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Combined metallic nano-rings and solid-immersion lenses for bright emission from single InAs/GaAs quantum dots

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Solid-state single-photon emitters are key components for integrated quantum photonic devices. However, they can suffer from poor extraction efficiencies, caused by the large refractive index contrast between the bulk material they are embedded in and air: this results in a small fraction (that can be as low as $\sim 0.1\%$) of the emitted photons reaching free-space collection optics. To overcome this issue, we present a device that combines a metallic nano-ring, positioned on the sample surface and centered around the emitter, and an epoxy-based super-solid immersion lens, deposited above the ring devices. We show that the combined broadband lensing effect of the nano-ring and the super-solid immersion lens significantly increases the extraction of light emitted by single InAs/GaAs quantum dots into free space: we observe cumulative enhancements that allow us to estimate photon fluxes on the first collecting lens approaching 1×10^6 counts per second, from a single quantum dot in bulk. The combined broad-band enhancement in the extraction of light can be implemented with any kind of classical and quantum solid-state emitter and opens the path to the realisation of scalable bright devices. The same approach can also be implemented to improve the absorption of light, for instance, for small-area broadband photo-detectors. *Published by AIP Publishing*. https://doi.org/10.1063/1.5023207

Improving the efficiency in the extraction of light from solid-state emitters into free space is a challenge that often needs to be overcome when dealing with photonic devices. Total internal reflection due to the refractive index contrast between different materials results, in some cases, in even less than 0.1% of the emitted photons to be collected into free space. This is a clear drawback if one wants, for instance, to use light for quantum information processing, where high fluxes of single photons and minimised losses are required.¹ Furthermore, the shape of the emitted light beam and its angular divergence are important parameters to control in order, for instance, to optimise the coupling efficiency of light into optical elements like objectives and single-mode fibers that can be used for propagating classical or quantum light over long distances with low loss. To improve the extraction efficiency of light, several photonic devices have been realised: optical cavities, which have the advantage of Purcell enhancements of the spontaneous emission rate,² achieving extraction efficiencies up to 65%, although limited to relatively narrow bandwidths;³ nanowire geometries that allow guiding and shaping of the emitted light in a broad-band device, reaching efficiencies as high as 72%. Optical microlenses that provide broad-band improvements in the brightness of the emitted light of up to one order of magnitude are also widely used and have been deterministically fabricated around specific emitters^{6,7} or deposited from the liquid phase.⁸ We have recently demonstrated that, by using an all-optical positioning technique, metallic nano-rings can be fabricated around specific emitters in order to obtain broad-band increases in the

brightness of the emitted light, reaching enhancement factors as high as ~20.9 Here, we combine such metallic nano-rings with epoxy-based super-solid immersion lenses (super-SILs)⁸ to combine the broad-band enhancement effects of the two devices and obtain bright emission from single InAs/GaAs quantum dots (QDs). We note that this technique is non-destructive and that both the ring and the super-SIL can be removed if required, leaving the properties of the emitters unchanged. Furthermore, such a combined approach can be also implemented to improve the absorption of light in small-scale broadband photo-detectors.

A schematic of the sample under study, containing a single layer of low density (\sim 1 per 1000 μ m⁻²) InAs/GaAs QDs, is shown in Fig. 1(a). We use a photoluminescence imaging technique, based on a double light-emitting diode (LED) illumination¹⁰ of a sample containing one layer of single InAs/ GaAs QDs. A 455 nm-LED is used to excite the QD emission, and a 940 nm-LED illuminates metallic markers that are deposited on the sample surface in order to locate single QDs. An example of a photoluminescence image, collected with an electron-multiplying charge coupled device (EMCCD), of metallic alignment marks deposited on the sample surface and of the emission of single QDs is shown in Fig. 1(b). By means of aligned electron-beam lithography, metal evaporation, and chemical lift off, gold nano-rings are then deposited on the sample surface, centred around specific emitters [see Fig. 1(c)—for more details, see Ref. 9].

To evaluate potential losses due to the presence of the metal, we have calculated, via finite-difference time-domain simulations, the total emission of a dipole before and after the metallic ring is deposited on the sample surface. This

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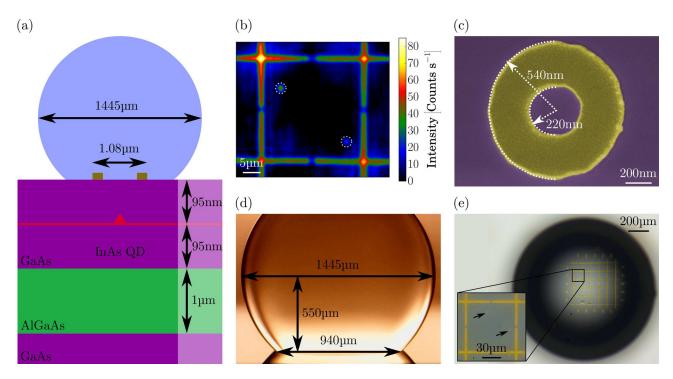


FIG. 1. (a) Schematic of the sample (vertical section, not to scale). (b) Example of a photoluminescence image, obtained under double LED illumination on an EMCCD camera (see main text for more details), showing metal alignment markers and the photoluminescence emission from two QDs, highlighted by the dashed lines (before super-SIL deposition, EMCCD settings: 2 s integration, 100 acquisitions). (c) False colour scanning electron micrograph of a metallic nano-ring, with critical dimensions shown. (d) Side profile of a super-SIL with critical dimensions shown. (e) Optical micrograph of a super-SIL deposited on a sample in correspondence to metallic alignment markers and nano-rings. Inset: Zoom-in of a single field showing metallic alignment markers and nano-rings positioned around selected QDs (highlighted by the arrows).

shows that, given the distance between the emitter and the metallic ring, losses are negligible.

A super-SIL¹¹ is formed by dispensing liquid ultraviolet (UV)-curable epoxy (with a refractive index of 1.54) onto a substrate over the region of the sample containing the metallic nanorings [see Figs. 1(d) and 1(e)]. The dispensing environment is filled with a liquid phase medium (glycerol), designed to increase the contact angle between the droplet edge and the substrate, by modification of the interfacial surface tensions, ¹² and, subsequently, the lens is solidified by exposure to UV light.

By carrying out micro-photoluminescence spectroscopy, we measure the intensity of light emitted by selected QDs at cryogenic temperatures (~10 K), under non-resonant laser excitation (with a continuous-wave laser with an emission wavelength of 780 nm), as a function of pump power. We report the flux of photons on the first lens as a way to evaluate the emission brightness independently of the specific setup and detector used. We measure the transmission $(\sim 17.5\%)$ and detection efficiency $(\sim 7.3\%)$ of our setup and use them to extract the photon flux on the collection objective with a numerical aperture of 0.65. The results of our measurements are shown in Fig. 2: the intensity of a single QD emission line is first enhanced by the metallic nano-ring and then further enhanced by the super-SIL. Such cumulative enhancements allow us to measure ~11000 counts/s (peak value) on the CCD and estimate photon fluxes on the first lens (with NA = 0.65) as high as \sim 900 000 counts/s from a single QD in bulk. By carrying out photoluminescence measurements under pico-second, above-band (780 nm) laser excitation, we can extract the fraction of emitted singlephotons collected on the first lens. Considering unitary efficiency in the QD emission process and taking into account that the excitation laser has a fixed repetition rate of 50 MHz, we calculate an efficiency at a saturation level of $\sim 1\%$. We would like to stress that this is a lower bound, given that the emission efficiency of the QD can be lower than one. ¹³ As shown in Fig. 3, different QDs in bulk show different levels

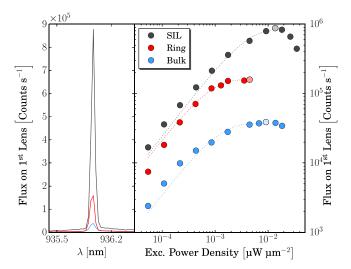


FIG. 2. Left: Photoluminescence spectra collected at saturation pump power for the emitter in bulk (blue line), after ring deposition (red line) and after super-SIL placement (black line). Right: Intensity of the emission lines shown in panel (a) plotted as a function of laser excitation power density (lighter dots correspond to the peak intensity measured from the spectra shown in the left panel, and dashed lines are fits to the data). Note that the photon fluxes on the first lens are estimated by taking into account the transmission of the setup and the sensitivity of the CCD camera (see main text for more details).

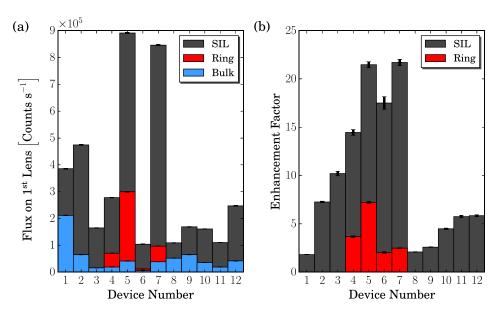


FIG. 3. Left panel: Histograms showing the photon flux on the first lens placed above the QD sample, at saturation pump powers, for the emitters in bulk (blue), with a nano-ring deposited on the sample surface (red), and with a super-SIL and/or nano-ring deposited on the sample surface (black). Right panel: Enhancement factors derived from the nano-rings and super-SILs, measured on the ODs shown in the left panel, using the same colour coding. (Note that for device numbers 4–7, the black bars represent the cumulative enhancement factor of the nano-ring and the super-SIL.)

of brightness: this is due to the different quantum efficiency of each emitter as grown. The enhancement factor derived from the super-SIL depends on the relative position of the QD with respect to the apex of the lens: therefore, we observe a range of enhancements that depend on the position of the emitter. Given the millimeter size of the super-SIL, the dependence of the light extraction enhancement factor on the distance of the emitter from the SIL⁵ is negligible in the present case. The statistics of the enhancement factors measured on QDs in bulk and within metallic nano-rings are shown in Fig. 3. It is worth noting that different QD emission lines are enhanced by different factors by the metallic nanorings, as already discussed in Ref. 9, while the super-SIL effect is measured to be the same for the different emission lines of a given QD.

From the analysis of Fig. 1(e), we can estimate the horizontal distance of the ring devices from the super-SIL apex, as shown in Table I. It is worth underlining that the super-SIL can create distortions in the top imaging of the grid, thus introducing errors that are difficult to quantify. Overall, the ring devices are within $100 \, \mu \text{m}$ from the apex, proving that the super-SIL can be placed with enough accuracy to obtain the significant enhancement factors shown in Fig. 3. Given that the QD emission pattern is also modified by the nanorings, the variations of the extraction efficiency as a function of distance from the super-SIL's apex cannot be easily simulated.

Overall, we measure enhancement factors as high as $\times 8$ by the nano-rings and up to $\times 10$ by the super-SIL. We note that previous work⁹ has shown enhancements as high as $\times 20$ by the nano-rings: therefore, for an ideally located super-SIL and nano-ring, cumulative enhancement factors as high as $\times 200$ could be potentially achieved. Further enhancements

TABLE I. Horizontal distance between the QD and the super-SIL apex (error $\sim\!\pm0.1\,\mu\text{m}).$

QD number	1	2	3	4	5	6	7	8	9	10	11	12
Distance (μm)	78.7	28.5	97.0	70.3	55.0	55.0	42.9	99.2	71.3	90.1	58.0	20.8

in the collected intensity can be obtained by introducing a bottom mirror (like a metallic layer or a distributed Bragg reflector) that would reflect the fraction of the light that is currently emitted towards the substrate towards the collection optics and using emitters closer to the sample surface.¹⁴

The combined lensing effect that we have reported, which derives from the deterministically fabricated metallic nano-ring and the super-SIL, is wavelength insensitive in a wide range of infra-red wavelengths, therefore broad-band, and compatible with any kind of solid-state emitter of classical or quantum light on any substrate. Furthermore, given that the emitters are in bulk, detrimental effects affecting, for instance, the coherence and/or stability of the light emission (that derive from the proximity to surface states due to the presence of etched surfaces¹⁵ that are needed when fabricating, for instance, pillar or photonic crystal structures) will be avoided in the emitters embedded within our devices.

In conclusion, we have shown how epoxy super-SILs can be implemented to enhance the extraction efficiency from single QDs and how such an effect can be combined to metallic nano-rings to realise broad-band devices allowing increases in the intensity of the light emitted by solid-state sources. We have implemented such a combined system to show bright emission from single QDs in bulk, reaching estimated photon fluxes approaching 1×10^6 counts per second, in a scalable and easy-to-fabricate device: the positioning of single QDs can be carried out in an automated system, ¹⁶ the dimensions of the rings are compatible with photolithography and nano-imprint, and the super-SIL deposition requires only optical microscopy tools.

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