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Solid phase epitaxy of molecular beam deposited amorphous GaAs on Si

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Solid phase epitaxial (SPE) crystallization of amorphous GaAs on (100) Si tilted by 4° toward ⟨011⟩ formed by molecular beam deposition (MBD) was first achieved by cw Kr laser irradiation for short durations. The ratio of As to Ga (y/x) in deposited amorphous Ga_xAs_y films was varied from 0.4 to 1.2. During the laser irradiation, movement of the amorphous/crystalline interface was measured using time-resolved optical reflectivity (TROR). It was found from TROR and micro-Raman scattering measurements that hetero-SPE is attained in samples with As/Ga ratios ranging from 0.8 to 1.1 and that the interface roughness is larger than that observed in homo-SPE (e.g., MBD GaAs on GaAs and P^+ ion-implanted GaAs).

Solid phase epitaxial (SPE) growth[1] is a unique technology for growth of thin films at relatively low temperatures. Recently, many studies on the formation of GaAs films on Si have been performed using molecular beam epitaxy (MBE)[2,3] or metalorganic chemical vapor deposition (MOCVD)[4] methods. However, there are several problems associated with these growth techniques including (1) thermal stress caused by the difference of thermal expansion between the film and the substrate, (2) degradation of surface morphology during deposition, and (3) generation of a large density of dislocations. In order to overcome these problems, SPE growth has been found to be useful and interesting because it is a low-temperature process. If a localized area of amorphous GaAs is selectively crystallized by using a cw laser beam, warping of the Si wafer can be avoided and a flat surface can be obtained.

In the two-step growth process,[4] an amorphous GaAs buffer layer is first deposited at a low temperature (< 120 °C). The layer has good surface morphology because the deposited atoms do not easily migrate on the Si surface.[4,5] Before the device layer is deposited, the wafer is annealed to crystallize the buffer layer by SPE. Good surface morphology is maintained even after annealing.

In this letter we report the hetero-SPE of molecular beam deposited (MBD)[5] amorphous GaAs on Si by laser beam irradiation. Hetero-SPE has been investigated by using time-resolved optical reflectivity (TROR)[6-9] measurements for SPE dynamics and by using micro-Raman scattering measurements for evaluation of crystallinity after SPE growth. The As to Ga ratios over which hetero-SPE growth is possible have been determined, and the roughness of the amorphous/crystalline interface during the SPE growth has been measured.

The substrate was a (100) Si wafer tilted by 4° toward ⟨011⟩. Before loading the substrate into a MBE chamber, the Si wafer was oxidized and etched in solutions of boiling HNO_3 and 50% HF. This process was repeated five times. Finally to protect the surface, the Si wafer was oxidized in a solution of boiling $HCl:H_2O_2:H_2O = 3:1:1$. In the MBE chamber at 10^{-9} Torr, the SiO_2 layer was removed by thermal etching at 800 °C. The structural change in the Si surface was observed by reflection high-energy electron diffraction (RHEED). After thermal etching, a 2×1 streak pattern of the (100) Si surface was observed. A monolayer of As atoms was deposited on the cleaned substrate at 700 °C and a 1×2 pattern[2] was observed from the deposited layer. Following the deposition of the As film, the amorphous Ga_xAs_y layer was deposited. The deposition was performed at 80 °C, and the thickness was 80 nm. The ratio (y/x) was determined using Auger electron spectroscopy.

Laser irradiation of the heterostructure was performed in an air ambient. An encapsulation layer was not used because the duration of irradiation was very short (from 10 ms to 20 s). A cw Kr laser ($\lambda = 647.0$ nm and 676.0 nm) beam was used to heat the sample and a cw He-Ne laser ($\lambda = 632.8$ nm) probe beam was used for the TROR measurement.[9] The cw Kr laser beam was elliptically focused to a spot $\sim 60\,\mu$m in the long axis and $\sim 40\,\mu$m in the short axis, and the irradiation time was controlled by a mechanical shutter. The size of the probe laser beam was 5 μm. The heating laser power was varied from 3.5 to 6.5 W and various irradiation times were selected for each laser power. The irradiation time was selected to be between the time when the SPE growth is completed and the time when surface degradation begins to occur.

Raman spectra[3] were measured in a backscattering geometry using a JASCO TRS-501S spectrometer with a R-MRS-11B microscope. The scattered light was detected by using photon counting techniques at -35 °C, and the detected signal was processed with a multichannel analyzer. An Ar laser with a wavelength of 514.5 nm served as the excitation source for the Raman measurements. The penetration depth at 514.5 nm is approximately 100 nm in the crystalline GaAs. The Ar laser beam was focused down to about 1 μm and the center in each crystallized region was irradiated by the focused beam.

In order to investigate the dependence of the SPE quality on the As/Ga ratio (y/x), amorphous Ga_xAs_y samples with different As/Ga ratios ranging from 0.4 to 1.2 were irradiated with Kr laser. Typical TROR signals are shown in Fig. 1. The heating laser power was kept at 5.8 W. The onset and end times of the laser irradiation are indicated by arrows in Fig. 1. The temperature of the GaAs layer is immediately raised within 1 ms from the onset.[6,7] The temperature during laser irradiation was estimated to be 380 °C by simply comparing the SPE velocity measured at $y/x = 1.0$ with the rate

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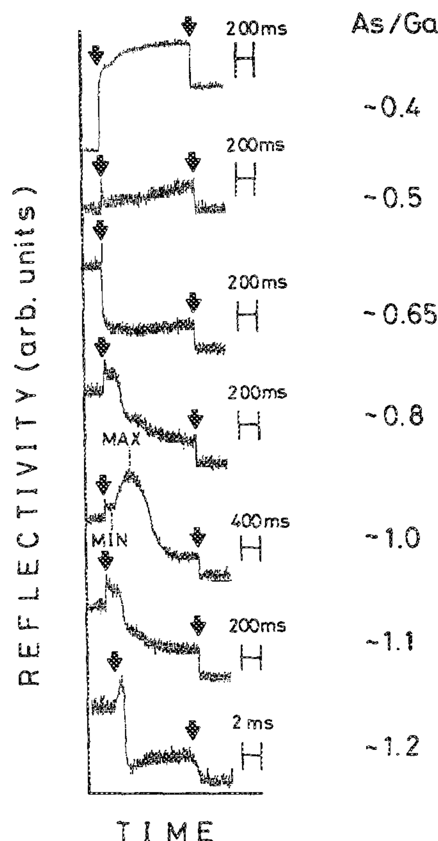

FIG. 1. TROR signals for various amorphous GaAs on Si samples with As/Ga ratios from 0.4 to 1.2 during laser irradiation. Laser power is 5.8 W and the temperature is estimated to be ~380 °C.

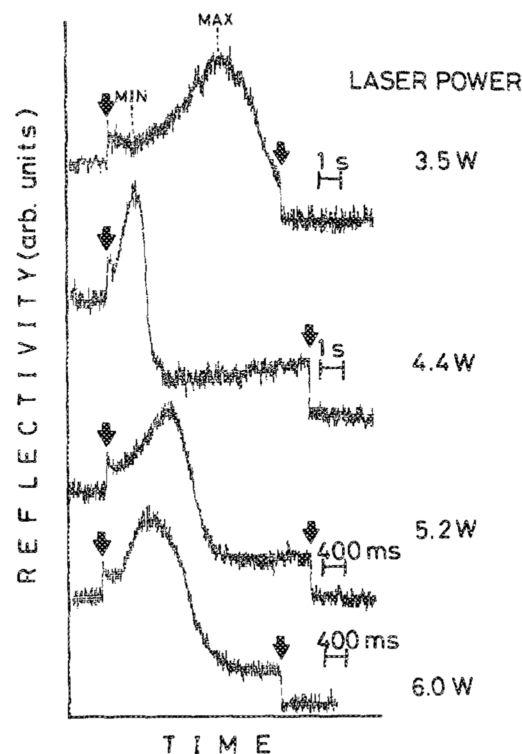

FIG. 2. Laser power dependence of TROR signals for amorphous Ga_1As_1 on Si. Laser power is varied from 3.5 to 6.0 W. The amplitude of TROR signal is strongly correlated with the roughness of amorphous/crystalline interface.

versus temperature data obtained previously for amorphous GaAs on GaAs.[5,6,8,9] The estimated temperature is higher than those ($\lesssim 300$ °C) previously used for SPE of amorphous GaAs on GaAs.[5] A clear interference indicating SPE growth was observed only for the case of $y/x = 1.0$, while small interferences were observed for both cases of 0.8 and 1.1. For other ratios such an interference was not evident, suggesting that another type of crystallization such as polycrystallization had occurred. It is found that SPE growth is successfully attained when the As/Ga ratio is between 0.8 and 1.1. This range is similar with that reported for SPE of amorphous GaAs on GaAs.[5] It is also noted that for $y/x = 1.2$, the change in reflectivity is very fast (<1 ms) and can be observed even at lower laser powers (1.2 W).

At the ratio y/x of 1.0, laser irradiation with various laser powers was performed in order to change the annealing temperature. Figure 2 shows TROR signals for various laser powers ranging from 3.5 to 6.0 W. As the laser power is increased, the amplitude of the TROR signal becomes smaller. It was observed in a cross-section view obtained by a transmission electron microscopy (TEM) measurement that formation of polycrystalline material ahead of the interface does not occur, but SPE with a rough interface takes place. These indicate that the roughness of an amorphous/crystalline interface increases with increasing the temperature. However, the surface morphology was not changed at higher laser powers used here.

In Table I, the interface roughness derived from TROR signals is compared for three types of amorphous GaAs: (1) amorphous GaAs produced by P^+ implantation (50 kV, 2×10^{15} cm^{-2}), (2) MBD amorphous GaAs on crystalline GaAs, and (3) MBD amorphous GaAs on Si. The thicknesses of the amorphous layers are ~80, ~65, and ~80 nm, respectively. The parameter in Table I is an estimated annealing temperature. We evaluated the interface roughness by a simple model in which the interface roughness is represented by Gaussian distribution[6,9] with standard deviation being defined as the interface roughness. The interface roughness of MBD amorphous GaAs on Si is found to be about two times larger than the others at the same temperature. This enhancement of roughness in the GaAs/Si system seems to be due to the initial heterointerface including the difference of lattice constant. In studies of the initial growth of MBE GaAs on tilted Si, it is known from high-resolution TEM that misfit dislocations are generated at the step edge which exist every ~4 nm.[10] Furthermore, there is 4% lattice

TABLE I. Interface roughness δ(nm) for three types of amorphous GaAs. In the case of MBD GaAs on Si, the roughness is much larger than other homo-SPE cases at the same temperature.

T_{sub} (°C)	P^+ implanted GaAs	MBD GaAs/GaAs	MBD GaAs/Si
340	35	<35	40
350	45	35	70
370	45	<45	100
380	50	40	110
400	60	50	···
450	90	>50	···

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that of the MBE GaAs layer on Si,[11] since the maximum laser output was limited to 6.5 W. It was recently found that laser irradiation at high powers for shorter times ($\lesssim 10$ ms) significantly improved the original crystallinity for the amorphous GaAs layer produced by P^+ implantation.[11] This suggests that very rapid SPE will yield better crystallinity also for MBD amorphous GaAs on a Si system, although the interface roughness during SPE becomes larger.

In summary, we have investigated laser solid phase epitaxy of the MBD amorphous GaAs on Si using TROR and micro-Raman scattering measurements. From the TROR measurements on amorphous GaAs on Si samples with different ratios of As to Ga, it is concluded that SPE growth can be successfully achieved for ratios between 0.8 and 1.1, while another type of crystallization occurred at other ratios. The roughness of an amorphous/crystalline interface for hetero-SPE is larger than that for homo-SPE.

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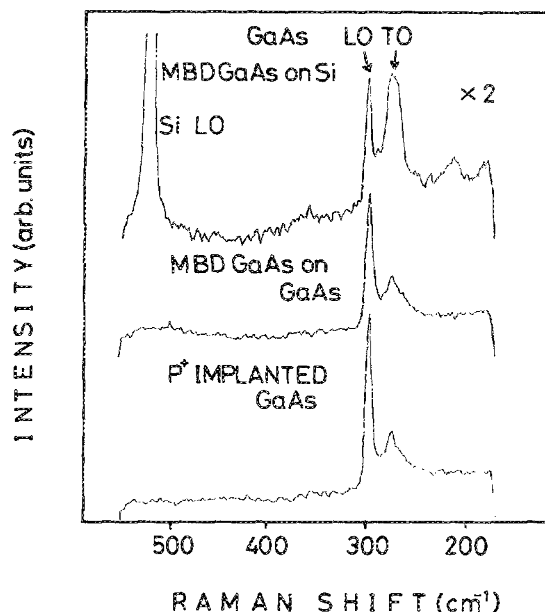

FIG. 3. Typical Raman spectra for MBD and ion-implanted GaAs crystallized by SPE. Amorphous GaAs on Si was annealed with a 6.5 W Kr laser beam for 20 s. Amorphous GaAs on GaAs substrate and ion-implanted amorphous GaAs layer were annealed with 1.8 W for 1.0 s and 2.2 W for 0.25 s, respectively.

mismatch between Si and GaAs, so that the generation of dislocations every ~ 10 nm[10] is needed to relax the lattice misfit.

We evaluated the crystallinity at the laser-annealed spot of amorphous GaAs by a micro-Raman scattering measurement. Typical Raman spectra are shown in Fig. 3. A LO phonon line of the Si substrate is also observed. For this backscattering geometry with (100) GaAs, only LO phonons are allowed from the selection rules. However, the forbidden TO phonons are observed as well as LO phonons. The TO phonons originate from regions with defects (e.g., twins and stacking faults) in the crystallized layer. With increasing laser power, the LO-phonon signal becomes larger, indicating better crystallinity. The best crystallinity obtained in this study (the top of Fig. 3 indicating the result for a sample annealed with 6.5 W for 20 s) was still worse than

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