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Regular Article

Influences of thickness and temperature of low temperature GaAs buffer layer on two-step MOVPE grown GaAs/Ge heterostructures

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Abstract. We investigate influence of GaAs buffer layer (BL) growth parameters such as temperature and thickness on the structural, morphological, crystalline and optical quality of metal organic vapor phase epitaxy (MOVPE) grown heterostructures of GaAs on Ge. It was found that the optimal BL conditions significantly decrease the effects of anti-phase boundaries (APBs) even when grown on offcut Ge substrate by two-step growth technique with AsH_3 pre-flow to promote double atomic step formation. It is observed that as the growth temperature increases, the growth rate of the GaAs BL increases, too. Improvement on the structural quality is observed up to BLs temperature of 535 °C, then it decreases. On the other hand, as the different thick BLs, 12, 25, 75 nm are considered, the epilayer grown on the 25 nm thick BL has shown the lowest full width at half maximum (FWHM) value, large photoluminescence peak intensity and internal quantum efficiency (IQE).

1 Introduction

Assembling of group III-V compounds and group IV elemental semiconductors, like GaAs-Ge, GaAs-Si, InP-Si etc, may form an impact in material features to be established on a single platform. This will permit devices to be used in upgraded functions and profit from various material systems to be developed [1]. The basic need for the integration of these material systems is high-quality growth of GaAs epilayer on Ge substrates. At this point, GaAs and Ge materials meet the major requirements to grow high-quality epitaxial films. Lattice mismatch (<0.1%) and coefficient of thermal expansion (<1.6%) between GaAs and Ge are very small [2,3]. However, the growth of polar GaAs epitaxial film on nonpolar Ge substrate forms several serious problems due to the formation of anti-phase domains (APDs). The reason is that when polar materials are grown epitaxially on nonpolar substrates, the position of cation and anion atoms in the two sublattices can be reversed from region to region in the grown epilayer, which results in the formation of APDs [4]. Domains of different sublattice positions are separated by anti-phase boundaries (APBs) consisting of wrong bonds of As–As or Ga–Ga [4]. APBs leads to decrease of photoluminescence intensity owing to creation of recombination centers [5,6], with high root mean square (rms) roughness of surface [7] and reduced crystal quality of GaAs epilayer [8]. Double atomic steps of Ge surface are needed to suppress or reduce the APBs [9]. Some different techniques have been introduced to decrease APBs on polar epilayers having double step surface on nonpolar substrates. The earliest one was to use {111} or {211} surfaces of Ge substrate, however (100) oriented Ge wafers were preferable due to their affordable price, considerable availability. This might open up a way for the integration of conventional III–V devices on Si (100) substrates that are used in CMOS [3].

Except for the different orientation of Ge substrates, there are some major approaches to obtain minimized APBs in GaAs epilayers. One of them is GaAs epitaxial layer growth on off-cut (2°–9°) Ge substrate. The influences of off-cut on the formation of APBs and dislocations were demonstrated by using 6° off-cut Ge substrate [10,11]. They have grown GaAs epilayers on Ge substrates with different off-cuts and demonstrated that GaAs growth on 6° off-cut Ge substrate resulted in high crystalline quality, smooth surface and reduced anti-phase domains. Although usage of off-cut Ge substrate is crucial for reducing APBs in GaAs epilayers, it is not enough to grow without APBs. The other approach is two-step GaAs growth on Ge with two different growth temperature regimes. Two-step growth method includes low-temperature 475–550 °C [12–14] and high-temperature 600–730 °C [13–15] growths of BL and following GaAs epitaxial layers. It is recognized that the low temperature grown GaAs layer in two-step growth method reduces the effect of Ge, Ga and As diffusions across the interface of epilayer and substrate. Additionally,

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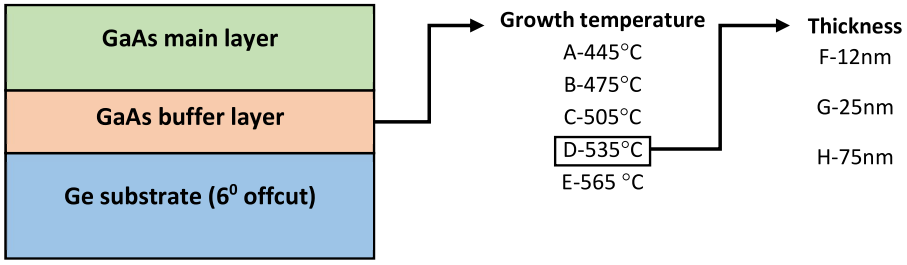


Fig. 1. Simplified schematic representation of GaAs epilayers on Ge substrate.

high-temperature growth of GaAs epitaxial layer reduces the carbon contamination [13,16,17]. The inter-diffusion of Ga, As, and Ge throughout the growth of high-temperature GaAs layer is source of a problem, which might result in unwanted n- and p-type doped areas [13]. Better device applications have been demonstrated on the two-step grown GaAs layer compared to that of homoepitaxial growth of GaAs [18]. In this study, both precautions, two-step growth method, and growth on an offcut Ge substrate have been considered. As it is introduced the buffer GaAs layer is very important for the subsequent main GaAs layer and the other epilayers which form the ultimate device structure. Non-adjusted conditions for growing of BL lead to increased number of APBs, high density of dislocations and rough surface morphology [18]. The growth temperature and the thickness of the GaAs BL are important growth parameters to obtain high-quality GaAs-Ge heterostructures. GaAs growth at very low growth temperature results in excess point defects of As which enhances the dislocation loops of nucleation [2,19,20]. These loops expanded during the growth of following high-temperature GaAs creates a many dislocations in thick GaAs epilayer. On the other hand, growth at very high temperature leads to the inter-diffusion problem and it reduces the efficiency of devices [21]. The growth of the GaAs BL at low temperature treats as a nucleation layer and it grows as an island-like structure. It has been demonstrated that GaAs can be grown pseudo-homoepitaxially on thickness optimized BL [22].

In this work, we investigate an extensive growth and characterization study of GaAs heterostructures which was grown on Ge substrate by two-step growth method via MOVPE on off-cut Ge substrates. The growth temperature and thickness effects of GaAs BL on following high-temperature GaAs epilayer were investigated. High-resolution X-ray diffraction (HRXRD), in-situ optical reflectance, room temperature-low temperature (10K) Photoluminescence (PL) and Raman spectroscopy measurements were used to examine the influences of growth parameters of MOVPE.

2 Experimental details

All GaAs epilayers were grown on a piece of 2 inch, 6° off-cut (001) Ge by means of horizontal flow MOVPE reactor of AIXTRON 200/4 RF-S. The reactor has in-situ reflectometer which provides knowledge about the growth rate of grown epilayer and optical quality of surface at the real growth time. High purity arsine (AsH_3) and opto-grade trimethylgallium (TMGa) were utilized as As and Ga sources, respectively. The Ge was desorbed thermally at 710 °C for 5 min right before the growth in order to obtain a single domain of GaAs epilayer and enhance double step formation [3,23]. Then, the temperature of the reactor was decreased to the growth of GaAs BL and five GaAs BL were grown at temperatures of 445, 475, 505, 535 and 565 °C. Then three different samples were grown with three different GaAs BL thickness 12, 25 and 75 nm. A sufficiently thick, low temperature GaAs layer was grown first to obtain the accurate thickness and growth rate of this layer by using LUXTRON commercial software. Finally, the growth temperature was increased to 650 °C under AsH_3 flow to prevent the surface from As deficiency and then 1 µm thick of main GaAs epilayer was grown on each BL. The reactor pressure was maintained at 50 mbar during growth of all samples. Figure 1 represents the simplified schematic of the grown GaAs samples.

HRXRD, PL, Raman spectroscopy, and in-situ optical reflectance measurements were performed on all the grown epilayers to investigate structural and optical properties of samples.

Raman measurements were performed with Witec Confocal Raman Microscopy system which has equipped with 514 nm wavelength. Measurements were conducted relatively low power level, under 5 mWatt, condition in order to prevent extra heating the surface. Low-temperature (10K) PL measurements were performed by using a 325 nm HeCd laser equipped with a 75 cm monochromator. The signals were detected by Si photodedector.

3 Results and discussion

3.1 Optimization of growth temperature for GaAs buffer layer

The epilayer surface was monitored during the MOVPE growth of all GaAs/Ge heterostructures were analyzed by means of optical reflectance during the growth as demonstrated in Figure 2. All growth stages can be tracked from the figure during the growth. Growth stages of GaAs can be separated to the following steps: high-temperature desorption (1), arsenization of Ge surface (2), GaAs BL growth at low-temperature (3), GaAs epilayer growth at high temperature (4) [24].

Figure 2 reveals the reflectance curves for varied growth temperatures of the GaAs BL. All reflectance curves are symmetric and showing a large amplitude of oscillations.

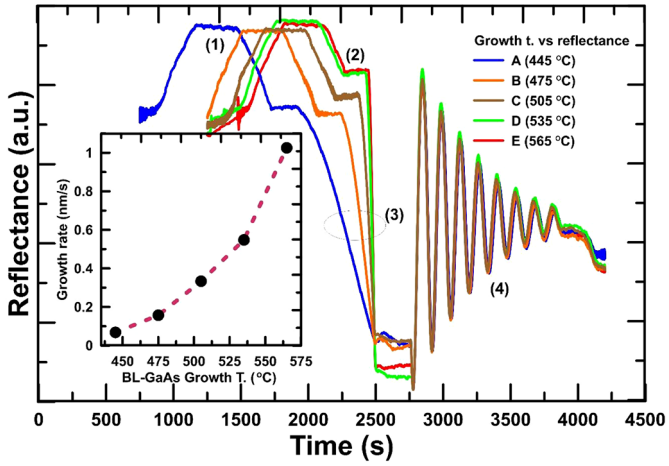


Fig. 2. GaAs in-situ optical reflectance. Epilayers were grown with increased BL temperatures. The inset shows the growth rate variation of GaAs bls with the increase of growth temperature.

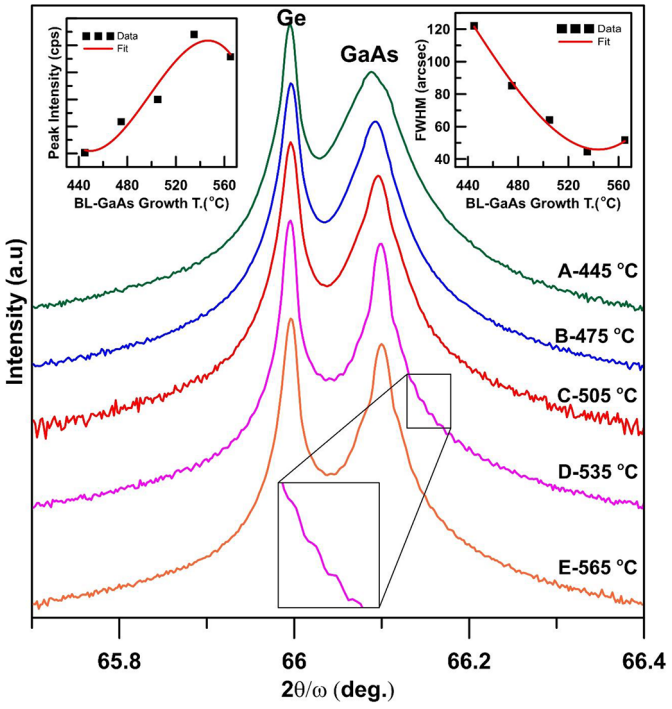


Fig. 3. $2\theta/\omega$ measurements of HRXRD for GaAs/Ge heterostructures which grown with different BL temperatures. The inset indicates the peak intensities (left) and FWHMs (right) of GaAs peaks.

Also, we have seen that there is no surface degradation of the GaAs epilayers, since the amplitudes of the reflectance oscillations remain similar for the samples grown at different temperatures. Although general behavior of in-situ reflectance for the GaAs/Ge heterostructures look similar, the amplitude of oscillations for sample D is larger than the others which prove the surface quality of the grown GaAs epilayer. The inset of Figure 2 shows the variation of growth rate for low-temperature GaAs BL with the increase of growth temperature. It indicates an increasing behavior from ∼0.1 nm/s to over 1 nm/s with the increase of growth temperature from 445 °C to 565 °C. This might show that growth is limited by the kinetics of cracking of growth species in the low-temperature range [25].

Figure 3 demonstrates $2\theta/\omega$ measurements of grown GaAs epilayers on Ge substrates for various growth temperature of GaAs BL. Right and left insets of Figure 3 indicate the change of FWHMs and peak intensities of GaAs diffraction peaks, respectively, for the increase of growth temperature of GaAs BL. The increasing growth temperature of GaAs BL leads to reduce the FWHM value (also increases peak intensity) down to 40 arcsec for 535 °C which in fact verifies the self-annihilation of APBs [26]. Then FWHM value starts to increase with further increase of growth temperature.

GaAs BL grows like nucleation layers on Ge substrate at lower growth temperatures which is similar to GaN (or AlN) on sapphire (silicon) substrate [13,27,28]. The reactor temperature is increased after the growth of the GaAs BL and this step promotes island grow of the grow GaAs BL. The optimized GaAs islands on Ge substrate prepare ideal templates for the epitaxial GaAs layers. It is clear from the measurements that the optimum growth temperature of GaAs BL is 535 °C owing to the observed lowest FWHM value. Furthermore, the HRXRD result of the sample D (grown at 535 °C) shows oscillations which are known Pendellösung fringes which points out the interface abruptness between GaAs and Ge and improved crystalline quality of GaAs epilayers as shown inset in Figure 3 [29].

GaAs epitaxial layers were analyzed by Raman Spectroscopy to obtain better information about the structural quality. Longitudinal Optical (LO) and Transverse Optical (TO) phonon modes of GaAs layers were observed at 288.8 cm^1 and, 265.7 cm^{-1} respectively, as revealed in Figure 4. These values seem to be 2–3 cm^{-1} lower compared to that of bulk GaAs [10,30]. The shift might be due to the strain state caused by Ge-GaAs lattice mismatch. On the other hand, no remarkable change in positions of TO and LO modes has been noticed among the GaAs epilayers grown on BLs with various growth temperatures. It might be concluded that no clear strain state change observable between these samples, as the Raman measurements are considered. It might be also possible that Raman spectroscopy is not able to characterize the interface between Ge-GaAs. This is due to the penetration depth of the 514 nm light, which is used to probe for Raman spectra, that is roughly 100 nm as the absorption coefficient of ∼10^4 cm^{-1} of GaAs is considered. However, as it can be understood from the figures that ratio of the integrated intensities of the TO to LO modes, I_{TO}/I_{LO}, varies. I_{TO}/I_{LO} is mostly used to figure out the structural quality of GaAs epi-layers. It is recognized that larger the ratio is, lower the epitaxial layer structural feature [31]. In this case TO mode is an unwanted mode which corresponds to the disordered structure in GaAs. As seen in Figure 4a, the I_{TO}/I_{LO} is the smallest, 0.056, for the sample grown at 535 °C, while it is 0.086 highest for the sample grown at 445 °C. There is also a difference between

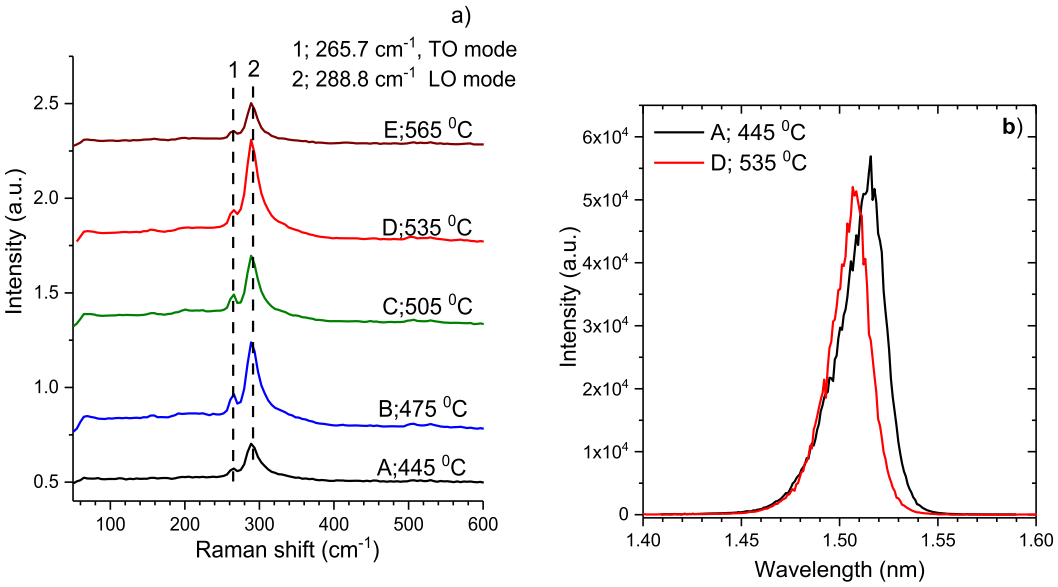


Fig. 4. (a) Raman; (b) PL results of the GaAs epilayers grown on various BL temperatures.

FWHM values of LO peak for these GaAs epitaxial layers. FWHM value changes between 19.4 and 21.9 cm^{-1} for the GaAs layer grown 535 °C and 445 °C, respectively. The Raman measurements conducted on the GaAs epilayers follows the result obtained from the XRD results.

Figure 4b demonstrates 10 K PL spectra for samples grown on Ge with various GaAs BL temperatures. For clarity, only the samples showing highest and the lowest structural quality has been included into the figure to show the variation better. The figure indicates near band edge emissions (NBE) of GaAs. It is easy to understand that there is change among the peak energies of epilayers. While the samples were grown at 535 °C has shown peak maximum at 1.508 eV, it is 1.516 eV for the sample grown at the lowest BL temperature indicating the shift about 8 meV. This change in the peak positions might be connected with the strain induced a shift. The analysis of the FWHM value of NBE PL emissions is one of the main indications of structural quality. It is 21 meV and 27 meV for the sample grown at 535 °C and 445 °C, respectively. Furthermore, internal quantum efficiency (IQE) value has been used for quality assessment parameter. IQE of the GaAs layers was calculated from the integrated intensity ratio of the band edge emissions at 10 K to 300 K. The values are 1.87% and 1.35% for sample D and A which were grown at 535 °C and 445 °C, respectively. The features of PL emission have noticed that the BL temperature is a significant growth parameter to govern optical and structural properties of GaAs epilayers on Ge substrates.

3.2 Optimization of thickness for GaAs bl thickness

The thickness of BL, which is another crucial parameter of growth to obtain pseudo- homoepitaxial growth, was altered from 12 nm to 75 nm with keeping all the other growth parameters fixed. Three (Sample F, G, and H) different GaAs BLs were grown with following thicknesses

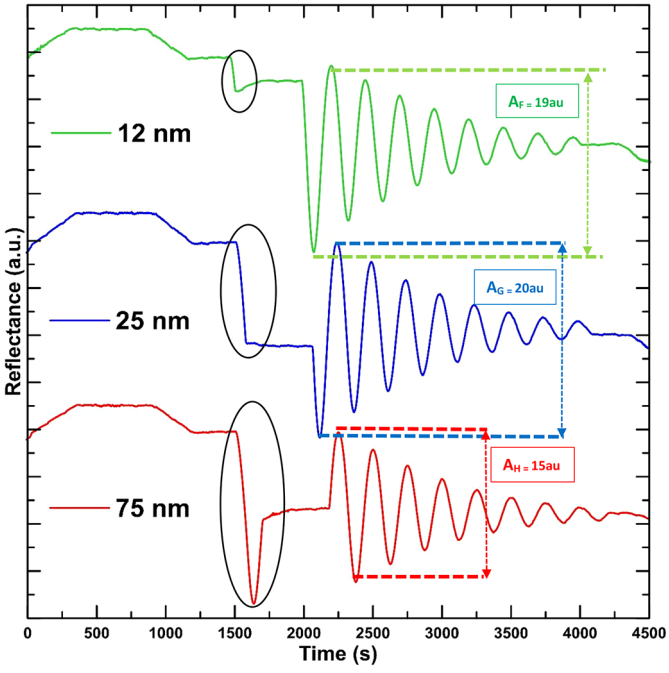


Fig. 5. GaAs in-situ optical reflectance. Epilayers were grown with various bl thicknesses. "A" represents the amplitude of the optical reflectance oscillations.

12, 25 and 75 nm on Ge substrate with an optimized growth temperature of 535 °C. GaAs main layer with 1 μm thickness was grown on every GaAs BL/Ge structure. Figure 5 demonstrates the optical reflectance of sample F (green), G (blue) and H (red). Ellipsoidal features indicate the thickness difference between samples. The in-situ reflectance behaviors of grown samples almost similar to the previous study. Even overall behavior of in-situ reflectance for the GaAs/Ge heterostructures look similar

during the growths with increasing thickness of the GaAs BL, the amplitude of oscillations for sample G ($A_G = 20$ au (arbitrary unit)) is larger than the others ($A_F = 19$ au and $A_H = 15$ au) which confirms the surface quality of the sample G is optically smoother than the others.

Similar to Figure 3, the HRXRD was performed on GaAs on Ge structures to analyze the crystal quality of GaAs epilayers. Figure 6 reveals $2\theta/\omega$ scans of GaAs/Ge structures for the increased thickness of the GaAs BL from 12 nm to 75 nm. The peak positions of the substrate and GaAs are almost the same as indicated in Figure 3. The increasing thickness of GaAs BL reduces the FWHM value (also increases peak intensity) down to 40 arcsec for 25 nm which in fact again confirms the self-annihilation of APBs [25]. The FWHM value changes its direction and begins to increase with the further increase of thickness. It can be figured out from the HRXRD scans that non-ideal BL thickness cannot promote subsequent GaAs to grow pseudo-homoepitaxial and lead to high-quality epilayers. Sample G has the most periodic and clearest oscillations among the samples, which point out the interface abruptness between GaAs and Ge and improved crystalline quality of GaAs epilayers [29].

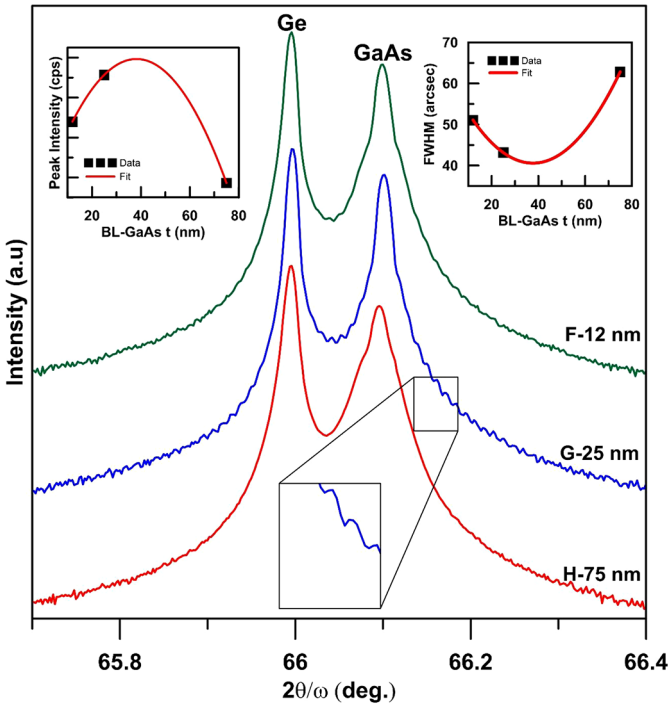


Fig. 6. $2\theta/\omega$ measurements of HRXRD for GaAs/Ge heterostructures which grown with different bl thicknesses. The inset indicates the peak intensities (left) and FWHMs (right) of GaAs peaks.

The I_{TO}/I_{LO} ratio also varies in this GaAs epitaxial layers grown on different BL thickness as seen in Figure 7a. No shift has been observed between the TO and LO modes similar to the different set of GaAs samples. The TO and LO values have been observed in 265.6 cm^{-1} and 289.1 cm^{-1}. Integrated intensity ratios are 0.080, 0.059, 0.094 for the samples F (12 nm), G (25 nm) and H (75 nm), respectively. It is important to note that the ratio is largest for the sample H, which is sample grown on the 75 nm thick BL. This confirms that the structural quality of the sample H is relatively poor compared to the samples grown with lower BL thicknesses. It is also clear that the smallest ratio has been observed for sample G. The FWHM values are also indicative for the structural qualities. It is 22.1 cm^{-1} for sample H, while it is 20.5 for both sample F and G. These results obtained from the Raman measurements show similar results claimed by the XRD data as demonstrated in Figure 6.

Figure 7b demonstrates the PL results required at 10 K. It is clear from the figure that sample H shows the lowest PL intensity. It is also clear from the figure that there is a shift between the PL emissions energies obtained for the epitaxial GaAs layers. The shift is about 3 meV which is not

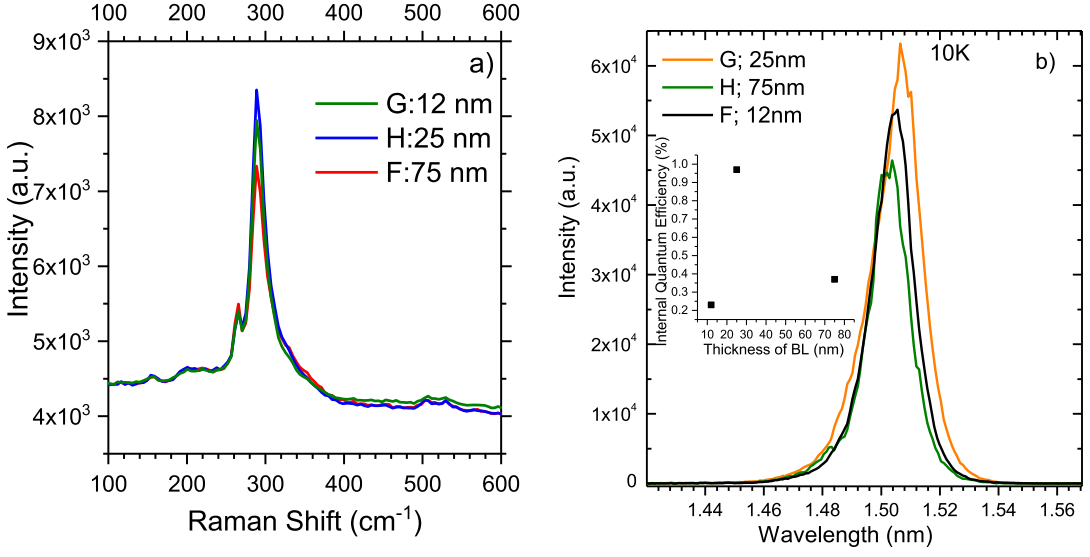


Fig. 7. (a) Raman; (b) 10 K PL results of the GaAs epilayers grown with various bl thicknesses.

very significant compared to that of GaAs epitaxial layer grown on different BLs temperatures. The inset shows the IQEs of the samples grown at different thick BLs. As indicated in the figure, for the sample grown on top of the 25 nm BL has indicated almost 3–4 times larger value compared to the other epilayers. This is also very similar indication obtained from the XRD results in which the FWHM and the peak intensity results are considered.

4 Conclusions

As a conclusion, we carried out the growth temperature and thickness influences of GaAs BL on crystalline, optical, structural and morphological quality of GaAs epilayers which were grown on Ge substrate heteroepitaxially. It is concluded that the thickness and growth temperature of the BLs are crucial for the epitaxial GaAs layer quality. APBs were reduced significantly by the optimal growth parameters. It was obtained that the growth temperature of 535 °C and 25 nm of layer thickness for the GaAs BL brought into high interface-crystalline quality with the lowest FWHM value and explicit-regular oscillations. Direct correlation between increasing crystalline-interface quality and a decrease in I_{TO}/I_{LO} ratio of GaAs epitaxial layers was observed. It was demonstrated that reduced APBs affected the optical quality of epitaxial GaAs layers regarding both increased PL peak intensities and IQE values.

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Author contribution statement

I.D. designed, planned and performed the growths and XRD measurements; A.E.K and E.G performed Raman measurements; H.F.B and E.G performed PL measurements; I.D and E.G wrote the manuscript; S.E supervised the study.

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