levels and $i, j \in$ [electron, hole]. Coulomb terms apply to e-e, h-h and e-h interaction, while exchange terms only apply to e-e and h-h interactions with identical spin. For two interacting excitons, the Coulomb and exchange corrections are given by $V_c^{p/q} = V_{c,ee}^{p/q} + V_{c,hh}^{p/q} + 2V_{eh}^{p/q}$ and $V_x^{p/q} = V_{x,ee}^{p/q} + V_{x,hh}^{p/q}$, respectively. All recombination energies are summarized in Table I, with E_g the bandgap and E_e^{11} and E_h^{11} the electron and hole confinement energies. $V_c^{11/11}$ is also known as the bi-exciton binding energy. Specific values, extracted from a fit to the spectrum, are listed in Table II.

PL transition	PL energy (peak)	
$1X \to v$	$E_{1X} = E_g + E_e^{11} + E_h^{11} + V_{c,eh}^{11/11}$	(a)
$1X^* \to v$	$E_{1X^*} = E_g + E_e^{12} + E_h^{12} + V_{c,eh}^{12/12}$	(e)
$1X^{**} \to v$	$E_{1X^{**}} = E_g + E_e^{21} + E_h^{21} + V_{c,eh}^{21/21}$	(f)
$2X \to 1X$	$E_{2X} = E_{1X} + V_c^{11/11}$	(b)
$2X^{*s} \to 1X$	$E_{1X^*} + V_c^{11/12}$	(h)
$2X^{*t} \to 1X$	$E_{1X^*} + V_c^{11/12} + V_x^{11/12}$	(g)
$2X^{*s} \to 1X^*$	$E_{1X} + V_c^{11/12}$	(d)
$2X^{*t} \to 1X^*$	$E_{1X} + V_c^{11/12} + V_x^{11/12}$	(c)
$3X \to 2X$	$E_{3X} = E_{1X^*} + 2V_c^{11/12} + V_x^{11/12}$	(j)
$3X \to 2X^{*t}$	$E_{2X} + V_c^{11/12}$	(i)
$3X \to 2X^{*s}$	$E_{2X} + V_c^{11/12} + V_x^{11/12}$	(-)
$4X \to 3X$	$E_{3X} + V_c^{12/12}$	(k)

Table I: Various (multi)-excitonic transition energies for a QD occupied with up to 4 excitons in energy levels (11) and (12). Labels 'a' through 'k' mark the spectral transitions in Fig. 1 (transition '-' is located outside the observed spectral range).

Term	Energy [eV]
E_{1X}	1.6320
E_{1X^*}	1.6462
$E_{1X^{**}}$	1.6479
$V_c^{11/11}$	$-2.5 \cdot 10^{-3}$
$V_c^{11/12}$	$-2.6 \cdot 10^{-3}$
$V_c^{12/12}$	$-3.0 \cdot 10^{-3}$
$V_x^{11/12}$	$-3.6 \cdot 10^{-3}$

Table II: Extracted values from our fit to the data in Fig. 1.

Once E_{1X}, E_{1X^*}, $E_{1X^{**}}$ and $V_c^{11/11}$ are established, 6 other peaks in the spectrum (labeled c, d, i, j, g, h) can be fitted using $V_c^{11/12}$ and $V_x^{11/12}$ as additional parameters. The $4X \to 3X$ recombination (line k) requires one additional free parameter $V_c^{12/12}$. Fig. 2a and b show some of the various recombinations. The transitions (g) and (h) represent triplet 't' and singlet 's' excited 2X recombinations. The other two excited 2X recombinations (c,d) are visible as a shoulder of the 2X recombination line (b).

A clear non-linear behavior can be seen from all multi-excitonic lines in the spectrum. They are only visible at higher excitation power, as opposed to the 1X and excited 1X recombination lines. Furthermore, they rise faster with increasing excitation power.

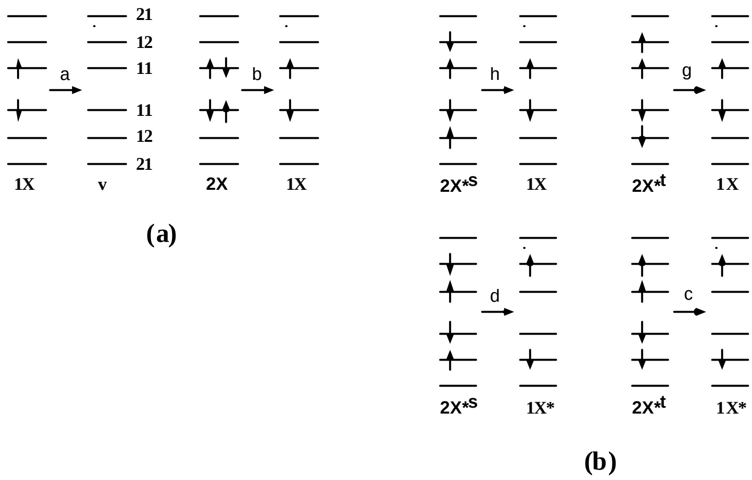

Fig. 2. (a) Ground state exciton 1X ® v(acuum) and bi-exciton 2X ® 1X recombinations, showing the ground (11) and excited (12) and (21) QD-states. Spin states are represented by vertical arrows. (b) Various relevant excited 2X transitions. Superscripts 's,t' indicate singlet and triplet configurations. Labels 'a' - 'h' are assigned in Table I.

Discussion

It is remarkable that we obtain the best fit by including excited 1X and excited 2X transitions, pointing towards an incomplete carrier relaxation within this specific strain-free QD sample. In SAQDs, a strain-induced polarized exciton is present which is expected to enhance the Fröhlich interaction [4]. If one believes that the fast carrier relaxation in SAQDs is due to the strain-induced polarized exciton, the carrier relaxation in our strain-free QDs will be considerable slower. We believe that such an incomplete carrier relaxation within our QD-arrays is the only way to explain the very rich µPL spectra observed in this sample.

From the application point of view, we observe clear excited bi-exciton and multi-exciton features which only appear at increased excitation density. The multi-excitonic features appear as sharp lines and are thus very promising for all-optical nonlinearities.

References

[1] M. Bayer, T. Gutbrod, A. Forchel, V.D. Kulakovskii, A. Gorbunov, M. Michel, R. Steffen, K.H. Wang, "Exciton complexes in $In_xGa_{1-x}As$/GaAs quantum dots", Phys.Rev.B58, pp. 4740-4753, 1998.

[2] K. Brunner, U. Bockelmann, G. Abstreiter, M. Walther, G. Böhm, G. Tränkle, G. Weimann, "Photoluminescence from a single GaAs/AlGaAs quantum dot", Phys.Rev.Lett. 69, pp. 3216-3219, 1992.

[3] R. Nötzel, Z. Niu, M. Ramsteiner, H.-P. Schönherr, A. Trampert, L. Däweritz, K.H. Ploog, "Uniform quantum-dot arrays formed by natural self-faceting on patterned substrates", Nature 392, pp. 56-59, 1998.

[4] A.W.E. Minnaert, A.Yu. Silov, W. van der Vleuten, J.E.M. Haverkort, J.H. Wolter, "Fröhlich interaction in InAs/GaAs self-assembled quantum dots", Phys. Rev. B63, pp. 075303-075306, 2001.

[5] G. Bacher, R. Weigand, J. Seufert, N.A. Gippius, V.D. Kulakovskii, A. Forchel, K. Leonardi and D. Hommel, "Dynamics of Excitons and Biexcitons in One Single Quantum Dot", Phys. Stat. Sol. (b) 221, pp. 25-29, 2000.

[6] P. Hawrylak, "Excitonic artificial atoms: Engineering optical properties of quantum dots", Phys. Rev. B60, pp. 5597-5608, 2000.