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PROBLEM RELATED TO THE MBE GROWTH AT HIGH SUBSTRATE TEMPERATURE FOR GaAs-Ga\textsubscript{1-x}Al\textsubscript{x}As DOUBLE HETEROSTRUCTURE LASERS

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Abstract - The MBE growth at high substrate temperature (T\textsubscript{S}>600°C) is now often used to obtain GaAs and Ga\textsubscript{1-x}Al\textsubscript{x}As layers with improved electrical and optical properties. In the case of GaAs-Ga\textsubscript{1-x}Al\textsubscript{x}As DH lasers, the optimisation of the threshold current density is obtained by increasing T\textsubscript{S}; the ultimate value we reached is l.5KA/cm\textsuperscript{2} for an active layer thickness of 0.16μm. Cependant au-delà d'une valeur critique de T\textsubscript{S} d'environ 670°C, nous avons observé une augmentation de J\textsubscript{s} puis une disparition de l'émission laser. Cet effet est corrélat à un problème particulier de la croissance MBE à très forte température qui sera étudié.

1. Introduction - During last years, molecular beam epitaxy was considered like a low temperature growth process compared to liquid phase and vapour phase epitaxy. Thus the MBE temperature growth range for GaAs is limited by the minimal temperature to obtain a not insulating semiconductor (= 480°C [1]) and the congruent sublimation temperature of GaAs (= 640°C [2]). However this last critical temperature is increasing for Ga\textsubscript{1-x}Al\textsubscript{x}As versus the x concentration; it is also possible to grow above this temperature with larger group V/III ratio due to the reduced As lifetime on the substrate surface.

It has been found that transport and optical properties of GaAs, Ga\textsubscript{1-x}Al\textsubscript{x}As and GaAs-Ga\textsubscript{1-x}Al\textsubscript{x}As heterostructures for many devices, improve when grown at high substrate temperature T\textsubscript{S} (> 600°C). This is due to a reduced incorporation of impurities with relatively high vapour pressures, with increasing T\textsubscript{S}[3], particularly for the incorporation of O in Ga\textsubscript{1-x}Al\textsubscript{x}As. As these impurities could act as non radiative...
centers, the photoluminescence intensity of GaAs, Ga$_{1-x}$Al$_x$As layers \[4\] and multi-quantum well structures GaAs-Ga$_{1-x}$Al$_x$As \[5\] is found to be higher with increasing $T_S$. This growth condition improves also the interface properties between Ga$_{1-x}$Al$_x$As and GaAs when the binary compound is grown on the ternary like in the inverted modulation doped heterostructure Ga$_{1-x}$Al$_x$As/GaAs\[6\] or in MESFET structure with Ga$_{1-x}$Al$_x$As buffer layer.\[7\].

In this study we report the optimisation of double heterostructure lasers GaAs-Ga$_{1-x}$Al$_x$As properties versus substrate temperature $T_S$. A degradation of these properties is observed above a critical value of $T_S$ due to a peculiar problem related to the MBE growth of GaAs and Ga$_{1-x}$Al$_x$As in this extremely high substrate temperature range.

2. Experimental - The growth experiments are realized in a first generation MBE chamber equipped of ionic pumped airlock, a liquid nitrogen shroud between and above the crucibles and a cooling of the growth chamber walls.

GaAs substrates are degreasing in chloroform, etch-polished with Br$_2$ methanol and rinsed in methanol. Indium was used to mount the substrate on a molybdenum block; to obtain a good uniformity of In layer and then of substrate heating, the back of the sample is also etch-polished. For high substrate temperature experiments, some requirements are necessary to obtain a good sticking of the sample because of the relatively high vapor pressure of In ($3.10^{-5}$torr at 700°C); Ga which has a lower vapor pressure could be an alternative solution to this problem, but Ga is highly reactive at high temperatures with Mo.

After the substrate is loaded into the airlock, this one is evacuated to a pressure of $10^{-8}$torr, such as the pressure in the growth chamber increases only to $6.10^{-10}$torr when the substrate and its block are transferred. The usual conditions for epilayer growth were reported elsewhere \[8\]; the growth rate usually used in this study is relatively high: about 2.8$\mu$/h for GaAl$_{0.32}$As and 1.9$\mu$/h for GaAs.

The substrate temperature which is the important growth parameter throughout the investigation is measured by a thermocouple close to the backside of Mo block. The heating of the substrate is realized with a regulated continuous intensity power. Due to a variation of the thermal emissivity of Mo block at the beginning of the growth, we observe a decrease of $T_S$ during the first hour before it was stabilized; this time was used to grow a buffer layer.

3. Optimisation of DH lasers growth - We have already reported that high growth temperature increase the 300K photoluminescence intensity of GaAs layers lightly doped with Be \[9\]. We observe also a similar effect for Ga$_{0.88}$Al$_{0.12}$As doped with Sn; but for the same range of $T_S$, the increase of photoluminescence intensity is ten times more important like we can see on figure 1. For double heterostructures lasers, the growth of such high optical quality Ga$_{1-x}$Al$_x$As is essential in reducing the contribution of bulk non-radiative recombination near the heterojunctions by Ga$_{1-x}$Al$_x$As confinement layers \[10\].

Since photoluminescence yield makes semiconductor growth at high $T_S$ advisable, DH structures are grown only in the range $T_S>$600°C. The semiconductor layer structure consists of a n$^+$ Ga$_{1-x}$Al$_x$As buffer layer ($n=10^{16}$cm$^{-3}$ ~2$\mu$m thick), a first n Ga$_{1-x}$Al$_x$As cladding layer ($n=5.10^{16}$cm$^{-3}$ ~2$\mu$m thick), a n GaAs active layer ($n=5.10^{16}$cm$^{-3}$), a second Ga$_{1-x}$Al$_x$As cladding layer ($p=5.10^{17}$cm$^{-3}$ ~2$\mu$m thick) and a p$^+$ GaAs contact layer ($p > 10^{19}$cm$^{-3}$ ~0.7$\mu$m thick). The Sn flux is decreased before the active layer growth in order to reduce the doping level of GaAs active layer and to minimize the time delay in Sn incorporation \[8\]; thus the p-n junction is at the proper location as we can see on figure 2.
Fig. 1: The substrate temperature dependence of 300K integrated PL intensity of GaAs and Ga$_{0.88}$Al$_{0.12}$As single layers.

Fig. 2: (a) Shallow angle cross section (12) of a typical MBE DH structure. (b) Scanning electron micrograph of the cross section of the structure with a superimposed electron beam induced current trace (c) a typical I-V characteristic of the laser diode.

For current threshold density evaluation broad area Fabry Perot lasers are fabricated. Broad area laser technology consists of first to thin the wafer down to 100µm, second to electroplate with Au and alloy n contact and then to electroplate with Au the p side. 100µm width stripes are then photolithographic defined in the p contact. Light output characteristics of individual laser are tested under pulsed conditions (500ns, 1KHz) on cleaved barrets of several individual laser.

We observe on figure 3 a linear output characteristic up to more than 30mW per facet and also a good uniformity of laser characteristic across the wafer, what is expected for MBE growth ; the ultimate average $J_{th}$ value we reached is 1.5kA/cm$^2$ for an active layer thickness of 0.16µm. External quantum efficiency $\eta_{ext}$ for the 2 mirrors is around 40% for these broad area contact lasers. We have recently measured on proton bombarded stripe geometry laser 10µm width $\eta_{ext}$ up to 80%.

Figure 4 shows the decrease of threshold current density with consecutive MBE runs after the growth chamber is opened to atmosphere ; this is due to the outgas improvement of effusion cells and substrate holder [11, 12]. We have noticed a similar behaviour for the 77K mobility of modulation doped single heterostructure GaAs-Ga$_{1-x}$Al$_{x}$As (Sn).

The substrate temperature dependence of the threshold current $J_{th}$ of DH lasers is shown on figure[5] for two series of consecutive growth runs. We observe until a critical value $T_S$ of about 670°C a decrease of $J_{th}$ ; we can notice that this effect is minimized for the second serie because we start form the high to the lowest substrate temperature in the contrary order of the improvement reported in figure 4. This improvement of $J_{th}$ is correlated to an increase of photoluminescence intensity of Ga$_{1-x}$Al$_x$As confinement layer and GaAs active layer with increasing $T_S$. We observe
ourselves an increase of GaAs active layer PL intensity of about a factor of 30 between 620 and 640°C and 150 between 620 and 680°C. These observations were first reported by Tsang et al which attributed this improvement of $J_{th}$ to a decrease of bulk nonradiative recombination near the heterojunctions in the wide bandgap regions.\[13\].

**Fig. 3:** Light current characteristics under pulsed operation of laser diodes of the same sample with different cavity lengths.

**Fig. 4:** The order of growth run dependence of the averaged $J_{th}$ of DH lasers $(W = 100 \, \mu m \; L = 400 \, \mu m)$.

**Fig. 5:** The substrate temperature dependence of the average $J_{th}$ of the consecutive series of DH lasers grown at different $T_S$; the number of each sample is related to the order of growth run.
However we observe on figure 5 above the critical $T_S$ value of about 670°C a sudden increase of $J_{th}$ until a non laser emission; this is confirmed on both series of growth runs.

4. Problem related to the MBE growth at high $T_S$ - In order to explain this effect, a specific structure is grown with changes of $T_S$ during the same growth. This structure consists of several alternate Ga$_{1-x}$Al$_x$As and GaAs layers; the time of growth of each kind of layers and all molecular fluxes ($F_{Ga}$, $F_{Al}$, $F_{As}$) are kept constant; and the substrate temperature $T_S$ is changed by steps after each second interface GaAs/Ga$_{1-x}$Al$_x$As. The thickness of GaAs layers is of the same order of magnitude than for the active layer of DH laser; on the contrary the thickness of Ga$_{1-x}$Al$_x$As is ten times more important.

A first characterization of this structure is an optical microscope observation of a shallow angle cross section (F) obtained by mechanical polish as shown on figure 6. The growth rate of GaAs and Ga$_{1-x}$Al$_x$As versus $T_S$, deduced from the thickness of each layer are reported on figure 7.

![Fig. 6: (a) Temperature time cycles for the continuous MBE growth of a specific multilayers structure with $T_S$ changes ($F_{Ga}$, $F_{Al}$, $F_{As}$ are kept constant) (b) Shallow angle cross section (12) of this structure.](image)

![Fig. 7: The substrate temperature dependence of GaAs and Ga$_{1-x}$Al$_x$As growth rate deduced from fig. 6b.](image)

We can observe a very important decrease of GaAs and Ga$_{1-x}$Al$_x$As growth rates above 670°C. This could be attributed to a preferential reevaporation of Ga atoms at the surface of the sample for high $T_S$ in accordance with the vapor pressure of Ga two orders of magnitude higher than that of Al in this extremely high $T_S$ range.
The both growth rate curves being similar, the Al concentration X of the ternary compound Ga$_{1-x}$Al$_x$As must increase with $T_S$. This could be already seen on the shallow-angle cross section of the structure by an increasing contrast for the three last Ga$_{1-x}$Al$_x$As layers. This confirmed by an Auger analysis (5KeV 0.5μA electron beam with a 0.50μm diameter) of a chemical bevel of the same structure produced by the technique originally developed by Mellet [14]. The curves of Auger high energy peaks Al (1395eV) and Ga (1070eV) reported on figure 8 show effectively a decrease of Ga and an increase of Al. We must notice that for the last Ga$_{1-x}$Al$_x$As layer grown at 735°C, we don't observe on the Auger spectra Ga transitions; a very important 0 peak is present and this one doesn't desappeared after many ionic bombardments. This is related to the formation of Al$_2$O$_3$ by oxydation to the air of a ternary compound Ga$_{1-x}$Al$_x$As with very high Al concentration (X=1)[15]. The X measurements by electronic microprobe on a single layer Ga$_{1-x}$Al$_x$As grown at 640°C in the same conditions of fluxes than the multilayer structure gives a X concentration of 0.31; so in this case the X concentration of Ga$_{1-x}$Al$_x$As would be about three times more important between 640 and 735°C. The variation of this effect is higher than that reported very recently by Wood et al. [16] for a lower substrate temperature.

In the case of DH structure versus $T_S$, we must observe in the same manner, first a decrease of the thickness of active layers and second an increase of X concentration of Ga$_{1-x}$Al$_x$As confinement layers. The first effect couldn't explain entirely the curve $J_{th}=f(T_S)$ because the normalized threshold current density curve $J_{th}=f(T_S)$ shows the same behaviour, and the active layer thickness is higher than 900Å for the three DH lasers 709, 715 and 719 grown in the critical substrate temperature range. So the degradation of $J_{th}$ above 670°C which is correlated to a decrease of external quantum efficiency $\eta_{ext}$ (fig. 5) could be due to an increase of losses of the cavity. It has been checked that the confinement layer thickness is always higher than 0.8μm such as coupling loss couldn't be a factor to the increase of $\eta_{ext}$ and $J_{th}$. It is the same case for free carrier losses, the carrier concentration of active layer and confinement layers being lower than 10$^{18}$/cm$^3$. In these conditions the increase of $J_{th}$ could be attributed to internal losses of the cavity due to scattering by roughness at the interfaces of the active layer; however no degradation of interface quality was observed by different authors in this substrate temperature range [5, 6]. An other explanation would be an increase of non radiative centers density in Ga$_{1-x}$Al$_x$As confinement layers with higher X concentration, allowing non radiative recombination via these centers near the heterojunctions. More work is presently in progress for a better understanding of the involved mechanisms.

Fig. 8 : The substrate temperature dependence of Ga(1070eV) and Al(1396eV) Auger peak measured on the multilayer structure of Fig.6b.
In this study, we have investigated the improvement of GaAs-Ga$_{1-x}$Al$_x$As DH properties obtained by MBE growth at high substrate temperature ($T_S > 600^\circ$C).

The ultimate value of threshold current density we reached is 1.5 kA/cm$^2$ for an active layer thickness of 0.16 μm. However, above a critical value of $T_S$ of about 670°C, an increase of $J_{th}$ was observed until a non laser emission. This seems to be correlated with an increase of the x concentration of Ga$_{1-x}$Al$_x$As confinement layers due to a reevaporation of Ga atoms at the surface of the sample during the growth at extremely high $T_S$.

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