Growth by molecular beam epitaxy and characterization of high purity GaAs and AlGaAs

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We report on the growth by molecular beam epitaxy of high-quality GaAs and $Al_xGa_{1-x}As$ ($x \lesssim 0.43$), and discuss the effect of system parameters on material quality. The highest Hall mobility in GaAs at 77 °K was 144 000 cm^2/V sec, and the photoluminescence spectra of undoped layers exhibited a strong free exciton line and a much reduced carbon peak with no carbon-related defects. A slow growth process at a substrate temperature of 600 °C produced excellent $Al_xGa_{1-x}As$ whose luminescence spectrum showed a distinct excition peak 4 meV wide. This $Al_xGa_{1-x}As$ is compared to layers grown at a faster rate at substrate temperatures of 700 °C.

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I. INTRODUCTION

Many papers have been published recently on the preparation and characterization of high-quality GaAs and $Al_xGa_{1-x}As$, and references can be found in excellent review articles.[1–4] We present here what we believe is a state of the art GaAs and $Al_xGa_{1-x}As$ material grown by the molecular beam epitaxy method (MBE). We show in some detail the steps taken to achieve the purest possible materials and the result of their characterization.

Since the introduction of the "load-lock transfer system" (samples are introduced without breaking the vacuum in the main chamber), the quality of MBE-fabricated GaAs has improved steadily, reaching its best published Hall mobility of 126 000 cm^2/V sec at 77 °K in the work of Hwang *et al.*[5] Excellent photoluminescence data (PL) has been published by Dingle *et al.*[6] and Temkin *et al.*,[7] showing that the GaAs epilayers are virtually free of defects and with carbon as the only notable impurity. [Those materials are still not as pure as material grown by vapor phase epitaxy (VPE) and liquid phase epitaxy (LPE).[8–10]] We report here on a maximum Hall mobility of 8600 cm^2/V sec at 300 °K and 144 000 cm^2/V sec at 77 °K, and show PL spectra of unintentionally doped GaAs, which exhibit a dominant free exciton peak and its excited state, and a much smaller carbon acceptor related peak.

$Al_xGa_{1-x}As$ is mostly used as a barrier for confining electrons, and its quality manifests itself in device characteristics only in second order effects. But when trapping of electrons is of concern, or current flows through the $Al_xGa_{1-x}As$ layer, its quality is of prime importance. From work reported on $Al_xGa_{1-x}As$-GaAs interfaces, superlattices, and injection lasers[11–14] came the conclusion that best quality $Al_xGa_{1-x}As$ is grown at substrate temperature of $\gtrsim$700 °C, which unfortunately is not compatible with the optimum growth temperature for GaAs ($\approx$600 °C). We present here PL data of high-quality $Al_xGa_{1-x}As$, grown at 600 °C, with a narrow exciton peak ($\approx$4 meV wide), and compare it to $Al_xGa_{1-x}As$ grown at 700 °C. This will enable the growth of GaAs-$Al_xGa_{1-x}As$ heterojunctions at 600 °C without sacrificing either material's quality.

In Sec. II we discuss in some detail the basic operation procedure of the system and the most important variables which we have tried to optimize. Sections III and IV are devoted to optical and electrical characterization of GaAs and $Al_xGa_{1-x}As$, respectively, with the main conclusions drawn in Sec. V.

II. IMPORTANT SYSTEM PARAMETERS

In this section we describe the most important steps we took in order to achieve good quality epilayer material. We present the operating procedure and demonstrate the importance of the substrate material and the influence of the As_4/Ga flux ratio on carrier concentration.

A. Preparation and growth procedure

The MBE system is a RIBER 1000-1 growth chamber with a rotating substrate holder of 1.5 in. diam, equipped with High Energy Electron Diffraction apparatus (HEED) and a Residual Gas Analyzer (RGA). Preparation and introduction chambers are situated between the MBE chamber and the atmosphere, both pumped by cryopumps. The MBE system is pumped simultaneously by a 400 liter/sec ion pump, a cryopump, and a titanium sublimation pump (TSP), and does not contain any polymer material. A shroud containing liquid nitrogen (LN_2) surrounds the effusion cells, growth area, and the TSP. Effusion cells with pyrolitic boron nitride (PBN) crucibles are outgassed for 48–72 h in the preparation chamber at temperatures of 1400–1600 °C, then charged with Ga, As, Al, and Si material and loaded into the growth chamber, which subsequently is pumped and baked at $\approx$220 °C for 72–84 h, while being pumped by an auxiliary ion pump of 200 liter/sec (which is later valved off the system), and the TSP. The shrouds are then filled with LN_2 and cell charges are outgassed at about 100 °C above the maximum operation temperature (except As). Between growths the cells are left idling at $\approx$700 °C (As at 150 °C) and the LN_2 cools the system continuously. With all cell shutters closed, the chamber pressure—in the idling condition—is less than 5×10^{-11} Torr, and its environment contains hydrogen and less than 0.01% of carbon monoxide.

Polished substrates are cleaned by the procedure prescribed by Cho *et al.*,[15] and subsequently mounted with Ga on a tantalum holder and placed in the introduction chamber. After evacuation to $\sim 10^{-8}$ Torr, they are moved into the preparation chamber (which idles at 1×10^{-10} Torr). After outgassing them at 400 °C for 1 h they are loaded into the growth chamber and kept at 400 °C until growth is initiated.

It was found that layers grown on substrates which were introduced into the growth chamber just prior to growth, or were in the chamber while the LN_2 shroud warmed up accidentally, contained more carbon and donor impurities than the others.

Since the substrate holder is rotatory, the temperature-controlling thermocouple is not in direct contact with it. Consequently, the substrate temperature drops by 50–80 °C during the first 1–2 h of the growth, due to an increase in the emissivity of the bare tantalum holder, which gets coated with GaAs. The temperature is monitored from one of the cell ports by a single wavelength infrared pyrometer, through a sapphire window (at a wavelength of 2.5 μm the emissivity is set at 0.6, and is reduced gradually when the window is coated).

Surface oxide usually desorbs at 570–600 °C, and the substrate is heated up momentarily to ≈ 650 °C under As flux, thereafter GaAs growth is initiated at 600 ± 5 °C. Beam fluxes are adjusted with an ion gauge in the growth position, and for a distance between sources and substrate of ≈ 12 cm and flux incidence angle of 30° to the normal of the substrate, a growth rate of ~ 1 μm/h is achieved with Ga Beam Equivalent Pressure (BEP) of $\simeq 1.8 \times 10^{-7}$ Torr (Ga cell tempeature is ~ 960 °C, and its orifice area is 2.8 cm^2). An As stable surface (2×4) or (3×2) is maintained with As BEP of $\approx 3.2 \times 10^{-6}$ Torr.

When $Al_{0.3}Ga_{0.7}As$ is grown, an Al BEP of 4×10^{-8} Torr leads to growth rates of 1.4 μm/h at 600 °C and 1 μm/h at 700 °C (at 700 °C the sticking coefficient of Ga is 75%).

B. Substrate material

The epilayers were deposited either on undoped substrates grown by the liquid encapsulated Czochralski (LEC) method or on Cr-doped substrates grown by the horizontal Bridgman (HB) technique. In both cases, the substrates were (100) oriented or 2° off (100) towards the (110) direction. The surface morphology of all layers grown on (100) substrates exhibited fine ripple ("orange peel" appearance), while the layers grown on the 2° off (100) substrates had a ripple-free surface. The LEC crystals had about 10^5 cm^{-2} dislocations, which seemed to be correlated with the number of local defects on the surface, almost absent in layers grown on HB crystals (which had less than 10^3 cm^{-2} dislocations). The features mentioned above are seen on Fig. 1. Layers grown after the Ga crucible was filled above $\sim 80\%$ showed a decrease of large local defects, in agreement with the theory of "spitting" suggested by Wood.[16] (The Ga cell is tilted 30° from the horizontal plane.) The number of local defects in samples grown after ~ 150 μm of previous growths increased, simultaneously with the number and size of Ga dro-

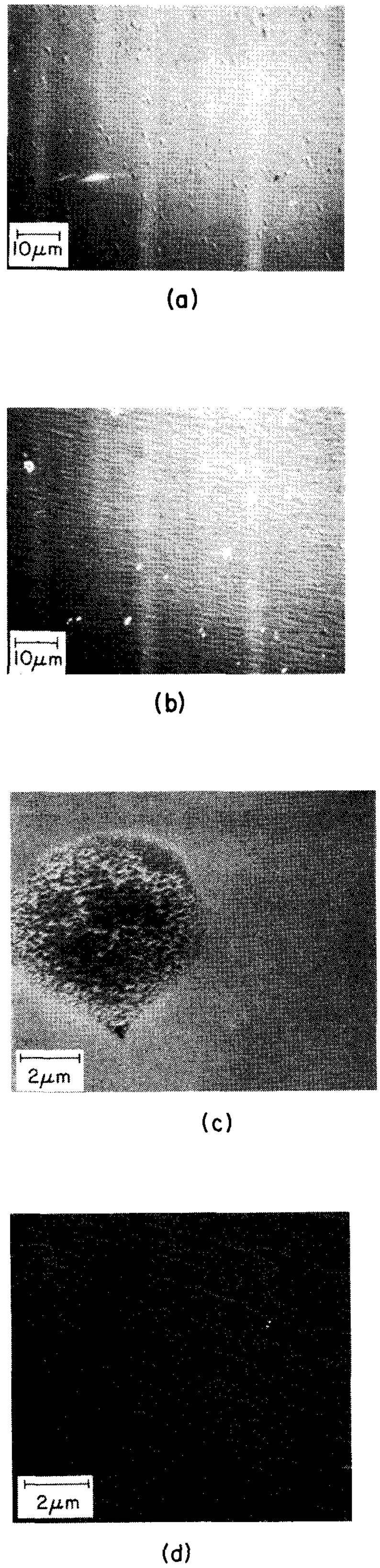

FIG. 1. Surface morphology of GaAs epilayers, ~ 6 μm thick, as revealed by optical microscope with $\times 1120$ magnification [(a) and (b)], and SEM [(c) and (d)]. Pictures (a) and (c) correspond to a layer grown on LEC-undoped substrate, oriented 2° off (100), and (b) and (d) to a layer grown on HB-Cr doped substrate, (100) oriented. The former layer shows many "oval defects" (a) and some of the larger defects attributed to Ga "spitting" (c). The latter presents an "orange-peel" morphology [(b) and (d)].

plets at the edge of the PBN crucible.

Unintentionally doped epilayers were grown simultaneously on undoped and Cr-doped substrates. The layers on the undoped substrates were p type, with a carrier concentration of $\sim 1\times 10^{14}$ cm^{-3} (determined from C-V measurements with a Hg probe), while the layers grown on the Cr-doped substrates were semi-insulating, probably due to Cr outdiffusion from the substrates. We believe that similar compensatory effects can explain the high resistivity of layers reported by Hwang *et al.*[5] (rather than uncompensated concentration of $\sim 10^{13}$ cm^{-3}, which would in turn lead to a mobility exceeding 200 000 cm^2/V sec at 77 °K).[17,18]

We have found that p-type impurities diffused from the substrate (probably Mn), as demonstrated in Fig. 2. Unintentionally doped epilayers grown on as-received ("virgin") LEC substrates had a peak hole concentration of $\approx 10^{15}$ cm^{-3} at the layer-substrate interface, and an average Hall mobility of ~ 2 000 cm^2/V sec at 77 °K. When those substrates had been baked at 750 °C for 24 h in H_2 atmosphere, and then repolished (by about 25 μm), a uniform hole concentration of $\sim 10^{14}$ cm^{-3} was measured (as seen in Fig. 2), with a Hall mobility of 7500 cm^2/V sec at 77 °K. Similar unintentionally doped material grown on "baked" HB-Cr doped substrates resulted in semi-insulating layers,[19] demonstrating that the "bake-polish" procedure reduces some p-type impurities in the substrate (like Mn), but does not affect the Cr concentration in the substrate and outdiffusion into the epilayer.

C. As_4/Ga flux ratio

Si is an amphoteric impurity in GaAs, but always exhibits a net n-type behavior in layers grown on (100) substrates. It has been reported that with increasing As_4/Ga flux ratio, Si doping results in higher electron concentration, due to a greater substitution of Ga sites. In a single continuous growth we have kept the Si cell temperature constant and varied the As flux. Subsequent etching of the layer and C-V profiling resulted in a peak carrier concentration for a flux ratio $F(As_4)/F(Ga)\simeq 1$, for two different doping levels, in the 10^{14} and 10^{16} cm^{-3} ranges. The electron concentration is much more sensitive to the As_4 flux in the 10^{14} cm^{-3} range of doping, as demonstrated in Fig. 3. The strong influence of As_4 at low doping levels[20,21] can also result from a difference in the incorporation of carbon in the layer. PL experiments at 10^{16} cm^{-3} doping levels showed that the ratio between the carbon and Si acceptor peaks increases monotonically with increasing As_4 flux,[22] suggesting that more carbon is incorporated with higher As_4 flux, thus reducing the net electron concentration in the 10^{14} cm^{-3} doping level.

Flux ratios are calculated from the BEP of Ga and As_4, and their relative angles of incidence on the substrate, assuming 30% cracking of $As_4 \rightarrow As_2$ by the ionization gauge,[23] which has a sensitivity proportional to the total number of electrons in the molecule.[24]

This optimum doping condition can be identified by the HEED pattern, which is on the verge of converting from a (2×4) to a (3×2); both are As stable conditions. At this growth condition the material is the least compensated for a particular electron concentration.

Different growth rates of GaAs were experimented with no significant differences in the PL spectra. Films grown at a rate of ≈ 0.25 μm/h exhibited excellent PL spectra. When growth temperature exceeded 620 °C the As_4 flux had to be increased substantially to maintain a (3×2) diffraction pattern and smooth surface morphology. However, as will be shown later, excellent GaAs layers were grown at 670 °C with $F(As_4)/F(Ga)\simeq 2$.

III. CHARACTERISTICS OF GaAs LAYERS

The highest theoretical Hall mobility of GaAs at 77 °K with no compensation $[(N_D+N_A)/(N_D-N_A)=1]$ is 244 000 and 296 000 cm^2/V sec, according to Wolfe *et al.*[17] and Rode,[18] respectively. Experimentally, maximum mobilities at 77 °K of GaAs layers grown by VPE were 210 000 cm^2/V sec for $N_D+N_A \simeq 2\times 10^{14}$ cm^{-3} and a compensation of 1.35,[17] and for those grown by LPE 240 000 cm^2/V sec for $N_D+N_A \approx 1\times 10^{13}$ cm^{-3} with compensation of 2.6.[25] At those low concentration levels, the measured mobilities approach the lattice scattering mobility, and are weakly sensitive to the total number of impurities (note that layer

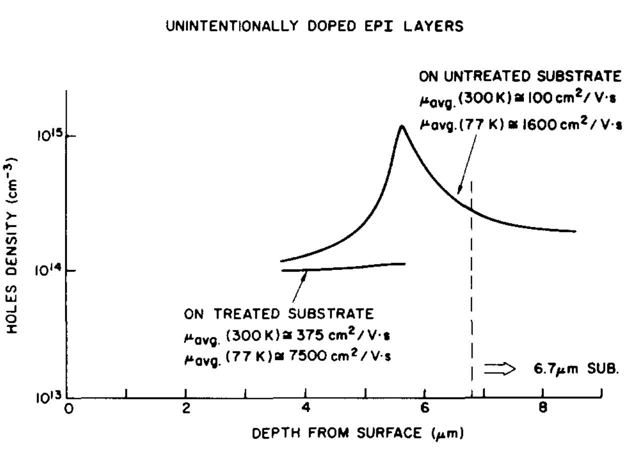

FIG. 2. Carrier concentration (measured with Hg probe) of unintentionally doped 6-μm-thick layers, grown on "virgin" and "baked" LEC-undoped substrates (see text). The layer grown on the "virgin" substrate shows a p-type accumulation near the interface.

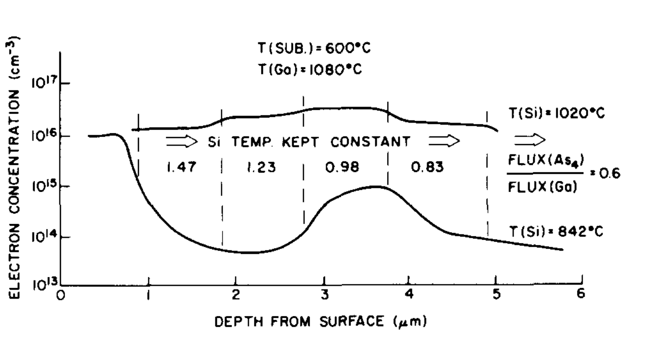

FIG. 3. Effect of As_4/Ga flux ratio on electron concentration in GaAs, resulting from Si doping. The Si source temperature was kept constant while the As_4 pressure was changed. A flux ratio of ~ 1 produced the highest electron concentration.

thickness was tens of microns). In MBE, since the practical layer thickness is less than 7–10 μm, carrier densities less than 10^{14} cm^{-3} are exceedingly difficult to measure, due to carrier depletion at the surface and substrate interfaces. Generally the purest unintentionally doped layers are of p-type nature with hole density $\sim 10^{14}$ cm^{-3}, with carbon as the main acceptor as identified by PL, and noted in Sec. II B.

Experimenting with different growth rates 0.1–1.5 μm/h did not result in a significant difference in the amount of carbon detected, suggesting that it is not incorporated from the chamber environment, or that it is growth-limited incorporated. On the other hand, when one of the cells was accidentally contaminated and produced large amounts of CO impinging on the sample, all layers were p-type doped in the $\sim 10^{17}$ cm^{-3} range. It is possible that when the levels of CO and CH_4 in the MBE chamber are very low, the carbon in the epilayers results partly from the surface or the substrate interior, and partly from the hot sources. Also, the rate of incorporation is limited by the surface population of the III–V's or the dopants.

For a doping level of $N_D \simeq 1\text{–}2 \times 10^{14}$ cm^{-3} and $N_A \simeq 1 \times 10^{14}$ cm^{-3} compensation levels of 2–3 are expected, and 77 °K mobilities could reach $\sim 150\,000$ cm^2/V sec. Calawa *et al.*[26] reported Hall mobilities exceeding 10^5 cm^2/V sec at 77 °K, using AsH_3, and attributed their success to the availability of As_1 species. Hwang *et al.*[5] published a 77 °K mobility of 126 000 cm^2/V sec using As_4, achieving these results by paying special attention to system cleanliness.

A. Electrical characteristics

Hall mobilities were measured at 300° and 77 °K—in the dark—by the van der Pauw method,[27] on a lithographically-fabricated 1×1-mm^2 mesa, with AuGeNi-alloyed contacts, at a magnetic field of 0.4 T. The epilayers were 5–6 μm thick lightly doped with Si on top of unintentionally doped buffer layer 0.5–1 μm thick grown on a virgin LEC substrate.

Carrier concentration was monitored by a Hg probe at 300 °K and assumed to be invariant down to 77 °K. Figure 4 summarizes the Hall mobilities of lightly doped layers. The lower shaded collection of points was achieved at the initial part of the study, for substrates introduced just before each growth with the ion gauge operating during growth period. When the procedure prescribed in Sec. III A was followed (especially after a more intensive outgassing of cells and charges, and a thorough bakeout of the chamber) the upper shaded region (between the two open rectangles) was measured. The highest mobility was measured on a sample with $n \simeq 2 \times 10^{14}$ cm^{-3}, and was 144 000 cm^2/V sec at 77 °K suggesting a compensation of ~ 2. This sample had a 300 °K mobility of 8560 cm^2/V sec.

B. Photoluminescence

The PL measurements were done by exciting the epilayers placed in a variable-temperature (1.9–300 °K) cryostat, with the 5145 Å line of an Ar^+ laser. The induced luminescence was focused into a double-pass 3/4 meter spectrometer and detected by a photomultiplier with a cooled GaAs cathode.

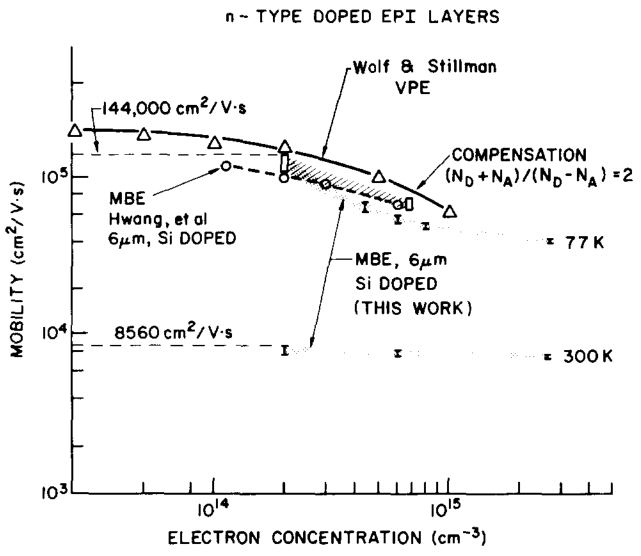

FIG. 4. Summary of the Hall mobility results of lightly-doped GaAs layers. The two shaded regions demonstrate the effects on the mobility of the procedures described in the text. The results of Wolfe *et al.* (Ref. 17) with their theoretical prediction and Hwang *et al.* recent results (Ref. 5) are shown for comparison.

Several undoped and lightly doped n-type samples, grown at various temperatures (in the range 590–670 °C), were studied. No significant decrease of the PL intensity was observed in samples grown at the highest temperature. Above 670 °C, a rapid deterioration of the PL has been reported[22] in layers grown under slightly different conditions.

A typical PL spectrum is shown in Fig. 5, corresponding to an undoped sample grown at 670 °C on a semi-insulating Cr-doped substrate. The main peak, at 1.5151 eV(F, X) is due to a free exciton recombination, its excited state ($n = 2$) being observed at 1.5181 eV. The well defined doublet at 1.5123 and 1.5125 eV has been identified as the $J = 5/2$ and 3/2 states of an acceptor-bound exciton.[28] A much weaker structure at 1.5128 eV has a similar origin, with $J = 1/2$. Recombination of exciton bound to neutral or ionized donors is responsible for the luminescence peaks at 1.5141 eV (D^0, X) and 1.534 eV(D^+, X), respectively. No evidence was found of a peak at 1.5145 eV, frequently observed in MBE- and VPE-grown GaAs,[29,30] and attributed to an antisite defect complex.[30] We believe that the spectrum of Fig. 5 shows the best resolved exciton peaks, of MBE-grown GaAs published to date, and is comparable to those reported in high-purity VPE material.[28]

The insert in Fig. 5 presents the PL intensity in the 1.49 eV region, where two peaks are observed, associated with acceptor-level transitions. The structure at 1.4934 eV (e, C_{As}) identifies carbon as the main residual acceptor and is due to a conduction band-to-carbon level transition.[31] The peak at 1.4892 eV (D^0, C_{As}) is attributed to a donor-to-carbon level transition. It is worth noting the small intensity of these structures compared to the exciton peak, indicating the small carbon content in the layer grown at such high temperatures.

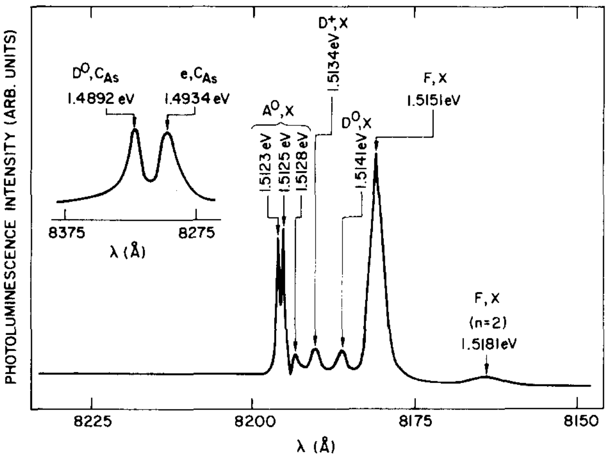

FIG. 5. Low-temperature ($T = 5$ °K) photoluminescence spectrum of an undoped GaAs sample grown at 670 °C. Excitation intensity was 0.5 W/cm^2. Perfect agreement was found with the energies of the various transitions listed in Ref. 28 and structures were labelled accordingly. The insert shows the spectral region corresponding to acceptor level recombination. The vertical scale is the same as in the main spectrum.

The PL spectra of undoped layers grown at lower temperatures (~ 600 °C), were very similar to that of Fig. 5. The only difference was the appearance of the so-called "defect excitons" in the region 1.504–1.511 eV, almost invariably present in MBE-grown GaAs.[6,29,32] (The exception is the data by Temkin and Hwang.[7]) In lightly Si-doped samples the (D^+, X) and (D^0, X) lines were dominant, all the other features remaining practically unchanged.

IV. CHARACTERISTICS OF $Al_xGa_{1-x}As$ LAYERS

The growth of high-quality $Al_xGa_{1-x}As$ is more difficult than that of GaAs due to the high reactivity of Al with oxygen, water vapor, and carbon monoxide, believed to result in deep levels in the grown $Al_xGa_{1-x}As$ layer.[33] Also, excess As seems to degrade the film quality due to the formation of Group III vacancies.[33,34]

Casey *et al.*[35] achieved good luminescence from $Al_xGa_{1-x}As$ layers grown with substrate temperature of ≈ 650 °C, but the material showed degraded surface morphology. Works which followed invoked higher substrate temperatures,[36] and resulted in reduced current threshold of heterojunction and quantum well lasers,[11,12] and improved PL of quantum wells[13] and bulk $Al_xGa_{1-x}As$.[33] The general belief is that the increase of the substrate temperature (up to ~ 700 °C) reduces the incorporation of impurities from the chamber environment and increases the desorption of As from the surface, preventing Ga vacancies. All reports show an enhanced luminescence efficiency and narrower spectra lines at higher temperatures. It was also claimed that interfaces between GaAs and $Al_xGa_{1-x}As$ are smoother at higher temprature,[13] leading for example, to better luminescence of quantum wells.

Here we show that a big improvement in the quality of the $Al_xGa_{1-x}As$ can be achieved by lowering the growth rate from ~ 1.4 to $\sim 0.14\,\mu$m/h, and maintaining the normal AS_4/Ga flux (≈ 1), and substrate temperature of ~ 600 °C.

$Al_xGa_{1-x}As$ layers with Al mole fraction betwen 0.3 and 0.43 have been grown. Three different conditions have been employed: $\sim 1.4\,\mu$m/h growth rate at 600 °C substrate temperature ("600 °C-Fast"), 1 μm/h growth rate at 700 °C ("700 °C-Fast"), and 0.14 μm/h growth rate at 600 °C ("600 °C-Slow"). While the surface morphology of the layers grown at "600 °C-Slow" and "700 °C-Fast" was excellent, with a clear reconstruction of (3×2), the layers grown at "600 °C-Fast" resulted in a poor surface morphology (even for layers as thin as ~ 1000 Å thick), and a spotty HEED pattern $\{$for $F(As_4)/[F(Ga) + F(Al)] \approx 0.7\}$.

All $Al_xGa_{1-x}As$ layers were grown on GaAs buffers grown at 600 °C, at a rate of $\sim 1\,\mu$m/h. When "700 °C-Fast" layers were grown the substrate temperature was raised to 700 °C in ~ 1 min and subsequently the $Al_xGa_{1-x}As$ deposition was initiated. The amount of As_4 needed to maintain a (3×2) surface reconstruction and a smooth surface morphology was less than half of the amount needed for growth of GaAs at 600 °C. For "600 °C-Slow" layers, Ga and As temperatures were reduced simultaneously to the desired rate, while the substrate temperature was maintained at 600 °C. All mole fractions were measured by microprobe and also deduced from the PL spectrum, which consistently gave a higher Al content (by 6%–8%). We quote here the "PL mole fraction."

A. PL spectra of $Al_xGa_{1-x}As$ grown at 700 °C

Figure 6 shows PL spectra at various temperatures of an undoped $Al_{0.34}Ga_{0.66}As$ layer grown at "700 °C-Fast." The low-temperature luminescence intensity was very

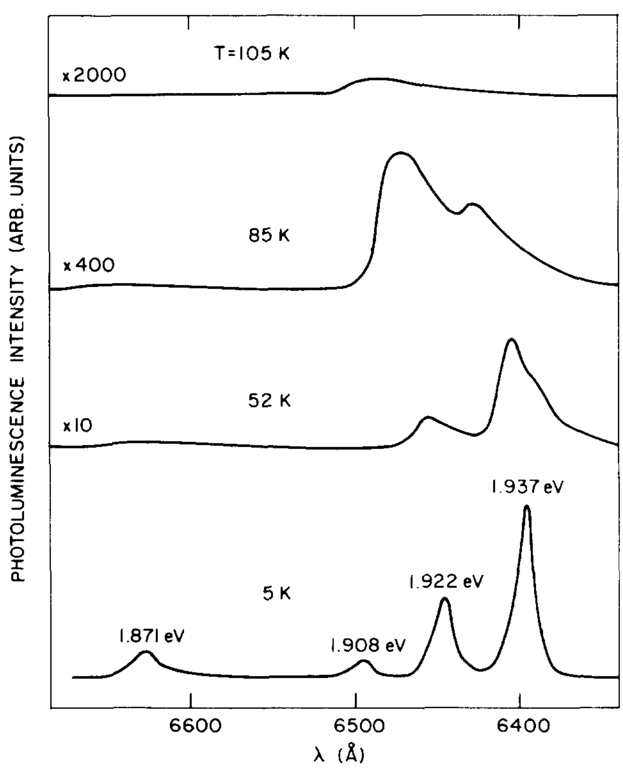

FIG. 6. PL spectra, at various temperatures, of an $Al_xGa_{1-x}As$ ($x \simeq 0.34$) epilayer grown at "700 °C-Fast" conditions on a conductive GaAs substrate. Identical results were obtained for layers grown simultaneously on Cr-doped and undoped substrates. Excitation intensity was 1 W/cm^2.

strong but it decreased fast with increasing temperature. The spectrum at 5 K consisted of four narrow peaks, whose relative amplitude did not change when the excitation intensity was varied by two orders of magnitude. The highest-energy structure, at 1.937 eV, and with a full width at half height (FWHH) of 4.5 meV, is believed to be due to excitonic recombination, possibly an acceptor-bound exciton. (Based on this assignment and on Stringfellow and Linnebach's data[37] we estimate $x \simeq 0.34$). At higher temperatures this peak develops a shoulder that can be attributed to an excited state of the exciton.

The 1.922-eV peak shows a high-energy tail at moderate and high temperatures, characteristic of free-to-bound transitions.[38] Although it is possible to assign it to conduction band-to-carbon level transitions (e,C_{As}), the energy separation with the exciton is too small in comparison with the values reported in the past.[39,40] More consistent with those data is the 1.908 eV feature, but its disappearance at relatively low temperature (~ 37 K) is puzzling. More work is being done to clarify the exact origin of these structures.

To check the effect of outdiffusion of impurities from the substrate, identical layers were grown simultaneously on three different substrates: LEC-undoped, HB-Cr doped, and HB-Si doped. No significant differences were observed among the PL of the three layers.

An unintentionally doped layer, ~ 4 μm thick with $x \cong 0.43$ grown at "700 °C-Fast" conditions, was *p* type with $\sim 5 \times 10^{14}$ cm^{-3} acceptors and 300 °K mobility of 370 cm^2/V sec (complete freeze out occurred at 77 °K). The *p*-type nature of the layer suggests that very little oxygen or other deep impurities exist in the layer (otherwise the nature of the layer would be most probably semi-insulating).

B. PL spectra of $Al_xGa_{1-x}As$ grown at 600 °C

High-quality $Al_{0.43}Ga_{0.57}As$ layers can also be grown at "600 °C-Slow" and "600 °C-Fast" growth conditions. The PL in Fig. 7 shows the PL of the "600 °C-Slow" layer. The spectrum consists of a peak at 2.055 eV, with a FWHH of 4 meV, a shoulder at 2.058 eV, and a broad peak at 2.001 eV. Under much higher excitation conditions or high temperatures, the relative intensity of the various structures remained unchanged. We attribute the high-energy peak to an exciton (from which we get $x \cong 0.43$). The energy of the low-energy structure seems inconsistent with an (e,C_{As}) transition, based in previous reports,[39,40] and its origin is unknown presently. Samples grown at "600 °C-Fast" exhibited a luminescence peak with a comparable intensity, contrary to Wicks *et al.* observations,[33] but with FWHH of 44 meV.

The most striking features of the spectrum of Fig. 7 are the narrowness of the excitonic peak for such a high x, which is comparable to undoped LPE material,[33] and the absence of defect-related excitons, seen in high-quality $Al_xGa_{1-x}As$.[33] These facts, together with a very strong luminescence intensity (similar to that from the layer grown at "700 °C-Fast"), indicate the high-quality of samples grown at low temperature under the "Slow" conditions.

The absence of impurities (carbon in particular) and the strong PL intensity suggest that the UHV environment is clean enough, or the sticking coefficient of the various impurities is low for a slow growth condition. Even more important, the results of the slow growth suggest that the reason for a higher film quality at 700 °C, reported in the past, is probably the increased mobility of Al atoms on the surface at high temperature. We achieved a similar effect by lowering the growth rate, enabling the Al atoms to find the right sites before the subsequent Al atoms arrive and thus minimizing clustering. This interpretation is consistent with the broad spectrum observed at "600 °C-Fast" conditions. The high PL intensity that we observed is probably the consequence of the clean environment eliminating the need for high substrate temperatures which was claimed to be required in order to retard the sticking of background impurities.

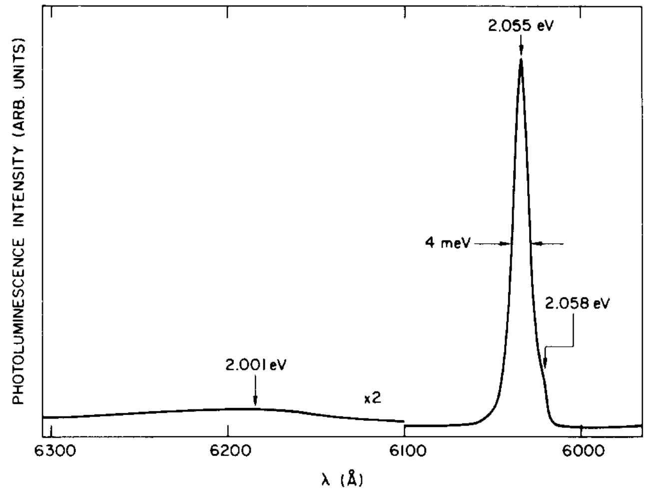

FIG. 7. PL spectra, at 5 °K, of an $Al_xGa_{1-x}As$ ($x \simeq 0.43$) layer grown at "600 °C-Slow" conditions, for an excitation of 1 W/cm^2. The shape of the spectrum remained unchanged when the excitation was raised over a three order of magnitude range.

V. CONCLUSIONS

We have presented the characteristics of GaAs and $Al_xGa_{1-x}As$ layers grown by MBE (a RIBER 1000-1 system), with overall quality approaching layers grown by VPE and LPE. Our effusion cells are constructed with Al_2O_3 parts, and contrary to previous work,[41,42] which predicted that layer quality should degrade due to incorporation of O and Al from the Al_2O_3 parts, the material is excellent. We atribute our success mainly to the systematic outgassing of the cells and their charges and, secondly, to the special care exercised in vacuum integrity and growth procedures.

We have measured the highest mobility reported to date on MBE-grown GaAs (at 77 °K, 144 000 cm^2/V sec), and presented the best PL spectra, regarding line width, excitonic levels, and their excited states. The high quality of the PL spectra was preserved for layers grown up to 670 °C.

By slowing the growth rate of $Al_xGa_{1-x}As$ to ~ 0.14 μm/h and maintaining the substrate temperature at 600 °C and As_4/Ga flux at ~ 1, an exciton peak of ~ 4 meV wide was measured for $x = 0.43$, with excellent surface morphology. This approach enables a continuous growth of GaAs and $Al_xGa_{1-x}As$ at 600 °C. Note that the MBE system environment must be very clean to avoid an enhanced incorporation of impurities at this slow rate of growth.

$Al_xGa_{1-x}As$ grown at substrate temperature of 700 °C showed PL line width and intensity comparable to the "600 °C-Slow" $Al_xGa_{1-x}As$, but the spectra exhibited more features, absent in the "600 °C-Slow" layers.

We do not advocate the necessity of all the steps taken for each device structure grown, since excellent device features produced by MBE, such as lasers, Schottky diodes, FET's, etc. have been reported in the literature, based on layers relatively heavily doped. On the other hand for structures like "selectively doped heterojunctions" and "ballistic devices," which operate at low temperatures and low intentional doping levels, attention to details will significantly improve their performance. In the near future, with the improvement in purification and growth techniques of Ga, As, Al, and GaAs substrates on one hand and Ta filaments and PBN (or another novel material) crucibles on the other material characteristics will probably approach theoretical predictions.

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