High Purity Single Photon Sources with Near Unity Collection Efficiencies by Deterministic Placement of Quantum Dots in Nanoantennas

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Abstract

Deterministic coupling between photonic nodes in a quantum network is an essential step towards implementing various quantum technologies. The omnidirectionality of free-standing emitters, however, makes this coupling highly inefficient, in particular
if the distant nodes are coupled via low numerical aperture (NA) channels such as optical fibers. This limitation requires placing quantum emitters in nanoantennas that can direct the photons into the channels with very high efficiency. Moreover, to be able to scale such technologies to a large number of channels and nodes, the placing of the emitters should be deterministic. In this work we present a method for placing single quantum emitters with high spatial accuracy at the center of highly directional bullseye metal-dielectric nanoantennas. We further employ non-blinking, high quantum yield quantum dots (QDs) to realize single-photon emission uncompromised by instability and non-radiative exciton recombination processes. This results in a record-high collection efficiency of 85% of the single photons into a low NA of 0.5, enabling efficient coupling between the on-chip, room temperature nanoantenna-emitter and a fiber or a remote free-space node without the need of any additional optics.

The physical realization of the photonic building blocks of future quantum technology and communication applications relies on the development of highly efficient and reliable quantum emitters at photonic nodes. Current state of the art sources are solid state emitters that operate at cryogenic temperatures where the technology is relatively well established. However there are many obstacles facing the scaling of these deployments and their implementation at room-temperature where sources face significantly different challenges. Hence, new approaches are needed to meet the demand for high-efficiency room-temperature sources. These must also address the need for scalability with precision in fabrication.

One impediment however, is the typical isotropic emission exhibited by quantum emitters which limits the practical useful brightness that can be collected into the photonic channels, and thus also limits the deterministic nature of the photon streams. In the past decade there have been considerable efforts to solve this deficiency by modifying the photonic environment near the quantum emitter. To achieve this, emitters were embedded in, or near metallic or dielectric nanostructures. Metallic and plasmonic nanostructures such as metal nanoparticles (MNPs), plasmonic patch antennas, and circular bullseye plasmonic nanoantennas generally have low mode volumes and quality factors enabling emission...
modification over a broadband spectrum. However, these structures also require high spatial accuracy and typically increase the rate of non-radiative recombination. On the other hand, dielectric structures, such as microcavities\textsuperscript{6,7,20–23} and photonic crystals\textsuperscript{24,25} that feature high radiative enhancement factors and low-loss, generally come with a limiting narrow frequency bandwidth, which is usually unsuitable for broadband room-temperature quantum emitters.

Therefore a hybrid metal-dielectric bullseye nanoantenna that combines the advantages of metallic and dielectric antennas but with fewer drawbacks was recently developed.\textsuperscript{26–32} In this structure (see Fig 1c) the photon source placed at the center emits into a dielectric layer that acts as a slab waveguide guiding the light radially outward towards the circular gratings. The parameters of the antenna are tuned in such a way that in the far-field the interference between the various diffracted waves occurs only at low angles thus resulting in a highly directional photon stream. In such a design, the emitter can be located at a large distance from the metal and still produce high directionality in a broad spectral range.

Nevertheless, it is necessary to center the emitter within the bullseye. To date, this has typically been accomplished by uncontrolled distribution of emitters into the nanophotonic structures, which yields a random statistical placement with low probability for single emitters being located at desired locations. Such approaches limit device scaling, and even if a single emitter is deposited, afford low placement accuracy within the target area. The latter impacts the quality of the achieved device, e.g., resulting in non-optimal directivity and limited photon collection into low NA.

Scanning probe techniques have recently been extensively used to place various quantum emitters into different nanostructures.\textsuperscript{29,33–37} Here, we employ a scanning probe technique that allows for directly placing quantum emitters into devices. We use a scalable method, dip-pen nanolithography (DPN), to achieve reliable single-emitter placement. With this method two requirements can be fulfilled: (1) The accurate targeting of small (<500 nm) regions in a predefined three-dimensional substrate; and (2) placing one emitter, rather than multiple, in a desired location, with a significant probability (currently >10\%). To fulfill the
Figure 1: Sketch of hybrid metal-dielectric bullseye structure used in this study and the DPN placement technique. (a) An AFM scan of the area is conducted (the right panel shows an actual image of the scanning of an antenna array), then (b) the tip is wetted and a droplet is placed at the center. After the fabrication process is finalized the resulting structure is shown in (c). (d) Statistics on placement probabilities. (see supplementary info for a more detailed description of the fabrication process)

first, the target region is scanned in its entirety with the AFM tip. This provides an image, effectively a topographical map, of the substrate to guide the writing step (Fig 1a). Then for placement of a single quantum emitter, in this case a non-blinking CdSe/CdS core/thick-shell giant QD (gQD), the tip is wetted with a gQD-solvent suspension (ink) and a droplet is placed in the predetermined location by contacting the AFM tip to the substrate. Several variables of the DPN process affect the area of the deposition spot and the number of nanocrystals that are deposited per spot. These include dwell time, ink-substrate interactions, and ink volume on the tip. In order to assess how these variables affect the probability of placing
Figure 2: (a) Spectrally resolved back focal plane PL image of a final device. The image is normalized with respect to the spectrum, i.e. each line (wavelength) is normalized to the max in that line. (b),(c) and (d) show back focal plane images at $\lambda = 630$, 660, and 690 nm respectively. (e) Simulated collection efficiency at various lateral displacements of the QD from the center of the bullseye antenna for NA = 0.22, 0.5. (f) Green: collection efficiency of same device in (a)-(d) spectrally filtered in the spectral range between 660-680 nm. Yellow: average measured collection efficiency of 9 fabricated single emitter devices, each integrated over the whole QD PL spectral range. Black: FDTD simulation of a reference QD placed on a glass slide. (g) Measured collection efficiency for each device for NA = 0.22 (orange triangles), and 0.5 (blue dots). Solid lines represent mean values and dotted lines are 1 standard deviation from the mean.
a single gQD, as opposed to QD clusters, on this substrate composition, an array of spots was "written" on a flat (featureless) section of the substrate. Out of 20 spots deposited in a single step, 3 spots contained small clusters (2-3 gQDs) and 2 spots contained single QDs (as determined by time-gated $g^{(2)}(0)$), giving the percentages shown in Fig. 1d. By combining optimal tip bleeding (release of excess ink from the AFM tip and upper regions of the cantilever prior to writing), short contact times, and a low concentration of QDs in the carrier solvent, single QDs can currently be placed with a 10% success rate, with small clusters placed 15% of the time. The detailed procedure is described in the Supplementary Information.

We found it necessary to modify the surface of the device to prevent the high-boiling ink solvent from facilitating transport of the placed QD from the surface to the underlying metal layers. Specifically, initial successful attempts at placing single gQDs into the bullseye antenna were thwarted by rapid photobleaching of the emitters. It was found that the polymer used to encapsulate the metal bullseye (serving as the 'lower half' of the dielectric waveguide layer) was softened by the DPN solvent (dichlorobenzene, DCB), causing the gQDs to sink through the dielectric layer toward the metal layer over time, which resulted in quenching of QD emission. This was confirmed in control experiments that compared gQD stability when deposited from DCB either on polymer-coated metal-on-glass or on polymer-coated glass. The nanocrystals placed on the polymer overcoating a Ag film quenched over time, while those placed on the polymer overcoating only glass did not quench (Supplementary Information Fig. S5). For this reason, we utilized a novel room-temperature pulsed gas-phase deposition technique process to deposit thin AlO$_x$ films onto the devices before conducting gQD deposition (see Supplementary Information for experimental details). This step effectively hardened the polymer layer to the DPN solvent, allowing sufficient stability to be maintained for extracting optical characteristics of the devices.

To characterize the overall performance of the emitter-antenna devices we conducted a series of directionality tests by imaging PL of the QDs in the different devices at the
back focal plane of our objective lens (NA=0.9) either on the slit of a spectrometer (with a CCD at the output) or directly onto a CMOS camera. In the first case the result will be a hyperspectral image of the back focal plane displaying the spectral dependence of the angular intensity distribution function which we will denote as $S(\lambda, \theta, \phi)$ where $\lambda$ is the wavelength and $\theta$ and $\phi$ are the polar and azimuthal angles respectively. Fig 2a is an image for $S(\lambda, \theta, \pi/2)$ of a typical device, which shows the broadband resonance of the antenna at between 660-680 nm. Moreover most of the QD emission over the entire 80 nm range shown in this image is within 10° from the normal emphasizing the highly broadband operation of these antennas. This is highlighted in the spectral sections in Fig 2b ($S(630nm, \theta, \phi)$), Fig 2c ($S(660nm, \theta, \phi)$), and Fig 2d ($S(690nm, \theta, \phi)$).

Having shown the broadband nature of our antenna we move on to analyse the performance over the spectrally integrated angular intensity distribution $I(\theta, \phi) = \int S(\lambda, \theta, \phi) d\lambda$. By integrating over all angles within a collection cone of a given NA one can calculate the ratio of photons collected; denoted as the collection efficiency:

$$\eta = \frac{\int_0^{2\pi} d\phi \int_0^{\theta_{NA}} d\theta \sin(\theta) I(\theta, \phi)}{\int_0^{2\pi} d\phi \int_0^{\pi} d\theta \sin(\theta) I(\theta, \phi)}$$

(1)

To demonstrate the reliability of our method we display (Fig. 2f & g) the measured collection efficiency for 9 different emitter-antenna devices (4 of which contained single QDs) made under identical conditions compared to an emitter in free space. The results show significant directionality enhancement (more details below) and good reproducibility among the devices. Furthermore to emphasize the importance of our high precision positioning on the performance of the devices we compare in Fig. 2e the collection efficiency of our devices with FDTD simulations of an emitter displaced laterally from the center. A significant degradation of performance is observed for even sub-micron displacements.

Fig. 3 displays the results for an antenna with a single QD placed in the center (Fig. 3a, b shows the spatial images of the device and of the PL distribution). We measure photon
Figure 3: Results from a single QD coupled to a bullseye nanoantenna. (a) Microscope image and (b) Spatial PL map of a single photon device. (c) Schematic displaying the time-gated filtering technique: the black pulses marks the laser excitation pulses, and the red and green lines mark the statistical temporal distribution of the bi-exciton and exciton emission respectively. Counts from \( t = 0 \) to \( t = T_F \) are post-rejected after the measurement (see supplementary infor for more details on the measurement scheme). (d) \( g^{(2)}(\tau) \) for three values of \( T_F = 0, 45, \) and 90 ns. (e) Brightness, and \( g^{(2)}_0 \) as a function of the time-gate delay \( (T_F) \) for a collection NA of 0.9 (solid lines) and 0.5 (dashed lines) compared to a similar QD placed on glass (black lines). (f) Brightness as a function of single photon purity. The inset represents the enhancement of the rate of a QD in our device compared to a similar QD on glass for an NA = 0.5 and 0.9.

brightness levels of up to 0.76 photons/pulse. This corresponds to a photon rate of 380 kHz (330 kHz) at an optical pump rate of 500 kHz or a maximal photon rate of \( \sim 11.5 \text{ MHz} \) (10.1 MHz) at the CW limit for a collection NA of 0.9 (0.5). Accordingly the bi-exciton and exciton quantum yields are 21% and 55% respectively (see Supplementary Information). With no temporal filtering the value of \( g^{(2)}(0) = 0.68 \). This high value of \( g^{(2)}(0) \) is a result of the measurements being mainly done near saturation where both biexciton and plasmonic emission play a role (see Supplementary Information form more details). Using
the time-filtered gating technique\cite{38,41,42} we confirm that the emitter is indeed a single QD by measuring the second order coherence starting from various times ($T_F$) after the excitation pulse. This helps in differentiating between the emission of bi-excitons (which have shorter lifetime) and the emission of multiple QDs.\cite{38} Using this technique the value of $g^{(2)}(0)$ quickly drops below 0.5 after only 2 ns of temporal filtering and continues falling as $T_F$ increases resulting in high single photon purity for larger filtering times reaching $g^{(2)}(0) < 0.01$ with 0.12 photons per pulse at $T_F = 90$ ns. Although this increase in single photon purity comes at the cost of brightness, one might tune this brightness-purity trade-off to optimize the specific application of interest (Fig.3f). Furthermore we recently proposed a new heralded scheme to overcome this limitation\cite{42} where we propose two methods (in addition to the time gated method used here) which utilize the difference in lifetime between the biexciton and exciton states and/or the cascaded nature of the emission to separate the resulting two photon emission in time. Critically we have shown that the use of these methods lead to better efficiencies depending on the original biexciton and exciton quantum efficiencies.

Despite this tradeoff (which is characteristic of most room-temperature sources) we emphasize that the benefit of our device over a similar QD on glass is maintained. To show this we compare in Fig. 3e,f between the rate of a QD in our device and on glass. We also display this enhancement of the rate for different $g^{(2)}(0)$ in the inset of Fig. 3f where a factor of 10 improvement in rate is displayed over a QD on glass even for the highest single photon purities.

The reasons behind this high relative photon fluxes even for low NAs can be determined by inspecting the back focal plane image from such a device. As can be seen in Fig. 4a,b, the emission displays strong directional behaviour as most of the emission is to low angles. This results in very high collection efficiencies into low NA (Fig. 4c). We compare our results to detailed FDTD simulations\cite{40} taking into account the broadband nature and random dipole orientation of the QDs and find good agreement between simulation and experiments, as is shown in Fig. 4d (for more details on the simulations refer to\cite{28}). Furthermore, we also
Figure 4: Results from a single QD coupled to a bullseye nanoantenna. (a) and (b) Back focal plane image of single photon device. (c) Collection efficiency of device compared to simulation measurements and various state-of-the-art platforms: Micropillar cavities,\textsuperscript{54,43,44} Tapered Nanowire,\textsuperscript{45} Photonic crystal cavities,\textsuperscript{46} Bullseye nanoantenna.\textsuperscript{26,31,32}
present in Fig. 4: a comparison with other state-of-the-art emitter-antenna platforms, including low temperature platforms. It is clear that the collection efficiencies measured in our devices are superior compared to various other platforms such as micropillar cavities, tapered nanowires, photonic crystal cavities, and bullseye antennas. Notably, most of these devices operate at cryogenic temperatures with cryogenic sources, where the emission lines are all much narrower. In contrast our sources (and most room-temperature sources) have a much broader spectrum (30-40 nm FWHM in our case see the supplementary information). Despite this, the design of our structure enables us to collect the photons efficiently throughout this spectral range (see Fig. 2 a-e). This is a key achievement for all room temperature sources.

While most of the platforms shown in Fig. 4 are focused on higher NAs, our devices are especially suited for collection into lower NAs such as is needed for direct coupling into optical fibers or for free space transmission to remote nodes. For example the number of collected photons into an NA of 0.12 (0.22), corresponding to the NA of a single (multi) mode fiber, is enhanced by a factor of 6 (5) compared to an emitter in free space. This is also highlighted in Fig. 3a in the small difference in the collected brightness into an NA of 0.5 and 0.9.

We also compare our current work with a previous work from our group on bullseye nanoantennas. While in the previous work a maximum of 37% collection efficiency was achieved at an NA of 0.65, in our current work we display a 90% collection efficiency into the same NA. This significant improvement was achieved due to multiple improvements including: (a) detailed optimization of the structure based on 3D FDTD simulations; (b) a new template stripping method used for the fabrication on the metal part of the antennas, yielding a much higher quality antennas (see supplementary information); (c) the novel placement method described above yielding a much better emitter positioning; and (d) a higher signal to noise ratio and better loss estimation methods due to the combined use of brighter and non-blinking QDs and a higher NA objective.
In summary we have put forward a method for placing single QDs onto predefined substrates with high spatial accuracy. By using this method with metal-dielectric hybrid bullseye antennas we were able to reproducibly show record-high directionalities of single photons at room temperature compared to other state-of-the-art platforms, with collection efficiencies exceeding 80% for NA=0.5 and brightness levels up to 0.76 photons/pulse, and high single photon purities using temporal post-selection. This approach is promising for both fiber-based and free space quantum communication applications.

Acknowledgements

This work was performed in part at the Center for Integrated Nanotechnologies (CINT), a Nanoscale Science Research Center and User Facility operated for the U.S. Department of Energy (DOE) Office of Science. AB and JW were funded by CINT. All other Los Alamos National Laboratory (LANL) authors were funded by the Laboratory Directed Research and Development (LDRD) program, grant 20170001DR. LANL, an affirmative action equal opportunity employer, is operated by Los Alamos National Security, LLC, for the National Nuclear Security Administration of the U.S. Department of Energy under contract DEAC52-06NA25396. R. B. and A.V.M. were supported by U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. de-sc0010697

Supporting Information Available

The following files are available free of charge.

The supplementary information contains details about the synthesis and fabrication in addition to experimental details such as the measurement scheme, photon rate calculation, and collection efficiency calculations.
References

(1) Lounis, B.; Orrit, M. Single-photon sources. *Reports on Progress in Physics* **2005**, *68*, 1129–1179.

(2) Aharonovich, I.; Englund, D.; Toth, M. Solid-state single-photon emitters. *Nature Photonics* **2016**, *10*, 631–641.

(3) Rodt, S.; Reitzenstein, S.; Heindel, T. Deterministically fabricated solid-state quantum-light sources. *Journal of Physics: Condensed Matter* **2020**, *32*, 153003.

(4) Laucht, A.; Pütz, S.; Günthner, T.; Hauke, N.; Saive, R.; Frédérick, S.; Bichler, M.; Amann, M.-C.; Holleitner, A. W.; Kaniber, M.; Finley, J. J. A Waveguide-Coupled On-Chip Single-Photon Source. *Physical Review X* **2012**, *2*, 011014.

(5) Gazzano, O.; Michaelis de Vasconcellos, S.; Arnold, C.; Nowak, A.; Galopin, E.; Sagnes, I.; Lanco, L.; Lemaître, A.; Senellart, P. Bright solid-state sources of indistinguishable single photons. *Nature Communications* **2013**, *4*, 1425.

(6) Ding, X.; He, Y.; Duan, Z.-C.; Gregersen, N.; Chen, M.-C.; Unslieber, S.; Maier, S.; Schneider, C.; Kamp, M.; Höfling, S.; Lu, C.-Y.; Pan, J.-W. On-Demand Single Photons with High Extraction Efficiency and Near-Unity Indistinguishability from a Resonantly Driven Quantum Dot in a Micropillar. *Physical Review Letters* **2016**, *116*, 020401.

(7) Somaschi, N. et al. Near optimal single photon sources in the solid state. *Nat. Photon.* **2016**, *10*, 340–345.

(8) Liu, F.; Brash, A. J.; O’Hara, J.; Martins, L. M. P. P.; Phillips, C. L.; Coles, R. J.; Royall, B.; Clarke, E.; Bentham, C.; Prtljaga, N.; Itskevich, I. E.; Wilson, L. R.; Skolnick, M. S.; Fox, A. M. High Purcell factor generation of coherent on-chip single photons. 2017,
(9) Wang, H. et al. Towards optimal single-photon sources from polarized microcavities. *Nature Photonics* **2019**, *1*–*6*.

(10) Yuan, C. T.; Yu, P.; Ko, H. C.; Huang, J.; Tang, J. Antibunching Single-Photon Emission and Blinking Suppression of CdSe/ZnS Quantum Dots. *ACS Nano* **2009**, *3*, *3051–3056*.

(11) Menagen, G.; Macdonald, J. E.; Shemesh, Y.; Popov, I.; Banin, U. Au Growth on Semiconductor Nanorods: Photoinduced versus Thermal Growth Mechanisms. *Journal of the American Chemical Society* **2009**, *131*, *17406–17411*.

(12) Ma, X.; Tan, H.; Kipp, T.; Mews, A. Fluorescence Enhancement, Blinking Suppression, and Gray States of Individual Semiconductor Nanocrystals Close to Gold Nanoparticles. *Nano Letters* **2010**, *10*, *4166–4174*.

(13) Ji, B.; Giovanelli, E.; Habert, B.; Spinicelli, P.; Nasilowski, M.; Xu, X.; Lequeux, N.; Hugonin, J.-P.; Marquier, F.; Greffet, J.-J.; Dubertret, B. Non-blinking quantum dot with a plasmonic nanoshell resonator. *Nature Nanotechnology* **2015**, *10*, *170–175*.

(14) Belacel, C.; Habert, B.; Bigourdan, F.; Marquier, F.; Hugonin, J.-P.; Michaelis de Vasconcellos, S.; Lafosse, X.; Coolen, L.; Schwob, C.; Javaux, C.; Dubertret, B.; Greffet, J.-J.; Senellart, P.; Maître, A. Controlling Spontaneous Emission with Plasmonic Optical Patch Antennas. *Nano Letters* **2013**, *13*, *1516–1521*.

(15) 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*.

(16) Dhawan, A. R.; Belacel, C.; Esparza-Villa, J. U.; Nasilowski, M.; Wang, Z.; Schwob, C.; Hugonin, J.-P.; Coolen, L.; Dubertret, B.; Senellart, P.; Maître, A. Extreme multiexciton emission from deterministically assembled single-emitter subwavelength plasmonic patch antennas. *Light: Science & Applications* **2020**, *9*, *33*.
(17) Yi, J.-M.; Cuche, A.; Devaux, E.; Genet, C.; Ebbesen, T. W. Beaming Visible Light with a Plasmonic Aperture Antenna. *ACS Photonics* **2014**, *1*, 365–370.

(18) Harats, M. G.; Livneh, N.; Zaiats, G.; Yochelis, S.; Paltiel, Y.; Lifshitz, E.; Rapaport, R. Full Spectral and Angular Characterization of Highly Directional Emission from Nanocrystal Quantum Dots Positioned on Circular Plasmonic Lenses. *Nano Letters* **2014**, *14*, 5766–5771.

(19) Andersen, S. K. H.; 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.

(20) Skolnick, M. S.; Fisher, T. A.; Whittaker, D. M. Strong coupling phenomena in quantum microcavity structures. *Semiconductor Science and Technology* **1998**, *13*, 645–669.

(21) Reithmaier, J. P.; Sęk, G.; Löffler, A.; Hofmann, C.; Kuhn, S.; Reitzenstein, S.; Keldysh, L. V.; Kulakovskii, V. D.; Reinecke, T. L.; Forchel, A. Strong coupling in a single quantum dotsemiconductor microcavity system. *Nature* **2004**, *432*, 197–200.

(22) Press, D.; Götzinger, S.; Reitzenstein, S.; Hofmann, C.; Löffler, A.; Kamp, M.; Forchel, A.; Yamamoto, Y. Photon Antibunching from a Single Quantum-Dot-Microcavity System in the Strong Coupling Regime. *Physical Review Letters* **2007**, *98*, 117402.

(23) Reitzenstein, S.; Hofmann, C.; Gorbunov, A.; Strauß, M.; Kwon, S. H.; Schneider, C.; Löffler, A.; Höfling, S.; Kamp, M.; Forchel, A. AlAs-GaAs micropillar cavities with quality factors exceeding 150000. *Applied Physics Letters* **2007**, *90*, 251109.

(24) Kaniber, M.; Laucht, A.; Neumann, A.; Villas-Bôas, J. M.; Bichler, M.; Amann, M.-C.; Finley, J. J. Investigation of the nonresonant dot-cavity coupling in two-dimensional photonic crystal nanocavities. *Physical Review B* **2008**, *77*, 161303.
(25) Liu, F.; Brash, A. J.; OHara, J.; Martins, L. M. P. P.; Phillips, C. L.; Coles, R. J.; Royall, B.; Clarke, E.; Bentham, C.; Prtljaga, N.; Itskevich, I. E.; Wilson, L. R.; Skolnick, M. S.; Fox, A. M. High Purcell factor generation of indistinguishable on-chip single photons. *Nature Nanotechnology* 2018, 13, 835–840.

(26) 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.

(27) Harats, M. G.; Livneh, N.; Rapaport, R. Design, fabrication and characterization of a hybrid metal-dielectric nanoantenna with a single nanocrystal for directional single photon emission. *Optical Materials Express* 2017, 7, 834.

(28) Abudayyeh, H.; Rapaport, R. Quantum emitters coupled to circular nanoantennas for high brightness quantum light sources. *Quantum Science and Technology* 2017,

(29) Nikolay, N.; Sadzak, N.; Dohms, A.; Lubotzky, B.; Abudayyeh, H.; Rapaport, R.; Benson, O. Accurate placement of single nanoparticles on opaque conductive structures. *Applied Physics Letters* 2018, 113, 113107.

(30) Rickert, L.; Kupko, T.; Rodt, S.; Reitzenstein, S.; Heindel, T. Optimized designs for telecom-wavelength quantum light sources based on hybrid circular Bragg gratings. *Optics Express* 2019, 27, 36824.

(31) Liu, J.; Su, R.; Wei, Y.; Yao, B.; Silva, S. F. C. d.; Yu, Y.; Iles-Smith, J.; Srinivasan, K.; Rastelli, A.; Li, J.; Wang, X. A solid-state source of strongly entangled photon pairs with high brightness and indistinguishability. *Nature Nanotechnology* 2019, 14, 586–593.

(32) Wang, H. et al. On-Demand Semiconductor Source of Entangled Photons Which Simultaneously Has High Fidelity, Efficiency, and Indistinguishability. *Physical Review Letters* 2019, 122, 113602.
(33) Dawood, F.; Wang, J.; Schulze, P. A.; Sheehan, C. J.; Buck, M. R.; Dennis, A. M.; Majumder, S.; Krishnamurthy, S.; Ticknor, M.; Staude, I.; Brener, I.; Goodwin, P. M.; Amro, N. A.; Hollingsworth, J. A. The Role of Liquid Ink Transport in the Direct Placement of Quantum Dot Emitters onto Sub-Micrometer Antennas by Dip-Pen Nanolithography. *Small* 2018.

(34) 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.

(35) Böhm, F.; Nikolay, N.; Pyrlik, C.; Schlegel, J.; Thies, A.; Wicht, A.; Tränkle, G.; Benson, O. On-chip integration of single solid-state quantum emitters with a SiO2 photonic platform. *New Journal of Physics* 2019, 21, 045007.

(36) Zadeh, I. E.; Elshaari, A. W.; Jöns, K. D.; Fognini, A.; Dalacu, D.; Poole, P. J.; Reimer, M. E.; Zwiller, V. Deterministic Integration of Single Photon Sources in Silicon Based Photonic Circuits. *Nano Letters* 2016, 16, 2289–2294.

(37) Kim, J.-H.; Aghaeimeibodi, S.; Richardson, C. J. K.; Leavitt, R. P.; Englund, D.; Waks, E. Hybrid Integration of Solid-State Quantum Emitters on a Silicon Photonic Chip. *Nano Letters* 2017, 17, 7394–7400.

(38) Mangum, B. D.; Ghosh, Y.; Hollingsworth, J. A.; Htoon, H. Disentangling the effects of clustering and multi-exciton emission in second-order photon correlation experiments. *Optics express* 2013, 21, 7419–26.

(39) Bose, R.; Dangerfield, A.; Rupich, S. M.; Guo, T.; Zheng, Y.; Kwon, S.; Kim, M. J.; Gartstein, Y. N.; Esteve, A.; Chabal, Y. J.; Malko, A. V. Engineering Multilayered Nanocrystal Solids with Enhanced Optical Properties Using Metal Oxides for Photonic Applications. *ACS Applied Nano Materials* 2018, 1, 6782–6789.

(40) Lumerical Solutions, Inc. [http://www.lumerical.com/tcad-products/fdtd/]
(41) Feng, S.-W.; Cheng, C.-Y.; Wei, C.-Y.; Yang, J.-H.; Chen, Y.-R.; Chuang, Y.-W.; Fan, Y.-H.; Chuu, C.-S. Purification of Single Photons from Room-Temperature Quantum Dots. Physical Review Letters 2017, 119, 143601.

(42) Abudayyeh, H.; Lubotzky, B.; Majumder, S.; Hollingsworth, J. A.; Rapaport, R. Purification of Single Photons by Temporal Heralding of Quantum Dot Sources. ACS Photonics 2019, 6, 446–452.

(43) Fischbach, S.; Kaganskiy, A.; Tauscher, E. B. Y.; Gericke, F.; Thoma, A.; Schmidt, R.; Strittmatter, A.; Heindel, T.; Rodt, S.; Reitzenstein, S. Efficient single-photon source based on a deterministically fabricated single quantum dot - microstructure with backside gold mirror. Applied Physics Letters 2017, 111, 011106.

(44) Liu, S.; Wei, Y.; Su, R.; Su, R.; Ma, B.; Chen, Z.; Ni, H.; Niu, Z.; Yu, Y.; Wei, Y.; Wang, X.; Yu, S. A deterministic quantum dot micropillar single photon source with 65% extraction efficiency based on fluorescence imaging method. Scientific Reports 2017, 7, 13986.

(45) Claudon, J.; Bleuse, J.; Malik, N. S.; Bazin, M.; Jaffrenrou, P.; Gregersen, N.; Sauvan, C.; Lalanne, P.; Gérard, J.-M. A highly efficient single-photon source based on a quantum dot in a photonic nanowire. Nature Photonics 2010, 4, 174–177.

(46) Madsen, K. H.; Ates, S.; Liu, J.; Javadi, A.; Albrecht, S. M.; Yeo, I.; Stobbe, S.; Lodahl, P. Efficient out-coupling of high-purity single photons from a coherent quantum dot in a photonic-crystal cavity. Physical Review B 2014, 90, 155303.

(47) Orfield, N. J.; Majumder, S.; McBride, J. R.; Yik-Ching Koh, F.; Singh, A.; Bouquin, S. J.; Casson, J. L.; Johnson, A. D.; Sun, L.; Li, X.; Shih, C.-K.; Rosenthal, S. J.; Hollingsworth, J. A.; Htoon, H. Photophysics of Thermally-Assisted Photobleaching in Giant Quantum Dots Revealed in Single Nanocrystals. ACS Nano 2018, 12, 4206–4217.
(48) Chen, O.; Zhao, J.; Chauhan, V. P.; Cui, J.; Wong, C.; Harris, D. K.; Wei, H.; Han, H. S.; Fukumura, D.; Jain, R. K.; Bawendi, M. G. Compact high-quality CdSe-CdS core-shell nanocrystals with narrow emission linewidths and suppressed blinking. *Nature Materials* 2013.