Microphotoluminescence characterization of cleaved edge overgrowth T-shaped In\(_x\)Ga\(_{1-x}\)As quantum wires

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Microphotoluminescence (micro-PL) characterization was performed for T-shaped In\(_x\)Ga\(_{1-x}\)As quantum wires (T-QWRs) fabricated by the cleaved edge overgrowth method with molecular beam epitaxy. The spatial distribution of optical properties in In\(_x\)Ga\(_{1-x}\)As T-QWRs was examined by means of PL intensity imaging and scanning micro-PL spectroscopy with about 1 \(\mu\)m spatial resolution. In the successfully fabricated 3.5-nm-scale In\(_{0.17}\)Ga\(_{0.83}\)As T-QWRs with Al\(_{0.3}\)Ga\(_{0.7}\)As barriers, uniform PL image and PL spectra from QWRs along the wire direction were observed, which indicates spatially uniform and high quality QWRs were formed. The effective lateral confinement energy of one-dimensional excitons was found to be 34 meV, showing the usefulness of In\(_x\)Ga\(_{1-x}\)As as a well material in T-QWR structures. On the other hand, in the unsuccessfully fabricated 4-nm-scale In\(_{0.09}\)Ga\(_{0.91}\)As T-QWRs with Al\(_{0.3}\)Ga\(_{0.7}\)As barriers, PL image and spectra were inhomogeneous. In addition, these results demonstrate the importance of flat cleaved surface and optimized overgrowth condition to fabricate uniform T-QWRs.

I. INTRODUCTION

Semiconductor quantum wire (QWR) structures and their one-dimensional (1D) electronic states have attracted great interest in a recent decade because of their promising superior performance as electronic and optoelectronic devices such as high mobility transistors and low threshold lasers.

Recently, T-shaped QWRs (T-QWRs)\(^1\)\(^\rightarrow\)^\(^5\) fabricated by the cleaved edge overgrowth (CEO) method\(^6\) with molecular beam epitaxy (MBE) have shown some improved optical properties of 1D excitons, which are the excitonic laser emission,\(^7\) the enhancement of exciton binding energy,\(^8\) and the concentrated oscillator strength.\(^9\) In the CEO method, quantum wells (QWs) firstly grown by the conventional MBE method are in situ cleaved, and the second QW is overgrown on the cleaved surface of the first QWs.\(^3\)^\(^5\) The 1D states are formed at the T-intersection between the first growth QWs and the overgrowth QW. This method has an advantage that the fabrication of arbitrary size of T-QWRs with atomic-size accuracy of 2D epitaxial growth is possible as the combination of the first growth QWs and the overgrowth QW.

In T-QWRs, the effective lateral confinement energy \((E_{1D-2D}^0)\) of excitons are defined as the energy separation of 1D excitons in QWRs and 2D excitons in the parent QWs. To stabilize 1D excitons and enhance their 1D characters, it is important to enhance \(E_{1D-2D}^0\) by increasing the potential barriers and reducing the size of T-QWRs. In the recent work, 5-nm-scale GaAs T-QWRs with AlAs barriers demonstrated \(E_{1D-2D}^0 = 38\) meV,\(^8\)^\(^1\)^\(^1\)^\(^1\) which is twice larger than \(E_{1D-2D}^0 = 18\) meV realized in 5-nm-scale GaAs T-QWRs with Al\(_{0.3}\)Ga\(_{0.7}\)As barriers.\(^8\)^\(^1\)^\(^2\)

In the view point of device applications, however, the use of AlAs should be avoided because the AlAs material has several problems, such as oxidation, interface roughness, and defects. In addition, the use of AlAs as a barrier material decreases the effective refractive index around the T-QWR region because of low refractive index in the AlAs material, which is not appropriate in optoelectronic devices.

Therefore, instead of the use of AlAs as the barrier material, we need to adopt In\(_x\)Ga\(_{1-x}\)As as a well material in T-QWRs with Al\(_{0.3}\)Ga\(_{0.7}\)As barriers and realize the tight confinement of 1D excitons.

The technical difficulty of this approach lies mostly in the overgrowth of In\(_x\)Ga\(_{1-x}\)As on the cleaved (110) surface. Firstly, the MBE growth condition for high quality In\(_x\)Ga\(_{1-x}\)As layer on (110) surface is very limited. The previous experiments for the growth condition of In\(_{0.15}\)Ga\(_{0.85}\)As layer on (110) GaAs surface have shown that slight deviation from the optimum substrate temperature of 430 °C causes the decrease of photoluminescence (PL) intensity and the
low quality of surface morphology. This temperature range is largely different from that of GaAs and Al$_{0.3}$Ga$_{0.7}$As layers, which is optimized at 500 °C. In addition, the uniformity of CEO critically depends on the flatness of the cleaved surface, so that incomplete cleavage is not allowed. Moreover, in such a critical growth condition, the uniform overgrowth can be easily disturbed by any difference in stacking coefficient, diffusion length of atoms, and/or lattice constant on the cleaved surface. Quantum microstructures consist of different component materials and compositions.

Under these circumstances, the fabrication of In$_{0.17}$Ga$_{0.83}$As T-QWRs with spatial uniformity and high crystal quality is important in the first place. Therefore, at least until the reproducibility of the CEO process is established, the microscopic characterization of the spatial uniformity of samples is required to support the estimation of 1D characters.

In the observation of spatial distribution of quality in quantum microstructures such as T-QWRs, optical measurements with high spatial resolution and high efficiency are required because of their small size. On the basis of micro-PL measurements with the spatial resolution of less than 1 μm, we can obtain spatially-resolved optical properties of these quantum microstructures. We characterize the spatial distribution of the sample structure and quality from the images of PL intensity. Detailed electronic structure in each region is given by the scanning micro-PL spectroscopy.

In this article, we describe the micro-PL characterization of In$_{0.17}$Ga$_{0.83}$As T-QWRs prepared by the CEO method with MBE. By micro-PL measurements, we were able to effectively obtain spatial distribution of optical properties in In$_{0.17}$Ga$_{0.83}$As T-QWRs in addition to the electronic states of 1D carriers.

In the next section, we describe a sample structure with preparation condition and detailed methods of micro-PL studies. In Sec. III, we present the results of micro-PL studies for the fabricated In$_{0.17}$Ga$_{0.83}$As T-QWRs. We compare the PL image and spectra of a successfully fabricated sample with those of an unsuccessful sample, to show the importance of the cleavage process and overgrowth condition. We demonstrate the uniformity of T-QWRs along the wire direction, and then discuss $E_{1D-2D}$ of excitons in In$_{0.17}$Ga$_{0.83}$As T-QWRs to show the usefulness of the In$_{0.17}$Ga$_{0.83}$As material. In Sec. IV, we summarize our experimental results and conclude.

II. EXPERIMENT

A. Sample structure and preparation

The In$_{0.17}$Ga$_{0.83}$As T-QWR sample was fabricated by the CEO method with MBE, following the CEO procedures described in the previous reports. Figure 1 schematically shows the structure of the T-QWR sample. The QWR states are formed at the T-intersections of the first growth QWs (denoted as QW1) and the second QW (denoted as QW2) overgrown on the cleaved edge surface after in situ cleavage. In the first growth, we grew a 500 nm GaAs buffer, a 5 μm Al$_{0.3}$Ga$_{0.7}$As layer, an In$_{0.17}$Ga$_{0.83}$As multiple-QW (MQW) layer with Al$_{0.3}$Ga$_{0.7}$As barriers, a 10 nm AlAs layer, a 5 μm Al$_{0.3}$Ga$_{0.7}$As layer, and a 50 nm GaAs cap layer onto a (001) GaAs substrate with the conventional MBE growth condition. The In$_{0.17}$Ga$_{0.83}$As MQW layer as QW1 has 10 periods of In$_{0.17}$Ga$_{0.83}$As QW with thickness $a = 3.7$ nm and Al$_{0.3}$Ga$_{0.7}$As barriers with thickness $c = 100$ nm. Total thickness of MQW region is about 1 μm. After in situ cleavage, we grew an In$_{0.17}$Ga$_{0.83}$As QW layer of thickness $b = 3.4$ nm as QW2, and 2 monolayer Al$_{0.3}$Ga$_{0.7}$As on the cleaved edge surface at the substrate temperature of 430 °C. The growth rate of In$_{0.17}$Ga$_{0.83}$As was 0.5 μm/hour, and the V/III flux ratio was 30. Then, the sample was heated up to 500 °C for the deposition of a 10 nm Al$_{0.3}$Ga$_{0.7}$As barrier layer and a 10 nm GaAs cap layer.

As a result, the sample has a 1-μm-wide region containing 10 periods of QWRs sandwiched by two 5-μm-wide regions of QW2 on the (110) CEO surface. This sample structure allows one to assign energy levels of QW1, QW2 and QWR, which is very important to determine the effective lateral confinement energy $E_{1D-2D}$ of 1D excitons precisely, by micro-PL measurements. We call this sample as S-1 in this article.

We also used another In$_{0.17}$Ga$_{0.83}$As T-QWR sample (S-2) for comparison with S-1 in the PL experiments. The sample structure was basically the same as that of S-1 except for In content of 0.09, well thickness of $a = 4.1$ nm. Since this sample was fabricated in early day before the optimum growth condition of In$_{0.17}$Ga$_{0.83}$As T-QWRs was established, the quality of T-QWRs in S-2 was not expected to be good.

B. Experimental setup

In order to evaluate sample quality and uniformity and to assign quantized energy levels of QWRs and the parent QWs, we performed micro-PL measurements with He-Ne laser as an excitation source using a micro-PL setup shown in Fig. 2(a). The configuration of the sample in the micro-PL
measurements is shown in Fig. 2(b). The micro-PL setup consists basically of an objective lens, an electrically-cooled CCD camera (Princeton Instruments, PentaMAX-1317-K) for PL imaging, a monochromator (Acton Research, SP300i) with a liquid-nitrogen-cooled CCD camera (Princeton Instruments, LN/CCD-1024EHRB) for PL spectroscopic measurement, and a monitoring CCD-TV camera with tungsten illumination lamp. Samples were mounted in a cryostat (Oxford Instruments, CF2102) with an optical glass window of 1.5 mm thickness. An objective lens (Union, PLLWDC40×) with nominal magnification factor of 40, working distance of 10 mm, and numerical aperture (NA) of 0.5, was used in this study. This objective lens has a function which compensates the aberration caused by a glass window of the cryostat existing between the objective lens and the sample.

In this micro-PL setup, two kinds of photoexcitation are available. One is point photoexcitation at a position on the sample surface. In this excitation mode, output of a He-Ne laser [denoted as He-Ne1 in Fig. 2(a)] was reflected by a beam splitter and focused, through the objective lens, into an almost-diffraction-limited spot of less than 0.8 μm diameter on the sample in the cryostat, in the normal-incidence configuration. Photoexcited position on the sample surface was monitored by the CCD-TV camera.

The other is uniform photoexcitation onto the whole of the sample surface, in which defocused light of a He-Ne laser (denoted as He-Ne2) was directly introduced to the sample surface from a space between the cryostat and the objective lens.

The PL from the sample was detected via the (110) surface in the backward scattering geometry, collected by the objective lens, reflected by a beam splitter, and focused on the electrically-cooled CCD camera for the PL imaging measurement, or focused in an entrance slit of the monochromator with the liquid-nitrogen-cooled CCD camera for the PL spectroscopic measurement. In the imaging configuration, we have obtained the spatial resolution of 1 μm and 0.65 μm at wavelength of λ=780 nm and 546 nm, respectively, through the glass window of 1.5 mm of the cryostat. These are almost the diffraction limit 0.61λ/NA of this objective lens.

III. RESULTS AND DISCUSSIONS

A. PL imaging with uniform photoexcitation

In order to characterize the uniformity and quality of fabricated In₀.17Ga₀.83As T-QWRs, we measured images of total PL intensity with uniform photoexcitation by He-Ne laser via a sharp-cut filter for excitation light. Figure 3(a) shows the PL image for In₀.17Ga₀.83As T-QWR sample S-1 at 5 K. Also shown in Fig. 3(b) is the reflection image at the same region obtained using the same experimental setup with illumination by a tungsten lamp filtered at 546 nm.

Figure 3(b) shows that the allow-like facet structures exist at the boundary of GaAs buffer layer and Al₀.5Ga₀.5As barrier layer. However, no facet structure or imperfection of cleavage was observed on the QWR region.

In Fig. 3(a), bright PL intensity was observed uniformly along the wire direction (x direction in Fig. 2), which is consistent with the good surface morphology on the QWR region shown in Fig. 3(b). Such uniform images were obtained from the whole region of the sample S-1. This result means that the uniform In₀.17Ga₀.83As QWRs and QWs were formed by the successful CEO process. The PL image consists of a brighter region in the middle and less bright regions in both sides, which coincides with the sample structure of the 1-μm-wide region with QWRs on QW1 and the two 5-μm-wide regions of QW2. Note here that the image of total PL intensity was detected without a bandpass filter. Thus, the brighter region of the PL image was a superposition of PL from QWRs and that from QW1. These two contributions
were resolved by the spatially-resolved PL spectroscopic measurements, which is described later.

For comparison, we studied the PL and reflection images of the In_{0.09}Ga_{0.91}As T-QWR sample S-2, which was fabricated before the optimization of preparation condition. Figure 4 shows the results for S-2 in the same measurements as those of Fig. 3. In Fig. 4(a), bright region of PL intensity was also obtained from 11-μm-wide region on the first growth layer structures. In S-2, however, PL intensity on the QWR region was not uniform along the wire direction. That on the QW2 region was also inhomogeneous.

In addition, stripe-structures of PL were observed on the substrate region in the first growth. Similar stripe-structures were also observed in the reflection image on the substrate region of S-2 as shown in Fig. 4(b), and the positions of these structures nearly coincide with that of PL. Note that these stripe-structures were not observed in both the PL and the reflection images of S-1 in Fig. 3. The observation of these stripe-structures in the reflection image indicates the existence of the large step edges on the overgrowth surface. We believe that such large steps were caused in the cleavage process.

When the step edges are formed on the cleaved surface by unsuccessful cleavage, it is expected that the anomalous facet growth occurs on the step edges, and that the growth condition is disturbed around there. As a result, the overgrowth layer with low quality would be fabricated. The unsuccessful cleavage shown on the GaAs substrate region in S-2 suggests the existence of small step-structures and/or the disturbed growth condition, also at the QWR region. Therefore, the inhomogeneous PL image observed on the QWR region in S-2 was most likely caused by the unsuccessful cleavage and/or the deviation from the optimum growth condition.

On the basis of this comparison, the uniform PL image on the QWR region in S-1 indicates successes both in the in situ cleavage process and the overgrowth on the cleaved surface. Hence, in order to fabricate the uniform In_{x}Ga_{1-x}As T-QWRs, the optimization of both the in situ cleavage process and the overgrowth condition are very important.

**B. PL imaging with point photoexcitation**

We performed PL imaging with point photoexcitation to reveal the effects of the diffusion of carriers in the point excitation, and to obtain the effective spatial resolution in the PL spectroscopic measurements under point photoexcitation.

Figures 5(a) and 5(b) show the images of PL intensity for S-1 at 5 K with the point excitation on the QWR region and on the 5-μm-wide QW2 region, respectively. Though the spot size of the point excitation was about 0.8 μm diameter, PL images were observed from more spread area than the spot size owing to the diffusion of the carriers. The PL image on the QWR region has smaller width across the wire direction. This means that the carrier diffusion takes place dominantly in QWRs and QW1 rather than on barriers, so that the diffusion across QWRs and QW1 is restricted. On
the other hand, the PL image on the 5-μm-wide QW2 region is almost circular, which indicates that the carrier diffusion is almost isotropic. The diffused length including the initial width 0.8 μm of excitation spot are estimated as the full width at the half maximum of PL intensity in Fig. 5. Its values are 1.3 μm for the QWR region along the wire direction, and 1.6 μm for the QW2 region.

C. Scanning micro-PL spectroscopy

It is important to precisely determine the quantized energy levels of QWRs, QW1, and QW2 to discuss the stability of 1D excitons. Furthermore, to assign PL signals unambiguously, we need spatially-resolved PL spectroscopic measurements with point photo-excitation on the overgrowth surface of the sample. Since the sample has a 1-μm-wide region with QWRs on QW1 and two 5-μm-wide regions of QW2, we are able to detect PL signals from QWRs, QW1, and QW2, separately by probing each region.

Figure 6 shows PL spectra of sample S-1 at 5 K where the probed position was scanned in the first growth direction (z direction in Fig. 2) by a step of 1 μm from the (001) top surface to the GaAs substrate. The probed positions and the sample structure are also schematically shown as an inset in Fig. 6. The distinct PL peaks corresponding to the sample structures were detected at each position on the overgrowth surface of the sample, as follows.

Curve a in Fig. 6 was obtained when the excitation laser was incident onto the QWR region. This PL spectrum shows two distinct peaks. The PL peaks at higher energy side was attributed to QW1 since a PL peak was observed at the same energy position in the sample without overgrowth layer. Then, the PL peak at lower energy side was attributed to QWR. Curves b and c in Fig. 6 were obtained on the QW2 region, where the PL peak came from QW2. Curve d was obtained with the excitation on the GaAs substrate region in the first growth. The PL peak shown in curve d came from the In0.17Ga0.83As overgrowth QW with barriers consist of an overgrowth Al0.3Ga0.7As layer and a GaAs of substrate.

Such unambiguous assignment of PL peaks to each quantum structure in In0.17Ga0.83As T-QWRs enables the exact estimation of each quantized energy level, and hence the $E_{1D-2D}^*$ of 1D excitons. This subject will be discussed in Sec. III D.

On the other hand, in the sample S-2, PL spectra do not show characteristic four distinct peaks expected for each quantum structure. Figure 7 shows PL spectra of sample S-2 at 5 K with point photoexcitation.

Curve a in Fig. 7 shows PL spectrum on the QWR region. The higher energy peak in curve a came from QW1 because of the same reason as that in S-1. The PL signal from QW1 shows clear peak and narrow width, which indicates the high quality of the QW1. However, the PL signals at the lower energy side are not simple, suggesting some unexpected complex structures formed in the CEO process.

Curve b in Fig. 7 was obtained on the QW2 region. The PL signal in curve b shows unclear peak and broad spectrum width in comparison with that from QW1. Curve c shows the same characteristics as those in curve b. Moreover, additional PL signals were observed at lower energy side (1.45–1.47 eV) in curve c. The peak position of these PL signals shifted continuously to far lower energy side and connected to the PL signals observed on the GaAs substrate region. These results indicate the low quality of overgrowth layers in S-2.

Curve d obtained on the GaAs substrate region has unclear peak and broad spectrum width as compared with curve d in S-1 shown in Fig. 6. The PL signals observed on the GaAs substrate region changed their peak position, spectrum width and intensity as the excitation position was scanned, and suddenly disappeared at a probed position. Similar abrupt change in the PL intensity was obtained across the stripe-structures on the substrate region in the PL image of S-2 shown in Fig. 4. These results indicate that the anomalous overgrowth occurred on the cleaved surface.

Next, we measured PL spectra on the QWR region with scanning the excitation position along the wire direction, in order to investigate the uniformity of the electronic states of the T-QWRs. Figures 8 and 9 show PL spectra on the QWR region for S-1 and for S-2, respectively, scanned along the wire direction by a step of 1 μm for 50 μm totally.

The PL spectra of the sample S-1 have two distinct PL peaks, attributed to QW1 and QWR, as shown in Fig. 8. The intensity of these PL peaks is nearly constant along the wire direction, and their peak positions do not move. This feature...
was reproduced in the whole range of S-1. The uniform PL spectra of QWRs along the wire direction confirm the high quality and uniformity of the fabricated In0.17Ga0.83As T-QWRs.

In the sample S-2, PL signals from QW1 have fairly uniform intensity and their peak positions do not move, reflecting the high quality of QW1. On the other hand, PL signals observed around 1.51 eV do not show a distinct peak. Moreover, their energy positions fluctuate and intensity varies along the wire direction, which is largely different from that in S-1. These results agree with the inhomogeneous PL image on the QWR region in S-2 shown in Fig. 4(a), and indicate that the expected T-QWRs were not formed.

On the basis of all the PL images and the micro-PL spectra with scanning, it is concluded that the sample S-1 has the intended T-QWR structure with high crystal quality and uniformity, and shows sharp well-resolved PL peaks from constituent quantum structures of QWR, QW1, and QW2. On the other hand, the CEO process for the sample S-2 was unsuccessful so that the anomalous overgrowth structures were formed inhomogeneously instead of the designed uniform layers. Since some of the anomalous structures make unexpected PL signals shown in Figs. 7 and 9, it is misleading to assign such signals to QWR or QW2 only from a PL measurement on few positions or a spatially-integrated PL measurement for a large region. Note that the high-quality samples like S-1 have been obtained after the refinement of the CEO process by the repeated fabrication and characterization of unsuccessful samples like S-2. Therefore, the characterization of quantum microstructures by the spatially-resolved PL measurement is important both to improve the sample fabrication method and to investigate their electronic properties.

D. Effective lateral confinement energy of excitons

The main purpose of the use of In0.17Ga0.83As as a well material in T-QWRs is the realization of stable 1D excitons and enhancement of 1D characters in QWRs. Thus, we look into the effective lateral confinement energy $E_{1D-2D}^*$ of excitons in T-QWRs, which represents the stability of 1D excitons in the QWRs.

Since the assignment of PL peaks for the In0.17Ga0.83As T-QWRs has been established in the sample S-1, the energy levels of QWR, QW1, and QW2 are determined from the PL peak energies. From Fig. 6, the $E_{1D-2D}^*$ was estimated to be 34 meV as the separation of PL peak energies of QWR and QW1.
The $E_{1D-2D}^*$ of 34 meV in 3.5-nm-scale In$_{0.17}$Ga$_{0.83}$As T-QWRs with Al$_{0.3}$Ga$_{0.7}$As barriers is far larger than the $E_{1D-2D}^*$ of 18 meV in 5-nm-scale GaAs T-QWRs with the same Al$_{0.3}$Ga$_{0.7}$As barriers. Note, however, that such a tight confinement of 1D excitons was achieved without the use of ALAs material.

This successful fabrication of uniform In$_{0.17}$Ga$_{0.83}$As T-QWRs with well-controlled cleavage process and optimized growth condition and the enhancement of confinement energy of 1D excitons indicate the usefulness of In$_{1-x}$Ga$_x$As as a well material in T-QWRs. By increasing In content $x$ of In$_{1-x}$Ga$_x$As QW and Al content $y$ of Al$_y$Ga$_{1-y}$As barriers and optimizing the structure of T-QWRs, tighter confinement of 1D excitons with $E_{1D-2D}^*$ far over the thermal energy $k_BT \sim 26$ meV at room temperature will be realized.

IV. CONCLUSIONS

The micro-PL characterization by imaging and scanning spectroscopy was performed at 5 K for the two samples of In$_{1-x}$Ga$_x$As T-QWRs fabricated by the CEO method with MBE.

The PL intensity images were measured with the NA = 0.5 objective lens and the cooled CCD detector realizing the almost-diffraction-limited resolution of 1 $\mu$m at $\lambda = 780$ nm under the uniform photoexcitation and the point photoexcitation with He-Ne lasers. The scanning micro-PL spectroscopy was performed by 1 $\mu$m step with the spatial resolution of 1.3–1.6 $\mu$m limited by the carrier diffusion. From the PL images and the PL spectra, the spatial distribution of the sample quality and the electronic energy levels is examined.

In the successfully fabricated 3.5-nm-scale In$_{0.17}$Ga$_{0.83}$As T-QWRs with Al$_{0.3}$Ga$_{0.7}$As barriers, uniform PL image and PL spectral peaks of QWRs along the wire direction were observed, which demonstrates the uniformity and the high quality of fabricated QWRs. Observed PL peaks are uniquely assigned to the constituent quantum structures. On the other hand, in the unsuccessfully fabricated 4-nm-scale In$_{0.09}$Ga$_{0.91}$As T-QWRs with Al$_{0.3}$Ga$_{0.7}$As barriers, homogeneous PL image and spectra were not obtained, proving that the anomalous overgrowth occurred in the unsuccessful CEO process. This difference in the micro-PL data shows the importance of flat cleaved surface and the optimized of overgrowth condition to fabricate uniform T-QWRs.

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14 The well thickness a and b are calibrated by comparing the PL data with calculated energy levels of QWs. The procedure will be shown in Ref. 17. Other thickness parameters are nominal values.
15 The well thickness b of the overgrowth QW in S-2 was designed as 4 nm. However, since the PL spectra did not show a distinct and spatially uniform peak from QW2 as described in Sec. III, the thickness b is not well-defined.