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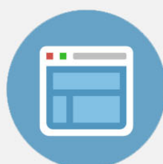

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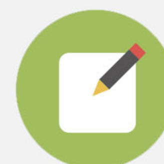

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The formation mechanisms and optical characteristics of GaSb quantum rings

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The growth mechanisms and optical characteristics of GaSb quantum rings (QRs) are investigated. Although As-for-Sb exchange is the mechanism responsible for the dot-to-ring transition, significant height difference between GaSb quantum dots (QDs) and QRs in a dot/ring mixture sample suggests that the dot-to-ring transition is not a spontaneous procedure. Instead, it is a rapid transition procedure as long as it initiates. A model is established to explain this phenomenon. Larger ring inner diameters and heights of the sample with longer post Sb soaking time suggest that As-for-Sb exchange takes places in both vertical and lateral directions. The decreasing ring densities, enlarged ring inner/outer diameters and eventually flat GaSb surfaces observed with increasing growth temperatures are resulted from enhanced adatom migration and As-for-Sb exchange with increasing growth temperatures. © *2013 AIP Publishing LLC*.
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I. INTRODUCTION

In the last two decades, a lot of effort has been devoted to the researches of quantum dots (QDs).[1–3] The unique optical characteristics and device applications of the nano-structure have been intensively studied. Among them, one of the most widely investigated structures is type-I InAs/GaAs QDs.[4–6] Different device applications such as 1.3 μm laser diodes (LDs), QD memory devices and QD infrared photodetectors (QDIPs) have been demonstrated.[7–9] The major advantage of the nano-structure for device applications lies on its Delta-function-like density of states such that low threshold current for LDs and normal incident absorption for QDIPs can be observed. However, for the memory device application, although the nano-structure does provide a superior carrier storage time compared with quantum-well structures, its type-I nature would be the major bottleneck for the practical application of QD memories. Therefore, it was proposed that type-II GaSb QDs can be an alternate choice for the storage medium of QD memories due to its separate electron and hole confinements and large valence band offset.[10] However, compared with the numerous publications of InAs QDs, only limited reports regarding GaSb QDs are published previously. The main reason responsible for this phenomenon is the difficult-to-control As/Sb interfaces such that defects are easily generated and therefore inferior optical and electrical characteristics are usually obtained for GaSb nano-structure.

In one of our previous publications, it has been demonstrated that by extending the post-growth Sb soaking time, significant photo-luminescence (PL) intensity improvement can be obtained for Gasb QDs.[11] Electro-luminescence (EL) of a GaSb QD light-emitting diode is also demonstrated at room temperature. The results suggest that the defects in the GaAs/GaSb interfaces have been greatly depressed with the long post-growth Sb soaking procedure. In the other publication, dot-to-ring transition is observed with the presentence of As atoms during the post-soaking procedure.[12] Similar quantum-ring (QR) structures formed after GaAs capping layer growth are also observed in literatures.[13,14] The results suggest that As-for-Sb exchange of the buried Gasb QDs with GaAs capping layer is the main mechanism responsible for the formation of GaSb QRs.[13] The optical characteristics of such structures are also observed.[15] Therefore, to maintain dot morphology for the buried GaSb QD layer, long-term post-growth Sb soaking procedure is proposed.[12] According to the observations in these publications and other earlier works,[16,17] although the buried GaSb QDs may transfer into QRs, it seems that the morphology of uncapped GaSb QDs can be well maintained. However, in yet another publication, uncapped GaSb QR structures are observed without the presence of As atoms during the GaSb deposition.[18] The results then led to the manipulation of full dot or ring morphologies by controlling the Sb/background As ratios during the post-growth Sb soaking procedure.[19] Superior PL intensities are also observed for GaSb QRs than QDs in the same paper. Although it has widely accepted that the optical recombination probability of type-II nano-structures is much lower than the type-I counterpart, compatible luminescence intensities of coupled GaSb QRs with InAs QDs are still observed with the improved electron confinement.[20] The results suggest that if interface defects are the main reason responsible the reduced luminescence intensities of the type-II nano-structures, the intense luminescence of GaSb QRs indicates a lower interface defect density for the structure. Therefore, more detailed investigations over GaSb QRs on its growth mechanisms and optical characteristics are essential for its practical applications.

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In this paper, samples with different Sb/background As flux ratios and time durations during the post Sb soaking procedure are investigated. The growth mechanism and the evolution progress of GaSb QRs are investigated based on the morphology changes of these samples. The PL spectra of the samples prepared at different growth temperatures are also demonstrated to verify the transition mechanisms responsible for the PL peaks.

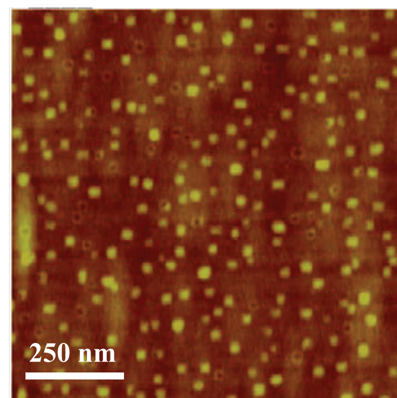
II. EXPERIMENTS

All the samples discussed in this paper are grown on (100)-orientated semi-insulated GaAs substrates by using Riber Compact 21 solid-source molecular beam eptaxy system (MBE). In this system, effusion cells with crackers and needle valves are adopted for Sb and As source to precisely control the atom flux. The cracker temperatures are set at 650 and 900 °C for As and Sb cells, which would result in the dominant group V materials to be As_4 and Sb_2, respectively. Samples with bi-layer structures are prepared to investigate the dot-to-ring transition and optical characteristics of GaSb QRs grown under different conditions. The sample structures are shown in Fig. 1. The growth procedure is as following: (a) 200 nm undoped GaAs buffer layer growth, (b) 15 s. Sb pre-soaking procedure to form 1 mono-layer (ML) GaSb on GaAs surface, (c) 2 ML GaSb deposition to form GaSb QDs, (d) post Sb soaking with controlled Sb/background As flux ratios, (e) 50 nm undoped GaAs separation layer growth, (f) repeat steps (b)–(d) to form the other GaSb layer for atomic-force microscopy (AFM) measurements. The 1 ML GaSb formed after the Sb pre-soaking procedure together with the deposited 2 ML GaSb would in total achieve 3 ML GaSb coverage on GaAs substrate. The GaSb QD growth rate and the V/III ratio are kept as 0.088 μm/h and 1.3, respectively. As have been discussed previously, QD, QR, or QD/QR mixture morphologies can be obtained with precise control of Sb/background As ratios.[19] Four samples with different Sb/background As ratios and post soaking times are prepared to investigate the formation mechanisms of GaSb QRs. Sample A is with the Sb/background As ratio 3.44 and post Sb soaking time 120 s. Samples B, C, and D are with Sb/background As ratio 0.25 and post Sb soaking time 60, 90, and 120 s, respectively. All the four samples are grown at 470 °C. To investigate the influence of growth temperatures on GaSb QRs, additional two samples with similar growth conditions as sample D are prepared at growth temperatures 495 and 520 °C, which are labeled as samples E and F, respectively.

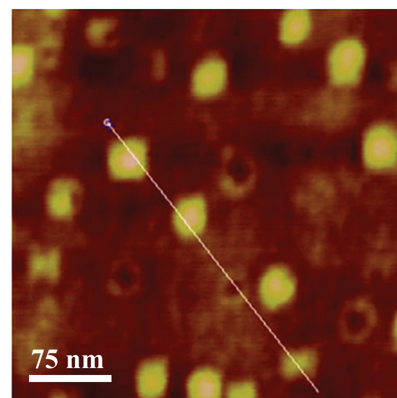
3 ML GaSb Nano-structure
50 nm undoped GaAs
3 ML GaSb Nano-structure
200 nm undoped GaAs
350 µm SI GaAs substrate

Samples	A	B	C	D	E	F
Growth Temperature (°C)						
	470	470	470	470	495	520
Post Sb soaking time (sec.)						
	120	60	90	120	120	120

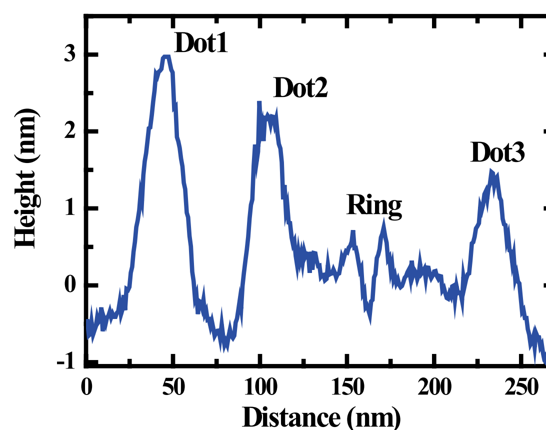
FIG. 1. The wafer structures of samples A–F. The Sb/background As flux ratios during the post Sb soaking procedure are 3.44 for samples A and 0.25 for rest of the samples.



(a)



(b)



(c)

FIG. 2. (a) The $1 \times 1\ \mu m^2$ and (b) $300 \times 300\ nm^2$ AFM image of sample A. (c) The surface profile of the line depicted on (b).

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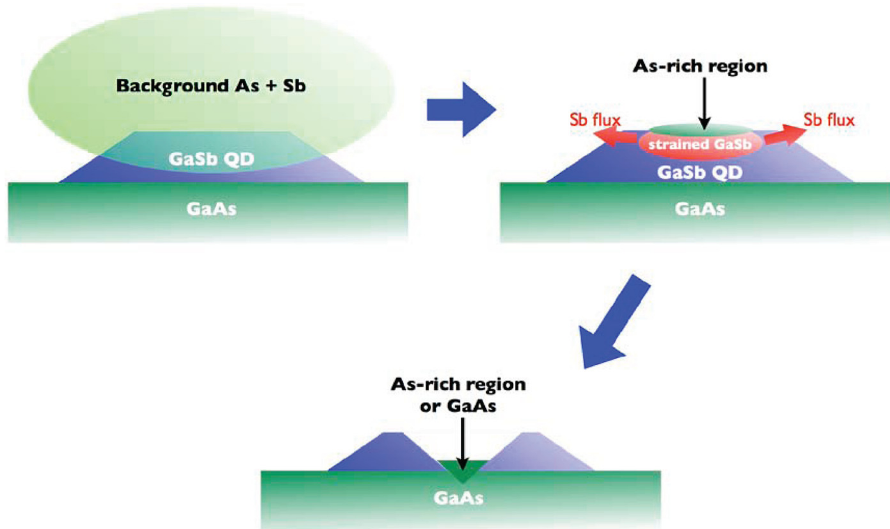

FIG. 3. A schematic figure describing the formation mechanisms of GaSb QRs.

The AFM measurements are performed by using Veeco Dimension ICON system. The PL measurements are performed by using the Jobin Yvon's NanoLog3 system coupled with a He-Ne laser as pumping source and a Janis cryostat to cool down the samples.

III. RESULTS AND DISCUSSIONS

As have been discussed previously, As-for-Sb exchange is the main mechanism responsible for the formation of GaSb QRs from QDs with the presence of As atoms.[12] Based on this concept, the manipulation of QD/QR morphologies through the control of Sb/background As ratios is demonstrated.[19] To investigate the evolution progress of GaSb QDs transferred to QRs, sample A with the morphology of QD/QR mixture is prepared. The $1 \times 1\,\mu\text{m}^2$ AFM image of the sample is shown in Fig. 2(a). As shown in the figure, around 40% GaSb QDs have been transferred to QRs. The results suggest that with the low background As pressure during the 120 s post Sb soaking procedure, not all GaSb QDs can be transferred to QRs. The phenomenon also indicates that the dot-to-ring transition may not be a spontaneous procedure. Instead, the procedure may require to surpass an activation barrier and would progress rapidly as long as it takes places. To further investigate this phenomenon, the zoom-in $300 \times 300\,\text{nm}^2$ AFM image of the sample is shown in Fig. 2(b). It is clear shown that there is a large height difference between QDs and QRs. The surface profile of the line depicted on Fig. 2(b) is shown in Fig. 2(c). As shown in the figure, the dot height is around 2–3 nm, while the ring height is only around 0.9 nm. The results suggest that although As-for-Sb exchange is the main mechanism responsible for dot-to-ring transition, the transition procedure is not a spontaneous procedure with constant As-for-Sb exchange rate.

To further explain the formation mechanisms of GaSb QRs, a model is established as shown in Fig. 3. During the Sb soaking processes, it is likely that the background As atoms condense onto the GaSb QD surface. Then, these As atoms aggregate and form an As-rich region on the summit of the QD. This region could be a small GaAs island, or As-induced surface reconstruction of GaSb (see the small green region atop the GaSb QD in the upper right panel of Fig. 3). Such an aggregation (or nucleation) process requires to overcome an activation energy barrier, and therefore, is not a spontaneous process. Then, such As-rich region will induce mismatch strain in the underlying GaSb QD (see the red region underlying the As-rich region in the upper right panel of Fig. 3), which will then force Sb atoms to diffuse away to relax the strain energy. This process will proceed until the QD transfers into QRs (lower panel in Fig. 3). Different from the previous publications of GaSb QRs formed after GaAs capping layer growth,[13,14] the growth method of GaSb QRs formed after post-Sb soaking procedure provides an alternate approach for GaSb QR preparation. This method provides a possibility of controlling the morphologies of GaSb nano-structures during MBE growth. However, more detailed investigations are still required in the future to fully understand the formation mechanism of GaSb QRs achieved by controlling Sb/background As flux ratios.

Since As-for-Sb exchange is still the main mechanism responsible for dot-to-ring transition during the post Sb soaking procedure, decreasing Sb/background As ratios is a reasonable approach to reduce the average dot-to-ring formation time and achieve full-ring morphology. According to our experience, with Sb/background As ratios reduced to 0.25 during the post Sb soaking procedure, the average ring formation time would be reduced to <60 s. Therefore, samples B, C, and D with Sb/background As ratio 0.25 and post Sb soaking time 60, 90, and 120 s are prepared to investigate the following evolution of GaSb QRs after its formation. The $1 \times 1\,\mu\text{m}^2$ AFM images of the samples are shown in Fig. 4(a). As shown in the figure, QR morphologies are observed for all the three samples. The results indicate that with well controlling Sb/background As ratios, QRs morphologies can be obtained when the post Sb soaking time exceeds 60 s. The statistic analysis of ring height, inner diameter, and outer diameter of the three samples extracted from the AFM images are shown in Fig. 4(b). Although the QR sizes are similar for the three samples, the values of ring heights and inner diameters are positively dependent on the soaking time durations, while no similar trend is observed for the outer diameter. As discussed in the previous publication, when do-to-ring transition

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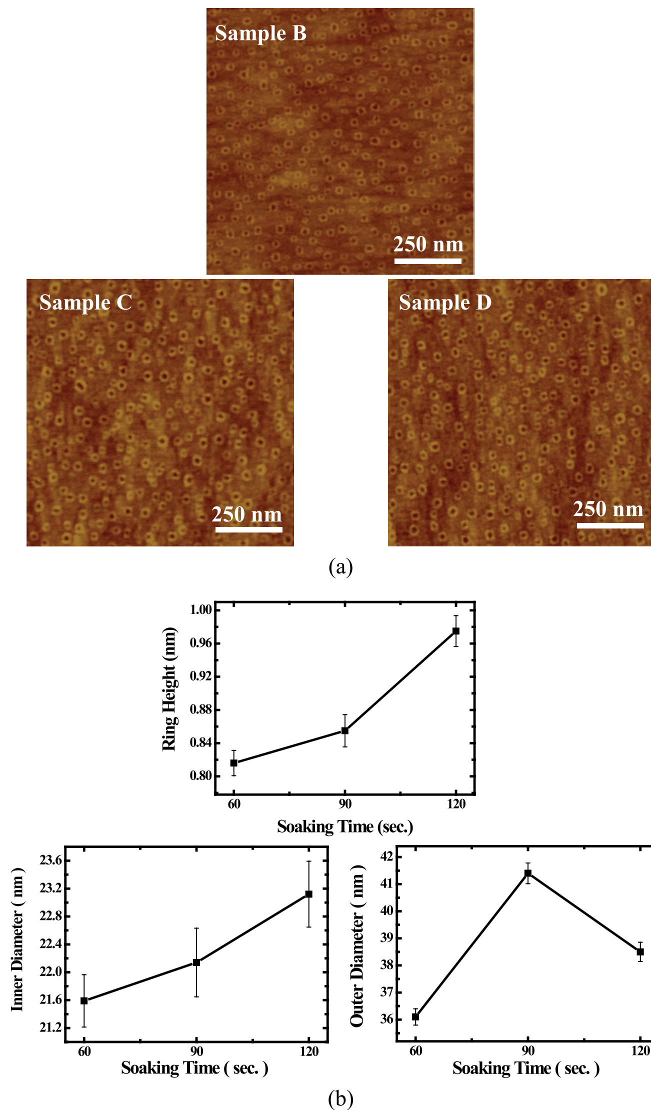

FIG. 4. (a) The $1 \times 1\,\mu m^2$ AFM images and (b) The corresponding ring heights (top), inner diameters (bottom-left) and outer diameters (bottom-right) of samples B, C, and D.

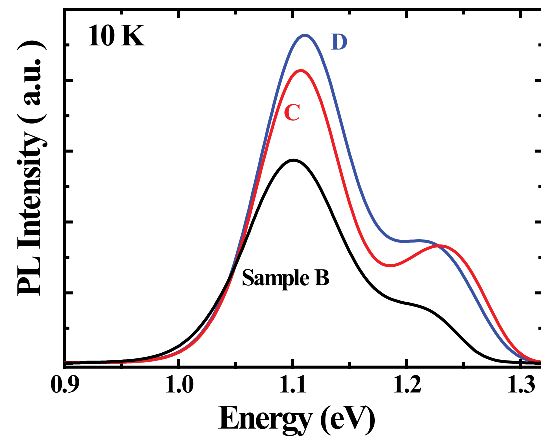

FIG. 5. The 10 K PL spectrums of samples B, C, and D.

initiates, the formation of QRs are dominated by the intense As-for-Sb exchange and following Sb out-diffusion on the QD summit.[12] In this case, the values of QR outer diameters should be determined by the QD diameters before ring formation. According to these results, the increasing inner diameters and heights of GaSb QRs with increasing post soaking time durations suggest that the As-For-Sb exchange happens on both vertical and lateral directions after ring formation. Although the exchange rate on the vertical direction is not as fast when dot-to-ring transition initiates, it is still be faster than in the lateral direction. The phenomenon may be resulted from the different binding energies of Sb atoms on (100) and (111) facets such that different As-for-Sb exchange rates would be obtained after QR formation.

Since full ring morphologies are obtained with the Sb/background As flux ratios 0.25 and post Sb soaking times longer than 60 s, the optimized Sb soaking time duration should be determined by the optical characteristics of the samples. The 10 K PL spectrums of samples B, C, and D are shown in Fig. 5. The phenomenon of PL peak blue shift with increasing pumping laser powers is also observed for the three samples, which confirm that the measured PL signals are from the type-II GaSb QRs.[19,20] As shown in Fig. 5, the PL intensities increase with increasing post soaking time. A possible mechanism responsible for this phenomenon is the less abrupt GaSb/GaAs interfaces resulted from the severe As-for-Sb exchange.[19] In this case, higher optical recombination probability will be obtained due to the increasing electron-hole wave function overlapping in the type-II hetero-structures. Also shown in the figure are two different PL peaks for the three samples located at ~1.1 and ~1.2 eV, respectively. The lower-energy PL peaks are attributed to the luminescence of the GaSb QRs. For the higher-energy ones, the luminescence was attributed to the GaSb WL.[17] To verify these attributions, samples E and F with higher growth temperatures are prepared.

The $1 \times 1\,\mu m^2$ AFM images of samples D, E, and F are shown in Fig. 6(a). As shown in the figure, with increasing growth temperatures, the ring densities reduce from 1.88 to 0.44×10^{10} cm^2 for samples D and E. For sample F, a flat surface without QRs on it is observed. Also observed are the larger QRs for sample E. It has been discussed previously that the QR outer diameters are determined mostly by the QD diameters. So, the larger QRs observed for sample E than sample D are simply attributed to the enhanced Ga and Sb adatom migration on the surfaces with increasing growth temperatures such that larger QDs are obtained. In this case, the outer diameter of GaSb QRs would be increased. The average QR outer diameters of samples D and E are 38.5 to 48.4 nm, respectively. The other phenomenon observed for the GaSb QRs is the same increase of inner diameters from 23.1 to 29.3 nm with increasing growth temperatures. The results suggest that with increasing growth temperatures, the enhancement of adatom migration would result in larger QDs and therefore larger QRs. The same high growth temperature parameter would also enhance As-for-Sb exchange and results in a larger ring inner diameter. Combined with these two mechanisms, with increasing growth temperatures, larger QRs with lower densities would be observed. However, with further increasing the growth temperatures, the As-for-Sb changes will eventually flatten the GaSb QRs

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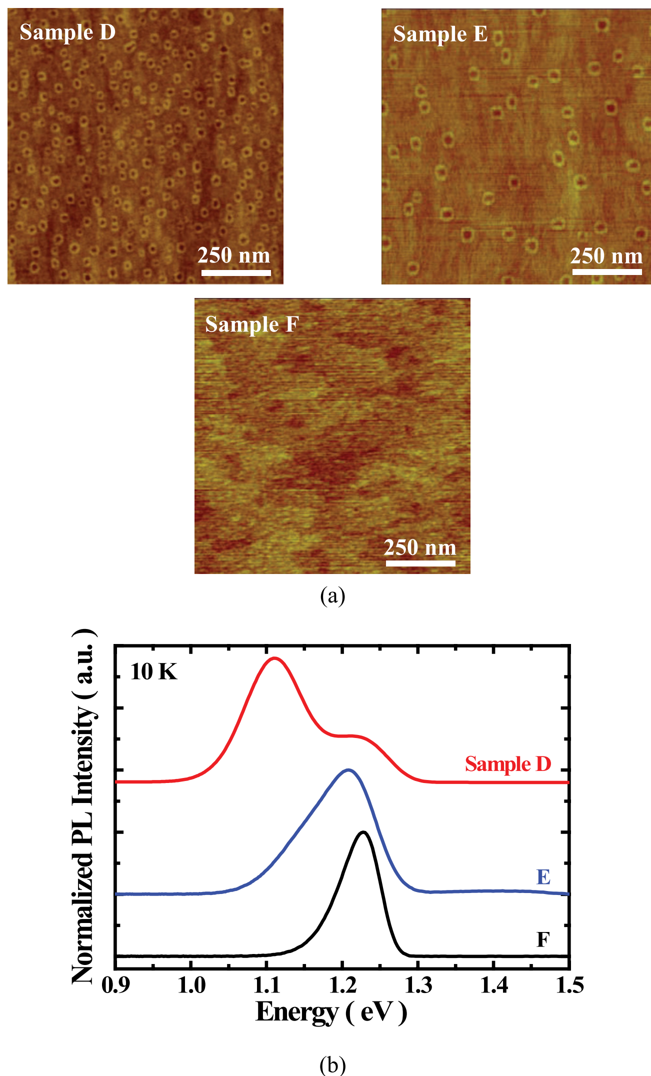

FIG. 6. (a) The $1 \times 1\,\mu m^2$ AFM images and (b) The 10 K PL spectrums of samples D, E, and F.

and, only GaSb WLs are remained on GaAs substrates as the case of sample F.

The 10 K PL spectrums of samples D, E, and F are shown in Fig. 6(b). As shown in the figure, the dominate PL peaks gradually change from the lower-energy one to the higher-energy one with increasing growth temperatures. The results suggest that PL peaks resulted from both QRs and WLs co-exist in the PL spectra. For sample D, the dominant PL peak should be resulted from the GaSb QRs and therefore, the lower-energy PL peak is attributed to the recombination of electrons in the GaAs layers and holes in GaSb QRs. For sample E, since the QR density reduces, the dominant PL peak should change to WLs. In this case, the higher-energy PL peak is attributed to the recombination of electrons in the GaAs layers and holes in GaSb WLs. The supporting evidence comes from sample F. Since there is no QR in the structure, the remaining PL peak could only be from the WLs. The observation of WL PL peak of sample F also confirms that although QRs are removed at high growth temperatures, the GaSb WLs still remain on the GaAs surfaces.

IV. CONCLUSIONS

In conclusion, the growth mechanisms and optical characteristics of GaSb QRs are investigated. Significant height difference between GaSb QDs and QRs in a dot/ring mixture sample suggests that the dot-to-ring transition is not a spontaneous procedure. Instead, it is a rapid transition procedure as long as it initiates. Through the observations over the PL spectra of the samples grown under different growth conditions, the transition mechanisms responsible for the each PL peak are verified in this paper. With the understandings over the growth mechanisms and evolution of GaSb QRs, the unique dot-to-ring transition observed for the GaSb nanostructures is further investigated. The knowledge over the growth mechanisms and optical characteristics of GaSb nano-structures are advantageous for their future applications in optical devices.

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