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Hole confinement in quantum islands in Ga(AsSb)/GaAs/(AlGa)As heterostructures

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Formation of self-organized Ga(AsSb) quantum islands during growth is shown to occur in a series of Ga(AsSb)/GaAs/(AlGa)As heterostructures, resulting in an in-plane hole confinement of several hundred meV. The shape of the in-plane confinement potential is nearly parabolic and, thus, yields almost equidistant hole energy levels. Transmission electron microscopy reveals that the quantum islands are 100 nm in diameter and exhibit an in-plane variation of the Sb concentration of more than 30%. Up to seven bound hole states are observed in the photoluminescence spectra. Time-resolved photoluminescence data are shown as function of excitation density, lattice temperature, and excitation photon energy and reveal fast carrier capture into, and relaxation within, the quantum islands. The advantages of such structures as active laser material are discussed.
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One of the remaining challenges of today's optoelectronics is the fabrication of emitters in the wavelength regime of 1.3–1.55 μm on GaAs substrates. One approach is to grow self-assembled quantum dots where material systems containing Sb have proven to be promising candidates: high dot densities[1] resulting in large peak modal gain[2] have been obtained. Problems related with the GaSb/GaAs system are the type-II alignment of the bands[2,3] and the subtle change between Stranski–Krastanov and interfacial-misfit growth mode.[4]

We study a series of 7 nm $GaAs_{0.64}Sb_{0.36}$ layers in between GaAs spacers of varying thicknesses. The spacer layer thicknesses are 0, 1, 2, 3, 6, and 9 nm for the six different samples which are labeled as samples 0, 1, 2, 3, 6, and 9, respectively. Carrier confinement layers of 75 nm $Al_{0.25}Ga_{0.75}As$ and 50 nm $Al_{0.5}Ga_{0.5}As$ on either side complete the structure. Details of sample structure and growth by solid-source molecular beam epitaxy are described in Refs. 5 and 6. Here, we show that this leads to the formation of quantum islands (QIs) in the Ga(AsSb) layer.

Time-resolved photoluminescence (PL) data are taken as function of excitation density and lattice temperature. The samples are excited by a 100 fs Ti:sapphire laser with an excitation energy of 1.476 eV, corresponding to a wavelength of 840 nm. Thus, the Ga(AsSb) layers are directly excited for temperatures below 170 K. The laser has a repetition rate of 80 MHz and its average power is varied from 3 to 250 mW. It is focused to a spot of 30 μm diameter at full width at half maximum. The excitation density is, thus, approximately varied between 4×10^{11} and 3×10^{13} cm^{-2}, taking into account reflection losses and assuming an absorption coefficient of 10^4 cm^{-1}.

The sample temperatures are set to 10, 30, and 290 K. The PL is imaged with three times magnification onto the 30 μm wide, vertical entrance slit of an imaging spectrometer where the PL is spectrally dispersed with a resolution of 3 nm. The horizontal entrance slit of the streak camera at the exit of the spectrometer is 30 μm wide, yielding a temporal resolution of 15 ps. The vertical and horizontal slits, thus, cut out an area of 10×10 μm^2 of the excitation spot on the sample. The QIs of one sample are also resonantly excited into the third excited state of the QIs at 1.094 eV using a synchronously pumped optical parametric oscillator (OPO) with all other experimental parameters kept identical.

In Fig. 1, we present a cross-section transmission electron microscopy (TEM) image of sample 6 for better understanding of the structures under investigation. It was performed in [010] projection using a JEOL JEM 3010 at an acceleration voltage of 300 kV. Dark field imaging using the chemically sensitive (002)-reflection was applied to derive the chemical composition of the mixed III/V alloys. The GaAs spacer layers are darker compared to Sb-rich material under the imaging conditions used. The brightness fluctuations in the quantum well (QW) between the dark GaAs spacer layers indicate a strongly inhomogeneous Sb profile in the QW plane, with up to 100 nm wide regions of high Sb concentration (~36%), the QIs, separated from each other by about 50 nm wide, Sb-poor regions (~0%). The fluctua-

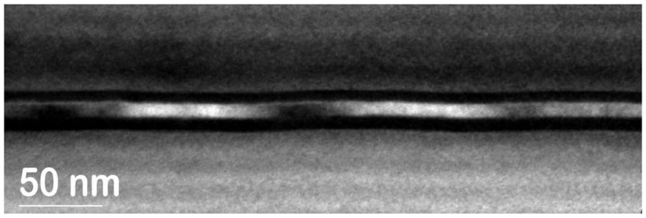

FIG. 1. Cross-section TEM of sample 6.

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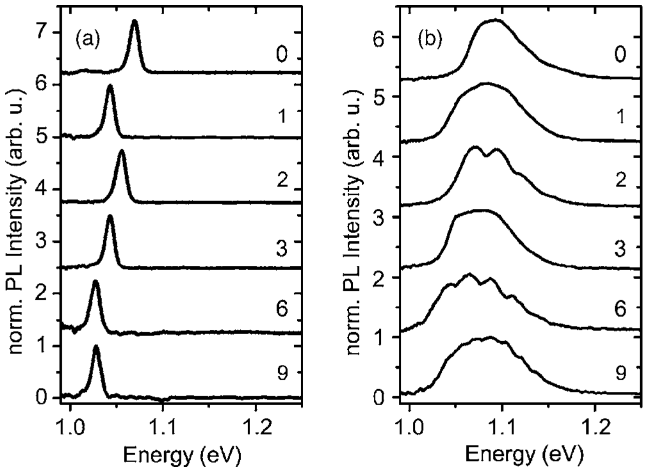

FIG. 2. Time-integrated photoluminescence spectra of the series of samples for lattice temperatures of $T=10$ K and an excitation power of (a) $\rho_{\mathrm{exc}}=3$ mW and (b) $\rho_{\mathrm{exc}}=250$ mW, respectively. The data are normalized to their relative maximum.

tions of Sb concentration cause a strong confinement of the holes in the Sb-rich regions. In contrast, the conduction-band offset of about 40 meV[7] is small, but yields a weak type-II confinement. The electrons are, therefore, only confined between the (AlGa)As layers and/or by the attractive Coulomb potential of the localized holes.

The structures behave like inhomogeneously broadened QWs in PL measurements and electroabsorption[3,7] when the samples are excited on a large area with low spatial resolution. However, taking a "closer look" with 10 μm spatial resolution reveals structured PL emission at high excitation densities and/or high temperatures, respectively. Figure 2 depicts time-integrated spectra of the various samples at (a) low and (b) high excitation densities. The data at low carrier density show, as a general trend, a decrease in transition energy with increasing GaAs spacer width. Sample 1 slightly deviates from that trend and shows a smaller transition energy than expected, which can be explained by internal electric fields as described in Refs. 7 and 8. Only one line is resolved for all samples at a photon density of 1.5×10^{13} cm^{-2}/pulse (a), whereas, up to seven almost equidistant lines are resolved at the highest density of 1.2×10^{15} cm^{-2} per pulse (b). As a general trend, the energy splitting between the almost equidistant lines decreases with increasing thickness of the GaAs spacer layer.

We now discuss the spatial variation of the PL across one sample. The corresponding spectra (not shown here) are taken at different sample positions close to the waver center at distances of 100 μm so that the excitation spots do not overlap. The distance and the relative intensities of the various peaks in the PL lines change with the position on the sample. This demonstrates an inhomogeneous size or shape distribution of the QIs. No structured PL is observed at the edges of the 2 in. wafers. For sample 9, one observes up to seven peaks, covering an overall energy range of 100 meV (not shown here), corresponding to recombination involving excited states. The peak positions are roughly equidistant in energy. Therefore, the shape of the additional confinement potential within the Ga(AsSb) layer should be close to a paraboloid. Similar results hold for all samples.

All these experimental features corroborate the interpretation of the various lines in the high density PL as recombination from the ground and excited states of holes in a QI confinement within the Ga(AsSb) layer. We now estimate the energy spacings of the excited hole states assuming a parabolic confinement potential, a hole mass of $0.08m_0$ (corresponding to the effective mass of a heavy hole in the $\mathrm{GaAs_{0.64}Sb_{0.36}}$ plane, the light hole is ~200 meV lower than the heavy hole, and is therefore, not taken into account) and a variation of Sb concentration from 0% to 36% (the corresponding band gap difference between GaAs and $\mathrm{GaAs_{0.64}Sb_{0.36}}$ is 0.49 eV at 30 K). Widths of a parabolic potential from 50 to 100 nm then deliver equidistant energies of the excited states of 15–40 meV. We, thus, get good agreement between the TEM results and the PL measurements.

Next, we investigate the carrier relaxation in the QI structures. Figure 3(a) depicts time traces taken at the spectral positions indicated by the dashed lines and symbols in the spectrum, which is shown in the inset. The excited state emission at higher energies decays faster. The lines at lower energy initially show a constant intensity caused by the re-

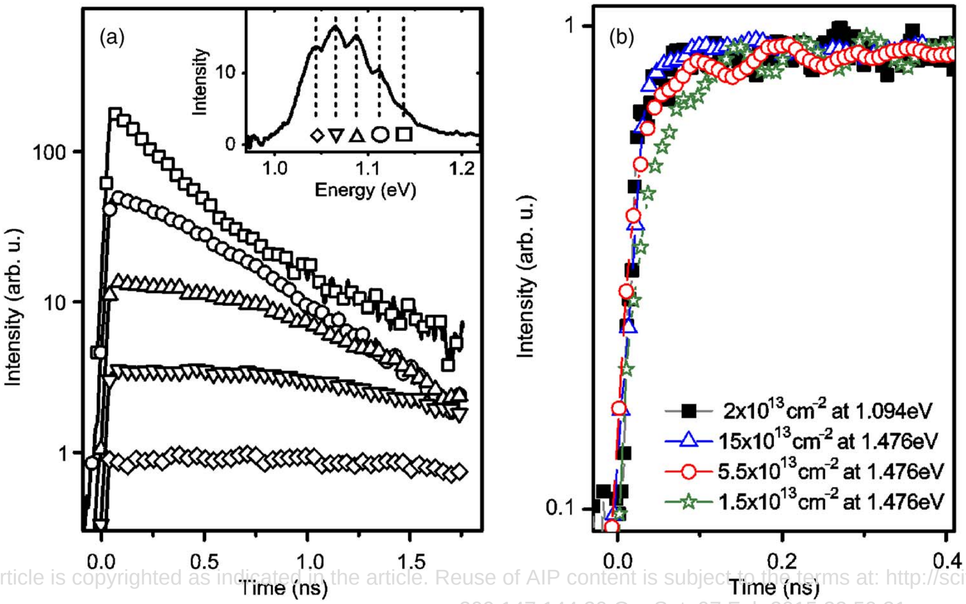

FIG. 3. (Color online) Time-resolved photoluminescence data of sample 6. (a) Temporal evolution of the photoluminescence at $T=30$ K for different energies, excited by the Ti:sapphire laser at 1.476 eV. The corresponding energetic positions are shown in the inset. (b) Excited at 1.094 eV (■) and 1.476 eV with different photon densities (○, ⋆, △) at $T=10$ K. The data are normalized to their relative maximum.

laxation of holes from the excited, high energy states to states at lower energy. This effect is well known and becomes less pronounced at higher temperatures since then the excited states are also thermally occupied.

Figure 3(b) shows the rise of the normalized PL of sample 6 excited with (i) the Ti:sapphire laser at 1.476 eV for three different photon densities and (ii) with the OPO at 1.094 eV for highest photon density. The rise time gets faster with increasing photon density in the case of 1.476 eV excitation. This observation corroborates that carrier-carrier scattering or Auger processes play a role in the capture process. The excitation at 1.094 eV yields a density independent, fast rise time. We conclude that the relaxation within the QI is dominated by phonon emission. This fast capture and relaxation is also corroborated by the effect that at low densities, only the ground state emission is observed.

We find the clear evidence for the QI formation only in the center region of the 2 in. wafers. This inhomogeneity on a large scale might be caused by a slightly inhomogeneous radial growth temperature profile in combination with the sensitivity of Sb-containing layers to changes of growth parameters.[4]

Finally, we discuss the potential advantages of the QI systems investigated here. The TEM measurements show distances of 100 nm of the QIs, yielding a QI density of about 10^{10} cm^{-2} and, thereby, a high density of states per volume. Such a high density obviously leads to a high modal gain of up to 200 cm^{-1}, which was recently obtained with exactly these structures containing only one active layer[9] and which is much larger than the 45 cm^{-1} recently reported by Tatebayashi *et al.*[2] or the 30 cm^{-1} reported by Borri *et al.*[10] Second, the QIs have a strong confinement due to the large in-plane variation of the Sb concentration. The high confinement potential of more than 100 meV is necessary for room temperature operation of QI lasers. Third, the spacing of the QI states is large, in the regime of a few tens of meV. In particular, the excited states in the QI are almost equidistant due to a parabolic confinement potential. We, therefore, expect that relaxation of holes between the QI states is fast since the problem of the phonon bottleneck in intraisland carrier relaxation is strongly reduced.[11] Relaxation of holes via emission of high energy phonons is very efficient and all energy spacings are neither too small nor larger than the maximum phonon energy. Fourth, carrier-carrier or Auger scattering enables fast capture of holes into the QIs at higher carrier densities. Fifth, the overlap between the wave functions of electrons and holes remains reasonably high since we have the additional confinement of the electrons in growth direction by the additional (AlGa)As barriers and by the variation of Sb concentration in the plane of the Ga(AsSb) layer and/or the attractive Coulomb potential of the localized holes which localizes the electrons at the GaAs/Ga(AsSb) interface.[2] This could be beneficial for both, large oscillator strength and fast capture of holes into the QIs via carrier-carrier scattering. The detrimental influence of a type-II structure can thus be reduced. A direct comparison of our QIs with conventional QW structures shows that actually, radiative recombination is comparably fast and efficient.

In summary, we conclude that the formation of Sb-rich QIs leads to a deep, nearly parabolic confinement potential for holes in the plane of a Ga(AsSb) layer surrounded by GaAs spacer layers. The energy separations of the excited states are in the order of a few tens of meV. The QIs have small distances with respect to each other. Barriers of (AlGa)As, the variation of Sb within the Ga(AsSb) layer, and/or the attractive Coulomb potential of the strongly localized holes lead to an additional confinement of the electrons. These effects, together, might prove as beneficial for a potential laser performance.

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