

# Enhancement of non-classical radiation from quantum dots embedded within semiconductor Huygens' metasurfaces

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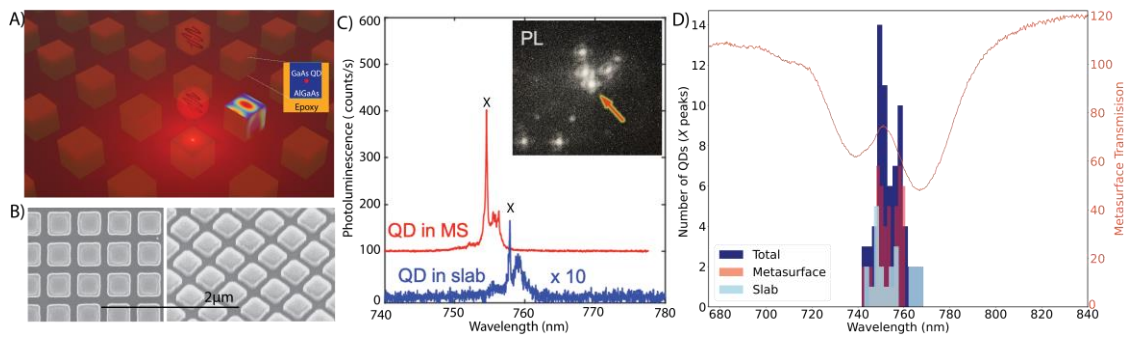
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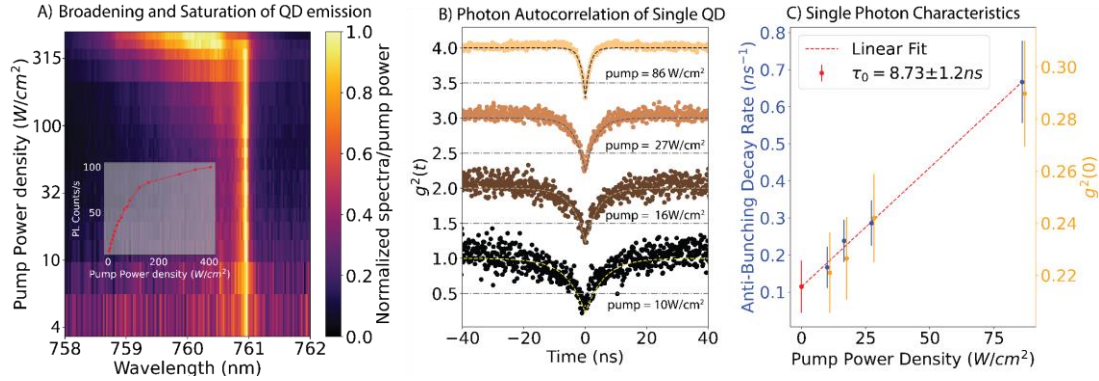
**Abstract:** We demonstrate position and size independent, order of magnitude increase in the collection efficiency and emission lifetime control of anti-bunched photons from local-droplet epitaxial GaAs quantum dots embedded within resonant semiconductor ( $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ ) Huygens' metasurfaces. © 2024 The Author(s)

The advancements in the field of photonic quantum information systems (QIS) has driven the development of high-brightness, on-demand, highly indistinguishable semiconductor epitaxial quantum dots (QDs) as single photon sources. Local-droplet epitaxy (LDE) QDs are particularly attractive as the growth is amenable to creating low density layers of strain free QDs, offer a path to greater emission tunability due to a larger freedom in choosing QD and host semiconductors, and the QDs offer deeper confinement potential than the strained Stranski-Krastanov (S-K) QDs [1]. However, the integration of the QDs to resonant nanophotonic cavities requires nm-scale precise positioning of the QDs and wavelength overlap between the QD emission and resonant cavity which remains a critical limitation for scaling up QD sources for QIS. Here we combine monodisperse, bright GaAs LDE QDs with a monolithic  $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$  Huygens' metasurface enabling us to increase by an order of magnitude the collection of efficiency and realize lifetime control of the anti-bunched photon emission from an isolated QD

Huygens' metasurfaces forms an ideal platform for embedding isolated semiconductor quantum dots because of the spatial uniformity of resonant modal profile (Fig 1A) within the resonators formed at the overlap of the fundamental electric and magnetic dipolar resonances [2]. This spatial uniformity of the electric and magnetic dipole resonant fields minimizes the effect of positioning the QD when compared with other higher-order multipolar Mie resonances whose field profiles have greater spatial non-uniformity [3]. The  $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$  thin films with embedded GaAs LDE QDs are epitaxially grown and fabricated into metasurface resonators using a flip-chip process [3] to remove the influence of the high index absorbing GaAs substrate on the embedded QD emitters (Fig 1B). Figure 1C shows that the PL spectra (measured at  $15 \pm 5\text{K}$ ) from an isolated GaAs LDE QD embedded within an exemplar Huygens' metasurface is



**Figure 1: Uniform Photoluminescence enhancement with Mie metasurfaces.** **A)** Schematic of Huygens' metasurface (MS) made of  $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$  resonators with an embedded GaAs local droplet epitaxy (LDE) grown quantum dot (QD) fabricated through a flip-chip process by encasing the resonators in epoxy on top of a glass substrate. The color-maps on the resonators' top and side shows the normalized electric field profiles (min – blue, max – red) of optical resonances. **B)** Scanning electron microscopy (SEM) images of fabricated metasurfaces with a top view (left) and tilted view (right). These images are taken prior to flip-chip bonding process. **C)** Measured photoluminescence (PL) spectra of GaAs LDE from a Huygens' metasurface and an unprocessed as-grown epitaxial film (slab) demonstrating an order of magnitude increase of emission into free space. The inset image shows the PL map from metasurface showing uniform enhancement of all of the QDs. **D)** Wavelength distribution of the charge narrow linewidth emission peak from 80 GaAs QDs from within the metasurface (red), the slab (light blue) and the total (dark blue) is plotted on the left axis. A typical transmission spectra (orange curve) of the metasurface normalized to transmission through the sapphire substrate is plotted on the right side axis. The dips in the spectra correspond to Mie resonances within the metasurface while the local peak at 750nm corresponds to the modal overlap between dipolar Mie modes. The QD emission statistics from the epitaxial growth shows a narrow distribution ( $< \pm 5\text{nm}$ ) aligned with the Huygens' point of modal overlap in the metasurface.



**Figure 2: Single photon characteristics of an Isolated QD.** **A)** The emission spectra from a single QD within the metasurface under continuous wave (CW) pump at 515nm is plotted post normalization for each pump power. The CW pump power is plotted on the y-axis on a log-scale. The bright streak at 760.9nm represents the narrow linewidth QD transition which undergoes spectral broadening under high pump powers. **Inset:** Saturation characteristics of the QD as a function of the CW pump power at 515nm. The QD emission rate starts saturating after 100W/cm<sup>2</sup> of focused illumination at 100 PL counts/s. **B)** Waterfall style plot of the photon auto-correlation from the same single QD under varying pump powers (10W/cm<sup>2</sup> – 86W/cm<sup>2</sup>). The dashed lines represent the single exponential fit to the rising edge of the  $g^2(t)$  plots. **C)** Plot of the anti-bunching rate (left axis, blue dots - fitted with the dashed lines in panel C) as a function of the pump power (CW at 515nm). The red dashed line represents a linear fit to the blue dots which is extrapolated to find the intercept at 0 pump power to extract the intrinsic exciton lifetime of the QD at  $8.73 \pm 1.2$ ns (Red dot). The anti-bunching dip at time 0ns is plotted in orange on the right axis to demonstrate that the QD behaves like a single photon source under these pump powers.

enhanced by an order of magnitude when compared to the QD emission from an unpatterned slab. Furthermore, the PL image (inset in Fig 1C) demonstrates the enhancement in emission from the QDs is agnostic of the position of the QDs within the metasurface. The transmission spectra of the Huygens' metasurface show the local maxima in the spectra at 745nm between the dips in the transmission spectra overlaps well with the narrow emission line from the QDs present on the wafer (red – collected within the metasurface, light blue – from the unpatterned slab). This alignment specifically highlights the capability of the low-Q semiconductor Huygens' metasurface to enhance QD emission regardless of the size distribution and randomness in position inherent to the LDE growth process.

To evaluate applications of QD embedded in MSs for QIS, we demonstrate single photon or anti-bunched emission from isolated GaAs LDE QDs embedded within the Huygens' resonators and its dependence on non-resonant continuous wave (CW) pump intensity at 515nm. We observe resolution limited spectra at low pump powers ( $< 100$ W/cm<sup>2</sup>) that broadens and saturates as the pump power is increased (Fig 2A). We perform correlated photon counting measurements in a Hanbury-Brown-Twiss (HBT) interferometer using two super-conducting nanowire broadband detectors to estimate the purity of the GaAs single photon source using the second order auto-correlation statistics ( $g^2(\tau)$ ) between the two detectors as a function of the CW pump power (Fig 2B). We measure anti-bunched ( $g^2(0) \sim 0.2$ - $0.3$ ) photo emission from an isolated QD within the metasurface under CW non-resonant pumping conditions with varying pump powers densities ( $< 100$ W/cm<sup>2</sup>). We track the photon emission rate ( $=1/\text{lifetime}$ ,  $\tau$ : fit using the equation  $g^2(t) = 1 - a \cdot e^{-t/\tau}$ , where  $a$  represents the purity of the single photon source such that  $a = 1 - g^2(t=0)$  and  $\tau$  represents the lifetime) as a function of the optical pump power to find a linear trend which when extrapolated to 0 optical pump power yields an intrinsic lifetime of the QD-resonator system of  $8.7 \pm 1.2$ ns (Fig 2C). Surprisingly, we discovered that at the low ( $< 100$ W/cm<sup>2</sup>) CW-pump (at 515nm) power densities incident on the metasurface, the effective lifetime of single photon emitters can be dynamically controlled by over an order of magnitude (8ns – 0.8ns) with the pump power. We expect the combination of semiconductor resonant metasurfaces with embedded quantum dot emitters will enable us to explore multiple particle quantum phenomena with the potential of novel quantum sensing and communication technologies.

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[1]. Gurioli, M., Wang, Z., Rastelli, A., Kuroda, T., & Sanguinetti, S. (2019). Droplet epitaxy of semiconductor nanostructures for quantum photonic devices. *Nature materials*, 18(8), 799-810.

[2]. Khoury, M., Quard, H., Herzog, T., Meijer, J., Pezzagna, S., Cuff, S., ... & Wood, T. (2022). Light Emitting Si-Based Mie Resonators: Toward a Huygens Source of Quantum Emitters. *Advanced Optical Materials*, 10(21), 2201295.

[3]. Santiago-Cruz, T., Gennaro, S. D., Mitrofanov, O., Addamane, S., Reno, J., Brener, I., & Chekhova, M. V. (2022). Resonant metasurfaces for generating complex quantum states. *Science*, 377(6609), 991-995.