Heralded single-photon absorption by a single atom

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Emission and absorption of single photons by single atoms is a fundamental limit of matter-light interaction, manifesting its quantum mechanical nature. As a controlled process, it is also a key tool in quantum optical information technology^{1–3}. Controlled single-photon emission is well advanced^{4–14}; for controlled single-photon absorption by a single atom, proposals exist but only preliminary experimental steps have been taken^{15–19}. Here we report the absorption of single photons by a single trapped ion: employing a photon pair source, detection of the quantum-correlated partner photon heralds the presence of the resonant photon at the atom. We find clear correlations between the detection of the herald and the absorption process in the atom; we also demonstrate polarization control of this process. Our experiment evidences previously unexplored interaction between a single absorber and a quantum light source; with improved control over the coupling, it will open up new avenues in quantum technology.

Single trapped atomic ions provide optimal conditions for quantum information processing, meeting the requirements of high-fidelity state manipulation and detection schemes, as well as controlled interaction of the quantum bits^{20–22}. At the same time, single photons are ideal carriers for transmitting quantum states and distributing entanglement over long distances²³. Establishing quantum correlations between single atoms and single photons allows one to carry out non-local atomic quantum gates mediated by photonic degrees of freedom, a key ingredient in quantum networks^{1,3,24,25}; this was demonstrated recently in experiments that created and employed entanglement between two remotely trapped ions^{26–28}. These operations are based on the underlying entanglement between a single atom and its emitted photons^{12–14}. A fully bidirectional atom/photon interface implies transfer of quantum correlations also in the absorption of a photon; then entanglement can be distributed in a network by making two distant atoms interact with an entangled photon pair^{29,30}. Control over single-photon absorption by means of strong focusing is being addressed in several experiments^{15–19}. We realized a different approach, the interaction between a single trapped ion and resonant heralded single photons from an entangled-photon source based on spontaneous parametric down-conversion (SPDC). We show that the time correlation shared by SPDC photon pairs is preserved in the interaction process, that is, that absorption of a single photon by the ion is marked by the coincident detection of the second photon from the pair. In addition, polarization control of the absorption process is demonstrated after suitable preparation of the ion in Zeeman sub-levels, which is important for future incorporation of our approach in quantum communication schemes. More

Figure 1 | Experimental set-up. A single $^{40}\text{Ca}^+$ ion is confined in a radiofrequency ion trap placed between two HALOs. The ion is laser-cooled by laser light at 397 nm and 866 nm, entering the trap from the side. Lasers at 850 nm and 854 nm are used for state preparation; relevant atomic levels and transitions are schematically represented in the inset. A magnetic field provides a quantization axis along the optical axis of the HALOs. Each HALO collects about 4% of the 397 nm fluorescence photons, which are thereafter detected by two PMTs (for simplicity, only one PMT is shown). One HALO is also used to focus photons from the pair source onto the ion. The photon pair source is based on a narrowband, frequency-stabilized diode laser (master laser) tuned to the $D_{5/2} - P_{3/2}$ transition in $^{40}\text{Ca}^+$ at 854 nm. After frequency doubling (second harmonic generation, SHG), the 427 nm light is focused into a periodically poled potassium titanyl phosphate (PPKTP) nonlinear crystal designed to produce photon pairs around 854 nm by SPDC. The crystal is operated in the type-II collinear phase-matching configuration, such that pairs of orthogonally polarized photons are generated in a single spatial mode. A polarizing beam splitter (PBS) spatially splits the photon pairs. One output mode is coupled through a single-mode fibre and focused onto the single ion through one HALO. In the second output we employ a spectral filtering stage, consisting of two cascaded Fabry–Perot cavities. The transmission frequency of this filter is actively stabilized to the 854 nm master laser, and its transmission bandwidth of 22 MHz is tailored to match the one of the 854 nm atomic transition^{32,33}. The source was operating at a rate of about 3,000 resonant fibre-coupled pairs per second. Photons transmitted through the filter are sent to an APD, and detection events are correlated with the arrival times of blue fluorescence photons at the PMT. Quarter-wave plates (QWP) allow control of the polarization of the SPDC photons and of the 854 nm laser light.

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Figure 2 | Interaction process. **a**, Periodic laser pulse sequence used to control and observe the ion–photon interaction. It first consists of a cooling phase lasting 5 ms followed by state preparation and detection phases of 5 and 60 ms respectively. At the end of the preparation phase, the internal state of the ion is deterministically initialized in sub-levels of the D_{5/2} manifold with magnetic quantum numbers $m = \{(3/2), (5/2)\}$ (or $\{-(3/2), -(5/2)\}$, not shown), which couple to the excited P_{3/2} state through σ^- (σ^+) transitions; thereby the possible absorption of a photon in the detection phase is controlled. An absorption event results in the onset of emission of 397 nm fluorescence by the ion, as illustrated in **b**. **b**, Time trace of fluorescence counts on the PMT with 1 ms time resolution (blue line) and detected trigger photons transmitted through the filtering cavities (red). **c**, Increasing the time resolution to 8 μ s, we note that the photon absorption event, marked by the first detected 397 nm photon, is coincident with the detection of its trigger photon. **d**, The correlation between the two events is statistically verified by a strong peak at zero time delay in the corresponding $g^{(2)}(\tau)$ -correlation function, shown for a total recording time of 30 min.

generally we demonstrate the interaction in a hybrid quantum system involving atoms and quantum light at the most fundamental limit of individual quantum particles.

Figure 1 shows schematically our experimental set-up. It combines a single trapped atomic ion with a continuous-wave source of entangled photon pairs. The $^{40}\text{Ca}^+$ ion is confined and laser-cooled in a linear Paul trap that is placed between two high-numerical-aperture laser objectives³¹ (HALOs) collecting the ion's laser-excited fluorescence. The photon source emits coincident, frequency-correlated photon pairs with orthogonal polarizations in a 200 GHz spectral band centred at the frequency of the D_{5/2}–P_{3/2} electronic transition of $^{40}\text{Ca}^+$, at 854 nm wavelength. For our experiments, the source is designed to interact resonantly with the $^{40}\text{Ca}^+$ ion^{32–34}: the photon pairs are split, and on one of them we impose a frequency filtering to select ‘trigger’ photons that match in frequency and bandwidth the D_{5/2}–P_{3/2} transition. The second, unfiltered photon is coupled to the ion through an optical fibre and one of the HALOs. As the two coincident photons are frequency-correlated³², absorption events should be accompanied by the simultaneous detection of a trigger photon that has passed the filter. The degree of such correlation between the detection of

Figure 3 | Dependence on photon polarization. The ion is prepared in an incoherent superposition of magnetic sub-levels $m = \{(3/2), (5/2)\}$ of the D_{5/2} manifold before exposure to the SPDC photons. For various settings of the SPDC photon polarization at the ion (QWP angle), the number of coincidences between absorption events and heralding photons is measured during 10 min, corresponding to the value of the $g^{(2)}$ correlation function at $\tau = 0$ (circles). The coincidence rate reaches a maximum value of $\approx 5 \text{ min}^{-1}$ for σ^- polarized photons (QWP angle of 0°) and reduces to the background level (red crosses) for σ^+ polarized photons (90°). The error bars correspond to one standard deviation assuming Poissonian counting statistics. A visibility of $90 \pm 1\%$, without subtracting accidental coincidences, is derived from a least-squares sinusoidal fit (solid curve).

trigger photons and the recording of absorption events is controlled by the detuning of the filtering cavities with respect to the atomic resonance, and by the polarization of the photons exciting the ion.

Figure 2a shows the excitation sequence used to control the photon–ion interaction. Each period starts with a time interval during which the motion of the ion is laser-cooled. Excitation parameters of this part, that is, intensities and detunings of the lasers and duration of the pulse, are optimized experimentally to ensure that the ion is well localized and Doppler effects are negligible. Thereafter, the internal state of the ion is prepared using an optical pumping pulse: circularly polarized laser light at 854 nm, propagating along the quantization axis and resonant with the D_{5/2}–P_{3/2} electronic transition, pumps the ion into one of the two outer Zeeman sub-levels of the D_{5/2} manifold, where it remains without scattering further photons. The appropriate helicity of the pump beam allows us to prepare an incoherent superposition of the states either with magnetic quantum numbers $m = \{(3/2), (5/2)\}$, or with $m = \{-(3/2), -(5/2)\}$. Finally, in the detection phase of the sequence, the ion is exposed to the unfiltered, polarization-controlled SPDC photons, while the photodetectors are activated (PMT and APD in Fig. 1).

Absorption events during the detection phase are signalled by the onset of fluorescence, following the transfer of the electronic population from the D_{5/2} to the P_{3/2} manifold and spontaneous decay towards the S_{1/2} level or the D_{3/2} level (with 93.5% and 0.7% probability, respectively³⁵); this then leads to steady blue fluorescence induced by the laser excitation at 397 nm and 866 nm and recorded by the photomultiplier tube (PMT). Meanwhile, the filtered trigger photons are detected on the avalanche photodiode (APD). Figure 2b shows the signal of the two photodetectors to illustrate this process. For sufficiently high time resolution, as shown in Fig. 2c, we note that the arrival time of the first blue photon on the PMT coincides with the detection of a trigger photon on the APD. Hence, the time correlation initially shared by the photon pair has been transferred to the

Figure 4 | Correlation spectroscopy with single photons. The rate of coincidences between absorption events and trigger photon detection is varied by controlling the central frequency of the filter cavities. The red squares (blue triangles) show data taken with the 854 nm pumping laser σ^+ (σ^-) polarized and the SPDC photons set to σ^- (σ^+). Each data point corresponds to the value of $g^{(2)}(\tau = 0)$ obtained after 10 min of acquisition. Poissonian error bars are also shown. The centre frequency of the $D_{5/2} - P_{3/2}$ transition is set to 0 MHz and deduced from fluorescence spectroscopy with a linearly polarized 854 nm laser beam and without optical pumping (green circles). The solid lines show least-squares Lorentzian fits to the data points.

ion–photon system in this particular absorption event. To confirm this coincidence statistically, we compute the second-order time correlation function, $g^{(2)}(\tau)$, between detection events on the two detectors. Figure 2d shows that it exhibits a large peak at zero time delay showing that absorption of a single SPDC photon by the ion is marked by the coincident detection of a trigger photon. In the data presented in Fig. 2d we observed around 2,256 such absorption events, at an average rate of 1.57 s^{-1} (see the Methods section for details). These led to 175 coincidences, 20 being accidental, such that we deduce an overall $\sim 7\%$ probability for the transfer of the temporal correlation from photon pairs to ion and photon. The absorption efficiency with which a resonant photon reaching the ion induces a quantum jump is $> 3 \times 10^{-4}$, in agreement with our expectation (see the Methods section).

With the ion optically pumped into the outer Zeeman sub-levels before the interaction, the rate of coincidences between absorption events and trigger photons is controlled by the polarization of the SPDC photons exciting the ion. We varied the polarization of the photons from right-circular to left-circular, thereby selecting excitation of transitions between $D_{5/2}$ and $P_{3/2}$ that involve a change of the magnetic moment by $\Delta m = -1$ or $+1$, respectively. Transitions with $\Delta m = 0$ are suppressed by exciting along the quantization axis. Figure 3 shows the observed dependence for preparation of the ion in one of the two possible initial states, $m = \{(3/2), (5/2)\}$, from which $\Delta m = 1$ transitions are not possible. The expected sinusoidal variation is found with $90 \pm 1\%$ visibility. Such polarization-selective absorption of a single photon is the key to transferring the polarization degree of freedom from the photon to the ion, and is therefore the first step towards mapping photonic quantum information, including photonic entanglement, onto atomic qubits.

We also studied the influence of varying the transmission frequency of the filtering cavities. It should be noted that in this measurement the ion interacts always with the same broadband SPDC photons. Nevertheless, the frequency correlation between the trigger photon detected on the APD and its partner photon that may be absorbed by the ion makes the coincidence probability of

these events depend on the filter frequency setting. This is shown in Fig. 4, which shows the coincidence counts, $g^{(2)}(\tau = 0)$, as a function of the centre frequency of the cavity filters. The two data sets correspond to the two different initial states of the ion, after optical pumping with σ^+ or σ^- light (see Fig. 2a). Coincidences are observed within the range of frequencies expected from the convolution of filter and transition bandwidth. The two spectra are displaced to higher and lower energies compared with the bare resonance of the $D_{5/2} - P_{3/2}$ transition. This is a consequence of the energy shift that the magnetic sub-states of the $D_{5/2} - P_{3/2}$ manifold experience in the applied magnetic field. The measured splitting of $9(2) \text{ MHz}$ between the centres of the two spectra is compatible, within the statistical error and some experimental unknowns, with the value of $\sim 12 \text{ MHz}$ expected for the applied magnetic field of 5 G . Like the polarization dependence, the control of the coincidence rate through the frequency of the trigger photons is another manifestation of the transfer of photon properties to the atom required in bidirectional photon–atom interfaces.

Methods

In 30 min we carried out about 25,700 experimental cycles of 70 ms duration each, and measured 3,548 quantum jumps. In each cycle the ion was exposed to SPDC light for 60 ms, while at the same time spontaneously decaying with a lifetime of about 1,110 ms. (This lifetime was experimentally determined and is slightly shorter than the literature value of 1,168 ms (ref. 36), owing to spurious quenching processes.) The combined decay probability of $3,548/25,700 = 0.138$ in 60 ms yields a total decay rate of 2.475 s^{-1} , and hence an SPDC-induced absorption rate (more precisely, rate of $D_{5/2}$ to $P_{3/2}$ absorptions that are followed by decay to either $S_{1/2}$ or $D_{3/2}$) of 1.574 s^{-1} . Thus, about 2,256 of the measured quantum jumps are attributed to absorption of SPDC photons, and about 1,292 are due to spontaneous decay. Errors are in the few-percent range.

The background of the correlation function (Fig. 2d), of 19.8 events on average in $8 \mu\text{s}$ time bins, is mainly produced by uncorrelated SPDC photon detections on the APD (that is, heralds with lost partners). APD dark counts of about 30 s^{-1} and PMT dark counts of about 50 s^{-1} contribute negligibly to this background.

The transfer efficiency, that is, the probability that an absorption event is accompanied by the detection of a heralding photon, is determined by the losses of the latter, the main contributions being $\sim 60\%$ fibre coupling efficiency, $\sim 50\%$ filter transmission and $\sim 30\%$ detection efficiency of the APD.

For estimating the absorption efficiency, we divided the measured absorption rate of 1.57 s^{-1} by the approximate rate of $5,000 \text{ s}^{-1}$ fibre-coupled SPDC photons that we send to the ion. The value of 3×10^{-4} is a lower bound because it does not include loss in the optics between the fibre output and the ion, which is hard to measure. The main contributions to this absorption efficiency are (see also ref. 34): the oscillator strength of the $D_{5/2}$ to $P_{3/2}$ transition, of $\sim 6\%$, the average Clebsch–Gordon coefficients for $\Delta m = \pm 1$ transitions out of the prepared mixture of Zeeman sub-states, $\sim 50\%$, the branching ratio for decay of $P_{3/2}$ to either $S_{1/2}$ or $D_{3/2}$, $\sim 94\%$, and the fibre-to-ion imaging. For the imaging, which comprises the mode matching from the fibre output to the absorption characteristic of the atom as well as losses in the steering mirrors, the vacuum window and the HALO, this implies a reasonable value of $\sim 1\%$.

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Author contributions

N.P., F.R., C.S. and M.A. contributed equally to the work. N.P. and A.H. built the photon pair source; F.R., C.S., M.A. and M.H. built the ion trap; N.P., J.H., J.G. and F.D. prepared the experiment and acquired and analysed the data; J.E. planned and supervised the project.

Additional information

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