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Progress in quantum-dot single photon sources for quantum information technologies: A broad spectrum overview

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ABSTRACT

Semiconductor quantum dots (QDs) of various material systems are being heavily researched for the development of solid state single photon emitters, which are required for optical quantum computing and related technologies such as quantum key distribution and quantum metrology. In this review article, we give a broad spectrum overview of the QD-based single photon emitters developed to date, from the telecommunication bands in the IR to the deep UV.

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I. INTRODUCTION

The development of photonics based quantum information technologies is poised to usher in a paradigm shift in data processing and communication protocols, leading to the establishment of a quantum network in which photonic qubits are created and manipulated on demand for computational applications and the secure transmission of data. One of the fundamental device elements of such technologies will be the single photon emitter: the quantum light source that provides single photons. Particular applications include optical quantum computing,[1–4] quantum key distribution for secure communications,[5] quantum random number generation,[6] and quantum optical metrology using entangled photon N00N states.[7]

There are several known methods of generating single photons. These include the trapping and subsequent optical excitation of individual atoms/ions,[8] the use of electronic states in isolated molecules[9] and isolated defects in semiconductors,[10] parametric downconversion of laser pulses in non-linear crystals,[11,12] and the use of optical transitions between quantum confined, isolated, and electronic states in semiconductor quantum dots (QDs). Semiconductor quantum dots can be formed as colloids[13] or can be grown epitaxially.[14] Epitaxially grown semiconductor QDs offer several advantages for application as single photon sources, including the possibility of site control during growth[15] and their fabrication inside optical cavities or photonic structures for emission enhancement and control.[16,17] Furthermore, the emission energy of the QDs can be finely tuned by applying external electric/magnetic fields,[18,19] and the emitters can be electrically contacted with relative ease, allowing for the fabrication of single photon emitting diodes.[20] Moreover, the electronic states confined in QDs can be utilized and coherently controlled as matter qubits in their own right,[21–27] providing a means to interface matter-based quantum memories with photonic flying-qubits for the possible realization of a quantum network,[28,29] and the generation of polarization-entangled photon pairs can also be realized from QDs with appropriate symmetry.[30] In this review, we provide a comprehensive overview of the epitaxially grown semiconductor quantum dot single photon sources developed to date over a wide range of wavelengths and operation temperatures.

II. SEMICONDUCTOR QUANTUM DOTS FOR SINGLE PHOTON EMISSION

Since the proposal of semiconductor quantum dots and their application to lasers for improving the lasing characteristics of semiconductor laser diodes in the early 1980s,[31] many research groups around the world have made great progress in the development of fabrication methods for quantum nanostructures and also the development of consistent theoretical models to explain their underlying physical properties. A semiconductor quantum dot typically consists of a small region of the semiconductor (∼10 nm in size) surrounded by another semiconductor with a larger bandgap. The surrounding material provides a confining potential barrier, such that electrons/holes in the conduction/valence band can be confined within the quantum dot. Due to the three-dimensional nature of the quantum confinement, the confined electrons and holes exhibit a delta-function-like density of states, which, when coupled with the fermionic nature of electrons, allows for the spectral isolation of individual optical transitions involving single electrons and hence the realization of single photon emission. Due to their corresponding atomic-like discrete energy levels, semiconductor quantum dots are often given the moniker *artificial atoms*. The absolute energy of the emitted photons is determined by the bandgaps of the chosen material system, the size of the QD, and induced strain effects, such that single photon emission from semiconductor QDs can be realized over a range of energies, tailored for specific applications.

Figure 1 shows a selection of images of epitaxially grown QDs from the literature. Growth is typically performed using Molecular Beam Epitaxy (MBE) or Metal-Organic Chemical Vapor Deposition (MOCVD). While these processes are generally used to grow semiconductor materials layer-by-layer, three-dimensional nanoscale structures can be formed using several techniques, including careful control of the reactor conditions and material strain during growth, the use of pre-growth substrate patterning to promote growth in selected areas, and the use of post-growth processing such as lithography and wet/dry etching. To date, the majority of studies on semiconductor QDs have been performed on self-assembled structures, which form due to a partial release of built-up strain when the QD material is coherently grown layer by layer on top of a material with a differing in-plane lattice constant [the Stranski–Krastanow (SK) growth mode]. This growth mechanism was first noted by Stranski and Krastanow in 1938.[32] It was not until 1985, however, in a seminal work by Goldstein *et al.* that the formation of SK-grown QDs was discovered by electron

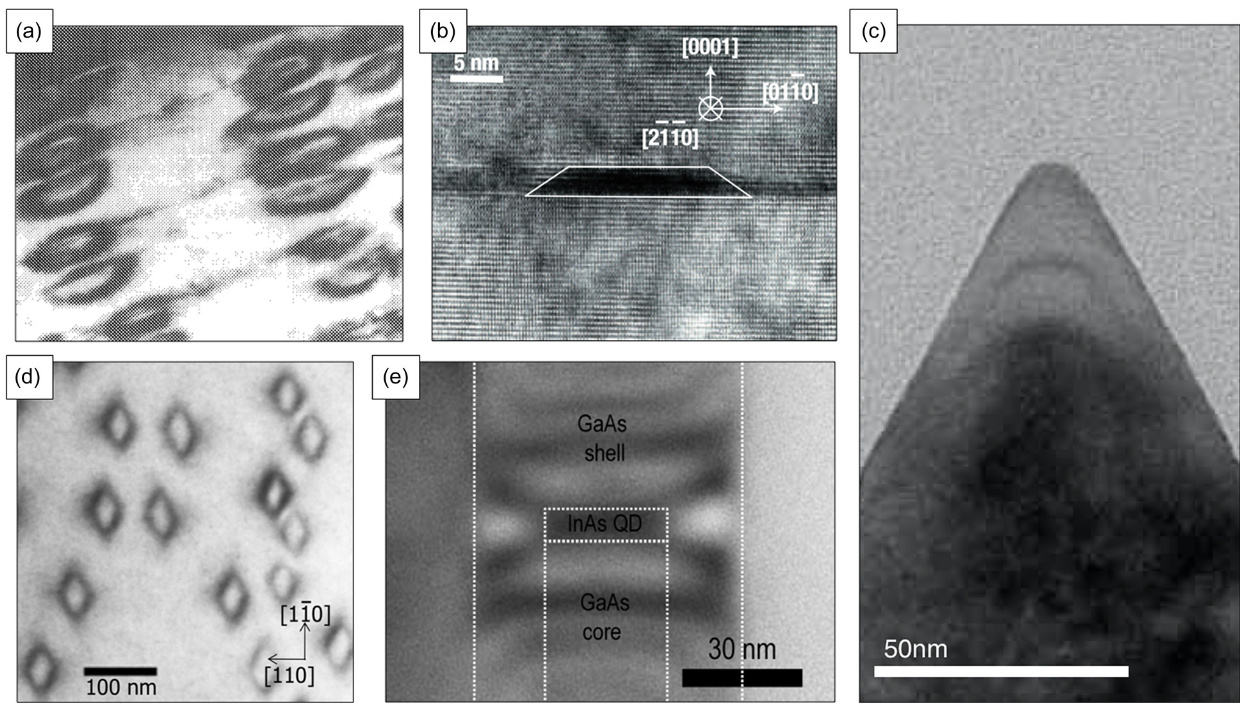

FIG. 1. Example electron microscopy images of epitaxially grown quantum dots. (a) An early example of InAs/GaAs QDs grown by MOCVD. Reprinted with permission from Goldstein *et al.*, Appl. Phys. Lett. **47**(10), 1099–1101 (1985). Copyright 1985 AIP Publishing.[14] (b) A single self-assembled GaN/AlN QD. Reprinted with permission from Kako *et al.*, Nat. Mater. **5**(11), 887–892 (2006). Copyright 2006 Springer Nature.[170] (c) A single site controlled GaN QD located near the tip of an AlGaN nanowire. Reprinted with permission from Holmes *et al.*, Nano Lett. **14**(2), 982–986 (2014). Copyright 2014 American Chemical Society.[38] (d) Self-assembled InAs/InP QDs. Reprinted with permission from Miyazawa *et al.*, Phys. Status Solidi C **8**(2), 417–4199 (2011). Copyright 2011 John Wiley and Sons.[103] (e) A single InAs/GaAs QD grown in a site controlled nanowire. Reprinted with permission from Tatebayashi *et al.*, Appl. Phys. Lett. **100**(26), 263101 (2012). Copyright 2012 AIP Publishing.[142]

microscopy and photoluminescence measurements.[14] Such QDs exhibit fine optical properties, with the drawback that they form at random locations and with a distribution in size (and hence a distribution in emission energy). Much work has been performed on controlling the density of self-assembled QDs by the SK modes via MBE and MOCVD growth techniques,[33–36] such that single structures can be isolated and used for the generation of single photons. In parallel, other growth methods such as droplet epitaxy have also been studied to control the QD symmetry, and efforts have also been made to fabricate site-controlled structures, such that a single QD can be fabricated in a pre-determined location for the deterministic fabrication of a single photon emitter.[37]

Research into epitaxially grown semiconductor QDs has progressed at an impressive rate, and single photon emitters based on structures of various materials and geometries have been developed operating in a wide range of wavelengths from the telecommunication band in the IR at 1.55 μm to the deep UV region at $<$280 nm. The majority of experimental demonstrations of single photon emission using semiconductor QDs are performed with the assistance of cryogenic cooling (typically using liquid helium at 4 K). This cooling suppresses the effects of phonons and electronic noise, freezing out non-radiative pathways and allowing for brighter emission with a higher degree of coherence and purity. However, in recent years, single photon emission from semiconductor QDs has also been realized at temperatures up to room temperature[38,39] and beyond.[40] Emission at high temperatures is accompanied by a large degree of phonon-related linewidth broadening, which drastically reduces the coherence of the emission.

A schematic of the single photon emission process using the confined electronic states of a quantum dot is shown in Fig. 2. Examples of the confined state wavefunctions can be found in Refs. 41–43. Optical excitation, which can be performed using either pulsed or continuous wave lasers, is typically used to excite electrons and holes into the dot (a brief discussion on other excitation methods is given at the end of this section). The optical excitation may be performed non-resonantly (by excitation of electrons/holes into the surrounding bulk material or wetting layer), by quasi-resonant excitation into excited states of the QD [usually detected by photoluminescence excitation spectroscopy (PLE)[43–49]], or by direct resonant excitation of single photon emitting transition,[50–53] also known as resonance fluorescence (RF). In the case of non-resonant or quasi-resonant excitation, an additional relaxation process is required: the excited electrons become trapped in the potential of the QD and rapidly relax to the lowest empty state via the emission of acoustic phonons.[46] Non-resonant excitation and quasi-resonant excitation are experimentally convenient in that the excitation wavelength can be easily filtered from the emission but result in reduced emission coherence and photon indistinguishability due to time uncertainty introduced by the relaxation process. Moreover, high energy excitation is likely to induce charge noise in the barrier material in the vicinity of the QD, leading to undesirable spectral diffusion.[54–56] Purely resonant excitation largely overcomes these problems but typically requires an additional use of crossed polarizers to filter out the excitation laser from the emission,[57] which necessarily block some of the emitted light. Recent advances in dichromatic spectral pulse-shaping may be able to overcome this drawback.[58]

Once an electron hole pair is formed in the dot, they will recombine to emit a single photon with an energy determined by their respective energy levels and any mutual coulomb interactions (excitonic effects) and with an emission lifetime determined by the inverse of the dipole matrix element considering the overlap of the electron and hole wavefunctions (although the emission rate may be enhanced via the use of optical cavities to control the available optical modes). In the case of purely resonant excitation of the two-level system transition with a pulsed laser, it is possible to deterministically excite a single electron/hole pair in the QD via application of an optical π-pulse. Such an excitation pulse can in effect be used as a trigger for a single photon gun (although true deterministic photon emission will also require photon extraction/collection with zero loss). The energy levels illustrated in Fig. 2 are each doubly degenerate due to the different projections of the electron/hole spin states into the z-direction.

When the QD is occupied with two electrons and two holes, a biexciton is formed. The total energy of the system differs from twice the exciton energy due to additional mutual interactions between all

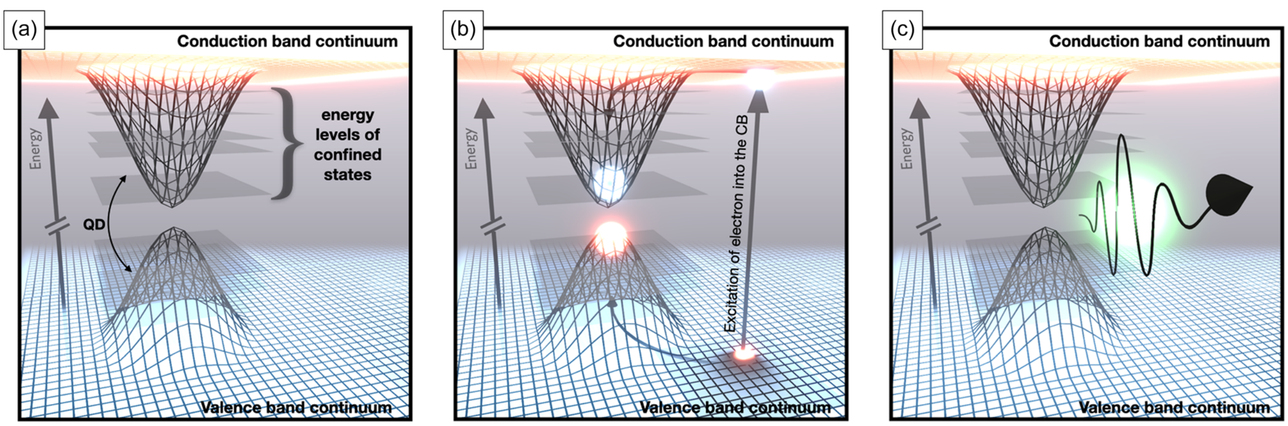

FIG. 2. QD states and the single photon emission process. (a) Empty QD. (b) Excitation of an electron (hole) into the conduction (valence) band. The carriers are captured in the potential of the QD and relax to the lowest (highest) energy empty energy level. (c) Recombination of the electron hole pair results in the emission of a single photon, returning the QD to its empty state, from which it cannot emit another photon until it is re-excited with another electron–hole pair.

the electrons/holes. A cascaded emission process ensues: the biexciton state decays via the emission of a single photon to one of the two possible exciton states [which may be energetically split by anisotropy: the so-called fine structure splitting (FSS)], and the exciton state then decays to the crystal ground state by the emission of another single photon. In general, the energies of the two emitted photons in the cascade are different due to the biexciton binding energy, although they can be quantumly entangled under the condition of vanishing FSS. Resonant 2-photon pulsed excitation of the biexciton state[59–62] (with each photon having half of the biexciton energy) has also proven extremely useful as a means to directly excite carriers into the QD and as a means to directly generate pairs of entangled photons using the biexciton cascaded emission, with the benefit that the excitation laser can be spectrally filtered from the emission. Two-photon resonant excitation provides an additional benefit in that it strongly suppresses re-excitation of the states from the laser pulse, as the whole cascade process must occur before the excitation photons can re-excite the dot, allowing for single photon generation with high purity.

It is also possible to electrically inject electrons into QDs by forming them within a light emitting diode structure and applying a potential difference across the dot. Direct charge injection using an electron beam microscope can also be used for excitation, which has been proved useful for studies on QD properties with high spatial resolution.[63,64]

The electron–hole recombination acts as an optical dipole, such that the photon emission essentially occurs in all directions. Moreover, due to the refractive index of the host material/structure, the majority of emitted photons from an as-grown QD are lost due to internal reflection before they can be collected. Great efforts have been made to enhance the emission directionality and overcome these losses via the fabrication of a variety of cavities and photonic structures,[16,65–75] in some cases leading to theoretical photon extraction efficiencies up to $\sim$90%.[76] Work has also progressed on the direct coupling of QDs to waveguides, such that the excitation/emission can be efficiently controlled.[77–81]

III. CHARACTERIZATION OF SINGLE PHOTON EMISSION

Single photon emission is typically confirmed via a measurement of the second order correlation function of the optical field at time delay τ,

$$g^{(2)}(\tau) = \frac{\langle \hat{a}^\dagger(t)\hat{a}^\dagger(t+\tau)\hat{a}(t+\tau)\hat{a}(t)\rangle}{\langle \hat{a}^\dagger(t)\hat{a}(t)\rangle^2}, \quad (1)$$

where $\hat{a}^\dagger$ and $\hat{a}$ are the creation and annihilation operators of the optical field, respectively. Experimentally, this is performed using a Hanbury Brown and Twiss (HBT) setup.[82] The time intervals between photon detection events at the exit ports of a 50/50 beam splitter are measured and then plot in a histogram, as illustrated in Fig. 3(a). Such a measurement is equivalent to $g^{(2)}(\tau)$ under the conditions that the reciprocal of the count rate is lower than the average time between emitted photons, i.e., in the low detection efficiency regime.[83,84] Quantum light is characterized by $g^{(2)}(0) < 1$ [as opposed to coherent light, which exhibits $g^{(2)}(0) = 1$, and thermal light, which exhibits $g^{(2)}(0) > 1$]. A pure single photon emitter will never emit more than one photon at a time, and its emission is characterized by $g^{(2)}(0) = 0$. In terms of the HBT measurement, this can be understood that with only a single photon entering the system at any given instant, the probability of detecting photons at both detectors at the same time ($\tau = 0$) is zero. In the theoretical case of a two-photon emitter that emits photon pairs into the same optical mode, $g^{(2)}(0) = 0.5$. The value of 0.5, therefore, provides an upper limit on an experimentally measured value of $g^{(2)}(0)$ required to verify the presence of single

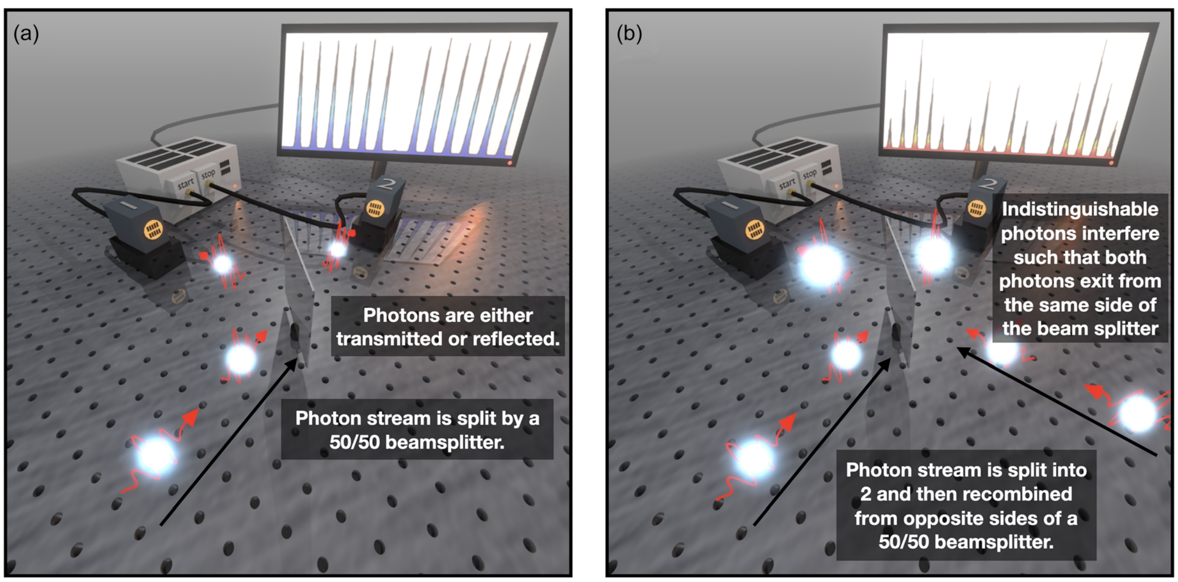

FIG. 3. Experiments for characterizing single photon emission: (a) HBT measurement of photons arriving on a beam splitter. (b) HOM measurement whereby the emission has been split into beams, which are recombined from opposite sides of a beam splitter. In the case of purely indistinguishable photons perfectly overlapping in time and space on the beam splitter, both particles will leave the beam splitter from the same side (i.e., one of the photons is reflected, and the other is transmitted.).

photon emission. A measurement of $0 < g^{(2)}(0) < 0.5$ implies the measurement of a single photon emitter with some degree of contamination, for example, by imperfect isolation of the optical transition being measured. The measurement can be performed using CW or pulsed excitation, and the resulting data will differ accordingly. Under pulsed excitation, the histogram will consist of a series of peaks separated by the reciprocal of the excitation laser repetition rate, with no peak at time delay zero for a pure single photon emitter. For the case of CW excitation, the histogram will typically exhibit continuous data with an exponential dip in coincidence counts at time delay zero, where the reciprocal of the time constant of the antibunching dip is equal to the sum of the reciprocals of the emission lifetime and the pumping time. Examples of both types of dataset can be found in the figures of this review.

For the majority of quantum information processing (QIP) applications (requiring two-photon interference), single photon emission alone is not enough: photon *indistinguishability* is also required, such that the photons cannot be distinguished by an experiment in which their different emission times are negated (although indistinguishability is not strictly required for applications such as QKD). The indistinguishability of photons is confirmed via the measurement of Hong–Ou–Mandel (HOM) interference with a modified HBT setup configured in such a way that the emitted photon stream is split into two paths and then recombined from opposite sides of the 50/50 beam splitter before the two detectors, as shown in Fig. 3(b). Two identical photons impinging on opposite inputs of a beam splitter with perfect spatial and temporal overlap interfere such that both must exit from the same side of the beam splitter (i.e., one photon is transmitted, and the other is reflected).[85] Therefore, a reduction in correlation counts at time delay zero between two detectors at the outputs of the beam splitter quantifies the degree of two-photon overlap or indistinguishability of the emitted photons. If the photons are somehow distinguishable (e.g., having different polarizations or energies), they will not interfere, and simultaneous detection at both detectors will be possible. The HOM measurement can be performed using either pulsed[86] or CW[87] excitation, although pulsed excitation is more common. In this case, a delay line in the excitation path is used to split each laser pulse into a pair of temporally separated pulses to excite the QD in rapid succession such that the dot emits pairs of temporally separated photons (the delay between pulses must be set longer than the QD emission lifetime). The emission is subsequently directed to an unbalanced Mach–Zehnder interferometer (with a long/short path difference equal to the delay between the excitation pulses) and is interfered on the exit beam splitter before being detected and correlated at the two detectors. The resulting histogram comprises clusters of peaks corresponding to the different possible combinations of paths that the first and second photons take, respectively. In the case where the first emitted photon travels down the long path and the second emitted photon travels down the short path, resulting in perfect temporal overlap from opposite sides of the beam splitter, the photons interfere with each other (if they are indistinguishable) and leave the beam splitter in the same exit arm (i.e., one is transmitted and one is reflected), and no coincidence counts at the two detectors are measured for time-delay zero. The amplitudes of the surrounding peaks are determined by the probabilities of the corresponding other path combinations (i.e., both photons travel down the same path, or the first photon takes the short path and the second photon takes the long path). Perfect indistinguishability and two-photon overlap will result in complete suppression of coincidence counts at time delay zero.

A wide range of detector technologies are regularly employed for converting the photons into electronic signals for the correlation measurement.[88,89] These include avalanche photodiodes (APDs), photomultiplier tubes (PMTs), and superconducting nanowire single photon detectors (SNSPDs).[90] The detector efficiency, temporal resolution, and dark count rates vary wildly between the different types of detectors,[88] and the choice of detector usually depends on the photon wavelength. SNSPDs must be cooled below the critical temperature of their superconducting element (usually to liquid helium temperatures), but recent advances in their design allow for high efficiency (>90%) detection[91] with a low timing jitter,[92] and they can be designed to operate over a wide range of wavelengths from the UV to the infrared.[93,94] It is also possible to fabricate them into waveguides for photonic applications with minimal coupling loss.[95–97]

IV. THE STATE OF THE ART IN QD-BASED SINGLE PHOTON EMITTERS

Single photon emission has been successfully realized with a wide variety of QDs made from various materials, operating over a large range of emission wavelengths (see Fig. 4 for information on the bandgaps of the materials discussed in this review). In this section, we discuss some of the major recent advances in the development of QD-based single photon emitters by the emission wavelength region. We note that significant progress is also being made in wavelength tuning/conversion techniques, whereby large shifts in photon energy can be achieved using non-linear frequency conversion techniques,[98,99] and fine-tuning can be performed using techniques such as strain control[100] and/or the application of electric/magnetic fields.[18,19]

A. The telecommunication bands

Optical fibers used for telecommunications have low optical losses in the wavelength regions of ~1300 nm and ~1550 nm, the so-

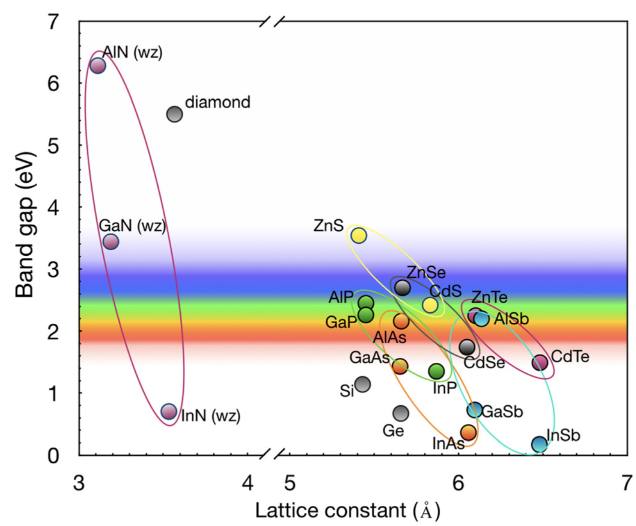

FIG. 4. Various semiconductor bandgaps, plot as a function of their in-plane lattice parameter.

called O-(original) and C-(conventional) bands. Of these, the lowest transmission losses can be found in the C-band. Therefore, developing single photon emitters that operate near this wavelength is essential for the realization of applications that require long-distance transmission, such as quantum key distribution. Moreover, the C-band offers an additional benefit in that it is promising for satellite-based quantum communication due to both reduced atmospheric scattering and a relatively low-background solar irradiance.[101] In principle, these wavelength bands can be covered by InAs/InP, InAs/GaAs, InSb/GaSb, and InN/GaN QDs. To date, there have been many reports on the optical properties of QDs emitting in these bands[102–105] and several reports on the realization of efficient single photon emission with high purity using InAs/InP or InAs/GaAs structures.[47,66,106–117]

Using InAs/InP QDs, Takemoto *et al.* developed and used an optical horn structure to increase the emitted photon extraction efficiency[66] [see Fig. 5(a)] and experimentally demonstrated a working QKD system capable of distributing secret keys at distances up to 120 km,[112] i.e., enough to cover a metropolitan area. In their demonstration, they employed quasi-resonant excitation of the quantum dot p-shell states and used an operational $g^{(2)}(0)$ value of 0.0051 (though they note that they were able to realize a lower $g^{(2)}(0)$ value with stricter spectral filtering). SNSPDs were used on the detection side, which provided both high enough detection efficiency (~10%) and low dark count rates down to 20 cps. During this experiment, both the QD and the SNSPD were cooled to liquid helium temperatures. Following this demonstration, Miyazawa *et al.* were able to report a $g^{(2)}(0)$ value of 4.4×10^{-4} from an InAs/InP QD in an optical horn at 8 K under quasi-resonant excitation,[47] which, they anticipate, may allow for QKD at distances up to 200 km [see Fig. 5(b)].

Muller *et al.* also used highly symmetric InAs/InP QDs, grown using MOVPE droplet epitaxy, to develop a single photon emitting diode operating in the C-band.[114] The structure comprised an electrically contacted asymmetric 2-λ cavity surrounded by distributed Bragg reflector (DBR) mirrors and exhibited a $g^{(2)}(0)$ value of 0.11, without any corrections at 4 K [see Fig. 5(c)]. Furthermore, the highly symmetric QDs exhibited a low fine structure splitting such that polarization entangled photons can be generated using the biexciton–exciton cascaded emission process. Moreover, it was shown that this photon entanglement can be maintained up to a temperature of 93.4 K, accessible with liquid nitrogen cooling. It was subsequently shown that under non-resonant optical excitation, similar QDs could exhibit a long coherence time of ~1 ns, and a successful demonstration of quantum teleportation was made.[118]

A source of indistinguishable photons in the telecommunication C-band, using InAs/InGaAs/GaAs QDs emitting at ~1550 nm, was reported by Nawrath *et al.*[119] In this case, they employed two-photon resonant excitation of, and measured the emission from, the biexciton state of an isolated QD, and were able to measure a $g^{(2)}(0)$ value of 0.072 (corrected for the time resolution of their experimental setup). They were then able to measure the degree of indistinguishability of post-selected photons to be 0.713 (0.894 when corrected for the detector response).

Kim *et al.* also realized a source of indistinguishable photons in the telecommunication O-band using InAs/InP QDs in two-dimensional photonic crystal cavities exhibiting a photon outcoupling efficiency of >36%. Cavity-enhanced emission lifetimes down to 400 ps, with $g^{(2)}(0)$ values down to 0.085, were measured at 4 K under non-resonant excitation. Two photon interference measurements revealed a photon indistinguishability of 0.18, limited by dephasing and timing jitter.[120] Post-selection of coincidences that occurred on timescales shorter than the coherence time (i.e., selecting only the data points corresponding to pairs of photons that arrived on the beam splitter within ~160 ps of each other) resulted in an improved indistinguishability of 0.67, with a reduction in brightness to <5% of the non-post-selected data.

It should be noted that these demonstrations of high quality single photon emission have been achieved using QDs that effectively form at random locations, requiring significant post-growth effort to

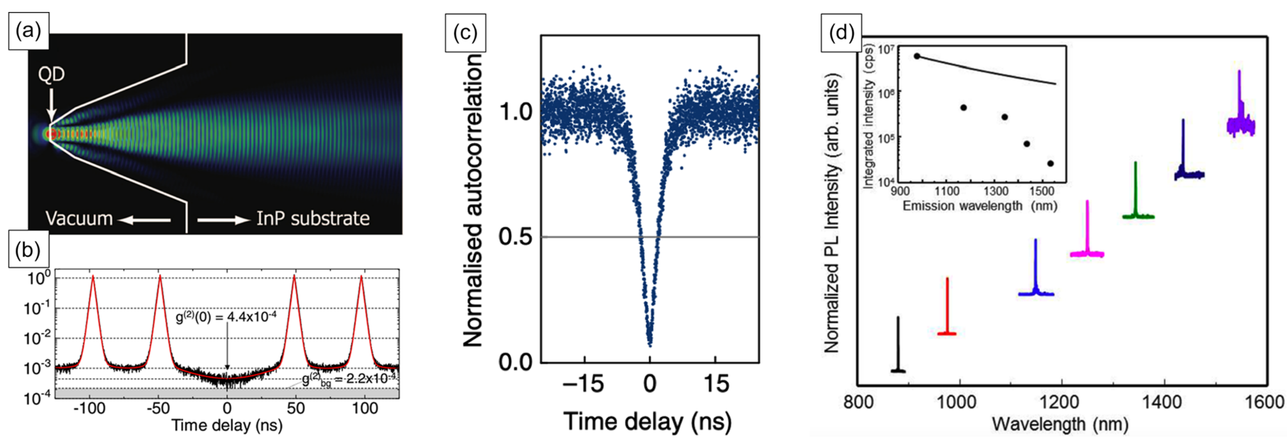

FIG. 5. (a) Emission profile from an InAs/InP QD in a photonic horn structure. Reprinted with permission from Takemoto *et al.*, J. Appl. Phys. **101**(8), 081720 (2007). Copyright (2007) AIP Publishing.[66] (b) Single photon emission exhibiting a $g^{(2)}(0)$ value of 4.4×10^{-4} measured from a QD in a photonic horn under quasi-resonant excitation. Reprinted with permission from Miyazawa *et al.*, Appl. Phys. Lett. **109**(13), 132106 (2016). Copyright (2016) AIP Publishing.[47] (c) $g^{(2)}$ data for quantum LEDs based on InAs/InP QDs operating at 1550 nm capable of generating entangled photons at temperatures up to 90 K. Reproduced with permission from Müller *et al.*, Nat. Commun. **9**(1), 862 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution 4.0 License.[114] (d) Emission from individual site-controlled InAsP QDs over a wide range of wavelengths in the telecom bands. Reprinted with permission from Haffouz *et al.*, Nano Lett. **18**(5), 3047–3052 (2018). Copyright 2018 American Chemical Society.[117]

locate high quality dots and/or perform any subsequent non-trivial device processing. In order to overcome this fundamental drawback, effort has also been made into the development of deterministic, site controlled, quantum dot single photon emitters operating in the telecom bands. Haffouz *et al.* recently reported bright single photon emission from InAsP/InP QDs in site-controlled InP nanowires grown using MOCVD on patterned substrates.[117] In addition to allowing for the position control of a single QD, the nanowire also acted as a waveguide to direct the emission from the QD. A $g^{(2)}(0) = 0.02$ value was obtained for an emitter in the O-band at 1342 nm, and the emission wavelength could be tuned from 880 nm to 1550 nm by changing the sizes of the QDs/nanowires during growth [see Fig. 5(d)].

B. The near infrared

Emission in the near infrared is typically covered by the III-arsenide material system, which has been investigated for use in transistors and used in laser diodes since the early 1960s.[121–124] Due to this historical maturity, growth techniques of the bulk semiconductor are well established, and III-arsenide quantum dots have been one of the most heavily studied types of epitaxial quantum dots to date.[14] It is no surprise, then, that the first report of single photon emission from an epitaxially grown quantum dot was from an InAs/GaAs dot operating at ∼940 nm.[83] Since then, there have been several significant developments with regard to single photon emitters, including the fabrication of single photon and entangled photon emitting diodes,[20,125] demonstrations of quantum logic gate operation,[52,126] and also quantum teleportation.[127–129]

Indistinguishable photon emission from InAs/GaAs QDs in nanopillars was reported by Santori *et al.*[86] Their QDs, emitting at ∼930 nm, exhibited a $g^{(2)}(0)$ value of ∼0.05 and a mean two-photon overlap up to 0.81 under non-resonant excitation with pairs of laser pulses separated by 2 ns. Many other groups have since reported on the realization of indistinguishable photon emission from III-As QDs using various sample geometries and excitation methods,[53,81,130–136] leading to demonstrations of near perfect indistinguishability in studies by He *et al.*[52] and Somaschi *et al.*,[74] which were achieved using purely resonant excitation of QDs in DBR microcavities. He *et al.* achieved a $g^{(2)}(0)$ value of 0.012 under π-pulse excitation using InAs/GaAs QDs emitting at ∼940 nm. They reported a two-photon interference visibility of 0.97 and then used two successively emitted photons (separated in time by 2 ns) to make an experimental demonstration of a working controlled-NOT gate. Somaschi *et al.* reported an indistinguishability of 0.989 from InGaAs/GaAs QDs in a DBR micropillar cavity [see Fig. 6(a)]. Their device employed tuning of the QD emission peak into resonance with the cavity at ∼925 nm via the application of an external bias, and they reported a $g^{(2)}(0)$ value of 0.0028 under these conditions. The resonant excitation removes the timing jitter of the carrier relaxation process involved in non-resonant excitation, allowing for a larger temporal overlap of the photons when recombined in the HOM interferometer.

Using a similar structure of a single InAs/GaAs QD in a micropillar cavity under resonant excitation (albeit without bias tuning), Wang *et al.* reported on the indistinguishability of photons at different emission delays,[133] demonstrating an indistinguishability of ∼0.9 even with photon separations of over 14 μs [see Fig. 6(b)]. Then, they went on to demonstrate a boson sampling experiment in which they demultiplexed the emitted photons and their single QD [$g^{(2)}(0) = 0.027$] into different spatial modes.[137] The demultiplexing process effectively produced multiple single photon sources (each with a reduced emission rate), which could then be interfered in a downstream photonic circuit.

Recently, Schweickert *et al.* used two-photon excitation of the biexciton state in a GaAs/AlGaAs QD to realize $g^{(2)}(0)$ values as low as 7.5×10^{-5} at a wavelength of ∼790 nm. High quality SNSPD detectors (also cooled to 4 K) with low dark count rates (≪1 cps) were used to enable accurate measurement of the photon purity. The two-photon resonant excitation heavily suppresses QD re-population and also uncorrelated background emission, enabling the clean isolation of a single excitonic transition in the QD.[138] In a similar study, Hanschke *et al.* reported a $g^{(2)}(0)$ value of 9.4×10^{-5}, using short pulses to further suppress any re-excitation of the dot.[139]

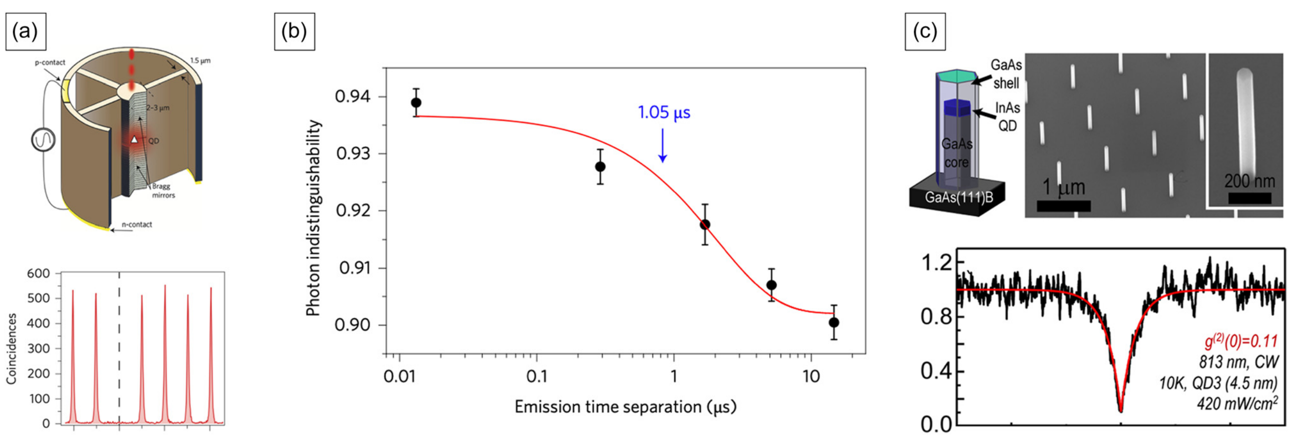

FIG. 6. (a) Schematic and $g^{(2)}$ data of a near optimal single photon emitter. Reprinted with permission from Somaschi *et al.*, Nat. Photonics **10**(5), 340–345 (2016). Copyright 2016 Springer Nature.[74] (b) Change in photon indistinguishability with delay between emission. Reprinted with permission from Wang *et al.*, Nat. Photonics **11**(6), 361–365 (2017). Copyright 2017 Springer Nature.[137] (c) Site-controlled nanowire InAs/GaAs QDs and a corresponding $g^{(2)}$ measurement. Reprinted with permission from Tatebayashi *et al.*, Appl. Phys. Lett. **100**(26), 263101 (2012). Copyright 2012 AIP Publishing.[142]

There has also been significant progress in the development of single photon emitting diodes using III-Arsenide materials. Yuan *et al.* reported the development of a device in which the emission from a self-assembled InAs QD grown in the active region of a GaAs *p–i–n* diode could be isolated and measured under electrical carrier injection. The device exhibited a $g^{(2)}(0)$ value of <0.07 at a temperature of 5 K, when corrected for a finite temporal resolution of the HBT. Since then, there have been many technological advances, leading to the report from Salter *et al.* of a device emitting pairs of entangled photons from the biexciton–exciton cascade.[140] The entangled-photon emitting diode also consisted of InAs QDs emitting at ∼885 nm, with very low FSS (∼0.4 μeV) such that entangled photon pairs from the biexciton cascade could be realized. The QDs were located in a planar DBR microcavity to enhance the emission output. Later demonstrations of indistinguishability between subsequently generated photons (two-photon interference visibility of 0.6),[141] and quantum teleportation using such devices has been reported.[127,128]

III-Arsenide materials emitting in the NIR also lead the way in terms of site-controlled QD growth. Although the above examples utilized self-assembled QDs, there have also been several reports of the successful site-control of single photon emitters using this material system. For example, Tatebayashi *et al.* developed a method to deterministically site control individual InAs QDs in GaAs nanowires,[142] enabling the fabrication of an array of quantum emitters [see Fig. 6(c)]. They realized single photon emission at 10 K at a wavelength of 813 nm with a $g^{(2)}(0)$ value of 0.11. It is also expected that control of the nanowire morphology will allow for deterministic control of the emission polarization.[143]

Similarly, Dalacu *et al.* also performed site controlled growth of InAsP QDs in InP nanowires and realized single photon emission at ∼950 nm with $g^{(2)}(0) < 0.005$ (T = 4 K).[144] The growth process resulted in defect-free nanowires to heavily suppress the electronic charge noise that often broadens QD emission linewidths via spectral diffusion.[55,56] They reported narrow emission linewidths down to 30 μeV.

In an alternative approach, Juska *et al.* formed arrays of site-controlled InGaAsN QDs in inverted pyramids that had been etched into a GaAs substrate.[15] The simultaneous site-control and geometry-control of the QDs achieved with this growth method allowed for the realization of highly symmetric QDs emitting at 865 nm with vanishing fine structure splitting. This, in turn, enabled the successful realization of polarization entangled photon emission at a temperature of 7 K from the biexciton-> exciton->empty QD cascade.

The choice of substrate for epitaxial growth is also important. While many of the above examples involved growth on native GaAs or InP substrates, it is also possible to grow QDs on silicon substrates, which paves the way for direct integration with silicon photonics. To that end, Kwoen *et al.* have reported the successful realization of an InAs/GaAs nanowire QD single photon emitter grown on Si and exhibiting $g^{(2)}(0) = 0.18$ at ∼905 nm at cryogenic temperatures (<10 K).[145]

C. The visible

Quantum dots formed from InGaN, InP, and various combinations of type II–VI semiconductors such as CdSe/ZnSe can be used to generate single photons with wavelengths in the visible region of the spectrum. Relatively low-cost and resilient plastic optic fibers (POFs) have transmission maxima in this region, making it an interesting wavelength region for possible applications that do not require extremely long transmission distances. Moreover, in contrast to the long wavelength emitters discussed above, single photon emission in the visible and ultraviolet has regularly been reported from QDs at ambient temperatures of 300 K and above.

Although II–VI QDs are often fabricated and studied in colloidal form,[13] there have also been several studies on epitaxially grown structures. Bounouar *et al.* realized single photon emission at 300 K with a $g^{(2)}(0)$ value of 0.48 when they isolated the biexciton emission line from a single CdSe QD grown epitaxially in a ZnSe nanowire [see Fig. 7(a)].[39] Emission lifetimes as short as 300 ps were measured under pulsed optical excitation. Fedorych *et al.* also realized room temperature single photon emission from CdSe/ZnSSe quantum dots.[146] Their structures were embedded in MgS barriers, and a $g^{(2)}(0)$ value of 0.16

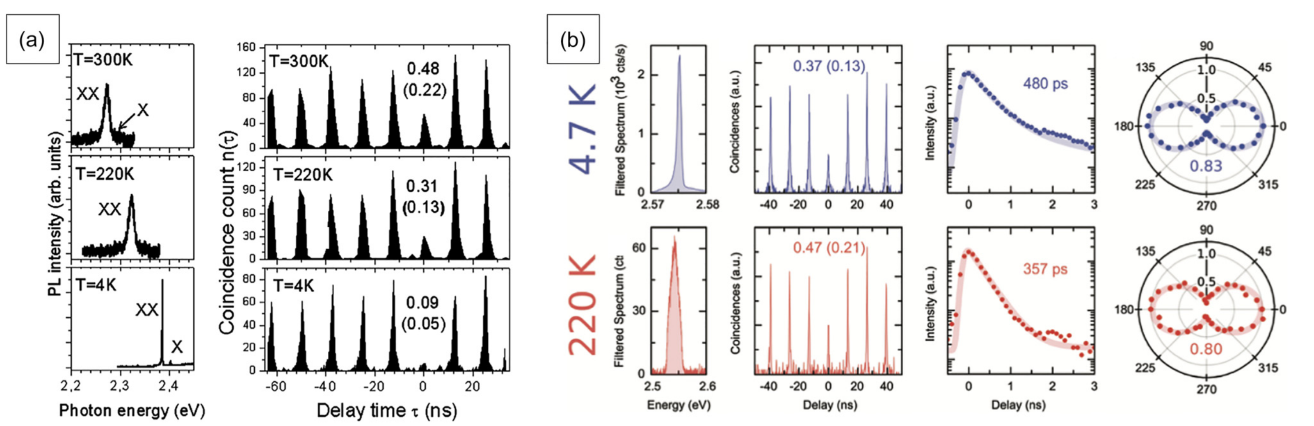

FIG. 7. (a) Emission spectra and autocorrelation data from the biexciton state of a CdSe QD in a ZnSe nanowire at various temperatures. Reprinted with permission from Bounouar *et al.*, Nano Lett. **12**(6), 2977–2981 (2012). Copyright (2012) American Chemical Society.[39] (b) Emission, autocorrelation, lifetime, and polarization from a non-polar InGaN/GaN QD at temperatures up to 220 K. Reproduced with permission from Wang *et al.*, Nanoscale, **9**, 9421 (2017). Copyright 2017 The Royal Society of Chemistry, licensed under a Creative Commons Attribution 3.0 Unported License.[164]

was reported after strict spectral filtering was used to isolate the emission from a single QD.

InGaN/GaN QDs are of interest as they can in principle be used to realize single photon emission over a wide range of wavelengths[147] depending on the indium mole fraction of the material. To date, most of the published examples of InGaN QD single photon emission have been realized in the visible, from a wide range of QD host structures, including the as-grown QDs,[148] nanowires,[149–154] micro/nano-pyramids,[155–157] QDs in microcavity structures,[158–160] and QDs formed by etching techniques.[161,162] Deshpande *et al.* realized an InGaN/GaN single photon emitting LED operating at a wavelength ~620 nm with operational $g^{(2)}(0)$ values of 0.37 (under pulsed current injection) and 0.32 (CW) at temperatures up to 280 K.[163] Wang *et al.* used InGaN QDs grown on a non-polar crystal plane that emit at ~490 nm to realize single photon emission with deterministic polarization (along the crystal *m*-direction) at temperatures up to 220 K [$g^{(2)}(0) = 0.47$].[164,165] These temperatures are reachable with thermoelectric cooling, and the degree of polarization could be maintained at 0.8 [see Fig. 7(b)]. Kocher *et al.* also used non-polar InGaN QDs emitting at ~420 nm to develop a deterministically polarized single photon emitting diode exhibiting a degree of linear polarization of 0.94 and a $g^{(2)}(0)$ value of 0.18 (corrected for the system response) at 4.5 K.[166] Deterministic and controllable single photon polarization was also realized using site-controlled InGaN QDs by Teng *et al.* via the development of elliptical QDs in shaped nanopillars.[161,167] Their structures, excited optically, exhibited a degree of linear polarization of 0.9 and a $g^{(2)}(0)$ value of 0.26 at 10 K. The polarization angle was controlled by choosing the major axis of the QDs' elliptical shape during device fabrication. It is anticipated that an electrically contacted device consisting of 4 such emitters oriented with appropriate polarizations could be used for QKD applications without the need for polarization filters.

D. The ultraviolet

The UV region of the spectrum is currently dominated by the III-nitrides, in particular GaN/AlGaN QDs.[147] This material system, which is technologically less mature than the III-arsenides, is now being intensively developed and is backed by a large industrial infrastructure due to its heavy use in solid state lighting and power electronics applications. While such short wavelength emission is

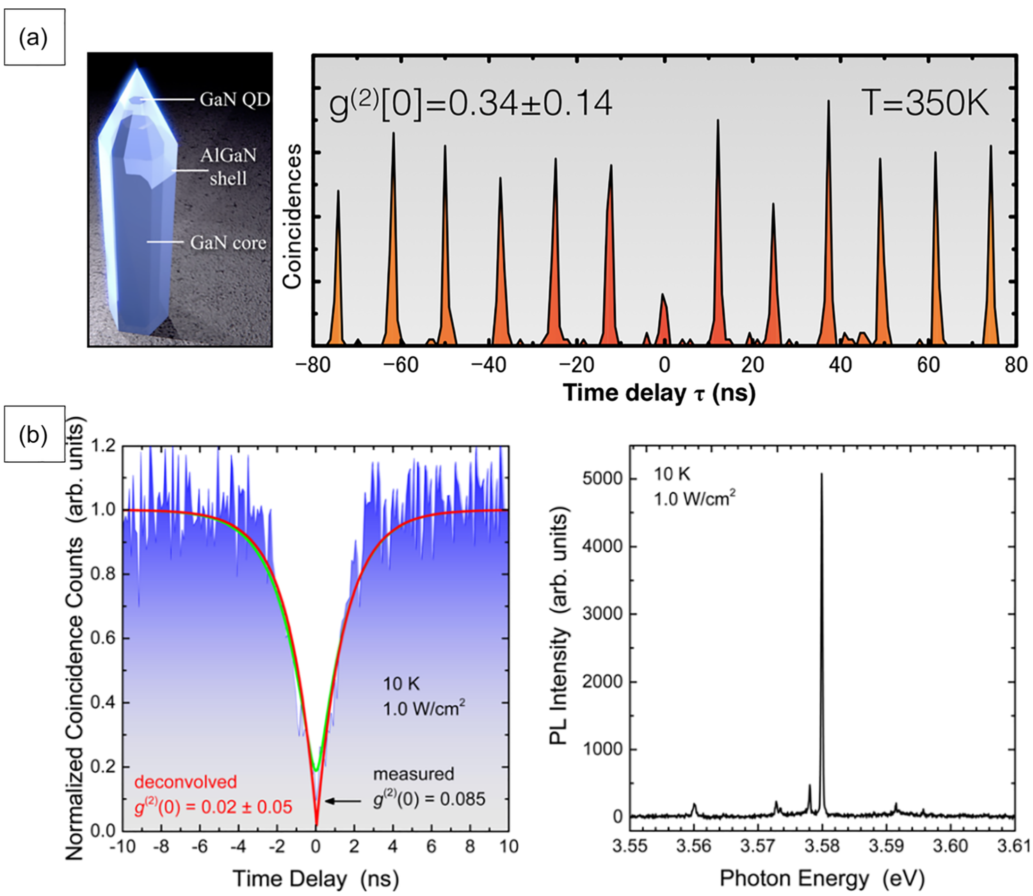

FIG. 8. (a) Schematic and g(2) data for a GaN nanowire QD single photon emitter operating at 350 K. Reprinted with permission from Holmes *et al.*, Nano Lett. **14**(2), 982–986 (2014). Copyright 2014 American Chemical Society.[38] Reprinted with permission from Holmes *et al.*, ACS Photonics **3**(4), 543–546 (2016). Copyright 2016 American Chemical Society.[40] (b) $g^{(2)}$ data and emission spectra for a GaN fluctuation QD at 10 K. Reprinted with permission from Arita *et al.*, Nano Lett. **17**(5), 2902–2907 (2017). Copyright 2017 American Chemical Society.[173]

currently not suitable for long distance fiber-based transmission, it is possible that single photon emitters in this wavelength range will be useful for on-chip applications (although direct conversion to the telecom c-band is also, in principle, possible[168,169]). Furthermore, the short wavelengths may allow for use of smaller transmission and receiving optics for free space communications,[170] particularly in the vacuum of space where atmospheric scattering will not be an issue. Interfacing with trapped ion based quantum memories, which typically have transmissions in the UV,[171,172] may also be possible. III-nitride QDs are of particular interest due to their large band offsets, which allow for strong quantum confinement and hence high temperature operation: to date, there have been several reports of high temperature single photon emission from III-nitride QDs,[38,40,170] including emission at temperatures up to 350 K[40] [see Fig. 8(a)]. For this study, Holmes *et al.* used GaN/AlGaN QDs formed near the apices of low-density site-controlled nanowires, such that single QDs could be readily identified, excited, and measured using an optical microscope. The $g^{(2)}(0)$ value at 350 K, with no applied corrections, is 0.34, which is limited by imperfect isolation of the emission peak caused by spectral contamination from the neighboring biexciton emission. During the same study, it was shown that luminescence from the QDs persisted up to temperatures as high as 400 K.

Relatively pure single photon emission from GaN/AlGaN QDs has been reported by Arita *et al.*, with a $g^{(2)}(0)$ value of 0.02 for interface fluctuation QDs[173] cooled down to a temperature of 10 K [see Fig. 8(b)]. These samples were grown at a slightly higher temperature in order to reduce the defect density in the AlGaN capping layer, leading to a higher spectral purity of the emission (and emission linewidths lower than 100 μeV). However, due to the weak lateral confinement provided by this particular QD geometry, the emission from the QDs could not be sufficiently isolated at temperatures above 70 K.[174]

III-nitrides generally crystalize in the non-centrosymmetric wurtzite structure, meaning that they are piezoelectric in nature. Strain due to lattice mismatch between the QDs and the barrier material therefore

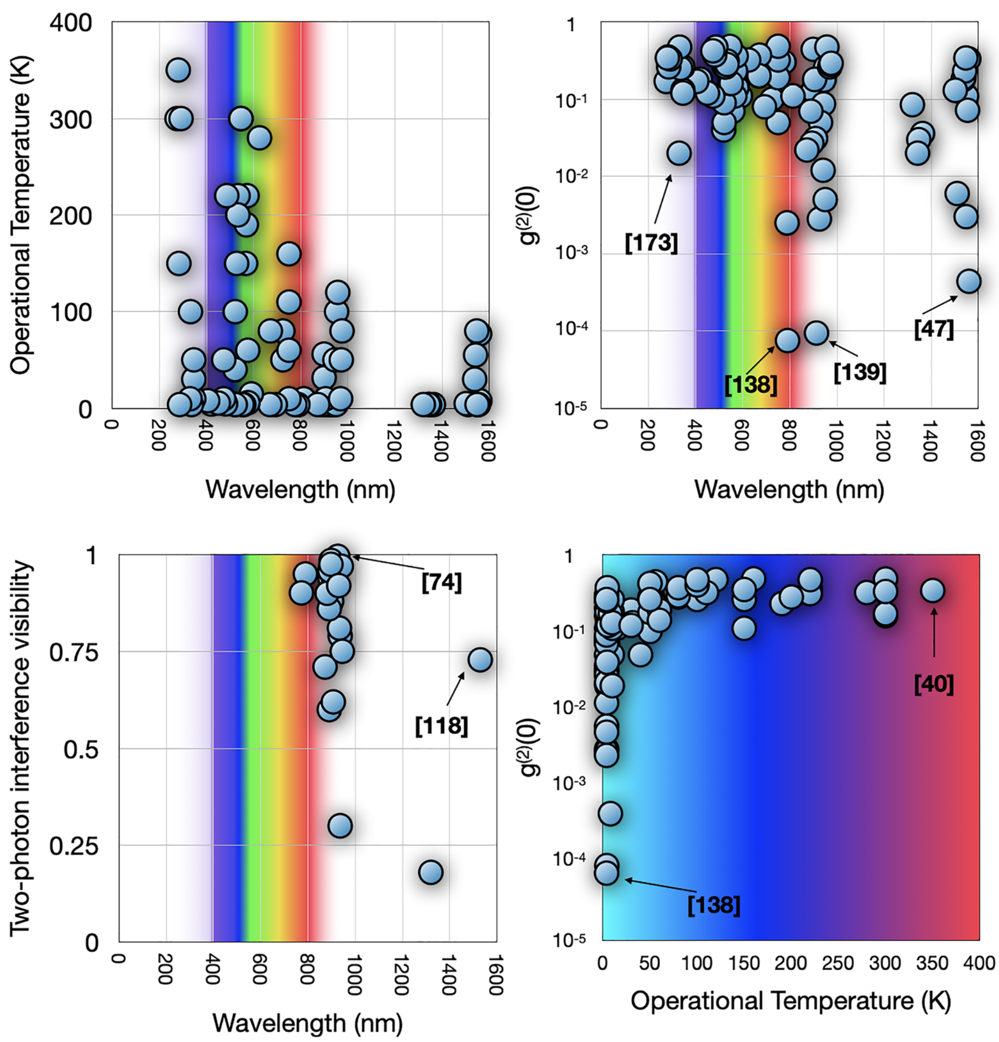

FIG. 9. Summary of epitaxially grown QD single photon emitters selected from the literature (as referenced in the main text).

necessarily invokes a large internal field of order MV cm^{-1} (Ref. 175) in the QDs, which leads to spatial separation of the electron and hole wavefunctions and a strong dependence of the emission lifetime and emission energy on the QD size: an intrinsic quantum confined Stark effect.[176,177] Indeed, emission lifetimes varying from 1000 ns to 0.5 ns, over a wavelength range of 496 nm to 275 nm, have been reported from size differences alone in *binary* GaN/AlN QDs.[178,179] However, the formation of GaN quantum dots in the Zincblende crystal phase is also possible,[180,181] resulting in relatively fast single photon emission. Using GaN QDs formed with this crystal structure, Kako *et al.* were able to demonstrate single photon emission at 336 nm with a $g^{(2)}(0)$ value of 0.25 and an emission lifetime of 360 ps at 4 K.[182]

Similar to the III-arsenides, III-nitride single photon emitting QDs can also be grown on silicon substrates[183] and have been shown to maintain their fine optical properties, exhibiting $g^{(2)}(0)$ values as low as 0.17 at 300 K.[184]

V. SUMMARY AND OUTLOOK

As can be seen in the previous sections, great progress is being made toward the development and control of epitaxially grown QD-based single photon emitters for quantum information processing applications. In Fig. 9, we present a summary of single photon purities and operational temperatures from the literature. Data points were taken from the articles discussed above and others.[20,38–40,47,52,62,74–76,86,110,111,113–117,119,120,130,133,136–139,142,144–146,157,159,163,164,166,173,174,182,185–203] The majority of advances in high quality single photon emitters have been made in the infra-red using III-As or III-P QDs, from which several examples of indistinguishability have been reported (with the exception of a few studies in the telecom bands, almost all experiments on photon indistinguishability have been performed using III-arsenide QDs emitting in the vicinity of 900 nm). The well-established growth and sample processing techniques have contributed significantly to this. Studies on QDs that emit in the visible and UV have shown high temperature operation, with the caveat of significantly reduced coherence due to phonon-related linewidth broadening (typical room-temperature emission linewidths are ~30 meV). Large biexciton binding energies,[147] which allow for the spectral isolation of individual emission peaks even in the presence of such large phonon-broadening, contribute to the high temperature operation of these devices. It should be noted, however, that the majority of studies of QDs operating in these wavelength regions are also on testing the properties of the chosen material system and are experiments to discover the limits of what is possible with different kinds of QDs.

To date, research has largely focused on the development of single photon emitting devices, and while this research is likely to continue for some time as we continue to develop new advanced materials and improved fabrication techniques, we have now reached the stage where groups are beginning to use their QD-based single photon emitters for experimental demonstrations of quantum technologies. Indeed, recent demonstrations of boson sampling,[137] metropolitan scale QKD,[108,112] quantum logic gate operation,[52,126] and quantum teleportation[118,127–129,204] are just some of the examples, leading the way for this exciting race to develop the necessary components of a full scale quantum network. Developments in photon frequency conversion techniques are also of increasing importance: recent demonstrations include the quantum downconversion of QD single photons and their subsequent interference with laser photons[205] and also the erasure of the frequency difference (and subsequent observation of two photon interference) between two remote emitters by frequency-conversion of the emitted photons from both emitters into the telecom C-band.[206] It is likely that as conversion techniques and their efficiencies improve, we may find ourselves in the situation where the material-determined nominal wavelength of the emitter is of little importance, and the quantum engineers of the future will be able to choose emitters depending on their required properties such as operational temperature and purity/indistinguishability. Another exciting area of increasing importance is the growing field of silicon quantum photonics,[207–209] whereby Si-based photonic circuits can be used for the generation of general quantum logic gates,[210] allowing for the realization of fully integrated on-chip QIP systems. Direct growth of QDs

TABLE I. Overview of properties of various epitaxially grown QD single photon emitters.

Material system	Wavelength range	Example $g^{(2)}(0)$ values	Operating temperature	Notes
InAs/InP	~1.5 μm	0.00044[47] ($T = 8$ K), 0.006[115] (4.2 K)	Up to 80 K[185] [$g^{(2)}(0) = 0.34$]	
InGaAs/GaAs	~900 nm–1.55 μm	0.012[52] 0.03[186]	Up to 120 K[189] [$g^{(2)}(0) = 0.47$]	$g^{(2)}(0) = 0.38$ at $\lambda = 1300$ nm[211]
GaAs/AlGAs	~800–1000 nm	0.000075[138]	Up to 80 K[188] [$g^{(2)}(0) = 0.28$]	
GaAs/GaAsP	~750 nm	0.08[190]	Up to 160 K[190] [$g^{(2)}(0) = 0.48$]	
InP/AlInGaP	~640 nm–690 nm	0.05,[212] 0.08,[193] 0.24[194]	Up to 50 K[212] up to 80 K[193]	
CdSe/ZnSe	~520 nm–570 nm	0.07[196]	Up to 220 K[196] [$g^{(2)}(0) = 0.36$] up to 300 K[39] [$g^{(2)}(0) = 0.48$]	
CdSe/ZnSSe	~550 nm	0.04[197] ($T = 4.5$ K), 0.16[198] ($T = 4$ K)	Up to 300 K[146] [$g^{(2)}(0) = 0.16$]	Electrically driven emission at $T = 200$ K[199]
CdTe/ZnTe	~550 nm	0.14[195] ($T = 6$ K)		
InGaN/GaN	~400–600 nm	0.11[157] ($T = 10$ K), 0.13[159] ($T = 5.6$ K)	Up to 280 K[163] [$g^{(2)}(0) = 0.32$]	Electrically driven emission at $T = 280$ K[163]
GaN/AlGaN (wz)	~280 nm–450 nm	0.02[173] ($T = 10$ K)	Up to 350 K[40] [$g^{(2)}(0) = 0.34$]	
GaN/AlGaN (zb)	~330 nm	0.25[182] ($T = 4$ K)	Up to 100 K [$g^{(2)}(0) = 0.47$]	

on Si substrates, or the post-fabrication coupling QDs into Si waveguides, are promising avenues for integrating QD-based single photon emitters into such quantum photonic circuits.

Finally, in Table I, we present a selection of emitter properties separated by the material system.

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DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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