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Lateral quantization in the optical emission of barrier-modulated wires

Ch. Gréus
Technische Physik, Universität Würzburg, D-8700 Würzburg, Germany

L. Butov
Technische Physik, Universität Würzburg, D-8700 Würzburg, Germany
and Solid State Physics Institute of the Russian Academy of Sciences, Chernogolovka 142432, Russia

F. Daiminger and A. Forchel
Technische Physik, Universität Würzburg, D-8700 Würzburg, Germany

P. A. Knipp*
Naval Research Laboratory, Washington, D.C. 20375

T. L. Reinecke
Technische Physik, Universität Würzburg, D-8700 Würzburg, Germany
and Naval Research Laboratory, Washington, D.C. 20375
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The optical properties of barrier-modulated $In_{0.18}Ga_{0.82}As/GaAs$ quantum wires with widths down to 25 nm have been studied in photoluminescence spectroscopy. For wires with widths less than 50 nm the lowest-lying transition shows a systematic shift to higher energy corresponding to lateral quantization. Higher-lying laterally confined states have been observed in time-dependent photoluminescence experiments. Detailed calculations have been made of the electronic states of these systems, and the results are in agreement with experiment for the dependence of the transitions on wire width using the widths measured in scanning electron microscopy.

Quasi-one-dimensional and zero-dimensional semiconductor heterostructures currently are of considerable interest both because of the opportunities for studying phenomena in solid-state physics and also for possible device applications. It has been suggested that such systems should show optical and transport properties such as enhanced optical nonlinearities and high mobilities as well as unusual single-particle and collective excitations. A key to progress in this area, particularly for optical studies, is the ability to produce high-quality structures with well-controlled lateral dimensions on the scale of the de Broglie wavelength. It is important that the electronic properties of the systems can be obtained quantitatively from their measured structure, as is the case, for example, with quantum wells, in order that experimental results can be understood fully and new phenomena predicted. A number of techniques[1-6] involving different physical mechanisms have been used recently to achieve lateral confinement and have met with varying degrees of success.

High-resolution electron-beam lithography and etching currently is the most commonly used technique to define and fabricate quantum-wire and quantum-dot structures. A major advantage of this technique is that it can be used readily to make arbitrarily shaped patterns. To date, the most severe drawback of this approach has been that a substantial amount of damage is introduced into the structures by the fabrication process. This leads to the formation of optically inactive dead layers at the wire or dot sidewalls and to strong degradation of their quantum efficiency. The nature and size of these dead layers are not known, and this prevents a quantitative understanding of the basic electronic properties of the systems.

In the present work we have performed photoluminescence studies of a type of quantum wire produced by lithography and etching in which the lateral confinement arises from the modulation of a barrier on a quantum well. We have made calculations of the electronic states of these systems using only the wire widths measured by scanning electron microscopy. We find that the calculated transition energies are in good agreement with experiment. This suggests that these structures are free from significant amounts of process-induced defects and dead layers and that their electronic and optical properties can be understood quantitatively and straightforwardly. To the best of our knowledge, these are the first semiconductor wire structures produced by lithography and etching for optical studies with widths down to ≈ 25 nm for which the electronic properties can be understood quantitatively based on their physical dimensions.

The wire structures studied here were fabricated in a particularly simple way with a single etch step, and they provide effectively buried wires with low defect densities in the optically active region. Strips of a GaAs layer covering an $In_{0.18}Ga_{0.82}As$ quantum well on a GaAs substrate are etched away. The cross section of the wire structure is shown schematically in the inset in Fig. 1. The physical origin of the lateral confinement can be seen by noting that the energy of the ground state in a $GaAs/In_{0.18}Ga_{0.82}As/GaAs$ quantum well is lower than that in a vacuum/$In_{0.18}Ga_{0.82}As/GaAs$ quantum well. This suggests that the carriers effectively experience a lateral potential increase near the edges of the GaAs barrier and that they may be confined under the barriers.

The starting material for the present structures is a 5-nm $In_{0.18}Ga_{0.82}As$ quantum well covered by a 20-nm

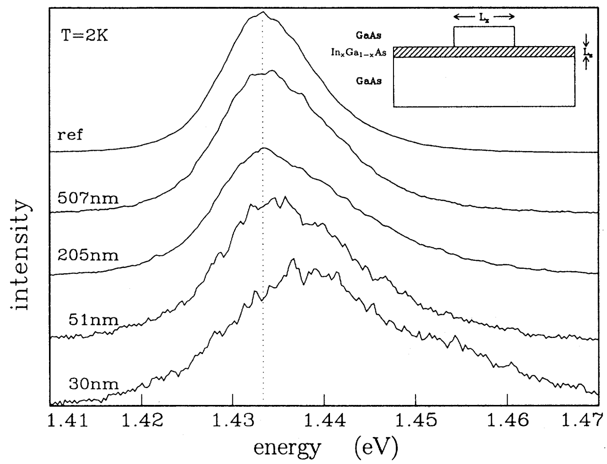

FIG. 1. Emission spectra of modulated barrier wires with varying widths. The wire widths have been measured by scanning electron microscopy. The inset shows the geometry of the modulated barrier wire structure.

GaAs barrier layer grown by molecular-beam epitaxy on a (100) GaAs substrate. The wires were formed by high-resolution electron beam lithography and selective wet etching with H_2O_2 buffered with NH_4OH in a procedure described in more detail elsewhere.[7] From studies of the etch rates in the two materials we estimate that in the etched regions the GaAs top layer is removed completely and the $In_{0.18}Ga_{0.82}As$ well is etched down by approximately 1.5 nm.[7,8] In this way wires have been formed having GaAs barrier widths from a few micrometers down to 25 nm as measured by scanning electron microscopy. The wires are formed in grid patterns having dimensions of $100\times100\ \mu m^2$.

The photoluminescence studies were made using either an Ar-ion laser (typical excitation power density 400 W/cm^2) or a frequency-doubled mode locked Nd:YAG (yttrium aluminum garnet) laser (peak power density about 1 MW/cm^2, pulse length 3 psec) for excitation. The emission was spectrally dispersed by a 30-cm monochromator. In the cw experiments an optical multichannel analyzer with $S25$ characteristics was used as a detector. Time-resolved spectra were recorded using a streak camera with approximately 20-psec time resolution. In these experiments the bath temperature was kept at 2 K.

Figure 1 shows typical photoluminescence spectra for several $In_{0.18}Ga_{0.82}As$ wire widths compared to that for an unetched quantum-well reference. The wire widths given in Fig. 1 are the geometrical widths measured by scanning electron microscopy. For wire widths of 50 nm or less the peak in the spectrum shifts to higher energy which we attribute to lateral quantization. The broadening of the linewidths for smaller wire widths results from fluctuations on the order of 5 nm in the wire widths. We find that the luminescence efficiency is reduced only slightly for wire widths down to the smallest studied[7] which suggests that there are no appreciable dead layers at the edges of the wires.

Figure 2 displays the systematic increase of the emission energy with decreasing wire width. There is little change in the emission energy for wire widths down to 50 nm. For widths of less than 50 nm there is a pronounced increase of the emission energy with shifts reaching ≈ 10 meV for widths of 25 nm. This steep increase of the emission energy for small wire widths indicates that the effective lateral potential has an abrupt spatial variation and that its maximum spatial extent corresponds to the measured wire width. This is consistent with the data for intensity as a function of wire width[7] and gives further evidence that there are no appreciable dead layers in these systems.

Pulsed high-excitation experiments have been used to obtain information on the higher-lying states. In this way both the lowest- and higher-lying wire states can be occupied. The time-resolved data were integrated within a time window of approximately 80 psec placed at the temporal position of the maximum of the intensity (typically ≈ 100 psec after the laser pulse). Figure 3 shows the emission spectra for several wire widths with the peak positions indicated by arrows. Once again, the lowest-lying transition shifts to higher energies with decreasing wire widths. In addition, for wire widths of less than 50 nm a second feature occurs at higher energy, and the energy difference between the two peaks increases with decreasing wire width. The higher-lying peak is found to disappear in times ≈ 100 psec, which is consistent with the carriers relaxing from the higher-lying state to the lower. The presence of the higher-lying transition indicates that the laterally confined states are quantized and have energy splittings on the order of 10 meV for the smaller wire widths. The experimental results for the

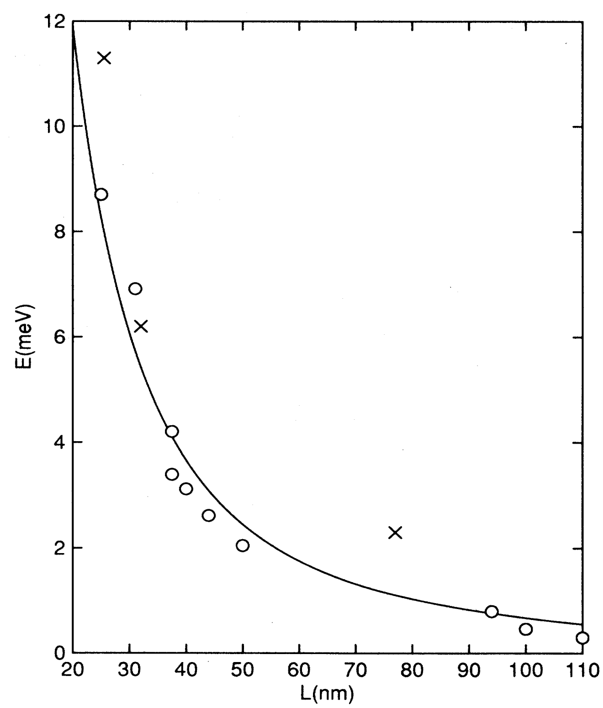

FIG. 2. The dependence of the lowest-lying quantum-wire transition energy on the wire width. Circles and crosses correspond to different samples. The solid line gives the results of calculations described in the text.

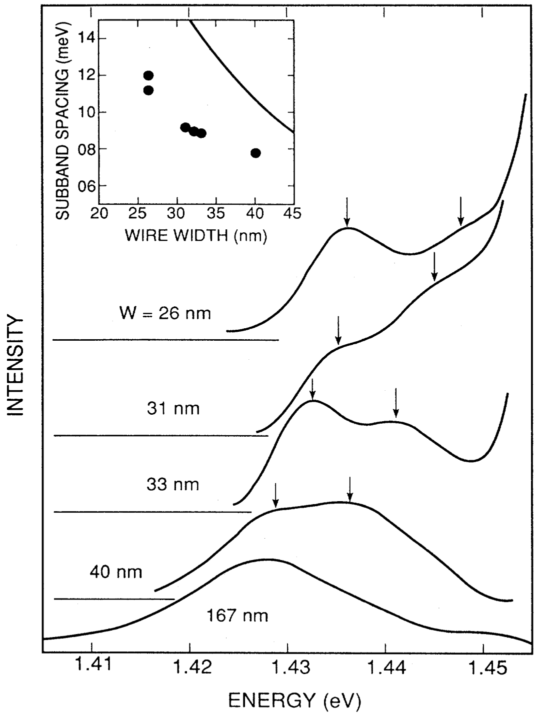

FIG. 3. Luminescence spectra of wires after pulsed excitation at 2 K. In the inset the dots give the splittings between the two transitions in the experimental spectra, and the solid line gives the results of the calculations described in the text.

splittings between the two transitions are given as a function of wire width by the dots in the inset in Fig. 3.

We have made calculations of the energies and wave functions of the electrons and holes in this wire system within the effective-mass approximation. Schrödinger's equation and the boundary conditions, which require the continuity of the carrier wave function and velocity, are not separable in the plane perpendicular to the wire, so the full two-dimensional problem must be solved. This has been done numerically by discretizing the Schrödinger equation following a method suggested by Press *et al.*[9] The usual effective masses and semiconductor band offsets have been used.[10] The effective potential for the interface between the $In_{0.18}Ga_{0.82}As$ quantum well and vacuum is less well known. Recent ultraviolet photoemission spectroscopy data[11] for this surface indicate that the Fermi level lies ≈ 5 eV below the vacuum. This value has been found to be typical of many III-V semiconductor surfaces and surfaces with overlayers.[11] For simplicity we use this value for both the electron and hole barriers.[12,13]

The calculated results for the confined electron wave functions in the quantum wire for a wire of 32 nm width are shown in Fig. 4. For this wire width only two electron states and two hole states are laterally confined. The lowest-lying electron state shown in Fig. 4(a) has no nodes. The next-higher-lying electron state shown in Fig. 4(b) has one nodal plane perpendicular to the quantum well. States with nodes parallel to the surface derive from higher-laying quantum-well states and lie in the quantum-wire continuum. The confined quantum-wire heavy-hole states are similar to those for the electrons shown in Fig. 4.[14] By symmetry only two optical transitions are allowed for this wire width. These transitions are that between the lowest-lying electron and lowest-lying hole states [Fig. 4(a)] and that between the first excited electron and first excited hole states [Fig. 4(b)]. From the present calculations for the wave functions we have found that the physical origin of the lateral quantization and of the wire width dependence of the quantization energy arises from the wave function's being increasingly pushed back into the higher-energy substrate material by the vacuum interface as the wire width decreases.

The calculated energies for the lowest-lying transitions for the $GaAs/In_{0.18}Ga_{0.82}As$ wires are shown by the solid

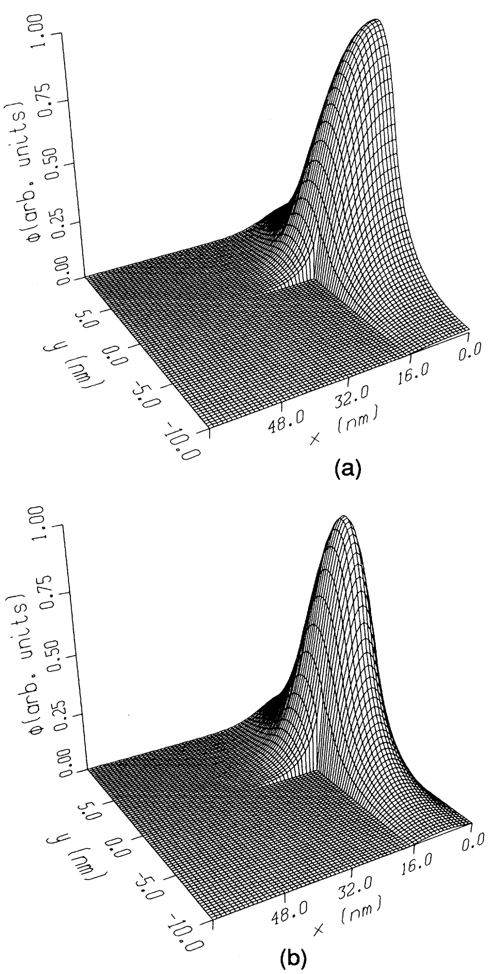

FIG. 4. The calculated wave functions corresponding to the (a) lowest-lying and (b) higher-lying laterally confined electron states for a modulated barrier quantum wire of width 32 nm. The $In_{0.18}Ga_{0.82}As$ quantum well lies between $0<y<5$ nm, the GaAs barrier lies between $-16<x<16$ nm and $-20<y<0$ nm, and the GaAs substrate lies in the region $y>5$ nm. The lowest-lying state has no nodes and is symmetric around $x=0$, and the higher-lying state has one node and is antisymmetric about $x=0$. Only the $x>0$ halves of the wave functions are shown here.

curve in Fig. 2. These theoretical results were obtained using no fitable parameters and using the measured GaAs barrier widths. The overall agreement between experiment and the calculations is seen to be good. We have also made similar experimental and theoretical studies of the low-lying wire transition energies as a function of wire width for $GaAs/In_{0.18}Ga_{0.82}As/Al_{0.15}Ga_{0.85}As$ wires. These systems are of interest because the higher potential barrier of the $Al_{0.15}Ga_{0.85}As$ substrate layer gives a greater lateral quantization. The agreement between the photoluminescence data for the wire width dependence of the emission energy and the calculations is comparable to that shown in Fig. 3. For the $GaAs/In_{0.18}Ga_{0.82}As/Al_{0.15}Ga_{0.85}As$ wires the calculated difference between the lowest-lying wire state and the quantum-well continuum is found to be ≈ 5 meV higher that for wires on GaAs substrates.

The two transitions seen in the time-dependent photoluminescence experiments shown in Fig. 3 correspond to that between the lowest-lying electron and hole states and that between the next-higher-lying electron and hole states as noted above. The splittings between these two transitions obtained from the present single-particle calculations are shown by the solid curve in the inset in Fig. 3. The calculated results show a qualitatively similar dependence on wire width as do the experimental results, which tends to confirm the present interpretation of the transitions. For these experiments, however, the densities are sufficiently high that many-body shifts of the transition energies due to exchange-correlation effects are expected, and it is expected that these shifts will depend on the transition involved as is the case in quantum wells.[15] We believe that this accounts for at least part of the differences between the experimental and theoretical results in Fig. 3.

The good overall agreement between the calculated results and the experimental data for both the $GaAs/In_{0.18}Ga_{0.82}As/GaAs$ and the $aAs/In_{0.18}Ga_{0.82}As/Al_{0.15}Ga_{0.85}As$ quantum wires indicates that the mechanism described here accounts well for the lateral confinement and that the effects of other lateral confinement mechanisms are small. In particular we find that in photoluminescence excitation experiments there is no significant wire width dependence of the splittings between the heavy- and light-hole transition energies. This indicates that the effects of any lateral inhomogeneities in the strain in the $In_{0.18}Ga_{0.82}As$ layer are small. In addition, the high quantum efficiency of these system suggests that the effects of variations in the depletion layer do not play a role in the lateral confinement.

The overall picture given here suggests that these modulated barrier quantum wires are of high quality and that the laterally confined states are quantized with splittings on the order of 10 meV. Their optical properties can be understood on the basis of a physical mechanism in which the vacuum interface causes the carriers to be confined below the GaAs barriers. Furthermore, the good agreement between the experimental results and the theoretical calculations shows that the electronic properties can be understood quantitatively on the basis of the measured physical dimensions of the structures. Based on this work we have been able to suggest that the lateral confinement in these modulated barrier structures can be modified in interesting ways by changing the substrate material or the quantum-well width or by covering the exposed regions of the quantum well with other materials with high potential barriers.

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*Present address: Dept. of Physics and Computer Science, Christopher Newport University, Newport News, VA 23606.

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