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Retrapping of carrier emission in photoluminescence decay time in InAlAs quantum dots with different thickness

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ABSTRACT

The effect of thickness have been investigated on the optical properties of InAlAs/GaAlAs quantum dots (QDs). The photoluminescence (PL) and time-resolved photoluminescence (TRPL) have been studied on temperature dependences. A sudden decrease in the PL energy emission with increasing InAlAs thickness indicates that the optical emission comes from the wetting layer to QDs, while the full width at half maximum (FWHM) abruptly increases due to the efficient relaxation process. The density of InAlAs QDs decreases as the InAlAs thickness increases from 4.2 to 6.4 monolayers (ML). With increasing excitation power, the QD emission energy was red-shifted for the highest ML, attributed to the carrier trapping. A strong decrease in the exciton lifetime with increasing ML has been observed on temperature dependence induced by the QD zero-dimensional confinement.

1. Introduction

Most of III–V materials have direct energy bandgaps and high carrier mobility, which are attractive to both electronic and optoelectronic devices. Optical memories based on quantum dots (QDs), for example, promise to yield ultra-high density storage media [1]. Another concept that uses large spread of optical transitions is a QD spectrometer proposed by Jimenez et al. [2]. Light of different wavelength can be absorbed to generate carriers inside QDs depending on the energy levels and therefore their size. A large inhomogeneous broadening will therefore be necessary for a device detecting broad spectral ranges. When two excess electrons are introduced, Coulomb repulsion forces them into opposite corners. By choosing which of the corners are occupied, logic states “0” and “1” can be realized [3]. The spin states of coupled single-electron QDs can be used for quantum computation [4]. Since the dimensions of QDs are extremely small, a precise control over the manufacturing process is essential. In early experiments, QDs were produced by lithographical methods as for example described by Reed et al. [5]. Another possible method that was carried out already very early is the growth of semiconductor nanocrystallites in solution [6] or in a dielectric matrix [7]. One of the most appealing approaches, however, is the use of special combinations of substrate and growth material to realize semiconductor QDs on a semiconductor surface without the need of lithography [8]. Depending on the experimental conditions (like substrate temperature, rate of deposition) and material parameters (lattice mismatch between substrate and deposited material, surface energies), molecular beam epitaxy is a precise control over the amount of material deposition. The situation is quite different in the presence of strain caused by a mismatch of the lattice constants of substrate and epilayer. The commonly used InAlAs/GaAlAs features, for example, a 3.5% lattice mismatch [9]. When the material is deposited slowly, one or two monolayers (the so-called wetting layer) are formed. Once a critical thickness is reached it becomes energetically more favorable to form separated islands. This process is sometimes referred to as 2D to 3D transition and is the so called Stranski-Krastanov (SK) mode of growth. The transition, occurring at typical thicknesses between 1.4 and 1.7 monolayers (MLs) of deposited material [10], is rather sharp as observed by reflection high-energy electron diffraction (RHEED). In this paper, the effect of InAlAs layer thickness have been investigated on temperature and excitation density dependences.

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2. Experimental procedure

The three samples studied in this work follow a similar growth procedure. The QD sample was prepared by molecular-beam epitaxy, and is similar to previously studied samples [16]: the growths of the $In_{0.62}Al_{0.38}As$ QDs was performed on a 100 nm-thick $Ga_{0.67}Al_{0.33}As$ epilayer, and was monitored by reflection high-energy electron diffraction (RHEED). The QDs were covered by a $Ga_{0.67}Al_{0.33}As$ epilayer of same composition and same thickness. The growth temperature is around 560 °C for all samples. The growth rate is similar for all samples, and is about 0.92 monolayer per second (ML/s). The critical thickness, at which the growth turns from two-dimensional (deposition of $In_{0.62}Al_{0.38}As$ layers) to three-dimensional (appearance of $In_{0.62}Al_{0.38}As$ QDs), is detected on the RHEED pattern. It is measured during the growth process at about 3.7 ML. Photoluminescence (PL) is a straightforward method to measure the material quality by receiving a wavelength spectrum. By exciting a high power laser beam whose wavelength shorter than the energy bandgap of sample; a 405 nm diode laser excited the InAlAs/GaAlAs QDs and focused on the sample in a 100 μm-diameter spot. The resulting PL was dispersed by a monochromator blazed at 0.6 μm and detected by a silicon avalanche photodiode. The excitation power is taken equal to 5 mW at the sample surface. During the PL measurement, the sample was in a variable-temperature He-cryostat (4–150 K), on a cold finger.

3. Results and discussion

Fig. 1 shows the energy bandgap of InAlAs, for different monolayers, at room temperature (T = 10 K). The PL can be attributed in part to the finite intrinsic emission of each single QD, which produces a collective emission background. Curves 1(a), 1(b) and 1(c) concern the 6.4, 5.5 and 4.2 ML samples, respectively. Because the QDs height of InAlAs is smaller than that of AlGaAs, the emission energy of these barriers becomes sufficiently long without significant changes in the morphology. The narrow line systematically measured at 1.94–2.00eV, with full width at half maximum (FWHM) 12–15 meV, is attributed to the PL of the $Ga_{0.67}Al_{0.33}As$ symmetric barrier. The spectrally broad PL signal observed at lower energies comes from fundamental optical transitions within the $In_{0.62}Al_{0.38}As$ QDs. In curve 1(a), such a PL signal is centred at 1.747 eV and possesses a FWHM of 91 meV. In curve 1(b), the maximum of the low energy PL signal is located at 1.786 eV, and the corresponding FWHM is 99 meV. In curve 1(c), this PL maximum is observed at 1.886eV, and the FWHM is 22.5 meV. The low-energy PL signal shifts toward lower energies and broadens as the amount of $In_{0.62}Al_{0.38}As$ deposition is increased.

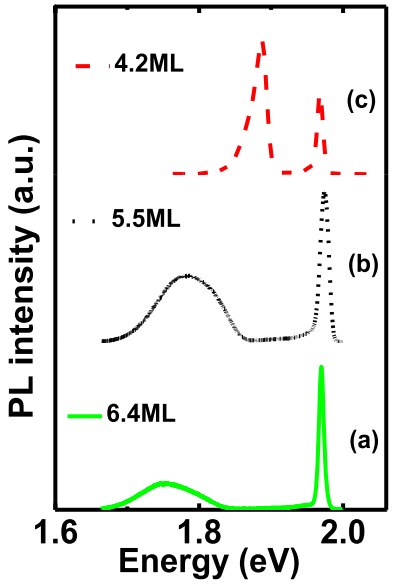

Fig. 1. Low-temperature PL spectra of the studied samples (a) 6.4 ML; (b) 5.5 ML and (c) 4.2 ML. The narrow line near 2 eV is due to the $Ga_{0.67}Al_{0.33}As$ barrier; the low-energy PL signal, spectrally broad, arises from the $In_{0.67}Al_{0.33}As$ QDs.

We first qualitatively interpreted the low-energy signal in the PL spectra shown in Fig. 1, following two guidelines: (1) in samples in which this structure is narrow (typically 22.5 nm), we suspect that the optical emission comes from wetting layers (WL), possibly with roughness and/or with QDs of so small sizes that they confine no photocreated carriers; (2) in samples in which this structure is broad 5.5 and 6.4 ML, the optical emission is probably due to confining QDs, the size distribution of which explains the inhomogeneous spectral broadness. Specifically, for the three x = 0.38 samples (see Fig. 1), the plots in Fig. 2 appear to confirm a transition from regime (1) to regime (2): as the total amount of deposited In0.62Al0.38As alloy is increased from 4.2 to 5.5 and 6.4 ML, the central position of the low-energy PL signal suddenly decreases in energy (from 1.89 to 1.77 eV), while its FWHM abruptly increases (from 20 to 100 meV). Another feature of QDs must be taken into account: As of today, no set of self-assembled QDs consists of identically shaped and sized QDs. Due to the nature of the growth process, they exhibit a stochastic distribution of sizes, which in turn changes their confinements and spectral parameters [11]. This can be measured as a broadening of the collective spectrum, referred to as 'inhomogeneous broadening' (see Fig. 1), as compared to the natural linewidth of a single QD (called 'homogeneous broadening').

A 532-nm green laser has been used to excite the PL sample in a 100 μm diameter spot, which is pasted in a variable-temperature (10–250 K) closed cycle He cryostat. The PL signals are dispersed by a 0.5 μm grating monochromator and recorded by a silicon avalanche photodiode. The power of 532-nm laser is varied to obtain different power density for PL measurement. The spectrum could illustrate the emission energy, the intensity of spectrum, bandwidth of emission and different performance from low temperature (~10 K) to high temperature (~150 K).

Fig. 3 (a), (b) and (c) show the temperature dependent PL spectra for sample A (6.4 ML), sample B (5.5 ML) and sample C (4.2 ML). The PL intensity measurement is attributed to the emission from the ground state QD samples. As usually observed in semiconductors [11], we see a regular red shift when the temperature increases from 10 K to 150 K (Fig. 3(a)), 10 K–110 K (Fig. 3(b)) and

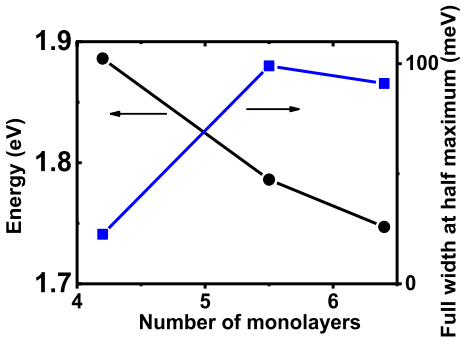

Fig. 2. Central energy E (full circles, left vertical axis) and FWHM (full squares, right axis) of the low-energy PL signal shown by the three samples (see Fig. 1); the horizontal axis represents the total amount of deposited $In_{0.67}Al_{0.33}As$. The optical emission appears to turn from a one due to a WL to another arising from inhomogeneous QDs.