Optical imaging spectroscopy of V-groove quantum wires: from localized to delocalized excitons

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Abstract

The exciton localization and delocalization is studied in GaAs/GaAlAs V-shaped quantum wires (QWRs) by microscopy and spectroscopy. Scanning optical imaging of different generations of samples shows that the localization length has been enhanced as the growth techniques were improved. In the best samples, excitons are delocalized in islands of length of the order of 10\textmu m, and form a continuum of 1D states in each of them. On the opposite, in the previous generation of QWRs, the localization length is typically 50 nm and the QWR behaves as a collection of quantum boxes. These localization properties are compared to structural properties and related to the progresses of the growth techniques. The presence of residual disorder is evidenced in the best samples and explained by the separation of electrons and holes due to the large built-in piezo-electric field in the structure.

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At low temperature the optical properties of excitons in semiconductor quantum wires are usually governed by localization effects [1]. It is mainly due to wire thickness fluctuations of ±1 monolayer and therefore to the roughness of the heterointerfaces. Recent progresses of growth techniques allow to obtain samples with longer terraces on the interfaces of the V-QWR [2] and therefore with longer correlation lengths for the localization potential [3]. We report here on the observation of the localization properties of excitons in V-shaped QWRs through our scanning optical imaging technique. A transition from a localized to a delocalized regime for the excitons is shown as the structural properties of the samples are enhanced.

The nanostructures studied experimentally are undoped 5 nm thick GaAs/Ga\textsubscript{0.57}Al\textsubscript{0.43}As V-shaped QWRs. They are grown on a 4.8\textmu m pitched V-grooved GaAs substrate by flow rate modulation epitaxy which is a modified metal organic vapor phase epitaxy technique [4]. The main features are a very strong lateral confinement leading to energy separation between subbands up to 60 meV and a large optical anisotropy, characteristic of the valence band mixing in 1D structures.
The effect of disorder in the optical processes of these structures has first been studied by a new imaging technique, named scanning optical imaging spectroscopy, that we developed recently. This is the equivalent in far-field optics to the scanning near-field optical microscopy. The sample is mounted on the cold finger of a Helium cryostat and cooled down to 10 K. The laser beam is focused on the sample by a microscope objective with a large numerical aperture (0.6), and the diameter of the laser spot is 1 μm, limited by the diffraction of the objective. The signal is analyzed through an imaging spectrometer coupled to a liquid nitrogen cooled CCD detector and the spectral resolution of the detection system is 50 μeV.

The excitation energy of 1.77 eV creates carriers resonantly in the wire transitions. Since the wire spacing is 4.8 μm, only one single wire is excited over 1 μm in our experiments. The excitation spot can then be translated with respect to the sample by moving the microscope objective with piezo-electric actuators with a precision better than 200 nm. At each position the microphotoluminescence (μPL) spectrum is recorded. We obtain in this way, a high resolution spatial image along the wire axis and a high resolution spectrum of a single QWR.

The localization properties of samples corresponding to two different generations of quantum wires have been investigated by scanning optical imaging spectroscopy, as presented in Fig. 1. Each white spot corresponds to a localization site, whose linear density is on the order of 10 μm⁻¹ for QWRs of the previous generation (Fig. 1a) and 1 μm⁻¹ for the new generation of QWRs (Fig. 1b). Their extension along the wire axis can therefore be estimated to about 100 nm and 1 μm for the previous and new generations, respectively. Extended sites can even be resolved spatially in the new generation of QWRs: for example at x = 15 μm in Fig. 1b, the length of the emitting site is 3 μm, larger than the optical resolution (1 μm).

These changes in the typical length scale of the localization in the wires imply not only a quantitative change of their electronic properties, but also a qualitative one, as depicted in the insets of Fig. 1. Whereas excitons are localized in quantum boxes with a discrete spectrum of energy levels in the previous generation of QWRs—told to be in the 0D regime—and form a quasi-continuum of 1D states. In this latter case, each emitting site can be considered as a portion of real quantum wire and will be called “island” in analogy to the situation in quantum wells.

Most of the spectroscopic studies performed up to now on GaAs QWRs confirmed the localized character of the excitons, and we were able to study in 0D regime QWRs the dependences on the localization length of the radiative lifetime, the relaxation time through acoustical phonons [5] and the fine structure of the excitons [6]. In 1D regime QWRs the 1D density of state of the excitons in each island has been probed by measuring the temperature dependence of the radiative lifetime, which follows a $T^{1/2}$ law as expected [7,8].

In order to evaluate the mean properties of the localization sites observed on scanning images, we performed a statistical analysis by auto-correlation techniques. This way we were able to define a “mean
Fig. 2. Statistical mean properties of the islands in the previous generation of QWRs (open dots) and in the new one (plain dots).

localization site”, i.e. its spectrum (Fig. 2a) and the spatial distribution of its luminescence (Fig. 2b). The “mean” spectrum strongly depends on the localization regime: it is very sharp, with coherence times greater than 30 ps, and limited by the spectral resolution for localized states in 0D regime QWRs, as expected for discrete levels. But it is broader (1 meV) and can be perfectly fitted by a lorentzian (in plain line in Fig. 2a) for islands in 1D regime QWRs. We will show in the last part of this paper that this broadening is due to the residual disorder experienced by the excitons as they are delocalized.

The “mean” spatial distribution of the luminescence emitted by the islands is slightly broader in 1D regime QWRs than in the 0D regime ones. Indeed quantum boxes in 0D regime QWRs are small (< 100 nm) compared to the spatial resolution of the microscope, so that the distribution corresponds to the spatial response of the microscope. The measured spatial resolution is equal to 0.8 µm (FWHM). In 1D regime QWRs the broadening of the “mean” spatial distribution (1.2 µm FWHM) implies that the extension of the emitting islands is not negligible compared with the spatial resolution of the setup, and it can be estimated to about 400 nm. This is compatible with the linear density of islands measured by scanning images (1 µm⁻¹).

The enhancement of the localization length of excitons in the wires has been related to the structural properties of the samples. Indeed the localization of the excitons is mainly due to the monolayer fluctuations of the interfaces of the wire (Fig. 3a), which have been identified as (1 0 0), (3 1 1) and (1 1 1) facets by transmission electron micrographs (TEM) of the section of the wire. The fluctuation of the confinement energy of the exciton associated to a monolayer step is about 9 meV on the central facet (1 0 0) and equals 3 meV for steps on the lateral facets (3 1 1) and (1 1 1). These fluctuations can be identified on the images of the new generation QWRs: the ones of the order of 3 meV are quite frequent (1–2 per µm) whereas the larger ones (about 10 meV, e.g. at x = 11 µm in Fig. 1b) are fewer (1 every 5 µm). These values correspond to the linear density of monolayer steps on the central and lateral facets, respectively. The samples of the new generation are therefore of very high quality.

Three main changes in the fabrication of the samples have been developed in the past years. First, the chemical etching allowed to get smoother hetero-interfaces between the well and the barrier materials, as shown by atomic force microscopy images [2]. Second, the arsenic source for the epitaxy has been changed and tertiarybutylarsine (TBAs) is now used instead of the very toxic arsenic (AsH₃). This leads to a strong diminution of the non-radiative recombination at room temperature, which is interpreted as the reduction of the impurity concentration in the sample [9]. The last and the most crucial parameter is the misalignment angle θₐ between the V-groove and the crystallographic direction [1–10] (Fig. 3b). Scanning optical imaging spectroscopy has been performed on samples with different misalignments, and the measured linear density of islands increases from 1 to 4 µm⁻¹ as the angle θₐ is increased from less than 0.004° to 0.1°. This confirms that the localization length in good samples is governed by the distance between monolayer steps on the lateral interfaces of the V-groove, i.e. by the alignment of the V on the crystallographic direction.

The homogeneous broadening (1 meV) of the µPL spectrum of single islands in 1D regime QWRs has to be studied in much more details. It is too large to be due to intrinsic dephasing mechanisms, such as the interaction with acoustic phonons (60 µeV at T = 10 K) and the radiative recombination (2 µeV). We attribute the observed broadening to the local disorder present in each islands. The perfectly lorentzian
lineshape of the “mean” spectrum (Fig. 2a) is characteristic of an homogeneous broadening and invites us to consider dynamical disorder, like collisions to impurities or charges, instead of static disorder, like alloy disorder or atomic roughness of the interfaces. The other signature of this disorder has been found in the µPL spectra under very weak excitation.

Fig. 4a presents the spectra of a single extended island in a 1D regime QWR for different excitation powers. The observed peak gets narrower as the excitation power is decreased, until it is limited by our resolution (100 µeV in this experiment) at the lowest power. We estimated that the excitation power required to create in average one exciton every radiative lifetime (300 ps) in an island is about 400 W cm$^{-2}$. The narrowing of the µPL peaks is therefore observed as one exciton is created every 500 ns in the island. Such long timescales can only be explained if electrons and holes are separated in the island. This charge separation is attributed to the large internal piezo-electric field (5000 V cm$^{-1}$) which exists in the wire [10]. This field is sufficient to separate electrons and holes only in 1D regime QWRs, in which the localization length is large enough, as depicted in Fig. 4b. As the excitation power is increased, fewer electron–hole pairs accumulate in the island and the effect of the piezo-electric field gets smaller, allowing the radiative recombination of the excitons.

In conclusion, we showed by a local probe technique (µPL) that the progresses of the growth techniques allowed to obtain QWRs in the 1D regime, where excitons are delocalized over long distances of a few hundred nanometers in regions free of monolayer fluctuations. The spectral broadening of the µPL lines in the best samples reflects the presence of a residual disorder due to the internal piezo-electric field in the wire.
References