Hybrid dielectric-plasmonic nanoantenna for efficient, bright and unidirectional photon sources

Pavel A. Dmitriev,†‡ Emmanuel Lassalle,∗,+ Lu Ding,† Darren C. J. Neo,†
Vytautas Valuckas,† Ramón Paniagua-Dominguez,† Joel K. W. Yang,¶,+†
Hilmi Volkan Demir,‡§ and Arseniy I. Kuznetsov∗,†

†Institute of Materials Research and Engineering, A*STAR (Agency for Science,
Technology and Research), 2 Fusionopolis Way, #08-03 Innovis, 138634 Singapore
‡LUMINOUS! Centre of Excellence for Semiconductor Lighting and Displays, School of
Electrical and Electronic Engineering, School of Physical and Mathematical Sciences,
School of Materials Science and Engineering, Nanyang Technological University, Singapore
639798, Singapore
¶Singapore University of Technology and Design, 8 Somapah Road, 487372, Singapore
§Department of Electrical and Electronics Engineering, Department of Physics, UNAM –
Institute of Materials Science and Nanotechnology, Bilkent University, Ankara 06800,
Turkey

E-mail: Emmanuel_Lassalle@imre.a-star.edu.sg, Arseniy_Kuznetsov@imre.a-star.edu.sg

Abstract

In this paper we propose and demonstrate single subwavelength hybrid dielectric-
plasmonic optical nanoantennas coupled to localised quantum dot emitters that con-
stitute efficient and bright unidirectional photon sources under optical pumping. To
achieve this, we devised a silicon nanoring sitting on a metallic (gold) mirror with a 10 nm gap in between, where an assembly of quantum dots is embedded. Such a structure supports both gap mode and (radiative) antenna mode resonances. We experimentally show that the assembly of quantum dots localised within the nanogap can efficiently couple to these two kind of modes, the resonant gap modes and the antenna modes, for the dual purpose of enhancing the absorption of the optical pump by the emitters, and for out-coupling the light into the far-field with high directionality. Moreover, almost independent control of the resonance spectral positions is achieved by simple tuning of geometrical parameters such as the ring outer and inner diameters, which is not possible with other, simpler, particle-on-mirror nanoantennas. Using the proposed architecture, we obtain average fluorescence enhancement factors of the assembly of emitters up to $654 \times$ folds together with a high radiation efficiency, and a directional emission of the photoluminescence into a cone of $\pm 17^\circ$ in the direction normal to the sample plane.

Introduction

Enhancing the fluorescence of localised light sources is important for both fundamental science, e.g. for study of quantum emitters and strong coupling effects, and for practical applications, e.g. for detection of fluorescent molecules for biological sensing and imaging.

Fluorescence enhancement can be achieved by different factors, such as local enhancement of the pump field intensity to enhance the absorption of the incident pump by the fluorescent emitters, modifying the electromagnetic environment via the local density of optical states (also known as “Purcell effect”) to make the light emission process faster and/or more efficient, and/or shape the emitted fluorescence towards the collection system to increase the collected signal. Nanostructures used for fluorescence enhancement usually aim to leverage on combining these methods.

Traditionally, plasmonic nanostructures have been used as a platform for these fluores-
cence enhancement applications, however, despite demonstrating very strong field confine-
ment,\textsuperscript{13,14} plasmonic nanoantennas suffer from large dissipation losses,\textsuperscript{15,16} which remains 
a fundamental problematic. For example, particle-on-mirror nanoantennas (also known as 
nanopatch antennas), which detain current record-breaking performances, have shown fluo-
rescence enhancement factors of $\sim 1000 - 2000 \times$ with Purcell factors of $\sim 1000 \times$\textsuperscript{17-23} but 
with very modest radiation efficiencies due to large non-radiative losses\textsuperscript{18,19}.

High-index dielectric nanophotonic structures,\textsuperscript{24-32} on the other hand, can nearly elim-
ninate dissipative losses and offer comparable scattering strengths and even greater flexibil-
ity in tuning the emission resonance and directionality,\textsuperscript{11,33,34} but are unable to compete 
with the field confinement of their plasmonic counterparts.\textsuperscript{30} Thus, in general, experi-
mental fluorescence enhancements and Purcell factors demonstrated by subwavelength dielectric 
nanostructures are well below the ones exhibited by plasmonic nanostructures,\textsuperscript{35-39} with best 
performance reported for silicon dimers,\textsuperscript{35,37} showing $\sim 500 \times$ fluorescence enhancement and 
$\sim 20 \times$ radiative decay rate enhancement, even though gallium phosphide dimers have been 
recently claimed to show much higher, over 3600 $\times$, fluorescence enhancement,\textsuperscript{40} with $\sim 22 \times$
total decay rate enhancement. Nevertheless, this field has mainly been driven by single 
molecule microscopy rather than photonic integration.

Recently, hybrid plasmonic-dielectric structures aim to take the best of both worlds, that 
is the high field enhancement and confinement of plasmonic structures and the low losses 
and greater flexibility of dielectric structures.\textsuperscript{41,42} Several hybrid nanoantenna designs have 
been proposed,\textsuperscript{43-50} promising in theory fluorescence enhancement factors over 1500 $\times$\textsuperscript{46} and 
Purcell factors over 5000 $\times$.\textsuperscript{43} Yet there are very few successful experimental demonstrations 
of such designs. Existing experimental works on hybrid nanostructures\textsuperscript{51-58} tend to focus 
on fabrication methodologies,\textsuperscript{51,52} second-harmonic generation\textsuperscript{53-55} or, when demonstrating 
directivity\textsuperscript{56,57} or Purcell enhancement (via cathodoluminescence),\textsuperscript{59} use intrinsic photolumi-
nescence of part of the antenna (e.g., using randomly dispersed dielectric particles\textsuperscript{59}). Only 
in a few works\textsuperscript{58,60} are quantum emitters successfully integrated and selectively coupled to
the antenna. These, however, typically involve micron-scaled hybrid bullseye antennas to demonstrate antenna directivity, but such systems cannot be shrink down to subwavelength sizes by their nature. The absence of more experimental demonstrations could be because the integration of emitters with the nanoantenna is on its own a very complex problem, usually requiring either precise manipulation or precise measurement of existing emitters location.

In this work, by sandwiching quantum dots between a silicon nanoring antenna and a gold mirror, within a nanogap of about ~10 nm, we alleviate the problem of quantum emitters localisation and obtain self-localised emitters located underneath the nanoantenna, without complex emitter manipulation or characterization. Thanks to the introduction of the metallic mirror, we achieve a very efficient coupling at the emission wavelength between the localised emitters and a strongly radiative mode supported by the nanoring antenna, that also provides strong out-of-plane directivity. Moreover, the existence of gap mode resonances enable high field confinement of the pump and thus absorption enhancement at the excitation wavelength. The hybrid dielectric-plasmonic nanoantenna reported here provides high degree of tunability of its optical resonances by simply changing its geometrical parameters, such as the inner and outer diameters of the nanoring. By carefully designing the silicon nanoring parameters to match its resonances with the quantum emitters emission and absorption wavelengths, we demonstrate experimentally a directional fluorescence enhancement over 650 that presents a large radiation efficiency and strong out-of-plane directionality, by a combination of three main fluorescence enhancement mechanisms — excitation, radiative decay and directivity enhancements.

**Design and fabrication**

The nanoantenna design, as shown schematically in Fig. 1a, consists of an amorphous silicon (aSi) nanoring on a gold (Au) mirror substrate, with an alumina (Al₂O₃) spacer in between
where the quantum emitters are embedded. Scanning electron microscopy (SEM) images of our fabricated samples are shown in Fig. 1b and as insets on Fig. 1c. On one hand, this design is inspired by a hybrid dielectric-metal resonator proposed in Yang et al.\textsuperscript{43}, that supports “gap mode resonances” which are created by surface plasmon polaritons bouncing back and forth at the dielectric particle termination.\textsuperscript{43,72} Thus, the resonance wavelengths have a geometric origin and are essentially related to the diameter of the particle with a similar condition as for Fabry-Perot resonances.\textsuperscript{43,72} Moreover, since these gap modes are strongly confined inside the nanogap, they can be exploited to enhance the absorption of the quantum emitters, which is what we do in this work. On the other hand, silicon nanorings can present strong scattering properties, with geometrically tunable “antenna resonances” (also called “Mie resonances”) that provide control over the scattering directionality, and potential ability to tailor the emission of electric or magnetic dipole emitters.\textsuperscript{73–76} In this work, we focus on the emission enhancement of electric dipole emitters by such antenna resonances.

We use CdSe/ZnS quantum dots (QDs) as quantum emitter model system. Transmission electron microscope image of the QDs is shown in Fig. 1c, and we obtained a rather good homogeneous distribution of QDs obtained after spin-coating, as shown in Fig. 1d. The QDs are chosen to have an emission peak centered around 650 nm.\textsuperscript{77} Their emission and absorption spectra on top of a glass substrate are given in Supp. Info. Section 1.

In order to separate the QDs from Au and to prevent quenching,\textsuperscript{78} a \( \approx 3 \) nm layer of \( \text{Al}_2\text{O}_3 \) was first deposited, and after spin-coating the QDs, a second \( \text{Al}_2\text{O}_3 \) layer of same thickness was deposited to protect the QDs from the following CVD deposition of aSi (see Methods section). All the QDs which were not precisely located between the Au mirror and aSi nanoantenna were etched out; hence, we managed to create spatially self-aligned and localized QDs in a nanogap of \( g \approx 10 \) nm, without requiring any complex emitter manipulation or characterization, similar to what was previously reported.\textsuperscript{36} A fabrication process flow schematics can be found in Supp. Info. Section 2.
Since the fabrication process exerts thermal and chemical stress that can potentially degrade the QDs, we characterized optically the QD layer at all steps of the fabrication process to quantify the changes in photoluminescence (PL) and fluorescence lifetimes. We observed that the intensity of the QD PL is reduced by almost 98% after depositing the second layer of Al₂O₃, that is in the configuration Au/Al₂O₃/QDs/Al₂O₃ (under the same excitation power). Moreover, time-resolved PL experiment (see Methods section) revealed that this drop in PL is correlated with a reduction of the QDs lifetime from $\tau_0 = 5.00 \text{ ns}$ to $\tau_0 = 0.65 \text{ ns}$ (see Supp. Info. Section 3). From these observations, we concluded that the QDs are degrading because of thermal stress during the ALD process (see Methods section), which makes the quantum yield drop from $\text{QY}^0 = 0.2$ (that we take according to Lim et al., and which matches our measured lifetime of $\tau_0 = 5.00 \text{ ns}$) to $\text{QY}^0 = 2.6 \times 10^{-3}$. Therefore, in the rest of this article, we choose to compare any PL enhancements when the aSi nanoantenna is present to the configuration Au/Al₂O₃/QDs/Al₂O₃ in which the QDs quantum yield is decreased to $\text{QY}^0 = 2.6 \times 10^{-3}$, and that we will call the “Reference” hereafter.

We designed and fabricated the aSi nanorings to have a fixed outer diameter of $D_{\text{out}} \approx 380 \text{ nm}$, chosen to support a gap mode resonance at around $\lambda_{\text{exc}} \approx 570 \text{ nm}$ (see Supp. Info. Section 4), in order to enhance locally the laser (pump) intensity at that excitation wavelength. The height of the nanoring was fixed to $H \approx 230 \text{ nm}$, chosen to support an antenna mode resonance around the emission wavelength of the QDs $\lambda_{\text{em}} \approx 650 \text{ nm}$ that strongly radiates in the upward direction (for out-of-plane emission). We fabricated different samples with approximately same outer diameter and height, but with different inner diameters $D_{\text{in}}$ (ring hole) varying between $\approx 60 \text{ nm}$ to $\approx 140 \text{ nm}$, in order to tune (shift) the antenna mode resonance (SEM images of the antennas are shown as insets on Fig. 1e). As we will see, because of the presence of these two classes of modes — the gap modes, which are mostly sensitive to $D_{\text{out}}$ of the ring, and the antenna modes that are strongly sensitive to $D_{\text{in}}$ — we are able to almost independently control the spectral positions of both types of resonances,
allowing us to adapt the design to match the excitation and emission wavelengths of the pump and QDs, respectively.

The fabricated nanoantennas were first characterized with dark-field scattering measurements (see Methods section). Experimental scattering spectra are shown in Fig. 1f and reveal that the scattering features are blue-shifted, with increasing $D_{\text{in}}$. This is in fair agreement with the scattering cross-section simulations spectra (see Methods section) shown in Fig. 1f. On the other hand, when increasing $D_{\text{out}}$ and fixing all other quantities, one observes a red-shift of the resonances (Supp. Info. Section 5).

We next computed the resonant optical modes $E_{\alpha}$ of the system as well as their complex eigenwavelengths $\lambda_{\alpha} = \lambda_{\alpha}^\prime + i \lambda_{\alpha}^\prime\prime$, with $\alpha$ labelling the mode, using quasi-normal mode (QNM) calculations (see Methods section). We identified that the main features appearing in the scattering spectra are associated to the excitation of three particular antenna modes, labelled $\alpha_1$, $\alpha_2$ and $\alpha_3$ in Fig. 1f.

The mode $\alpha_1$ has a $Q$-factors of about $Q \sim 30$, which is higher than for $\alpha_2$ and $\alpha_3$ (with $Q \sim 10$ and $Q \sim 20$, respectively), as can be seen from the sharper features in the scattering spectra. Moreover, its spectral position is more sensitive to $D_{\text{in}}$ than the two other QNMs, allowing for a higher degree of tunability. For these reasons, we choose the antenna mode $\alpha_1$ for the purpose of enhancing the emission of the QDs. The field profile of the mode $\alpha_1$, shown in Supp. Info. Section 6, reveals that most of the electric field is located inside the hole of the nanoring, which explains why this mode is highly sensitive to the parameter $D_{\text{in}}$.

Moreover, a comparison with the case of a standalone nanoring shows that the presence of the metallic mirror contributes to more than doubling the $Q$-factor of the mode $\alpha_1$, in addition to slightly blue-shifting its spectral position (see Supp. Info. Section 6, Fig. S6). Even more importantly, we found that, while for the standalone nanoring it is mostly the in-plane dipoles that couple to this mode, and only within a small area (located below the hole of the nanoring), the presence of the mirror makes the out-of-plane dipoles to mostly couple to this mode, within a much larger area (that forms a circular “band” surrounding the
hole of the nanoring), and with much higher coupling strength (see Supp. Info. Section 6, Fig. S7). In practice, since it is very challenging to control the position and orientation of the emitters precisely, the PL signal coming from the assembly of emitters is averaged out over emitters spatially distributed over the nanoantenna area with random dipole orientations. Therefore, a nanoring on top of a metallic mirror is expected to give a significantly higher PL enhancement compared to the case of a standalone nanoantenna, due to higher coupling strength and the more spatially “extended” coupling.

Results

In the following, we present the results obtained for the selected nanoantenna case having $D_{in} = 60$ nm, which we call “Antenna A” hereafter, for which we obtained the highest total fluorescence enhancement. To characterize the total fluorescence enhancement of the QDs with the nanoantenna compared to the Reference situation (in the absence of nanoantenna, denoted by the superscript “0” hereafter), we use the well established fact that, in the low excitation regime, the fluorescence enhancement per emitter (located at $r$) is proportional to the gains in excitation rate, collection efficiency and quantum yield:

$$EF_{th}(r) = \frac{\gamma_{exc}(r)}{\gamma_{exc}^0} \frac{D_{em}(r)}{D_{em}^0} \frac{QY(r)}{QY^0}$$  

The first ratio represents the excitation enhancement of the QDs, which corresponds to the excitation rate $\gamma_{exc}$, and is directly proportional to the local enhancement of the pump intensity at the position of the emitter $r$. This quantity depends therefore on the wavelength used for the pump laser $\lambda_{exc}$. The second ratio represents the collection efficiency enhancement, which corresponds to enhancement of the collected signal into a given numerical aperture (NA). It thus depends on the collection NA, denoted by $NA_{col}$, and also on the wavelength of fluorescent emission $\lambda_{em}$. Finally, the last ratio represents the enhancement of the emitter quantum yield (QY), the QY quantifying the radiation efficiency (i.e. the probability that
the excitation of the emitter actually results in the emission of a photon in the far field), which also depends on the emission wavelength $\lambda_{\text{em}}$. More specifically, in the Reference situation, $QY^0$ reads $QY^0 = \gamma_r^0/(\gamma_r^0 + \gamma_{\text{nr}}^0)$, with $\gamma_r^0$ and $\gamma_{\text{nr}}^0$ being the radiative and non-radiative decay rates of the QDs, respectively. With the introduction of the nanoantenna, the QY is modified as $QY(r) = \gamma_r(r)/[\gamma_r(r) + \gamma_{\text{nr}}^0 + \gamma_{\text{abs}}(r)]$, with $\gamma_r(r)$ is the new radiative decay rate modified in the presence of the nanoantenna, $\gamma_{\text{abs}}(r)$ is a new nonradiative decay channel that takes into account the absorption by the nanoantenna, and $\gamma_{\text{nr}}^0$ is assumed to remain unaffected by the introduction of the nanoantenna.

Note that all of these quantities are defined for a single electric dipole emitter with a given position $r$ and also having a fixed orientation of its dipole moment along the unit vector $u$, but for the sake of readability, we omit the dependence on the parameter $u$ in the above quantities. Also, it is interesting to note from Eq. (1) that the total fluorescence enhancement $EF_{\text{th}}$ depends on the type of emitter used in the Reference situation through $QY^0$, and is thus not an absolute figure-of-merit to characterize the performance of a given antenna.

After introducing the above quantities, we now use the fact that $QY/QY^0 = [\gamma_r(r)/\gamma_r^0][\tau(r)/\tau^0]$, where we further introduce the fluorescence lifetimes defined as $\tau(r) \equiv 1/[\gamma_r(r) + \gamma_{\text{nr}}^0 + \gamma_{\text{abs}}(r)]$ and $\tau^0 \equiv 1/(\gamma_r^0 + \gamma_{\text{nr}}^0)$ in the nanoantenna and Reference situations, respectively, to recast Eq. (1) into the following form, which will be more convenient for our purpose of characterising the underlying mechanisms at play in our nanoantenna:

$$EF_{\text{th}}(r) = \frac{\eta_{\text{exc}}(r, \lambda_{\text{exc}}) \eta_{\text{em}}(r, \lambda_{\text{em}}, NA_{\text{col}})}{\eta_{\text{tot}}(r, \lambda_{\text{em}})}$$

In Eq. (2), we defined the quantities $\eta_{\text{exc}} \equiv \gamma_{\text{exc}}/\gamma_{\text{exc}}^0$ and $\eta_{\text{em}} \equiv (D_{\text{em}}/D_{\text{em}}^0)(\gamma_r/\gamma_r^0)$, which quantify the enhancements in excitation and emission, respectively, and also the quantity $\eta_{\text{tot}} \equiv \tau_0/\tau$, which quantifies the fluorescence lifetime modification (or equivalently the total decay rate enhancement $\eta_{\text{tot}} = \gamma_{\text{tot}}/\gamma_{\text{tot}}^0$ with $\gamma_{\text{tot}} = \tau^{-1}$ and $\gamma_{\text{tot}}^0 = (\tau^0)^{-1}$ — also known as
the “Purcell factor”), and we made the dependence in terms of $\lambda_{\text{exc}}$, $\lambda_{\text{em}}$ and $\text{NA}_{\text{col}}$ of the terms $\eta_{\text{exc}}$, $\eta_{\text{em}}$ and $\eta_{\text{tot}}$ explicit. We emphasize that, through Eq. (2), the total fluorescent enhancement is related to but different from the Purcell factor.

**Excitation enhancement and gap resonance.** Experimentally, we first optimized the excitation wavelength $\lambda_{\text{exc}}$ to maximize the excitation enhancement $\eta_{\text{exc}}$. For that, we recorded the PL signal as we varied the pump wavelength $\lambda_{\text{exc}}$ from 488 to 588 nm, while maintaining a constant pump power, by collecting the light radiated into air using an objective lens with $\text{NA}_{\text{col}} = 0.9$. Even though the QDs absorb shorter wavelength light more efficiently (as shown in Supp. Info. Section 1), optimal pumping conditions for PL enhancement in the case of the nanoantenna were found to be at approximately $\lambda_{\text{exc}} = 570$ nm, as shown in Figs 2a. In all the subsequent measurements, we therefore fix the pump wavelength at $\lambda_{\text{exc}} = 570$ nm.

We confirmed by numerical simulations (see Methods section) the presence of a local maximum for the excitation enhancement around 583 nm, shown in Figs 2b. We obtain that the theoretical excitation enhancement factor $\langle \eta_{\text{exc}} \rangle$ — the bracket denoting position and orientation averaging of the QDs — is $\langle \eta_{\text{exc}} \rangle = 7.3$ at the pump wavelength $\lambda_{\text{exc}} = 583$ nm. We also show the field intensity in the horizontal cross-section located in the middle of the gap, and in vertical cross-section passing through the center of the nanoantenna, in Figs 2c, d, respectively, which reveal that most of the intensity is located in “hot spots” formed within the nanogap. We also checked in simulations that the field profile of this gap mode matches the one expected in theory for the gap mode resonance of symmetry ($n = 3, m = 1$) (see Supp. Info. Sections 4 and 7), for which the nanoantenna was designed, as mentioned earlier.

Finally, note that the intensity distribution of this gap mode (exploited for absorption enhancement), is mostly located around the hole of the nanoring, and thus presents a rather good spatial overlap with the area within which the mostly coupled emitters to the antenna mode (exploited for emission enhancement) are located, as discussed earlier.

**Emission enhancement and antenna resonance.** We next measured the angle-resolved
PL spectra of the nanoantennas, using back focal plane imaging technique (see Methods section), using the same objective lens with NA_{col} = 0.9. We show in Fig. 3a and b the back-focal-plane and angle-resolved PL of Antenna A, respectively. The corresponding images in Reference situation are shown in Fig. 3c and d. One can see that the nanoantenna situation looks brighter and present more directivity than the Reference situation.

Since our nanoantennas are designed for emission in the upward direction (out-of-plane), and in order to quantify the emission enhancement in the upward direction, we choose to integrate the angle-resolved PL of Fig. 3 over a collection NA_{col} = 0.3, which corresponds approximately to \pm 17\degree. The corresponding PL spectra is shown in Fig. 4a (blue line), together with the Reference PL (red line). In order to deduce the experimental total fluorescence enhancement from these PL spectra, we deconvoluted the PL signal of the nanoantenna with the Reference PL, according to the formula:

\[
\langle \text{EF}_{\text{exp}} \rangle = \frac{I}{I_0} \frac{A_0}{A}
\]  

(3)

where \( I \) (resp. \( I_0 \)) is the PL intensity collected in the nanoantenna situation (resp. in the Reference situation) — as shown in Fig. 4a — and \( A \) (resp. \( A_0 \)) is the area corresponding to the ring horizontal cross-section that reads \( A = \pi(D_{\text{out}}/2)^2 \) with \( D_{\text{out}} = 380 \text{ nm} \) where the QDs are located (resp. the area of the excitation spot \( A_0 = \pi(D_{\text{spot}}/2)^2 \) which is estimated to \( D_{\text{spot}} \approx 1.37 \mu\text{m} \); see Supp. Info. Section 8). The obtained experimental total fluorescence enhancement spectra is plotted in Fig. 4b. One can see one main peak around the emission wavelength of the QDs \( \lambda_{\text{em}} = 652 \text{ nm} \), for which we get a maximum total fluorescence enhancement factor of \( \langle \text{EF}_{\text{exp}} \rangle = 654 \) and also a secondary peak at 717 nm.

We corroborated these experimental total fluorescence enhancement spectra with numerical simulations (see Methods section). We computed in Fig. 4c the emission enhancement < \( \eta_{\text{em}} \) > in the upward direction (corresponding to NA_{col} = 0), because it corresponds to the direction of maximum directivity, and the bracket denotes once again position and orienta-
tion averaging of the QDs. One can see a good qualitative agreement with the experiment, with also the presence of two main peaks at $\lambda_{\text{em}} = 680\,\text{nm}$ and $\lambda_{\text{em}} = 725\,\text{nm}$. The shift of the resonances in experiment compared to simulation can be attributed to size variations in the height and outer diameter of the ring (even though we tried to fabricate all samples with these parameters being fixed, slight variations are unavoidable). The maximum emission enhancement at the main peak at $\lambda_{\text{em}} = 680\,\text{nm}$ is $\langle \eta_{\text{em}} \rangle = 206.2$.

Moreover, from the previous mode analysis shown in Fig. 1, we identify the two resonances at 680 nm and 725 nm with the antenna modes $\alpha_1$ and $\alpha_3$, respectively, as highlighted in Fig. 4c. One can even see a weaker third resonance around 700 nm, which we identify with the antenna mode $\alpha_2$ from Fig. 1f, as highlighted in Fig. 4c.

Finally, we obtained the experimental radiation patterns of the nanoantenna from the angle-resolved PL of Fig. 3a at the emission wavelengths $\lambda_{\text{em}} = 652\,\text{nm}$ and $\lambda_{\text{em}} = 717\,\text{nm}$. Their radiation patterns are presented in Fig. 5a and b, respectively (dark lines). The radiation pattern of the Reference situation is also shown (light blue lines). One can clearly see that each antenna resonance reshapes the emission into a main lobe oriented in the upward direction. The collection efficiency enhancement at $\lambda_{\text{em}} = 652\,\text{nm}$ is estimated to be $< D_{\text{em}}/D_{\text{em}}^0 > = 1.43$ in the collection NA $\text{NA}_{\text{col}} = 0.3$ (see Eq. (6) in Methods section for the formula used).

We also computed the theoretical radiation patterns (see Methods section) at the two main resonances exhibited in the simulated emission enhancement spectrum. Fig. 5c and d display the simulated radiation patterns at the resonances 680 nm and 725 nm, respectively (dark lines). The simulated reference situation is also shown (light blue lines). One can see a good qualitative agreement between experiment and simulation, showing strong out-of-plane directivity. We calculated from these simulation results a collection efficiency enhancement at $\lambda_{\text{em}} = 680\,\text{nm}$ of $< D_{\text{em}}/D_{\text{em}}^0 > = 1.31$ in the normal direction (see Methods section), from which together with the previously calculated maximum emission enhancement, we deduced from Eq. (2) the radiative decay rate enhancement to be $\langle \gamma_r/\gamma_r^0 \rangle = 156.2$ at $\lambda_{\text{em}} = 680\,\text{nm}$. 

12
Discussion

In order to account more quantitatively for the experimental total fluorescence enhancement, and to interpret the main physical mechanisms involved based on Eq. (2), it remains to determine the average lifetime reduction term \( \langle \eta_{\text{tot}} \rangle = \tau^0/\tau \). Since it is usually tricky to obtain this quantity theoretically,\textsuperscript{22} as it also depends on experimental input parameters such as the intrinsic non-radiative decay rate of the emitters \( \gamma_0^0_{\text{nr}} \), we choose to use the experimentally obtained value from the measured lifetimes with a time-resolved PL setup (see Methods section). After fitting the experimental data (see Supp. Info. Section 9, Fig. S12), we found a total decay rate enhancement or lifetime reduction of \( \langle \eta_{\text{tot}} \rangle = 1.79 \), which is a very modest value of Purcell factor, but is due to a certain extend to the very small value of the quantum yield of our emitters \( \text{QY}^0 = 2.6 \times 10^{-3} \), and hence is not entirely characteristic of the nanoantenna. Note that all other terms in Eq. (2) calculated theoretically are entirely characteristic of the nanoantenna and do not depend on the type of emitter, which is not the case for the term \( \langle \eta_{\text{tot}} \rangle \).

By then applying Eq. (2), and only using the experimentally measured lifetime reduction \( \langle \eta_{\text{tot}} \rangle = 1.79 \) to feed the theoretical formula, the other gains \( < \eta_{\text{exc}} > = 7.3 \) and \( < \eta_{\text{em}} > = 206.2 \) being all computed independently with full-wave simulations, we obtain a theoretical total fluorescence enhancement factor of \( \langle \text{EF} \rangle_{\text{th}} = 841 \) (corresponding to a collection in the upward direction), which is slightly higher than the experimentally measured one \( \langle \text{EF} \rangle_{\text{exp}} = 654 \) (for a collection NA of \( \text{NA}_{\text{col}} = 0.3 \) in the upward direction), but in good qualitative agreement. One can infer from this analysis that the emission enhancement \( < \eta_{\text{em}} > \) — which we recall is the product of the collection efficiency and radiative decay rate enhancements — is the main mechanism responsible for the total fluorescence enhancement observed here, with also a smaller but non-negligible contribution of the absorption enhancement.

We also analyse in the Supp. Info. Section 9 the results obtained for two other nanoantennas, called Antenna B and C, having identical outer diameters and heights as Antenna A whose results were presented here, but larger inner diameters, \( D_{\text{in}} = 80 \text{nm} \) and
$D_{in} = 110\,\text{nm}$, respectively. We found similarly good qualitative agreements between experiment and theory, by again only feeding the theoretical formula of Eq. (2) with the experimentally measured lifetime reduction for $\langle \eta_{\text{tot}} \rangle$. Moreover, this comparative study between Antennas A, B and C highlights that one can tune (blue-shift) the resonance exploited for emission by varying (increasing) $D_{in}$ with respect to the resonance exploited for absorption, as already anticipated from the mode study in Fig. 1f.

**Conclusion**

In this work, we experimentally demonstrated a highly efficient silicon nanoring on gold mirror nanoantenna coupled to localised quantum dots in a nanogap of $\sim 10\,\text{nm}$, which transformed very poor emitters into bright and nearly unidirectional sources, with total fluorescence enhancement factor of $654 \times$. Different mechanisms were exploited to increase the brightness of our nanoantennas. Firstly, the metallic mirror creates a nanogap that supports “gap” modes,$^{43,72}$ whose resonance wavelength mostly depends on the ring *outer diameter* and therefore can be easily tuned using this parameter. These were used to create “hot spots” in the nanogap that enhance the local field intensity, leading to an overall gain in the excitation/absorption efficiency of the QDs. Secondly, the nanoring antenna, designed to have strong scattering properties, increases the radiative rates and improves the directionality in the upward direction, with a resonance wavelength that can be tuned using the ring *inner diameter* to match with the emission wavelength of the emitters, and in an almost independent way from the resonance exploited for the excitation enhancement. This is not the case for most other particle-on-mirror studies,$^{18,20–22,59}$ where the particle has only one degree of freedom, giving very limited capability to tune the resonant responses of the whole system.

Despite the very modest Purcell factor reported here (partially due to the very low intrinsic quantum yield of the emitters used), it has been predicted that such hybrid nanoantennas
could potentially increase the Purcell factor by three orders of magnitude by further reducing of the gap size.\textsuperscript{43} Also, mode hybridization between “antenna” mode and “gap” mode could be engineered to retain the best properties of both worlds, that is a large Purcell factor and a large radiative efficiency and directionality.\textsuperscript{83} The experimental demonstration provided in this paper confirms the relevance of nanorings in hybrid dielectric-plasmonic nanostructures as highly tunable nanoantennas with subwavelength size, to create efficient, bright and directional sources in the visible spectral range.

**Methods**

**Fabrication**

To fabricate the nanoantenna structure, we deposited a 100 nm thick film of gold onto a silicon substrate with a 5 nm titanium adhesion layer by Electron-beam Physical Vapor Deposition (EBPVD, Denton Explorer) at a rate of 0.1 Å/s.

Next, we deposited a first layer of alumina (Al\textsubscript{2}O\textsubscript{3}) with thickness of \(\approx 3\) nm on the gold using Atomic Layer Deposition (ALD, Beneq TFS 200),\textsuperscript{84} from trimethylaluminum and H\textsubscript{2}O precursors at 120°C. After that, a layer of CdSe/ZnS alloyed quantum dots, synthesized according to,\textsuperscript{77} were spin-coated at 2000 rpm for 1 minute from a solution of 5 mg QDs per ml in toluene. The quantum dots were then covered by another \(\approx 3\) nm thick layer of alumina, this time using ALD at a temperature of 80°C. The final Al\textsubscript{2}O\textsubscript{3}/QDs/Al\textsubscript{2}O\textsubscript{3} sandwich structure has a total thickness of approximately 10 – 15 nm (ellipsometry measurements estimated the thickness, assuming a homogeneous alumina layer, of 13 nm), prior to patterning the silicon (Si) ring nanoantenna.

For the ring structure, we deposited a 230 nm thick film of amorphous silicon by Induction-Coupled Plasma Chemical Vapor Deposition (ICP-CVD, Oxford PlasmaPro 100) at 80°C from a SiH\textsubscript{4} precursor. Hydrogen silsesquioxane e-beam resist (Dow Corning XR-1541-06), spin coated at 5000 rpm for 1 minute and a change dissipation layer (Espacer 300AX01),
spin coated at 1500 rpm for 1 minute were used for the Electron Beam Lithography (EBL) writing (Exlionix ELS-7000), with a dose of $\approx 300 \text{ mC/cm}^2$. The sample was then developed by a NaOH/NaCl salty solution (1% wt. /4% wt. in de-ionized water) for 60 s and then rinsed by de-ionized water to stop the development. The final structures was created by Induction-Coupled Plasma Reactive Ion Etching (ICP-RIE, Oxford Plasmalab 100) using chlorine gas, with a slight over etch to etch any quantum dots not protected by the silicon structures.

**Optical characterization**

All optical measurements were performed in a microspectrometer setup, based on an inverted microscope (Nikon Ti-U) and a spectrometer system (Andor SR-303i spectrograph with a 150 lines/mm grating coupled to a $400 \times 1600$ pixel Andor Newton 971 EMCCD). Incident light was focused on the sample by a $100 \times$ objective lens with a 0.9 NA (Nikon LU Plan Fluor). Signal collected by the same objective lens was then projected onto the spectrograph entrance slit with a width of 250 $\mu$m.

- **Dark-field scattering measurements:** For dark-field scattering, white light from a halogen lamp was used to excite the sample, with the central low-$\vec{k}$ portion of the beam blocked from entering the objective lens, meaning only light scattered by the nanoantennas was collected and sent to the spectrograph. Reflectance of an silver mirror was used as the Reference.

- **Photoluminescence spectroscopy:** For photoluminescence measurements, a supercontinuum source (SuperK Power, NKT Photonics) with band-pass filter (SuperK Varia, NKT Photonics), pulse duration 70 ps, 78 MHz repetition rate was used to excite PL. The band-pass filter was used to scan the pump wavelength from 488 to 588 nm with a 10 nm bandwidth. Average pump power was maintained at $\approx 250 \mu$W. The pump laser was focused onto the sample substrate by the same $100 \times$ 0.9 NA objective lens,
resulting in an approximately 1.2 µm diameter laser spot (see Supp. Info. Section 7 for details on the method used to estimate the laser spot size). A 610 nm long pass filter was used to cut off any pump laser light in the collection beam path, the 610 nm cut-off can be clearly seen in all the photoluminescence curves on Figure 4 and in Supp. Figs S1, S3a and S10.

- **Back-focal-plane imaging:** To capture back-focal-plane images, the same 100× 0.9NA objective lens was used to collect light emitted by the nanoantennas, except that, instead of the image plane, the back focal plane of the objective was projected onto a CCD. The maximum collected angle, according to $NA = n \sin \theta$, and in our case, $n = 1$ (air), is about $\theta = 64.2^\circ$.

- **Back-focal-plane spectroscopy:** To measure angle-resolved PL spectra, the same 100× 0.9NA objective lens was used to collect light emitted by the nanoantennas, except that, instead of the image plane, the back focal plane of the objective was projected onto the spectrograph entrance slit. The maximum collected angle, according to $NA = n \sin \theta$, and in our case, $n = 1$ (air), is about $\theta = 64.2^\circ$.

- **Lifetime measurements:** Time-resolved photoluminescence was studied using a Picoquant Microtime 200 TCSPC system coupled to our microspectrometer setup. The same supercontinuum source was used to excite the sample. Spectrally integrated PL in a narrow 5 nm range, centered at 650 nm was collected using a Si single photon avalanche photodiode. The instrument response function (IRF) was recorded using excitation light scattered from the sample, where the IRF was measured to be 77 ps. PL decay measurements were fit using reconvolution with the measured IRF by a bi-exponential function.85
Numerical simulations

- **Scattering cross-sections:** To compute the scattering cross-sections, we used the finite-difference time-domain (FDTD) method in Ansys Lumerical FDTD. The Si particle (nanoantenna) in the presence of Au mirror was surrounded by a Total-Field Scattered-Field (TFSF) source, which simulates a planewave excitation and allows to separate directly the scattered field from the incident field. The distance between the TFSF box and nanoantenna was set to be larger than 100 nm. The incident wave was chosen to be linearly polarized and coming at normal incidence from the top of the nanoantenna. The scattering cross-section (i.e. the power flowing out of the particle divided by the source intensity, with a unit of $m^2$) was calculated using six plane monitors that formed a closed box surrounding the particle and located outside the TFSF source. The distance between the monitor box and the TFSF box was about 50 nm. Note that we consider for the simulations that the gap between the Si particle and Au mirror is filled with a homogeneous medium of refractive index corresponding to the one of alumina, i.e. $n = 1.77$.

- **Mode calculations:** The quasi-normal modes (QNMs), denoted $E_{\alpha}(r)$, can be defined as an eigenvalue problem of the solution of Maxwell equations in the absence of sources:

$$\nabla \times \frac{1}{\mu_0} \nabla \times E_{\alpha}(r) = \omega_{\alpha}^2 \varepsilon(r, \omega_{\alpha}) E_{\alpha}(r)$$

where $\omega_{\alpha} = \omega'_{\alpha} + i \omega''_{\alpha}$ denotes the complex eigenfrequency associated with the eigenmode $E_{\alpha}(r)$, and supplemented by outgoing boundary conditions (also known as the Sommerfeld radiation condition as $|r| \to \infty$). Note that $\omega''_{\alpha} < 0$ due to the convention $"e^{-i\omega t}"$ used for the time-harmonic fields. Here, the system is considered nonmagnetic with a vacuum permeability $\mu_0$, and $\varepsilon(r, \omega)$ denotes the relative permeability of the medium. the complex eigenwavelengths are defined as $\lambda_{\alpha} \equiv 2\pi c/\omega_{\alpha}$.

In order to solve Eq. (4) and obtain the eigenmodes and eigenfrequencies in the con-
figurations shown in Fig. 1g, we employed the “QNMEig solver”, developed by IOGS-CNRS,\textsuperscript{86} which, computes and normalizes the QNMs of plasmonic and photonic resonators, implemented using COMSOL Multiphysics. The QNMEig solver needs all dispersive material permittivities to be modelled by a $N$-pole Lorentz-Drude model, in order to reformulate Eq. (4) into a linear eigenvalue problem (see, for example, Yan et al.\textsuperscript{86}). The parameters of the Lorentz-Drude model that we used for the dispersive permittivities of amorphous silicon (nanoantenna) and gold (substrate) can be found in Supp. Info. Fig. S11.

\begin{itemize}
  \item \textit{Excitation enhancement simulations:}
    
    To compute the term $\eta_{exc}(r, \lambda_{exc})$ in Eq. (2), shown in Fig. 2, we use the fact that the normalized excitation rate can be readily expressed as:\textsuperscript{87}
    \begin{equation}
    \eta_{exc}(r, \lambda_{exc}) = \frac{|u \cdot E(r, \lambda_{exc})|^2}{|u \cdot E_0(r, \lambda_{exc})|^2}
    \end{equation}
    where we recall that $u$ is a unit vector showing the orientation of the emitter dipole moment, $r$ is the position of the emitter, and $E(r)$ (resp. $E_0(r)$) is the electric field at the emitter position $r$ for a given excitation source in the nanoantenna case (resp. in the Reference case). In our case, we consider illumination as a linearly polarized planewave coming at normal incidence from the top of the nanoantenna. We computed, using Ansys Lumerical FDTD, this enhancement factor by integrating the electric field intensity in the horizontal plane (i.e. parallel to the substrate) and located in the middle of the nanogap over an area corresponding to the ring horizontal cross-section, which we obtained the averaged excitation enhancement $\langle \eta_{exc} \rangle$ after averaging over all directions to account for randomly distributed and oriented dipole emitters, and normalizing by the case without Si nanoantenna.

  \item \textit{Emission enhancement simulations:} The calculations of the radiative emission enhancement $\eta_{em}(r, \lambda_{em})$ from Eq. (2) and shown in Figs. 4, were carried out using the
reciprocity principle, following the method well described in Ref.88. This method was implemented in Ansys Lumerical FDTD, where planewave sources were used with two orthogonal linear polarizations to excite the nanoantenna, using the TFSF source tool. Then, the near-field response was recorded in a plane located in the middle of the gap by point monitors, distributed in an area with the same size as the nanoantenna cross-section, and with a density of 3600 µm⁻² (i.e. we use approximately 408 point monitors homogeneously distributed below the ring within an area of πR² with R = D_out/2 = 190 nm). By reciprocity, the power recorded in each point monitor and calculated from the projection of the electric field along axis i (i = x, y or z) is equal to the emission power of light with the same polarization as the source from a point electric dipole oriented along i and located at the same position as the monitor. To obtain the averaged emission enhancement ⟨η_em⟩ of the assembly of electric dipoles randomly oriented and distributed uniformly under the nanoantenna, the power over all orientations i and over the spatial distribution of monitors was integrated, averaged over two orthogonal linear polarizations, in the direction out-of-plane (normal incidence), and normalized to the case without nanoantenna.

• Directional enhancement simulations:

To obtain the directivity patterns shown in Fig.5, we use the same reciprocal simulations as mentioned above, and made a sweep over all angles of incidence. In Fig.5, we show the emission angular power distribution (averaged over two orthogonal linear polarizations).

In order to quantify the percentage of light that can be collected in the upward direction with given collection NA NA_col, we use the relation (see e.g.44):

\[
D_{em} = \frac{\int_0^{2\pi} \int_0^{\theta_{col}} p(\theta, \phi) \sin(\theta) d\theta d\phi}{\int_0^{2\pi} \int_0^{\pi} p(\theta, \phi) \sin(\theta) d\theta d\phi} \tag{6}
\]

with \(p(\theta, \phi)\) being the angular power radiated into a certain solid angle (parametrized
by $\theta$ and $\phi$), and $\theta_{\text{col}}$ is defined as $\theta_{\text{col}} = \sin^{-1}(NA_{\text{col}})$. We consider only one direction for the calculated radiation patterns, which is the upward direction to be consistent with the enhancement spectra shown in Fig. 4, that is we set $\theta_{\text{col}} = 0$ in Eq. (6).

**Acknowledgement**

We would like to thank Sergey Gorelik and Zhaogang Dong for helpful discussions regarding the fluorescence lifetime measurements. We would also like to thank Jinfu Ho for assistance with nanofabrication.

We would like to acknowledge the Facility for Analysis, Characterisation, Testing and Simulation, Nanyang Technological University, Singapore, for use of their electron microscopy facilities.

This work was supported by the A*STAR SERC Pharos program (grant number 1527300025) and MTC Programmatic Grant No. M21J9b0085.

**Author contributions**

P.D., E.L., R.P.-D., H.V.D. and A.I.K. developed the concept. R.P.-D., J.K.W.Y., H.V.D. and A.I.K supervised and coordinated the work. P.D. fabricated all the nanostructures and performed the SEM and all the optical measurements of the fabricated structures. E.L. developed the code based on reciprocity principle for the emission calculations and performed the numerical simulations of radiative emission spectra and emission patterns and designed the nanoantenna. E.L. also performed the quasi-normal mode analysis and the computation of the scattering cross-sections spectra. P.D. also performed the calculations of the field enhancement in excitation configuration, some scattering cross-section calculations, as well as some emission spectra and emission patterns calculations. L.D. provided support and supervised some of the optical measurements. D.C.J.N. synthesized the QDs and helped to spin coat them onto the samples. V.V. helped with SEM measurements. P.D. and E.L.
wrote the manuscript, and all co-authors participated in results interpretation and read and reviewed the manuscript.

Supporting Information Available

Additional experimental details: laser spot size estimation, quantum dot lifetime measurements. Additional numerical simulation details: Fitting parameters for material models, ring mode simulations.
Figure 1: **Design and fabrication of hybrid dielectric-plasmonic nanoantennas.** a. Artist’s impression of the aSi nanoring placed on a Au substrate, with a Al₂O₃ spacer layer containing the embedded QDs (represented by the red dots). b. Tilted SEM image of a typical fabricated sample. The scale bar represents 400 nm. c. TEM image of the CdSe/ZnS quantum dots. The scale bar represents 10 nm. d. SEM image of the Au substrate coated by a first layer of Al₂O₃ itself covered with spin-coated QDs. The scale bar represents 400 nm. e. Experimental scattering measurements for different nanoantennas with same $D_{\text{out}} = 380$ nm and $H = 230$ nm, but increasing $D_{\text{in}}$ (from bottom to top). The insets show SEM images of the corresponding fabricated nanoantennas. The scattering for the case $D_{\text{in}} = 60$ nm (chosen to be discussed in the rest of this work and called "Antenna A"), is highlighted by a thicker line. The scale bars represent 100 nm. f. Simulated scattering cross-sections (lines, left axis) and calculated eigenwavelengths (real part $\lambda'_{\alpha}$) of three resonant modes labelled $\alpha_1$, $\alpha_2$, $\alpha_3$ (points, right axis) for different nanoantennas with same $D_{\text{out}} = 380$ nm and $H = 230$ nm, but increasing $D_{\text{in}}$ from $D_i = 0$ to $D_i = 120$ nm (by steps of 20 nm for the former case, and by steps of 10 nm for the latter case, from bottom to top). The mode widths (determined as two times the imaginary part $2\lambda''_{\alpha}$) are also shown by the shaded areas. The lines joining the points are guide-to-the-eye, and the horizontal dashed line shows the case $D_i = 60$ nm (Antenna A).
Figure 2: Excitation enhancement for Antenna A and comparison with simulations. 

a. Experimental PL intensity spectra in counts (cts) from Antenna A, showing their dependence on the pump excitation wavelength $\lambda_{\text{exc}}$ having a linear polarization and normal incidence. The maximum emission peak is obtained for $\lambda_{\text{exc}} = 570$ nm. 

b. Simulated average excitation enhancement $\langle \eta_{\text{exc}} \rangle$ for Antenna A as a function of the excitation wavelength $\lambda_{\text{exc}}$, calculated as the intensity enhancement of the pump electric field in the nanogap, according to Eq. (5) (see Methods section). The maximum intensity enhancement is obtained for $\lambda_{\text{exc}} = 583$ nm in the simulation. The vertical dashed line denotes the excitation wavelength at $\lambda_{\text{exc}} = 570$ nm. 

c. and d. Simulated electric field intensity distribution at $\lambda_{\text{exc}} = 570$ nm in the horizontal cross-section passing through the middle of the nanogap and in the vertical cross-section passing through the middle of the nanoantenna, respectively. In the simulations, the excitation source has its electric field linearly polarized along the x-axis, and comes at normal incidence, like in the experiment.
Figure 3: Brightness enhancement for Antenna A and comparison with Reference. a. Back focal plane image of the emission PL of Antenna A taken with a bandpass filter centered around $\lambda_{em} = 650$ nm with a FWHM of 30 nm and using a high-NA collection objective lens of $NA_{col} = 0.9$. b. Angle-resolved PL spectra of Antenna A using the same collection objective lens and projecting the image onto a spectrometer. c. and d. Back focal plane image and angle-resolved PL spectra of the Reference situation (absence of nanoantenna), respectively.
Figure 4: Emission enhancement for Antenna A and comparison with simulations. 

a. Experimental emission PL spectra for Antenna A (blue curve) and Reference (red curve), integrated over $NA_{\text{col}} = 0.9$. 

b. Emission PL enhancement $\langle EF_{\text{exp}} \rangle$ spectra obtained after deconvoluting the PL from Antenna A from the Reference PL, according to Eq. (3), and restricting the integration over $NA_{\text{col}} = 0.3$ to capture the directional emission only. A 610 nm long pass filter was used to cut off any pump laser light in the collection beam path (denoted by the blue area in the plots which are filtered in the collection channel while the red area reaches the spectrometer). 

c. Simulated emission enhancement $\langle \eta_{\text{em}} \rangle$ for Antenna A in the upward direction (see Methods section). In all plots, the vertical dashed line indicates the pump excitation wavelength $\lambda_{\text{exc}} = 570$ nm.
Figure 5: Directional enhancement for Antenna A and comparison with simulations. a. and b. Experimental radiation angular patterns for Antenna A (dark curves) at the two peaks of the PL enhancement $\langle EF_{\text{exp}} \rangle$ of Fig. 4b, located at $\lambda_{\text{em}} = 652$ nm and $\lambda_{\text{em}} = 717$ nm, respectively. The Reference case at $\lambda_{\text{em}} = 650$ nm is also shown (light blue curves). Grey dashed lines show the maximum collection angle corresponding to $\theta = 64.2^\circ$. c. and d. Simulated radiation angular patterns for Antenna A (dark curves) at the two peaks of the PL enhancement $\langle \eta_{\text{em}} \rangle$ of Fig. 4c, located at $\lambda_{\text{em}} = 680$ nm and $\lambda_{\text{em}} = 725$ nm, respectively. The Reference case at $\lambda_{\text{em}} = 650$ nm is also shown (light blue curves).
References

(1) Straubel, J.; Filter, R.; Rockstuhl, C.; Slowik, K. Plasmonic nanoantenna based triggered single-photon source. Physical Review B 2016, 93, 195412.

(2) Gwo, S.; Wang, C.-Y.; Chen, H.-Y.; Lin, M.-H.; Sun, L.; Li, X.; Chen, W.-L.; Chang, Y.-M.; Ahn, H. Plasmonic metasurfaces for nonlinear optics and quantitative SERS. Acs Photonics 2016, 3, 1371–1384.

(3) Curto, A. G.; Volpe, G.; Taminiau, T. H.; Kreuzer, M. P.; Quidant, R.; van Hulst, N. F. Unidirectional emission of a quantum dot coupled to a nanoantenna. Science 2010, 329, 930–933.

(4) Vladimirova, Y. V.; Zadkov, V. N. Quantum optics in nanostructures. Nanomaterials 2021, 11, 1919.

(5) Li, N.; Lai, Y.; Lam, S. H.; Bai, H.; Shao, L.; Wang, J. Directional control of light with nanoantennas. Advanced Optical Materials 2021, 9, 2001081.

(6) Lakowicz, J. R.; Shen, B.; Gryczynski, Z.; D’Auria, S.; Gryczynski, I. Intrinsic fluorescence from DNA can be enhanced by metallic particles. Biochemical and Biophysical Research Communications 2001, 286, 875–879.

(7) Geddes, C. D.; Cao, H.; Gryczynski, I.; Gryczynski, Z.; Fang, J.; Lakowicz, J. R. Metal-enhanced fluorescence (MEF) due to silver colloids on a planar surface: Potential applications of indocyanine green to in vivo imaging. The Journal of Physical Chemistry A 2003, 107, 3443–3449.

(8) Darvill, D.; Centeno, A.; Xie, F. Plasmonic fluorescence enhancement by metal nanostructures: shaping the future of bionanotechnology. Physical Chemistry Chemical Physics 2013, 15, 15709–15726.
(9) Punj, D.; Regmi, R.; Devilez, A.; Plauchu, R.; Moparthi, S. B.; Stout, B.; Bonod, N.; Rigneault, H.; Wenger, J. Self-assembled nanoparticle dimer antennas for plasmonic-enhanced single-molecule fluorescence detection at micromolar concentrations. *ACS photonics* **2015**, *2*, 1099–1107.

(10) Strobbia, P.; Languirand, E. R.; Cullum, B. M. Recent advances in plasmonic nanostructures for sensing: a review. *Optical Engineering* **2015**, *54*, 100902.

(11) Krasnok, A.; Caldarola, M.; Bonod, N.; Alú, A. Spectroscopy and biosensing with optically resonant dielectric nanostructures. *Advanced Optical Materials* **2018**, *6*, 1701094.

(12) Koenderink, A. F. Single-photon nanoantennas. *ACS photonics* **2017**, *4*, 710–722.

(13) Lozano, G.; Louwers, D. J.; Rodríguez, S. R.; Murai, S.; Jansen, O. T.; Verschuuren, M. A.; Rivas, J. G. Plasmonics for solid-state lighting: enhanced excitation and directional emission of highly efficient light sources. *Light: Science & Applications* **2013**, *2*, e66–e66.

(14) Schuller, J. A.; Barnard, E. S.; Cai, W.; Jun, Y. C.; White, J. S.; Brongersma, M. L. Plasmonics for extreme light concentration and manipulation. *Nature materials* **2010**, *9*, 193–204.

(15) Caldarola, M.; Albella, P.; Cortés, E.; Rahmani, M.; Roschuk, T.; Grinblat, G.; Oulton, R. F.; Bragas, A. V.; Maier, S. A. Non-plasmonic nanoantennas for surface enhanced spectroscopies with ultra-low heat conversion. *Nature communications* **2015**, *6*, 1–8.

(16) Albella, P.; Alcaraz de la Osa, R.; Moreno, F.; Maier, S. A. Electric and magnetic field enhancement with ultralow heat radiation dielectric nanoantennas: considerations for surface-enhanced spectroscopies. *Acs Photonics* **2014**, *1*, 524–529.
(17) Baumberg, J. J.; Aizpurua, J.; Mikkelsen, M. H.; Smith, D. R. Extreme nanophotonics from ultrathin metallic gaps. *Nature materials* **2019**, *18*, 668–678.

(18) Akselrod, G. M.; Argyropoulos, C.; Hoang, T. B.; Ciraci, C.; Fang, C.; Huang, J.; Smith, D. R.; Mikkelsen, M. H. Probing the mechanisms of large Purcell enhancement in plasmonic nanoantennas. *Nature Photonics* **2014**, *8*, 835–840.

(19) Akselrod, G. M.; Ming, T.; Argyropoulos, C.; Hoang, T. B.; Lin, Y.; Ling, X.; Smith, D. R.; Kong, J.; Mikkelsen, M. H. Leveraging nanocavity harmonics for control of optical processes in 2D semiconductors. *Nano Letters* **2015**, *15*, 3578–3584.

(20) Akselrod, G. M.; Weidman, M. C.; Li, Y.; Argyropoulos, C.; Tisdale, W. A.; Mikkelsen, M. H. Efficient nanosecond photoluminescence from infrared PbS quantum dots coupled to plasmonic nanoantennas. *Acs Photonics* **2016**, *3*, 1741–1746.

(21) Hoang, T. B.; Akselrod, G. M.; Mikkelsen, M. H. Ultrafast room-temperature single photon emission from quantum dots coupled to plasmonic nanocavities. *Nano letters* **2016**, *16*, 270–275.

(22) Hoang, T. B.; Akselrod, G. M.; Argyropoulos, C.; Huang, J.; Smith, D. R.; Mikkelsen, M. H. Ultrafast spontaneous emission source using plasmonic nanoantennas. *Nature communications* **2015**, *6*, 1–7.

(23) Sugimoto, H.; Yashima, S.; Fujii, M. Hybridized plasmonic gap mode of gold nanorod on mirror nanoantenna for spectrally tailored fluorescence enhancement. *Acs Photonics* **2018**, *5*, 3421–3427.

(24) Yang, Y.; Zenin, V. A.; Bozhevolnyi, S. I. Anapole-assisted strong field enhancement in individual all-dielectric nanostructures. *Acs Photonics* **2018**, *5*, 1960–1966.

(25) Krasnok, A.; Li, S.; Lepeshov, S.; Savelev, R.; Baranov, D. G.; Alú, A. All-optical
switching and unidirectional plasmon launching with nonlinear dielectric nanoantennas. 

*Physical Review Applied* **2018**, *9*, 014015.

(26) Feng, T.; Zhang, W.; Liang, Z.; Xu, Y. Unidirectional emission in an all-dielectric nanoantenna. *Journal of Physics: Condensed Matter* **2018**, *30*, 124002.

(27) Lepeshov, S.; Krasnok, A.; Alù, A. Enhanced excitation and emission from 2D transition metal dichalcogenides with all-dielectric nanoantennas. *Nanotechnology* **2019**, *30*, 254004.

(28) Zhang, T.; Li, X.; Xu, J.; Zhang, X.; Deng, Z.-L.; Li, X. Subwavelength Silicon Nanoblocks for Directional Emission Manipulation. *Nanomaterials* **2020**, *10*, 1242.

(29) Peter, M.; Hildebrandt, A.; Schlickriede, C.; Gharib, K.; Zentgraf, T.; Förstner, J.; Linden, S. Directional emission from dielectric leaky-wave nanoantennas. *Nano letters* **2017**, *17*, 4178–4183.

(30) Albella, P.; Poyli, M. A.; Schmidt, M. K.; Maier, S. A.; Moreno, F.; Sáenz, J. J.; Aizpurua, J. Low-loss electric and magnetic field-enhanced spectroscopy with subwavelength silicon dimers. *The Journal of Physical Chemistry C* **2013**, *117*, 13573–13584.

(31) Bakker, R. M.; Permyakov, D.; Yu, Y. F.; Markovich, D.; Paniagua-Domínguez, R.; Gonzaga, L.; Samusev, A.; Kivshar, Y.; Luk’yanchuk, B.; Kuznetsov, A. I. Magnetic and electric hotspots with silicon nanodimers. *Nano Letters* **2015**, *15*, 2137–2142.

(32) Kuznetsov, A. I.; Miroshnichenko, A. E.; Brongersma, M. L.; Kivshar, Y. S.; Luk’yanchuk, B. Optically resonant dielectric nanostructures. *Science* **2016**, *354*, aag2472.

(33) Kuznetsov, A. I.; Miroshnichenko, A. E.; Fu, Y. H.; Zhang, J.; Luk’Yanchuk, B. Magnetic light. *Scientific reports* **2012**, *2*, 1–6.
(34) Krasnok, A. E.; Miroshnichenko, A. E.; Belov, P. A.; Kivshar, Y. S. All-dielectric optical nanoantennas. *Optics Express* 2012, 20, 20599–20604.

(35) Regmi, R.; Berthelot, J.; Winkler, P. M.; Mivelle, M.; Proust, J.; Bedu, F.; Ozero, I.; Begou, T.; Lumeau, J.; Rigneault, H., et al. All-dielectric silicon nanogap antennas to enhance the fluorescence of single molecules. *Nano Letters* 2016, 16, 5143–5151.

(36) Cihan, A. F.; Curto, A. G.; Raza, S.; Kik, P. G.; Brongersma, M. L. Silicon Mie resonators for highly directional light emission from monolayer MoS 2. *Nature Photonics* 2018, 12, 284–290.

(37) Cambiasso, J.; König, M.; Cortes, E.; Schlücker, S.; Maier, S. A. Surface-enhanced spectroscopies of a molecular monolayer in an all-dielectric nanoantenna. *ACS Photonics* 2018, 5, 1546–1557.

(38) Rutckaia, V.; Heyroth, F.; Novikov, A.; Shaleev, M.; Petrov, M.; Schilling, J. Quantum dot emission driven by Mie resonances in silicon nanostructures. *Nano Letters* 2017, 17, 6886–6892.

(39) Kolchin, P.; Pholchai, N.; Mikkelsen, M. H.; Oh, J.; Ota, S.; Islam, M. S.; Yin, X.; Zhang, X. High Purcell factor due to coupling of a single emitter to a dielectric slot waveguide. *Nano Letters* 2015, 15, 464–468.

(40) Cambiasso, J.; Grinblat, G.; Li, Y.; Rakovich, A.; Cortés, E.; Maier, S. A. Bridging the gap between dielectric nanophotonics and the visible regime with effectively lossless gallium phosphide antennas. *Nano Letters* 2017, 17, 1219–1225.

(41) Lepeshov, S. I.; Krasnok, A. E.; Belov, P. A.; Miroshnichenko, A. E. Hybrid nanophotonics. *Physics-Uspekhi* 2019, 61, 1035.

(42) Barreda, Á.; Vitale, F.; Minovich, A. E.; Ronning, C.; Staude, I. Applications of Hybrid
Metal-Dielectric Nanostructures: State of the Art. *Advanced Photonics Research* 2021, 2100286.

(43) Yang, Y.; Miller, O. D.; Christensen, T.; Joannopoulos, J. D.; Soljacic, M. Low-loss plasmonic dielectric nanoresonators. *Nano letters* 2017, 17, 3238–3245.

(44) Barreda, A.; Hell, S.; Weissflog, M.; Minovich, A.; Pertsch, T.; Staude, I. Metal, dielectric and hybrid nanoantennas for enhancing the emission of single quantum dots: A comparative study. *Journal of Quantitative Spectroscopy and Radiative Transfer* 2021, 276, 107900.

(45) Zeng, X.; Yu, W.; Yao, P.; Xi, Z.; Lu, Y.; Wang, P. Metallo-dielectric hybrid antenna for high Purcell factor and radiation efficiency. *Optics express* 2014, 22, 14517–14523.

(46) Sun, S.; Zhang, T.; Liu, Q.; Ma, L.; Du, Q.; Duan, H. Enhanced directional fluorescence emission of randomly oriented emitters via a metal–dielectric hybrid nanoantenna. *The Journal of Physical Chemistry C* 2019, 123, 21150–21160.

(47) Sun, S.; Li, R.; Li, M.; Du, Q.; Png, C. E.; Bai, P. Hybrid mushroom nanoantenna for fluorescence enhancement by matching the stokes shift of the emitter. *The Journal of Physical Chemistry C* 2018, 122, 14771–14780.

(48) Zhang, T.; Xu, J.; Deng, Z.-L.; Hu, D.; Qin, F.; Li, X. Unidirectional enhanced dipolar emission with an individual dielectric nanoantenna. *Nanomaterials* 2019, 9, 629.

(49) Rusak, E.; Staude, I.; Decker, M.; Sautter, J.; Miroshnichenko, A. E.; Powell, D. A.; Neshev, D. N.; Kivshar, Y. S. Hybrid nanoantennas for directional emission enhancement. *Applied Physics Letters* 2014, 105, 221109.

(50) Sinev, I.; Iorsh, I.; Bogdanov, A.; Permyakov, D.; Komissarenko, F.; Mukhin, I.; Samusev, A.; Valuckas, V.; Kuznetsov, A. I.; Luk’yanchuk, B. S., et al. Polarization control
over electric and magnetic dipole resonances of dielectric nanoparticles on metallic films. *Laser & Photonics Reviews* 2016, 10, 799–806.

(51) Zuev, D. A.; Makarov, S. V.; Mukhin, I. S.; Milichko, V. A.; Starikov, S. V.; Morozov, I. A.; Shishkin, I. I.; Krasnok, A. E.; Belov, P. A. Fabrication of Hybrid Nanostructures via Nanoscale Laser-Induced Reshaping for Advanced Light Manipulation. *Advanced Materials* 2016, 28, 3087–3093.

(52) Ray, D.; Raziman, T.; Santschi, C.; Etezadi, D.; Altug, H.; Martin, O. J. Hybrid metal-dielectric metasurfaces for refractive index sensing. *Nano Letters* 2020, 20, 8752–8759.

(53) Aouani, H.; Rahmani, M.; Navarro-Cia, M.; Maier, S. A. Third-harmonic-upconversion enhancement from a single semiconductor nanoparticle coupled to a plasmonic antenna. *Nature nanotechnology* 2014, 9, 290–294.

(54) Shibanuma, T.; Grinblat, G.; Albella, P.; Maier, S. A. Efficient third harmonic generation from metal–dielectric hybrid nanoantennas. *Nano letters* 2017, 17, 2647–2651.

(55) Renaut, C.; Lang, L.; Frizyuk, K.; Timofeeva, M.; Komissarenko, F. E.; Mukhin, I. S.; Smirnova, D.; Timpu, F.; Petrov, M.; Kivshar, Y., et al. Reshaping the Second-Order Polar Response of Hybrid Metal–Dielectric Nanodimers. *Nano letters* 2019, 19, 877–884.

(56) Ho, J.; Fu, Y. H.; Dong, Z.; Paniagua-Dominguez, R.; Koay, E. H.; Yu, Y. F.; Valuckas, V.; Kuznetsov, A. I.; Yang, J. K. Highly directive hybrid metal–dielectric Yagi-Uda nanoantennas. *ACS nano* 2018, 12, 8616–8624.

(57) Ramezani, M.; Casadei, A.; Grzela, G.; Matteini, F.; Tütüncüğlu, G.; Rüffer, D.; Fontcuberta i Morral, A.; Gomez Rivas, J. Hybrid semiconductor nanowire–metallic Yagi-Uda antennas. *Nano letters* 2015, 15, 4889–4895.
(58) Kan, Y.; Ding, F.; Zhao, C.; Bozhevolnyi, S. I. Directional Off-Normal Photon Streaming from Hybrid Plasmon-Emitter Coupled Metasurfaces. *ACS Photonics* 2020, 7, 1111–1116.

(59) Maimaiti, A.; Patra, P. P.; Jones, S.; Antosiewicz, T. J.; Verre, R. Low-Loss Hybrid High-Index Dielectric Particles on a Mirror for Extreme Light Confinement. *Advanced Optical Materials* 2020, 8, 1901820.

(60) Andersen, S. K.; Bogdanov, S.; Makarova, O.; Xuan, Y.; Shalaginov, M. Y.; Boltasseva, A.; Bozhevolnyi, S. I.; Shalaev, V. M. Hybrid plasmonic bullseye antennas for efficient photon collection. *ACS Photonics* 2018, 5, 692–698.

(61) Li, L.; Chen, E. H.; Zheng, J.; Mouradian, S. L.; Dolde, F.; Schröder, T.; Karaveli, S.; Markham, M. L.; Twitchen, D. J.; Englund, D. Efficient photon collection from a nitrogen vacancy center in a circular bullseye grating. *Nano letters* 2015, 15, 1493–1497.

(62) Choy, J. T.; Bulu, I.; Hausmann, B. J.; Janitz, E.; Huang, I.-C.; Lončar, M. Spontaneous emission and collection efficiency enhancement of single emitters in diamond via plasmonic cavities and gratings. *Applied Physics Letters* 2013, 103, 161101.

(63) Livneh, N.; Harats, M. G.; Istrati, D.; Eisenberg, H. S.; Rapaport, R. Highly directional room-temperature single photon device. *Nano letters* 2016, 16, 2527–2532.

(64) Stella, U.; Boarino, L.; De Leo, N.; Munzert, P.; Descrovi, E. Enhanced directional light emission assisted by resonant Bloch Surface Waves in circular cavities. *ACS Photonics* 2019, 6, 2073–2082.

(65) Livneh, N.; Harats, M. G.; Yochelis, S.; Paltiel, Y.; Rapaport, R. Efficient collection of light from colloidal quantum dots with a hybrid metal–dielectric nanoantenna. *ACS Photonics* 2015, 2, 1669–1674.
(66) Makarov, S.; Sinev, I.; Milichko, V.; Komissarenko, F.; Zuev, D.; Ushakova, E.; Mukhin, I.; Yu, Y.; Kuznetsov, A.; Belov, P., et al. Nanoscale generation of white light for ultrabroadband nanospectroscopy. *Nano letters* 2018, 18, 535–539.

(67) Bek, A.; Jansen, R.; Ringler, M.; Mayilo, S.; Klar, T. A.; Feldmann, J. Fluorescence enhancement in hot spots of AFM-designed gold nanoparticle sandwiches. *Nano Letters* 2008, 8, 485–490.

(68) Schell, A. W.; Kewes, G.; Schröder, T.; Wolters, J.; Aichele, T.; Benson, O. A scanning probe-based pick-and-place procedure for assembly of integrated quantum optical hybrid devices. *Review of Scientific Instruments* 2011, 82, 073709.

(69) Schroder, T.; Schell, A. W.; Kewes, G.; Aichele, T.; Benson, O. Fiber-integrated diamond-based single photon source. *Nano letters* 2011, 11, 198–202.

(70) Werschler, F.; Lindner, B.; Hinz, C.; Conradt, F.; Gumbsheimer, P.; Behovits, Y.; Negele, C.; De Roo, T.; Tzang, O.; Mecking, S., et al. Efficient emission enhancement of single CdSe/CdS/PMMA quantum dots through controlled near-field coupling to plasmonic bullseye resonators. *Nano letters* 2018, 18, 5396–5400.

(71) Liu, S.; Srinivasan, K.; Liu, J. Nanoscale Positioning Approaches for Integrating Single Solid-State Quantum Emitters with Photonic Nanostructures. *Laser & Photonics Reviews* 2021, 2100223.

(72) Filter, R.; Qi, J.; Rockstuhl, C.; Lederer, F., et al. Circular optical nanoantennas: an analytical theory. *Physical Review B* 2012, 85, 125429.

(73) Kuznetsov, A.; Fu, Y. H. Antenna, assembly, and methods of forming the same. 2020; US Patent 10,873,135.

(74) Yik, J.; Bai, P.; Liu, Z.; Png, C. E.; Fu, Y. H.; Kuznetsov, A. Antenna, antenna
array, and methods of forming the same. 2020; International Publication Number, WO2020171771A1.

(75) Feng, T.; Xu, Y.; Liang, Z.; Zhang, W. All-dielectric hollow nanodisk for tailoring magnetic dipole emission. *Optics letters* **2016**, *41*, 5011–5014.

(76) Zenin, V. A.; Garcia-Ortiz, C. E.; Evlyukhin, A. B.; Yang, Y.; Malureanu, R.; Novikov, S. M.; Coello, V.; Chichkov, B. N.; Bozhevolnyi, S. I.; Lavrinenko, A. V., et al. Engineering nanoparticles with pure high-order multipole scattering. *ACS Photonics* **2020**, *7*, 1067–1075.

(77) Lim, J.; Jeong, B. G.; Park, M.; Kim, J. K.; Pietryga, J. M.; Park, Y.-S.; Klimov, V. I.; Lee, C.; Lee, D. C.; Bae, W. K. Influence of Shell Thickness on the Performance of Light-Emitting Devices Based on CdSe/Zn1-XCdXS Core/Shell Heterostructured Quantum Dots. *Advanced Materials* **2014**, *26*, 8034–8040.

(78) Anger, P.; Bharadwaj, P.; Novotny, L. Enhancement and quenching of single-molecule fluorescence. *Physical review letters* **2006**, *96*, 113002.

(79) Lyons, T. Y.; Williams, D. N.; Rosenzweig, Z. Addition of fluorescence lifetime spectroscopy to the tool kit used to study the formation and degradation of luminescent quantum dots in solution. *Langmuir* **2017**, *33*, 3018–3027.

(80) Lalanne, P.; Yan, W.; Vynck, K.; Sauvan, C.; Hugonin, J.-P. Light Interaction with Photonic and Plasmonic Resonances. *Laser & Photonics Reviews* **2018**, *12*, 1700113.

(81) Aouani, H.; Mahboub, O.; Bonod, N.; Devaux, E.; Popov, E.; Rigneault, H.; Ebbesen, T. W.; Wenger, J. Bright unidirectional fluorescence emission of molecules in a nanoaperture with plasmonic corrugations. *Nano letters* **2011**, *11*, 637–644.

(82) Matsuzaki, K.; Liu, H.-W.; Götzinger, S.; Sandoghdar, V. On quantum efficiency measurements and plasmonic antennas. *ACS Photonics* **2021**, *8*, 1508–1521.
(83) Zhang, C.; Hugonin, J.-P.; Greffet, J.-J.; Sauvan, C. Surface plasmon polaritons emission with nanopatch antennas: enhancement by means of mode hybridization. *ACS Photonics* **2019**, *6*, 2788–2796.

(84) Jin, H.; Moon, H.; Lee, W.; Hwangbo, H.; Yong, S. H.; Chung, H. K.; Chae, H. Charge balance control of quantum dot light emitting diodes with atomic layer deposited aluminum oxide interlayers. *RSC Advances* **2019**, *9*, 11634–11640.

(85) Dmitriev, P. kitchenknif/lifetime_reconvolution: v0.9. 2022; https://doi.org/10.5281/zenodo.6198822.

(86) Yan, W.; Faggiani, R.; Lalanne, P. Rigorous modal analysis of plasmonic nanoresonators. *Physical Review B* **2018**, *97*, 205422.

(87) Chen, H.; Yang, J.; Rusak, E.; Straubel, J.; Guo, R.; Myint, Y. W.; Pei, J.; Decker, M.; Staude, I.; Rockstuhl, C., et al. Manipulation of photoluminescence of two-dimensional MoSe 2 by gold nanoantennas. *Scientific reports* **2016**, *6*, 1–11.

(88) Zhang, S.; Martins, E. R.; Diyaf, A. G.; Wilson, J. I.; Turnbull, G. A.; Samuel, I. D. Calculation of the emission power distribution of microstructured OLEDs using the reciprocity theorem. *Synthetic Metals* **2015**, *205*, 127–133.
Supporting Information:

Hybrid dielectric-plasmonic nanoantenna for efficient, bright and unidirectional photon sources

Pavel A. Dmitriev,†‡ Emmanuel Lassalle,*† Lu Ding,† Darren C. J. Neo,†
Vytautas Valuckas,† Ramón Paniagua-Dominguez,† Joel K. W. Yang,¶†
Hilmi Volkan Demir,‡§ and Arseniy I. Kuznetsov*†

†Institute of Materials Research and Engineering, A*STAR (Agency for Science,
Technology and Research), 2 Fusionopolis Way, #08-03 Innovis, 138634 Singapore
‡LUMINOUS! Centre of Excellence for Semiconductor Lighting and Displays, School of
Electrical and Electronic Engineering, School of Physical and Mathematical Sciences,
School of Materials Science and Engineering, Nanyang Technological University, Singapore
639798, Singapore
¶Singapore University of Technology and Design, 8 Somapah Road, 487372, Singapore
§Department of Electrical and Electronics Engineering, Department of Physics, UNAM –
Institute of Materials Science and Nanotechnology, Bilkent University, Ankara 06800,
Turkey

E-mail: Emmanuel_Lassalle@imre.a-star.edu.sg; Arseniy_Kuznetsov@imre.a-star.edu.sg
S1  Quantum dot emission and absorption spectra

Figure S1: Absorption (green) and emission (purple) spectra of the CdSe/ZnS quantum dots used. Pump laser wavelength was 570 nm and a 610 nm long pass filter was used to cut off any pump laser light in the collection beam path.
Figure S2: Schematic showing nanoantenna fabrication steps.

The whole antenna structure process flow is shown in Figure S2. Details of each step of the process are given below:
1. We deposited a 100 nm gold thin film onto a silicon substrate with a 5 nm titanium adhesion layer by EBPVD (Denton Explorer) at a rate of 0.1 Å/s.

2. Next, using ALD (Beneq TFS 200), we deposited several nanometers (30 cycles) of alumina from trimethylaluminum and H$_2$O precursors at 120°C.

3. After that, a layer of CdSe/ZnS alloyed quantum dots$^{[4]}$ were spin-coated at 2000 rpm for 1 min from a solution of 5 mg QDs per mL in toluene (shown on Figure 1c in the main text).

4. The quantum dots were then covered by another layer of alumina (30 cycles), this time using ALD at a temperature of 80°C.

5. For the ring structure, we deposited an $\approx$ 230 nm thick film of amorphous silicon by ICP-CVD (Oxford PlasmaPro 100) at 80°C from a SiH$_4$ precursor (45 sccm SiH$_4$ and 30 sccm Ar at 8 mTorr process pressure, 50 W RF power at 20 DC forward bias and 3000 W ICP RF power).

6. Hydrogen silsesquioxane e-beam resist (Dow Corning XR-1541-06), spin-coated at 5000 rpm for 1 min, and a change dissipation layer (Espacer 300AX01), spin coated at 1500 rpm for 1 min, were used for the EBL writing.

7. EBL (Elionix ELS-7000) was performed at an acceleration voltage 100 kV and a current of 500 pA, with a dose of $\approx$ 300 mC/cm$^2$. The sample was then developed in a NaOH/NaCl salty solution (1% wt./4% wt. in de-ionized water) for 60 s and then rinsed by de-ionized water to stop the development.

8. The final structure created by ICP-RIE etching (Oxford Plasmalab 100) using chlorine gas, with a slight over etch to etch any quantum dots not protected by the silicon structures (22 sccm Cl$_2$ at 5 mTorr process pressure, 100 W RF power and 300 W ICP RF power).
S3  Lifetime and PL measurements during fabrication process

Time-resolved PL decay shown in Fig. S3 was fit by a bi-exponential function using re-convolution. S2 Fitting parameters are shown in Table S1. For all measured samples, the time-resolved decay showed two characteristic decay rates. For the case of quantum dots on a substrate, this is commonly interpreted as quantum dots with different orientations exhibiting different decay rates because of the substrate interactions. S3

![Figure S3: Photoluminescence (a.), lifetimes (b.) and fit decay components (c.) of reference quantum dots. Purple lines - quantum dots on SiO₂. Green lines - quantum dots directly on Au mirror, with noticeable quenching. Blue lines - quantum dots on Au mirror but with Al₂O₃ spacer separating them from the gold, which completely stops quenching. Red lines - damaged quantum dots after second Al₂O₃ deposition.](image-url)
Table S1: Amplitude ($a_1, a_2$) and decay ($\tau_1, \tau_2$) components of the bi-exponential fitting used to the experimental lifetime measurements.

|          | $a_1$, a.u. | $\tau_1$, ns | $a_2$, a.u. | $\tau_2$, ns |
|----------|-------------|--------------|-------------|--------------|
| QDs on SiO$_2$ | 0.05        | 0.27         | 0.019       | 3.80         |
| QDs on Au    | 0.08        | 0.17         | 0.012       | 1.30         |
| QDs on Al$_2$O$_3$/Au | 0.04       | 0.27         | 0.022       | 5.00         |
| Al$_2$O$_3$ on QDs on Al$_2$O$_3$/Au | 0.10        | 0.10         | 0.006       | 0.65         |
S4  Gap mode resonance

Figure S4:  a. Spatial distribution of the mode of a circular resonator of symmetry \((n = 3, m = 1)\), calculated from Eq. (S1). Redder colours denote positive values, and bluer colour negative ones.  

b. Schematic of the metal/dielectric 1/dielectric 2 configuration.  

c. Multilayer dispersion relation given by Eq. (S2) for a nanogap of thickness \(g = 10\) nm and permittivity \(\varepsilon_1 = 1.77^2\) sandwiched between an aSi semi-infinite covering layer of permittivity \(\varepsilon_2 = \varepsilon_{\text{Si}}(\omega)\) given below without losses (i.e. \(\gamma_{\text{Si}} = 0\)) and a semi-infinite gold mirror whose dispersive permittivity \(\varepsilon_m\) is the one given below without losses (i.e. \(\gamma_{1,\text{Au}} = 0\) and \(\gamma_{2,\text{Au}} = 0\)). The oblique dark line corresponds to the light cone in a homogeneous aSi medium \(\omega = kc/\sqrt{\varepsilon_{\text{Si}}(\omega \to 0)}\) with \(\sqrt{\varepsilon_{\text{Si}}(\omega \to 0)} = 3.3\). The horizontal black line corresponds to the cutoff frequency \(\omega_c(\varepsilon_1)\) obtained as the zero of the standard dispersion relation \(k = \omega/c\sqrt{\varepsilon_1\varepsilon_m/(\varepsilon_1 + \varepsilon_m)}\), that is by solving: \(\varepsilon_1 + \varepsilon_m(\omega_c) = 0\).

It is not very convenient to identify the gap modes using the QNM formalism because in this spectral region (around and below 600 nm), the number of modes is very large and they are not spectrally well separated, which makes their analysis quite tedious. Instead, we use a more intuitive analytical approach to find the modes, based on geometrical considerations only.\(^\text{S4,S5}\)
As mentioned in Yang et al.\textsuperscript{55}, the gap resonances of our system can be understood as “the surface plasmon of a planar multilayer metal-dielectric system restricted to specific quantized wavevectors”. The cylindrical symmetry of our system implies that resonances can be labeled with indices \((n, m)\), enumerating field variations in the radial and azimuthal directions, respectively. The wavevectors \(k_{nm}\) are “quantized” due to the geometry of the above nanoantenna, which reflects the surface plasmon at its boundary, similarly to the modes of a Fabry-Perot cavity.

The gap modes can be modelled as the modes of a two dimensional circular resonator, which are found by solving the wave equation in polar coordinates \((r, \phi)\) — which becomes a Bessel’s equation — with Dirichlet boundary conditions.\textsuperscript{56} The solutions of this wave equation take the form:\textsuperscript{56}

\[
E(r, \phi, t) = A \cdot J_m(kr) \cdot \cos(m\phi) \cdot e^{-i\omega t} \tag{S1}
\]

where \(A\) is the amplitude of the mode, \(k\) is the wavevector, and \(m\) is the azimuthal number. Due to the Dirichlet boundary conditions, the wavevector is quantized according to:

\[
k_{nm}R_{\text{out}} = J_n,\]

where \(J_n\) is the \(n^{th}\) zero of the Bessel’s function of the first kind of order \(m\) (note that in reality, the solutions for a ring are a combination of Bessel functions of the first and second kind, and the discrete wave-vectors \(k_{nm}\) must be found numerically,\textsuperscript{56} but as a first approximation, we consider the ring inner diameter negligibly small compared to the outer diameter \(D_{\text{in}} \ll D_{\text{out}}\), which allows us to solve these equations to the first order and to obtain the same results as for a circular resonator, i.e. a plain disk).

Next, the resonance frequency associated to these quantized wavevectors is found by “sampling” the multilayer dispersion relation of surface plasmons in the configuration metal/dielectric 1/dielectric 2 according to Yang et al.\textsuperscript{55}, where in our case the metal is gold, dielectric 1 is alumina and dielectric 2 is silicon (configuration shown on Fig. S4a, assuming the thickness of Si semi-infinite, which is justified if we are looking at the surface plasmon
We thus calculate the multilayer dispersion relation given by solving the following transcendental equation:\textsuperscript{57}

\[
\tanh (k_1 g) = -\frac{1 + \varepsilon_2 k_m/(\varepsilon_m k_2)}{\varepsilon_2 k_1/(\varepsilon_1 k_2) + \varepsilon_1 k_m/(\varepsilon_m k_1)}
\]  

(S2)

where “tanh” is the hyperbolic tangent function, \( k_i = \sqrt{k^2 - \omega^2/c^2} \varepsilon_i \), with \( i = 1, 2 \) for dielectrics 1 or 2 and \( i = m \) for the metal substrate, and \( g \) is the thickness of the dielectric 1 (the metal and dielectric 2 are considered semi-infinite). This dispersion is plotted in Fig. S4c, corresponding in our case to \( g = 10 \) nm, \( \varepsilon_1 = 1.77^2 \) (permittivity of Al\(_2\)O\(_3\)), \( \varepsilon_2 = \varepsilon_{Si}(\omega) \) (dispersive permittivity of aSi given below), and the metal permittivity \( \varepsilon_m = \varepsilon_{Au}(\omega) \) (dispersive permittivity of Au given below).

We then apply the above theory to our nanoantenna geometry. In our case, since we are looking at a gap mode which is excited by the pump laser, which has a linear polarization in our experiment, only modes with \( m = 1 \) can be excited due to symmetry compatibility. We find that the discrete wavevector \( k_{31} \) (which corresponds to the third zero of the Bessel function of order \( m = 1 \)) for a radius \( R_{out} = 190 \) nm (vertical dashed red line in Fig. S4c), intersects with the dispersion curve (blue curve in Fig. S4c), at the resonance frequency 574 nm, which is in quantitative agreement with the observed resonance wavelength of the pump around \( \lambda_{exc} \approx 570 \) nm. The spatial profile of this mode with symmetry \( (n = 3, m = 1) \), calculated from Eq. (S1) with \( t = 0 \), is shown in Fig. S4a.
Figure S5:  

**a.** Experimental scattering spectra from antennas with an inner radius of about 40 nm and height of about 230 nm while increasing the outer radius. 

**b.** FDTD-simulated scattering spectra of antennas with a inner radius of 40 nm, height of 230 nm while increasing the outer radius.
S6  Comparison between “antenna” mode of the nanoring in free space and on gold mirror cases

Figure S6: Comparison between the Quasi-Normal Mode (called “antenna mode” in the main text) of the ring (outer diameter $D_{\text{out}} = 380\,\text{nm}$ and height $H = 230\,\text{nm}$) standing in free space (blue dots) and on top of a gold substrate (red stars).  

a. and b. Schematics of the standalone nanoantenna in free space and of the nanoantenna on top of gold substrate, respectively.  
c. Inner (hole) diameter $D_{\text{in}}$ vs. eigenwavelength (real part) of the mode.  
d. Inner diameter $D_{\text{in}}$ vs. quality factor $Q$ of the mode. The cases of $D_{\text{in}} = 60, 80$ and $110\,\text{nm}$, (corresponding to what we call Antenna A, B and C later on, respectively) are also highlighted (horizontal black dashed lines). The insets represent the norm of the QNM mode $|E_\alpha|$ for the disk (lower inset) and the ring with $D_{\text{in}} = 60\,\text{nm}$ (upper inset) in the plane perpendicular to the substrate, passing through the center of the nanoantenna.

To understand how the presence of the metallic (gold) mirror affects the antenna mode (labelled $\alpha_1$ in the main text), we calculate the eigenfrequency and $Q$-factor of this mode when the nanoring is standing in free space, see Fig. S6a and b, respectively. The field
profiles of this mode $E_\alpha$ are also shown as insets.

Now, in order to understand how quantum emitters might couple to this antenna mode, we compute the mode volume $V_\alpha(r)$ associated with this QNM, which is given by the relation:

$$V_\alpha(r) = \frac{1}{2\epsilon_0 (E_\alpha(r) \cdot u)^2}$$

with $\epsilon_0$ being the vacuum permittivity. One can see from Eq. S3 that the volume quantifies the interaction between electric dipole emitters with dipole orientation along unit vector $u$ and the QNM field $E_\alpha(r)$ at the position of the emitter $r = (x, y, z)$. The smaller the mode volume, the stronger the interaction.

We calculate the spatial distribution of the mode volume associated with the antenna mode, across the $xy$ plane located 5 nm underneath the ring, in the case of a disk (lower panel) and a ring with $D_{in} = 60$ nm (upper panel), related to dipoles oriented out-of-plane (i.e. perpendicular to the plane, denoted by the symbol $\perp$) and dipoles oriented in-plane (i.e. parallel to the plane, denoted by the symbol $\parallel$).

For the standalone nanoring/nanodisk, shown in Fig. S7a, in both cases of the disk and the ring, dipole emitters whose dipole moment orientation is in-plane are more efficiently coupled to the nanostructure in the central area, compared to emitters with out-of-plane orientation.

In stark contrast, for the nanoantenna on top of gold mirror, shown in Fig. S7b, one can see that the coupling of in-plane dipoles, is extremely small, while the coupling of out-of-plane dipoles is substantial within a circular “band” surrounding the central area (corresponding to the hole area). This behavior can be understood partially by the fact that the presence of the metallic substrate forces the tangential component of the electric field to be near zero at the interface.

We therefore expect to induce a larger enhancement of the PL of quantum emitters distributed in the nanogap between the nanoantenna and gold mirror even after averaging over random orientations and spatial distribution, compared to the antenna alone situation,
owing to a stronger coupling and also “delocalised” coupling over a larger area beneath the ring, where the emitters are localised.

Figure S7: Spatial distribution of the real part of the inverse mode volume $\text{Re}(1/V_\alpha)$ associated with the chimney mode, calculated in a plane located 5 nm below the nanoantenna, in the two cases of a disk and a ring with $D_{in} = 60$ nm, both having an outer diameter $D_{out} = 380$ nm and height $H = 230$ nm in free space. We discriminate between the out-of-plane oriented dipoles (symbol $\perp$) and the in-plane oriented dipoles (symbol $\parallel$, where in this case we averaged over $x$ and $y$-orientations). The dotted white lines represents the projection onto the plane of the outer and inner diameters of the ring.
S7  Field profile at the pump excitation wavelength

The local field enhancement shown in the Fig. 2 in the main text is mostly due to the $z$-component of the electric field, which we show in Fig. S8. One can note the same symmetry as the mode profile of a circular resonator shown in Fig. S4a.

Figure S8: Component $E_z$ of the electric field at excitation wavelength $\lambda_{\text{exc}} = 570$ nm in (a) the horizontal cross-section 5 nm underneath the nanoring and (b) in the vertical cross-section passing through the middle of the nanoring.
S8  Laser spot size estimation

Figure S9:  

a. SEM of a marker on the sample, $10 \times 5 \mu m^2$. 

b. Image of the same marker through $100 \times 0.9$NA objective lens used to focus laser. Image scale is approximately 7 pixels per micron. 

c. Image of the laser spot using the same optical setup. Measured spot diameter at intensity $I = \frac{1}{e^2} I_{\text{max}}$ is $1.37 \mu m$. 

Comparison between antennas A, B and C

Figure S10: a, b, c Experimental PL signal for the nanoantennas (red) compared to Reference quantum dots (blue), d, e, f PL enhancement factor estimated from dividing nanoantenna PL by Reference PL. g, h, i Simulated enhancement factor for the nanoantennas. Dashed line in the plots indicates the pump wavelength. Blue area in the plots is filtered in the collection channel; only signal from the red area reaches the spectrometer.

In this Section, we compare three cases experimentally and in simulations of nanoantenna on top of gold mirror with same height $H = 230$ nm, outer diameter $D_{\text{out}} = 380$ nm, but different inner diameters: $D_{\text{in}} = 0, 60, 80, 110$ nm, which we will call hereafter “Antenna A”, “Antenna B” and “Antenna C”, respectively.

Their experimental total enhancement spectra are shown in Fig. S10 for a collection $\text{NA}_{\text{col}} = 0.3$ and compared with simulations of the emission enhancement in the normal
direction in Fig. S10. Experimentally, around the 650 nm, we have enhancement factors of 654, 349 and 98 for Antenna A, B and C, respectively. One can note that the positions of the peaks are slightly blue-shifted as one moves from Antenna B to Antenna C in experiment. Antenna A does not follow this trend, but this is attributed to slight variations in the height and outer diameter compared to the nominal values.

![Figure S11: Experimental and simulated radiation patterns for the nanoantennas at resonant modes with the highest enhancement. Grey dashed lines show the maximum collection angle.](image)

In simulations, the peaks are blue-shifted as one goes from Antenna A to C as expected, and the emission wavelengths corresponding to the maximum of the first resonance are at 680, 665 and 639 nm, respectively. The calculated emission enhancements $\langle \eta_{em} \rangle$ in the normal direction are about 206, 136 and 54, respectively.

We also show the measured and calculated radiation patterns in Fig. S11, that reveal directivity in the out-of-plane direction in all cases.

In order to account for the different values of enhancement, we compute the Purcell factor $F_\alpha(r)$ associated to the antenna mode responsible for the peak around 650 nm (labelled $\alpha_1$...
in the main text), according to the formula:

\[
F_\alpha(r) = \frac{6\pi c^3}{\omega_\alpha^3} Q_\alpha \text{Re} \left( \frac{1}{V_\alpha(r)} \right)
\]  

(S4)

The Purcell factor quantifies the decay rate enhancement due to the coupling with QNM \(\alpha\) compared to free space, for an emitter located at position \(r\), and for a perfect matching of the emission frequency of the emitter with the real part of the QNM eigenfrequency. We found that the maximum Purcell factor \(F_\alpha^{\perp}(r^{\text{max}})\), corresponding to a dipole emitter with an out-of-plane orientation (\(\perp\)) and located at the position \(r^{\text{max}}\) where \(\text{Re}(1/V_\alpha(r^{\text{max}}))\) is maximum (that is in the regions corresponding to the yellow/orange bands in Fig. S7), increases as the hole diameter decreases — and is maximized for the disk case; see Table S2. This indicates a stronger coupling of quantum emitters with the QNM as the hole diameter decreases. We also calculated the averaged Purcell factor denoted \(\langle F_\alpha \rangle\) for emitters spreading over the entire area below the ring (corresponding to the geometric cross-section of the ring \(\sigma_{\text{geo}} = \pi R_{\text{out}}^2\) with \(R_{\text{out}} = D_{\text{out}}/2 = 190\,\text{nm}\)) and with random orientations; these values are displayed in Table S2 together with the quality factors, \(Q_\alpha\). One can see that the averaged Purcell factor associated to this antenna mode decreases as the inner diameter increases, in agreement with the measured and simulated enhancement factors.

Table S2: Quasi-normal modes related quantities for four antennas configurations on gold mirror: Disk, Antenna A, Antenna B and Antenna C. Shown in the table are: the real and imaginary parts of the complex eigenfrequencies \(\omega_\alpha\), the \(Q\)-factors associated with each mode calculated as \(Q_\alpha = -\omega_\alpha'/(2\omega_\alpha'')\), the maximum Purcell factor associated to each mode calculated for a dipole with out-of-plane orientation and located in the positions where the coupling is maximum, and the averaged Purcell factor associated to each mode calculated after averaging over dipole orientations and emitter positions in the plane located below the antenna and in the middle of the gap.

|        | \(\omega_\alpha'\) [rad/s] | \(\omega_\alpha''\) [rad/s] | \(Q_\alpha\) | \(F_\alpha^{\perp}(r^{\text{max}})\) | \(\langle F_\alpha \rangle\) |
|--------|-----------------------------|-----------------------------|-------------|-----------------------------------|-----------------------------|
| Disk   | \(2.654 \times 10^{15}\)    | \(-4.402 \times 10^{13}\)  | 30.1        | 41.6                             | 6.5                         |
| Antenna A | \(2.748 \times 10^{15}\)    | \(-4.849 \times 10^{13}\)  | 28.3        | 27.4                             | 4.4                         |
| Antenna B | \(2.811 \times 10^{15}\)    | \(-5.188 \times 10^{13}\)  | 27.1        | 22.0                             | 3.4                         |
| Antenna C | \(2.923 \times 10^{15}\)    | \(-5.438 \times 10^{13}\)  | 26.9        | 14.1                             | 2.2                         |
We also measure the lifetime for the nanoantennas A, B and C, which is shown in Fig. S12 with the fitting parameters being displayed in Table S3. The “fast” decay rate enhancement is about 1.43, 1.11 and 1.43 for Antennas A, B and C, respectively and the “slow” decay rate enhancement is 1.86, 1.18 and 1.3 for A, B and C, respectively. Assuming an homogeneous distribution of QD orientations, the average decay rate enhancement is just the average of the “fast” and “slow” enhancements, giving 1.65, 1.15 and 1.37 average decay rate enhancements for Antennas A, B and C, which corresponds to the averaged Purcell factor discussed in the main text. Note that this is the "total" Purcell factor, which is different from the "modal" Purcell factor calculated in Table S2 which is calculated for the antenna mode only.

Figure S12: a. Measured and fit lifetime signal from nanoantennas and reference quantum dots. Grey curve is the instrument response function. Blue curve is freshly spin-coated quantum dots lifetime. Red curve is the same quantum dots after ALD. Dark blue, violet and orange curves are the lifetime signals from Antennas A, B and C, respectively. Grey curve is the instrument response function. PL decay was measured for a narrow spectral band, centered at 650 nm.

We summarized the different calculated and measured enhancement factors in Table S4 and found very good qualitative agreement between experiment and calculations.
Table S3: Amplitude ($a_1, a_2$) and decay ($\tau_1, \tau_2$) components of the bi-exponential fitting used to the experimental lifetime measurements.

| Antenna | $a_1$, a.u. | $\tau_1$, ns | $a_2$, a.u. | $\tau_2$, ns |
|---------|-------------|--------------|-------------|--------------|
| A       | 0.12        | 0.07         | 0.004       | 0.35         |
| B       | 0.12        | 0.09         | 0.004       | 0.55         |
| C       | 0.12        | 0.07         | 0.003       | 0.60         |

Table S4: Comparison between calculated and measured averaged fluorescence enhancement factors. The “theoretical” enhancement factor ($\langle EF_{th}\rangle$) is calculated based on Eq. (2) from the main text, where all the contributing ratios are computed theoretically, except the ratio between the lifetimes $\tau_0/\tau$, which is taken from the experimental measured values. The “experimental” enhancement factor ($\langle EF_{exp}\rangle$) is extracted from the measured photoluminescence based on Eq. (3) from the main text. All the quantities that depend on the excitation wavelength and emission wavelength are taken at $\lambda_{exc} = 570$ nm and $\lambda_{em} \approx 650$ nm, respectively. The theoretical enhancement factor is calculated for a collection NA$_{col} = 0$, while experimentally we integrate the signal over a non-null NA of NA$_{col} = 0.3$.

| Antenna | $\langle \gamma_{exc}/\gamma_{0,exc}\rangle$ | $\langle D_{em}/D_{em}^0\rangle$ | $\langle \gamma_r/\gamma_r^0\rangle$ | $\langle \tau_0/\tau\rangle$ | $\langle EF_{th}\rangle$ | $\langle EF_{exp}\rangle$ |
|---------|---------------------------------|---------------------------------|---------------------------------|------------------------------|--------------------------|--------------------------|
| A       | 7.3                             | 1.31                            | 156.2                           | 1.79                         | 841                      | 654                      |
| B       | 5.0                             | 1.26                            | 107.9                           | 1.17                         | 585                      | 349                      |
| C       | 3.8                             | 1.21                            | 43.9                            | 1.12                         | 183                      | 98                       |
S10 Optical constants of amorphous silicon and gold

We fit our experimental data of the optical constants of our amorphous Si (i.e. refractive index $n$ and extinction coefficient $\kappa$), obtained via ellipsometry measurements, shown in Fig. S13a,b, by a Lorentz model with 1 pole (or oscillator), which is in a quantitative agreement with the experimental data for wavelengths $\lambda > 550$ nm. The complex permittivity $\varepsilon = \varepsilon' + i\varepsilon''$, which is related to the optical constants by the relations $\varepsilon' = n^2 - \kappa^2$ and $\varepsilon'' = 2n\kappa$, that we use in the case of Si reads:

$$
\varepsilon_{\text{Si}}(\omega) = \varepsilon_{\infty,\text{Si}} \left[ 1 - \frac{\omega_{p,\text{Si}}^2}{\omega^2 - \omega_{0,\text{Si}}^2 + i\omega\gamma_{\text{Si}}} \right]
$$

(S5)

with $\varepsilon_{\infty,\text{Si}} = 1$, $\omega_{p,\text{Si}} = 15 \times 10^{15}$ [rad/s], $\omega_{0,\text{Si}} = 5 \times 10^{15}$ [rad/s], and $\gamma_{\text{Si}} = 0.3 \times 10^{15}$ [rad/s].

In the case of the Au substrate, we did not measure the optical constants and assumed that they correspond closely to the values measured by Johnson and Christy. In order to fit these experimental data, we used a Lorentz-Drude model with 2 poles (oscillators), which is also in a quantitative agreement with the experimental data for wavelengths $\lambda > 550$ nm (see Fig. S13c,d). The complex permittivity that we use in the case of Au reads:

$$
\varepsilon_{\text{Au}}(\omega) = \varepsilon_{\infty,\text{Au}} \left[ 1 - \frac{\omega_{p,1,\text{Au}}^2}{\omega^2 + i\omega\gamma_{1,\text{Au}}} - \frac{\omega_{p,2,\text{Au}}^2}{\omega^2 - \omega_{0,2,\text{Au}}^2 + i\omega\gamma_{2,\text{Au}}} \right]
$$

(S6)

with $\varepsilon_{\infty,\text{Au}} = 6$, $\omega_{p,1,\text{Au}} = 5.37 \times 10^{15}$ [rad/s], $\gamma_{1,\text{Au}} = 6.216 \times 10^{13}$ [rad/s], $\omega_{p,2,\text{Au}} = 2.2636 \times 10^{15}$ [rad/s], $\omega_{0,2,\text{Au}} = 4.572 \times 10^{15}$ [rad/s], and $\gamma_{2,\text{Au}} = 1.332 \times 10^{15}$ [rad/s].
Figure S13: Fitting of refractive index $n$ (a) and extinction coefficient $\kappa$ (b) of the amorphous silicon used for the fabrication of the nanoantennas, and of the gold used for the substrate (c and d). The fitting parameters for the analytical curves (red lines) are given in the Methods section in the main text. The experimental data (blue points) were obtained via ellipsometry measurements in the case of silicon, and taken from Ref. S10 in the case of gold. The wavelength of maximum emission of the quantum dots at 650 nm is also shown (vertical black dashed line).
References

(S1) Lim, J.; Jeong, B. G.; Park, M.; Kim, J. K.; Pietryga, J. M.; Park, Y.-S.; Klimov, V. I.; Lee, C.; Lee, D. C.; Bae, W. K. Influence of Shell Thickness on the Performance of Light-Emitting Devices Based on CdSe/Zn1-XCdXS Core/Shell Heterostructured Quantum Dots. Advanced Materials 2014, 26, 8034–8040.

(S2) Dmitriev, P. kitchenknif/lifetime_reconvolution: v0.9. 2022; https://doi.org/10.5281/zenodo.6198822.

(S3) Hoang, T. B.; Akselrod, G. M.; Mikkelsen, M. H. Ultrafast room-temperature single photon emission from quantum dots coupled to plasmonic nanocavities. Nano letters 2016, 16, 270–275.

(S4) Filter, R.; Qi, J.; Rockstuhl, C.; Lederer, F., et al. Circular optical nanoantennas: an analytical theory. Physical Review B 2012, 85, 125429.

(S5) Yang, Y.; Miller, O. D.; Christensen, T.; Joannopoulos, J. D.; Soljacic, M. Low-loss plasmonic dielectric nanoresonators. Nano letters 2017, 17, 3238–3245.

(S6) Ceperley, P. 2016; http://resonanceswavesandfields.blogspot.com/2016/03/all-postings-by.html#eqn43.

(S7) Karalis, A.; Lidorikis, E.; Ibanescu, M.; Joannopoulos, J.; Soljačić, M. Surface-plasmon-assisted guiding of broadband slow and subwavelength light in air. Physical review letters 2005, 95, 063901.

(S8) Sauvan, C.; Hugonin, J. P.; Maksymov, I. S.; Lalanne, P. Theory of the Spontaneous Optical Emission of Nanosize Photonic and Plasmon Resonators. Phys. Rev. Lett. 2013, 110, 237401.

(S9) Lalanne, P.; Yan, W.; Vynck, K.; Sauvan, C.; Hugonin, J.-P. Light Interaction with Photonic and Plasmonic Resonances. Laser & Photonics Reviews 2018, 12, 1700113.
(S10) Johnson, P. B.; Christy, R.-W. Optical constants of the noble metals. *Physical review B* 1972, 6, 4370.