Cathodoluminescence study of band filling and carrier thermalization in GaAs/AlGaAs quantum boxes

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We have examined carrier thermalization, recombination, and band filling in GaAs/AlGaAs quantum boxes with low-temperature cathodoluminescence (CL). The temperature dependence of the quantum box CL intensity for $T \leqslant 90$ K exhibits an Arrhenius behavior, as a result of carrier thermalization between the quantum box and surrounding barrier regions. The width of the quantum box luminescence is found to increase rapidly with an increasing excitation density and reveals an enhanced phase-space and real-space filling, in comparison to the behavior observed for quantum wells. © *1997 American Institute of Physics.* [S0021-8979(97)05404-2]

I. INTRODUCTION

The realization of GaAs/Al$_x$Ga$_{1-x}$As nanostructures (quantum wires and boxes) has attracted a great deal of interest, as they possess unique nonlinear optical properties which are amenable for advanced applications in optical communication that involve switching, amplification, and signal processing.[1–7] An important nonlinear optical property is the change in the emission of light (in energy, polarization, and intensity) that results from phase-space filling of carriers in quantum wells, wires, and boxes. This property is akin to the Burstein–Moss band filling which occurs in bulk semiconductors. Recently, the *in situ* growth of thin III-V layers on patterned GaAs substrates has demonstrated the potential for the creation of nanostructures in a *single-step* growth. This procedure eliminates the need for post-growth etching and patterning on a nanoscale, which are known to degrade the optical and structural quality of the nanostructure.[2–6] Previously, we examined the kinetics of carrier relaxation in *multiple* GaAs/AlGaAs layers grown on a pre-patterned GaAs(001) substrate and demonstrated evidence for an enhanced collection of carriers into smaller GaAs volumes.[7]

In this article, we have examined the band filling and carrier thermalization in isolated GaAs/AlGaAs quantum boxes (QBs). The QBs were grown via deposition of a *single* GaAs/Al$_{0.25}$Ga$_{0.75}$As quantum well on a patterned GaAs(001) substrate, resulting in an array of optically active QBs. The maturation of the etching and growth procedures used for the size reducing growth has facilitated the fabrication of this simple array of QBs, from which fundamental optical properties may be examined using spatially and spectrally resolved cathodoluminescence (CL). A cornerstone in establishing true quantum box behavior is the confirmation of an enhanced phase-space filling of carriers due to a narrowing of the density of states (DOS) relative to a higher dimensionality system, such as a quantum well or wire. We further explore the details of carrier relaxation in the QBs using CL with a variable sample temperature and show that the structural design permits a high degree of carrier collection into the box from the surrounding barriers. A large electron-hole capture rate into the active region is a key parameter in optimizing the design of QB lasers.

II. EXPERIMENT

The details of the sample preparation and procedure for size-reducing molecular beam epitaxial growth on GaAs(111)B and GaAs(001) substrates have been previously reported.[2–4] For this study, GaAs(001) substrates were patterned along $\langle 100 \rangle$ directions using conventional photolithography followed by wet chemical etching to give square mesas with ~ 3 μm long edges and ~ 2 μm in vertical height. The growth consisted of a size-reducing buffer consisting of 16 periods of 195 ML GaAs/5 ML Al$_{0.25}$Ga$_{0.75}$As followed by a quantum well (QW) structure composed of a 100 ML Al$_{0.25}$Ga$_{0.75}$As layer, a 20 ML GaAs QW, a 200 ML Al$_{0.25}$Ga$_{0.75}$As layer, and a 50 ML GaAs capping layer (1 ML$=$2.83 Å). The evolution of the size-reducing epitaxy has been revealed in considerable detail previously, using various Al$_x$Ga$_{1-x}$As marker layers.[4] Figure 1(a) shows a cross-sectional transmission electron microscopy (TEM) image of a typical as-grown mesa demonstrating negligible growth of GaAs layers on the $\{101\}$ sidewalls and the formation of a three dimensional (3D) confined GaAs volume, as identified by the arrow at the mesa top just below the pinch-off layer. The GaAs volume with lateral dimensions of ~ 100 and ~ 1200 Å for the height and width, respectively, delineates a region expected to exhibit QB effects.

The CL experiments were performed with a modified JEOL-840A scanning electron microscopy (SEM) using a $E_b = 8$ keV electron beam with a probe current I_b varying from 10 pA to 5 nA. The sample temperature was varied between 18 and 200 K, using a liquid helium cryogenic specimen stage. The luminescence signal was dispersed by a 0.275 m focal length spectrograph and detected by a cooled Si close coupled diffraction (CCD) array.

III. RESULTS AND DISCUSSION

CL spectra were acquired with a spectral resolution of ~ 0.3 nm. CL spectra at various temperatures, with the e-beam ($I_b = 150$ pA) rastered about a $\sim 1 \mu\text{m} \times 1 \mu\text{m}$ region centered at the mesa top, are shown in Fig. 2. The high-

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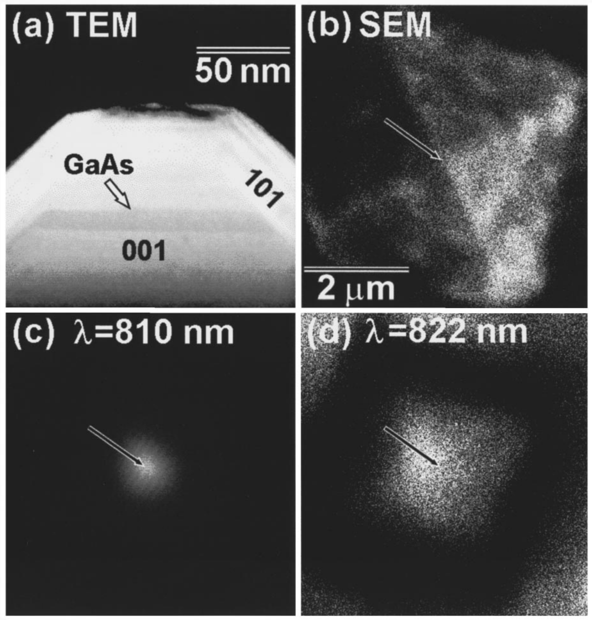

FIG. 1. (a) TEM, (b) SEM, and monochromatic CL images at (c) $\lambda=810$ nm and (d) $\lambda=822$ nm taken with an electron beam of $E_b=8$ keV and $I_b=150$ pA at $T=87$ K. The length scale for CL images (c)–(d) is indicated in the SEM image. The arrows in (b)–(d) point to the mesa top where pinch-off occurred.

energy peak, varying from 807 to 827 nm for temperatures ranging from 18 to 200 K, is attributed to emission from the QB. The CL spectra from the planar (nonpatterned) region show a relatively blue-shifted QW peak at $\lambda=778$ nm for $T=87$ K, revealing that a positive Ga migration to the mesa top occurred and led to a thicker GaAs layer along the [001] growth direction in the QB relative to that for the QW in the planar region. Also, a low-energy luminescence feature, resulting from recombination in the GaAs/AlGaAs buffer layer, is observed to move from ~817 to 843 nm as temperature is increased from 18 to 200 K. An additional impurity-related feature, likely due to carbon in bulk GaAs, is observed at ~830 nm for $T\leqslant 41$ K. In order to further confirm the origin of the luminescence features, monochromatic CL imaging was performed at $T=87$ K. The CL images are shown in Figs. 1(c) and 1(d). In comparison with the SEM image of the mesa in Fig. 1(b), the QB luminescence at $\lambda=810$ nm is evidently localized at the mesa apex. Moreover, the CL image at $\lambda=822$ nm in Fig. 1(c) reveals the broad underlying size reducing GaAs/AlGaAs buffer layer. Thus, the CL imaging results confirm the spatial localization of the QB luminescence.

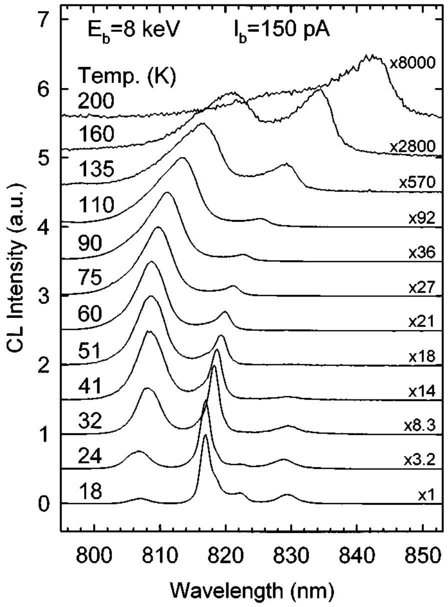

FIG. 2. CL spectra taken at various sample temperatures in the $18\leqslant T\leqslant 200$ K range with the beam rastered about a 1 μm×1 μm region centered at the mesa top.

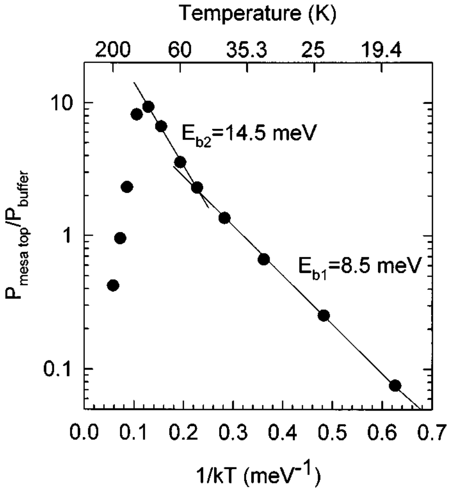

FIG. 3. Ratio of CL intensity for the quantum box to that for the GaAs/AlGaAs buffer, R_{QB}, vs $1/kT$.

The temperature dependence of the CL spectra shown in Fig. 2 reveals striking aspects concerning the thermalization and collection of carriers into the QB. The ratio of the integrated CL intensity from the QB to that from the GaAs/AlGaAs buffer, R_{QB}, is plotted as a function of $1/kT$ in Fig. 3. The ratio increases by more than two orders of magnitude as the temperature increases from 18 to 90 K, and sharply reduces as the temperature further increases to 200 K. In the lower temperature range (i.e., $T\leqslant 90$ K), an Arrhenius dependence for R_{QB} is observed and is characterized by two activation energies $E_{b1}=8.5$ meV and $E_{b2}=14.5$ meV for $T\leqslant 50$ K and $T\geqslant 50$ K, respectively. The energy difference between the QB and GaAs buffer peaks is ~17.5 meV, which is close to E_{b2}. These results demonstrate that carrier collection into the QB occurs primarily via thermal activation over the AlGaAs barriers separating the QB from the GaAs buffer. As the temperature increases from 18 to 90 K, the increasing thermal energy facilitates an enhanced collection into the QB. The QB is expected to exhibit a larger quantum capture rate compared to the thick GaAs buffer, and will therefore dominate the luminescence at temperatures sufficiently large to permit an equilibration of carriers throughout the e-beam excitation volume. For $T\geqslant 90$ K, the ratio R_{QB} decreases as temperature increases, which cannot

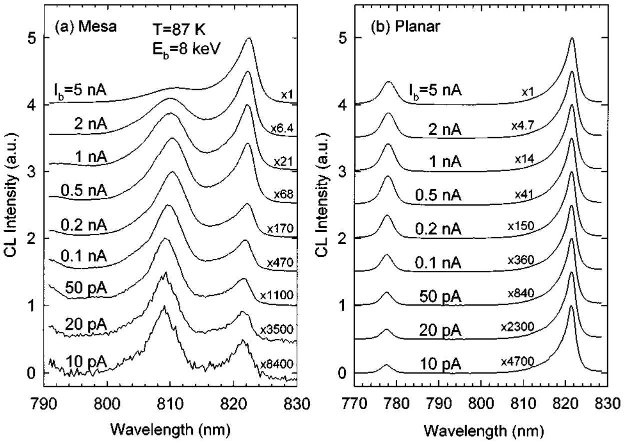

FIG. 4. CL spectra for (a) QB (the mesa top), and (b) the QW (the planar region) with an electron beam of $E_b = 8$ keV and I_b ranging from 10 pA to 5 nA.

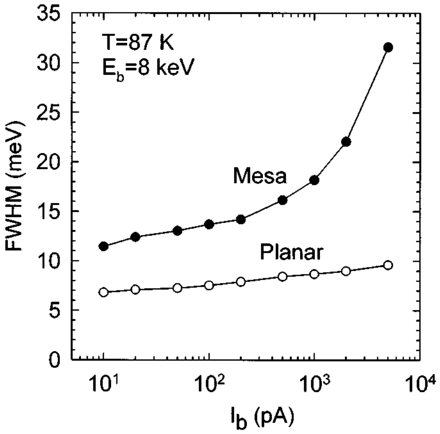

FIG. 5. Full width at half maximum (FWHM) of CL peaks associated with the QB and the GaAs/AlGaAs QW vs I_b.

be explained by the above thermalization process. At higher temperatures, other defect-related channels become active. Such nonradiative recombination is also thermally activated and can be enhanced by nearly two orders of magnitude as the temperature increases from 85 to 300 K.[8,9] A similar temperature dependence of the luminescence efficiency in nanostructures has been observed by Zhang *et al.*,[10] who examined the photoluminescence of strain-confined GaAs/AlGaAs quantum wires and QBs. It is thus evident that barriers which inhibit carrier collection can be found in a variety of nanostructure systems; the barrier height depends on the method and materials employed in the fabrication of the nanostructure.

In order to examine the possibility of enhanced phase-space filling effects associated with the QB, local CL spectra were acquired for regions in close proximity to the mesa top (the QB) and the planar region (the QW), as shown in Figs. 4(a) and 4(b), respectively. In these measurements, the e-beam was rastered about a $1\,\mu\text{m}\times 1\,\mu\text{m}$ region and the beam current was varied from 10 pA to 5 nA. The high-energy emission feature with a peak at $\lambda = 778$ nm in the planar region [Fig. 4(b)], as previously noted, is the luminescence associated with a single AlGaAs/GaAs QW. The luminescence peaks were fitted with a simple Gaussian function with variable width to determine the full width at half maximum (FWHM) of the QB and QW peaks. The FWHM of the QW and QB spectra are shown as a function of beam current I_b in Fig. 5 and increases from 6.7 to 9.6 meV and 11.3 to 31.5 meV, respectively, as I_b increases from 10 pA to 5 nA. We attribute the relatively rapid increase in the QB width to two possible effects. First, the narrowing of the step-like density of state (DOS) of the QW into the spike-like DOS of the QB is expected to result in a rapid filling of single particle states in the QB leading to the intense broadening. A similar excitation dependent broadening in a micro-photoluminescence study has been observed in GaAs/AlGaAs QBs and QWs by Nagamune *et al.*,[11,12] who used metalorganic chemical vapor deposition and a selective growth approach.

The minimum hole intersubband energy separation, E_{hh2}-E_{hh1}, is ~1 meV for a 550-Å thick GaAs/$Al_{0.25}Ga_{0.75}As$ QW. The joint DOS for the QBs in this study will lead to optical transitions likewise possessing an energy separation of $\lesssim 1$ meV. Thus, for the sample temperatures and e-beam excitation conditions employed in this study, we should not expect to observe the extremely narrow (<0.1 meV) spike-like features attributed to optical transitions between the lowest confined e-h states in these QBs. Second, the efficient carrier collection into the QB, as ascertained from the above thermalization data, leads to a real-space filling effect in which a large density electron-hole plasma can be generated in the QB for an extremely weak e-beam excitation. The real space filling will enhance the local e-h plasma density in close proximity to the QB relative to the QW, thereby concomitantly leading to a large phase-space filling in the QB at even the lowest beam current (10 pA) utilized in this study.

IV. CONCLUSION

In conclusion, these results further underscore the potential of size-reducing growth techniques in realizing optically active nanostructures. The temperature dependent CL spectra of these QB samples show that carrier collection into the QB is a thermally activated process for $T \leqslant 90$ K. A large broadening of the luminescence spectra of the QB relative to that for the QW (in the unpatterned region) was observed as the excitation density increased, revealing that the effects of an enhanced real-space and phase-space filling are operative for these QBs. The present findings, which show a large carrier capture and collection into these nanostructures, is a first step in demonstrating the viability for applications in future QB lasers.

ACKNOWLEDGMENT

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