

Nanowires for Quantum Optics

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We present optical measurements on nanowire heterostructured quantum dot devices. Quantum dots defined with hetero-material structures are presented as well as a novel type of quantum dot defined by the crystal phase of the semiconductor. We demonstrate the possibility of electrically addressing a single quantum dot with photocurrent and perform electroluminescence measurements on a single nanowire light emitting diode. The applicability of our devices to advanced quantum optics and quantum plasmonics measurements is discussed.

Nanowire quantum dots offer a range of advantages over the well established self-assembled quantum dots. The light extraction efficiency can be far higher because the dot is not fully embedded in a high refractive index material and the nanowire, given the right diameter, can act as a waveguide for the quantum dot emission. The position of the nanowire can be controlled by simply positioning the gold seed particles that are used as catalysts to grow the nanowire heterostructure. The lateral size of the quantum dot can be controlled at will with the nanowire diameter from a few to tens of nanometers and the height of the quantum dot is simply set by the growth time. Stacking two quantum dots in a nanowire is straightforward, making quantum dot molecules easily realized. The possibility to grow a shell around the nanowire enables surface passivation as well as strain engineering. Because of the very small diameters of nanowires, strain at heteroepitaxial boundaries is easily accommodated by lattice deformation instead of defect formation as is the case for two-dimensional growth. This versatility enables the combination of a vast range of materials, even including the combination of silicon and III-V materials. Last but not least, a single nanowire can be electrically contacted and all the current flowing through the device will then flow through the quantum dot.

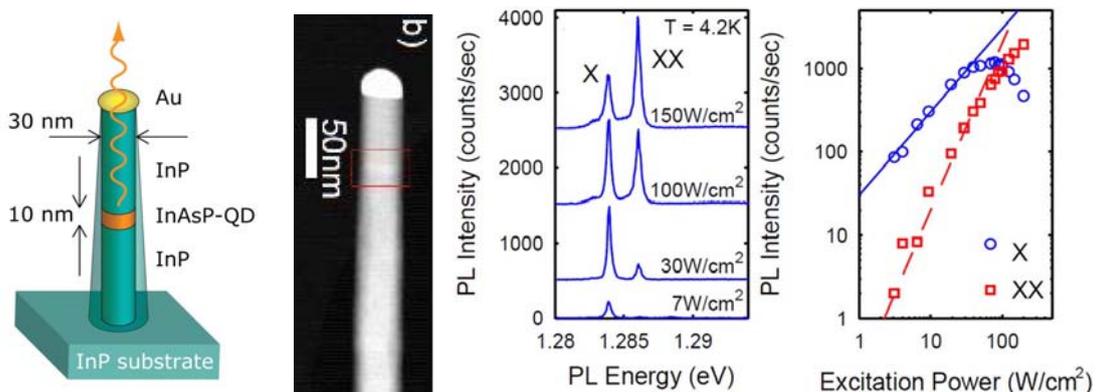


Fig 1. Left: structure of an InAsP/InP nanowire quantum dot, a transmission electron microscope image of a nanowire quantum dot is shown, the quantum dot is highlighted by the red rectangle. Photoluminescence spectra taken under increasing laser power showing the exciton (X) and biexciton (XX) emission. Right: Integrated power dependence for the exciton and biexciton.

Our first goal was to define a high quality nanowire quantum dot heterostructure, where the emission linewidth would be narrow enough to enable quantum optics experiments. We obtained high quality quantum dots made of InAsP in an InP nanowire with a thin InP shell acting as a surface passivation for the nanowire. A schematic of the structure is shown in fig 1a. A transmission electron microscope image confirms the presence of the quantum dot, as shown in fig 1b.

Optical measurements were performed at the single quantum dot level at cryogenic temperatures using a micro-photoluminescence setup with a high numerical aperture objective (0.85 NA), sensitive CCD and spectrometer with a resolution of 25 \AA eV . Spectra taken under increasing laser excitation power density are shown in fig 1c where the exciton and biexciton emission lines are observed. The identification of the exciton and biexciton lines is confirmed by a power dependence shown in fig 1d where the exciton shows a linear power dependence while the biexciton shows a quadratic power dependence. This behavior is very much reminiscent of the usual self-assembled quantum dot behavior.

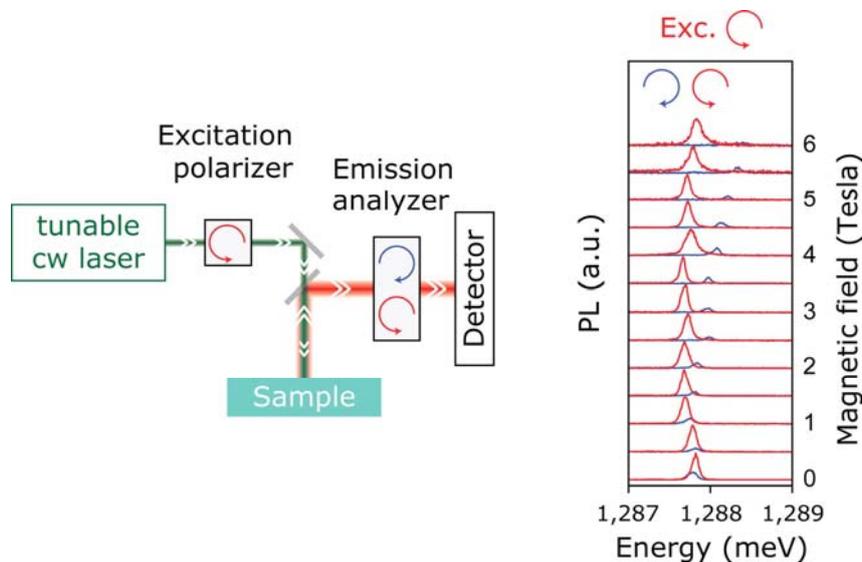


Fig 2 Left: Schematic of the single spin memory experiment: a circularly polarized laser pulse creates an exciton. The photoluminescence polarization is then analyzed. Right: Photoluminescence spectra under right circularly polarized excitation measured under increasing magnetic field from 0 to 6 T. The photoluminescence is always right circularly polarized.

The narrow linewidth of the exciton emission (typically of the order of 100 \AA eV) enables the observation of a Zeeman splitting at easily accessible magnetic fields. We tuned the excitation laser to the p-shell of the quantum dot for quasi-resonant excitation and set its polarization to right circular polarization. This then creates a given spin orientation of the electron and hole. No spin flip is to be expected from the p to s shell relaxation and the luminescence should therefore retain the same polarization than the incoming excitation laser pulse. This experiment was performed with the setup schematically shown in fig 2a, where the laser and photoluminescence polarization is precisely measured. The measured exciton photoluminescence is shown in fig 2b as a function of magnetic field intensity for right circular excitation. The photoluminescence is dominantly right circular polarized, demonstrating that the exciton spin is conserved during relaxation. Our nanowire quantum dots can therefore act as spin memories for time scales in excess of their radiative lifetimes (of about 1 ns). The same experiment was performed with left circularly polarized light and demonstrated left circularly polarized photoluminescence. [1]

Quantum dots are usually defined with material heterostructures such as InAs in GaAs. This material combination has been a very successful and thoroughly studied quantum dot system. Nanowires offer a new route to charge confinement at the nanometer scale: the crystal phase is an additional degree of freedom: both zincblende and wurtzite crystal structures are stable at the nanometer scale. While both crystal structures have the exact same chemical compositions, their band structure differ in bandgap as well as in band alignment. A zincblende InP region in a wurtzite InP nanowire confines electrons in the conduction band. We show InP nanowire in fig 3a, transmission electron microscopy images are shown in fig 3b. The red arrows indicate zincblende regions. While the zincblende segments are not yet fully controlled in terms of position and width during growth, the density is low enough to study a very small number of such structures optically. In fig 3c, we show an atomic resolution image, where the narrow zincblende region that can confine electrons is visible.

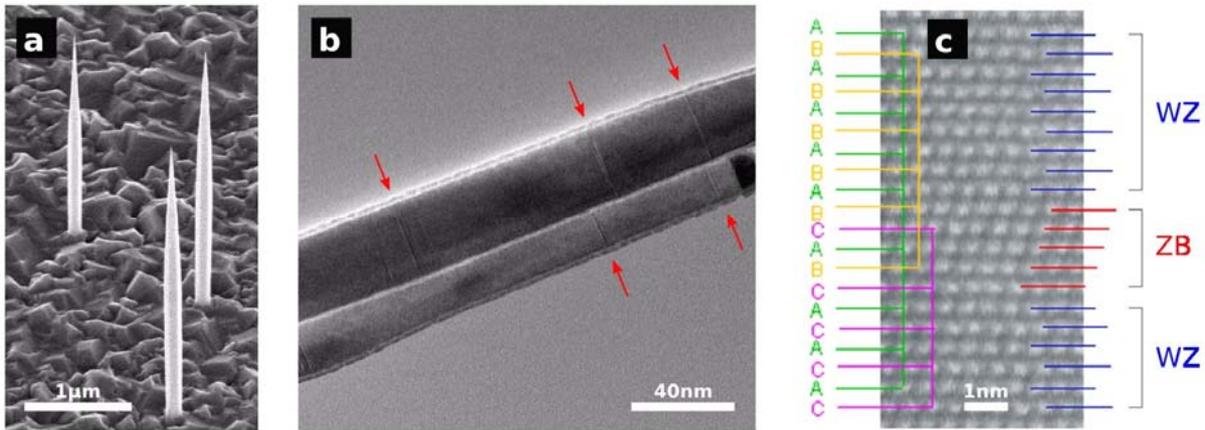


Fig Fig 3 (a) Scanning electron microscope image of an InP nanowire. (b) Transmission electron microscope image of two InP nanowires. The red arrows indicate zincblende segments. (c) atomic resolution image showing a zincblende segment.

A photoluminescence spectrum taken on a single InP nanowires containing zincblende segments is shown in fig. 4. The emission linewidth is very narrow ($24 \mu\text{eV}$, limited by our spectrometer resolution) and very intense with an integrated intensity above 10^4 detections/s. This demonstrates the great value of this novel type of quantum dots where only the crystal phase is used to define the confinement potential. We have demonstrated that these crystal phase quantum dots act as single photon sources, and that the radiative lifetimes are very long, as expected from the type II recombination in this system [2].

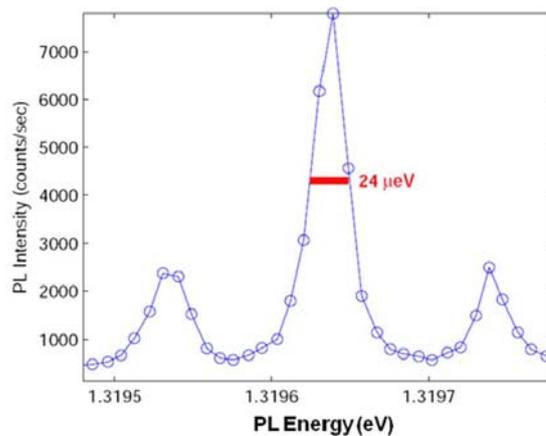


Fig 4 Photoluminescence spectrum measured on a single zincblende segment in a wurzite InP nanowire.

The high quality InAsP quantum dots in InP nanowires were then positioned in nanowires with a p doped extremity on one side and n doping on the other to define a pin structure. This structure can be used both as a detector by reverse biasing the pn junction or as a light emitting diode under forward bias. A schematic view of the band structure of the device is shown in the top of fig 5. A photocurrent image was obtained by applying a constant reverse bias on the nanowire structure and measuring the current while a laser spot was scanned across the device. The resulting image is shown at the bottom of fig 5 where the surface reflection was also added to make the contacts visible. An intense photocurrent spot is clearly visible in the middle of the nanowire, exactly where the quantum dot is expected. This demonstrates that absorption takes place in the quantum dot and that under reverse bias the photogenerated electron-hole pairs tunnel out and contribute to a current.

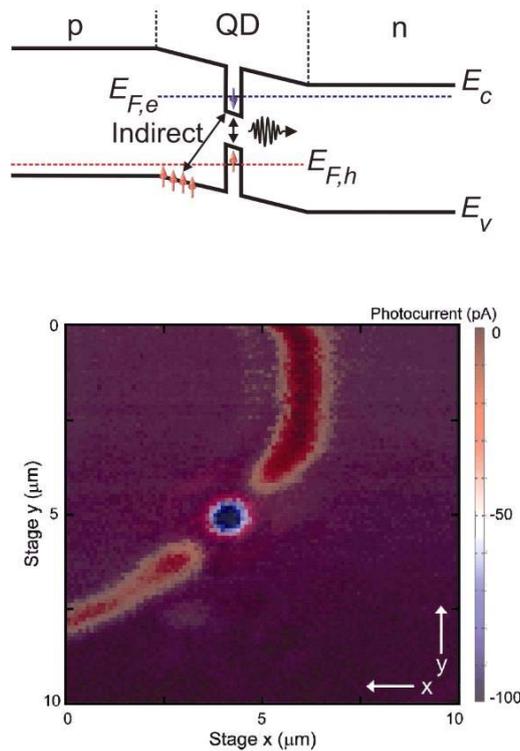


Fig 5 Top: band structure of a nanowire quantum dot light emitting diode. Bottom: photocurrent image of a nanowire device under reverse bias. A reflection image was superimposed with the photocurrent to make the electrical contacts visible.

The very same device that was used for photocurrent measurements can also be used for electroluminescence experiments. Here a single quantum dot embedded in a nanowire is electrically excited; this represents an important step towards the realization of a practical single photon turnstile device where single photons would be generated on demand under the injection of single electrons. In fig 6 (left), we present a schematic of the device we have realized where ohmic electrical contacts are made to both the p and n doped regions and electroluminescence occurs at the pn junction under forward bias. A microscope image of the real device under white light illumination and zero bias is shown in fig 6 (center), here the device is held at liquid helium temperature. Fig 6 (right) shows the device under a forward bias of 2 V. Electroluminescence is clearly observed from the center of the nanowire where the pn junction is expected. This constitutes the operation of the smallest light emitting diode reported so far where the active region is only one single quantum dot.

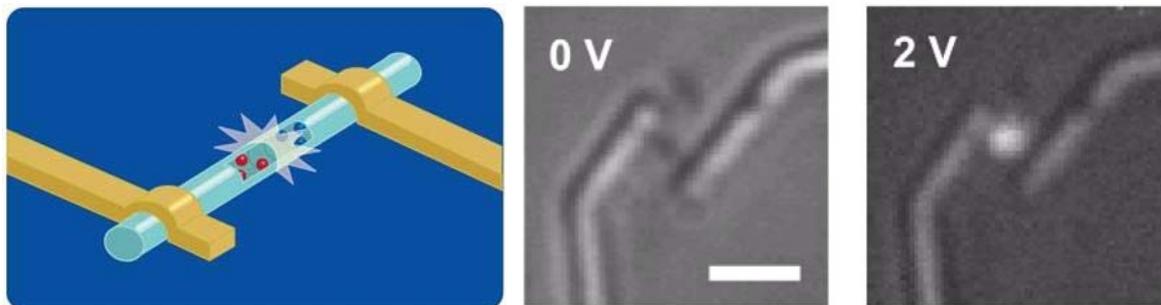


Fig 6. Left: schematic of a single nanowire light emitting diode. Center and right: microscope image of a nanowire light emitting diode, from ref [3].

Our single quantum dot nanowire offers great application perspectives in the field of integrated quantum optics. We show in fig 7a a schematic representation of an all on-chip quantum plasmonics device where a single nanowire light emitting diode is used as a source and is coupled to a plasmon waveguide. A superconducting nanowire is then used to detect single plasmons in the near field. A plasmonic waveguide coupled to a single plasmon detector was fabricated and successfully operated in our group [4]. Fig 7b shows an integrated plasmonic Hanbury-Brown Twiss interferometer where two plasmonic waveguides are coupled over 5 microns to obtain a 50-50 coupling according to simulations. Here, both output arms are coupled to superconducting nanowire single plasmon detectors. Merging optically active nanowire structures with plasmonic structures offers a very strong potential for on chip-quantum optics experiments.

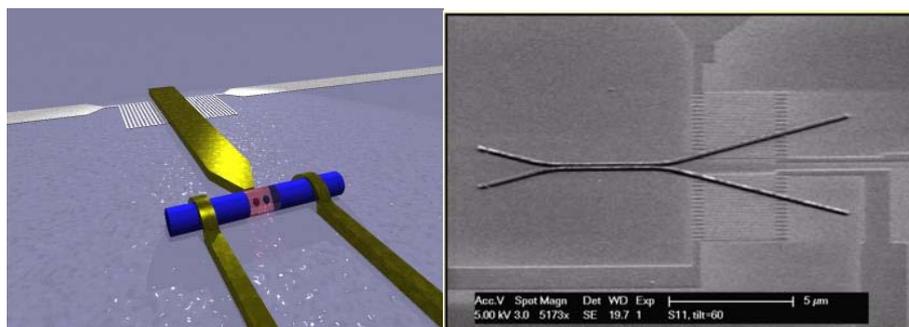


Fig 7. Left: Integrated quantum plasmonics concept. A nanowire light emitting diode is coupled to a plasmonic waveguide. Single plasmons are detected with a meandering superconducting nanowire. Right: A plasmonic beam splitter (coupled waveguide) coupled to two independent single plasmon detectors.

In conclusion, we have demonstrated that optically active quantum dots in nanowires are very good nano-optical systems with properties and functionalities that offer advantages over more conventional self-assembled quantum dots. In particular, the possibility of merging nanowire quantum dot light emitting diodes with plasmonics circuits offers a wide field of applications and experiments.

References

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