Investigation of size fluctuations in quantum wires by micro-luminescence

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Abstract

Disorder and spectral broadening of vertically-stacked InGaAs/GaAs V-grooved quantum wires have been investigated by means of micro-probe luminescence. We show that the main spectral broadening mechanism originates from monolayer fluctuations at the bottom of the wire. A direct evidence of monolayer height islands of area 40×40 nm formed at the bottom of the grooves is provided by atomic force microscopy. Lateral and vertical wire-to-wire fluctuations are found to be negligible on the micron-scale.

1. Introduction

Size fluctuations and inhomogeneous size distribution cause important broadening in the optical spectra of nanostructures, particularly in conventional spatially integrated experiments, in which a large laser spot, of typical 100 micron diameter, excites simultaneously thousands of wires/dots. Micro-probe spectroscopy allows one to drastically reduce the effects of the size distribution and to study the optical transitions of individual wires/dots, thus measuring the intrinsic linewidth of the structure [1]. In quantum wires, in addition to the wire-to-wire fluctuations, one has to consider the presence of one degree of freedom for the electron motion which allows the diffusion of carriers in the wires on the micron-scale [2]. In this work we have studied high-quality vertically stacked V-shaped InGaAs/GaAs quantum wires by sub-micron spatially resolved luminescence. The aim of the work is to elucidate the impact of wire to wire fluctuations (either vertical and lateral) and of diffusion on the broadening of the optical spectra.

2. Experimental

The samples investigated in this paper were vertically stacked (3 to 6) V-shaped InGaAs/GaAs quantum wires grown by sub-atmospheric pressure metal-organic chemical vapour deposition (MOCVD) on patterned (100) undoped GaAs. The substrates were patterned by holographic lithography followed by anisotropic wet chemical etching resulting in V-shaped grooves along the the [01-1] direction with lateral periodicity of 700 nm and depth 250 nm. Single InGaAs wires were also grown for reference. Due to the small In content these quantum wires have lateral confinement potential of the order of 60 meV for the electrons, with intersubband splitting

of the order of 6 meV. However, the excellent morphology of these structures results in extremely low injection current for the quantum wire electroluminescence (below $300 \mu\text{A}/\text{cm}^2$) and in very narrow luminescence linewidth (below 6 meV) at low temperatures [3-5]. The confocal micro-photoluminescence (micro-PL) set-up had a spatial resolution of $0.7 \mu\text{m}$, sufficient to excite one individual groove. A $30 \mu\text{m}$ pin-hole placed in the image plane of the system was scanned across the magnified image of the luminescence spot with a resolution of $0.3 \mu\text{m}$ in order to probe the density profile across the excited area and the diffusion of carriers far away from the spot center [2]. The samples were excited with the light propagation vector parallel (top excitation) or perpendicular to the growth axis (edge excitation), exciting the sample from the cleaved edge. In all measurements the power density was kept carefully constant so that the energy position of the luminescence energy was not changed by the screening of the internal piezoelectric field (causing a well resolved blue-shift with increasing carrier density [6])

3. Results and discussion

The spatially integrated PL spectra of the samples exhibit a spectral broadening as low as 6 meV at 10 K, which is due to both wire-to-wire fluctuations and microscopic disorder within each individual wire. In our structures, we have to distinguish between the lateral size inhomogeneity (size fluctuations of wires in adjacent grooves) and the vertical inhomogeneity (differences between stacked wires in the same groove). The results of the micro-PL experiments are exemplified in Fig.1, where we compare the

luminescence spectra of the vertically stacked wires taken under top and edge excitation.

Fig.1 - Micro-PL spectra recorded under edge or top excitation. The dotted curves represent the lorentzian deconvolution of the micro-PL spectrum collected under edge excitation. The dashed-dotted curve is the total line-shape fitting. The temperature is 10 K.

The main results are :

- (i) there is no substantial narrowing of the micro-PL linewidth even though the laser spot excites a single groove. The spectra indeed exhibit the same linewidth of the spatially integrated PL, both under top and edge excitation.
 - (ii) there is no change in the micro-PL linewidth of the single and vertically stacked wires (see Fig.2).
 - (iii) a clear shift of the micro-PL band is seen depending on the propagation direction of the exciting beam (top or edge excitation). In particular in edge configuration the luminescence of the $n=2$ excited state is dominant [7].
- Points (i) and (ii) indicate that both the lateral and the vertical wire-to-wire fluctuations are irrelevant on the micron scale. This is clearly demonstrated in Fig.2 where we show the energy position and the linewidth of the micro-PL taken from different individual grooves of a single wire structure (under top excitation). Both the $n=1$ and $n=2$ states (obtained after careful deconvolution of the spectra) exhibit a variation of less than 1 meV over several tens of investigated grooves. Similarly the full width at half maximum of the single wires ranges between 5 and 6.2 meV (top panel of Fig.2). The measured FWHM values are very similar to those obtained in the vertically stacked wires or by the conventional spatially integrated PL (empty symbols in Fig.2 demonstrating the excellent lateral homogeneity of the investigated samples. We can conclude that the wire-to-wire fluctuations introduce a spectral broadening of

the order of 1 meV in the spatially integrated PL spectra.

Fig.2 - Energy of the $n=1$ and $n=2$ transitions of the wires (full dots in the bottom panel and left axis) measured in different individual grooves. The top panel displays the full width at half maximum (FWHM) of the ground level emission band measured in each individual groove (full squares and right axis). The empty symbols refers to the spatially integrated luminescence values

Another broadening mechanism originates from carrier diffusion. Assuming a mean thermal velocity of carriers of 10^6 cm/sec and a recombination time of 350 ps (measured in ref.[8]) we obtain an upper limit for the diffusion length of electrons of the order of 3.5 microns. This value reduces to about 1 μ m if we account for the heavy-hole diffusion and the eventual formation of excitons. This is clearly seen by scanning the confocal pin hole far away from the spot center across the magnified image of the luminescence and along the wire axis. The carrier density reduces progressively due to diffusion, and the concomitant reduction of the screening of the internal piezoelectric field causes a red-shift of the spectrum (due to stronger Stark shift induced by the built-in field). For displacements of about 1 micron away from the spot center we indeed measure a red shift of the micro-PL of about 2 meV.

These measurements are shown in Fig.3. They indicate that diffusion along the wire axis spreads the carrier distribution over an area which is larger than the excitation spot [9]. In the diffusion process carriers undergo localization at

Fig.3 - Red shift of the quantum wire ground level emission measured by scanning the detection pin-hole (empty circle in the inset) far away from the center of the excitation spot (full circle in the inset) along the infinite quantum wire axis.

disorder, resulting in a broadening of the optical spectra, somehow limiting the effective spatial resolution of the experiment. In order to suppress the effect of diffusion, we have also fabricated isolated square-mesas of submicron size by lithography and wet chemical etching. Since the size of the mesa was comparable to the laser spot, a homogeneous carrier density without diffusion was obtained under optical excitation. Under this condition the luminescence spectra again showed a peak at the ground level energy with a linewidth of the order of 6 meV. This experiment confirmed that the spectral broadening does not originate from the localization of carriers spread by the diffusion in different regions of the quantum wires.

The results presented so far unambiguously indicate that:

- 1) lateral and vertical wire-to-wire fluctuations are not responsible for the spectral broadening of the quantum wire luminescence, and,
- 2) diffusion does not influence the spectral broadening of the emission.

This suggests that the actual disorder occurs on a scale length considerably shorter than one micron in quantum wires, so that sub-micron luminescence of individual grooves does not provide a reliable identification of the disorder mechanism. Compositional disorder has been carefully studied in our InGaAs quantum wells, and is found to be controlled within $\pm 0.1\%$ in the range $x=0.1$ [3]. This is supported by the observation of typical linewidth below 2 meV in the absorption and luminescence spectra of our $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ quantum wells. Therefore other broadening mechanisms like the thickness fluctuations in the single wire profile have to be

accounted for in order to understand our results. Since the lateral potential profile originates from the local variation of thickness occurring at the bottom of the groove with respect to the sidewalls, we can write a simple one-dimensional model in which the lateral potential $V(y)$ is a function of the thickness of the bent quantum L_z well at different lateral positions in the grooves. The fluctuations in the wire profile are reflected in random variations of L_z at different y positions ($\delta L_z(y)$). The disorder induced change in the potential is thus given by

$$V_{\text{dis}}(y) = V(y) \pm \frac{dV(y)}{dL_z} \delta L_z(y)$$

putting V_{dis} in the Schrodinger equation we find that fluctuations at the bottom of the groove ($y=0$) introduce an eigenstate variation of the order of 3 meV/monolayer, whereas fluctuations along the sidewalls ($y \neq 0$) have much less impact (0.2 meV/monolayer). This is expected since the thickness fluctuations at the sidewalls cause variations of the lateral potential height, which in turn cause only small variations of the ground level. Though with strong approximation, we might expect that 2 monolayers fluctuations in the wire thickness at the bottom of the groove originate a spectral broadening of the order of 6 meV, i.e. comparable to the measured value. Similar thickness variations along the sidewalls would affect the micro-PL linewidth much less. These results indicate that broadening in the optical spectra of our quantum wires mainly comes from thickness fluctuations of few monolayer height and lateral extension well below $1 \mu\text{m}$ (i.e. much smaller than our spatial resolution), occurring at the bottom of the bent quantum well [10]. The existence of monolayer steps is inherent to the step-flow nature of the MOCVD growth adopted for the fabrication of our wires. In order to confirm our assumptions we have performed atomic resolution AFM experiments on a single quantum wire structure grown without topmost barrier, probing the monolayer planarity of the InGaAs wire at the bottom of the groove. Measurements performed with atomic resolution evidence the presence of 1 monolayer and 2 monolayers high islands in the bottom of the grooves extending over areas of the order of $40\text{nm} \times 40\text{nm}$. A typical image of the wire surface at the bottom of the groove and of its cross section is shown in Fig.4. Steps of the order of 0.35nm and 0.72nm are clearly observed thus confirming the interpretation given above. Clearly the lateral size of the island cannot be resolved

individually by the sub-micron-probe used in our experiment, and can be considered as a form of microscopic disorder which broadens the optical spectra already on the micron-scale (which adds to compositional disorder).

Fig4 Atomic resolution AFM image (40nm*40nm) and cross section taken at the bottom of the wire.

4. Conclusions

In conclusion we have investigated the optical properties of vertically stacked InGaAs/GaAs quantum wires by micro-probe luminescence. We demonstrated that the broadening of the optical spectra comes from thickness fluctuations of monolayer height and lateral extension of about 40 nm, mainly occurring at the bottom of the groove. Wire-to-wire fluctuations, either lateral or vertical, were found to be less important for the observed spectral broadening.

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