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1 Fabrication and tuning of plasmonic optical nanoantennas around droplet 2 epitaxy quantum dots by cathodoluminescence

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We use cathodoluminescence to locate droplet epitaxy quantum dots with a precision $\lesssim 50$ nm before fabri-
cating nanoantennas in their vicinity by e-beam lithography. Cathodoluminescence is further used to evidence
the effect of the antennas as a function of their length on the light emitted by the dot. Experimental results
are in good agreement with numerical simulations of the structures.

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13 Ever decreasing sizes and low dimensionality of semi-
14 conducting heterostructures makes it possible to operate
15 them in the quantum regime where single photons are
16 produced and/or detected, paving the way to applica-
17 tions in quantum information processing and communi-
18 cation. In this context a key issue of nanooptics con-
19 cerns the possibility to modify and control the proper-
20 ties of the light coming from a single solid-state emit-
21 ter (direction, polarization, temporal profile). A poten-
22 tial route towards this goal is to embed it in a dielectric
23 microcavity¹ or a photonic waveguide². Another promis-
24 ing strategy is to extend the know-how of RF electrical
25 engineering to the optical domain by placing metallic
26 nanostructures in the vicinity of the emitter^{3,4}. They
27 act as plasmonic optical nanoantennas able to enhance
28 the emission rate of the emitter^{5,6} and control its radi-
29 ation pattern⁷⁻⁹. From a technological point of view, it
30 is paramount to control both the emitter-antenna dis-
31 tance with a precision $\lesssim 10$ nm, and the characteristic fre-
32 quency of the plasmonic mode to tune the antenna to
33 the emission wavelength. Precise positioning can be
34 achieved with the help of AFM manipulation^{6,10} or chem-
35 ical functionalization⁸, while standard top-down fabri-
36 cation techniques are better suited to define the shape
37 of the metal nanostructure and hence its resonant fre-
38 quency. In this letter, we demonstrate that cathodolumi-
39 nescence (CL) combined to standard e-beam lithography
40 is a powerful tool to achieve both goals. We use this tech-
41 nique to locate droplet epitaxy quantum dots (QDs) be-
42 fore fabricating nanoantennas of different length around
43 them. CL is further used to characterize the effect of the
44 antennas on the QDs’ emission properties as a function
45 of the length.

46 Our target structure is shown in Fig. 1(c-d). It
47 has already been shown to enhance the fluorescence of

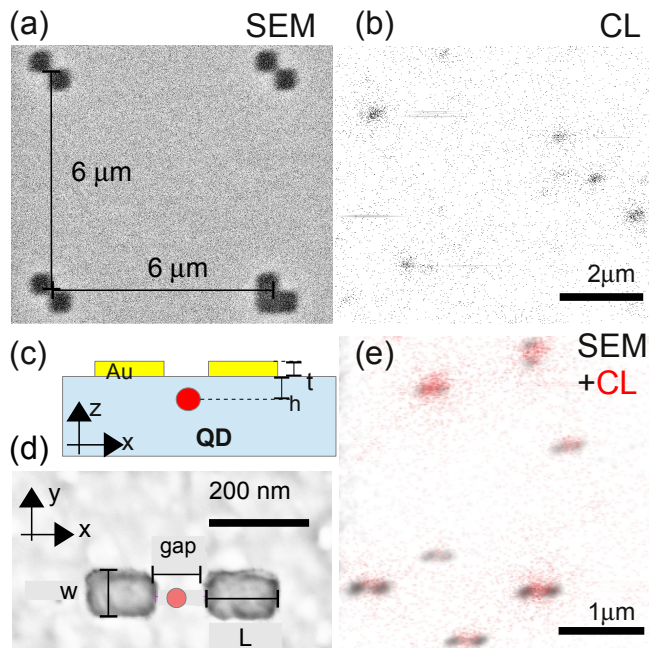


FIG. 1. Simultaneously recorded images of (a) the SEM signal of the Au alignment marks deposited on the sample and (b) the CL signal of the droplet epitaxy quantum dots ($\lambda_{em}=750$ nm). (c) Cut and (d) top field effect SEM microphotograph of the target QD/nanoantenna assembly; $h=50$ nm, gap 100 nm, $w=100$ nm, $t=35$ nm, L varies from 80 to 350 nm. (e) Composite image of the fabricated Au nanoantennas observed in SEM (black channel) and of the QD spots (red channel).

48 an ensemble of dye molecules¹¹. In our case a single
49 GaAs/AlGaAs quantum dot is buried under the gap be-
50 tween two Au nanostrips. QDs are produced using the
51 modified droplet epitaxy method¹². Atomic force mi-
52 croscopy measurements show a density of emitters
53 about $3 \mu\text{m}^{-2}$. Each dot has a typical 20 nm diame-

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54 ter and a height of 10 nm. Capping layers of different
 55 thickness h from 22 to 70 nm are overgrown on top of
 56 the QD layer. Thermal annealing is necessary to activate
 57 the luminescence of the dots¹³. Ensemble photolu-
 58 minescence measurements show that QDs emit between
 59 750 and 800 nm. Microphotoluminescence and cathodo-
 60 luminescence allow us to observe that typical signal in-
 61 tensity from individual QD is dramatically reduced for
 62 thicknesses $h \leq 40$ nm. We attribute this observation to
 63 additional nonradiative losses generated by surface in-
 64 duced recombinations. Nanoantennas are fabricated on
 65 a sample with $h = 48$ nm in order to ensure the smallest
 66 QD/antenna distance without having extra non-radiative
 67 losses. Before localization of individual quantum dots,
 68 Au alignment marks are fabricated onto the substrate
 69 by e-beam lithography and sputtering of a Au layer fol-
 70 lowed by lift-off [Fig. 1(a)]. Marks are placed on a regular
 71 square array of period $6 \mu\text{m}$.

72 The sample is then observed in CL at 5 K. Light emit-
 73 ted by the excited dots is collected by a parabolic mirror
 74 and analyzed by a spectrometer. It is then detected by an
 75 avalanche photodiode (APD) at the output of the spec-
 76 trometer. A standard SEM image of the marks [Fig. 1(a)]
 77 is simultaneously recorded with the CL image at emission
 78 wavelength $\lambda_{em} = 750$ nm [Fig. 1(b)]. We limit the image
 79 acquisition time to a few seconds in order to limit the ef-
 80 fect of thermal drift, surface contamination and charging
 81 of the substrate in the vicinity of the quantum dot. The
 82 presence of a QD is hence revealed by a cloud of single
 83 photon detection counts from the APD (black pixels on
 84 the image, typical cloud diameter ~ 400 nm). The cen-
 85 ters of the marks are determined by an edge detection
 86 algorithm on the SEM image. The CL signal intensity
 87 of each QD is obtained by summing all its corresponding
 88 pixels and its position is determined by their centroid.
 89 Combination of those informations yields the absolute
 90 coordinates of each dot with respect to the alignment
 91 mark array.

92 Nanoantennas are fabricated by e-beam lithography on
 93 a 100 nm-thick PMMA layer followed by sputtering of a
 94 35 nm thick layer of Au and lift-off. Before exposing
 95 antennas the SEM e-beam is aligned by observing neigh-
 96 bouring marks. For all antennas the gap between the
 97 strips is 100 nm, the width $w = 100$ nm and the strips
 98 have a variable length L ranging from 80 to 360 nm.

99 After fabrication of the nanoantennas, a second CL
 100 experiment is undertaken. Figure 1(e) presents a com-
 101 posite image of the SEM signal (black channel) and CL
 102 signal (red channel). It clearly shows that each an-
 103 tenna coincides with a QD. In order to assess the perfor-
 104 mances of our fabrication process, we have fabricated 123
 105 QD/antenna assemblies. For each system we measure the
 106 QD's position by its centroid as well as the antenna gap
 107 center on the SEM image. Figure 2(a) shows a scatter
 108 plot of the relative positions of the QDs with respect to
 109 the nanoantennas and figure 2(b) displays an histogram
 110 of their distance distribution. 70% of the dots are within
 111 the $100 \text{ nm} \times 100 \text{ nm}$ square defined by the gap between

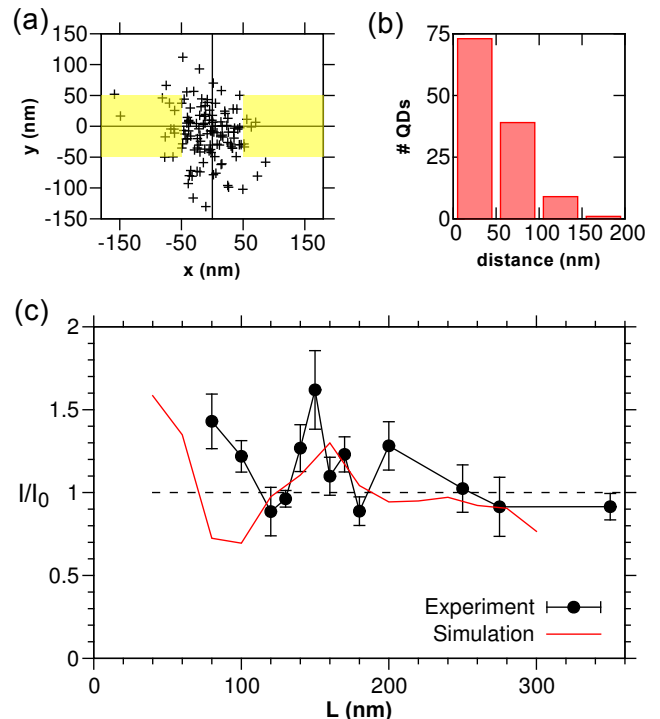


FIG. 2. (a) Scatter plot of the QD positions measured by CL with respect to the nanoantenna center. The position of the strips is shown by yellow rectangles. (b) Histogram of QDs' distance distribution from antenna center. (c) Luminescence enhancement ratio I/I_0 as a function of antenna length L . Experimental points are compared to numerical simulations assuming a random distribution of dipole orientations

112 the two nanostrips. Less than 8% of them are at distances
 113 larger than 100 nm. A qualitative study of those poorly
 114 fabricated assemblies shows that they correspond to sys-
 115 tems located close to the edges of the images or to other
 116 dots. The centroid algorithm does not work properly in
 117 those conditions. The average distance to the antenna
 118 center is 48 nm. Assuming that it is the sum of three
 119 independant equal sources of error (first localization, e-
 120 beam realignment before fabrication and second local-
 121 ization), one infers a typical localization error of 28 nm.
 122 Our present error is larger than the target precision for
 123 fabricating state of the art plasmonic structures, but it
 124 is nonetheless good enough to evidence coupling of the
 125 QD to the antenna in the case of a 100 nm gap as is our
 126 case.

127 In order to evidence the effect of the nanoantennas
 128 on the QDs, we compare their CL signal in presence of
 129 the antenna I to the one measured before fabrication I_0 ,
 130 with the same parameters of beam current, dwell-time
 131 and magnification. Hence, the excitation rate γ_{exc} is the
 132 same for the two experiments. Moreover it is low enough
 133 to ensure that the quantum dots are not saturated, i.e.
 134 $\gamma_{exc} \ll \gamma_{tot}$ where γ_{tot} is the relaxation rate of the quan-
 135 tum dot. Between 6 and 27 QD/antenna assemblies have

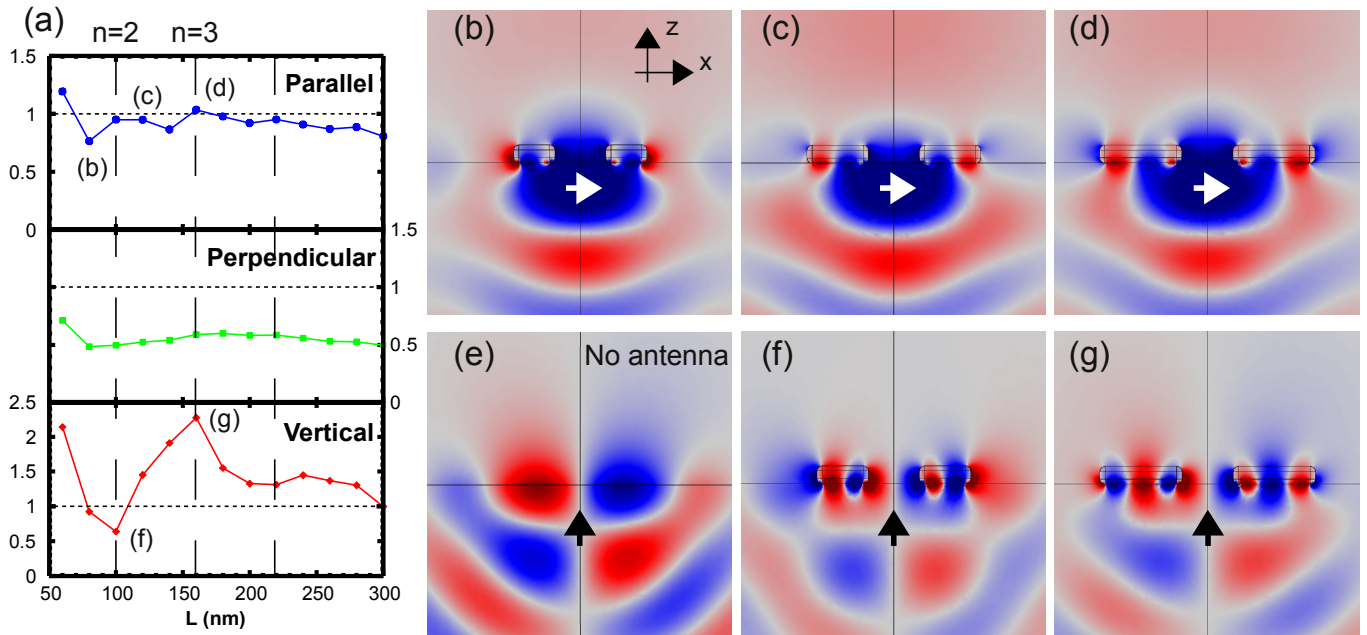


FIG. 3. (a) simulated enhancement factors as a function of antenna length L for a quantum dot corresponding to an oscillating dipole along the parallel (x), perpendicular (y) or vertical (z) direction. Maps of $Re[E_x]$ in the (x, z) plane for a parallel dipole (b-d) and a vertical dipole (f-g) for different antenna lengths. The corresponding points are shown on figure (a) except for (e).

136 been fabricated for each antenna length L . Figure 2(c) 166
 137 presents the average ratio I/I_0 as a function of L . The 167
 138 corresponding error bars represent the statistical standard 168
 139 error on this value. One observes an enhancement 169
 140 of the signal (about 50%) below 80 nm and in the 140- 170
 141 170 nm range. This proves that it is possible to tune 171
 142 the antenna length in order to optimize the emission of 172
 143 light towards the CL detector. In our setup, the collec- 173
 144 tion mirror is placed just above the sample and we can 174
 145 assume that all the light emitted on the air side of the 175
 146 sample is collected. The measured intensity I is then¹⁴ 176
 147 $I = hc/\lambda_{em} \cdot \gamma_{exc}\eta$, where η is the quantum efficiency 177
 148 $\eta = \gamma_r / \gamma_{tot} = \gamma_r / (\gamma_r + \gamma_{nr})$. γ_{nr} 178
 149 is the non-radiative relaxation rate of the quantum dot. 179
 150 The radiative relaxation rate γ_r is the sum of the light 180
 151 emitted on the air-side and on the substrate side of the 181
 152 sample $\gamma_r = \gamma_{r, air} + \gamma_{r, sub}$.

153 A deeper understanding of our results arises from nu- 183
 154 merical simulations of the fabricated QD/antenna sys- 184
 155 tem with the finite-element software COMSOL. The QD 185
 156 is replaced by an electric dipole oscillating at frequency 186
 157 $\nu_{em} = c/\lambda_{em}$. We compute the power $P_{r, air}$ (resp. 187
 158 $P_{r, sub}$) radiated in the far-field towards the air side (resp. 188
 159 the substrate side) of the sample. We also evaluate the 189
 160 power P_{abs} absorbed in the metallic nanoantennas due 190
 161 to the imaginary part of the index of refraction of Au 191
 162 at λ_{em} ¹⁵. As we have previously checked that surface 192
 163 induced nonradiative recombinations are negligible for 193
 164 $h = 50$ nm, we assume that all nonradiative losses are 194
 165 caused by absorption, i.e. $\gamma_{nr} \propto P_{abs}$. We also have 195

166 $\gamma_{r, air} \propto P_{r, air}$ and $\gamma_{r, sub} \propto P_{r, sub}$. It is therefore pos- 166
 167 sible to evaluate the quantum efficiency before and after 167
 168 the antenna fabrication. We perform these calculations 168
 169 for fixed values of $w = 100$ nm, $t = 35$ nm, gap = 100 nm 169
 170 and with various conditions of strip length L or dipole 170
 171 orientation. We also plot the maps of the corresponding 171
 172 electromagnetic fields.

173 The simulations show that the dipole couples to an- 173
 174 tenna modes exhibiting 1 to 3 antinodes in each nano- 174
 175 strip along the x axis (figure 3). From one mode to the 175
 176 next one, L is increased by 60 nm. This is in good agree- 176
 177 ment with half the expected surface plasmon wavelength 177
 178 λ_p for the Au/GaAs interface at frequency ν_{em} ^{16,17}. For 178
 179 a dipole oriented along the y axis, the coupling to the 179
 180 antenna mode is poor. Hence the quantum efficiency is 180
 181 essentially flat and degraded to about 0.5 due to extra 181
 182 absorption in the metal [Fig. 3(a)]. In the case of a dipole 182
 183 along the x axis, the excited antenna mode radiates ei- 183
 184 ther in phase with the dipole, resulting in an increase of 184
 185 the field received by the detector [Fig. 3(b)], or in phase 185
 186 opposition, leading to the reverse effect [Fig. 3(c)]. The 186
 187 relative phase of the plasmon oscillation is shifted by π 187
 188 as L spans across the resonance condition. As a conse- 188
 189 quence the luminescence enhancement is modulated with 189
 190 a period $\lambda_p/2$ with peaks at $L = 100, 160$ and 220 nm. 190
 191 The effect of the antenna is dramatic in the case of a 191
 192 dipole along the z axis, which does not radiate efficiently 192
 193 towards the detector in absence of antenna [Fig. 3(e)]. 193
 194 In this configuration the dipole excites antenna modes 194
 195 which efficiently radiate on the air side if they have an

196 odd number of antinodes [Fig. 3(g)]. For even number of
 197 antinodes [Fig. 3(f)], the contributions of each antinode
 198 cancel in the far field and absorption dominates. As a
 199 result one observes a modulation of the emitted intensity
 200 versus L with a period λ_p .

201 As opposed to stransky-Krastanov quantum dots, the
 202 orientation of the electric dipole in droplet epitaxy dots
 203 is not strongly constrained due to the absence of internal
 204 stress, relative large size and smooth interface. The com-
 205 parison of the experimental results of figure 2(c) with the
 206 simulations of figure 3(a) suggests that a large fraction of
 207 systems have a dipole vertically aligned. The simulation
 208 data plotted on figure 2(c) correspond to an average of
 209 the three orientations. It is in good agreement with the
 210 experiment.

211 In summary, we have demonstrated the controlled cou-
 212 pling and tuning of Au nanoantennas to droplet epitaxy
 213 QDs using cathodoluminescence and standard electron-
 214 beam lithography. Our method offers the advantage of
 215 being spectrally selective and has a higher throughput
 216 than AFM nanomanipulation techniques. The enhance-
 217 ment factor of luminescence which we observe is well ex-
 218 plained by numerical simulations. It could be dramati-
 219 cally increased with a smaller QD/antenna distance. We
 220 plan to improve the QD signal while reducing the cap-
 221 ping layer. We have also developed a method for di-
 222 rectly aligning the e-beam lithography setup on the CL
 223 signal, thus suppressing the realignment step onto ancil-
 224 lary marks¹⁸. With those two improvements, we expect
 225 to fabricate coupled QD/antennas with typical depth and
 226 antenna gap of the order of 20 nm. It is also possible
 227 to fabricate more complicated structures like clusters of
 228 nanoantennas in order to optimize the emission quantum
 229 efficiency¹¹ and reach higher enhancement factors.

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