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Surface photovoltage spectroscopy and photoluminescence study of vertically stacked self-assembled InAs/GaAs quantum dots

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Abstract — **We present a study of two 30-layer stacks of self-assembled InAs/GaAs quantum dots with different spacer layer (SL) thickness using surface photovoltage spectroscopy (SPS) and photoluminescence (PL) at room temperature. Both PL and SPS spectra of stacked QDs structure with a thinner spacer layer in comparison to other structures show additional feature. QD features are more clearly visible in SPS spectra and show more features in comparison to PL ones. This study demonstrates the considerable potential of SPS and PL for the contactless and nondestructive characterization of QDs structures at room temperature.**

Index Terms — **Optical spectroscopy, photoluminescence, quantum dots, semiconductor devices, surface photovoltage.**

I. INTRODUCTION

Multilayer quantum dot (QD) structures are of growing interest in the last few years. It has been reported that when two or more layers of self-assembled QDs are deposited under appropriate growth conditions, the QDs develop in self-aligned columns where the electronic coupling between the dots is possible if a separating layer is thin enough [1,2].

The use of vertically stacked QDs is very important for optimizing the light emitting diodes [3] and laser devices [4] performance. Usually a single sheet of quantum dots is insufficient for laser applications and thus a larger total volume of stacked dots in the active region of the laser is required for a higher modal gain [5].

The QD energy levels previously have been investigated using PL, photoluminescence excitation, photocurrent, capacitance, resonant tunneling techniques [6], but the information gained is often limited to only lower energy states, which does not allow a precise deduction of the shape of the QD potential or of the strength of the electronic coupling in stacked structures. Such information can be obtained using electromodulation techniques [7,8] or surface photovoltage spectroscopy (SPS) [9]. SPS is like electromodulation techniques (e.g. photoreflectance –PR and contactless electroreflectance –CER), being nondestructive and contactless, but possesses some advantages over PR and CER. For instance, there are no below bandgap oscillations originating from the doped substrates [10,11] and in which case, the PR or CER techniques are not useful.

In this paper we present an investigation of vertically stacked InAs/GaAs QD systems with different spacer layer thickness using room temperature SPS and PL techniques. In comparison to PL spectra SPS spectra show more QD related features and features are clearly resolved. The PL and SPS spectra for structure with the thinner GaAs spacer layer show an additional feature at energy higher than QD ground state energy comparing with spectra of the other structures. For the presence of this additional feature may be responsible coupling effect between QDs in neighboring layers. Both PL and SPS spectra show that QD size distribution is very similar in all samples. We found very good agreement between transition energies of features present in PL and SPS spectra. These experimental observations will be analyzed and discussed.

II. EXPERIMENTAL

The QD samples were grown on n^+ (100) GaAs substrate by solid source MBE on Riber Epineat machine. An undoped GaAs buffer layer was followed by 30 nm AlAs layer, 60 nm GaAs cladding layer and 30 layers of 2.6 ML of InAs separated by GaAs spacer layers (SLs). QDs were covered by 60 nm GaAs cladding layer and 30 nm AlAs layer. All structure was covered by 50 nm of GaAs cap. Under investigations were two samples with

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SLs width of 30 nm and 20 nm, respectively. As a reference the structure with only one QD layer was used.

All measurements were performed at room temperature. PL measurements were made using 532nm line of Nd-YAG laser and the signals detected by a thermoelectrically cooled InGaAs diode. The SPS measurements, which used normalized incident light intensity [12], were performed at normal incidence using a fixed grid and probe light chopped at 200 Hz [13].

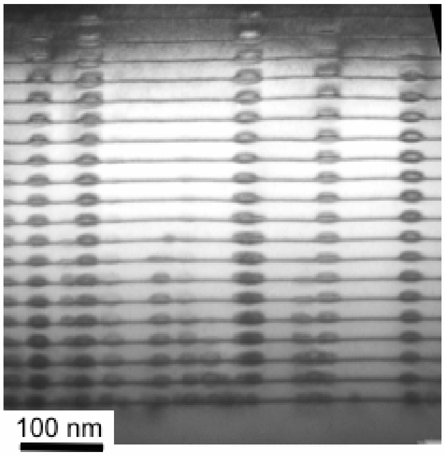

Fig. 1. Cross-sectional TEM image of InAs/GaAs QDs with 20 nm GaAs spacer layer.

Figure 1 is cross-sectional transmission electron microscope (TEM) bright-field image of structure (lower 20 stacks) with QDs separated by 20 nm GaAs SLs. We can see good stacked one over another well formed lens-shaped QDs with similar size. In the case of Fig. 1 the estimated QD sheet density is 10^{10} cm^{-2}.

III. RESULTS AND DISCUSSION

In Fig. 2 we show the PL spectra of structure with one QD layer (reference) and two structures with 30 layers of vertically stacked InAs/GaAs QDs with different SL. Arrows indicate the QD related transition energies obtained from the fitting procedure. The best results were found for fitting to experimental data with Gaussian profile with temperature dependent Bolzmann distribution factor [14]. Obtained from the fitting QD related transition energy values are listed in Table 1. For the reference structure and for the structure with SL = 30 nm we can see two peaks. The lower energy peak described as QD1 corresponds to the ground state quantum dot transition and the second described as QD2 is first excited state. The PL spectrum of structure with SL = 20 nm exhibits an additional feature named QDA and localized in-between QD1 and QD2 features.

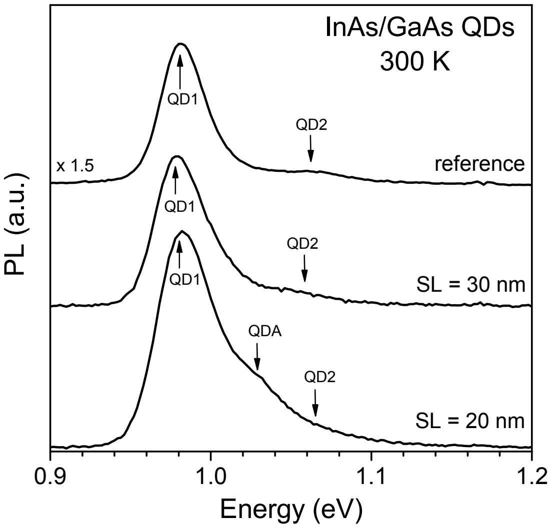

Fig. 2. PL spectra of 30 layers vertically stacked InAs/GaAs QDs with different SL and reference structure with one QD layer. Arrows show QD related transition energies obtained from the fit.

Both QD1 and QD2 peaks for all samples are localized almost in the same energy positions. The FWHM value changes from 32 meV for reference sample to 39 and 44 meV for sample with SL = 30 nm and 20 nm, respectively. All above proves that for all structures QDs has very similar size distribution. As illustrated in Fig. 2, the most prominent feature is related to the fundamental QD1 transition for structure with SL = 20 nm. This is probably because for that structure a higher number of QDs is involved in the PL process. The smallest intensity of QD1 peak demonstrates the spectrum for reference sample, what is obvious because there is only one QD layer in this structure.

Room temperature surface photovoltage spectra of all QD structures being investigated are shown in Fig. 3. The highest energy region is marked GaAs which indicates the region of absorption edge in GaAs and immediately below the GaAs edge is the region of optical transitions originating from the InAs wetting layer and labeled WL.

In order to determine the values of the QD related transition energies we fit the SPS data with the Gaussian line shape [9]. The arrows in Fig. 3 indicate the QD transition related energies obtained from the fitting procedure. Energy values are listed in Table 1.

As an example of fitting to SPS data in Fig. 4 we can see the decomposition (dotted lines) of SPS spectrum for QD structure with GaAs SL = 20 nm into four Gaussian lines.

TABLE 1
THE TRANSITION ENERGY VALUES OBTAINED FROM FITTING TO PL AND SPS DATA

Transition	Transition energy (eV)					
	Reference sample		SL = 30 nm		SL = 20 nm	
	PL	SPS	PL	SPS	PL	SPS
QD1	0.981 ± 2	0.994 ± 2	0.978 ± 2	0.995 ± 2	0.980 ± 2	0.990 ± 2
QDA	–	–	–	–	1.029 ± 2	1.034 ± 2
QD2	1.062 ± 3	1.080 ± 3	1.060 ± 3	1.075 ± 3	1.068 ± 3	1.087 ± 3
QD3	–	1.162 ± 3	–	1.163 ± 3	–	1.156 ± 3

In comparison with the PL data, SPS spectra showed in Fig. 3 present more QD related transitions. For the reference structure and structure with SL = 30 nm we can clearly observe three features denoted as QD1, QD2 and QD3. In the SPS spectrum for structure with GaAs spacer layer width of 30 nm we can observe that QD1 and QD2 transitions are broadened comparing to the reference sample, where the line shape is excitonic. Like in PL the SPS spectrum for structure with SL = 20 nm shows an additional feature labeled QDA at energy between the ground and first excited quantum dot states. The transition energies of QD ground state and excited states are almost unchanged comparing spectra for all structures. The same behavior was observed for PL spectra and proves again that QD size distribution in all the samples is very similar.

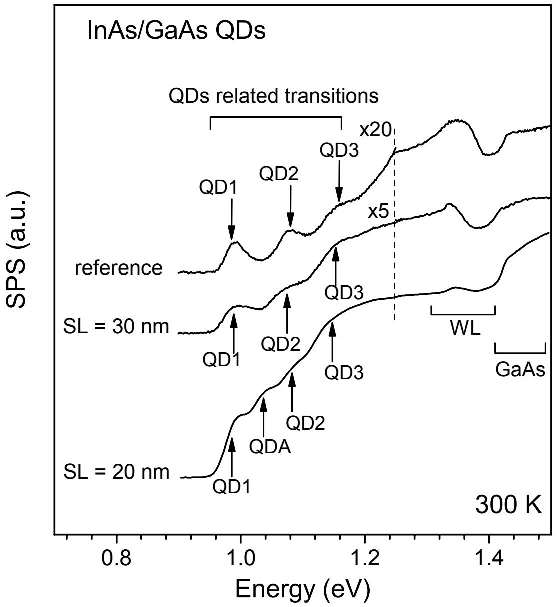

Fig. 3. Room temperature SPS spectra of all QD structures. Arrows indicates the QD transition related energies obtained from the fitting procedure to SPS data.

In Fig. 4 we can see the comparison of PL and SPS for structure with SL = 20 nm. On the SPS spectrum, corresponding to the structure with GaAs spacer layer width of 20 nm, all transitions are clearly separated – step like spectrum, in comparison with PL data where e.g. QD2 peak intensity is very small and hard to localize without fitting procedure.

The splitting between QDA and QD1 obtained from SPS spectrum is 44±4 meV and agree very well with value obtained from the PL spectrum which is 49±4 meV. This splitting seems to be larger than expected comparing to that of reference [8]. The reason can be understood by noting that the coupling induced effects depend on the effective separation of the QDs, which depends on the relative height of QDs and SL. Comparing the transition energies obtained from the fitting procedures to PL and SPS data shown in Table 1 we can find that all QD related transition energies obtained from PL are red shifted about 10 meV comparing to those taken from SPS.

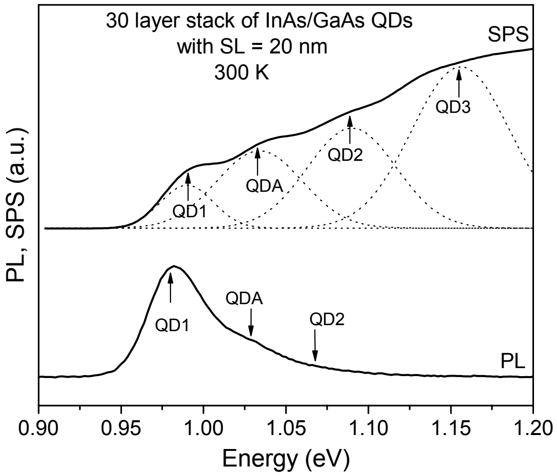

Fig. 4. The comparison of PL and SPS for vertically stacked QD structure with GaAs SL = 20 nm. With dotted lines is shown the decomposition of SPS spectrum into four Gaussian peaks.

IV. CONCLUSION

In this paper we have presented a study of two 30 layers vertically stacked InAs/GaAs QD systems with different spacer layer thickness. To identify the coupling effects between the stacked QDs we have used a structure with one QD layer as a reference. All SPS and PL measurements have been realized at room temperature. In comparison with the PL spectra, the SPS spectra show more QD related features and features are clearly resolved. The SPS spectra for structure with SL = 20 nm in comparing with other structures show an additional feature which can be related to the excited state due to the coupling between QDs from neighboring layers. The accurate locations of the transition energies can be determined from the Gaussian line shape fit. The splitting energy between QD ground state transition QD1 and QDA feature is the same at PL and SPS. The ground state and excited states related transition energies has almost the same values for all structures what proves very similar QD size distribution for all samples. This study demonstrates the considerable potential of surface photovoltage spectroscopy for the contactless and nondestructive characterization of QD structures at room temperature.

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