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Optical and Structural Properties of MOCVD Grown $In_xGa_{1-x}As$ Epilayers

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Abstract: $In_xGa_{1-x}As$ layers on undoped InP (100) substrates were grown with Aixtron 200-4 RF/S horizontal Metal Organic Chemical Vapour Deposition (MOCVD) reactor. All the epilayers have been grown with different indium compositions. Thickness of the samples were measured via Scanning Electron Microscopy (SEM). Indium concentrations were defined by High Resolution X-ray Diffraction (HRXRD) and optical measurements were done with spectroscopic ellipsometry in order to obtain refractive index (n) and thickness of the samples. In-situ reflectance is used to measure thickness of samples. Then all of the thicknesses are compared.

Keywords: Metal Organic Chemical Vapour Deposition MOCVD, Ellipsometry, HRXRD, SEM, In-situ Measurement.

MOCVD ile Büyütülen $In_xGa_{1-x}As$ Epikatmanların Optik ve Yapısal Özellikleri

Özet: $In_xGa_{1-x}As$ tabakaları katkısız InP (100) alttaş üzerine Aixtron 200-4 RF/S yatay Metal Organik Kimyasal Buhar Depolama (MOCVD) sistemi ile büyütülmüştür. Bütün epikatmanlar farklı indiyum konsantrasyonlarında büyütülmüştür. Katmanların kalınlıkları Taramalı Elektron Mikroskobu (SEM) ile ölçülmüştür. İndiyum konsantrasyonları Yüksek Çözünürlüklü X-ışını Kırınım (HRXRD) cihazı ile tayin edildi ve kırılma indisi (n) ve kalınlıkların belirlenmesi için optiksel ölçümler spektroskopik elipsometre ile yapıldı. In-situ yansıma, örneklerin kalınlıklarının belirlenmesi için kullanılmıştır. Son olarak bütün kalınlıklar karşılaştırılmıştır.

Anahtar Kelimeler: Metal Organik Kimyasal Buhar Depolama MOCVD, Elipsometre, HRXRD, SEM, In-situ Ölçüm

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1. INTRODUCTION

Compound semiconductors are very attractive materials not only for their tunable band gap energy but also for tuning their lattice constant to the commercially available substrates such as, GaAs, InP, Ge etc. $In_xGa_{1-x}As$ is one of these compound semiconductor materials. It can be grown lattice matched on InP ($In_{0.53}Ga_{0.47}As$) [1]. Meanwhile, $In_xGa_{1-x}As$ can also be grown on these substrates slightly lattice mismatched with high crystal quality by avoiding relaxation and dense defect density by keeping thickness relatively thin and lower than the critical thickness. Band gap energy value of $In_xGa_{1-x}As$ can be tuned from 0.37 eV to the 1.42 eV by changing indium concentration [2]. This energy range spans near infrared to the mid infrared region which makes $In_xGa_{1-x}As$ a suitable material for infrared device applications. Not only had those properties mentioned above makes $In_xGa_{1-x}As$ usage popular in device structure, but also it has a direct band gap from x=0 to x=1 and that makes $In_xGa_{1-x}As$ very suitable for several optoelectronic device applications. For instance, it is commonly used in tandem solar cell structures [3], laser diodes [4], and p-i-n photodiodes [5]. $In_xGa_{1-x}As$ is mostly grown with MOCVD and MBE (Molecular Beam Epitaxy) on single crystal substrates [3-5]. MOCVD has advantageous for example, it is possible to obtain high crystal quality layer at very high growth rate with multiple growths at once. That makes MOCVD suitable for mass production. However, a major disadvantages of MOCVD reactors due to their growth ambient, they prohibits a wide usage of in-situ measurements tools. Even though, new tools are under development, the only tool commonly used nowadays is in-situ reflectance measurements and from these measurements one can calculate growth rates and thickness. Therefore, to determine the structural properties of MOCVD growths, further ex-situ measurements are required. This is somehow problematic since during the growth because grower depends on in-situ measurements which are not very precise and also done under elevated growth temperatures and ex-situ measurements generally more precise but done at room temperature. So it is important to quantitatively determine how different these measurements can be? Until we began to use improved in-situ measurement techniques a clear understanding of connecting in-situ and ex-situ measurements are very crucial for MOCVD growths.

In this study, four InGaAs samples have been grown by MOCVD technique on InP wafers with different indium concentrations. During the growths, in-situ reflectance measurements are done at growth temperatures. Post growth measurements such as thicknesses of the films are done using SEM and spectroscopic ellipsometry at room temperature. Also derived properties such as refractive index was determined for both in-situ and ex-situ methods and compared with each other.

2. EXPERIMENTAL STUDIES

All the samples are studied in this work are grown using AIXTRON 200-4 RF/S horizontal reactor MOCVD system. TMIn, TMGa bubblers are used as indium (In) and gallium (Ga) precursors, AsH_3 is used as arsenic (As) source for growth of $In_xGa_{1-x}As$. Samples are grown as intrinsic layers so no doping sources are used. During the growth, high purity H_2 is used as a carrier gas. Before and after each growth the reactor chamber is pumped out and refilled with high purity N_2 in order to satisfy proper growth condition and clean reactor ambience. Four $In_xGa_{1-x}As$ samples are grown on (001) oriented single crystal 3 inches epi ready semi-insulating InP substrates. Prior to growth, substrates are heated up to growth temperature 640 °C under H_2 ambient. During the heating PH_3 line is also activated at 300 °C to clean the surface of substrate via desorption without any surface deterioration. In order to minimize defects propagating from substrate a thick 1µm InP buffer layer is grown homoepitaxially on substrate before InGaAs layer growths. Details of growth parameters are summarized in Table 1. During the growth, we kept the following growth parameters constant; Reactor pressure is at 50 mbar, sample rotation is at 60 rpm and total carrier gas flow is at 6000 sccm.

Table 1. Details of growth parameters.

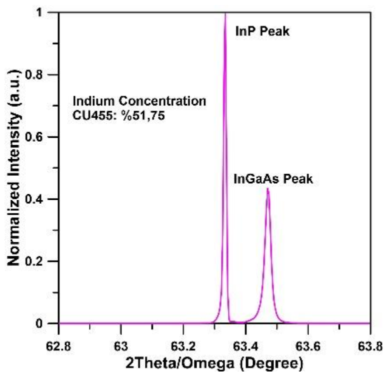
Sample Name	Flow Rates			Temperature (°C)	V/III Ratio
	TMIn (sccm)	TMGa (sccm)	AsH_3(sccm)		
CU455	165	4.2	100	640	70
CU475	227	9	200	640	88
CU476	114	4.5	100	640	87
CU477	103	4.5	100	640	92

In-situ reflectance measurement is used to determine the $In_xGa_{1-x}As$ layer thickness and will be discussed later. Rigaku SmartLab High Resolution X-ray Diffraction system is used to measure alloy composition of the $In_xGa_{1-x}As$ layers. Optical properties of the epitaxial layers are obtained with OPT-S9000 Spectroscopic Ellipsometry. Δ and ψ were measured by spectroscopic ellipsometry in the energy range of 0.756 eV - 2.067 eV (1640 nm-600 nm) at an angle of 65°. FEI Nova NanoSEM 450 Scanning Electron Microscopy System (SEM) is used as a direct thickness measurement system.

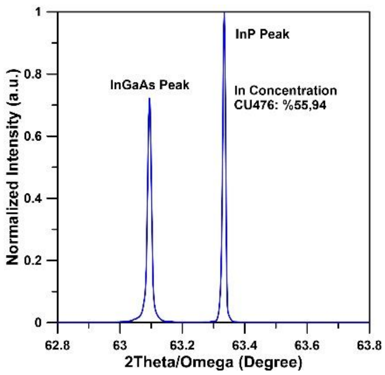
3.RESULTS AND DISCUSSIONS

3.1.X-ray Diffraction Results

Alloy composition of the $In_xGa_{1-x}As$ films have been investigated by means of high resolution x-ray diffraction via out-plane direction (004), symmetric 2-Theta/Omega measurements and result are given in Figure 1.



(a)



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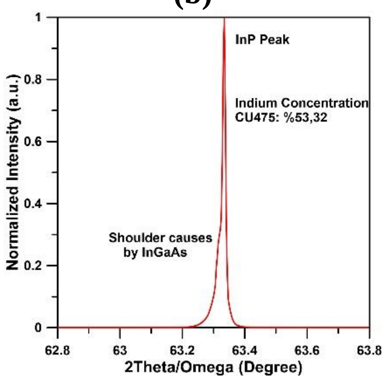

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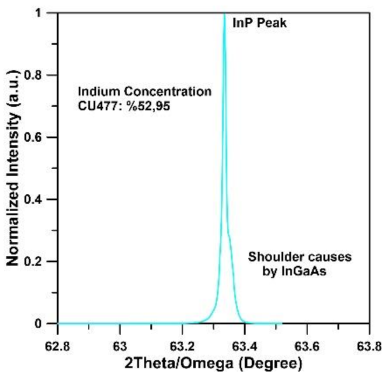

(d)

Figure 1. Symmetric out-plane 2Theta/Omega measurements.

According to the x-ray diffraction measurements, $In_xGa_{1-x}As$ peaks are on the right side of InP peak for the sample CU455 (Figure 1a), it means that crystal is under compressive strain through out-plane direction. Indium concentration of this sample is calculated as 51.75%. CU476 is the second lattice mismatch sample and $In_xGa_{1-x}As$ peaks are on the left side of InP peak so this crystal is grown under tensile strain Indium concentration of this sample is calculated as 55. 94%. CU475 and CU477 are lattice matched as clearly seen from Figure 1 b and d. One is shown up slightly on the left shoulder and the other is on the right shoulder of InP peak and their indium concentrations are 53.32% and 52.95%, respectively.

3.2.Optical Properties

Ellipsometry is a very sensitive technique to measure optical properties of the thin films and is mostly used to calculate thickness, refractive index etc. using polarized light. There are two polarization directions, if the polarization direction is perpendicular to the incident direction it is symbolized as s and if it is parallel as p. Complex Fresnel reflection coefficient (ρ) is determined by the ratio of polarized reflected lights r_s and r_p as [6]

$$\rho = \frac{r_p}{r_s} = (\tan \Psi) \exp(i\Delta) \quad (1)$$

ψ and Δ are measurable quantities and described as amplitude ratio and phase shift. Those quantities are used to calculate film thickness and refractive index (n) by using well known Fresnel equations [7].

In order to calculate those quantities, suitable mathematical model and Brewster angle knowledges are needed. In this study, Cauchy model [8] is used and related formula is given below.

$$n(\lambda) = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4} \quad (2)$$

In this model, A, B and C variables and those defines refractive index of the measured film. Most suitable values of the variables are found by fittings.

Δ and Ψ measurements and fits are given in Figure 2 and Figure 3. Measurements and fits are matching quite well, so accuracy of the obtained data are good.

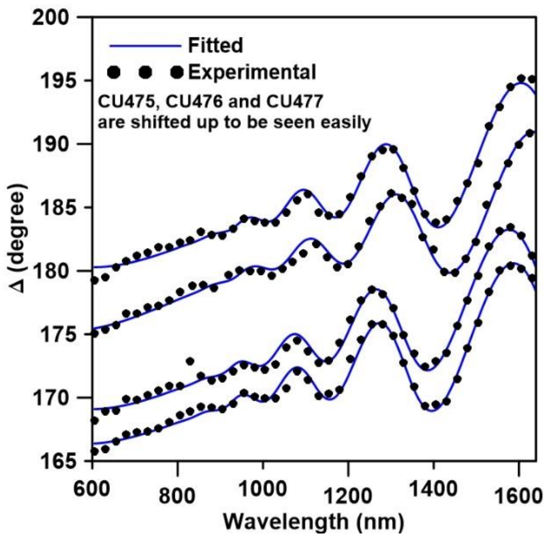

Figure 2. Experimental and fitted ellipsometric parameter (Δ) as a function of wavelength in the order from bottom to top CU455, CU475, CU476, CU477.

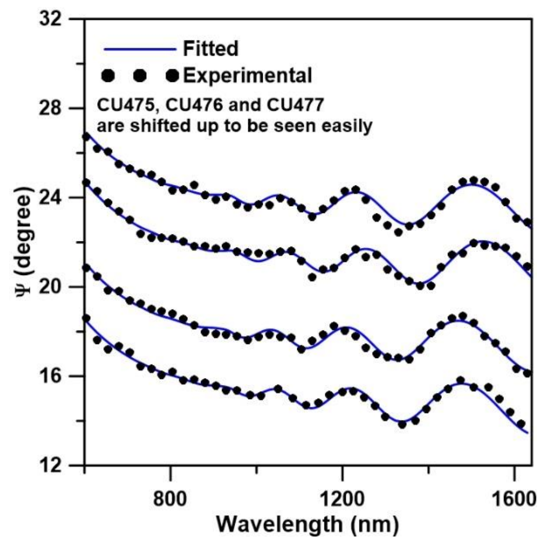

Figure 3. Experimental and fitted ellipsometric parameter (Ψ) as a function of wavelength in the order from bottom to top CU455, CU475, CU476, CU477.

As expected refractive index values of CU475 and CU477 are quite close to each other because their indium concentrations are almost the same. When indium concentration increases, refractive indices of the material increases and similar behavior is also reported in literature [9]. The results show that while CU455 has smallest refractive index CU476 has the highest as result of their indium concentrations as shown in Figure 4.

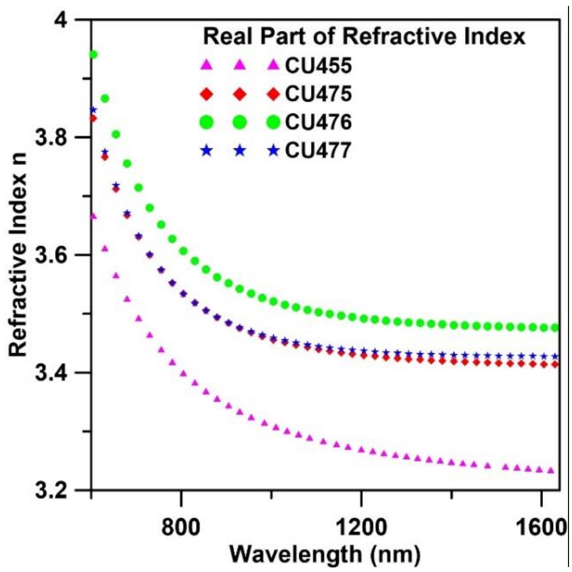

Figure 4. Wavelength versus refractive index for $In_xGa_{1-x}As$ films.

3.3.In Situ Reflectance Measurement

Real-time in-situ reflectance measurement during the growth is used to calculate thicknesses by measuring surface reflectance. 880 nm wavelength laser light is sent to the sample surface at 7 degree and then reflected laser light from the surface are detected as shown in Figure 5.

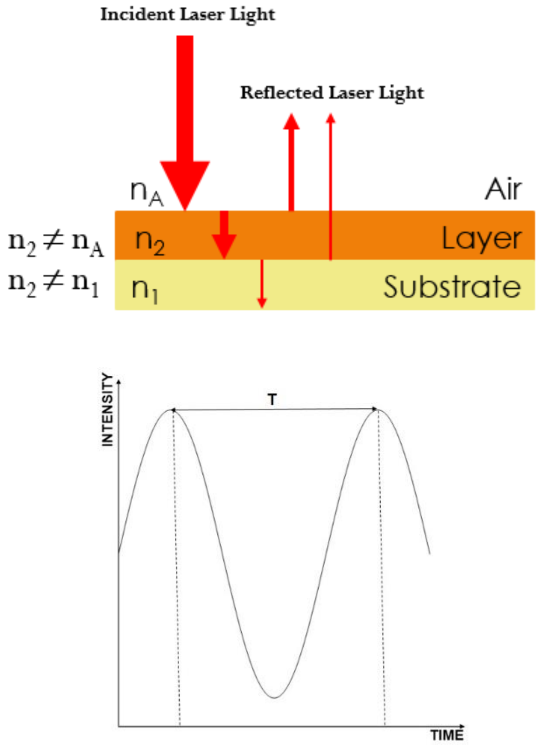

Figure 5. Representation of in situ reflectance measurement and Fabry-Perot oscillation.

n_1, n_2, n_A are refractive indices of the substrate, grown layer and air (air of reactor ambient). If refractive index of the substrate and grown layer are different, Fabry-Perot oscillations are observed as shown in Figure 6 [10]. Thickness of the one oscillation can be calculated by using refractive index of the grown layer and the wavelength of the laser light. Δd is the one oscillation thickness, λ is the wavelength of laser light, n is the refractive index of the layer, r is the growth rate and T is the one period oscillation time.

$$\Delta d = \frac{\lambda}{2n} \quad (3)$$

$$r = \frac{\Delta d}{T} \quad (4)$$

Intensity of the Fabry-Perot oscillations are directly related with the value of the refractive index difference and surface quality. If refractive index of the grown layer is higher than refractive index of the substrate, higher reflection is obtained. If not lower reflection is observed. Refractive index of the grown InGaAs samples in this study are higher than InP so that higher oscillations are seen. Within the frame of these rules, concentration of indium directly affects the refractive index of the grown layer [9, 11]. The lower indium concentration means relatively lower oscillation intensity. On the other hand, surface quality of the grown crystal is also important if surface roughness is high, then the laser light scatters from the surface decreasing the number of detected photon. That also causes a reflectance drop thus obtaining alloy concentration via reflection intensity is not conclusive. In Figure 6, in-situ reflectances of the samples are shown.

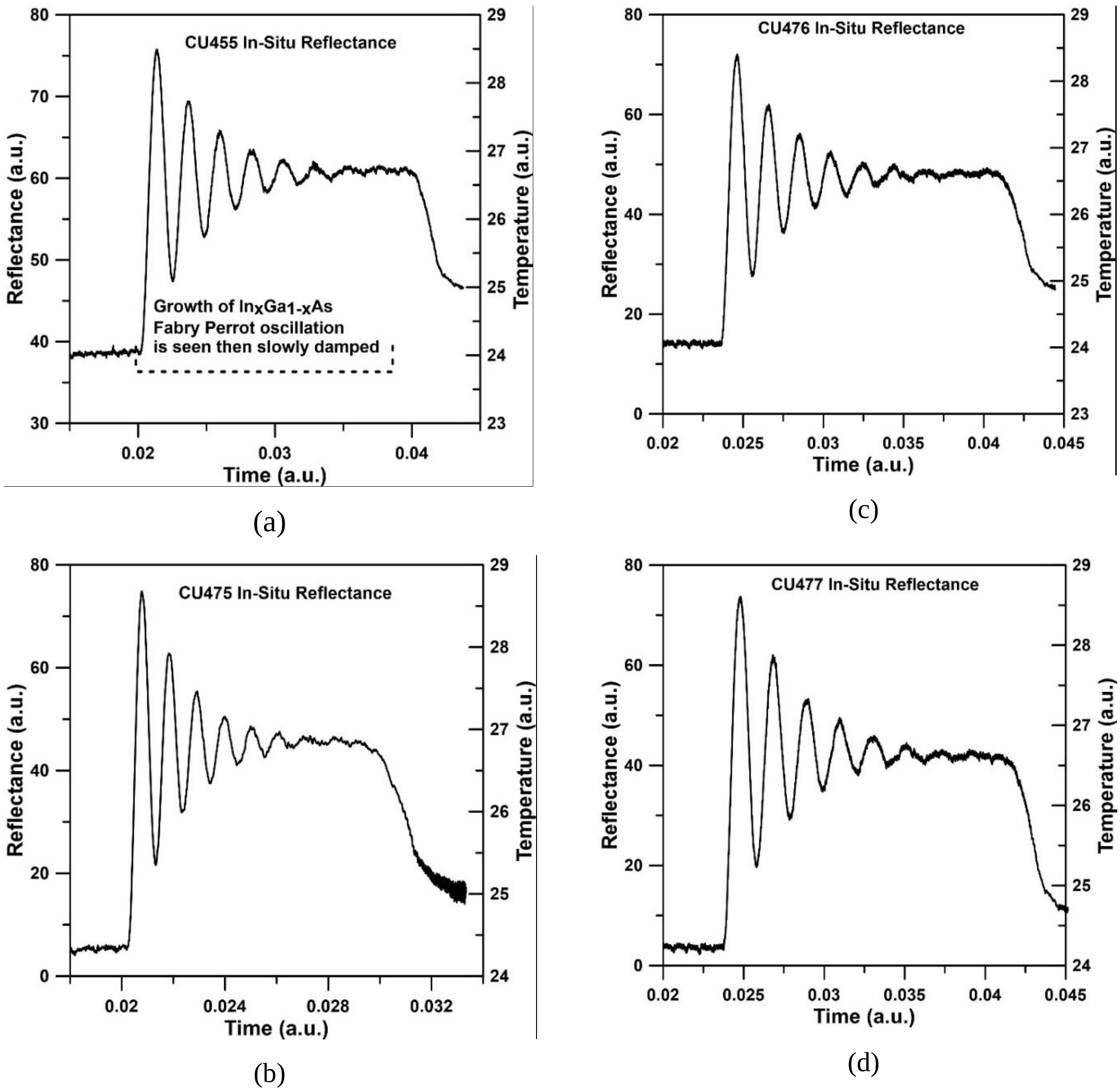

Figure 6. In-situ reflectance measurement of samples from top to bottom a) CU455, b) CU475, c) CU476 and d) CU477.

Fabry-Perot oscillations are seen after InGaAs grown started and as mention above each period has the same thickness so total thickness of the InGaAs layer can be estimated. As the layer thickness increases the thickness oscillations damps down because of the absorption of the laser light.

3.4. SEM Results

After optical studies, thickness confirmation is needed so that SEM measurement is done and related measurements are shown in Figure 7.

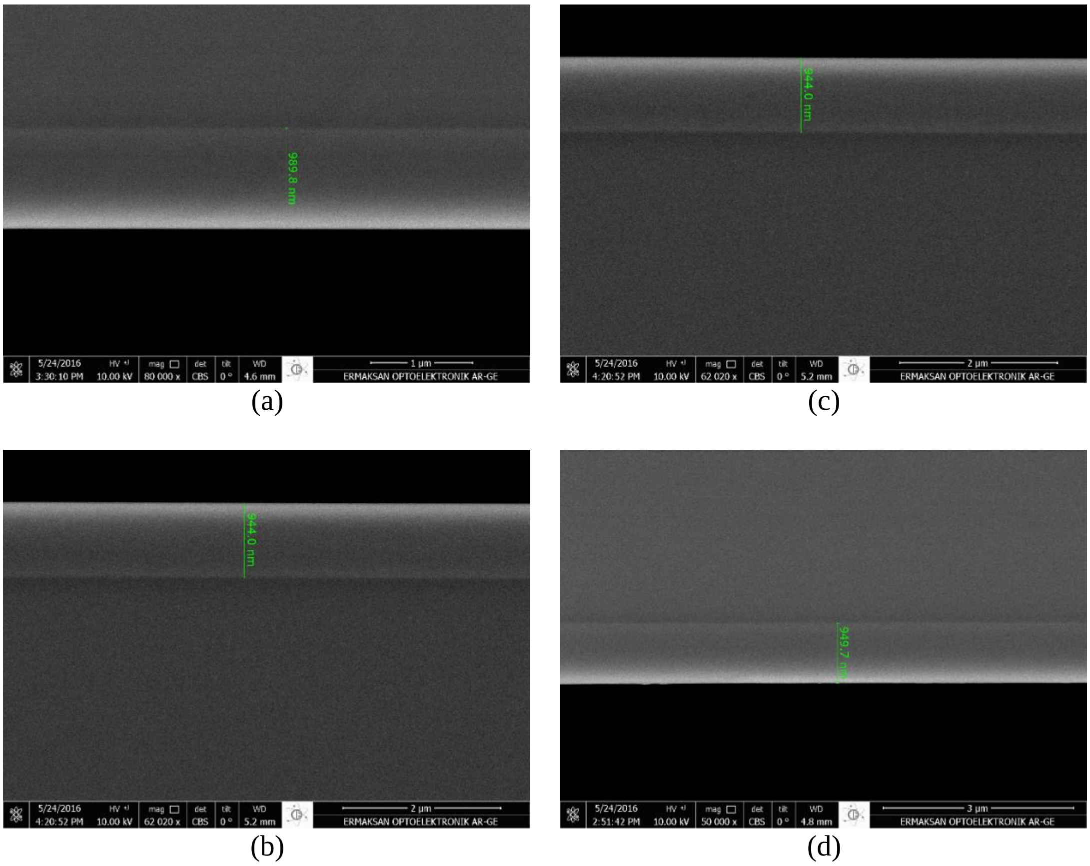

Figure 7. SEM measurements (a) CU455 (b) CU475 (c) CU476 (d) CU477.

All thicknesses are given in Table 2. SEM is direct thickness measurement system so that real thicknesses should be around those values and our results for two sample are in quite good agreement with the ellipsometry. Projected refractive indices are used to calculate thicknesses with in-situ measurement system because of the uncertainty about indium concentration of $In_xGa_{1-x}As$ layers during the growth. So, that affects the results. In order to avoid this error, thicknesses are also calculated after growth with refractive indices which are found with ellipsometry at room temperature. Those result are shown in Table 2 and values are higher than SEM results because room temperature refractive index values are lower than growth temperature ones. In order to overcome this error, we estimated growth temperature refractive index values (given in Table 2) to give the same thicknesses as SEM thickness values.

Table 2. Thicknesses and growth rates found with in-situ reflectance, ellipsometry and SEM.

Sample	Thicknesses (nm)			Refractive Indices at 880 nm	
	In-situ Reflectance at Growth Temperature	Ellipsometry	SEM	Ellipsometry	In-situ Reflectance at Growth Temperature
CU455	1088	990	990	3.354	3.685
CU475	1041	992	944	3.494	3.854
CU476	1070	1012	1014	3.562	3.760
CU477	1044	1009	950	3.493	3.841

Both the variation refractive index values obtained at room temperature using ellipsometry measurements and estimated refractive index values at growth temperature versus Indium concentration is given in Figure 8, respectively.

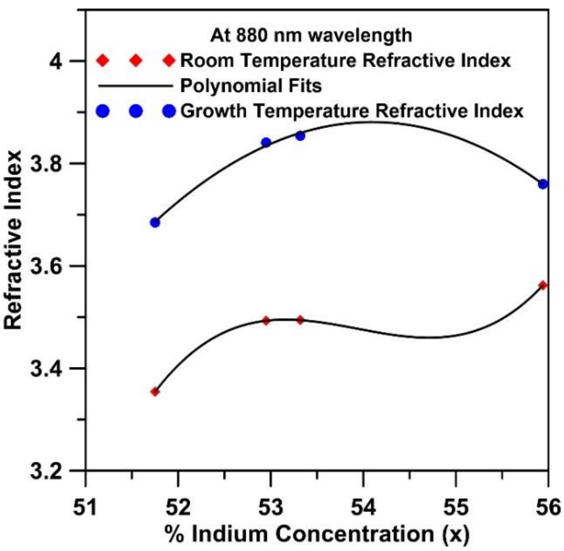

Figure 8. Dependence of the refractive index (n) at 880 nm of In_xGa_{1-x} films for room and growth temperature.

3. CONCLUSIONS

Four InGaAs samples were successfully grown by MOCVD technique on InP wafer with various indium concentrations. Indium concentrations are calculated by HRXRD system and strain types of the films determined. We obtain three different strain types to clearly see their effects on measurements, namely; compressive strain, tensile strain and strain-free (lattice matched). The effect of the indium concentration on the optical properties, refractive indices, of the films are measured with spectroscopic ellipsometry. Beside this, InGaAs layer thicknesses are estimated from in-situ reflectance measurements and ellipsometry and compared with SEM measurement results. The thicknesses obtained from SEM and the other measurement techniques must converge to each other. It is found that spectroscopic ellipsometry results are in good agreement with SEM results. However, the thickness values obtained from in-situ measurements using room temperature reflectance values are higher than the values obtained both SEM and ellipsometry measurements. As a result, it is clear that to use in-situ measurements effectively, refractive index values for the related growth temperature must be known otherwise in-situ measurements results must be confirmed and/or corrected by related ex-situ measurements.

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