Time-resolved cathodoluminescence study of carrier relaxation in GaAs/AlGaAs layers grown on a patterned GaAs(001) substrate

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We have examined the kinetics of carrier relaxation in three-dimensionally confined GaAs/AlGaAs layers obtained by growth on prepatterned GaAs(001) with time-resolved cathodoluminescence (CL). Time-delayed CL spectra at 87 K reveal that (i) relaxation of hot carriers into the largest 3D confined regions occurs on a time scale of a few hundred ps during the onset of luminescence, and (ii) the luminescence decay time also increases for these larger confined regions, owing to thermal reemission from QWs, diffusion across Al,Ga 1−x As barriers, and carrier feeding from surrounding thinner QWs. © 1996 American Institute of Physics.

The growth of GaAs/Al,Ga 1−x As nanostructures (quantum wires and boxes) on patterned GaAs substrates has attracted much attention in recent years. The high quality of the heterlayers and interfaces that can be achieved during a single growth step circumvents the need for postgrowth etching and patterning on a nanoscale that can inevitably degrade the optical and structural quality of the nanostructure and its surrounding barriers. As a result of facet-dependent growth and cation migration rates, growth on prepatterned substrates can be used to attain a size reduction in the as-patterned micron-scale GaAs mesas so that the resulting size of three-dimensionally confined regions are on the order of ~100 Å. From a fundamental perspective, the study of quantum boxes fabricated on isolated mesas should permit a simple understanding of the temperature and excitation dependence of its optical properties. Such isolated nanostructures on regular arrays inherently avoid a more complex environment of thermally excited electrons and holes that can potentially interact with several quantum boxes, as in the cases for the self-organized InAs island growth. A basic issue which needs to be addressed with the growth of nanostructures on mesas concerns the tailoring of the structure in a manner that maximizes carrier capture and collection into the quantum box. Studies of carrier thermalization and collection have been performed for GaAs/Al,Ga 1−x As quantum wires grown into V-shaped grooves. From a device viewpoint, the efficiency of carrier collection in the nanostructure determines the quantum efficiency of the quantum wire or box laser.

In this letter, we examine the kinetics of carrier relaxation and spatial distribution of luminescence in 3D confined GaAs/Al,Ga 1−x As regions using time-resolved cathodoluminescence (CL). We show that thermalization of carriers in GaAs/Al,Ga 1−x As layers comprising the mesa sidewalls influence both the carrier capture time and luminescence decay time in the 3D confined GaAs volumes situated under the mesa pinch-off region. These results are expected to influence current thinking in the design and growth of 3D confined GaAs volumes so that a maximum collection efficiency of carriers in the quantum box can be achieved.

The details of the sample preparation and procedure for size-reducing molecular beam epitaxial growth have been previously reported for growth on GaAs(111)B and GaAs(001) substrates. For this study, GaAs(001) substrates were patterned along (100) directions using conventional photolithography followed by wet chemical etching to give square mesas with ~3−4 μm long edges and ~2−3 μm in vertical height. The growth consisted of a size-reducing buffer consisting of four periods of 175 ML Al,Ga 0.25 Ga 0.75 As/525 GaAs followed by a 40 period multiple quantum well (MQW) composed of 40 ML Al,Ga 0.25 Ga 0.75 As/20 ML GaAs (1 ML = 2.83 Å). A detailed study of this growth procedure, utilizing transmission electron microscopy (TEM), showed that the evolution of the growth on the mesa sidewalls involves the formation of {103} and {101} facets bounding the (001) surface layers on the mesa top. A cross-sectional TEM of a typical mesa after growth is shown in Fig. 1(a). A pinch-off region is identified by the arrow in Fig.1(a), which shows the final full (001) GaAs layer prior to mesa shrinkage to an apex. The lateral confinement in the final stage of pinch-off due to the negligible growth on the {101} sidewalls is also observed. The

![FIG. 1. (a) TEM, (b) SEM, and monochromatic CL images at various wavelengths (c)–(h) of a typical mesa. The arrows in (b)–(h) point to the mesa top where pinchoff occurred. The length scale for images (c)–(h) is indicated in the SEM image. CL imaging parameters are indicated in the upper-right-hand side of the figure.](http://example.com/figure1.png)
arrow indicates a GaAs volume whose lateral dimensions are 440, 275, and 100 Å for the bottom, top, and height of the trapezoid, respectively, thereby clearly delineating a GaAs region expected to exhibit quantum confinement effects. Owing to the interfacet cation migration and facet-dependent growth rates, an increase in the QW and barrier thickness of the (001) layers relative to the as-deposited MQW is observed in Fig. 1(a). Subsequent growth after pinch-off results in a ~0.2-µm-thick short period GaAs/AlGaAs MQW on the sidewalls whose width, w, varies mainly from ~10–50 Å.

The CL experiments were performed with a modified JEOL-840A scanning electron microscope (SEM) using an 8 keV electron beam with probe current of 150 pA, corresponding to a low excitation condition.9 The temperature of the sample was varied from 87 to 200 K in this study. Monochromatic CL images of the mesas were obtained at 87 K. Time-resolved CL experiments were performed with the method of delayed coincidence in an inverted single photon counting mode, with a time resolution of ~100 ps.10 Electron beam pulses of 50 ns width with a 1 MHz repetition rate were used to excite the sample. The luminescence signal was dispersed by a 1/4 monochromator and detected by a cooled GaAs:Cs PMT. Time-delayed CL spectra were acquired with a spectral resolution of ~1 nm.

Results of constant excitation CL imaging and spectroscopy are shown in Figs. 1 and 2, respectively. A SEM image of a different but again typical mesa is shown in Fig. 1(b). The arrows in Figs. 1(b)–1(h) point to the mesa top. The CL images of this mesa are shown for increasing wavelengths in the 763–826 nm range in Figs. 1(c)–1(h). The CL spectra were taken with the beam rastered about a ~1 µm×1 µm region centered at the mesa top, thereby giving a spatial average over this region. Various sample temperatures were examined, as shown in Fig.2. Three identifiable peaks, labeled p1–p3, are observed to lie within the intense broad luminescence feature located in the 780≤λ≤815 nm range. A fourth peak, p4, has its peak emission at 826 nm for T =87 K, and is identified as emission from the underlying 525 ML GaAs layers. Peaks p1–p3 are observed to follow the shift of the GaAs peak with temperature, and originate from the various GaAs/AlxGa1−xAs layers grown on the mesa. As observed in the CL imaging, emissions at different wavelengths have varying degrees of localization about the mesa top. Images in Figs. 1(c) and 1(d) show that the luminescence at 763 and 788 nm, respectively, is primarily from regions along the sidewalls away from the mesa top. As the wavelength increases from 798 to 814 nm [Figs. 1(e)–1(g)] we observe a greater localization of luminescence about the mesa top. Calculations of the n =1 electron to heavy-hole (e1–hh1) transition energy in Al0.32Ga0.68As/GaAs QWs at T =87 K show that the 780≤λ≤815 nm range corresponds to QW widths in the 48≤w≤130 Å range. Additional smaller peaks are observed in the 723≤λ≤760 nm range, corresponding to 16≤w≤32 Å. Thus, in connecting the CL observations with the above TEM measurements, we conclude that emission involving λ≤800 nm (w≤75 Å) is attributed to luminescence from the sidewall QWs, while emission in the 805≤λ≤815 nm range originates from the thick (90≤w≤130 Å) QWs below the pinch-off region.

The temperature dependence of the CL spectra shown in Fig. 2 reveals salient aspects concerning the thermalization of carriers. For temperatures increasing from 87 to 165 K, the peak intensity of p1 decreases rapidly relative to that for p3. This is consistent with a larger thermal reemission of carriers from thinner QWs relative to thicker QWs for this temperature range. Despite the larger carrier capture rate in short period MQWs due to quantum capture,11 the larger e1–hh1 energies in thinner QWs likewise reduce the barrier height for thermal reemission,12 leading to a marked reduction in QW luminescence efficiency at these higher temperatures. The relative increase in p3 therefore suggests that
In order to further examine these thermalization effects, we have measured time-delayed CL spectra at $T=87$ K for the various time windows indicated in Fig. 3. The time windows $O6–O1$ and $D1–D5$ denote time windows relative to the beginning of onset and decay, respectively, of the luminescence, as referred to the beginning and end of the electron beam pulses. The CL intensity of peaks $p1–p4$ is shown versus time for both the decay and onset stages in Figs. 4(a) and 4(b), respectively. It is evident that rapid capture occurs for the carriers giving rise to the $p1$ feature, as this is the strongest emission for the $O6$ window centered at 150 ps. The other features, $p2–p4$, rapidly grow in the 0–2 ns range; the peak intensity ratios, $p2/p1$, $p3/p1$, and $p4/p1$, versus onset time are shown in Fig. 4(c). Peak $p1$ is also visibly blue shifted $\sim$10 nm during the $O6$ and $O5$ windows (centered at 150 and 450 ps) and gradually toward the 788 nm position observed in spectra measured during constant excitation (Fig. 2) and in the center of the 50 ns excitation pulse (labeled in pulse in Fig. 3). Again, the narrower QWs and barriers comprising $p1$ are expected to exhibit the faster quantum capture times. Subsequent re-emission of these carriers on a time scale of a few hundred ps and thermalization into the thicker QWs comprising $p1$ give rise to the gradual red shift observed for windows $O6–O1$. Likewise, these hot carriers are able to diffuse over sub-$\mu$m distances, further facilitating collection in the 3D confined regions beneath the pinch-off layer.

In conclusion, these results illustrate convincingly that the collection of carriers into the larger QWs and 3D confined GaAs volumes occurs as a result of thermalization, re-emission, and diffusive transport of carriers from the thinner QWs comprising the sidewalls. The thinner sidewall QWs in this study have conveniently served as optical markers, in addition to their intended purpose of structural markers, thereby providing key signatures in the wavelength and time domain that allow for a visualization of the carrier relaxation. It is anticipated, therefore, that a suitable choice of cladding layers during future designs of size-reducing growth will enhance the efficiency of carrier collection into quantum boxes residing just below the pinch-off region.

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