Micro-photoluminescence characterization of local electronic states in a (110) GaAs quantum well fabricated by cleaved-edge overgrowth

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Abstract

Local electronic states due to characteristic surface islands and pits formed on an atomically flat (110) GaAs interface of a quantum well fabricated by a cleaved-edge-overgrowth method with high-temperature growth-interrupt annealing are characterized by micro-photoluminescence (PL) imaging and spectroscopy. With overall observation of the 6-nm-thick (110) quantum well with the area of 6.8 µm width and 3 mm length, we found local PL signals due to isolated 2- or 3-monolayer (ML) islands, isolated 1-ML pits, 1-ML islands and pits formed along cleavage atomic-step lines, and gradual change of the states across the full range of the sample.

PACS numbers: 79.60.Jv, 78.67.De, 78.55.-m, 68.37.-d, 68.37.Ps

Keywords: local electronic states, atomic steps, atomically flat (110) GaAs interface, cleaved-edge-overgrowth, growth-interrupt annealing,
A cleaved-edge-overgrowth method, in which two molecular-beam-epitaxy (MBE) growth steps are separated by an *in situ* wafer cleavage process [1], has become of increasing importance for fabricating novel semiconductor nanostructures. This method has been used to fabricate a host of low-dimensional quantum structures including precisely spaced quantum dots [2], modulation-doped quantum wires showing nearly ideal one-dimensional (1D) quantum transport characteristics [3], and T-shaped quantum wires formed at the right angle T-shaped intersection of two quantum wells [4].

The overgrowth on a cleaved (110) surface requires a low substrate temperature range (470 - 510°C) and a very high As₄-vapor overpressure (V/III beam-equivalent-pressure ratio of about 70) for GaAs [1], and results in rough growth [5]. However, it is found that a growth-interrupt annealing at 600°C for 10 min turns a rough (110) GaAs surface to an atomically flat GaAs surface [6]. This annealing method has enabled us to realize quantum wires and wells with unprecedentedly high quality to study various 1D and 2D optical properties [7, 8].

In our previous work [8], effects of deviation from integral-monolayer (ML) deposition on the annealed surface were studied by atomic force microscope (AFM) and photoluminescence (PL). When we supply Ga flux either slightly in excess or insufficient to grow an integral number of MLs, the growth-interrupt annealing makes the upper GaAs surface atomically flat except for formation of µm-lateral-scale islands or pits with characteristic stable shapes. The islands are shaped like *boats*, 2- or 3-ML high, and typically smaller than 1 µm in lateral
size. The pits are shaped like tropical fish, 1-ML deep, and typically as small as a few µm in lateral size. When quantum wells or wires are formed by the cleaved-edge-overgrowth method with growth-interrupt annealing, the islands and the pits generate local electronic states with low and high energies, respectively, and make bright PL spots and dark PL regions in PL images.

Further experiments [9] analyzing the shapes of each island and pit, and first principles calculations [10] on adatom-migration energies on a (110) GaAs surface reveal basic aspects of the formation of atomically flat surfaces, islands, and pits. From application point of view, furthermore, we need to characterize wide sample regions to survey all local electronic states in samples, because device performance is determined by all electronic states in a whole device.

In this paper, we investigate local electronic states in a (110) GaAs quantum well fabricated by the cleaved-edge-overgrowth method with growth-interrupt annealing. We measured wide sample regions over a few mm by means of micro-PL imaging and scanning micro-PL spectroscopy. We monitored PL images corresponding to continuous evolution from the islands to the pits, and found high and low energy regions along atomic steps most probably formed in the in-situ cleavage. Connected structures of the islands, the pits, and the atomic steps were also found. We demonstrate that all these structures contribute to the corresponding PL spectra, and as a whole make an overall spectrum as a device.
A 6-nm-thick GaAs quantum-well sample [8] was fabricated by the cleaved-edge-overgrowth method with growth-interrupt annealing. In the first growth, we successively grew, by conventional MBE on a (001) GaAs substrate, a 90 nm (GaAs)$_4$/(AlAs)$_4$ superlattice layer, a 6.8 $\mu$m Al$_{0.29}$Ga$_{0.71}$As barrier layer, a 90 nm (GaAs)$_4$/(AlAs)$_4$ superlattice layer, and a 4.5 $\mu$m GaAs cap layer. For the second MBE growth, we cleaved the (001) GaAs substrate at the growth position of our MBE chamber. On the cleaved (110) surface, we overgrew a 6-nm-thick (30-MLs) GaAs layer under a substrate temperature of 490$^\circ$C and a growth rate of 0.43 $\mu$m/h. Right after the growth, we shuttered Ga flux, raised the substrate temperature to 600$^\circ$C, and annealed the sample for 10 minutes under an As$_4$ flux in the MBE chamber [6]. We then grew a 10 nm Al$_{0.33}$Ga$_{0.67}$As barrier layer and a 10 nm GaAs cap layer, after we lowered the substrate temperature back to 490$^\circ$C. During the growth of the 6-nm-thick quantum-well layer, the substrate was not rotated but aligned along the Ga flux gradient, which introduced spatial distribution of GaAs layer thickness around the nominal thickness by Ga flux gradient of 1 %/mm with respect to the local positions on a sample. Because the quantum well is formed on the cleaved edge of a 6.8 $\mu$m Al$_{0.29}$Ga$_{0.71}$As barrier layer, the quantum well is 6.8 $\mu$m wide in the [001] direction and 3 mm long in the [110] direction with the layer-thickness gradient. Schematic diagram of the quantum well structure is shown in Fig. 1.

Measurements of micro-PL images and scanning micro-PL spectra [11] were performed on
the (110) quantum well sample at 4 K. In the imaging measurements, excitation light from a Ti:Sapphire laser with 730-nm wavelength irradiated the quantum well uniformly. Spatial resolution of the measurements was 0.8 \( \mu \)m with a 0.5 numerical-aperture objective lens. We measured 157 regions \((33 \mu \text{m} \times 26 \mu \text{m})\) to cover entire sample of 3 mm length. We also measured local PL spectra under 0.8-\( \mu \)m-diameter point excitation scanned by 1 \( \mu \)m steps along [T10] direction, where excitation intensity was 170 nW into 0.8-\( \mu \)m-diameter spots, or 30 W/cm\(^2\). In the spectrum measurement, excitation is made into a 0.8-\( \mu \)m-diameter spot, but PL detection covers a wider region over 10 \( \mu \)m. Therefore, PL comes from positions where carriers have finally reached after diffusion.

Figures 2 and 3 show PL images of various regions of 100 \( \mu \)m \( \times \) 6.8 \( \mu \)m areas in the (110) GaAs quantum well, where each region is denoted by longitudinal position measured from one end of the sample. With increasing position indices, averaged thickness of the quantum well increases by 0.3 ML/mm due to the Ga flux gradient in the overgrowth.

PL images in 0 - 500 \( \mu \)m regions shown in Fig. 2 consist of bright or dark regions and uniform background regions. The uniform background regions, which occupy major areas in Fig. 2, correspond to the quantum-well regions with atomically flat interfaces [8]. The bright and dark regions correspond to lower- and higher-energy regions than surrounding regions, respectively, and hence, to ML islands and ML pits formed on the quantum-well surface [8]. Since all the PL images were taken under uniform excitation, PL inhomogeneity
is mostly caused by carrier migration from higher- to lower-energy regions. Image resolution is limited by both carrier-diffusion length [12] and the 0.8 µm resolution of our microscope.

Most of the dark or bright regions in 0 - 500 µm regions are aligned along lines. These lines reflect most probably atomic-step lines generated in an in-situ cleavage process. Slightly-tilted imperfect cleavage causes "hackling", that is the formation of single- or multiple-atomic-step lines, where each region separated by the lines has an atomically flat (110) surface. Such repeated atomic steps cause additional thickness variations in the quantum well after the overgrowth and growth-interrupt annealing. While the lines in 0 - 200 µm region are mostly dark, the lines in 400 - 500 µm region are mostly bright. A bright line at 510 µm position is connected and uniform, while the lines in 400 - 500 µm region have array of bright spots.

Our interpretation is that the 0 - 200 µm region had overgrowth of GaAs slightly less than integer MLs, and the surface flattening by growth-interrupt annealing generated thinner region along the atomic steps. On the contrary, in the 400 - 500 µm region, overgrowth of GaAs was slightly more than integer MLs, and thicker regions were formed along the steps. Apparently, the thicker regions can form either arrays of isolated islands or connected lines along the steps.

In 300 - 400 µm region, which had overgrowth of integer MLs, both bright and dark PL occur only along the cleavage lines, and wide uniform PL regions are found between the lines.
This demonstrates that perfect cleave without atomic steps due to hackling and overgrowth of integer MLs are both necessary to make devices with ideal atomically flat interfaces by the cleaved-edge-overgrowth method.

The 510 - 600 µm region has no atomic-step line, but random distribution of bright PL spots. Observed sizes of the bright spots are limited by our microscope resolution of 0.8 µm. As shown in our previous work [8], the bright PL spots are from local low-energy states caused by the islands formed on the quantum-well surface with excess supply of GaAs from integer MLs. We do not call these states as quantum dots, because the lateral size of the islands measured by AFM is mostly 0.1 - 1 µm, and still too large for quantum confinement.

Figure 3 demonstrates evolution from slightly excess coverage than integer \( n \) MLs to slightly insufficient coverage than the next \( n+1 \) integer MLs.

The PL image of the 700 - 800 µm region in Fig. 3 is similar to that of the 520 - 600 µm region in Fig. 2, and indicates the island formation due to slightly excess coverage than integer \( n \) MLs. A bright PL line at the 720 µm position is similar to that at the 510 µm position. In the 1250 - 1350 µm and 1600 - 1700 µm regions, the isolated bright spots become fewer, while connected dark and bright areas increase. In the 1900 - 2000 and 2300 - 2400 µm regions, dark areas are surrounded by bright areas and become isolated, indicating the pit formation due to slightly less coverage than \( n+1 \) integer MLs. The image of the 2700 - 2800 µm region is similar to that of the 200 - 300 µm region. The observed evolution is
consistent with our previous results measured by AFM and PL images [8]. It is additionally found that a few number of bright spots remain in the region in 1600 - 2400 µm positions, and that atomic steps formed in the cleavage process modify formation of islands and pits.

Figures 4 and 5 show spatially resolved micro-PL spectra scanned by 1 µm steps along the center of the 6.8 µm-wide area of the quantum-well sample. The longitudinal positions are indicated by the unit of µm, similarly to Figs. 2 and 3. At the bottoms of PL spectra covering 100 µm regions, averaged PL spectra are also shown.

Peaks appear at discrete energies, as marked by vertical dashed lines, which indicates that the PL peaks originate from regions with discrete thickness of integer \((n, n+1, n+2, \text{ and } n+3)\) MLs. Small PL peaks with fluctuating intensity reflect islands or pits formed on the quantum well surface, and the peak positions indicate their height or depth. The PL peaks for \(n, n+1, n+2, \text{ and } n+3\) MLs are separated by about 3.7 meV, which is consistent with thickness difference by 1 ML. Their energy positions are typically at 1.591, 1.587 eV, 1.583 eV, and 1.579 eV, though they fluctuate by ±1 meV with positions in several hundred µm. The reason for the fluctuations is currently not known, but might be due to strain induced by glue or scratches on the back of the sample.

Figure 4 shows PL spectra in the 200 - 500 µm regions. Note that PL peaks of \(n\) MLs dominate PL spectra in these regions, proving that the continuous PL signal from flat regions in Fig. 2 corresponds to the \(n\)-ML peak. On the other hand, small PL peaks of \(n+1\) MLs
are found at positions in 290 - 300 µm, 300 - 320 µm, 360 - 380 µm, and 420 - 490 µm, which correspond to the positions of local bright PL spots along the atomic-step lines in Fig. 2. This shows that islands formed along the lines are 1-ML higher than surrounding regions.

In the PL spectra of Fig. 4, there was no feature (such as appearance of other peaks or drop of n-ML peak intensity) corresponding to the dark regions in Fig. 2. This is because carriers excited into those regions migrate to the n-ML regions and emit PL there. This proves that the dark regions in Fig. 2 are caused by carrier migration instead of non-radiative decay.

In the 500 - 600 µm region shown in Fig. 5, spectra are still dominated by n-ML peak, which indicates that the major region of the quantum well has thickness of n MLs. Local PL peaks of n+1 MLs are found only around 510 µm. In the 520 - 600 µm region, local PL peaks of n+2 and n+3 MLs are found instead of those of n+1 MLs. Comparison with the PL image in Fig. 2 shows that the n+1-ML peak corresponds to a bright line in the image, while the n+2- and n+3-ML peaks correspond to isolated bright spots. This proves that the islands formed along the cleavage line are 1-ML high, while the isolated islands formed on the flat region are 2- or 3-ML high. The latter is consistent with our previous results of AFM that the isolated islands are 2- or 3-ML high [8].

In the 1250 - 1350 µm region, where a mean thickness of n+0.3 ML is expected, PL peaks are found at all the energy positions of n, n+1, n+2, and n+3 MLs in Fig. 5. Note that the
PL image in Fig. 3 has horizontal stripes and bright spots. Though detailed correspondence between the PL image in Fig. 3 and the spectra in Fig. 5 is difficult, our previous results of AFM [8] suggest that the dark and bright horizontal stripes are due to laterally elongated terraces with 1-ML steps, and that bright spots are due to isolated islands with 2- or 3-ML steps.

In the 2300 - 2400 µm region, spectra are dominated by \( n+1 \)-ML peak, which indicates that the major region of the quantum well has now thickness of \( n+1 \) MLs. Local PL shows peaks of \( n \) MLs, which means that the local PL is from pits with 1-ML depth. The \( n+1 \)- and \( n \)-ML peaks in Fig. 5 correspond to PL from continuous flat region and PL from the dark isolated triangular regions, respectively. This result is consistent with our previous AFM results [8] that isolated pits with such triangular shapes are 1-ML deep.

At the bottoms of respective panels in Figs. 4 and 5, averaged PL spectra are shown. Note that the averaged PL spectra reflect all the characteristic features measured in scanning PL spectra and PL images. Hence, we are now able to infer characteristic local electronic states formed in cleaved-edge-overgrowth quantum wells and wires, on the basis of averaged PL spectra obtained for overall samples or devices.

In this work, (110) quantum wells were formed by the cleaved-edge-overgrowth method, where the quantum wells are grown on the cleaved exact (110) surfaces. Such an exact crystal surface is hardly achieved on polished wafers. However, we also studied quantum
wells on the surfaces resulted from imperfect cleavage, which have atomic-step lines. Growth on such surfaces is very similar to that on the polished wafers. Therefore, results of this work can be extended to investigation of MBE growth and growth-interrupt annealing on (110) surfaces in general.

In summary, the whole region of a GaAs quantum well fabricated by the cleaved-edge-overgrowth method with growth-interrupt annealing was characterized by micro-PL images and scanning micro-PL spectra. We observed local PL signals due to isolated 2- or 3-ML islands, isolated 1-ML pits, 1-ML islands and pits formed along cleavage atomic-step lines, and evolution from islands in $n$-ML background to pits in $n+1$-ML background. These local PL features characteristic to structures formed by the cleaved-edge-overgrowth method with growth-interrupt annealing are reflected in averaged PL spectra over 100 $\mu$m regions. These results are useful to understand inhomogeneity in future nanostructure devices fabricated by (110) MBE.

This research is partly supported by Ministry of Education, Culture, Sports, Science and Technology, Japan, and by Japan Society for the Promotion of Science.


Figure Captions

Fig. 1: A schematic cross-sectional image of a quantum well structure fabricated by the cleaved-edge-overgrowth method and growth-interrupt annealing.

Fig. 2: PL images at 4 K of a (110) GaAs quantum well fabricated by the cleaved-edge-overgrowth method and growth-interrupt annealing. Each image show a 100 µm × 6.8 µm area of the quantum well in 0 - 600 µm longitudinal positions. Deposition thickness of the quantum well increases approximately by 0.3 ML/mm with positions. The 300 - 400 µm region is expected to have an integer ML thickness.

Fig. 3: PL images at 4 K of a (110) GaAs quantum well fabricated by the cleaved-edge-overgrowth method and growth-interrupt annealing. Each image show a 100 µm × 6.8 µm area of the quantum well in 700 - 2800 µm longitudinal positions. Deposition thickness of the quantum well increases approximately by 0.3 ML/mm with positions.

Fig. 4: Scanning micro-PL spectra measured by 1 µm steps over 100 µm in 200 - 500 µm regions at 4 K are displayed in a row. The bottom spectra show averaged spectra in the regions.

Fig. 5: Scanning micro-PL spectra measured by 1 µm steps for 500 - 600 µm, 1250 - 1350 µm, and 2300 - 2400 µm regions at 4 K are displayed in a row. The bottom spectra show averaged spectra in the regions.
Cross-sectional image

10 nm GaAs cap layer
10 nm AlGaAs barrier
0.33 0.67
6 nm GaAs QW
Cleaved surface
Annealed surface
[110]
[001]
[110]

Fig. 1 Oh et. al, for JAP
Fig.2 Oh et.al JAP
Fig. 3 Oh et al. JAP
Fig. 4 Oh et al JAP
Fig. 5 Oh et al. JAP