InGaAs quantum dots grown with GaP strain compensation layers

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A major obstacle in the growth of stacked dot structures with a large number of layers is the high degree of strain in the dot layers. Strain buildup can affect the nucleation of the dots, which may cause defects that are detrimental to device performance. In this work, thin GaP layers are inserted below the quantum dot (QD) layer in single and stacked $In_{0.5}Ga_{0.5}As$/GaAs QD structures. These layers counterbalance the strain caused by the previous layers. Changes in dot nucleation are observed for dots grown directly on GaP layers. The QDs are found to be smaller in width and height. The luminescence from these dots is blueshifted due to interdiffusion between the dots and the GaP buffer layer. In a single layer of dots, no change in dot formation is seen when a thin GaAs barrier layer is deposited between the GaP layer and the QD layer. However, in stacked structures, the addition of a GaP layer below each QD layer is found to increase the density of the dots in the top layer of the stack. The room-temperature photoluminescence intensity is also increased. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1707230]

I. INTRODUCTION

In recent years, quantum dot (QD) devices have been shown to provide performance gains over quantum well devices. However, there are some challenges in fabricating these devices. QD devices, such as lasers and photodetectors, require multiple layers of dots to create enough gain to operate. It is difficult to grow stacked dot structures in which each dot layer has a similar size and density and is without defects.[1,2] Each layer is highly strained and, after several layers, dislocations start to form in the upper layers.[3,4] These dislocations are detrimental to device performance. Even with large separations between the layers, dislocations occur when many layers are stacked. Large numbers of dot layers (>10) are required for photodetectors. Lasers[5] and photodetectors[6] with small numbers of multiple layers of QD layers have been demonstrated. However, only a few groups have reported devices grown by metalorganic chemical vapor deposition (MOCVD).[7–9] MOCVD is the preferable growth technique for manufacturing, but it is more difficult to grow QD device structures using MOCVD. One reason is because of the higher growth temperatures (700–750 °C) required to grow the AlGaAs layers included in the device structure. This produces an annealing effect on the QD layers that have been grown at a lower temperature (500–550 °C). The effect of this annealing has been studied and shown to reduce the photoluminescence (PL) intensity dramatically as well as causing a blueshift and closer energy spacings between the excited states.[10,11] The thermal stability of these stacked structures is also affected by the strain. Both interdiffusion and strain relaxation processes occur during annealing. In this work the use of GaP as a strain compensation layer is investigated. The GaP layers are in tensile strain with respect to GaAs, with a lattice mismatch of 4%, whereas the $In_{0.5}Ga_{0.5}As$ dots are in compressive strain. By growing a thin layer of GaP the strain caused by the GaP layer should partially compensate the strain caused by the dot layer. This should stabilize the system during annealing and should also stop the strain field from the previous layer affecting the nucleation of the dots in the upper layers of a stacked structure.

II. EXPERIMENT

The samples were grown using a low-pressure horizontal flow MOCVD reactor. The sources used were tri-methyl gallium, tri-methyl indium, arsine (AsH_3), and phosphine (PH_3). For the study of the dot nucleation on a tensile strained layer, a single layer of dots was grown with a GaP layer underneath the dots. Firstly, a buffer layer of GaAs was grown at 650 °C, and the growth temperature was then reduced to 550 °C for the dot growth. The dot layer was formed from 5.8 monolayers of $In_{0.5}Ga_{0.5}As$ using a V/III ratio of 15. A thin (5–20 Å) GaP layer was grown either directly before the dots or with a small (50 Å) GaAs barrier between the dots and the GaP layer. After the dot growth, the temperature was ramped back up to 650 °C during growth of a 3000 Å GaAs capping layer. The temperature was then reduced again for a second layer of dots, which was left uncapped for the purposes of atomic force microscopy (AFM) measurement.

As well as single-layer structures, stacked structures were grown to demonstrate the reduction in strain buildup. Two sets of samples were grown. Samples with a 2000 Å cap were grown for luminescence measurements, and samples with the top layer of the stack left uncapped were used for AFM measurements. In all samples, three dot layers, separated by 300 Å of material, were grown. The growth of the dots is the same as was just described. The GaP layer in all samples is 5 Å thick. For sample A, there are no GaP layers; in sample B, each GaP layer is 150 Å below each dot layer; for sample C, they are 50 Å below; and in sample D; they are 10 Å below each dot layer. Sample E has a GaP above each

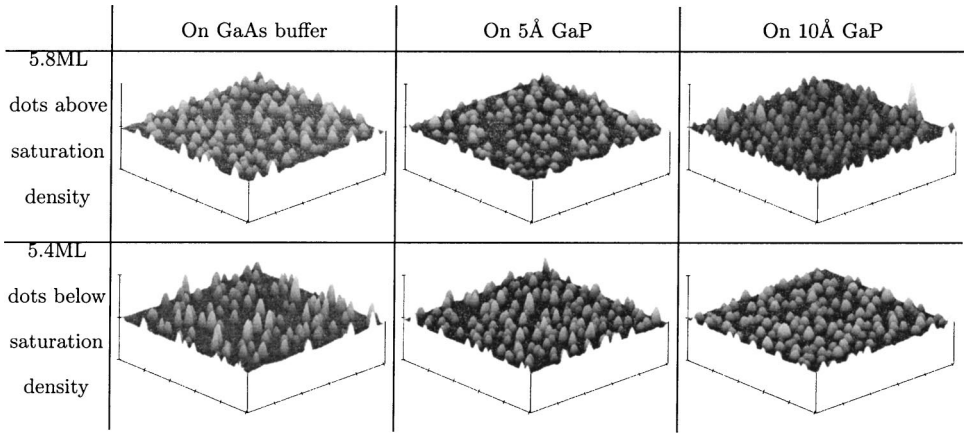

FIG. 1. AFM pictures of InGaAs dots, all images are 500 nm×500 nm and the height scale is 20 nm. The scale is not proportional to illustrate the height change in the dots.

dot layer approximately 100 Å above the wetting layer (WL). In sample F, the dots are grown directly on GaP layers.

The AFM measurements were carried out on a multimode Nanoscope III™ machine in contact mode. The room-temperature PL spectra were measured by exciting with a green diode pumped solid-state laser source. The PL was dispersed through a 0.5 m monochromator and collected with an InGaAs detector.

III. CHANGES IN NUCLEATION OF A SINGLE LAYER

The QD nucleation (size and density of the dots) depends on the surface energy as well as the strain difference of the system. It is expected that the GaP surface energy will be different from that of GaAs, which will change the nucleation of the dots. It was found that the two (2D)- to three-dimensional (3D) transition occurred sooner on the GaP buffer. There was also a larger range in the amount of deposited material in which coherent dots were formed. Normally, coherent dots are formed until a saturation density, after which the density of dislocated clusters increases. In this case, more material can be deposited without a high density of dislocated clusters forming. Figure 1 compares InGaAs dots of two different amounts of material grown on GaAs and on GaP buffer layers. The dots grown on a thin GaP (5 Å) buffer layer are slightly smaller in width and height than the dots grown on GaAs. For the larger dots, as the buffer layer thickness increases, taller dots are formed. The height of self-assembled dots is normally associated with the strain in the system (e.g., InGaAs are much flatter than InAs dots on GaAs). Here, there are two competing processes. Firstly, the GaP buffer layer increases the surface energy and strain, which should increase the height of the dots. The transition from 2D to 3D growth also occurs at a smaller thickness, so that for the same amount of material deposited, the dots are more evolved. For small amounts of material, the dots become denser when grown on GaP buffer layers, but once the saturation density is reached, the dots become larger and less uniform. The saturation density was the same for the dots with or without the GaP buffer layer; however, it occurs at a smaller amount of deposited material on a GaP buffer.

A second process causes the height reduction in the case of a small amount of GaP; namely, the interdiffusion occurring with the GaP buffer during growth. This reduces the strain in the WL and smaller dots are formed. During growth, interdiffusion occurs between the InGaAs dot material and the buffer layer. Interdiffusion during the growth of dots has been studied previously, and it is especially noticeable in the InAs system, in which studies have demonstrated a composition gradient in uncapped dots.[12] In our case, both the InGaAs dots and the GaP buffer layer are strained. The dots are under compressive strain while the GaP buffer is under tensile strain, so that an increase in the amount of interdiffusion is expected. Both group III and group V interdiffusion will be taking place, thereby lowering the In content of the dots and promoting As/P exchange in the dots. Therefore, InGaAsP dots (with a lower In content) are created. These are less strained in the GaAs matrix than the InGaAs dots, so that they can be expected to have a lower height.

It was found that the PL of the dots grown directly on GaP was shifted towards shorter wavelengths. Figure 2 shows the PL from dots grown on GaAs and GaP layers, and then capped with GaAs. The same amount of material was used to form the dots in each case. As discussed earlier, we believe some group V interdiffusion is occurring, as well as an enhancement of the group III interdiffusion. This reduces the strain in the dots, which could create a small redshift in the dots. However, the dots have slightly smaller height, with decreased In and a small amount of P. These factors all lead to a blueshift in the luminescence. Interdiffusion will also occur during the growth of the GaAs capping layer, and an

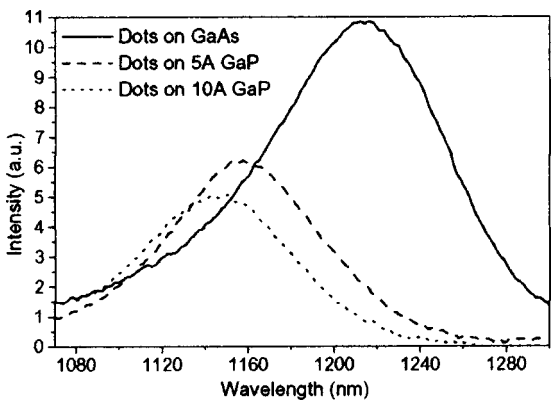

FIG. 2. PL spectra at RT for InGaAs dots grown on various buffer layers.

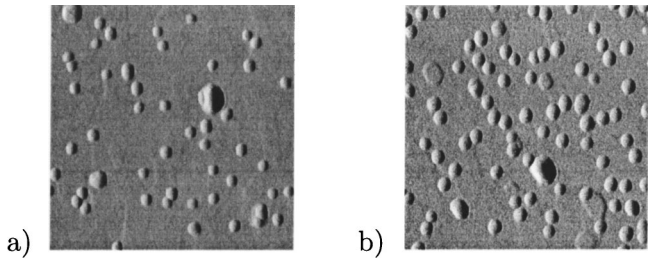

FIG. 3. AFM images of the third layer of dots (a) no GaP and (b) GaP 50 Å below the dots. Both images are 500 nm×500 nm.

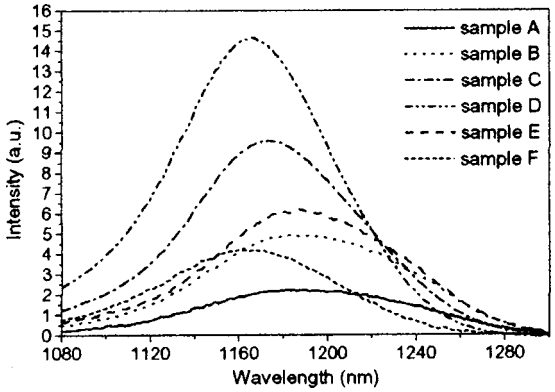

FIG. 4. PL spectra at RT of stacked InGaAs dots grown with GaP strain compensation layers at various distances away from the dots layers. Sample A is without GaP; B has 5 Å GaP 150 Å underneath dots; C has 5 Å GaP 50 Å underneath dots; D has 5 Å GaP 10 Å underneath dots; E has 5 Å GaP 100 Å above the WL; and in F, the dots were grown directly on top of the GaP layer.

increase in this interdiffusion would also cause a blueshift in the luminescence. Although small changes in the interdiffusion will take place due to the change in shape of the dots, the In–Ga interdiffusion during capping layer growth is not expected to increase in the case of the dots grown on the GaP buffer layer. The reduction in PL intensity in the samples on GaP buffer layers may be due to a shallower confining potential, which allows more carrier leakage at room temperature.

If the amount of dot material is kept constant, the linewidth of the PL spectra of dots grown on the GaP buffer layers decreased. When a thin (50 Å) GaAs layer is grown between the GaP layer and the dots, an increase in the PL linewidth and a small shift to the red is observed. This is consistent with a larger size distribution of the dots due to surface roughness. No difference between this sample and one without GaP was seen by AFM. Therefore, the blueshift observed in the other samples must be due to the changes in nucleation and interdiffusion during formation of the QDs on the GaP buffer layer.

IV. STACKED STRUCTURES

It is difficult to grow uniform stacked dot structures as the strain from each dot layer affects the nucleation of the next dot layer. This occurs noticeably when small separations are used. In this case the subsequent layers of dots form on top of the first layer of dots, forming dot columns.[13] Each subsequent layer of dots becomes more uniform and the dots are generally larger. This is caused by the underlying dots creating small regions of reduced strain which act as nucleation sites for the next layer of dots. However, in this study there is a much larger separation between the dot layers, so that the strain from the previous layer will be uniform and no nucleation sites are present. There will be a general buildup of strain and strain compensation layers can be used to limit this strain buildup. In a three-layer stacked structure with no strain compensation, the dots in the uppermost layer of the stack were found to be smaller in width and height than in a single layer of dots. Figure 3 shows AFM images of the top layer of a three-layer QD stack, in which each layer is separated by 300 Å. In image (a) there is no strain compensation (sample A), whereas in image (b) there is a 5 Å GaP layer 50 Å below each dot layer (sample C). The uppermost layer of dots in a stacked structure with GaP layers is similar in width to a single layer of dots, but are slightly smaller in height. The stacked structure grown with the dots directly on the GaP layer had only a small increase in the dot density of the uppermost layer compared to the structure with no strain compensation. This is because, although the overall structure will have less strain, there is still a buildup of strain that is seen at the time of nucleation of each dot layer. In the case of the GaP layer separated from the dot layer, the tensile strain field in the GaP layer counterbalances the strain field from the previous dot layers. The dot layer is then formed on a GaAs buffer, which has a reduced strain.

The other major factor in the dot nucleation is the roughness caused by each dot layer. The GaP layers seem to smooth out the surface as well as provide strain compensation. This can be seen in AFM images of the uncapped samples, but for quantitative data the surface roughness of capped samples was measured. A three-layer stack structure was grown at 550 °C, and a 2000 Å GaAs cap was then grown while the temperature was increased to 650 °C. The surface roughness (rms value) was measured by AFM. For a three-layer stack with no strain compensation, the value is 19 Å, which shows that even after 2000 Å of capping growth, there is still significant surface roughness. With GaP layers included 10 Å away from each dot layer, the surface roughness reduced to 3.4 Å, which is close to a monolayer roughness. The closer the GaP layer was to the dots, the smoother the surface became. This is mainly due to the lower density of large dislocated clusters in the structures, with GaP layers very close to each dot layer. The other factor is the strain during the growth of the GaAs barrier. When the GaP was placed close to the dot region, there was less strain on the GaAs barrier layer while it was grown.

The RT PL of each structure is shown in Fig. 4. The intensity of the luminescence greatly increases as the GaP strain compensation layer is placed closer to the dot region. There is also a small blueshift and a large narrowing in the PL of the structures with GaP layers. The narrowing is larger than that expected by interdiffusion. The narrowing is caused by the changes in the nucleation of the second and third layers. The structures with GaP close to the dot region have dots of a similar size in each layer of the stack, whereas in

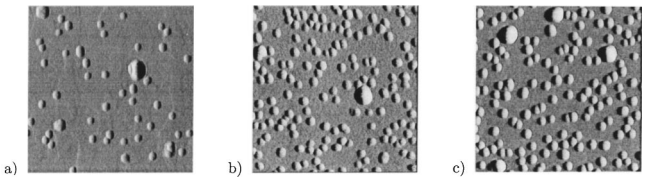

FIG. 5. AFM images of the third layer of dots in (a) previous sample, (b) smoothed barrier layers, and (c) smoothed barrier layers and a GaP layer 50 Å below the dots. All images are 500 nm×500 nm.

TABLE I. The energy shift in RT luminescence due to annealing with various layer structures. The anneals were carried out in the MOCVD reactor under Arsine ambient for 40 min.

Sample	Placement of GaP	Energy shift (meV) 650 °C	Energy shift (meV) 700 °C
A	no GaP layers	54.9	102.4
B	150 Å below dots	50.2	104.1
C	50 Å below dots	39.5	88.9
D	10 Å below dots	44.9	110.0
E	100 Å above WL	51.7	106.5

the sample without GaP layers, the strain builds up and changes the nucleation in the upper dot layers. The blueshift is probably caused by the dots having a slightly smaller height in the samples with GaP due to the reduction in strain during the nucleation process.

Sample E is a little different. Here, the GaP layer was deposited on top of the dots, 100 Å away from the WL, which is 200 Å below next dot layer. As the dots are roughly 40–50 Å high, this GaP layer is a distance away from the dots that is similar to the case with the GaP layer 50 Å underneath the dots. Therefore, one would expect a similar strain reduction behavior. The PL of this sample is not blueshifted when compared to sample A, but it is narrower. This indicates that the size is not changed, due to the GaP being far away from the dot region at the time of nucleation, but a reduction in the overall strain has helped to reduce the change in sizes of the dots in the upper dot layers. This sample also did not show any reduction in surface roughness, indicating that there is no reduction in the formation of dislocated clusters.

The main reason for the increase in dot density in the upper layer of the stack with GaP is the increased smoothness of the GaAs prior to the dot deposition. After each dot layer, there is an increase in step bunching during the GaAs barrier growth due to the uneven growth caused by the dots underneath. This is also enhanced due to the low growth temperature. Step bunching will cause an increase in the surface energy and if dots are formed on this surface they will be smaller in the width.[14] For the dot layer to have a large density of coherent dots, the surface prior to formation of the dots must be as smooth as possible. Additional uncapped and capped three-layer stacked samples were grown with growth interrupts and increased arsine flow in the barrier to help smooth out the barrier layer. Samples, one without GaP layers and one with GaP layers 50 Å below the dot layer, were grown. The AFM pictures of the uppermost layer in the three-layer stack is shown in Fig. 5 compared to the previous sample without any GaP. The density of the dots is much higher in the samples with smoothed barrier layers it is similiar to that of a single layer. The density in the sample with GaP is even slightly higher again. The PL shows the same trend of increased intensity and blueshift as seen before, indicating that these effects are related to the strain reduction rather than any size change due to rough surfaces. The PL linewidths of both samples are narrower, indicating that the size distribution caused by the different nucleation in the upper layers has been reduced. These samples also show the same thermal stability as is shown by the other samples that are discussed in Sec. V.

V. THERMAL STABILITY

For actual device structures to be grown by MOCVD, the temperature must be raised above 650 °C for the growth of the upper cladding layers. This creates an annealing effect on the dots. The stacked dot structures, with and without GaP, were annealed at 650 or 700 °C for 40 min under an AsH_3 overpressure. In Table I, the energy shift of the RT PL caused by this anneal is presented. The structures with the GaP strain compensation layers close underneath the dot layer have a much reduced energy shift. The introduction of the GaP layers creates a compensating strain on the dot region, thus reducing the strain gradient for interdiffusion. Sample C shows a reduction of the energy shift caused by interdiffusion by 15%–20%. However, in sample D, the energy shift increases compared to samples with slightly larger separation between the dot layer and the GaP layer. This is an indication that with a long anneal, there is phosphorous diffusion into the dot region. Interestingly, in sample E, the structure with GaP layer close to the top of the dots (100 Å away from the WL, 200 Å below next dot layer) does not have a large reduction in the interdiffusion. In this structure, the GaP layer will be around 50 Å from the top of the dots in each layer. The sample with a GaP layer 50 Å away from the bottom of the dots shows a large reduction in interdiffusion, whereas when the GaP is a similar distance away on the top of the dots, there is no reduction in the interdiffusion. This indicates that the strain during the formation process has the most influence on the stability of the dots.

VI. CONCLUSION

Single layers of InGaAs dots grown directly on GaP strain compensation layers are smaller both in width and height. The interdiffusion between the InGaAs dot material and the GaP buffer leads to the formation of InGaAsP dots that are less strained inside the GaAs matrix. The PL peak of these dots is blueshifted from the PL peak of the InGaAs dots grown on GaAs. GaP layers inserted below the dot region reduce the height of the dots slightly but do not affect the density of the dots. The luminescence from this sample is very similar to that of a structure without GaP.

Stacked dot structures with GaP layers inserted into the barriers have much narrower and more intense luminescence peaks. It is found that the density of dots in the top layer of the stack is increased as the GaP layer is deposited closer to

the dot layer. The GaP layers reduce the strain buildup so that the nucleation of each layer of dots is similar to that of a single layer, producing stacked dot layers that have a more uniform dot size and a higher dot density. These effects are reduced when smoother barrier layers are grown between each layer of dots. A small blueshift is seen in the PL of the structures with GaP layers, due to a slight reduction in the height of the dots. There is an increase in the thermal stability of the dot structures that have GaP strain compensation. It was found that the thermal stability of the dots is influenced mostly by the strain in the dots at the time of formation, rather than post-formation strain compensation. The use of GaP layers is promising to create stacked dot structures with a large number of layers.

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