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Lateral absorption measurements of InAs/GaAs quantum dots stacks: potential as intermediate band material for high efficiency solar cells

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Abstract

Prototypes based on InAs/GaAs QDs have been manufactured in order to realize the theoretically predicted high efficiency intermediate band solar cells (IBSCs). Unfortunately, until now, these prototypes have not yet demonstrated the expected increase in efficiency when compared with reference samples without IB material. One of the main arguments explaining this performance is the weak photon absorption in the QD-IB material, arising from a low density of QDs. In this work, we have analyzed the absorption coefficient of the IB material by developing a sample in an optical wave-guided configuration. This configuration allows us to illuminate the QDs laterally, increasing the path length for photon absorption. Using a multi-section metal contact device design, we were able to measure an absorption coefficient of ~100cm^{-1} around the band edge (~1eV) defined by the VB→IB transition in InAs/GaAs QD-IB materials. This figure, and its influence on the IBSC concept, is analyzed for this system.

Keywords: Intermediate band; Quantum dots; Absorption; Characterization; Intermediate band solar cell

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1. Introduction

The intermediate band (IB) materials rely on the creation of a half-filled electronic band within the otherwise conventional bandgap of a semiconductor. Sandwiching the IB material between appropriate *p* and *n* emitters, it is expected to realize the so called intermediate band solar cells (IBSCs). For ideal IBSCs, up to 63.2% of ideal conversion efficiency has been predicted theoretically [1], a figure which represents a notable increase when compared with the 40.7% of efficiency for conventional single gap solar cells.

Among the different approaches suggested for putting into practice the IB concept [2-6], systems based on quantum dots (QDs) have been proposed as suitable candidates for the realization of IBSCs [7]. At present, prototypes based on InAs/GaAs QDs have been manufactured in order to study principles of operation of these kinds of novel cells [8-13]. Unfortunately, these prototypes have not yet demonstrated the expected increase in efficiency when compared with reference samples without IB material [14]. QD-IBSCs have shown a poor increase in the photocurrent and voltage degradation. The voltage degradation arises from the fact that we do not have a true zero density of states between the IB and the CB, something which degrades the wider bandgap transition of the host material. The poor increase in photocurrent has been attributed to weak photon absorption in the QD-IB material, arising from a low volume density of QDs. To overcome the problem of low absorption by the potential QD-IB material, several solutions have been proposed: (i) increase the number of stacked QDs layers [12,15]; (ii) the employment of light management structures within the IB material [16]; (iii) the use of diffraction grids that illuminate laterally the stack of QDs at the wavelengths of interest [17].

In this paper we will discuss the solution of lateral illumination of InAs/GaAs QD-IB materials. For doing so, we have analyzed the absorption coefficient for the VB→IB transition by developing a sample in which the QDs are sandwiched between AlGaAs barriers in an optical wave-guided configuration. This configuration allows us to illuminate the QDs laterally, increasing the path length for photon absorption. Experimental details about the fabrication and operation of these devices (from here, optically coupled devices, OCDs) are presented in the next section.

2. Experimental

The samples were made at Glasgow University by molecular beam epitaxy (MBE). The InAs/GaAs QDs forming the IB material were grown by the Stranski-Krastanow growth mode. In this mode, the QDs appear spontaneously after the deposition of a quite thin quantum well (known as wetting layer) as a consequence of the strain relaxation induced by the different lattice constants of dot (InAs) and barrier (GaAs) materials. The sample employed in the development of the OCDs is a stack of 10 QDs layers sandwiched between ~1.5 microns thick *p* and *n* AlGaAs graded barriers. This structure allows the electrical confinement of carriers within the QD-IB material as well as the lateral optical confinement for photons emitted by the QD stack (wave-guided configuration). Every layer of QDs was grown, at 530ºC, depositing 3.2 monolayers (MLs) of InAs and capped with 8.5 nanometers of GaAs.

After the growth of the wafer, we proceed with the fabrication of the OCDs. By applying photolithography, ~150nm of the *p*+ top layer was removed for defining a ridge for lateral light guiding. 200nm of SiN where deposited in order to isolate electrically the ridge and help for the encapsulation of the QCDs during wire bonding process. Pd+Au based back and front metal contacts were deposited. The frontal metal design consists on 7 isolated metal (of 300μm lengths) paths in order to bias every section independently. Finally, a chemical wet etching was employed ($5H_2SO_4$-H_2O_2-H_2O) on the back facet, obtaining an angle of 57 degrees observed by scanning electron microscopy (SEM). This guarantees the operation of our device in a light single pass mode (not a Fabry-Perot resonant cavity).

Figure 1 shows a sketch of the optically coupled device (right), and the principle of operation (left). By biasing every section (emitter) we obtain an emission which travels through the device (not biased sections operating as absorbers) until leaving the OCD. The emitted light is coming from the ground state of the QDs stack (which corresponds to the IB position in the IBSC model). The emitted light from one section is able to be absorbed by pumping carriers from the VB to the IB and from the IB to the CB (figure 1(right)). In a more realistic scenario (as it is plotted in fig. 1 right), the absorptions through the IB (VB→IB and IB→CB) are in fact transitions between the QD hole states (ΔEv in fig. 1) to the QD electron states (ΔEc in fig. 1) and to the QD electron states to the CB continuum of the host semiconductor (GaAs). Given the inter-sub-band transition strength into continuum states, the latter process is expected to be very unlikely to happen.

Following the Beert-Lambert law which defines the decay of light intensity when it is passing through an absorbing media, we are able to test the absorption coefficient of our 10 QDs layers stack under lateral illumination.

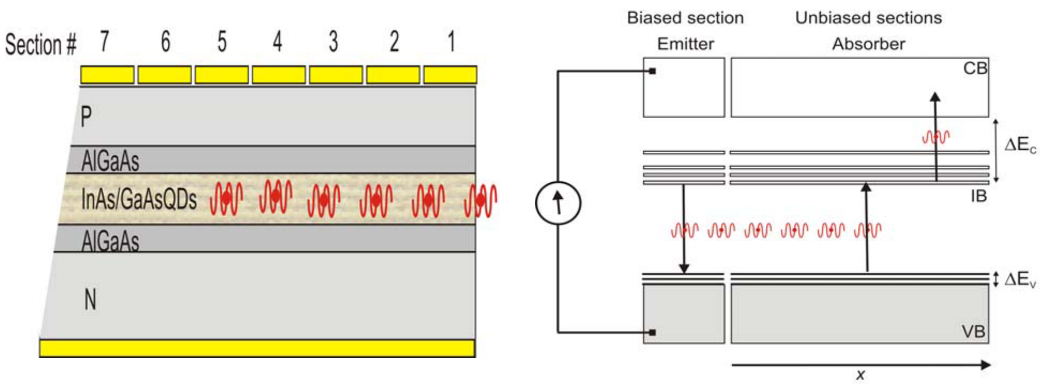

Figure 1: Sketch of the so called optically coupled device (left); and the expected behaviour under operation (right). The output emission of every biased section can be analysed as a function of distance, *x*. In this respect, we are able to measure directly the absorption coefficient of the QD-IB material.

3. Results and discussion

Figure 2 represents the results obtained after the operation of the OCDs. In the bottom graph we observe the emission output for the device when biasing each one of its isolated sections. This emission corresponds to the radiative recombination between the InAs/GaAs QDs ground and first excite state to the VB (or in terms of the ideal IB model, from the IB to the VB). The sections with an area of 300x550µm^2 where injected up to 60mA (36A/cm^2). Due to problems related to contacting metal path 6 (see fig. 1 right), emission data from section is not presented in figure 2.

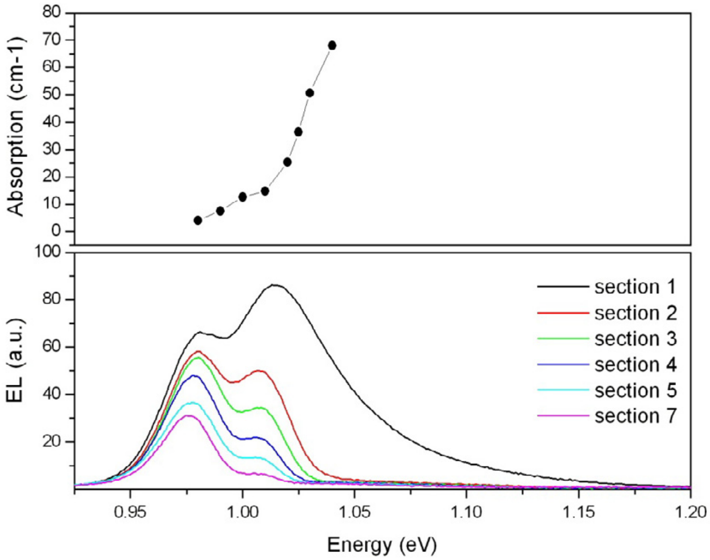

Figure 2: Electroluminescence (EL) emission output signal when biasing each section (bottom); and absorption coefficient for a stack of 10 QDs layers stack under lateral illumination (top).

Following the Beert-Lambert law which describes the intensity light decay when it passes through an absorbing media;

$$I(x) = I_0 \cdot exp(-\alpha \cdot x) \tag{1}$$

we have calculated the absorption coefficient (α) for the energy range in between 0.975eV and 1.05eV for the QD-IB material (top of figure 1). In equation 1, $I(x)$ represents the light intensity as a function of distance x, and I_0 is the value of $I(x=0)$.

The QDs ground state emission for the IB system is located at 0.975eV (Figure 2 bottom). Below this energy no photons are emitted and we do not have data in order to obtain the absorption coefficient. If we consider, as the IB ideal model does, photon absorption selectivity [18], for energies above 0.975eV we are only testing the band edge absorption for the VB→IB transition (or from the VB to the QDs electron states). In a more real scenario, with no photon absorption selectivity, we are actually obtaining the absorption coefficient for the combination of all the accessible absorptions, we mean from the VB→IB and from the IB→CB (the latter is contributed also by the excited estates generated by the dots within that transition, ΔEc in fig. 1 right). Another concern is that a red shift of QDs emission of 40meV (Stokes shift) when compared with the absorption levels estimated by photoreflectance [19] have been observed in previous works for similar samples. Having the same Stokes shift in our OCD sample, this implies that we have photon emission below the VB→IB transition defining the absorption. In other words, part of the emitted light is only able to pump electrons from the IB to the CB (and the QDs excited states therein). Considering this scenario (Figure 3) we have that the emission below 1.015eV (ground state emission minus the Stokes 40meV shift) corresponds with photons only able to pump electrons from the IB to the CB. On the other hand, photons with energy above 1.015eV are able to pump electrons from the VB→CB and IB→CB transitions.

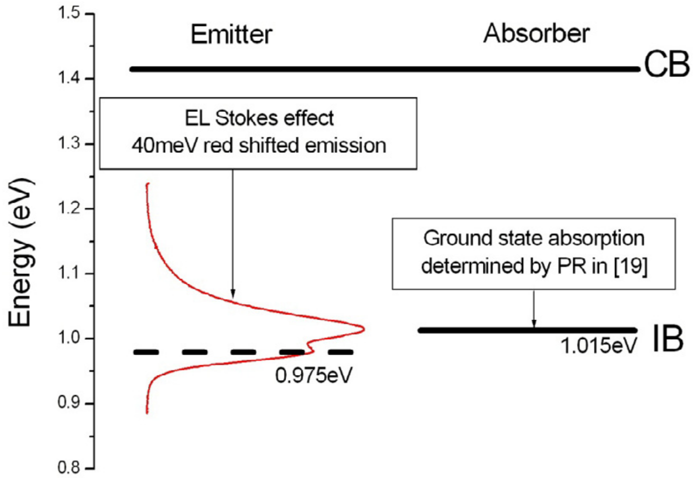

Figure 3: Photons from each biased section (emitter) below 1.015eV are only able to be absorbed by the IB→CB transition. Photons with energy above 1.015eV are able to be absorbed by the VB→IB and IB→CB transitions simultaneously.

For the energy range analyzed we have an indication of the lateral absorption coefficient for both transitions through the intermediate band. In this respect, we obtained (see figure 2) a value of ~10cm^{-1} for the IB→CB transition (far from its energy band edge around 0.4eV) and ~100cm^{-1} for the VB→IB transition in the energy relating band edge absorption. The figure of 10cm-1 (that we have estimated) has to be considered as an upper limit for the absorption within that energy range in the following sense: even if the IB→CB transition would be responsible of this absorption, this would imply an absorption coefficient as low as 10 cm-1 for that transition. Although we do not have data for the analysis of the absorption coefficient above 1.05eV, it is evident following Figure 2 that the absorption coefficient is still growing for higher energies.

The absorption coefficient for the band edge of GaAs (the barrier material of our QD system) is ~10000cm^{-1}, so the absorption coefficients estimations for the transitions through the IB are very much lower figures. The obtained estimations for the absorption coefficients through the IB are in agreement with published results for three InAs/GaAs QDs layers stack samples [20]. Our results explain properly the poor enhancement in photocurrent observed in similar 10 layers stacks QD-IBSCs prototypes [14].

The different values obtained for the absorption coefficient through the IB can imply that the IB operation should be limited by the lower figure of them (~10cm^{-1} for the IB→CB transition). In this respect, it is quite important to test in future works the recombination lifetimes for the different transitions for describing the dynamic operation of IBSCs. In this way, the filling of the IB can extinguish the absorption of carriers from the VB to the IB. On the other hand, an empty IB will be detrimental for the pumping of carriers from the IB to the CB. In this point, it is important to note that our measurements were done for undoped QDs (so the IB is expected to be empty at room temperature before illumination). How the absorption coefficients can be affected by the particular filling conditions (illumination conditions and doping) of the IB is out of the scope of this work.

Another aspect to have into account for our measurements is that the IB-QD material is within the space charge region (SCR) generated by the *p* and *n* emitters which sandwich the IB material. This fact implies that our QD-IB material is within a built-in potential region permanently [19]. Our estimations for the lateral absorption coefficient of the QD-IB material have to be considered then within this condition. The focus of this paper was to analyze the absorption coefficient of QDs under lateral illumination. Considering an ideal scenario of perfectly spherical and distributed QDs in the volume which define the IB material, identical absorption coefficients trough the IB for lateral and normal incidence of light should be expected. If this is close to the real case, our results implies that 2mm thick QD-IB material was unable to absorb all the photons coming from the biased section, and then, the aim of stacking QDs in order to increase the absorption for future devices (based on InAs/GaAs QDs) has to be combined with light management structures [16], in order to confine the photons which have to be absorbed through the IB transitions.

The structural quality of the IB material was analysed by transmission electron microscopy (TEM). A number of TEM images (such as that shown in Figure 4) of the active region were obtained using dark field (DF) 002 imaging conditions to determine the quantum dot growth evolution in the stack structure. The DF 002 images indicate the presence of some areas with QD spatial ordering, i.e. vertically aligned columns of QDs. Nevertheless, there are also areas where alignment of QDs has not been achieved. QDs with lens-shaped (height ~3nm and base length ~10nm) are also revealed. One of the main features to note is that the spacer layers (~7nm) are not flat, probably due to the effect of strain on growth rates. This could be improved through different growth approaches, either using thicker GaAs spacer layers or higher temperature during spacer layer growth. No dislocation or threading defects were observed in the TEM images recorded under bright field 220 and 004 imaging conditions (not shown in this work) in either the active region or the AlGaAs barrier layers. This means the defect density of the system is lower than 10^7 cm^{-2}.

The QDs stacking failures evidenced by TEM in figure 4 surely have had some influence in our estimations for the absorption coefficients. Some authors have reported the improvement in PL efficiency when comparing samples without and with this kind of stacking failures [21]. However, as we commented before, TEM images taken under bright field conditions of the active region showed no dislocations or threading defects within the IB material. These kinds of failures also could modify the QDs density tested and/or can introduce dispersion in the dots sizes that have been analyzed. The inhomogeneous distribution in dot sizes will affect the number of levels between the IB and CB transition, and then, the results presented here (for the absorption coefficients) have to be considered as a particular example for the growth recipe presented in section 2. In any case, the employment of the so called optically coupled devices (OCDs) for the analysis of absorption coefficients for IB related transitions was successful.

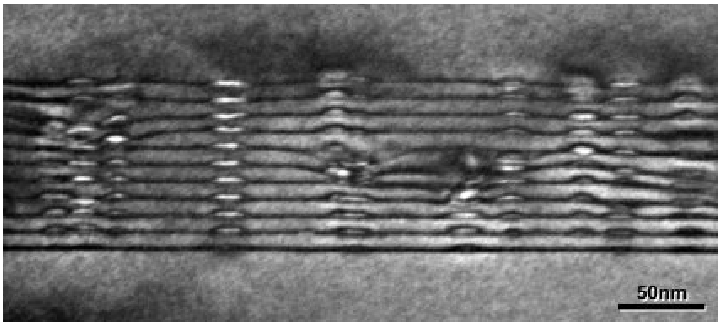

Figure 4: TEM image showing vertical assembling of QDs in the stack (QD-IB material). Several areas where alignment has not been achieved are also observed.

4. Conclusions

Optically coupled devices were developed (and successfully employed) with the aim of estimating the absorption coefficient under lateral illumination of potential QD-IB materials.

The absorption coefficient evolution in between 0.975eV and 1.05eV (in the vicinity of the VB→IB energy band edge) was determined for a stack of 10 InAs/GaAs QDs layers. Values for α up to 100cm^{-1} were obtained within the energy range of analysis. These results have to be considered carefully attending the particular recipe employed for the growth of the QDs within the devices. However, the figures obtained are in agreement with those presented by other authors.

5. Acknowledgements

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