Optical properties and Stokes shifts in lamp-annealed InGaAs/GaAs strained layer superlattice

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The effect of incoherent lamp annealing on the photoluminescence and optical absorption characteristics in $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}(x = 0.2$ and $0.24)$ strained layer superlattices grown by molecular-beam epitaxy has been investigated. The annealing time and temperatures were varied from 5–25 s and 850–950 °C, respectively. It is seen that the main photoluminescence and absorption peaks a low temperatures (11.5 K) shift to higher energies with increase in annealing temperatures. We believe this is due to In-Ga atomic interdiffusion across the heterointerfaces and have estimated the magnitude of this interdiffusion by solving the appropriate Schrodinger equation for this region. The estimated interdiffusion constants D are $\sim 10^{-16}$–10^{-15} cm^2/s for the above annealing conditions, which are about three orders of magnitude higher than those reported for long-term furnace annealed $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}(x = 0.13$–$0.15)$. Optimal rapid thermal annealing has a significant effect in improving the quality of the superlattices in terms of Stokes shift and absorption coefficients. Extremely small Stokes shift ~ 1.1 meV was observed for $\mathrm{In}_{0.2}\mathrm{Ga}_{0.8}\mathrm{As/GaAs}$ superlattices annealed at 890 °C for 5 s.

I. INTRODUCTION

Strained layer superlattices (SLS) have attracted considerable interest in recent years not only because of their potential application in electronic and optoelectronic devices,[1-6] but also because of their interesting properties arising as a result of lattice mismatch.[2,4,7-10] It has been reported that strained layer superlattices have good structural integrity even after ion implantation[11] and long-term thermal annealing[12] but are unstable against high level laser excitation.[13] In the present work we have investigated the effects of halogen lamp annealing on InGaAs/GaAs strained layer superlattices by the measurement of photoluminescence (PL) and optical absorption in these materials at low temperatures. It is seen that the energies of the main PL and absorption peaks shift to higher energies with annealing temperature. We believe this is due to In-Ga atomic interdiffusion across the heterointerfaces and have estimated the magnitude of this interdiffusion. The PL linewidth initially decreases with annealing temperature and then increases at higher annealing temperatures. The absorption peaks become narrower and the absorption coefficients increase after rapid thermal annealing (RTA) under optimal conditions. In addition, the $\mathrm{In}_{0.2}\mathrm{Ga}_{0.5}\mathrm{As/GaAs}$ SLS exhibits extremely small Stokes shift (~ 1.1 meV) after annealing at 890 °C for 5 s, indicating a marked improvement in the quality of the superlattice.

II. EXPERIMENT

$\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ strained layer superlattices $(x = 0.20,\ 0.24)$ were grown by molecular-beam epitaxy (MBE) on (100) Si-doped GaAs substrates at 520 °C. The growth rates for GaAs and InGaAs were approximately 1.45 and 1.9 μm/h, respectively. The schematics of the experimental samples are shown in Figs. 1(a) and 1(b).

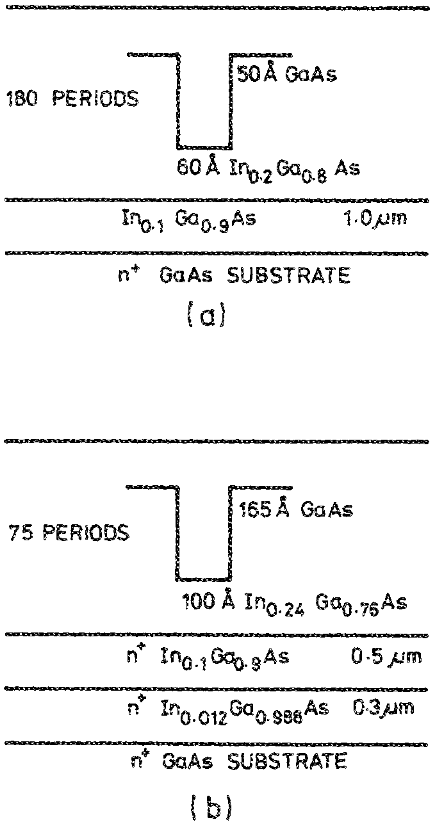

FIG. 1. Schematic of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ SLSs samples with different indium compositions. (a) $x = 0.2$, and (b) $x = 0.24$.

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The samples were annealed in a halogen lamp annealing station with a protective GaAs cap under flowing argon. The anneal time and temperature were varied from 5–25 s and 850–950 °C, respectively. Low-temperature (11.5 K) photoluminescence measurements were made with the samples being photoexcited with an argon laser (5145 Å). The luminescence was analyzed with a 1-m Jarrell–Ash spectrometer and detected with a cooled photomultiplier tube. The spectra were recorded on a strip chart recorder after suitable amplification by a lock-in amplifier. For absorption studies, a tungsten-halogen lamp was used as the radiation source while the detection and recording techniques remained the same. Photoluminescence and absorption measurements were done on the same samples for a meaningful determination of the Stokes shift.

III. RESULTS AND DISCUSSION

Photoluminescence and absorption spectra for as-grown samples and those annealed at different temperatures for a fixed duration of time (5 s) were measured for $In_xGa_{1-x}As/GaAs$ MQW with $x = 0.20$ and 0.24. It was observed that the transition peaks in both the spectra shift to higher energies with increasing annealing temperatures. The linewidths initially decrease and then increase with increasing annealing temperature. Representative PL and absorption data for as-grown and annealed samples (880–900 °C) are shown in Figs. 2 and 3, respectively. The main PL transition and the absorption peak at the lowest energy are both believed to be due to $n = 1$ electron heavy-hole excitonic resonances in the strained quantum wells. It is also seen from Fig. 3 that there is significant enhancement in the absorption coefficient α for both $x = 0.20$ and 0.24 after an optimal anneal cycle.

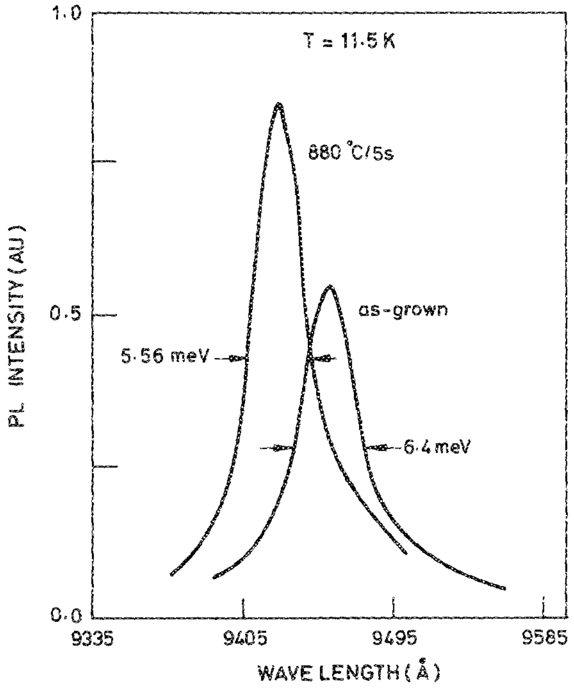

FIG. 2. Low-temperature photoluminescence spectra of as-grown and annealed samples of $In_{0.2}Ga_{0.8}As/GaAs$ strained layer superlattices.

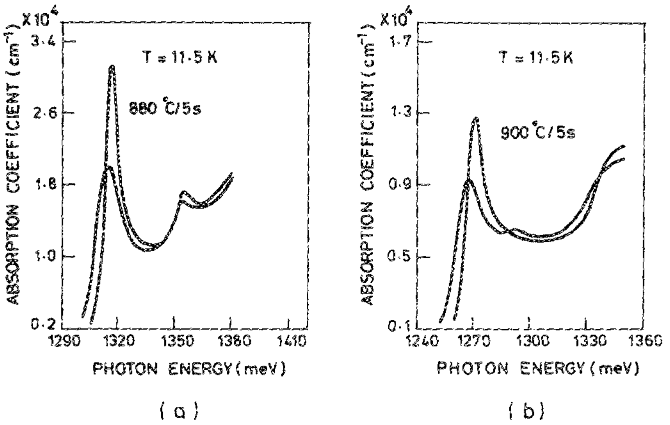

FIG. 3. Measured absorption characteristics for as-grown and annealed SLS: (a) $In_{0.2}Ga_{0.8}As/GaAs$ and (b) $In_{0.24}Ga_{0.76}As/GaAs$.

The measured variation of the peak PL energies upon annealing (ΔE), as compared to the as-grown samples ($x = 0.24$), is depicted in Fig. 4. The shift of the excitonic peak to higher energies can be related to an increase of the electron and hole subband level energies, possibly due to a modification in well shape. This modification possibly results from In-Ga atomic interdiffusion[12] across the heterointerfaces of the SLS. The diffusions are driven by the concentration gradients and mobilities of the individual species. The mobilities of the group III metals are inversely proportional to the bonding strengths in the lattice. Overall, the interdiffusion in the multicomponent interface is controlled by the stoichiometry boundary condition. The interdiffusion is expected to be relatively larger at higher anneal temperatures leading to a larger shift, and this is borne out experimentally. The shift of the main peak at different annealing temperatures was modeled by taking into account error function diffusion and solving the Schrodinger equation for the quantum well with the graded interfaces caused by In-Ga atomic interdiffusion. The graded ternary $(In_{1-y}Ga_y)_xGa_{1-x}As$ interface region is characterized by a spatial profile given by

$$x = 1 - \frac{1}{2}\left(\mathrm{erf}\,\frac{h-z}{2\sqrt{Dt}} + \mathrm{erf}\,\frac{h+z}{2\sqrt{Dt}}\right), \qquad (1)$$

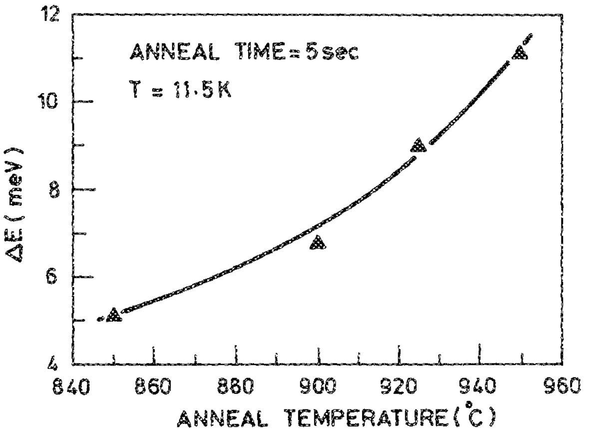

FIG. 4. Low-temperature PL peak energy shift (ΔE) compared to as-grown sample of $In_{0.24}Ga_{0.76}As/GaAs$ SLS with anneal temperature for a fixed anneal time of 5 s.

where z is the distance measured from the center of the $In_xGa_{1-x}As$ well, h is the half-width ($L_z/2$) of the well in an as-grown sample, and t is the annealing period. It may be noted that in this study we report on strained quantum wells with $x = 0.20$ and 0.24. It is also important to note the width of the graded region is much smaller than L_z. The dependence of the mismatch (strain) on composition is given by

$$\epsilon = 0.07x \tag{2}$$

and the dependence of E_g on z and composition is given by

$$E_g(z) = 0.36 + 1.064(1-y)x + 9.8y\epsilon. \tag{3}$$

The band-gap discontinuities in strained MQWs are not definitely known. However, recent measurements by us[14] with $In_{0.2}Ga_{0.8}As/Al_{0.16}Ga_{0.84}As$ quantum wells indicate that $\Delta E_c = 0.204$ eV, which indicates that $\Delta E_c \simeq 0.638\,\Delta E_g$. Recent theoretical work also indicates that the lattice-mismatch effects tend to increase the value of ΔE_c.[15] For this study, we have assumed

$$\begin{aligned} \Delta E_c(z) &= 0.65\Delta E_g(z), \\ \Delta E_v(z) &= 0.35\Delta E_g(z). \end{aligned} \tag{4}$$

This is the only way that we have taken into account the effect of strain. The grading will cause a change in the alloy composition, and, therefore, the mismatch. The effect of this on $\Delta E_c(z)$ and $\Delta E_v(z)$ has been neglected. The calculated peak energy shift ΔE as a function of $2\sqrt{Dt}$ (D is the diffusion constant) is depicted in Fig. 5(a). The interdiffusion coefficient D at various temperatures is obtained by fitting experimental data with the theoretically calculated ones as indicated in Fig. 5(a). The estimated values of D are plotted against inverse temperature in Fig. 5(b), from which an activation energy of 1.22 eV is obtained. The estimated interdiffusion coefficients are reduced by 25% when rise and fall sections of the anneal cycle are taken into account.[16] The values of D obtained in this study are about three orders of magnitude higher than those reported by Joncour, Charesse, and Burgeat.[12] The diffusion coefficient of In in annealed $In_xGa_{1-x}As$/GaAs SLS ($x = 0.13$–0.15) has been determined by these authors by x-ray diffraction measurements. A value of $\sim 10^{-18}$ cm^2/s was derived after annealing at 850 °C for 15–71 h. However, the diffusion coefficient during initial stages of annealing is normally higher because of contribution from defects and/or compositional inhomogenieties of the interfaces (defect assisted diffusion). As the annealing period and temperature increase, the defects are annealed and consequently the diffusion coefficient can decrease. A decrease in the interdiffusion coefficient by an order of magnitude by annealing only for 30 min at 750 °C was observed by Seo *et al.*[17] in the case of $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$ quantum wells. We believe this is the main cause of the discrepancy. Slight differences in strain ($x = 0.2$ vs 0.15) may also play a minor role.

The photoluminescence linewidth is principally determined by the ternary well and heterointerface qualities.[18,19] We see from Fig. 2 that the as-grown sample with $In_{0.2}Ga_{0.8}As$ wells exhibits a very narrow linewidth (~ 6.4 meV) compared to the sample with $In_{0.24}Ga_{0.76}As$ wells (13.2 meV). The linewidth in the former case is among the best reported[6] value for this SLS material. The second sample was grown at a different time and probably under inferior ambient conditions. The increase in alloy composition of the well material can only be partly responsible for higher linewidth in the second case. Although the linewidth of this sample was higher, it was selected for the present study in order to highlight the effect of RTA on the Stokes shift in samples having different photoluminescence linewidths.

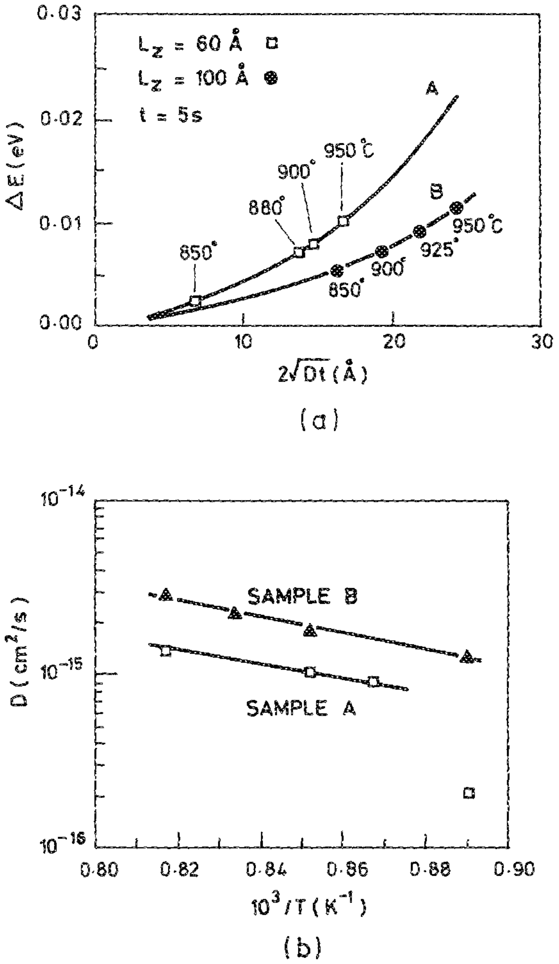

FIG. 5. (a) Calculated continuous lines and measured (□●) variation of the shift in energy position of the main photoluminescence transition with interdiffusion distance $2\sqrt{Dt}$; in $In_xGa_{1-x}As$/GaAs SLS and (b) estimated diffusion constants as a function of inverse temperature in $In_xGa_{1-x}As$/GaAs SLS, obtained from the data shown in (a) the values of $x = 0.20$ and 0.24 in samples A and B, respectively.

The dependence of the PL linewidth on anneal temperature for both $x = 0.20$ and $x = 0.24$ in the well regions is shown in Fig. 6. The linewidth initially decreases with anneal temperature and then increases. The initial decrease could be due to reduction in fluctuations in alloy composition originally present. For annealing at temperatures higher than 880–900 °C, the interdiffusion of In and Ga might result in an inhomogeneous alloy compositon in the wells due to differential rate of interdiffusion, which is expected due to different atomic radii of Ga and In.

The effect of annealing period at a fixed anneal temperature (850 °C) on the PL and absorption peaks was also stud-

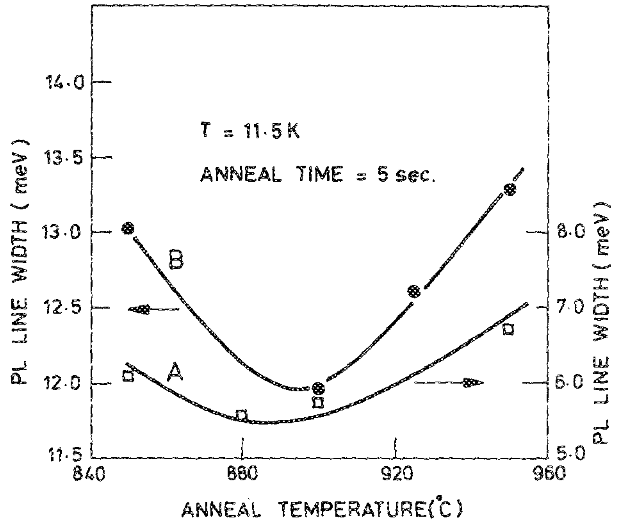

FIG. 6. Variation of PL linewidth with anneal temperature in $In_xGa_{1-x}As/GaAs$ SLS, (a) $x = 0.2$ and (b) $x = 0.24$. The linewidths in the two as-grown samples are 6.4 and 13.2 meV, respectively.

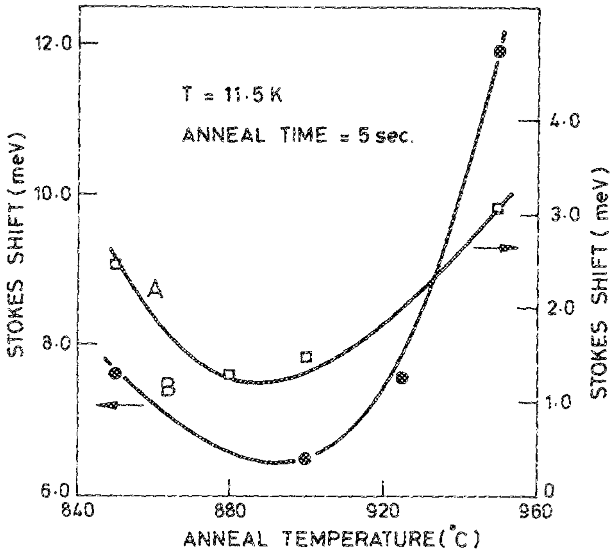

FIG. 8. Variation of measured Stokes shift with anneal temperature in lamp-annealed $In_xGa_{1-x}As/GaAs$ SLS (sample A: $x = 0.20$, sample B: $x = 0.24$) for a fixed anneal period of 5 s. The Stokes shift in the corresponding as-grown samples are 7.35 and 9.74 meV, respectively.

ied. The shift in peak energies is depicted in Fig. 7 and does not appear to be large at 850 °C. It seems that the energy shift of PL and absorption peaks is more sensitive to temperature than time. For example, annealing at 850 °C for 25 s produces more or less the same effect as annealing at 900 °C for 5 s. However, the linewidth in the former case is higher than in the latter. This indicates that rapid thermal annealing of SLS at a higher temperature for a shorter period is more advantageous than at lower temperature for a longer period.

The difference between the optical emission and absorption energies is usually referred to as the Stokes shift. This degradation of the optical energy at low temperatures, among other parameters, depends upon the defects at the heterointerfaces in MQW or superlattice structures. Therefore, the magnitude of the Stokes shift can be taken as an index of heterointerface quality. We have plotted in Fig. 8 the Stokes shift for both types of samples as a function of anneal temperature. It is seen that the Stokes shift initially decreases with anneal temperature in a manner similar to the PL linewidth (Fig. 6) and then increases at higher anneal temperatures. It may also be noted that the Stokes shift (1.1 meV) in the $In_{0.2}Ga_{0.8}As/GaAs$ SLS, which exhibits very narrow PL linewidth (5.56 meV at 11.5 K) after an optimal anneal cycle, is considerably smaller than the shift observed in the $In_{0.24}Ga_{0.76}As/GaAs$ SLS (~ 6.4 meV). Furthermore, the improvement in the optical quality in terms of Stokes shift in the first case is about 85%, while there is only 34% decrease in Stokes shift in the second sample. The increase of the Stokes shift at higher anneal temperatures reflects the deterioration of heterointerfaces due to interdiffusion which, in turn, introduces compositional inhomogeneity. It is, therefore, evident that high-quality SLS or MQW material of good optical quality can be achieved by post-growth RTA and our current study shows that 890–900 °C/5 s anneals give the best result for the SLS systems studied here.

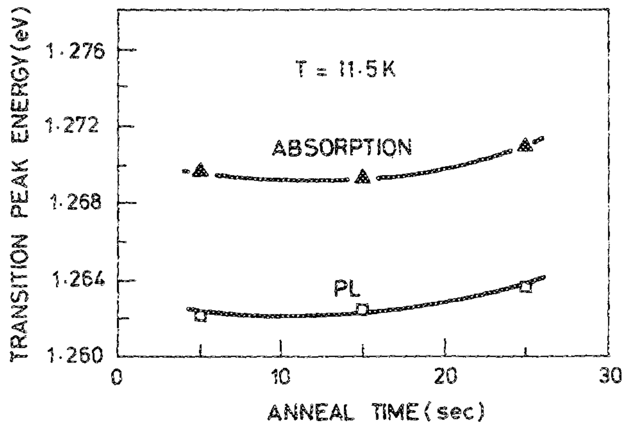

FIG. 7. Variation of PL and absorption peak energies with anneal time at 850 °C for $In_{0.24}Ga_{0.76}As/GaAs$ SLS.

IV. CONCLUSIONS

The effects of halogen lamp annealing on the PL and absorption characteristics of $In_xGa_{1-x}As/$ GaAs ($x = 0.20$, 0.24) strained layer superlattices was studied. The results clearly indicate that the optical quality of SLS can be improved in terms of PL linewidth and Stoke's shift under optimal annealing conditions. The $In_{0.2}Ga_{0.8}As/$ GaAs SLS shows extremely small Stoke's shift (~ 1.1 meV) when annealed at 890 °C for 5 s. Improvement in the quality of interfaces and significant enhancement in absorption coefficient after annealing makes these SLS materials suitable for high performance optical devices such as modulators and detectors. The results of the present study further support the previous observations on the stability of strained layer superlattices subjected to long-term annealing.

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