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Optical effects of GaAs Spacer Thickness on InAs/InGaAs QDs Intermediate-Band Solar Cells

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Abstract— **We report the effects of spacer thickness on the optical and spectral response characteristics of five stacked InAs/InGaAs/GaAs quantum dots intermediate band solar cells heterostructures grown by Solid-Source Molecular Beam Epitaxy (SS-MBE). The samples were investigated via photoluminescence (PL) and photocurrent (PC). Distinctive asymmetric shape located at the high energy side is observed in PL spectra of the QD solar cells samples, which can be de-convoluted in two sub-bands. From the excitation power-dependent and the temperature-dependent PL measurement, the two sub-bands are associated to the ground-state transition from InAs QDs with a bimodal size distribution of dots. In addition, the PL spectrum exhibits a blue shift as the spacer thickness decrease and the vertical coupling between QDs layers is more effective due to the strain-driven In/Ga intermixing between QDs and spacer. Solar cells containing quantum dots show a broader photo-response spectrum at longer wavelength into the near-IR extending up to 1280 nm, compared to GaAs solar cell response.**

Keywords—**InAs quantum dots, molecular beam epitaxy, intermediate- band solar cell, spacer layer and In/Ga intermixing.**

I. INTRODUCTION

Solar energy is widely considered to be one of the most promising energy sources. However, compared with conventional energy sources, solar energy is presently inferior because of the restrictions of conventional solar cells. Thus attention should be given to the development of new approaches to improve solar cell, (IBSC) based on quantum dots (QD) is one of the most promising methods to achieve this [1]. This intermediate band solar cell comprised of an ideal 3-dimensional semiconductor quantum dot (QD) super-lattice incorporated in the active region of a p-i-n structure has attracted intense research [2]. However the properties of QDs are greatly influenced by their growth conditions such as growth rate, fluxes substrate temperature as well as spacer material and thickness [3]. Particularly, realizing such high performance QD photovoltaic devices requires close-coupled QD layers for the formation of an intermediate band (IB). However, the strong strain accumulation caused by a reduced spacer layer thickness between two adjacent QDs can lead to the generation of dislocations. This significantly degrades the crystalline quality of vertically aligned QD structures [4].

In this work, we investigate the optical properties of multilayer-stacked InAs/InGaAs QDs with different GaAs spacer layer thickness. From the results we investigate the different origins and carrier's dynamics of distinctive double peaks and asymmetry shape observed from the five-stacked InAs/InGaAs QDs samples with different GaAs spacer layer. The optical transition and the carrier's behaviours in the i-region were investigated by photoluminescence (PL) and photocurrent spectroscopy.

II. EXPERIMENT

Fig.1 illustrates a schematic diagram of three solar cell samples: S0 homojunction solar cell (no QDs), two QDs solar cells which differ by the GaAs spacer layer thickness and the In composition in the $In_xGa_{1-x}As$ strain compensated layer respectively (S1: 7nm of GaAs spacer layer with X (In) =11% and S2:33nm with 9% of In composition).

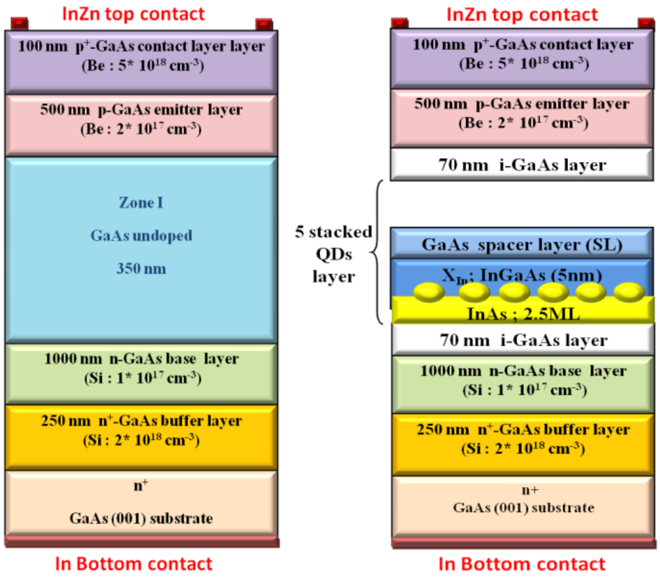

Fig. 1 Schematic layer structure of sample: **(a)** GaAs solar cell (reference), **(b)** quantum dots solar cell (S1 and S2)

The epitaxial layers were grown on epiready n^+- GaAs (100) substrate using solid-source molecular beam epitaxy (SS-MBE) with Riber 32P system. Following oxide desorption at 600°C under As_4 ambient condition, in-situ reflexion High Energy Electron Diffraction (RHEED) was used to observe the surface reconstruction. When a clear (2x4) reconstruction appeared, the temperature was decreased to 520°C to start growth of 250 nm n^+- doped GaAs buffer with a doping density of $2.10^{18}cm^{-3}$ followed by a 1000 nm n-doped GaAs base layer with a doping density $1.10^{17}cm^{-3}$.The substrate temperature is then lowered and stabilized to 500°C for the deposition of the active region. This later consists of 5 stacks $InAs/In_xGa_{1-x}As/GaAs$ QD heterostructures sandwiched by 70 nm intrinsic GaAs layer. For the InAs QD, we deposited normally 2.5(ML) with growth rate of 0.085 ML/s. Since the InAs QDs were formed in the Stranski-Krastanov growth mode, this has led to a wetting layer of about 1.7 thickness and the QDs on top of it. After, a 5nm $In_xGa_{1-x}As$ strain compensated layer and 7 nm and 33nn of GaAs spacer layer respectively for S1 and S2 were deposited. At the end, a 500 nm p-doped GaAs emitter layer ($2.10^{17}cm^{-3}$) followed by a 100 nm GaAs p^+- doped contact layer ($5.10^{18}cm^{-3}$) were grown on top of the structure. Silicon (Si) and Beryllium (Be) were used respectively as n- and p- type dopants. During the growth, the temperature was calibrated by a pyrometer. The growth rate was measured by RHEED specular spot oscillations. A reference sample (S0) of the same p-i-n structure without QD was grown, with a 350 nm-thick i-layer (Fig.1, (a)). The photoluminescence (PL) and the spectral response (SR) measurements were performed to examine the optical transition QDSCs. In PL measurements, an argon ion laser with a wavelength of 514.5 nm was used as an excitation source to generate electron-hole pairs, and its intensity was $150W/cm^2$. The luminescence light from the samples was dispersed by a high-resolution spectrometer and detected by a thermoelectrically cooled InGaAs photo-detector with a built-in amplifier. For the excitation power-dependent and temperature-dependent PL, the samples were mounted in a closed-cycle temperature-controlled helium cryostat. The PL spectra were taken in the nominal output power range of 0.3-300mW and the temperature range 11-300K.

For the spectral response measurements, Indium Zinc (InZn) alloys was used for the top ohmic contact and indium (In) for the bottom contact, without antireflection coating (ARC) layer. The SR measurement was performed at 300K using a photocurrent spectroscopy bench comprising a 100W tungsten halogen lamp, CVI CM110 1/8 monochromator and a kiethley 6485 picoammeter.

III. RESULT AND DISCUSSION

The normalized via the maximum of PL spectra from the two QDs solar cells samples S1 and S2 respectively with 7nm and 33nm of GaAs spacer layer thickness are shown in Fig 2. The normalized PL spectra were measured at low temperature at 11K and an excitation power range from 0.3 mW to 175 mW. The Gaussian fitting lines shapes of each PL spectrum from the active region in the samples at low excitation power. There are two peaks in PL spectra for the two QDs samples at relatively low excitation power. These double-emission peaks are located around 1.12 eV, 1.16 eV and 1.02 eV, 1.04 eV respectively for S1 and S2 samples. With an increase in the excitation power, the overall intensity is enhanced indicating a prominent band filling effect caused by the induced photocarriers [5] and the shape is not significantly changed. The relative intensity between two peaks in PL spectra is the same for S2 sample and changed for S1 sample. We note that the S1 sample exhibits an asymmetrical broadening to high-energy side at 1.18 eV which increase in intensity with increasing the excitation power and with increasing the excitation power, the ground-state emission of L-QDs begins to saturate and then the first excited-state emission (at 1.18eV) of L-QDs increase in intensity with an energy difference of around 60 meV.

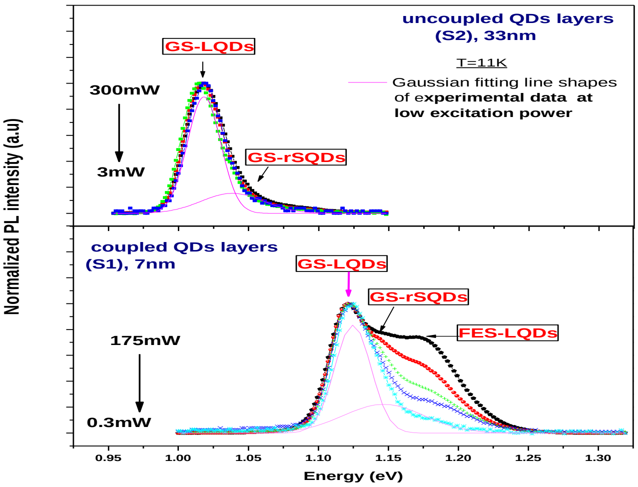

Fig.2 Excitation power-dependent normalized PL spectra from S1 and S2 samples measured at 11K accompanied with the Gaussian fitting line shapes of each PL spectrum from the sample at low excitation power.

This result indicates that these two peaks are largely related respectively to bimodal size distribution of QDs for S2 and to the first excited state which is attributed to larger LQDs located in 1.18eV with a bimodal distribution of QDs in the ground state for S1(LQDs located in 1.12eV and SQDs in 1.16eV).

In addition, we observe that with decreasing the GaAs spacer layer from 33nm to 7nm we note that the FWHM increase by 37%.Also, we note an abnormal blue-shift with 100meV as the spacer thickness decrease and the vertical coupling between QDs layers is more effective which is mainly explained by strain-driven In/Ga intermixing between QDs and spacer layers [6]-[7]-[8]which overcompensates the effects of electronic and structural couplings between QD layers [9]which was attributed to the formation of mini-band caused by the electronic coupling in the vertical direction. Hence, QDs must be intermixed with the adjacent GaAs spacer layers to reduce the total energy, leading to coherently relaxed strain surrounding the QDs. The increase in the band gap energy due to In/Ga intermixing overcompensates for the effect of the size increase and causes a blue-shift, suggesting that the large strain energy, which increases as the thickness of the spacer layer decreases, may drive considerable intermixing between InAs QDs and the GaAs spacer layer.

A supplementary behaviour relevant the strain-driven In/Ga intermixing between QDs and spacer layer can be found in the temperature-dependent PL spectra. Fig.3 (a) and (b) depicts the temperature-dependent PL spectra at 150mWfor S1 and S2 samples.With increasing temperature, all spectra red-shifted to low energies side, which is explained by the dilatation of lattice parameter and electron-phonon interaction effects [10].

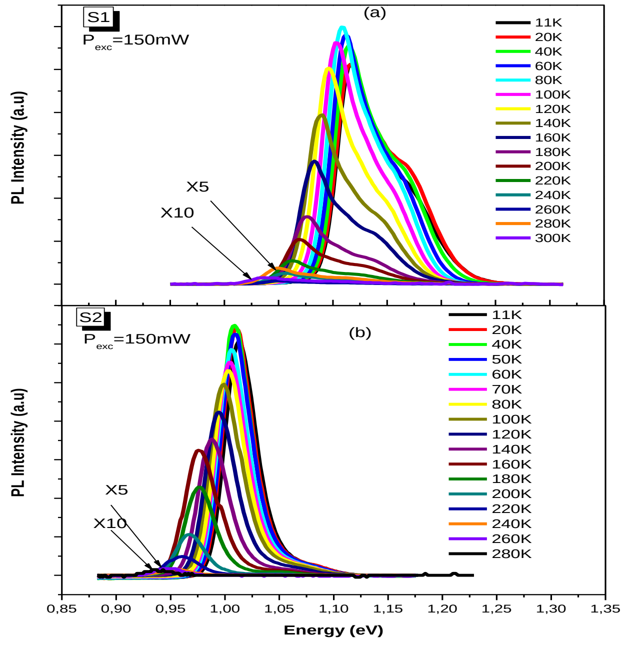

Fig .3 Temperature-dependent PL spectra from (a) the S1 sample and (b) the S2 sample measured at an excitation power of 150mW.

Fig.4 shows the temperature-dependence of the emission peaks for the different-energy side peaks from the PL spectra for the S1 and S2 sample measured at an excitation power of 150 mW. As clearly show in figure 4(a) and 4(b), for the sample S2 all the curves on the low- and high-energy side peaks follow the Varshni equation expressed as

$$E_g\,(T) = E_g\,(0) - \alpha\, T^2/\,(T+\beta) \quad \text{(blue line).}$$

We used the parameter α=3.158 10^{-4} eV/K and the Debye temperature β=93 K for InAs bulk [11]. In contrast, for the sample S1, the comparison between these two fits has shown that for these two QDs family LQDs and SQDs especially for LQDs, α for InAs QDs (3.6 10^{-4} eV/K) is bigger than the corresponding value in the InAs bulk crystal (α=3.158 10^{-4} eV/K). The Debye temperature (β) in InAs QDs (100 K) is also bigger in comparison with bulk InAs (93 K). These both facts suggest the assumption that the material composition of the studied structure has changed due to In/Ga Interdiffusion

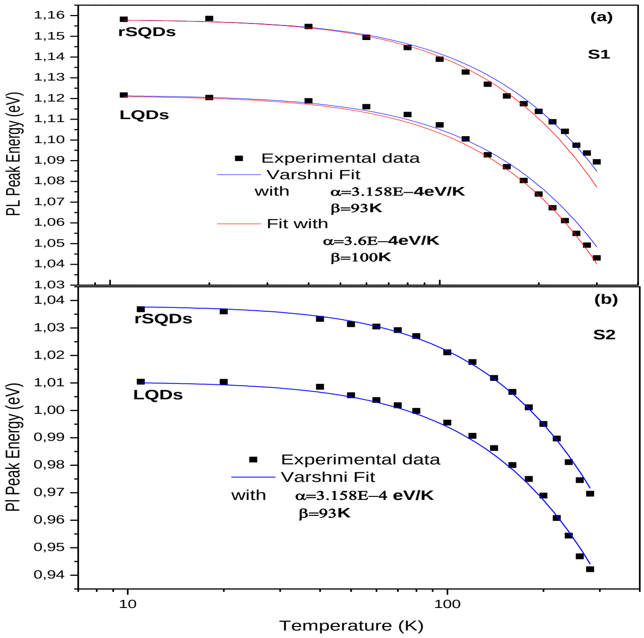

Fig.4 The Temperature-dependence of PL Peak energy for S1(a) and S2 (b) measured under 150mW, The blue line show the Varshni fit of bulk InAs band gap and the red one is a varshni fit with the suitable parameter with give the best correspondence with the experimental data.

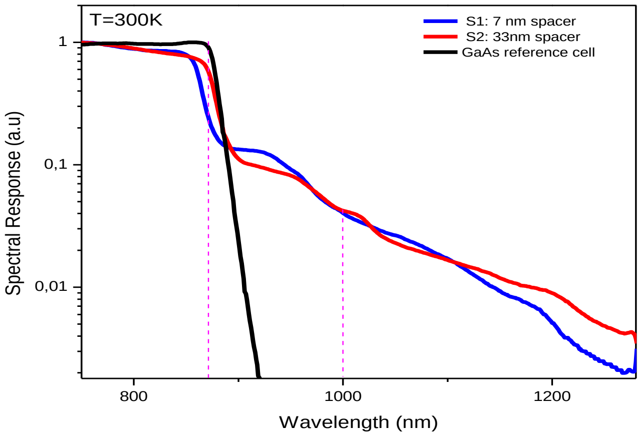

Fig.5 Spectral Response of the GaAs reference solar cell and of the InAs/InGaAs coupled (S1) and uncoupled (S2) QDs.

during growth. This inter-diffusion process creates defects that work as non-radiative center which confirm the increase of FWHM due to the creation of non-radiative defects in the samples and explain the reduction of the spectral response of (S1) in Fig.5.

The spectral response (SR) as a function of wavelength is depicted in Fig.5 which gives further insight into the absorption qualities of solar cells. The spectral response in shorter wavelength below 870nm is mainly the contribution from p-emitter layer, while the peak structure observed around 950nm is from the InGaAs wetting layer [12]-[13].Further, spectral response for above 1000nm is thus solely due the contribution from InAs QDs[14]-[15]. We note that both of IB-QDSCs (S1 and S2) present an extend response up to 1280nm due to the absorption of lower energy photons through the IBQDs. In contrast the response of GaAs reference cell drops sharply at 870nm (1.42eV) indicating that absorption of photons is taking place only above the band gap GaAs. Moreover, S1 has a lower spectral response than S2 in the range of QDs absorption, due to the generation of non-radiative centers as the coupling between the QDs is more effective. Also due to the inter-diffusion process which creates defects that work as non-radiative center which explain the reduction of the spectral response.

IV. CONCLUSION

In summary, we have successfully fabricated GaAs (001)-based SC with a 5 multistacked InAs QDs by capping an InGaAs layer on the QDs and GaAs spacer layer. The optical properties QDs structures with various GaAs spacer thickness were investigated. The PL measurements demonstrate a significant blue-shift (100meV) as the GaAs spacer thickness decreased (7nm), because of the strain-driven intermixing of In in QDs and Ga in the GaAs spacer layer which is gradually increased in the growth direction. The spectral response measurement of InAs QDs SC demonstrates an extended photoresponse up to 1280 nm due to the absorption of low-energy photon by the QDs. Also the SR measurements shows that the spectral response increase with increasing the In composition in the InGaAs absorption range and decreases as the coupling QDs is more effective in the QDs absorption domain.

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