Quantization energy mapping of single V-groove GaAs quantum wires by femtosecond near-field optics

M. Achermann,[a] B. A. Nechay, and U. Siegner[b]
Institute of Quantum Electronics, Swiss Federal Institute of Technology Zurich, ETH Honggerberg-HPT, CH-8093 Zurich, Switzerland

A. Hartmann, D. Oberli, and E. Kapon
Department of Physics, Swiss Federal Institute of Technology Lausanne, CH-1015 Lausanne, Switzerland

U. Keller
Institute of Quantum Electronics, Swiss Federal Institute of Technology Zurich, ETH Honggerberg-HPT, CH-8093 Zurich, Switzerland

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We demonstrate that femtosecond pump–probe spectroscopy in the optical near field is well suited to study the intrinsic properties of single V-groove GaAs quantum wires. Temporally and spatially resolved experiments show that the shape of near-field pump–probe traces sensitively depends on the detuning between the laser photon energy and the lowest exciton resonance of a quantum wire. This detuning dependence allows one to map the quantization energy fluctuations along a single quantum wire with 200 nm spatial resolution. We measure fluctuations of about 12 meV over 2 μm wire length, resulting from wire thickness variations of 1 ML. © *2000 American Institute of Physics.* [S0003-6951(00)03219-8]

Low-dimensional semiconductors are highly interesting systems, both from the point of view of fundamental physics and with regard to applications in photonics.[1,2] In particular, V-groove semiconductor quantum wires (QWRs) are promising one-dimensional systems due to their large confinement energy.[1] Typical samples contain a large number of QWRs with submicron spacing. The properties of the QWRs sensitively depend on the sample structure and may vary over a sample due to growth inhomogeneities. Therefore, the interpretation of optical far-field experiments is complicated by averaging over an inhomogeneous ensemble of QWRs. As a consequence, experimental methods for the study of single QWRs and for the quantitative analysis of growth inhomogeneities are highly desirable. Near-field scanning optical microscopes (NSOMs) can provide information about optical properties of laterally structured semiconductors with nanometer-scale spatial resolution.[3,4] In combination with femtosecond pump–probe techniques, the NSOM allows for the study of ultrafast carrier dynamics in nanostructures with high spatial and temporal resolution.[5–7]

In this letter, we present femtosecond-resolved degenerate pump–probe measurements of single V-groove GaAs QWRs using an ultrafast NSOM system.[8] Moreover, we demonstrate a method for the mapping of quantization energy fluctuations along single quantum wires with nanometer-scale spatial resolution. This method takes advantage of the sensitive photon energy dependence of near-field pump–probe traces around the lowest exciton resonance of the QWRs.

The measured sample consists of a 0.5-μm-pitch lateral array of single V-groove GaAs/$Al_xGa_{1-x}As$ ($x=0.33$) QWRs, grown by low-pressure organometallic chemical-vapor deposition on a GaAs substrate.[9] The nominal GaAs epilayer thickness is 1.7 nm, resulting in a QWR thickness of 4.2 nm at the crescent center. To prepare the sample for the NSOM measurements, it was mounted on a glass disk and the GaAs substrate was removed by selective wet etching, leaving a 35-nm-thick $Al_xGa_{1-x}As$ barrier layer on top of the QWRs (see the inset of Fig. 1). This layer thickness

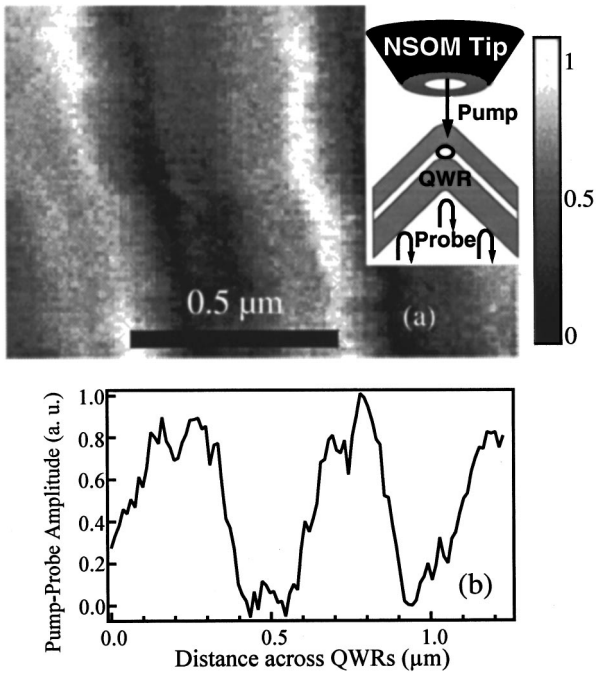

FIG. 1. (a) Two-dimensional image of the pump–probe amplitude at zero time delay. The bending of the wires in the center of the image is due to scanner drift. Inset: schematic of the back-etched V-groove QWR sample (gray: AlGaAs; white: GaAs) and the experimental configuration. (b) Line scan of the pump–probe amplitude perpendicular to the quantum wires.

[a]Electronic mail: achermann@iqe.phys.ethz.ch
[b]Present and permanent address: Physikalisch–Technische Bundesanstalt, Bundesallee 100, D-38116 Braunschweig, Germany.

ensures both QWR energy confinement and good spatial resolution in NSOM measurements. The bottom barrier layer is 200 nm thick to provide mechanical stability. At 20 K, the sample was characterized by photoluminescence (PL) measurements, showing the QWR peak at 1.675 eV with a full width at half maximum (FWHM) of 14 meV. From these PL measurements and PL excitation spectra, we estimate a transition energy of 1.59–1.60 eV of the lowest exciton at room temperature.

Femtosecond NSOM degenerate pump–probe measurements are performed at room temperature with 200 fs pulses, corresponding to a spectral FWHM of 10 meV, from a mode-locked 100 MHz Ti:sapphire laser. The orthogonally polarized pump and probe pulses are modulated at 1 and 1.05 MHz, respectively. The pump–probe signal is detected with an avalanche photodiode at the difference frequency of 50 kHz. Before coupling into the NSOM fiber, the laser pulses are precompensated with a prism pair setup to ensure high temporal resolution. A more detailed description of the femtosecond NSOM can be found in Ref. 8.

In a first experiment, the pump pulse with center photon energy 1.58 eV is transmitted through the NSOM tip to excite the QWRs locally. The pump-induced changes of the reflected probe pulse intensity are globally measured in the far field (see the inset of Fig. 1). The excited carrier density was calculated to be about 10^5 cm^{-1}. Figure 1(a) shows a two-dimensional image of the amplitude of the pump–probe signal at zero time delay. The quantum wires, separated by a distance of 0.5 μm, are well discernable, proving high spatial resolution. Along a single wire, intensity variations of the pump–probe signal indicate fluctuations of the energy or the oscillator strength of the QWR excitonic resonance. Figure 1(b) shows a line scan perpendicular to the QWRs. Between the QWRs the pump–probe signal disappears completely, proving that high spatial resolution and strong contrast is obtained with the applied method. In contrast, if one measures the transmitted pump intensity, one finds that its modulation is only a few percent. This means that the coupling of the near-field pump and the sample changes only slightly across the sample. Therefore, the topographical artifacts[10] give negligible contribution to the nonlinear probe modulation shown in Fig. 1(b). Especially, if we scan along a single QWR, the topographical artifacts will be even more strongly reduced in comparison to a scan perpendicular to the QWRs. Thus, nonlinear optical near-field spectroscopy lends itself well to the characterization of these QWR samples.

For the mapping of quantization energy fluctuations, the sample is excited in the far field and the pump-induced transmission changes are locally detected with a probe pulse, which is sent through the ~200 nm aperture NSOM tip and the sample (see the inset in Fig. 2). For resonant excitation of a QWR, the measured differential transmission is mainly due to pump-induced absorption changes and its decay reflects the carrier dynamics in the QWR.[7] Figure 2(a) shows near-field pump–probe traces for different laser photon energies around the lowest exciton resonance. The traces are taken at a fixed position on a single wire. The measurements are performed with constant laser excitation power, resulting in a carrier density of 2×10^6 cm^{-1} for resonant excitation of the exciton. For all photon energies, a positive bleaching signal

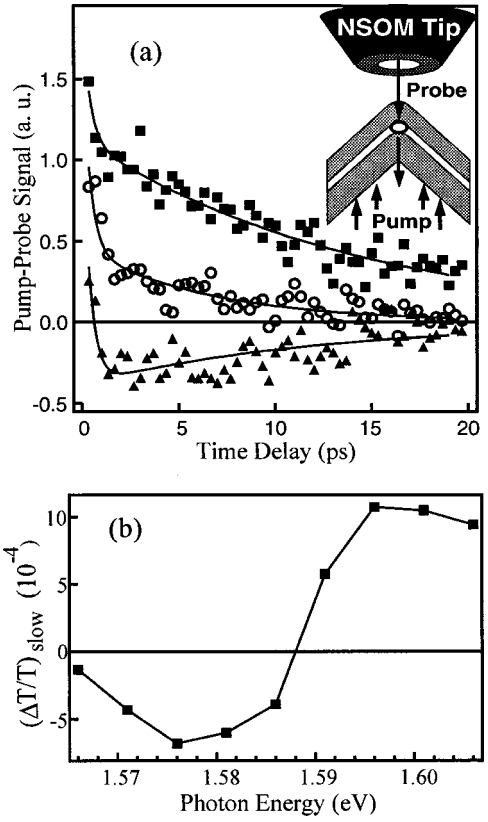

FIG. 2. (a) Near-field pump–probe traces taken at different photon energies around the lowest exciton resonance of the QWR: 1.596 eV (■), 1.586 eV (○), and 1.576 eV (▲). All traces have been obtained at the same position of the QWR. Inset: schematic of the experimental configuration. (b) amplitude $(\Delta T/T)_{\text{slow}}$ of the slowly decaying contribution of near-field pump–probe traces vs photon energy for a fixed QWR position.

is observed at early times due to excitonic phase-space filling and screening. The bleaching signals show a fast initial decay with a time constant of 300–400 fs. We attribute this decay to exciton ionization by LO phonons which reduces the phase-space filling contribution to the pump–probe signal.[11]

After the initial fast decay, the pump–probe traces sensitively depend on the laser photon energy. At higher photon energies close to the exciton resonance, the nonlinear signal is positive, indicating a bleaching process. In contrast, at photon energies well below the exciton resonance, the signal becomes negative, demonstrating induced absorption. We attribute the induced absorption to pump-induced broadening of the exciton resonance. For all photon energies, the pump–probe signal approaches zero with a second time constant of 10–12 ps. Since radiative recombination occurs on a much longer time scale,[12] the slow decay is due to nonradiative processes, such as fast carrier trapping by activated interface impurities.[13]

In order to quantify the detuning dependence, we fit the pump–probe traces to double-exponential functions $(\Delta T/T)_{\text{fast}}\exp(-\Delta t/\tau_{\text{fast}})+(\Delta T/T)_{\text{slow}}\exp(-\Delta t/\tau_{\text{slow}})$. In Fig. 2(b), we show the extracted amplitude $(\Delta T/T)_{\text{slow}}$ of the slowly decaying contribution of a set of pump–probe traces versus laser photon energy. The measurements are performed at a fixed position on the QWR, i.e., for a fixed exciton resonance energy E_x.[14] In a photon energy region around the zero crossing of the amplitude $(\Delta T/T)_{\text{slow}}$, the shape of the

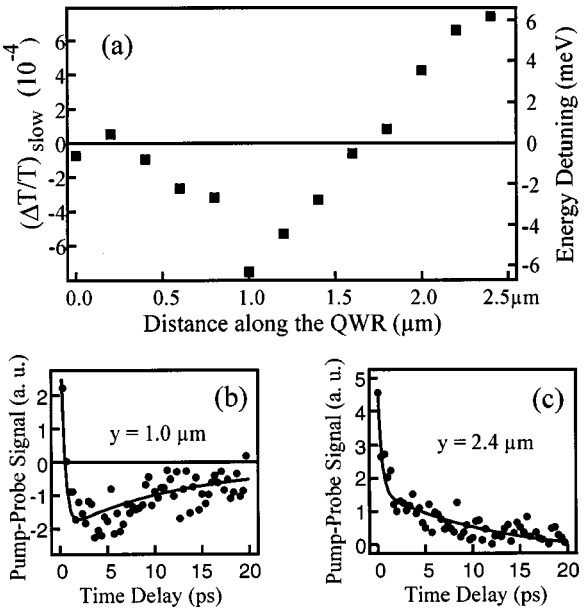

FIG. 3. (a) Amplitude $(\Delta T/T)_{\text{slow}}$ of the slowly decaying contribution of near-field pump–probe traces vs position along a single QWR at fixed center laser photon energy of 1.578 eV. The right axis indicates the calculated energy detuning variation along the wire. (b) and (c) are pump–probe traces at two different positions, 1.0 and 2.4 μm, respectively.

pump–probe signal is very sensitive to the photon energy. In this region, we obtain, in a first approximation, a linear relation between the amplitude and the detuning between the laser photon energy $\hbar\omega$ and E_x with the following slope:

$$\Delta[(\Delta T/T)_{\text{slow}}]/\Delta[\hbar\omega - E_x] = 1.2\times 10^{-4}\ \text{meV}^{-1}. \quad (1)$$

From this slope, one can calculate the variation of the energy detuning for a given change of the amplitude. Equation (1) is valid for a certain excitation power. If this excitation power is used, Eq. (1) allows for the mapping of quantization energy fluctuations along a single quantum wire. This will be discussed in the following.

If the NSOM tip is scanned along a QWR for a fixed laser photon energy, the detuning between the photon energy $\hbar\omega$ and the exciton resonance energy E_x may change due to fluctuations of the exciton quantization energy. In turn, the change of the detuning would result in a change of the amplitude $(\Delta T/T)_{\text{slow}}$ according to Eq. (1). Figure 3(a) shows $(\Delta T/T)_{\text{slow}}$ versus position along a single QWR for a fixed photon energy of 1.578 eV. The values of $(\Delta T/T)_{\text{slow}}$ are calculated from pump–probe traces taken every 200 nm along a single QWR with a NSOM tip providing a spatial resolution of ~200 nm. Apparently, $(\Delta T/T)_{\text{slow}}$ varies with position. This fact is highlighted in Figs. 3(b) and 3(c), which show pump–probe traces at two different positions. The variation of $(\Delta T/T)_{\text{slow}}$ in Fig. 3(a) is due to detuning variations, which result from the fluctuation of the exciton resonance energy since the photon energy is constant.

The fixed photon energy of 1.578 eV is chosen to give the smallest amplitude $(\Delta T/T)_{\text{slow}}$ at the start position of the scan. This ensures that Eq. (1) is valid. Taking Eq. (1), we can calculate the change of the detuning between photon energy and exciton resonance energy from the variation of $(\Delta T/T)_{\text{slow}}$, see the right vertical axis of Fig. 3(a). In that way, the quantization energy fluctuations are determined along the wire. An increase (decrease) of $(\Delta T/T)_{\text{slow}}$ reflects a redshift (blueshift) of the excitonic resonance and, therefore, a smaller (larger) quantization energy.

Energy variations of 12 meV are observed over a 2 μm wire length. Such energy variations are often deduced from the linewidth of spatially integrated PL spectra since they cause inhomogeneous broadening. PL measurements of the QWR sample at low temperatures show a FWHM of the PL peak from the wires of 14 meV, directly giving the inhomogeneous broadening. From the comparison to the measured quantization energy fluctuations of 12 meV, we conclude that the inhomogeneous broadening mainly arises from energy fluctuations along the wires. The quantization energy fluctuations result from variations of the wire thickness. In a simple finite-potential model for our QWRs, considering the first electron–heavy-hole transition, an energy variation of 12 meV corresponds to a wire thickness fluctuation of around 3 Å or 1 ML of GaAs. It is noteworthy that energy fluctuations of less than 12 meV, corresponding to thickness variations of less than 1 ML, are resolved in Fig. 3(a). This shows that monolayer fluctuations occur on a length scale smaller than the tip diameter.

In conclusion, we have presented a method for measuring quantization energy fluctuations along single QWRs with high spatial resolution. The method is based on nonlinear femtosecond pump–probe spectroscopy in the optical near field.

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