Large terrace formation and modulated electronic states in (110) GaAs quantum wells

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Interface roughness and local electronic states formed in GaAs quantum wells (QWs) grown on (110) cleaved surfaces were characterized by high-resolution microscopic photoluminescence (micro-PL) imaging and spectroscopy assisted by a solid immersion lens. From macroscopic PL spectra and micro-PL images, it was found that a well width fluctuation of about 0.6 to 0.7 nm, which corresponds to 3 to 3.5 monolayers (ML), with laterally sub-μm- to μm-scale ML terraces existed in the (110) GaAs/AlAs QWs, and this was also supported by the results of atomic force microscopy measurements performed on the (110) GaAs surface. In high-resolution solid-immersion micro-PL spectroscopy, sharp PL peaks were observed at low excitation power and their temperature and excitation power dependence showed a localized quantum-dot-like (QD-like) nature in the (110) GaAs QWs. The spatial and spectral distribution of these sharp PL peaks indicated that the localized QD-like states were formed by short-scale roughness in the bottom GaAs-on-AlAs interface of the (110) GaAs/AlAs QWs on each μm-scale large ML terrace in the top AlAs-on-GaAs interface.

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I. INTRODUCTION

The molecular beam epitaxy (MBE) growth of GaAs-related materials on non-(001) surfaces has increasing importance in fabrication of novel quantum nanostructures such as quantum wells (QWs), wires (QWRs), and dots (QDs). The growth on the (110) cleaved surface, in particular, has been a key process in the cleaved-edge overgrowth (CEO) method, a two-step-growth method combined with in situ cleavage, by which T-shaped QWRs (T-QWRs) and atomically precise QDs have been produced.

On the other hand, the growth of high-quality GaAs layers on the (110) surface is still difficult compared with that on the (001) surface, first because the acceptable growth conditions deviate from those on the (001) surface and are very narrow, and also because the growth mechanism is not fully understood. For instance, reflection high-energy electron diffraction (RHEED), scanning tunneling microscope, and atomic force microscope (AFM) measurements on (110) MBE growth have shown that there exist growth-condition-sensitive monolayer and bilayer growth modes simultaneously with step growth and triangular-island formation, which modify RHEED oscillation patterns, and that a topographically smooth surface was formed under As-rich conditions where RHEED intensity oscillations were not sustained. Furthermore, in spite of much effort to reduce photoluminescence (PL) linewidth of (110) GaAs QWs by optimizing the growth condition, it has been always broader than that of well-controlled (001) QWs for unknown reasons. In order to clarify the reasons for the broad PL linewidth and improve the quality of the (110) GaAs QW, studies on the local electronic states and interface properties in the (110) QW are quite important. However, a detailed investigation of the local electronic states in the (110) GaAs QW has not been performed so far.

To investigate the local electronic states in the nanostructures, microprobing techniques such as micro-PL and near-field scanning microscopy are powerful tools. In fact, these techniques have been extensively applied to the characterization of thin (001) GaAs QWs, revealing QD-like localized electronic states due to interface roughness.

In this study, we investigated the local electronic states in (110) GaAs QWs by the micro-PL technique with the help of a solid immersion lens (SIL) and attempted to clarify the reason for the broad PL line width, in order to improve and understand the CEO method and T-shaped QWRs.

This paper is organized as follows: in Sec. II, we describe the growth of (110) and (001) single QW (SQW) samples, and their characterization via conventional PL (macro-PL) and AFM measurements, which suggest that monolayer-step (ML-step) terraces with laterally sub-μm to μm width were formed in the (110) GaAs interface and that totally 3 to 3.5 well-width fluctuation existed in the (110) GaAs QW. We then point out questions or expectations raised by these measurements. In Sec. III, we describe the results obtained in micro-PL spectroscopy and imaging analyses of a (110) SQW sample using a (001) SQW sample as a reference sample, to show the temperature-dependent spatial inhomogeneity of PL images, spectrally-resolved PL images, and the sharp-line features of the PL spectra under point excitation via SIL at various excitation power levels, temperature levels, and positions. Our results demonstrate the QD-like localized nature of electronic states with an energy distribution suggesting the contribution of interface roughness on both the top and the bottom of the (110) QW. In Sec. IV, based on the micro-PL results obtained, we discuss the interface
roughness and electronic states which would be formed in the (110) QW by CEO and the influence on the T-shaped QWRs. Our conclusion is summarized in Sec. V.

II. SAMPLES

A. Sample structure and growth

We grew several samples of 5 nm GaAs SQW structures with AlAs barriers on (110) and (001) surfaces in separate growth runs with MBE. (110) GaAs SQW samples were grown on the cleaved (110) edge of a (001) GaAs substrate. The preparation procedure was as follows. After cleavage in air, the substrate was loaded into the MBE chamber and the top oxide layer was removed at 580°C. We grew the (110) SQW samples at a substrate temperature ($T_s$) between 490°C and 510°C. Setting the V/III flux ratio to 30 and the GaAs growth rate to 0.35 μm/hr, we successively grew 200–500 nm GaAs and GaAs/AlAs superlattice buffer layers, a 5 nm GaAs SQW sandwiched by 10 nm AlAs barriers, and a 10 nm GaAs cap layer without growth interruption.

As a reference, a 5 nm GaAs SQW of the same structure with AlAs barriers was grown on a GaAs (001) substrate under the conventional growth conditions with $T_s = 605°C$, a V/III flux ratio of 5, a GaAs growth rate of 1.0 μm/hr, and a growth interruption time at the GaAs surface of 45 s.

B. Macro-PL characterization

We first characterized the samples by conventional macro-PL measurements at 77 K under photoexcitation by a He-Ne laser with a spot size of about 100 μm. The solid and dashed curves in Fig. 1 show the macro-PL spectra of 5 nm GaAs/AlAs SQWs formed on (110) and (001) surfaces, respectively. The difference in peak energy was due to anisotropy in the heavy hole mass. Note that the PL spectrum of the (110) SQW had broader width than that of the (001) SQW. The full width at half maximum (FWHM) was 27 meV for the (110) SQW, and 7.2 meV for the (001) SQW. The PL width of 7.2 meV in the (001) SQW was smaller than the energy separation of about 15 meV in the quantization energy due to a 3-ML difference in well width. On the other hand, the PL width of 27 meV in the (110) SQW was greater than the energy separation corresponding to a 3-ML difference in well width, which suggested the existence of greater interface roughness in the (110) SQW.

The inset in Fig. 1 shows the FWHM of the PL spectra as a function of PL peak energy for seven (110) GaAs/AlAs SQW samples grown at different substrate temperatures. The same symbols at several PL peak energies show different excitation positions in the case of the same sample. The larger FWHM of the closed triangles as compared to the others was probably due to lower sample quality with slightly lowered growth temperature, because the sample had higher-density triangle-shaped facet structures on the surface as observed under an optical microscope. Lines in the inset indicate calculated PL-energy difference due to the well width deviations of $±0.1$ to $±0.4$ nm from the centered well width. Note that most samples follow the calculated lines with the deviation being about $±0.3$ to $±0.35$ nm. This result indicates that the (110) QWs had almost the same quality at growth substrate temperatures within the range of 490 to 510°C, and that a well width fluctuation of about 0.2 to 0.35 ML [1 ML = 0.2 nm in the (110) surface] existed in the (110) QW.

The dash–dotted curve in Fig. 1 shows the macro-PL spectrum of an additional reference sample of 5 nm GaAs SQWs with Al$_{0.3}$Ga$_{0.7}$As barriers on a (110) surface grown under the same conditions as those in the case of the (110) SQWs with AlAs barriers. Its FWHM was 13.5 meV and the corresponding well width fluctuation was 0.74 nm or 3.7 ML, which indicates that the large well width fluctuation in the (110) QW was independent of the barrier material.

C. AFM characterization

Figure 2 shows AFM images of the surface morphology on the top GaAs cap layers and cross sections of the images obtained for a (110) GaAs/AlAs SQW sample [Figs. 2(a) and 2(b)] and a (001) GaAs/AlAs SQW sample [Fig. 2(c)]. In the case of both samples, monolayer steps were clearly observed.

The (001) GaAs surface, shown in Fig. 2(c), had monolayer steps, which were regularly aligned and went down in one direction. The angle of tilt from the (001) direction was estimated to be 0.16°, which was within the range specified for the GaAs (001) wafer used in this study.

On the other hand, the (110) GaAs surface, shown in Figs. 2(a) and 2(b), had the monolayer or bilayer steps irregularly...
aligned. An 8-ML height difference in total (that is, a height fluctuation of about 4 ML) was formed in a 5 \( \mu \text{m} \times 5 \mu \text{m} \) region, shown in Fig. 2(a), though the substrate prepared by cleavage had just the (110) surface, suggesting that some instability existed in the GaAs growth on the (110) surface under the present growth conditions. Note that the lateral width of monolayer terraces in the island structures was very large and was sub-\( \mu \text{m} \) to \( \mu \text{m} \) scale.

Though the AFM measurements were performed examining the GaAs surface of the 10 nm cap layer, we expect that the top GaAs surface of the buried GaAs/AlAs SQW had similar morphology.

D. Motivation for the micro-PL study

In view of the results obtained in the macro-PL and AFM measurements, we had the following expectations or questions concerning the (110) GaAs/AlAs SQW.

First, a height fluctuation as large as 4 ML was expected in the upper GaAs surface of the (110) GaAs/AlAs SQW, which should cause well thickness fluctuation, and hence broad PL linewidth, in the SQW. Since the irregular patterns of ML steps and terraces had quite large lateral width of sub-\( \mu \text{m} \) to \( \mu \text{m} \) scale, we may spatially resolve the inhomogeneous electronic states through micro-PL measurements.
Second, the interfaces of the (110) SQW are expected to have large atomically-flat terraces with a lateral size of \( \mu \text{m} \) scale, which is much larger than the Bohr radius of two-dimensional (2D) excitons, so the ideal 2D electronic states might be selectively observed in such regions through micro-PL measurements. This microscopic picture is quite different from that in the case of very thin \( \text{GaAs}/\text{AlAs} \) SQWs, where localized QD states are formed due to interface roughness.\(^{12-17}\)

To clarify these expectations, in the experiments described in the following sections we investigated the interface roughness and the modulated electronic states in the (110) \( \text{GaAs} \) QW by micro-PL imaging and spectroscopy.

### III. MICRO-PL IMAGING AND SPECTROSCOPY

#### A. Inhomogeneity in PL images of (110) and (001) GaAs SQWs

Figure 3 shows PL images at 4.7 and 60 K obtained for the (110) \( \text{GaAs}/\text{AlAs} \) SQW [Fig. 3(a)] and the (001) \( \text{GaAs}/\text{AlAs} \) SQW [Fig. 3(b)] under uniform excitation by a He-Ne laser, and the cross-sectional intensity profiles of the PL images at the position shown by dotted lines at four [or three in Fig. 3(b)] different temperatures. Each intensity profile was normalized by the averaged PL intensity at each temperature. The spatial resolution of this micro-PL measurement was 0.8 \( \mu \text{m} \). The experimental setup has been presented elsewhere.\(^{19}\)

In the case of (110) SQW, the PL images showed spatial inhomogeneity with a lateral size of \( \mu \text{m} \) scale, whereas the profile was almost uniform in the case of (001) SQW. The peak and valley positions in the cross-sectional intensity profiles in the case of (110) SQW were the same at all four temperatures and only the relative intensity comparing the intensity at the peaks to that at the valleys changed, which most probably reflected an inhomogeneous distribution of the electronic states in the sample.

#### B. Spectrally-resolved PL images of the (110) GaAs SQW

To reveal the origin of inhomogeneity in the PL images, we measured spectrally-resolved PL images adding an interference filter with a bandwidth of 2 nm, corresponding to 4 meV, in front of the charge coupled device (CCD) camera. Figure 4(a) shows spectrally-resolved PL images filtered at three different energies, as well as an integrated PL image obtained without the interference filter (denoted as ‘‘All’’), in the same region of the (110) \( \text{GaAs}/\text{AlAs} \) SQW at 60 K. Simultaneously monitored PL spectra without (solid line) and with the filter at three different energies (dashed lines) are also shown in Fig. 4(b).

At the high energy of 1.648 eV, the PL intensity was fairly spatially uniform. At the low energy of 1.628 eV, on the other hand, the PL intensity contrast increased and the bright regions in the PL image were strongly spatially localized. The positions of the bright regions coincided with those in the integrated PL image. This means that the spatial inhomogeneity in the PL intensity was caused by the inhomogeneous spatial distribution of the quantization energy in the SQW.

Note that the lateral spatial size of the PL intensity fluctuation was sub-\( \mu \text{m} \) to \( \mu \text{m} \) scale, which was similar to that of monolayer terraces on the (110) \( \text{GaAs} \) surface observed by AFM. Together with the finding that the result obtained by AFM, indicating that there was height fluctuation of about 4 ML on the (110) \( \text{GaAs} \) surface, was consistent with the well width fluctuation of about 3 to 3.5 ML estimated from the broad macro-PL line width, this similarity suggested that the spatial inhomogeneity in the PL intensity reflected the spatial distribution of the quantization energy due to monolayer terraces with laterally sub-\( \mu \text{m} \) to \( \mu \text{m} \) width existing at an interface of the SQW.

The temperature dependence of the PL intensity contrast shown in Fig. 3 is explained by the distribution of the quantization energy in the SQW and the temperature-dependent carrier migration processes as follows.

At the lowest temperature, the PL intensity contrast was weak due to the short migration length of photoexcited carriers. When the temperature was raised from 4.7 to 60 K,
question as to whether the ideal 2D states are formed or not, we performed micro-PL spectroscopy measurements and our findings concerning the local electronic states in the (110) GaAs QW are discussed in the next subsection. Based on the results obtained by micro-PL spectroscopy concerning the local electronic states and interface roughness, moreover, we again discuss the inhomogeneous PL images and their temperature dependence.

C. Point spectroscopy of local electronic states in the (110) GaAs SQW

To investigate the local electronic states in the (110) GaAs QWs, we performed point-excitation micro-PL spectroscopy with high-spatial and high-spectral resolution. In order to enhance the spatial and spectral resolution, we used the micro-PL setup combined with a SIL and a 76-cm monochromator with a liquid nitrogen-cooled CCD camera. The spatial resolution and the spectral resolution were enhanced to 0.4 μm and 0.2 meV, respectively. For the point excitation, light from a He-Ne laser was focused on the sample surface with a spot size of 0.4 μm through an objective lens and a SIL.

Moreover, to obtain local electronic states at a selected position on the sample exactly, we first obtained PL images under uniform excitation using the CCD camera as shown in the upper part of Fig. 5(a) prior to the spectroscopy, and selected an excitation position. Then, we changed the excitation mode to point excitation and obtained PL images as shown in the lower part of Fig. 5(a) and the PL spectra. This procedure enabled us to maintain the excitation at the selected position of the sample surface precisely even though the sample moved due to thermal drift or other extrinsic effects.

Figure 5(b) shows PL spectra at 4.8 K at the position shown in Fig. 5(a) for various excitation power levels. Here, the intensity of each PL spectrum was calibrated in terms of the excitation power and the acquisition time of the CCD camera, so that the PL intensity becomes constant if it is proportional to the excitation power.

Note the PL spectra under low excitation power, where only a single sharp line appeared at low temperature (150 K). The PL intensity contrast became weak again, most likely due to the thermal population effect of the carriers. As the temperature increased and the thermal energy became close to the difference in the quantization energy in the SQW, carriers were thermally activated, and occupied surrounding higher energy states, which reduced the PL intensity contrast.

D. Point spectroscopy–excitation-power dependence

In the PL spectra at higher excitation power in Fig. 5(b), additional peaks (open symbols), which we assigned to biexcitons, came up on the low energy side of the original peaks remaining from low excitation power (solid symbols). To demonstrate it more clearly, the excitation-power dependence of a strong PL peak at a different position is shown in Fig. 6(a), where only a single sharp line appeared at low excitation power. Figure 6(b) shows the intensity of the

![Image](https://via.placeholder.com/150)

**FIG. 4.** (a) Spectrally-resolved micro-PL images obtained at three different wavelengths selected by means of an interference filter and an integrated micro-PL image obtained without the filter at the same position (denoted as “All”) for the (110) GaAs/AlAs SQW at 60 K. (b) Simultaneously monitored PL spectra obtained without (solid line) and with the filter at three different wavelengths (dashed lines).

However, the PL contrast became stronger. This was due to enhanced carrier migration. With enhanced carrier migration at high temperature, photoexcited carriers preferably flowed from surrounding higher energy states into spatially-localized lower energy states formed in the island regions due to interface roughness and radiatively recombined there. Then the PL intensity contrast was enhanced. At higher temperature (150 K), the PL intensity contrast became weak again, most likely due to the thermal population effect of the carriers. As the temperature increased and the thermal energy became close to the difference in the quantization energy in the SQW, carriers were thermally activated, and occupied surrounding higher energy states, which reduced the PL intensity contrast.

In order to verify the hypothesis that the PL image reflects the well width fluctuation with μm scale and to answer the
original peaks and the additional peaks at various excitation power levels. The new emission line increased superlinearly with the excitation power, whereas the original peak showed linear dependence. The energy separation between the new and the original peaks was about 2.5–3.0 meV. This value is similar to the binding energy of biexcitons in the naturally-formed QD states in the (001) GaAs QWs$^{13}$ and that in the self-organized QDs.$^{25}$

The number of incident photons at the lowest excitation power of 1.2 W/cm$^2$ in Fig. 5 was $5 \times 10^9$ photons/s in the
excitation spot area with \( f = 0.4 \) \( \mu \)m, which corresponds to an averaged exciton number of 0.04 in the excitation spot assuming that the reflection at the surface is 0.3, the absorption in the SQW is 3\%, and the radiative lifetime of excitons is about 0.4 ns. This supports the view that the emission was from single excitons at the lowest excitation level. The new lines begin to appear at an excitation power of about 10 W/cm\(^2\) corresponding to the averaged carrier number of 0.3, which is a reasonable value for the appearance of biexcitons. The broad background PL that appeared at very high excitation power above 1000 W/cm\(^2\) must originate from interaction between many carriers, or plasma emission.

Note in Fig. 5(b) that biexciton peaks were observed not only for the lowest energy PL peak but also for the high energy peaks, which supports our assignment that each PL peak observed at the low excitation power was from the ground state in each local minimum in energy.

E. Spectral and spatial distribution of local electronic states in the (110) GaAs SQW

To see the spatial distribution of the local energy minima in the (110) QW, we observed the PL spectra under weak point excitation at several positions on the sample. Figure 7(a) shows the PL peak positions observed at ten different positions; two (a1 and a2) at bright PL positions, four (b1–b4) at middle intensity positions, and four (c1–c4) at dark positions, which are marked by crosses in Fig. 7(b). In Fig. 7(a), the size of the dots represents the PL intensity of each sharp peak. Vertical dashed lines show the calculated PL peak positions for GaAs/AlAs (110) SQWs differing in thickness by 0.2 nm steps (a ML step).

In Fig. 7(a), the bright and dark regions in the PL image of Fig. 7(b) have different PL peak energy distributions. In brighter regions (a1, a2), relatively strong PL peaks were dominantly observed on the lower energy side. In darker regions (c1–c4), on the other hand, peaks were located on the higher energy side with weak intensity. This is consistent with the spatially-resolved PL images in Fig. 4(a) and is explained by the spatial distribution of the quantization energy in the (110) QW and the carrier migration from high energy states to low energy states.

It is important to note the energy distribution of PL peaks in Fig. 7(a). As indicated by circles drawn to guide the eyes, three to five neighboring PL peaks made groups, and those groups had energy spacing almost equal to the separation of the PL peak energy in (110) GaAs QWs with one ML difference in well width as marked by vertical dashed lines.

To explain such unique energy distribution, we assumed the following interface model for the GaAs (110) SQW. As was observed in the AFM study on the GaAs surface, the top surface of GaAs/AlAs SQW, where AlAs covered the GaAs surface, should have large ML terraces with sub- \( \mu \)m to \( \mu \)m width. On the other hand, the bottom surface, where GaAs covered the AlAs surface, should have shorter-scale roughness due to the low mobility of Al atoms. Hence, several localized QD-like states due to small-scale interface roughness in the bottom surface should exist in each large ML terrace with sub- \( \mu \)m to \( \mu \)m scale on the top surface.

Since the excitation spot size was comparable to this terrace size, it is expected that one or a few terraces contributed to the PL spectra. Then, each observed PL peak group might originate from one \( \mu \)m-scale terrace in the top interface, and the sharp PL peaks in each group might originate from localized QD-like states due to interface roughness in the bottom surface within each ML terrace in the top interface, contrary to our expectation for ideal 2D formation.

FIG. 7. (a) Peak positions of micro-PL spectra observed at ten different excitation positions on the GaAs/AlAs (110) SQW sample and (b) their excitation positions shown as crosses on the micro-PL image via SIL under uniform excitation. The size of the dots in (a) represents the PL intensity of each peak. Vertical dashed lines in (a) show the calculated PL peak positions for GaAs/AlAs (110) SQWs with well width of every ML step.

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gration from the groups of the localized states in the surrounding energetically higher ML terraces to the groups in the lower energy terraces.

Moreover, note in Fig. 8 that the linewidth of the sharp PL peaks was independent of the temperature. This also supports the view that the electronic states in the (110) QW were not 2D but QD states.\cite{27}

IV. INTERFACE ROUGHNESS IN (110) QW AND T-QWR GROWN BY CEO

From the results of micro-PL spectroscopy and imaging, we found that localized QD states were formed in the (110) GaAs/AlAs QW with $\mu$m-scale large ML terraces on the top AlAs-on-GaAs interface and shorter-scale roughness on the bottom GaAs-on-AlAs surface.

In order to realize ideal 2D states with $\mu$m-scale ML terraces on the GaAs surface, we have to reduce the roughness of the bottom AlAs barrier surface, for example, by the CEO method. The (110) GaAs QW grown by the CEO method with in situ cleavage is expected to have a top interface with $\mu$m-scale ML terraces and a bottom cleaved flat surface.

Hasen et al. performed a micro-PL experiment on a single T-QWR grown by the CEO method. They observed discrete sharp emission peaks from the QWR and concluded that these sharp peaks originated from localized QD states due to fluctuation of the monolayer thickness of the two constituent QWs; the first growth (001) QW and the CEO (110) QW.\cite{28} However, the individual contribution of the interface roughness in the two QWs to the localized states has never been clarified. Furthermore, AFM study has never been performed on structures formed by the CEO method. To solve this problem, microscopic spectroscopy studies described in this paper are necessary for (110) QWs grown by the CEO method in comparison with our results on (110) QW grown by the conventional method. Especially, it should be elucidated that how the growth conditions affect the growth surface and the microscopic electronic states in the (110) QWs formed by both methods.

V. CONCLUSIONS

We have performed macro- and micro-PL spectroscopy and imaging analyses of the GaAs SQWs grown on the (110) cleaved surface and characterized the well width fluctuation and the spatial and spectral distribution of local electronic states in the (110) GaAs QWs. We have found that the well width fluctuation in the GaAs (110) SQW was about 0.6 to 0.7 nm, which corresponds to 3 to 3.5 ML in the (110) surface, with the lateral size of sub-$\mu$m to $\mu$m scale. This was also supported by AFM measurements of the (110) GaAs surface of the same sample.

In the high-resolution micro-PL spectroscopy assisted by the SIL, sharp PL peaks were observed from the localized electronic states formed at local energy minima in the (110) GaAs QWs, which showed a QD-like nature in terms of their dependence on excitation power and temperature. The
spectral and spatial distribution of the sharp PL peaks demonstrated that the localized QD states were formed as a result of the short-scale roughness in the bottom AlAs surface of the (110) GaAs/AlAs SQW on each large ML terrace with sub-µm to µm scale in the top GaAs surface.

Therefore, the broad PL linewidth in the (110) GaAs/AlAs SQW should be attributed to the spectrally and spatially inhomogeneous PL from local electronic states as a whole, including a plasma emission effect, reflecting the unique interface roughness in the (110) GaAs QW.

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