Strain and Relaxation Processes in $In_{1-x}Ga_xAs_yP_{1-y}$/ InP Single Quantum Wells Grown by LP-MOVPE

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Received February 6, 1999

Strained and partially relaxed $In_{1-x}Ga_xAs_yP_{1-y}$/InP single quantum wells (SQWs) with different cap layer thicknesses and biaxial strain values grown by low-pressure metalorganic vapor phase epitaxy (LP-MOVPE) were investigated by double crystal X-ray diffraction, photoluminescence microscopy (PLM) imaging and photoluminescence spectroscopy techniques. Our results indicate a significant improvement of the optical quality of the quaternary wells with increasing values of the cap layer thickness. Tensile and compressive strained $In_{1-x}Ga_xAs_yP_{1-y}$/InP SQWs grown with the same structure exhibited different relaxation processes, even when the strain magnitude was the same. PLM images of highly compressive quantum wells exhibited a large number of dark lines corresponding to misfit dislocations as a result of the partial relaxation process in the well material. PLM images of similar tensile strained samples revealed only the presence of dark spots with no evidence of misfit dislocations.

I Introduction

Strained-layer $In_{1-x}Ga_xAs_yP_{1-y}$ quaternary alloys grown on InP substrates have been widely used as active layers in optoelectronic devices, mainly in long wavelength semiconductor lasers and electroabsorption modulators for optical communication systems. The introduction of a biaxial strain into the active region of multiple-quantum well laser diodes and modulators resulted in improved electro-optic performances as compared with similar ones employing lattice-matched quaternary materials[1]. Although high quality InGaAsP strained layers have been successfully obtained by different growth techniques, the strain relaxation mechanisms involved in these structures are still unclear. If a critical thickness is exceeded, plastic relaxation of the elastic energy can occur through generation of misfit dislocations. More recently, relaxation through elastic mechanisms was observed in tensile strained InGaAsP/InP structures, where the strain relief takes place via surface undulations along [110] direction. This elastic relaxation process was found to be strongly dependent on the growth temperature and on the growth rate[2-4]. It has been reported evidence of strain recovery in partially relaxed InGaAs/GaAs single quantum wells (SQWs) with increasing cap layer thickness, suggesting that the cap layer thickness should be also considered as a parameter to determine critical layer thickness in lattice mismatched SQWs[5].

The aim of this contribution is to investigate the effects of the cap layer thickness and the sign of the strain on the relaxation processes of strained and partially relaxed InGaAsP/InP SQWs grown by low-pressure (LP) metalorganic vapor phase epitaxy (MOVPE). Our analysis is mainly focused on the optical properties of quaternary strained materials using photoluminescence microscopy (PLM) imaging and photoluminescence (PL) spectroscopy techniques.

II Experimental details

Strained InGaAsP/InP SQWs were grown in a Thomas Swan LP-MOVPE horizontal reactor equipped with a fast vent-run switching manifold. The samples were grown on InP:S (100)-oriented substrates at 70 Torr pressure and at a growth temperature of 670^{o}C. Typical strained InGaAsP/InP heterostructures consisted of a 300nm thick undoped InP buffer layer followed by an undoped 30nm thick strained quaternary InGaAsP well material, with emission wavelength at room temperature in the range of 1250-1350nm and nominal strain values (ϵ) of $\epsilon = \pm 0.75\%$ and $\epsilon = \pm 1.0\%$.

The structures were ended with an undoped InP cap layer (dcap) with thicknesses varying from 5 to 500nm.

The PLM imaging system uses the 514.5nm line of an argon ion laser as the excitation source. The spatial resolution of the PLM imaging apparatus is about 1μm. Integral luminescence radiation emitted from the samples was collected and imaged into a lead-sulfide infrared (IR) camera. The laser excitation power used was in the range of 1 to 500W/cm^2. All PLM measurements were performed at room temperature.

Spectrally resolved PL measurements were performed at 12K temperature using the 514.5nm line of an argon ion laser with an excitation power in the range of 10^{-3}% to 10^{+2} W/cm^2. The emitted PL radiation was dispersed by a 0.75m grating monochromator and the detection was obtained by a liquid-nitrogen cooled Ge photodetector using a standard lock-in technique. Double crystal X-ray diffraction experiments were carried out on (200) reflection using Cu-Kα1 radiation. $In_{1-x}Ga_xAs_yP_{1-y}$ biaxial strain values and alloy compositions were determined by combining results of X-ray and PL measurements.

III Results and discussion

Fig. 1 shows the PLM images of compressive ($\epsilon = +0.90\%$) and tensile ($\epsilon = -0.98\%$) strained InGaAsP/InP SQWs with different cap layer thicknesses. PLM images of compressive strained SQWs (Figs.1a-1c) revealed a large number of dark lines that were attributed to misfit dislocations. The occurrence of plastic relaxation in these samples indicates that the well thickness exceeded the critical layer thickness. As the cap layer is increased the density of dark lines slightly decreases. This suggests an increase of the residual strain in compressive strained SQWs. PLM images of tensile strained SQWs (Figs.1d-1f) exhibited the presence of dark spots with no evidence of misfit dislocations. The dark spots were attributed to crystal defects acting as non- radiative centers, strongly quenching the luminescence in the extent of the carrier diffusion length. The dark spot density is almost three orders of magnitude higher than the etch pit density measured on the InP:S substrate, indicating that they are not correlated with threading dislocations originating at the substrate[6]. PLM images (not shown) of InGaAsP/InP SQWs with strain values of $\epsilon = +0.70\%$ and $\epsilon = -0.75\%$ exhibited only the presence of dark spots without any evidence of misfit dislocations.

The results shown in Fig.1 indicate a significant difference in the strain relaxation processes in InGaAsP/InP SQWs grown with essentially the same structure and strain magnitude but having opposite signs. In the case of compressive strained SQWs with $\epsilon = +0.90\%$ a plastic deformation occurs trough the creation of misfit dislocations. On the other hand, strain relaxation in tensile structures with $\epsilon = -0.98\%$ takes place via elastic processes and can be explained by the lateral composition modulation caused by the miscibility gap of the quaternary alloy, which favors phase separation during growth into InAs- and GaP-rich regions[7].

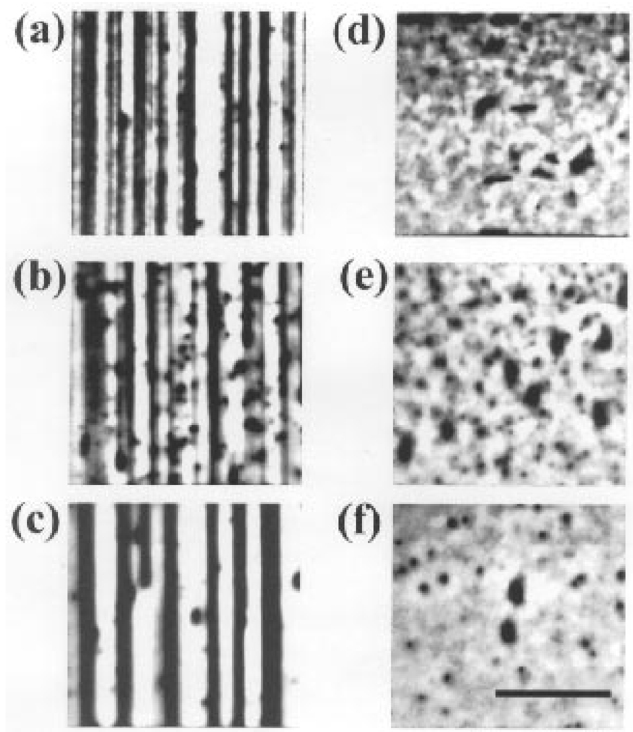

Figure 1. PLM images of compressive strained, $\epsilon = +0.90\%$, InGaAsP/InP SQWs with cap layer thicknesses of (a) 5nm, (b) 100nm and (c) 400nm, and tensile strained, $\epsilon = -0.98\%$, InGaAsP/InP SQWs with cap layer thicknesses of (d) 5nm, (e) 100nm and (f) 400nm. Marker represents 100μm.

In order to provide additional information about the relaxation processes on strained InGaAsP/InP SQWs, we show in Fig.2 the results of low temperature PL measurements (normalized to unity) performed on the same samples analyzed in Fig.1. The PL spectra of tensile strained SQWs (Fig.2a) exhibited a single emission line corresponding to the fundamental transition between first electron and light-hole states ($e_1 - lh_1$). When the cap layer is increased from 5 to 400 nm, the PL intensity increases and the full width at half maximum (FWHM) decreases from 21.1 meV to 10.6 meV, respectively. The PL spectra of compressive strained SQWs (see Fig.2b) exhibited three emission lines. The main peak located at the high-energy side was attributed to the transition between first electron and heavy-hole states ($e_1 - hh_1$). The remainder peaks labeled as **A** and **B** are located at $\sim$ 14 meV and $\sim$ 8 meV below the fundamental QW $e_1 - hh_1$ transition, respectively and their origin will be discussed later. Again, the increase of cap layer thickness in compressive strained SQWs resulted in improved material quality. As the cap layer is increased from 5 to 400 nm the PL intensity increases and the FWHM corresponding to the $e_1 - hh_1$ transition is reduced from 10.6 meV to 3.9 meV, respectively. The PL results exhibited in Fig.2 clearly indicate that samples grown with larger d_{cap} values exhibited narrower PL linewidths. Variations in PL peak positions with increasing d_{cap} values shown in Fig.2 are attributed to differences in the alloy compositions between samples. A larger PL linewidth was observed for all tensile strained samples grown with different d_{cap} values when compared to similar compressive strained samples. This behavior is due to larger composition fluctuations caused by phase separation during growth of tensile quaternary alloys into of InAs- and GaP-rich regions [7].

The remarkable reduction of PL linewidth with cap layer thickness can be explained by an improvement of the interface quality and to a decrease of the quaternary alloy composition fluctuation during growth of the binary cap layer. Samples grown with larger cap layer thicknesses require longer growth times. As a consequence, strained quaternary well materials experience longer annealing times. A possible explanation for the observed PL linewidth reduction with cap layer thickness is the sharpening of lateral composition modulation interfaces caused by the thermal annealing process[8]. Also, the annealing process tends to homogenize regions of different alloy compositions.

In order to identify the origin of the additional features observed in compressive strained SQWs (see Fig.2b), the excitation power dependence of the PL spectra at low temperatures of these samples, and the corresponding tensile strained ones, were investigated. Figs.3a and 3b shows the PL spectra (normalized to unity) of tensile strained SQWs with d_{cap} = 5 and 100nm, respectively, as a function of excitation intensity. When the excitation intensity is increased by four orders of magnitude, the $e_1 - lh_1$ PL peak energy of the tensile strained SQW sample with d_{cap}=5nm (Fig.3a) shifted by 14.0 meV towards higher energies. At the same excitation intensity range a negligible shift of the $e_1 - lh_1$ PL peak energy was observed for the tensile strained sample with d_{cap} = 100nm. The excitation power dependence of the compressive strained SQW sample with d_{cap} = 5nm (Fig.3c) showed a shift of the $e_1 - hh_1$ PL peak energy as large as 31.1 meV towards higher energies. Also, as the excitation intensity increases the relative intensity of the spectral feature **A** decreases and almost vanishes at higher excitation power levels. Similar to the tensile strained SQW with large d_{cap} layer, a negligible PL peak energy shift was observed on the compressive strained sample with d_{cap} = 5nm. However, the relative intensity of transitions **A** and **B** decreases with the excitation power intensity.

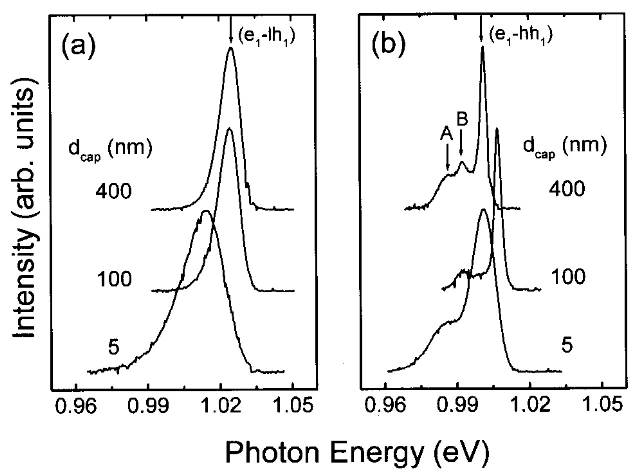

Figure 2. PL spectra at T=12K of (a) tensile strained, $\epsilon = -0.98\%$, and (b) compressive strained, $\epsilon = +0.90\%$, InGaAsP/InP SQWs with different cap layer thicknesses.

The intensity saturation of low-energy **A** and **B** PL emissions (see Figs.3c and 3d) suggest that defects might be involved in these transitions[9]. Similar PL experiments were performed on compressive strained SQWs with $\epsilon = +0.70\%$ and with different dcap values. The PL spectra (not shown) of those samples exhibited only a single emission band corresponding to the $e_1 - hh_1$ transition and no evidence of low- energy features were observed. The above results and those obtained in Figs.1a-1c provide additional support that transitions observed below the $e_1 - hh_1$ peak energy (see Figs. 2 and 3) are related to defects.

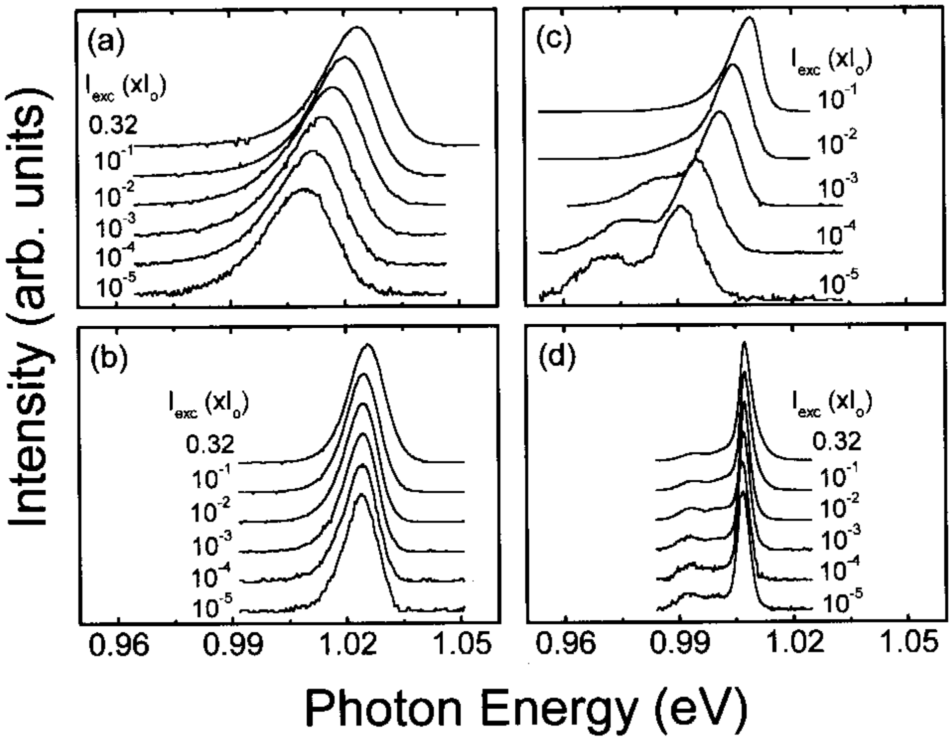

Figure 3. PL spectra at T=12K at different excitation power levels of tensile strained InGaAsP/InP SQWs, $\epsilon = -0.98\%$, with (a) $d_{cap} = 5$nm and (b) $d_{cap} = 100$nm, and compressive strained InGaAsP/InP SQWs, $\epsilon = +0.90\%$, with (c) $d_{cap} = 5$nm and (d) $d_{cap} = 100$nm.

The strong PL peak energy shift with excitation power observed on strained InGaAsP/InP SQWs samples with $d_{cap} = 5$nm (see Figs. 3a and 3c) can be tentatively explained by a decrease of the surface band bending. As the excitation intensity increases, optically created minority carriers move toward the surface and recombine with charges trapped in surface states. This reduces the net charge at the cap layer surface and then reduces the built-in electric field. As a consequence, the PL peak energy shifts to higher energies. On the other hand, strained quantum wells grown with larger d_{cap} values are less sensitive to the built-in electric field and exhibited a negligible PL energy shift with excitation intensity.

IV Conclusions

Strained and partially relaxed strained InGaAsP/InP single quantum wells with different strain values and different cap layer thicknesses were investigated by PL, PLM and high resolution X-ray diffraction techniques. Plastic relaxation was observed in highly compressive strained SQWs through the generation of misfit dislocations. Similar tensile strained SQWs exhibited elastic relaxation processes via composition fluctuations. Increased values of the cap layer thickness resulted in improved optical quality of the analyzed samples. Near-surface strained InGaAsP/InP SQWs exhibited pronounced PL peak energy shift with power excitation. This effect was attributed to a reduction of the surface built-in electric field.

Acknowledgements

The authors gratefully acknowledge the technical assistance of Mr. L.C. Silveira Vieira and Mr. H. Gazetta Filho.

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