Type-II GaSb/GaAs quantum rings: charging mechanisms and the bimolecular recombination approximation

Peter D. Hodgson,[1] Robert J. Young,[1] Mazliana Ahmad Kamarudin,[1,2]
Peter J.Carrington,[1] Anthony Krier,[1] Qian D. Zhuang,[1]
Erwin P. Smakman,[3] Paul M. Koenraad[3] and Manus Hayne[1]

1 Department of Physics, Lancaster University, UK
2 Department of Physics, Universiti Putra Malaysia, Malaysia
3 Department of Applied Physics, Eindhoven University of Technology, The Netherlands

i. Introduction

The results presented here are for GaSb/GaAs self-assembled quantum dot (QD) samples that were grown by molecular beam epitaxy. A summary of the growth conditions are shown in Table 1 and Fig. 1. Transmission electron microscopy has revealed the presence of quantum rings (QRs) as well as quantum dots in several of the samples [1]. The GaAs cold cap temperature and thickness affects the ratio of QDs to QRs in the sample. The GaSb/GaAs system has a type-II band alignment, where holes are confined to the GaSb nanostructures, whilst electrons populate the GaAs.

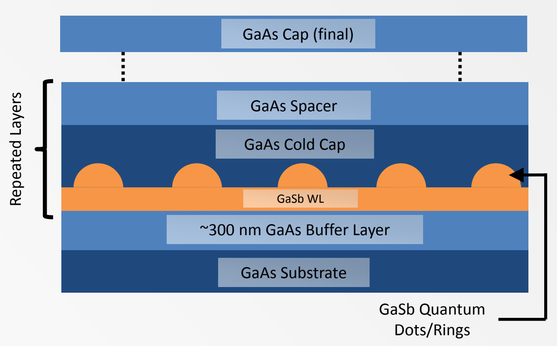

Figure 1. Typical sample structure.

Sample	Growth Method	GaSb (ML)	GaSb (°C)	V/III ratio for QD/QR	GaAs Cold Cap (nm)	GaAs Cold Cap (°C)	GaAs Spacer (nm)	GaAs Spacer (°C)	# of QD/QR layers	GaAs Cap [final] (nm)	GaAs Cap [final] (°C)
A	SK	2.1	490	10	5	430			×1	245	580
B	SK	2.1	490	10	5	430			×1	250	500
C	SK	2.1	490	10	5	430	30	570	×10	100	570
D	SK	2.1	490	10	5	430	30	500	×10	100	500
E	SK	2.1	490	2	9	430			×1	100	580
F	SK	2.1	490	10	8	450	13	510	×3	300	465
G	MEE	1.5	480	10	10	480	40	580	×10	100	580
H	SK	2.1	490	10	10	490			×1	250	570
I	SK	2.1	490	10	10	490			×1	250	500
J	SK	2.1	490	10	10	490	30	570	×10	100	570
K	SK	2.1	490	10	5	430			×1	250	570

Table 1. Summary of sample growth conditions.

ii. Simple Charging Models

The type-II nature of GaSb/GaAs QRs leads to a characteristic blueshift in emission energy with increasing laser power. This blueshift is commonly observed in the literature and attributed to either **capacitive charging** [2] or **band bending** [3] effects. **But which effect dominates?**

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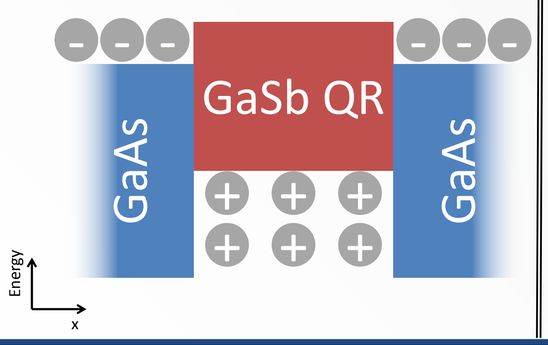
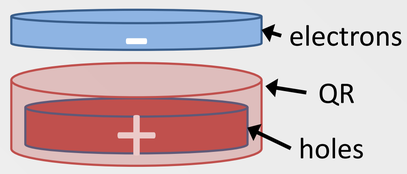


Capacitive Charging

Type-II QRs act as nano-capacitors. After the dot has been charged with one hole, the addition of subsequent holes results in a charging energy:

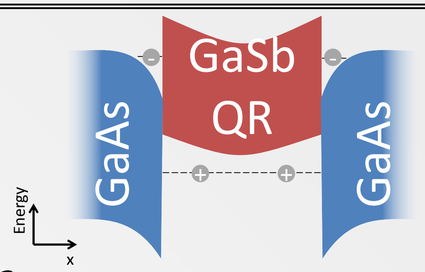
$$\Delta E_{CC} = \frac{e^2 d}{2\pi r^2 \varepsilon_r} n_h$$



e = charge of electron
r = radius of QR
ε_r = relative permittivity
n_h = number of holes in QR
P = incident laser power

Band Bending

The spatial separation of charge carriers in type-II systems creates an electric field which bends the bands to produce an approximately triangular well [4].



$$\Delta E_{BB} = \left(\frac{\hbar^2}{2m}\right)^{\frac{1}{3}} \left(\frac{9}{8}\pi\right)^{\frac{2}{3}} \left(\frac{2\pi e^2}{\varepsilon_0 \varepsilon_r}\right)^{\frac{2}{3}} n^{2/3}$$

ε_0 = permittivity of free space
P = incident laser power
m = effective mass of carrier
n = carrier density

iii. Bimolecular Approximation

The carrier density and QR occupancy, n, are usually not known in photoluminescence experiments. Therefore emission intensity, I, or laser power, P, are often used as more convenient indicators of carrier density. They are linked to the carrier density using the bimolecular rate equation [5], as follows:

$$I = \frac{dn_e}{dt} = \frac{dn_h}{dt} = -bn_e n_h$$

From charge neutrality, $n_e = n_h$, and so

$$I = bn^2,$$

It is often assumed that $I \propto P$. Therefore we can write the energy shifts of the two charging models as:

$$\Delta E_{CC} \propto I^{1/2} \propto P^{1/2}$$
$$\Delta E_{BB} \propto I^{1/3} \propto P^{1/3}$$

iv. Results: Sample A

Uniquely, we observed discretely charged dot states in this sample (labelled peaks in Fig. 2(a)). Analysis of the intensity variation with power for these sub-peaks (see Young et al, Ref: [6]) shows that they are due to discretely charged states.

Fig. 2(a) also shows that the energy shift of the centre of mass (~34 meV) comes from the increase in intensity of peaks at higher energy (greater occupancy). The separation of these sub-peaks is 24 meV and can only result from capacitive charging. Furthermore, as Fig 2 (b) shows, $\Delta E_{CC} \propto n$, which proves that **the capacitive charging model is correct**.

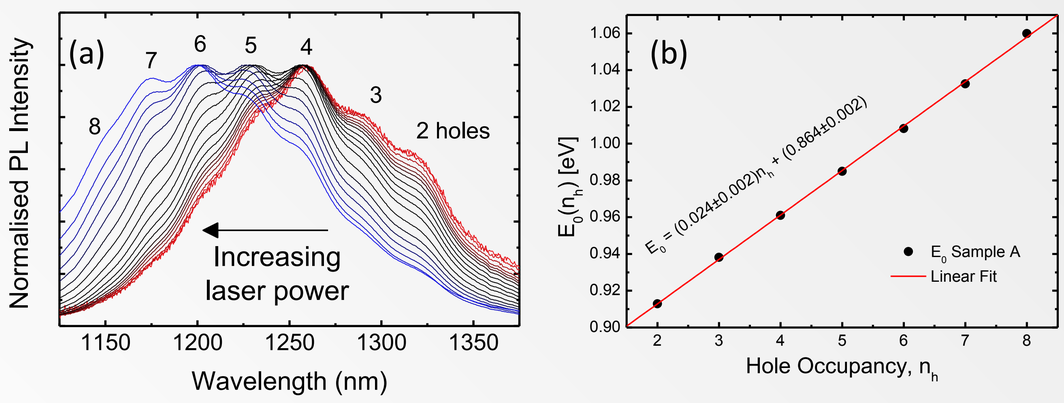


Figure 2. Direct analysis of Sample A. (a) QR emission spectra at different laser powers. (b)The energy separation of the charge-state sub-peaks

The 7 sub peaks have nearly identical energy shifts with laser power, which must result from the effect of unconfined charge in the proximity of the QRs. Therefore the energy shift of the individual charge-state sub-peaks must come from band bending. Furthermore the magnitude of the energy shift for the sub-peaks is only ~3 meV, confirming that **capacitive charging dominates the blueshift of the ensemble spectrum**.

v. Results: All Samples

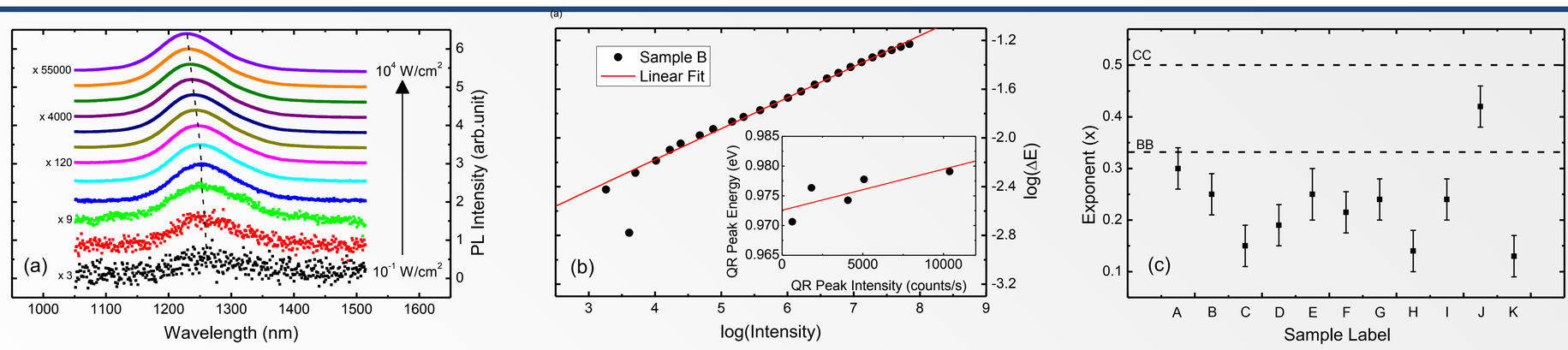


Figure 3.
(a) Example QR spectra from Sample B.
(b) Extracting the exponent for Sample B.
(c) Exponents for all samples. CC and BB show the expected values for capacitive charging and band bending respectively.

Using the information that we have already learned from Sample A:

- Capacitive charging dominates the energy shift.
- The capacitive charging model is correct ($\Delta E_{CC} \propto n_h$).

We can now test if $\Delta E_{CC} \propto I^{1/2}$ is true, as predicted by the bimolecular approximation.

To do this:

1) Find the emission energy, E, and intensity, I, from the emission spectra [Fig 3(a)].
2) Find the minimum emission energy by extrapolating the low power data points to zero [Fig 3(b) inset].
3) Plot a log-log graph of the energy shift, ΔE, against I [Fig. 3(b)].
4) The gradient of this graph is the exponent, x, in $\Delta E_{CC} \propto I^x$

The exponents for all samples are plotted in Fig. 3(c). All of these values are below the expected value of 0.5 (capacitive charging). Therefore it is clear that **the bimolecular recombination approximation cannot be applied to type-II QD/QRs**.

We suggest that the bimolecular recombination coefficient, b, should include an n_h dependence to account for the increased coulomb attraction of electrons to highly charged QDs/QRs.

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