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Using room-temperature surface photovoltage spectroscopy, we have characterized several 30-layer stacked self-assembled InAs/GaAs quantum dot (QD) structures with different spacer layer (SL) thicknesses. Signals from every relevant portion of the samples, including QDs, wetting layer, and GaAs barrier have been observed. The strain-induced field for thinner SL is responsible for a significant modification of the band structure, possibly resulting in the appearance of an additional excited state lying higher than the second excited QD state. A peculiar feature below the fundamental transition is tentatively attributed to the optical absorption from uncoupled dots of which the density is significantly lower than that of vertically coupled ones. The spectra show blueshifted features with a decrease of the SL thickness, indicating that the materials intermixing between InAs QDs and GaAs SL are strongly driven by strain. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2221402]

Recently, a great deal of attention has been focused on the investigation of three dimensional spatial confinement of carriers in quantum dot (QD) heterostructures. Such structures are of growing interest from the viewpoint of fundamental physics and are very promising for optimizing the light emitting diode[1] and laser device performance.[2] The application of QDs as the active region in a semiconductor laser should lead to ultralow threshold current and extremely high thermal stability because of its delta functionlike density of states. Electronic vertically coupled QD (EVCQD) structures have been extensively studied with the aim of increasing active volume,[3] reducing the emission linewidth, and shifting the emission wavelength to longer wavelength.[4] Although it is possible to grow multilayer EVCQD samples, a complicated situation where the structural and electronic couplings correlate closely with spacer layer (SL) thickness may arise. A full understanding and control of it is critical for design and performance of devices.

Optical properties of InAs/GaAs EVCQD structures have already been studied mainly through photoluminescence[5–8] (PL) and photoluminescence excitation (PLE) measurements.[9,10] PL typically offers information only about the lower lying quantum states, and little on the position of the quantum level relative to the band structure of the host matrix. PLE is an efficient technique, but it requires the use of a tunable laser and is laborious to achieve. The surface photovoltage spectroscopy (SPS) has been shown to be a powerful technique, which is simple, contactless, and nodestructive.[11] It can be performed in a great variety of materials and at any reasonable temperature (including room temperature). There are some reports available in literature on QD structures using SPS,[12–14] but less on multilayer EVCQD structures, particularly on the investigation of SL thickness on the electronic characterization of EVCQDs using SPS not yet found elsewhere.

The layer structures used in this study were grown by solid source molecular beam epitaxy (SSMBE) on (001) n^+ GaAs substrate in Riber Epineat machine. An undoped GaAs buffer was followed by 30 nm AlAs layer, 60 nm GaAs cladding layer, and 30 layers of 2.6 ML (mono layers) of InAs separated by GaAs SL. QDs were covered by 60 nm GaAs cladding layer and 30 nm AlAs layer. All structures were covered by 50 nm of GaAs cap. Under investigations were four samples with SL widths of 30, 20, 15, and 10 nm. As a reference the structure with only one QD layer was used for the purpose of comparison.

The apparatus used in SPS measurements was described in detail in literature.[15] The monochromatic light chopped at 200 Hz was impinged uniformly on the sample with the light density of $\sim 10^{-5}$ W/cm^2. Owing to the pinning of the Fermi level at the surface, a built-in electric field exists across the undoped layer and QDs. The optically excited carriers were separated by the electric field and produced a periodic photovoltage variation with the same frequency as the chopped light. Pulses of photovoltage induced on the electrodes were detected by a lock-in amplifier. All measurements were performed at room temperature.

Figure 1 shows the SPS spectra of the four EVCQD samples and a reference sample with only one QD layer at room temperature as a function of photon energy. On the higher energy side near 1.43 eV, a steep absorption edge corresponding to the band edge absorption of GaAs barrier is observed. Below the GaAs edge is the optical transition from the InAs wetting layer labeled as WL. Three QD related features denoted as QD1, QD2, and QD3 are clearly visible

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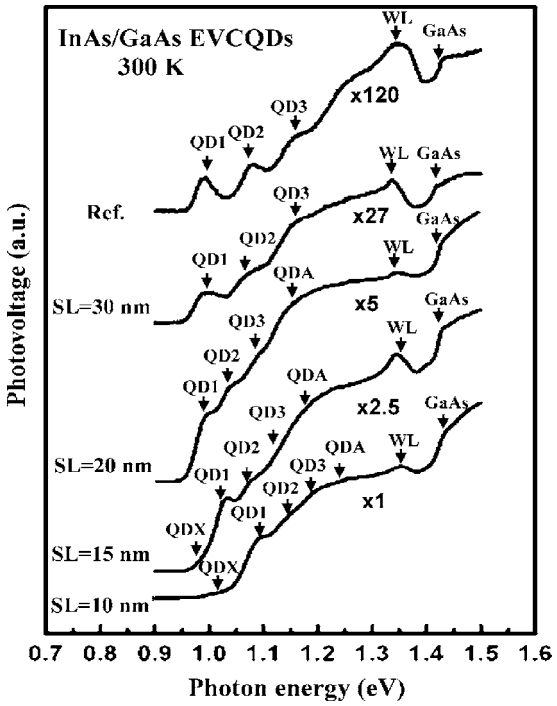

FIG. 1. Room-temperature SPS spectra of 30-layer vertically stacked InAs/GaAs QDs with different SLs and the reference structure with one QD layer. Arrows show QD related transition energies determined from the fitting.

in the reference sample, reflecting the higher homogeneity of the single dot layer. The energy positions of all transition peaks for the 30 nm SL sample are nearly identical to those of the reference sample since the thick separation prevents any electronic coupling of neighboring layers. The line shape of the QD emission for the 30 nm SL sample is more energetically broadened than that for the reference sample. Energy level variation in stacked dot layers is mainly caused by dot size fluctuations. As illustrated in Fig. 1, the most prominent feature, QD1, related to the fundamental transition for EVCQD structures is remarkably larger in intensity than the corresponding one for the reference sample. The higher absorption strength for EVCQD structures indicates the increase of optically absorbing QD volume and the effectiveness of the vertically aligned EVCQD system. Moreover, the increase of QD1 intensity with the decreasing thickness of SL is exhibited. The working characteristic of SPS is a two-step process, involving both absorption and carrier escape from the dots. For thinner SL the better transport behavior is achieved due to essentially overlapped wave functions between neighboring dots, thus yielding the greater SPS signal. A new signal labeled QDA at energy lying above QD3 arises for SL thickness less than 30 nm. The structural coupling for thinner SL complicates the strain field distribution in and around closely stacked QDs, and has a significant modification of the band structure, possibly giving rise to the presence of QDA.[16] An extraordinary feature, QDX, occurring at energy below QD1 for 10 and 15 nm SL samples is seen in SPS spectra. QDX is not treated as the fundamental transition of EVCQDs; otherwise a significant signal should exist there. The prominent feature lying above QDX is therefore attributed to QD1. From the observations that the intensity of QDX is much weaker than that of QD1 and that there is a closer energy difference between QDX and QD1 of the reference sample, it is presumed that QDX is connected with some uncoupled dots which occupy a smaller quantity of dots than coupled ones in the stacked columns where QD1 dominates.

TABLE I. Transition energies (in eV) of EVCQD structures for various SL thickness (in nanometers) and the reference sample determined from the SPS.

Sample	QD1	QD2	QD3	QDA	QDX
Reference	0.994	1.080	1.162	···	···
SL=30	0.995	1.075	1.163	···	···
SL=20	0.990	1.034	1.087	1.156	···
SL=15	1.025	1.078	1.116	1.176	0.985
SL=10	1.092	1.144	1.188	1.241	1.019

In order to determine the values of QD related transitions we have fitted the SPS data with a Gaussian line shape.[13] The arrows in Fig. 1 indicate the QD related transition energies obtained from the fitting procedure. The energy values are listed in Table I. As an example of fitting to the SPS data in Fig. 2, the decomposition of a SPS spectrum for the 10 nm SL sample into five Gaussian lines is illustrated. A satisfactory fit obtained by using a Gaussian line shape indicates that the broadening is inhomogeneous and is probably due to fluctuations in the QD size.

Note that the QD signals are redshifted as SL thickness is reduced from 30 to 20 nm and then blueshifted significantly below 20 nm. Previous reports on EVCQDs mainly demonstrated a redshift[12,17] but a few blueshift of QD transitions.[18] It is generally deduced that the smaller the SL width is, the stronger the interacting strain field resulting in the stronger vertical electronic coupling is. Moreover, high strain field will favor the formation of large QDs due to an increased elastic strain relaxation. These will result in an energetic redshift, which contradicts with our observation. To explain this anomaly, another effect must be taken into account. We have tentatively suggested that the blueshift is associated with the strain driven material intermixing.[17] The overall strain field is superimposed by the individual field from the constituent dot, which enhances the total strain energy during overgrowth. Therefore QDs have to intermix with GaAs SL to reduce the total energy, and this leads to a coherently relaxed strain surrounding QDs. The increase of

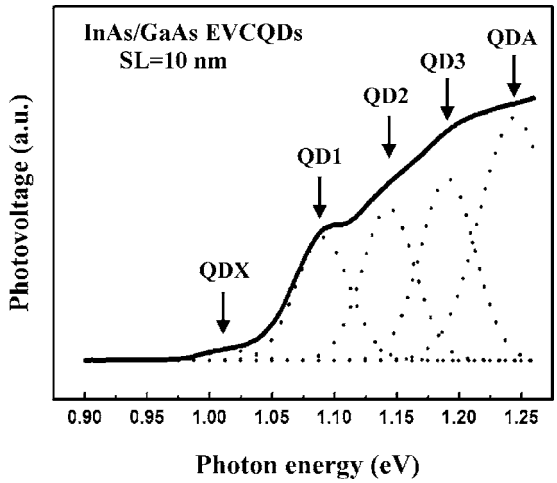

FIG. 2. The SPS spectrum for the InAs/GaAs EVCQD structure with GaAs SL thickness of 10 nm. The dotted lines show the deconvolution of the SPS spectrum into five Gaussian features.

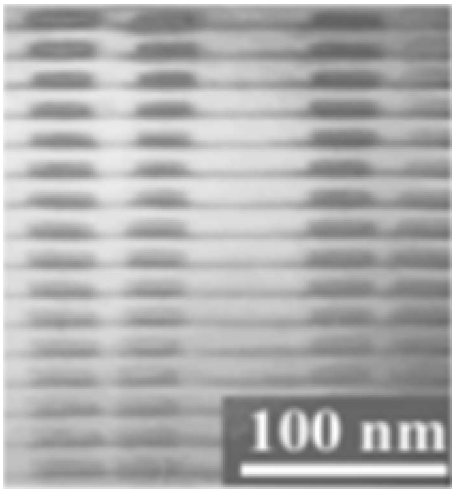

FIG. 3. The TEM image of the EVCQD structure with 15 nm SL thickness reveals very good vertical alignment without any dislocation. Besides, the image displays a strain field distribution of rectangularlike shape for each QD instead of a conventional pyramid shape, revealing the occurrence of material intermixing.

the Ga content overcompensates for the size increase of QDs and leads to a blueshift due to the increment of band gap energy. A possible clue to material intermixing is revealed in the transmission electron microscopic (TEM) image of the 15 nm SL sample as depicted in Fig. 3 where a strain field distribution leads to the formation of rectangular shaped QDs instead of conventional pyramid shaped ones. This stems from the strain reduction of the top of QDs through the intermixing of In atoms of InAs QDs and Ga atoms of GaAs SL. It means that the large strain energy may serve as a driving force for substantial intermixing between InAs QDs and GaAs SL and becomes more pronounced as the number of deposited dot layers increases with thinner SL.

As noted earlier, the SPS spectrum is determined by the following two factors: (1) the optical absorption in QDs and (2) the escape of carriers from QDs and their separation in the electric field. Although the change of QD shape would influence the absorption strength, the major contribution on SPS comes from the second factor for reasons stated below. The intensity of QD1 for thinner SL samples is dramatically larger than that for 30 nm SL one. TEM of thinner SL samples reveals a minor increase of dot size, which is unable to explain such intensity relation. For thinner SL, the raised states caused by material intermixing are expected to have higher probability for carriers to escape from QDs. Besides, the electronic coupling between QDs facilitates carrier transport, thus resulting in greater SPS intensity.

In conclusion, we have studied InAs/GaAs EVCQD system with varying SL thickness using the room-temperature SPS technique. QD transitions, two dimensional wetting layer, and GaAs barrier are well resolved in the SPS spectra. The SPS spectra for EVCQD structures with thin SL in comparison with other structures show an additional feature lying above the second excited QD state due to the modified band structure caused by the strain-induced field. An extraordinary feature below the fundamental transition is probably associated with the ground state transition from uncoupled dots of which the density is significantly lower than that of vertically coupled ones. With the decrease of SL thickness, the spectrum exhibits a blueshift, indicating that the material intermixing is strongly driven by strain.

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