Molecular beam epitaxy growth of GaAs on an offcut Ge substrate*

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Molecular beam epitaxy growth of GaAs on an offcut Ge (100) substrate has been systemically investigated. A high quality GaAs/Ge interface and GaAs film on Ge have been achieved. High temperature annealing before GaAs deposition is found to be indispensable to avoid anti-phase domains. The quality of the GaAs film is found to strongly depend on the GaAs/Ge interface and the beginning of GaAs deposition. The reason why both high temperature annealing and GaAs growth temperature can affect epitaxial GaAs film quality is discussed. High quality $In_{0.17}Ga_{0.83}As$/GaAs strained quantum wells have also been achieved on a Ge substrate. Samples show flat surface morphology and narrow photoluminescence line width compared with the same structure sample grown on a GaAs substrate. These results indicate a large application potential for III–V compound semiconductor optoelectronic devices on Ge substrates.

Keywords: molecular beam epitaxy, anti-phase domain, GaAs/Ge interface
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1. Introduction

III–V compound semiconductor materials grown on Ge substrates have received much research interest in recent years. The main driving force is the development of InGaP/(In)GaAs/Ge multi-junction high efficiency solar cells.[1−7] On the one hand, a Ge junction can absorb low energy photons that are transparent to InGaP and (In)GaAs material, so the conversion efficiency can be enhanced. On the other hand, a Ge substrate has the advantages of larger size, lighter weight, and higher mechanical strength than a GaAs substrate.[8] These two aspects are very important for solar cell applications. Ge-based solar cell efficiency has been improved from 25.3% in 1988[7] to over 40% since 2007.[9,10]

GaAs and Ge have almost the same lattice constants, but GaAs is a type of polar semiconductor, while Ge is non-polar. When GaAs is deposited on a Ge substrate, the main problem is to avoid the formation of an anti-phase domain (APD), which is very common in polar-on-non-polar epitaxy.[11,12] A simple solution is to make use of double-atom steps. Several groups have suggested that offcut Ge (100) substrates with an appropriate offcut angle could lead to double-atom steps on the surface.[13,14] However, it has been found that offcut Ge substrates cannot guarantee APD-free GaAs deposition. The quality of epitaxial GaAs also strongly depends on molecular beam epitaxy (MBE) or metalorganic chemical vapour deposition (MOCVD) growth conditions.[14−19] In the present work, MBE growth of GaAs on an offcut (100) Ge substrate is investigated. High temperature annealing before GaAs deposition is found to be indispensable to avoid the occurrence of APD. The quality of GaAs film is found to strongly depend on growth temperature.

2. Experiments

A Veeco Mod Gen-II solid source MBE system was used for all growth processes. As_4 was used as the arsenic species. Epi-ready (100) 6° offcut towards ⟨111⟩ Ge substrates, were bought from Axt Company and one of the substrate sides was chemical mechanical polished. Substrates were first baked at 180 °C for 3 h in an intro-chamber, then degassed at 420 °C for 0.5 h in a buffer chamber, and finally transferred into a growth chamber. In the growth chamber, the substrates were first annealed at a temperature T_1 for 20 min. Then the substrate temperature was low-

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ered to temperature T_2 with a rate of 30 °C/min, and the substrate was exposed to As_4 species (beam equivalent pressure of 8.25×10^{-4} Pa for 2 min. Then, a 400 nm GaAs layer was deposited with a growth rate of 0.8 μm/h. Finally, the substrate temperature was decreased to T_3 and an $In_{0.17}Ga_{0.83}As$/GaAs quantum well (QW) structure was grown on the 400 nm GaAs layer to characterise the quality of the GaAs film indirectly. The $In_{0.17}Ga_{0.83}As$/GaAs QW also has potential applications for semiconductor lasers and detectors. T_3 was chosen as 472 °C in all the samples grown on the Ge substrates. The thickness of the $In_{0.17}Ga_{0.83}As$ potential well layer was 6 nm, less than the theoretical critical thickness.[20] Figure 1 shows the schematic diagram of a sample structure.

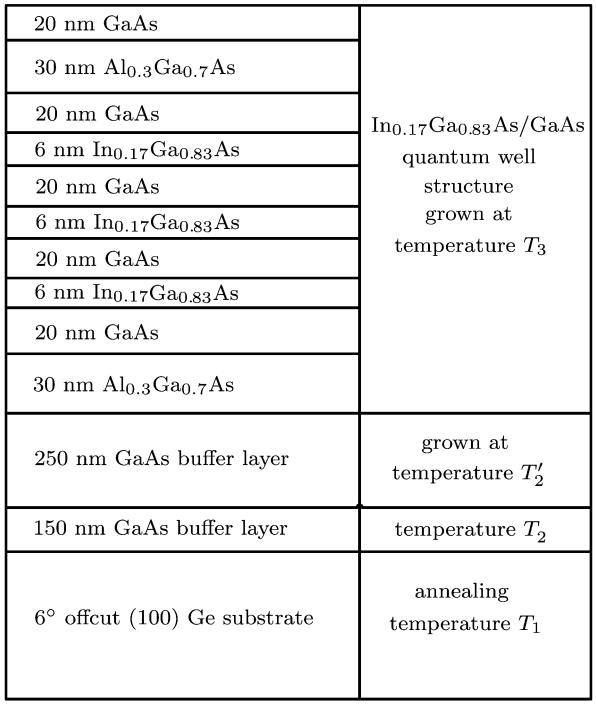


Fig. 1. Schematic diagram of a sample structure. Different temperatures for different epitaxial layers are illustrated on the right-hand side.

Another sample, with the same epitaxial structure, was grown on a GaAs (100) substrate for comparison. An InGaAs/GaAs QW laser structure sample was also grown under the optimised conditions. Table 1 gives the growth parameters of all the samples.

A Nanonavi Esweep S-II Atomic Force Microscope (AFM) was used to measure the morphologies of the samples in contact mode. A Philips Tecnai F30 Field Emission Gun Transmission Electron Microscope (FEG-TEM) was used to evaluate the microstructure and crystal quality of epitaxial layers. Photoluminescence (PL) spectra of the QW structure were measured at room temperature using a Nicolet FTIR760 Fourier transform spectrometer and detected by a liquid nitride cooled Ge detector. The excitation light source was the 632.8 nm line from a He–Ne Laser and the power was about 11 mW.

Table 1. Growth temperatures of different epitaxial layers in different samples. The absence of growth temperature T_3 for sample S20 means that no quantum wells are grown. The annealing temperature T_1 for sample S32 is absent as there is no need for high temperature annealing when the GaAs substrate is used. The highest T_1 is chosen as 735 °C because of the limitation of the substrate heater in our system.

sample number	substrate	annealing temperature T_1/°C	growth temperature T_2/°C	growth temperature T_2'/°C	growth temperature T_3/°C
S11	Ge	680	526	562	472
S12	Ge	698	526	562	472
S13	Ge	716	526	562	472
S14/S21	Ge	735	526	562	472
S22	Ge	735	544	544	472
S23	Ge	735	562	562	472
S24/S31	Ge	735	580	580	472
S25	Ge	735	600	600	472
S20	Ge	680	500	500	
S32	GaAs		580	580	472
S24L	Ge	735	580	580	480

3. Results and discussion

3.1. High temperature annealing

To avoid APD, several groups have suggested that offcut Ge (100) substrates with an appropriate offcut angle could lead to double-atom steps on the surface.[14,15,18] In our experiment, it is found that high temperature annealing (HTA) is indispensable for generating double-atom steps on an offcut Ge substrate surface. This is consistent with the results of Sieg *et al.*[18] Comparing the AFM images of the Ge substrate (Fig. 2(a)) and sample S24 (Fig. 2(b)), it can be seen that lines along a particular direction in Fig. 2(b) are much clearer and more uniform than those in Fig. 2(a). The origin of these lines contributes to uniform double-atom steps. This can be explained as follows. The chemical mechanical polishing (CMP) partly destroys the double-atom step structure of the offcut substrate so that the substrate surface contains not only double-atom steps but also single-atom steps. The coexistence of different atom steps weakens the step uniformity so that clear lines cannot be seen in Fig. 2(a). The uniform double-atom steps can originate only from HTA, because the epitaxy of GaAs and $In_{0.17}Ga_{0.83}As$/GaAs QW structure (strained, not relaxed) cannot generate new double-atom steps and the uniformity of double-atom steps depends merely on the surface atom structure before the process of epitaxy. The only experience of a Ge substrate before GaAs epitaxy is HTA as well as a two-minute exposure to As_4. However, the exposure to As_4 cannot generate new double-atom steps either. So, the only origin of uniform double-atom steps is HTA.

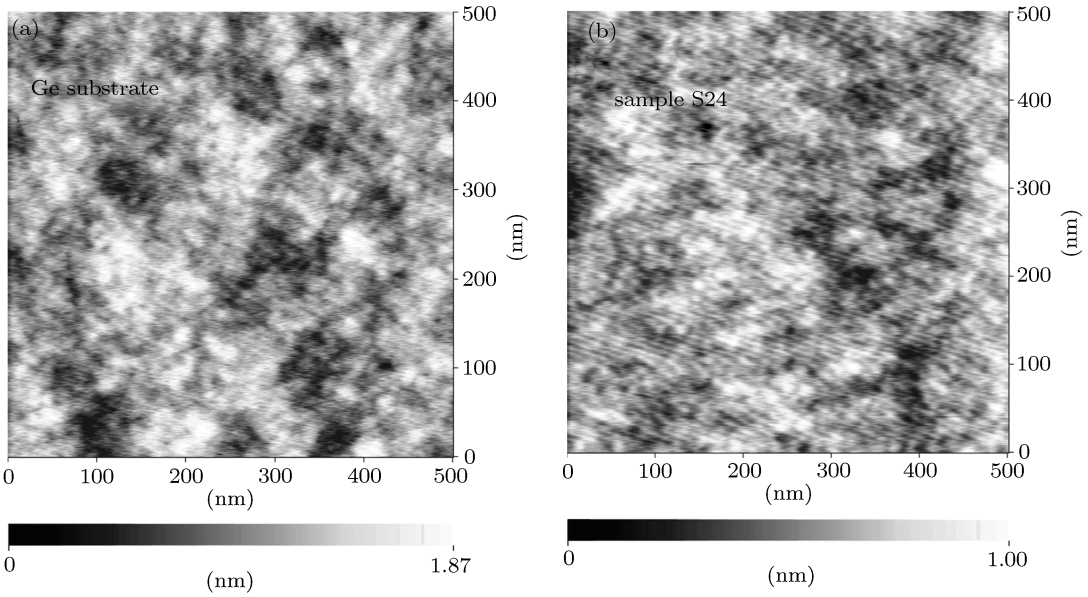


Fig. 2. AFM images (500 nm × 500 nm), where panel (a) is for the Ge substrate and panel (b) is for the sample S24.

From Fig. 3, it is found that the root mean square (RMS) roughness (10 μm×10 μm) decreases substantially from 2.47 nm to 0.73 nm when T_1 increases from 680 °C to 735 °C. The AFM image of sample S11 (Fig. 4(a), T_1 = 680 °C) shows several APDs, but these disappear when T_1 increases to 735 °C from Fig. 4(b), the AFM image of sample S14. This proves the necessity of HTA for uniform double-atom steps. The mechanism whereby HTA helps the offcut Ge substrate surface to generate uniform double-atom steps is believed to be related to the thermal active energy.

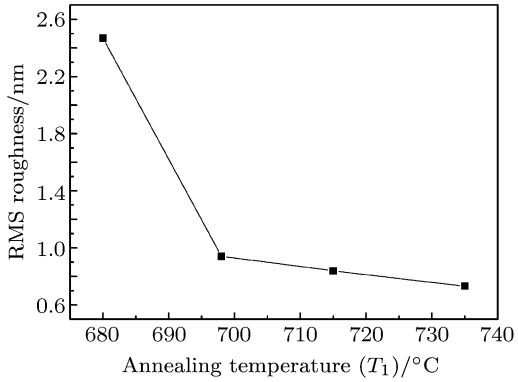


Fig. 3. RMS roughnesses (10 μm×10 μm) for samples S11, S12, S13 and S14 annealed at different annealing temperatures.

At higher temperature, the Ge atoms have higher thermal active energy to enable them to move from non-double-atom step sites to appropriate sites to form double-atom steps. This means that non-double-atom steps can be converted into double-atom steps in the HTA process.

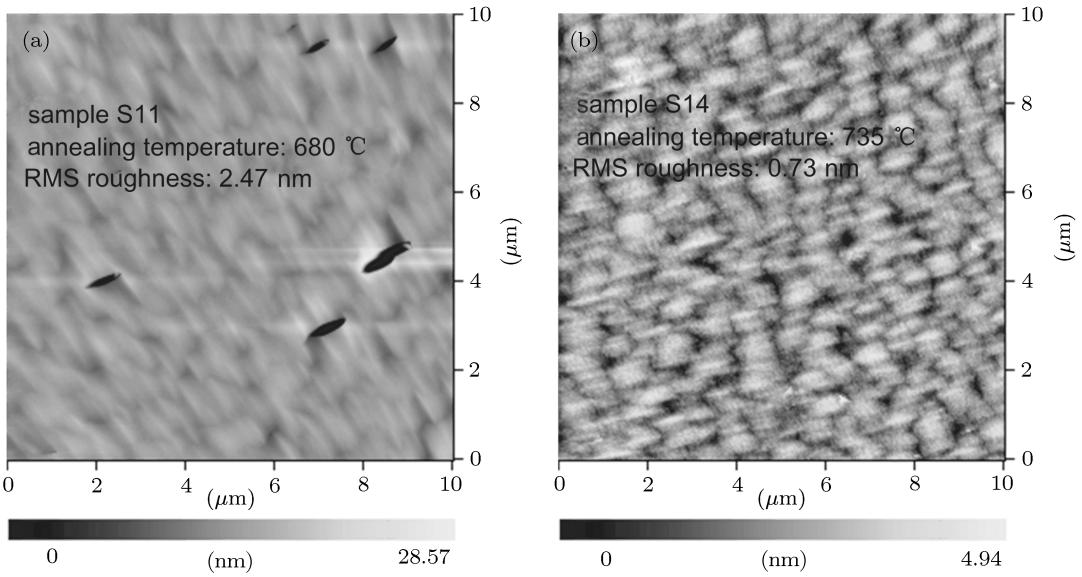


Fig. 4. AFM images (10 μm × 10 μm), where panel (a) is for sample S11 and panel (b) is for S14.

3.2. Exposing the Ge substrate to As_4

Exposing the Ge substrate to As_4 can form an As-terminated surface, which will lead to an ordinal deposition of Ga and As atoms. This step is important for avoiding APD.[21] Without this exposure the orders of Ga and As atoms will become worse on the Ge surface, thus increasing the possibility of APD formation.

3.3. 400 nm GaAs film growth

According to previous results, the annealing temperature was set as 735 °C during the subsequent experiments. Other series samples were grown in order to optimise the 400 nm GaAs buffer growth. Optimisation was focused on the growth temperature (T_2). In previous samples S11, S12, S13 and S14, T_2 was 526 °C for the first 150 nm and then changed to 562 °C (T_2') for the next 250 nm. In the following samples, i.e., S22, S23, S24 and S25, T_2 was kept constant for the whole 400 nm GaAs layer. Sample S14 is renamed here S21 for convenience of comparison.

From Fig. 5, on the one hand, it can be seen that RMS roughness (10 μm × 10 μm) decreases from 0.73 nm to 0.32 nm when T_2 is elevated from 526 °C to 600 °C, even smaller than that of the Ge substrate (0.34 nm). On the other hand, the PL intensity first reaches a peak at 580 °C (sample S24) and then becomes lower when T_2 further increases to 600 °C. Considering the negligible RMS roughness difference between sample S24 and S25, 580 °C is thought to be the most appropriate temperature for GaAs buffer growth. It should be noted that 580 °C is also the most commonly used growth temperature for MBE growth of GaAs on the GaAs substrate. High resolution transmission electron microscope (HRTEM) measurement of sample S24L also indicates a high quality GaAs/Ge interface and GaAs film. From Table 1, it can be seen that sample S24L was grown at the same

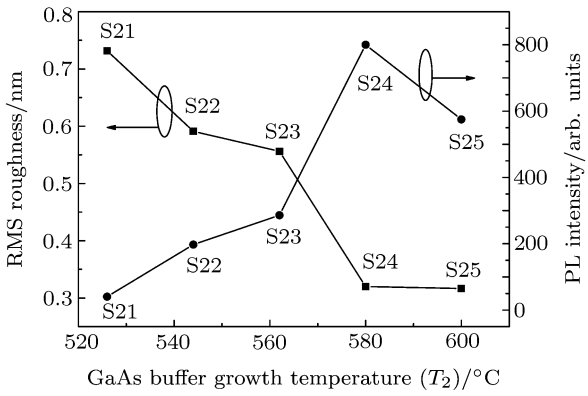


Fig. 5. RMS roughnesses (10 μm × 10 μm) and PL intensities for samples S21, S22, S23, S24 and S25 grown at different GaAs buffer temperatures.

temperatures T_1 and T_2 as sample S24, so these two samples should have the same qualities of GaAs/Ge interface and GaAs buffer layer. Figure 6(a) directly shows the flat GaAs/Ge interface, and a high crystal quality InGaAs/GaAs QW structure can be seen in Fig. 6(b).

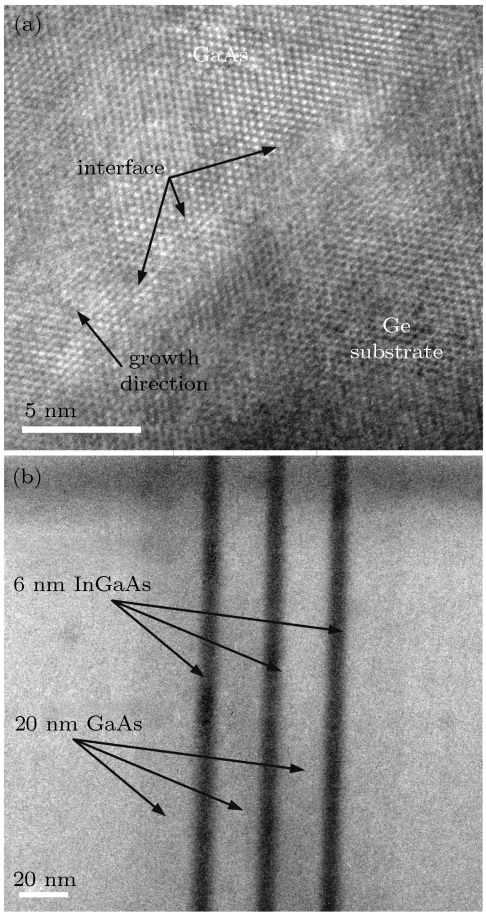


Fig. 6. HRTEM images of sample S24L, where panel (a) is for the GaAs/Ge interface and (b) is for the InGaAs/GaAs quantum well structure.

Another point should be noted, which is the difference between sample S21 and S23: the first 150 nm GaAs was grown at 526 °C for S21 but it was grown at 562 °C for S23. However, this difference in growth temperature results in an obvious distinction in RMS roughness and PL intensity. The PL intensity of sample S23 is six times higher than that of S21, and the RMS roughness of S23 is 0.56 nm, much smaller than that of S21, 0.73 nm. This directly proves that the GaAs/Ge interface and the initial epitaxial GaAs film near the interface both play dominant roles in the quality of the 400 nm GaAs layer.

If the growth temperature is too low, the Ga and As atoms cannot migrate for a sufficient time to find their appropriate sites before the deposition of subsequent atoms. The case of sample S20 confirms this point. Compared with sample S11, S20 has the same T_1 but lower T_2 of 500 °C. After growth, S20 has a gray surface, not a mirror-like surface. The gray surface indicates high density APDs. This is also confirmed by the reflection high energy electron diffraction (RHEED) pattern. 2×4 reconstruction could be seen for S11, but the RHEED pattern became spotted for S20 (RHEED pictures are not shown here). At higher temperature, the Ga and As atoms have longer migration times to find their appropriate sites to avoid APD formation and thus the epitaxial film has better quality. The trends of RMS roughness and PL intensity changing with the increase of T_2 from 526 °C to 580 °C indicate this point. However, when the temperature becomes too high, the intermixture and interdiffusion among Ga, As and Ge atoms becomes severe so that the film quality deteriorates. This leads to a decrease of PL intensity when T_2 is 600 °C.

3.4. Comparison between samples grown on Ge and GaAs substrates

Comparing the samples grown on different substrates in Fig. 7, it can be found that the PL intensity of $In_{0.17}Ga_{0.83}As$/GaAs QW structure of S32 (grown on the GaAs substrate) is about three times higher than that of S31 (grown on the Ge substrate, sample S24 renamed), which means a better optical quality of QW on the GaAs substrate. However, it should be noted that the growth temperature was fixed at 472 °C which was not optimised and the optical quality of $In_{0.17}Ga_{0.83}As$/GaAs QW structure could be improved further, because the optical quality strongly depends on QW growth conditions.[20,22,23] Another

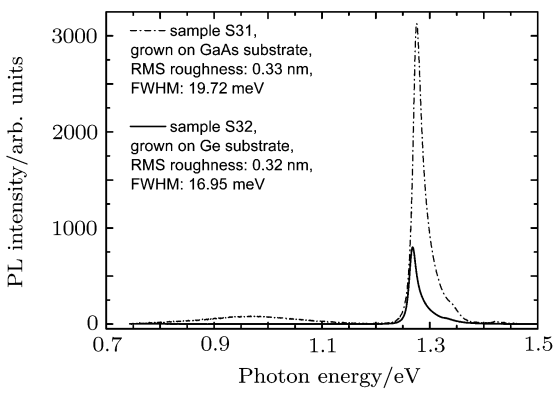


Fig. 7. Photoluminescence spectra of samples S31 and S32, where RMS roughness and FWHM data are also shown.

point that should be noted is that the RMS roughness and full width at half maximum (FWHM) of S31 are even a little better than those of S32. This indicates the good quality of GaAs buffer layers on a Ge substrate. It also provides good potential applications for III–V compound semiconductor optoelectronic devices on Ge substrates, such as solar cells, lasers and detectors.

4. Conclusion

Molecular beam epitaxy growth of GaAs on an offcut Ge (100) substrate has been systemically investigated. High temperature annealing before GaAs deposition is found to be a key element in avoiding APD because it is an indispensible step in generating uniform double-atom steps on the substrate surface. The quality of the GaAs film strongly depends on the GaAs/Ge interface and the beginning of GaAs deposition. Growth temperature plays an important role during GaAs film epitaxy. Low temperature growth can result in APD, but serious intermixing and interdiffusion will reduce the quality of the GaAs/Ge interface and GaAs film when growth temperature becomes too high. High quality GaAs/Ge interfaces and GaAs films have been achieved on Ge by optimising growth condition. High quality $In_{0.17}Ga_{0.83}As$/GaAs strained quantum wells are also grown on a Ge substrate. Samples show flat surface morphology and narrow photoluminescence line width compared with those of the same structure grown on a GaAs substrate. This provides huge application prospects for III–V compound semiconductor optoelectronic devices on Ge substrates.

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