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Doubly heralded single-photon absorption by a single atom

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Abstract We report on a single-photon-to-single-atom interface, where a single photon generated by spontaneous parametric down-conversion (SPDC) is absorbed by a single trapped ion. The photon is heralded by its time-correlated partner generated in the SPDC process, while the absorption event is heralded by a single photon emitted in its course. Coincidence detection marks doubly heralded absorption, enabling photon-to-atom quantum state transfer (Kurz et al. in Nat Commun 5:5527, 2014; Müller and Eschner in Appl Phys B 114:303, 2013). Background in the coincidence measurement is strongly suppressed by a new method that discriminates real absorption events from dark-count-induced coincidences.

1 Introduction

A major goal in quantum technologies is to integrate quantum communication and information processing into quantum networks [1]. One promising approach is using single atoms and photons as nodes and channels of the network [2–4]. Single photons are able to transmit quantum information between different nodes of the network, while single atoms serve as high-fidelity, long-storage-time memories; single trapped ions, in particular, may additionally be used as quantum processors [5–8]. In this context, the use of heralding photons for repeaters and memories schemes is very promising [9, 10]: Successful entanglement transfer or information storage onto a single atom is signaled by the detection of a photonic herald emitted as a consequence of the process. The detection of such heralds permits one to discriminate successful events and thereby enables high-fidelity protocols, e.g., for photon-to-atom quantum state transfer [11, 12, 13] or distant ion entanglement [9].

Previously, we have demonstrated the absorption of single SPDC photons in a single ion, signaled by the onset of fluorescence, i.e., by a quantum jump [14]: Absorption of a photon excites the ion out of a metastable state into a fluorescence cycle, and a large number of emitted photons herald the absorption event with near 100 % efficiency. This method has also been used to herald single-photon interaction between two distant ions [4] and to manifest the polarization entanglement of a SPDC photon pair in single-photon absorption [15]. By generating fluorescence, however, the state of the ion after the single-photon absorption is not preserved, and transferred quantum information is lost. This limitation is overcome when a single scattered photon is used to herald the absorption process [16, 12, 13, 17, 18]. In [11], we implemented a specific protocol based on such heralding that enabled high-fidelity transfer of a photon polarization onto an atomic qubit and that may also be used for photon-to-atom entanglement mapping [18]. While the experimental demonstrations so far [11, 13] were carried out with laser photons, here we extend them to the absorption of single photons from an SPDC source, thus proving the feasibility of photon-to-atom quantum state conversion with our interface.

The principle of our single-photon-to-single-atom interface is as follows: A single trapped ion is exposed to a single resonant photon, generated via SPDC and heralded by its partner; successful absorption is heralded by the detection of a single Raman scattered photon released as a

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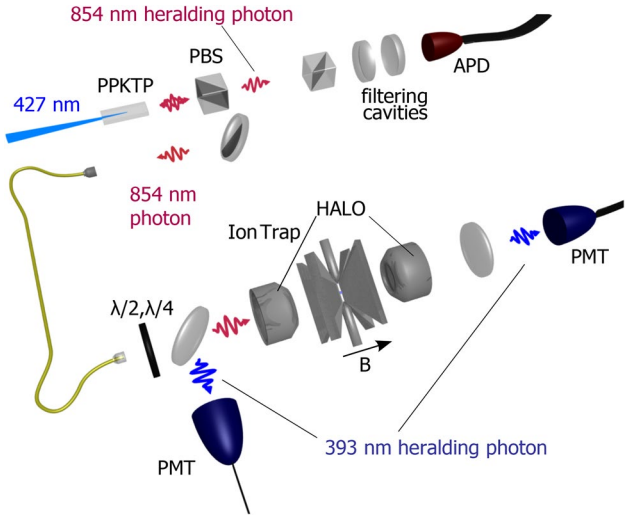

Fig. 1 Experimental setup. The *top part* shows the SPDC source of resonant, narrowband, heralded single photons; the *lower part* shows the ion trap setup consisting of a linear Paul trap with two high numerical aperture laser objectives (HALOs). A single-mode optical fiber connects the two setups. Further details are given in the text

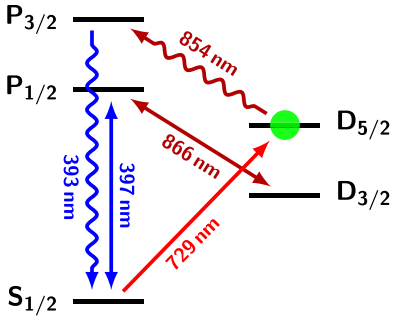

Fig. 2 Relevant levels and transitions of the $^{40}Ca^{+}$ ion. Lasers at 397 and 866 nm are used for cooling and fluorescence detection. Lasers at 729 and 854 nm are used for optical pumping and state preparation. The single ion is prepared in the $D_{5/2}$ metastable state for absorption of 854 nm SPDC photons. Absorption heralds are emitted at 393 nm

result of the absorption. Coincident detection of the partner SPDC photon and the photon emitted by the ion marks doubly heralded single-photon absorption by a single atom. Furthermore, we present a method for discriminating real absorption events from dark-count-induced coincidences, tackling the main source of background.

2 Experimental setup

The experimental apparatus, sketched in Fig. 1, consists of the SPDC source setup [19, 20] and the ion trap setup and is similar to the one used in [14, 15]. Degenerate photon pairs at 854 nm are created collinearly by spontaneous parametric down-conversion in a PPKTP (periodically poled potassium titanyl phosphate) nonlinear crystal. The crystal is pumped with a continuous laser beam at 427 nm obtained by second harmonic generation of an amplified 854-nm diode laser (Toptica, TA-SHG pro), which is stabilized to the $D_{5/2} \rightarrow P_{3/2}$ transition in the $^{40}Ca^{+}$ ion via a transfer lock [21]. The two photons of a pair have perpendicular polarizations due to type II phase matching. Their polarization entanglement is projected out when they are split by a polarizing beam splitter (PBS), in order to use them as a heralded single-photon source. The vertically polarized photons are coupled into a single-mode fiber and transmitted to the ion trap setup for absorption; their polarization at the ion is adjusted using waveplates ($\lambda/4$, $\lambda/2$). Utilizing the time correlation of the SPDC pair, the horizontally polarized photons are used to signal the presence of their partners. They are filtered by two cascaded Fabry-Pérot cavities, which only transmit the 22-MHz bandwidth of the $D_{5/2} \rightarrow P_{3/2}$ transition, and which are actively stabilized to resonance. Transmitted photons are detected with an avalanche photodiode (APD, Perkin-Elmer, ~30 % efficiency). This selects only ion-resonant photons from the broadband (~200 GHz) SPDC emission. More details are explained in Refs. [19, 20].

The linear Paul-type ion trap holds a single $^{40}Ca^{+}$ ion, whose relevant levels and transitions are shown in Fig. 2. The ion is initially prepared in the $D_{5/2}$ level, from where it can be excited to $P_{3/2}$ by absorption of a single 854-nm photon. Following absorption, it decays to the $S_{1/2}$ ground state with 93.5 % probability (according to the branching ratio of the $P_{3/2}$ state), thereby releasing a single photon at 393 nm. If detected, this photon heralds the single-photon absorption. For efficient ion-photon coupling, two in-vacuum high numerical aperture laser objectives (HALOs, NA = 0.4) are placed along one of the radial trap axes, which coincides with the quantization axis (magnetic field direction, **B** in Fig. 1). One of the HALOs is used to focus the 854-nm SPDC photons onto the ion, while both of them collect the 393-nm absorption heralds, which are extracted with dichroic mirrors and detected by two photomultiplier tubes (PMTs, Hamamatsu). Including collection and detection efficiency, 1.86 % of the emitted heralds are finally detected. The ion is cooled, pumped and prepared for absorption by several lasers as described in Fig. 2.

3 Experimental sequence

The experimental sequence runs at 401 Hz repetition rate and consists of several phases. First, Doppler cooling is applied for 100 μs using 397 nm and 866 nm laser light, which brings the ion to the Lamb-Dicke limit. After

optical pumping to the $|m = -\frac{1}{2}\rangle$ Zeeman substate of the $S_{1/2}$ ground state for 60 μs, the ion is prepared for the absorption of 854 nm SPDC photons by coherent excitation to the $|m = -\frac{5}{2}\rangle$ substate of the $D_{5/2}$ metastable manifold (lifetime $\tau_D = 1.17$ s) with a 7-μs-long π-pulse of a narrow-band 729-nm laser. Subsequently, a detection window is open for 2 ms, while photons from the SPDC source are sent to the ion at a rate of $\sim 3500\,s^{-1}$ resonant photons. These photons are set to be σ^+ polarized in the ion's reference frame, thereby exciting the ion to the $|P_{3/2}, m = -\frac{3}{2}\rangle$ level, which corresponds to the transition with the highest absorption probability between $D_{5/2}$ and $P_{3/2}$. During the interaction phase, detection events of 854 nm photons on the APD and 393 nm photons on the PMTs are recorded and time-tagged for their later correlation analysis. After an experimental cycle of cooling, pumping, preparation and interaction has finished, a series of 729 nm π-pulses is used for discrimination of the background, as explained below. The effective single-photon-single-atom interaction time is 68 % of the measurement time, resulting from the 2-ms interaction period per 2.5 ms cycle repetition, and reduced by the periodically applied stabilization of the cavity filters [19, 20].

4 Experimental results

Time correlation histograms of the detections are shown in Fig. 3. The peak around zero time delay corresponds to coincident photon detection on the APD and on one of the PMTs and thereby marks doubly heralded single-photon absorption. In the histogram of Fig. 3a shown in blue, 88.9(6.1) doubly heralded absorption events (on top of 37.1(6.1) background counts) were detected in 10 h (24 540 s interaction time), meaning an average coincidence rate of $3.6 \cdot 10^{-3}$ per second. With an average background of 37.1 counts per 50 ns bin and Poissonian noise, the signal-to-background ratio (SBR) amounts to 2.4 and the signal-to-noise ratio (SNR) is 14.6. Higher time resolution of the coincidence peak using 3 ns binning (Fig. 3b) reveals a symmetric double-sided exponential of about 7 ns 1/e-time, originating from the temporal shapes of the two heralding photons: The shape of the 393-nm herald represents the lifetime of the $P_{3/2}$ state ($\tau_P = 7.2$ ns), while the shape of the 854-nm photons corresponds to the ring-down time of the filtering cavities, designed to match the atomic spectral shape [19, 20]. The red line in the plot represents the expected shape of the correlation function, showing very good agreement with the experimental findings.

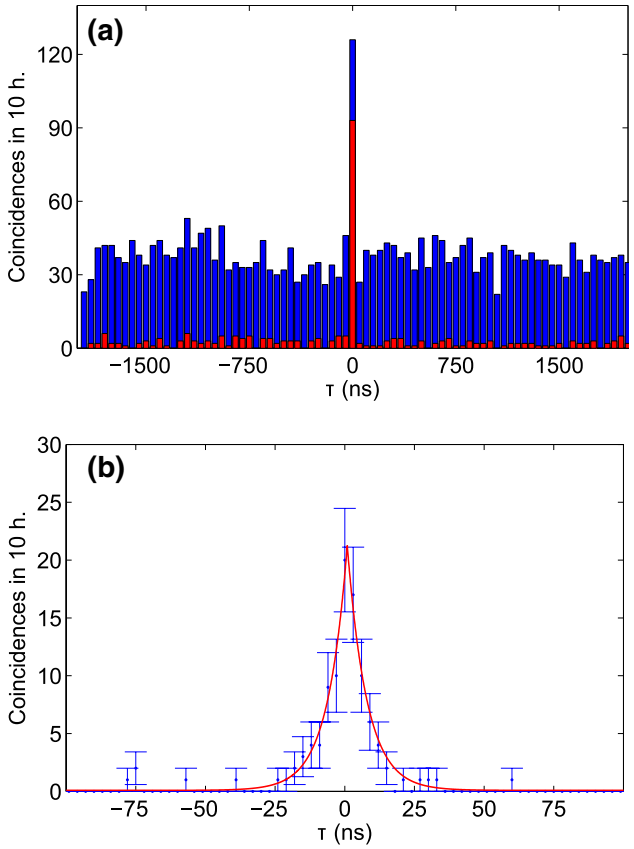

Fig. 3 **a** Time correlation between photon heralds (854 nm) and absorption heralds (393 nm) with 50 ns binning and 10 h total measurement time (24,540 s interaction time). The peak at zero time delay corresponds to doubly heralded single-photon absorption. The *blue and red histograms* show, respectively, the same data without and with additional background reduction (see text). **b** Coincidence peak after background reduction and with 3 ns binning. The *red line* represents the expected correlation function

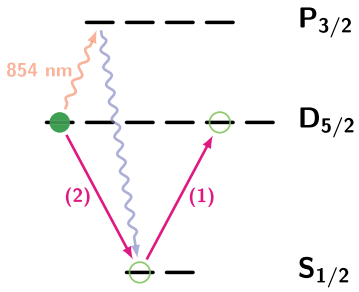

Fig. 4 Experimental scheme and background reduction. The scheme shows the absorption of an 854-nm photon (*wavy red arrow*) and subsequent emission of the photon at 393 nm that heralds the absorption process (*wavy blue arrow*). State measurement for background reduction employs two coherent pulses at 729 nm (*arrows* labeled *1* and *2*) after the single-photon interaction period, and subsequent state-selective fluorescence, as explained in the text

An absorption efficiency per source photon of $\sim 1 \cdot 10^{-3}$ is derived for this experiment, which is higher than the value reported in [4, 15], which we attribute to better preparation of the ion in the $|D_{5/2}, m = -\frac{5}{2}\rangle$ state by using the 729-nm laser.

5 Background reduction

In a further step of the data analysis, a discrimination method is included to reduce the most important source of background, i.e., events where a photon detection on the APD is coincident with a dark count on the PMT. Reducing the background is important since it decreases the measurement time for a target SNR or SBR and increases the fidelity of quantum state transfer schemes [11], by disregarding accidental coincidences that mimic a real absorption. Our method to achieve this is to measure the atomic state after the interaction and consider only those coincidence detections where the atom has indeed made the transition from $|D_{5/2}, m = -\frac{5}{2}\rangle$ to $|S_{1/2}, m = -\frac{1}{2}\rangle$. For this purpose, a series of laser pulses (altogether $\sim$300 μ s) is included in the experimental sequence after the ion–photon interaction phase. First, as shown in Fig. 4, two 729 nm π-pulses are applied to the ion. The first pulse takes the population from $|S_{1/2}, m = -\frac{1}{2}\rangle$ to $|D_{5/2}, m = +\frac{3}{2}\rangle$, which is used as an auxiliary metastable state. This population corresponds to the case where absorption of an 854-nm photon has happened. The following pulse transfers the population from $|D_{5/2}, m = -\frac{5}{2}\rangle$ to $|S_{1/2}, m = -\frac{1}{2}\rangle$, which corresponds to the case where no photon was absorbed. Subsequently, by switching on the cooling lasers for 250 μs, both cases are discriminated. The onset of fluorescence (ion turning bright) means that the ion remained in $D_{5/2}$ during the interaction, and no 854 nm photon was absorbed. In the case where an absorption event happened, the ion will stay dark. By selecting only those 393 nm heralds that go along with a dark ion, we disregard coincidence events triggered by dark counts of the PMTs. While the coherent pulses, which in effect swap the bright and the dark result of the state measurement, do not seem to be strictly necessary for the state discrimination in this particular experiment, they are required in order to protect any quantum information transferred in the absorption event [11], and furthermore, they are part of a more extended method which will be explained below.

The result for the background reduction method as described so far is shown in Fig. 3a, where the red bars correspond to the correlation measurement for the same data as the blue bars, but with postselection of the relevant events. The average background per bin is reduced from 37.1 to 2.3 events, a 16-fold improvement of the SBR, which translates into a 4.1-fold increase in the SNR. After applying the background reduction as described, there are still three cases which account for most of the remaining background counts, namely failed initial state preparation in $D_{5/2}$, failed $D_{5/2} \rightarrow S_{1/2}$ transfer (pulse 2 in Fig. 4) before the state detection, and spontaneous decay from $D_{5/2}$ to $S_{1/2}$. The measured background level is compatible with the probability that these errors occur. Since in these cases the ion will be in $D_{5/2}$ (i.e., dark) during the state-selective fluorescence phase, they will be counted as successful absorptions events when they coincide with a double-herald detection. For the particular measurement of Fig. 3, these "false positives" are not critical, as verified by the low observed background (2.3 events for 50 ns bins, or 0.14 events for 3 ns bins, as in Fig. 3b). Nevertheless, our background reduction method can be extended to also eliminate the background related to failed preparation: By applying a 397-nm laser pulse after the 729-nm preparation pulse, remaining $S_{1/2}$ population can be stored in the $D_{3/2}$ metastable state (see Fig. 2) until the end of the interaction period. The coherent pulses after the interaction period leave the $D_{3/2}$ population untouched, such that it will correctly contribute to the bright cases, i.e., to the no-absorption result of the state measurement.

6 Conclusion

As a summary, we have realized an interface where a single emitted photon heralds the absorption of a single SPDC photon by a single atom. This demonstrates that our protocol for high-fidelity heralded photon-to-atom quantum state transfer, presented in [11], can be extended to single photons. Successful absorption is heralded by the coincident detection, within <50 ns, of the emitted photon at 393 nm and the SPDC partner photon at 854 nm. The time correlation displays the expected double-exponential temporal structure. Furthermore, we presented a way to efficiently reduce background counts by verifying the atomic state after the detection of a herald, which is important as the fidelity of photon-to-atom state conversion may be reduced by dark counts that mimic heralds. With small time overhead and no additional lasers, we decreased the background counts by a factor of 16, leading to a 4.1-fold improvement of the signal-to-noise ratio. These results add another proof-of-principle to constructing a quantum network toolbox with single photons and single atoms.

As an outlook, a brighter entangled photon source will allow us to implement full photon-to-atom quantum state transfer and to work toward entanglement mapping from photon pairs to distant atoms [11, 16, 22]. Moreover, quantum frequency conversion of heralding photons or nondegenerate SPDC pairs with one partner in the telecom range [23] will connect atomic quantum memories and processors

with long-haul quantum communication and with different quantum systems such as solid-state memories and color-center- or semiconductor-based single-photon sources [24], in order to construct a multiplatform hybrid quantum network.

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