Position-Controlled Telecom Single Photon Emitters Operating at Elevated Temperatures

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ABSTRACT: A key resource in quantum-secured communication protocols are single photon emitters. For long-haul optical networks, it is imperative to use photons at wavelengths compatible with telecom single mode fibers. We demonstrate high purity single photon emission at 1.31 μm using deterministically positioned InP photonic waveguide nanowires containing single InAsP quantum dot-in-a-rod structures. At excitation rates that saturate the emission, we obtain a single photon collection efficiency at first lens of 27.6% and a probability of multiphoton emission of $g^{(2)}(0) = 0.021$. We have also evaluated the performance of the source as a function of temperature. Multiphoton emission probability increases with temperature with values of 0.11, 0.34, and 0.57 at 77, 220 and 300 K, respectively, which is attributed to an overlap of temperature-broadened excitonic emission lines. These results are a promising step toward scalably fabricating telecom single photon emitters that operate under relaxed cooling requirements.

KEYWORDS: nanowire quantum dot, photonic waveguide, selective-area vapor–liquid–solid epitaxy, telecom single photon source
are dictated by the core diameter \( D_c \sim 20 \text{ nm} \). The core was clad with an InP shell to produce a photonic waveguide targeting a base diameter of \( D_b = 310 \text{ nm} \) tapered to 20 nm over the 12 \( \mu \text{m} \) length of the waveguide. A schematic of the dot-in-a-rod structure is shown in Figure 1b and scanning electron microscopy images of the clad nanowire devices are shown in Figure 1a.

Figure 1. (a) Scanning electron microscopy image at 45° of an array of nanowires pitched at 7.5 \( \mu \text{m} \). Inset: close-up of a single nanowire. Scale bar is 1 \( \mu \text{m} \). (b) Schematic of a cutaway cross-section of the dot-in-a-rod structure. (c) PL spectra from 4 to 300 K of a single nanowire excited at \( P_{\text{sat}} \) with a CW laser. Inset: zoom-in of the PL spectrum at 4 K.

Figure 2. Low temperature (4 K) measurements: (a) Detected counts as a function of excitation power using 80 MHz (black) and 20 MHz (red) pulse rates showing a saturation count level of 1.86 Mcps and 0.507 Mcps, respectively. (b) Coincidence counts measured at 0.1 \( P_{\text{sat}} \) pumped at 20 MHz for which \( g^{(2)}(0) = 0.007 \). (c) Integrated counts in the \( g^{(2)}(\tau) \) zero-delay peak relative to side peaks as a function of excitation power at 20 MHz. (d) Zoom in of the correlation peak around \( \tau = 0 \text{ ns} \) as a function of excitation power showing evidence of re-excitation.
Temperature-dependent optical measurements were made with the device in a closed-cycle helium cryostat. The nanowire was excited through a 100x cryogenic objective (numerical aperture NA = 0.81) located in the cryostat. Spectrally resolved photoluminescence (PL) measurements were performed using a continuous-wave (CW) laser while for time-resolved photoluminescence (TRPL) measurements, a pulsed laser operating at 10 to 80 MHz was used. In both case, excitation was above-band at $\lambda = 670$ nm. The PL was collected through the same objective and directed to a spectrometer with a liquid nitrogen-cooled InGaAs detector for the spectrally resolved measurements. For TRPL measurements, the emission was coupled to a fiber and sent to two superconducting nanowire single photon detectors (SNSPDs) via a 50/50 beamsplitter. For the latter, the emission line to be measured was isolated using a filter with a bandwidth appropriate for the measurement, discussed below.

Typical PL spectra as a function of temperature from a device emitting around 1300 nm are shown in Figure 1b at an excitation power $P = P_{sat}$ which corresponds to the power required to saturate the excitonic transition. At 4 K the PL spectrum is characteristically dominated by a narrow single peak with a full width half-maximum value of 44 $\mu$eV, limited by the resolution of the spectrometer. The peak is attributed to an excitonic transition within the quantum dot and, with increasing temperature, shifts continuously to longer wavelength, starting at 1301.28 nm at 4 K and reaching 1397.8 nm at 300 K. As the temperature is increased the predominant peak broadens and starts to overlap with the adjacent higher energy transition line that rises up, becoming one single broad peak above 150 K. Above 100 K a broad short wavelength peak associated with p-shell transitions can also be seen. It should be noted that emission from a single quantum dot can be readily observed at room temperature.

Exciting at 80 MHz we measured, at saturation, 1.86 Mcps on the SNSPDs (3.8 Mcps under CW excitation) after filtering with a 0.1 nm filter. To facilitate the discrimination between coincidence and background counts in the correlation measurements discussed later, we have also made measurements at lower repetition rates. For example, using 20 MHz we measure count rates at saturation of 0.507 Mcps, approximately equal to the expected 4-fold reduction compared to 80 MHz. Integrated count rates versus excitation rate for both laser repetition rates are shown in Figure 2a. The end-to-end source efficiency, which is defined by dividing the maximum collected count rates at the SNSPDs by the repetition rate of the pumping laser (20 MHz was used) is thus 2.54% at saturation. If we account for the detector efficiency of 90%, the end-to-end efficiency is 2.82% and by taking into account a throughput of 10% for the experimental setup a collection efficiency at the first-lens of 28.2% is obtained. This value is reduced to 27.6% after correcting for the multiphoton emission probability measured at the above count rate (see below). The efficiency increases to 34.5% if we include photons emitted into the phonon sidebands, estimated to be 20% of the total emission at 4 K, which we have filtered out using the narrow passband filter. In principle the efficiency can be nearly doubled using a mirror (Au layer) at the bottom of the nanowire to allow collection of the photons that are emitted toward the InP substrate. A theoretical model reflectivity of about 0.91 can be achieved using a gold layer under the nanowire base.

To assess the single-photon purity, second-order correlation measurements, $g^{(2)}(\tau)$, were performed in a standard Hanbury–Brown and Twiss experiment. As mentioned above, a pulse

![Figure 3. Second-order correlation measurements of the source at different temperatures. The excitation power was 0.5 $P_{sat}$ at temperatures up to 120 K and reduced to $\sim 0.25 P_{sat}$ at 220 K and above. The red line is a curve fit to the data and the blue dashed line is the fitted background count level. A separate measurement of the lifetime was made to allow the background level to be determined.](https://doi.org/10.1021/acs.nanolett.2c04375)
repetition rate of 20 MHz was used to avoid significant overlap between consecutive pulses and a 0.1 nm bandpass filter was used to isolate the emission from a single transition. A typical correlation measurement is shown in Figure 2b when the source is excited at ~0.1 $P_{sat}$. We fit the $g^{(2)}(\tau)$ curves using the expression $BG + A[g^{(2)}(0)\exp(-|\tau|/\tau_d) + \sum_{n\neq 0}\exp(-|\tau| + nT/\tau)]$, where BG is the background counts, $A$ is the normalization constant, $\tau_d$ is the time delay, $\tau_i$ is the measured lifetime, and $T$ is the pulse period. The integrated counts in the zero-delay peak relative to the side peaks gives a probability of multiphoton emission of $g^{(2)}(0) = 0.007$. As the pump power is increased the multiphoton probability increases, as shown in Figure 2c, with a value $g^{(2)}(0) = 0.021$ at saturation. The reason for this increase is the presence of re-excitation of the dot due to the use of above band excitation, with a characteristic dip in the center of the peak observed around zero delay, \cite{24,41}. A behavior is a consequence of an excess of carriers in the barrier material at high pump powers that can be captured by the dot after the first photon emission event has occurred, leading to the re-excitation of the dot. The rise time from 0 ns is associated with the carrier capture and subsequent relaxation within the dot, while the decay time is associated with the lifetime of the exciton.

Next we assess the temperature dependence of the single-photon purity through coincidence measurements from 4 to 300 K. In Figure 3 we show correlations at four selected temperatures that span the measured range. We have adjusted the pulse excitation rate between 10 and 40 MHz depending on temperature in order to minimize the overlap between adjacent correlation peaks while maintaining sufficient count rates to limit excessive integration times. The increased peak-to-peak overlap and decreased count rates with increasing temperature is discussed below.

We first note that as the temperature was increased the emission lines broadened and started to overlap, as shown in Figure 1b. Importantly, the line width broadening with increasing temperature will have two effects on the second-order correlation measurements: First the use of a narrow pass band filter will reduce the count rates arriving at the SNSPDs. To maintain reasonable count rates for the measurements, the bandpass of the filter employed was widened as the temperature was raised, from 0.1 to 12 nm at 175 K and to 25 nm at 300 K. Second, the overlap between the adjacent optical emission lines will make the selection of a single transition difficult. Both of the above can potentially result in photons from different transitions reaching the detectors and affecting the $g^{(2)}(0)$ value. If two transitions are not the result of a sequential decay in a cascade process, for example, a neutral and charged exciton, then $g^{(2)}(0)$ can remain zero. On the other hand if the two transitions correspond to a cascade process, such as for the biexciton and exciton, then the value of $g^{(2)}(0)$ will rise.

To obtain accurate values of the coincidence counts in the zero delay peak, the curve fitting procedure described above (which included an uncorrelated background signal) was applied to the measured $g^{(2)}(\tau)$ curves. To enable accurate fitting, a TRPL measurement was made before each correlation measurement to determine the lifetime to be used for each fit. Without this extra piece of information a wide range of background levels and lifetimes could be used to fit the same data, in particular at high temperature. The fits are plotted in Figure 3 (red curves) and the extracted $g^{(2)}(0)$ values are plotted in Figure 5 (red stars).

Before discussing the observed $g^{(2)}(0)$ dependence on temperature, we first look at the lifetime dependence extracted from the TRPL measurements, plotted in Figure 4a. We observed a dramatic increase in lifetime with increasing temperature, from 2.1 ns at 4 K to 10.8 ns at 300 K. We attribute this increase in part to a dependence of the spontaneous emission rate into the detected waveguide mode, HE_{11s}, on the emission wavelength.\cite{24} In Figure 4b, we plot the lifetime as a function of emission wavelength which redshifts with increasing temperature as shown in Figure 1c. In the inset, we show the calculated wavelength-dependent spontaneous emission rate normalized to that for bulk, as a function of wavelength for three different nanowire diameters. For the nanowire diameters used here the shift in emission wavelength from 1300 to 1400 nm, as seen when going from 4 to 300 K, results in a drop in emission rate or equivalently an increase in lifetime (shaded region in inset).

This alone is insufficient to account for the observed increase and we consider a second mechanism proposed in ref 42, to describe the increase in lifetime with temperature observed in InAs/GaAs quantum dots for temperatures up to 250 K. The authors attributed the increase to Boltzmann spreading over dark states, i.e. thermal excitation of electrons and holes out of the ground state into higher lying states that do not have allowed optical transitions. Based on the s-p level spacing of 60 meV in our dot, this mechanism alone underestimates the increase in lifetime observed. If, however, we include both of the above mechanisms we can reach qualitative agreement with the observed lifetimes.
We consider now the temperature dependence of $g^{(2)}(0)$ plotted in Figure 5 that shows a steady rise as the temperature is increased from 10 to 120 K. Above 120 K it saturates at just under 0.5 until 300 K where it is just over 0.5. A measured nonzero $g^{(2)}(0)$ for a quantum dot can arise from a number of mechanisms; detector dark counts, scattered excitation laser, multiple independent emitters, re-excitation of the dot from the same excitation pulse, and multiple emission lines from a single dot. The increase in $g^{(2)}(0)$ with increasing temperature is clearly not a consequence of dark counts (which would be a uniform background), or scattered laser emission (which would be a narrower peak with a width limited by the detector timing jitter of 60 ps). Multiple independent emitters are also very unlikely since the nanowire can contain one and only one dot by design and has a very low $g^{(2)}(0)$ at low temperature. This leaves re-excitation and multiple emission events from the dot as the likely causes. Re-excitation of the dot typically results in a dip in the $g^{(2)}(\tau)$ value around zero delay, which is not observed here, although the time scale for re-excitation at higher temperatures may be too short to resolve.

Multiple emission events from the dot, such as bieexciton followed by exciton emission are also quite likely. At low temperature these transitions can be separated spectrally. With increasing temperature these transitions broaden and eventually overlap so they can no longer be isolated, increasing $g^{(2)}(0)$. By dropping the excitation power at high temperatures the $g^{(2)}(0)$ should improve due to the decreased probability of creating a bieexciton, and indeed this is observed, but at the expense of overall count rates. Considering the apparent saturation $g^{(2)}(0)$ at a value of ~0.5 (i.e., two photons per excitation pulse) observed in Figure 5, it is likely that this last mechanism, involving only a single additional transition, dominates the observed temperature dependence.

For comparison, we also plot literature values of $g^{(2)}(0)$ from other quantum dot-based single photon sources emitting at telecom wavelengths. Only results obtained from nonpost-selected measurements (i.e., using pulsed excitation) are included. Measurements where the excitation power was specified to be at or close to $P_{\text{sat}}$ as is the case here, are indicated by an asterisk in the legend. This applies strictly to the low temperature measurements since at higher temperatures it is difficult to observe saturation of a single transition when it overlaps with another.

We first consider experiments performed using above-band excitation, as is the case here, indicated in the figure by filled symbols. The sources in this study, under these operating conditions, outperform, to our knowledge, all other existing approaches based on quantum dots. Next we consider experiments performed using quasi-resonant excitation (i.e., excitation via a p-shell level in the dot), indicated in the figure by open symbols. Here we observe several sources that display reduced multiphoton emission probabilities compared to the devices in this study. In approaches utilizing randomly nucleated quantum dots such that multiple emitters may be simultaneous probed, quasi-resonant excitation will mitigate both spectral pollution from other emitters as well as re-excitation from the same emitter. For the sources studied here, which are fabricated using a site-selection technique that assures each device contains only one emitter, quasi-resonant excitation is also expected to improve the single photon purity by mitigating re-excitation process responsible for the nonzero $g^{(2)}(0)$ values, specifically at lower temperatures (see Figure 2d).

For completion, we also include other solid-state 2-level systems that have demonstrated telecom single photon emission, specifically at room temperature, indicated in the figure by crosses. These include sources based on defects in SiC, GaN, and carbon nanotubes. Although the nanowire-based sources described here technically generate nonclassical light up to temperatures of 220 K, devices with such high multiphoton emission probabilities are of limited practical application. To achieve high temperature operation with reduced multiphoton emission probability to levels comparable to these defect-based systems will require engineering of the quantum dot electronic levels such that they are sufficiently separated when severely broadened to eliminate any overlap between them.

In conclusion, we have demonstrated high purity telecom single photon emission from devices operating with high efficiency and grown using a position-control technique. We have also evaluated the temperature-dependent performance of the sources, quantifying the degradation of single photon purity with temperature. Finally, we note that the structures described can be incorporated in a hybrid on-chip platform to provide a stable and robust plug and play field-ready source that will be required to scalably build a future telecom quantum network.

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