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RHEED Oscillation-Based Optimization of Growth Conditions for Gas-Source MBE Growth of InGaP Using Tertiarybutylphosphine

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1. Introduction

$In_{0.48}Ga_{0.52}P$ is a promising material for optoelectronic devices in the visible and long wavelength ranges and for high-speed electron devices. Although gas-source MBE growth (GSMBE) using tertiarybutylphosphine (TBP) is potentially attractive growth method of InGaP combining advantages of MBE, including a monolayer-level thickness controllability, easy access to UHV-based processing tools and surface science-type characterization tools etc. with low toxic nature of TBP. However, electronic and optical properties of InGaP grown by GSMBE using TBP[1] have so far been inferior to those by GSMBE using PH_3[2], and those by MOCVD[3].

The purpose of this paper is to optimize the growth conditions for GSMBE of InGaP using TBP by performing a detailed RHEED oscillation study. By using optimized growth conditions, $In_{0.48}Ga_{0.52}P$ layers having the electronic and optical properties comparable to the best data reported for MOCVD[3] have been achieved for the first time.

2. Growth Procedure and Initial GaAs Surface

Undoped InGaP layers with thickness between 0.9-1.3μm were grown on GaAs buffer layers within a substrate temperature range from T_g=450°C to 640°C. Metallic In, Ga and As were used as source materials for group III elements and As_4. On the other hand, to obtain P_2 flux, 100% TBP was decomposed in a thermal cracker cell. Flow rate of TBP, F_{TBP}, was varied within a range from 0.5 to 4sccm.

Preparation of appropriate initial GaAs surface was found to be essentially important for realizing stable layer-by-layer growth of InGaP on GaAs. Namely, InGaP growth on As-stabilized (2x4)-GaAs surfaces resulted in observation of (2x4) or (2x1) pattern with clear and persistent RHEED oscillations as shown in Fig.1(a), whereas growth on As-rich (2x1) surfaces usually resulted in growth without RHEED oscillations as shown in Fig.1(b). Therefore, in this study, all the InGaP growth were performed on the (2x4)-GaAs surfaces.

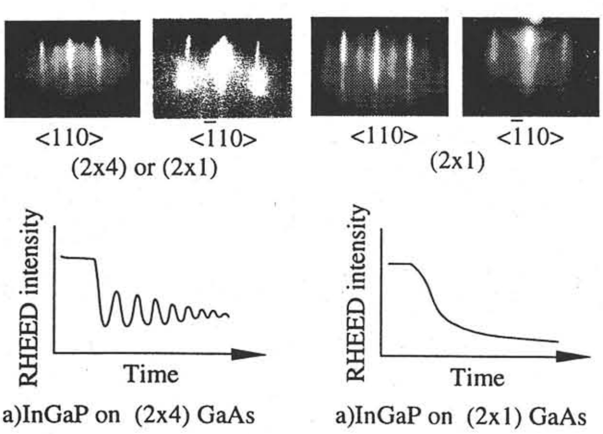
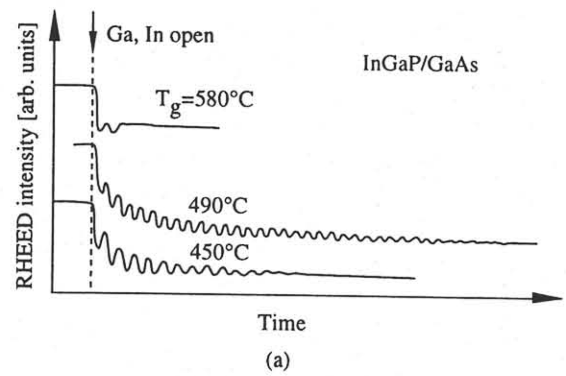


Fig. 1 RHEED patterns and oscillations during InGaP growth a) on As-stabilized (2x4)-GaAs, and b) on As-rich (2x1)-GaAs.

3. RHEED Study

Figure 2(a) shows typical RHEED oscillations observed during the growth of InGaP at T_g=450, 490 and 580°C at F_{TBP}=4sccm. Although persistent RHEED oscillations were observed at 490°C, they decreased more quickly at other temperatures. The observed dependence of the number of RHEED oscillations, N, on T_g is summeruzed in Fig. 2(b). The result shows that stable layer-by-layer growth can only be realized within a limited temperature range from 480°C to 510°C.

Figure 3 shows a schematic illustration of F_{TBP} dependences of the InGaP growth rates and number of



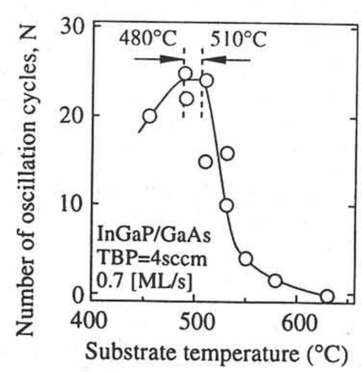


Fig. 2 (a)Typical RHEED oscillations during the InGaP growth on (2x4)-GaAs at 450, 490 and 580°C. (b)Dependence of number of RHEED oscillation cycles on growth temperature.

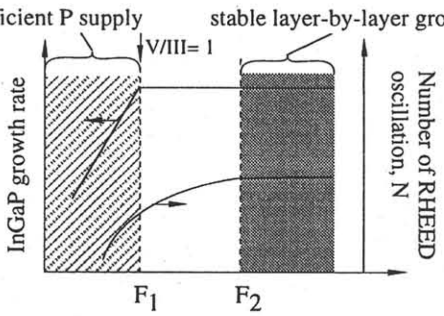


Fig. 3 Commonly observed F_{TBP}-dependence of InGaP growth rate and RHEED oscillation numbers.

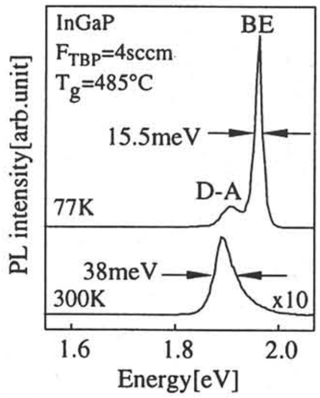


Fig. 4 PL spectra of InGaP layer grown at 485°C measured at 77 and 300K.

RHEED oscillations commonly observed in the entire T_g range studied here. Increase of T_g only shift the both curves to the righthand side. Similar to the GSMBE growth of InP using PH_3[4], the constant growth rate region above a certain TBP flow rate of F_1 corresponds to the group-III-limited growth region, and the lower growth rate region below F_1 to the P-limited-growth region due to an insufficient P supply, respectively. The intersection between these two region (=F_1) occurs when the effective supply of group III and V sources are the same (III/V=1) and located between 1 and 1.5sccm for T_g between 490°C and 510°C.

On the other hand, the number of RHEED oscillation, N, usually does not saturate even at F_{TBP}=F_1 and somewhat higher flow rate (>F_2) is necessary for saturation of N where most stable layer-by-layer growth was realized. More specifically, such a growth mode was realized at F_{TBP}=4sccm within a temperature rage form 470-490°C, resulting in achievement of extremely smooth InGaP surfaces.

4. Optical and Electrical Properties

Figure 4 shows PL spectra of InGaP layer grown at T_g=485°C measured at 77 and 300K. The PL spectrum taken at 77K have two peaks around 1.91 and 1.96eV whereas that at 300K has a single peak at 1.89eV. The main peaks around 1.96eV at 77K and 1.89eV at 300K can be assigned as the InGaP band-edge emissions. The side peak at 1.91eV observed at 77K is due to donor-acceptor (D-A) pair emission

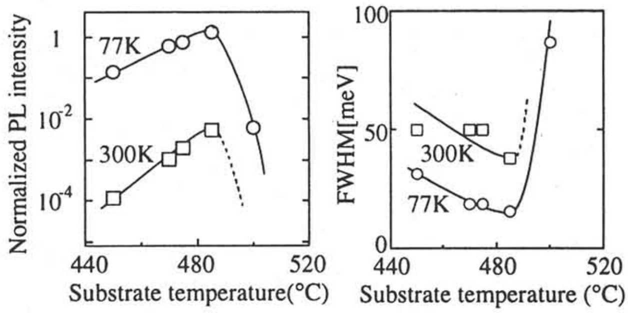


Fig. 5 Substrate temperature dependence of (a) PL intensity and (b) FWHM of PL in InGaP measured at 77 and 300K

Table I Best data of carrier concentration and electron mobility at 300K for undoped InGaP grown by various growth methods.

	n[cm^{-3}]	μ[cm^2/V·s]	Ref.
Present work (GSMBE TBP)	$5x10^{14}$	3300	
GSMBE(TBP)	$4x10^{14}$	1900	[1]
GSMBE(PH_3)	$1.3x10^{15}$	3200	[2]
MOCVD(PH_3)	$1.3x10^{15}$	3300	[3]
MBE(GaP soure)	$5x10^{15}$	1000	[6]

[5]. Figure 5 summarizes T_g dependence of a)PL intensity and b)FWHM for InGaP band-edge emission. The PL intensity and FWHM values were found to be strongly dependent on T_g and took maximum and minimum, respectively, at T_g=485°C. The minimum FWHM values of 15.5meV at 77K and 38meV at 300K, respectively, are comparable to the best values reported for InGaP layers grown by other methods, and are narrowest of all the reported values for InGaP layers grown by GSMBE using TBP.

As for the electrical properties, although growth below 475°C resulted in high resistive InGaP layers, the growth above 485°C led to growth of InGaAs layer with extremely high electron mobilities and low carrier concentrations. The highest electron mobility of 3300cm^2/V·s at 300K and that of 21000cm^2/V·s at 77K at the low carrier concentration value of $5x10^{14}$-$1x10^{15}cm^{-3}$ were achieved in the present InGaP layer grown above 485°C. These data are not only the best of TBP-based MBE but also among the best data obtained by various growth methods[1],[2],[3],[6] as shown in Table I.

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