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Near-Field Optical Spectroscopy of Carrier Exchange between Quantum Wells and Single GaAs Quantum Wires

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Real-space transfer and trapping of carriers into single GaAs quantum wires on patterned (311)A GaAs surfaces are studied by near-field luminescence and luminescence excitation spectroscopy with a spatial resolution of 250 nm at temperatures of 8 and 300 K. At room temperature, real-space transfer of carriers in continuum states occurs over a length of several micrometers determined by the mobility of holes and the nanosecond carrier lifetimes. At low temperature, local energetic barriers in the vicinity of the wire suppress the capture process.

The optical and transport properties of semiconductor nanostructures have attracted much interest as tailoring the dimensions and/or compositions of such materials leads to new physical properties and optimized performance of devices. While quasi-two-dimensional (2D) systems have been studied in great detail, carrier dynamics in quasi-onedimensional (1D) quantum wires are less well characterized. In part, this is due to the limited spatial resolution of conventional optical techniques, which allows only to study ensembles of nanostructures, making a separation of real-space transfer and local relaxation processes of carriers difficult. Here, experiments with single quantum wires should provide much more specific information. In this paper, we report the first study of carrier transport and capture in a single quantum well (QW) embedded quantum wire (QWR) nanostructure by means of near-field photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy in a wide temperature range.

The QWR sample was grown by molecular beam epitaxy on patterned GaAs (311) substrates at the sidewall of 15 to 20 nm high mesa stripes oriented along [01-1]. It consists of a nominally 6 nm thick GaAs well layer (QW) clad by 50 nm thick $Al_{0.5}Ga_{0.5}As$ barriers. Formation of an up to 13 nm thick sidewall wire (QWR) arises from the preferential migration of Ga atoms within the QW layer towards the sidewall and results in a 1D confinement over a lateral QWR width of about 50 nm [1,2].

A novel near-field scanning optical microscope [3,4] for sample temperatures between 10 and 300 K was used to record optical spectra of a single QWR [5,6]. A spatial resolution of better than 200 nm is provided by the nanometer-sized aperture of a near-field fiber probe. The sample was excited through the fiber tip and photoluminescence was recorded through a conventional microscope objective. The PL dispersed in a double



Fig. 1. Upper part: near-field PLE spectrum recorded at a sample temperature of 10 K for a detection energy of 1.544 eV and for spatially resolved excitation of the OWR sample through the fiber tip. The detected PL is plotted as a function of the excitation energy and the lateral separation between the QWR position and the fiber tip. Only carriers excited at the QWR position at y = 0 contribute to the signal. Lower part: cross section through the upper part of Fig. 1 at y = 0(solid line) and luminescence spectrum (dash-dotted line). The arrows indicate the position of interband transitions in the QWR sublevel system derived from a theoretical model

monochromator (resolution 1 nm) was detected with a silicon avalanche photodiode, either in steady state or with a time resolution of 250 ps. The excitation density was between 10^4 and 10^5 cm⁻¹.

The high spatial resolution of the near-field technique allows one to separate the photoluminescence excitation spectra of both the QWR and the embedding QW structure and thus provides direct information on the local energetics of this nanostructure. In Fig. 1 we present the spatially resolved PLE spectrum of a single QWR. In Fig. 1, (upper part) the intensity of QWR photoluminescence at 1.544 eV is plotted as a function of excitation energy (abscissa) and of lateral distance of the exciting fiber tip from the QWR position at y = 0 (ordinate). We find a PLE spectrum that is spatially confined to the QWR, i.e. luminescence occurs exclusively for excitation at the QWR location. The width of this region (FWHM) has a value of about 300 nm and is limited by the finite distance of 70 nm between QWR and sample surface. The cross section through this image in Fig. 1, (lower part) exhibits several maxima below the onset of QW absorption. Such peaks are due to the quasi-1D subband structure of the QWR and closely related to the QWR absorption spectrum. Preliminary calculations of the optical transition energies suggest that the first three peaks are due to the n = 1, n = 2, and n = 3heavy hole to conduction band transitions (arrows in Fig. 1, (lower part) [7,8]). From the separation between the first peak and the onset of QW absorption, one derives a confinement energy of about 80 meV. The small Stokes shift of 10 meV between the first peak and the PL maximum and the narrow PL spectrum (dash-dotted line) indicate the high structural quality of the sample. The small inhomogeneous broadening in the spectra is - most probably - related to exciton localization induced by interface roughness on a length scale between the exciton diameter and the spatial resolution of 250 nm.

In Fig. 2a, we present the spatially resolved PLE spectrum of a single QWR at 300 K. For $E_{\rm ex} < 1.515$ eV, photoexcitation occurs exclusively in a narrow region along y = 0,



Fig. 2. a) Room temperature near-field PLE spectrum. The intensity of QWR luminescence at 1.459 eV is plotted as a function of excitation energy (abscissa) and of lateral distance between the QWR located at y = 0 and the fiber tip (ordinate). For excitation energies below 1.515 eV, electronhole pairs are excited exclusively in the QWR. At higher photon energies, carriers excited in the quantum well contribute to the PL signal. b) Near-field PL line scan for excitation at 1.52 eV. c) Near-field PL line scan for excitation at 1.96 eV

similar to the low-temperature data. At photon energies higher than 1.515 eV, however, QWR luminescence is also observed for QW excitation. In Fig. 2a, this gives rise to the bright areas at positive and negative y values. A cross section through this image at $E_{\rm ex} = 1.52$ eV, the onset of QW absorption at 300 K, is shown in Fig. 2b. Carriers generated by localized excitation of the QW undergo diffusive transport over a length of several microns until they are trapped and contribute to the QWR emission. Carrier trapping is dominated by phonons whereas carrier-carrier scattering makes a negligible contribution at the very low excitation densities of our experiments. In the vicinity of the QWR, two local minima of the signal occur, corresponding to the dark areas in Fig. 2a. This is due to local changes of the interband transition energy in our sample. The formation of the QWR by Ga atom migration in the growth process results in a thinning the GaAs QW in the vicinity of the QWR and a blue shift of the local interband absorption. As a result, the QW absorption at 1.52 eV and - thus - the PL signal are reduced at the minima in Fig. 2b. For carrier transport, the bandgap increase close to the QWR represents a local barrier. At 300 K, however, carriers diffusing to the QWR move through this region because of their relatively large thermal energy (kT = 25 meV).

For a quantitative analysis of carrier diffusion, we performed experiments with a high excitation energy of $E_{\rm ex} = 1.96$ eV where QW absorption and – thus – excitation density are not affected by the local barriers. The PLE data in Fig. 2c display a peak at the location of the QWR that is due to direct excitation and exponentially decaying wings which are indicative of the diffusion process. From the exponential decrease of the PLE signal with distance, a diffusion length $L_{\rm d}$ of 1.6 µm is deduced. Using the measured recombination lifetime of 2D carriers of $\tau_{\rm rec} = 2.1$ ns, the relation $D = (L_{\rm d})^2/\tau_{\rm rec}$ and the Einstein relation $\mu_{\rm eff} = De/(kT)$ for the effective mobility of the carriers, one derives values of D = 12.2 cm²/s and $\mu_{\rm eff} = 488$ cm²/(Vs) at T = 300 K. This $\mu_{\rm eff}$ is very close to the mobility of holes in GaAs at room temperature, whereas electron mobilities are significantly higher. We conclude that hole diffusion determines the real-space transfer preceding carrier capture into the QWR. It should be mentioned that the inverse process of carrier transfer from 1D states in the QWR into the 2D continuum mediated by absorption of optical phonons was observed at room temperature [6].

In summary, we demonstrated the potential of near-field photoluminescence excitation spectroscopy for characterizing the nanoscopic optical properties of single semiconductor nanostructures. The high spatial resolution of the NSOM technique allows us to resolve the optical spectra of QWR and embedding QW and to separate real-space transfer and capture of carriers into the QWR in a broad temperature range.

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