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Growth and characterization of InAs columnar quantum dots on GaAs substrate

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The growth of InAs columnar quantum dots (CQDs) on GaAs substrates by molecular beam epitaxy was investigated. The CQDs were formed by depositing a 1.8 monolayer (ML) InAs seed dot layer and a short period GaAs/InAs superlattice (SL). It was found that the growth of the CQDs is very sensitive to growth interruption (GI) and growth temperature. Both longer GI and higher growth temperature impact the size dispersion of the CQDs, which causes the broadening of photoluminescence (PL) spectrum and the presence of the additional PL peak tails. By properly choosing the GI and the growth temperature, CQDs including GaAs (3 ML)/InAs (0.62 ML) SL with period number up to 35 without plastic relaxation were grown. The corresponding equivalent thickness of the SL is 41 nm which is two times higher than the theoretical critical thickness of the strained InGaAs layer with the same average In composition of 16%. The increase of the critical thickness is partially associated with the formation of the CQDs. Based on a five-stack CQD active region, laser diodes emitting around 1120 nm at room temperature were demonstrated, indicating a high material quality. CQDs with nearly isotropic cross section (20 nm × 20 nm dimensions) were formed by depositing a 16-period GaAs (3 ML)/InAs (0.62 ML) SL on an InAs seed dot layer, indicating the feasibility of artificial shape engineering of QDs. Such a structure is expected to be very promising for polarization insensitive device applications, such as semiconductor optical amplifiers. © 2007 American Institute of Physics. [DOI: [10.1063/1.2764212](https://doi.org/10.1063/1.2764212)]

I. INTRODUCTION

Semiconductor quantum dots (QDs), created by epitaxial growth in Stranski–Krastanow (S-K) mode, have attracted considerable interest in the last two decades. The three-dimensional confinement potential in a single QD results in a discrete energy spectrum with δ -like density of states.^{1,2} They have already been extensively utilized in the electronic and optoelectronic devices^{3–8} including single electron transistors, lasers, superluminescent emitting diodes, infrared detectors, and single photon emitters, delivering an improvement in several device characteristics over conventional technology. Recently, development of a novel QD semiconductor optical amplifier (SOA) is expected to bring promising characteristics such as broadband amplification, high saturation output power, and ultrafast response.⁹ However, the optical gain in those QD SOAs is polarization dependent, which limits their applicability in general fiber communication since the polarization of the optical signals is random in the fiber. This polarization sensitivity could be resolved by polarization diversity configuration or large optical cavity waveguide structure,^{10,11} but it still remains necessary to investigate the possibility and method of controlling polariza-

tion properties in QDs themselves. Generally, the anisotropy of confinement potential and strain in a nanostructure provides a strong polarization dependence of the dipole moment. In the conventional S-K growth of QDs, the effects of strong electronic confinement in the growth direction and compressive strain, both related to the anisotropic dot shape, induce a splitting of the heavy-hole (HH) and light-hole (LH) states. The lowest energy transitions have then electron-HH characters, and couple to an in-plane transverse-electric (TE) polarization. Modification of the anisotropy in the QD system is expected to substantially improve the polarization sensitivity of a QD SOA.^{12–15} QD shape modification can be achieved by layer-by-layer closely stacking of QDs in forming columnar QDs (CQDs)^{12–14} and first evidence of polarization insensitivity of optical gain in the CQD SOA was reported recently.¹⁵ In this growth mode, a first layer of seed QDs is formed, followed by a short period GaAs/InAs superlattice (SL), with a growth interruption after each InAs layer. The in-plane strain distribution created by the first QD layer favors In incorporation on top of QDs,¹⁶ thereby resulting in an In-rich column-shaped nanostructure. According to the calculation based on the elastic continuum theory for the strain distribution and the eight-band $\mathbf{k} \times \mathbf{p}$ theory for the electronic structures,¹⁷ the increase of transverse-magnetic (TM) component in the CQDs is considered to be due to the

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enhanced HH-LH mixing because of a reduction of the biaxial strain in the central portion of the CQDs. The CQDs are thus promising for the polarization insensitive QD SOA, but there are few reports on the growth of this unique type of QDs.^{14,18–20} In this article, we report the growth of InAs CQDs on GaAs substrate by solid-source molecular beam epitaxy (MBE). The effects of growth parameters on the optical and structural properties of the CQDs were investigated by photoluminescence (PL), high-resolution x-ray diffraction (HRXRD) and transmission electron microscopy (TEM). Based on a CQD active region, laser diodes with emission wavelength of 1120 nm were fabricated, indicating a very high material quality.

II. EXPERIMENTAL PROCEDURE

The samples used in this work were grown on (001)-oriented GaAs substrate by using MBE. The growth procedure included a 500-nm-thick GaAs buffer layer grown at 600 °C, an array of CQDs formed by depositing a 1.8 monolayer (ML) InAs QD seed layer and a short period GaAs (3 ML)/InAs (x ML) superlattice grown at various temperatures ranging from 490 to 520 °C, and a 100 nm GaAs capping layer grown at the same temperature as the dot. After each growth of InAs layer, a growth interruption (GI), 5 s if not specified, was applied in order to make the QD size more uniform. The number of SL periods was varied from 0 (reference) to 50. The growth rate of GaAs and InAs were 0.7 and 0.1 ML/s and As₂ beam equivalent pressure was 1×10^{-5} Torr. The QD evolution was monitored *in situ* by reflection high-energy electron diffraction (RHEED).¹⁸ After growth of the 1.8 ML InAs QD layer, characteristic chevrons along the [0–11] direction are observed. They evolve into short and streaky diffraction rods during the growth of the 3 ML GaAs layers. When the subsequent InAs layers are grown, the RHEED pattern changed back and during GI clear chevrons redevelop. Generally, the redeveloped characteristic chevron is a very important hint in obtaining a high quality material. After growth, all the samples were characterized by room temperature (RT) PL measurements, and some of them were also characterized at low temperature (5 or 77 K). The PL was excited with a 632.8 nm He-Ne laser and measured by an uncooled InGaAs detector. The estimated excitation power density on the samples is about 200 W/cm².

III. RESULTS AND DISCUSSION

In order to confirm the formation of the CQDs, TEM results are first presented. The measurements were performed on the sample including a 16-period GaAs (3 ML)/InAs (0.62 ML) SL grown at 505 °C. Figure 1 shows (a) the [001] bright field plan-view, (b) micrograph and (c) magnified $g = \langle 002 \rangle$ dark-field cross-sectional TEM images, and (d) the In composition profiles across the center of the CQDs and quantum well (QW) layer. The technical details of the TEM experiment and derivation of the layer composition have been described elsewhere.^{21,22} From the plan-view, the size of the CQDs is very uniform and the dot density is about 1.7×10^{10} cm⁻². The CQDs have a parallelogram shape with

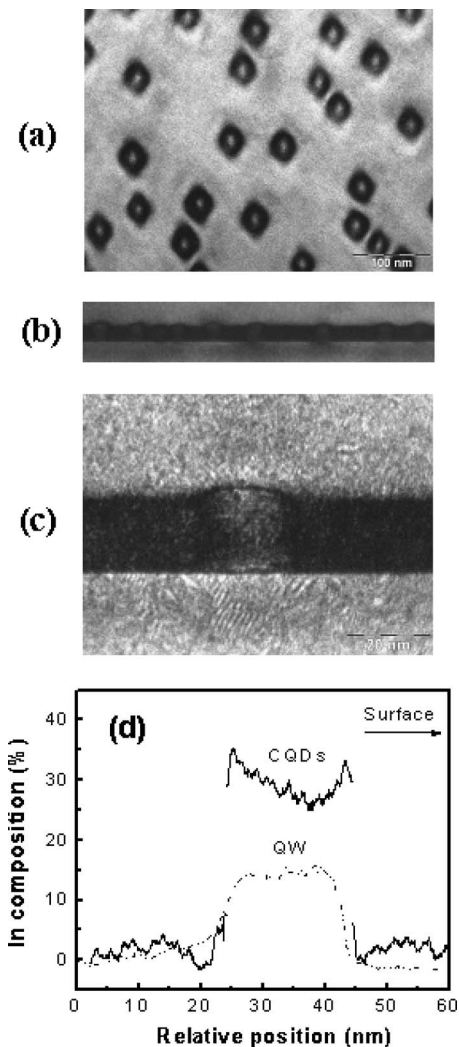


FIG. 1. TEM of the sample including a 16-period SL. (a) [001] Bright-field plan-view image, (b) micrograph and (c) magnified $g = \langle 002 \rangle$ dark-field cross-sectional images, and (d) In composition profiles across the center of the CQDs and QW layer.

the diagonals exactly along the $\langle 110 \rangle$ and $\langle 1-10 \rangle$ directions of the growth plane. The length of these two diagonals is anisotropic which can be explained in term of the $\{136\}$ model developed by Lee *et al.*²³ The average ratio between the long diagonal and the short one is 1.22 with a standard deviation of 0.05. From the observations of the cross-sectional images, the whole structure consists of two parts, i.e., the CQDs and a two dimension (2D) layer around them. The 2D layer consists of an InGaAs QW with an average thickness of about 20 nm. Its composition is uniform and has an average value of about 16%. The achieved high uniform In composition across the QW is mainly due to the In-Ga intermixing during the growth. Therefore, the whole structure is a QD in QW structure. Although the CQDs are slightly higher than the QW, the interfaces between GaAs and the QW layer are sharp and planar, no dislocation and QD plastic relaxation were observed, indicating an excellent material quality. The aspect ratio of the CQDs is close to one ($20 \text{ nm} \times 20 \text{ nm}$), which is promising for polarization insensitive device applications. However, the In composition across the center of the CQDs is not uniform, a vertical com-

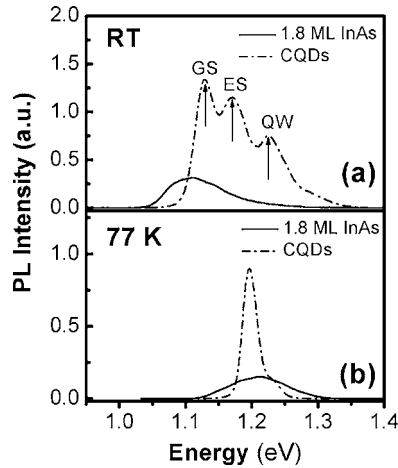


FIG. 2. (a) RT and (b) low temperature (77 K) PL spectra of the reference sample with 1.8 ML InAs QDs (no SL) and the CQDs including a ten-period GaAs (3 ML)/InAs (0.7 ML) SL grown at 500 °C.

position modulation is observed. The average In composition of 35% at the bottom decreases to 25% at around the 2/3 of the total thickness and then increases again to about 33% near the top of the structure. The obtained unique shape of the CQDs is very different from that of the conventional S-K growth of QDs, indicating the feasibility of artificial shape engineering of QDs. We suggest that even vertical InGaAs quantum wires or rods can be formed by optimizing the growth conditions and increasing the SL period number.

Figure 2(a) shows the RT PL spectra of the reference sample with 1.8 ML InAs QDs (no SL) and the CQDs including ten-period GaAs (3 ML)/InAs (0.7 ML) SL grown at 500 °C. The PL spectral width of 97 meV from the 1.8 ML InAs QD sample is very broad, which is a typical feature of the conventional S-K growth of QDs with InAs thickness just above the critical value.²⁴ The broad PL spectrum can be attributed to the large QD size inhomogeneity and to thermal population of close-spaced excited states (ES). When the 1.8 ML InAs QDs were capped by the GaAs/InAs SL, a well-developed PL spectrum with three distinct peaks along with a strong increase in PL intensity can be observed. The two peaks at around 1.127 and 1.169 eV originate from two bound states of the CQDs, i.e., ground state (GS) and the first ES. The 1.224 eV peak and its high energy tail are related to the transitions from the 2D QW surrounding the CQDs, which were confirmed by PL measurements of a control sample (data not shown). The observed strong emission from the QW originates mainly from the thermal population of the high 2D density of states. Figure 2(b) shows the PL spectra taken from the same samples at 77 K. PL spectral width of the CQD sample drastically decreases due to the suppressed thermal population of the QD ES and the QW, whereas the PL spectral width of the 1.8 ML InAs QDs is nearly the same as at RT. For the CQD sample, three PL peaks at RT evolve into a single GS peak with a high energy tail at 77 K. The PL spectral width is only about 26 meV, which is much smaller than the value of 97 meV obtained from the 1.8 ML InAs QDs and thereby demonstrates a relatively small inhomogeneous broadening. The small inhomogeneous broadening is related partly to the uniform size distribution evidenced by

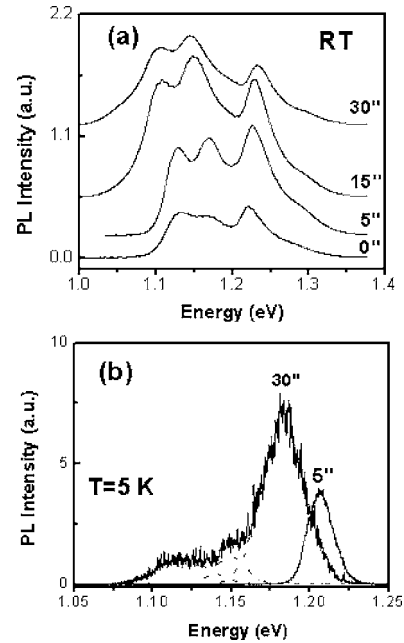


FIG. 3. (a) RT PL spectra of four CQD samples including a ten-period GaAs (3 ML)/InAs (0.7 ML) SL grown at 500 °C with different GI and (b) PL spectra taken from the samples with GI of 5 and 30 s under very low excitation power at 5 K.

TEM, and partly to the large QD dimensions and small confinement energy, which make the energy levels less sensitive to size fluctuations.

As one of the important growth parameters, GI affects the morphological and optical properties of the In(Ga)As/GaAs QD systems.^{25–27} The QD size can be controlled by GI and a uniform QD ensemble can be obtained by a proper choice of the GI time. To investigate the effects of GI on the CQDs, samples with different GI time were grown. Figure 3(a) shows the RT PL spectra of four CQDs including ten periods of GaAs (3 ML)/InAs (0.7 ML) SL. These samples were grown at 500 °C with nominally identical growth conditions except for the GI after each InAs layer growth. The GI was set to 0, 5, 15, and 30 s. Upon increasing the GI, the PL intensity of the samples first increases and then decreases; the PL peaks related with the CQDs redshift and a low-energy peak tail appears. The redshift of the PL peaks can be attributed to an increase of the average QD volume.^{28–30} The PL peaks related with the QW slightly blueshift, indicating that the thickness of the InAs layer in the QW partly reduces during the GI to provide material for the CQD growth.^{29,30} However, this transfer of InAs material from the QW may only play a minor role in achieving the larger PL peak redshift for the CQDs. The volume increase of the CQDs can be mainly ascribed to material exchange among different CQDs.²⁸ During the GI, a certain fraction of CQDs dissolve to enable the growth of other CQDs. This procedure will induce CQD size dispersion/multimodal distribution and consequently lead to a composite PL spectrum.^{29,30} The low-energy peak tail probably originates from the emission of larger size CQDs. To confirm this point, PL spectra were taken from the samples with GI of 5 and 30 s under very low excitation power at 5 K, as shown in Fig. 3(b). The PL peaks related with the QW cannot be observed any more for both

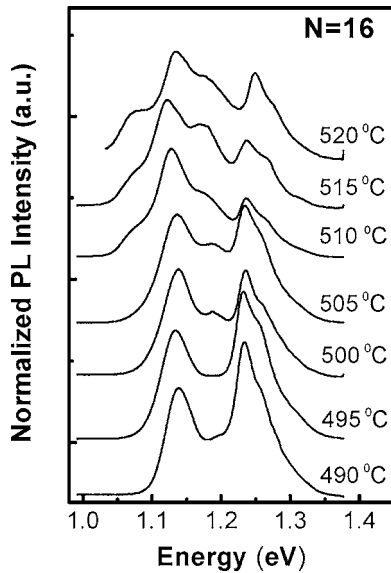


FIG. 4. Normalized RT PL spectra of the CQD samples with 16-period SL grown at different temperatures.

samples as discussed earlier. Only a single Gaussian peak with a narrow PL linewidth of 18 meV was observed from the sample with the GI of 5 s, indicating the size of the CQDs is very homogeneous. In contrast, a composite PL spectrum including several peaks from the sample with the GI of 30 s was observed. The composite PL spectrum can be fitted by three Gaussian peaks (dash line) peaked at 1.119, 1.15, and 1.183 eV, respectively. The multiple peaks are attributed to the emission from several families of CQDs with different sizes because at low temperature, carriers are highly localized and captured in the individual QDs.³¹ Thereby, in order to avoid dot size dispersion and improve the optical properties of the CQDs, a shorter GI should be used.

The temperature-dependent surface diffusion of the adatoms affects the structural and the optical properties of the QDs. In order to investigate these effects, a set of CQD samples were grown at different growth temperatures ranging from 490 to 520 °C. In these samples, the number of periods in the GaAs (3 ML)/InAs (0.62 ML) SL is 16 and the GI is 5 s. The reduced InAs thickness of 0.62 ML was used to increase the number of SL periods and consequently the aspect ratio of the CQDs. The RT PL spectra taken from these samples are shown in Fig. 4. They are normalized to the low-energy PL peak emitting from the GS of the CQDs. The PL peaks related with the CQDs slightly redshift with increasing substrate temperature up to 515 °C and then blue-shift at the growth temperature of 520 °C. The redshift can be attributed to the increasing size of CQDs which is the consequence of the increased diffusion length of the adatoms at higher growth temperature.³² The blueshift at 520 °C can be explained by the enhanced In desorption. A low-energy peak tail appears at higher temperatures, which is very similar to that of the samples grown with longer GI. In addition, the PL spectrum of the sample grown at higher temperature is broader than that of the sample grown at lower temperature. Indeed, the material exchange among different CQDs during the GI may be accelerated due to the increasing

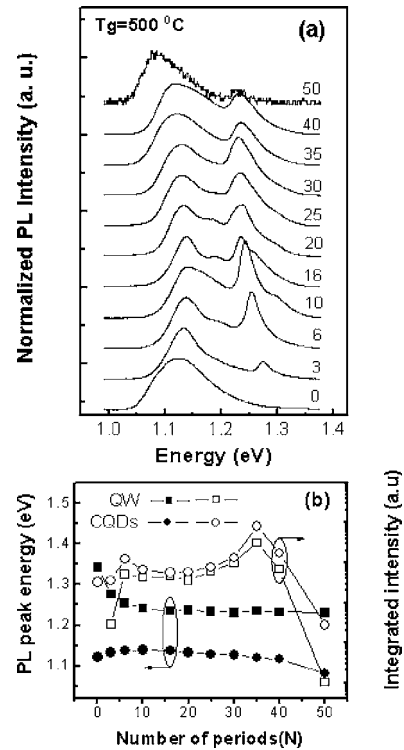


FIG. 5. (a) Normalized RT PL spectra of the CQD samples including different number of periods of the GaAs (3 ML)/InAs (0.62 ML) SL grown at 500 °C. The number of periods was varied from 0 (reference) to 50. (b) Variation of PL peak energies and integrated intensities from the QW and the CQDs as the function of the SL period number.

growth temperature, which induces dot size dispersion as discussed before. This explains the appearance of the low-energy peak tail and the spectral broadening at higher growth temperature.

The target of the CQD growth is to modify the shape anisotropy of conventional asymmetric QDs and consequently the PL polarization. Such modification can be realized by adjusting the GaAs/InAs SL period number. Indeed, the number of SL periods determines the height of the CQD. This control of the QD vertical dimension is unique to CQDs. With increasing period number, polarized emission along the growth direction (TM mode) overtakes the in-plane polarized emission (TE mode).¹² Kita *et al.* demonstrated that switching from TE to TM could be achieved with about nine periods of the SL.¹⁵ It is interesting to investigate the effects of SL period number. For this purpose, a series of CQD samples including a different number of periods in GaAs (3 ML)/InAs (0.62 ML) SL was grown at 500 °C. The number of periods was varied from 0 (reference) to 50. The RT PL spectra of these samples are shown in Fig. 5(a). They are normalized to the low-energy PL peak emitting from the GS of the CQDs. In the Fig. 5(b), the dependences of the PL peak energies and the PL integrated intensities on the number of periods are summarized. The PL peak energies and the PL integrated intensities of the QW and the CQDs were, respectively, derived from fitting of the experimental PL spectra with multiple peaks. With increasing number of periods, the PL peak related with the QW redshifts and then keeps nearly constant after a certain number, indicating the evolution of

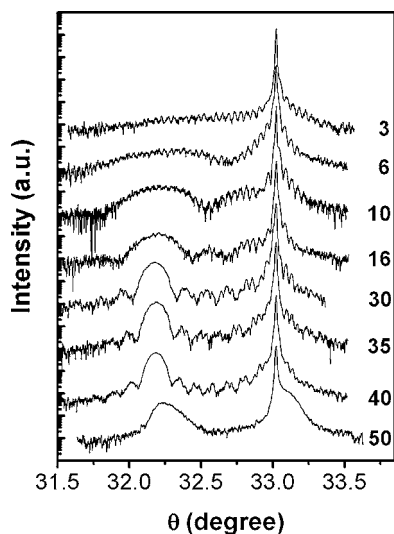


FIG. 6. HRXRD diffraction curves recorded near the (004) GaAs from the CQD samples including different number periods of the GaAs (3 ML)/InAs (0.62 ML) SL grown at 500 °C.

the SL equivalent thickness; whereas the PL peaks related with the CQDs first blueshift and then redshift systematically. The initial blueshift can be qualitatively understood according to the following simple model. Generally, In-Ga atoms intermixing and In segregation during the growth of In(Ga)As/GaAs material system play the very important roles in determining the In composition profile along the growth direction.^{33–37} For the growth of the 1.8 ML InAs QDs which are directly capped by the thick GaAs capping layer after 5 s GI, In-rich QDs are formed and consequently a longer emission wavelength results due to the smaller effects of the In-Ga atoms intermixing and the In segregation. For the CQD samples including the SL with a lower number of periods, the In-Ga atoms intermixing and the In segregation of the whole CQD structure are enhanced due to the increasing reaction time. The In composition of the initial In-rich QDs is decreased, thereby a PL peak blueshift from the CQDs is observed. However, with increasing the stacking number, the vertical electronic coupling in the system becomes more effective and the equivalent island height increases, and consequently the PL peak redshifts and the PL integrated intensity increases.³⁸ The PL energy shift from the samples including the SL with the number of periods less than 40 is small probably due to the low average In composition in the CQDs (as determined from TEM). As an exception, the abrupt redshift of the PL peak energy and the drastic decrease of the PL integrated intensity for the sample including the SL with a period number of 50 are attributed to the plastic relaxation probably due to the total equivalent thickness of the SL exceeding the corresponding critical value. This is confirmed by the following HRXRD measurements.

HRXRD diffraction curves recorded near the (004) GaAs from the earlier samples are shown in Fig. 6. The peaks on high-angle side ($\sim 33^\circ$) correspond to the diffraction from the GaAs substrate and the peaks on low-angle side ($\sim 32.2^\circ$) correspond to the composite diffraction from the CQDs and the QW. Given the small surface coverage of the CQDs (about 5%), the low-angle diffraction peak, in fact,

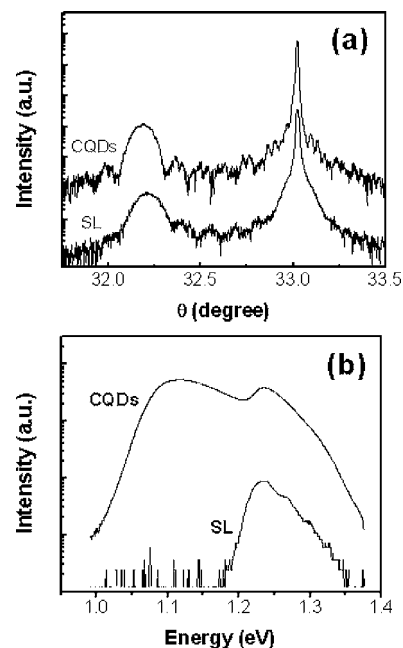


FIG. 7. (a) HRXRD diffraction curves and (b) RT PL spectra from the CQD sample including 35-period of the GaAs (3 ML)/InAs (0.62 ML) SL and the control sample with only 35-period SL.

mainly originates from the QW part. The absence of SL satellites and the gradually increasing diffraction intensity with the SL period number on the low-angle side confirm the formation of the uniform InGaAs QW layer, which is consistent with the TEM observations. The presence of numerous Pendellosung fringes at the high-angle side for the samples with the number of periods up to 35 indicates that the interfaces between the GaAs and the QW layer of these samples are sharp and planar.^{39,40} This feature is a strong evidence of high crystal perfection. For the samples with period number over 35, Pendellosung fringes gradually disappear, implying the onset of plastic relaxation of the QW. In particular, for the sample with 50 periods, the diffraction peak shifts abruptly to the high-angle side, clearly indicating plastic relaxation. From the angular difference between the low- and high-angle peaks, the average In composition and the equivalent thickness of the QW for these samples can be estimated by using dynamical x-ray diffraction theory. As examples, average In composition around 16% and equivalent thicknesses of 20 and 41 nm for the QW part of the CQD samples including 16- and 35-period SL were estimated. These values correspond very well with the TEM results. According to Matthews model,⁴¹ the critical thickness of a strained InGaAs layer with composition of 16% is 18 nm which is two times lower than the value of 41 nm achieved in the sample including a 35-period SL. The increase of the critical thickness is unusual and partially associated to the formation of the CQDs, although the mechanism is not clear. To confirm this point, a control sample with only 35-period SL and without the seed dot layer was grown. Figure 7 shows (a) the HRXRD diffraction curve and (b) the RT PL spectrum of the control sample. The spectra of the CQDs including 35-period SL are also shown for comparison. For the control sample, the disappearance of the Pendel-

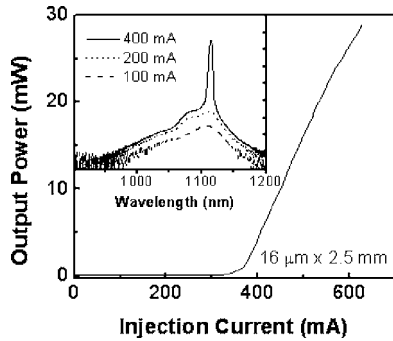


FIG. 8. Output power against drive current (L - I) characteristic from a $16\ \mu\text{m} \times 2.5\ \text{mm}$ CQD laser stripe under pulsed operation at RT. Inset shows the emission spectra (log scale) of the same device at different injection currents.

losing fringes at the high-angle side implies the inferior interface quality; the degradation of the PL efficiency implies the existence of non radiative recombination centers. Both of them are probably related to the production of the misfit dislocation or the occurrence of plastic relaxation.

To validate the device applications of the CQDs, a five-stack of CQDs including a ten-period GaAs (3 ML)/InAs (0.7 ML) SL was inserted into a conventional AlGaAs/GaAs waveguide structure with n - and p -type electrodes. A $16\ \mu\text{m} \times 2.5\ \text{mm}$ etched laser stripe with cleaved facets was tested under pulsed operation at room temperature. Figure 8 shows the output power against drive current (L - I) characteristic for such a device. The laser operates with a threshold current of 360 mA, corresponding to a threshold current density around $900\ \text{A}/\text{cm}^2$. The emission spectra of the same device at different injection currents are shown in the inset. Above threshold, the peak emission wavelength is 1120 nm, corresponding to the GS transition in the CQDs. The polarization behaviors of these devices have been addressed and the results will be presented elsewhere.

IV. CONCLUSION

In conclusion, we reported the growth of CQDs on GaAs substrate by MBE. It was found that the CQD growth is very sensitive to the GI and growth temperature. A longer GI and higher growth temperature induce size dispersion of the CQDs, which produce the broadening PL spectrum and the presence of additional PL tails. By properly choosing the GI and the growth temperature, CQDs including the GaAs (3 ML)/InAs (0.62 ML) SL with period number up to 35, corresponding to an equivalent height of 41 nm was grown, without plastic relaxation. This value is two times higher than the theoretical critical thickness of a strained InGaAs layer with the same In composition of 16%. The increase of the critical thickness is partially associated to the formation of the CQDs. Based on a five-stack CQD active region, laser diodes emitting around 1120 nm at RT were demonstrated, indicating a high material quality. CQDs with nearly isotropic cross section ($20\ \text{nm} \times 20\ \text{nm}$ dimensions) were formed by depositing a 16-period SL on an InAs seed dot layer. It is thereby expected that such a structure will be very promising for the polarization insensitive SOAs and specific applications requiring the controlling of the QD aspect ratio.

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