# Two-dimensional mapping of composition in $In_xGa_{1-x}As$ using photoluminescence

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A computer-controlled scanning photoluminescence (PL) mapping system has been developed to investigate the two-dimensional variation of composition in bulk $In_xGa_{1-x}As$ crystals. The accuracy of PL results was confirmed by comparing with those obtained from the standard technique of chemical analysis. It was also confirmed that PL results were not limited by the sample size, shape and surface conditions. Besides the compositional homogeneity examination, the usefulness of PL peak intensity map was also found to check the crystallinity in $In_xGa_{1-x}As$ crystals.

## 1. Introduction

The composition of bulk $In_xGa_{1-x}As$ crystals must be uniform in microscopic range for using as a lattice-matched substrate material in various InGaAs-based laser diodes and optoelectronic devices. In order to investigate the spatial variation of composition in bulk $In_xGa_{1-x}As$ crystals, it is required to establish two-dimensional mapping system using a suitable nondestructive technique. Several techniques, such as electron probe microanalyser (EPMA), energy dispersive X-ray (EDX), Raman scattering (RS), and photoluminescence (PL) can be used for two-dimensional mapping of composition in bulk $In_xGa_{1-x}As$ crystals. Among them, the EDX and EPMA techniques are not convenient for large samples, because they require the measurements to be done under high vacuum. In our previous studies [1-5], it was found that RS is a good technique to estimate the composition as well as residual strain in bulk $In_xGa_{1-x}As$ crystals with high spatial resolution and accuracy. However, to get the better accuracy in RS measurements, it is necessary to record Raman spectra for longer time in $In_xGa_{1-x}As$ crystals, which makes it slower for mapping

of composition compared to other techniques. On the other hand, PL is a convenient fast characterization technique to investigate the two-dimensional variation of composition in $In_xGa_{1-x}As$ crystals with high spatial resolution and accuracy. Especially, if the sample is large, PL is the best technique to investigate the entire sample with the spatial resolution of a few micron. Both peak wavelength and intensity in PL spectra are changed, respectively, due to the change in composition and crystal orientation. Therefore, the two-dimensional PL map is suitable for simultaneous determination of compositional homogeneity and crystallinity. For the first time, we develop a computer-controlled scanning PL mapping system to characterize the two-dimensional variation of composition in $In_xGa_{1-x}As$ crystals. Using various standard polycrystalline samples, the reliability of PL results was confirmed by comparing with those obtained from the standard technique of chemical analysis (CA). Although CA gives us good results, being a destructive nature, it is not suitable for routine characterization. After confirming the accuracy of PL results with CA, the compositional profiles from a

73

cylindrically-shaped polycrystalline feed material with as-grown surface conditions were measured both by PL and by RS. A good comparison between the estimated compositional profiles indicates that PL results are not limited by the sample size, shape and surface conditions. It is noted here that RS characterization of composition in polycrystalline feed material was already confirmed by CA in our previous studies [2,3]. In this report, we also present some results of the two-dimensional variations of composition and integrated PL peak intensity for various wafers obtained from a crystal grown by the normal freezing technique. The PL maps obtained from various wafers indicate that the composition was changed drastically at the centers of the wafers, where PL peak intensity was also changed drastically.

The drastic change in PL peak intensity is associated with the change in crystal orientation, which was confirmed by X-ray measurements. At present; our experimental setup is limited to $0 \leq x \leq 0.5$ due to the detector sensitivity, which can be extended up to the whole range in composition with the combination of InGaAs and InAs detectors. Instead of further improvement, the established PL experimental setup is enough to investigate the compositional homogeneity and crystallinity in ground- and space-grown crystals having the composition $x = 0.3$. Therefore, PL and RS is a good experimental combination to characterize the compositional profile in feed material for ground and space experiments as well as to check the compositional homogeneity in grown crystals.

## 2. Experimental setup

Figure 1 shows the block diagram of our PL apparatus developed in the present work.
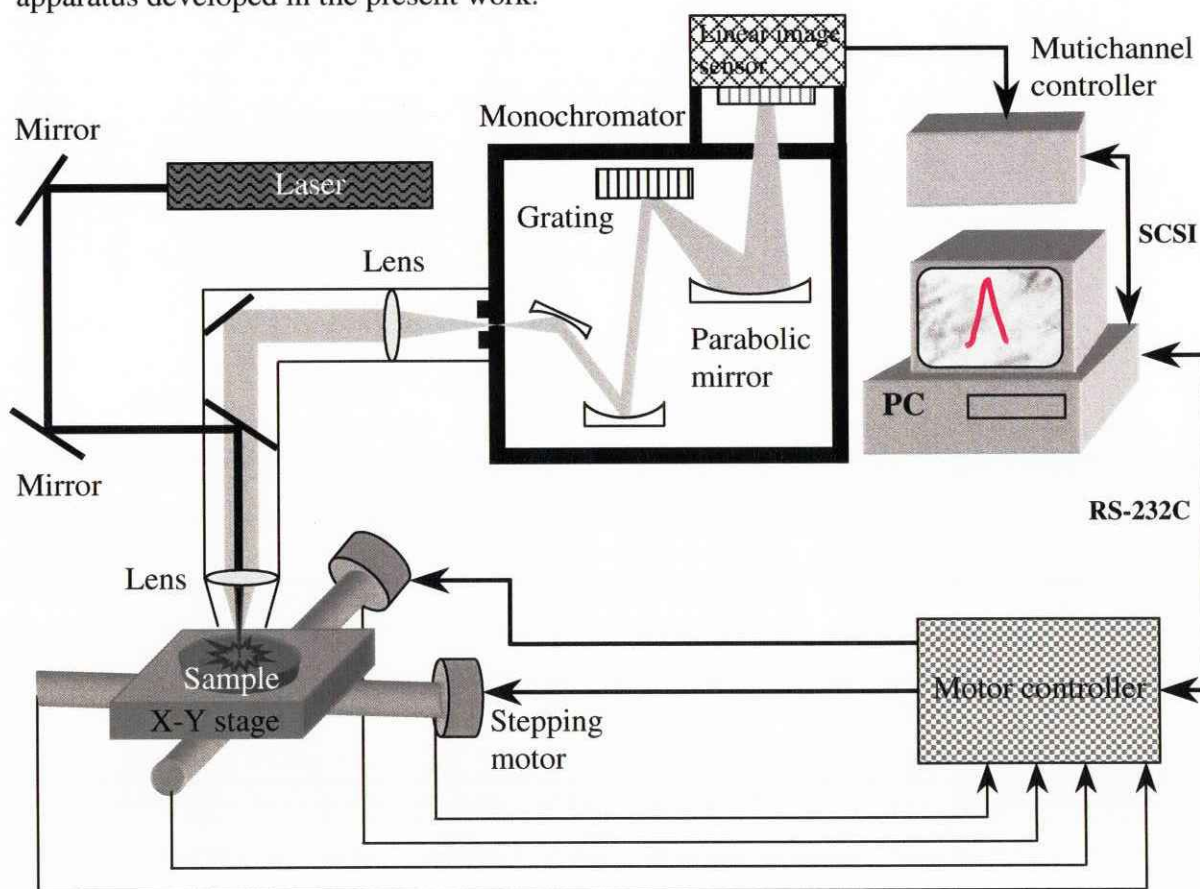


Fig. 1 Block diagram of scanning PL apparatus.

74

Fig. 2 Picture of scanning PL apparatus.

The scanning PL mapping system consists of a 17 mW He-Ne laser, computer controlled X-Y translational stage, motor controller, monochromator, InGaAs detector, and A/D converter. The working power and probing beam diameter of the laser is about 8 mW and 10 μm, respectively. The sample is scanned by a computer-controlled automatic X-Y translational stage, whose minimum step is about 2.5 μm. The optical configuration of the apparatus is shown in the block diagram. A cooled linear-array InGaAs detector is used to detect the PL signal. The wavelength range of the detector is from 0.9 to 1.55 μm, which corresponds to the composition range of $0 \leq x \leq 0.5$. The estimated composition range can be extended up to the whole range by using a InAs detector with sensitivity in the range from 1 to 3.8 μm. Therefore, with the combination of InGaAs and InAs detectors, PL can be used to measure the entire range of composition in $In_xGa_{1-x}As$ crystals.

## 2. Experimental procedure

In the present study, PL experiments were performed in polycrystalline and crystalline bulk $In_xGa_{1-x}As$ in two steps. In the first step, standard polycrystalline samples, which were prepared with various compositions from $x=0.06$ to $x=0.5$ to confirm the PL results with CA. The standard samples were prepared in the form of square shaped from a polycrystalline ingot, which had the graded composition along the growth direction. After confirming the PL results with CA, a cylindrically-shaped polycrystalline feed material with the diameter of 32 mm and the length of 140 mm length was measured with as-grown surface conditions. The feed material had the gradient in composition along the growth direction. The polycrystalline sample was prepared by using a vertical heating furnace [6].

In the next step, an $In_xGa_{1-x}As$ single crystal of 15 mm diameter was grown by the normal freezing technique, which had a graded composition along the growth direction. This crystal was sliced into several wafers perpendicular to the growth direction at 5, 15, 25 and 40 mm growth lengths from the seed. The surfaces of the wafers were mirror polished to characterize the spatial variation of composition.

The wafers were investigated at room temperature using the PL mapping system developed in the present work. The wafers were scanned by the computer-controlled X-Y translational stage at 50 μm spatial resolution. Peak position of PL spectrum was determined by line-shape fittings.

## 4. Results and discussions

### 4.1 Standard polycrystalline samples

Figure 3 shows PL spectra, which were measured from the centers of various standard polycrystalline samples. The peak wavelengths measured from various samples were found to vary from sample to sample in Fig. 3, because the peak wavelength position in PL spectra is strongly associated with the alloy composition. It is possible to estimate the composition from the PL spectra using the compositional dependence of band gap energy. The band gap energy was assumed to be at the peak wavelength of the spectra and then using the equation: $E_g(x) = 1.424 - 1.564x + 0.494x^2$, which was expressed at room temperature in a previous study [7], the composition as estimated for various polycrystalline samples. It is noted here that the estimated range of composition was limited to $0 \le x \le 0.5$ due to the detector sensitivity, which corresponds to the wavelength range from 0.9 to 1.55 μm in our experimental setup. Compositions in these samples were also determined independently by the standard method of CA. Figure 4 shows a good comparison between the
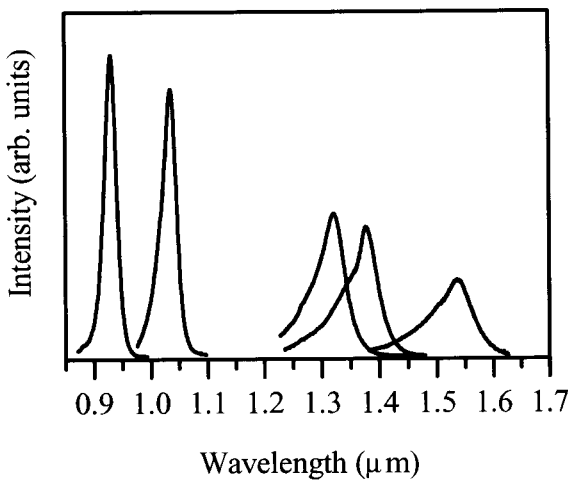
compositions estimated by PL and CA. The average deviation of the PL results from those obtained from the CA was found to be 7.5%. The compositions measured by PL could be deviated from CA, because these were estimated by assuming that peak wavelength positions are corresponding to the band gap of the material. It is observed in Fig. 3 that the PL lineshape is asymmetric with tail in the higher energy side. This is because, at room temperature, energy states are occupied by carriers following Boltzman distribution. Therefore, the peak wavelength positions in PL spectra could be shifted slightly from the band gap of the material. However, since the compositions estimated from the peak wavelength positions in PL spectra show small deviation with CA, PL can be used to estimate the composition in $In_xGa_{1-x}As$ crystals.

### 4.2 Polycrystalline feed material

After confirming the accuracy of PL results, a cylindrically shaped polycrystalline feed material of 32 mm diameter and 140 mm length was measured with as-grown surface conditions. Figure 5(a) shows the image of the feed material, where the filled white circles indicate some of the measurement points. The compositional profiles in this feed material
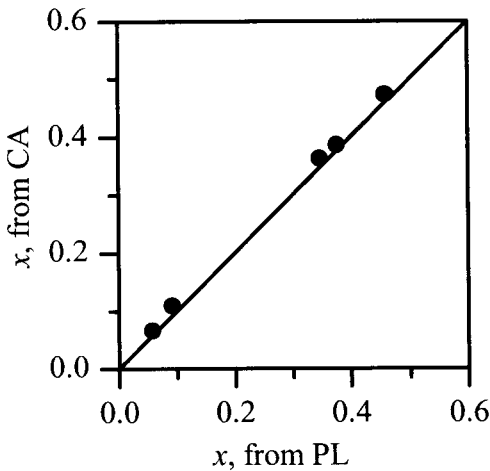


Wavelength (μm)

Fig. 3 PL spectra measured in polycrystalline standard samples.



$x$, from PL

Fig. 4 Comparison between the compositions determined by PL and CA measurements in polycrystalline standard samples.

76
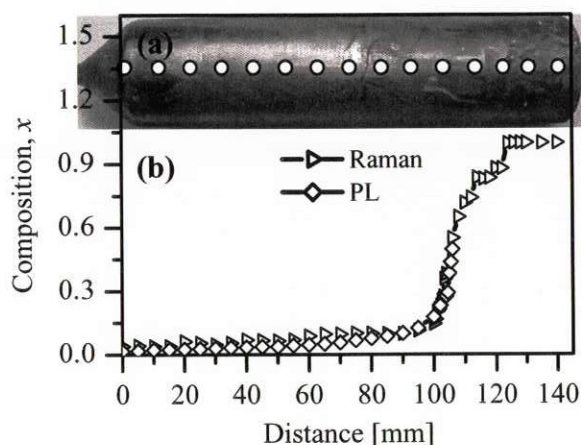
Fig. 5(a) Polycrystalline In$_x$Ga$_{1-x}$As feed material. Symbols represented by filled white circles indicate some of the measurement points (b) Composition profiles measured by RS and PL. PL measurements are limited to $0 \leq x \leq 0.5$ due to the detector sensitivity.

were determined independently by PL and RS. Figure 5(b) shows a good comparison between the compositional profiles estimated by PL and RS. It is observed in Fig. 5(b) that the estimated compositional profiles vary from 0.02 to 1.0, where their variations were found to be very slow from one end of the sample, however, they start changing drastically close to the other end. It is noted here that Raman results in standard polycrystalline In$_x$Ga$_{1-x}$As materials were confirmed by the CA in our previous studies [1-3], where Raman measurements were not limited to the sample size, shape, and surface conditions. Due to the detector sensitivity as mentioned earlier, PL measurements were limited to $0 \leq x \leq 0.5$. However, with the combination of InGaAs and InAs detectors, it is possible to measure the whole range of composition in In$_x$Ga$_{1-x}$As crystals. Since the compositions were estimated with as-grown surface condition of the sample and the estimated compositional profile shows good agreement with that obtained from RS, it was confirmed that PL measurements are also not limited by the sample size, shape and surface conditions. Therefore, as a fast characterization and accurate technique, PL is well-situated to characterize the compositional profile in feed materials, which is required to optimize the growth parameters in traveling liquidus zone (TLZ) growth method.

### 4.3 Crystalline wafers

Various In$_x$Ga$_{1-x}$As wafers were measured at spatial resolution of 50 µm using the two-dimensional PL mapping system. Figure 6 shows peak wavelength (composition) and integrated intensity profile maps corresponding to the wafers sliced at the 5, 15, 25 and 40 mm growth lengths of the crystal. The PL maps plotted for various wafers indicate that peak wavelength (composition) is increasing slowly from $\lambda = 0.911$ to 0.959 µm ($x = 0.04$ to 0.085) along the growth direction of the crystal. It is observed in Fig. 6 that the composition map obtained from the wafer sliced at the 5 mm growth length from the seed shows the existence of compositional homogeneity throughout the entire wafer. However, the maps obtained from the wafers sliced at the 15, 25 and 40 mm growth lengths from the seed show lower In-content at the central regions of the maps, where it changes drastically compared to the surrounding. Apart from the central regions, the variation of composition is also observed in other regions of the maps. It is found in Fig. 6 that some peak wavelength (composition) maps show sharp white lines, which were plotted for the wafers sliced at the 5 and 15 mm growth lengths, however, such lines were not found in the same maps plotted for the wafers sliced at the 25 and 40 mm growth lengths. These white lines could be appeared due to the scratches on the sample surface. The drastic fluctuation of composition in the local region could be originated due to the convection-induced or constitutional

77

From seed ⇩

Peak wavelength

λ=0.922μm (*x*=0.05)

λ=0.911μm (*x*=0.04)

5mm

λ=0.927μm (*x*=0.055)

15mm

λ=0.917μm (*x*=0.045)

λ=0.932μm (*x*=0.06)

25mm

λ=0.922μm (*x*=0.05)

λ=0.959μm (*x*=0.085)

40mm

λ=0.943μm (*x*=0.07)

Integrated intensity

800 c/s
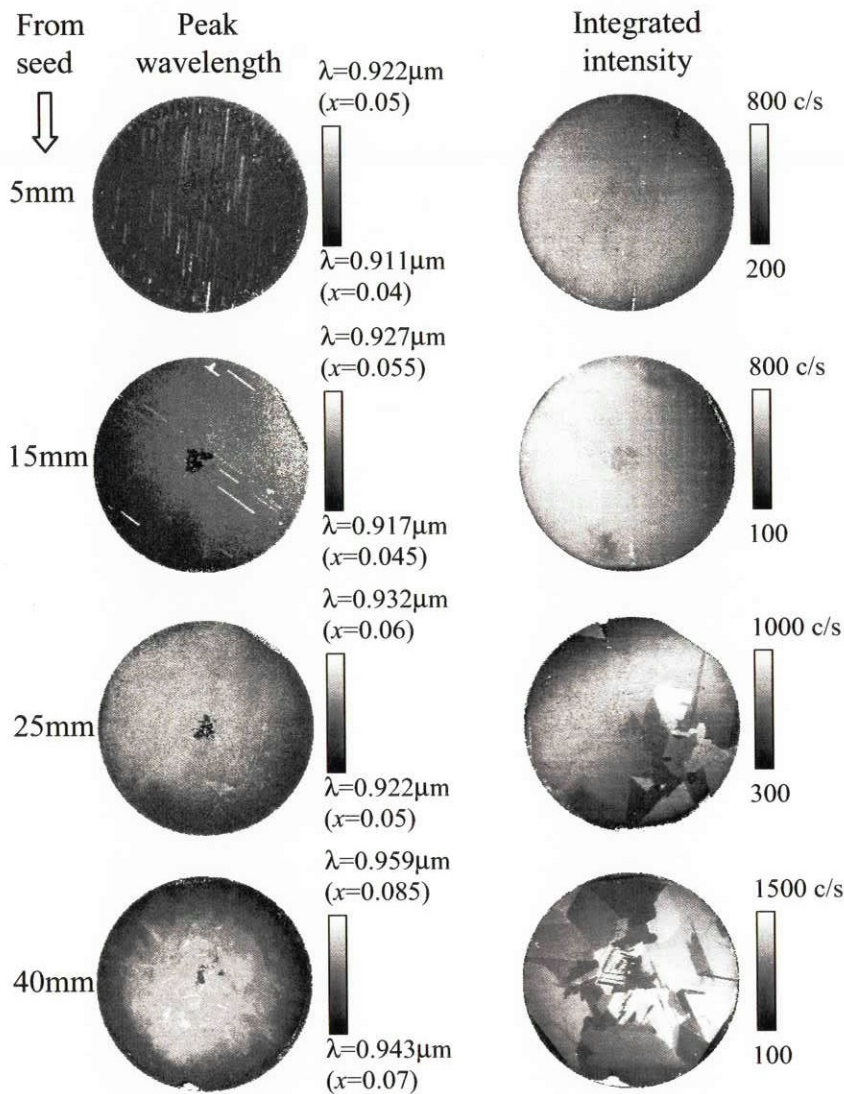
200

800 c/s

100

1000 c/s

300

1500 c/s

100

Fig. 6 2D-maps of PL peak wavelength (composition) and integrated intensity measured in various samples.

supercooling, which was suggested in our previous study [8]. In another recent study [4, 5], it was suggested that residual strain is induced in bulk $In_xGa_{1-x}As$ single crystals due to the compositional inhomogeneity. Since compositional inhomogeneity exists in the investigated wafers, the measured PL peak wavelengths should have the influence of strain. The integrated intensity profile maps shown in Fig. 6 indicate that the variation of PL peak intensity is almost smooth in the wafer, which was sliced from the 5 mm growth length from the seed. However, it changes drastically at the centers of the wafers, which were sliced at the 15, 25 and 40 mm growth lengths from the seed. The drastic fluctuation of PL peak intensity is associated with the change in crystal orientation, which

was confirmed by X-ray measurements for a wafer, where crystal orientation was changed by about 1.5 degree. It is observed in Fig. 6 that polycrystallization was initiated at the center of the wafer, where the composition was changed drastically. It is also observed that polycrystalline region starts extending from the center to the periphery, which is clearly appeared in the intensity maps plotted for the wafers sliced at the 25 and 40 mm growth lengths from the seed. Therefore, two-dimensional PL maps of composition and intensity are very important to investigate the bulk $In_xGa_{1-x}As$ crystals. Although, the present experimental setup is limited to estimate the composition $0 \le x \le 0.5$, the present experimental setup is enough to characterize the spatial variation of

78

composition of space- and ground-grown crystals having the composition $x = 0.3$.

## 5. Conclusions

Using the computer-controlled scanning PL mapping system, two-dimensional variations of composition and integrated intensity profile have been investigated in various wafers obtained from a bulk $In_xGa_{1-x}As$ crystal grown by the normal freezing technique. The accuracy of PL results was confirmed by using various standard polycrystalline samples. The compositions estimated at the centers of various polycrystalline samples show a good agreement with those determined by the standard technique of CA. Since PL results are not limited by the sample size, shape and surface conditions, as a fast characterization technique it is suitable to characterize the feed materials for TLZ growth method.

Two-dimensional variation of PL maps indicates that the composition is increasing slowly along the growth direction of the crystal. The drastic change in composition was found at the centers of the wafers, where integrated PL peak intensity was also changed drastically indicating that polycrystallization was initiated from the centers of the wafers. This polycrystalline region was found to increase in size with increasing the growth length. Our previous study [8] suggested that the drastic fluctuation of composition in the local region could be originated due to the supercooling resulting in polycrystallization in $In_xGa_{1-x}As$ crystals. Using two-dimensional

PL mapping system, the spatial variations of composition and crystal orientation were investigated simultaneously, which are vital information for the crystal grower to optimize the growth parameters, and hence to improve the crystal quality.

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