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2007 Nanotechnology 18 015401

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Optical and structural properties of vertically stacked and electronically coupled quantum dots in InAs/GaAs multilayer structures

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Received 4 September 2006, in final form 24 October 2006
Published 8 December 2006
Online at stacks.iop.org/Nano/18/015401

Abstract
This work systematically investigated the optical and structural properties of multilayer electronic vertically coupled InAs/GaAs quantum dot (QDs) structures grown by molecular beam epitaxy for long-wavelength applications. A significant energy blue-shift in the photoluminescence (PL) spectra from 30-period InAs/GaAs QDs structures was observed as the GaAs spacer thickness was decreased. Transmission electron microscopy (TEM) and PL measurements indicated that the abnormal blue-shift can be attributed to the strain-driven In/Ga intermixing between QDs and spacer layers, which overcompensates for the effects of electronic and structural couplings between QD layers. Moreover, this study demonstrates that increasing the growth rate of InAs QDs can prevent intermixing. A PL emission wavelength of 1320 nm with strong luminescence at room temperature, which corresponds to an energy red-shift of 50 meV from that of the single QD layer sample, was achieved in a 10-period InAs/GaAs QD superlattice with a spacer thickness of 16 nm.

1. Introduction
Quantum dots (QDs) are important nanostructures owing to their superior characteristics and extensive applications. Their use in semiconductor lasers supports an ultra low threshold current density and extremely high thermal stability, because of the delta-function-like density of states [1]. 1.3 μm wavelength lasers, based on self-organized InAs QDs embedded in InGaAs quantum wells, have been demonstrated to have a very low threshold current density (16 A cm−2) [2]. The use of a ten-layer stack of long-wavelength QDs has enabled the external differential quantum efficiencies to reach 88% in edge-emitting lasers [3] and 1.3 μm vertical cavity surface-emitting lasers (VCSELs) to be formed monolithically on GaAs substrates [4]. On the other hand, small InAs QDs have been vertically stacked to increase the active volume; increase the emission wavelength and reduce the emission linewidth [5]. As the spacer layers between the dots are thinned to a few nanometres, electronic coupling is observed. By
performing an analysis based on the coupled rate equations, Shi and Xie [6] predicted that electronic vertically coupled QD (EVCQD) lasers exhibit a higher single-mode output power than uncoupled lasers, especially when the dot size fluctuates greatly. Moreover, the optical modal gain can be increased by using a thinner EVCQD active region, improving the momentum matrix element for the recombination of carriers. Additionally, the EVCQD active region improves the carrier injection efficiency by the tunnelling of carriers through the QD layers. However, the specific range of application is governed by the characteristics of the QD laser—in particular, the emission wavelength. 1.3 μm emission with high luminescence intensity from a vertical stack of small InAs QDs in GaAs has not yet been obtained. Our previous work [7, 8] demonstrated high-performance EVCQD lasers with lasing wavelengths of around 1.23 μm using a ten-stack InAs/GaAs superlattice with a spacer thickness of 17 nm. However, fully understanding structural and electronic coupling between dot layers is essential to device design and growth condition optimization, especially for long-wavelength applications.

This work systematically investigates the optical and structural properties of multilayer electronic vertically coupled InAs/GaAs QDs structures. The effects of spacer thickness and growth rate were studied. At a low growth rate, a significant blue-shift in the photoluminescence (PL) spectra of 30-period InAs/GaAs QDs structures occurred as the thickness of the GaAs spacer decreased. The results of transmission electron microscopy (TEM) and PL measurements indicate that the blue-shift is caused by In/Ga intermixing between InAs QDs and GaAs spacer layers, driven by the accumulation of strain energy in the multilayer QDs structures. We have also demonstrated that a higher growth rate of InAs QDs can prevent intermixing. An energy red-shift of 50 meV caused by the formation of a mini-band was observed for a 10-period InAs/GaAs QD superlattice with a spacer thickness of 16 nm. Strong room-temperature PL emission at 1320 nm was achieved from this sample.

2. Experiment

The structures were grown by molecular beam epitaxy (MBE) on GaAs substrates. The QDs were grown in Stranski–Krastanow growth mode by depositing 2.6 monolayers (MLs) of InAs with a growth rate of 0.028 or 0.085 ML s⁻¹ and a substrate temperature of 490–510 °C. The formation of QDs was controlled in situ by monitoring the diffraction pattern of high-energy electrons (RHEED). The surface density of the QDs, estimated from atomic force microscopic (AFM) images, was 2–5 × 10¹⁰ cm⁻². Following the deposition of QDs, a 10 nm thick GaAs layer grew uninterruptedly at the same temperature to cover the surface; then, the growth temperature of the remaining layers was changed to 600 °C, to improve the crystalline quality. Table 1 summarizes the detailed structures and growth parameters. The thickness of the GaAs spacer layer between the InAs QD layers is defined as the thickness between the adjacent wetting layers, and not the thickness between QDs. For instance, in sample D, the thickness of the GaAs spacer layer is 16 nm. Since the height of the InAs QD is about 7 nm, as shown in figure 1, the separation between the coupled QDs is approximately 9 nm. PL measurements at 10 K and room-temperature were made using a 632.8 nm He–Ne laser as the excitation source and detecting the emission using an InGaAs photodetector.

3. Results and discussion

Figure 1 shows cross-sectional TEM images of two 30-period InAs/GaAs QD samples (a) with 30 nm thick GaAs spacer

Table 1. Number of periods, growth parameters and PL (10 K) data of InAs/GaAs QDs multilayer structures.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Spacer type</th>
<th>Number of periods</th>
<th>Growth rate (ML s⁻¹)</th>
<th>Growth temperature (°C)</th>
<th>Main peak energy (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>n-type</td>
<td>1</td>
<td>0.028</td>
<td>490</td>
<td>1.065</td>
</tr>
<tr>
<td>B</td>
<td>n-type</td>
<td>30</td>
<td>0.028</td>
<td>490</td>
<td>1.058</td>
</tr>
<tr>
<td>C</td>
<td>n-type</td>
<td>30 20</td>
<td>0.028</td>
<td>490</td>
<td>1.062</td>
</tr>
<tr>
<td>D</td>
<td>n-type</td>
<td>30 16</td>
<td>0.028</td>
<td>490</td>
<td>1.127</td>
</tr>
<tr>
<td>E</td>
<td>n-type</td>
<td>30 10</td>
<td>0.028</td>
<td>490</td>
<td>1.157</td>
</tr>
<tr>
<td>F</td>
<td>S.I.</td>
<td>1</td>
<td>0.085</td>
<td>510</td>
<td>1.068</td>
</tr>
<tr>
<td>G</td>
<td>S.I.</td>
<td>10 16</td>
<td>0.085</td>
<td>510</td>
<td>1.018</td>
</tr>
<tr>
<td>H</td>
<td>S.I.</td>
<td>30 16</td>
<td>0.085</td>
<td>510</td>
<td>1.016</td>
</tr>
</tbody>
</table>

layers, sample B, and (b) with 16 nm thick GaAs spacer layers, sample D. The size of the InAs QDs was determined to be about 7 nm high and 22 nm wide. As presented in figure 1, no clear correlation exists between the QD layers in the 30 nm spacer sample, while complete vertical alignment is evident in the 16 nm spacer sample and a dot size increase with a layer increase was unobserved. The vertical alignment is caused by the strain field of the buried dots in the underlying layer. Therefore, the vertical pairing probability can be controlled by adjusting the spacer thickness [9–11]. During the growth of the QDs in the upper layer, the strain minima that result from the underlying QDs typically cause many atoms to migrate to the regions, making the dots in the upper layer slightly larger than in the lower layer [9, 11, 12]. Besides, smaller uncoupled QDs in the first layer were observed, as indicated by the arrow in figure 1(b).

The electronic states of EVCQDs can acquire a wire-like character due to vertical dot-to-dot coupling. This behaviour can be easily examined by optical anisotropy [13–15]. Figure 2 shows the dependence of the edge-emitted PL spectra of sample A, B, C, D and E on polarization. The degree of polarization is defined as $P = (I_\parallel - I_\perp)/(I_\parallel + I_\perp)$, where $I_\parallel$ ($I_\perp$) is the PL intensity polarized in (normal to) the surface plane. For a semiconductor QD, the optical transition is well known to be polarized in the elongated dot direction. As presented in figure 2, the polarization of the sample with a single layer of dots is perpendicular to the growth direction (TE polarization) with $P \sim 21\%$, corresponding to a larger in-plane dimension. As the spacer thickness declines, the polarization exhibits TM behaviour, in the growth direction. The degrees of polarization of the 16 nm and 10 nm samples are $P \sim -18\%$ and $-26\%$, respectively. These results are direct evidence that vertical electronic coupling occurs when the spacer thickness is $\leqslant 16$ nm and the electronic states are strongly elongated in the growth direction. The results are also consistent with the results of surface photovoltage spectroscopy [16].

Figures 3(a) and (b) display the 10 K PL spectra of samples grown at low and high growth rates, respectively. The main peak energies are summarized in table 1. As presented in figure 3(a), the dots in the 30 nm spacer sample (sample B) are almost all isolated. Therefore, the peak energy is close to that of the reference sample, with a single layer of dots (sample A). However, for samples C–E, the energy is considerably blue-shifted as the spacer thickness declines. This finding is inconsistent with the observed red-shift, described in previous reports, which was attributed to the formation of a mini-band caused by the electronic coupling in the vertical direction [5] or/and the increase in the size of InAs QDs caused by strain field coupling [17, 18]. The blue-shift is suggested to be associated with the strain-driven material intermixing [16, 19–21]. The individual field from the constituent dot is superimposed on the overall strain field, increasing the total strain energy during overgrowth. Hence, QDs must be intermixed with the adjacent GaAs spacer layers to reduce the total energy, leading to coherently relaxed strain
surrounding the QDs. The increase in the band gap energy due to In/Ga intermixing overcompensates for the effect of the size increase and causes a blue-shift, suggesting that the large strain energy, which increases as the thickness of the spacer layer decreases, may drive considerable intermixing between InAs QDs and the GaAs spacer layer. Another weak peak was observed at 1.070 and 1.076 eV from samples D and E, respectively. Since the peak energies are close to the ground state transition energy of the single-layer sample (sample A), this lower energy transition is attributed to the uncoupled QDs in the first layer, as shown in figure 1(b). The lack of uniformity of the sizes causes some of the smaller QDs to be uninvolved in the vertical alignment. The critical size of these uncoupled QD should drop as the spacer thickness decreases, slightly increasing the transition energy. However, the PL peak from the high-growth-rate samples, as shown in figure 3(b), is red-shifted by 50 meV from the corresponding position for the single QD layer, sample F, for the 10-period EVCQD structure (sample G) and by an additional 2 meV for the 30-period EVCQD structure (sample H). However, the PL signal of the uncoupled QDs merges into the signal of the excited state transition of EVCQDs. The red-shift behaviour differs from that of the QD samples grown at low growth rate. This discrepancy is attributed to the effect of the growth rate on the strain-driven In/Ga intermixing between QDs and spacer layers. Increasing the growth rate, however, caused growth farther from the thermal dynamic equilibrium condition, alleviating intermixing.

Parts of QD layers were etched from sample D and the PL spectra were measured to elucidate further the strain-driven intermixing. Figure 4 presents the results. The remaining number of periods was estimated from the etching depth, which was determined using a surface profiler. The energy of the aforementioned low energy peak, attributed to the uncoupled QDs in the first layer, essentially remains constant as the etching depth increases, while the relative intensity gradually increases, finally dominating the PL spectrum. While the high energy peak which is assigned to the EVCQDs gradually red-shifts as the etching depth increases. This finding indicates that as the number of periods increases, the accumulating strain energy increases the degree of intermixing, increasing the transition energy in the growth direction.

The evolution of transition energy throughout the structure can also be investigated by comparing the PL excited from the front side with that excited from the back side. In the back side PL measurement, the excited light was incident on the back surface of the substrate. Figure 5 presents the results.
The solid lines refer to normal excitation while the dotted lines refer to excitation from the side of the substrate. The higher background signal represented by the dotted line in figure 5(a) is from the n-type GaAs substrates while the other two back side spectra from the samples grown on Si substrates do not have the problem. All samples give one peak (on the low energy side for sample D, and the high energy side for samples G and H) at essentially the same energy; the intensity in the cases of excitation from the substrate side markedly exceeds that of in the case of normal excitation. This energy peak is attributed to the uncoupled QDs in the first layer. The position and behaviour of the peaks are consistent with our aforementioned observations. Moreover, for sample D, a sample grown at a low growth rate of 0.028 ML s\(^{-1}\), the results show that the transition energy of the coupled QDs in the upper layers considerably exceeds those in deeper layers. This result provides evidence of gradually enhanced strain-driven intermixing in the growth direction. However, the peaks from sample G, a sample grown at a high growth rate of 0.085 ML s\(^{-1}\), are essentially at the same position, indicating constant transition energies of the coupled QDs throughout the multilayer structure. Accordingly, higher growth rates of QDs could greatly suppress the intermixing, driven by the accumulative strain energy. Furthermore, the energetic red-shift of 50 meV, observed in the 10-period InAs/GaAs QD superlattice with a spacer thickness of 16 nm, is attributed to the formation of a mini-band. Moreover, as presented in figure 6, an emission wavelength of 1320 nm with strong luminescence at room temperature was achieved. The results reveal that the active region of the EVCQD superlattices is very promising for supporting high-quality 1.3 \(\mu\)m lasers.

4. Summary

The structural and optical properties of multilayer electronic vertically coupled InAs/GaAs QD structures grown by MBE with various GaAs spacer thickness and InAs QDs growth rates were investigated. In samples grown at a low growth rate, a significant blue-shift was observed as the GaAs spacer thickness decreased, because of the strain-driven intermixing of In in QDs and Ga in the GaAs spacer layer, which is gradually increased in the growth direction. Some uncoupled QDs in the first layer were also observed. However, using a higher growth rate can greatly suppress the intermixing effect. Therefore, an energy red-shift of 50 meV caused by the formation of a mini-band was obtained in a 10-period InAs/GaAs QDs superlattice with a spacer thickness of 16 nm, and the emission wavelength of 1320 nm with strong luminescence at room temperature was achieved. The results reveal that the electronic vertically coupled InAs/GaAs QD superlattice is a very promising active medium for high-quality 1.3 \(\mu\)m lasers.

Acknowledgments

The authors are grateful to K Y Hsieh for his help in TEM measurement, and K F Lin for performing the chemical etching. This work was supported by the National Science Council of the Republic of China, Taiwan, under contract No NSC 95-2112-M-033-008-MY3 and NSC 95-2221-E-002-368.

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