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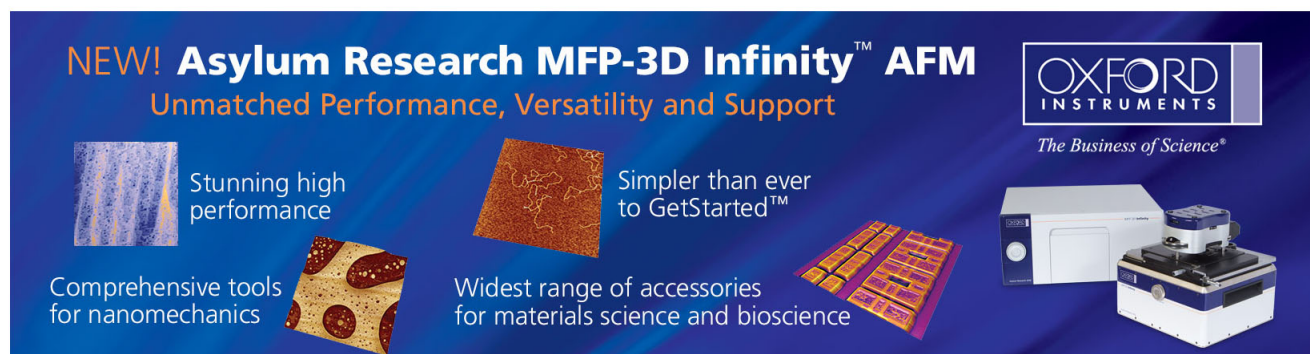
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Type-II GaSb/GaAs coupled quantum rings: Room-temperature luminescence enhancement and recombination lifetime elongation for device applications

Wei-Hsun Lin,¹ Kai-Wei Wang,² Shu-Wei Chang,³ Min-Hsiung Shih,³ and Shih-Yen Lin^{3,a)}

¹*Institute of Electronics Engineering, National Tsing Hua University, Hsinchu 300, Taiwan*

²*College of Photonics, National Chiao-Tung University, Tainan 711, Taiwan*

³*Research Center for Applied Sciences, Academia Sinica, Taipei 11529, Taiwan*

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Type-II GaSb/GaAs coupled quantum rings have exhibited two-order-of-magnitude luminescence enhancement and ten-times elongation of recombination lifetime at room temperature as compared with regular rings. The longer lifetime suggests that a significant amount of electrons are confined in coupled rings rather than simply leaking away. These phenomena indicate that type-II nanostructures can be potentially utilized for room-temperature luminescence and carrier storage applications. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4737443>]

It is generally accepted that compared with type-I hetero-structures, the radiative recombination in type-II hetero-structures is less efficient. Applications of type-II structures in light-emitting devices (LEDs) such as type-II quantum cascade lasers^{1,2} are therefore not as common as their type-I counterparts. Under such circumstances, the more emphases on type-II nanostructures such as GaSb/GaAs quantum dots (QDs) are focused on applications in memory devices.^{3,4} As expected, photoluminescence (PL) spectra of GaSb/GaAs QD samples are mostly observable at low temperatures in literatures due to the intrinsic separation of electrons and holes in type-II structures.^{5,6} Nevertheless, in our recent works,^{7,8} the clear room-temperature PL and electroluminescence (EL) from GaSb QDs, of which the morphologies are maintained with the procedure of long post-growth Sb soaking, were observed. It has also been demonstrated that by controlling the ratio between Sb and background As flux during the post-growth Sb soaking procedure, either dots or rings can be formed.⁹ The formation mechanism and the growth procedure of the GaSb quantum rings (QRs) discussed in this manuscript are quite different from the previous report.¹⁰ Moreover, GaSb QDs exhibit the more intense PL than QDs do. The blue shifts of PL/EL peaks with the increasing pumping power/injection current confirm that the luminescence indeed originates from the radiative recombination around type-II QRs. These results indicate that with well-controlled GaSb/GaAs interfaces, photonic applications based on the luminescence from type-II nanostructures at room temperature are plausible. Therefore, in addition to memory devices, if the luminescence is further enhanced, the type-II nanostructures can be promising for LED applications with unique characteristics distinct to those of type-I counterparts.

In this paper, with two GaSb layers separated by a thin GaAs layer, we observe the self-aligned growth of QRs formed in the two GaSb layers, namely, coupled QRs. The sample exhibits over two-order-of-magnitude enhancement in the PL intensity and about ten-time elongation of the over-

all decay time (nanosecond range) in the time-resolved PL (TRPL) when compared to the standard QR sample (uncoupled QRs). The phenomena suggest that the enhanced PL intensity may not be completely due to the increase of radiative recombination probability. The more significant blue shifts in PL spectra of coupled QRs than those of the standard ones in the same span of pumping power indicate that more electrons can contribute to the radiative recombination without rapidly escaping from the type-II structure in the former case. Therefore, the amount of electron-hole pairs in coupled QRs may greatly outnumber that in standard rings, which brings about significant differences between room-temperature PL intensities of these two type-II nanostructures. This picture is also consistent with the longer decay time of the coupled ring structure in the TRPL measurement. Since the time-dependent intensity (averaging over all spectral components) of TRPL is a measure of the overall decay rate (radiative/nonradiative recombination and leakage) of carriers around the type-II structure, the much shorter decay time of the standard ring structure reflects the fast escape of electrons near each independent QR. On the other hand, a decay time of a few nanoseconds for coupled QRs is closer to the timescale of radiative recombination in generic type-II nanostructures.

The samples investigated in this paper are grown on (100)-orientated semi-insulating GaAs substrates using the Riber C21 solid-source molecular beam epitaxy system. In this system, effusion cells with crackers and needle valves are adopted for both the As and Sb sources. Two samples with standard and coupled GaSb QR layers are prepared, which are referred as samples A and B, respectively. Additional uncapped QR layers are grown on the two samples for atomic-force-microscope (AFM) measurements. For sample A, there are three single GaSb QR layers while for sample B, three periods of two GaSb QR layers separated by a 5 nm GaAs layer are prepared. The sample structures are shown in Table I. To form the GaSb QR structure, the post-growth soaking with a specific ratio of Sb and background As is carried out after the pre-growth of GaSb QDs. Details on the growth procedures of GaSb QRs can be found in Ref. 9. The

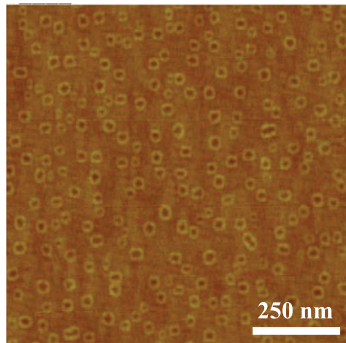
^{a)}Electronic mail: shihyen@gate.sinica.edu.tw.

TABLE I. The wafer structures of samples A and B.

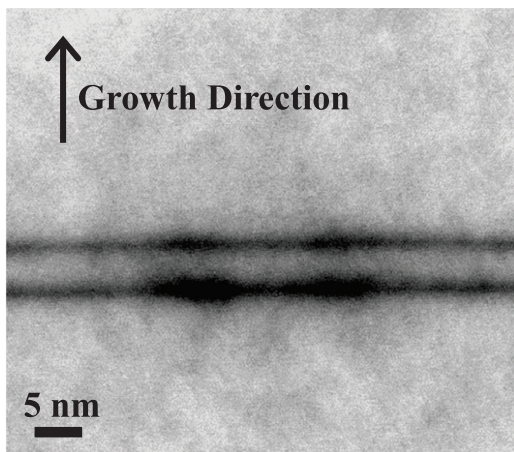
Sample	A	B
	3.0 ML GaSb/GaAs QR	
	40 nm GaAs	
	10 nm GaAs	3.0 ML GaSb QRs
		5 nm GaAs
	3.0 ML GaSb QRs	3.0 ML GaSb QRs
	200 nm undoped GaAs	
	350 μm GaAs S-I substrate (001)	

cross sectional scanning transmission electron microscopy (STEM) is performed with the Jeol 2100F system. The TRPL measurements are taken with a picosecond pulsed laser diode at a wavelength of 860 nm and the picoharp 300 photon counting system.

The $1 \times 1 \mu\text{m}^2$ AFM image of sample A is shown in Fig. 1(a). From this image, the clear ring morphologies are present. The average height and outer diameter of rings are 1.5 and 46.7 nm, respectively. The result indicates that by well controlling the ratio between Sb and background As during



(a)

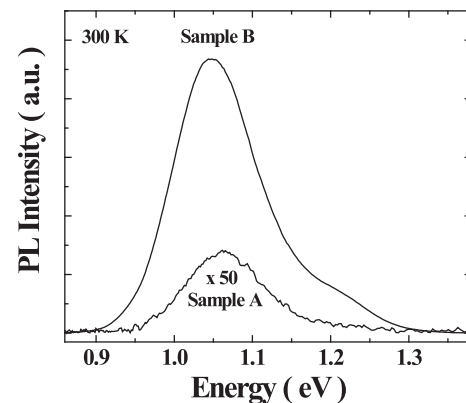


(b)

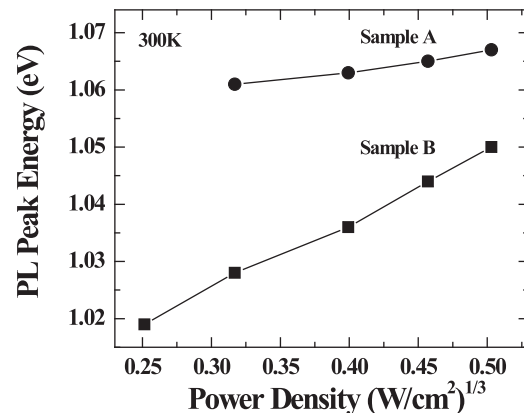
FIG. 1. (a) The $1 \times 1 \mu\text{m}^2$ AFM images of sample A and (b) the STEM image of sample B taken in bright-field condition.

the post-growth soaking procedure, GaSb QDs can be completely transformed into QRs. The cross sectional STEM image of sample B taken in the bright-field imaging condition is shown in Fig. 1(b). In this cross sectional view, one QR in the upper GaSb layer is self-aligned with the other in the lower layer. This phenomenon indicates that the 5 nm GaAs layer does not completely relax the strain of the underneath QRs so that QRs in the upper layer are preferentially formed right above the lower ones. In addition, the unit cells of sandwiched GaAs regions may be also expanded by the larger cells of GaSb nanostructures and become tensile-strained. The two QRs together turn into a pair of coupled rings. A similar phenomenon is also present in InAs QDs.¹¹ The heights/outer diameters of the upper and lower QRs in Fig. 1(b) are 1.6/21.9 and 2.5/25 nm, respectively. Compared with the values obtained from the AFM image, outer diameters of the two rings in the STEM image are smaller, which is attributed to the overestimation in AFM measurements.¹²

The room-temperature PL spectra of samples A and B at a pumping intensity of 0.5 W/cm^2 are shown in Fig. 2(a). The lineshapes and peak energies of the two PL spectra are similar at this pumping level. However, the PL intensity of sample B is about two orders of magnitude higher than that of sample A. The similar PL lineshapes of the two samples



(a)



(b)

FIG. 2. (a) The room-temperature PL spectra at a pumping intensity of 0.5 W/cm^2 , and (b) the PL peak energies of samples A and B as a function of the pumping intensity.

suggest that although strain-induced self-aligned growth is present in sample B, the energy distribution of electronic states due to size, shape, and composition fluctuations in coupled QR layers may remain close to that in uncoupled ones. Nevertheless, each pair of GaSb coupled rings provides three-dimensional barriers for electrons in the sandwiched GaAs layer, namely, coupled rings effectively function as depositories for carriers and maintain a decent level of carrier density around the nanostructures. In this way, even though the radiative recombination probability of each electron-hole pair may not increase significantly, the more electron-hole pairs tremendously enhance the PL intensity from sample B. To verify this attribution, the PL peak energies of samples A and B as a function of the pumping intensity are shown in Fig. 2(b). Sample A has the higher peak energies than those of sample B, indicating that the GaAs unit cells around coupled QRs may be more tensile-strained than those around standard QRs. For both samples, the blue shifts of PL peak energies are proportional to the cube root of pumping intensities, which confirms that the luminescence indeed originates from type-II transitions.¹³ On the other hand, the larger energy blue shift of sample B than that of sample A in the same pumping power span is observed. According to the theoretical prediction, if more carriers are present in the type-II hetero-structure, the larger Coulomb force between electrons and holes steepens the confinement potential, which increases energy differences between electron and hole states and leads to the more prominent blue shift in the PL peak energies.¹³ Therefore, the larger PL peak blue shift of sample B suggests an increasing number of electrons accumulated near coupled QRs.

Both the PL intensity and blue shifts in the peak energies are indicative of the increased electron accumulation in coupled QRs at room temperature. The difference of carrier accumulations between the standard and coupled QRs can be understood from the band diagrams around the two nanostructures shown in Fig. 3. From the band diagram, for standard QRs at room temperature, the leakage time τ_{leak} due to various scattering and nonradiative processes is significantly shorter than the radiative recombination lifetime τ_{rad} so that electrons tend to escape from QRs and leak into the GaAs continuum states thermionically. As a result, the smaller number of electrons around regular QRs brings about the weaker room-temperature PL intensity for Sample A. As for the coupled QR in Fig. 3(b), the tensile strain in the GaAs region sandwiched between two aligned GaSb rings may shrink the bandgap energy and lower the potential experienced by electrons there. As a result, a bound state absent in standard QRs emerges due to the enhanced confinement potential in the conduction band. Electrons in this bound state are well confined inside coupled QRs and stay there much longer than those outside the nanostructures do. Since the leakage and thermalization of carriers in this bound state are much less significant, the recombination is dominated by the radiative process and characterized by the radiative lifetime $\tau_{\text{rad,b}}$ of the bound state. This picture is consistent with the lower peak energy of the PL spectrum from sample B than that from sample A due to the presence of tensile strain in the sandwiched GaAs region, and a detailed modeling examining this viewpoint is under investigation.

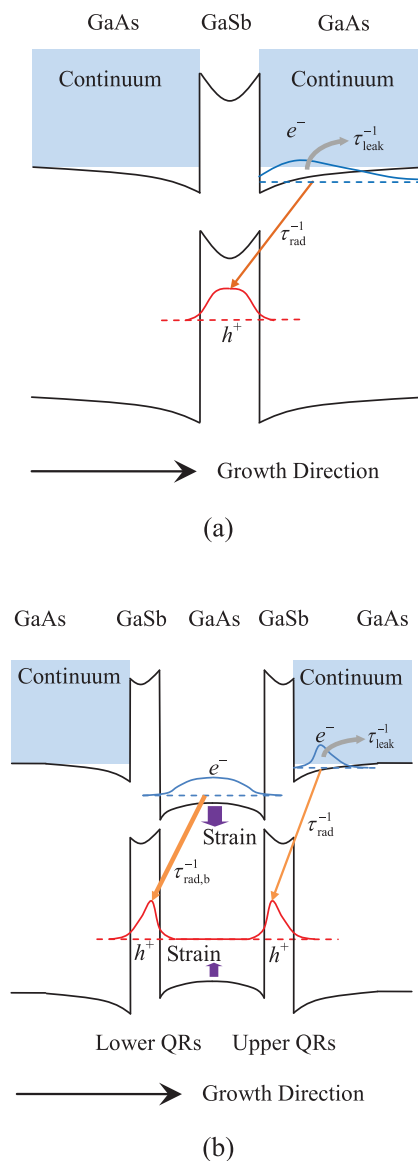
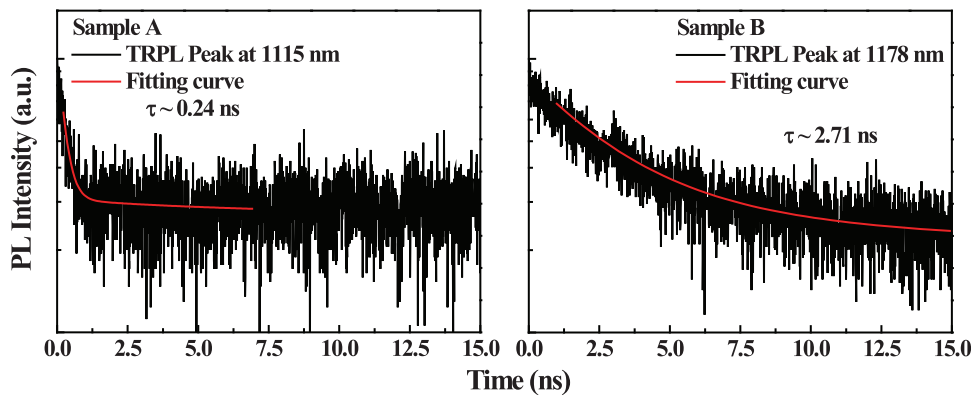
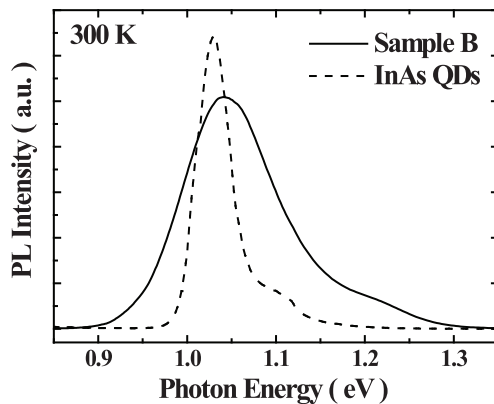


FIG. 3. (a) The band diagram near a standard QR and (b) the band diagram near a pair of coupled QRs at room temperature.

From the above scenario, the overall decay time of TRPL measurements would qualitatively reflect differences of carrier dynamics between the standard and coupled GaSb QRs. Since electrons and holes are confined at different locations, the longer radiative recombination lifetimes of type-II hetero-structures (above the nanosecond range) than those in type-I structures are always expected. The TRPL decay traces of the two samples at room temperature are shown in Fig. 4(a). Assume that the decay traces follow a single exponential function $\exp(-t/\tau)$ of time t , where τ is the overall PL decay time which reflects the quench rate of carriers around type-II nanostructures.¹⁴ The decay times τ of samples A and B are 0.24 and 2.71 ns, respectively. A decay time in the nanosecond range for sample B suggests that a significant portion of the recombination in coupled QRs remains radiative and type-II in nature at room temperature. On the other hand, the 10-time shorter decay time of sample A than that of sample B indicate that other mechanisms such as thermalization, leakage/diffusion away from nanostructures, and nonradiative recombinations, dominate over the



(a)



(b)

FIG. 4. (a) The decay traces of room-temperature TRPL for samples A and B, and (b) the room-temperature PL spectra of single-layer InAs QDs (2.4 monolayers) and sample B.

radiative recombination of standard QRs at room temperature. For luminescence applications, although the radiative recombination probability of coupled QRs may not be significantly enhanced, the better-confined bound state of electrons due to the tensile-strained GaAs inside GaSb coupled QRs improves the luminescence of the type-II nanostructure. The direct comparison between room-temperature PL spectra of a generic sample of single-layer InAs QDs (2.4 monolayers) and sample B in Fig. 4(b) demonstrates that the luminescence intensity of type-II nanostructures can be comparable to that of type-I counterparts.

In conclusion, we have experimentally demonstrated that the self-aligned growth of type-II GaSb/GaAs QRs tremendously increases the luminescence compared with standard QRs. An enhancement over two orders of magnitude is observed in PL intensities from the two nanostructures. The improved luminescence may originate from the electron bound state due to the tensile-strained GaAs inside coupled QRs. The compatible PL intensities of type-II coupled GaSb QRs and type-I InAs QDs have exhibited potential applications of such structures in LEDs. The recombination lifetime elongation of coupled QRs is advantageous for the memory device application.

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