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Nanowire Quantum Dots Tuned to Atomic Resonances

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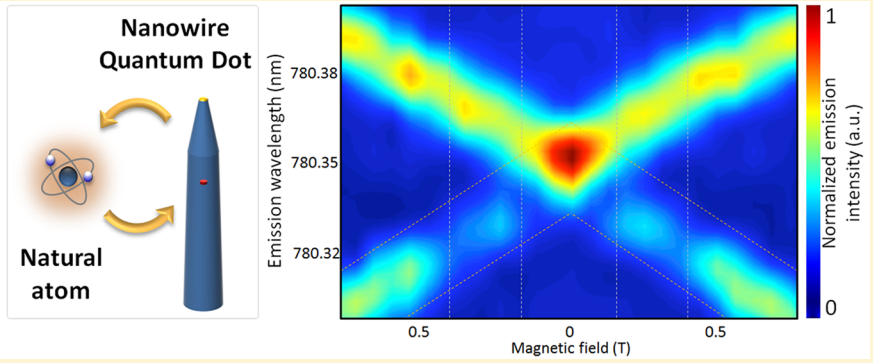
Supporting Information



ABSTRACT: Quantum dots tuned to atomic resonances represent an emerging field of hybrid quantum systems where the advantages of quantum dots and natural atoms can be combined. Embedding quantum dots in nanowires boosts these systems with a set of powerful possibilities, such as precise positioning of the emitters, excellent photon extraction efficiency and direct electrical contacting of quantum dots. Notably, nanowire structures can be grown on silicon substrates, allowing for a straightforward integration with silicon-based photonic devices. In this work we show controlled growth of nanowire-quantum-dot structures on silicon, frequency tuned to atomic transitions. We grow GaAs quantum dots in AlGaAs nanowires with a nearly pure crystal structure and excellent optical properties. We precisely control the dimensions of quantum dots and their position inside nanowires and demonstrate that the emission wavelength can be engineered over the range of at least 30 nm around 765 nm. By applying an external magnetic field, we are able to fine-tune the emission frequency of our nanowire quantum dots to the D_2 transition of ^{87}Rb. We use the Rb transitions to precisely measure the actual spectral line width of the photons emitted from a nanowire quantum dot to be 9.4 ± 0.7 μeV, under nonresonant excitation. Our work brings highly desirable functionalities to quantum technologies, enabling, for instance, a realization of a quantum network, based on an arbitrary number of nanowire single-photon sources, all operating at the same frequency of an atomic transition.

KEYWORDS: *Nanowires, quantum dots, hybrid systems, VLS growth, GaAs/AlGaAs*

Exploiting the advantages of atomic and semiconducting systems opens new possibilities and research directions. For instance, quantum dots have been frequency tuned and locked to atomic resonances to provide a single photon source with stable emission frequency.[1−3] Such hybrid systems enable multiple emitters working at the exact same wavelength, which is one of the major challenges in quantum communication and quantum information processing. Recently, a different material system has been used to tune InGaAs quantum dots to Cs atoms, optically with a laser[4] and by electrically induced strain.[5,6] Furthermore, electrically pumped quantum dots were developed to make the first quantum-LED tuned to atomic lines,[7] paving the way to on-chip electrically controlled tunable single and entangled photon sources. In addition, coupling between a single quantum dot and a single ion has been shown,[8] allowing for direct transfer of quantum information from a single quantum dot to a single atom.

Nevertheless, all previous works have used quantum dots in materials grown in bulk, while embedding the quantum dots in nanowires provide additional important functionalities to the system, making them one of the most promising systems for numerous applications in quantum nanophotonics.[9−13] Over the past few years several groups have successfully demon-

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strated nanowire quantum dots in various material systems.[14,15] Recently, the exciton energy of single nanowire quantum dots on piezo crystals was tuned into resonance by applying strain,[16] and Bell's inequality violation[17] and entanglement[18,19] have been also demonstrated. The advantages of the nanowire structure are, among others, high extraction efficiency,[10,11,20,21] the possibility of maximizing the coupling to optical fibers,[10,22,23] the positioning of nanowires in highly uniform arrays with higher uniformity than what is achievable with self-assembled quantum dots,[24−28] and the deterministic and scalable integration with photonic nanostructures that can lead to on-chip photonic circuits.[16,29,30]

GaAs quantum dots in AlGaAs structures are our preferred material system; their emission energy can be engineered close to the D_2 transitions of ^{87}Rb,[1] they are the most pure single photon sources reported to date,[31] and they show the highest degree of polarization entanglement of photons[32] generated from semiconductor quantum dots. Furthermore, GaAs quantum dots grown in AlGaAs nanowires are expected to have much sharper interfaces between the two materials, since aluminum crystallizes at least 100 times faster than gallium,[33,34] quickly leaving the droplet without aluminum once the source is closed. This allows for a higher control of the quantum dot shape, compared to the case of other material systems. The first works have already shown the growth of GaAs quantum dots in AlGaAs nanowires.[35,36] However, the optical properties were not yet optimal to address quantum applications, for instance, the emission line width spans from 95 μeV[35] to 2 meV.[36] As of today, realization of a controlled growth of high optical quality nanowire quantum dots working at atomic frequencies requires development of the material system. Here, we demonstrate precisely engineered GaAs quantum dots in AlGaAs nanowires with nearly pure crystal structure, high optical quality, and frequency-tuned to the optical transition of Rb atoms.

We grow our GaAs quantum dots in AlGaAs nanowires on a Si wafer using the vapor liquid solid technique in a Molecular Beam Epitaxy reactor, using a gold particle as catalyst. The growth takes place just below the catalyst particle, where the atoms from the vapor crystallize, forming the nanowire. To grow a quantum dot, we stop the Al source for a few seconds (e.g., 5, 7, or 15 s). This brief growth of only GaAs creates a quantum dot confined between AlGaAs segments of the nanowire. We set the growth parameters to allow for a simultaneous axial and radial growth, resulting in tapered nanowires (for details see Methods section). The radial growth results in an essential shell that protects the quantum dot from the environment outside the nanowire. We grow several sets of nanowires, with heights in the range of 3−6 μm and with diameters (core plus shell) in the range of 150−200 nm at the position of a quantum dot, depending on the growth conditions. The quantum dots have diameters defined by the size of the gold catalysts, which is about 20 nm.

In Figure 1a we present a schematics showing the quantum dot and the core−shell structure of the nanowire. Parts b and c of Figure 1 depict 2D energy-dispersive X-ray spectroscopy (EDS) elemental maps for Ga and Al, respectively. As expected, we observe an increase in the Ga content with a corresponding decrease of Al content at the quantum dot position. To clearly show the quantum dot, in Figure 1d we present 1D EDS scans along the nanowire core. We note that the expected sharp interfaces are not observed here due to the small concentration difference between GaAs and AlGaAs and

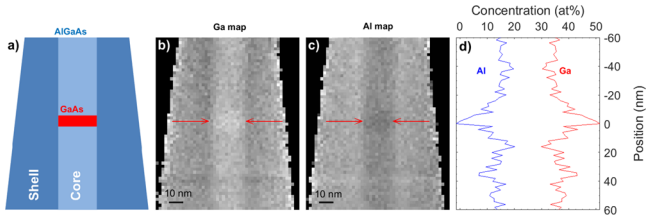

Figure 1. GaAs quantum dots in AlGaAs nanowires. (a) Schematics of a GaAs quantum dot (red) in a AlGaAs nanowire (blue), composed of a core surrounded by a shell. (b) and (c) Concentration maps of Ga and Al in a nanowire measured using STEM-EDS, respectively. The quantum dot is seen as an increase in Ga concentration and a corresponding decrease in Al concentration. (d) Concentrations of Ga (red) and Al (blue) in the nanowire core along its length, corrected for imperfect measurement technique and low material contrast (see Supporting Information section 1). All measurement in this figure are done on a sample specifically grown to have the quantum dot closer to the top of the nanowire to reduce the effect of the shell on the measurements (see Supporting Information section 1).

a beam broadening inside the material which reduces the sensitivity and the spatial resolution of the measurement.

In Figure 2a we show the photoluminescence (PL) spectra at 4.2 K from three quantum dots, grown for different times: 5,

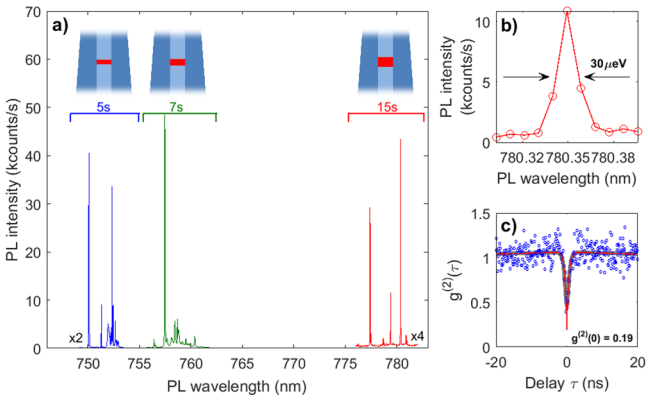

Figure 2. Optical characterization of nanowire quantum dots. (a) Photoluminescence (PL) spectra at 4.2 K of three samples with different growth times of the quantum dot: 5, 7, and 15 s (for more details see Supporting Information section 3). A schematic of the quantum dot in nanowire structure is shown above each corresponding spectra. (b) Spectral zoom on the exciton line at ∼780 nm (D_2 line of ^{87}Rb). (c) Autocorrelation measurements of an exciton (750 nm) in a nanowire quantum dot (5 s). The blue circles are the measured data and the gray line is the fit, which takes into account the finite time resolution of our detectors. The red line shows how the measurement would appear for an ideal system response (for more details see Supporting Information section 4).

7 and 15 s. As we expect, increase in the growth time increases the height of the quantum dot, which decreases the emission energy (increases the emission wavelength) of the quantum dot. As we show in Figure 2b, the PL spectra of our nanowire quantum dots has narrow line width, limited by the resolution of our spectrometer (30 μeV). In Figure 2c we plot an autocorrelation measurements of the exciton emission of a quantum dot, showing antibunching with a fitted $g^{(2)}(\tau=0) = 0.19$ (see Supporting Information section 4). Our results show that we can engineer the wavelength of the emission from our nanowire quantum dots over a range of 30

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nm around 765 nm, by selecting the growth time from 5 s (emission at 750 nm) to 15 s (emission at 780 nm). Our nanowire quantum dots are spectrally narrow and bright sources of single photons, with rates of 2.2 Mcounts/s under continuous wave excitation (see Supporting Information section 3), even without any intentional cavity integration. The count rates can be further increased, for instance, by implementing mirrors at the bottom of the nanowires[37,38] and by optimizing the waveguide design of the nanowire.[11,39] The narrow and intense lines also suggest that our nanowires are defect free and of a very high crystal purity, in some cases down to only 10 stacking faults per micrometer (see Supporting Information section 1). We note, however, that not all of our nanowires are free from defects, and not all quantum dots have excellent optical properties.

Even if one can precisely control the dimensions of quantum dots, no growth method allows obtaining systematically identical heterostructure quantum dots. In virtually all cases, the emission frequency is slightly different from one quantum dot to the other, not allowing for exchange of photons between distant quantum dots. Therefore, a reliable postgrowth technique is necessary to fine-tune each quantum dot to a frequency reference provided, for instance, by an atomic transition line. To demonstrate such tuning with our nanowire quantum dots, we interface them with a vapor of Rb atoms, similarly to our earlier work on quantum dots grown in a multistep self-assembly process in bulk.[1] In the absence of strong electric or magnetic fields, these atomic transition energies are the same, independently on time, location and number of emitters.

We use the D_2 optical transition of ^{87}Rb atoms as established frequency references to which we fine-tune the emission from our nanowire quantum dots, using an external magnetic field. This is done by means of the Zeeman effect and diamagnetic shift. During the experiment, we excite the nanowire quantum dot with a laser and then direct its emission through a cell with a vapor of ^{87}Rb (see Figure S11 for schematics). Varying the applied magnetic field, we change the frequency of the emitted photons. The photons propagate through the Rb cell and as their frequency matches the atomic transitions, they get absorbed or scattered. The transmission of photons through the Rb cell is then measured.

In Figure 3, we show the results of our experiment. We scan the magnetic field from 0.7 to 0 T and then back to 0.7 T for reproducibility. We then trace the short-wavelength branch of the Zeeman-split lines (highlighted by dashed lines in Figure 3a) and analyze the transmitted intensity, shown on Figure 3b. We clearly resolve two transmission dips due to the hyperfine structure of the D_2 lines of ^{87}Rb. Since the experiment is repeated twice (from 0.7 to 0 T and from 0 to 0.7 T), Figure 3b shows four dips. We then fit the data to our model[1] (see Supporting Information section 6) and take advantage of our hybrid system to perform high-resolution spectroscopy of our nanowire quantum dot, similarly to laser spectroscopy.[3] The fit, which has only one fitting parameter (the spectral line width of the emission) reveals a narrow exciton line of 9.4 μeV. To the best of our knowledge, this is the narrowest line width reported for GaAs nanowire quantum dots[15,40,41] and can be further improved by exciting the quantum dots resonantly.

In summary, we developed a technique for controlled growth of GaAs quantum dots in AlGaAs nanowires on Si substrates, where we precisely engineer the size and position of the dots inside the nanowire. Our nanowires are nearly defect free, and we can design the quantum dot to emit single photons anywhere in the range of 30 nm around 765 nm. These nanowire quantum dots are bright emitters with very narrow spectral line width, down to 9.4 μeV. We demonstrated the tuning of their emission to the D_2 lines of ^{87}Rb, enabling operation of our nanowire quantum dots at a stable and precise reference frequency. The hybrid system we present in this work combines the reliability and stability of natural atoms with a variety of functional advantages of nanowire quantum dots. Nanowire quantum dots tuned to atomic resonances could be one of the key elements in quantum information networks, on-chip photonic circuits, and in generation of multidimensional photonic cluster states.[42]

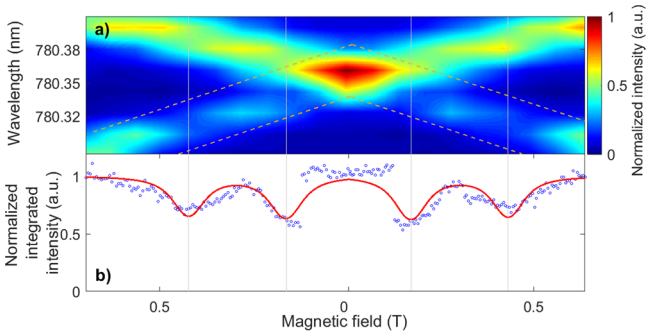

Figure 3. Transmission through Rb vapor. (a) PL spectra of an exciton in a nanowire quantum dot scanned through the atomic transitions. The experiment is done by swiping the magnetic field from 0.7 to 0 T and then back to 0.7 T. Colors represent the normalized PL intensity. The data are then interpolated for clarity (see raw data in Figure S12). (b) Transmission of the exciton line through the Rb vapor, obtained by tracing the short wavelength branch of the Zeeman-split exciton in the raw data, marked by dashed lines in (a). Blue points refer to the experimental data, while the red line represents the fit to our model. The fit reveals a very narrow emission line width of 9.4 ± 0.7 μeV.

Methods. We grow our nanowires on a Si(111) wafer. The wafer is first outgassed at 850 °C and then the Au is deposited with total thickness of 0.1 nm at 550 °C. After deposition, the Au layer will form droplets of about 20 nm in diameter, as this formation minimizes the potential energy. In order to improve the homogeneity of the Au droplet size, the substrate is kept for 1 min at 550 °C; this time is long enough to ensure that the droplets become similar in size. Afterward the substrate is cooled to room temperature and transferred to the growth chamber with no vacuum brake. During the nanowire growth the substrate temperature is set at 510 °C, the V/III flux ratio is 3, and the total AlGaAs growth rate is fixed at 0.3 nm/s, for all samples in this work. The AlGaAs part of the nanowire is grown for 20 or 25 min, depending on the desired height of the final nanowire, and then the Al source is closed for 5, 7, or 15 s in order to form the GaAs quantum dots. Afterward, the supply of Al is opened again for 5 min to continue the nanowire growth. The growth method results in tapered core−shell nanowires (see Figure 1a). The Au droplet diameter will determine the nanowire core diameter, and hence the quantum dot diameter. As the distribution of the size of the Au droplets is ensured to be homogeneous, the nanowire core and the quantum dot for all samples will have a diameter close to 20 nm. The diameter of the total (core+shell) nanowire is in the range 150−200 nm at the position of the quantum dot, depending on the total AlGaAs growth time. The height of the quantum dot inside the nanowire is given by the time in which

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the Al shutter is closed (5, 7, or 15 s in these samples). The Al shutter closing time is 0.3 s and thus allows accurate control the quantum dot height. The density of the nanowires was kept low during growth (see Figure S1), in order to be able to distinguish individual nanowire quantum dots.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.8b03363.

Electron microscopy images of the samples, EDS line scan corrections, further optical characterization measurements, magneto-luminescence data, fitting procedures and details on data interpolation used in the main text (PDF)

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Notes

The authors declare no competing financial interest.

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