Japanese Journal of Applied Physics, **29** (8) (1990) 1435-1436

## Characterization of GaAs and AlGaAs Layers Grown by Laser Atomic Layer Epitaxy

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(Received April 19, 1990; accepted June 16, 1990)

Raman spectra were measured at 300 K to characterize GaAs and AlGaAs layers grown by laser atomic layer epitaxy. The quality of the GaAs patterned layer grown by laser scanning was uniform in spite of the laser intensity profile. The molar fraction of Al in the peripheral region of the AlGaAs layer is affected by the intensity profile of the laser beam.

KEYWORDS: Raman scattering, atomic layer epitaxy, MOVPE, gallium arsenide, aluminum gallium arsenide Atomic layer epitaxy of the GaAs/AlGaAs system is an attractive method since this method seems to be a promising candidate for producing thin epitaxial layers and abrupt interfaces controlled in the one-atomic-layer scale. To achieve atomic layer epitaxy, laser ALE (laser atomic layer epitaxy) has been used for GaAs crystal growth.<sup>1)</sup> Application of laser ALE to line patterning is possible by scanning laser beam. The thickness of the epitaxial layer is expected to be uniform since the growth rate is independent of the laser power. However, the crystal quality of the small area grown by laser ALE is not measured. In this paper, we describe the characterization of thin and small regions of epitaxial layers using Raman scattering.

Epitaxial layers were grown on (100)-oriented Si-doped (n =  $10^{18}$  cm<sup>-3</sup>) GaAs substrates in a low-pressure MOVPE (metalorganic vapor-phase epitaxy) system. The growth condition of epitaxial layers is listed in Table I. The TEG (triethylgallium) and AsH<sub>3</sub> were switched on and off alternately. A laser beam from an Ar laser (NEC GLG-3300,  $\lambda$  = 488.3 nm and 514.5 nm) was also switched by a shutter and was introduced into the reactor for irradiating a substrate surface. One cycle for epitaxial growth consists of a supply of TEG for 1 s followed by a purge for 1 s and a supply of AsH<sub>3</sub> for 1 s followed by a purge for 1 s. Thus, one cycle was completed in 4 s. Laser irradiation was performed concurrently with the introduction of TEG. The growth rate of the epitaxial layer was 0.28 nm/cycle (one monolayer/cycle), so that the layer with a thickness of about 0.4 µm was grown with a 1500 cycle.

A GaAs line pattern was grown by scanning the laser beam (scanning rate = 10 Hz). The size of the epitaxial line was 0.1 mm x 3.5 mm.

The AlGaAs was grown by introducing TEA (triethylaluminium) and TEG simultaneously. The laser beam was not scanned, so that the shape of the epitaxial layer was elliptical. The growth area was about 0.4 mm<sup>2</sup>.

Raman spectra have been measured at room temperature in backscattering geometry, using an Ar laser (spectra Physics 168B,  $\lambda = 514.5$  nm). The diameter of the laser beam was 0.1 mm. Laser power was about 100 mW at the sample surface. The scattered light was analyzed by a 1 m double monochromator (Jovin Yvon U-1000). The spectra were measured with a photomultiplier (Hamamatsu R464SS), a photon counter (Hamamatsu C1230-S), a computer and an X-Y plotter.

Figure 1 shows Raman spectra of the patterned epitaxial line of GaAs: a) the central part, b) the peripheral region and c) the GaAs substrate. A pure LO (longitudinal optical) phonon line at 292.5 cm<sup>-1</sup> and a small peak at 269 cm<sup>-1</sup> are observed in the GaAs substrate. The frequency shift of the 269  $\text{cm}^{-1}$  line is close to the TO (transverse optical) phonon mode and LO phonon-plasmon coupled mode.<sup>2)</sup> The TO phonon mode is forbidden for backscattering from a (100) surface. If the crystal orientation is different from the expected (100) direction, the TO phonon mode is observed. However, the 269 cm<sup>-1</sup> line is not observed in the epitaxial layer. Therefore, this line is not ascribable to the TO phonon mode. The LO phonon-plasmon coupled mode is observed in GaAs with a high carrier density. The observed frequency of the coupled mode depends on carrier density. The coupled mode is observed at about 269 cm<sup>-1</sup> in n-GaAs with a donor density in the range of  $n \ge 2 \times 10^{18}$  cm<sup>-3</sup>.<sup>3</sup> Since donor density in the substrate is almost in this range, the 269 cm<sup>-1</sup> line is considered to be attributable to the LO phonon-plasmon coupled mode. The pure LO phonon mode at 295.2 cm<sup>-1</sup> originates from the surface depletion layer. The depth of the depletion layer is about 20 nm for the electron density 2 x  $10^{18}$  cm<sup>-3</sup>.<sup>4)</sup> Since the penetration depth of 514.5 nm light is about 100 nm, both the pure LO phonon mode and the coupled mode are observed.

Only a pure LO-phonon mode is observed in the epitaxial layer, as shown in Figs. 1(a) and 1(b). The spectral shape of the 295.2 cm<sup>-1</sup> line of the epitaxial region is similar to that of the substrate. The Raman spectrum of the central part (a) is also similar to that of the peripheral part (b). These results indicate that the quality of the epitaxial line is almost the same as that of the bulk crystal and that the patterned layer is of uniform quality. The carrier density in the epitaxial layer is less than 5 x  $10^{17}$  cm<sup>-3</sup> since the LO phonon-plasmon coupled mode is not observed.

Figure 2(a) shows the Raman spectrum of the central part of the AlGaAs epitaxial layer. Two lines are observed: GaAs-type LO phonon mode (287 cm<sup>-1</sup> line) and AlAs-type LO phonon mode (365 cm<sup>--1</sup> line). The dependence of frequency shifts on the molar fraction x of Al in Al<sub>x</sub>Ga<sub>1-x</sub>As has been reported by Abstreiter et al.<sup>2)</sup> The value of x = 0.15 is obtained with the frequency shifts of the two Raman lines. Figure 2(b) shows a Raman spectrum of the peripheral region of the epitaxial layer. Two lines are observed at 284.5 cm<sup>-1</sup> (GaAs type) and 369cm<sup>-1</sup> (AlAs type). These Raman shifts lead to the

value of x = 0.21. The Al concentration is related with the spatial profile of the laser beam. It is expected that the Al/Ga ratio in the laser-irradiated region is less than that in the nonirradiated region since the TEG decomposition is enhanced by the irradiation more than that of TEA. Since the spectral shapes of the GaAs-type LO phonon mode are similar to that of the GaAs substrate shown in Fig. l(c), the quality of the AlGaAs layer is almost the same as that of the GaAs bulk crystal. Detailed results on the AlGaAs layer are reported elsewhere.<sup>5)</sup>

In summary, Raman spectra were measured in a thin GaAs patterned layer and in an AlGaAs layer grown by laser atomic layer epitaxy. The spectral shape of the Raman line shows the GaAs patterned layer to be uniform and of high crystalline quality in spite of the intensity profile of the laser beam. Shifts of the Raman lines show that the molar fraction of Al in the peripheral region of the AlGaAs layer is influenced by the laser intensity profile.

## References

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  S. Hunklinger, W. Ludwig and G. Weiss (World Scientific, Singapore, 1990) Vol. 2, p. 1445.

## **Figure captions**

- Fig. 1. Raman spectra of the GaAs line pattern grown by laser atomic layer epitaxy: a) the central part, b) the peripheral region and c) the GaAs substrate.
- Fig. 2. Raman spectra of Al<sub>x</sub>Ga<sub>1-x</sub>As epitaxial layer grown by laser atomic layer epitaxy:a) the central part, b) the peripheral region.

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	GaAs line pattern	AlGaAs
Laser power	140	200 W/cm <sup>2</sup>
Growth temperature	360	350–355°C
Flux of TEG	4.7 x 10 <sup>-7</sup>	$2 \times 10^{-7}$ mol/cycle
Flux of TEA	0	1.2 x 10 <sup>-7</sup> mol/cycle
Flux of AsH <sub>3</sub>	3 x 10 <sup>-5</sup>	3 x 10 <sup>-5</sup> mol/cycle
Total flow rate of		
H <sub>2</sub> carrier gas		2.8 x 10 <sup>-3</sup> m <sup>3</sup> /min

Table I. Growth conditions of epitaxial layers.

Fig. 1



Fig. 2

