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Temperature-dependent modulated reflectance and photoluminescence of InAs-GaAs and InAs-InGaAs-GaAs quantum dot heterostructures

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Abstract Optical transitions and electronic properties of epitaxial InAs quantum dots (QDs) grown with and without InGaAs strain-relieving capping layer within GaAs/AlAs quantum well (QW) are investigated. Modulated reflectance and photoluminescence (PL) spectroscopy is used to probe the QD- and QW-related interband optical transitions over the temperature range of 3–300 K. The observed spectral features in QDs are identified using numerical calculations in a framework of 8-band $k \cdot p$ method. It is found that covering the dots by a 5 nm-thick InGaAs layer yields the energy red-shift of ground-state transition by ~ 150 meV. Moreover, the analysis of interband transition energy dependence on temperature using Varshni expression shows that material composition of InAs QDs significantly changes due to Ga/In interdiffusion. A comparison of emission- and absorption-type spectroscopy applied for InAs-GaAs QDs indicates a Stokes shift of ~ 0.02 meV above 150 K temperature.

Keywords InAs quantum dots · Modulated reflectance · Photoluminescence · Optical transitions · Electronic structure

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1 Introduction

Semiconductor nanostructures based on quantum dots (QDs), which possess a unique atomic-like density of states, have been studied extensively from fundamental and application perspective. Indeed, self-assembling QDs are particularly attractive for infrared applications in optoelectronics, such as optical-fiber lasers (operating at $1.3\,\mu$m,

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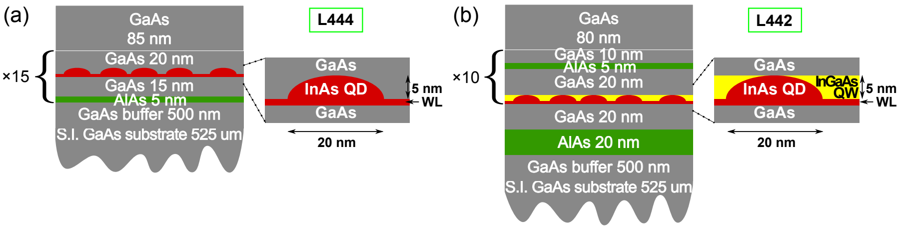

Fig. 1 Full-sample sketches and active region layouts corresponding to the GaAs-capped (a) [sample L444] and InGaAs-capped (b) [sample L442] QD structures. Lens-shaped QDs of 5 nm height, 20 nm base diameter, and $\sim 10^{10}\,\mathrm{cm}^{-2}$ sheet density were intentionally grown. QDs were directly Si-doped at a level corresponding to about one electron per dot.

1.55 μm wavelength) [1] and photodetectors (3–5 μm, 8–12 μm) [2]. In particular, InAs-GaAs quantum dots (QDs) have been used for QD infrared photodetectors (QDIPs), exploiting intraband electronic transitions between bound-state of QD and continuum. In recent years, an advanced InAs-InGaAs dots-in-a-well (DWELL) quantum heterostructure was implemented, where InAs QDs are covered by InGaAs strain-relieving layer (SRL), forming a quantum well (QW) [2]. This new QDIP concept is then based on intraband optical transitions between bound-states of QD and QW, thus allowing a spectrally adaptive optical response through adjusting QD/QW parameters and/or applying a bias voltage. To optimize operation of such novel devices it is essential to know in detail their optical properties and electronic structure over wide temperature range. This information can be obtained by photoluminescence (PL) and modulated reflectance spectroscopy techniques — photoreflectance (PR), contactless electroreflectance (CER), and phototransmittance (PT) [3–10].

This work presents a comparative spectroscopic study of self-assembled InAs QDs grown with and without InGaAs SRL and embedded in GaAs-AlAs QW. Interband optical transitions of these QD structures are studied using modulated reflectance and PL methods over the temperature range of 3-300 K with a particular interest on QD electronic states. Experimental data are then verified by numerical calculations in a framework of 8-band $k \cdot p$ method under fully 3-dimensional approach.

2 Samples and experimental

The structures studied were grown by molecular beam epitaxy on semi-insulating (100) GaAs substrate and compose of InAs QDs (10/15 periods) embedded in 30/35 nm-thick GaAs wells and surrounded by 5 nm-thick AlAs barriers. InAs QD “seed” layer (2.4 monolayers) was covered either by GaAs (sample L444) or by 5-nm thick $\mathrm{In_{0.15}Ga_{0.85}As}$ SRL (sample L442). Thereby, two different QD structures were formed: InAs-GaAs-AlAs (L444) and InAs-InGaAs-GaAs-AlAs (L442) [see Fig. 1].

Spectroscopic measurements were carried out in the temperature range 3–300 K by mounting the samples on the cold finger of a closed-cycle helium cryostat. A diode-pumped solid-state laser emitting a wavelength of 355 nm or 532 nm was used as the modulation source in PR, and as the excitation source in PL. Laser light was modulated by a mechanical chopper ($\sim$ 277 Hz) and the laser power density was varied using neutral density filters and/or an attenuator.

PR spectra were measured using a synchronous double-monochromator system performing with a better signal-to-noise ratio compared to the use of low-pass filter, and allowing simultaneous collection of the PL spectrum [3,10]. Room temperature CER experiments were carried out using a capacitor-like system [5,7,8]. To provide a modulating electrical field, a sine-type AC signal from a high voltage amplifier (Trek 609E-6) of $\sim$ 1.5 kV peak-to-peak amplitude and $\sim$ 190 Hz frequency was applied to the top transparent electrode. A phase-sensitive detection technique was used in all spectroscopic measurements.

3 Spectroscopic results and discussion

Room temperature CER and PL spectra for the GaAs-capped (sample L444) and InGaAs-capped (sample L442) QDs are shown in Fig. 2(a) and (b), respectively. Three major sets of features related to various excitonic transitions can be identified. In the low photon energy range of 0.9–1.25 eV, CER and PL spectra exhibit several broadened features due to QD ground-state (GS) and excited-state (ES) transitions. Further, in the energy range between 1.25–1.42 eV, CER spectrum of sample L444 shows heavy- (11H) and light-hole (11L) related transitions from InAs-GaAs wetting layer (WL) QW. In case of DWELL sample L442, sharp and intensive CER lines correspond to optical transitions in the InAs-InGaAs binary quantum well (bi-QW) formed by InAs WL and InGaAs SRL. The high energy features above the bandgap of GaAs (1.42 eV) are due to interband transitions in GaAs-AlAs QW, which are superimposed by Franz-Keldysh oscillations from the GaAs buffer layer.

To determine transition energies and broadening parameters, first derivative of a Lorentzian-type function was used to fit CER spectra (see fit components in Fig. 2) [4]. Three QD-related optical transitions were estimated for the GaAs-capped structure L444: GS $=$ 1.101 eV, ES1 $=$ 1.162 eV, and ES2 $=$ 1.212 eV. While for the InGaAs-capped structure four QD-related transitions were recorded at GS $=$ 0.953 eV, ES1 $=$ 1.018 eV, ES2 $=$ 1.085 eV, and ES3 $=$ 1.160 eV.

The presence of the InGaAs cap layer induce a red-shift by $\sim$ 150 meV and decrease broadening parameter by 20 meV of GS optical transitions in InAs QDs. This significant energy shift could be explained in terms of reducing strain, suppressing compositional mixing and increasing dot height [11–13].

To verify spectroscopic observations of QD ensembles, we have compared experimental transition energies with numerical calculations in a framework of 8-band $k \cdot p$ method using nextnano^3 software [14]. We have used a uniform In distribution within the dot as an approximation of rather complicated In composition profile [15]. Best agreement with experimental data was obtained assuming a lens-shaped $\mathrm{In_xGa_{1-x}As}$ QD with a significantly reduced In percentage down to $x = 55\%$ (82%) for L444 (L442) structure (see simulated absorption spectra in Fig. 2). Indeed, material composition of GaAs-capped QDs significantly change in a growth process due to Ga/In interdiffusion, while compositional intermixing can be considerably suppressed in InGaAs-capped QDs [11,13].

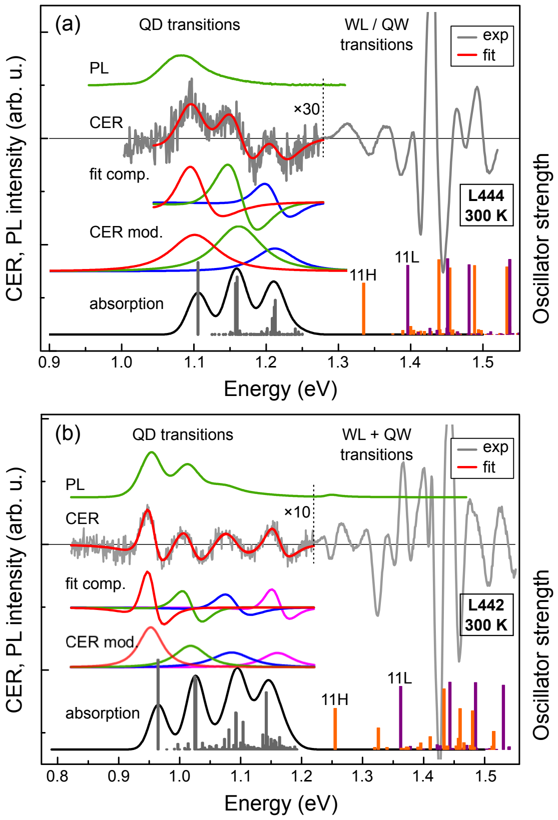

Fig. 2 Room temperature PL and CER spectra of the GaAs-capped (a) [sample L444] and InGaAs-capped (b) [sample L442] QD structures. Fitting components and the corresponding CER moduli are also provided. The position and amplitude of vertical bars for QD- and QW-related features represent calculated optical transition energies and intensities, respectively. QD transition energies and intensities were estimated using 8-band $k \cdot p$ method, while QW features were established in a framework of effective mass approximation. Each individual transition in both simulated QD absorption spectra is broadened by a FWHM of 20 meV. $In_xGa_{1-x}As$ QD parameters used: $x = 0.55$ (0.82), 20 nm base diameter, 4 (7) nm height for L444 (L442) structure, WL = 0.5 nm, 5 nm-thick $In_{0.13}Ga_{0.87}As$ SRL assumed to cover QD with even thickness.

Temperature-dependent PR and PL spectra for as-grown L444 and L442 structures in the photon energy region of QD and QW optical transitions are presented in Fig. 3. The photomodulated signal for as-grown QD structures is actually a superposition of PR and PT signals due to back-surface reflection effect [10]. The blue-shift of PR features expected as the temperature is reduced can be attributed mainly to the bandgap change of the QD/QW materials. While decrease of GS PR signal intensity at low temperature is governed by state-filling effect. Also, the PR/PL linewidth is much narrower for the InGaAs-capped sample, owing to a more uniform size distribution of QDs [9].

The temperature dependence of GS and ES transition energies for both types of QD ensembles obtained from the PR and PL (if observed) spectra as well as numerical calculation results are presented by symbols in Fig. 4. QD optical transition energies were fit according to the Varshni expression, $E(T) = E_0 - \alpha T^2/(T+\beta)$, (lines in Fig. 4) [16] with parameters showing strong InGaAs alloying of InAs dots in agreement with calculation results. The Stokes shift between the emission-like PL and the absorption-like PR spectra seems not to be significant for the InGaAs-capped QDs, while for the GaAs-capped QDs the Stokes shift is apparent (at temperatures

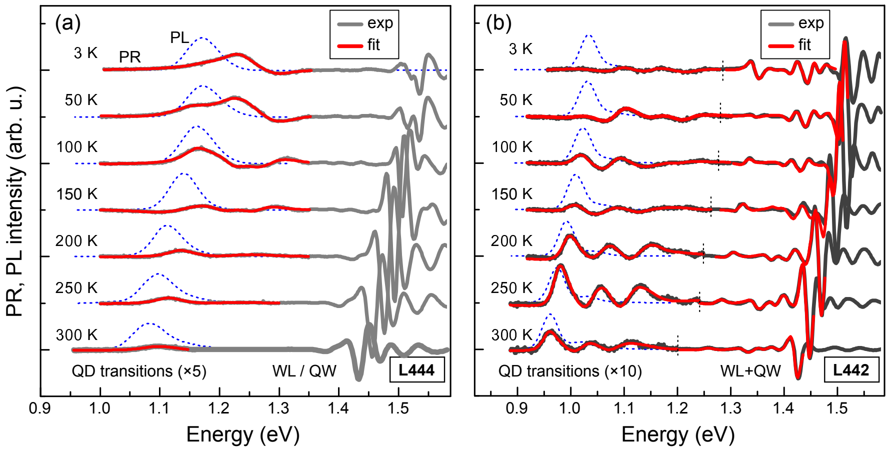

Fig. 3 PR (solid line) and PL (dotted line) spectra for the QD structure without (a) and with (b) InGaAs SRL over the 3–300 K temperature, showing the evolution of optical transitions originating within the InAs QDs and WL-QW (InAs-GaAs QW; sample L444) or bi-QW (InGaAs-GaAs QW; sample L442).

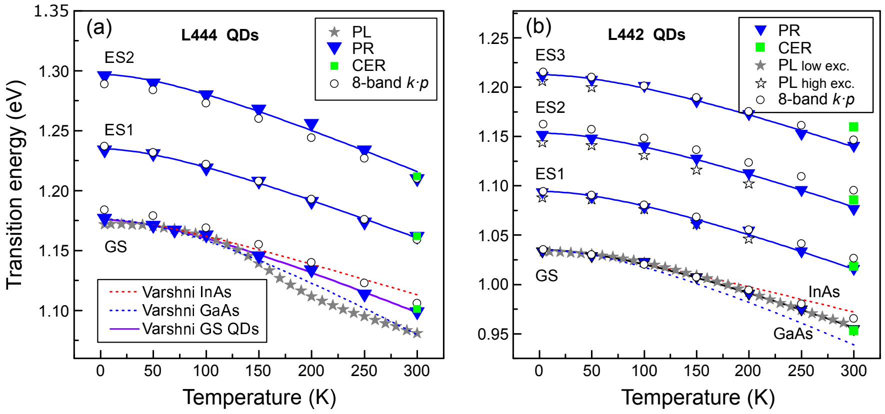

Fig. 4 Energies of QD optical transitions as a function of temperature for the QD structure without (a) and with (b) InGaAs SRL. The symbols are experimental data, whereas the solid lines are fitted using the Varshni formula. Dashed lines indicate energy bandgap variation with temperature in the bulk InAs and GaAs. 8-band $k \cdot p$ numerical calculations (open circles) match reasonably well the experimental data.

$>$ 150 K). This shift can be attributed to thermal redistribution of carriers between small and large QDs at elevated temperature [17].

4 Conclusions

The effect of 5 nm-thick InGaAs capping layer on optical properties and electronic structure of epitaxial InAs quantum dots (QDs) was studied. Temperature-dependent (3–300 K) modulated reflectance and photoluminescence (PL) spectroscopy was used with particular interest to investigate the interband optical transitions in QDs. Spectroscopic features observed were identified using numerical calculations in a framework of 8-band $k \cdot p$ method. It was shown that covering the dots with InGaAs strain-relieving layer induce the decrease of QD ground-state transition energy by ~ 150 meV. It was found that temperature dependence of QD interband transition energy can be fit using Varshni expression with parameters larger than used for bulk InAs due to Ga/In interdiffusion during epitaxial growth. Furthermore, contrary to InAs-InGaAs QDs, for InAs-GaAs dots a significant Stokes shift appeared at elevated temperature (150 K) owing to carrier thermal redistribution effect among QDs of different size.

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