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Relaxation study of $In_xGa_{1-x}As/GaAs$ quantum-well structures grown by MBE on (0 0 1) and (1 1 1)B GaAs for long wavelength applications

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Abstract

A comparative study of the relaxation mechanisms of thin, single $In_xGa_{1-x}As/GaAs$ quantum wells (QWs) grown simultaneously on (0 0 1) and (1 1 1)B GaAs substrates is presented. Transmission electron microscopy (TEM) studies indicate that the primary relaxation mechanism for high-indium content (0 0 1) samples is the formation of 3D islands. For the growth conditions used, this occurs at an indium content above 0.24 in our samples. Although initially coherent with the substrate, above a certain critical thickness catastrophic relaxation occurs through a very high density of edge dislocations ($>10^{12}\ cm^{-2}$) as the islands coalesce. A similar relaxation mechanism is not observed in the (1 1 1) orientation. Here, 3D islanding is suppressed and instead a novel configuration of misfit dislocations (MD) appears at $x>0.24$. This new MD configuration presents an additional misfit-relieving component not taken into account in the previous theoretical analysis of the critical layer thickness (CLT) for (1 1 1)B. However, the mechanism is relatively inefficient when compared to the catastrophic relaxation in the (0 0 1) case, and therefore, the (1 1 1)B substrate still offers considerable advantages. This is illustrated by photoluminescence results, which show that (1 1 1)B $In_xGa_{1-x}As$ QWs are able to reach wavelengths as long as 1.1 μm at room temperature. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Highly strained $In_xGa_{1-x}As/GaAs$ quantum wells (QWs) on GaAs substrates continue to be investigated for potential optoelectronic devices

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operating at wavelengths > 1 μm. Layers grown on (0 0 1)-oriented substrates have been studied in much greater detail due to the greater availability of substrates and the good epitaxial layer quality that can be achieved using a relatively wide range of growth conditions. There are, however, very few reports of optoelectronic devices, e.g. lasers operating beyond 1 μm [1–3]. It would seem that, apart from some few isolated reports, strain relaxation leads to a critical layer thickness for $In_xGa_{1-x}As$ QWs (CLT) that sets an upper wavelength limit for (0 0 1) devices at about 1 μm.

A number of groups have reported a theoretical and experimental critical layer thickness for strain relaxation of $In_xGa_{1-x}As$ layers grown on (1 1 1)B orientation which is larger than that for (0 0 1) [4–6]. This implies that, for a given quantum well width, a higher In content can be used and that, $In_xGa_{1-x}As$/GaAs heterostructures on (1 1 1)B oriented substrates offer a potential for the development of optoelectronic devices at wavelengths beyond the 1 μm limit in the (0 0 1) case, or for improved reliability in (1 1 1)B devices working around 1 μm. It has been difficult to evaluate the potential improvements to be gained using (1 1 1)B growth because there exists a wide spread of data in the literature concerning the CLT for this growth orientation. These range from reports indicating that the CLT for the (1 1 1)B orientation is three times larger than that for (0 0 1) [5] to reports suggesting a similar CLT for (0 0 1) and (1 1 1) [6].

One fundamental problem with all these studies is the definition of "critical layer thickness" (CLT). Relaxation of thin strained layers is often highly discontinuous. There are a number of possible definitions of CLT that we must consider [7]. With increasing strain the first is (i) the thickness at which the first misfit dislocation segments are formed through the turning over of threading dislocations in the interface plane. Nevertheless, not only the number of threading dislocations that rise from the substrate is low (around $1000\,cm^{-2}$) but also the density of point sources (defects, inclusions, etc.) in good-quality epitaxial growth. Dislocation levels are low, the dislocations are spatially isolated, so the overall relaxation is relatively insignificant. The second two CLTs we consider are more in terms of their influence on the optical quality. These are: (ii) the thickness at which significant relaxation occurs as new misfit dislocations (MD) are generated and (iii) the onset of a 3D growth mode [8]. In the latter case, the 3D growth mode relieves misfit through surface strain energy and not by dislocations (at least initially).

For the (0 0 1) orientation, the onset of relaxation is determined by the formation of misfit dislocations when the indium content is lower than 0.25 and by 3D-island formation for indium contents above 0.30 [9]. However, the relaxation mechanisms for the (1 1 1) orientation are less well known. Besides, there are some indications of 3D-growth inhibition in strained layers on (1 1 1) GaAs [10], which would reduce the relaxation mechanisms for this orientation to those due to generation of dislocations. For the (1 1 1) orientation, a recent study on multiple quantum well (MQW) InGaAs/GaAs structures shows that depending on the degree of strain existing in the wells, two different kind of MD are generated [11]. According to this study, the conventional MD configuration [12] governs relaxation when the indium content is low, and a new type of MD configuration appears for indium contents higher than 0.22. This new MD configuration controls the relaxation process in high indium content MQW structures. The objective of this paper is to investigate further the relaxation of (1 1 1)B single $In_xGa_{1-x}As$ QW's in the range $0.1 < x < 0.35$, to compare these results with the (0 0 1) case and to show which of the two orientations is more suitable for device applications at wavelengths beyond 1 μm. Low-temperature photoluminescence (PL) and transmission electron microscopy (TEM) have been used to determine the mechanism resulting in the degradation of the optical properties of high In content QW's and to elucidate its origin.

2. Experimental procedures

MBE growth was performed in a Riber 2300 system using two different substrates mounted side-by-side in each MBE run. A (0 0 1) GaAs on-axis substrate was mounted with a (1 1 1)B GaAs substrate of orientation 1 degree off $(\bar{1}\,\bar{1}\,\bar{1})$ towards $[\bar{2}\,1\,1]$. Surfaces were deoxidised by thermal annealing at ~ 630°C for 20 min under As_4 flux. The structures subsequently grown on both substrates consisted of a 100 Å thick $In_xGa_{1-x}As$ single quantum well embedded between a 0.3 μm thick GaAs buffer layer and a 0.3 μm GaAs cap layer. The nominal indium content in the well was varied in the range $x = 0.1$–0.3. The growth rate was about 1.1 Å/s and the buffer/cap layer were grown at 610–615°C. In the last 1000 Å of the buffer layer the temperature was continuously ramped down to 500°C. After growing the quantum well at 500°C, the substrate temperature was continuously increased to 610°C in the first 1000 Å of the cap layer. The beam equivalent pressure for the $In_xGa_{1-x}As$ layer was 30, which we assume to be equivalent to an atomic flux ratio of about 2. The growth conditions have been optimised for simultaneous growth on both substrate orientations and details of this can be found elsewhere [13].

PL studies were performed both at low and at room temperature using a He–Ne laser ($\lambda = 6328$ Å). The emission was dispersed through a Jobin-Yvon H25 spectrometer and detected by either an S1 photomultiplier or a Ge detector. For the TEM studies, specimens were prepared by mechanical thinning and chemical etching. The observations were performed in a JEOL 1200EX at 120 kV accelerating voltage.

3. Experimental results and discussions

Fig. 1 shows the 23 K full-width at half-maximum (FWHM) of the PL peaks as a function of the In-content for the structures grown on both orientations. For the (0 0 1), it can be seen that for an In-content up to 0.21, the FWHM is low at 4–8 meV. Since interfacial roughness fluctuations of ± 1 monolayer (ML) can give rise to a 3–4 meV linewidth component, the additional influence of MDs on the quantum well emission remains relatively small. The data show an abrupt change in FWHM for 0.24 In content. Here, the linewidth broadening is accompanied by a decrease in the PL peak height compared to PL peaks for QWs with lower indium content, as can be seen in Fig. 2a. However, the integrated emission intensity of the QWs with $x = 0.24$ is similar to those with lower indium content. From the last two results, we infer that the onset of plastic relaxation in our structure occurs at an indium content between 0.22 and 0.24. The constant integrated intensity implies that the number of non-radiative recombination centres does not vary very much. We assume that the origin of this effect could be a transition from layer-by-layer growth to a 3D Stranski–Krastanov growth mode. This observation is confirmed below, by the TEM results.

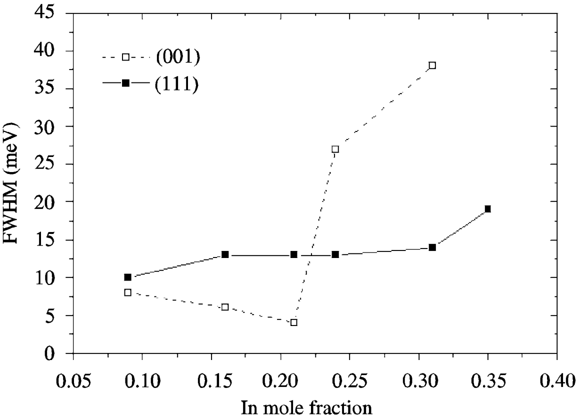

Fig. 1. FWHM of the PL lines as a function of indium content in the series of $In_xGa_{1-x}As/GaAs$ single QW samples ($T = 23$ K). Open and solid squares represent the values for (0 0 1) and (1 1 1), respectively.

For the (1 1 1) orientation, the structures show a FWHM of about 13 meV up to $x = 0.31$ indium content (see Fig. 1). However, the PL peak height decreases as the indium content increases (Fig. 2b). This PL peak height reduction, which is not observed in the (0 0 1) case, might be explained by a degradation of the radiative to non-radiative recombination ratio due to an increased plastic relaxation taking place in the QW as the indium content increases. This would imply a notorious quality degradation of the samples. However, we should remark that the obtained FWHM values in these (1 1 1) samples are quite narrow since they have an added PL peak broadening due to the piezoelectric field in the (1 1 1) strained wells, not present in the (0 0 1) case. This added broadening effect is stronger for wide wells with high indium contents, and its influence on the transition energies is about 11 meV in similar structures [14]. Therefore, the

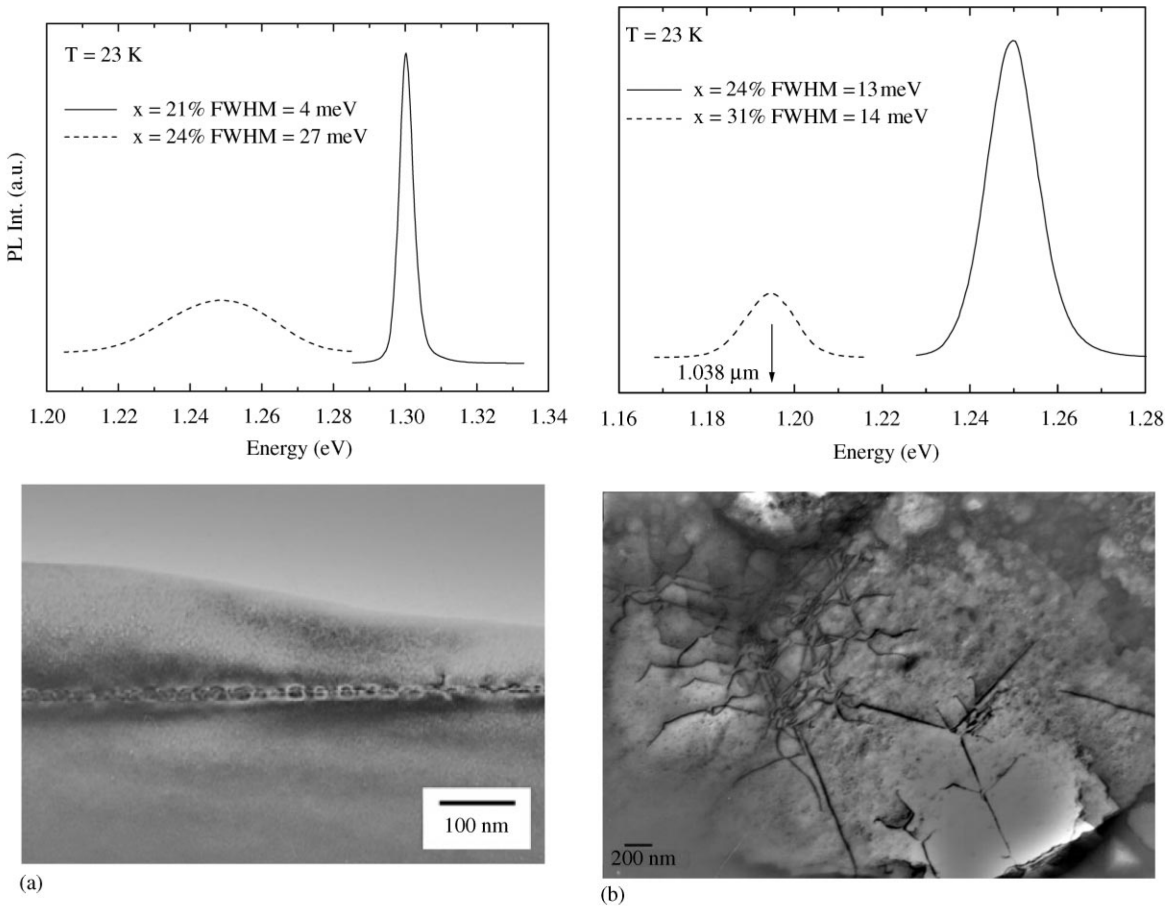

Fig. 2. (a) $In_xGa_{1-x}As/GaAs$ single QW on (0 0 1) GaAs: upper part, 23 K PL spectrum of samples with $x = 0.21$ and 0.24; lower part, cross-section TEM image of $x = 0.24$ sample. An irregular $In_{0.24}Ga_{0.76}As/GaAs$ interface but no indications of MDs can be observed. (b) $In_xGa_{1-x}As/GaAs$ single QW on (1 1 1)B GaAs: upper part, 23 K PL spectrum of samples with $x = 0.24$ and 0.31; lower part, PVTEM image of $In_{0.31}Ga_{0.69}As/GaAs$ QW. A three pointed star shaped MD dislocation can be seen with its branches parallel to the $\langle \bar{2}\,1\,1 \rangle$, directions contained in the growth

above PL peak height reduction would have a quality-independent origin, as the extra band-bending barriers for electrons (bottom of the QW) and holes (top of the QW) induced in the surrounding GaAs by the strong piezoelectric field in the well. This would decrease the QW ability to capture electrons and holes from both GaAs sides, thus reducing the QW recombination and hence PL signal. Also the reduction of the electron-heavy hole overlap taking place as the In content is increased has also been considered, but it is not sufficient to account for the observed decrease. Nevertheless, the PL data clearly indicate the onset of plastic relaxation in (1 1 1) orientation by the increase of the FWHM value, which occurs between $x = 0.31$ and 0.35 indium content.

Plan view (PV-) TEM observations of the samples grown on the (0 0 1) orientation did not reveal any MDs even for indium contents as high as $x = 0.31$. Nevertheless, cross-section analysis (XTEM) indicated some type of irregular interface morphology for indium contents higher than $x = 0.24$ (see micrograph in Fig. 2a). This result has already been interpreted by Yao et al. [15] as due to the transition from 2D to 3D growth mode. A correlation with our PL results leads to the assumption that the CLT for the (0 0 1) orientation is determined by the formation of 3D islands when the indium content is 0.24 or higher. This result is in close agreement with Wang et al. [9], who indicated that initial strain relaxation in $In_xGa_{1-x}As$/GaAs single-strained quantum wells is dominated by the formation of MDs for $x < 0.25$ and by 3D islands for $x > 0.30$. Both processes are concomitant in the range $0.25 < x < 0.30$.

PVTEM observations of the (1 1 1)B samples indicate that no MDs are present in our structures until we reach to 0.31 indium content, where the MD configuration observed is not due to 60° mixed-type MD as usually observed in low indium content $In_xGa_{1-x}As$ layers once CLT is surpassed [12]. It is, instead, of a novel type (Fig. 2b) first observed in MQW structures [11], consisting of isolated three-pointed, star-shaped, dislocation grouping with 120° angle between its arms, which lay on the $\langle \bar{2}\,1\,1 \rangle$, directions contained in the growth plane. Finally, when the indium content is increased, the density of these configurations also increases such that they may interact and multiply their branches. The resulting polygonal network, which extends over the bottom $In_xGa_{1-x}As$/GaAs interface, is shown in Fig. 3. The behaviour in our single QW differs from that observed in multiple QW structures in two respects. First, the evolution of the relaxation in the MQW structures implies the multiplication of the MD parallel to the $\langle \bar{2}\,1\,1 \rangle$, directions, which often changes their lines rotating 90° towards $\langle 0\,1\,\bar{1} \rangle$, directions. In this case, two MD networks could clearly be seen, a fact not observed in our single QW samples, where the branches of the stars develop a polygonal network at the bottom interface of the $In_xGa_{1-x}As$/GaAs epilayers. Secondly, the $\langle \bar{2}\,1\,1 \rangle$, dislocation network of the MQW structures could be localised crossing the different quantum wells, but in our single QW structures, this network is localised only at the bottom $In_xGa_{1-x}As$/GaAs interface. Further work is needed to understand the nucleation of this novel configuration and the dependence of its relaxation dynamics on the number of quantum wells.

From the above results and the PL FWHM data of Fig. 1, we can conclude that the MDs present in the 0.31 indium content QW are not detected by the PL technique. Whilst a few misfit dislocations are observed in this 0.31 indium case, the optical properties are relatively unaltered. Only at 0.35 indium content, a clear increase in the PL FWHM is seen. These observations reflect the limited resolution of the PL technique to strain relaxation [16]. The well thickness of the 0.31 indium content sample is close to the predicted CLT of the Matthews and Blakeslee equilibrium model [17], which normally predicts quite accurately the onset of relaxation. We believe at this point, however, that only a few MD's can originate from substrate threading dislocations (density 10^3–10^4 cm^{-2}) or extrinsic defects. They appear to be too few in number to significantly relax the sample. Arnaud et al. [18] have shown that in this case there is a very limited effect on the electronic band structure of $In_xGa_{1-x}As$/GaAs. Of more concern for optoelectronic devices is the CLT at which significant plastic relaxation has occurred [19]. Our 100 Å well width is the above CLT for an In composition ranging from $0.31 > x > 0.35$.

Fig. 3. PVTEM image of the bottom interface of a relaxed $In_{0.35}Ga_{0.69}As$/GaAs single QW on (1 1 1)B GaAs substrate. A novel MD configuration is operating as a dislocation multiplication source. plane.

The $\boldsymbol{g \cdot b}$ analysis of the branches of the novel MD configuration shows that the Burgers vector is a $\frac{1}{2}\langle 1\,\bar{1}\,0 \rangle$ type *contained in the growth plane* and forming a 30° angle with the dislocation lines. The

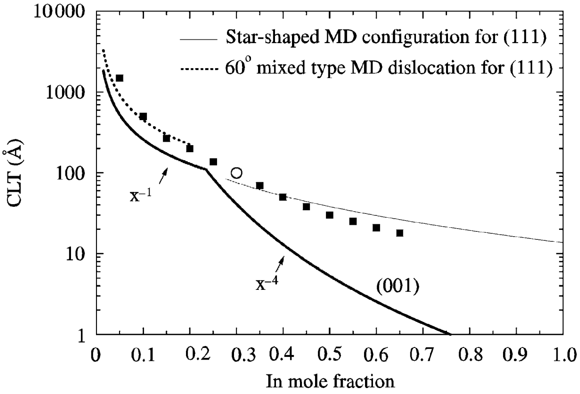

Fig. 4. Summary of calculated CLT as a function of In content for the (0 0 1) and (1 1 1) orientations. Solid squares give estimates to achieve emission at 1.1 μm at room temperature from $In_xGa_{1-x}As/GaAs$ QWs on (1 1 1)B GaAs. The open circle is an experimental value whose PL emission is given in the next figure.

strain relaxation through MDs is proportional to the misfit-relieving component of the Burgers vector, b_r. For this novel configuration, b_r is $\sqrt{3}$ times larger than those usually considered in the literature for the (1 1 1) orientation [12]. This result has an immediate consequence: The CLT for $In_xGa_{1-x}As/GaAs$ heterostructures on the (1 1 1) orientation may be smaller than expected for high indium contents, where the novel MD configuration appears. Assuming a Matthews–Blakeslee relaxation mechanism [17], Fig. 4 shows the calculated CLT in (1 1 1) orientation, for In contents below 0.20 (conventional MD configuration; dashed line) and for In contents above 0.25 (novel MD configuration; thin solid line). In the same figure, we have also plotted a set of points relating the well width and In content needed to reach 1.1 μm wavelength emission (e1-hh1 transition) at 300 K, using $In_xGa_{1-x}As/GaAs$ quantum wells grown on the (1 1 1) orientation. From this figure, we can deduce that it is not advisable to use low indium contents with wide wells because we would be very close to the CLT given by the conventional MD dislocation mechanism. It is preferable to use high indium contents and narrow wells because we are below the CLT given by this novel MD configuration.

For the (0 0 1) orientation, the thick solid line in Fig. 4 represents the calculated CLT as previously discussed. The breakpoint at 0.24 In content corresponds to the transition from a CLT set by conventional MD generation (x^{-1} analytical dependence) to a CLT being dominated by 3D islanding, for which an x^{-4} analytical dependence is expected [9]. We are considering also that our 0.24 (0 0 1) sample (see Fig. 2) is already in this 3D-growth regime. As seen in Fig. 4, the CLT for (1 1 1) growth always exceeds the CLT for (0 0 1) growth. It is interesting to consider the thickness and composition of $In_xGa_{1-x}As$ QWs required for long wavelength device operation. For the (0 0 1) case, the Matthews and Blakeslee equilibrium CLT is always exceeded for structures designed for 1.1 μm wavelength emission. The layers lie within a regime in which the first misfit segments have probably operated and are close, or beyond, the point at which significant relaxation has taken place through misfit generation. For the (1 1 1) case, all the points depicted in Fig. 4 for In-contents higher than 0.40 are within the calculated CLT. It is important to note that they are calculated for (1 1 1)-oriented QWs in which a quantum confined stark effect exists due to the strain-induced piezoelectric field and which is not present in samples grown on the (0 0 1) orientation. The effect provides extra assistance to reach longer wavelengths. Therefore, $In_xGa_{1-x}As/GaAs$ QWs on the (1 1 1) orientation, using high indium contents and narrow wells, offer a considerable potential to exceed (0 0 1) device performance for long wavelength optoelectronic applications.

We have observed a good room temperature PL intensity at 1.1 μm from a 0.31 nominal indium content sample, as shown in Fig. 5. This sample has already started relaxation and shows a very low dislocation density in PVTEM. Yet its narrow FWHM, from PL measurements at low temperature, indicated no significant relaxation, at least not enough to affect the electronic states in the quantum well (see Figs. 1 and 2b). Hence, our experimental observations are in agreement with the analysis of Fig. 4 discussed above, at least for samples grown under the growth conditions used here.

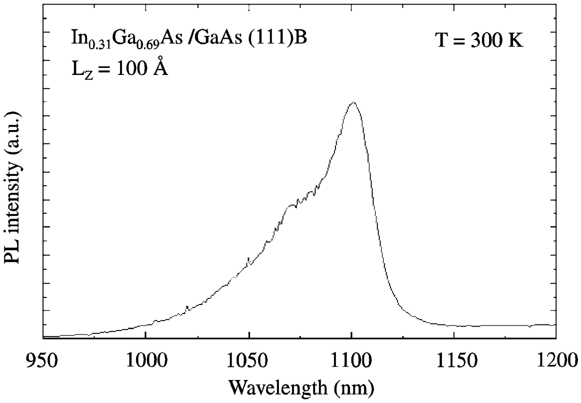

Fig. 5. Room-temperature PL emission from $In_{0.31}Ga_{0.69}As$/GaAs single QW on (1 1 1)B GaAs substrate.

4. Conclusions

We have performed a PL and TEM study of a series of $In_xGa_{1-x}As$/GaAs single QWs ($x \leqslant 0.35$) with a fixed well thickness grown by MBE on (0 0 1) and (1 1 1)B GaAs substrates mounted side-by-side. For the growth conditions used, our results indicate that the effective CLT for structures grown on the (0 0 1) direction is a consequence of three-dimensional islanding (3D) when the indium content exceeds 0.24. In contrast, in (1 1 1) direction the relaxation is dominated by the formation of a novel MD configuration when the indium content reaches 0.31. The appearance of this novel relaxation mechanism, which seems to operate only at high indium contents, reduces the CLT from that previously expected for (1 1 1) structures. Nevertheless, the absence of a 3D-growth mode gives an effective CLT for (1 1 1), which is larger than that for (0 0 1). For long wavelength optoelectronic applications, $In_xGa_{1-x}As$/GaAs QW grown on (1 1 1) substrates seems to be the best choice. Strong room temperature PL emission at 1.1 μm has been achieved for a (1 1 1) $In_xGa_{1-x}As$/GaAs QW with a 0.31 nominal indium content.

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