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Perspective

Single-Photon Generation Engineering

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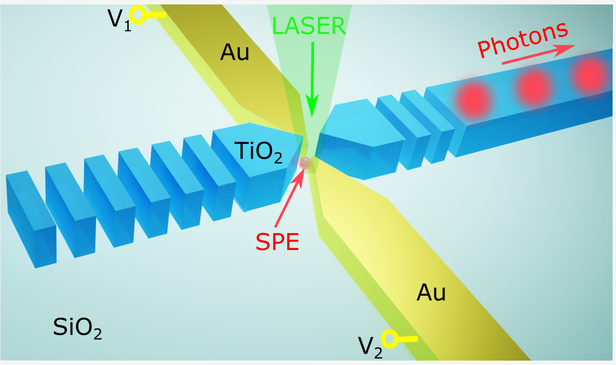

ABSTRACT: Single-photon generation by quantum emitters of specially designed nanostructures can be strongly influenced by the immediate environment, resulting in dramatically enhanced emission rates, along with funneling emission into a well-defined (in terms of propagation direction and polarization state) radiation channel. Single-photon generation engineering thus compels one to search for the configuration incorporating a highly resonant nanocavity coupled to a quantum emitter and to a directional antenna or an optical waveguide. Significant progress has been achieved in understanding physical mechanisms involved in single-photon generation, and impressive experimental demonstrations of ultrabright, polarized, and directional single-photon sources have been reported. At the same time, targeting highly desirable room-temperature generation of indistinguishable single photons, especially when efficiently funneled into single-mode optical waveguides, poses many serious challenges, which are discussed in this Perspective, along with possible ways to tackle these challenges.

KEYWORDS: *quantum plasmonics, quantum optics, plasmonic cavity, dielectric cavity, single-photon emitter*

Single-photon emitters (SPEs) constitute important building blocks for emerging photon-based quantum information and communication technologies.[1−3] Especially solid-state SPEs are considered apposite for scalable photonic quantum technologies.[4] In general, stand-alone SPEs have many shortcomings that prohibit their direct use in quantum networks. The main SPE limitations are low emission rates (due to intrinsic radiative lifetimes of the order of 10 ns), emission in practically all directions (due to its electric dipole nature) and not into a particular optical mode, and decoherence processes (due to phonon-mediated interactions) leading to spectral broadening of emission spectra. The SPE spontaneous emission can significantly be sped up by increasing the local density of photon states at the SPE location and frequency with a suitably designed optical cavity.[5] Combining optical cavities with antenna configurations ensuring efficient directional emission many of the above shortcomings can be tackled, making SPEs suitable for photonic quantum information processing.[6] Note that the antenna choice is determined not only by the requirement of directional photon emission but also the necessity to ensure a sufficiently fast radiation rate commensurate to the enhanced relaxation rate.[7,8] In this Perspective, we first briefly look at the cavity structures that have been used to improve the SPE properties. Then, we consider the directions along which cavities can be developed to further the emission characteristics. Probably the most enticing perspective in accelerating the photon emission rate is to reduce the emission time below the dephasing time even at room temperatures, opening thereby a doorway toward room-temperature photonic quantum networks.[9] The challenges on the way and possible ways to tackle those challenges are discussed.

There are two classes of configurations that can be exploited for designing optical cavities: photonic configurations using (primarily) dielectric materials and plasmonic configurations based on differently shaped metal−dielectric interfaces. Dielectric cavities, having practically no absorption, can dramatically enhance the spontaneous emission rate by employing a high ratio between the cavity quality factor and its volume (i.e., the Purcell enhancement) although their volumes are diffraction limited.[5] SPEs such as defect centers in diamonds and quantum dots have been coupled to dielectric cavities to enhance their emission properties. For example, color centers in diamond thin films were efficiently coupled to fiber microcavities.[10−15] These microcavities are flexible, exhibit high quality factors, and the emission can conveniently be collected in an optical fiber (Figure 1a). Other advantages of such cavities include easy alignment of emitters to the cavity mode and tunability for matching the emission wavelength of SPEs. Such a cavity has been utilized for converting an organic molecule into a

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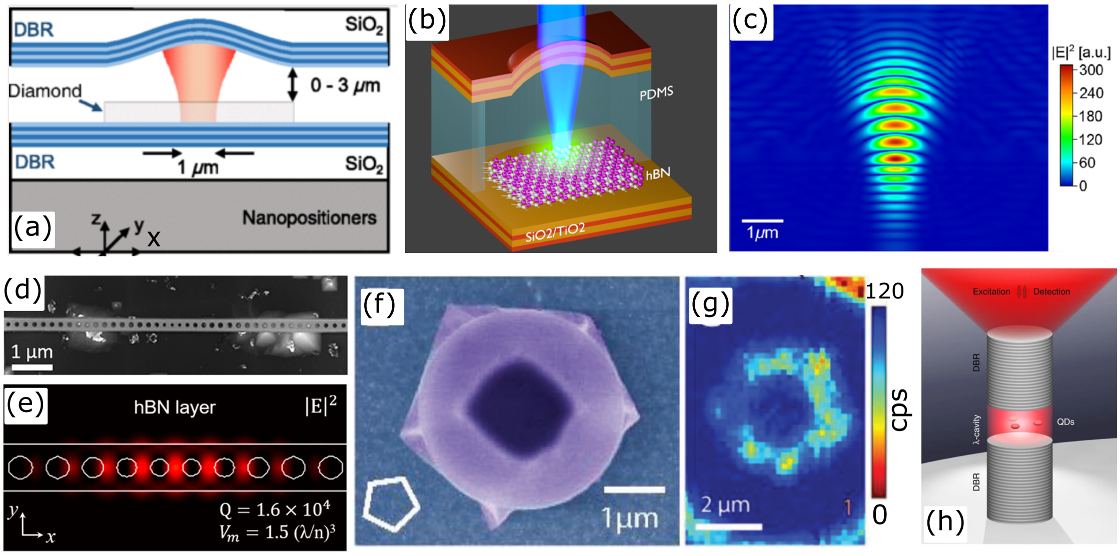

Figure 1. Examples of SPE-dielectric cavity coupled systems. (a) Color center in a diamond coupled to a fiber microcavity.[12] (b) Microcavity consisting of a hemispherical and a flat mirror with the quantum emitter hosted by hexagonal boron nitride (hBN). PDMS spacer sets the cavity length. (c) Electric field mode profile of the cavity in (b) obtained using finite difference time domain (FDTD) simulations.[17] (d) SEM image and (e) mode supported by the photonic crystal cavity coupled to an SPE in hBN.[18] (f) SEM image of 20 nm thick hBN on a silicon nitride microdisk. The shape of the hBN crystal is indicated in the figure. (g) Confocal fluorescence image of a microdisk resonator.[26] (h) Schematic of a quantum dot coupled to a micropillar Bragg cavity.[29] Panel (a) Reprinted with permission from Riedel et al. Deterministic Enhancement of Coherent Photon Generation from a Nitrogen-Vacancy Center in Ultrapure Diamond. *Phys. Rev. X* **2017**, 7, 031040 (DOI: 10.1103/PhysRevX.7.031040). Copyright 2017 American Physical Society. Panels (b) and (c) Reprinted with permission from Vogl et al. Compact Cavity-Enhanced Single-Photon Generation with Hexagonal Boron Nitride. *ACS Photonics* **2019**, *6* (8), 1955−1962. Copyright 2019 American Chemical Society. Panels (d) and (e) Reprinted with permission from Fröch et al. Hexagonal Boron Nitride Quantum Emitters to Photonic Crystal Cavities. *ACS Nano* **2020**, *14*, 7085−7091. Copyright 2020 American Chemical Society. Panels (f) and (g) Reprinted with permission from Proscia et al. Microcavity-coupled emitters in hexagonal boron nitride. *Nanophotonics* **2020**, 9 **2020**, *9*, 2937−2944 (DOI: 10.1515/nanoph-2020-0187). Copyright 2020 De Gruyter. Panel (h) Reprinted with permission from Albert et al. Microcavity controlled coupling of excitonic qubits. *Nature Communications* **2013**, *4*, 1747 (DOI: 10.1038/ncomms2764). Copyright 2013 Nature Publishing Group.

two-level quantum system as well as for enhanced interaction of single photons with a single molecule.[16] Drawbacks of this cavity design include relatively large mode volumes and problematic scalability. Utilizing focused ion beam (FIB) milling lower cavity mode volumes have been obtained and these cavities have been coupled to various emitters (Figure 1b, c).[13,17] A dielectric cavity formed in a photonic crystal waveguide (PCW) is an alternative way to combine an optical cavity and an optical waveguide (Figure 1d,e).[18] PCW-based cavities are attractive because their mode volumes are decreased down to the diffraction limit in combination with high quality factors (Q), while PCWs can naturally be integrated into on-chip quantum networks. Quantum dots, color centers in diamonds and SPEs in two-dimensional materials have been coupled to PCW cavities.[18−22] PCWs with cavities containing SPEs can also be efficiently coupled to optical fibers. Because of the efficient coupling to on-chip networks and optical fibers as well as enhanced SPE properties, several promising SPE-PCW-based fiber-coupled configurations have been developed, demonstrating the application potential for scalable photonic quantum networks.[23,24] PCW-cavity coupled color centers in diamonds can also be tuned by applying strain to the diamond, a feature that enhances their prospects for being used in scalable systems.[25] Cavity structures supporting whispering gallery modes, such as microdisks, can be used for obtaining very high Q, and have been utilized for coupling to quantum emitters (Figure 1f,g).[26,27] Such cavities can have a very high Q (in excess of 10^7), but their limitation from the viewpoint of Purcell factor is related to rather large mode volumes.[28] Another promising optical cavity configuration represents a microcavity pillar with a quantum dot located in the center of a Bragg cavity (Figure 1h). Such cavities are capable of greatly enhancing the SPE emission rate while also channeling the emitted photons into a single optical mode.[29,30] Moreover, quantum dots in the microcavity pillars can be electrically controlled to emit long chains of indistinguishable photons.[31] Therefore, these configurations have been utilized for photonic quantum applications, such as boson sampling.[32−34] It should however be noted that all these remarkable achievements can be demonstrated only at very low (liquid helium) temperatures at which the influence of phonon-mediated dephasing can be neglected. The fact is that, despite significant enhancements of photon emission rates demonstrated with photonic configurations, the fundamental limit of the emission enhancement related to the diffraction limit in cavity volumes do not allow to go even close to dealing with the decoherence problem at room temperature,[9] simply because high-quality cavities cannot emit photons sufficiently fast.[7]

Plasmonic cavities, representing resonant configurations for surface plasmon-polariton modes, offer a clear advantage over dielectric cavities because their mode volumes can be ultra-small.[7,8] Metallic structures supporting gap-plasmon modes with gaps down to few nanometers have been experimentally realized and coupled to SPEs.[35−40] Most dramatic enhancement of the photon emission rates have been demonstrated with SPEs, such as a nitrogen vacancy center (Figure 2a) or a quantum dot (Figure 2b) coupled to the gap plasmon mode supported by a silver nanocube and silver[37] or gold[36] surfaces. Such cavities offer high decay-rate enhancements, amounting to ∼3 orders of

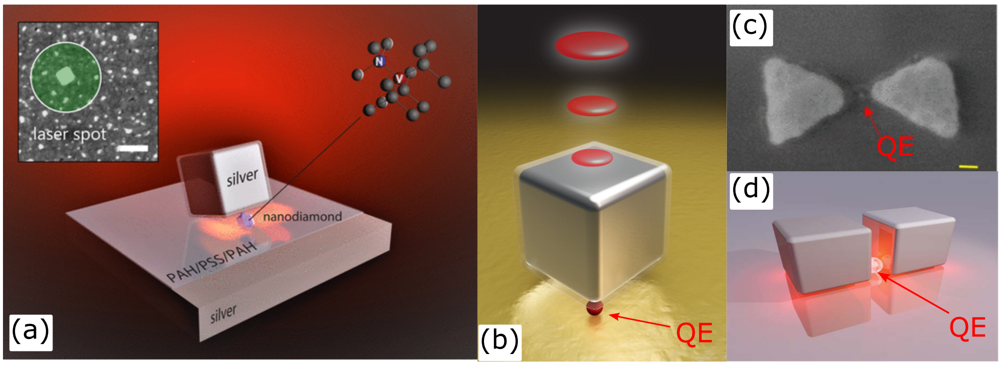

Figure 2. Examples of SPE-plasmonic cavity coupled systems. (a) Color center in a diamond coupled to a gap-plasmon cavity supported by a silver nanocube and a silver surface.[37] (b) Schematic of a quantum dot coupled to a gap-plasmon cavity supported by a silver nanocube and a gold surface.[36] (c) SEM image of quantum dot coupled to a plasmonic bow-tie cavity.[38] (d) Schematic of an NV center in a nanodiamond coupled to a nanocube dimer.[41] Panel (a) Reprinted with permission from Bogdanov et al. Ultrabright Room-Temperature Sub-Nanosecond Emission from Single Nitrogen-Vacancy Centers Coupled to Nanopatch Antennas. *Nano Letters* **2018** *18*, 4837−4844. Copyright 2018 American Chemical Society. Panel (b) Reprinted with permission from Hoang et al. Ultrafast Room-Temperature Single Photon Emission from Quantum Dots Coupled to Plasmonic Nanocavities. *Nano Letters* **2016**, *16*, 270−275. Copyright 2016 American Chemical Society. Panel (c) Reprinted with permission from Gupta et al. Complex plasmon-exciton dynamics revealed through quantum dot light emission in a nanocavity. *Nature Communications* **2021**, *12*, 1310 (DOI: 10.1038/s41467-021-21539-z). Copyright 2021 Nature Publishing Group. Panel (d) Reprinted with permission from Andersen et al. Ultrabright Linearly Polarized Photon Generation from a Nitrogen Vacancy Center in a Nanocube Dimer Antenna. *Nano Letters* **2017**, *17*, 3889−3895. Copyright 2017 American Chemical Society.

magnitude, together with well-directed emission. Similarly, bow-tie antennas (Figure 2c) have also been utilized for obtaining high decay-rate enhancements together with efficient collection of photons.[35,38] Plasmonic cavities can also be utilized for obtaining highly polarized single photons from emitters, which do not emit photons with a well-defined polarization. Thus, a plasmonic cavity formed by the gap of a nanocube dimer allows one to both enhance the photon emission rate by placing an SPE in the gap (Figure 2d) and obtain a linearly polarized stream of photons with the polarization dictated by the dimer axis.[41] We consider all these experimental demonstrations as only a beginning, with the full potential of plasmonic cavities yet to be realized, as discussed below.

Given the freedom of optimizing the plasmonic cavity configuration for the photon emission enhancement, several theoretically proposed plasmonic structures were found to promise the decay-rate enhancements exceeding 5 orders of magnitude.[42−44] It has also been predicted that, in principle, the emission rate enhancement can be so large that it would overcome the decoherence problem (i.e., the emission time would become shorter than the dephasing time), resulting in a Fourier Transform limited emission spectrum and the generation of indistinguishable photons at room temperature.[9,45] Importantly, the use of gap-plasmon modes is common in all these proposals, implying that with further improvements in design possible decay-rate enhancements can exceed one million. Here, it should be commented that the emission rate enhancement in simple plasmonic configurations, although offering 2 orders of magnitude of improvement over dielectric cavities, is still limited to 5 orders of magnitude, because very small cavities are poor emitters of radiation.[8] The configurations involving gap-plasmon modes are attractive also because they typically involve a very small cavity (such as a gap) volume to boost the enhancement and a large antenna (such as a nanocube) volume to efficiently outcouple gap plasmons into propagating away photons.[46] This is the underlying mechanism resulting in plasmonic cavity-antenna hybrid structures to offer promise of large emission rate enhancements, even exceeding one million.[35−37,44−46]

There are still, however, a few challenges to be dealt with. One worry with plasmonic cavities is quenching of emission near the metal surfaces. However, in appropriately designed plasmonic cavities, quenching can be minimized and made negligible even when the gap widths are below 2 nm. This is achieved due to the hybridization of plasmonic modes that decreases quenching near metals in ultranarrow gaps between two metal surfaces.[47,48] This suppression of quenching makes possible the total decay-rate to be enhanced and the photon emission to be efficiently funneled into a single optical mode.[49]

Efficient generation and channeling of photons from SPEs coupled to plasmonic cavities will be the key to its application in photonic quantum technologies. For designing such structures appropriate methods should be utilized in order to incorporate nonclassical effects, as they become important when the gap sizes are below 2 nm.[50,51] With properly designed plasmonic cavity, emission from SPEs can be efficiently channelled into an optical mode, where a dielectric cavity can be used to further enhance the emission at its resonance and the emission directed in one path. A schematic of such a system is shown in Figure 3. In the schematic, gold antennas are used to enhance and direct the emission into a titanium oxide dielectric cavity. In addition, the same antennas can be utilized to tune the emission wavelength of SPEs by applying suitable voltages and making use of the Stark shift.[52,53] A titanium oxide cavity is utilized to further enhance and direct the emission into a single mode photonic waveguide. Such a structure can form a node in a complex photonic quantum network.

Another important issue that needs to be tackled for obtaining indistinguishable photons at room temperature is availability of suitable SPEs that can be placed in ultrasmall plasmonic gaps. Molecules have been utilized in such gaps to obtain strong coupling,[54] for example. However, the fluorescence spectrum of molecules is very broad and tend to bleach-out very fast at room temperature.[54,55] Defects in two-dimensional (2D) materials have the potential to be used in such gap-plasmonic structures, and this has been demonstrated as well.[56] Some of the SPEs in hBN have been shown to have Fourier transform (FT) limited emission lines at room temperature.[57] SPEs in 2D materials are

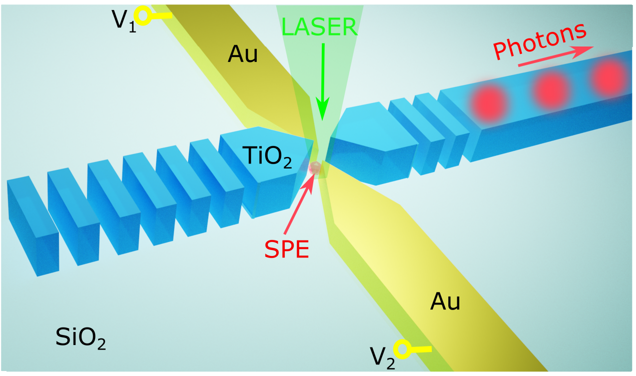

Figure 3. Schematic of the proposed experiment. An SPE is coupled to a gap-plasmonic mode which channels photons into a dielectric cavity. The dielectric cavity, in turn, couples the emitted photons to an optical waveguide.

promising for quantum technologies.[58] However, the atomic structure for many of the SPEs in 2D materials are not known.[59] Therefore, further exploration of SPEs in 2D materials is required to discover an emitter with spectrally narrow (preferably FT limited) and stable emission at room temperature. Defects in diamonds have been shown to be promising for quantum technologies. Nitrogen vacancy (NV) centers, in particular, have been used for many quantum technological demonstrations, including being used as nodes in a quantum network.[60] NV centers in diamonds, however, have a broad emission spectrum (~200 nm) at room temperature,[61] a feature that can be detrimental for some applications. Group IV defects in diamonds, such as silicon vacancy (SiV) centers and germanium vacancy (GeV) centers, have relatively narrow spectrum even at room temperature. Therefore, with enough decay-rate enhancement, they can be made to emit indistinguishable photons at room temperature. SiV centers in small nanodiamonds, sizes down to 2 nm, have been shown to be stable emitters at room temperature.[62] However, these nanodiamonds are not easily available, even though there is some encouraging development in this direction.[63]

With metallic structures and suitable SPEs available, it will also be a challenge to assemble them when the accuracy required for assembly will be of the nanometer scale. Therefore, new methods to place SPEs in the desired locations are encouraging.[64] A combination of methods, such as optical and plasmonic trapping of nanoparticles, electron-beam lithography, and directed self-assembly of nanoparticles, can be used to achieve assembly of SPEs in the gap between plasmonic structures in a scalable way.[65,66] For proof-of-principle experiments manipulation of nanoparticles by the tip of an atomic force microscope (AFM), in combination with other methods, can be used.[67]

In conclusion, in the future we expect to see many realizations where a hybrid cavity will be utlized with very small gap-plasmon mode in combination with dielectric cavities to harness the advantages of both kinds of cavities. This will eventually enable photonic quantum technologies to work at room temperature.[68] This possibility relies on many developments such as availability of SPEs in ultrasmall sizes, their positioning in ultrananogaps, and also the fabrication technology to scalably and reproducibly produce such structures.

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Notes

The authors declare no competing financial interest.

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