Materials for Quantum Technology

PERSPECTIVE

The heat is on: towards the realization of non-cryogenic photonic quantum technologies

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Abstract

Solid-state devices capable of emitting single photons on demand are poised to allow the development of several photonic quantum technologies. Although high-performance devices have been reported in laboratory settings, the vast majority of experimental demonstrations performed to date have required the assistance of cryogenic cooling. In this perspective article we discuss the general progress and future challenges for the development of single photon emitters capable of operation at higher temperatures: negating the need for costly and cumbersome cryogenic cooling systems and their related vacuum requirements.

1. Introduction

Scientists around the globe are devoting considerable time and effort into realizing the hardware that will be used to power the quantum devices of the future. In particular, there has been a rapid increase in the volume of research into the development of solid-state single-photon emitters and their related technologies, such as photonic circuits and detectors. These devices are considered key enabling technologies for the realization of photonic quantum systems [1] and for the eventual development of a quantum internet [2, 3].

This intense research has directly resulted in the development of single-photon emitters based on a range of systems. Examples include the heavy attenuation of laser pulses, spontaneous parametric down conversion in non-linear crystals [4], and the isolation of single-quantum emitters such as isolated atoms/ions [5, 6], molecules [7], defects and colour centres in bulk crystal and 2D materials [8], and semiconductor quantum dots (QDs) [9]. Solid-state quantum emitters are of particular interest because the isolation of a single optical transition in such a structure guarantees the emission of a single photon, they can offer bright and stable emission, and in principle they can be also be contacted electrically.

Requirements on the properties of single-photon emitters vary depending on the application in mind [10], but in principle there are 3 main properties of such an emitter that must be optimized [11]: the *emission purity* [characterized by the second order intensity correlation function at time delay zero: $g^{(2)}(0)$], the *photon indistinguishability* (characterized by the visibility of 2-photon interference), and the *emission brightness* (which can be characterized generally as a rate at which photons can be extracted from the device, either under continuous wave excitation at emission saturation, or under pulsed excitation at a given repetition rate). Note that indistinguishability is not strictly required for some applications such quantum key distribution using the BB84 protocol [12].

There is no doubt that the best performing solid-state single-photon emitters to date, in terms of both indistinguishability [13] and emission purity [14], have been achieved using epitaxially-grown QDs that have been cryogenically cooled with liquid helium to temperatures of ∼4 K. Indeed, many would therefore argue that such cryogenic cooling is a necessity, both for the successful isolation of a single quantum emitter in the first place, and also for the successful suppression, or freezing-out, of the interactions between the emitter and its local environment (see below). However, in recent years, significant advances have also been made towards the realization/discovery of single-photon emitters that are operational at room temperature and above (or at least to temperatures accessible using thermoelectric cooling), and therefore essentially free from the constraints of

cryogenic cooling systems. Although device performance is still not at the level set by cryogenically-cooled state-of-the-art emitters, single-photon emission at high temperature has now been realized from a diverse range of structures, including isolated molecules of organic compounds [7], strongly interacting Rydberg atoms in vapour cells [15], optically-active isolated defects/color-centers/rare earth ions in a variety of crystals [16–24], defects in 2-D materials such as hexagonal boron nitride [25–27], nanostructures such as perovskite and colloidal/epitaxially-grown semiconductor QDs [28–36], carbon nanotubes [37, 38], and other nanostructures with more exotic geometries [39]. These different emitters all have particular emission wavelengths, which will allow for different applications. For example, room-temperature single-photon emission from defects in GaN [24], SiC [40, 41] and carbon nanotubes [38] has been realized at telecommunications wavelengths, which will allow compatibility with the existing telecommunications infrastructure, whereas emitters at shorter wavelengths may allow for direct interfacing with ion-based quantum memories.

In the following sections we discuss in general terms how the properties of high-temperature emitters compare to state-of-the-art low-temperature emitters, and discuss the possible future routes of development in the three key areas of purity, indistinguishability, and brightness.

2. Purity

Unfortunately, the reported single photon emission purities of high-temperature emitters are currently not as good as those reported from the best cryogenically-cooled devices, and are generally not yet at the level required for their realistic use in various quantum technologies such as optical quantum computers and quantum repeaters [10] (although other issues such as indistinguishability are currently more pressing concerns, as discussed in the next section). The highest purity single photon emission from solid state emitters has been achieved using cryogenic cooling of III-As QDs (and pulsed 2-photon resonant excitation of the biexcitonic resonance), leading to $g^{(2)}(0)$ values on the order of 10^{-5} [14]. In contrast, $g^{(2)}(0)$ values from all kinds of high-temperature emitters still tend to be several orders of magnitude higher: typically on the order of $10^{-2} \sim 10^{-1}$. For example, relatively pure room-temperature emission from a perovskite quantum dot has recently been reported with a $g^{(2)}(0)$ value of 0.05 [34], and emission from defects in carbon nanotubes has been reported with a $g^{(2)}(0)$ value of 0.01 [38].

In principle, however, these higher $g^{(2)}(0)$ values are not *intrinsically* limited by the choice of quantum emitter or indeed the ambient temperature, and are likely measured due to a combination of a finite temporal resolution of the experimental setups (in the case of continuous wave excitation) and imperfect experimental isolation of the emitter due to a residual overlapping spectral contamination from other emitting states. Such spectral contamination is exacerbated at high temperatures due to thermal broadening of the emission lines (see below), requiring stricter filtering than could be used at low temperatures. Future improvements in the purity of single photon emitters at high temperature can therefore be expected via a combination of improved spatial/spectral isolation of the emitter, and the deterministic fabrication of structures with low areal density. This could be achieved, for example, via site-controlled ion implantation for defect-based emitters [42, 43], or by the formation of site-controlled QDs that are uncontaminated by related wetting layer or quantum well emission. The combined use of optical cavities and spectral mode filtering could also be employed, and has been recently demonstrated experimentally to increase the emission purity from defects in diamond emitting at room temperature [44]. In addition to improved spatial and spectral isolation, temporal isolation can also be used to improve the emission purity further (with the caveat of reduced intensity), by gating the emission to filter out degenerate spectral contamination with differing time constants. Such methods have recently lead to the realization of $g^{(2)}(0)$ values <0.01 from colloidal QDs at room temperature [45]. It is likely that a combination of these techniques, along with improved pulsed optical excitation to suppress re-excitation of the emitting states, will be used to improve the emission purity of high temperature emitters in the near future.

3. Indistinguishability

Although high-temperature single photon emission with a reasonable purity has been achieved, quantum photonics applications such as optical quantum computing are usually based on photon interference, and thus require sources of *indistinguishable* single photons: individually emitted quanta must be indistinguishable to any experiment that negates the difference in their emission time and/or location. Indistinguishable photon emission is in principle achieved by having transform-limited emission, whereby the emission linewidth is determined by the Fourier transform of the emission lifetime, rather than additional processes such as homogeneous linewidth broadening (typically induced by fast phonon interactions), and inhomogeneous linewidth broadening induced by charge fluctuations in the emitter's local environment that occur on slower

timescales. Such an achievement is a tall order indeed for devices operating in a noisy high-temperature environment, where phonon interactions and charge noise can rapidly broaden the emission linewidth to values far exceeding its transform-limited value (often by several orders of magnitude). In particular, acoustic phonon interactions lead to a pure dephasing such that room-temperature emission linewidths can be broadened to reach values on the order of a few 10 s of meV (whereas purely lifetime limited linewidths could be on the order of $\sim$1 μ eV). Moreover, phonon-assisted recombination can result in broad phonon side bands that can dominate the emission spectra of various defect related emissions (widths vary wildly between various emitters, but can reach up to $\sim$200 meV for some NV centres in diamond). The situation is compounded by the charge noise in the fluctuating electronic environment, which can lead to a temporal fluctuation in the emission energy (spectral diffusion) that typically occurs on time scales varying from nanoseconds to milliseconds [46–53].

Whilst spectral diffusion may allow for the emission of a series of indistinguishable photons from an individual emitter on timescales faster than the fluctuations in the environment, it will prevent 2-photon interference between photons emitted from two, otherwise identical, spatially separate emitters. In principle, however, this charge noise can be suppressed by improving the material quality and cleaning up the electronic environment. Phonon-induced linewidth broadening, on the other hand, is a much more fundamental/difficult issue to overcome, and remains a significant challenge. However, recent research provides hope that there may indeed be indistinguishable light at the end of the proverbial tunnel.

One promising method to overcome broadening and increase the indistinguishability of photons from a given emitter is to couple the emitter with an optical cavity [54, 55]. The cavity can enhance emission into the cavity resonance via the Purcell effect, and simultaneously suppress emission in the phonon side-bands (which are off-resonance with the mode), in effect funneling the emission into a narrow spectral range of the cavity resonance. An additional benefit can be achieved by designing the cavity such that the emission is directed into a narrow solid angle, allowing for more efficient photon collection. The choice of the cavity parameters is crucial such that timing jitter in the emitted photons is limited, but the use of cascaded cavities has also been recently proposed in order to remove some stringent constraints on the cavity quality [56, 57]. In such a geometry the emitter is first coupled to an initial cavity with a small mode volume and modest Q-factor that enables efficient coupling of the emitted photons, which are then coupled into a second, higher quality cavity, to funnel the emission. Such cavity coupling schemes provide a clear path for developing high temperature indistinguishable photon sources using existing technology, although simultaneously maximizing indistinguishability and efficiency remains an issue.

Another possible path towards the generation of indistinguishable photons from high-temperature solid state systems may simply be the development of an emitter that does not couple to the acoustic phonons in the first place! Interestingly, researchers studying optically active defects in layered hexagonal boron nitride at room temperature have recently reported the observation of a remarkable decoupling of the emitters from low-frequency acoustic phonons [58]. The decoupling, which the authors claim could be due to the spatial geometry of the confined carrier wavefunctions between the layers of the hBN, allowed for the measurement of transform-limited linewidths of $\sim$0.25 μeV ($\sim$60 MHz) during photoluminescence excitation scans performed on timescales fast enough to negate spectral diffusion effects. However, it should be noted the emitters do still exhibit a pronounced spectral diffusion, leading to inhomogeneously broadened linewidths of $\sim$17 meV (4 THz).

The generation and measurement of indistinguishable photons from an emitter at high temperature must now be a high priority, for until high degrees of indistinguishability can be achieved, the use of high temperature single photon emitters will be limited to applications that do not require 2-photon interference, such as quantum key distribution and quantum random number generation.

4. Brightness

In terms of emission brightness, ideally we would like to evaluate and compare the flux of useable photons from the emitter that are be coupled into a fibre/waveguide or free space channel, or at least the rate collected by the first lens with a known transmission efficiency. However, direct comparisons between various reported emitters become difficult due to different reporting methods and experimental techniques. Typical differences include reporting photon count rates at varying stages of the setup, the use of different objective lenses with vastly different numerical apertures (NA), and different excitation techniques (particularly the differences between pulsed and continuous wave excitation: the former leading to photon emission rates ultimately limited to the laser repetition rate). However, to the extent that comparisons can be made (at a stretch, admittedly), it seems that high-temperature solid-state emitters are not too far behind their low-temperature brethren. For example, defects in diamond nanocrystals have been shown offer very bright room-temperature single photon emission. Indeed, under continuous wave excitation, detection rates of $\sim$3 MHz have been reported from silicon-vacancy

centres [59] (NA = 0.8), and rates of ∼3 MHz have also been detected from chromium related defects [60] (NA = 0.95). Similar photon count rates of ∼2 MHz have also been detected from defects in bulk SiC [19] (NA = 1.4) and 1.5 MHz from defects in bulk GaN [24] (NA = 1.35). However, and in fairness, these rates must be compared to the highest quality emitters at low-temperature (III-As QDs coupled to cavities or photonic structures to enhance the collection efficiency), which are typically measured using lower NA lenses (often due to limitations on the lens working distance, imposed by the cryostat), and under pulsed excitation (using Ti: sapphire lasers at ∼80 MHz). Typical photonic structure designs include nanowires to direct the emission [61], the coupling of QDs to nanophotonic waveguides [62, 63], planar 2D photonic crystal cavities [64], bullseye structures [65, 66], and cavities with distributed Bragg reflector mirrors [13, 67, 68]. Even under the restrictions of using lower NA lenses and pulsed excitation, the brightest of such emitters can still offer high photon count rates of, for example, ∼4 MHz (NA = 0.4) [69], 0.7 MHz (NA = 0.4) [70], 4.4 MHz (NA = 0.68) [66], and in some cases values exceeding 8 MHz [67] (including corrections for the detector dead time), whilst simultaneously maintaining high photon indistinguishability and purity. Recently, photon count rates of ∼13.7 MHz on fibre-coupled detectors (under π-pulse excitation at 76 MHz) were realized from cavity-coupled QDs, corresponding to an extraction efficiency *into the fibre* of ∼24% (using a lens with numerical aperture 0.65) [71]. Of course, due to the fact that the QD emission lifetimes are much lower than the excitation pulse intervals (∼12 ns), the photon emission rates of these low-temperature QDs could be increased trivially by pumping at higher repetition rates. It seems entirely plausible that such high values of brightness will also be achievable using emitters at high temperature, providing that suitable high-efficiency photon extraction can be developed.

Somewhat unfortunately, quantum dot based single photon emitters at high temperature have not yet reached the same levels of brightness. This can be ascribed to factors such as thermally activated non-radiative recombination that results in a reduced emission efficiency. However, even in the ideal case of no emission quenching at high temperature, increased linewidth-broadening will still reduce the spectral overlap between the emitter and any cavity [72], ultimately leading to a reduction in the photon extraction rate unless cavities with both large coupling strengths and broad resonances are used [54] (which are extremely challenging to experimentally realize for emitters with large linewidths). It is possible that high-efficiency and relatively broadband structures such as nanowires and bullseyes could be used for maintaining a high brightness at elevated temperatures (although this would result in limited indistinguishability, as discussed above). Indeed, we note that recent advances in the development of GaN QDs in nanowires have led to the observation of almost no reduction in integrated emission intensity between 4 K and 300 K [33, 73], and furthermore that bright emission at 300 K has recently been reported from GaN QDs in bulk [74], although optical systems and detectors used in the UV for this type of QD tend to be less efficient, and therefore currently limit the actual detected count rates to $\sim 10^4$ Hz (NA 0.55). The sub-nanosecond emission lifetimes of these QDs will in principle allow for operation at high repetition rates.

5. Summary

The future development of single photon emitters that operate at high temperature will involve improvements in all three of the areas discussed above. Whilst we can expect to see regular improvements in photon purity and emission brightness via brute-force incremental gains in the efficiencies of optical setups and optimization of enhanced photon extraction structures, more delicate (or fundamentally radical) work will be required to develop high temperature sources of indistinguishable photons. However, we firmly believe that the international research community is up to the challenge, and we anticipate a bright future for the development of non-cryogenic photonic quantum technologies.

Data availability

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

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