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Self-assembled quantum dots in a nanowire system for quantum photonics

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Quantum dots embedded within nanowires represent one of the most promising technologies for applications in quantum photonics. Whereas the top-down fabrication of such structures remains a technological challenge, their bottom-up fabrication through self-assembly is a potentially more powerful strategy. However, present approaches often yield quantum dots with large optical linewidths, making reproducibility of their physical properties difficult. We present a versatile quantum-dot-in-nanowire system that reproducibly self-assembles in core–shell GaAs/AlGaAs nanowires. The quantum dots form at the apex of a GaAs/AlGaAs interface, are highly stable, and can be positioned with nanometre precision relative to the nanowire centre. Unusually, their emission is blue-shifted relative to the lowest energy continuum states of the GaAs core. Large-scale electronic structure calculations show that the origin of the optical transitions lies in quantum confinement due to Al-rich barriers. By emitting in the red and self-assembling on silicon substrates, these quantum dots could therefore become building blocks for solid-state lighting devices and third-generation solar cells.

Semiconductor quantum dots have been shown to be excellent building blocks for quantum photonics applications, such as single-photon sources and nano-sensing. Desirable properties of a single-photon emitter include high-fidelity anti-bunching (very small $g^2(t=0)$), narrow emission lines (ideally transform limited to a few microelectronvolt) and high brightness (>1 MHz count rate on standard detector). For simplicity, these properties should be achieved either with electrical injection or non-resonant optical excitation. Desirable properties of a nano-sensor include a high sensitivity to local electric and magnetic fields, with the quantum dot located as close as possible to the target region. A popular realization involves Stranski Krastanow InGaAs quantum dots embedded in a three-dimensional matrix, which are excellent building blocks for the realization of practical single-photon sources[1]. However, the photon extraction out of the bulk semiconductor is highly inefficient on account of the large mismatch in refractive indices of GaAs and vacuum. An attractive way forward is to embed the quantum dots in a nanowire[2]. To solve the extraction problem, the nanowire is designed to operate as a single-mode waveguide, a so-called photonic nanowire, with a taper as photon out-coupler[3]. Also, for nano-sensing applications, a quantum dot in a nanowire can be located much closer to the active medium. Top-down fabrication of the photonic waveguide is technologically complex, however. Bottom-up fabrication of the photonic waveguide is very attractive[4 6], but it is at present challenging to self-assemble quantum dots in the nanowires with narrow linewidths and high yields[7,8]. Nano-sensing applications are at present not highly developed. Other degrees of freedom of the quantum-dot-in-nanowire system that can be usefully exploited are the mechanical modes for optomechanics, and doping for p n junctions with applications in light harvesting[9,10].

Here we present a promising new quantum-dot-in-nanowire system. A schematic of the physical structure is shown in Fig. 1a. The structure consists of Al-poor $Al_xGa_{1-x}As$ ($x \sim 10\%$) quantum dots in an Al-rich $Al_xGa_{1-x}As$ ($x \sim 60\%$) barrier wrapped in an intermediate Al-content matrix ($x \sim 33\%$). The quantum dots form in the ridge of an AlGaAs nanowire. The self-assembly is driven by the different Ga and Al adatom mobilities on the nanowire surface, leading to Al segregation. The quantum dots can be positioned close to the nanowire surface or close to the nanowire core during the growth simply by choosing the growth mode, lateral or radial, and the overall diameter of core and shell. We note that the quantum dot size is independent of the core diameter. Significantly, the nanowire growth is not complicated by fluctuations in crystal structure (polytypism). We find that the quantum dots are very stable, surviving prolonged electron beam bombardment, exposure to air and so on, and that the quantum-dot-in-nanowire growth is very reproducible from one run to the next: there is a wide window of parameters under which they form. The quantum dots have excellent optical properties even when they are located just a few nanometres from the surface: individual quantum dots are very bright (Megahertz count rate) even without engineering the

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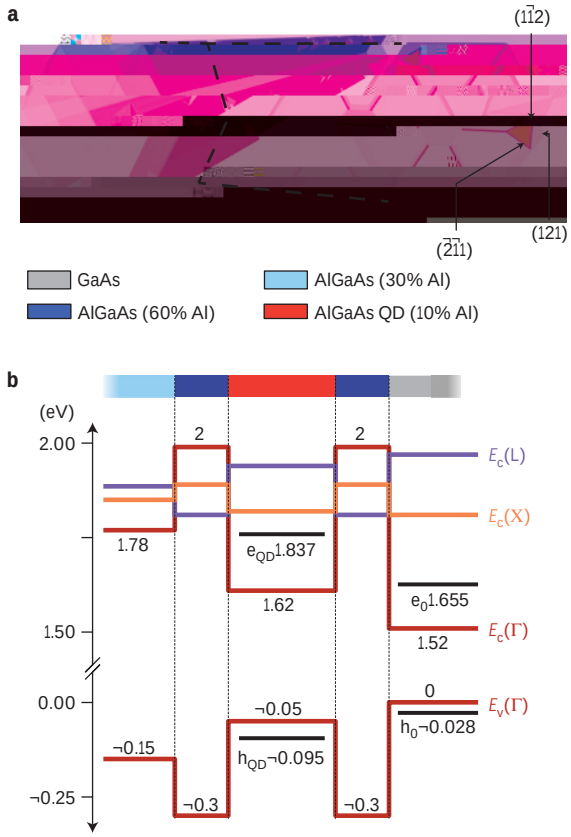

Figure 1 | Schematics of the quantum-dot-in-nanowire system. **a**, The nanowire consists of GaAs. The quantum dot forms close to the outer edge of an $Al_xGa_{1-x}As$ shell. Aluminium segregates at the nanowire edges owing to the lower mobility of aluminium. At the outer edge of the $Al_xGa_{1-x}As$ layer, Al segregates further in the [112] directions, leading to Al depletion and the formation of a nanoscale inclusion, an Al-poor $Al_xGa_{1-x}As$ quantum dot. **b**, The band edge diagram showing from left to right the $Al_xGa_{1-x}As$ matrix and barriers, the lowest energy states confined to the quantum dot and the external GaAs capping taken from atomistic pseudopotential theory.

Figure 2 | Structure of quantum-dots-in-a-nanowire. **a**, Aberration-corrected high-angle annular dark-field STEM image of the entire cross-section of a GaAs nanowire coated with multiple $Al_{0.33}Ga_{0.77}As$/GaAs shells. **b**, Zoom-in of **a**. **c**, Detail of the Al-poor

photonic modes, the linewidths are small (sub-100 eV) and the photons are highly anti-bunched (the upper bound on $g^2(t=0)$ is just 2%) even with intense non-resonant excitation.

An unusual feature is the blue-shift of the quantum dot emission relative to emission from electrons and holes in the lowest energy continuum states, in this case emission from the GaAs substrate, the core. This wavelength ordering of quantum dot and continuum emission is reversed relative to Stranski Krastanow quantum dots. We interpret this unusual result with large-scale calculations using both the empirical pseudopotential method (modelling explicitly 500,000 atoms in this dot-in-wire structure) and density functional theory (modelling a wire geometry with up to 12,000 atoms). The calculations show that whereas the states at the band edge of the system as a whole are indeed located in the GaAs layers, states at higher energy exist, confined to the quantum dot. The results are summarized in an energy level diagram, Fig. 1b, which shows the band edge valence and conduction states, h_0 and e_0, and the quantum dot valence and conduction states, h_{QD} and e_{QD}. For Stranski Krastanow dots, the continuum states are associated with the wetting layer, a thin 2D layer connecting the quantum dots, and lie at higher energy than the lowest energy quantum dot states, e_0 and h_0. For the quantum-dot-in-nanowire system presented here, this energy reversal of quantum dot and continuum emission represents a new Ansatz for a solid-state single-photon emitter.

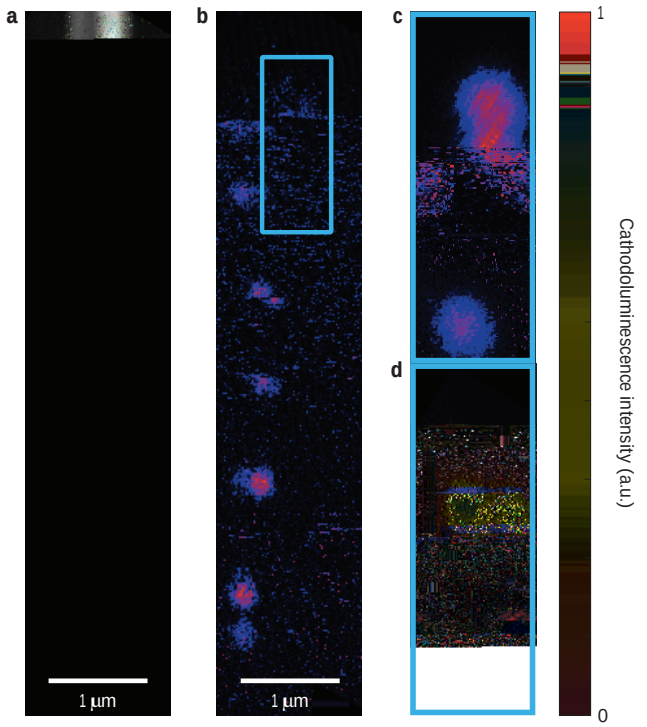

Figure 3 | Cathodoluminescence of a single nanowire. a, Electron microscopy image of a GaAs nanowire with a $Al_{0.75}Ga_{0.25}As$ shell. **b**, Cathodoluminescence mapping of a nanowire detecting emission at 677 nm. **c**,**d**, Detailed cathodoluminescence map showing spatially localized cathodoluminescence centres corresponding to quantum dots less than 200 nm apart on two adjacent edges of the nanowire emitting at 677 nm.

nanometres in extent with low Al content: this region constitutes the quantum dot. Figure 2c shows how the Al-rich stripe following the (101) plane bifurcates into two Al-rich stripes parallel to the (112) and (211) planes, forming a Y-like shape, terminated on a

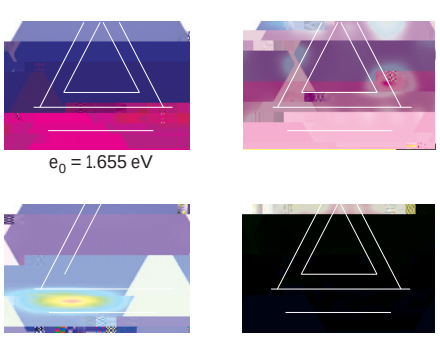

Figure 5 | Atomistic calculations of electronic states in a quantum-dot-in-nanowire system. a, Pseudopotential eigenstate densities for a 10% Al content $Al_xGa_{1-x}As$ quantum dot, 9 nm high, surrounded by a 2.5 nm thickness 60% AlGaAs barrier embedded in an 30% AlGaAs matrix and sitting on a pure GaAs substrate (4 nm thickness in computational cell). Plotted are the lowest energy conduction and valence states (e_0 and h_0, respectively) and the lowest energy quantum dot-bound conduction and valence states (e_{19} and h_{67}, respectively). **b**–**d**, The localized Kohn–Sham states nearest the band edges, computed using self-consistent linear-scaling density functional theory, and viewed along the [1$\bar{1}$1]

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