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Photonic jets writing of quantum dots with intrinsic photoluminescence collection enhancement(*)

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Summary. — Owing to their ability to generate non-classical light states, quantum dots (QDs) are ideal candidates for the large-scale deployment of quantum information technologies. However, semiconductor QDs alone lack the high photon collection efficiency needed by these technologies. In this work we present a laser writing technique for the fabrication of QDs self-aligned with dielectric microspheres, which, in turn, increase the collection efficiency of the system of a factor 7.3 ± 0.7. That technique exploits the use of photonic nanojets, produced by illuminating the microspheres, to selectively break the N-H bond in a GaAs/$GaAs_{1-x}N_x$:H/GaAs quantum well, thus fabricating $GaAs_{1-x}N_x$ QDs.

Semiconductor quantum dots (QDs) are ideal candidates as future building blocks for the development of quantum information technologies [1] thanks to their ability to act as single-photon emitters. In particular, the most promising ones are the epitaxially grown QDs, which are fabricated within a solid-state matrix, that fixes their position and makes them uniquely suitable for integration in electronic devices [2]. However, the presence of an embedding material, typically characterized by a high refractive index, strongly limits the QD luminescence extraction due to total internal reflection [3]. Moreover, the numerical aperture (NA) of the collecting optics limits the collected luminescence even further.

One of the approaches developed to solve this issue is the use of solid immersion lenses (SILs). SILs are lenses of various sizes and shapes placed on the sample, on top of the emitters [4], and they are spectrally and spatially tolerant tools. However, they are characterized by a limited performance (they lead to a maximum collection enhancement

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of about the square of their refractive index [5]), they can be bulky (mm size) and they are sensitive to the air gap which can occur between them and the sample surface [5].

In this work, we present a technique to fabricate $GaAs_{1-x}N_x$ QDs self-aligned to dielectric microspheres, obtaining a reversible, low-cost and broadband solution to the problem of light collection [6]. Indeed, it is known that the presence of a microsphere on top of a QD helps the collection process due to an increase in the NA of the system [7]. However, to maximize this effect, a precise positioning of the microsphere on top of the QD is required; we circumvented this issue by exploiting the microspheres not only for the collection process, but also for the QD fabrication. In particular, harnessing the properties of hydrogenated $GaAs_{1-x}N_x$ [8] in conjunction with the so-called *photonic nanojets* (PJs) [9] our novel technique allows us to "laser write" $GaAs_{1-x}N_x$ QDs exactly below the microspheres, self-aligned with their central axis.

Our technique is based on controlling the hydrogen removal process in $GaAs_{1-x}N_x$:H/-GaAs ($x = 0.011$) quantum well (QW) by using PJs. A PJ is a highly intense light beam characterized by a sub-wavelength spatial extension, obtained below a microsphere illuminated with a plane wave of proper λ [9]. By illuminating a microsphere, we were able to remove hydrogen from a nanometric region of the QW, creating a $GaAs_{1-x}N_x$ ($E_g < 1.5\,\text{eV}$ at 10 K [10]) nanocrystal surrounded by $GaAs_{1-x}N_x$:H ($E_g \approx 1.5\,\text{eV}$ at 10 K [11]) in the lateral direction and GaAs ($E_g = 1.519\,\text{eV}$ at 10 K [12]) in the vertical direction, obtaining the 3D energy confinement necessary to define a QD.

The $GaAs_{1-x}N_x$ QDs obtained in this work were created at room temperature and in air, by using a diode-pumped solid-state laser at 532 nm with different powers, to obtain QDs of different sizes. The QDs were characterized by their photoluminescence (PL) at low temperature ($T = 10\,\text{K}$), measured with a home-made confocal microscope setup equipped with an objective with NA = 0.7. The PL spectra of the most representative QDs are reported in fig. 1 along with the spectrum of the QW before hydrogenation, for comparison. From the measured QD emission energy it is possible to roughly estimate their size, after some reasonable assumptions (*e.g.*, cylindrical shape) and using the simple model developed in ref. [13]. From the calculation we find that the QD diameters

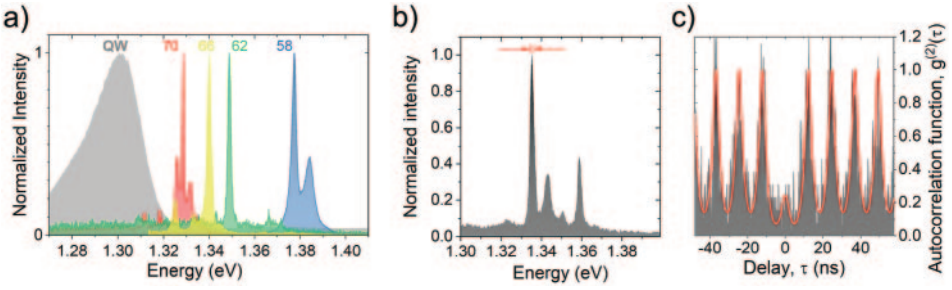

Fig. 1. – (a) PL spectra of four different QDs fabricated at different fabrication powers, distinguished by colors. The fabrication powers, in units of mW, are provided as labels. The shaded spectrum corresponds to the PL spectrum of the $GaAs_{1-x}N_x$/GaAs QW before hydrogenation. (b) PL spectrum of QD1. The interval indicated by two red arrows represents the spectral range (2 meV) used for the measurement of the second-order autocorrelation function, $g^{(2)}(\tau)$. (c) $g^{(2)}(\tau)$ measured just below the saturation power (dark histogram) and its fit (red line) with the model described in the text. It is clear that the $g^{(2)}(0) < 0.5$, confirming the single-photon emitter nature of this QD. All the PL spectra and the $g^{(2)}(\tau)$ measurements were taken at 10 K.

span the range between 6.8 nm and 10 nm, obtained with fabrication powers between 58 mW and 70 mW, respectively. This proves our ability to remove hydrogen from an area much smaller than the diffraction limit by employing PJs as well as our control over the QDs size, and thus over their emission energy, by changing the fabrication power.

In order to test the ability of these QDs to emit single photons, we measured their second-order autocorrelation function, $g^{(2)}(\tau)$. In fig. 1(b) we report the PL spectrum of a QD (labeled QD1), while in fig. 1(c) we display the $g^{(2)}(\tau)$ relative to the emission line of the same QD, under pulsed excitation regime. Clearly, the raw value of $g^{(2)}(\tau)$ at zero delay ($\tau = 0$) is about 0.25, much lower than 0.5, showing the single-photon emitter nature of the QD. The $g^{(2)}(\tau)$ was fitted with a curve (reported as a solid red line in fig. 1(c) based on the solution of a system of rate equations, developed by taking into account the main processes leading to the capture, relaxation and recombination of carriers in and out of the QD (for a more detailed description of the model see ref. [14]). It also comprises a term describing an uncorrelated background, which is found to account for about $\sim 20\,\%$ of the measured coincidences, likely due to the broad emission overlapping with the QD line under consideration. When removing this uncorrelated background, the value of $g^{(2)}(0)$ results lower than 0.1.

After demonstrating the possibility of "laser writing" $GaAs_{1-x}N_x$ QDs, we verified another "optical advantage" that the presence of the microspheres brings to the table, *i.e.*, the collection enhancement (CE), which is defined as the ratio between the collected power with and without the microsphere on top of a QD. To perform the measurement of the CE, we measured a QD (labeled QD2) before and after the removal of the sphere on top of it. However, to properly compare the intensities in the two situations, the PL spectra were acquired as a function of the excitation power to find the saturation power. Indeed, the saturation power corresponds to an identical excitation condition. The integrated intensity as a function of the excitation power before and after sphere removal is reported in fig. 2(a), combined with the fitting curves obtained with the model described in ref. [13]. The CE observed at saturation is about 7.3±0.7. For the sake of completeness, in fig. 2(b) we have reported the PL spectra of the QD before and after sphere removal with an excitation power as close as possible to the saturation power. The enhancement of the collected signal is clearly observed.

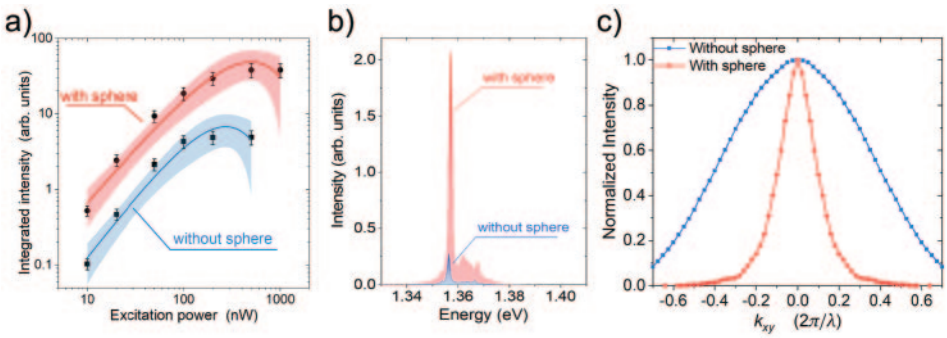

Fig. 2. – (a) Integrated intensity at 10 K of the main transition (X) of a QD (labeled QD2) with a sphere on top (circles) and after sphere removal (squares), as a function of the excitation power. The solid lines are the best fits obtained with the model described in ref. [13] (the filled areas represent 95% confidence bands). At saturation, the value obtained from the ratio of the intensities of the two fitting curves is 7.3±0.7. The CW spectra at $P = 500$ nW are reported in panel (b) for comparison. (c) Measured angular profiles (red circle and blue squares) of QD3 (with a microsphere on top of it) and of the QW (without the microsphere), respectively.

Finally, in order to investigate the physical origin of the CE, we measured the angular distribution of the emission of a QD (labeled QD3), with a microsphere on top. Indeed, we believed that the CE should be mainly due to an improved directionality of the emission. To verify this, we measured the angular emission pattern of QD3 and of the $GaAs_{1-x}N_x$/GaAs QW. The latter was performed to mimic a point-like emission (like a QD) without a microsphere. The resulting measurements are reported in fig. 2(c), which clearly shows the predicted increase in directionality due to the presence of the microsphere.

In conclusion, in this work we have demonstrated the possibility of creating $GaAs_{1-x}N_x$ QDs by exploiting PJs to locally tune the hydrogen content in dilute nitrides. This fabrication method also leads to a broadband enhancement of the collection efficiency ($\times 7.3$) for the light emitted by the QD.

This work paves the way for several possible developments: from the realization of complex structures such as QD arrays, by packing up the microspheres in a honeycomb structure [15], to the development of innovative approaches to the creation of site-controlled QDs, by controlling microsphere deposition or by mounting a microsphere at the end of an optical fiber (which, in turn, could be mounted on a nanopositioner).

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