Tunable ion–photon entanglement in an optical cavity

A. Stute¹, B. Casabone¹, P. Schindler¹, T. Monz¹, P. O. Schmidt²⁻³, B. Brandstätter¹, T. E. Northup¹ & R. Blatt¹-⁴

Proposed quantum networks require both a quantum interface between light and matter and the coherent control of quantum states. A quantum interface can be realized by entangling the state of a single photon with the state of an atomic or solid-state quantum memory, as demonstrated in recent experiments with trapped ions¹⁻³, neutral atoms⁴⁻⁰, atomic ensembles¹⁻³ and nitrogen-vacancy spins¹⁷. The entangling interaction couples an initial quantum memory state to two possible light–matter states, and the atomic level structure of the memory determines the available coupling paths. In previous work, the transition parameters of these paths determined the phase and amplitude of the final entangled state, unless the memory was initially prepared in a superposition state (a step that requires coherent control). Here we report fully tunable entanglement between a single ⁴⁰Ca⁺ ion and the polarization state of a single photon within an optical resonator. Our method, based on a bichromatic, cavity-mediated Raman transition, allows us to select two coupling paths and adjust their relative phase and amplitude. The cavity setting enables intrinsically deterministic, high-fidelity generation of any two-qubit entangled state. This approach is applicable to a broad range of candidate systems and thus is a promising method for distributing information within quantum networks.

Optical cavities are often proposed as a means to improve the efficiency of atom–photon entanglement generation. Experiments using single emitters⁴⁻⁵,³⁰ collect photons over a limited solid angle, with only a small fraction of entanglement events detected. However, by placing the emitter inside a low-loss cavity, it is possible to generate photons with near-unit efficiency in the cavity mode¹⁰⁻¹². Neutral atoms in a resonator have been used to generate polarization-entangled photon pairs⁶⁻¹¹, but this has not yet been combined with coherent operations on the atomic state. Trapped ions have the advantage of well-developed methods for coherent state manipulation and readout¹⁻³,¹³. Using a single trapped ion integrated with a high-finesse cavity, we generate maximally entangled states with fidelities up to 97.4 ± 0.2% (here the number in parentheses indicates the uncertainty in the last digit).

In initial demonstrations of atom–photon entanglement, the amplitudes of the resulting state are fixed by atomic transition amplitudes⁵⁻⁸,¹¹,¹². If the final atomic states are not degenerate, as in the case of a Zeeman splitting, the phase of the atomic state after photon detection is determined by the time at which detection occurs. In contrast, we control both amplitude and phase via two simultaneous cavity-mediated Raman transitions. The bichromatic Raman fields ensure the independence of the atomic state from the photodetection time; their relative amplitude and phase determine the state. Within a quantum network, such a tunable state could be matched to any second state at a remote node, generating optimal long-distance entanglement in a quantum-repeat architecture⁴¹.

A tunable state has previously been used as the building block for long-distance entanglement in a quantum-repeater architecture¹⁴. A tunable state has previously been used as the building block for long-distance entanglement in a quantum-repeater architecture¹⁴. A tunable state has previously been used as the building block for long-distance entanglement in a quantum-repeater architecture¹⁴. A tunable state has previously been used as the building block for long-distance entanglement in a quantum-repeater architecture¹⁴.
nine combinations of ion Pauli bases $[\sigma_x, \sigma_y, \sigma_z]$ and photon polarization bases $[H/V, \text{diagonal/antidiagonal, right/left}]$

In order to measure the ion in all three bases, we first map the superposition of $|D', D\rangle$ onto the $|S, D\rangle$ states [12,13]. We then perform additional coherent operations to select the measurement basis and discriminate between $S$ and $D$ via fluorescence detection [13]. Each sequence lasts 1.5 ms and consists of 800 μs of Doppler cooling, 60 μs of optical pumping, a 40 μs Raman pulse, a 4 μs mapping pulse, an optional 4.3 μs rotation, and 500 μs of fluorescence detection. The probability of detecting a photon in a single sequence is 5.7%; we thus detect on average 40.5 events s$^{-1}$. Note that the photon is generated with near-unit efficiency, and detection is primarily limited by the probability of the photon leaving the cavity (16%) and the photodiode efficiencies (40%).

In a first set of measurements, we choose the case $\alpha = \pi/4$, corresponding to a maximally entangled state $|\psi\rangle$. From the tomographic data, the density matrix is reconstructed as shown in Fig. 2a. Here we have tuned $\Omega_1$ and $\Omega_2$ so as to produce both photon polarizations with equal probability, corresponding to maximal overlap of the temporal pulse shapes of $H$ and $V$ photons (Fig. 2b). In order to demonstrate that the photon-detection time does not determine the phase of the state, we extract this phase from state tomography as a function of the photon time bin (Fig. 2c). Because the frequency difference of the bichromatic fields $A_1 - A_2$ is equal to the level spacing between $|D\rangle$ and $|D'\rangle$, the phase $\phi = 0.25\pi$ remains constant. Further details are given in the Methods section.

Tomography over all time bins yields a fidelity of $F = \langle \psi | \rho | \psi \rangle = 97.4 \pm 0.2\%$ with respect to the maximally entangled state, placing our system definitively in the non-classical regime $F > 50\%$. Another two-qubit entanglement witness is the concurrence [23], which we calculate to be $95.2 \pm 0.5\%$. The observed entanglement can also be used to test local hidden-variable models (LHVs) via the violation of the Clauser-Horne-Shimony-Holt (CHSH) Bell inequality [24]. Entanglement of a hybrid atom–photon system holds particular interest, because it could be used for a loophole-free test of a Bell-type inequality [25]. Whereas LHVs require the Bell observable of the CHSH inequality to be less than 2, we measure a value $2.75 \pm 0.01\% > 2$, where quantum mechanics provides an upper bound of $\sqrt{2}$. We now establish that we can prepare $|\psi\rangle$ with high fidelity over the full range of the Raman phase $\phi$. We repeat state tomography for seven
additional values of the relative Raman phase. As a function of $\phi$, the real and imaginary parts of the coherence $\rho_{ij} = \langle DH|\rho|DV \rangle$ vary sinusoidally as expected (Fig. 3a). The fidelity has a mean value of 96.9 ± 0.1% and does not vary within error bars over all target phases (Fig. 3c).

A second measurement set demonstrates control over the amplitudes $\cos \alpha$ and $\sin \alpha$ of the entangled ion–photon state. After selecting three target amplitudes $\cos \alpha = \{1/\sqrt{2}, 1/\sqrt{3}, 1/\sqrt{8}\}$, we generate the corresponding state by adjusting the Raman field amplitudes, since $\alpha$ is a function of the ratio $\Omega_2/\Omega_1$. The density matrix for each state is then measured. In Fig. 4a, we see that the populations $\rho_{11} = \langle DH|\rho|DH \rangle$ and $\rho_{22} = \langle DV|\rho|DV \rangle$ for the three target amplitudes agree well with theoretical values.

The fidelities of the asymmetric states (Fig. 4b) are as high as those of the maximally entangled states and are limited by the populations, that is, by errors in tuning the Raman fields to match the target values.

Errors in atomic state detection, atomic decoherence and multiple excitations of the atom reduce the fidelity of the atom–photon entangled state by $\ll 1%$. Imperfect initialization and manipulation of the ion due to its finite temperature and laser intensity fluctuations decrease the fidelity by 1%. The two most significant reductions in fidelity are due to dark counts of the avalanche photodiodes at a rate of 36 Hz (1.5%) and imperfect overlap of the temporal pulse shapes (1%).

To our knowledge, the measurements reported above represent both the highest fidelity and the fastest rate of entanglement detection to date between a photon and a single-emitter quantum memory. This detection rate is limited by the fact that most cavity photons are absorbed or scattered by the mirror coatings, and only 16% enter the output mode. However, using mirrors with state-of-the-art losses and a highly asymmetric transmission ratio, an output coupling efficiency exceeding 99% is possible (see Methods). In contrast, without a cavity, a highly asymmetric transmission ratio, an output coupling efficiency exceeding 99% is possible.

In the experiments of refs 3, 6 and 9, although the phase of the cavity output, $\phi$, is in this sense predetermined and can be stored in, or extracted from, a quantum memory in a time-independent manner. The bichromatic Raman process used here provides a basis for coherent atom–photon state mapping as well as one- or two-dimensional cluster state generation.

**Methods Summary**

**Detection and state tomography.** The cavity output path branches at a polarizing beamsplitter into two measurement paths, and the detection efficiencies of these paths are unequal. We compensate for this imbalance by performing two measurements for a given choice of ion and photon basis and sum the results; between these measurements, a rotation of the output waveplates swaps the two paths.

Correlations of the photon polarization and the atomic state are the input for maximum likelihood reconstruction of the most likely states. Error bars are one standard deviation derived from non-parametric bootstrapping assuming a multinomial distribution.

**Time independence.** In the experiments of refs 3, 6 and 9, although the phase of the entangled state is time independent before photon detection, the phase of the atomic state after photon detection is determined only by the start time of the experiment and not by the photon-detection time. The state $|\psi\rangle$ is in this sense predetermined and can be stored in, or extracted from, a quantum memory in a time-independent manner. The bichromatic Raman process used here provides a basis for coherent atom–photon state mapping as well as one- or two-dimensional cluster state generation.

**Methods**

**Detection and state tomography.** The cavity output path branches at a polarizing beamsplitter into two measurement paths, and the detection efficiencies of these paths are unequal. We compensate for this imbalance by performing two measurements for a given choice of ion and photon basis and sum the results; between these measurements, a rotation of the output waveplates swaps the two paths.

Correlations of the photon polarization and the atomic state are the input for maximum likelihood reconstruction of the most likely states. Error bars are one standard deviation derived from non-parametric bootstrapping assuming a multinomial distribution.

**Time independence.** In the experiments of refs 3, 6 and 9, although the phase of the entangled state is time independent before photon detection, the phase of the atomic state after photon detection is determined only by the start time of the experiment and not by the photon-detection time. The state $|\psi\rangle$ is in this sense predetermined and can be stored in, or extracted from, a quantum memory in a time-independent manner. The bichromatic Raman process used here provides a basis for coherent atom–photon state mapping as well as one- or two-dimensional cluster state generation.
Full Methods and any associated references are available in the online version of the paper at www.nature.com/nature.

Received 10 February; accepted 11 April 2012.

1. Cirac, J. I., Zoller, P., Kimble, H. J. & Mabuchi, H. Quantum state transfer and entanglement distribution among distant nodes in a quantum network. Phys. Rev. Lett. 78, 3221–3224 (1997).
2. Kimble, H. J. The quantum internet. Nature 453, 1023–1030 (2008).
3. Blinov, B. B., Moehring, D. L., Duan, L. M. & Monroe, C. Observation of entanglement between a single trapped atom and a single photon. Nature 428, 153–157 (2004).
4. Olmschenk, S. et al. Quantum teleportation between distant matter qubits. Science 323, 486–489 (2009).
5. Volz, J. et al. Observation of entanglement of a single photon with a trapped atom. Phys. Rev. Lett. 96, 030404 (2006).
6. Wilk, T., Webster, S. C., Kuhn, A. & Rempe, G. Single-atom single-photon quantum interface. Science 317, 488–490 (2007).
7. Matsukevich, D. N. et al. Entanglement of a photon and a collective atomic excitation. Phys. Rev. Lett. 95, 040405 (2005).
8. Sherson, J. et al. Quantum teleportation between light and matter. Nature 443, 557–560 (2006).
9. Togan, E. et al. Quantum entanglement between an optical photon and a solid-state spin qubit. Nature 466, 730–734 (2010).
10. Law, C. & Kimble, H. Deterministic generation of a bit-stream of single-photon pulses. J. Mod. Opt. 44, 2067–2074 (1997).
11. Weber, B. et al. Photon-photon entanglement with a single trapped atom. Phys. Rev. Lett. 102, 030501 (2009).
12. Leibfried, D., Blatt, R., Monroe, C. & Wineland, D. Quantum dynamics of single trapped ions. Rev. Mod. Phys. 75, 281–324 (2003).
13. Häffner, H., Roos, C. & Blatt, R. Quantum computing with trapped ions. Phys. Rep. 469, 155–203 (2008).
14. Briegel, H.-J., Dur, W., Cirac, J. I. & Zoller, P. Quantum repeaters: the role of imperfect local operations in quantum communication. Phys. Rev. Lett. 81, 5932–5935 (1998).
15. Mauriz, P. et al. Heralded quantum gate between remote quantum memories. Phys. Rev. Lett. 102, 250502 (2009).
16. Russo, C. et al. Raman spectroscopy of a single ion coupled to a high-finesse cavity. Appl. Phys. B http://dx.doi.org/10.1007/s00340-011-4861-0 (published online, 13 January 2012).
17. Sfute, A. et al. Toward an ion–photon quantum interface in an optical cavity. Appl. Phys. B http://dx.doi.org/10.1007/s00340-011-4861-0 (published online, 13 January 2012).
18. James, D. F. V., Kwiat, P. G., Munro, W. J. & White, A. G. Measurement of qubits. Phys. Rev. A 64, 052312 (2001).
19. McKeever, J. et al. Deterministic generation of single photons from one atom trapped in a cavity. Science 303, 1992–1994 (2004).
20. Keller, M., Lange, B., Hayasaka, K., Lange, W. & Walther, H. Continuous generation of single photons with controlled waveform in an ion-trap cavity system. Nature 431, 1075–1078 (2004).
21. Hijkema, M. et al. A single-photon server with just one atom. Nature Phys. 3, 253–255 (2007).
22. Barros, H. G. et al. Deterministic single-photon source from a single ion. N. J. Phys. 11, 103004 (2009).
23. Wootters, W. K. Entanglement of formation of an arbitrary state of two qubits. Phys. Rev. Lett. 80, 2245–2248 (1998).
24. Clauser, J. F., Horne, M. A., Shimony, A. & Holt, R. A. Proposed experiment to test local hidden-variable theories. Phys. Rev. Lett. 23, 880–884 (1969).
25. Rosenfeld, W. et al. Towards a loophole-free test of Bell’s inequality with entangled pairs of neutral atoms. Adv. Sci. Lett. 2, 469–474 (2009).
26. Economou, S. E., Lindner, N. & Rudolph, T. Optically generated 2-dimensional photonic cluster state from coupled quantum dots. Phys. Rev. Lett. 105, 093601 (2010).
27. Ježek, M., Fiurášek, J. & Hradil, Z. Quantum inference of states and processes. Phys. Rev. A 68, 012305 (2003).
28. Davison, A. & Hinkley, D. Bootstrap Methods and Their Application (Cambridge Univ. Press, 1997).
29. Shore, B. The Theory of Coherent Atomic Excitation (Wiley, 1990).

Acknowledgements We thank J. Barreiro, D. Nigg, K. Hammmer and W. Rosenfeld for discussions. This work was supported by the Austrian Science Fund (FWF), the European Commission (AQUTE), the Institut für Quanteninformation GmbH, and a Marie Curie International Incoming Fellowship within the 7th European Framework Program.

Author Contributions Experiments were performed by A.S., B.C. and T.E.N., with contributions from P.S. to the set-up. Data analysis was performed by A.S., B.C. and T.M. The experiment was conceived by P.O.S. and R.B. and further developed in discussions with A.S., B.B., B.C. and T.E.N. All authors contributed to the discussion of results and participated in manuscript preparation.

Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of this article at www.nature.com/nature. Correspondence and requests for materials should be addressed to T.E.N. (tracy.northup@uibk.ac.at).
METHODS

Detection and state tomography. The cavity output path branches at a polarizing beamsplitter into two measurement paths, and the detection efficiencies of these paths are unequal. We compensate for this imbalance by performing two measurements of photon polarization and atomic-state phase in order to measure the same atom–photon state before photon detection and the phase of the atomic state after detection are independent of photon-detection time. The cavity mirrors have transmission $T_1 = 1.3$ p.p.m. and $T_2 = 1.3$ p.p.m., with combined losses of 68 p.p.m. State-of-the-art combined losses at this wavelength are $L = 4$ p.p.m. (ref. 30). In our cavity, these losses would correspond to an output coupling efficiency of $T_1/(T_1 + T_2 + L) = 71\%$. To improve this efficiency, an output mirror with higher transmission $T_1$ could be used; for example, $T_1 = 500$ p.p.m. corresponds to an efficiency of 99%. The cavity decay rate $\kappa$ would also increase, but single-photon generation with near-unit efficiency is valid in the bad-cavity regime$^{30}$.

We define a model system with bases $\{|S, n\}, |D, n\rangle, |D', n\rangle$, where $n = 0, 1$ is the photon number in either of the two degenerate cavity modes. The excited state has been adiabatically eliminated, so that $g_{eff}$ couples $|S, 0\rangle$ to $|D, 1\rangle$ and $g_{eff}$ couples $|S, 0\rangle$ to $|D', 1\rangle$. After transformation into a rotating frame $U = e^{\pi i (\delta_1/2)} e^{\pi i (\delta_2/2)} e^{\pi i (\delta_1/2)} |D, 1\rangle, 1\rangle$, the Hamiltonian is

$$
\begin{align*}
\langle o_1 \rangle |S, 0\rangle (|S, 0\rangle + o_1 |D, 1\rangle |D, 1\rangle + (o_1 - o_2 |D', 1\rangle |D', 1\rangle + c_1 |1\rangle |1\rangle + g_{eff} |1\rangle |1\rangle + h.c.,
\end{align*}
$$

where $\hbar = 1, \{o_1, o_2\}$ are the state frequencies, $o_1$ is the cavity frequency, and terms rotating at $|o_1 - o_2| \approx g_{eff}$ are omitted$^{30}$. In this frame, the couplings $g_{eff}$ are time-independent, and the states $|D\rangle$ and $|D'\rangle$ are degenerate. Therefore, the phase between $|D, 0\rangle$ and $|D', 0\rangle$ remains fixed during Raman transfer, and the phase between $|D, 0\rangle$ and $|D', 0\rangle$ stays constant after photon detection.

Cavity parameters. The cavity mirrors have transmission $T_1 = 13$ p.p.m. and $T_2 = 13$ p.p.m., with combined losses of 68 p.p.m. State-of-the-art combined losses at this wavelength are $L = 4$ p.p.m. (ref. 30). In our cavity, these losses would correspond to an output coupling efficiency of $T_1/(T_1 + T_2 + L) = 71\%$. To improve this efficiency, an output mirror with higher transmission $T_1$ could be used; for example, $T_1 = 500$ p.p.m. corresponds to an efficiency of 99%. The cavity decay rate $\kappa$ would also increase, but single-photon generation with near-unit efficiency is valid in the bad-cavity regime$^{30}$.

30. Rempe, G., Thompson, R. J., Brecha, R. J., Lee, W. D. & Kimble, H. J. Optical bistability and photon statistics in cavity quantum electrodynamics. Phys. Rev. Lett. 67, 1727–1730 (1991).