A high-rate source for single photons in a pure quantum state

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Abstract. We report on the efficient generation of single photons, making use of spontaneous Raman scattering in a single trapped ion. The photons are collected through in-vacuum high-numerical-aperture objectives. Photon frequency, polarization and temporal shape are controlled through appropriate laser parameters, allowing for photons in a pure quantum state. These photons are suitable heralds for single-photon absorption in a single-ion quantum memory.

Quantum networks allow for quantum communication between distant locations (nodes) where local quantum information processing is carried out. The key ingredient for this kind of network architecture is to establish entanglement between network nodes [1]. Its scalability is facilitated by employing quantum repeaters [2, 3].

Different platforms are pursued for implementing quantum networks, and various schemes exist to generate entanglement between their nodes. In the field of trapped single atoms and ions as nodes [4], one approach is to use pairs of entangled photons, split them up and have them absorbed by two separate atoms, thus transferring the photonic entanglement onto

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the atoms [5]. Alternatively, each atom is first entangled with a single photon through an emission process [6–8]. The photons are then brought to interference on a beam splitter, and a measurement is performed that projects the atoms into an entangled state [9–11]. A hybrid version of these two approaches is to have one atom emit a single photon (with which it is entangled) that is absorbed by the other, thereby establishing entanglement between the atoms [12]. All these schemes are probabilistic, but if there is a suitable process heralding the successful creation of entanglement, subsequent information processing operations can be performed deterministically.

For these schemes to succeed in practice, a high rate of generated and collected single photons in a well-defined quantum state is necessary. This requires their spectral–temporal structure to be at the Fourier limit, their polarization to be pure and their spatial shape to be single mode. Two main approaches to achieve this have been followed so far. One is the use of optical cavities strongly coupled to the atomic systems and to optical fibers (operating as quantum channels) [8, 13–16]; this allows for efficient exchange of light between atoms and single-mode fibers. Alternatively, free-space coupling employing high-numerical-aperture (NA) objectives is pursued [17–20], trading better fiber coupling for higher rates.

Trapped ions are a particularly promising physical platform for quantum networks, as they offer the integration of atom–photon interfaces with local multi-qubit information processing. In this context, cavity-enhanced single-photon generation has been explored in various experiments [8, 14]. There is a limitation, however, to the achievable enhancement or generation rate in this system, since the size of a cavity around an ion trap cannot be reduced easily below the mm-scale. Using the alternative free-space coupling approach, we have previously implemented various experimental building blocks of trapped-ion-based quantum networks. Pairs of entangled photons from spontaneous parametric down-conversion were split up and one of the photons of a pair was absorbed by a single ion [21, 22]; temporal, frequency and polarization correlations between the absorption process and the partner photon showed that this is a promising approach to create entanglement between remote quantum systems. We also demonstrated the generation of single photons tunable in bandwidth and temporal shape [23]. The individual and mutual coherence properties of photons emitted by single ions in two spatially separated traps were investigated through two-photon interference; coherence within a factor of two from the Fourier limit was attained, limited by the generation of a mixed frequency state. Here we demonstrate additional engineering of the atomic quantum state which enables generation of single photons in a pure state of both polarization and frequency, without the need for optical filtering elements. Moreover, we achieve an increased rate of fiber-coupled photons as well as higher tunability of the photon wave packets compared to our previous work. Owing to our free-space approach and atomic state manipulation techniques, we report the highest rate of pure-state photons obtained so far with ion-based quantum technology. These photons are also suitable heralds for single-photon absorption in an ion-based quantum memory.

Our experimental setup is sketched in figure 1. Single $^{40}\text{Ca}^+$ ions are trapped in a linear radio-frequency (Paul) trap and Doppler cooled by frequency-stabilized diode lasers at 397 and 866 nm wavelength [24]. Additional laser beams at 850 and 854 nm are used for state preparation and single-photon triggering. This is facilitated by a magnetic field, also defining a quantization axis. The ion is positioned between two high-NA laser objectives (HALOs, NA = 0.4) for fluorescence collection and laser focusing, each covering $\sim$4.2% of the total solid angle [25]. Blue fluorescence light (at either 397 or 393 nm) is coupled to optical fibers and detected by photomultiplier tubes (PMTs) with $\sim$28% quantum efficiency. Their output pulses
Figure 1. Experimental setup. HALO: high-NA laser objective; PMT: photomultiplier tube; QWP: quarter-wave plate; PBS: polarizing beam splitter; $\vec{B}$: magnetic-field direction. The ion is trapped between the HALOs.

Figure 2. Top: $^{40}$Ca$^+$ level scheme including Zeeman substates and transition wavelengths. Bottom: sequence of laser pulses; in the cooling phase, the ion is Doppler cooled by lasers at 397 and 866 nm. For state preparation, an additional laser at 850 nm is switched on, transferring the population to the $D_{5/2}$ state. After switching off the three lasers, the 854 nm transition is driven, releasing a single photon at 393 nm wavelength.

are time-tagged and stored for later processing. Wave plates are used to adjust the polarizations of the laser beams and to analyze the polarization of the fluorescence.

The experimental sequence is outlined in figure 2. First, the ion is Doppler cooled by the 397 and 866 nm lasers for 600 ns. Then the 850 nm laser is switched on for 2 $\mu$s, transferring the ion to the long-lived $D_{5/2}$ state. Here we make use of a three-photon resonance excited
by the three lasers, coupling the $S_{1/2}$ ground state directly to the $P_{3/2}$ excited state, i.e. with minimized population of the $P_{1/2}$ and $D_{3/2}$ levels. This brings the rate with which population is transferred to the $D_{5/2}$ state close to the limit set by the lifetime and branching ratio of the $P_{3/2}$ state. Subsequently, the 854 nm trigger laser is switched on for a variable time (800 ns–35 µs) while all the other lasers are off, driving the $D_{5/2}$–$P_{3/2}$ transition and releasing a single Raman-scattered photon at 393 nm wavelength.

For several values of 854 nm laser power, a histogram of arrival times of the single 393 nm photons is displayed in figure 3(a), reflecting control of the temporal shape of the photonic wave packet. The higher the laser power, the faster the $D_{5/2}$ state is depopulated and hence the shorter is the wave packet. The inset shows a lifetime-limited photon of $\sim$14 ns length, twice as long as the decay time of the $P_{3/2}$ excited state. As the 854 nm laser beam is focused onto the ion through one of the HALOs, an optical power of 3.1 µW is sufficient. On the other hand, the longest photon created has a duration of 1.03 µs at 13 nW power of 854 nm.

To assess the single-photon character of the scattered light, we measure the second-order time correlation ($g^{(2)}$) function (figure 3(b)). 393 nm light is collected from both HALOs and detected with individual PMTs. This is equivalent to a Hanbury–Brown–Twiss setup, where the ion itself plays the role of the beam splitter [26]. The detected photon arrival times on the two PMTs are then correlated. For a perfect single-photon source, we do not expect any coincidences at zero time delay. From the measured residual counts, we deduce a multi-photon to single-photon ratio for light emitted by the ion of 1.34(63)% , corrected for accidental coincidences caused by stray light and detector dark counts. This small amount of multi-photon events is attributed to the acousto-optic modulators switching the lasers in the state preparation phase. If a single photon is emitted while these lasers are not yet fully switched off, a non-zero probability to re-excite the ion to the $P_{3/2}$ state arises, sometimes resulting in a second photon.

The highest repetition rate in these experiments is 230 kHz, given by optimizing the state preparation time for the highest rate of generated photons. Together with a total detection
Figure 4. (a) Depopulation rate (inverse photon duration) of the D$_{5/2}$ state as a function of the 854 nm quarter-wave plate angle for non-saturating 854 nm laser power during the photon generation phase. (b) Number of photon detection events as a function of the 393 nm quarter-wave plate angle for two different D$_{5/2}$ Zeeman states ($|m = \pm \frac{5}{2}\rangle$). Error bars are smaller than the size of symbols.

efficiency of 1.18% from both HALOs (including the PMT quantum efficiencies and using multi-mode optical fibers), we obtain a rate of fiber-coupled single photons of 5.46 kHz (1.53 kHz detected). For single-mode fiber coupling, these rates are multiplied by 25–30%.

In the results shown so far, the ion was prepared in a statistical mixture of all six Zeeman states of the D$_{5/2}$ manifold. Repumping back to the S$_{1/2}$ ground state then happens through a multitude of Raman transitions, resulting in a single photon in a mixed polarization and frequency state. In schemes involving two-photon interference, this limits the indistinguishability of the two photons [23]. Therefore, we implemented an excitation scheme that populates only one specific D$_{5/2}$ Zeeman sub-level. This is achieved by using an additional 854 nm laser beam during state preparation, entering the trap at 45° with respect to the quantization axis. Its elliptical polarization is tailored such that only $\pi$ and $\sigma^+$ transitions are driven, effectively pumping all population to the $|D_{5/2}, m = +\frac{5}{2}\rangle$ state. After preparation of this pure atomic state, the 854 nm trigger laser is turned on to drive a Raman transition via the $|P_{3/2}, m = +\frac{3}{2}\rangle$ state to the $|S_{1/2}, m = +\frac{1}{2}\rangle$ ground state, thereby generating a single 393 nm photon in a pure polarization ($|\sigma^+\rangle$) and frequency state. By adjusting the laser polarizations appropriately, we are also able to generate single photons in the $|\sigma^-\rangle$ polarization state. Hence, the additional atomic state engineering removed the main limitation to creating Fourier-limited photons that we experienced in our previous experiment [23]. Residual incoherence of the photons is now mainly due to the 6% decay probability from P$_{3/2}$ back to D$_{5/2}$ during their generation.

To verify the preparation of a single D$_{5/2}$ Zeeman sub-level, we change the polarization of the trigger laser by rotating the quarter-wave plate in front of the HALO through which the light is focused onto the ion. As the HALO optical axis is also the quantization axis, we smoothly change between driving $\sigma^+$ and $\sigma^-$ transitions (and superpositions of the two) in the ion. Figure 4(a) shows the rate at which the D$_{5/2}$ state is depopulated during the photon generation phase as a function of the quarter-wave plate angle. The clear sinusoidal behavior with a visibility compatible with unity confirms that only one specific $\sigma$ transition can be driven.
This would still be the case if the ion were in one of the two outer Zeeman states of \( D_{5/2}, |m = +\frac{5}{2}\rangle \) or \( |m = +\frac{3}{2}\rangle \). However, considering also that the pump beam enters at 45° during the state preparation, we conclude that only one specific \( D_{5/2} \) Zeeman state (\( |m = +\frac{5}{2}\rangle \)) is populated.

In order to analyze the polarization of the emitted single photons, we place a 393 nm quarter-wave plate behind one of the HALOs, followed by a polarizing beam-splitter cube and a PMT. The number of photon detection events for different wave-plate angles is shown in figure 4(b). The two data sets correspond to the two possible \( D_{5/2}, m = \pm \frac{5}{2} \) states that we prepare. A mean visibility of \( V = 90.5(6)% \) indicates an almost pure circular photon polarization, emitted through the transitions \( |D_{5/2}, m = \pm \frac{5}{2}\rangle \rightarrow |P_{3/2}, m = \pm \frac{1}{2}\rangle \rightarrow |S_{1/2}, m = \pm \frac{1}{2}\rangle \), respectively. The non-unity visibility is caused by the non-perfect 393 nm quarter-wave plate. The pure \( |\sigma\rangle \) polarization is corroborated by an increase of the total detection efficiency from 1.18 to 1.55%, as light from \( \sigma \) transitions is preferentially emitted along the quantization axis (which coincides with the optical axis). Thus, we have verified that we generated single photons in a pure polarization and frequency state.

The duration of the atomic state-preparation step was set to \( \sim 100 \mu s \), thereby reducing the repetition rate from 230 to 10 kHz. Combined with the increased collection efficiency, we reach a rate of fiber-coupled pure-state photons of 540 Hz. This rate compares favorably to ion-based cavity experiments (e.g. 95 Hz in [8]). Shortening of the preparation phase from \( \sim 100 \mu s \) to below 50 \( \mu s \) will be possible without compromising the purity of the photonic state and will allow for pure-state photons at \( >1 \) kHz rate.

In conclusion, we generated single photons from a single ion at 5.5 kHz fiber-coupled rate with very small multi-photon contribution. The temporal shape of the photonic wave packet was tuned between 14 ns and 1 \( \mu s \) by appropriate laser parameters. Importantly in the context of quantum networks, the ion was made to emit photons of single frequency and polarization at \( >0.5 \) kHz fiber-coupled rate. For the future, we aim at realizing a quantum memory that stores the polarization of a photon absorbed on the 854 nm transition in the \( S_{1/2} \) Zeeman substates. One possible scheme is to first prepare the ion in a coherent superposition of the two \( |D_{5/2}, m = \pm \frac{5}{2}\rangle \) states [27, 28]. The absorption of an 854 nm photon will then transfer the photonic state into the ion’s ground state. Successful state transfer is heralded by the detection of the emitted 393 nm photon in a linear polarization basis. Here we have shown that this heralding process is successful with up to 1.55% probability.

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