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Investigation of the energy spectra and the electron–hole alignment of the InAs/GaAs quantum dots with an ultrathin cap layer

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ABSTRACT

The effects of indium composition and the thickness of the combined InGaAs/GaAs thin cap layer on the energy spectra and relative electron–hole alignment of InAs quantum dots (QDs) grown by metal organic vapor phase epitaxy (MOVPE) are investigated by photoelectrical spectroscopy in a semiconductor/electrolyte system. In structures with InAs QDs and an InGaAs strain reducing layer, the shift of the hole's wave function to the QDs' top was revealed, which indicates In enrichment of the area near the top of QD'. In structures with an ultrathin GaAs cap layer a change of the sign of the built-in dipole moment was observed. This is explained by coupling effects of quantum-confined electrons with surface states.

1. Introduction

The high probability of a radiative recombination in the quantum dot (QD) nanostructures based on A3B5 direct band gap semiconductors, in particular the InAs/GaAs QDs, has attracted tremendous interest among researchers during the last few decades. Due to the possibility of obtaining optical emissions of a wide wavelength range from 1.0 to 1.8 μm [1], covering the main transparency windows of quartz optical fiber (1.3 μm and 1.55 μm), the interest in studying this system has become keener. One of the approaches to tuning the QDs' ground state transition energy is to vary the thickness and composition of the cap layer. Several factors concerning the influence of the cap layer parameters on the QDs' energy spectrum are known from the literature. When the InAs QDs layer is covered first with an $In_xGa_{1-x}As$ quantum well (QW) layer and then with a GaAs cap layer a redshift of the QDs' emission energy occurs for two reasons. The first reason is the dissolution of the QW material, stimulated by the presence of InAs clusters, thereby inducing the formation of an indium enrichment area near the top of QD' [2], which further leads to the augmentation of the QDs' effective height. The second reason is the relaxation of elastic strains in the QDs, induced by reduction of the lattice mismatch on the QW/QDs interface [3,4]. When the thickness of the GaAs cap layer is less than 20 nm, a redshift of the emission occurs, which is induced by a decrease of elastic strains in the thin cap layer [5]. Furthermore, the authors of the work [6] pointed out that in the InAs/GaAs QD's nanostructures where the thickness of the cap layer is small (< 10 nm) there is a variation of the confining potential barrier height from the side of the QDs' top, due to the change of elastic strains in the cap layer, thereby inducing a variation of the band gap. With smaller GaAs cap layer thickness (< 5 nm) a blueshift of the emission was observed [6], which can be explained by an increase in the potential barrier from the side of the QDs' top while approaching the vacuum level.

When the cap layer thickness is small enough to allow tunneling, an interaction between the quantum-confined states and the localized surface states becomes possible [7,8], which should also provoke a variation of the QDs' energy spectrum.

All the above-mentioned factors should affect not only the QDs' energy spectrum, but the electron–hole spatial alignment [9] as well, and the latter one can be analyzed by the quantum-confined Stark effect [10]. The dipole moment defined by the electron–hole vertical separation is an important characteristic of QDs which can be obtained experimentally. It is sensitive to the shape and the composition gradient of the QDs, whereas the ground state transition energy mainly depends on the QDs' mean size and composition. The work [11] demonstrated the high sensitivity of the

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built-in dipole moment to confining potential profile features in the growth direction.

Therefore, herein we are examining the influence of the cap layer parameters on the QDs' electronic structure by investigating both the energy spectrum and the quantum-confined Stark effect, which also allows us to determine the factors affecting the QDs' energy spectrum variation in each case.

2. Experimental techniques

This study investigates nanostructures with single InAs QDs layers and with combined InGaAs/InAs QW/QDs layers on the GaAs substrate. The samples were grown by metal organic vapor phase epitaxy (MOVPE) at the atmospheric pressure of hydrogen. Initially, a 0.6 μm thick n-GaAs buffer layer with the electron density of $\sim 10^{16}$ cm^{-3} on (100) GaAs substrate was formed at 600 °C. Subsequently, an InAs QDs layer and a GaAs cap layer were deposited at 520 °C. In samples with the combined QW/QDs layers the $In_xGa_{1-x}As$ QW ($x=0.2$, 0.3) layer with the thickness (L_{QW}) from 2 to 12 nm was formed prior to deposition of the cap layer. The thickness of the GaAs cap layer (d_{GaAs}) varied from 3 to 30 nm. The InAs layer was doped with bismuth in order to increase the QDs' uniformity [12]. On the surface of some investigated structures a ~ 1 mm^2 circle area semi-transparent rectifying Au Schottky contact was deposited by thermal evaporation in vacuum at 100 °C. Ohmic contacts to the buffer layer and the substrate were formed by burning in tin foil using electric sparks.

The photosensitivity spectra of the nanostructures with the Schottky barrier and the spectra of the structures with the semiconductor/electrolyte contact were investigated at 300 K. An MDR-2 monochromator equipped with a 100 W tungsten halogen lamp and a grating of 300 g/mm operating in a range from 780 to 2530 nm was used as a monochromatic light source. The dispersion of the system was 4 nm/mm, which gave a resolution of 3 meV with slit width of 1 mm with the energy $h\nu=1$ eV. Measurements were performed on an alternating signal. A mechanical optical chopper operating at 130 Hz frequency was used. Detection of the alternating electrical signal was carried out according to the standard lock-in technique using the Stanford Research Systems 510 selective amplifier.

The spectral dependence of the relative photosensitivity $S_{ph}(h\nu)=I_{ph}(h\nu)/L(h\nu)$ was plotted, where $I_{ph}(h\nu)$ stands for a photo current, and $L(h\nu)$ stands for the light intensity (in arbitrary units).

A mixture of 0.5 M KCl solution and glycerin (1:1) was used as an electrolyte. The structures were photo-excited from the bottom side in order to avoid the influence of light absorption by electrolyte, which is essential at energies less than 1 eV. To change the electric field strength, the external bias voltage of the inverse polarity (+ to the heterostructure of n-type) was applied to the semiconductor/electrolyte junction. After each measurement of the Stark effect at certain applied voltages a reproducibility test was performed to prevent irreversible changes of the photosensitivity spectrum, related to the photoelectrochemical activity of the electrolyte.

The electric field strength in the QDs layer was estimated from the surface barrier height ψ_0, shallow donor concentration in the buffer layer i_d and the value of the external applied bias U by the following equation, using the standard Schottky barrier model with the assumption of fully ionized impurities:

$$F(U)=\frac{eN_d}{\epsilon_r\epsilon_0}\left[\sqrt{2\epsilon_r\epsilon_0(\psi_0-U)/eN_d}-d_c\right], \quad (1)$$

where, e is the electron's charge, ϵ_r is the dielectric permittivity of GaAs, ϵ_0 is the vacuum permittivity, $d_c=d_{GaAs}+L_{QW}$ is the resulting cap layer.

3. Results and discussion

In this research the photoelectric spectroscopy was used on the semiconductor/electrolyte junction to study the properties of produced nanostructures [13]. This method allows the researcher to obtain photoelectrical spectra as well as to study the quantum-confined Stark effect on structures with QDs, covered by an ultrathin cap layer. Along with the semiconductor/electrolyte junction, in some cases the semiconductor/metal junction was used to explore photosensitivity spectra. Fig. 1 shows photosensitivity spectra obtained both on the semiconductor/metal and the semiconductor/electrolyte junctions. The measured structures differed in the thickness of the combined cap layer, consisting of InGaAs QW and GaAs layers. For structures with resulting cap layer thickness d_c of 13 and 17 nm, the spectra obtained on both junctions were similar (Fig. 1). In the case of $d_c=10$ nm the photocurrent measured at the energy of the QDs' ground state transition on the semiconductor/metal junction was drastically lower compared to the one measured on the semiconductor/electrolyte junctions. At $d_c=7$ nm the photocurrent obtained on the latter one was still visible, whereas the signal measured on the semiconductor/metal junction disappeared. This effect can be explained as follows. When the cap layer thickness is small enough to allow tunneling, the emission of the excited electrons from the QDs' ground state quantum-confined level (E_{e0}) to the semiconductor volume (inset in the Fig. 1, transition 2), which is responsible for the photocurrent can no longer compete with more feasible emission to the metal (inset in the Fig. 1, transition 1). It does not occur on the semiconductor/electrolyte junction due to the lack of a high density quasi-continuous spectrum of the allowed energy levels in the electrolyte. The drop of the QDs' photocurrent value on the semiconductor/metal junction can be estimated quantitatively by comparing the resulting emission lifetime of electrons [14] with the tunneling time into the metal (τ_{e0}^{tun}). The latter can be calculated by the Wentzel–Kramer–Brillouin approximation for one-dimensional potential [15].

$$\tau_{e0}^{tun}=\left[\frac{\pi\hbar}{2m_eL^2}\exp\left(-\frac{2}{\hbar}\int_0^{d_c}\sqrt{2m_e(E_c(z)-E_{e0})}dz\right)\right]^{-1} \quad (2)$$

where, $\hbar$ is the Planck constant, m_e is the effective mass of the electron in 1D potential, L is effective QD's height, $E_c(z)$ is the conduction band profile, and E_{e0} is the electron ground state energy.

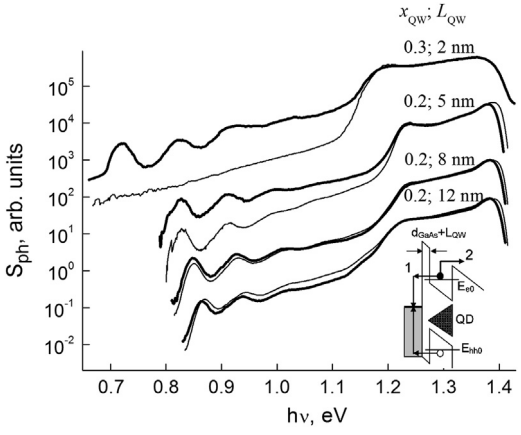

Fig. 1. Comparison of the photosensitivity spectra on the semiconductor/electrolyte junction (bold curves) and on the semiconductor/metal junction (dashed curves) of the structures with $In_xGa_{1-x}As$/InAs combined QW/QDs layer with different thicknesses of the double cap layer ($d_{GaAs}+L_{QW}$). $d_{GaAs}=5$ nm. In the inset: process 1 – tunneling of the electron from the ground state E_{e0} into the metal; process 2 – emission of the electron to the semiconductor's matrix, inducing the photocurrent.