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Photoluminescence characterization of InAs/GaAs quantum dot bilayers

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Abstract

We have investigated the emission from InAs/GaAs quantum dots (QD) bilayer samples with different GaAs spacer thickness. For large spacers, one peak is observed, and the layers are independent. For small spacers, one peak is observed and the layers are then electronically coupled. For intermediate spacers (≈ 120 Å), two emission peaks are observed and these can be made coincident by tuning the amount of InAs deposited in the second layer. We present a technique using two different excitation wavelengths, which enables us to attribute the emission to each layer, and to show that the shifts are not due to electronic coupling. Moreover, resonant excitation shows that the wetting layers are also different in each layer. These results indicate that the presence of the first QD layer strongly influences the growth of the second one, leading to very different properties.

Keywords: Quantum dots; Multilayer; Photoluminescence

1. Introduction

Self-assembled quantum dots (QD) are predicted to produce lasers with low, temperature-independent thresholds. However, the gain of a single QD layer is usually insufficient for lasing from the ground state (GS) of the QDs. Several groups have investigated the structural changes that occur when QD layers are deposited sequentially. Imaging techniques have demonstrated vertical ordering (stacking) [1–3] of the islands, and the ordering is lost when the thickness of the GaAs spacer region exceeds 200 Å [2]. The consensus is that stacking results from a strain interaction during growth [2,4]. Stacking can also have a significant effect on the electronic properties; for spacer layers up to ≈ 100 Å, a redshift of the emission (compared with that of a single layer) is usually observed and attributed to electronic coupling or tunneling between the QD layers [5–10]. However, in some cases blueshifts have been reported [11–13]. In some laser structures the QD layers are separated by thin spacer layers and are designated 'columnar'. It is arguable that these structures are quasi-1D with strong electronic coupling along the column and negate, in part, the benefits of ideal QDs [14].

In this paper we report changes in the emission properties of QD bilayer samples where the dots are grown at a low-growth rate and have the benefit of a relatively small linewidth and room temperature emission close to 1.3 μm [15]. The impetus for the present study arises from a perceived need to have all the QD layers emitting at the same wavelength. This can be achieved by adopting relatively large spacers between the QD layers [13]. However, large spacers may not be the optimum choice for some applications such as vertical cavity surface emitting lasers (VCSELs) where it is advantageous to have the layers grown closely spaced within the cavity. We have therefore studied changes in the emission between QD layers having a fixed intermediate (120 Å) GaAs spacer layer and tuning the emission from the second layer by changing the

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InAs coverage. We have used a variable-pump wavelength (VPW) technique, which enables us to unambiguously correlate the emission with a particular layer and rule out the possibility of electronic coupling.

2. Experimental details

The samples were grown by solid source molecular beam epitaxy using As_2 on n^+ (Si-doped) (001) GaAs substrates. The Ga and In fluxes were calibrated using RHEED oscillations observed during GaAs and InAs homoepitaxy and the 2D–3D transition was monitored during growth of the QD structures. After growth of a 0.6 μm GaAs buffer layer at a substrate temperature of $T_S = 580$ °C, T_S was reduced to 500 °C for growth of a further 500 Å of GaAs. InAs was then deposited at a relatively low-growth rate of 0.016 monolayer per second (ML s^{-1}). The amount of InAs deposited was fixed at $\theta_1 = 2.3$ ML for the first layer and varied between 1.9 and 5.0 ML for the second layer (θ_2). The QD layers were separated by GaAs spacer layers of 120, 240 or 450 Å. For the larger spacer layers the 2D–3D transition time was the same for both layers, but for the 120 Å spacer the second layer transition occurred slightly earlier ($\approx 10\%$). After deposition of the final QD layer the structure was capped with 500 Å of GaAs at the lower value of T_S and the temperature ramped to 580 °C for deposition of a final 1500 Å of GaAs. All the deposited layers were undoped. The growth system has a facility for rapid cool-down (within a few seconds) and transfer to a scanning tunneling microscope (STM). The STM images were used to determine the island size and density for each QD layer. The details of these results will be the subject of another paper. Photoluminescence (PL) measurements were made at low temperature (10 K) using an Ar^+ or HeNe laser, dispersing the light with a SPEX 0.5 m monochromator and detecting with a cooled Ge diode. In order to avoid any emission from the excited states, all the PL experiments were performed at low-excitation densities.

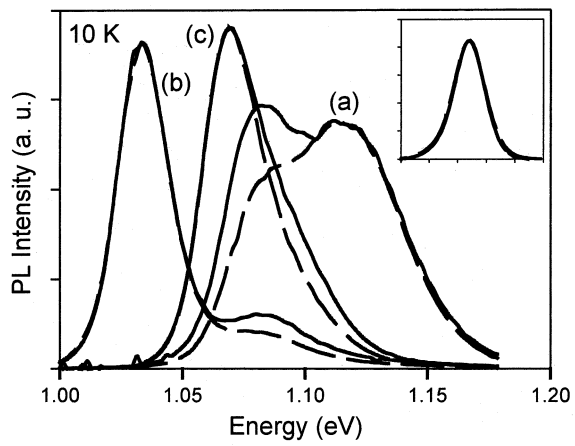

Fig. 1. Low-temperature PL spectra excited with a HeNe laser (solid lines) and Ar^+ laser (dashed lines) from bilayer samples with a spacer of 120 Å, $\theta_1 = 2.3$ ML, and $\theta_2 =$ (a) 2.3, (b) 3.6, and (c) 2.7 ML, respectively. The inset shows the same spectra for a sample with a spacer of 100 Å. They are then indistinguishable.

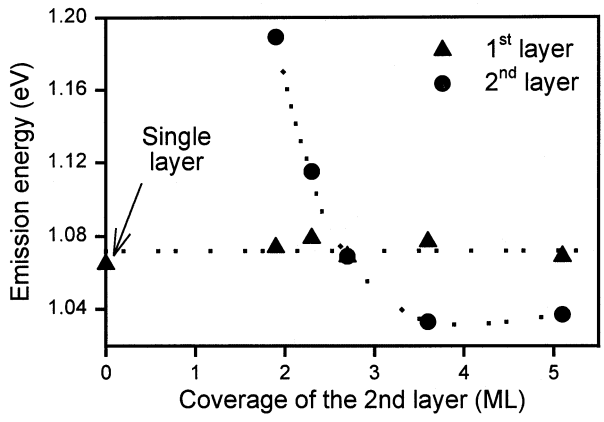

Fig. 2. Emission energies of first and second QD layers as a function of the second layer coverage θ_2 for a fixed spacer of 120 Å. The emission energy from the first layer is roughly constant whereas the second layer peak energy decreases strongly with coverage, saturating around 1.03 eV for the highest coverages. The two curves intersect for $\theta_2 = 2.7$ ML.

3. Results and discussion

We first studied the low-temperature PL of a single QD layer formed during the deposition of 2.3 ML of InAs. The emission (not shown here) occurs around 1.07 eV at 10 K with a linewidth of ≈ 40 meV. The peak is slightly asymmetric on the high-energy side which is due to the presence of some smaller dots (visible in STM images of an equivalent uncapped sample). For a bilayer sample with $\theta_1 = \theta_2 = 2.3$ ML and a 450 Å spacer layer, the emission feature is practically indistinguishable from that of the single layer indicating that each QD layer is unaffected by the presence of the other. A slightly broader feature is observed when the spacer is reduced to 240 Å and we believe that this is indicative of a weak interaction between the two QD layers [13]. Reducing the spacer to 120 Å results in two peaks as shown in spectrum (a) of Fig. 1. The peak around 1.07 eV is attributed to the first layer while the second layer emission is blueshifted. Recent experiments have shown that the emission energy of single layers is sensitive to the amount of InAs deposited [16]. In order to make the two peaks coincident we have therefore deposited different amounts of InAs in the second layer. As illustrated in Fig. 1, a peak around 1.07 eV is always observed and again attributed to the first QD layer. The other peak is attributed to the upper layer. These assignments will be confirmed by

VPW optical experiments discussed later. Fig. 2 shows the emission peaks from the first and second QD layers for different values of θ_2. The lines intersect around 2.7 ML and this combination; $\theta_1 = 2.3$ ML and $\theta_2 = 2.7$ ML produces coincident peaks as shown in spectrum (c) of Fig. 1. This demonstrates how the amount of indium deposited can be used to tune the second layer emission.

The variation of the emission peak from the second layer with InAs coverage is quite different to that observed for single layers [16]. These results demonstrate a complicated interaction occurring between adjacent layers at least for the spacer thickness adopted here. STM images obtained from these samples (not shown here) show that the dots in the second layer are quite different to those in the first, even when the deposition is nominally the same: the dot density is reduced by a factor of 2 and the dots in the second layer are much larger (the average volume is multiplied by ≈ 2). Given the structural differences it is then not surprising that the emission properties of the second layer are different. However, larger dots should lead to a redshift, which is the opposite of what we observed and can only be accounted for by the changes in the strain or indium composition [13] of the dots in the second layer. Our studies of the capping of low-growth rate dots (typically larger than conventional dots) shows that the surface, which is flat before the growth of the first layer, is undulating before the growth of the second layer, due to the presence of dots below [17]. Although the mechanisms involved are still under investigation, we attribute the differences between the layers to the surface morphology and local strain [18] present prior to the growth of the second QD layer. Smoothing of this surface is therefore another strategy to obtain coincident emission even for $\theta_1 = \theta_2$, and is currently under investigation.

In Refs. [9,12], electronic coupling was proposed to explain the PL of multilayers with spacers as large as 100 Å. Tunneling was also invoked [8] to understand the PL of bilayers with 100 Å spacers. Low-growth rate dots are typically bigger (height ≈ 60 Å) than conventional dots [15,16], and therefore, even for the relatively large spacers used here, the tunneling barrier remains small and the QDs may be electronically coupled. To test this we have adopted VPW techniques that enable us to probe the properties of each layer individually, allowing unambiguous identification of the first and second layers. The technique utilizes the change in penetration depth with excitation wavelength and has been used to probe multiple quantum well structures [19]. PL experiments were performed with an Ar^+ laser (emitting at 488 nm) and an HeNe laser (emitting at 632.8 nm). Because these two lasers have different absorption coefficients in GaAs (and therefore different penetration depths), they lead to different distributions of carriers along the growth direction. The Ar^+ ion laser (with smaller penetration depth) will favor the topmost layer compared with the HeNe laser. It is important that the spectra are obtained under low-excitation density, where the shape of the spectrum is unaffected by state filling effects. The spectra obtained from the single layer sample and those with large spacer layers do not change with excitation wavelength, as expected. However, for each bilayer sample, changes in the shape are always observed. Fig. 1 illustrates this for three of the samples. The spectra are superimposed and their intensities normalized. We clearly see evidence for repartition of carriers between the two QD layers. The Ar^+ laser (dashed spectra) favors the layer closest to the surface and this allows us to identify the emission peak for each layer, confirming the previous assignments. In addition, the fact that the relative intensity of the peaks can be changed demonstrates that the two layers do not behave as a single entity. They are, therefore, not electronically coupled and tunneling between dots is not significant. Even when the two peaks coincide (spectrum (c)), we clearly observe a reduction of the FWHM when using the Ar^+ laser showing that the FWHM of the top layer is reduced compared with the first layer, and that the layers are not coupled. The inset to Fig. 1 shows the same study for a bilayer sample where the spacer layer was reduced to 100 Å. Only one peak is observed and its shape does not change when changing excitation wavelength. Because it is highly unlikely that the layers are perfectly identical, it indicates that these two layers are coupled (electronic coupling/tunneling). These results demonstrate the benefits of VPW techniques for the study of multilayer samples and highlight the fact that care must be exercised when measuring PL spectra obtained with Ar^+ lasers in structures with many layers, since the spectral shape of the PL does not reflect the spectral shape of the gain.

Finally, the inset of Fig. 3 shows emission in the vicinity of the wetting layer (WL) obtained under higher excitation conditions from the sample with $\theta_1 = \theta_2 = 2.3$ ML. The shape of this spectrum suggests the presence of two overlapping features, which we tentatively attribute to emission from different WLs associated with each QD layer. To confirm this, we collected the PL from the dots when resonantly exciting at energies between 1.414 and 1.458 eV. Some of the spectra obtained are presented in Fig. 3. Spectrum (a) corresponds to excitation above both WLs and emission from both QD layers is present. Decreasing the excitation energy results in an increase in the QD emission followed by a decrease. However, the emission from each QD layer does not reach its maximum intensity at the same excitation energy. Spectrum (b) corresponds to the maximum of the upper QD layer whilst (c) corresponds to the maximum for the lower.

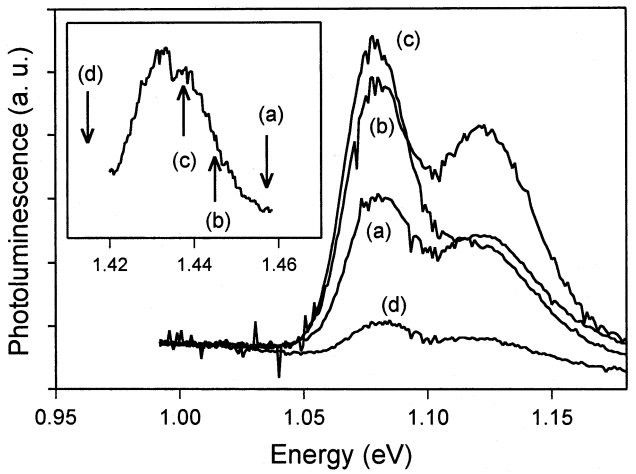

Fig. 3. Spectra obtained for the sample with nominally identical layers ($\theta_1 = \theta_2 = 2.3$) at excitation energies of: (a) 1.458, (b) 1.446, (c) 1.438, (d) 1.414 eV. Spectra (a), (b) and (d) exhibit roughly equal contributions from each QD layer, whereas the first layer strongly dominates in spectrum (c). In this case, the excitation energy is below the absorption edge of the WL of the second layer. The inset shows the PL spectrum in the region of the WL, and excitation energies for each resonant PL spectrum are indicated by arrows. A Stokes shift of 5–9 meV is observed.

Note that emission from the upper layer has almost disappeared in spectrum (c) demonstrating that excitation at 1.438 eV is below the absorption edge of the WL of the second layer but not of the first. Decreasing the excitation energy below both WLs (spectrum (d)) there is only direct absorption into the QDs and a consequent reduction in the PL intensity. From this series of spectra, we can conclude that the energy of the second WL is slightly higher (≈ 8 meV) than that of the first layer. The second layer turns 3D slightly earlier than the first and we can expect there is less indium in the upper WL. This is consistent with the results but other differences in the morphology of the WLs can be expected. Finally, this study of the WLs shows that we can selectively excite one of the layers using resonant excitation, confirming the conclusions obtained from VPW experiments, namely the absence of electronic coupling and tunneling.

4. Conclusions

We have studied the emission properties of bilayer QD samples grown at a low-growth rate. For relatively large GaAs spacer layers, the emission energy is similar to that of single layers. In order to stack as many layers as possible (for VCSEL, for example), spacers with thickness 120 Å are desirable. For 120 Å spacers, the emission peaks of two nominally identical layers were resolved with the second layer being blueshifted with respect to the first. We attribute this change to the different surface morphology and local strain prior to growth of the second layer. Changing the coverage in the second layer, it is possible to tune the emission to be coincident with the first. We used a technique based on the comparison of the PL obtained with two different lasers, enabling us to identify unambiguously the peak arising from a particular layer. In addition we have shown that there is negligible electronic coupling between the two layers for the spacer thickness investigated here. Resonant excitation PL confirms these results and highlights differences in the WL properties of each layer.

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