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# Enhanced photon extraction from a nanowire quantum dot using a bottom-up photonic shell

Mathieu Jeannin,<sup>1,\*</sup> Thibault Cremel,<sup>2,\*</sup> Teppo Häyrynen,<sup>3</sup> Niels Gregersen,<sup>3</sup> Edith Bellet-Amalric,<sup>2</sup> Gilles Nogues,<sup>1,†</sup> and Kuntheak Kheng<sup>2</sup>

<sup>1</sup>*Univ. Grenoble Alpes, CNRS, Institut Néel, "Nanophysique et semiconducteurs" group, F-38000 Grenoble, France*

<sup>2</sup>*Univ. Grenoble Alpes, CEA, INAC, PHELIQS, "Nanophysique et semiconducteurs" group, F-38000 Grenoble, France*

<sup>3</sup>*DTU Fotonik, Department of Photonics Engineering, Technical University of Denmark, Ørsted's Plads, Building 343, DK-2800 Kongens Lyngby, Denmark*

## Abstract

Semiconductor nanowires offer the possibility to grow high quality quantum dot heterostructures, and in particular CdSe quantum dots inserted in ZnSe nanowires have demonstrated the ability to emit single photons up to room temperature. In this letter, we demonstrate a bottom-up approach to fabricate a photonic fiber-like structure around such nanowire quantum dots by depositing an oxide shell using atomic layer deposition. Simulations suggest that the intensity collected in our NA=0.6 microscope objective can be increased by a factor 7 with respect to the bare nanowire case. Combining micro-photoluminescence, decay time measurements and numerical simulations, we obtain a 4-fold increase in the collected photoluminescence from the quantum dot. We show that this improvement is due to an increase of the quantum dot emission rate and a redirection of the emitted light. Our ex-situ fabrication technique allows a precise and reproducible fabrication on a large scale. Its improved extraction efficiency is compared to state of the art top-down devices.

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\* contributed equally to this work

† gilles.nogues@neel.cnrs.fr

## 23 I. INTRODUCTION

24 Controlling and enhancing the spontaneous emission of quantum emitters is one of the  
25 current key issues in the field of nanophotonics. Semiconductor quantum dots (QDs) are  
26 considered as promising and efficient single-photon emitters for quantum optics applications.  
27 [1–6] Over the past few years, several approaches have been pursued to control their emis-  
28 sion properties, from the use of photonic crystals [7, 8] to top-down photonic wires [9–11]  
29 and trumpets.[12, 13] These strategies are based on the early work of Purcell[14] which  
30 demonstrated that the spontaneous emission of an emitter can be modified by engineering  
31 its electromagnetic environment. They rely on a waveguiding approach to increase the cou-  
32 pling between a well-defined propagating optical mode and the QD while simultaneously  
33 reducing the coupling between the QD and background radiation modes, offering control of  
34 both the optical mode profile and the QD spontaneous emission rate.

35 In this context, the interest of the dot-in-a-nanowire configuration fabricated using  
36 bottom-up methods naturally arises because it provides a simple way to ensure the center-  
37 ing of a single quantum emitter in the photonic structure.[15–17] The bottom-up fabrication  
38 method also avoids heavy processing, like etching the semiconducting material, that is often  
39 detrimental to the QDs optical properties. However, the main realizations up to now concern  
40 III-V semiconductors, [15–17] limiting the operation range to the cryogenic temperature.  
41 Tackling this issue, the potential of II-VI materials, in particular CdSe QDs inserted inside  
42 ZnSe nanowires (NWs) has been demonstrated in previous studies. They allow for robust  
43 high temperature single-photon emission using heteroepitaxial [18] or homoepitaxial [19]  
44 nanowire growth. Contrary to all the aforementioned systems where the photonic wire  
45 structure has a diameter comparable to the wavelength  $\lambda/n$  of the guided light which allows  
46 for highly efficient coupling to the  $HE_{11}$  mode[20], the diameter of the II-VI NW embedding  
47 the QD ( $\sim 20$  nm) is much smaller than the wavelength of the emitted light (530 nm). It  
48 leads to light emission predominantly into non-guided radiation modes and a low collection  
49 efficiency. An additional fabrication effort has thus to be made to ensure an efficient coupling  
50 to the collection optics.

51 In a previous report[21] we have theoretically investigated the potential of using an oxide  
52 shell deposition on a bare ZnSe NW to form a thick photonic wire structure. In this article,  
53 we experimentally demonstrate the use of atomic layer deposition (ALD) to fabricate a

conformal aluminum oxide ( $\text{Al}_2\text{O}_3$ ) shell around ZnSe NWs containing a single CdSe QD. We show that the oxide shell drastically enhances the light intensity emitted by the QD, and we use time-resolved microphotoluminescence to systematically study the effect of the shell thickness on the nanowire quantum dot (NWQD) emission rate. Our results are compared to numerical simulations accounting for the real NW geometry, evidencing the different physical mechanisms leading to the enhancement of the spontaneous emission from the QD and to the improved light collection from the emitting structure.

## II. PRINCIPLES OF OPERATION

To illustrate the effect of the NW and its surrounding medium on the QD emission rate, let us consider a QD placed inside an infinitely long cylinder as illustrated in Fig. 1(a) radiating a field at a wavelength  $\lambda$ . The cylinder is made of a dielectric material (refractive index  $n$ ) and has a diameter  $d$ . We first consider a dipole orientation perpendicular to the NW axis in order to use the NW as a propagation medium for the emitted light. In the limit where  $d \ll \lambda/n$ , the dielectric screening effect[11] reduces the spontaneous emission rate  $\gamma$  by a factor:

$$\frac{\gamma}{\gamma_0} = \frac{4}{n(n^2 + 1)^2}, \quad (1)$$

where  $\gamma_0$  is the radiative emission rate in the bulk material of index  $n$ . [22] For a ZnSe cylinder ( $n_{\text{ZnSe}} = 2.68$  at  $\lambda=530$  nm), the screening factor is  $\sim 1/45$ . If the NW is surrounded by a shell of refractive index  $n_s$  instead of vacuum, equation 1 remains valid by replacing  $n$  with the index contrast  $n/n_s$ . For an  $\text{Al}_2\text{O}_3$  surrounding medium ( $n_s = 1.77$ ), the screening factor becomes  $\sim 1/4.1$ , resulting in an order of magnitude larger radiative rate.

In addition to changing the dielectric screening, the  $\text{Al}_2\text{O}_3$  shell also influences the guiding of light along the NW. We have computed the total emission rate  $\gamma$  and the emission rate  $\gamma_{\text{HE11}}$  into the fundamental  $\text{HE}_{11}$  waveguide mode from a radial dipole as function of the shell thickness  $t_s$  [see Fig. 1(a)] using a semi-analytical approach[23] combined with an efficient non-uniform discretization scheme in  $k$  space.[24] The results are plotted in Figure 1(b). We observe that the shell thickness of  $\sim 120$  nm not only leads to an increased total emission rate, it also allows for confinement of the fundamental  $\text{HE}_{11}$  mode to the core-shell NW leading to a preferential coupling of the emitted light to this mode. Figure 1(c) presents the spontaneous emission  $\beta$  factor representing the fraction  $\beta = \gamma_{\text{HE11}}/\gamma$  of emitted light coupled

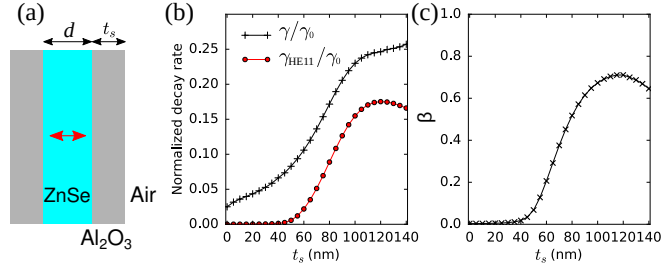


Figure 1. (a) Geometry of the infinite NW. (b) Total spontaneous emission rate (black +) and spontaneous emission rate into the first guided mode HE<sub>11</sub> (red •) as a function of shell radius for a radial dipole. (c) Fraction  $\beta$  of power radiated into the HE<sub>11</sub> mode.

83 to the HE<sub>11</sub> mode. We observe indeed that up to 71% of the emitted light is coupled to this  
84 mode for  $t_s=120$  nm. The dipole thus becomes coupled to the equivalent of a monomode  
85 photonic wire[9–11, 15–17] paving the way to the control of its far-field radiation pattern.

### 86 III. SAMPLE FABRICATION

87 Our emitters are CdSe QDs embedded inside a ZnSe NW with a thin, epitaxial passivation  
88 Zn<sub>0.83</sub>Mg<sub>0.17</sub>Se shell grown around the NW. They are grown by molecular beam epitaxy on a  
89 GaAs(111)B substrate. A ZnSe buffer layer is first grown on the GaAs substrate after which  
90 a thin layer of Au (less than one monolayer thick) is evaporated on the sample surface and  
91 dewetted at 510 °C to form small ( $\sim 10$  nm diameter) Au droplets that serve as a catalyst  
92 for the NW growth. The substrate temperature is then set at 400 °C and a flux of Zn and Se  
93 atoms with an excess of Se is used, inducing preferential growth of vertical ZnSe NWs. The  
94 NWs are in wurtzite phase and their diameter is the same as the droplet ( $\sim 10$  nm diameter).  
95 The thickness of the initial Au layer is chosen to ensure a low NW density ( $\leq 1$  NW per  
96  $\mu\text{m}^2$ ). After the growth of a 400 nm high NW, the atom fluxes are stopped to allow the  
97 evacuation of residual Se atoms inside the droplet. Then, the QD is grown under a flux of Cd  
98 and Se atoms for 20 s. The fluxes are interrupted again before the ZnSe growth is resumed,  
99 resulting in an expected QD height of 2-3 nm inserted in a  $\sim 700$  nm high NW. Finally, an  
100 epitaxial Zn<sub>0.83</sub>Mg<sub>0.17</sub>Se shell (5 nm thick) is grown around the NW at 220 °C. A scanning  
101 electron microscope (SEM) image of such a CdSe/ZnSe/ZnMgSe core/shell NWQD system  
102 is presented in Figure 2(a). The flag-shape termination of the NW is formed during the

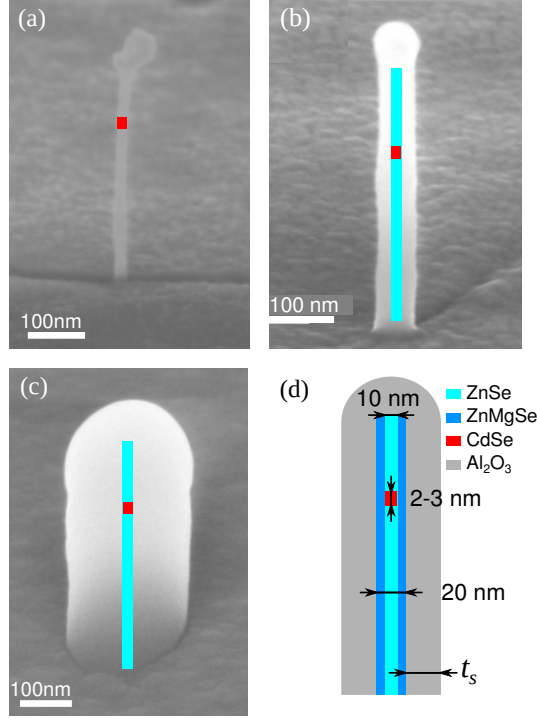


Figure 2. (a) SEM image of a standing ZnSe/ZnMgSe NW embedding a CdSe QD. The QD position is marked by the red square. (b,c) Tilted SEM image of a ZnSe NW after a 20 nm and 110 nm thick  $\text{Al}_2\text{O}_3$  shell deposition respectively. The NW is sketched on the SEM image. Note the circular shape of the shell as well as its hemispherical termination above the NW apex. (d) Sketch of the NWQD geometry, indicating the QD height (2-3 nm), the NW diameter ( $\simeq 10$  nm), the epitaxial shell thickness ( $\simeq 5$  nm) and the ALD shell thickness  $t_s$ .

103 growth of the ZnMgSe shell. It is present in some NWs.

104 The higher bandgap of  $\text{Zn}_{0.83}\text{Mg}_{0.17}\text{Se}$  shell prevents the charge carriers to recombine  
 105 non-radiatively on the ZnSe NW sidewall and hence improves the quantum yield of the  
 106 CdSe emitter. In principle, it could directly be used to grow a photonic wire of diameter  
 107  $\sim \lambda/n_{\text{ZnSe}}$  around the NWQD. However, during the epitaxial shell growth two phenomena  
 108 are competing: the radial growth of the shell around the wurtzite NWs, and the vertical  
 109 growth of a 2D  $\text{Zn}_{0.83}\text{Mg}_{0.17}\text{Se}$  layer on the sample surface. The radial shell growth rate is  
 110 very low because the growth of ZnSe on WZ surfaces is not favourable. Because of this low  
 111 shell growth rate, a trade-off has to be found to avoid burying the NWs in a  $\text{Zn}_{0.83}\text{Mg}_{0.17}\text{Se}$   
 112 matrix. As a result, only thin epitaxial shells can be fabricated.

113 The complexity of creating a thick epitaxial shell is one of the reasons why we fabricate

114 the photonic structure by depositing an oxide shell around the NW using ALD. Another  
 115 reason is that, since this process step can be done *separately* from the NW growth process,  
 116 it allows to tune ex situ the shell parameters after a first optical characterization of the  
 117 QD. Indeed, due to its slow deposition rate, the ALD process allows to precisely control the  
 118 deposited thickness, which can also be finally verified using scanning electron microscopy.  
 119 We have tested several oxide materials, and selected  $\text{Al}_2\text{O}_3$  because it produced very smooth  
 120 and conformal, amorphous shells. Figure 2(b) and (c) show two SEM images of the result-  
 121 ing oxide shell deposition (20 nm and 110 nm), and the complete structure is sketched in  
 122 Fig. 2(d). We note that the conformal deposition allows to end the NW+shell structure by  
 123 an almost perfect half-sphere as can be seen in Fig. 2(b,c). ALD also buries the Au droplet  
 124 under the shell. The latter might interact with the field emitted by the QD through its  
 125 localized plasmon resonance. Considering its small diameter it will essentially absorb the  
 126 incoming field. Moreover the guided HE11 mode profile presents a minimum on the NW  
 127 axis. This is why we neglect the droplet influence in the following.

#### 128 IV. EXPERIMENTAL RESULTS

129 A sample from a single epitaxial growth process is cut in pieces, and photonic structures  
 130 with different oxide shell thicknesses are fabricated. Taking advantage of the low NW den-  
 131 sity, individual structures are optically characterized directly on the growth substrate. The  
 132 samples are mounted on the cold finger of a He-flux cryostat and cooled down to 4 K. Indi-  
 133 vidual photonic structures are probed using confocal microphotoluminescence ( $\mu\text{PL}$ ). They  
 134 are excited by a supercontinuum pulsed laser (Fianium WhiteLase, 10 ps pulse duration,  
 135 repetition rate 76 MHz) and a spectrometer selecting a 10 nm bandwidth centered around  
 136 485 nm. This excitation energy, below the ZnSe gap, allows us to induce crossed transitions  
 137 between delocalized states in the NW 1D continuum and a discrete confined 0D state in the  
 138 NWQD band structure[25]. In this configuration, the NW axis is aligned with the optical  
 139 axis and emission from the QD is collected by a  $NA = 0.6$  objective. A typical NWQD  
 140 spectrum is presented in Figure 3(a) as a function of the pump laser power. Three lines  
 141 can be identified and are attributed to the exciton (X), the charged exciton (CX) and the  
 142 bi-exciton (XX) respectively. The total emission intensity of the X line as a function of the  
 143 pump power is reported in Figure 3(b). It shows a linear increase at low pumping power, and

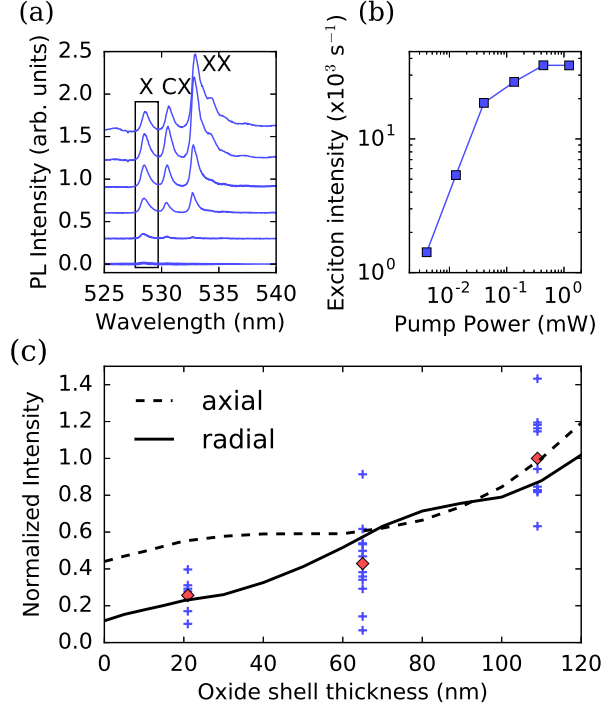


Figure 3. (a)  $\mu$ PL spectrum of a NWQD with 120 nm thick photonic shell for different pumping powers. It shows a exciton (X), charged exciton (CX) and biexciton (XX) lines. The corresponding pumping powers are reported in panel (b). The black rectangle indicates the integration bandwidth used to extract the total exciton emission intensity (X line). (b) Integrated exciton emission intensity as a function of pumping power, in a log-log scale. (c) Blue crosses: Total exciton emission intensity for different NWQDs as a function of the oxide shell radius. Red diamonds: average of the experimental data points. Data are normalized to the average intensity at  $t_s = 110$  nm Black lines: results of the numerical simulations for a radial (solid line) and an axial (dashed line) dipole. They are normalized to the axial intensity at  $t_s = 110$  nm

144 a constant plateau at high pumping powers corresponding to the saturation of the exciton  
 145 level.[26] Under pulsed excitation, we note that changing the shell thickness might modify  
 146 the laser power in the NW and the excitation probability of the QD. Hence it affects the  
 147 slope at low power in Fig. 3(b). It has however no effect on the saturation plateau which only  
 148 depends on the QD emission rate and light collection efficiency. This allows us to compare  
 149 statistical sets of nanostructures with different oxide shell thicknesses. The total integrated  
 150 emission at saturation as a function of the oxide shell thickness is reported in blue markers  
 151 for each NWQD in Figure 3(c). The values have been normalized to the average intensity



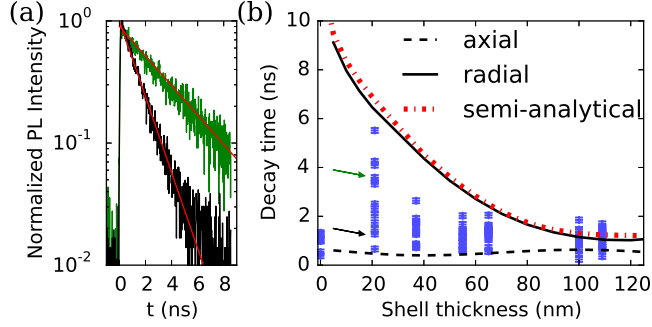


Figure 4. (a) Example of TRPL signal versus time for 2 NWs with a  $t_s=20$  nm shell. Background counts are measured for  $t < 0$  and subtracted. Amplitude of counts are normalized to 1 to compare the 2 datasets. Red lines are mono-exponential fits, whose corresponding points in (b) are shown by arrows. (b) Blue crosses: experimental exciton decay times for several QDs as a function of the oxide shell radius. The vertical error bars represent the fit error. Black lines: numerical simulation results for a radial dipole (solid line), or an axial dipole (dashed line). Red dashed-dotted line: Semi-analytical calculations for the infinite NW.

at  $t_s = 110$  nm. For NWs without an oxide shell, the luminescence intensity is very low and we were never able to reach the saturation regime, this is why we do not report the corresponding points in Fig. 3(c). For each shell thickness, we observe a large spread in exciton saturation intensity. However, we note a general trend of increasing saturation intensity with increasing shell thickness, as demonstrated by the red markers which show the position of the average intensity of our measurements for each shell thickness. On average, the position of a 110 nm thick shell results in the experiments in an almost 4-fold enhancement of the collected intensity with respect to the 20 nm thick shell case. The semi-analytical calculations show that this enhancement is 10-fold when we compare to a NW without oxide shell.

The observed increase in intensity at saturation corresponds to the combination of improved collection efficiency through light redirection from the structure and enhancement of the spontaneous emission rate. In the latter case, a modification of the QD dynamics is expected to be detected by measuring the exciton decay rate. Time-resolved measurements were carried out using a low pump power as compared to the exciton saturation power to avoid any repopulation of the X level. The measured decay transients are thus monoexponential. The fitted decay constant is the total exciton decay time  $\tau$ . The experiment

169 was carried out in another setup on a different set of photonic structures compared to the  
 170 one of figure 3(c). The same excitation laser was used, the QD fluorescence was spectrally  
 171 filtered in a spectrometer (500gr/mm grating) and integrated on an avalanche photodiode  
 172 in a photon correlation setup, using the exit slit of the spectrometer as a spectral bandpass  
 173 filter. The results of these measurements, presented in Figure 4 show also a great dispersion  
 174 in decay time. One observes however that longer lifetimes are observed for smaller shell  
 175 thickness (up to 5.9 ns). Increasing the shell thickness leads to an overall decrease in the  
 176 measured exciton lifetime, hence an enhancement of the exciton decay rate in agreement  
 177 with the results of the numerical simulations. For systems without an oxide shell, only a  
 178 few NWQDs give a large enough signal to be properly measured. They yield a much smaller  
 179 dispersion of short decay times.

## 180 V. DISCUSSION AND COMPARISON TO NUMERICAL SIMULATIONS

### 181 A. Dispersion of the results

182 For each oxide shell thickness, the large variations of the experimental results in both  
 183 Figs. 3(c) and 4 have several possible origins. First, the presence of non-radiative recom-  
 184 bination channels can reduce the intensity at saturation and change the decay time. The  
 185 non-radiative recombination rate can vary from QD to QD because of fabrication inhomo-  
 186 geneities, leading to a spread in the measured values.[27] Second, variations in the QD aspect  
 187 ratio and piezoelectric fields induced internal strain applied by both the ZnSe core and the  
 188  $\text{Zn}_{0.83}\text{Mg}_{0.17}\text{Se}$  shell lead to different overlap of electron and hole wavefunctions and hence  
 189 different exciton oscillator strengths. Finally, considering the QD aspect ratio and internal  
 190 strain, we expect a heavy-hole exciton type for our QDs.[28–31] Heavy-hole exciton recom-  
 191 bination results in a mixture of circularly polarized emission, composed of two degenerate  
 192 out-of-phase radial dipoles. However, strain and confinement effects might lead to valence  
 193 band mixing between light hole and heavy hole levels,[32–34] resulting in an emission com-  
 194 posed of a mixture between axial and radial dipoles and hence to a spread in total emitted  
 195 intensity, as we discuss later. Additional measurements on NWQDs grown in similar condi-  
 196 tions and mechanically dispersed on a substrate (i.e. lying horizontally on it) revealed that  
 197 one NWQD out of 6 emit light polarized along the NW axis, while others emit light polarized

perpendicularly to the NW axis, evidencing the presence of both kinds of dipoles. Due to the  $\text{Zn}_{0.83}\text{Mg}_{0.17}\text{Se}$  shell and low temperature of observation, we expect that non-radiative effects play a minor role. The epitaxial shell prevents non-radiative decay channels owing to surface traps. Additional measurements as a function of temperature show that both the emission intensity and the decay time do not change significantly up to 150-200 K (not presented here). This indicates that the non-radiative effects are not dominating at low temperature, as in the present experiment. While we cannot yet completely rule out the contribution of non-radiative effects, we think that the major effect to explain the dispersion of the results comes from variations in valence band mixing and oscillator strength due to the local environment of the QD. Finally let us stress that the shortest decay times (1-2 ns) we measure remain longer than the decay time of CdSe self-assembled QD embedded in bulk ZnSe ( $<1$  ns)[35]. The reduction of the dielectric screening effect is a main effect we evidence.

## B. Collected intensity and radiative lifetime

To better understand the effect of the shell deposition on the NWQD emission, we perform numerical simulations of the photonic structure formed by the full NW + oxide shell geometry [see Fig. 2(d)]. It takes into account the presence of the ZnSe substrate, and the  $\text{Al}_2\text{O}_3$  shell and layer deposited on the NWs and substrate. The QD is modeled as an oscillating electric dipole, either in the axial direction (along the NW axis) or in the radial direction (orthogonal to the NW axis). We perform finite-element method simulations (Comsol v4.1) to compute the total field radiated by the dipole.[34] For each shell thickness and dipole orientation, we evaluate the power radiated towards the objective by computing the flux of the Poynting vector over a surface limited by its numerical aperture ( $\text{NA}=0.6$ ) in a region far from the NW where near field can be neglected. The results of these simulations are reported in Figure 3(c) in black lines for an axial (dashed line) or radial (solid line) dipole. The results are normalized to the axial intensity at  $t_s=110$  nm. Comparing the simulated integrated intensity in the case of a 20 nm and 110 nm reveals an enhancement factor less than 2-fold for an axial dipole and almost 4-fold for a radial dipole. The 4-fold enhancement observed in our measurements suggests that on average, the dominant emitting dipole in our structure is radial, in good agreement with the recombination of a heavy hole exciton.

228 The theoretical limits for the radiative lifetimes is extracted from the numerical sim-  
 229 ulations by integrating the total power radiated over every direction for the two dipole  
 230 orientations (radial and axial)  $P$ . We normalize this value by the same quantity computed  
 231 for a dipole in bulk ZnSe  $P_0$ . For a purely radiative system we have  $P/P_0 = \gamma/\gamma_0 = \tau_0/\tau$  [36],  
 232 where  $\tau$  and  $\tau_0$  are the radiative lifetime for the nanostructure and for bulk ZnSe respectively.  
 233 Radiative times are presented in black lines in Figure 4, where we have chosen  $\tau_0 = 300$  ps  
 234 in good agreement with previously reported radiative lifetime of CdSe QD in bulk ZnSe[37].  
 235 The axial dipole radiates with an almost constant decay time as a function of the oxide shell  
 236 thickness, while the radial dipole decay time strongly decreases with increasing oxide shell  
 237 thickness  $t_s$ . Additionally, we compare the decay time for the radial dipole computed for the  
 238 full geometry to the semi-analytical calculations for the infinite NW presented in fig. 1(b)  
 239 with the same  $\tau_0$  value. The agreement is excellent indicating that interference effects due  
 240 to reflections from the substrate and from the top hemispherical termination are negligible.

241 Comparing the trends of the simulations, we can confirm that our emitters bear a strong  
 242 radial dipole character. The measurements dispersion can be well understood by considering  
 243 that the real emitters are a mixture of radial and axial dipoles radiating with a characteristic  
 244 decay time comprised between the simulated lifetimes of the pure radial and axial dipole.  
 245 We do not observe long decay time for NWQDs without an oxide shell in Fig. 4. For these  
 246 systems, it is very difficult to find emitters which are bright enough to be detected is because  
 247 both the laser absorption and the emission rate of a radial dipole are very weak for such  
 248 small NW diameters. We think that the emitters which have been selected correspond to  
 249 NWQDs having a large fraction of axial dipole character as they are the brightest ones when  
 250 no oxide shell is present.

### 251 C. Radiation pattern

252 To analyze the mechanisms leading to the increase in collected intensity with increasing  
 253 shell thickness, we present in Figure 5 several simulated radiation patterns. They are rep-  
 254 resented as polar plots of the far-field intensity  $I(\theta)$  in the top  $(x, z)$  plane,  $\theta$  is the angle  
 255 between the direction of observation and the vertical  $z$  axis. Simulations are made using  
 256 respectively a radial dipole [along  $x$ , Figures 5(a-c)] or an axial dipole [along  $z$ , Figures 5(d-  
 257 f)].

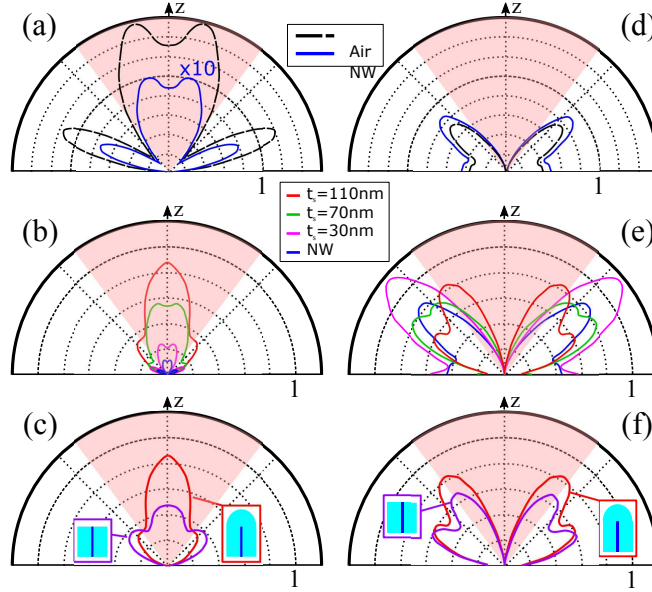


Figure 5. Radiation patterns from numerical simulations for a radial dipole placed at 470 nm above the substrate. The experimental NA region is shaded and indicated in red. (a,d) Comparison between the case of a free-standing emitter in air (black dashes), and embedded inside the NW (blue solid line) for a radial (a) or axial (d) dipole. They evidence the effect of the dielectric screening from the NW on the radial dipole and the absence of screening for the axial dipole. (b, e) Comparison of the total emitted intensity versus  $t_s$  for a radial (b) or axial (e) dipole. A combined effect of reduced dielectric screening and light guiding and redirection towards small angles is observed. (c, f) Effect of the shell layer termination shape for a radial (c) or axial (f) dipole. The hemispherical shape increases the fraction of light that is redirected towards the  $z$  direction.

258 Figures 5(a,d) show the effect of the NW structure alone (no oxide shell being present)  
 259 on such dipoles by comparing it to the case of a free standing dipole in vacuum above  
 260 the same ZnSe substrate. One can see that the presence of the NW does not affect the  
 261 shape of radiation diagram, which is essentially determined by the interferences between the  
 262 directly radiated field and its reflection on the substrate. Most remarkably, in the case of the  
 263 radial dipole the presence of the NW dramatically reduces the emission intensity through  
 264 the dielectric screening effect discussed earlier. Simulations show a radiative rate reduction  
 265 by a factor  $\sim 1/16 \simeq n_{\text{ZnSe}}/45$  in agreement with the dielectric screening value predicted by  
 266 Eq. (1). In contrast, in the case of the axial dipole it can be seen that the presence of the  
 267 NW only slightly increases the emitted intensity.

268 Figures 5(b,e) show the computed radiation patterns of the NWQD for increasing oxide  
 269 shell thickness  $t_s$ . In the case of the radial dipole, the shell first reduces the index contrast  
 270 between the NW and the surrounding medium (cf. Eq. 1), resulting in a strong reduction of  
 271 the emitter lifetime and thus in an increased total emitted intensity as seen in in Fig. 3(c)  
 272 and Fig. 4. Note that the the intensity pattern shown in the polar plot must be multiplied by  
 273 the solid angle  $\sin \theta d\theta$  if one wants to evaluate the power radiated in the numerical aperture.  
 274 This is why the intensity for an axial dipole can be larger than for a radial one, as seen in  
 275 Fig. 3(c). Second, as shown in Figure 1(c), the shell presence ensures preferential emission  
 276 into the guided  $\text{HE}_{11}$  mode for increasing shell thickness. As a consequence a near-Gaussian  
 277 far-field emission pattern corresponding to the far-field emission profile of the  $\text{HE}_{11}$  mode[38]  
 278 is observed for  $t_s=110$  nm, contrary to the structures with a smaller oxide shell thickness  
 279 where one observes the presence of two closely-spaced lobes at small emission angles ( $\pm 10^\circ$   
 280 with respect to the  $z$ -axis). The resulting emission into the 0.6 NA cone is maximum for  
 281  $t_s = 110$  nm, where the emission into the  $\text{HE}_{11}$  mode is nearly maximum [cf. Fig. 1(b)].  
 282 The effect of the oxide shell thickness on the axial dipole is completely different. While the  
 283 total emitted intensity does not vary much, and hence the emitter lifetime stays constant (as  
 284 noted in Fig. 4), the light emitted by the axial dipole does not couple to the  $\text{HE}_{11}$  mode but  
 285 is emitted exclusively into radiation modes. Thus the fraction of intensity emitted towards  
 286 the collection lens increases only slightly as the oxide shell thickness increases [cf. Fig.5(e)].  
 287 This intensity increase for the axial dipole also presented in Fig.3(c) is not due to a change  
 288 in the spontaneous emission rate of the emitter, but rather to a slight redirection of the  
 289 emitted light.

290 Finally, Figures 5(c,f) compare the actual hemispherical geometry of the oxide shell ter-  
 291 mination to the flat end of a simple lateral shell. They show that the presence of the  
 292 hemisphere is beneficial to the radiation pattern for both kinds of dipole. For the radial  
 293 dipole, the hemisphere enables a near-adiabatic expansion of the  $\text{HE}_{11}$  mode[38] leading to  
 294 a narrowing of the far-field emission pattern and an increased collection by the numerical  
 295 aperture. The axial dipole benefits less from the hemispherical termination of the photonic  
 296 structure since no light from this dipole is coupled to the  $\text{HE}_{11}$  mode. We also note that half  
 297 of the emitted light propagates towards the growth substrate and due to the index-matching  
 298 condition between the NW and the substrate, this light is predominantly lost.

299 In order to assess the performances of our device we compute the ratio  $\eta$  between the

300 power radiated into a 0.6 NA to the one radiated into the top air side hemisphere. This  
 301 parameter is a good figure of merit for the antenna redirection effect although it cannot be  
 302 directly related to the overall collection efficiency because of the power lost in the substrate.  
 303 For our full photonic structure and a radial dipole one has  $\eta \simeq 80\%$  for  $t_s=110$  nm. This value  
 304 reduces to  $\simeq 66\%$  for a flat terminated core-shell photonic wire illustrating the importance of  
 305 the adiabatic expansion of the  $\text{HE}_{11}$  guided mode at the end of the wire. For the dipole in the  
 306 NW without shell  $\eta \simeq 55\%$ . We have also simulated a structure inspired by state-of-the-art  
 307 devices fabricated by top-down methods in Ref. [9]. In this case we simulate a 110 nm oxide  
 308 shell photonic wire where the hemispherical termination is replaced by a conical taper of  
 309  $\text{Al}_2\text{O}_3$  whose radius progressively decreases from 120 to 10 nm in  $1.5\text{ }\mu\text{m}$ . In this case one  
 310 has  $\eta \simeq 94\%$ , showing that although beneficial our hemispherical termination is not optimal.

## 311 VI. CONCLUSION

312 In summary, we have presented a bottom-up approach to fabricate a dielectric antenna  
 313 around a QD inserted inside a NW. This method allows for both reproducible and very  
 314 precise fabrication of the structure on a large ensemble of emitters at once. It is based on the  
 315 deposition of a thick oxide shell around the NW using atomic layer deposition. Experiments  
 316 show a 4-fold enhancement of the QD photoluminescence shown in Fig. 3(c) between a  
 317 20 nm and a 110 nm thick shell. Semi-analytical calculations and numerical simulations of  
 318 the structure reveal that the oxide shell thickness strongly acts on the radial dipole emission  
 319 through two main phenomena: the reduction of the dielectric screening, which increases the  
 320 spontaneous emission rate from the QD, and the redirection of light through a waveguiding  
 321 effect. Simulations suggest that the collected intensity is multiplied by a factor 7 with respect  
 322 to the bare NW case. The fabrication process of the photonic shell is very simple and can be  
 323 applied to QDs emitting single photons up to room temperature. Although not optimal, the  
 324 resulting structure is a step towards the best nanowire single photon sources operating at  
 325 low temperature[9]. Dielectric screening could be further reduced by growing an oxide shell  
 326 of higher index matching  $n_{\text{ZnSe}}$  like  $\text{TiO}_2$ . We note also that in our system a large fraction  
 327 of the emitted power is radiated in the substrate. This loss channel could be reduced by  
 328 having a mirror at the bottom of the structure.[15, 39] Moreover, to fully benefit from the  
 329 waveguiding approach, a better control on the intrinsic QD properties has to be reached

to ensure the presence of radial dipoles, which radiate more efficiently in the experimental collection aperture.

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