Optical investigations of the dynamic behavior of GaSb/GaAs quantum dots

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Time-resolved radiative recombination measurements on GaSb quantum dots have been performed. The GaSb quantum dots are grown by molecular beam epitaxy on (100) GaAs through a self-assembly process. Time-resolved measurements show that, after a rapid hole capture process, the photoluminescence decays with a fast and a slow component. The fast component is shortened significantly with higher excitation intensity while the slow component is roughly constant. The radiative lifetimes are much longer than the lifetimes of ordinary GaSb quantum wells with a straddling band lineup. These results support a staggered band lineup and space charge induced band-bending model. © *1996 American Institute of Physics.* [S0003-6951(96)04611-4]

Zero-dimensional (0D) quantum dot (QD) structures have attracted great interest in recent years due to their δ-function-like density of states, strong carrier localization, increased exciton binding energies, and enhanced oscillator strength. Most of the QD research[1–3] has focused on material systems with a straddling (type I) band lineup, with spatially direct transitions. On the other hand, a staggered (type II) band lineup heterojunction accommodates spatially indirect transitions. This will drastically influence optical and electrical properties[4,5] and their fundamental features need further studies.

In this letter, we report time-resolved photoluminescence measurement on staggered-lineup QDs. We choose the GaSb/GaAs system because we can create highly strained and staggered-lineup heterostructures composed of direct-gap materials.[6] At the GaAs-GaSb interface, a large valence band offset of 0.81 eV has been suggested[7] which results in strong hole wavefunction localization in the GaSb region. This large valence band offset implies a staggered band lineup at the GaSb/GaAs interface accompanied by a spatially indirect radiative recombination process[7] [Fig. 1(a)]. Recently, Hatami *et al.* published photoluminescence (PL) measurements on GaSb/GaAs quantum dots.[8] Their data support a staggered-lineup band structure. However, no time-resolved PL has been reported, which would provide more direct information on the nature of the radiative recombination and on a possible phonon bottleneck effect[9] for hole capture processes.

The GaSb QDs studied in this paper were grown by molecular beam epitaxy (MBE). The surface morphology was monitored *in situ* during the growth by a reflection high energy electron diffraction (RHEED) facility. In this letter we report on two different sample structures, referred to as A and B. They were both grown on semi-insulating GaAs (100) substrates after depositing a 300 nm GaAs buffer layer grown at a substrate temperature of 600 °C.

On sample structure A, GaSb was deposited at 530 °C directly on the GaAs buffer layer. Monitoring the RHEED reconstruction pattern, there appeared, in addition to a pure streak pattern indicating the two-dimensional growth of the GaAs layer underneath, a spotty pattern with a larger lattice constant indicating that the deposition of the GaSb was taking place in a 3D growth mode. The growth was interrupted after 1.4 monolayer deposition. In order to prepare the samples for the photoluminescence (PL) measurements, we overgrew the dot layer with a 350 nm GaAs cap. High overgrowth temperatures (>500 °C) resulted in immediate disappearance of the spotty RHEED pattern, even when exposing the surface to a very low As flux. This was not observed at lower overgrowth temperatures. The growth temperature was thus kept at 460 °C to avoid a possible replacement of the antimony by the arsenic at the dot interfaces. During the overgrowth, the spotty pattern in the RHEED reconstruction vanished slowly and disappeared only after the dots were completely capped with GaAs.

On sample structure B (depicted in the inset of Fig. 2), the GaSb dot layer was deposited in the center of a 15 nm GaAs quantum well (QW) with 200 nm thick $Al_{0.5}Ga_{0.5}As$ barriers on both sides. Similar to structure A, the RHEED indicated a formation of the GaSb dots during the GaSb

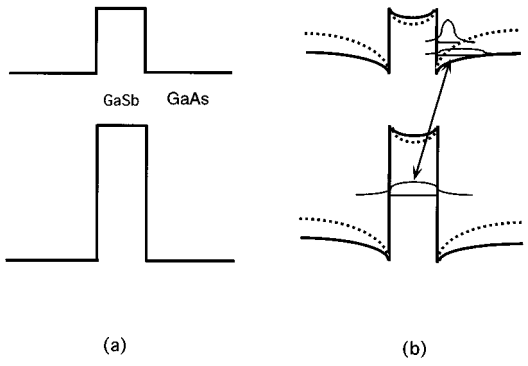

FIG. 1. Schematic drawings of the spatial dependence of the conduction and valence bands for the staggered band lineup (a) and including a space-charge-induced band-bending effect (b). The dashed line in (b) shows a stronger band-bending effect.

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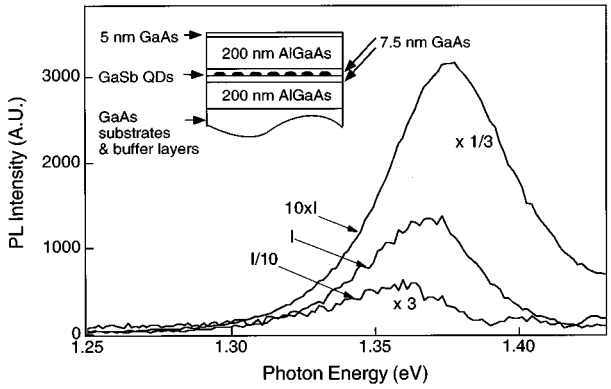

FIG. 2. Quantum dot photoluminescence spectra of structure B with different excitation intensities. Inset shows the schematic diagram of structure B.

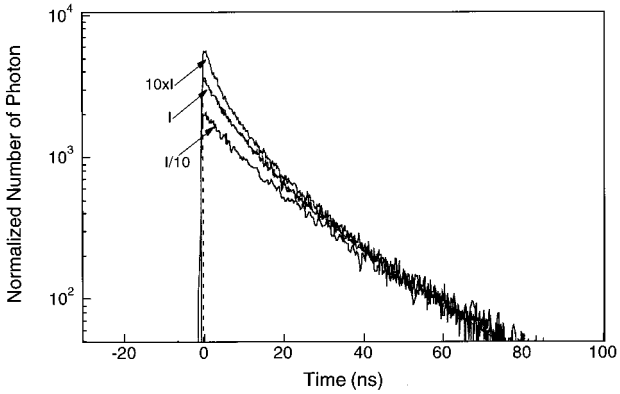

FIG. 3. Time-resolved PL at 1.348 eV on structure B samples at 10 K with different excitation intensities. The data are normalized with the tail exponential parts. The dashed line on top of the middle trace is the double-exponential fit.

deposition and their slow and well-controlled disappearance during the low-temperature GaAs overgrowth.

Atomic force microscope (AFM) imaging of QDs was performed on a sample identical to structure A samples, except that the growth was terminated after the GaSb layer deposition. The dots have a height of 10 nm and a lateral diameter of 80 nm on the (100) surface, with a standard deviation of 7 nm. The dot density is on the order of $4 \times 10^9 \mathrm{cm}^{-2}$.

Photoluminescence (PL) measurements were performed at $T=10$ K using a Ti:sapphire laser as the excitation source which was operated at 790 nm wavelength (1.57 eV). The output of the Ti:sapphire oscillator was injected into a regenerative Ti:sapphire amplifier with a 250 KHz repetition rate. Besides the strong GaAs bandgap luminescence, the low-temperature PL spectrum of structure B shows an efficient luminescence centered at 1.37 eV with 10 mW average excitation power. This luminescence is attributed to the radiative recombination between holes confined in the 0D GaSb QDs and electrons in the surrounding 2D GaAs QW. This spectrum has a full width at half-maximum (FWHM) linewidth of 46 meV, which is attributed to the dot size distribution. A PL spectrum of structure A shows a similar behavior except the dot luminescence intensity is weaker by about a factor of 5 and is centered at 1.322 eV. Higher efficiency of the QD luminescence in structure B samples is due to the separate confinement structure formed by AlGaAs barriers. Higher emission photoenergies in structure B samples might be attributed to quantization of the electron population in the GaAs well, which are involved in the radiative recombination transitions. Compared with other results,[8] the somewhat higher emission energy of 1.37 eV is probably due to the exchange between antimony and arsenic on the dot surfaces during GaAs overgrowth or smaller effective dot size after overgrowth.

Figure 2 shows the QD luminescence with excitation intensities I (10 mW average excitation power), $10\times I$, and $I/10$ on structure B. With 10 mW average excitation power and a spot size on the order of 300 μm, the generated carrier densities in the GaAs QW region are on the order of 10^{11} cm^{-2}. Similar to the report of Hatami *et al.*,[8] we observe a 23 meV blue shift of the dot spectra with increasing excitation power. The center wavelength of the QD luminescence is shifted from 1.355 eV with $I/10$ excitation intensity to 1.378 eV with $10\times I$ excitation intensity. This shift is characteristic for staggered lineup heterostructures,[8,10] and is attributed to the dipole layers formed by the spatial separation of holes in the GaSb dot and the attracted electrons confined in nearby GaAs regions. With higher hole population in the dot region (higher excitation intensity), stronger band-bending is created, induced by the static electric field of the dipole layers. The band-bending effect will push the quantized electron level toward higher energy and cause the observed blue shift, as indicated in Fig. 1(b).

Time-resolved PL measurements were performed using time-correlated photon counting (TCPC) techniques.[11] The system was driven by regenerative amplified Ti:sapphire pulses at 790 nm wavelength with a 1 picosecond pulsewidth and a 250 KHz repetition rate. With this excitation wavelength, no carriers are generated in the $Al_{0.5}Ga_{0.5}As$ barrier region and the measured lifetime is not limited by the slow time constant in the $Al_{0.5}Ga_{0.5}As$ region due to indirect k-space transitions. The time resolution of the system is about 250 ps.

Figure 3 shows the time-resolved PL of structure B samples at 10 K, performed at different excitation intensities, $10\times I$, I, and $I/10$. The measured traces are normalized by the tail part of the decay. A resolution-limited rise is followed by a radiative decay that cannot be fitted with a single exponential. The resolution-limited rise time (<250 ps) indicates a fast hole capture process in GaSb QDs. Combined with the observed bright luminescence, this result supports the concept of an efficient hole capture process even at a large valence band offset. There is no indication of a phonon bottleneck for hole relaxation. Temperature-dependence study indicated that the recombination is dominated by radiative processes at low temperatures. The radiative decay is composed of a faster initial component and a slower tail component. In order to extract time constants from the data, we fit the data with the sum of two exponential functions. The fit for the experimental data with excitation intensity I is shown as the dashed line on top of the trace with time constants τ_1 and τ_2 of 7.5 and 23 ns respectively. The measured lifetimes are much longer than the radiative lifetimes of spatially direct GaSb/AlSb quantum wells (600 ps).[12] These results

support the staggered lineup model (Fig. 1) and the longer time constants are attributed to reduced spatial overlap between electron and hole wavefunctions.

The observed non-single-exponential decay can be explained with the same space charge model used for the blue shift in the intensity-dependent PL measurement [Fig. 1(b)]. After the excitation, electron and holes are generated in the GaAs region. The process of hole capturing into the GaSb dot will compete with carrier recombination in GaAs. Within 250 ps, most of the holes are captured and subsequently a dipole layer is formed between the holes in the GaSb dots and the electrons attracted fromthe surrounding GaAs regions. This field-induced band-bending will confine the electron wave function closer to the GaSb dots. The faster initial time constant is attributed to the increased spatial overlap due to the band-bending-induced electron confinement. As the carrier density decreases with time, the time-dependent band-bending effect is also reduced and the electron wave function is more spread out into the GaAs QW. This reduces the spatial overlap between the electron and hole wave functions and decreases the time-dependent recombination rate. When the majority of the carriers has recombined, the band-bending effect is negligible and the electrons will only be confined in the GaAs QW. An exponential-like decay with a fixed time constant of 23 ns is thus observed. This long decay time is attributed to the slow radiative recombination between Γ state holes in GaSb QDs and Γ state electrons in the GaAs QW.[13] This exponential decay probably indicates a type-II excitonic recombination process. Similar behavior is also observed in structure A samples, in which the tail part shows a longer time constant of 30 ns, whichis due to even weaker electron confinement in the bulk GaAs region.

From the above discussion, we expect a faster initial decay time if the excitation intensity is increased. As we can see from Fig. 3, τ_1 is getting shorter with higher excitation intensity (5.5 ns for $10\times I$ and 9 ns for $I/10$) while τ_2 is roughly constant. This agrees with our model. With higher excitation, the stronger band-bending will confine electrons closer to the GaSb QDs and enhance the carrier recombination rate. At longer time, we are looking at the recombination rate of holes in GaSb QDs and electrons in the GaAs QW, the concentration of which does not depend on the initial hole population. On the other hand, with even higher excitation intensity, we should be able to expect a stronger electrostatic confinement where spatially direct Γ state transition in GaSb QDs should be eventually observed.

In conclusion, we present time-resolved radiative recombination measurements in a staggered line-up quantum dot structure. The meaurements are performed on GaSb quantum dots grown by MBE on (100) GaAs through a self-assembly process. Time-resolved photoluminescence shows a resolution-limited rise time which indicates a fast hole capture process in GaSb QDs. Combined with the observed bright luminescence, this study supports the concept of an efficient hole capture process even at a large valence band offset. The decay following the capture is composed of a faster initial component and a slower exponential-like tail component. The time constant of the initial component is shortened with increased excitation intensity while the time constant of the tail component shows weak dependence on the excitation intensity. Intensity-dependent PL shows a blue shift of the centered emission energies. These results support a staggered-lineup and band-bending models in which the observed luminescence originates from the radiative recombination of 0D holes GaSb dots and 2D or 3D electrons in surrounding GaAs regions.

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[13] This might support the existence of type-II excitons at lower carrier density. For definition of type-II excitons in QDs, see Ref. 5. At higher carrier density, whether they still persist is questionable. The observed initial non-exponential decay does not support the existence of excitons.