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Influence of the size of self-assembled InAs/AlAs quantum dots on photoluminescence and resonant tunneling

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Abstract

Self-assembled InAs quantum dots (QDs) grown on AlAs have been embedded in three sample structures for atomic force microscopy (AFM), photoluminescence (PL) and resonant-tunneling measurements. The amount of deposited InAs was varied between nominally 1.6 and 2 monolayers. This leads to a shift of the QD-related PL peak from about 1.9 to 1.6 eV that is due to an increasing QD size as confirmed by AFM images of the reference wafer. With increasing InAs coverage, the onset voltage for single-electron tunneling shifts from 0.3 to 0 V which is explained by the lower lying ground state in the larger QDs. The observed strong dependence of the QD size on the InAs coverage offers up a simple way to adjust the onset voltage for single-electron tunneling via growth parameters. © 2002 Elsevier Science B.V. All rights reserved.

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Keywords: Self-assembled quantum dots; Atomic force microscopy; Photoluminescence; Resonant tunneling; Single-electron tunneling

1. Introduction

Self-assembled InAs quantum dots (QDs) embedded in an AlAs matrix are of increasing interest [1–3]. The smaller size and the deeper confinement potential within the high-band gap material AlAs instead of GaAs shift the photoluminescence (PL) energy of the QDs into the visible range and offer some potential for opto-electronic applications [4]. In contrast to InAs/GaAs, little is known about the growth, the properties and the electronic structure of the InAs/AlAs QD system. Nevertheless, this material system has been used for several years in resonant-tunneling diodes for investigations of single-electron tunneling through zero-dimensional (0D) dot states [5–9]. In this study, we present results of resonant tunneling experiments and PL spectroscopy on InAs/AlAs QD samples concerning the influence of the dot size.

2. Sample preparation

InAs/AlAs QDs were embedded in three different sample structures all fabricated with molecular-beam epitaxy. The layer sequence of wafer 1 (AFM structure) with uncapped QDs on top is as follows: a semi-isolating GaAs(1 0 0) wafer, a 500 nm GaAs buffer layer and 20 nm of AlAs grown at 600°C. Wafer 2 (PL structure) is similar to wafer 1 except that the QDs were additionally capped with 20 nm AlAs and 10 nm GaAs. In wafer 3 (resonant-tunneling diode) for vertical transport experiments the QDs are embedded symmetrically in two 5 nm AlAs barriers, 15 nm GaAs pre-wells and 1 μm n^+GaAs emitter

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and collector, respectively, grown at 600°C on an n^+GaAs substrate [8]. The wet-chemically etched tunnel diodes of 40×40 μm^2 base length are equipped with standard AuGeNi contacts.

The growth conditions of the QDs were the same for all wafers. During a growth interruption the substrate temperature (T_S) was lowered to 500°C and the substrate rotation was stopped in order to achieve a gradient in the InAs coverage across the wafer. The gradient across a 2″ wafer was estimated from reference samples to about 30%. Then, InAs was deposited with a growth rate of 0.05 monolayers (ML)/s. A change of the reflection high electron energy diffraction (RHEED) pattern indicated the beginning of QD formation at 1.6 ML InAs. Nominally 1.8 ML of InAs were deposited in the center of the wafer which results in a gradient of 1.55–2.05 ML InAs. Pieces with different InAs coverage were cut from the wafers.

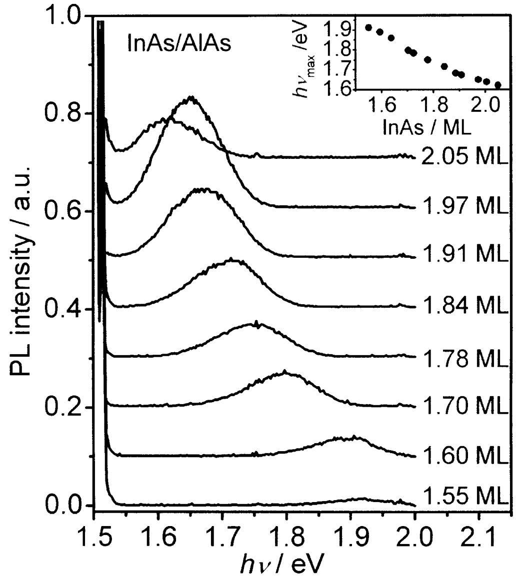

Fig. 2. PL spectra of InAs/AlAs QDs measured at $T = 6$ K for samples with different InAs coverages. The spectra are shifted for clarity. The inset shows the dependence of the PL peak maximum $h\nu_{max}$ on the InAs coverage.

3. Experimental results

AFM images measured on the uncapped QD sample (wafer 1) in Fig. 1 show a high QD density of about 3×10^{11} cm^{-2} which is nearly constant for InAs coverages between 1.6 and 2 ML in agreement with Ref. [2]. With increasing InAs supply, an increasing size of the QDs is observed from about 10 to 14 nm in diameter and up to 3 nm in height. This behavior is quite different from that of QDs grown on GaAs where the dot density increases with InAs coverage. Compared to InAs/GaAs QDs, the size distribution is broader and it somewhat improves at higher InAs coverage. For a coverage of 2.0 ML InAs, the majority of the dots reaches their maximum size of about 14 nm in diameter before coalescence occurs.

PL spectra of wafer 2 with the QDs buried in AlAs measured at $T = 6$ K under cw-excitation ($\lambda = 514$ nm, $P = 10$ mW cm^{-2}) of an Ar^+ laser are plotted in Fig. 2. The emission around 1.95 eV is attributed to PL from the wetting layer (WL) [3]. With increasing InAs coverage (across the wafer), a gradual decrease of the PL peak energy $h\nu_{max}$ from about 1.9 to 1.6 eV is observed; see inset of Fig. 2. This red shift is explained by a lower ground state energy in the larger QDs at higher InAs coverage which agrees with the AFM result from the uncapped QDs (wafer 2). The full-width at half-maximum

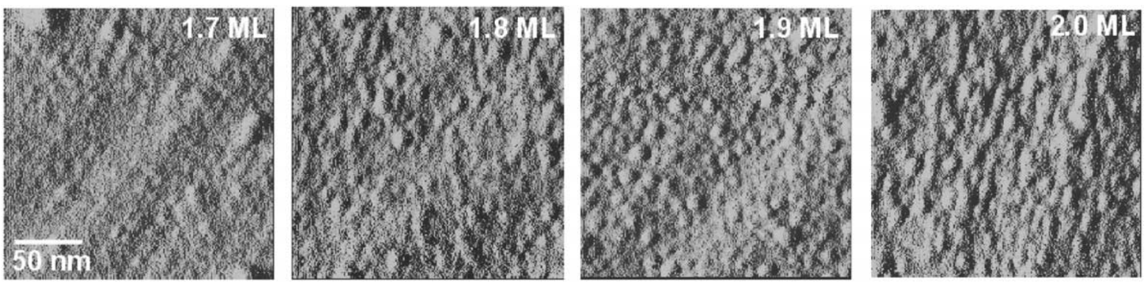

Fig. 1. AFM deflection images of InAs QDs grown on AlAs with different InAs coverages.

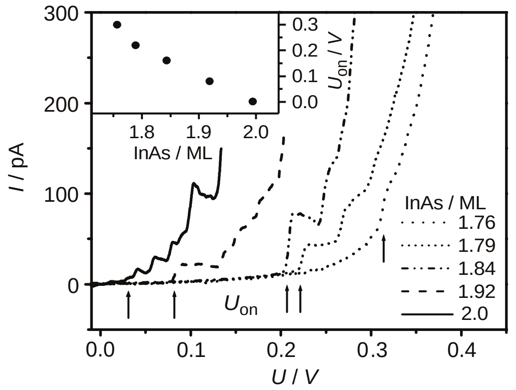

Fig. 3. $I(V)$ characteristics at $T = 4.2$ K of resonant-tunneling diodes with InAs QDs grown with different InAs coverages. The arrows mark the onset of single-electron tunneling at U_{on}. The inset shows the dependence of U_{on} on the InAs coverage.

(FWHM) of the QD-related PL peak of more than 100 meV reflects the broad size distribution of the QDs. The slight decrease from 120 to 100 meV FWHM with increasing InAs supply is also seen in AFM images as an improvement of the dots size distribution. The smallest linewidth of FWHM ≈ 100 meV and the highest quantum efficiency is obtained for the sample with the large dots at high InAs coverage of about 1.9 ML. Above 2 ML, the PL signal is quenched, probably by the introduction of dislocations.

Current–voltage characteristics $I(V)$ of the resonant tunnel diodes cut from wafer 3 with different amounts of InAs incorporated in the AlAs barriers are plotted in Fig. 3. For all samples we observe steps in $I(V)$ which we attribute to single-electron tunneling from the 3D emitter into 0D states of the QDs (electron tunneling into the base of the QDs) [8,9]. With increasing bias voltage, the emitter shifts upwards in energy. When the Fermi level (E_F) in the emitter is in resonance with the lowest QD states (large dots), steps in the $I(V)$ curves occur at the onset voltage (U_{on}). A further increase of the voltage shifts E_F towards the maximum of the QDs energy distribution which strongly enhances the tunneling current and single steps can no longer be resolved.

The observed shift of U_{on} to lower voltages in the samples with higher InAs coverage is due to lower electronic levels in the larger dots in agreement with the PL and the AFM results. Then, resonant tunneling occurs at lower bias voltage and a gradual shift from about $U_{on} \approx 0.3$ V to nearly 0 V for the samples with 1.76–2.0 ML InAs is observed; see inset of Fig. 3. The fact that U_{on} is a function of the InAs coverage and the QD size, respectively, offers a simple way to tune the voltage range of single-electron tunneling. With the adjustment of the bias voltage, the large QDs are selected for single-electron tunneling undisturbed by the large number of the smaller QDs. In that sense, the broad size and energy distribution of the InAs/AlAs QDs is not disadvantageous to tunneling experiments and the reduced dot size (compared to InAs/GaAs) put the electronic levels to reasonable energies accessible by resonant tunneling.

4. Comparison of PL and resonant tunneling data

The absence of resonant tunneling close to $U = 0$ show that the electronic QD levels are located above the emitter Fermi energy (E_F) which is about 15 meV above the conduction band (E_C) in the n^+GaAs layer [9]. The energy of the resonant QD level (E_{QD}) can be estimated from the voltage U_{on} with $E_{QD} = E_F = \alpha e U_{on}$ (α is the relative voltage drop across the first tunneling barrier and e is the electron charge). On the assumption of a symmetric voltage drop of $\alpha = 0.5$, the energetic position of the largest QDs with respect to E_C in the emitter (E_{QD}–E_C) is plotted in Fig. 4. It can be seen that E_{QD} strongly depends on the InAs coverage. From the $I(V)$ curves of resonant-tunneling diodes incorporating only an InAs-WL (kink at about 0.6 V [8]) the WL energy is estimated at $E_{wl} - E_C \approx$ 300 meV. Knowing E_{wl}, we can fit this energy scale to that estimated from the PL spectra. From the maximum of the QD PL peak energies ($h\upsilon_{max}$, inset of Fig. 2) and the WL emission at $h\upsilon_{wl} = 1.95$ eV the maximum of the electronic QD state distribution (E_{max}) with respect to E_{wl} is calculated according to $E_{max} - E_{wl} = \beta(h\upsilon_{wl} - h\upsilon_{max})$. ($\beta$ is the portion due to conduction band states.) With the assumption of $\beta = 0.7$ and $E_{wl} =$ const. in all samples the result is plotted in Fig. 4. It can be seen that the QD energies derived from the PL data also depend strongly on the InAs coverage. These values are about 50 meV higher in energy compared to those from U_{on} which is a reasonable value for the difference between the maximum (measured by PL) and the low energy end (measured

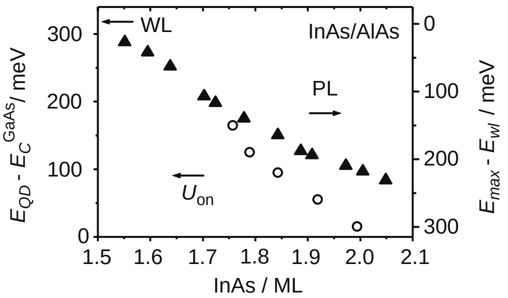

Fig. 4. Electronic ground state energies of the QDs estimated from U_{on} of resonant tunneling (open circles) and PL peak energy $h\upsilon_{max}$ (solid triangles) as a function of the InAs coverage. The arrow marks the energetic position of the wetting layer as deduced from [8].

by resonant tunneling) of the energy distribution of the electronic QD levels. In spite of the rough assumptions, both experiments give a consistent picture for the energy band diagram of the InAs/AlAs QD system.

5. Summary

The onset voltage for the first current step, U_{on}, in the $I(V)$ characteristics of resonant-tunneling diodes with InAs QDs embedded in the AlAs-tunneling barrier strongly depends on the InAs coverage. With increasing InAs supply from about 1.6 to 2.0 ML, U_{on} is decreasing from about 0.3 to 0 V. As suggested by AFM and PL results obtained from reference samples, this is explained with lower lying electronic states in larger QDs that allows resonant tunneling at lower bias voltage. This offers a simple way to control the onset of single-electron tunneling in resonant-tunneling devices via growth parameters.

Acknowledgements

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