

Lateral subband transitions in the luminescence spectra of a one-dimensional electron-hole plasma in $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ quantum wires

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$\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ quantum wires with widths down to about 10 nm have been investigated by high-excitation luminescence spectroscopy. The one-dimensional electron-hole plasma spectra of quantum wires with widths between 50 and 30 nm exhibit shoulders on the high-energy edge of the emission. With decreasing quantum-wire widths the shoulders shift consistently to higher energy. The low- and high-energy features are identified by model calculations with the first, second, and third lateral-subband transitions in the structures. A simple model based on a square-well confinement by the semiconductor vacuum interface and the measured widths of the structures is in quantitative agreement with the experimental data.

In the past few years quasi-one-dimensional systems have been intensely investigated regarding their basic physical properties as well as potential applications. In structures with lateral extensions on the order of the de Broglie wavelength, one should be able to control the band gap over a wide energy range and to observe a lateral subband structure due to lateral quantization effects.¹⁻⁴ Up to now a lateral confinement has been obtained mainly in rather wide structures with dimensions on the order of a few times that of the de Broglie wavelength. As a consequence the lateral quantization effects have generally been rather small. This is an important restriction for the use of the structures at room temperature, because the simultaneous population of several wire states tends to suppress one-dimensional effects in the density of states or regarding the influence of the reduced phase space for scattering.^{5,6} In particular, the energy splitting of a one-dimensional system should be significantly larger than the characteristic energy describing the carrier distribution (e.g., thermal energy or Fermi energy). Therefore it is necessary to develop structures with lateral extensions significantly below 50 nm. Up to now there exist several approaches to realize such wire structures.⁷⁻¹⁷ However, in deep-etched structures independent of the material system investigated, there have been no reports on strong lateral subband splittings up to date.

In the present work we report on the optical properties of ultranarrow $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ quantum wires with lateral dimensions down to about 10 nm. The wires are fabricated by high-resolution electron-beam lithography and wet chemical etching. The confinement potential in the structures is given to a first approximation by the electron affinity. Due to the large value of the electron affinity (≈ 5 eV),¹⁸ the wires show strong lateral quantization effects, which are comparable in size to quantization effects observed in quantum wells. High-excitation spectra show up to three lateral subband transitions, which

vary consistently as the wire width is reduced. Because of the large energy splitting of about 70 meV between the heavy-hole and light-hole states in the 5-nm-thick $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ quantum well, the spectral features in the energy range up to 60 meV above the two-dimensional (2D) band edge are clearly associated with lateral subband transitions in the wires. The experimentally observed lateral quantization effects are well described by a theoretical model based on the measured wire widths and the electron affinity.

The wires are defined on $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ single-quantum-well structures, which include a 5-nm $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ layer below an 8-nm-thick InP top barrier layer. The high-resolution electron-beam exposure is performed on a 100-nm-thick layer of polymethylmethacrylate (PMMA) electron-beam resist at an acceleration voltage of 200 kV. By a lift-off process gold wires are obtained, which serve as a mask for deep wet chemical etching. Details of the fabrication process have been published elsewhere.¹⁹ The widths of the quantum wires were determined by high-resolution scanning electron microscopy (SEM).

In order to analyze the optical properties of the wet etched wires, photoluminescence spectroscopy was performed at a temperature of 2 K. The wires were excited with normally incident light of the 514-nm line of a cw argon-ion laser. The photoluminescence signal was detected by a liquid-nitrogen-cooled germanium detector using the lock-in technique. The experiments were carried out on wire arrays of about $50 \times 50\text{-}\mu\text{m}^2$ size. In addition to the wire arrays, mesa structures of the same size were placed on the samples serving as two-dimensional references. The distance between the different arrays was about 200 μm . The low excitation photoluminescence spectra of wires with widths down to 60 nm have a similar shape and position to the spectra of the 2D references. For wire widths smaller than 60 nm an increasing blue-shift of the photoluminescence signal is observed, which

reaches up to 80 meV for wire widths between 8 and 10 nm.¹⁹

Figure 1 displays photoluminescence spectra from 28-nm-wide $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ quantum wires for different power densities of the exciting cw laser. The excitation densities were varied from 12 W/cm^2 up to 2.8 kW/cm^2 . At the lowest excitation density only the e_{11} - hh_{11} transition between the lowest subbands is observed. Here the first index gives the quantum number of the vertical confinement parallel to the growth direction, and the second index characterizes the lateral quantization. With increasing laser power a one-dimensional electron hole plasma is formed in the wires. In consequence, the linewidth of the emission increases due to phase-space filling at the quantum-wire band edge. When the excitation power is sufficiently high to create a band filling over an energy range of about 45 meV (238 W/cm^2 trace in Fig. 1), the emission of the 28-nm-wide wires includes a high-energy shoulder. It should be pointed out that only remarkably low laser powers are required in order to observe strong band-filling effects. A further increase of the excitation power density leads to an extension of the band filling to higher energies. For power densities in excess of 1 kW/cm^2 a further high-energy feature appears. Furthermore, the slow decrease of the emission intensity on the high-energy side indicates a significant increase of the carrier temperature. As will be discussed in more detail in conjunction with high-excitation spectra of wires with different widths, we associate the high-energy features with the e_{12} - hh_{12} and e_{13} - hh_{13} transitions between the second and third lateral subbands, respectively.

Additionally to the band-filling effects, the low-energy edge of the luminescence gradually shifts to lower energies with increasing excitation density. This effect indicates a renormalization of the band gap due to many-

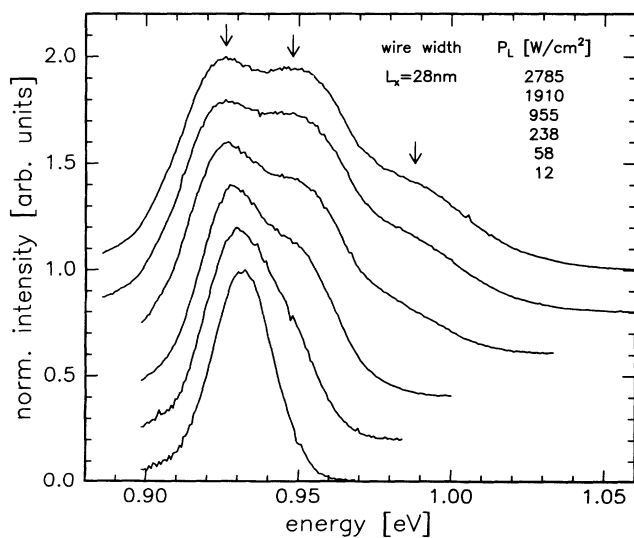


FIG. 1. Photoluminescence spectra of 28-nm-wide $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ wire structures for different excitation densities taken at a temperature of 2 K. For reasons of clarity the spectra are normalized and shifted relative to each other in the vertical direction.

body effects in the one-dimensional electron-hole system. The band-gap renormalization results from the exchange and correlation interaction in the one-dimensional plasma.

Figure 2 shows photoluminescence spectra for different wire widths taken at a fixed excitation density of 350 W/cm^2 . For comparison, a spectrum from a quasi-two-dimensional mesa structure is included at the bottom of the figure. The energy of the photoluminescence emission remains constant for wire widths down to about 60 nm. For smaller wire widths a significant blueshift is observed, which increases systematically with decreasing wire widths and amounts to about 70 meV for the lowest (e_{11} - hh_{11}) transition of 11-nm-wide wires. For wires with widths between 45 and 20 nm the electron-hole plasma spectra exhibit one or more spectral features on the high-energy edge of the emission. That is, the data for the 33-nm-wide wires in Fig. 2 clearly show transitions within the first three lateral subbands. With decreasing wire width the emission features due to higher lateral subband transitions shift strongly to higher energies. Simultaneously we observe a decrease of the emission intensity. The lateral quantization increases the energetic distance between the e_{11} and hh_{11} ground states and the higher subbands in the wires, resulting in a decrease of the occupation of the higher subbands in the narrow wires. The e_{12} - hh_{12} transition vanishes at wire widths

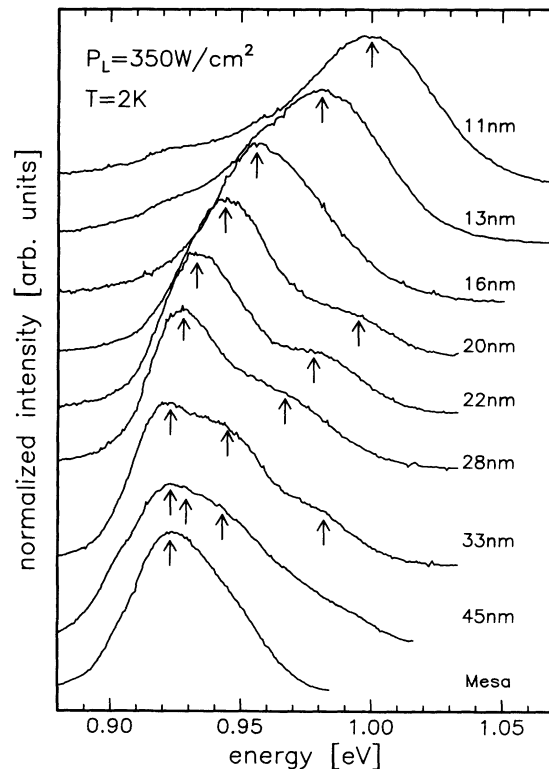


FIG. 2. Photoluminescence spectra of $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ wire structures with different wire widths taken at an excitation density of 350 W/cm^2 . The approximate positions of the different lateral subband transitions are indicated by arrows.

smaller than 20 nm. For the smallest wire widths only the lowest-lying e_{11} - hh_{11} transition is observed.

The arrows in Fig. 2 display the approximate position of different subband transitions in the wires. The positions have been determined by fitting Gaussian line shapes to the experimental spectra. From the systematic increase of the linewidth of the luminescence spectra with decreasing wire width, the roughness of the wires can be estimated. Based on the calculation of the lateral quantization energies discussed below, the average value for the wire width fluctuations is estimated to about ± 3 nm, which is in good agreement with results obtained from SEM micrographs.

Figure 3 displays the energy shift of the different photoluminescence transitions of the wires with respect to the two-dimensional reference signal as a function of the SEM measured geometrical wire widths. The different symbols correspond to the experimental data for the three lateral subband transitions. The energetic distance between the first and second lateral subband transitions increases from a few meV for 45-nm-wide wires to about 40 meV for 20-nm-wide structures. For the energy separation between the second and third subband transitions, we obtain experimental values increasing from about 15 to 45 meV as the wire width is reduced from 45 to 30 nm. In conjunction with the large quantization-induced shift of the band edge by up to 80 meV for 8-nm-wide wires, the present wire structures are well suited to investigate properties of the carrier system in the one-dimensional limit. For wire widths smaller than 25 nm the lateral subband splitting exceeds, e.g., the Fermi energy for an electron-hole pair density of $1 \times 10^6 \text{ cm}^{-2}$ as well as the thermal energy at room temperature and typical LO-phonon energies.

The different curves represent the calculated width dependences for the e_{11} - hh_{11} transition (solid line), e_{12} - hh_{12} transition (dashed line), e_{13} - hh_{13} transition (dotted dashed line) and e_{11} - lh_{11} transition (dotted line). The calculation is based on a simple theoretical model using the standard material parameters and assuming a lateral square-well potential at a finite height of 5 eV.²⁰ This value for the vacuum work function has been found to be typical for many III/V semiconductor surfaces.¹⁸ For simplicity we used this value for both the electron and the hole barrier. The lateral quantization and the quantization in the growth direction are treated to a first approximation as being separable. As shown in Fig. 3 the measured photoluminescence energy shifts are in good quantitative agreement with the theoretical predictions for all three subband transitions. This suggests that the magnitude of other effects, such as surface states, which might reduce or enhance the lateral confinement, are small. Furthermore, the good agreement between the measured and calculated lateral quantization energies indicates that there are no significant dead layers at the sidewalls of the wires.

In conclusion, we have investigated the photoluminescence emission of a one-dimensional electron-hole plasma

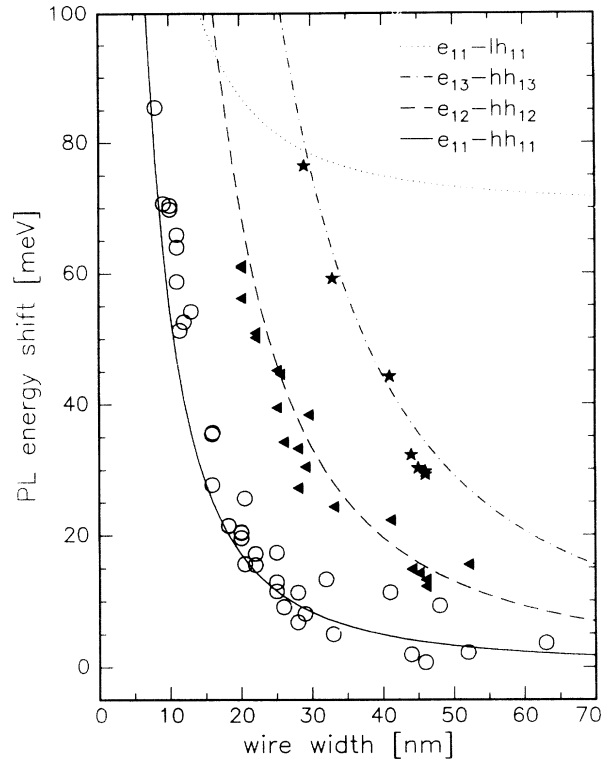


FIG. 3. Wire width dependence of the lateral subband transition energies observed in the photoluminescence spectra of the $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ wires. The curves represent the calculated energy shift of the first three lateral subband transitions based on a simple theoretical model: e_{11} - hh_{11} (solid line), e_{12} - hh_{12} (dashed line), e_{13} - hh_{13} (dashed dotted line), and e_{11} - lh_{11} (dotted line).

in wet chemically etched $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ quantum wires with lateral widths down to about 10 nm. These wires show strong optical emission even without having been overgrown. For increasing excitation densities we observe up to two spectral features on the high-energy side of the 1D plasma spectra. They are attributed to transitions between higher lateral subbands, which are occupied due to the high carrier concentrations. The energetic positions of the three observed lateral subband transitions depend systematically on the widths of the wires. The experimental shifts of the transition energies are in quantitative agreement with the predictions of a simple theoretical model. Our results indicate that electron-beam lithography in combination with wet chemical etching is well suited in order to realize lateral quantum structures with extensions in the 10-nm range.

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