Investigation of interface intermixing and roughening in low-temperature-grown AlAs/GaAs multiple quantum wells during thermal annealing by chemical lattice imaging and x-ray diffraction

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The effect of thermal annealing on the interface quality in undoped, AlAs/GaAs multiple quantum well (MQW) structures grown at a low substrate temperature (310 °C) by molecular beam epitaxy has been investigated using chemical lattice imaging and high resolution x-ray diffraction. The low-temperature-grown MQW is of high crystalline quality comparable to the standard-temperature-grown MQW. However, significant interface roughening and intermixing occurs at the quantum well heterointerface when the structures are annealed beyond 700 °C. The effective activation energy for interdiffusion is estimated as 0.24 ± 0.07 eV. The structural properties observed here suggest that the excess arsenic associated with the low-temperature growth substantially enhances the diffusion of column III vacancies across an interface, which leads directly to intermixing of Al and Ga.

The low-temperature-grown (LTG) $Al_xGa_{1-x}As$–GaAs system has attracted much attention for ultrafast photodetector applications.[1–3] Recently, Lahiri *et al.* reported multiple quantum well (MQW) AlAs/GaAs *p-i-n* transmission modulators grown by molecular beam epitaxy (MBE) at low substrate temperatures (310 °C).[4] Sharp excitons with ultrafast lifetimes were demonstrated for the first time. Low-temperature growth results in excess arsenic incorporated into the AlAs/GaAs MQW.[5] During anneal, the excess arsenic collects into precipitates[6,7] that deplete free carriers from the surrounding material, rendering it high resistivity.[8] The ultrafast lifetimes in the LTG quantum wells are achieved by the arsenic precipitates, which serve as efficient recombination sites.[3] The sharp excitons in LTG quantum wells are made possible by narrow barriers of AlAs and high-quality well–barrier interfaces. These high-quality, well–barrier interfaces grown at low substrate temperatures challenge the general belief that high quality Al(Ga)As/GaAs MQWs require high substrate growth temperatures in the neighborhood of 600 °C. However, it was found that anneals of the LTG quantum wells at temperatures greater than 700 °C produced significant broadening to the exciton and shifts of exciton transition energy.[4,9]

A smooth and abrupt heterointerface is crucial to quantum well devices.[10] Interface intermixing results in a nonrectangular or rounded well, which causes a shift of exciton energy levels to higher energies. Interface roughness generates inhomogeneous broadening of the exciton due to an uncertainty of the well width. The heterointerface of $Al_xGa_{1-x}As$/GaAs heterostructures grown at standard substrate temperatures is one of the systems that has been extensively investigated structurally and optically.[10–14] It has been shown that growth interruptions,[12] dopants,[13] and anneal ambients[14] have effects on interface roughening or intermixing. In all cases, enhanced diffusion of column-III vacancies across an interface leads directly to intermixing of Al and Ga. Out-diffusion of gallium vacancies from LTG GaAs during annealing has been detected optically and electrically.[15–17] In this letter, we present the structural characterization of these undoped, LTG, AlAs/GaAs MQWs subjected to different anneal temperatures and compare them with identical structures grown at a standard temperature (600 °C). The focus is on the AlAs/GaAs heterointerface on an atomic scale. The investigation was performed by the (010) illumination in high resolution transmission electron microscopy (HRTEM) or chemical lattice imaging, and high resolution x-ray diffraction.

Two identical *p-i-n* structures were grown by MBE, with the *i* region containing an undoped 150-period multiple quantum well (MQW) of 10 nm GaAs wells and 3.5 nm AlAs barriers. All layers of the control structure were grown at a substrate temperature of 600 °C, which we will refer to as the standard-temperature-grown (STG) sample. The MQW in the second structure was grown at a substrate temperature of 310 °C, resulting in 0.2% excess arsenic in the MQW, which we will refer to as the LTG sample. The top *p*

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region of the LTG sample was grown at 450 °C, which acts as a weak *in situ* anneal of the LTG-MQW region, and results in the formation of arsenic precipitates in the MQW region as verified by TEM. Therefore, the only difference between the LTG and STG samples is that the LTG sample has excess arsenic on the MQW. Growth interruptions, which are sometimes used to provide smoother interfaces in an MQW, were not utilized in growing our MQWs.

Samples from each growth were subjected to rapid thermal anneal at temperatures ranging from 600 to 900 °C for 30 s to control the size and spacing of the arsenic precipitates using precipitate engineering.[7] Electro-optic characterization was performed on these samples and has been published elsewhere.[4,9] Structural characterization was conducted on four samples subjected to different growth and anneal conditions: the as-grown or *in situ* 450 °C annealed LTG, the 900 °C annealed LTG, the as-grown STG, and the 900 °C annealed STG samples. [010] cross sections of the samples were prepared by standard Ar milling, and then examined by transmission electron microscopy (TEM) using a Jeol 2000EX microscope operated at 200 kV. X-ray rocking curves (XRCs) were recorded using a high-resolution x-ray diffractometer with a four-reflection monochromator.[18]

The (010) illumination is superior to the (110) illumination in HRTEM of the GaAs/AlAs heterointerface.[11] Figures 1(a), 1(b), and 1(c) show [010] cross-sectional high-resolution TEM images of the 450 °C annealed LTG, the 900 °C annealed LTG, and the 900 °C annealed STG samples, respectively. The defocusing and the thickness of the areas for these images were chosen to yield the dominance of 200-type reflections from the AlAs side and 220-type reflections from the GaAs side. The GaAs layer appears in a finer structure pattern than the AlAs layer, which allows the evaluation of the interface. Interfaces appear to be less well-defined and rougher in the 900 °C annealed LTG sample compared to the 450 °C annealed LTG and the 900 °C annealed STG samples.

A transmission electron diffraction (TED) pattern can also be a sensitive measure of interface quality because it represents a Fourier transform of the crystal potential associated with the alternating AlAs and GaAs layers in the MQW.[19] The TED patterns of the samples all show characteristic satellite spots due to the superstructure of the MQW, but the number of satellite spots does vary for each sample. Satellite spots up to $n=13$, 8, 21, and 21 were seen in the *in situ* 450 °C annealed LTG, the 900 °C annealed LTG, the as-grown STG, and the 900 °C STG samples, respectively. This supports the HRTEM results that thermal annealing causes deterioration of the interface in the LTG but not in the STG samples.

Figures 2(a) and 2(b) show the (002) XRCs of the *in situ* 450 °C annealed LTG and the 900 °C annealed STG samples, respectively. Sharp and well-defined satellite peaks could be seen up to $n=-6$ in the *in situ* 450 °C annealed LTG sample. The presence of a large number of satellite peaks indicates a well-defined periodic structure. However, after a 900 °C anneal for 30 s, all the satellite peaks in the LTG sample had their intensities reduced by 50–60% and

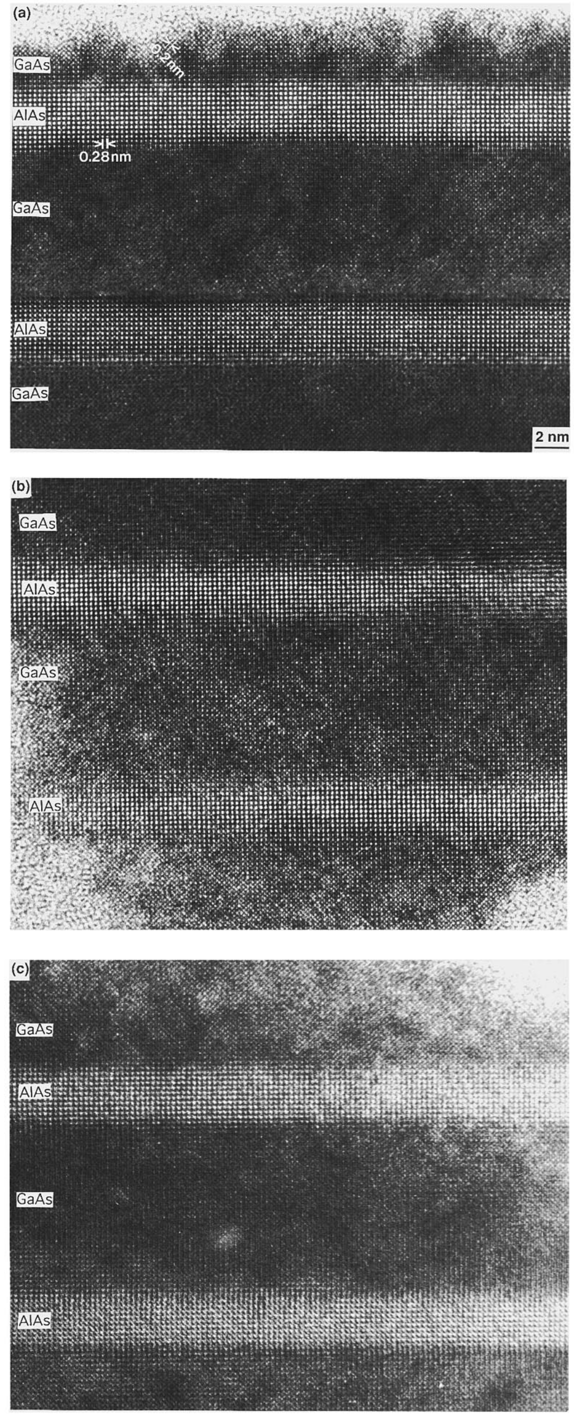

FIG. 1. Chemical lattice images of (a) *in situ* 450 °C/24 min annealed LTG, (b) 900 °C/30 s annealed LTG, and (c) 900 °C/30 s annealed STG 3.5 nm AlAs/10 nm GaAs multiple quantum wells. The AlAs layer appears in a brighter contrast than the GaAs layer. The interface in (b) is deteriorated.

linewidths broadened by 15–20%. As a result, satellites of higher orders, e.g., $n=-5$ and -6, are undetectable in Fig. 2(b). The decrease of intensities implies that the interface is not as abrupt as before, probably caused by intermixing.[20] The broadening of linewidths suggests a random variation of the MQW-period length, which was possibly caused by interface roughening.[20] On the other hand, the XRCs of the STG samples do not show noticeable differences after annealing. The shape of the x-ray satellite envelope and the

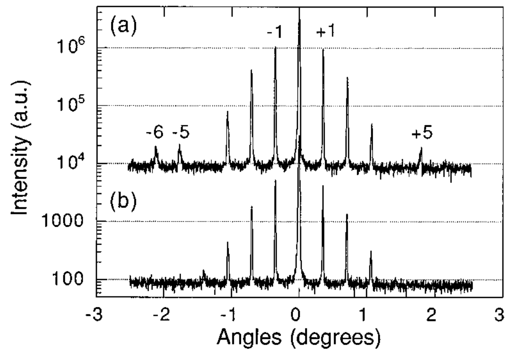

FIG. 2. (002) x-ray rocking curves of (a) *in situ* 450 °C/24 min annealed, and (b) 900 °C/30 s annealed low-temperature-grown AlAs/GaAs multiple-quantum well structures. The satellite peaks, $n=-5$ and -6, are undetectable in (b).

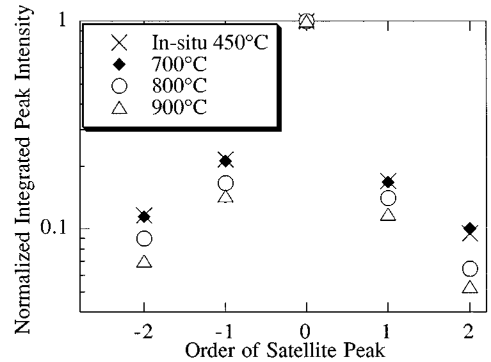

FIG. 3. Normalized integrated satellite peak intensities vs order of satellites ($n=-2$, -1, $+1$, and $+2$) for the LTG samples subjected to different anneals. The LTG MQW is stable up to 700 °C.

maximum position of each satellite are very similar in the *in situ* 450 °C annealed LTG and STG samples. The linewidths of the satellites do appear narrower in the STG sample. For example, the linewidths of the $n=1$ peak are 40 and 27 arcsec for the LTG and STG samples, respectively. All these observations suggest that interdiffusion occurs in the LTG sample but not in the STG sample after a 900 °C anneal, and that the as-grown LTG AlAs/GaAs MQW is of high crystalline quality comparable to the STG MQW.

Quantitative analysis of the x-ray data for several isochronal anneals was conducted to determine the temperature for which the interfacial mixing initiates and the activation energy for the diminution of satellite peak intensities. The satellite peak intensities were numerically integrated, and normalized to the integrated central peak intensity with a superlattice Debye–Waller factor. As shown in Fig. 3, the LTG MQW is stable up to 700 °C, above which the integrated intensities for $n=-2$, -1, $+1$, and $+2$ satellites decrease as anneal temperatures increase. The activation energy, 0.24 ± 0.07 eV, was extracted from the plot of the integrated peak intensities versus 1/anneal temperatures for several satellites. This value is in agreement with our optical analysis.[9]

In conclusion, we report on the structural characterization of undoped, LTG, and STG AlAs/GaAs MQWs annealed at different temperatures. Chemical lattice imaging, electron diffraction, and x-ray diffraction experiments consistently show that a 900 °C anneal for 30 s results in significant interface intermixing and roughening in the LTG MQW but not in the STG MQW. Quantitative analysis of x-ray data for several anneals further indicates that at temperatures above 700 °C, the interdiffusion becomes important in the LTG MQW and the effective activation energy is estimated as 0.24 ± 0.07 eV. Since the only difference between these two growths is the excess arsenic associated with the low-temperature growth, it is reasonable to deduce that the excess arsenic is responsible for enhanced diffusion of column-III vacancies across an interface, which leads directly to intermixing of Al and Ga.

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