Application of selective epitaxy to fabrication of nanometer scale wire and dot structures

John A. Lebens, Charles S. Tsai, and Kerry J. Vahala Department of Applied Physics, Mail Stop 128-95, California Institute of Technology, Pasadena, California 91125

T. F. Kuech

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

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The selective growth of nanometer scale GaAs wire and dot structures using metalorganic vapor phase epitaxy is demonstrated. Spectrally resolved cathodoluminescence images as well as spectra from single dots and wires are presented. A blue shifting of the GaAs peak is observed as the size scale of the wires and dots decreases.

Conventional crystal growth techniques, such as molecular beam epitaxy (MBE) and metalorganic vapor phase epitaxy (MOVPE), have achieved atomic scale compositional control in one dimension, the growth direction. This capability has resulted in a wealth of new physical phenomena and device applications, producing structures exhibiting quantum confinement in one direction (e.g., quantum wells). Recently there has been interest in developing techniques enabling equivalent control in the lateral directions. Such a capability would greatly enhance the variety of possible structures as well as provide advancements in applications such as monolithic chip integration. A major goal of this effort is to fabricate structures exhibiting quantum confinement in two or three dimensions, so-called quantum wires and quantum dots. An important application of these structures would be their incorporation into the active region of a laser diode, which has been predicted to greatly improve present performance. 1,2

To achieve lateral confinement a variety of approaches is being studied. Lateral confinement has been produced in an already existing quantum well by defining a confinement pattern using techniques such as electron or ion beam lithography and then transferring this pattern into the quantum well by etching, 3-6 impurity-induced disordering using ion beam implantation, or selective diffusion. However, such methods suffer from free surface effects, creation of a damage field during implantation, or interface control due to the random nature of the disordering mechanism. Recently, methods have emerged which allow the formation of nanostructures during the epilayer growth process itself. Growth on patterned substrates has recently shown interesting compositional modulation effects⁹ and has demonstrated stimulated emission from a single quantum wire. 10 Wires grown by migration enhanced epitaxy on a tilted substrate have exhibited optical anisotropies associated with confinement effects. 11 These latter methods have not yet been used to produce quantum dot structures however.

We report here the application of selective epitaxy to the fabrication of arrays of nanometer scale wires and dots. Selective epitaxy refers to lateral spatially controlled growth of epitaxial material within openings of a masking material. In the work presented here, the structures grown are determined by patterning a masked substrate using electron beam lithography. This allows the selective growth of GaAs wires and dots which can be followed by single-crystal growth of $\mathrm{Al_xGa_{1-x}As}$ to embed the structures. Using cathodoluminescence scanning electron microscopy (CL), spectra of single dots and wires as well as spectrally resolved CL images will be presented.

The samples prepared for the selective epitaxy process consisted of a 2 μ m undoped Al_{0.38}Ga_{0.62}As layer followed by a 100 Å GaAs cap layer grown by MOVPE on a semi-insulating (100)GaAs substrate. For the masking material 200 Å of Si₃N₄ was formed by plasma deposition on the sample. Arrays of 100- μ m-long wires with widths ranging from 900 to 3000 Å and dots with diameters ranging from 700 to 3000 Å were patterned into the nitride using electron beam lithography and plasma etching. The wires were oriented in the [011], [011], and [001] directions. Larger areas were also written into the sample as a control.

The selective growth of GaAs was carried out in a low-pressure horizontal MOVPE reactor. Details of the selective growth process have been given elsewhere. A range of samples was grown with the growth temperatures varying from 600 to 750 °C. The growth time gave approximately 1000 Å of GaAs as measured from the cross section of a larger area growth on the sample. The actual growth thickness of the dots and lines may be different and depends on the orientation of the growth as seen below. The GaAs was found to grow selectively within the openings of the mask and this selectivity did not depend on growth temperature. We observed growth of GaAs in all openings, including the smallest.

Figure 1(a) shows a micrograph of an array of dots grown by this technique. The growth displays a highly faceted profile with excellent size uniformity. The exact orientation of these bounding planes is difficult to measure for such small structures. In general the slow growth planes determine the facet profile of the dot. For the halide-based growth of GaAs these have been observed to be the {110} and {111} families of planes. ^{12,13} Selectively grown lines also exhibited a faceted profile with bounding planes depending on line orientation. ¹³ Figure 1(b) shows wires grown oriented in the [001] direction. For the three

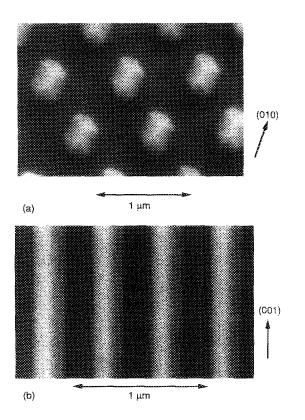


FIG. 1. Scanning electron micrographs of selectively grown GaAs wires and dots. Both the dots in (a) and the wires in (b) show faceted growth profiles. Crystal orientation is indicated in the figures.

wire orientations used in this study we observed a uniform facet profile along the wire.

To obtain good luminescence from such small structures it is important that no free surface exist that would contribute to nonradiative recombination centers. Also, both carrier types (electrons and holes) should be confined to the dot or wire by surrounding the structure with a higher band-gap material. This was accomplished in a second set of samples in which the GaAs selective growth (identical to the first set) was followed by a growth of $Al_xGa_{1-x}As$ (nominally x=0.2) with an approximate thickness of 1000 Å. These samples contained patterns with similar exposure conditions to the first set.

The luminescence properties of such passivated structures were investigated using cathodoluminescence. Details of the CL system are given elsewhere. 14 CL spectra of single wires and dots were measured at 77 K as a function of lateral width and diameter, respectively. These spectra are shown in Figs. 2(a) and 2(b). For comparison the spectrum of a selectively grown broad area on the same sample is included in the figures. We note that the luminescence intensity of these large areas was comparable to conventionally grown GaAs layers we have measured. The exact size of the GaAs dot or wire is difficult to obtain for the capped samples since the lateral growth of the Al_xGa_{1-x}As cap layer is dependent upon the GaAs structure size, line orientation, pattern density, and growth conditions. We estimate the sizes using the first set of samples which have similarly exposed patterns in which only the GaAs was grown. Numerous spectra of dots and wires

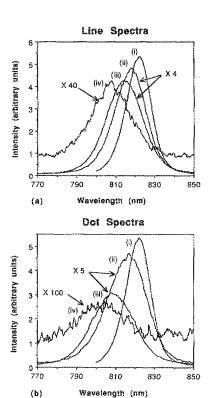


FIG. 2. Cathodoluminescence spectra of wires and dots for three different wire widths and dot diameters, respectively. Plot (a) shows the wire spectra. Spectra (i) is from a large-area control region and is included for reference. The estimated widths of the wires are (ii) 1600 Å, (iii) 1400 Å, and (iv) 1000 Å. Plot (b) shows the dot spectra. Again (i) is the control region. The estimated dot diameters are (ii) 1400 Å, (iii) 1100 Å, and (iv) 800 Å.

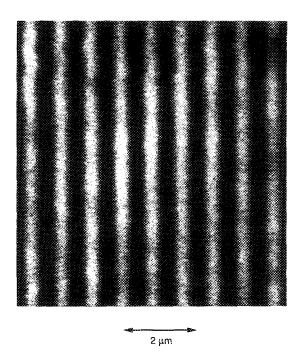
were taken for each size range, showing only a small distribution in peak position about the luminescence peaks in the figures. As the dimensions of the structures decrease, the luminescence peaks show an increasing blue shift.

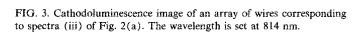
A possible cause of the blue shift is the increasing quantum confinement of the carriers in the GaAs structures. The shifts we see, however, are larger than those predicted for confinement at the estimated structure sizes. There are other possible effects that might contribute to lateral confinement. Electrostatic band bending arising from the free surface on top of the capping $Al_xGa_{1-x}As$ layer will alter the confining potential. Strain created by the surrounding nitride layer could modulate the band gap in the wires and dots. Both effects could potentially increase the quantum confinement of the structures. In order to attribute this shift to quantum confinement with certainty we are currently studying samples with even smaller sizes and at lower temperatures in an attempt to observe subband structure.

Spectrally resolved CL images with the wavelength set at the emission peak of the arrays of dots and wires were also taken. Typical images of wires and dots are shown in Figs. 3 and 4, respectively. The images shown consisted of 1100-Å-diam dots with 0.66 μm spacing and 1400-Å-wide wires with 0.83 μm separation. The wires and dots exhibit good uniformity in emission intensity. This uniformity decreased somewhat for the smallest dots and wires.

In conclusion, we have demonstrated the use of selective epitaxy for fabrication of nanometer scale wires and

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dots. Wires and dots with diameters as small as 700 Å were prepared. Spectrally resolved CL showed luminescence from single wires and dots whose emission was blue shifted with respect to the bulk GaAs. Further investigations are under way in fabricating smaller structures. The success of this growth process shows promise for many possible device applications including dot or wire laser structures.

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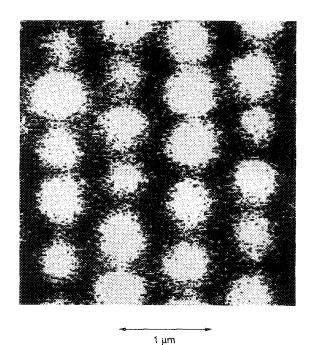


FIG. 4. Cathodoluminescence image of an array of dots corresponding to spectra (iii) of Fig. 2(b). The wavelength is set at 809 nm.

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