## Collecting and Manipulating Single Photons with Near-Unity Efficiency

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**Abstract** In this tutorial I will first review various approaches which have been employed to efficiently collect single photons from a single emitter. In the second part I will discuss experiments where a single molecule provides an optical nonlinearity strong enough to manipulate single photons.

Single-photon sources will play a major role in quantum technologies<sup>[1],[2]</sup>. emerging Applications range from quantum communication and computation to quantum imaging, sensing and novel intensity standards for metrology. The defining feature of such a source is that it emits exactly one photon after being triggered. A simple attenuated laser can unfortunately not fulfil this requirement. The reason is that the underlying photon statistics of a laser is inherently Poissonian. A single quantum system, like a single atom or molecule, on the other hand can naturally only emit one photon at a time on a given transition. Single atoms, molecules, quantum dots or defect centers are therefore primary candidates for the realization of true single-photon sources. During the last twenty years researchers have demonstrated numerous single-emitter based single-photon sources in a variety of material systems. Despite all the progress today's single-photon sources are still not mature enough to be used in real world applications.

Although the ability to emit exactly one photon at a time is the defining property of a single-photon source, it is not the only property that has to be considered. Efficiency, photon indistinguishability and brightness might be equally important. Most of the so far demonstrated single-photon sources are non-ideal in the sense that they have a nonnegligible probability for multi-photon emission. This means that in contrast to the simplistic picture of an isolated two-level quantum system which emits exactly one photon upon excitation, there are sometimes two or even more photons emitted. The purity of the source is therefore spoiled to a certain degree. A measurement of the second-order correlation function  $g^2(\tau)$  at time zero provides a direct measure of the purity of a source. Deviations from zero indicate multiphoton emission due to unavoidable background fluorescence, residual laser light or even other single emitters that contribute to the collected signal. This correlation function can distinguish a single-photon source from other types of sources. It is, however, insensitive to photon loss. This means that a measurement of  $g^2(0)$  cannot distinguish between a perfect single-photon source and an inefficient source which is most of the time dark.

While it is obvious that one ideally wants to have a source, which excels in every aspect, it is also clear that this is not necessary. Realistically a single-photon source does not need to be perfect in every aspect to be useful in a certain application. While photon indistinguishability is for example important for quantum computation schemes it might be completely irrelevant for imaging, metrology quantum and communication. Efficiency, defined as the probability to have a photon at disposal after an excitation certainly helps in any application. In some cases a certain efficiency is even required for example to ensure scalability in quantum computation schemes<sup>[3]</sup> or for some types of quantum imaging. Therefore, researchers have put a lot of effort in devising devices which improve the photon collection efficiency from a single emitter. A quick overview of the most common currently used photon collection methodologies and their performance can be found in Reference [4].

A prominent approach employs optical microcavities. These resonators feature high quality factors and low mode volumes<sup>[5]</sup>. The spontaneous emission rate into the cavity mode is increased due to the Purcell-effect, which in turn leads to a directional emission into the cavity mode. Photons are then collected via a suitable out-coupler. Single emitters coupled to microcavities have come a long way. Micropillars

and more recently in-plane structures such as bull's eye resonators have shown remarkable photon collection efficiencies of 80% and 85%, respectively<sup>[6,7,8]</sup>. Other photonic structures which have been investigated include parabolic mirrors<sup>[9]</sup>, tapered nanowires<sup>[10]</sup> and plasmonic antennas<sup>[11]</sup>. However, the largest collection efficiencies have so far been demonstrated with planar antenna structures<sup>[12,13]</sup>. With collection efficiencies close to 100% it is for example possible to demonstrate a sub-shot noise single emitter quantum light source which can be used illumination source in quantum an as imaging<sup>[14,15]</sup>. A drawback of the planar antenna structures so far has been their non-Gaussian radiation pattern, which prevents for example an efficient coupling of the collected photons into a single mode fiber. The concept of the planar antenna has therefore been recently extended. The main idea is to create an omnidirectional photonic bandgap by a lateral structuring to inhibit unwanted large-angle emission and to small-angle defect-guided-mode enhance emission.



Fig.1:(a) Proposed omnidirectional reflector for collecting single photons with 95% efficiency into a fundamental Gaussian mode. The structure consists essentially of a hollow circular dielectric grating made out of TiO<sub>2</sub> (orange) and SiO<sub>2</sub> (purple) placed on a silver mirror. The main purpose of the cubic zirconia solid immersion lens (SIL) on top is to keep the maximum emission angle below 40°. Note that the red arrow indicates the dipole orientation of a single quantum emitter in the center of the structure. Parameters R=590nm, h=280nm and d=180nm have been optimized using numerical simulations, relevant refractive indices  $n_0=1$ ,  $n_1=2,58$ ,  $n_2=1.5$ , important dimensions D=100nm, t<sub>1</sub>=55nm, n₃=2.15. t<sub>2</sub>=110nm. (b) Angular emission density and angle-dependent collection efficiency. The inset is displaying the 2D angular emission pattern.

This truncated metallo-dielectric omnidirectional reflector can be realized for various experimental conditions and promises again near-unity collecting efficiency while having a highly Gaussian emission profile<sup>[4]</sup>. Figure 1 displays such an antenna with a solid immersion lens as an integral part of the design. Note the low divergence angle of less than 40°.



Fig.2: A single molecule coupled to an open Fabry-Perot microcavity blocks the transmission of a weak laser beam when the laser is on resonance with the optical transition of the molecule (magenta). The black symbols where taken at a detuning of 20 GHz with respect to the cavity resonance and act as a reference. Inset: schematics of the experiment, SM: single molecule.

Given the fact that one can nowadays efficiently extract single photons from a single emitter it should, simply by the argument of reciprocity, also be possible to observe interactions of single photons with a single emitter. If the interaction is strong enough it can in fact be exploited to manipulate single photons<sup>[16]</sup>. Such a scenario actually describes the ultimate limit of light-matter interaction, which is not only interesting from a fundamental perspective. On the contrary interesting applications arise if one could steer one or two photons with the help of a single emitter. Such interactions are usually based on the optical nonlinearity provided by the quantum emitter<sup>[17]</sup>. Note that this nonlinearity is not necessarily small, because the underlying interaction mechanism is related to the cross section of the quantum emitter, which can actually be quite large. One can increase the interaction strength further by employing the same structures used for increasing the photon collection efficiency. Microcavities are again a prime example. Figure 2 shows how the transmission of a weak laser beam through a Fabry-Perot microcavity drops when the laser is on resonance (0 GHz detuning) with a single molecule<sup>[18]</sup>. This can be interpreted as follows: while an empty Fabry-Perot would transmit all the light on resonance, the single molecule-cavitysvstem reflects the incoming photons. Experiments with single photons emitted by another molecule show identical results<sup>[18]</sup>. These types of experiments demonstrate that one can indeed manipulate single photons with a single quantum emitter. The situation gets even more interesting if one could put two or three photons with certain frequency detunings into the cavity. Then one could observe nonlinear effects, like three-photon amplification and four-wave mixing at the level of individual photons<sup>[19]</sup>. Particularly exciting is the perspective to build a device where a single photon can switch another photon.

In recent years we have seen the demonstration of several basic building blocks essential for a technology, be it classical or quantum, based on single or few photons and single quantum systems as active elements. Now it is time to put these individual components together, possibly on an integrated chip and start to build small networks.

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## References

- B. Lounis *et al.*, Single-photon sources, Rep. Prog. Phys. **68**, 1129–1179 (2005).
- [2] S. Scheel, Single-photon sources-an introduction. J. Mod. Opt. 56, 141–160 (2009).
- [3] M. Varnava *et al.*, How Good Must Single Photon Sources and Detectors Be for Efficient Linear Optical Quantum Computation?, Phys. Rev. Lett. **100**, 060502 (2008).
- [4] W. Lie *et al.*, Truncated Metallo-Dielectric Omnidirectional Reflector: Collecting Single Photons in the Fundamental Gaussian Mode with 95% Efficiency, ACS Photonics **7**, 2474–2481 (2020).
- [5] K.J. Vahala, Optical microcavities, Nature 424, 840-846 (2003).
- [6] X. Ding *et al.*, On-Demand Single Photons with High Extraction Efficiency and Near-Unity Indistinguishability from a Resonantly Driven Quantum Dot in a Micropillar, Phys. Rev. Lett. 116, 020401 (2016).
- [7] O. Gazzano *et al.*, Bright solid-state sources of indistinguishable single photons, Nat. Commun. 4, 1425 (2013).
- [8] J. Liu *et al.*, A solid-state source of strongly entangled photon pairs with high brightness and indistinguishability. Nat. Nanotechnol. **14**, 586–593 (2019).

- S. Morozov *et al.*, Metal–Dielectric Parabolic Antenna for Directing Single Photons, Nano Lett. **18**, 3060– 3065 (2018).
- [10] J. Claudon *et al.*, A highly efficient single-photon source based on a quantum dot in a photonic nanowire, Nat. Photonics 4, 174–177(2010).
- [11] A.G. Curto *et al.*, Unidirectional Emission of a Quantum Dot Coupled to a Nanoantenna, Science **329**, 930-933 (2010).
- [12] K.G. Lee *et al.*, A planar dielectric antenna for directional single-photon emission and near-unity collection efficiency, Nat. Photonics **5**, 166–169 (2011).
- [13] X.-W. Chen *et al.*, 99% efficiency in collecting photons from a single emitter, Opt. Lett. **36**, 3545–3547 (2011).
- [14] X.-L. Chu *et al.*, Experimental realization of an optical antenna designed for collecting 99% of photons from a quantum emitter, Optica 1, 203 (2014).
- [15] X.-L. Chu *et al.*, A single molecule as a high-fidelity photon gun for producing intensity-squeezed light, Nat. Photonics **11**, 58–62 (2017).
- [16] Y. L. A. Rezus *et al.*, Single-Photon Spectroscopy of a Single Molecule, Phys. Rev. Lett. **108**, 093601 (2012).
- [17] D.E. Chang *et al.*, Quantum nonlinear optics photon by photon, Nature Photonics 8, 685–694(2014).
- [18] D. Wang *et al.*, Turning a molecule into a coherent twolevel quantum system, Nature Physics **15**, 483 (2019).
- [19] A. Maser *et al.*, Few-photon coherent nonlinear optics with a single molecule, Nature Photonics **10**, 450-454 (2016).